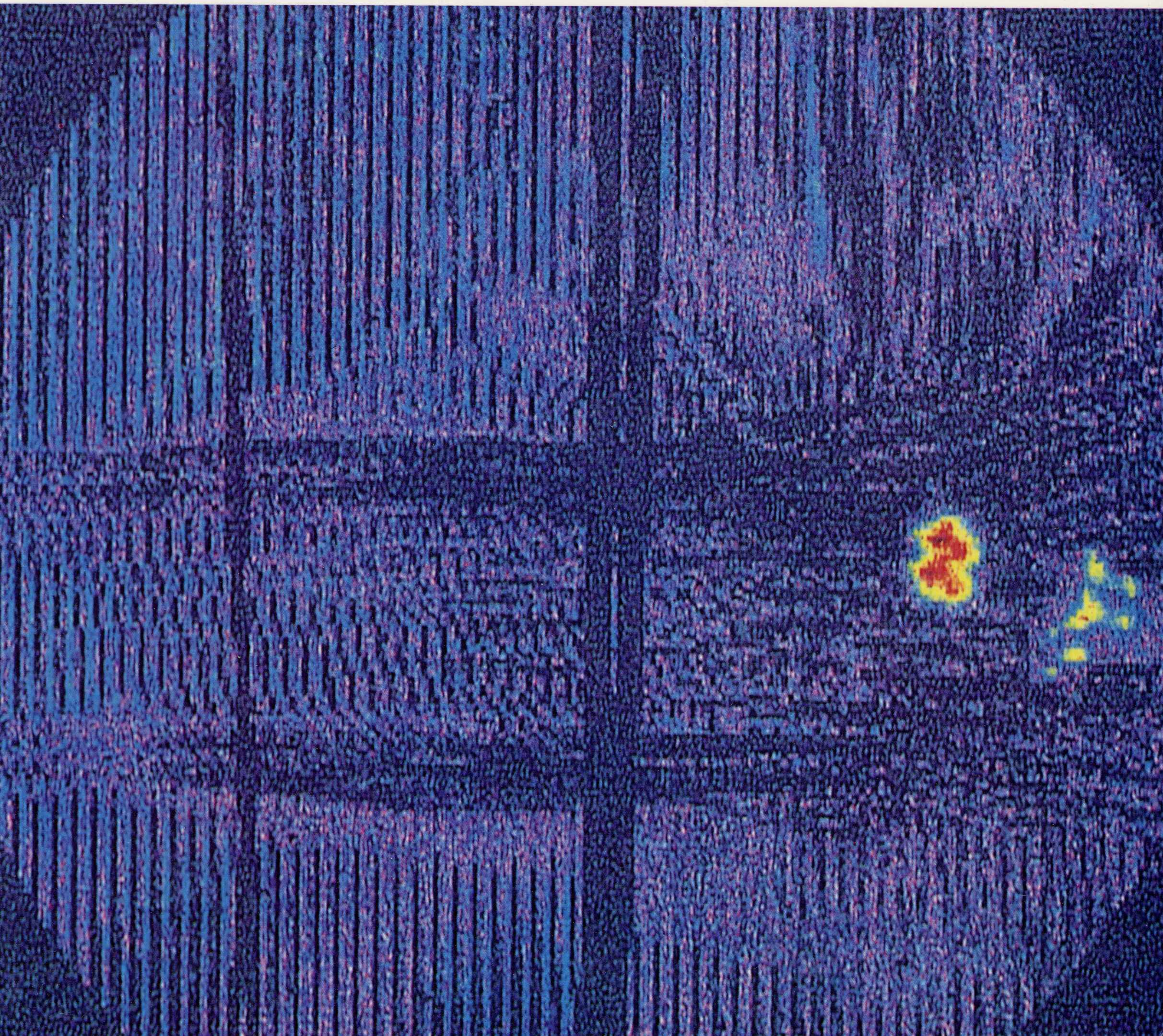




CLRC

COUNCIL FOR THE CENTRAL LABORATORY
OF THE RESEARCH COUNCILS

Central Laser Facility Rutherford Appleton Laboratory Annual Report 1994-95



Central Laser Facility

Rutherford Appleton Laboratory

Annual Report 1994 - 95

Central Laser Facility
Rutherford Appleton Laboratory
Chilton, Didcot
Oxfordshire OX11 0QX
Tel. (0)1235 - 445582
Fax. (0)1235 - 445888

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*The cover shows a Moiré deflectometer
image of a high intensity beam propagating
in a preformed plasma channel (see p.34)*

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W T Toner

This report is the product of a team effort.

The production was organised by Nic Allen and Katharine Hale with Chris Reason overseeing the technical specification.

The Chapter Editors were Peter Norreys, Tony Parker and Mick Shaw, Section Editors were Colin Danson, Edwin Divall, Abdes Djaoui, Chris Edwards, Chris Hooker, Graeme Hirst, Andy Kidd, Andrew Langley, Dave Neely, Steve Rose, Waseem Shaikh, Phil Taday, Sue Tavender, Edmond Turcu and Barry Walsh. The important task of collecting publications was overseen by Chris Hooker with the help of Dave Neely, Sue Tavender and Andy Kidd.

Most of the text has been provided in camera-ready format by the Facility users and they deserve particular thanks for supplying their contributions to our production deadline.

The document has been produced by the RAL Reprographics Section.

PREFACE

This report contains scientific accounts of the work of the Central Laser Facility for the financial year 1994/5. It has been an outstandingly fruitful year and also very eventful. The granting of a Royal Charter to the new Council for the Central Laboratory of the Research Councils and the changes which flow from it are described on the next page. Scientific excellence will continue to be the goal.

Chirped pulse amplification was extensively used in complex experiments on both Vulcan and Sprite throughout the year. Electrons were accelerated to over 45 MeV in a distance of a few tenths of a millimetre; high neutron yields were observed from CPA interactions with deuterated plastic targets; the shortest wavelength harmonic yet to be recorded was produced; a high harmonic was amplified in a laser-produced plasma; channeling by a preformed plasma was observed; high gain was demonstrated at 11 nm wavelength in a recombination X-ray laser. Long pulse experiments also yielded new results: improved X-ray laser gain from the use of separated pulses to form and heat the plasma and the demonstration of foam smoothing of laser ablative acceleration, for example.

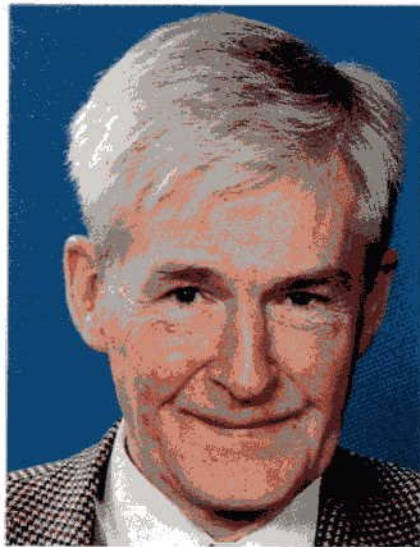
The Titania amplifier module was thoroughly tested in over 1000 shots, coming fully up to design expectations. The start of the construction phase of the laser system was marked by the firing of Sprites last shot as a laser in its own right, on the 31st March, by Paul Williams. He is scheduled to fire the first shot of the new Titania system on 1st April, 1996.

Sprite has delivered 21000 shots since its first, in 1982. Picosecond operation began in 1987. Sprite was the only high power KrF laser to be used for plasma experiments and more recently was the worlds brightest UV laser. It will now become the preamplifier for Titania, designed to be ten times brighter still.

Highlights of the very diverse programme of the Laser Support Facility include high rates of reaction at low temperatures in the gas phase; the observation that vitamins E and C act to prevent as well as cure cell damage; the production of transistors whose 200 nm gates are defined by X-ray lithography; the generation of soft X-rays at an average power of one watt. Recently awarded grants have enabled three new lasers to be purchased for the loan pool, one incorporating OPO technology. A femtosecond OPA system for transient Raman and absorption spectroscopy at RAL is under test.

The increasingly international nature of the programme can be seen from the authorship of the reports, which includes the names of many visiting collaborators. Some reports describe work done abroad by the UK community. Work funded from abroad includes experiments on X-ray lasers and on ceramic film deposition carried out by European visiting groups under the HCM programme and also investigations of KrF gain bandwidth, funded by ONRL, and the development of the plasma X-ray source, funded commercially.

The Longair Review Panel reported to the EPSRC in February following a thorough study of the Facility and its community. It found that outstanding science was being produced in all parts of the facility and recommended that it continue to operate from the Rutherford Appleton Laboratory at the current level of baseline funding. The policy to operate both Vulcan and Titania was endorsed and a proposal to increase the peak power of Vulcan towards the Petawatt level was given a high priority. Additional funding has now been obtained for the first phase of this upgrade. The report is to be published by the EPSRC, which has accepted its recommendations. I would like to express my thanks to Professor Longair and the Panel for the open and constructive manner in which the review was conducted.



It is a pleasure to record the election to the Royal Society of Professors G Pert and I W M Smith, both of whom are contributors to this report, and have long associations with the Facility. I would also like to congratulate Mike Key on the award of the Max Born prize of the Institute of Physics and the Deutsche Physikalische Gesellschaft, for his contributions to laser science. He is on sabbatical this year.

It has been a great privilege to lead the facility, particularly in a vintage year such as this has been, and my thanks are due to everyone in the community and on the staff for a superb performance. There is every reason for confidence that there are many more years of excellent science yet to come.

William T. Toner

W T Toner

INTRODUCTION TO THE CENTRAL LASER FACILITY

C B Edwards

The Central Laser Facility was established in 1977 at the Rutherford Appleton Laboratory to provide advanced lasers and associated instrumentation for use by UK university researchers. Today it is one of the world's leading centres for high power laser research.

Laser Facilities for Users

Two unique large scale, ultra-high intensity laser installations, **Vulcan** and **Sprite**, provide ultra-short pulse laser irradiation facilities at the 10^{19}W.cm^{-2} level, and multi-beam nanosecond pulses at the 3kJ level for a user programme in basic plasma physics research, inertial confinement fusion studies, x-ray lasers, particle acceleration and high harmonic generation.

In the **Laser Support Facility** (LSF) a suite of specialised tunable laser systems provides laser pulses with durations from 100fs to 20ns and a wavelength coverage which spans the spectrum from the ultra violet to the infra-red for studies in photo-chemistry, biology and photo-physics. The LSF specialises in the application and development of advanced diagnostic and analytical techniques including transient Raman spectroscopy.

Stand-alone, commercial laser systems are available on loan from the **Laser Loan Pool** for periods of up to 6 months at the user's home laboratory.

A high repetition rate (100Hz) laser-produced **x-ray source** provides radiation for applications in contact and scanning X-ray microscopy, lithography, and radio-biology.

A vigorous development programme ensures that the facilities maintain their international competitiveness. Titania, the new KrF facility currently under construction at the CLF, will come on line in 1996 as a successor to SPRITE with peak powers in excess of 10TW. An upgrade is underway to the Vulcan laser to enhance its performance to the 250TW level over 2 years. The LSF is currently being extended with the purchase of new lasers funded from grants.

Council of the Central Laboratory of the Research Councils

During the period covered by this report, and following the change in the structure of the Research Councils arising from the White Paper on Science, the Rutherford Appleton and Daresbury Laboratories have been operated as a single entity by the Engineering and Physical Sciences Research Council.

This transitional arrangement ceased on 1st April 1995, when the two laboratories transferred to the ownership of the Council for the Central Laboratory of the Research Council (CCLRC), formed by a Royal Charter approved on 14th December 1994. This year's Annual Report has been issued following the change to the new arrangements, and is presented in the new corporate style of the CCLRC.

Operating within the new council, the CLF is currently negotiating Service Level Agreements (SLA's) with EPSRC, BBSRC etc. which specify facility availability, the level of user support, funding and reporting mechanisms under the new arrangement.

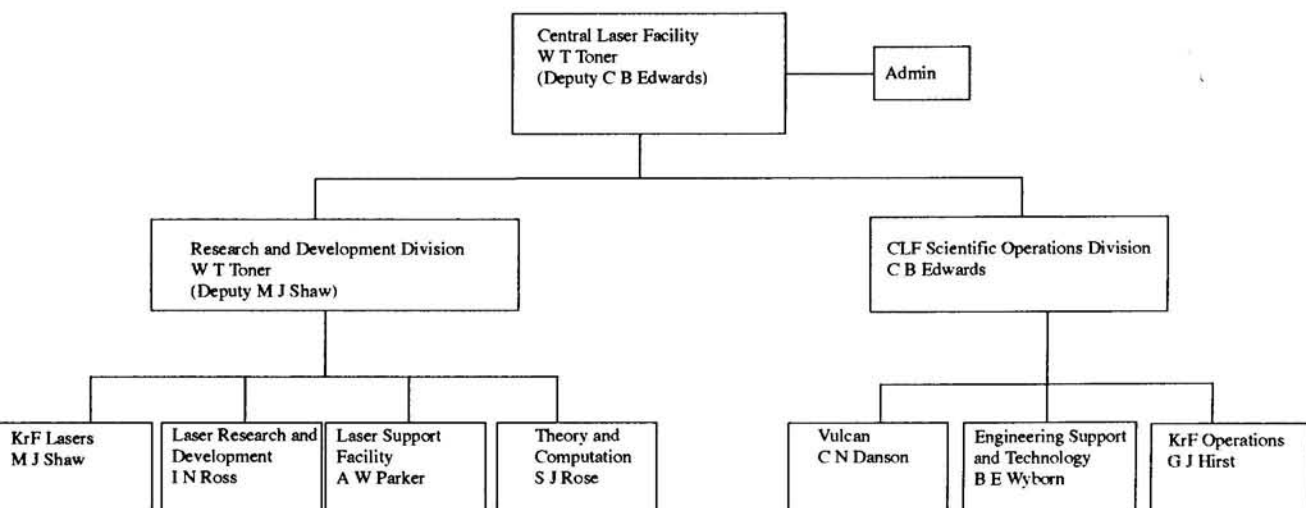


Fig.1 Divisional and Group Structure of the Central Laser Facility

Access to Facilities at the CLF

There are three calls per year for proposals for experiments requiring access to LSF and High Power Laser facilities. Each proposal is assessed by a specialist panel (see Section IV for membership of the LSFP and HPLP) which determines scientific priorities and advises on strategic lines of research for the CLF programme.

Proposals will normally be for research themes already supported by grants from the Research Councils, though a small proportion of beam time is available for proof-of-principle experiments to assist in the preparation of full grant applications. Commercial access is also available on a full costs basis.

Researchers from Europe have the opportunity to bid for beam time at the CLF with funding from the E.C. through a Large Facility Access contract with the C.L.F. Several European groups have already performed extremely successful experiments under this arrangement. More E.U. funded experiments are scheduled in the coming year.

Potential new users should contact the Head of the CLF in the first instance for further information.

Facility Management

The divisional and group structure of the facility staff for the year to March 1995 is shown in Fig.1.

During Prof. Key's absence on sabbatical leave, the CLF has been led by Mr WT Toner who has also headed the Research and Development Division; Dr CB Edwards has acted as deputy Head of the CLF and head of the Scientific Operations Division.

Dr AW Parker has led the LSF; Mr CN Danson and Dr GJ Hirst have led the Vulcan and Sprite Operations Groups respectively.

Facts and Figures

Baseline funding of the CLF from the Research Councils in the FY to March '95 was £3.8 (EPSRC) and £140k (BBSRC) with an additional £358k from Research Council grants.

During the period covered by this report, 16 experiments were scheduled on the Vulcan facility, involving a total of 89 weeks of beam time including experiment set-up. Seven experiments were performed on SPRITE which provided 34 weeks of beam time, plus a further 5 weeks of facility development. In the LSF 48 experiments were performed during a total of 153 weeks of laser access. A total of six years of laser access was made available from the Laser Loan Pool involving 15 U.K. universities.

In terms of scientific publications, the year has been extremely productive with 156 articles published and a further 22 in press. This performance continues the excellent publication record of the facility which is shown in Fig. 2

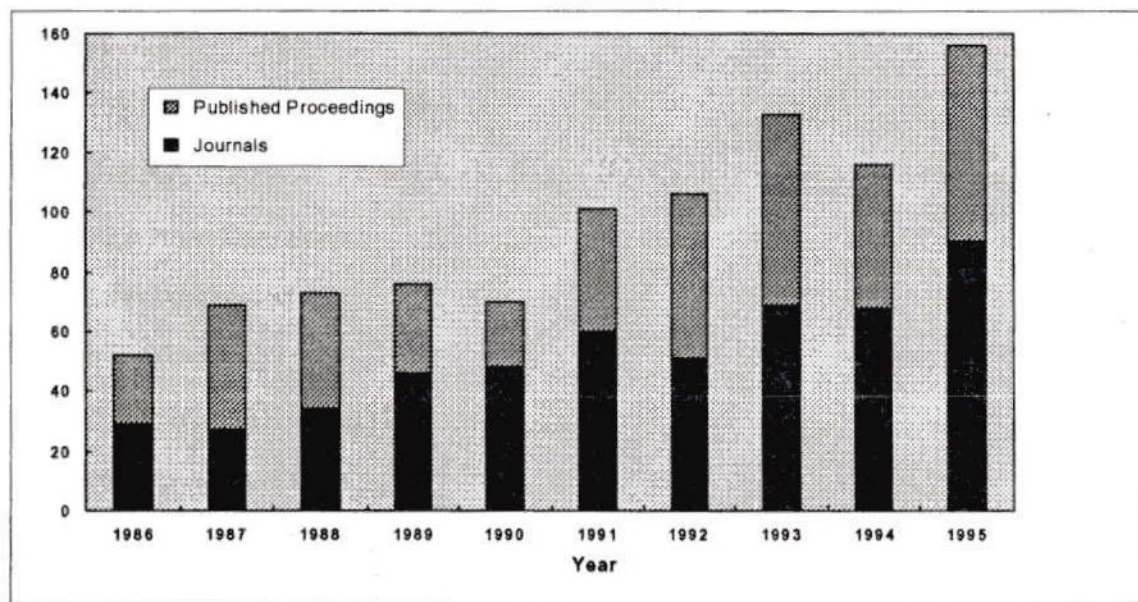


Fig.2: Refereed publications
& published conference proceedings

SPATIAL DISTRIBUTION OF THE C VI LASING GAIN REGION

J Zhang¹, M H Key^{1,2}, D Neely², P A Norreys², G J Tallents³, A Behjat³, C Danson², A Demir³, M Holden³, P B Holden⁴, C L S Lewis⁵, A G MacPhee⁵, G J Pert⁴, S A Ramsden⁴, S J Rose^{2,6}, Y F Shao⁷, F Walsh², Y L You⁸

¹Department of Atomic and Laser Physics, University of Oxford, Oxford, OX1 3PU, UK

²Central Laser Facility, Rutherford Appleton Laboratory, Chilton, OX11 0QX, UK

³Department of Physics, University of Essex, Colchester, CO4 3SQ, UK

⁴Department of Physics, University of York, York, YO1 5DD, UK

⁵Department of Pure and Applied Physics, Queen's University, Belfast, BT7 1NN, UK

⁶Department of Physics & Space Science, University of Birmingham, Birmingham, B15 2TT, UK

⁷Scientific visitor from Institute of Applied Physics and Computational Mathematics, Beijing 100088, China

⁸Scientific visitor from Institute of Nuclear Physics and Chemistry, Chengdu 610003, China

INTRODUCTION

Adiabatically cooled recombination x-ray lasers show promise as efficient producers of high gain at short wavelengths. An exceptionally high gain operation has been demonstrated at 18.2 nm in a C VI recombination laser driven by a 20 J, 2 ps pulses at RAL [1]. In order to obtain information on the uniformity, size and distribution of the high gain region in the lasing plasma, an imaging experiment was conducted.

EXPERIMENT

The experimental set-up was similar to that in the previous work [1]. A 2 psec, 20 TW CPA laser beam at 1.053 μm was generated using the Nd:glass laser VULCAN which was equipped with a chirped pulse amplification (CPA) system [2]. The final pulse recompression grating was located inside the target chamber under vacuum. The recompressed beam of diameter 140x88 mm was incident onto a partially reflecting turning mirror which transmitted 5% of the recompressed pulse for measurements of the laser pulse length, prepulse (contrast ratio), spectral distribution and divergence. An f/4 off-axis parabolic mirror reflected the beam to a spot focus which was imaged by an f/4 off-axis spherical mirror to produce a line focus 7 mm long and 20 μm wide.

It is important to minimise any pre-pulse and post-pulse to avoid pre-forming a plasma and post-heating of the plasma respectively. Pre-forming a plasma may reduce the laser gain and both pre- and post-pulses can be expected to increase the plasma x-ray emission which is not associated with x-ray lasing. For this experiment, a large background contrast ratio of between 10^6 to 10^7 was measured [2]. The pulse length was monitored during the experiment by a single shot third order auto-correlator and was typically in the range of 2 ± 0.6 ps. The incident spatially averaged irradiance in the line focus was $(6 \pm 1) \times 10^{15} \text{ W/cm}^2$.

The targets used in the experiment were 7 μm diameter carbon fibres supported at one end [1]. The fibre targets were positioned with better than $\pm 2 \mu\text{m}$ spatial accuracy and $\pm 1 \text{ mrad}$ angular accuracy using a modified split-field microscope system [1]. The irradiation of the fibre targets was accomplished by placing the fibres in the 20 μm wide line focus using a CW alignment laser. The free end of the fibre target was always placed well within the irradiated line focus length so as to avoid a cold end of the line plasma, which could absorb the x-ray radiation along the fibre length.

A 250 mm focal length concave x-ray multilayer mirror (central wavelength at 18.2 nm, bandwidth of 2.0 nm) [3] situated at a distance 320 mm from the free end of the fibre target imaged at a

magnification of 3.8x the output end of the lasing plasma onto a Kodak 101-04 film with a spatial resolution of $\sim 2 \mu\text{m}$ or an XUV sensitive phosphor that was coupled to a computer controlled CCD detector. A 1.5 μm thick Al filter provided rejection of optical emission.

Off-axis diagnostics included a flat-field grating spectrometer with a 2400 line/mm grating at 45 degree off the fibre axis to record C V and C VI resonance spectra in the range of 2.0 nm - 10.0 nm. These spectra were used to diagnose the characteristics of plasma which could effect the soft x-ray lasing performance during adiabatic expansion [1,4]. The length and uniformity of the plasma were monitored with an x-ray pinhole camera recording an image of emission at wavelengths below 1 nm. A group of Faraday cups/calorimeters gave an estimate of the absorbed energy in the plasma and of the angular variation of ion emission.

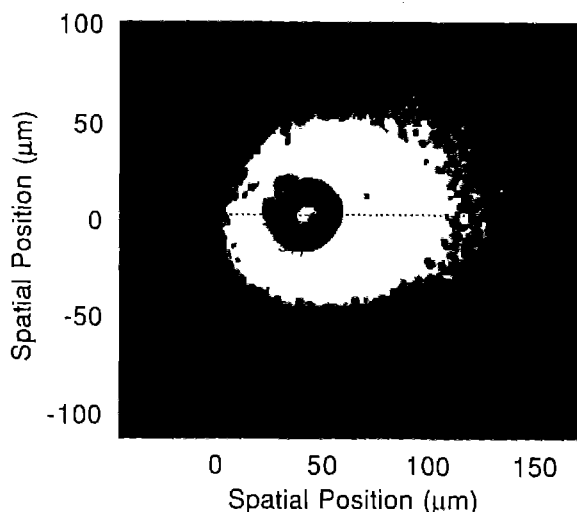


Figure 1. Image showing the intensity distribution of the C VI x-ray laser at the exit plane of the recombining plasma.

RESULTS

An image on a Kodak film of output end of the C VI laser is shown in figure 1. The image is dominated by the 18.2 nm laser line emission rather than continuum as the line emission measured in the experiment [1] is at least an order of magnitude stronger than the continuum emission when integrated over the 2.0 nm multilayer mirror bandwidth [1,4]. The peak intensity region has a structure with a diameter around 35 μm . The emission from the region was so intense that the recording film was bleached. Figure 2 is a horizontal scan along the central line of the output end of

the C VI laser. It is not clear why the part of film recording emission from the most central region ($\sim 5 \mu\text{m}$ diameter) was not bleached. Images recorded on the CCD detector, which had a thicker Al filter, however suggested that the most central region had the strongest emission [1]. It is therefore suspected that the most central region on the film may be somewhat damaged due to the strong emission from this region. The outer region of the image has full widths at half maximum of $120 \mu\text{m}$ and $80 \mu\text{m}$ in the horizontal and vertical directions respectively. This part of film recorded the emission from the outer region was saturated.

The origin of the abscissa in figure 1 is however not the position of the target surface because the spatial position was not properly referenced for the shots on films. The spatial location of the peak emission was estimated from CCD images to be displaced by $100 \pm 50 \mu\text{m}$ towards the laser from the surface of the fibre by comparison with a reference CCD image of continuum emission from a Cu slab target [1]. This displacement is broadly consistent with the simulation prediction within the $\pm 50 \mu\text{m}$ error in the referencing of the spatial location [5].

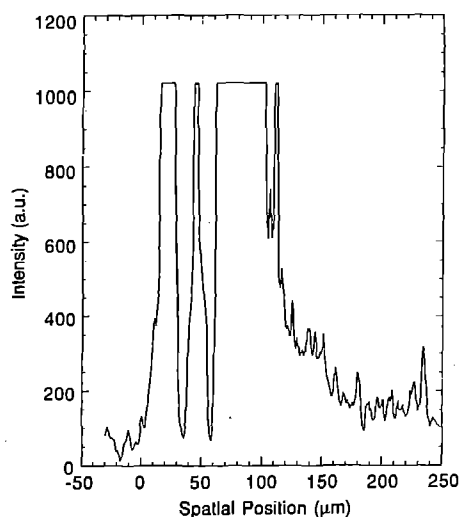


Figure 2. Horizontal scan of the output end of the C VI laser.

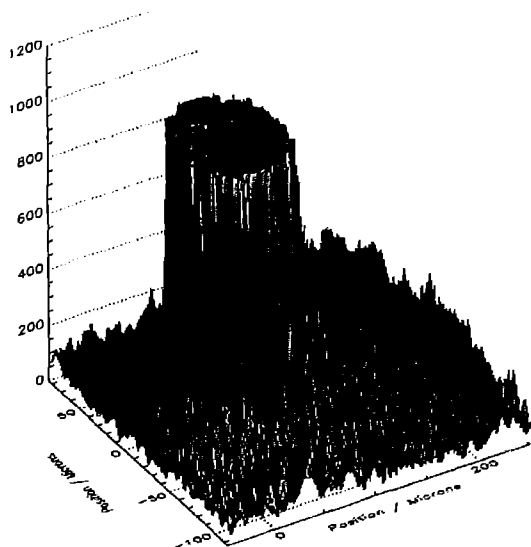


Figure 3. Three-dimensional plot showing the intensity distribution of the C VI laser vs the spatial position at the exit plane. The central region of the peak emission has bleached the film giving an apparent hollow to the "tree trunk" intensity variation.

The intensity distribution of the output end of the laser is shown three-dimensionally in figure 3. The image looks rather like a "hollow tree trunk". The top of the trunk is "cut" by the saturation level of the recording film. The hollow caused by the bleaching of the film represents the highest gain region.

The output energy from the high gain region of the 5 mm long lasing plasma was estimated from the photon flux for bleaching the recording film, solid angle of the detector and reflectivity of the mirror to be larger than 7.8 nJ. Similarly, the output energy from the outer region was estimated from the photon flux required to saturate the film to be larger than 7.7 nJ. The output apertures can be estimated from the image in figure 1 & 2 to be $9.61 \times 10^{-6} \text{ cm}^2$ and $7.54 \times 10^{-5} \text{ cm}^2$ respectively. The duration of the emission was not measured but modelling predicts it to be about 50 ps [6]. The power densities on these two output apertures were then determined from the data to be $1.6 \times 10^7 \text{ W/cm}^2$ and $2.0 \times 10^6 \text{ W/cm}^2$.

The power density from the high gain region was about 2 orders of magnitude higher than the averaged output level (which corresponds to a space-averaged gain coefficient of 12.5 cm^{-1}) of the C VI laser in the previous experiment [1]. This fact suggests that a much higher gain coefficient ($\sim 17 \text{ cm}^{-1}$) has been achieved in this high gain region.

It is of interest to compare this with the saturation intensity of the laser predicted by modelling. If the lifetime of the $n = 3$ level is assumed to be dominated by Lyman β decay, and the line width of the transition is taken to be due to Doppler broadening at 10 eV temperature, the saturation intensity is calculated to be $2 \times 10^9 \text{ W/cm}^2$. The onset of saturation for this laser is predicted to occur when the plasma length is about 8 mm. This onset of saturation for a small increase in length reflects the fact that the peak gain is higher than the apparent gain so that with increasing length there should be an increase of apparent gain. A saturated laser of this type would generate a peak power of about 5 kW with a pulse length of about 50 ps and energy of about 200 nJ.

CONCLUSION

In conclusion, we have measured the near field distribution of the C VI recombination x-ray laser in the exit plane. The output end of the C VI laser has been imaged with a high spatial resolution multilayer mirror system. The highest gain coefficient of 17 cm^{-1} has been found in the central region with diameter of about $35 \mu\text{m}$. The output power density from this high gain region of a 5 mm long plasma was estimated from experimental data to be $1.6 \times 10^7 \text{ W/cm}^2$.

ACKNOWLEDGEMENT

The authors would like to thank the VULCAN laser operations, the target preparation and engineering groups of the Central Laser Facility for their help and cooperation.

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- [2] C D Danson, L J Barzanti et al, Opt. Commun., **103**, 392 (1993).
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- [4] J Zhang and M H Key, Appl. Phys. B58, 13 (1994).
- [5] A V Borovskiy, P B Holden et al, J. Phys. B 25, 4991 (1992).

AMPLIFICATION OF XUV HARMONIC RADIATION IN A GALLIUM AMPLIFIER

T. Ditmire¹, M. H. R. Hutchinson², M. H. Key³, C. L. S. Lewis⁴, A. MacPhee⁴, I. Mercer²,
D. Neely³, M. D. Perry¹, R. A. Smith², J. Wark⁵ and M. Zepf⁵

¹Laser Program, Lawrence Livermore National Laboratory, U.S.A.

²Imperial College of Science, Technology and Medicine, London.

³Rutherford Appleton Laboratory, Chilton, Didcot, Oxon.

⁴Queens University of Belfast, Belfast, N. Ireland.

⁵University of Oxford, Oxford.

INTRODUCTION.

In recent years the development of sources of intense, coherent XUV radiation has been carried out using two distinct approaches. With intense, short optical pulses from chirped pulse amplification (CPA) lasers [1], ultra-high harmonics of the fundamental have been observed [2,3] extending to wavelengths below 7.0 nm [4]. In the so called plateau region of harmonic generation, (where successive orders have similar intensities) conversion of optical energy to xuv with an efficiency up to 10^{-7} has been achieved [5]. With weak focusing conditions and intensities low enough that phase matching is not destroyed by plasma formation, these harmonics appear to exhibit a high degree of spatial coherence [6,7]. Since harmonics are produced only during the laser pulse, the xuv emission has a duration determined by the driving laser. As a result xuv pulses can be produced at any length from a few tens of femtoseconds to near 100 picoseconds.

Higher pulse energy and shorter wavelength radiation than currently obtainable from high harmonics has been demonstrated with electron collisionally pumped laser schemes in neon-like [8] and nickel-like [9] ions and in recombination schemes [10]. These sources have produced amplified spontaneous emission in the range 43-400 Å with output energy as high as a millijoule at 21 nm [11]. However they generate relatively long pulses typically on the order of a few hundred picoseconds. Due to the spontaneous nature of the emission and the lack of a cavity, they also have low spatial coherence, although this issue has to some extent been addressed by using injection seeding of light from one plasma into another.

EXPERIMENT AND RESULTS

Here we report the first observation of amplification of a high harmonic in a collisionally pumped X-ray amplifier. This was achieved by injecting harmonic radiation from a frequency doubled Nd:Glass CPA laser into a Neon-like Gallium plasma. The 21st harmonic of the frequency doubled Nd:Glass laser was tuned to give a spectral overlap with the J=2-1 transition of GaXXII at a wavelength of 25.11 nm. For these experiments, the Vulcan laser was configured to operate with one arm in CPA mode for the generation of harmonics and three arms to provide long pulse pumping beams for the X-ray amplifier. A 1 nJ, 850 fs duration pulse was stretched in a dispersive delay line, amplified to 10-30 J in one arm of Vulcan, and then recompressed using a single-pass, two-grating system to a final pulse length of 1.3 ps. Light entering the amplifiers was tuned by spectral windowing in the Fourier plane of the grating stretcher, and this, in conjunction with gain pulling in the glass amplifier chain determined the output spectrum of the amplified pulse. This allowed us to wavelength tune the CPA laser and hence the high harmonics to give a spectral overlap with the x-ray laser line. The beam was then frequency doubled in a 1 cm thick KDP crystal giving a conversion efficiency of 30% and up to 5 J of second harmonic light. This second harmonic beam was then focused with an $f = 3.5$ m singlet lens to an approximately 4x diffraction limited spot with a peak intensity of $\sim 5 \times 10^{15}$ W cm⁻². At focus this beam traversed 1 mm of helium gas at a density of $\sim 2 \times 10^{19}$ cm⁻³ produced by a hypersonic gas jet.

To produce the x-ray amplifier gallium stripe targets up to 18 mm long were irradiated from one side with three overlapped line foci, and particular care was taken to ensure that both ends of the stripe were well illuminated to avoid absorption of the input or output radiation. The separation between the gas jet and the entrance to the gallium amplifier was 20 mm, which is comparable to the Rayleigh range of the harmonic beam (~ 15 mm). This allowed for good coupling of the harmonics into the approximately 80 μ m diameter gain region of the plasma, while being far enough removed to avoid preionization of the gas jet by blackbody radiation. The experimental arrangement for the target chamber is shown below. (Figure 1)

The harmonic beam was analysed with an axial 1200 l/mm flat-field grating spectrometer placed 2 m downstream from the gas jet to avoid damage from the driving laser. Spectra were recorded with either a calibrated time integrated CCD camera or an x-ray streak camera. For x-ray laser or amplified harmonics shots, the instrument was shifted slightly off axis in the horizontal plane to allow for refraction of x-rays by the density gradient in the Gallium plasma.

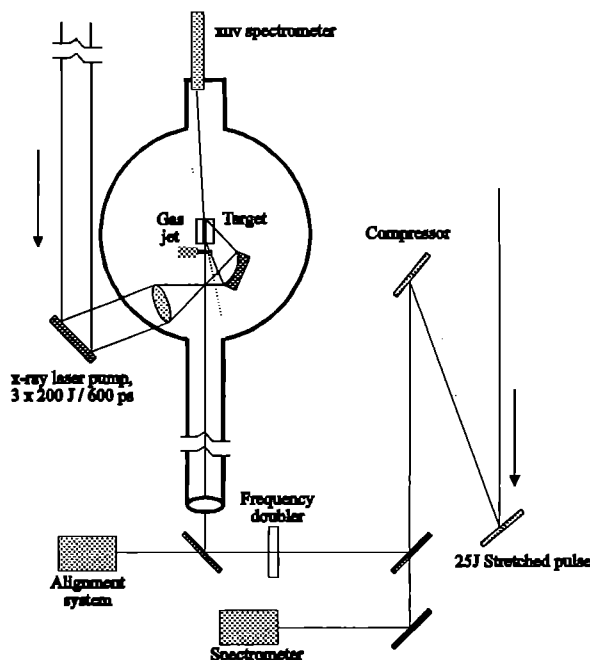


Figure 1. Experimental arrangement

In time integrated measurements the single shot energy contained in the 21st harmonic was found to be between 0.5 and 2 nJ. From this the efficiency of harmonic production is estimated to be $\sim 10^{-9}$, which is substantially less than has been previously demonstrated for optimum conditions in the plateau region. We believe that spatial modulation of the pump beam degraded the phase matching of the harmonics and result in a much greater divergence and lower

conversion efficiency than achieved with a Gaussian profile. As well as the 21st harmonic which we wish to amplify, the spectrometer covers sufficient wavelength range to show adjacent harmonic orders at the same time. These should remain unamplified when traversing the Ga plasma and thus act as an intensity fiducial. Figure 2 compares different single shot harmonic spectra demonstrating the good shot to shot reproducibility in relative intensity of harmonic orders.

Time Integrated Harmonic Spectra

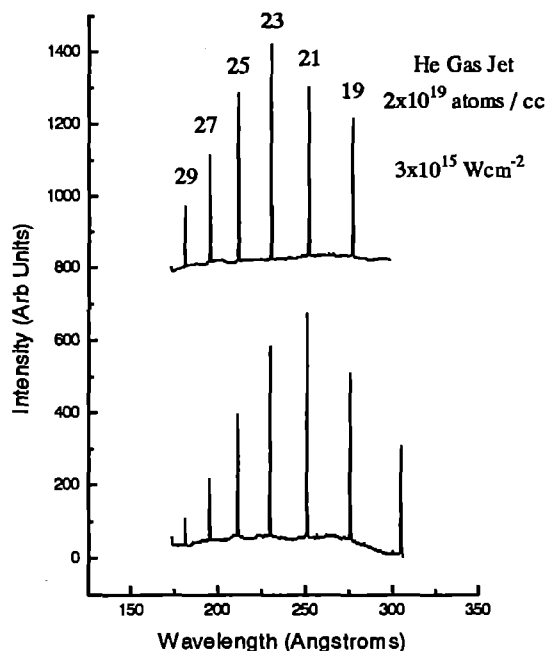


Figure 2. Comparison of single shot harmonic spectra

Using the axial spectrometer the length dependence of the gallium J = 2-1 laser line at 25.11 was measured for plasmas up to 18 mm showing gains of approximately 3cm^{-1} . This is the first time that the Ga XXII laser system has been investigated in any detail and figure 3 shows a plot of the exponential growth of the j = 2-1 at 25.11nm.

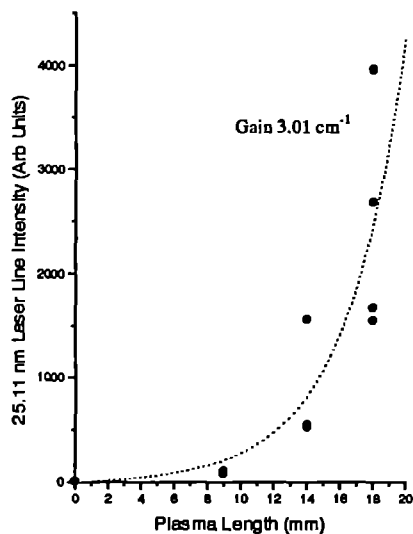


Figure 2. Plasma length study of the Ga XXII laser line

The time integrated output of the plasma in the 25.11 nm spectral region is substantially greater than that of the harmonic, and to distinguish the amplified 21st harmonic from the longer lived background plasma emission we used a streak camera to time resolve the spectra. Figure 4 shows a time trace taken at 25.11 nm from a combined harmonic / x-ray amplifier shot showing a clear spike in time when the harmonic was injected.

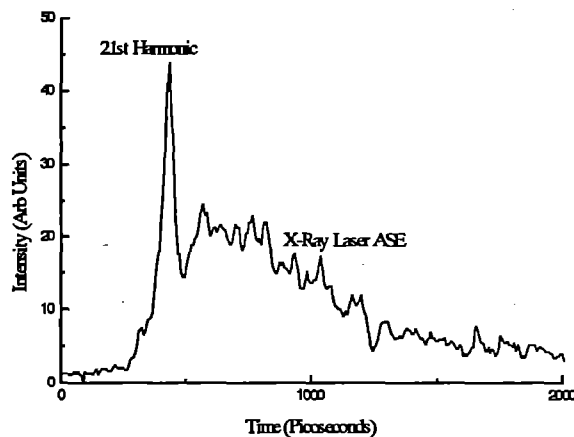


Figure 4. Time history of 25.11 nm signal

Figure 5 compares the spectra from a time resolved harmonic shots with no plasma present, and after traversing an 18 mm Ga plasma. The harmonic was tuned to give optimum spectral overlap with the 25.11 nm laser line. These two spectra were averaged over a 30 ps time window which was the resolution of the streak camera. The instrument and filter response have been deconvoluted and the background, found from averaging time slices 30 ps either side of the harmonic signal, has been subtracted. There is a clear enhancement of approximately three of the 21st harmonic relative to the 19th and 23rd harmonics in the case where the harmonic propagates through the amplifying plasma.

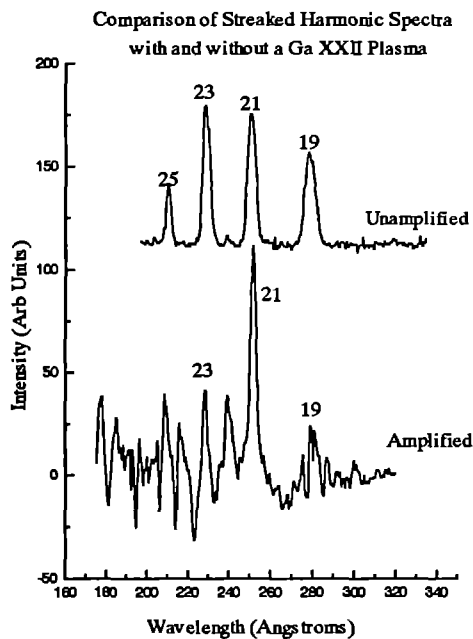


Figure 5

We can estimate the expected gain for the 21st harmonic based on the measured 3 cm^{-1} gain coefficient for the 25.11 nm GaXXII laser line and the coupling efficiency between the harmonic and the plasma amplifier. For complete coupling between the harmonic and the 18 mm x-ray amplifier, we would expect a small signal gain of $\exp[\alpha l] \sim 220$. However, this will be reduced by both the spectral and spatial overlap of the harmonic with the gain region of the plasma.

Based on previous measurements of harmonic linewidths for 140 fs pulses at 825 nm we estimate the linewidth of the 21st harmonic to be $\sim 0.01 \text{ nm}$ (FWHM) which is well below the resolving power of our XUV spectrometer. Measurements of the linewidths of similar collisionally pumped neon-like x-ray laser schemes suggest that the linewidth is predominately Doppler broadened with small corrections for Stark and lifetime broadening. From this work we estimate that the linewidth of the Ga XXII, $J=2-1$ transition is $\sim 0.005 \text{ nm}$. Thus, with optimum spectral overlap of the harmonic to the laser line, we calculate that the coupling efficiency will be $\sim 25\%$ assuming a convolution of Gaussian spectral profiles. This lowers the maximum gain to ~ 50 . The observable harmonic gain will be further reduced because only a fraction of the harmonic energy can be coupled into the plasma since the spatial extent of the harmonic exceeds that of the gain region. We estimate from far field measurements that the beam waist of the harmonics at the plasma is $\sim 300 \mu\text{m}$ while the diameter of the gain region is only $\sim 80 \mu\text{m}$. This will further reduce the observable gain to ~ 3.5 which is in good agreement with what we measure for optimum shots.

In conclusion we have demonstrated the injection and amplification of harmonic radiation in a collisionally-pumped soft x-ray amplifier. A net energy gain of three was achieved for the 21st harmonic of a frequency doubled Nd:glass laser in a neon-like gallium plasma at 25.11 nm. The measured gain is in good agreement with that calculated when the spatial and spectral overlap of the harmonic with the amplifier is accounted for. In an optimized experiment using a more tunable source and better mode matching between the harmonic source and amplifier considerably higher amplification could be possible. We believe that this can provide a method of achieving short pulse operation of soft x-ray lasers with good spatial coherence for use in applications such as ultra-short pulse probing of laser plasmas. In addition this technique can be used for direct measurements of the spatial and temporal distribution of gain in x-ray laser plasmas.

We gratefully acknowledge the efforts of the Rutherford Appleton Laboratory target preparation group and the Vulcan laser operators. This work was supported by an EPSRC grant and the US Dept. of Energy under contract W-7405-Eng-48. R. A. Smith is supported by an EPSRC Advanced Fellowship.

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USING LOW AND HIGH PREPULSES TO ENHANCE THE J=0-1 TRANSITION AT 19.6nm IN THE Ne-LIKE GERMANIUM XUV LASER

G.F. Cairns¹, C.L.S. Lewis¹, M.J. Lamb¹, A.G. MacPhee¹, D. Neely⁴, P. Norreys⁴, M.H. Key^{2,4},
S.B. Healy³, P.B. Holden³, G.J. Pert³, J.A. Plowes³, G.J. Tallents⁵ and A. Demir⁵

¹ Department of Pure and Applied Physics, Queen's University of Belfast, Belfast BT7 1NN.

² Clarendon Laboratory, University of Oxford, Oxford, OX1 3PU, UK

³ Department of Physics, York University, York, YO1 5DD, UK

⁴ Central Laser Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxon, OX11 0QX, UK

⁵ Department of Physics, University of Essex, Colchester, CO4 3SQ, UK

INTRODUCTION

Significant progress has been made in our efforts to produce efficiently a bright, monochromatic, and highly coherent XUV laser beamline below 25 nm. This has been achieved by using prepulses and multi-pulses with the Ne-like ions of Ni, Zn and Ge, to enhance the J=0-1 ($2s^2 2p_{1/2} 2p_{3/2}^4 3p_{1/2}$ J=0 to $2s^2 2p_{1/2} 2p_{3/2}^4 3s$ J=1) transition^{1,2,3}. Of particular interest has been the dramatic increase in the lasing output of the J=0-1 line at 19.6nm in the germanium XUV laser with both low (~0.02%) and high (~2%) level prepulses. The levels are quoted as the fractional energy in the prepulse compared with that in the main pulse arriving on target. A summary is given here of the main results from two experiments using different prepulse conditions and the comparison made between them. Modelling has improved significantly our understanding of the mechanisms involved in these schemes with general agreement between the model predictions and the experimental observations⁴.

EXPERIMENTAL DETAILS

The experiments were carried out in Target Area East using the Vulcan glass laser at 1.06 μm wavelength. A more detailed description of the setup has been previously reported including how the prepulses were generated. Given here in table 1 is a summary of the main experimental parameters used in both cases. To allow for measurements of the effective gain observed on both the J=0-1 (19.6 nm) and J=2-1 lines (23.2 & 23.6 nm), target lengths of 9 and 14mm were used as well as those given in the table.

RESULTS AND DISCUSSION

A significant enhancement of the J=0-1 line relative to the J=2-1 lines was observed when the prepulse was applied in both cases. The temporal characteristics were similar, with the J=0-1 line lasing more intensely than the J=2-1 lines, earlier in time and for a considerably shorter duration. Evidence from the beam pointing, which was reduced with the prepulse, suggested a relaxation of the electron density gradients within the plasma column. In summary for both prepulse levels, -the onset of lasing tended to occur earlier in time compared to the no prepulse case, -the J=0-1 lasing occurred earlier in time compared to that on the J=2-1 lines with the J=0-1 line peaking typically 300ps before that of the J=2-1 lines, whose peak in turn corresponded closely to the peak of the drive pulse, -the FWHM of the J=0-1 line was much shorter with the prepulse, ~100ps, compared with J=2-1 lines whose FWHM remained similar to that with no prepulse at typically 500ps, -the average output power in the J=0-1 line was nominally ~6X that of the average of the J=2-1 lines, -the peak radiant density ($\mu\text{J}/\text{st}$) on the

PARAMETERS	LOW PP	HIGH PP
Prepulse relative intensity (%)	~ 0.02	~ 1.8
Target Length (mm)	22±0.5	18±0.5
Target Width (μm)	100±10	120±10
Drive pulse duration FWHM (ps)	700±50	825±125
Average Irradiance ($\times 10^{13} \text{Wcm}^{-2}$)	1.8±0.4	1.6±0.5
Delay after prepulse (ns)	5	3.2
Detector	Ilford Q plates	Kodak 101.04

Table 1 Main experimental parameters

J=0-1 line was increased to a level similar to that of the J=2-1 lines, - the beam pointing was reduced from typically 10mrad without any prepulse to a few mrad with the low prepulse and 5-9 mrad with the high prepulse.

It was necessary to adjust the results from each prepulse experiment to allow for a comparison, taking into account the variation of the gain with irradiance on target. The gain dependence on irradiance for the J=0-1 line was assumed similar to that for the J=2-1 lines which has been previously characterised⁶. The results for the low case were also adjusted for their longer target lengths of 22mm. To minimise the effects of diffraction gratings, reflection filters, transmission filters and detectors, which were also different between the experiments, the results were normalised to the peak radiant densities and powers observed when no prepulse was applied in the two cases. The results are given in table 2. They show that the high (~1.8%) prepulse delivered at 3.2ns was more effective than the low (~0.02%) prepulse delivered at 5ns in increasing the output on the J=0-1 line at 19.6nm. Its peak energy was estimated to increase by a factor of ~4.5X and its peak power by a factor of ~40X. The output power for the J=2-1 lines was also increased with the high level of prepulse. There was ~2x more radiant density and ~7x more power for both J=0-1 and J=2-1 lines with the high prepulse compared with the low prepulse. Due to the limited data set and the inherent fluctuations from shot to shot, it was not possible to deduce what effect if any the low prepulse had on the J=2-1 lines output. The overall observations supported by modelling suggest that the prepulse leads to a) enhanced absorption of the drive energy, b) increase in the volume of the gain zone, and c) a relaxation in the density gradients. This provides the necessary high density and temperature conditions for accessing gain on the J=0-1 transition.

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	BOTH NO PP	LOW PP	HIGH PP	RATIO PP/NoPP LOW	RATIO PP/NoPP HIGH	RATIO HIGH PP /LOW PP
Peak J=0-1 radiant density	0.2±0.1	0.5±0.2	0.9±0.3	2.5	4.5	1.8
Peak J=2-1 radiant density	1.0	0.4±0.2	0.7±0.2	0.4	0.7	1.8
Average (0-1) power	0.2±0.1	1.4±0.8	8±5.5	7.0	40.0	5.7
Average (2-1) power	1.0	0.2±0.1	1.7±0.8	0.2	1.7	8.5
Rad. Density ratio E(0-1)/E(2-1)	0.2	1.25	1.3			
Power ratio P(0-1)/P(2-1)	0.2	7	5			

Table.2 A summary of the normalised results for the low and high prepulse cases where scaling of gain with irradiance and different target lengths have been taken into account. A unit of power, as scaled to an 18mm long target irradiated at $1.6 \times 10^{13} \text{Wcm}^{-2}$ with no prepulse corresponds to 0.035kW. Peak radiant density for the same conditions are in units of $26 \mu\text{J}/\text{st}$. The large uncertainties are primarily due to the inherent shot to shot fluctuations in output coupled with a small sample number of shots for each case.

EXPERIMENTAL STUDY OF A SCALING CONCEPT FOR INCREASING THE GAIN EFFICIENCY OF THE NI-LIKE TA LASER USING A KRF RAMAN LASER DRIVER.

C G Smith¹, M H Key^{1,2}, D Neely², P A Norreys², G Cairns³ and C L S Lewis³

¹ Clarendon Laboratory, Oxford

² Rutherford Appleton Laboratory

³ Queen's University Belfast

INTRODUCTION

There is a need to reduce the driver energy of XUV lasers to enable the development of a saturated laser at a wavelength suitable for holography of biological specimens.¹ The best candidate XUV laser is Ni-like Ta at a wavelength of 4.48 nm^2 , but it has not so far been possible to drive this laser to saturation because the required energy ($>10 \text{ kJ}$ by extrapolation from ref. 2) exceeds the capability of current drivers. One possible solution to this problem is to reduce the energy requirement by taking advantage of scaling characteristics which favour the use of a shorter duration pulse at a shorter wavelength in a narrower line focus. The basis of this scheme has been described elsewhere³, and it has been shown that the KrF laser pumped Raman laser is particularly suitable for this purpose. Estimates from the scaling analysis suggest the possibility of saturated output from the Ta laser with less than 700 J of driver energy at 268 nm in a 125 ps pulse and a 30 micron wide line focus at 10^{15} Wcm^{-2} . These parameters are close to the capability of the Titania laser system currently being constructed at RAL.⁴ Motivated by this attractive possibility we have conducted an experimental study of plasma production in a short line focus under relevant conditions using the available Sprite KrF laser which delivers up to 12 J in Raman laser mode of operation.⁵ The aim of the experiment was to verify the scaling concept by X-ray spectroscopy showing the production of the necessary degree of ionisation and excitation in the plasma and by X-ray imaging showing the production of the required narrow line focus.

THE EXPERIMENT

This experimental arrangement is shown as figure 1.

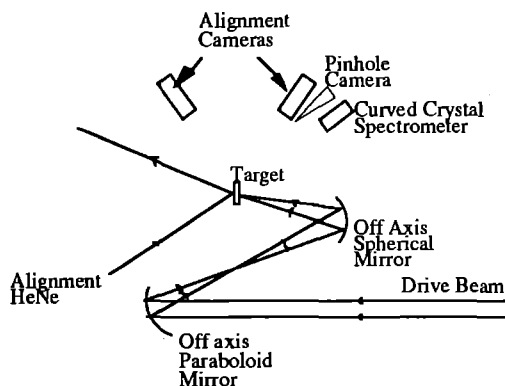


Figure 1. The experimental arrangement.

The 268 nm Sprite Raman beam of 10 cm diameter was focused to a line using an $f/4$ off axis parabola to produce a point focus which was imaged astigmatically by an off axis spherical mirror to give a 500 micron long line focus. The optical width of the line focus on the target was varied by defocusing, the minimum width being less than 10 microns . The targets were solid slabs or thin films of Ta or Ag (Ag being a Neon-like analogue of the Ta laser²). The targets were positioned using highly magnified images from orthogonal telemicroscopes and aligned in angle using reflection of a reference He-Ne laser beam. The pulse duration of the Raman beam was either 25 ps or 60 ps .

A pinhole camera equipped with a 12 micron pinhole and filtered by 25 micron Be and 400 micron mylar, imaged the line focus at an angle of 45 degrees in the plane of incidence of the drive beam. The image was recorded on film (Industrex C) at $4\times$ magnification.

A crystal spectrometer using a KAP crystal recorded the resonance lines from the plasma in the region $5.5 \text{ \AA}$ to $8 \text{ \AA}$; increased resolution was achieved by the use of a convex configuration (30 cm radius). The spectrometer was operated with either a $300 \text{ \mu m}$ slit (which gave an effectively spatially integrated output) or a $100 \text{ \mu m}$ slit which gave limited spatial resolution transverse to the line focus. The spectra were recorded on a CCD detector connected to a computerised image acquisition system.

RESULTS

The pinhole camera was used to record the width of the X-ray emitting line focus (FWHM) as a function of the displacement of the focal plane relative to the target as shown in figure 2 for 60 ps pulses on Ta. Similar plots were obtained with 25 ps pulses and when Ag was substituted for Ta.

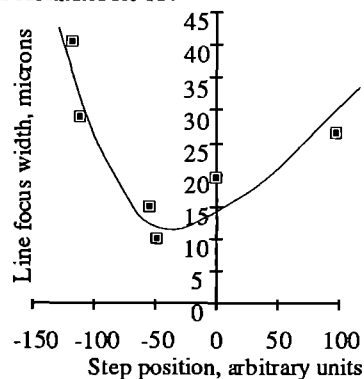


Figure 2. A graph of the width of the X-ray image of the line focus as a function of displacement of the target for Ta slabs irradiated by 60 ps pulses carrying 10 J .

Note that the minimum line focus width is around $12 \text{ \mu m}$ confirming our expectation that the scaling allows use of narrow line foci without significant lateral spreading of the energy flow. By contrast studies with 1.05 micron light at $3 \times 10^{13} \text{ Wcm}^{-2}$ in 0.6 ns pulses, driving the Neon-like Ge laser, have previously shown that lateral spreading limits the minimum effective line focus width to 50 micron .⁶

The resonance spectra at minimum line focus width and for a width of 60 microns are compared in figures 3a and 3b.

There is a clear difference between these spectra with an obvious increase in emission at shorter wavelengths for the narrow line focus compared to the wide line focus.

An identification of the spectrum was made^{7,8} by fitting a dispersion curve to the data using wavelength calibration lines from an Al target. This dispersion curve was used to make the

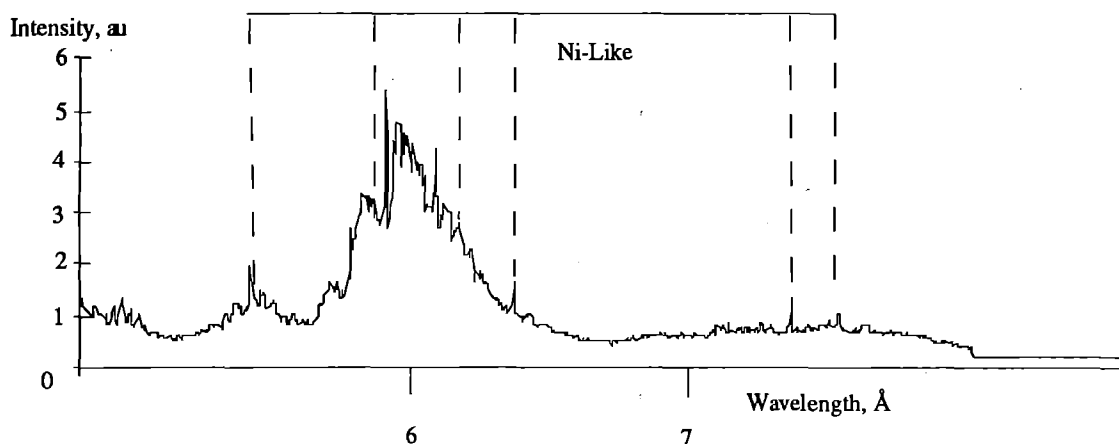


Figure 3a The resonance spectra recorded from a Ta slab target irradiated by a 60 ps pulse in a line focus with a 10 micron wide X-ray image. Intensity in this case was $3 \times 10^{15} \text{ Wcm}^{-2}$

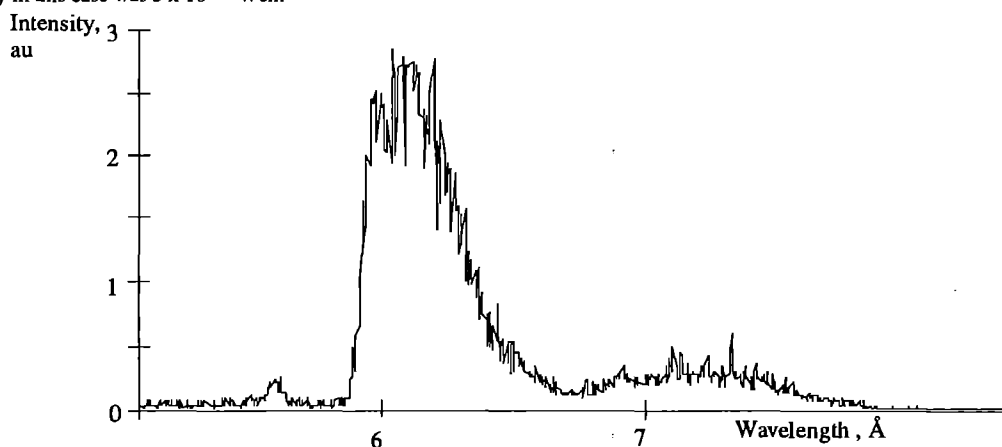


Figure 3.b The resonance spectra recorded from a Ta slab target irradiated by a 60 ps pulse in a line focus with a 60 micron wide X-ray image.

identification of Ni-like lines shown in Figures 3. The spectra obtained here were compared with spectra from a conventionally driven Ta laser³ (530 nm 500 ps drive at $2.5 \times 10^{14} \text{ Wcm}^{-2}$). It was noted that the narrow line focus result gave a very similar spectrum in the area of overlap of the data which is centred around the $3p^6 3d^{10} - 3p^5 (1/2) 3d^{10} 4s$ Ni-like line at 5.3 Å.

The intensity ratio of the Ni-like line ($3p^6 3d^{10} - [3p^6 3d^9 5/2 4p 3/2] J'=1$) at 7.3 Å and Co-like line ($3p^6 3d^9 - [3d^8 3/2, 5/2 (4) 4p 1/2] J'=7/2$) at 7.2 Å was measured for different line focus widths for tantalum slab targets irradiated by 60 ps pulses. In this way a relative measure of the ionisation and excitation of the plasma was obtained. The results of such a plot are shown as figure 4.

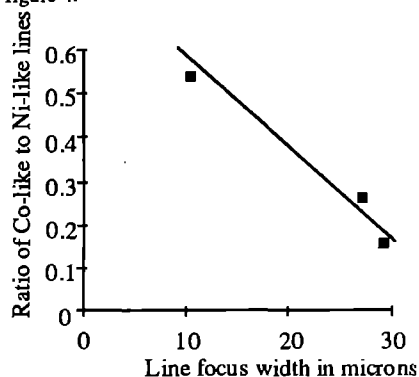


Figure 4. The ratio of Co-like resonance emission to Ni-like emission for Ta slabs irradiated by 60 ps pulses.

The monotonic change of the ratio shows the effectiveness of narrowing the line focus on increasing the excitation of the Ni-like ions.

CONCLUSION

The experiment demonstrates that a Ta plasma with ionization and excitation similar to that of an operational Ta laser is obtained with short pulse short wavelength drive in a narrow line focus as postulated in a scaling model. The results are an encouraging indicator of the possibility of future use of the Titania KrF pumped Raman laser system to drive the Ta laser close to saturation.

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OPTIMISATION OF DRIVE CONDITIONS FOR LOW Z NE-LIKE SYSTEMS

J. Warwick¹, C.L.S. Lewis¹, A. MacPhee¹, M. Holden², J. Krishnan², G. Tallents², P. Jaegle³, G. Jamelot³, A. Klisnick³, M. Nantel³, B. Rus³ and Ph. Zeitoun³

¹ Department of Pure and applied Physics, Queens University of Belfast, Belfast BT7 1NN, UK

² Department of Physics, University of Essex, Colchester CO4 3SQ, UK

³ Laboratoire de Spectroscopie Atomique et Ionique, Batiment 350, University de Paris-Sud, 91405 Orsay, France

INTRODUCTION

Low intensity prepulses preceding the main drive pulse have been used in the past to enhance the output and efficiency of low Z Ne-like x-ray lasers [1-5]. The most marked improvement occurs in the $J = 0-1$ lines; indeed in some cases lasing is not achieved in the absence of a prepulse. It is thought that they preform a cold plasma which is then heated by the main pulse, leading to an increase of the electron temperature and a relaxation of the density gradients which cause refraction of the x-rays out of the plasma. More efficient coupling of the main pulse into the plasma and a larger gain volume are also thought to occur.

However, as yet this effect has not been characterised in detail. We report the first parameter scan of prepulse fraction (prepulse / main pulse ratio), investigating the $J = 0-1$ lasing intensity in neon-like Zinc ($\lambda = 212\text{\AA}$), Copper ($\lambda = 221\text{\AA}$) and Nickel ($\lambda = 231\text{\AA}$).

EXPERIMENTAL DETAILS

The experiment was conducted in collaboration with the Laboratoire De Spectroscopie Atomique et Ionique using six beams of the LULI Nd:YAG laser at L'Ecole Polytechnique, Palaiseau, France. Fig. 1 shows the experimental setup.

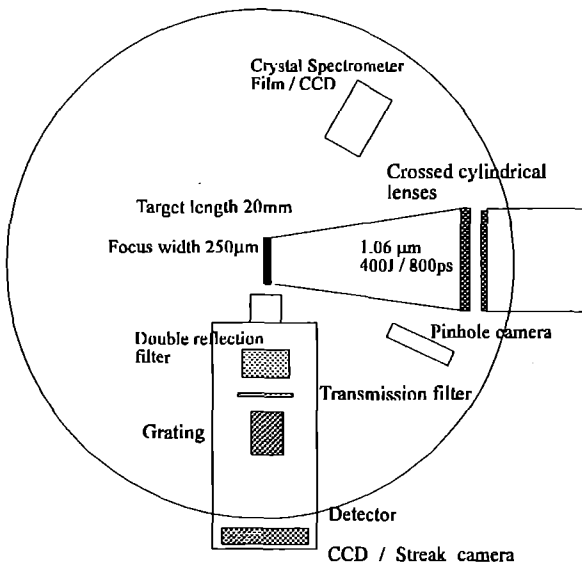


Fig. 1 Schematic diagram of experimental setup

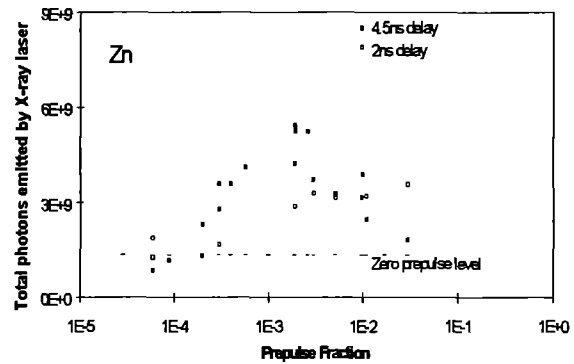
Pairs of crossed cylindrical optics were used to produce line foci $0.25\text{mm} \times 25\text{mm}$, at the fundamental, for an average intensity on target of $\sim 10^{13}\text{ W/cm}^2$. The pulse duration was measured to be $800 \pm 50\text{ps}$, with a total pulse energy of $400 \pm 50\text{J}$. Diagnostics included a flat-field spectrometer coupled either to a CCD camera or to a streak camera and a space resolving KeV crystal spectrometer. Prepulse fractions were induced at the 10^{-5} to 10^{-1} level using a polarisation sensitive optical beam splitter and delays of the main pulse relative to the prepulse of both 2ns and 4.5ns investigated. This apparatus was located after the first stage

amplifiers, prior to the splitting of the beam into separate beam lines, in order to induce an equal prepulse into all six pump beams. In order to eliminate any uncertainty in the prepulse fraction introduced by intensity dependent gain in the amplifier chain, the prepulse level on a single beam was monitored prior to entering the interactions chamber. The beam leakage through one of the final mirrors was split with a glass plate and taken to a pair of differentially filtered optical diodes, reverse biased and impedance matched to the inputs of a pair of 7GHz band-width digital oscilloscopes. The attenuation was adjusted to allow one to record the prepulse signal, whilst saturating on the main pulse, and the other to register the main pulse only. Cross calibration of the two channels was performed by firing laser shots, with the attenuation set to allow both channels to resolve the intensity of the prepulse. By inserting calibrated ND filters one channel was then attenuated sufficiently to observe the main pulse. Full experimental details are given in reference [6]

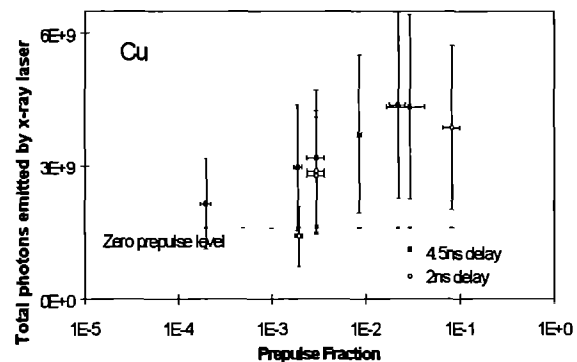
RESULTS

Time integrated data for Zn, Cu and Ni for both the 4.5 ns delay and the 2 ns delay are shown in Fig. 2.

(a)



(b)



(c)

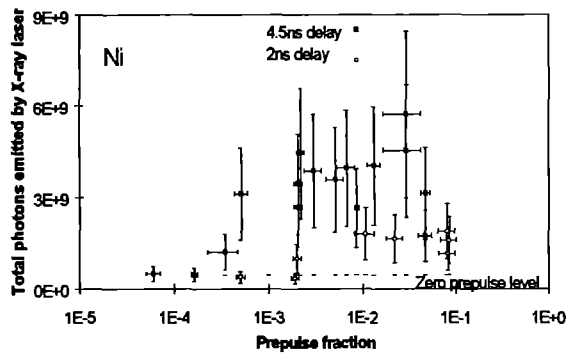


Fig.2 Time integrated emission as a function of prepulse fraction for (a) Zn, (b) Cu and (c) Ni. The error bars have been left off Fig. 2(a) for clarity but are comparable to those for Fig. 2(b) and (c)

The graphs for Copper and Nickel clearly show a trend of increasing output with prepulse fraction, up to the maximum prepulse investigated, 10^{-1} , for both 4.5ns and 2ns delays. The graph for Zinc is not so conclusive. While it appears to exhibit a peak at $\sim 2 \times 10^{-3}$ prepulse, there is a large scatter in the data due partly to shot to shot variations in the laser energy. This for example is evident for the Ni shots at prepulse fraction in the range around 0.05 in the 4.5ns delay case. It is possible that we were not working in a strong lasing regime, large variations in the output signal resulting from relatively small fluctuations of input intensity. It is our feeling that had a more extensive data set been built up at the higher prepulse fractions the trend would have been continuously increasing output above 10^{-3} . This is supported by the time resolved data, by the lack of peaks in both Cu and Ni for either 4.5ns or 2ns delay, and Zn itself for the 2ns delay.

For all three elements the 4.5ns delay is more efficient than the 2ns delay at producing lasing output. The $J = 2-1$ lines were barely distinguishable above noise and showed no significant response to prepulse fraction.

Fig. 3 shows the time resolved data for Zn. The signal has been spectrally and temporally binned to give a measure of the total lasing output. It shows a similar trend to that observed with the time integrated data; increasing output with increasing prepulse fraction and the 4.5ns delay being more effective than the 2ns delay.

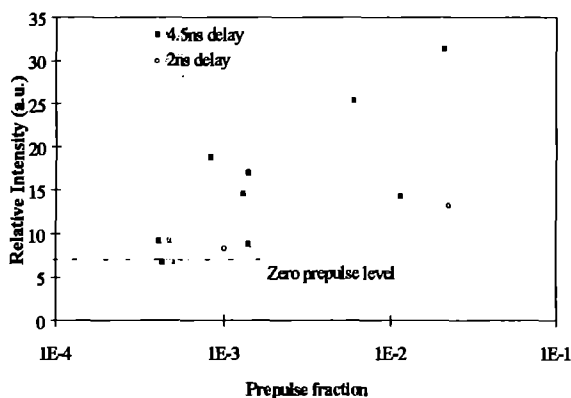


Fig. 3 The time resolved data for Zn.

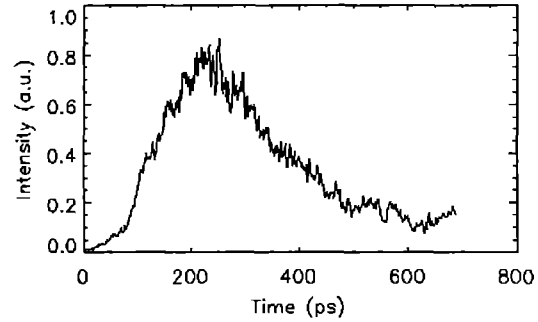


Fig. 4 Typical temporal history of Zn 212Å line.

The time integrated space-resolved KeV spectra of the 4.5ns delay Zinc data has been analysed [7] to infer the effect of the prepulses on the plasma conditions through spectroscopic line ratio measurements. It was observed that the plasma temperature and large scale homogeneity along the line focus were unchanged by varying the prepulse fraction from 10^{-3} to 10^{-2} . The lateral size of the Ne-like region of the plasma was seen to increase with the prepulse fraction, which is consistent with the general idea of the prepulse contributing to the formation of a larger volume gain zone with improved ASE propagation characteristics. The Ne-like ground state density in the plasma also increases with the prepulse level, creating more efficient pumping of the $J = 0-1$ upper level through direct collisional excitation from the Ne-like ground state. These effects help to explain the observed dependence of lasing output on prepulse fraction.

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SPACE-RESOLVED MEASUREMENT OF O VIII BALMER ALPHA FINE STRUCTURE IN EXPLODING FOIL PLASMAS

J Zhang¹, P A Norreys², G Cairns³, G J Tallents⁴, A. Demir⁴,
A Behjat⁴, M Holden⁴, M H Key^{1,2}, C L S Lewis³, C Smith¹

¹Department of Atomic and Laser Physics, University of Oxford, Oxford, OX1 3PU, UK

²Central Laser Facility, CLRC Rutherford Appleton Laboratory, Chilton, OX11 0QX, UK

³Department of Pure and Applied Physics, Queen's University, Belfast, BT7 1NN, UK

⁴Department of Physics, University of Essex, Colchester, CO4 3SQ, UK

INTRODUCTION

The strategic goal of the x-ray laser programme has been to realise saturated gain in the water window of the spectrum, preferably by using sub-kilojoule laser drivers that could be made available to the biological community on a facility basis. One way to achieve this goal is to provide short wavelength Ni-like x-ray lasers. Irradiating exploding foil targets with two pulses has two beneficial effects: the density gradients in the plasma are eliminated (reducing the refraction of the x-ray laser beam out of the high gain region); and to generate Ni-like ionisation at high densities, thereby increasing the gain coefficient [1,2].

We conducted an experiment on the VULCAN Nd-glass laser to investigate this scheme. A compact spectrometer consisting of a double cylindrical mirror coupled to a Hitachi flat field 2400 l/mm grating provided spatially resolved spectra at high spectral resolution ($\lambda/\Delta\lambda > 2000$). We report here on a space-resolved measurement of the O VIII Balmer alpha lines. The ratios of the intensities and line profiles of the Balmer alpha fine structure are of great interest both because of their value in diagnosing the plasma conditions and for ascertaining the plasma parameters suitable for recombination x-ray lasers [3-5].

EXPERIMENTAL SET-UP

Six opposing beams of the VULCAN laser operating at 0.53 μm were focused into a line on a 41.7 $\mu\text{g}/\text{cm}^2$ (60 nm thick) pure Yb foil with a 11.1 $\mu\text{g}/\text{cm}^2$ (100 nm thick) Formvar ($\text{C}_{11}\text{H}_{18}\text{O}_5$) backing. The width of the line focus was approximately 50 μm and the target length was about 6 mm. The line-focus length extended beyond the ends of the target, thus fully illuminating the target. A 150 ps FWHM (full width at half maximum) Gaussian pulse of 1×10^{13} W/cm² followed by a second Gaussian pulse of 20 ps duration and intensity 1×10^{14} W/cm² from the six opposing beams illuminated a common area from both sides of the target.

X-ray emission in the axial direction was measured by use of a flat-field grazing incidence XUV spectrometer with a 2400 line/mm aperiodically ruled grating [6]. It recorded the spectral range from 3.0 nm to 20.0 nm on Kodak 101-04 film. Spatial resolution in the spectrometer was provided by two cylindrical mirrors that compensated for the astigmatism in the focal plane [7], instead of toroidal mirrors that are usually used in astigmatic grating-incidence spectrometers. In the sagittal plane the first cylindrical mirror forms an image of the source on the focal plane of the spectrometer to provide one-dimensionally space-resolved spectra; in the meridional plane the source is focused through a 100 μm entrance slit to illuminate the grooved surface of the grating in order to obtain good spectral resolution and high throughput. The whole spectrometer has a spatial resolution better than 40 μm , a spectral resolution ($\lambda/\Delta\lambda$) higher than 2000 and a collecting solid angle larger than 2×10^{-4} sr.

RESULTS

The emission of the Balmer alpha spectral lines of hydrogen-like oxygen ions in the exploding foil plasma is shown in figure 1. The ordinate is the spatial expansion direction. The abscissa is wavelength. It is easily seen that the oxygen plasma, where the emission of the Balmer alpha lines come from, originates from the

back of the foil target and expands outwards. The Balmer alpha lines of the hydrogen-like ions consist of seven fine-structure components grouped to give the appearance of a doublet. The wavelength separation of the doublet was measured from figure 1 to be about 140 mÅ, which agrees approximately with the published data [4].

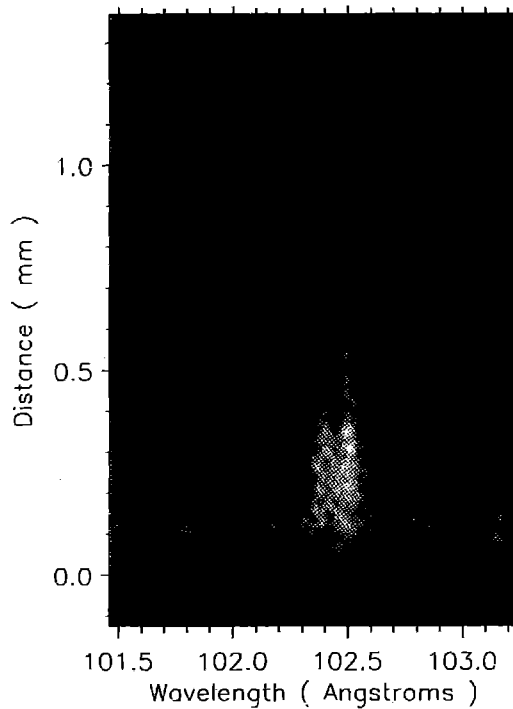


Figure 1. Space-resolved O VIII Balmer alpha line emission from an exploding foil plasma.

The Balmer alpha lines do not clearly split, though distinguishable, inside the exploding region (the continuum emission region 0.0 mm - 0.3 mm in Fig. 1), where the broadening effects of the spectral lines are so dominant that the separation between the doublet components become masked due to the high density and temperature in this region. The full width at half maximum (FWHM) of a wavelength component of the Balmer alpha doublet in this region was measured by deconvolution method to be about 120 mÅ. The fine structure splitting become progressively more pronounced with increasing distance corresponding to regions of decreasing density and temperature during the expanding stage. The line profiles shown in figure 2 for increasing distance from the target correspond to regions of decreasing density and temperature and consequently the various local broadening mechanisms, Stark and Doppler, decrease with respect to the fine-structure separation. The FWHM at a position of 0.5 mm from the target was measured to be 100 mÅ. For positions farther than 0.7 mm, the FWHMs remained approximately constant (80 mÅ).

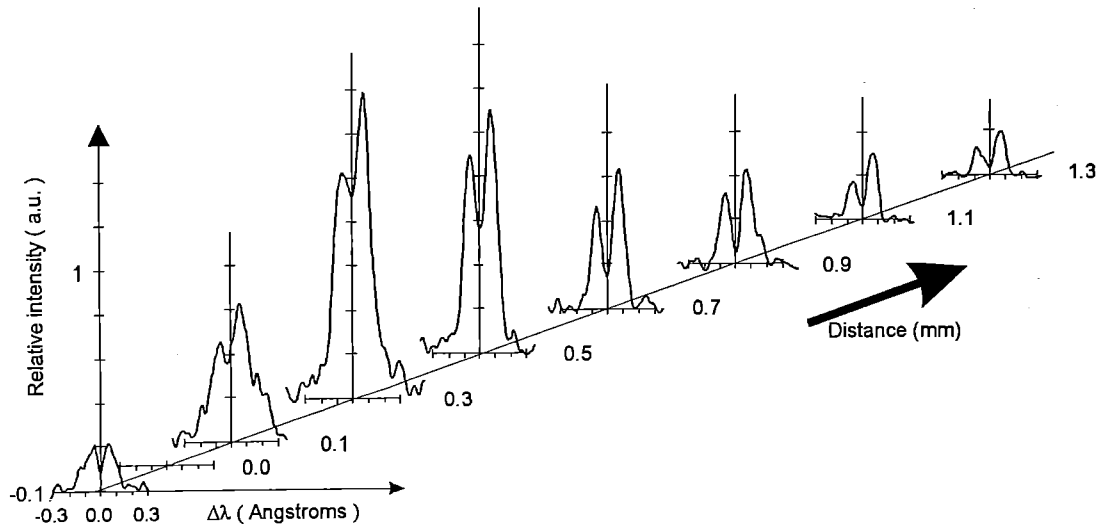


Figure 2. Profiles of the Balmer alpha line emission vs distance from the target in relative units of intensity.

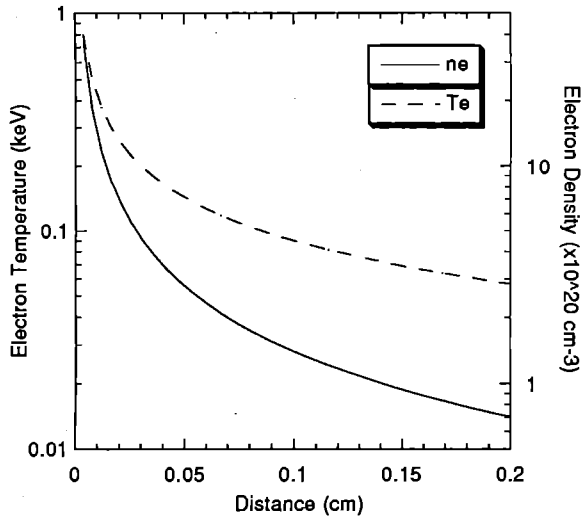


Figure 3. Theoretical prediction of the electron temperature and electron density vs distance from the target using an accurate simple model [8].

The intensity of the fine structure components of the Balmer alpha line depends on the population density of the $n = 3$ upper quantum sub-level associated with the transition and on the transition probability for the particular fine structure component [5]. Tallents investigated theoretically the dependence of the fine structure ratio on the electron density and on the ground-state density of the hydrogen-like ions. From the fine structure ratio of the Balmer alpha lines in figure 2, the electron densities at different positions of the expanding plasma can be deduced using Tallents's model. Inside the exploding region and part of the expanding region (< 0.9 mm), the ratio was measured to be about 1.3:1, which approximates the statistical value 1.24:1. This implies an electron density $> 1.3 \times 10^{20} \text{ cm}^{-3}$ for these regions [4], where the $n=3$ sublevel populations are proportional to their statistical weights. This estimate agrees approximately with a theoretical prediction using an accurate simple model assuming an isothermal, homogenous plasma expansion [8] (figure 3). At larger distances (> 0.9 mm) from the target, the fine structure ratio become somewhat a larger value in the range of 1.6-1.8, implying that a population inversion may have been formed due to the rapid recombination cascade into the $n = 3$ levels and a nearly empty

population of the ground state [9], which is a necessary condition for recombination lasers. With most recombination schemes, the peak gain of the Balmer alpha line occurs at densities $\text{Ne} \sim 10^{14} \text{ Z}^7 \text{ cm}^{-3}$ [9], which is about $2.0 \times 10^{20} \text{ cm}^{-3}$ for oxygen plasmas. This density corresponds to the expanding plasma regions of 0.7 - 0.8 mm, where the electron temperature is about 100 eV (figure 3), from the target. A detailed investigation is still in the process of analysis.

No gain was observed on the Ni-like Yb $J = 0-1$ transition at 50.23 Å during the experiment. Detailed modelling by MEDUSA for these target and irradiation conditions indicate that the electron temperature peaked at 800 eV at the end of the 20 psec heating pulse. However the flat density profile that is essential for this scheme was only established 15 psec later after the electron temperature had reduced to 600 eV, which is below that required for efficient collisional excitation. Thus demonstration of the feasibility of the double pulse technique must await the implementation of pulse compression techniques to increase the power to targets.

CONCLUSION

In conclusion, spatial transition of splitting of the Balmer alpha line emission has been observed from an exploding foil plasma. From the fine structure ratio of the Balmer alpha line, the electron density of plasmas with increasing distance from the target has been diagnosed. The deduced electron density agrees with the theoretical prediction using an accurate model. Evidence of population inversion has been observed at large distances from the target suggesting that Balmer alpha gain may occur in the plasma region of 0.7-0.8 mm from the target.

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A STUDY OF X-RAY LASER MEDIUM UNIFORMITY

A Behjat¹, G J Tallents¹, J Zhang², M H Key^{2,3}, P A Norreys³, C N Danson³, A Demir¹, L Dwivedi¹, M Holden¹, P B Holden⁴, M E Kurkcuoglu¹, C L S Lewis⁵, A G MacPhee⁵, D Neely³, G J Pert⁴, S A Ramsden⁴, S J Rose³, O Thomas³, F N Walsh³

¹ Department of Physics, University of Essex, UK.

² Department of Atomic and Laser Physics, University of Oxford, UK.

³ Central Laser Facility, Rutherford Appleton Laboratory, UK.

⁴ Department of Physics, University of York, UK.

⁵ Department of Pure and Applied Physics, Queen's University, UK.

INTRODUCTION

Evidence for recombination lasing at 18.2 nm on the CVI Balmer α -line was recently obtained⁽¹⁾ with 7 μ m diameter carbon fibre targets irradiated using the 2 ps, 20 TW, chirped pulse amplification (CPA) beam of the VULCAN laser facility. The laser line focus was created by the combination of off axis parabolic and spherical mirrors. Axial and transverse flat-field spectrometers and a footprint monitor were used to diagnose the plasma⁽²⁾. The uniformity of the x-ray laser medium is thought to be critical to the development of recombination x-ray laser⁽³⁾ and so a passive pin hole camera was used to monitor the uniformity and the length of the created plasma. In this report, we examine some of the issues relevant to uniformity in short pulse recombination x-ray laser experiments.

PARAMETER STUDY RESULTS

Assuming a flat topped laser beam profile, the intensity distribution varies approximately along the line focus as

$$I(x) = I(0) \left[1 - \left(\frac{2x}{L} \right)^2 \right]^{\frac{1}{2}} \quad (1)$$

where L is the length of the line focus and x is the distance from the centre of the line focus. The electron temperature along the line focus, $T_e(x)$, will thus be different along the plasma line, varying approximately as

$$T_e(x) = T_e(0) \left[\frac{I(x)}{I(0)} \right]^{\frac{2}{3}} \quad (2)$$

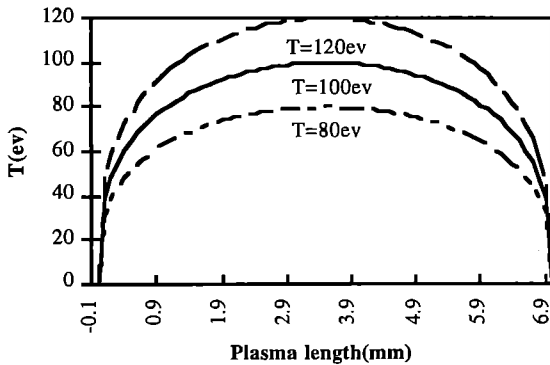


Figure 1 Variation of the electron temperature along the plasma length calculated using equation (1) assuming electron peak temperatures of 80, 100 or 120 eV.

where $I(0)$ is the laser beam intensity and $T_e(0)$ is the electron temperature at the centre of the line focus (Figure 1)⁽⁴⁾.

The RATION⁽⁵⁾ code was used to calculate the intensities of the sum of the CVI Lyman- α , CVI Lyman- β , CVI $1s^2$ - $1s2p$ and CV $1s^2$ - $1s3p$ spectral lines (I_{sum}) over a range of T_e up to 500 eV assuming an electron density of 10^{21} cm^{-3} and neglecting opacity effects. The transmission coefficient through the 0.5 μ m and 0.9 μ m thick aluminium filters (which were used in the pin hole camera) was taken into account at each wavelength using the SF code⁽⁶⁾. Resultant line intensities for the spectral lines were then summed and the variation of this total intensity (I_{sum}) with electron peak temperature plotted for each filter (Figure 2).

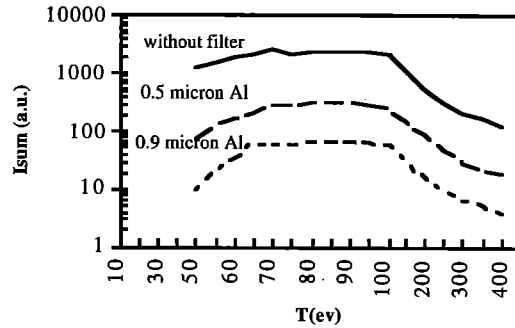


Figure 2 -A RATION code calculation of the intensities of the sum of the CVI Lyman- α , CVI Lyman- β , CV $1s^2$ - $1s2p$ and CV $1s^2$ - $1s3p$ spectral lines (I_{sum}) without and after filtering (0.5 and 0.9 micron Al filter) for an electron density of 10^{21} cm^{-3} .

The variation of the electron temperature along the plasma length was calculated using equation (2) for different peak electron temperatures (figure 1). These plots were used to deduce the total recorded x-ray emission along the plasma line for different temperatures (figure 3 and 4).

It can be seen that the spatially resolved x-ray emission recorded cannot be expected to be uniform. Increasing the laser irradiance and consequently the electron temperature produces apparent emission peaks at the irradiated fibre ends (figure 4). By increasing the filtering, the spatially resolved x-ray emission can be expected to become apparently more uniform (figure 3).

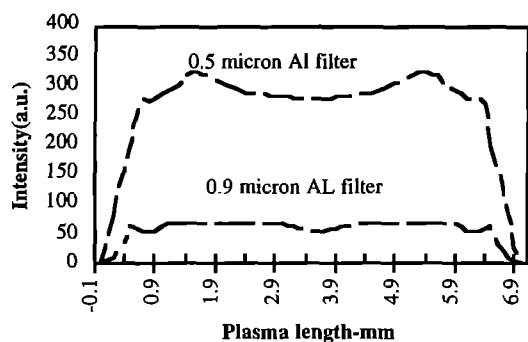


Figure 3 Calculated variation of recorded x-ray intensity along the plasma for an assumed peak plasma electron temperature of 100 eV.

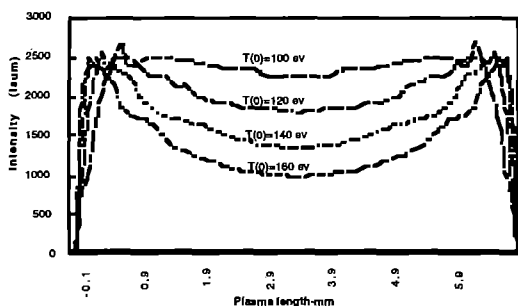


Figure 4 Calculated x-ray emission assuming different peak electron temperatures (0.9 micron Al filter).

An experimentally observed intensity variation along the plasma length is shown in figure 5. The emission peaks at the ends of the irradiated fibre in agreement with the modeling (figure 3 and 4).

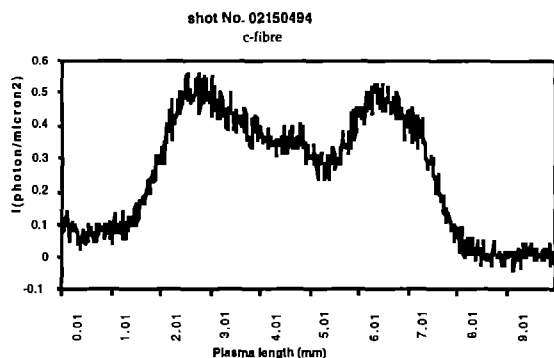


Figure 5 Typical densitometer trace of a pin hole camera image converted to x-ray intensity obtained from a 2 ps laser irradiated carbon fibre target with a 0.9 μm Al filter.

POLARISATION DEPENDENCE OF RESONANCE ABSORPTION

Resonance absorption varies along the line focus length if the incident beam polarisation is horizontal, but is constant for vertical polarisation (figure 6). With horizontal polarisation, the resonance absorption is at a minimum at the centre of the line focus where the irradiance is highest. Hence, the total resonance absorption integrated along the line focus is less for horizontal polarisation.

Experimental evidence for this was suggested by results reported by Holden et al.⁽⁷⁾ where ion probe signals for vertical polarisation indicated higher absorption (figure 3 of Holden et al. shows horizontal and vertical polarisation as the top and bottom traces respectively).

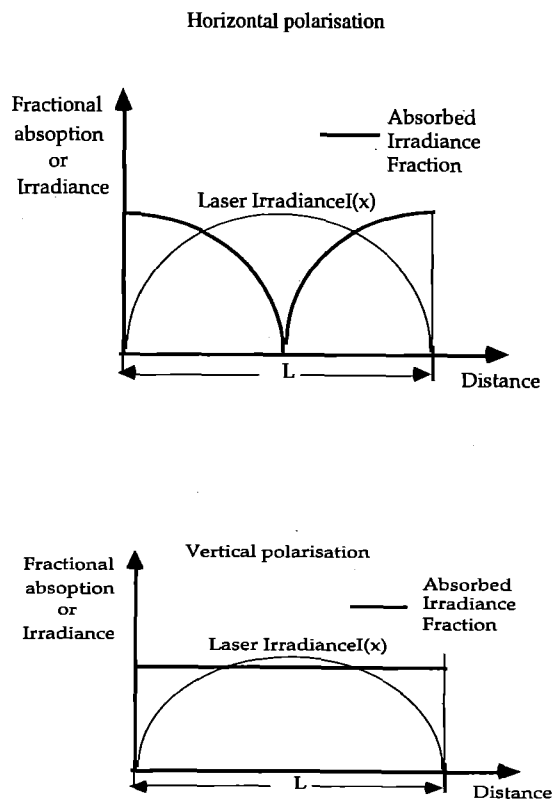


Figure 6 Schematic showing why resonance absorption is less for horizontal polarisation in a (horizontal) line focus. The fractional resonance absorption and target irradiance as a function of distance along the line focus for horizontally and vertically polarised laser light is shown.

CONCLUSION

A pinhole camera was used to study the uniformity of the line plasmas produced by 2ps, 10-30 Joules laser pulses and a model of the expected emission along the plasma length has been presented. It has been shown that better uniformity and more resonance absorption should be obtained with vertical polarisation.

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COMPARISON OF PLASMA EMISSION GENERATED BY NS AND PS PULSES

E E Fill¹, J Zhang², Y Li¹, D Schlögel¹, J Steingruber¹, H Arslan³, A Demir⁴, M Holden⁴,
M H Key^{2,5}, C L S Lewis⁶, A G MacPhee⁶, P A Norreys⁵, G J Tallents⁴, F Walsh⁵ and P Zeitoun⁴

¹ Max-Planck-Institut für Quantenoptik, D-85740 Garching, Germany

² Department of Atomic and Laser Physics, University of Oxford, Oxford, OX1 3PU, UK

³ Department of Physics, Zonguldak Karaelmas University, 67100 Zonguldak, Turkey

⁴ Department of Physics, University of Essex, Colchester, CO4 3SQ, UK

⁵ Rutherford Appleton Laboratory, Chilton DIDCOT Oxon, OX1 0QX, UK

⁶ Department of Pure and Applied Physics, Queen's University, Belfast, BT7 1NN, UK

INTRODUCTION

There is growing interest in the interaction of ps or fs pulses with matter, owing to the advent of short-pulse high-power lasers based on chirped-pulse amplification. In particular, more and more work is devoted to generating laser-produced plasmas with such pulses, in view of variety of applications such as laser fusion, short pulse x-ray emission and x-ray lasers [1-3].

While the generation of x-rays by ns laser plasmas is well investigated, and a considerable amount of work has also been expended on investigating x-ray emission from ps laser plasmas, only a few studies are devoted to comparing x-ray spectra generated by different pulse durations.

Short (ps or fs) laser pulses can heat targets significantly before any expansion occurs, generating plasmas at densities close to solid density, in contrast to the interaction of ns long pulse with targets where the main heating occurs at densities close to the critical density. Consequently, the densities of ps laser plasmas can be orders of magnitude higher than those of ns laser plasmas. The enormous difference of the densities and the different plasma processes will have dominant effects on the atomic energy structures and therefore spectra of the plasma emission. Comparison of plasma emission generated by ns and ps laser pulses would provide better understanding of different plasma physics in the ns laser plasmas and ps laser plasmas.

In this paper we present a comparison of plasma emission in the region $60 \text{ \AA} < \lambda < 220 \text{ \AA}$ generated by 1 ns and 2 ps laser pulses, with similar pulse energies. Thus, the intensities applied to the target differed by a factor of up to 500. The investigations were made with moderately high intensities up to $1 \times 10^{13} \text{ W/cm}^2$ for 1 ns and $5 \times 10^{15} \text{ W/cm}^2$ for 2 ps pulses.

EXPERIMENT

The experiment was conducted with the VULCAN high-power Nd-glass laser. The VULCAN laser can be operated either in the normal mode, in which mode-locked pulses of 1 ns duration are amplified or in the chirped pulse amplification (CPA) mode [4] in which ps pulses from a separate oscillator are stretched and recompressed after amplification. In both cases the precursor intensity is $< 10^{-6}$ of the intensity of the main pulse.

The pulses, with a maximum energy of 45 J, were focused by means of an off-axis parabola in conjunction with an off-axis spherical mirror to a line 6 - 7 mm long and 20 μm wide. Thus, for a 1 ns and a 2 ps pulse, after a 50% reflection loss by the compression gratings and focusing system, intensities up to 1×10^{13} and $5 \times 10^{15} \text{ W/cm}^2$, respectively, were reached in the focal region. Fibre targets with a diameter of 7 μm were irradiated, thus, only about one third of the laser energy reached the target. The fibres were supported on one end and positioned with better than $\pm 2 \text{ \mu m}$ spatial accuracy and $\pm 1 \text{ mrad}$ angular accuracy by using a modified split-field microscope system [2]. The targets were fabricated by coating Sigradur fibres [5], 7 μm in diameter, with a thickness $> 300 \text{ \AA}$ NaF, Al, Cu or Fe layer. It turned out that the deposition process frequently resulted in bending of the fibre targets. Only fibres with a deviation from straightness of less than 20 μm were used in the experiment.

To record the soft x-ray spectra, a flat-field spectrometer [2] coupled to a x-ray sensitive fluorescent-screen CCD camera was installed axially to the fibre target. For a number of shots Shanghai x-

ray film (SIOFM-5FW) or Kodak 101-05 plates were used as the detector, in which case the evaluation of the spectra was carried out by means of a digital densitometer. The densitometer yielded time-integrated spectra with a spectral resolution of about 0.2 $\text{\AA}$ and with a spatial resolution better than 40 μm . The data were Abel-inverted along the spatial co-ordinate to take the cylindrical symmetry of the experiment into account.

RESULTS

In figs. 1 - 3 spectra of NaF, Al and Cu plasmas generated with 1 ns and 2 ps pulses are shown. The pulse energies and the lengths of fibre are indicated on the spectra. These spectra are integrated over a spatial region with $r > 50 \text{ \mu m}$ (here r represents the radial distance from the fibre surface).

It is quite conspicuous that in all cases the ps spectra show a significantly higher ratio of continuum to line intensity. This can be attributed to a number of reasons related to opacity, line broadening effects etc.

The spectra allow comparison of the ionisation stages generated with the different pulse durations. In NaF the ns and ps spectra show that essentially the same ionisation stages are reached. Li-like Na and Li-like F lines predominate. H-like and He-like F can also be seen, while He-like and H-like Na lines are absent.

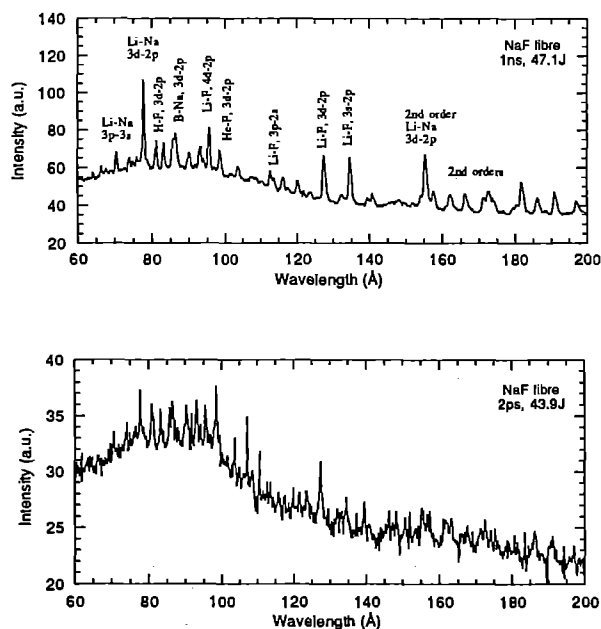


Fig. 1. Axial spectra of 7 μm diameter carbon fibres coated with 3000 $\text{\AA}$ of NaF. a) 1 ns duration. b) 2 ps duration.

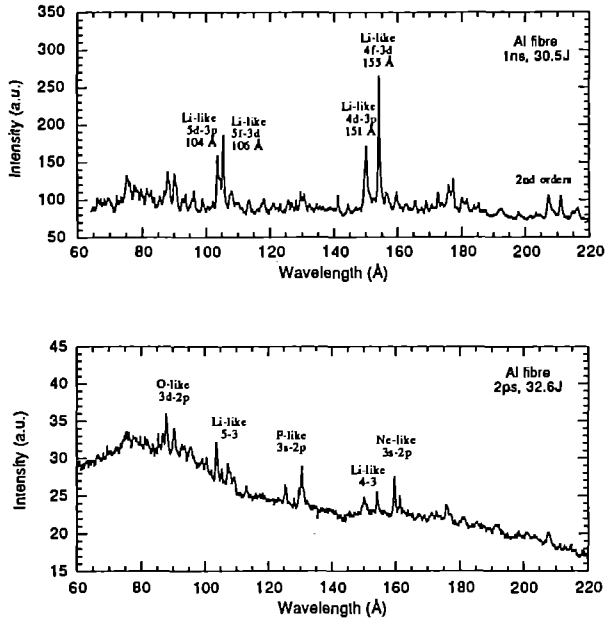


Fig. 2. Axial spectra of 7 μm diameter carbon fibres coated with 3000 Å of Al. a) 1 ns duration. b) 2 ps duration.

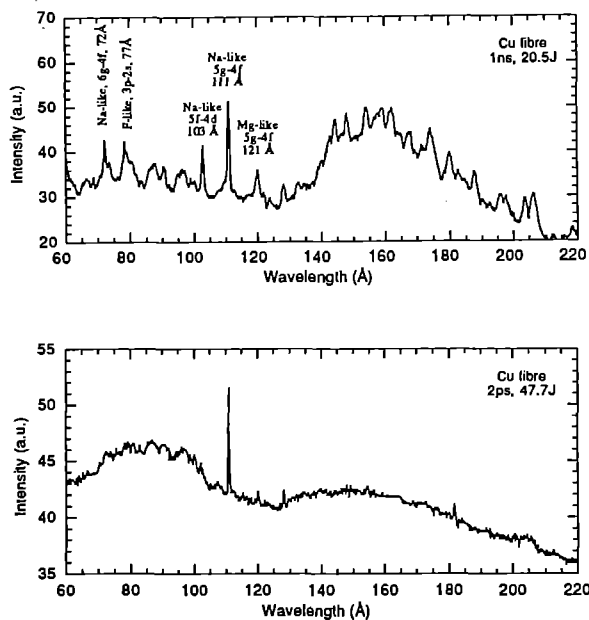


Fig. 3. Axial spectra of 7 μm diameter carbon fibres coated with 3000 Å of Cu. a) 1 ns duration. b) 2 ps duration.

For aluminium the situation is quite different: While the spectra with ns pulses show a predominance of Li-like recombination lines (in particular the gain lines 3d-4f and 3d-5f at 154 and 106 Å) the spectra with 2 ps pulse duration are dominated by lines emitted from relatively low ionisation stages, with Ne-like, F-like and O-like lines being the most prominent ones. The Li-like 3d-4f line appears only weakly in these spectra. The reason for this observation is not clear, since simulations would predict the ionisation of the aluminium to be similar for ns and ps pulses.

For the Cu and Fe (not shown here) spectra the degree of ionisation is again similar for ns and ps pulses. For both pulse durations Na-like recombination lines show the strongest emission. Spectra taken with a KAP crystal spectrograph observing transversally to the fibre axial direction revealed the presence of Ne-like and F-like and O-like resonance lines in both materials.

Special attention was paid to the Na-like recombination laser lines 5g-4f, 5f-4d, 6g-4f and 6f-4d. It was found that all four lines were strongly present in the ns spectra. In the spectra with 2 ps pulses only the 5g-4f line emission was seen, but with a high intensity. This line is by far the strongest one in the spectrum if the spectrum is integrated over the spatial region with $r > 50 \mu\text{m}$, thus suppressing the emission from the part close to the target surface. Figure 3b shows the corresponding spectra for Cu. The high intensity of 5g-4f line and the absence of the 5f-4d, 6g-4f and 6f-4d lines give a strong indication of gain on that line. A gain measurement will be presented in a following paper.

SUMMARY AND CONCLUSIONS

We have compared the axial soft x-ray emission from fibre targets of two low-Z materials (NaF and Al) and of two medium-Z materials (Fe and Cu) generated with 1 ns and 2 ps pulses with similar pulse energies. For the two pulse durations the pulse energy on target was kept constant, resulting in a factor up to 500 higher intensity for the ps pulse irradiation. For NaF, Fe and Cu, in spite of the large difference in intensity, the degree of ionisation for both durations were found to be rather similar and in accord with computer simulations. For aluminium the experimental spectra with ps pulse indicate a plasma of much lower ionisation stage than with ns pulses. Simulations predict however the ionisation stages to be rather similar in the two cases.

For all targets the ratio of continuum to line emission was considerably higher for the short pulse irradiation, a fact explained by the higher density from which the x-rays are emitted in the ps plasmas. For iron and copper strong emission on a recombination gain line, the 5g-4f line of the Na-like ion, was observed for ns and ps pulse irradiation. For ps pulses this line dominated the spectra if the spatially integrated emission over the region with $r > 50 \mu\text{m}$ from the target surface was considered.

The spatial extent of the emission was quite different for the two pulse durations. For ns pulses the time-integrated spatially resolved spectra show a typical fall-off of the line emission within a region of 300 μm , whereas for the ps pulses the emission terminates much closer to the target, typically around 100 μm from the target surface. This observation is explained by the simulations to be due to the sharp density gradients and the rapid decay of the density perpendicular to the target surface for ps pulses.

The anomalously strong emission on the 5g-4f line in Na-like Cu and Fe ions suggests that there is gain on that recombination line. This is accord with the simulation results, which predicts a gain coefficient of about 10 cm^{-1} under the conditions of the experiment.

ACKNOWLEDGEMENT

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EVIDENCE OF HIGH GAIN IN A SODIUM-LIKE COPPER X-RAY LASER AT 111 Å

J Zhang¹, E E Fill², Y Li², D Schlögel², J Steingruber², C Danson³,
A Demir⁴, M Holden⁴, M H Key^{1,3}, C L S Lewis⁵,
A G MacPhee⁵, P A Norreys³, G J Tallents⁴, F Walsh³ and P Zeitoun⁴

¹ Department of Atomic and Laser Physics, University of Oxford, Oxford, OX1 3PU, UK

² Max-Planck-Institute für Quantenoptik, D-85740 Garching, Germany

³ Rutherford Appleton Laboratory, Chilton DIDCOT Oxon, OX1 0QX, UK

⁴ Department of Physics, University of Essex, Colchester, CO4 3SQ, UK

⁵ Department of Pure and Applied Physics, Queen's University, Belfast, BT7 1NN, UK

INTRODUCTION

One of the primary objectives in x-ray laser research is biological applications in the so-called water window around 22-45 Å [1]. X-ray laser microscope could provide higher resolution than optical instruments and avoid much of the often destructive specimen preparation needed for an electron microscope. X-ray laser holograms of biological structure in living cells would be very helpful for investigating their functions.

The recombination scheme operating with adiabatic cooling is in principle more favourable than the collisional pumped scheme in extending x-ray lasers to shorter wavelength range because the recombination scheme requires a much lower driver energy. Systematic studies suggest that interaction of fibre targets and picosecond (ps) laser pulses produces a heating close to solid density, thus enhancing the adiabatic cooling. The rapid cooling in turn leads to a high gain in recombination lasers [2,3]. There is reason therefore to believe that it is possible to reduce the driver energy for x-ray lasers considerably by using ps high brightness laser pulses. Towards this goal, a series of experiments was conducted at RAL. An exceptionally high gain was recently demonstrated on the hydrogen-like carbon 3-2 transition using 20 J, 2 ps pulses focused on 7 µm diameter carbon fibres [4].

Lithium-like and sodium-like ions are analogous to the hydrogen-like ions, having one electron in the outer atomic shell but one or two inner atomic shells. For lasing transitions, lithium-like and sodium-like ions have somewhat higher quantum efficiency. Amplification has been observed for many transitions both in lithium- and sodium-like ions using nanosecond (ns) driver pulses [5,6].

With a view to a possible extension of high gain recombination x-ray lasers to shorter wavelengths, sodium-like copper was chosen for the present investigation. We report here experimental results of an evidence of high gain in a sodium-like copper recombination x-ray laser driven by 20 J, 2 ps pulses from the chirped pulse amplification (CPA) beam on the VULCAN laser facility.

EXPERIMENT

The set-up is similar to that used in the C VI x-ray laser experiment [4]. The 2 ps CPA beam was reflected by an f/4 off-axis parabolic mirror to a spot focus which was imaged by an f/4 off-axis spherical mirror to produce a line focus 7 mm long and 20 µm wide. The spatially averaged incident irradiance in the line focus was $(5 \pm 1) \times 10^{15}$ W/cm² for the data discussed here.

The targets used in the experiment were copper coated (thickness typically > 3000 Å) carbon fibres of 1 cm length and 7 µm diameter supported at one end. They were positioned better than ± 2 µm spatial accuracy and ± 1 mrad angular accuracy using a split-field microscope system. The free end of the target was placed well within the line focus to avoid creating a cold output end in the plasma, and the irradiated length was varied by moving

the line focus axially along the fibre which was always at the same location.

Two soft x-ray spectrometers viewed the line plasma in the axial direction from both sides as the primary diagnostics. One of the spectrometers was a flat-field grazing incidence soft x-ray spectrometer with a 1200 line/mm aperiodically ruled grating. The detector was an x-ray sensitive phosphor screen coupled to a charged-coupled device (CCD). The other spectrometer was a 5000 line/mm transmission grating. The length and the spatial homogeneity of the line focus were monitored by means of an x-ray pinhole camera, which viewed the line focus at 45 degree from above. A KAP crystal spectrometer was used at 45 degree to the fibre axis to measure the ionisation balance of the plasma.

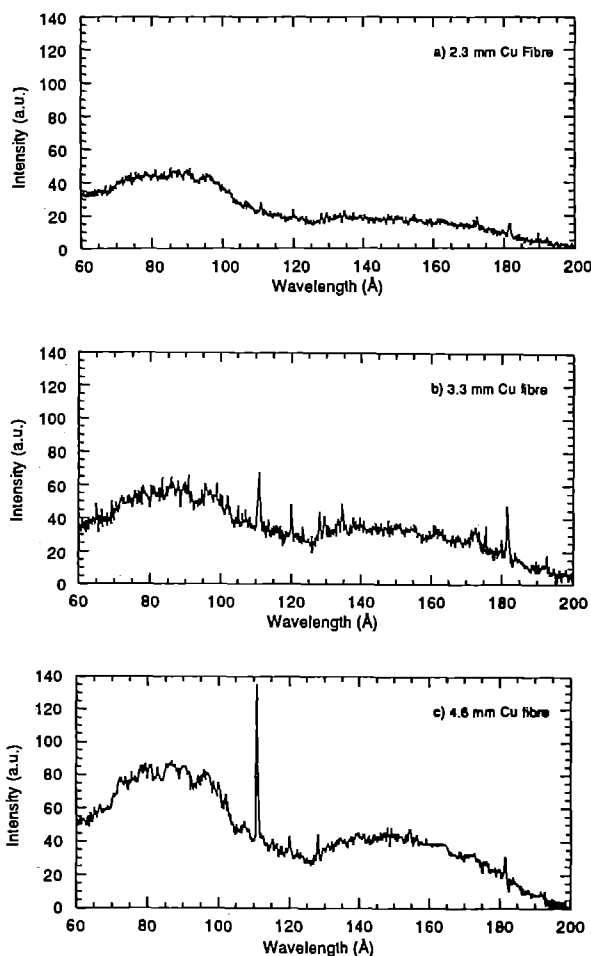


Fig. 1. On-axis spectra of the copper fibre target with 2.3, 3.3, 4.6 mm lengths irradiated by 20 J, 2 ps laser pulses.

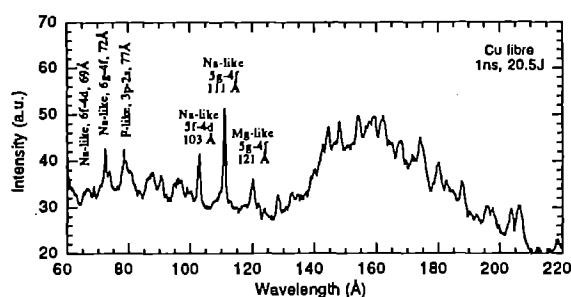


Fig. 2. On-axis spectra of the copper fibre target irradiated by 20 J, 1 ns laser pulses.

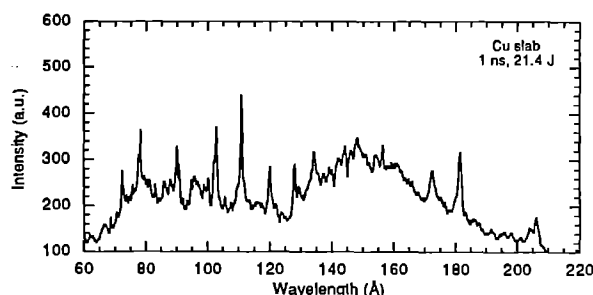


Fig. 3. On-axis spectra of the copper slab target irradiated by 21 J, 1 ns laser pulses.

RESULTS

A series of spectra from the on-axis flat-field spectrometer are shown in figure 1. The spectra correspond to three irradiated lengths of copper fibres: a) 2.3 mm, b) 3.3 mm, c) 4.6 mm respectively. In these spectra, only the strong 5g-4f line emission was observed. This is contrast to steady state ablation from either fibre or slab targets with long pulse (nsec) driver lasers where all of the four possible gain lines (5g-4f, 5f-4d, 6g-4f and 6f-4d) appear (figures 2 & 3). Similar features were observed in the spectra of iron fibre targets irradiated in the experiment. The irradiated intensity on fibres for the data in figure 1 was around 5×10^{15} W/cm², at which there were mainly neon-like resonance lines and only comparatively weak fluorine-like and oxygen-like lines in the crystal spectra. This plasma ionisation is near optimum for a sodium-like copper recombination laser according to the simulation result [7].

There is strong continuum emission background in the 2 ps spectra, compared with the ns spectra (figures 2 & 3). This maybe attributed to the ps pulse produced higher density at which the maximum electron temperature is reached. The higher density leads to a larger effects of opacity, line broadening etc. To suppress the strong continuum emission from the part close to the target surface, spectra in figure 1 were integrated over the spatial region in the radial direction with $r > 50$ μ m from the target surface.

The intensity of an individual line was obtained by subtracting the background continuum level and integrating spectrally and spatially over the spatial region with $r > 50$ μ m in the radial direction.

At very short length, the 5g-4f line transition at 111 Å is very weak compared with the continuum background emission. The intensity of this line emission increased with the plasma length non-linearly. This line emission became dominant in the spectra

for longer targets, giving a strong indication of stimulated emission on the 5g-4f transition. The results of these measurements were plotted in figure 4. The intensities of different shots on different detectors were cross-calibrated by the continuum emission, which increased linearly with the plasma length. The gain coefficient was estimated by least squares fitting of the Linford formula to the data giving 8.8 ± 1.4 cm⁻¹.

The fibre targets were fabricated by coating 7 μ m diameter carbon fibres with a layer (thickness > 300 nm) of copper. It turned out that the coating process frequently resulted in bending of the fibre targets. As a result, the longest straight length of the coated fibre targets was limited to 5 mm by the coating process. It has however been found out after the experiment that longer straight copper coated fibres could be manufactured by sputtering cold copper ions into the surface layer of carbon fibres.

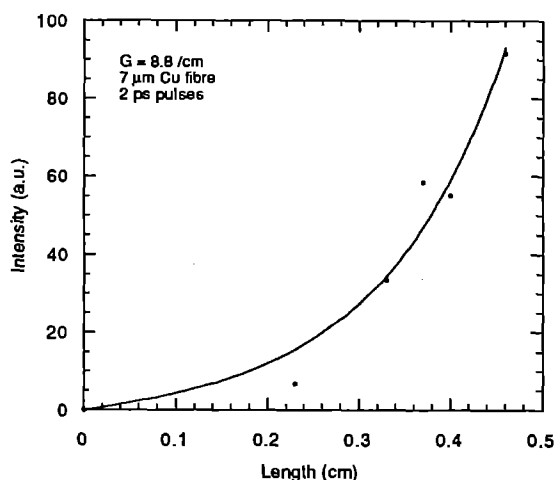


Fig. 4. The fitted gain to the data from figure 1 for 5g-4f transition in sodium-like copper ions using Linford formula.

CONCLUSION

In conclusion, we have measured high gain for the 5g-4f transition at 111 Å in sodium-like ions driven by 20 J, 2 ps laser pulses. It is anticipated that a better performance will be achieved for the sodium-like copper recombination x-ray laser by optimising the conditions and using longer straight fibres in future experiment.

ACKNOWLEDGEMENT

The skilful work of the target manufacturing group at the Rutherford Laboratory is gratefully acknowledged. we thank the laser staff for operating the laser under the different conditions. This work was performed under the European Large Facilities programme of Rutherford Laboratory.

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ENHANCED EFFICIENCY AND MONOCHROMATICITY OF THE J=0-1 196Å LASING LINE IN NE-LIKE GERMANIUM USING TWO 100PS PULSES.

A.G. MacPhee¹, A. Bhejat², M.J. Lamb¹, C.L.S. Lewis¹, D. Neely³, G.J. Tallents², P.J. Warwick¹, E. Wolfraam³

¹ Department of Pure and Applied Physics, The Queens University of Belfast, BT7 1NN, UK

² Department of Physics, University of Essex, Colchester, England, CO4 3SQ,

³ The Rutherford Appleton Laboratory, CLRC, Chilton, Didcot OXON, OX11 0QX, UK

INTRODUCTION

Continuing with the X-ray laser consortiums efforts to improve the characteristics of X-ray lasers in general, the latest in a series of experiments employing multiple pulse irradiance of slab targets for collisional excitation schemes has demonstrated further significant enhancements in output parameters. A pair of 100ps, 1.05µm pulses separated by 400ps were incident on two opposing 14mm germanium slabs, with a line focus width of 160µm. The intensity on target was $2.7 \times 10^{13} \text{ Wcm}^{-2}$. The contrast in the total energy content between the J=0-1 196Å and the J=2-1 236Å lines was measured at 2×10^2 (peak ratio $\sim 10^3$). The output from both ends of the target were spectrally resolved using flat field grazing incidence spectrometers, one of which employed a grazing incidence cylindrical focusing optic for imaging the exit pupil of the XRL in the horizontal plane, whilst the other operated in the conventional angular resolving mode (Figure 1).

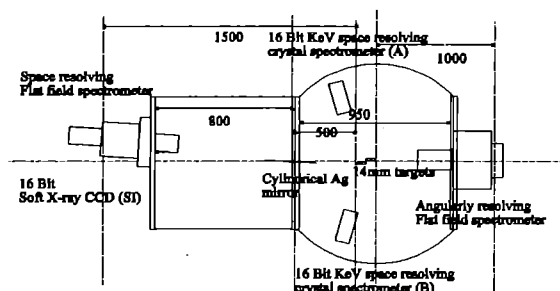


Figure 1 Experimental geometry

The data from the space-resolving spectrometer was collected using a 16 bit CCD system¹, the angularly resolved data was recorded on Ilford Q-plates. The target irradiance uniformity was monitored with time integrated, space resolving Bragg Crystal spectrometers incorporating 16 bit CCD detectors².

The grazing incidence cylindrical mirror collected all rays in the horizontal plane, parallel to the rulings on the flat field grating. From previous footprint measurements³, the vertical X-ray laser beam divergence is known to be similar to that in the horizontal plane. Hence, from the horizontal divergence, measured using the time-integrated angularly resolved output from the north spectrometer, an estimate was made of the vertical divergence (1:1). The fraction of the beam collected in the vertical plane by the grating in the imaging spectrometer was therefore established. From the absolute sensitivity of the soft X-ray CCD system (0.03 counts $\text{hv}^{-1} \mu\text{m}^{-2}$), and estimates of the reflectivity of the grating and silver coated mirror (5% and 80% respectively) and the transmission of the 1.6µm Al filter (2.8%), the average signal power on the 196Å line was measured to be 57kW, based on an estimated pulse width of 100ps. A spectrum from the space resolving XUV spectrometer is shown in Figure 2, where

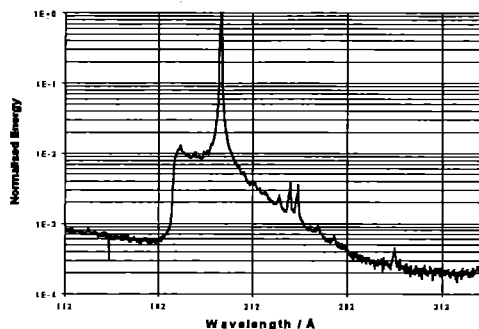


Figure 2 XUV spectrum from double 14mm target irradiated at $2 \times 10^{13} \text{ Wcm}^{-2}$, with $2 \times 100\text{ps}$ 1:1 contrast ratio pulses.

energy has been integrated in space across the detector plane. The beam flux was 148 mJsr^{-1} , with a total integrated signal of 5.7mJ. For the J=2-1, 232Å and 236Å lines, the integrated signals were each $\sim 28\text{nJ}$. By comparison, the maximum measured value for the average power on the brightest J=2-1 236Å line from a 3cm target irradiated at $2.5 \times 10^{13} \text{ Wcm}^{-2}$ with a 1ns pulse was 14kW^4 , with an associated beam flux of 74 mJsr^{-1} . For the 196Å line from recent asymmetric pre-pulse work⁵, the equivalent power was 28kW and the beam flux 8 mJ sr^{-1} .

Along with the improvement in the monochromaticity of the shorter wavelength emission in this mode, the enhanced integrated energy, higher peak power and shorter pulse duration make for a substantial increase in the usability of this beam for holographic and probing experiments. From recent work by Da Silva *et al*⁶, there is scope still for further enhancement using a travelling pump along the target axis. For a 100ps gain width we are limited to target lengths of $< 3\text{cm}$, whereas the modelling and experimental results in general suggest the gain width to be significantly shorter than this.

One explanation for this enhancement may be the effect of the movement of the plasma after the initial pulse, to a region of higher average intensity and narrower line focus during the second pulse. The F/2.5 lens/off-axis spherical mirrors systems used at RAL to generate the line foci can produce focal widths $\sim 25\mu\text{m}$. During this experiment, the focus was positioned $\sim 250\mu\text{m}$ in front of the target surface. From a Pollux simulation⁷, the critical surface of the plasma would be positioned $10\mu\text{m}$ in front of the original solid, with a ~ 0.1 critical density at the beam waists. This will have introduced a modulation in the vertical profile of the second drive pulse, resulting in possible gain guiding, although further ray tracing will be required to assess this.

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MULTIPLE PULSE EFFECTS IN NE-LIKE YTTRIUM.

D. Neely¹, G. Cairns², S. Healey³, M. H. Key^{2,5}, J. Krishnan⁴, M. J. Lamb², C. L. S. Lewis², A. G. MacPhee², P. N. Norreys¹, R. Parker¹, G. J. Pert³, J. Plowes³, S. A. Ramsden⁵ and G. J. Tallents⁴

- (1) Rutherford Appleton Laboratory, Chilton, Didcot, Oxon, OX11 0QX
- (2) Department of Pure and Applied Physics, Queens University of Belfast, Belfast, BT7 1NN
- (3) Department of Computational Physics, University of York, York, YO1 5DD
- (4) Department of Physics, University of Essex, Colchester, CO4 3SQ
- (5) Clarendon Laboratory, Oxford University, Oxford, OX1 3PU

INTRODUCTION

Soft X-ray collisionally excited lasing action within long pulse (≈ 1 ns) laser driven plasmas have been extensively studied in many laboratories world-wide. A significant restriction on readily achieving saturation is due to the limited photon propagation length within the active media caused by the refractive electron gradients present. Recent experimental advances in obtaining higher efficiency and shorter wavelength soft X-ray lasing action have concentrated upon compensating for¹, or reducing² the transverse electron density gradients present across the gain region of an ablated plasma.

Multiple pulse drive can be used to reduce the gradients present across the gain region of a plasma and give an additional degree of freedom in the design of soft X-ray lasers. Experimental developments³ have concentrated on optimising plasma and target conditions to minimise transverse beam refraction and maximise gain. In this report an additional effect of lateral density gradient modification is considered.

BACKGROUND

Multiple pulse irradiation of a target has significant advantages over a single pulse drive case. At intensities typically used to drive collisionally excited lasers of $10^{12} \leftrightarrow 10^{15}$ Wcm⁻² Inverse Bremsstrahlung is the dominant energy absorption mechanism within the plasma. The amount of laser energy absorbed is strongly dependent on the density gradient⁴ across the interaction region. More efficient coupling is achieved by having a lower gradient and a larger interaction zone. Such conditions can be efficiently generated by using a low fraction of the main laser energy to pre-form a plasma. Since the pre-formed plasma is initially cooler, the amount of energy lost through thermal emission during the time required for it to hydrodynamically expand to the required density will also be considerably reduced. When the second or later pulses come along, they can then ionise the plasma to the required state over a larger plasma volume, with more laser energy being absorbed and lower refractive gradients present. There is a trade off, since, if the plasma volume that energy deposition occurs over is increased by more than the extra laser energy absorption, the plasma temperature will not be as high as previously and this can reduce the gain. This is to some extent compensated for (in the observed output beam) by the reduced density gradients, which increases the volume within the plasma that can usefully contribute to the output.

EXPERIMENT

Yttrium stripe targets were irradiated using six 108 mm diameter 1.05 μ m wavelength beams in the Target Area East off-axis line focus chamber as depicted in Figure 1. The standard laser drive pulse used throughout the experiment was a 650 ps FWHM flat topped Gaussian shaped pulse. Both single (up to 9 mm) and double⁵ (up to 9 + 9 mm) targets were used to study the length scaling⁶ of the 15.5 nm lasing transition. The targets consisted of thermally evaporated 100 ± 10 μ m wide, 1 ± 0.2 μ m thick Yttrium, deposited onto a 0.1 μ m plastic overcoated 1.5 mm thick glass substrate. The double target's surface planes were set parallel to each other (± 1 mrad), with the stripes in the same horizontal plane (± 10 μ m), at a fixed separation along the plasma length of 0.58 ± 0.05 mm in an alignment jig. The principle diagnostics of target drive uniformity were space resolving crystal spectrometers which observed the Ne and F-like resonance line ratio⁷ and a crossed slit camera. A 1200 lines/mm flat-field grating spectrometer which used Kodak 104-02 film as the recording medium was used to examine the intensity and

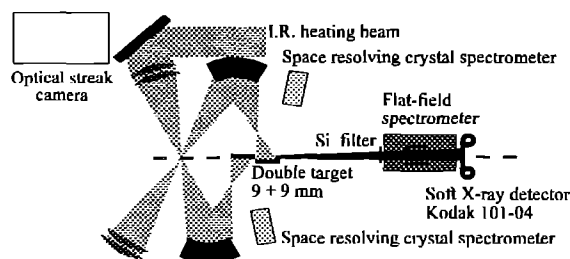


Figure 1. Schematic diagram showing the experimental geometry for the Ne-like Y double target experiment.

angular distribution of the lasing emission⁸. An optical streak camera was used to monitor the drive laser pulse profile and the output was captured using an 8 bit CCD and frame store system⁹.

RESULTS

Lasing action was observed on the 15.5 nm Ne-like Y soft X-ray laser transition with a measured gain coefficient of 3 cm⁻¹ from targets of up to 15 mm (9+6 mm double target). However, for the majority of shots no increase in the output signal was observed for target lengths up to the maximum examined, of 9 + 9 mm. The only exception to this was for one shot (Figure 2) where the drive laser pulse profile was not a flat topped Gaussian but was as depicted in figure 3. Target

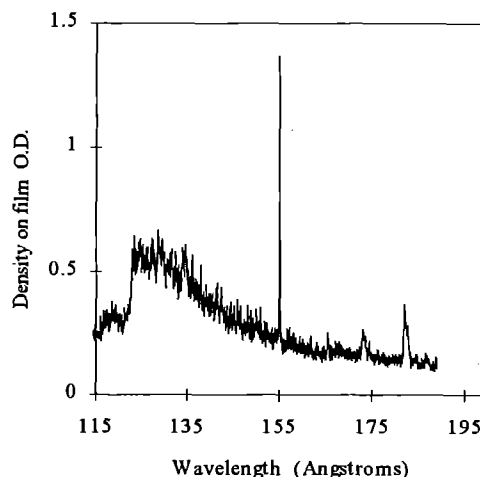


Figure 2 Densitometer trace showing the dominance of the 15.5 nm lasing line. The spectra was taken through a Si filter and the L edge at 123 Å is clearly observable.

illumination uniformity was examined but this suggested that the drive beam was satisfactory.

During the experiment there was some difficulty in reproducibly fabricating high quality Y target coatings. The experimental records and data were checked, but no evidence was obtained to suggest that the 9 + 9 mm double target which produced the high intensity output was any different physically, from the other targets which produced lower output signals.

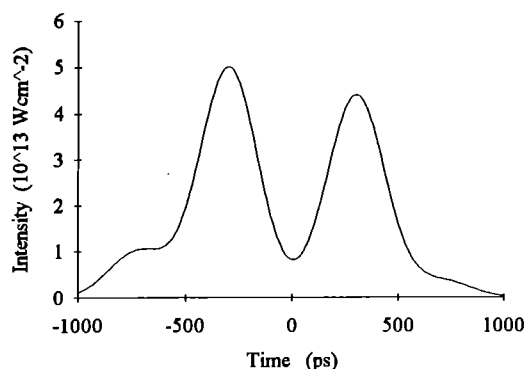


Figure 3. Optical streak camera trace showing the temporal profile of the multiple pulse.

DISCUSSION

Initial hydrodynamic modelling using 1D simulations of the plasma expansion showed three distinct advantages of using multiple pulse drive compared to the single pulse. These were; (1) The amount of absorbed incident laser energy is increased; (2) The size of the expected region for lasing action is increased and; (3) The density gradient across the region of interest is reduced by almost 50 %. These improvements in the hydrodynamic response of the Y target to the drive laser may have accounted for the increase in output. However, detailed modelling was not possible due to the lack of a complete atomic physics data base for Yttrium.

Experimental data from other soft X-ray laser double target studies had highlighted the possibility of lateral refraction¹⁰ playing a role in determining the characteristics of the output beam. Evidence for the refractive effects of propagating through an 18 + 18 mm Ge double target is shown in Figures 3(iii) and 3(iv) of reference (10) where a double lobbed structure in the lateral direction of the output beam is visible.

For 100 μm wide targets such as those typically used in soft X-ray laser experiments, the expansion gradients in the transverse direction (figure 4) are normally much larger than those in the lateral direction. It is therefore anticipated that lateral refraction effects should be most visible in the case where transverse beam refraction has been reduced or compensated for.

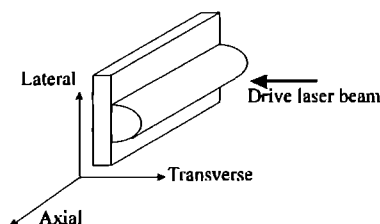


Figure 4. Schematic diagram showing the model target geometry where the soft X-ray beam propagates along the Axial direction

To study this, simulations were carried out using the POLLUX code which examined the 2 dimensional expansion of an Yttrium slab target irradiated using a Gaussian shaped spatial profile, where the density contours were examined at 25 ps intervals throughout the plasma expansion. For the temporal drive pulse profile shown in figure 3, the code predicted an interesting phenomena occurring near the peak of the second high intensity drive pulse (time $\approx$ 300 ps). The second high intensity pulse preferentially heats the centre of the ablated plasma and causes an additional lateral expansion of the plasma. This leads to a central electron density dip being formed. Such a density structure should be ideal for the propagation of a soft X-ray laser beam¹¹ providing that the spatial gain profile is not dramatically altered. Although compensation for lateral beam refraction has been proposed using a modified target substrate¹² these simulations would suggest a new route for obtaining such an effect without the complexity of a specialised target. For the case of the standard 650 ps flat topped Gaussian drive pulse the simulation

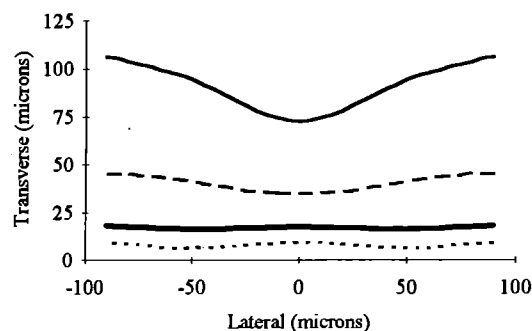


Figure 5. Numerical simulation showing the density gradients present when a multiple pulse is used to drive a slab Y target. The thick solid line shows the location of the critical surface for the 105 μm drive beam and each contour represents a factor of 2 change in density.

predicted an electron density gradient which peaks on centre and slowly falls away in the wings as expected.

CONCLUSION

Hydrodynamic modelling of a multiple pulse driven Yttrium target has indicated that two effects may account for the significantly larger lasing output observed, compared to the single drive pulse case. These are; (1) The reduction in the transverse density gradient and; (2) The production of a lateral density dip. The reduction of the transverse density gradients has been used by many groups world wide and is known to significantly increase the propagation length within a plasma. The modification to the lateral electron density profile using a structured drive profile producing a central dip is a new phenomena only predicted using 2 dimensional simulation. Further experimental studies including other modifications to the drive profile are planned in the near future to ascertain the presence of any dip and its magnitude.

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ACCELERATION TO 44 MEV OF ELECTRONS TRAPPED IN PLASMA WAVES GENERATED BY SELF-RESONANT WAKEFIELD AND FORWARD RAMAN SCATTERING

A. Modena¹, Z. Najmudin¹, A.E. Dangor,¹ C.E. Clayton², K.A. Marsh², C. Joshi²,
V. Malka³, C.B. Darrow,⁴ C.N. Danson¹, D. Neely¹, F.N. Walsh¹

¹Imperial College, London, UK

²UCLA, Los Angeles, USA

³Ecole Polytechnique, Palaiseau, France

⁴LLNL, Livermore, USA

⁵Rutherford Appleton Laboratory, Didcot, UK

INTRODUCTION

Particle acceleration¹ by laser-plasma interaction has gained a lot of interest lately due to new theoretical approaches^{2,3} and to the fast grow of short-pulse lasers which can finally implement these approaches. Till now only Beatwave experiments have given interesting results^{4,5} but the need of a very good matching of the density and a very homogenous plasma limited its performance to low densities.

With the advent of the new short-pulse lasers the possibility of the Wakefield method has been tried extensively but no definite result has been achieved as now. The new approach, Self-Resonant Wakefield, as opposed to the usual Wakefield does not need such a short pulse, so looks very promising. We report here of an experiment which proved this last method with great success, achieving acceleration of trapped electrons to the record energy of 44 MeV. The electric field >100 GV/m represents by far the highest collective-wave field ever produced in a laboratory.

The observation of wavebreaking at the higher density is also a very interesting feature which is the subject of another publication⁶.

SELF-RESONANT WAKEFIELD

This method^{2,3}, that came out only in the last years, involves pulses which are a few plasma periods long (ω_p^{-1}). In this case a small plasma wave is created by the Wake from the sharp rise of the laser pulse enhanced even more sometimes by the local pump-depletion due to Raman side-scatter that grows quickly at the beginning of the pulse. From this "noise" source Forward Raman Scattering (FRS) can grow. FRS is a 4-wave mixing process where the Stokes and anti-Stokes scattered waves (respectively down-shifted and up-shifted by ω_p) beat together with the laser wave to further stimulate the plasma wave as it propagates through the plasma. In the high intensity case (when Vosc, the electron quiver velocity in the laser field, is close to c) the plasma wave modulates the group velocity of the laser (similar to the photon acceleration model) at the plasma period and thus bunches the laser intensity longitudinally (as well as radially in case self-focusing is taking place⁷) and this effect drives even more the plasma wave. For this reason the plasma wave does not need a resonant laser pulse (as in the case of the Wakefield) but the laser itself gets bunched to provide a sort of series of micro-pulses matching the resonant condition.

EXPERIMENTAL SET-UP

The experiment was made using the Vulcan CPA 1 μ m laser⁸ which could give up to 25 J in 0.7-1 ps in a 20 μ m diameter spot with typical values of 20-25 TW of power and an intensity of 5×10^{18} W/cm².

The laser was focused onto the well-defined edge of a 4 mm diameter, laminar plume of helium gas from a pulsed, supersonic gas jet⁹ located 2 mm below the focal region. The maximum intensity of 6×10^{18} W/cm² is sufficient to doubly-ionise the helium gas over at least 2 mm into the jet. The plasma density which varied with the backing pressure of the jet was measured (through the frequency shift of the anti-Stokes sidebands) to be linear with backing pressure from 5 bar to at least 18 bar where the electron density was 1.5×10^{19} cm⁻³. The two key diagnostics were the energy spectra of trapped and accelerated electrons in an f/60 cone centred on the laser axis and the forward scattered electromagnetic spectra in the full f/5 for the incident laser cone. Due to the high intensity of the laser a huge amount of second harmonic was generated (conversion efficiency of ~10-3) and we measured it both, spectrally, in the forward direction (which was also modulated by the plasma wave) and at 30° imaging it onto a CCD.

RESULTS

Figure 1 shows the measured electron energy spectra corresponding to the range of plasma density scanned (i.e. from 0.5 to 1.5×10^{19} cm⁻³). All three cases were at a laser power of approximately 25 TW. The data represented by symbols was taken with biased silicon surface-barrier detectors while the solid curve was obtained using a fluorescer/film combination, giving a continuous spectrum from 13 to 32 MeV and the vertical scale was adjusted to overlay the solid triangles for the same shot. The number of accelerated electrons at a given energy and the maximum electron energy both show a dramatic increase as the plasma density is increased to 1.5×10^{19} cm⁻³: the number of electrons above 20 MeV increases by at least three orders of magnitude and the accelerated electron distribution is rather flat up to 30 MeV where it begins to decrease with energy up to 44 MeV, which is the detection limit. We interpret this sudden increase in accelerated electrons and maximum energy together with the broadening of the electromagnetic

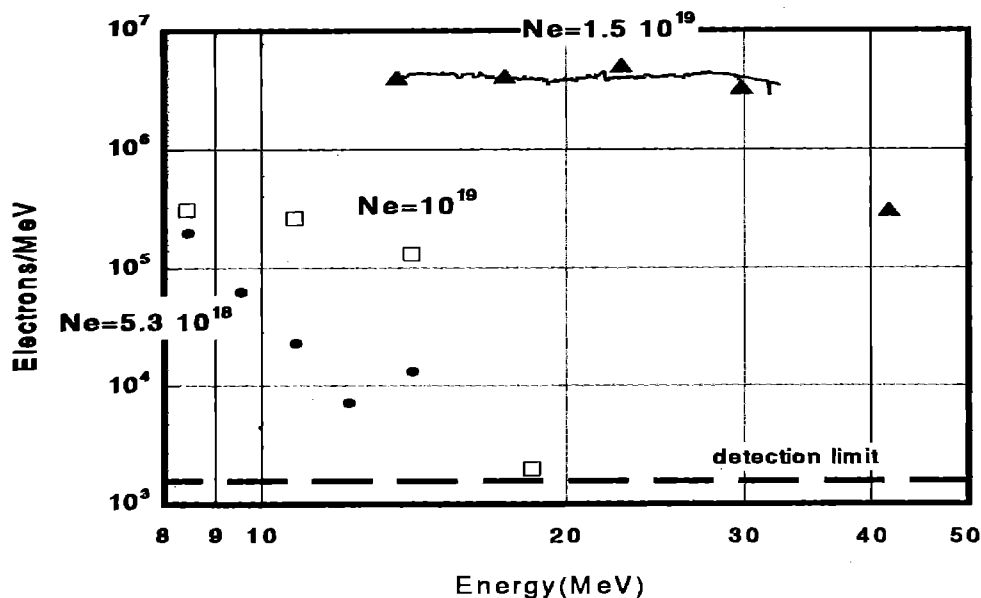


Fig. 1 Electron energy spectra at three different plasma densities

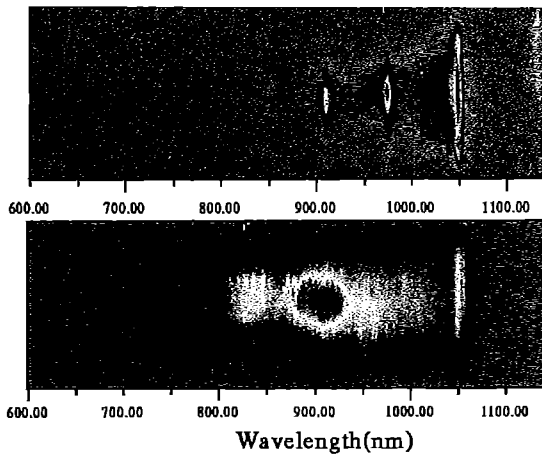


Fig. 2 Laser spectra images at $5.4 \times 10^{18} \text{ cm}^{-3}$ (above) and $15 \times 10^{18} \text{ cm}^{-3}$ (below)

spectrum as the signature that wavebreaking has occurred. The copious increase in the number of electrons accelerated at the highest plasma density is consistent with the wave trapping the bulk of the plasma thermal distribution function rather than a few tail particles at low plasma densities. Indeed the (detection limited) maximum electron energy of 44 MeV is not too far from the absolute maximum of 70 MeV that a test electron would obtain limited by dephasing in an ideal plasma wave with $\xi = 1$. A peculiar aspect of the measured spectrum at high density is that it is flat from 15 to 30 MeV. This is due to the fact that the observation is made in only an $f/60$ cone in the exact forward direction. The highest energy electrons are those that traverse the wave in the region of the highest longitudinal fields, i.e., right down the axis of the plasma wave. Lower energy electrons necessarily experience more of the radial fields and would tend to have a larger angular spread as a result.

The two pictures in Figure 2 represent the raw measured laser spectra at two different number densities (5.4 and $15 \times 10^{18} \text{ cm}^{-3}$), the pictures were processed, integrated vertically over space and corrected for the different transmission functions and sensitivity of the optics and replotted in Figure 3 as a function of frequency to highlight the periodicity of the sidebands

At the lower density the upshifted anti-Stokes and downshifted Stokes signals at $\Delta\omega/\omega_p = \pm 1$ are clearly visible as is the transmitted pump at $\Delta\omega/\omega_p = 0$ and the second and third anti-Stokes sidebands. These signals are clearly sharply peaked and their width indicates that the plasma wave which generated these signals must have a coherence time on the order of the laser pulse. The dashed curve in Fig. 3 shows the spectrum when the plasma density is increased to $1.5 \times 10^{19} \text{ cm}^{-3}$.

Because the scale has been normalised to ω_p the sidebands still have integer values on this scale. The most startling feature is the tremendous broadening of the individual anti-Stokes peaks at this higher density. This broadening is characteristic of wavebreaking and is mainly due to the loss of coherence as the wave breaks. From another point of view, as we approach wavebreaking the laser light no longer scatters off a collective mode of the plasma but instead scatters off the recently trapped electrons which are still periodically deployed in space. The Doppler shifts one expects for electrons moving at a relativistic $\gamma = 4$ within a range of angles corresponding to $f/5$ focusing is approximately $\pm \omega_p$ which could explain the "filling in" of the satellites.

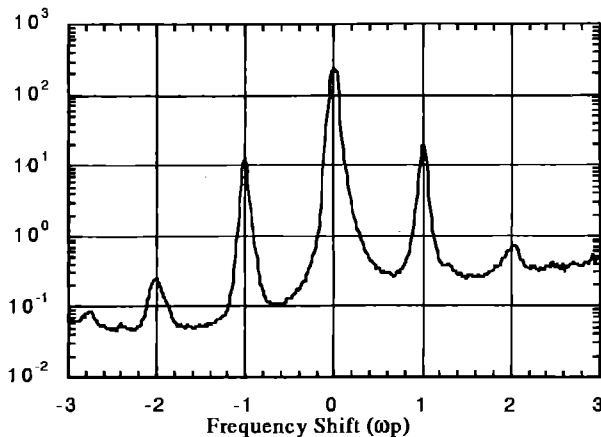


Fig. 4 Second harmonic forward spectrum at $\text{Ne} = 6 \times 10^{18} \text{ cm}^{-3}$

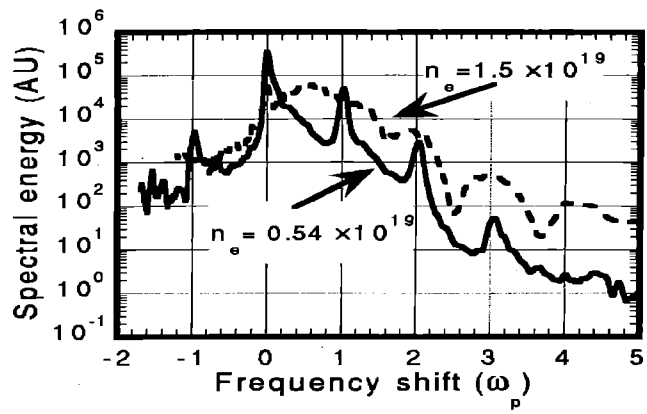
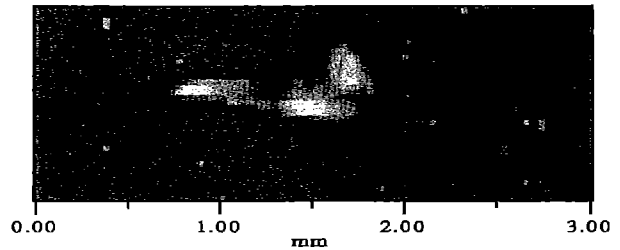


Fig. 3 Corrected laser spectra

OTHER DIAGNOSTICS

The creation and growth of the plasma wave was also monitored by its interaction with the second harmonic (0.5 mm) light generated by the laser, which we used like a probe. Figure 4 shows the second harmonic forward spectrum at a typical density of $6 \times 10^{18} \text{ cm}^{-3}$. As the sensitivity of our CCD decreased sharply at $1.1 \mu\text{m}$ we could not resolve the Stokes sidebands in the laser transmitted spectrum whereas in the visible range instead the sensitivity is flat and so we could get nice symmetric spectra. The second harmonic was emitted in all directions so we imaged the interaction area onto a CCD at 30° in green. The typical images in Figure 5 show very interesting features especially in the case of Helium where there is sign of filamentation which extends much further than the Rayleigh length ($300\text{--}350 \mu\text{m}$ in our case).

IN HELIUM



IN HYDROGEN

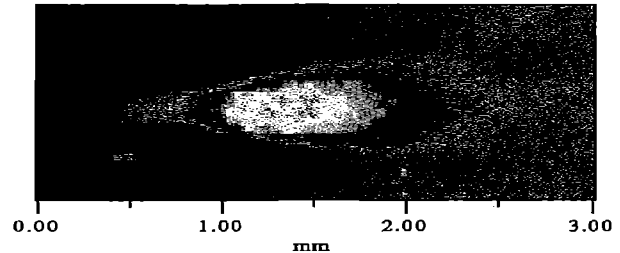


Fig. 5 Second harmonic images of the plasma

CONCLUSIONS

We observed the creation, growth and breaking of high amplitude plasma waves generated by self-resonant Wakefield and Forward Raman Scattering. These waves trapped electron from the bulk of the electron distribution function to give a large amount of electrons accelerated. Electrons, with an energy up to the detection limit of 44 MeV, were observed. The electric field implied in excess of 100 GV/m represents by far the highest collective-wave field ever produced in a laboratory. This observation has an important application in the areas of particle accelerators and energetic electron sources.

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ANGULAR DEPENDENCE OF THE ABSORPTION OF A 350 FS KrF PULSE ON SOLID TARGETS

M. Borghesi¹, D. Riley², S. M. Viana¹ and O. Willi¹

¹ Imperial College of Science, Technology and Medicine, London

² The Queen's University, Belfast

INTRODUCTION

The interaction of intense pre-pulse free, picosecond and subpicosecond laser pulses with solid targets opens up the possibility of producing hot, high density plasmas. These plasmas are relevant for astrophysical studies or for their application to the generation of bright, short-lived coherent and incoherent x-ray sources^{1,2}.

A key issue for the success of all such experiments is the understanding of the mechanisms through which the laser energy is transferred to the plasma. In a recent experiment the interaction of a 12 ps KrF Raman amplified pulse with solid targets was studied. It was observed that the absorption process could be reasonably well modeled by a combination of resonance and collisional absorption³. For p-polarized light, the maximum absorption was found at an angle of 10 degrees, giving a density scale length of about 4 μm .

EXPERIMENT ARRANGEMENT

The SPRITE laser produces, in the CPA configuration, KrF pulses of 350 fs duration at a wavelength of 248 nm. The duration of the pulse is short enough that no significant hydrodynamic motion occurs during the pulse. The short pulse is superimposed on an ASE pulse with an intensity of 10^9 Wcm^{-2} . The laser pulse is focused with an $f/3.5$ off axis parabola onto targets that consist of 300 nm thick Al layers overcoated onto a glass slide. The average irradiance on target is about 10^{17} Wcm^{-2} in a focal spot of 30 μm in diameter. The level of absorption was measured with an integrating Ulbrecht sphere. The targets were mounted on a translatable rotating stage, driven by a calibrated stepper motor. The target rotation axis was changed during the experiment from horizontal to the vertical direction allowing s and p polarization to be studied. The position of the target was controlled with a telescope alignment system. In addition, the light backscattered into the parabola was measured with a photodiode.

EXPERIMENTAL RESULTS

Only preliminary results are shown here since the data analysis is still in progress.

The level of the absorbed energy was measured as a function of the angle of incidence. Two different sets of measurements were performed, with s and p-polarized light. The results of these two series of measurements are shown in the graphs 1 and 2. The overall shot-to-shot energy variation of the data used in the graphs is less than 30%.

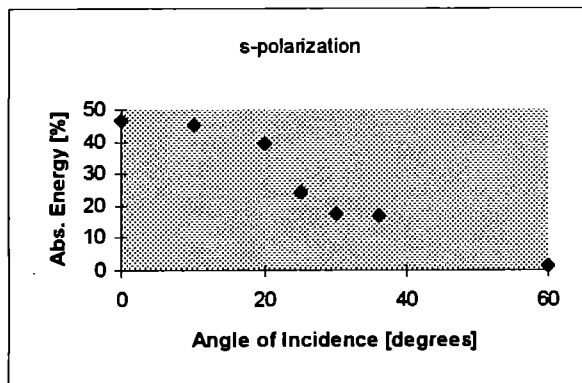


Fig. 1 Absorbed Energy fraction versus Angle of Incidence for s-polarized light.

Each point on the graph is the average of at least 3 measurements. The fraction of absorbed energy for normal incidence is about 47% of the incident energy. As expected, the absorption for s-polarized light decreases for larger angles of incidence. In fact, the absorption for this

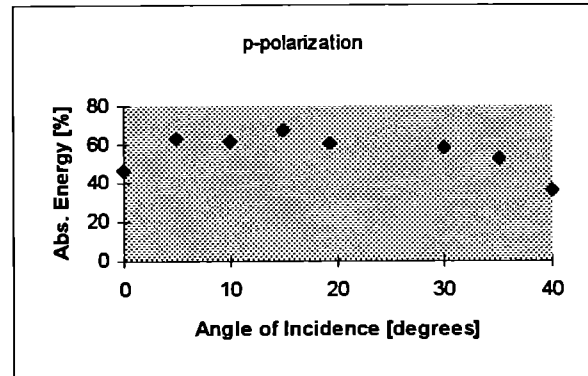


Fig. 2 Absorbed Energy fraction versus Angle of Incidence for p-polarized light.

polarization is mostly collisional. For p-polarized light, the graph of figure 2 shows a broad peak with a maximum absorption fraction of about 70% at 15 degrees. In this case the absorption will be both resonant and collisional.

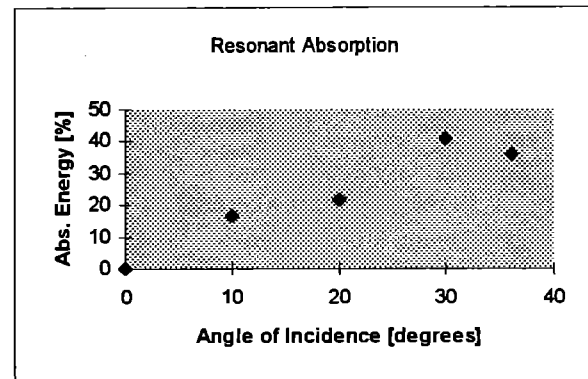


Fig.3 Angular dependence of the resonant absorption component, deduced by subtraction of the absorbed energy fraction for s-polarized light from that for p-polarized light.

Subtracting the absorption fraction for s-polarised light from that for p-polarized light, it is possible to deduce the angular dependence of the resonant absorption component, shown in the graph of figure 3. A peak with a maximum absorption fraction of about 40% at 30 degrees is evident from the graph. Taking into account that the optimum angular condition for resonance absorption⁴ is given by

$$\sin\phi = 0.8(\omega L/c)^{-1/3}$$

where ϕ is the angle of incidence, ω the laser frequency, the density scale length L of the plasma is estimated to be about 0.16 μm .

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GENERATION OF HIGH HARMONICS ON THE SPRITE CPA LASER SYSTEM.

S.G.Preston¹, M. Zepf¹, A.Sanpera¹, W.J.Blyth¹, C.G.Smith¹, D.Neely², K.Burnett¹, M.H.Key^{1,2}, A.A.Offenberger³, J.S.Wark¹.

¹Clarendon Laboratory, Department of Physics, University of Oxford, Parks Road, Oxford OX1 3PU, UK,

²Rutherford Appleton Laboratory, Chilton, Didcot, Oxon, OX11 0QX, UK,

³University of Alberta, Edmonton, Alberta, T6G 2G7, Canada.

INTRODUCTION

Recent advances in short-pulse laser technology have stimulated much research into the efficient production of coherent radiation in the extreme ultraviolet (XUV) by the generation of high odd-integer harmonic orders of a fundamental laser frequency. Most of this research has been performed using either 1.05 μm Nd:YAG or 800 nm Ti:sapphire systems. These exploit the frequency dependence of the plateau in the atomic harmonic response, given by $E_{\text{max}} = I_p + 3U_p$ ⁽¹⁾, where I_p is the ionisation potential and U_p is the ponderomotive energy ($U_p = e^2 E^2 / 4m_e \omega^2$), to produce very high harmonic orders. This limits the ultimate harmonic generation efficiency, both in phase matching the high harmonic with the fundamental, and in keeping the laser intensity below the saturation intensity to maintain a high population of atoms. We present experimental studies showing that a shorter wavelength laser, namely the Sprite 248 nm KrF laser operating in CPA mode is better suited to harmonic generation, producing a higher harmonic response and generating shorter wavelengths than previously seen. This is achieved in part by generating harmonics from ions rather than atoms: something that is not possible with long wavelength systems.

We report on quantitative studies of harmonic generation from a KrF laser, from 7th (355 Å) to 35th harmonic (71 Å) in Neon and to 37th harmonic (67 Å) in Helium. These are the shortest harmonic wavelengths generated to date. Further, we report high generation efficiencies of the 7th harmonic (355 Å) in Helium. These results are well matched to simulations of the single atom/ion response of Helium and Neon at the intensities present in our laser focus.

THE EXPERIMENT

The experiments were performed using the Sprite KrF laser at the Central Laser Facility at the Rutherford Appleton Laboratory, Chilton, Didcot, U.K. The laser, in chirped pulse amplification mode⁽²⁾, delivered up to 250 mJ on target in a 380 fs full-width at half-maximum (FWHM) pulse at 248.6 nm. The multimode beam was focused using a 100 cm OAP with a 10 cm diameter beam, producing a 6 times diffraction limited focal spot of 18 μm FWHM with a mean focused intensity of just over 10^{17} W/cm² over a length of approximately 700 μm .

A solenoid-valve gas jet operating with sub-atmospheric to 40 bar backing pressure provided a gas target of Helium or Neon with densities from 10^{17} to 10^{19} atoms/cm³ and gas lengths between 0.5 and 2 mm. Densities produced at the laser focus by the gas jet were determined by recording and analysing the Thomson scattering spectrum⁽³⁾ from the plasma formed by optical ionisation by the fundamental laser.

Harmonics generated at the focus were imaged using a curved gold-coated mirror through a flat-field XUV spectrometer containing a Hitachi variable line spaced 1200 g/mm grating and were time integrated onto a Galileo double microchannel plate (MCP). The MCP image was coupled to a Photonic Science DarkStar intensified CCD camera using a 4:1 reducing fibre optic coupler. The CCD output was stored using a computer controlled image acquisition device. (The absolute response of the MCP and CCD camera was calibrated against film⁽⁴⁾). Carbon or aluminium

filters were used to discriminate against the fundamental laser light and to attenuate the lower harmonics.

RESULTS

A typical lineout of a Helium harmonic spectrum is shown in figure 1. This plot is uncorrected for instrument or filter response. Note the exceptionally high level of the 2nd order spectrum, which is because the blaze angle of the grating becomes optimised for second order diffraction below about 100 Å. We saw no harmonics in first order above the 37th, as their intensities were below our detection threshold. The carbon filter cuts off wavelengths below 44.7 Å, which corresponds to harmonics above the 55th. Figure 2 shows the conversion efficiencies (harmonic energy/laser energy) for the harmonics above the 7th in Neon. These numbers take into account the absolute response of the MCP, the grating response⁽⁵⁾, the focusing mirror response, the filter transmission and the collection solid angle of the spectrometer.

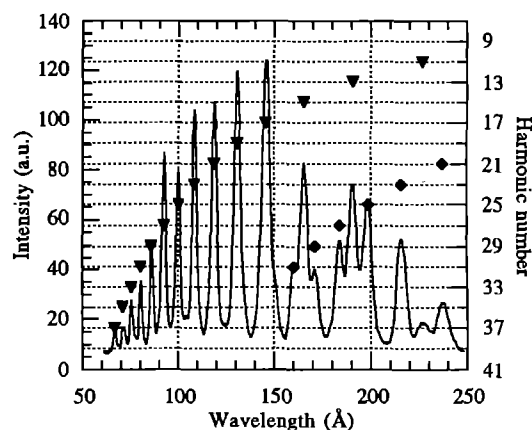


Figure 1. Lineout of a typical harmonic spectrum from Helium using the 100 cm OAP. The gas density is about 5×10^{17} atoms/cm³. The grating dispersion curve is shown as triangles (first order), and diamonds (second order). Note the high levels of the grating response in 2nd order. This spectrum has not been corrected for instrument or filter responses.

Measurements of the angular distribution of harmonics produced using a 33 cm OAP⁽⁶⁾ indicate that KrF harmonics up to at least the 19th cover the full solid angle of the laser beam. Because of the multimode beam structure, we do not expect this to change for higher harmonics. The 7th to 13th harmonics are taken from a different laser shot sequence than those from 15th upwards (and used Aluminium rather than Carbon filters). This is because the MCP detector had to be moved to cover this wavelength region. The simulated single atom/ion Neon responses for a Gaussian focus are shown in the same figure for comparison. Note the sharp change in the response for Neon at the 9th and 21st harmonic. This is due to the change from neutral Neon to Ne⁺ and from Ne⁺ to Ne²⁺ respectively as the dominant harmonic generator. A similar effect occurs for the transition from neutral Helium to He⁺ at the 13th harmonic. Also of note is the difference in conversion efficiencies between Helium and Neon. The 7th, 9th and 11th harmonic efficiencies for Helium are over an order of magnitude greater than those for Neon. After this Neon recovers, with both gases ending up with approximately the same efficiencies at the

35th harmonic, but with Helium showing a marginally higher conversion efficiency up until the 31st harmonic. This is contrary to the work of Kondo et al.⁽⁷⁾ which showed Helium and Neon with essentially the same response from the 9th to the 23rd harmonic, but with Helium marginally lower after the 13th.

The harmonic response of Helium exhibits several sharp changes in the slope of its curve. These discontinuities occur after the 13th, 19th, and 29th harmonics. Considering the absolute energy produced at the harmonics frequencies, we see that we generate up to 30 nJ at 355 Å (7th harmonic), 1 nJ at 191 Å (13th harmonic), 1.5 pJ at 85 Å (29th harmonic), and 0.25 pJ at 67 Å (37th harmonic). This corresponds to 5.5×10^9 , 2×10^8 , 6×10^4 , and 8.5×10^3 photons respectively.

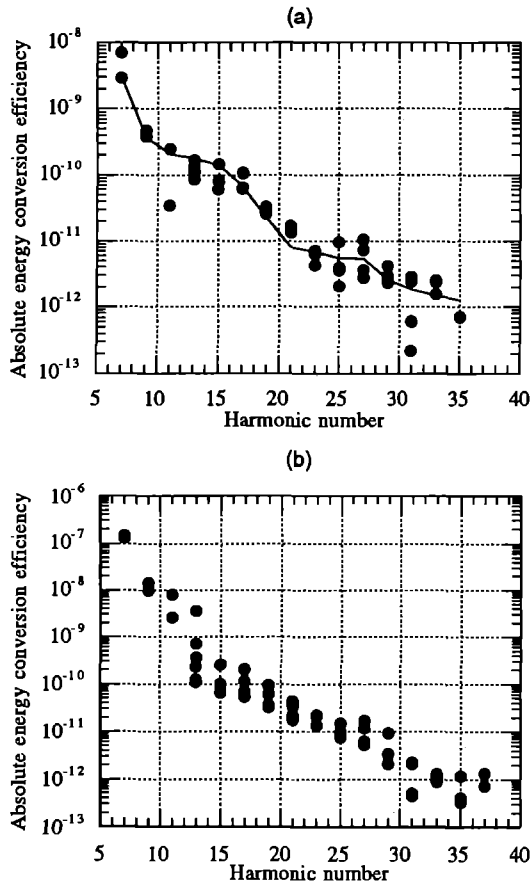


Figure 2. The absolute harmonic conversion efficiencies for (a) Neon, and (b) Helium. The simulated total Neon harmonic response to a Gaussian focus is shown as a solid line for comparison. The simulated response includes Neon, Ne^+ , and Ne^{2+} .

SIMULATIONS

When making a full simulation of the experiment, one should consider not only the single atom response but also all the issues of beam propagation. However, numerical simulations of the individual atoms can match the experiments reasonably well by combining the response of the neutral single atom response with that of their ions for a full pulse⁽⁸⁾.

Our numerical simulations are performed for a number of laser intensities in the range of 10^{15} - 10^{17} W/cm² at a fundamental wavelength of 248 nm for Helium and its ion and for the Neon atom and its charge states up to Ne^{3+} . The laser pulse length has a sine-squared temporal profile with a FWHM of 350 fs. These intensities range from close to well above the saturation intensity of

the neutral and it is therefore necessary to study the contributions from both neutral atoms and ions. The simulations are corrected for the variation of intensity in the laser focus. Only simulations for Neon are shown here in figure 2. Those for Helium will be included in a future publication⁽⁹⁾.

A comparison of the simulations with experiment indicates that we are seeing very strong ionic response from He^+ , Ne^+ , and Ne^{2+} . This is the first experiment to unambiguously show the generation of harmonics from doubly-ionised Neon. Analysis of 1st order spectra failed to reveal any indications of harmonics above the 37th. Interpretation of the 2nd order Helium spectrum is difficult because of the overlap with the 1st order spectrum, but there are indications of harmonics up to the 45th sitting on the wings of the first order spectrum. A full analysis of this second order data is currently underway.

CONCLUSION

In conclusion, we have studied the generation of high harmonics in underdense, optically-ionised Helium and Neon plasmas. We show that multiply charged ions play an important role in the generation of high energy harmonics from a short-pulse, high intensity, ultraviolet laser: generating to 37th harmonic (67 Å) in Helium, and 35th harmonic (71 Å) in Neon in 1st order and showing indications of harmonics up to 45th (55 Å) in Helium in 2nd order. Furthermore, we show that Helium atoms can produce very high conversion efficiencies for the lower harmonics of 248 nm laser radiation, producing energies up to 31 nJ, and powers up to 80 kW, for the 7th harmonic (355 Å). These results indicate the need for short wavelength driver lasers for the efficient production of high energy XUV radiation by harmonic generation. A more detailed discussion of these results will be presented in a forthcoming publication⁽⁹⁾ which is under preparation.

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HIGH FUSION YIELDS FROM PICOSECOND LASER IRRADIATED DEUTERATED TARGETS.

A.P.Fews¹, P.A.Norreys², A.E.Dangor³, A.R.Bell³, B.A.Hammel⁴, F.N.Beg³, M.Glinasy⁴, P.Lee³, M.Tatarakis³.

¹Department of Physics, University of Bristol, Tyndall Ave., Bristol BS8 1TL

²Central Laser Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxon OX11 0QX,

³Imperial College of Science, Technology & Medicine, Prince Consort Road, London SW7 2AZ

⁴Lawrence Livermore National Laboratory, Livermore, California 94551, USA

Fusion yields of up to 4.4×10^9 were measured when planar deuterated polystyrene targets were irradiated with $1.054 \mu\text{m}$, 1.3 psec laser radiation and a focused intensity of 1×10^{18} - $2 \times 10^{18} \text{ W cm}^{-2}$. The neutron energy spectra are consistent with an isotropised beam-target interaction, whereas a thermonuclear source is considered unlikely. The fusion yield represent Q values (fusion energy released/laser energy on target) of up to 2.2×10^{-4} .

The fast ignitor scheme for Inertial Confinement Fusion (ICF) has recently been proposed [1]. In this scheme, short pulse high intensity laser radiation creates energetic electrons at the critical density surface of an already compressed high density plasma. These electrons are transported into the deuterium-tritium fuel and heat it to thermonuclear temperatures before it has time to disassemble. There is a potential for a considerable saving of laser drive energy as the creation of a hot spark at the centre of the compressed fuel is no longer required and the symmetry requirements can be significantly relaxed. For this scheme, fundamental experiments need to be carried out to characterise the generation of both fast electrons and ions and their associated energy transport through-out the target.

Measurements of the number of fast electrons and their characteristic temperature can be achieved by the established technique of K-alpha spectroscopy [2]. However, measurement of fast ion transport is complicated by the extremely strong electrostatic fields that are generated. Ion measurements made outside the target will be characteristic not of their behaviour in the plasma during the interaction, but of the expansion of the plasma, which is driven by the fast electrons [3]. Thus to determine the fast ion behaviour in the plasma during the interaction, other more direct diagnostics need to be used.

Position sensitive neutron spectroscopy is just such a technique. Energetic deuterons, created during the interaction of a high intensity laser pulse, produce neutrons by fusing as a result of collisions, either thermally or by a beam-target type interaction. Neutron measurements in the forward and backward directions are a very sensitive measure of any systematic motion of the centre of mass of the reacting ions and will register either as an upshift or downshift in neutron energy, depending on the position of the detector. Even a very small systematic centre of mass motion will register as a shift in energy between the detectors and be observable. This is illustrated in figure 1 which shows the calculated energies of the emitted neutron as a function of the energy of a deuteron impacting a stationary deuteron, for neutrons emitted in the forward and backward directions with respect to the incident particle. The technique can detect very small relative motions - for instance, a 10 keV deuteron interacting with a stationary deuteron will give rise to a relative shift of $\sim 200 \text{ keV}$ between the forward and backward neutron energy spectra. It also has a very large dynamic range and will detect centre of mass motions from 10 keV - 10 MeV and beyond. It should differentiate between ion motion into and out of the target - 2.5 dimensional particle-in-cell kinetic simulations predict a substantial percentage of the laser energy is transferred to ions directed into the plasma [4]. Similarly, it can differentiate fast ions impacting deuterons deposited on both diagnostic equipment and the walls of the vacuum vessel [5].

The experimental configuration used was similar to that in our other experiments [6][7]. The VULCAN laser, operating at a wavelength of $1.054 \mu\text{m}$, gave pulses of 1.3 psec duration[8]. An equivalent plane monitor showed the laser was focused to a $30 \mu\text{m}$ focal spot. An off axis parabolic mirror of 44 cm focal length focused 8 - 20 J of p-polarised laser radiation at 30° angle of incidence onto massive flat deuterated polystyrene (C_8D_8) targets that were $120 \mu\text{m}$ thick.

The neutron detectors consisted of CR-39 nuclear plastic track detectors which were covered in 3mm of lead to stop all energetic charged particles from the plasma impinging on the detector. The neutrons can then be detected by observing the knock-on protons in the CR-39. CR-39 records charged nuclear particles as tracks which are etched into the surface of the detector after the exposure. Each incident ion produces a separate etched track, the dimensions and depth of penetration of which enable the energy and atomic number of the fast ions to be individually determined [9]. In this way, neutrons of a particular energy produce a characteristic distributions of proton track size which is different from that of neutrons of a different energy. These distribution shapes are extremely sensitive to the incident neutron energies $\leq 3 \text{ MeV}$. A maximum entropy unfolding technique, similar to that used in image deconvolution [10], is then applied to the recorded track size distribution to produce a neutron spectrum. This technique selects the optimum neutron spectrum by matching a predicted proton track size distribution to the measured data. The spectrum which is produced is the flattest (maximum entropy) neutron distribution where the measured and predicted proton distributions are consistent with each other. The resulting spectrum will then only contain those significant features which are required to fit the predicted proton track distribution to the measured data.

The detectors were calibrated for fusion yield principally with a test exposure to a known 2.5 MeV neutron flux from the National Physical Laboratory, United Kingdom. This conversion was confirmed by a calculated response using data from a variety of sources of proton exposures. These two values agree to within 20%. The track diameter of each individual proton track was recorded by automated image analysis, which can then be related to the particle energy [9]. The identity of the recorded tracks in the CR-39 was confirmed by individually labelling a selection of tracks and repeating the etching process in stages until all the tracks were "ranged out". This provided an unambiguous determination of the particle species, and confirmed that all of the sample were indeed protons.

The detector placed in front of the target (which defines the backward direction) was located 20 cm from the target at an angle of 22° to the horizontal and 35° to the vertical planes respectively and the forward direction detector was located 20 cm behind the target at an angle of 0° to the horizontal and 10° to the vertical planes respectively.

Other CR-39 detectors were fielded to measure the fast ion emission from the plasma itself. The detector in front of the target in the backward direction was located at the end of an extension tube at a distance of 3.0m from the chamber centre. Standard pinhole cameras recorded the x-ray emission and ion penumbral imaging provided images of ions $> 1 \text{ MeV}$.

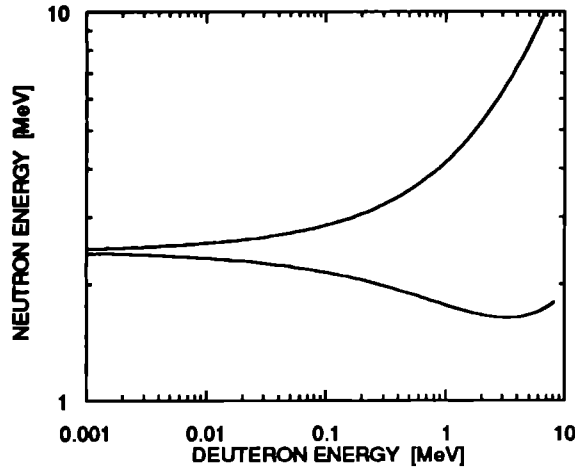


Figure 1. Neutron energies in the forward (top curve) and backward direction (bottom curve) as a function of incident deuteron energy.

Table 1 shows the observed neutron yields in the forward and backward directions (extrapolated into 4π from the measured value in the detectors) together with the laser energy incident on target. The neutron yields were in the range $4 \times 10^8 - 2.2 \times 10^9$ and do not differentiate between the forward and backward directions. The total fusion yields ranged from 8.0×10^8 to 4.4×10^9 allowing for the two branches of the $D + D$ fusion reaction. These yields represent Q values (fusion energy released / laser energy incident onto the target) of up to $Q = 2.2 \times 10^{-4}$.

The neutron spectra were calculated by unfolding the proton distribution by maximum entropy described above. Figure 2a shows the neutron spectrum in the forward direction and Figure 2b shows the neutron spectrum in the backwards direction. The spectra were integrated over six shots to improve the signal to noise ratio. The spectral distribution of the 2.45 MeV neutron from the $D + D \rightarrow {}^3\text{He} + n$ branch has a Gaussian profile. However, extensive calculations have shown that it is possible to generate a Gaussian-type neutron distribution even from very non-Maxwellian ion velocity distributions and it is not possible to deduce a thermonuclear from the neutron spectrum alone[11]. The neutron spectra both have an energy full width at half maximum (FWHM) of 680 keV. We estimate that the unfolded neutron spectra have a systematic centring error of ± 100 keV (corresponding to an upper limit in the centre of mass motion of the reacting ions of ± 8 keV) due to uncertainty in the proton track calibration in the CR-39.

The spectra are centred at 2.30 MeV and 2.35 MeV for the forward direction detector and backward direction detectors respectively, which is within the error of the technique for 2.45 MeV neutrons. These spectra are indicative of an isotropic source and cannot be of a single direction beam - target interaction, i.e. inwardly directed energetic ions interacting with cold deuterons of the target because this type of interaction would result in an up-shift of the neutron spectrum in the forward direction and a downshift in the backward direction. This equally applies to deuterons impacting the diagnostic equipment around the target. Neither can the fusion yield be produced by deuterons of energies greater than 10 keV interacting with a layer of deuterons deposited on the walls of the chamber because this would give a larger downshift in the neutron energy when observed near the chamber centre. Further, neutrons cannot be generated by lower energy deuterons impacting the walls of the chamber because a detector located at the wall showed a signal

$1/8$ of the level of the main detectors, confirming a $1/R^2$ fall off from the chamber centre.

Hot electron temperature measurements, made by analysis of K alpha x-ray spectra obtained under identical irradiation conditions but with multilayer metal targets, indicate that the electron temperature is between 50 - 100 keV provided that metal conductivity is assumed. However, if Spitzer conductivity is used, then resistive inhibition is increased and a larger electron temperature (~ 600 keV) is required to penetrate to a given depth in layered metal targets[12]. T.H. Tan et al [13] have used a simple isothermal plasma expansion model to relate the fastest ion velocities to the hot electron temperature by the expression $T_h = 0.0138 E_{\text{max}}$ (keV). We measured maximum ion energies of 8.0 (± 3.0) MeV at $2 \times 10^{18} \text{ W cm}^{-2}$ intensity, suggesting a hot electron temperature of 100 (± 40) keV for these conditions.

The neutron spectra strongly suggest their production by fusion of deuterium nuclei. Given the small Doppler shift and spectral width (680 keV from figure 2), the obvious hypothesis is that the neutrons originate in a thermonuclear plasma. If we hypothesise that (i) the neutrons originate in a hemisphere (radius R) of uniformly heated solid density plasma (temperature T_{plasma}) centred on the middle of the laser spot, (ii) the electron, deuteron and carbon temperatures are equal, (iii) the laser energy absorbed into this plasma is E_{absorbed} and (iv) fusion reactions occur during the time for hydrodynamic expansion $\tau_{\text{hydro}} \sim R / c_{\text{sound}}$ where c_{sound} is the isothermal sound speed, then the neutron yield should be:

$$N \approx \left(\frac{E_{\text{absorbed}}}{10 \text{ J}} \right)^{3/2} \left(\frac{T_{\text{plasma}}}{30 \text{ keV}} \right)^{-7/2} 1.4 \times 10^8 \quad (1)$$

where the approximation has been made that the neutron production rate is proportional to $T^{5/4}$ in the range of interest. If $T_{\text{plasma}} = 30 \text{ keV}$ and $E_{\text{absorbed}} = 10 \text{ J}$, then the radius of the plasma is $R = 10 \mu\text{m}$, and the expansion time is $\tau_{\text{hydro}} = 8 \text{ psec}$. The temperature dependence is relatively weak and a low value is indicated by the large neutron yield, but T_{plasma} cannot be much less than 30 keV because the fusion rate drops off rapidly below 20 keV and the neutron spectra imply a temperature close to 70 keV [11]. Hence a thermonuclear interpretation cannot easily explain the large neutron yield. Further reasons for rejecting the thermonuclear interpretation are that (i) the Coulomb mean free path of 100 keV fast electrons is around $100 \mu\text{m}$ (see also ref. 14) which is much greater than the $10 \mu\text{m}$ radius in which the energy must be deposited to heat the plasma to thermonuclear temperatures (ii) a thermonuclear plasma is not easily established because the electron-deuteron collisional energy equilibration time is many 100's of psec (iii) the cooling time of a 30 keV plasma of $10 \mu\text{m}$ radius due to the electron thermal conduction is less than 0.1 psec.

TABLE 1.

Shot Number	Laser energy (J)	Neutron yield (forward direction)	Neutron yield (backward direction)
21/22/6/94	15.2	2.2×10^9	2.2×10^9
23/22/6/94	7.6	1.2×10^9	1.7×10^9
8/23/6/94	11.3	1.4×10^9	8.4×10^8
12/23/6/94	7.6	9.1×10^8	3.5×10^8
19/23/6/94	11.2	1.7×10^9	1.8×10^9
25/23/6/94	12.9	1.6×10^9	1.7×10^9
27/23/6/94	20.7	7.1×10^8	4.8×10^8
3/24/6/94	15.9	4.1×10^8	4.6×10^8
6/24/6/94	19.3	7.5×10^8	5.4×10^8

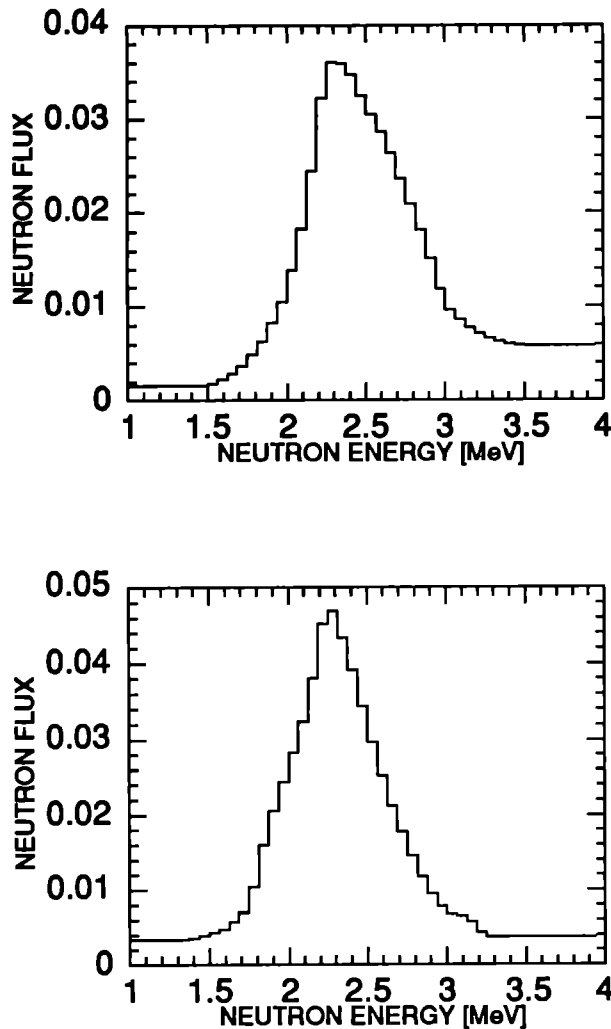


Figure 2. Neutron spectra in the forward (top) and the backward (bottom) directions.

We now analyse the possibility that a beam-target type of interaction could produce 2×10^9 neutrons. The electron density is seven times the deuteron density. If 10J of laser energy were divided in this proportion between the electrons and the deuterons, and the deuterons were heated to 100 keV (the minimum for efficient neutron production, in keeping with the ion measurements, and possibly arising from the electrostatic potentials near the target surface) then the number of energetic deuterons would be 8×10^{13} . The neutron generation cross-section [15] and the Coulomb cross sections for energy loss to cold background carbon ions, deuterons and electrons (multiplied by n_e/n_D to allow for their larger number density) are: $\sigma_{fusion} = 1.7 \times 10^{-30} \text{ m}^2$; $\sigma_{DC} = 4 \times 10^{-26} \text{ m}^2$; $\sigma_{DD} = 7 \times 10^{-27} \text{ m}^2$ and $(n_e/n_D)\sigma_{De} = (T_e/\text{keV})^{-3/2} 8 \times 10^{-25} \text{ m}^2$. The Coulomb cross sections are derived from the times given by Nishikawa and Wakatani [16] for relaxation of a 100keV Maxwellian distribution of deuterons with $\log \Lambda = 5$. If the electron temperature is greater than 7keV, the energetic deuterons lose their energy predominantly to background carbon ions. The ratio of the neutron production to the Coulomb cross section is 4×10^{-5} , so the expected neutron production would be 3×10^9 , which is close to the largest observed neutron yield. If the electron temperature is less than 7 keV, the Coulomb cross section is greater and the neutron yield less by $(T_e/7\text{keV})^{3/2}$. However,

an electron temperature of 7 keV or greater is not unreasonable since the deuterons themselves would heat the plasma as they lost energy. If the energy of the energetic deuterons were greater than 100 keV, the fusion cross section would be greater and the Coulomb cross section less, favouring an increased neutron production. We therefore conclude that a beam-target interpretation of the neutron yield is energetically feasible but requires efficient transfer of energy to deuterons, which is itself not easily explained, but might result from acceleration in the electrostatic potentials around the target surface. The isotropy of the neutrons and the lack of a large systematic Doppler shift may be taken to imply a thermonuclear origin, but equally this could be explained if the energetic deuterons were produced with some degree of isotropy or were isotropised by angular scattering off the carbon ions. Indeed, the cross section for isotropisation by angular scattering is about 6 (the carbon to deuteron mass ratio) times greater than the Coulomb scattering cross section energy for energy loss to background carbon ions. Thus a relatively isotropic deuteron, and therefore neutron, distribution would be expected.

In conclusion, we have observed fusion yields of up to 4.4×10^9 when deuterated polystyrene (C_6D_6) targets were irradiated with 8 - 20J of 1054nm laser light in a pulse length of 1.3 psec and focused intensity of $1 \times 10^{18} - 2 \times 10^{18} \text{ Wcm}^{-2}$. Overall, it appears that a thermonuclear source of neutrons is unlikely, but an isotropised beam-target interpretation may be feasible. In reality, the two possible interpretations are not distinct, since collision times are too long to ensure equipartition between species or thermalisation within species. We believe that the results presented here confirm that energy deposition from fast ions will play a significant role in the short pulse ignitor scheme for ICF.

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OPTICAL GUIDING OF 10TW,PICOSECOND LASER PULSES IN PREFORMED DENSITY CHANNELS AT HIGH INTENSITIES AND DENSITIES

A. J. Mackinnon, M. Borghesi, A. Iwase, F. Walsh* and O. Willi

The Blackett Laboratory, Imperial College

*Rutherford Appleton Laboratory,

1. INTRODUCTION

The propagation of an extremely intense laser pulse through underdense plasmas is an important issue in a number of possible applications. In the recombination X-ray laser scheme [1] a sub-picosecond laser is focused into a neutral gas to create an optically ionised (OFI) plasma. This forms a plasma where the ionisation stage is determined by the intensity of the laser and the electrons are heated predominately by ionisation heating (ATI) [2]. The efficiency and gain of the system are directly affected by propagation issues such as ionisation defocusing and filamentation which limit the length and uniformity of the plasma. In the fast igniter scheme [3] the issue of intensity dissipation in the underdense plasma of the guiding channel is also important. Any reduction in the laser intensity will affect the maximum density to which the channel forming pulse can propagate and will reduce the probability of ignition.

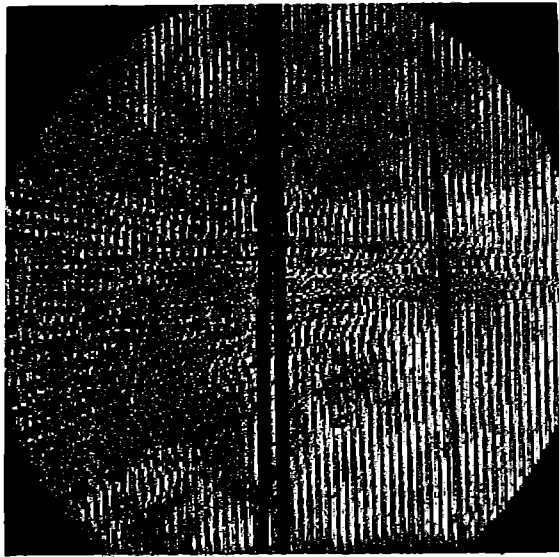
A study of the mechanisms affecting the propagation of extremely intense pulses through the underdense plasma is an interesting problem for both these applications. Refraction from density gradients caused by any ionisation will reduce the laser intensity more rapidly than diffraction. This ionisation defocusing mechanism has been studied theoretically by Rae [4] and by Burnett [5], who suggested the use of a preformed density channel as a method of overcoming it. The formation of the channel has been studied by Rankin et al [6] theoretically and the guiding of a 100ps pulse down a density channel has been experimentally observed by Milchberg and Freeman [7]. The effect of refraction on the length of the preformed channel has also been recently studied theoretically by Liu and Tripathi [8].

This paper will present experimental results that provided a measurement of the ionisation induced defocusing and studied the use of preformed channels to overcome it.

The experiment was undertaken at the Rutherford Appleton Laboratory using the VULCAN laser operated in chirped pulse amplification mode. Typical operating parameters were 15-40J on target in a pulse of temporal width (FWHM) of 1-4ps and of spatial width (FWHM) of 30 μ m (obtained with a F/4 off axis parabolic mirror). This typically gave a peak power and intensity of 5-10TW and $1-3 \times 10^{18} \text{ Wcm}^{-2}$ respectively. The laser pulse length, spectrum and focal spot were all monitored for each shot. The preformed channel was formed with co-linear pulse that was either a 1% or 10% split off the interaction beam. The laser was focused on the boundary of a solenoid operated gas jet target, with a 1mm diameter cylindrical nozzle. The plasma was diagnosed with a temporally independent probe pulse which was compressed with a separate set of gratings, the pulse was then frequency doubled using a KDP crystal. The duration of the probe beam was less than 10ps. A Moiré deflectometer [9] used the probe to detect density gradients in the plasma. A space resolved self emission channel was used to detect the time integrated plasma fluorescence normal to the propagation direction. The film pack was filtered using an interference filter with a centre wavelength of 620nm and a bandwidth of 25nm.

The problem of ionisation defocusing of the pulse is graphically illustrated in Fig 1 where a pulse with a vacuum intensity of $1 \times 10^{18} \text{ Wcm}^{-2}$ is focused into neon gas at an atomic density of $1 \times 10^{19} / \text{cc}$. It can be seen that the plasma expanded rapidly in the radial direction and broke into filaments as the laser propagated through the gas.

Abel inversion of Fig1 gave a peak electron density at the gas-vacuum boundary of $4.5 \times 10^{19}/\text{cc}$.



In order to overcome these very strong refractive effects the laser was focused into a preformed density channel.

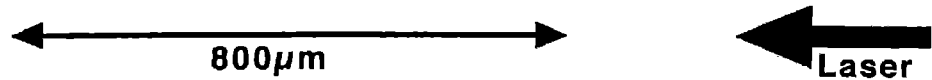
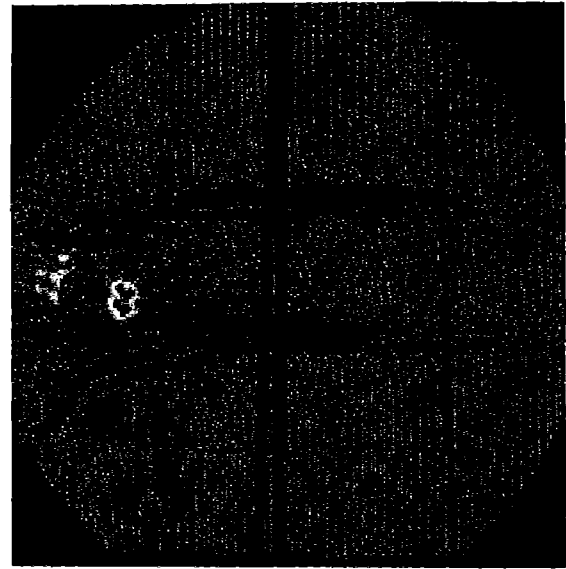


Figure 1 Strong refraction in Neon at 1×10^{19} atoms/cc.
Note : the spatial scale shown beneath Figs 1 and 3 applies to all the images in this report.

Figure 3 Main pulse focused into the density channel.

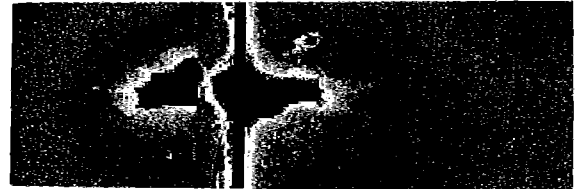
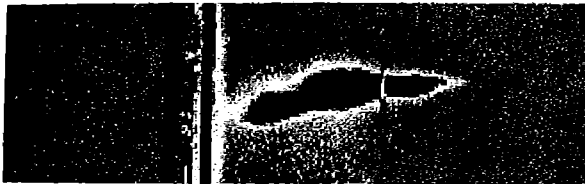


Figure 2 Self Emission confirming strong refraction.

Figure 4 Self emission showing guiding distance

The self emission in Fig 2 confirmed that the plasma expanded with propagation. A simple estimate of the refraction can be obtained by considering a ray refracted from a transverse density gradient of the same order of the laser focal spot. From fig 1 the expansion angle of the plasma was measured to be 27 degrees. The refraction angle for a plasma with an electron density of $5 \times 10^{19}/\text{cc}$, a density gradient of $30\mu\text{m}$ extending over $750\mu\text{m}$ was calculated to be 27 degrees. This agreed well with the conditions measured in Fig 1. It was also thought that refractive splitting of laser hot spots in small scale density gradients was responsible for the structure in Fig 1.

Fig 3 shows a 3ps beam focused at $1 \times 10^{18} \text{ Wcm}^{-2}$ into a preformed density channel. It can be seen that in the uniform part of the channel the refracted laser light was contained and the pulse was guided. Towards the end of the channel part of the pulse manages to escape through the channel walls while the rest of the light was effectively focused onto a high density unexpanded plasma that blocked the end of the channel.

DISCUSSION

In the absence of a plasma the diffractive spreading of a laser pulse with a gaussian spatial profile would be given by the Rayleigh length, $L_r = \pi a^2/\lambda$. For the laser parameters used in this experiment L_r ranged from 300-600 μm and the plasma would extend for over 2 Rayleigh lengths. The self emission shows that the plasma length was around 450 μm which was barely more than one Rayleigh length. It is clear from these two pictures that the intensity along the length of the plasma was severely modified by ionisation induced refraction. The length characterising the expansion of the plasma in this case was the defocussing length L_d given by [4], $L_d = (\lambda/2)N_c/N_e$. L_d was estimated to be around 50 μm for our plasma conditions, which was significantly less than a Rayleigh length. Fig 4 confirmed that the laser was guided for approximately 300 μm over Fig 2. This was equivalent to guiding over at least 6 defocussing lengths.

The deflectogram of the preformed plasma shown in Fig 5 showed that the channel had not expanded uniformly about the laser axis along the whole length of the channel.

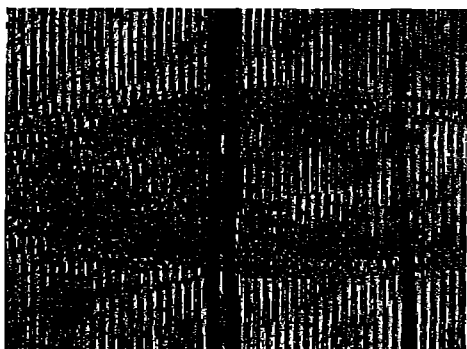


Figure 5 The preformed density channel just before the arrival of the main pulse

The non uniform axial expansion of the channel was caused by refractive defocusing of the preforming laser pulse. The plasma had a uniform width only over the first 400 μm and there was very little expansion towards the end of the

channel. The high density regions at the end of the channel corresponded exactly to the regions of self emission in fig 3. The channel closure was thought to be caused by ionisation induced refraction of the prepulse. The diminishing laser intensity as the laser propagated caused a reduction in the plasma temperature which led to very little or no expansion of the plasma at the end of the channel. Axial non uniformity of the preformed channel has been attributed to ionisation induced refraction in a recent theoretical paper by Liu [8]. It was found that the channel closure could be overcome simply by increasing the energy in the prepulse beam.

CONCLUSIONS

Ionisation induced refraction in the interaction of picosecond pulses with dense gas jet targets has been observed using time resolved optical probing. The refraction effects were overcome with the use of a preformed density channel, allowing propagation over many defocussing lengths. Further analysis and modelling is being carried out to give a more detailed insight into the feasibility of the role of preformed channels in the X-ray laser and fast igniter applications.

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ENERGY TRANSPORT IN PLASMAS PRODUCED BY ~ 300 fs KrF LASER PULSES FOCUSED ONTO SOLID TARGETS

G J Tallents¹, M G Holden¹, Y Al-Hadithi¹, P Fewes², K Nazir³, S Rose^{3,4},
P A Norreys⁴ and J Zhang⁵

¹Department of Physics, University of Essex, Colchester CO4 3SQ, UK.

²Department of Physics, University of Bristol, Bristol BS8 1TL, UK.

³Department of Physics, University of Birmingham, Birmingham B15 2TT, UK.

⁴Central Laser Facility, Rutherford Appleton Laboratory, Didcot OX11 0QX, UK.

⁵Clarendon Laboratory, University of Oxford, Oxford OX1 3PU, UK.

INTRODUCTION

The transport of energy from the focal region when high power lasers are focused onto solid targets can occur due to several different mechanisms. Thermal electron conduction axially into solid targets may be significantly modified by X-ray and superthermal electron penetration ('preheat'). However, radiation transport and heating in solid targets is effectively switched-off by using low atomic number targets such as carbon for which the X-ray emission is small. Superthermal electrons are produced when the target irradiance I for a laser wavelength λ is such that $I\lambda^2 > 10^{14} - 10^{15} \text{ Wcm}^{-2} \mu\text{m}^2$. For the experiment reported here, we could expect superthermal electron preheat as $I\lambda^2 \sim 10^{16} \text{ Wcm}^{-2} \mu\text{m}^2$, but as we shall discuss, classical flux-limited thermal conductivity describes the observed heat penetration.

METHOD

During its operation, the SPRITE CPA laser produced ≈ 0.8 TW pulses of 300 fs duration at a wavelength of $0.249 \mu\text{m}$ with high contrast ($\sim 10^{10}$) and low divergence ($\approx 20 \mu\text{rad}$)¹. Focused irradiances around 10^{18} Wcm^{-2} in a spot focus of $\approx 20 \mu\text{m}$ diameter were produced using an off-axis parabolic mirror.

We studied energy transport from the focal spot by recording X-ray emission from tracer layers of iron ($0.2 \mu\text{m}$ thick) buried below overlay plastic (CH) of variable thickness. As in previous studies², the laser was focused onto the overcoat and the penetration of the heat front was measured by recording the depth of overcoat for which the tracer layer could be observed to emit characteristic X-ray spectral lines or simply more emission due to its higher Z.

A flat crystal spectrometer with a $10 \mu\text{m}$ thick beryllium filter (for light tightness) was used to record emission from the buried layers using crystals of TLAP. The spatial distribution of the X-ray emission was measured using a pinhole camera with large pinhole ($80 \mu\text{m}$) and CCD recording. Sub-picosecond pulse experiments only produce small amounts of X-ray flux and so a penumbral X-ray imaging technique³ was used to deconvolve the X-ray pinhole images to give a final image with spatial resolution better than $5 \mu\text{m}$. Two X-ray photodiodes with filtering of $10 \mu\text{m}$ and $250 \mu\text{m}$ Beryllium were used to measure X-ray emission around energies of 1 and 3 keV respectively.

RESULTS

The X-ray image diameter as a function of the plastic overlay thickness is shown in figures 1. The depth of penetration of the heat front is $\approx 0.4 \mu\text{m}$ without significant lateral transport (the scatter in the diameters of figure 1 is believed to be mainly due to inadvertent target positioning outside the best focal position). The ratio of fluorine-like to neon-like iron emission indicates a rapid decrease of temperature with depth (figure 2).

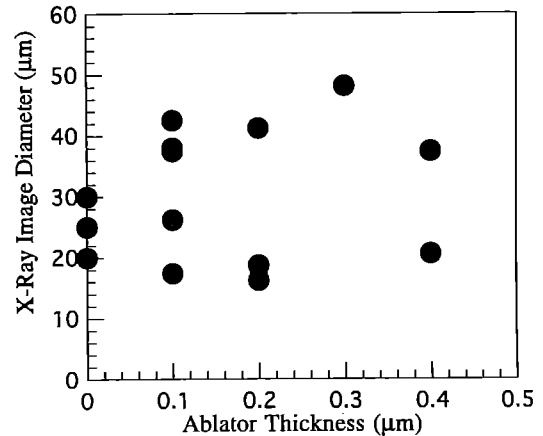


Figure 1. The measured X-ray emission diameter as a function of the depth of the signature layer. The focal spot diameter is $\approx 20 \mu\text{m}$.

DISCUSSION

One-dimensional Medusa simulations^{4,5} without superthermal electron production predict heat penetration into plastic of $\approx 1.6 \mu\text{m ps}^{-1}$ (assuming an absorption of 10^{17} Wcm^{-2} , a flux limiter of 0.1 and no lateral transport). For these 300 fs experiments, the heat penetration is expected to be $\approx 0.5 \mu\text{m}$ (for a rate of $1.6 \mu\text{m ps}^{-1}$) which is close to the measured value of $\approx 0.4 \mu\text{m}$.

The intensity contrast ratio between pre-pulse and main pulse has been measured¹ to be at least 10^{10} . Using published mass ablation rate data⁶, it can be readily shown that this level of pre-pulse will only ablate $\sim 0.01 \mu\text{m}$ of the target plastic overlay and so not affect the heat penetration measurements directly. However, the pre-pulse may assist the absorption of the laser energy by the generation of a density ramp which enhances resonance absorption during the 300 fs pulse.

Forslund, Kindel and Lee⁸⁾ have shown that a superthermal electron component of temperature (eV) $\approx 2 \times 10^{-3} (\Omega^2)^{0.33} T_c^{0.5}$ (where T_c is the background thermal temperature in eV) is generated by resonance absorption. For the experimental conditions presented here, we can expect a superthermal component of ~ 20 keV based on this scaling. The photodiodes measuring photon energies around 1 and 3 keV produced signal levels consistent with such a temperature. Superthermal electrons of this temperature can be expected to penetrate $\sim$ microns of plastic⁷⁾ and so modify thermal electron energy transport to enhance heat penetration. However, classical flux-limited thermal conductivity as modelled by the Medusa code⁵⁾ describes the observed axial heat penetration ($\approx 0.4 \mu\text{m}$) adequately.

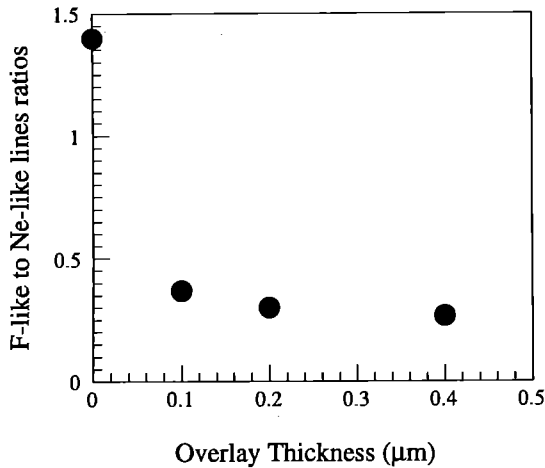


Figure 2 The ratio of the intensity of an FeXVIII (F-like) line (1.526 nm) and an FeXVII (Ne-like) line (1.2264 nm) as a function of depth of Fe signature layer.

Lateral transport of energy was small for these 300 fs pulse-length experiments (figure 1). Using the scaling for the distance Δr between the absorption and ablation front previously obtained²⁾, the lateral transport observed here follows the pattern found by Al-Hadithi et al²⁾ for $W/\Delta r$ as a function of W_{emission}/W (see figure 3), where W is the laser focal spot diameter and W_{emission} is the diameter of X-ray emission.

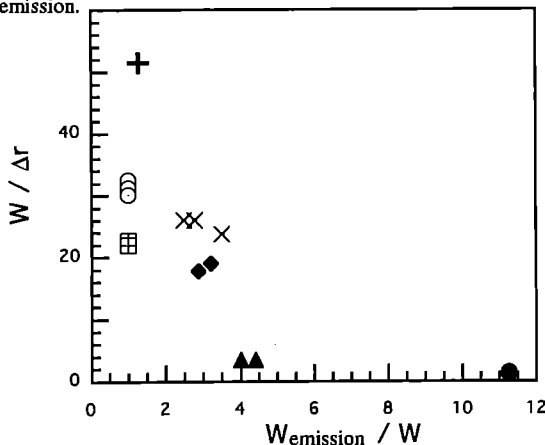


Figure 3 The focused laser diameter W divided by an estimate for the critical density to ablation surface distance Δr as a function of the width of X-ray emission W_{emission} divided by the focused laser diameter W . Results from previous experiments are shown (see Al-Hadithi et al²⁾) together with the data point for this experiment (+).

CONCLUSION

The transport of energy from the target surface focal region for high irradiance ($\sim 10^{18} \text{ Wcm}^{-2}$), high contrast ($\sim 10^{10}$), short pulse (300 fs) krypton fluoride laser pulses focused onto solid targets has been studied. Diagnostic buried layers of iron in the otherwise plastic (CH) targets have been used to deduce axial heat penetration by recording the characteristic Fe spectra for varying overlay thicknesses, while a pinhole camera/penumbral image technique has been used to search for lateral transport of heat. Classical flux-limited thermal conductivity describes the observed axial heat penetration ($\approx 0.4 \mu\text{m}$) adequately. The small amount of lateral heat transport is in good agreement with a previously obtained scaling.

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FEASIBILITY STUDIES OF OPTICALLY-IONISED RECOMBINATION XUV LASER SCHEMES ON THE SPRITE KRF CPA LASER SYSTEM.

S.G.Preston¹, W.J.Blyth¹, K.M.W. Zepf¹, J.Zhang³, A.E.Dangor², M.H.Key^{3,1}, A.Modena², Z.Najmudin², A.A.Offenberger⁴, J.S.Wark¹.

¹Clarendon Laboratory, Department of Physics, University of Oxford, Parks Road, Oxford OX1 3PU, UK,

²Department of Physics, Blackett Laboratory, Imperial College, London, UK,

³Rutherford Appleton Laboratory, Chilton, Didcot, Oxon, OX11 0QX, UK,

⁴University of Alberta, Edmonton, Alberta, T6G 2G7, Canada.

INTRODUCTION.

Much worldwide research interest has been concentrated on producing efficient XUV laser schemes. Most systems to date have required high pump energies supplied by a large laser system. The attraction of being able to generate coherent XUV sources using much lower pump energies has stimulated much theoretical research into the possibility of producing an optically-ionised XUV laser⁽¹⁻⁵⁾. Such a system uses of a short pulse (~100 fs) pump laser which attains a very high peak power with very little energy. This introduces the possibility of a table-top XUV laser system.

The laser is focused into a gas target, typically provided by a pulsed gas jet. At focus, the electric field of the laser becomes greater than the Coulomb potential binding the electrons to their atoms. These electrons can be liberated from the atomic potential either by quantum mechanical tunnelling through a finite potential barrier, or by classical tunnelling over the suppressed barrier. Figure 1 shows this schematically. With a high enough laser intensity, we can produce a plasma with a very cold electron temperature, and an extremely low ionic ground state population. The low electron temperature results in rapid collisional recombination and the electrons cascade down towards the ground state. If this cascade is fast enough, a population inversion is created between the ground state and an excited state and laser action occurs. This lasing to the ground state is desirable as it provides us with a high quantum efficiency for the laser, and produces a very short (self-terminating) XUV laser pulse.

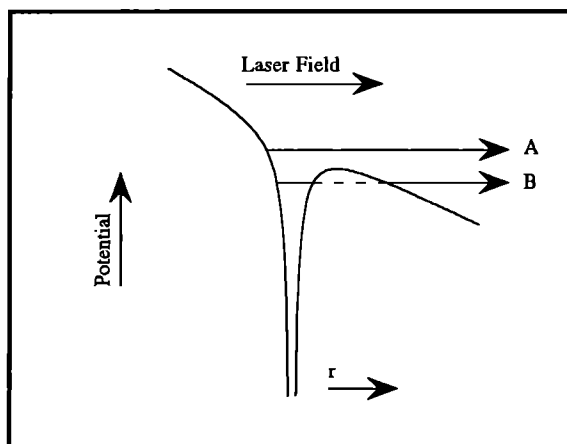


Figure 1. Schematic diagram of the process of optical ionisation of an atom. A depicts classical tunnelling and B depicts quantum mechanical tunnelling. This is a snapshot in time of the superposition of the laser potential and the Coulomb potential.

Most systems considered have been based on a Lithium-like scheme, where an atom is stripped to a Helium-like ion and recombination produces a population inversion between the 3d and 2p levels. Candidates for such systems include Ne VIII at 98 Å and N V at 248 Å. Details of these two schemes are summarised in table 1 below.

These systems are dependent on producing a very low electron temperature. It may be that it is not possible to attain such a cold plasma at the intensities required for the schemes to work. It has also been suggested that when heating by three body recombination is included in the simulations the gain is severely reduced. We report on experimental studies that we have performed to determine whether such low electron temperatures are produced using such a system and to determine whether gains in such systems are as high as simulations suggest.

Scheme	Ne VII	N V
Wavelength (Å)	98	248
Pump intensity (W/cm ²)	3×10^{17}	2×10^{16}
Electron density (cm ⁻³)	5×10^{20}	2.5×10^{19}
Electron Temperature (eV)	40	10
Gain (cm ⁻¹)	190	30

Table 1. Summary of two proposed optically-ionised laser schemes. The details come from simulations performed by Eder et al.^(2,3)

TEMPERATURE MEASUREMENTS.

The experiments were performed on the Sprite KrF laser system in chirped pulse amplification mode (CPA) producing 200mJ in 350fs at 248nm. Using 33 cm, 100 cm and 200 cm off-axis parabolas (OAPs), we produce focused intensities of up to 6×10^{17} W/cm², 10^{17} W/cm², and 3×10^{16} W/cm² respectively. Thomson scattering and stimulated Raman scattering (SRS) are used as diagnostics to determine the electron density and temperature during the laser pulse. These diagnostics have been described in previous reports⁽⁶⁻⁸⁾.

Study of the variation of electron temperature with density in both Helium and Neon for an intensity of 6×10^{17} W/cm² (shown in figure 2) indicates a rapid increase of temperature with density, moving from less than 10 eV at 10^{18} cm⁻³ to over 1000 eV at 10^{21} cm⁻³. The solid lines in the figure are Medusa simulations of the electron temperature due to above threshold ionisation and inverse bremsstrahlung. The Helium temperatures are consistently above the simulated values. This we attribute to the onset of SRS.

It is obvious that the 350 fs Sprite laser is unsuited to producing the conditions required for XUV gain in Neon, but a consideration of the variation of SRS heating with laser intensity (figure 3) shows the rapid reduction of heating with intensity. In the figure, 500 mJ corresponds to 6×10^{17} W/cm². We see that a reduction by a factor of two in intensity takes the level of SRS heating to a negligible level. Scaling the temperatures in figure 1 to intensities of 3×10^{16} W/cm² and electron densities of 2.5×10^{19} cm⁻³ shows us that conditions should be ideal for the Nitrogen laser scheme. Hence, a study of the XUV emission of N V should reveal whether the system exhibits the high gain predicted by Eder et al. or the lower gains.

The full implications of these electron temperature measurements are discussed elsewhere⁽⁹⁾.

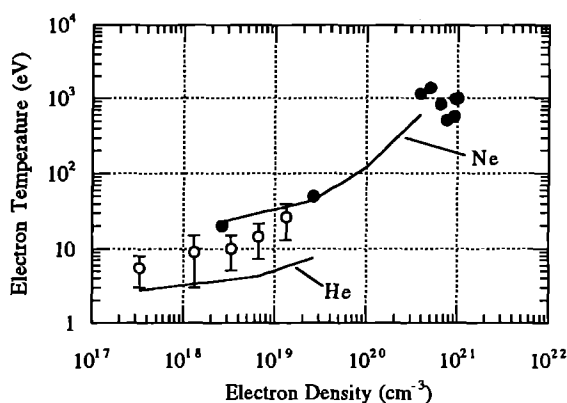


Figure 2. Electron temperature variation with density for an intensity of $6 \times 10^{17} \text{ W/cm}^2$. The solid circles are Neon, and the unfilled circles are Helium. Medusa simulations for the two gases are shown as solid lines.

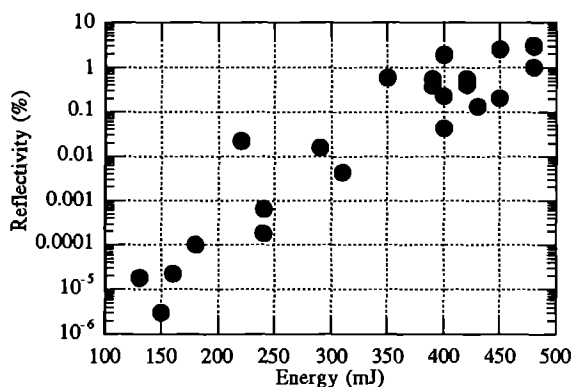


Figure 3 Growth rate of SRS backscatter reflectivity with laser energy from a 35 bar neon gas jet $100\mu\text{m}$ below the laser focus. Note that the laser energies quoted are energies before the compressor gratings.

GAIN MEASUREMENTS.

The emission from N V, optically ionised by the Sprite CPA laser focused with the 200 cm OAP, was focused using two cylindrical gold-coated mirrors onto a Hitachi variable spaced 1200 g/mm grating. The focused and spectrally resolved output from the grating was collected onto a double microchannel plate detector fibre-optically coupled to a CCD data acquisition system. A boron filter was used to eliminate laser light scattered from the grating zero order. Variables in the experiment were the gas length (0.5, 10, 20 mm), the gas density (gas jet backing pressures from 0.3 to 8 bar) and the laser intensity (provided by fluctuations in the output from Sprite, and by changing from a 200 cm to a 100 cm OAP, 10^{16} to 10^{17} W/cm^2).

Had the gain been as high as predicted by Eder et al. we should have seen a gain-length product of 8 for the 2 mm jet, and an enhancement of the 3d-2p line of almost 100 when compared to the 0.5 mm jet. No such enhancement was seen. This leads us to the conclusion that gains in these systems are much lower than previously predicted by theory and that heating by three body recombination is an important factor in limiting the maximum gain achievable in optically-ionised recombination XUV laser systems.

ACKNOWLEDGEMENTS.

We would like to express our thanks to the Sprite laser staff and the CLF target area support staff for their help in preparing and performing these experiments.

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HIGH CONTRAST MEASUREMENT OF STARTUP IMPRINTING AND INDUCED RAYLEIGH TAYLOR GROWTH

R J Taylor¹, J P Dahlburg², D E Fyfe², J H Gardner², A Iwase¹, O Willi¹

¹ Imperial College, London, SW7 2BZ, UK

² Naval Research Laboratory, Washington DC, USA

INTRODUCTION

Laser irradiation can accelerate a thin foil target through ablation of a hot plasma from the target surface, and has received considerable attention as a possible driver for Inertial Confinement Fusion (ICF). A perfectly smooth irradiation at high intensity has proven very difficult to obtain, however, and experimental [1] and computational [2] studies have demonstrated that the most sophisticated smoothing techniques available are not sufficient to eliminate the imprinting of the laser non-uniformity during the early phases of illumination. So-called "start-up imprinting" is thought to occur when the laser is incident on the initially cold target surface, before sufficient separation has developed between critical density and the ablation surface (D_{ac}). Once D_{ac} is established, electron thermal conduction helps to smooth the drive non-uniformities [3].

Since a high acceleration is associated with an ICF implosion, and the ablation surface is unstable to the Rayleigh-Taylor (RT) instability [4], even very small perturbations induced in the early phase can grow rapidly, leading to shell breakup and an ineffective implosion. To design a successful direct-drive implosion pellet, it is necessary that the sophisticated hydrocodes already in existence [5] for this purpose are benchmarked against detailed experimental data on the imprint phenomenon. Obtaining such data requires a diagnostic technique which is sensitive to small areal density perturbations on small spatial scales, such as are associated with the speckle pattern present in the focal spot of a typical high power laser. Conventional probing techniques are hampered by a combination of low spatial resolution ($\sim 10\mu\text{m}$) and low contrast, making them unsuitable for the study of very small perturbations. We present here the first demonstration of a technique which at once possesses the resolution and the contrast requisite for a study of startup imprinting.

XUV TRANSMISSION RADIOGRAPHY

XUV radiation (in this case, $h\nu=248\text{eV}$) possesses characteristics which make it ideal for the study outlined above. Multi-layer coatings are available with reflectivity of a few percent at this photon energy, hence allowing the use of reflective optical components for image formation. This at once has the advantage that a large entrance pupil to the imaging system can be used, hence increasing the x-ray flux relayed to the detector, and also provides us with the ability to generate high-magnification images at very high spatial resolution through the use of high-quality optical substrates, coated with the multilayer stack.

Figure 1 is a schematic of the layout used for the experiment reported here, where a single, spherical multilayer mirror was used to image the plane of the target foil onto a gated XUV camera with a magnification

of 70. The limiting resolution in the system was that of the detector, as demonstrated in shots where the target was replaced with a finely spaced copper mesh, leading to an overall resolution of $\sim 1\mu\text{m}$.

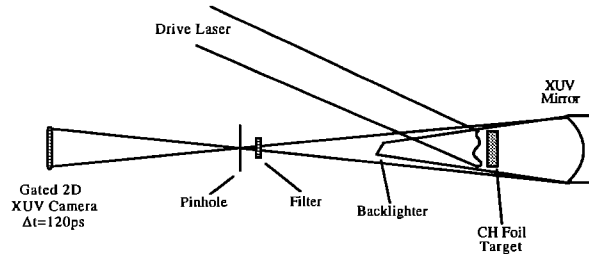


Figure 1 Experimental layout for face-on XUV radiography

The multi-layer coating is designed such that significant reflectivity is produced only in a narrow band ($\pm 5\%$) around the central photon energy of 248eV , with higher order reflections rejected due to the absorption and reflection coefficients of the layer materials (Platinum and Carbon). Some residual reflectivity is retained in the optical band, however, so a $1000\text{\AA}$ Aluminium foil is used to optically isolate the detector enclosure from the target chamber itself.

The XUV backlighting is provided by a separate high-Z (in this case Gold) plasma, separated from the target by a sufficient distance such that the level of radiation incident on the target itself is negligible with respect to the hydrodynamic evolution of the system. The O-band radiation of gold, centred around $\sim 300\text{eV}$, is spectrally filtered by the multi-layer mirror to provide a monochromatic image at 248eV .

The experiment reported here was designed to study the early phase of laser imprinting by using low irradiance levels ($\sim 5.10^{12}\text{W/cm}^2$) as would typically be used in the low-intensity foot of a direct-drive ICF pulse. CH target foils were used to represent the low-Z outer layer of a direct-drive ICF capsule. Study of hydrocode predictions and analytical models suggest that such irradiance conditions will generate a target compression of $\sim 3\times$ solid density, and minimal shock heating. The accelerated, shock compressed target region is expected to remain below 5eV in temperature, whereas the ablated plasma is expected to be heated to several hundred eV at a density of less than 0.002 g/cc . The intermediate region will be small due to the localised deposition of laser energy around critical density and the consequently steep density gradient up to the ablation surface. The ablated plasma is expected to be virtually transparent to the probe radiation, predominantly due to the low material density and high temperature, whereas the opacity of the compressed target lies around $2700\text{cm}^2/\text{g}$, with the exact level dependent on the prevailing plasma conditions. Opacities predicted by coupling the IMP [6] opacity code to hydrocode simulations suggest that the opacity varies by up to 10% through the accelerated portion of

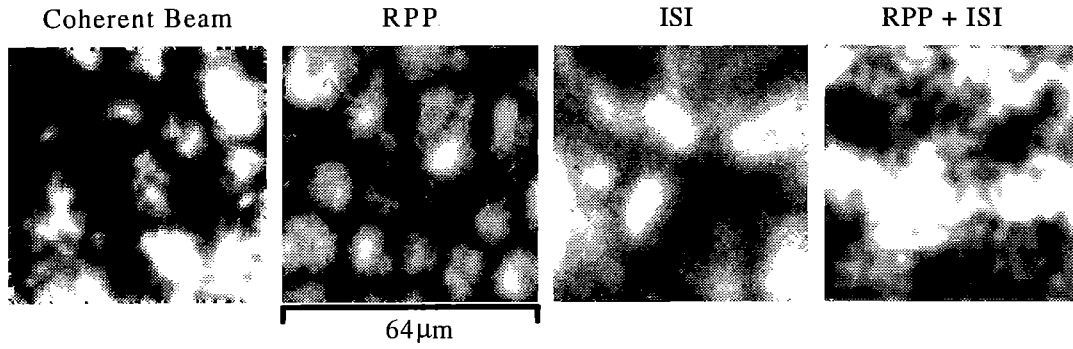


Figure 2 XUV Radiographs of CH foils driven by optically smoothed and unsmoothed laser irradiation.

the target, and so to gain detailed information as to the areal density profile in the target we must compare the measured optical depth with that predicted by various opacity codes coupled to hydrocodes.

To a first approximation, however, the sensitivity of the XUV probe can be estimated by assuming a constant target opacity at $K_{\text{cold}} = 2763 \text{ cm}^2/\text{g}$ (from Henke's tabulated data) and then evaluating the areal density perturbation which will generate the minimum detectable signal fluctuation. The XUV framing camera is capable of recording a ripple amplitude of $\sim 2.5\%$ without the signal being lost in noise, and hence through Equation 1 the minimum detectable areal density ripple amplitude ($\Delta \rho_{\text{min}}$) is $0.09 \mu\text{m g/cm}^3$.

$$\Delta \rho_{\text{min}} \approx \ln((1 + \Delta I_{\text{min}})/1) / K_{\text{cold}} \quad (1)$$

XUV RADIOGRAPHS OF STARTUP IMPRINTING

The technique detailed above has been implemented to obtain XUV radiographs of plastic foils ($3\text{--}8\mu\text{m}$ thick CH) irradiated with the VULCAN laser ($\lambda = 0.527\mu\text{m}$) at irradiances around 5.10^{12} W/cm^2 . Images were recorded both during the shock transit and at later times once the imprinted seed had grown by the RT instability. The coherent beam and beams smoothed with Random Phase Plate (RPP), Induced Spatial Incoherence (ISI) and hybrid RPP+ISI techniques were used. Figure 2 shows typical radiographs for each of these irradiation conditions, and it can be seen that none of these techniques were capable of generating a uniform acceleration, even for the modest conditions prevailing here. Similar shots using targets coated with $1000\text{\AA}$ Aluminium produced similar results, thus confirming that the imprinting process is a surface effect, and not strongly influenced by possible disruption deep inside a target, such as may be generated by the "shining-through" of the low-intensity leading edge of the laser pulse.

In contrast, however, the novel FDD [7] smoothing scheme was found to generate radiographs which were smooth to within the sensitivity of the XUV probe (see Figure 3), highlighting the promise FDD holds for direct drive ICF. The residual fractional ripple in the transmitted signal is within the noise:signal ratio of the camera. FDD smoothing is achieved by placing a layer of low-density foam before the target, overcoated with a high-Z element to provide a flash of soft x-rays at early time. This supersonically pre-forms a plasma from the foam [8], providing rapid thermal smoothing of the subsequent drive and enhanced dispersion of the initial non-uniform shock wave whilst D_{ac} is established, thus demonstrably suppressing the laser imprint. The x-

ray flash from the high-Z overcoat will inevitably heat the region around the ablation surface of the FDD package to a greater extent than for a plain foil, hence potentially reducing the sensitivity of the XUV probe when used with an FDD target. This heating is thought to be small, however, and rapidly swamped by PdV compression and shock heating as steady-state is approached. In support of this, target disruption with FDD targets was never observed, even after the prolonged (3ns) acceleration that was sufficient to destroy foils driven with optically smoothed beams.

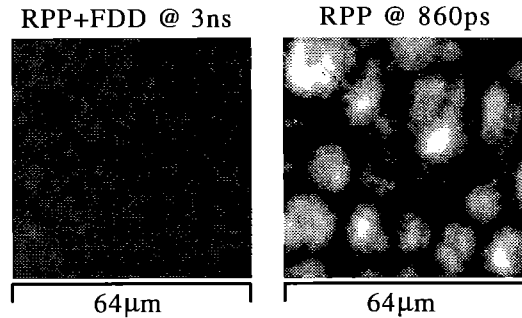


Figure 3 Comparison of FDD and non-FDD drive

CONCLUSIONS

Detailed analysis of the imprinting data is underway, involving comparison with sophisticated, multi-dimensional hydrocodes. The extent to which conventional hydrocode physics can reproduce the data acquired will provide a valuable insight into the physical processes involved, and benchmarking these codes on actual data such as that presented here will bring confidence in our ability to successfully design a high-gain direct-drive ICF pellet.

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DIAGNOSIS OF LASER DRIVEN SHOCKS IN FOAM USING ABSORPTION SPECTROSCOPY

D. Hoarty, A. Iwase, C. Meyer and O. Willi
Blackett Laboratory, Imperial College, London

INTRODUCTION

One of the major problems in inertial confinement fusion studies is the non uniformities in the laser beams used to irradiate the fusion capsule. These non uniformities imprint themselves onto the ablation surface of the capsule seeding instability growth which acts to impair symmetrical implosion.

A scheme proposed by the Imperial College plasma group is to isolate laser imprinting from the ablation surface by using a foam buffer around the capsule shell. This acts to smooth out the laser imprint non-uniformities without impairing the hydrodynamic efficiency of the implosion^{1,2}. A number of experiments have been carried out by the group investigating the physics of laser and X-ray interaction with foam targets. Studies so far have included the propagation of supersonic radiation waves in foam³, Rayleigh Taylor instabilities and recently shock and radiation waves produced by direct laser heating and collision with flyer plates⁴. These experiments have used imaging in the XUV using a spherical multilayer mirror coupled with a streak camera or gated micro-channel plate. The measurements reported here have applied the technique of point projection absorption spectroscopy to foam targets directly irradiated with the VULCAN laser. The technique allows measurement of the shock position and the distribution of ions in the shock from which the electron temperature in the shocked region can be inferred.

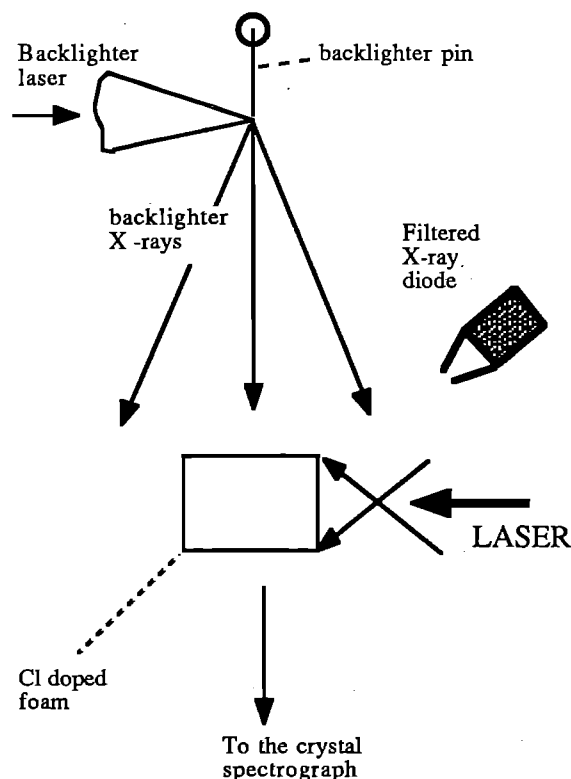
The targets used were doped with chlorine which when ionized is detectable by its characteristic absorption lines. The range of frequencies covered by the spectrometer was chosen to cover the range of K-shell transitions of chlorine ions. By comparing the ratio of transmission in the ion stages present in the shock region to a model the ionization can be used to infer the ionization temperature in the shock.

EXPERIMENTAL SETUP

Figure 1 shows a schematic diagram of the experimental setup.

Point projection spectroscopy^{5,6} involves the use of a pin backlighter irradiated at one end by a short pulse of laser light to produce a flash of X-rays. By using a Bragg crystal spectrometer a dispersed radiograph of the foam giving both spatial and spectral information is obtained. In this experiment the backlighter used $1\mu\text{m}$ bismuth coated onto gold pins. A TiAP crystal was used in the spectrometer at 11° to cover the range of 2.0-3.4keV.

FIG. 1 EXPERIMENTAL SETUP



The foam targets were of three types. In the first type the foam was coated with a thin layer of aluminium. This was coated onto the end of the foam irradiated by the laser and was varied between 0.1 and $0.03\mu\text{m}$ from target to target. The second type had a nominal $0.02\mu\text{m}$ gold coating on the end. In the third type the foam had no coating. The foams were approximately $250\mu\text{m}$ long.

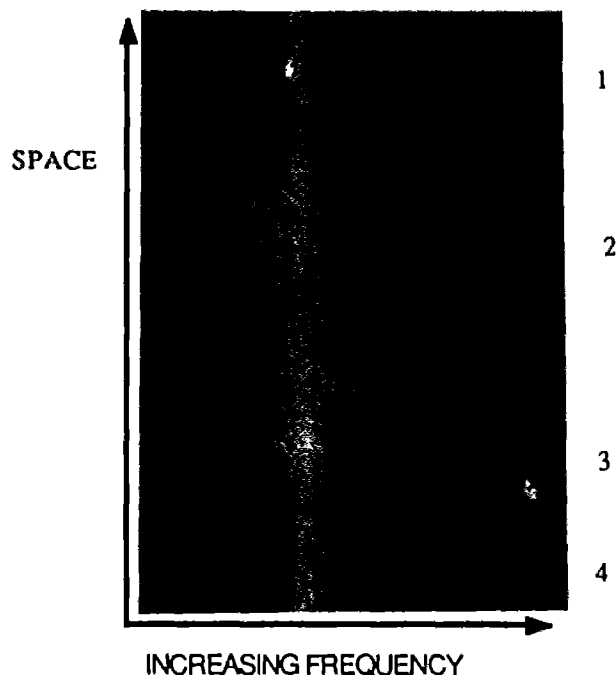
The target chamber was used in the cluster configuration. The pulse length was varied during the experimental run from 750ps to 2ns. The backlighter beam had a pulse length of 100ps for the shots presented here and was focussed with an f2.5 lens. The backlighter delay was varied from shot to shot between 350ps and 1300ps.

On some shots two shocks were launched into the foam by delaying the heating beams to study the behaviour of coalescing shocks in the foam. The coated foam shots are at a preliminary stage of analysis, the results reported here are from type three targets, ie uncoated foam with two of the six cluster beam used to irradiate the foam.

RESULTS

A radiograph of an uncoated foam target is shown in figure 2. The radiograph is divided into four areas. First the backlighter which has not passed through the foam, second the undisturbed foam, third the shocked region showing the absorption features characteristic of ionized chlorine and fourth the rarefraction region where there is material ablated by the laser beam. The delay of the backlighter on this shot was 350ps.

FIGURE 2



The radiograph is image processed to remove fog and emission from the foam image. Lineouts taken through the absorption region are then divided by unattenuated backlighter emission to give transmission. Figure 3 shows the absorption in the shocked region plotted against frequency in keV at 350ps delay. The 1s-2p and 1s-3p absorption lines in various ion stages can be clearly seen. The subtraction of bright chlorine emission from the laser corona limits the accuracy of 1s-3p absorption measurement but the 1s-2p lines are not affected. The position of the shock front relative to the end of the foam is also measured. Figure 4 shows the absorption at 850ps delay. Again the subtraction of bright chlorine emission has meant a loss of data in the region between the 1s-2p absorption features and the first strongly absorbing 1s-3p feature. However, as can be seen from the preliminary fits the data are sufficient to allow the ionization temperature to be inferred. The data have been compared to the LTE opacity code IMP written by Dr. S Rose⁷.

Fig. 3 Absorption in the shocked foam at 350ps delay

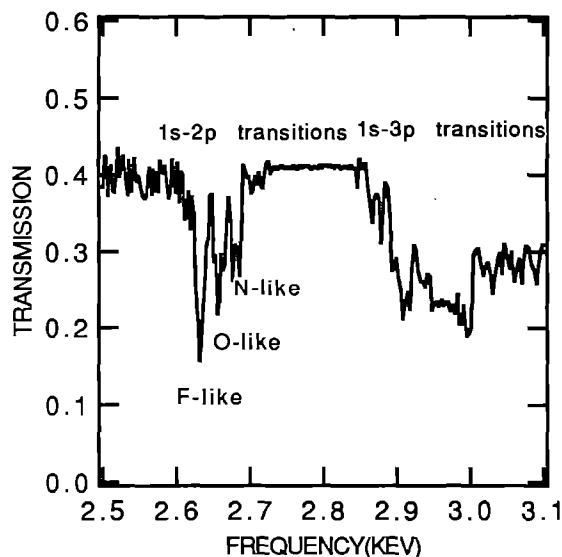


Fig. 4 Absorption in the middle of the shocked region 850ps.

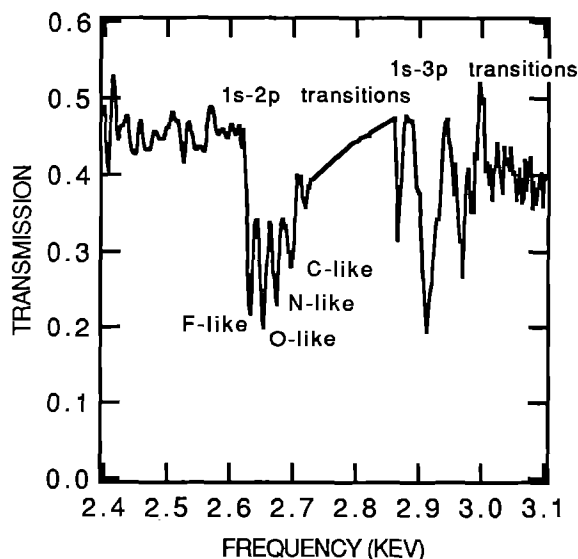


Figure 5 and 6 show the fits to the data shown in figure 3 and 4 respectively. Although the model calculation differs slightly from the experimental data the temperature can still be inferred. The data shows evidence of a slight temperature gradient at both 350ps and 850ps delays. At a later date a detailed MCDF data base will be constructed for chlorine to model the details of the spectrum in order to ascertain how much of the discrepancy between experiment and fit is due to detailed structure and the extent of the gradients.

Fig. 5 IMP calculation 55eV,0.24g/cc

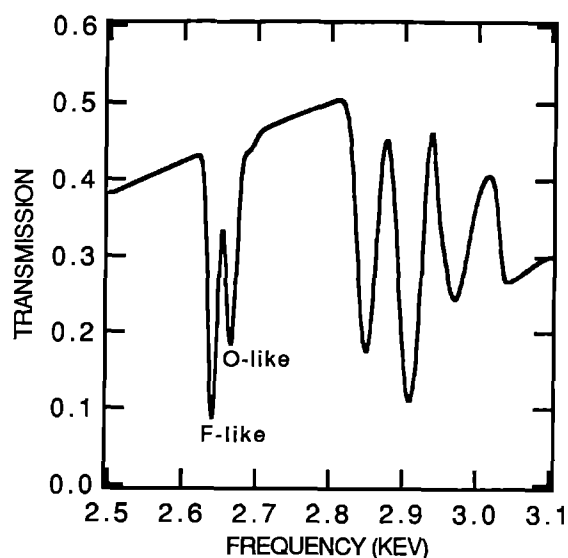
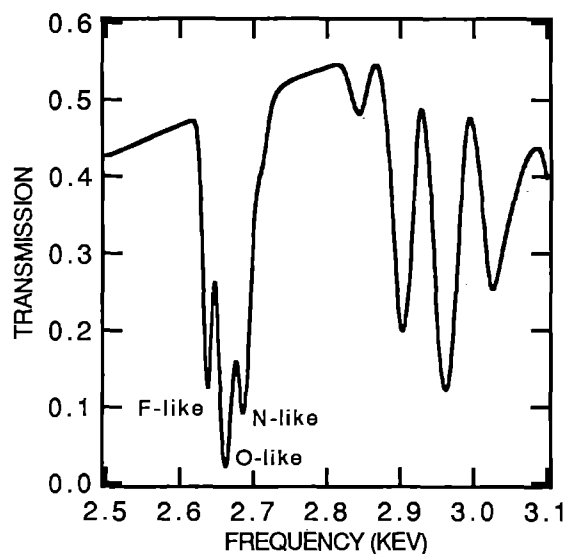


Fig. 6 IMP calculation 71eV,0.25g/cc



The density input to the IMP model was obtained by modelling using a 1D Lagrangian radiation hydrodynamics code.

CONCLUSION

The technique of absorption spectroscopy has been successfully applied to chlorine doped foam targets. Shock widths, positions and ionization stages within the shock have been measured allowing electron temperatures and shock velocities to be inferred. The absorption spectra show an ionization gradient through the shocked region. We have detailed quantitative

measurements of the propagation of laser driven shocks in foam targets. Together with the results of Iwase the data represents an exacting test of radiation hydrodynamics code modelling. The technique could be extended to further experiments on shock coalescence, foil impact and radiation transport with gated detectors to reduce emission from the laser corona.

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EXPERIMENTAL OBSERVATION OF SHOCK WAVE PROPAGATION THROUGH A LOW DENSITY FOAM TARGET INDUCED EITHER BY LASER ABLATION OR FOIL IMPACT

A. Iwase, C. Meyer, R. Taylor and O. Willi

The Blackett Laboratory, Imperial College of Science, Technology and Medicine, London, SW7 2BZ, U.K.

1. INTRODUCTION

Shock waves have been studied in an number of areas of physics for many years. In astrophysics, shocks occur through the interaction of interstellar clouds. In laser driven targets, shocks are launched by laser ablation of material. A shock is, by definition, a nonlinear process and its investigation is important not only from an application point of view but also from a fundamental physics point of view. In laser-produced plasma, the study of shock waves has almost entirely been limited to observations of shock break out at the rear of the target. Recently, a radiographic technique has been applied¹ to the temporal observation of the shock transit through the target driven by x-ray ablation. As a hard x-ray of 5 - 7 keV probe was used to diagnose the high density target only the shock front could be observed. However the use of low density foam targets allows the shock propagation to be studied in detail.

An experiment has been carried out to observe the temporal behaviour of shock wave propagation through a low density foam target using a soft x-ray imaging technique operating at 50Å² or point projection absorption spectroscopy.³ A shock wave was launched in the low density foam either by laser ablation of a high density layer (Au or Al) which was overcoated onto the foam or by the impact of a laser driven foil flyer.

2. EXPERIMENTAL ARRANGEMENT

The experiment was conducted on the VULCAN Nd: glass laser system. Figure 1 shows a schematic of the experimental setup. Six frequency-doubled laser beams were arranged in a cluster configuration. The laser beams were focused onto target with f/10 lenses and the pulse duration was between 400 and 700 ps. The foam target was imaged side-on by a spherical multilayered mirror onto a slit of a low magnification streak camera. The slit was positioned along the axis of the foam target. A gold disk, situated 4-5 mm from the foam target, was irradiated by a separate laser beam to produce a 3ns backlighter source. The mirror acted as a narrow band-pass filter in the XUV region, reflecting at a wavelength of 50Å ± 10%. Overall magnification achieved with this system was ~ 70x. The temporal and spatial resolutions were found to be about 60 ps and 1µm respectively. A 1000Å thick aluminium filter was used to block the optical light from reaching the streak camera. Direct x-rays were blocked by a thick metal disk. An S-20 optical streak camera was used to monitor the pulse shape and the pulse length.

Two series of experiments were carried out. In the first experiment, the shock was induced by the ablation pressure produced by irradiating the foam target, while in the second measurement, the shock was launched by the impact of a laser driven thin foil target with the stationary foam. In both cases, the shock was preceded by a supersonic ionisation wave driven by x-ray radiation which was emitted from the thin overcoated gold or aluminium layer.

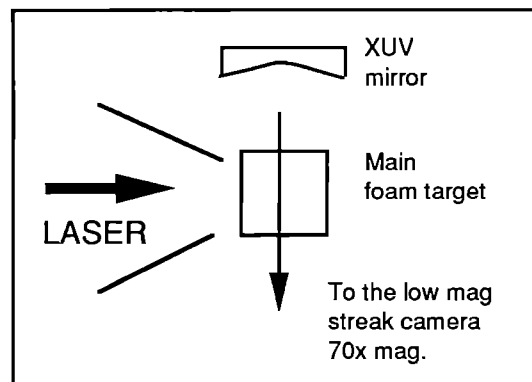


Figure-1 A schematic of the experimental setup. The slit of the streak camera was placed along the axis of the foam target to observe the temporal evolution of the shock.

A. SHOCK INDUCED BY LASER ABLATION

The target was a 50 mg/cc, 200 µm in diameter, 180 µm long triacrylate (C₁₅H₂₀O₆) cylindrical foam overcoated with 1000Å of aluminium or 200Å of gold on the side facing the laser light. Two laser beams were focused onto the coated side of the target. Figure 2 shows a streak record of a radiographic image of a target which was irradiated with a 470ps pulse at a laser intensity of 2x10¹⁴ W/cm². The laser is incident from left hand side of the picture. Several interesting features can be seen. Feature A) Supersonic ionisation wave induced by the soft x-ray radiation of the laser irradiated aluminium coating, B) shock front and C) second ionisation wave produced by radiation from the shocked region.

The supersonic ionisation wave (feature A) is seen to travel initially at a velocity of 3x10⁷ cm/s preionising the material before the shock front (feature B). The ionisation wave originates from the aluminium coating and slows down suddenly which is due to the burnthrough of the coating. The ionisation wave increased the transmission of backlighting x-ray prior to shock wave (B) which, in turn, reduces the transmission due to higher density. At ~ 1700ps the shock breaks out at the rear of the foam with an average velocity of 1x10⁷ cm/s.

B. FOIL IMPACT MEASUREMENTS

A foam with no coating on the front side was impacted by a 3 µm thick aluminium foil target which was separated from the foam by about 110 µm. Four ISI laser beams irradiated the foil for 500 ps FWHM at an irradiance of 2x10¹⁴ W/cm². This target geometry decouples the shock behaviour from laser ablation. The experimental setup was similar to the one shown in Figure 1. A total magnification of 50 x was used.

Figure 3 shows a streak record of an absorption radiograph. The laser irradiated foil impacted with the foam after 950 ps from the beginning

of the pulse with a velocity of 1.2×10^7 cm/s. The average shock velocity is 1.5×10^7 cm/s.

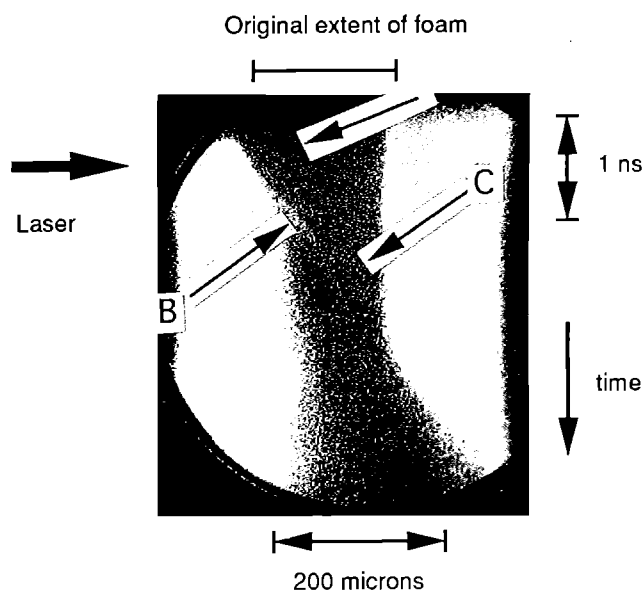


FIG 2. A time resolved radiographic image of a cylindrical foam, 50 mg/cc and 180 micron long initially, overcoated with 1000 Å of aluminium. The approximate pulse shape is shown on the right hand side.

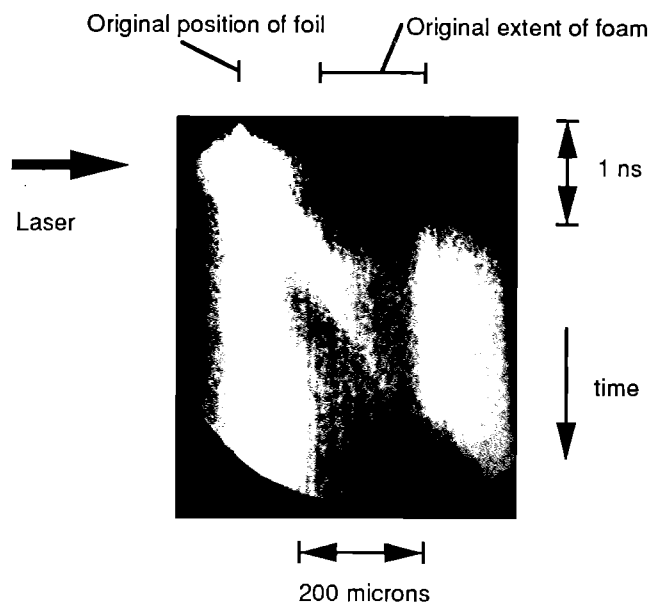


Figure 3. A time resolved radiographic image of an aluminium foil impacts onto a stationary foam.

SIMULATION AND DISCUSSION

Preliminary modelling was performed with 1D Lagrangian code MEDUSA⁴ for the shot shown in figure 2. No attempt was made to simulate the porous structure of the foam target.

Figure 4 shows a result of a MEDUSA run. The global shock behaviour was reproduced well although a lower laser irradiance had to be used in the simulation. This may be due to 2-dimensional effects in the experiment. Another reason for the lower laser irradiance in the simulation could be explained by the presence of the preionisation in front of the shock.

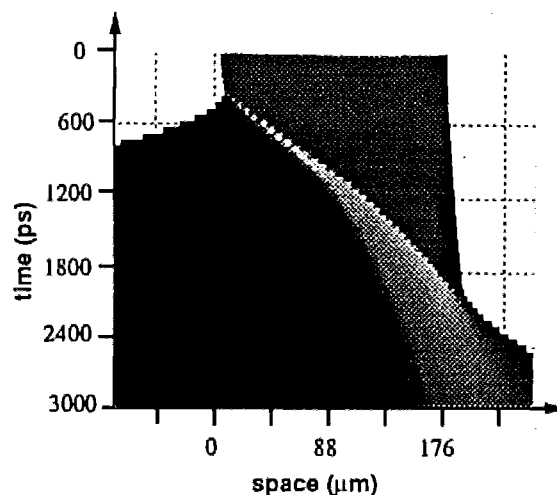


FIG 4. Temporal behaviour of the density as simulated by MEDUSA.

The first ionisation wave present in Figure 1 slows down after the laser burns through the aluminium coating. The MEDUSA simulation suggests that the laser burns through 1000 Å of aluminium after two hundred picoseconds with the laser conditions used, which seems to be consistent with the observation. This suggests that the extent of preionisation or preionised plasma buffer can be controlled by varying the thickness of the overcoat.

CONCLUSION

Detailed observations of shock propagation through low density foam targets are presented. Shocks are produced by laser ablation or foil impact. Shock waves were preceded by an ionisation wave originating from the laser irradiated metal overcoat. The ionisation wave is seen to be slowing down which corresponds to the burn through time of the metal overcoat. The experiment was modelled with the 1D MEDUSA hydrocode. Good overall agreement was obtained.

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UPGRADE OF TA4 TO PROVIDE INCREASED ENERGY FOR SOFT X-RAY CONTACT MICROSCOPY.

A.D. Stead¹, T.W. Ford,¹ C. Danson², D. Peplar², M. Ebbage²

¹ Dept. of Biology, Royal Holloway (University of London), Egham, Surrey, UK. TW20 0EX

² RAL, Chilton, Didcot, Oxon. UK. OX11 0QX

BACKGROUND

To image living biological material using soft X-ray contact microscopy it is necessary to use a very short pulse of soft X-rays because, during prolonged exposures radiation-induced structural changes may occur to the biological specimen. Indeed all estimates as to the likely doses needed to image specimens are equal to, or in excess of, the doses shown to cause gross structural changes. In experiments with *Chlorella* for example, exposure to 10^4 Gy caused significant radiation-induced structural damage³. Structural damage to Chinese hamster ovarian cells was detected at 10^5 Gy, but loss of membrane and enzyme function occurred at an order of magnitude lower¹¹. The contraction of rabbit muscle myofibrils after the addition of ATP was also prevented by 10^4 Gy^{1,9}. The ability of irradiated yeast cells to exclude the viability stain Evans Blue was greatly reduced (indicating a loss of membrane function) by similar doses (Fujisaki, pers. comm.). Significant mass loss was detected from wet *Vicia faba* chromosomes which were imaged successively with doses of 2×10^5 Gy¹⁴.

To avoid the problem of radiation damage we have chosen to use soft X-ray contact microscopy (SXCM) in which an X-ray absorption map of the specimen is recorded in a photosensitive resist. The advantage of this technique is that by using soft X-rays generated by a laser plasma source imaging times can be kept to a few nanoseconds so that neither radiation-induced damage nor natural movement can interfere with the imaging process⁷. It has been suggested that exposures of even a few nanoseconds may cause image blurring due to thermal expansion of the specimen, in fact at least one author has suggested that imaging times must be in the order of 30psec¹⁰. However, such blurring has not manifest itself in our previous studies even when the image resolution has equalled, or exceeded 50nm^{4,12}.

Although the exposure time must be short, the soft X-ray fluence must remain high in order that the development time needed to produce the relief map of the carbon distribution of the specimen in the photoresist is kept to a minimum. The consequences of prolonged chemical development are that small features in the resists, themselves corresponding to small structures in the original material, may be lost²⁷. Therefore, to ensure that both the exposure time and the subsequent photoresist development time are minimised it was decided to increase the laser energy reaching the target in order to increase the soft X-ray flux generated from the laser-produced plasma.

Three options were originally considered:

- introduction of a new dedicated amplifier into the beam path
- positioning a second beam such that two laser plasma sources could be employed
- reduction in the loss of energy between the rod chain output and target

METHODS

1. Physical measurements

The beam path into TA4 comprises some seven mirrors, a $\lambda/4$ waveplate, window and lens (Fig. 1); the laser energy was measured

both before and after each component and the % energy loss calculated for each component. For measurements of the focal spot size an equivalent plane monitor was set up.

2. Evaluation by imaging

Images of the unicellular green alga, *Chlamydomonas*, and of bacterial spores were taken as described previously⁶ and the photoresist developed in MIBK:IPA mixtures. The time needed for the imaged area to first be visible by incident light microscopy (Leitz Dialux) was recorded and, once the depth of development was considered satisfactory, the photoresist was examined by atomic force microscopy (Park Scientific Instruments or Burleigh Instruments). Development times and image quality were compared to those obtained prior to the upgrade.

RESULTS

Before embarking on a costly upgrade the various components of the beam path were initially tested. Compared to the output from the rod chain it was found that as little as 46.3% of the corner energy was reaching the target. When originally constructed a polariser was installed to protect the laser from back reflections, however, sufficient protection now exists within the laser area and so this plate was removed. Removal of the polariser increased the energy reaching the target to 52.2%

Calorimetry showed that most of the optical components transmitted approximately 90% of the laser energy, however two components were much worse (Table 1). The temporary replacement of these optics (mirrors 5 and 7) and the cleaning of the remaining optics increased the % laser energy arriving on target from 52.2 to 73.3%, thereby indicating the benefits of the installation of new optical components. Since this time the remaining optical components have also been replaced including the installation of an antireflection coated aspheric focussing lens. As a consequence of this the energy that can be delivered onto target in a 3nsec pulse now equals, or even exceeds, 16J. Measurements of the focal spot size showed that the FWHM was 118.2 μ m, or some 1.25X the diffraction limit (94.8 μ m). However, during these measurements it was discovered that the position of the focal spot for the CW:YLF differed from that of the rod chain shots by approximately 100 μ m and this has had to be corrected.

Given that the % conversion efficiency to soft X-rays increases as the energy on target increases this means that the flux of soft X-rays available for imaging should have increased very significantly. Future experiments will quantify this increase. However, imaging experiments, and the time needed for the development of the images, already indicate that there has been a very successful increase in soft X-ray fluence. The development of some of the images of the unicellular green algae, *Chlamydomonas*, shown in previous RAL reports⁶ and elsewhere^{3,8} required as long as 30mins development in a 50% mixture of MIBK and IPA. Images of the same organism taken since the replacement of the optical components in TA4 required, on average, 2mins to achieve a similar depth of development. Furthermore, the more recently obtained images have been taken with a specimen to target distance of 13mm (cf previous images taken at 8mm). Interestingly, in collaborative experiments, images of similar cells taken using the KrF laser, ASHURA, in Tsukuba, Japan required 2min

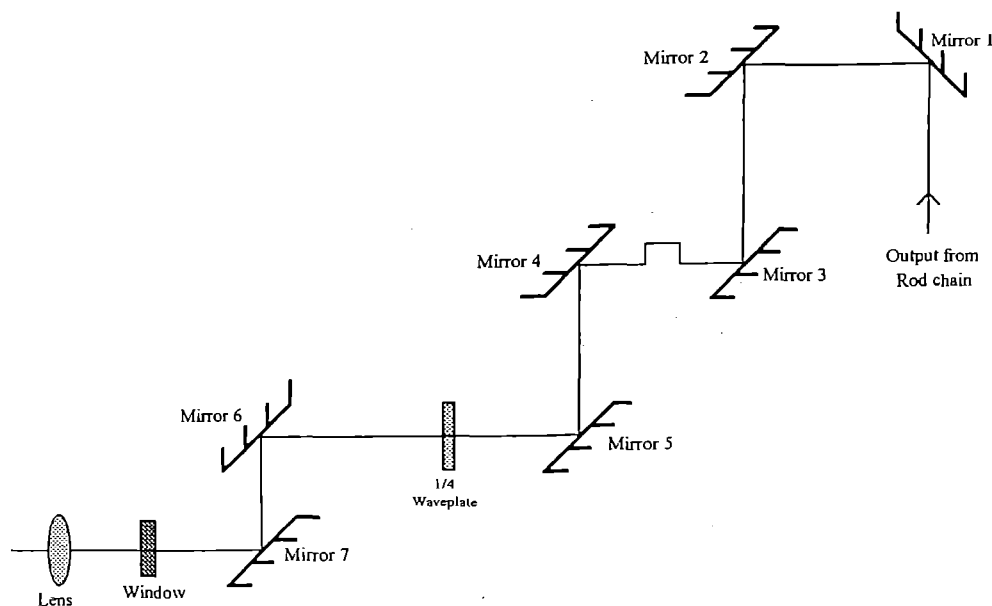


Figure 1 The configuration of the optical path into TA4

Position of measurement	Before optic mW	After optic mW	Optic loss %
Mirror 1	22.7	21.9	3.5
Mirror 2	21.9	21.5	1.8
Mirror 3	21.5	20.6	4.2
At TA4 shutter (air path)	20.6	19.1	7.3
Mirror 4	19.1	18.8	1.6
Mirror 5	18.8	15.8	16.0
$\lambda/4$ waveplate	15.8	14.6	7.6
Mirror 6	14.6	14.4	1.4
Mirror 7	14.4	13.3	7.6
Chamber window	13.3	13.2	0.8

Table 1 Quantification of the energy loss through the various optical components within TA4.

development in 50% MIBK when the laser energy was as low as 6J, however in these experiments the number of optical components was much lower and the target to specimen distance was only 7mm.

DISCUSSION

Although TA4 has been used successfully for the imaging of living biological specimens¹³ the development time of the exposed photoresists has often been excessive⁶. By increasing the available energy onto target this time has been significantly reduced and this should enable images with a higher resolution to be produced. At the present time it is clear that the development needed has been drastically reduced but it is impossible to ascertain if the resolution of the images has improved since images are now being recorded by atomic force microscopy which can, irrespective of other factors, produce images with a greater resolution than those previously produced using scanning electron microscopy. A further advantage of the increased soft X-ray flux has been that the distance between the specimen and the target has now been increased and this reduces

the problems of debris from the target and makes the placement of the specimen, relative to the target, a much easier operation.

ACKNOWLEDGMENTS

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STEREO IMAGING OF BIOLOGICAL SPECIMENS WITH SOFT X-RAYS

A.D. Stead¹, R. Bagby², R.A. Colton¹, D. Neely³, E. Miura⁴, A.M. Page¹, T.W. Ford¹

¹ Biology Department, Royal Holloway (University of London), Egham, Surrey, U.K. TW20 0EX

² Department of Zoology, University of Tennessee, Knoxville, Tennessee, TN 37996-0810. U.S.A

³ Rutherford Appleton Laboratory, Chilton, Didcot, Oxon. U.K. OX11 0QX

⁴ Electrotechnical Laboratory, 1-1-4 Umezono, Tsukuba, Ibaraki 305, Japan

INTRODUCTION

The attraction of imaging biological specimens with soft X-rays in the so-called "water window" (2.3 - 4.4nm) is that in this region the absorption of X-rays by oxygen is low compared to absorption by carbon⁶. It is therefore possible to discriminate between structural features of the cell which have a high carbon content and the cytoplasm which is relatively carbon-dilute. The technique requires that a high-brightness X-ray source and suitable detector be available and these requirements were not available when the prospects of X-ray microscopy were first discussed¹.

Throughout the development of soft X-ray microscopy the major goal, for the biologist at least, has been the ability to image living material without the need for either chemical fixation or tissue freezing, both of which can introduce artifacts¹⁷. However, unless the images are made with either a very low dose of X-rays, or with exposure to the X-rays being very short, then damage due to radiation or thermal expansion will occur and alter the specimen before it can be recorded^{1,7,9}. Synchrotron sources have advantages, particularly the ability to alter wavelength², but the flux of their soft X-rays is low enough that exposure times are long, and both radiation-induced^{9,11,20} and natural movement of the specimen will blur the image. To date the only sources that can produce sufficient soft X-ray fluence for the single-shot, short-duration exposures required to image living specimens are the pulse plasma sources (Z-pinch and laser-produced). Such images have been produced using soft X-ray contact microscopy^{6,18,19} in which the specimen is placed in close contact with a photosensitive resist - usually polymethylmethacrylate (PMMA). Exposure to X-rays results in bond breakage in the PMMA molecule¹³ and, after chemical development, the image in the resist can be viewed by scanning electron microscopy or atomic force microscopy (AFM). The topography of the image corresponds to the X-ray absorption by the specimen which in turn is a reflection of the original specimen carbon density.

Although laser-pulsed SXCM (soft X-ray contact microscopy) is able to give images of living biological specimens with resolution superior to that obtained with light microscopy, the resolution of structures deep within the specimen is compromised by the recorded image being an absorption map of everything in line with the path of X-rays. Superimposition of separate, spherical structures in different XY planes can lead to the impression of a larger, multi-lobed structure as imaged at the plane of the photoresist. The thicker the specimen, the more superimposition becomes a problem. Since living cells are usually greater than 1µm thick, superimposition is a problem which must be addressed if living cells are to be imaged with greater clarity. One way to solve the problem in light microscopy is to make "optical sections", assigning Z-coordinates to the separate XY planes, as is now done routinely with confocal light microscopes. This requires the ability to focus on separate planes in the specimen, something which cannot yet be done with sufficient resolution with X-rays as the numerical aperture of the focusing optics (usually zone plates) is too low. An alternative approach has been to record several different views with the specimen rotated between each image. Such tomographic studies can produce high resolution images of test objects¹² but as yet such techniques have not been applied to biological specimens other than those only requiring low resolution⁵. Furthermore, the need to take several

images increases the radiation received by the specimen to well beyond that which might be tolerated by even the most radiation-insensitive biological specimen. Similarly X-ray holography, which has been used to image biological specimens^{15,16} precludes the possibility of imaging living material without radiation-induced damage since the imaging times, and radiation doses, are excessive. One technique that has not, however, been explored is that which is widely used in many other forms of microscopy: stereo imaging, in which a pair of images are made from different perspectives. The two images may then be used to determine the relative positions of two structures in three-dimensional space, usually with sufficient Z-axis resolution to determine whether two structures are actually joined or merely appeared to be joined in single images because of superimposition of structures. Both light and electron microscopists have endeavoured to avoid the loss of 3D information by the use of stereo pairs, and stereo pairs have also enhanced the information available for X-ray projection microscopy²¹. Indeed the stereo images taken by X-ray projection microscopy often show details not usually seen by light microscopy¹⁴. Since SXCM has been able to provide better resolution than X-ray projection microscopy, single-shot, laser-pulsed, stereo SXCM should give the best 3D resolution of all the currently available techniques, and it has the advantage over other stereo X-ray methods published so far that it may be used on living cells to capture details before the energy of the X-rays distorts the structure.

In order to achieve stereo SXCM it is necessary to obtain two views from different perspectives simultaneously so as to avoid natural or radiation-induced movement; the rotation of the specimen, as occurs during tomographic studies, is not a viable alternative. Therefore, this paper reports the manufacture of contoured photoresists with V-shaped grooves in which the biological specimen can be placed so that the specimen could be irradiated simultaneously from two separate soft X-ray sources each at 90° to one of the surfaces of the groove (Fig. 1).

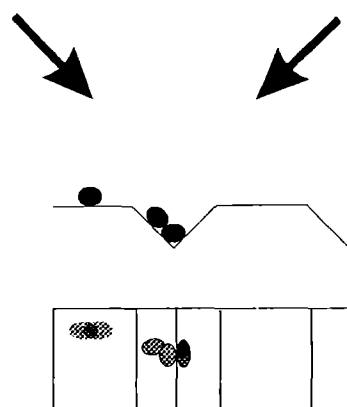


Fig 1 Conceptual diagram of stereo imaging experiment

MATERIAL AND METHODS

V-Grooved Photoresist

Commercial-grade perspex sheeting 1mm thick (Goodfellows, UK)

was used as the starting material since it has the same chemical structure as the spun PMMA (polymethylmethacrylate) photoresists usually used for SXCM recording. The 90° edge of a piece of cut plate glass was used as the tool to emboss a series of parallel 90° V-grooves, this produced grooves from 5-7µm wide at the top and 3-5µm deep. The desired groove depth was obtained by varying both the embossing time and the ambient temperature. The approximately 90° profile of the finished grooves was verified by light microscopy, and the roughness of the finished grooves was found to be about the same, or better, than the flat original surface of the perspex by AFM (atomic force microscopy).

Spun PMMA Photoresist

Some conventional flat PMMA photoresists spun 1µm thick on silicon wafers were used in the same experiment. It was anticipated that they would be used to calibrate exposures with the biological specimens prior to starting the more difficult stereo shots with the V-grooved resists. However, it was found that their sensitivity was about an order of magnitude less than that of the perspex sheeting. It was also found that at the angle used for stereo shots (45° to a flat surface), the increased path length of the X-rays through the water-immersed specimen made it very difficult to get sufficient exposure through the specimen to the photoresist, especially when the laser beam energy to an X-ray target was halved with the stereo setup.

Biological specimens

SXCM images of immersed, living cells on conventional PMMA resists gave several possibilities for specimens whose structure was sufficiently complex that stereo images would be very useful. However, a new problem emerged as these fully immersed specimens were loaded onto the V-grooved perspex. We had no way of preventing some specimens ending up on the top flat surfaces between the V-grooves. The delicate Si₃N₄ windows (Fastec, UK) could not be placed too near the surface of the V-grooved resist lest the specimens distort or break the window, compromising the pressure in the environmental chamber when the target chamber was evacuated to allow free passage of soft X-rays from the targets to the specimen chamber. The extra depth of water required to prevent breaking the window on the specimen would increase the path length to the point where sufficient exposure of the resist surface at the bottom of the V-grooves would be unlikely. The only solution was to switch to specimens which did not require total immersion so that the extra layer of water several microns thick would not be present to attenuate soft X-rays. Mushroom spores and diatoms were chosen since mushroom spores are able to withstand desiccation over long periods as part of the life cycle of a mushroom, and freshwater diatoms in nature survive ponds drying out, besides having a very ordered structure in their silica shells. Basidiospores from the edible mushroom *Agaricus campestris* and specimens of a diatom culture of *Navicula* sp. were found to have sizes such that they would fall to near the bottom of the V-grooves, making them ideal for the stereo SXCM experiments. Specimens were placed in the environmental chamber to ensure specimens did not dry further⁶ and the specimen holder was loaded into the target chamber using a TV monitor to ensure alignment of the V-grooves normal to the paths of X-rays from the two targets. The experimental layout is shown in Fig. 2. The target material was yttrium and the laser (wavelength 1.053µm) energy varied from 10-20J per beam (although for some shots this was increased considerably). The optimum specimen to target distance was approx. 13mm and the soft X-ray fluence at this distance was estimated between 50 and 150 mJcm⁻² at the resist surface. The photosensitive resists were chemically developed with 50% methylisobutyl ketone:isopropyl alcohol and examined initially by DIC light microscopy and finally, when the development depth was sufficient, by AFM (Park Scientific BD2).

RESULTS AND DISCUSSION

Comparison of perspex and spun PMMA as SXCM resists

Initial images suggest that whilst "perspex" sheeting is an extremely sensitive photoresist material it may have unacceptable surface scratches and roughness which make it generally unacceptable for high-resolution SXCM. Measurements of the surface area roughness, using AFM, indicate that figures of 100nm are typical for the exposed surface of commercially available perspex. Despite this criticism it was possible to produce some satisfactory images of some cells as long as the development time for the resist was kept to a minimum. Images of *Chlamydomonas*, very similar to those produced using conventional spun PMMA^{4,8,10} and clearly showing the flagella, the extracellular X-ray lucent area outside of the main cell body and several internal features, including several relatively large X-ray absorbing areas, were successfully obtained. The embossing of perspex therefore offers one way to create the necessary grooved surface which is required for stereo imaging. The use of glass knives with differing angles offers the possibility of producing grooves of differing angles, whilst the duration of embossing and the temperature of the photoresist, affect the depth of the grooves produced. The surface of the grooves produced in this way appears to be a considerable improvement over the unembossed surface (Fig. 3). However, during development of the exposed groove surface, surface cracks appear more often than in the spun PMMA, indicating a difference in the "grain-size" of the two materials which gives the desirable property of increased sensitivity but, undesirably, lower resolution. It may, however, be possible to get higher resolution V-groove surfaces by embossing thick-spun PMMA layers or by increasing temperatures during embossing of perspex.

Stereo SXCM of specimens on horizontal surfaces

An important part of the experiment was to demonstrate that one could obtain two equal, orthogonal soft X-ray beams which were at right angles to the V-grooves. Examination of specimen images on the top surfaces (between the V-grooves) parallel to the plane of the Si₃N₄ window showed this had been accomplished. Since the top of the grooved resist was some distance away from the window, typically 20-25µm, only the middle portion of the resist received X-rays from both targets, whilst the end regions were shaded by their respective soft X-ray-opaque window edges. Thus, at one end of the resist, one ought to see shadows of the specimen extending away from one X-ray source, and at the opposite end the shadows on the resist should extend in the opposite direction. In the middle portion, where both X-rays from both sources struck the same specimen, there ought to be two shadows for each specimen, each of which points away from its X-ray source. Images of mushroom spores (Fig. 4) show no internal detail but those of diatoms show, on one side at least, the substructure of the frustule (Fig. 5). In this case the separation of these features is about 0.6µm. Therefore all of the anticipated conditions were met.

Stereo SXCM of specimens in the V-grooves.

In attempting to get a reasonable proportion of moist biological specimens both in grooves and on horizontal surfaces, one is constantly battling with adhesive characteristics of the specimen and the surface tension of water. The fresh basidiospores of mushrooms are somewhat sticky, making it impossible to get individual spores by dropping them onto the resists. Suspending them in water allowed us to get a better dispersion, and their being slightly heavier than water caused them to fall from suspensions quite nicely into grooves and onto the horizontal surfaces. However, as the suspension dried the last bits of water tended to bead on the horizontal surfaces, and as the water beads dried, the surface tension of the water pulled the spores in the grooves up onto the horizontal surfaces. By the time the water had evaporated enough for the exposure to soft X-rays, none of the spores were in the grooves. By wicking the water away from the ends of the grooves with absorbent tissue it was possible to ensure that at least some spores remained within the grooves. It was easier to ensure that the diatoms remained in the grooves since these organisms secrete a sticky mucilage-like material which caused the cells to

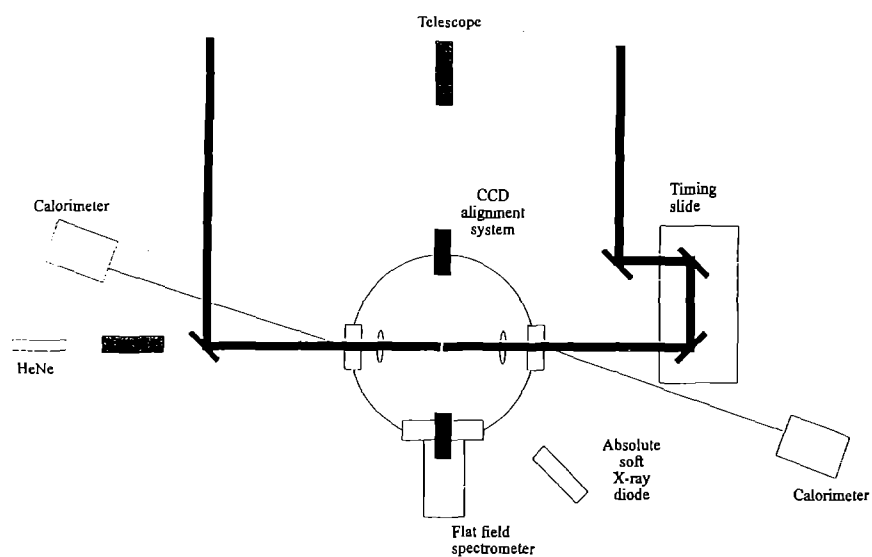


Fig 2 The experimental layout for stereo imaging

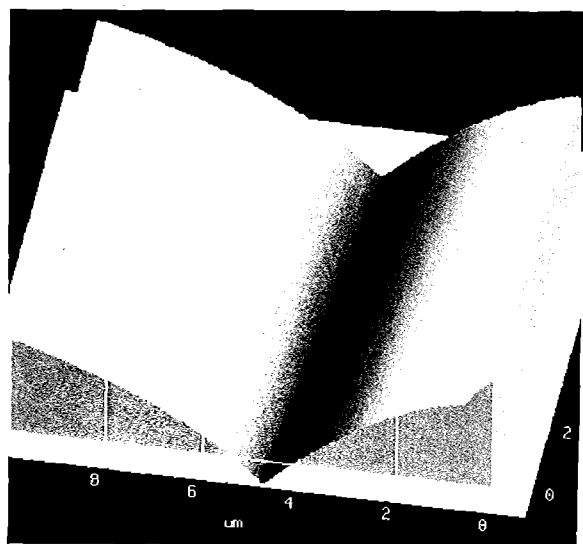


Fig 3 AFM of grooved perspex

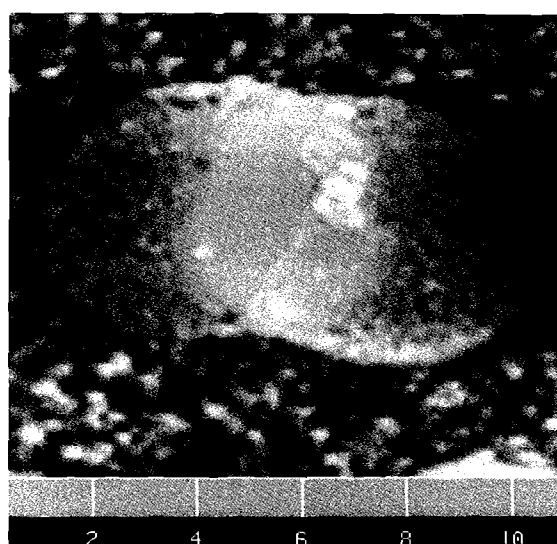


Fig 4 AFM of an image of a mushroom spore on the flat, horizontal surface of perspex

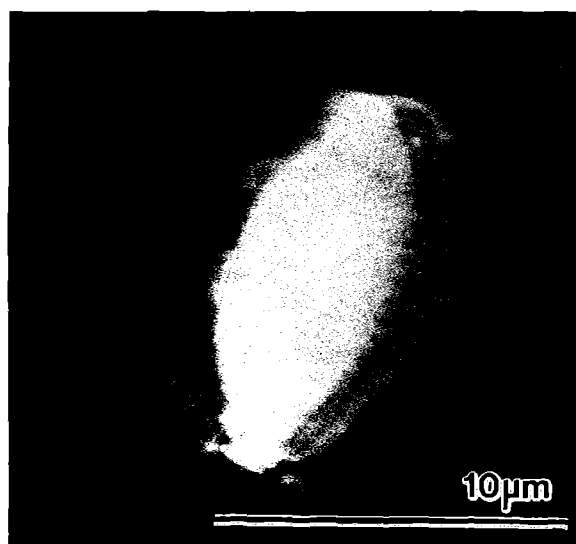


Fig 5 AFM of "paired images" of a diatom frustule on the flat, horizontal surface of perspex

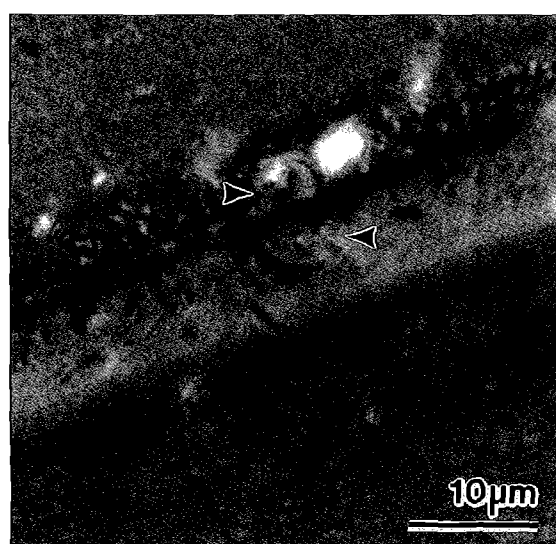


Fig 6 LM of "paired images" (arrowed) of a mushroom spore within a groove

adhere quite strongly to whatever surface they were in contact with. Therefore, once positioned in the grooves, the cells mostly remained there.

By exposing these grooved photoresists to soft X-ray sources, each orthogonal to one of the sides of the groove, it is possible to produce two images of the same specimen, one on each face of the groove (Fig. 6). Ideally these images should be studied using AFM to produce two high resolution images which can then form the basis of the production of the stereo pair. For most AFM's, however, the range of movement in the Z-direction is approximately 1/10 of the XY scan size, thus obtaining images with a small scan head is not possible but this difficulty should be surmountable. At the present time, therefore, we can demonstrate, by LM (Fig. 6), that it is possible to make stereo pairs of living biological specimens with SXCM.

Before this technique could be considered routine several aspects must be attended to. A more suitable starting material for making high-resolution V-grooved resists should be found, the use of thick, spun-coatings would seem the most appropriate. The presence of biological material on the upper, horizontal resist surface must be eliminated so that the distance between the Si_3N_4 window and the bottom of the grooves can be minimised. This will reduce the attenuation of the X-rays so that satisfactory images of the specimens can be obtained at the very bottom of the V-grooves even when the specimens are totally immersed.

We hope that our demonstration of stereo SXCM with living biological specimens will attract other biologists and hasten the technological developments necessary to make this technique more available and easier to perform.

ACKNOWLEDGMENTS

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INERTIALLY-CONFINED LASER-PRODUCED PARTIALLY-DEGENERATE STRONGLY-COUPLED PLASMAS

G. R. Bennett¹, J. S. Wark¹, D. J. Heading¹, R. W. Lee², N. C. Woolsey²,
R. Cauble², P. Young², and H. He¹

¹Department of physics, Clarendon Laboratory, University of Oxford,
Parks Road, Oxford, OX1 3PU, UK

²Lawrence Livermore National Laboratory, Livermore, CA 94551, USA

INTRODUCTION

The production of low-temperature high-density plasmas is of great interest to astro and plasma physicists alike. Such plasmas are generally strongly-coupled and partially-degenerate in character, and may be created in ion or electron beams, self-pinching discharges, laser fusion, capillary discharges, laser-produced plasmas, etc. Strongly-coupled plasmas (SCPs) are predicted to have equation of state (EOS) and transport properties quite different from those of classical plasmas; in addition, the various theories developed for such conditions differ quite dramatically in their predictions. Attempts by previous workers to measure such properties have always been limited by rather poor density and temperature uniformity; however, the plasma production method described below appears to overcome these uniformity limitations. This report describes the complete experimental investigations of the P, V, E equation of state and visible spectroscopy of the plasmas produced; using this method also described are preliminary experimental investigations into of x-ray probing, visible opacity, and d.c. electrical conductivity.

The SCPs were laser-produced and inertially confined between two X-cut quartz substrates which were separated by a known, well-defined micron-scale gap, and where one of the substrates formed a piezoelectric pressure transducer with nanosecond time resolution and a 1 - 40 kbar usable range. The target design is shown below in figure 1 and is described in other reports along with descriptions of the experimental details.¹⁻⁴ The results presented here were obtained from experiments recently carried out in the TA4 facility of the VULCAN laser.

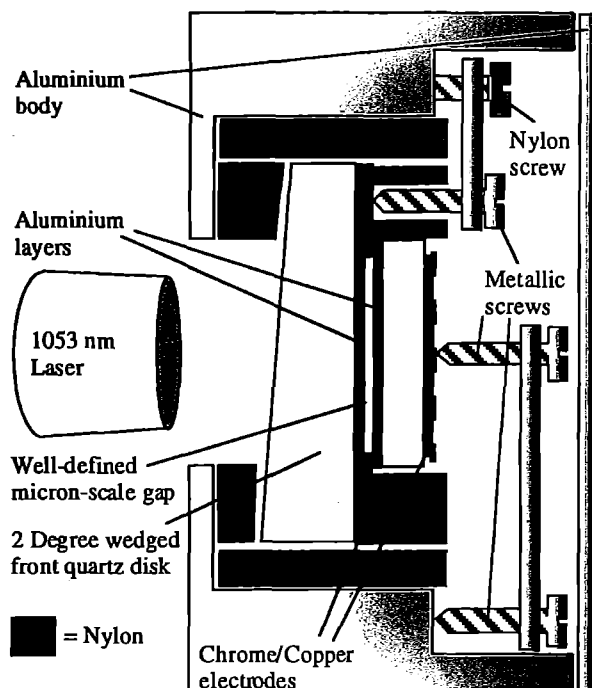


Figure 1 Complete target assembly used for the P, V, E measurements. The rear X-cut quartz disk serves as both a time-resolving piezoelectric pressure transducer, and a confining substrate. The SCP itself forms one of the transducer electrodes.

EQUATION OF STATE

In the ideal-gas limit, plasma pressure is described by

$$P = \frac{2}{3} U, \quad (1)$$

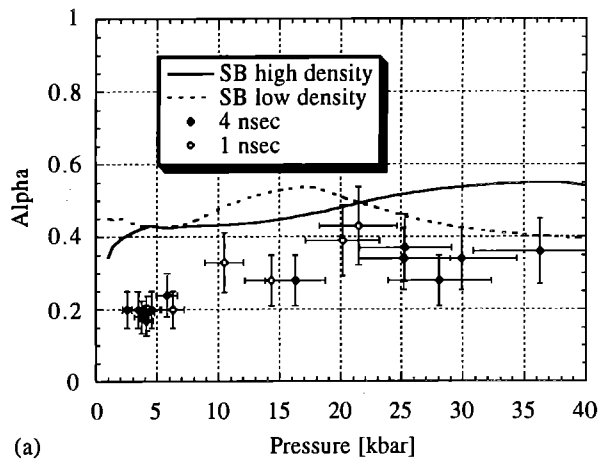
where U is the total internal energy per unit volume. For any real plasma, however, the pressure is related to energy per unit volume by

$$\alpha = \frac{3P}{2U}, \quad 0 < \alpha < 1 \quad (2)$$

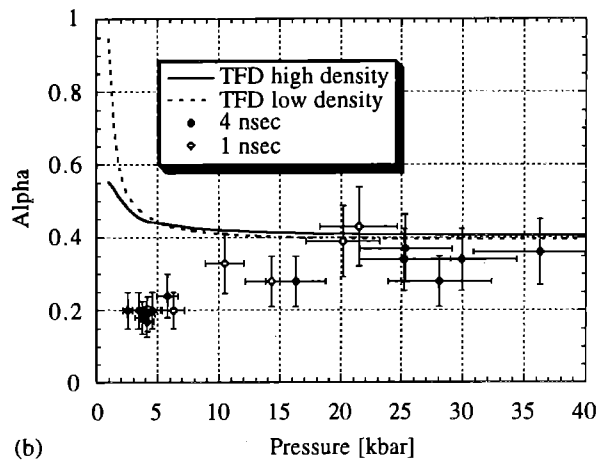
where $1 - \alpha$ is the fraction of energy used in ionisation, latent heats of melting and vapourisation, etc. It is useful here to present the P, V, E data as α versus pressure with α ranging from zero (a solid material), to one (an ideal gas). The data is compared - in figure 2 - with two EOS models of the SESAME library; i.e. the Thomas-Fermi-Dirac (TFD) and Saha-Boltzmann (SB) models. The measured densities at the time of peak pressures, as a fraction of solid aluminium, ranged from 0.46 % (low density) to 0.98% (high density) with both the TFD and SB models predicting intermediate densities to have α values resting between the high and low density curves. Two laser pulses of 1 or 4 nsec FWHM Gaussian were used throughout the experimental work. For the 2 - 20 kbar range the measured values of α were some what less than those predicted by TFD and SB; although for the 20 - 35 kbar range a good agreement with TFD - within the error bars - was observed.

If one assumes the SESAME tables to be correct, then the low α values observed must have occurred as a consequence of energy leaking out of the plasma. Interestingly, the fractional absorbed energy which was lost appeared to increase with decreasing pressure; therefore the actual energy lost may have been similar for all data shots. To address this issue, it was interesting to note that a certain amount of quartz was expected to melt, implying there existed a surface of constant temperature at the molten/solid quartz boundary. Also, the quantity of heat diffused into a semi-infinite strip of material - with one end held at constant temperature - is described by a very simple equation.⁷ Indeed this simple heat diffusion equation explained very well the apparent similarity in values of energies lost over a wide range of pressures. To this end, the diffusion model and the SESAME equation of state subroutines were incorporated into a code developed to include energy deposition by a Gaussian laser pulse, and both effects of the compression of the confining walls; i.e. the changing plasma density and the PdV work done.

Simulations using this code explained very well the apparent deviation of data from theory in the 2 - 20 kbar region, and the good agreement obtained in the 20 - 35 kbar range. A typical 20 kbar pressure shot agreeing with the TFD EOS is shown in figure 3 along with the pressure histories predicted by the two SESAME models. Notice the excellent agreement with theory. Such comparisons give confidence in the overall plasma uniformity and the validity of SESAME itself. One would expect the temperature history predicted by SESAME to be equally valid and this is shown in figure 4 for the same data shot. It is now clear that the production aluminium plasmas with parameters 20 - 35 kbar and 0.5 - 1.0% solid density can be reliably produced: plasmas in the 2 - 20 kbar range are not as interesting due to their rather weakly-coupled nature. The SESAME predicted electron densities and temperatures for these 2 - 35 kbar, 0.5 - 1.0 % solid density Al plasmas were $1 - 15 \times 10^{20} \text{ cm}^{-3}$ and 2 - 15 eV respectively.



(a)



(b)

Figure 2 (a) Comparison of the aluminium plasma peak pressure data with SESAME SB model. (b) As a (a) except SESAME TFD model; good agreement is observed for 20 - 37 kbar

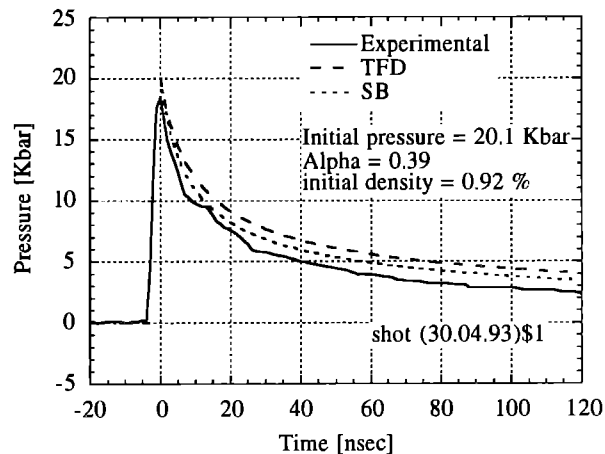


Figure 3 A 20 kbar experimental pressure history, which agrees with the TFD EOS model, along with two pressure histories predicted by the TFD and SB models

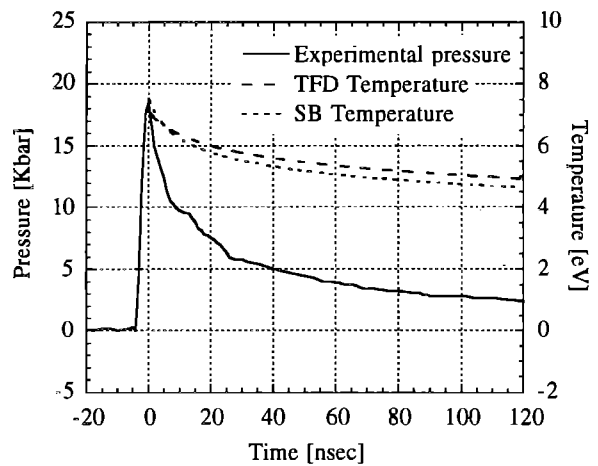


Figure 4 As figure 3 but with the predicted temperature histories

VISIBLE SPECTROSCOPY

The visible spectroscopy of the 20 - 35 kbar well-characterised plasmas formed the next stage of the project. Each spectral line was expected to be very broad due to the high electron density; therefore the relatively high dispersion of conventional grating spectrometers was expected to be of little use. Furthermore, the comparatively low luminosity of the plasmas themselves and the low throughput of such spectrometers may have resulted in a signal well below the detection level of the streak camera. A prism spectrometer, however, overcomes these two limitations since it has a very high throughput and a very low dispersion. To this end, such an instrument with a 400 - 700 nm wavelength span was built *in-situ* in TA4. Glass achromatic optics were used throughout and a 10x de magnified image of the plasma was focused onto the entrance slits of the spectrometer. The spectrometer was coupled to an Imacon 500 streak camera fitted with reflective streak optics and two image intensifiers in tandem to increase sensitivity.

Many high-quality low-noise streaks were obtained using 5 nsec/mm and 1 nsec/mm, with all streaks showing no evidence of spectral lines. For example, figure 5(a) shows the visible emission spectroscopy from an aluminium plasma (36.3 kbar pressure and 0.55% solid density at $t = 0$). In this example one can see no spectral structure even up to ~ 125 nsec after the laser pulse: figure 5(b) shows the corresponding simultaneous pressure measurement history. The featureless streaks observed from these plasmas were the result of spectral lines being so severely broadened by the high electron density, that they essentially merged into a continuum.³ Triggering the camera many 10s of nsec after the laser pulse yielded the same featureless spectra, indicating the electron densities were sufficient, even in cooling plasmas, to merge all lines.

In the light of these results a deliberate increase of the gap width to ~ 250 microns actually lowered the electron density, with the consequence that lines became distinguishable.³ Figure 6, for example, shows the time-resolved visible spectroscopy from such data shot, and the difference with figure 5(a) is immediately obvious. In particular, 30 nm FWHM lines are clearly observable; although at around $t = 0$ (the laser heating) until 40 nsec (the time for plasma to fill the gap and become homogenous) the spectrum is in the first instance difficult to model. For times greater than ~ 40 nsec, however, the spectrum emitted is that of cool, dense uniform optically-thin plasma. In fact, for these particular plasmas, the electron densities were measured from the line widths and line shifts,^{3, 5-6} and by using previously published line data it was possible to model the observed spectrum to yield an inferred temperature.^{3, 5-6} For example, at $t = 60$ nsec the electron density measured from figure 6 was $2.5 \times 10^{19} \text{ cm}^{-3}$, and the corresponding temperature was 4.3 eV^{3, 5-6} (inferred by entering temperature value into spectra code that yielded best match with experiment).

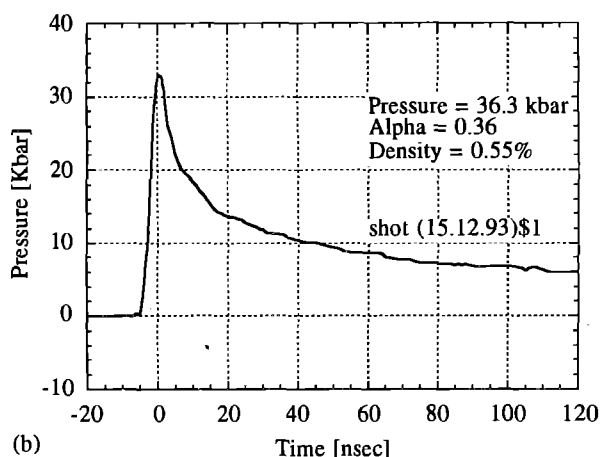
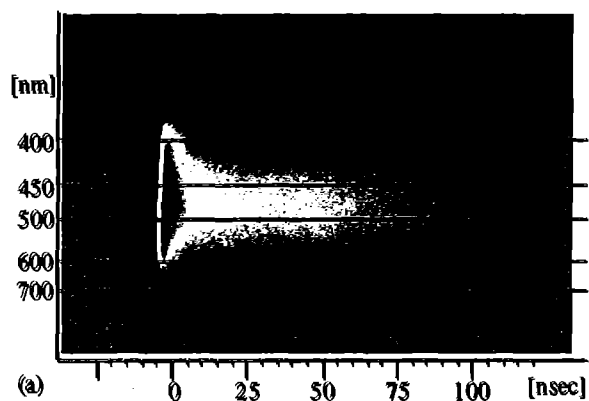


Figure 5 Simultaneous spectroscopy and pressure measurements of shot (15.12.93)\$1

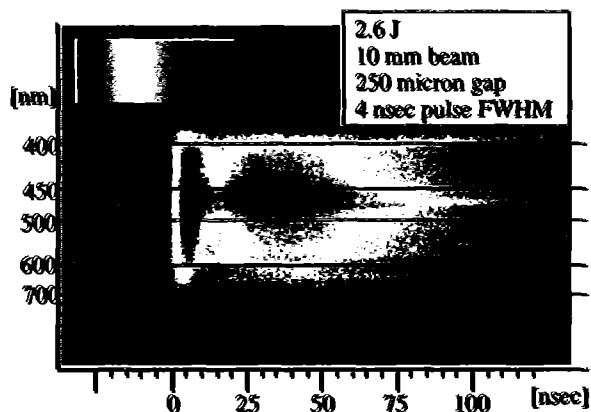


Figure 6 Time-resolved visible spectroscopy of a cool, dense aluminium plasma of order 10^{19} cm^{-3} electron density and several eV temperature. Notice the lines become narrower at later times because the plasma electron density reduces. At $t \sim 40$ nsec onwards the plasma is uniform across the gap width.

The $1 - 3 \times 10^{19} \text{ cm}^{-3}$ electron density $1 - 8 \text{ eV}$ aluminium plasmas created in the wider gaps have allowed one to study optical spectroscopy to the very limit of where lines may be distinguished. Furthermore, the electron density values for these optically thin, uniform plasmas are an order of magnitude larger than those measured in plasmas created using previous production techniques. Also, these plasmas can exhibit ionisation potential depressions,^{3, 5-6} larger than those of plasmas created in ICF experiments.

X-RAY AND VISIBLE PROBING

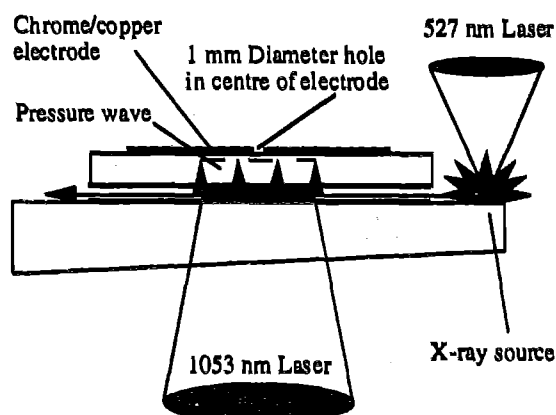


Figure 7 Target based on the design shown in figure 1. Allows x-rays to probe the laser-produced inertially-confined plasma

In addition to the pressure, mass density, internal energy per unit volume, and visible emission spectroscopy - there are at least three other fundamental plasma measurements that are possible using a target based on figure 1. For instance, a preliminary experiment performed in TA4 focused the single 1053 nm beam onto a metallic layer, as shown in figure 7, with a view to testing x-ray probing experiments. The side on probing with the x-rays created, was necessary because the x-cut quartz confining walls were opaque to wavelengths less than $\sim 220 \text{ nm}$. The experiment indicated that x-rays which were passed through a 5 micron gap, across the diameter of the target, could be easily detected. In a similar vein, it is possible to probe with optical light, through the 1 mm hole shown, in order to afford visible opacity measurements. Under certain conditions such optical depth measurements are equivalent to direct Coulomb logarithm (Cl) measurements. The importance of such investigations arises from the implicit inclusion of Cls into various transport property theories such as thermal conductivity, Inverse Bremsstrahlung absorption, d.c. electrical conductivity, etc.

D.C. ELECTRICAL CONDUCTIVITY

In this spirit, a further preliminary experiment performed in TA4 addressed the d.c. electrical conductivity directly. In particular, a known current was passed through a confined plasma during the laser heating and thereafter. Now because the plasma dimensions were known, a constant current through the plasma would have resulted in a voltage drop directly proportional to its resistance, allowing the resistivity to be determined directly. Indeed the current was observed to remain constant for several 10s of nsec after the laser pulse - which would not only simplify the data analysis - but would eliminate the problems associated with any inductance within the circuit, since any inductive device remains dormant until the current passing through it changes with time. To conclude now. In addition to the very successful EOS and spectroscopy work thoroughly investigated already, preliminary experiments have indicated the possibility of *simultaneous* pressure, mass density, internal energy per unit volume, visible spectroscopy, x-ray probing, visible opacity, and d.c. electrical conductivity measurements using a single target design.

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STUDIES OF CONFINED LASER PRODUCED PLASMAS

D.J. Heading, G.R. Bennett, J.S. Wark

Clarendon Laboratory, Department of Physics, University of Oxford, Parks Road, Oxford OX1 3PU, UK

INTRODUCTION

Recent work has shown that the generation of dense, cool aluminium plasmas by the technique of confined laser ablation can yield plasma electron densities of $\sim 10^{19} \text{ cm}^{-3}$ ^{1,2}. This is an order of magnitude greater than those previously observed in conventional plasma spectroscopy sources, such as z pinches or flash tubes^{3,4}. As the ionisation potential depression of an atom in a plasma is both temperature and density dependent, we can address issues of the ionisation balance in a plasma at these more modest densities, which include similar physics to those found in inertially confined fusion plasmas⁵. Furthermore, because of the low ionisation state of the plasma, the emitted spectral lines are observable using optical spectroscopy. This also raises the interesting question of the value of the ionisation potential depression of a neutral species immersed in a dense plasma.

The plasmas are generated in a target consisting of a confining wall transparent to the incident laser light, a gap, on the order of 250 microns wide, the target foil (in our case aluminium) and a supporting wall. The laser generates a plasma on the foil, which then expands into the gap, cooling as it does so. The plasma then hits the confining wall, and reheats. Homogeneity should be ensured by the hydrodynamic shocks passing through the plasma. Thus the time of interest for spectroscopic observation is after two transit times, which is around 60 ns for the plasmas of interest here.

We have pursued further studies in the generation of dense plasmas by confined laser ablation. Specific investigations of the homogeneity of the plasma and work on simulating the observed spectrum have been undertaken and are reported here. We find that the plasma has a high degree of homogeneity, and that simulations of the plasma emission can, when compared with the experimental data, be used to diagnose the experimental conditions.

HYDRODYNAMIC SIMULATIONS

A code has been written which incorporates the Sesame equation of states tables⁶ and thermal conductivities⁷ into the Medusa 1 dimensional Lagrangian hydrodynamic code⁸. This has been reported previously⁹. For the purposes of the simulation, the code was expanded to include 5 layers. For our simulation, these were set to be a layer of quartz, then solid aluminium, then hot aluminium, then a gap layer, filled with cool aluminium at low density, and finally a confining layer also of quartz. It was found difficult, because of the Lagrangian nature of the code, to write a code which simulated the laser ablation phase, and the subsequent expansion of the plasma into the gap, its contact with the wall and subsequent cooling. This is due to the reduction in size of the initially large cells which form the gap, which leads to the timestep in the code being radically reduced and the code running out of time. This is still the subject of investigation.

Running the code with a 250 micron gap, in which the first microns were filled with the hot plasma lead to the results shown in figures 1. The initial plasma temperature was set to be 8 eV. This is the temperature derived from the Sesame equations of state for a plasma with the internal energy derived for a typical experimental shot. As can be observed, after 30 ns, the plasma is quite homogenous. The cooler regions near to the walls are seen to be narrow and the region nearest to the confining wall is actually found to have a half width of the temperature gradient of 6 microns. The central region of the plasma is homogenous in temperature to 10%. The hydrodynamic confinement thus ensures

the high degree of homogeneity of the plasma. The cooling rate of the plasmas, which is caused by conduction into the walls, is in reasonable agreement with those observed in the experiment.

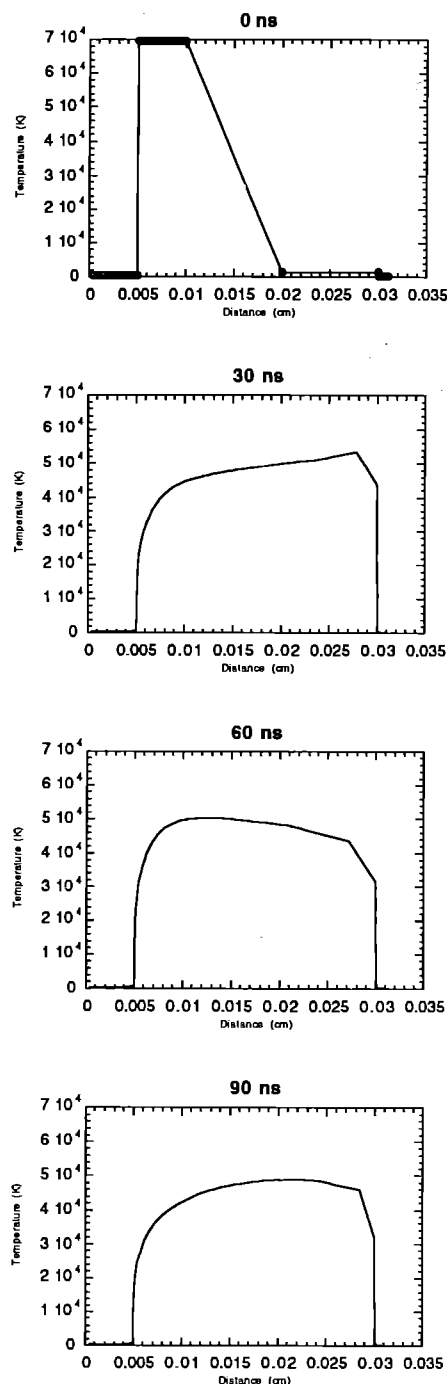


Figure 1: Simulations of the Temperature Distribution in a Confined Target for Various Times.

SPECTRAL SIMULATIONS

In order to understand the observed spectra, it is necessary to simulate the emission from the plasma. This is because the spectral lines are so broad that the underlie each other, thus making the spectrum difficult to interpret. In order to simulate the

spectrum, a large quantity of data for the atomic species is required. This includes ionisation potentials, energy levels, transition probabilities and line shape parameters. For aluminium this information can be found in the literature. The code has to calculate the ionisation balance and then the relative intensity, width and shift of the individual lines. This work is to be published in more detail elsewhere¹⁰.

Briefly, the program first calculates the ionisation balance in the plasma. This is done using the Saha-Boltzmann equation. For this we need the partition functions of the different species in the plasma. For these plasmas the normal approximation of using the ground state degeneracy to give the value of the partition function is inadequate, because higher lying states will be highly populated. Thus an explicit calculation of the partition functions is required. This is performed using data from the literature for the energy levels of the low lying states and a hydrogenic approximation¹¹ for the upper states. These calculations both require the calculation of the ionisation potential depression. This may be calculated using the Debye, ion sphere or Stewart-Pyatt¹² models. Once the ionisation balance is calculated, the relative intensity, line width and shift of each line may be calculated, assuming a Lorentzian line shape. Hence a spectrum is synthesised.

The input variables to the code are the electron density and the temperature. By varying these a fit to the experimental data can be achieved. In this way a diagnostic of the plasma can be undertaken. This method is very sensitive to the temperature, but not to the electron density. Thus, a measurement of the line widths directly from the data is also used to check with that derived from the code. These two values have been found to be in good agreement. Sample fits to the data are shown in figures 2 and 3. Also shown in figure 2 is the calculated spectrum using only the ground state degeneracies in the partition functions. It can be seen that the full model is a great improvement over these. In figure 3 are also shown plots of the spectrum with the temperature varied by 0.5 eV. The sensitivity of the data to the electron temperature is thus demonstrated.

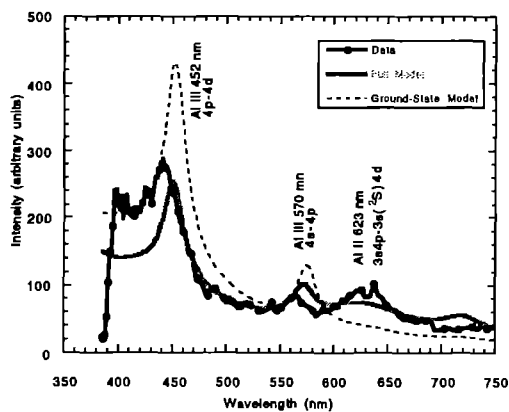


Figure 2: Experimental data from 60 ns after the laser pulse. Fits are using the full model and the ground states only model, $N_e = 2.5 \times 10^{19} \text{ cm}^{-3}$, $T_e = 4.3 \text{ eV}$

SUMMARY

Simulations of the hydrodynamics and emission from a dense cool plasma generated by confined laser ablation have been described. Despite difficulties inherent in using a Lagrangian code the hydrodynamic simulations have shown that the plasma has a remarkable degree of homogeneity. The wall temperature

gradient having a half width of 6 microns in a 250 micron gap. The simulations of the emission spectra have proved to be successful in the temperature diagnostic of the plasma. These simulations assume that the emission comes from a homogenous optically thin layer of plasma. Thus these tend to confirm the results of the hydrodynamic simulations, although it would be necessary to combine the two programs to prove this point.

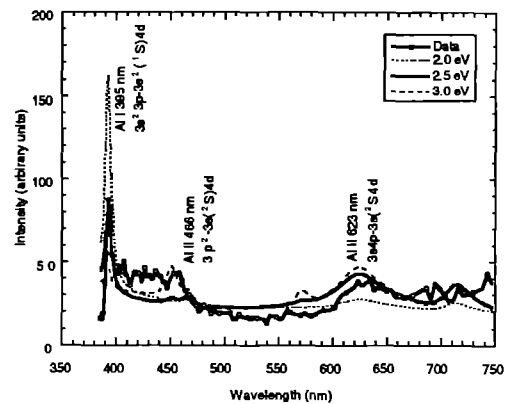


Figure 3: Experimental data from 120 ns after the laser pulse. Fits are using the full model varying the temperature, $N_e = 1.5 \times 10^{19} \text{ cm}^{-3}$, $T_e = 2, 2.5 \text{ and } 3 \text{ eV}$

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HIGH-POWER LASER-PLASMA MODELLING WITH RELEVANCE TO ASTROPHYSICAL PLASMAS

K Nazir¹, T D Beynon¹, S J Rose^{1,2} and A Djaoui²

¹School of Physics and Space Research, University of Birmingham

²Rutherford Appleton Laboratory

INTRODUCTION

The recent rapid progress in short pulse, high intensity lasers has made possible the generation of hot, dense plasmas consisting of matter in an extreme state of high energy concentration (Key 1991). It is the high material density, temperature and radiation flux of laser-produced plasmas which make them suitable for measurements relevant to certain astrophysical phenomena (Rose 1991, Ripin *et al* 1990).

We have performed soft X-ray emission spectroscopy of laser-irradiated targets, with a buried layer of iron. The experiments (Tallents *et al* 1995) were performed on the SPRITE CPA system and involved irradiation of the buried layer targets with 248nm light of 300fsec FWHM and a peak intensity of $10^{17} \text{ W cm}^{-2}$. We measured a large cluster of lines at photon energies ranging from approximately 800eV upwards.

The plasma conditions formed in the diagnostic layer were modelled using a hydrodynamic code coupled to an atomic physics package. The temperatures and densities achieved in the iron layer were characteristic of those found in the solar interior (Cox and Guilli 1968). Short pulse laser experiments thereby offer a possible direct approach to opacity measurements at higher density and temperature regimes than can be currently achieved with the technique of radiative heating (Rose 1991).

SHORT PULSE LASER EXPERIMENTS AND ASTROPHYSICS

When the laser pulse duration is significantly less than the hydrodynamic response time, the energy transport processes are quite different from those in steady-state ablation (Key 1991). The resulting hydrodynamic motion of the plasma can be insignificant during a short laser pulse and heat flow, through electron thermal conduction, occurs at densities close to solid in contrast to steady-state ablation where the main heating occurs at densities close to the critical density (Key 1991). The interaction of sub-picosecond laser pulses with solid material provides a possible method of measuring plasma properties at extremely elevated temperatures but nearly constant density (Rose 1991).

The measurement of opacity is complicated by gradients and transients produced initially through direct laser heating of the plasma, and later by rapid expansion and cooling (Key 1991). This problem is even more pertinent for plasmas produced in sub-picosecond laser pulse experiments due to the lack of available spectrometers with sufficient temporal resolution. This complication may be partially overcome by limiting the spatial and temporal conditions at which plasma emission occurs through the use of a thin buried tracer layer.

To perform astrophysically meaningful opacity measurements in the laboratory, the plasma should be in a state of local thermodynamic equilibrium (LTE) and have negligible gradients of density and temperature, especially for the duration of strong emission (Rose 1991). The target used in the SPRITE experiment consisted of three-layers in which a tracer foil of iron was placed between two layers of a lower-Z material (CH or Al) which act as tampers.

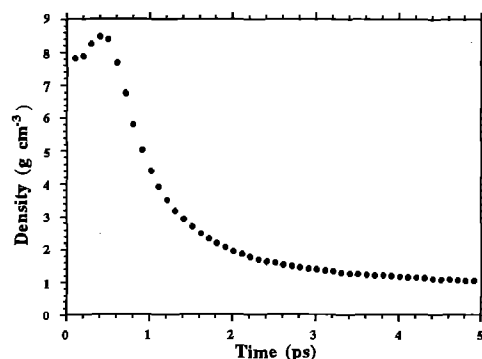


Figure 1: Time development of mean iron plasma cell density as predicted by MED103

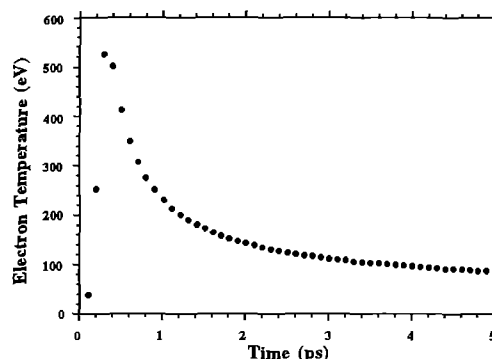


Figure 2: Time development of iron plasma cell electron temperature as predicted by MED103

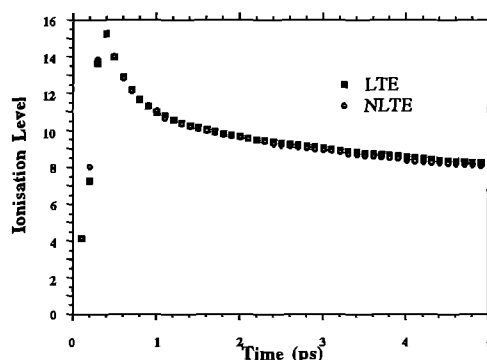


Figure 3: Time development of mean iron plasma cell ionisation level, as predicted by separate NLTE and LTE ionisation models coupled to MED103

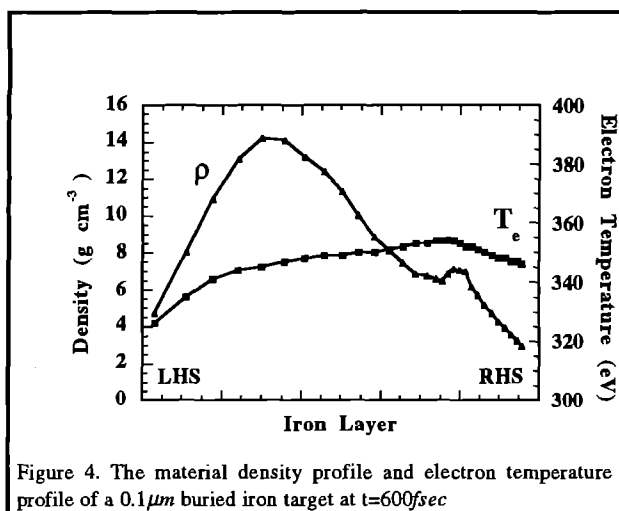


Figure 4. The material density profile and electron temperature profile of a $0.1\mu\text{m}$ buried iron target at $t=600\text{fs}$

HYDRODYNAMIC MODEL AND RESULTS

The modelling is based on a hydrodynamic code coupled to a time-dependent non-LTE ionisation model of the atomic kinetics. A planar target is irradiated in the axial direction and a 1-D Lagrangian hydrodynamic code (Christiansen *et al* 1974) (MEDUSA) is used to simulate the energy deposition and plasma expansion. In our modified version of MEDUSA, MED103, a fixed fraction (20%) of the laser energy absorbed at the critical density is deposited in the plasma by resonance absorption. Of this, a specific fraction is converted into hot electrons. The main process of laser energy absorption in the plasma is inverse Bremsstrahlung. The heat flow is modelled by the smaller of the Spitzer value and a flux limited (20%) free-streaming limit (Key 1991).

A total of 60 Lagrangian cells were used to model the different layers of the sandwich target. The laser beam is incident on the right hand side (RHS) surface of the target on the overcoat layer (figure 4). From the left hand side (LHS) surface of the target to the right: (a) cells 1 to 15 were assigned to the substrate (mylar) layer, at the target back (with respect to the incidence of the laser beam); (b) cells 16 to 45 were assigned to the buried diagnostic layer (Fe); (c) the remaining cells, 46 to 60, were allocated to the overcoat layer (CH or Al). From the RHS target surface to inner layers, the target layer composition was CH plastic overlay of thickness $0.1\mu\text{m}$; followed by a Fe buried layer (solid density) of thickness $0.1\mu\text{m}$; and finally, a $2\mu\text{m}$ CH plastic substrate (mylar).

The hydrodynamic results, depicted in figures 1, 2 & 3, show mean plasma values, as a function of time (ps), of density, electron temperature and ionisation level reached, respectively; we observe a close agreement of non-LTE ionisation dynamics to LTE predictions (figure 3). In figure 4, we depict plasma density and electron temperature as a function of space (axially over the buried iron layer) at a time of $t=600\text{fs}$ (i.e. 300fs after the switching off of the laser pulse).

We observe a peak in the mean plasma density of over 8gcm^{-3} at around $t=600\text{fs}$, reaching a minimum of 1gcm^{-3} (figure 1). At this point in hydrodynamic time ($t=600\text{fs}$) we observe a mean plasma electron temperature of over 300eV (figure 2). Ionisation levels (figure 3) reach a maximum of Ne-like Fe ($Z^*=16$), just after the laser pulse is switched off. From figure 4, we can observe a quasi-homogeneous iron plasma in temperature (i.e. variation in temperature across the layer is small), but a density gradient does exist. Simulations performed for an identical iron target with iron doped at 1gcm^{-3} density rather than solid, reveal a significant decrease in density gradient development across the buried layer (upon laser irradiation). The main disadvantage of using a doped iron layer at a significantly lower density is the reduction in photon signal from the target upon laser irradiation.

CONCLUSIONS

Numerical analysis using a Lagrangian hydrodynamic model coupled to NLTE ionisation physics was found to predict that the density and temperature of high-Z plasmas from high-intensity short-pulse lasers would be significantly higher than those currently achievable by other techniques, such as radiative heating. The period of strong X-ray emission is predicted to be around 300fs after switching off the laser pulse (at around $t=600\text{fs}$ in the above figures). From the hydrodynamic simulations (figures 1 & 2) we predict high material density and temperature in the plasma, in excess of 1gcm^{-3} and 300eV , at this point in hydrodynamic time.

The direct irradiation of solid sandwich targets was found to produce an iron plasma, quasi-homogeneous in temperature for the duration of strong X-ray emission. The data analysis of the SPRITE experiments is currently in progress. By comparison of experimental data with opacity calculations we may be in a position to comment on the applicability of current opacity models to astrophysical phenomena.

ACKNOWLEDGEMENTS

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Li-LIKE AND He-LIKE SATELLITE EMISSION FROM PLASMA PRODUCED BY A 12 ps LASER PULSE

A. Djaoui†, R. Evans†, T. A. Hall†, A. Badger‡, F. B. Rosmej§

† Rutherford Appleton Laboratory, Chilton, Didcot, Oxon, OX11 0QX, U.K.

‡ Department of Physics, University of Essex, Wivenhoe Park, Colchester, CO4 3SQ, U.K.

§ Ruhr-Universität Bochum, Institut für Experimentalphysik V, Universitätsstr. 150, D-44780 Bochum, Germany.

INTRODUCTION

Plasmas produced by picosecond and subpicosecond optical laser pulses interacting with solid targets at intensities greater than 10^{15} W/cm^2 , are currently the subject of numerous studies because of their unique properties: high energy density and highly transient, non-equilibrium behaviour. A very important diagnostic tool for these plasmas is K-shell spectroscopy of H-like and He-like ions and in particular satellite transitions from doubly excited autoionizing levels of He-like and Li-like ions (Gabriel 1972, Gabriel and Paget 1972, Jacobs and Blaha 1980). In this work we investigate the diagnostic potential of Li-like and He-like satellites and present results for a thin Al target (2000 Å) which is irradiated with a 12 ps KrF laser at intensities up to $\sim 10^{16} \text{ W/cm}^2$. During the early evolution of the plasma, time-dependent calculations of line ratios which post-process the hydrodynamic simulation of the laser target interaction are in overall good agreement with the experimental measurements.

EXPERIMENTAL SETUP

3J of laser energy was focused using an f/10 calcium fluoride lens on to 2000 Å coatings of Al deposited on a glass microscope slide. The laser pulses were 12 ps FWHM and were Raman shifted in methane to 268 nm wavelength. The ASE prepulse to main pulse power ratio was below 10^{-10} and consequently no plasma was formed before the arrival of the main pulse. By varying the focal spot size incident laser intensities from 10^{14} W/cm^2 to greater than 10^{16} W/cm^2 were obtained. The time resolved spectra were recorded using a streaked x-ray spectrograph. This consisted of a TiAP crystal which dispersed the incoming x-rays onto the entrance slit of a fast x-ray streak camera fitted with a KBr photocathode. The streak rate of the camera was $\sim 5 \text{ ps/mm}$ and combined with an input slit width of 1 mm gave a temporal resolution of around 5 ps. The spectral resolution of all the spectrometers used was dominated by the source size, which gave $\lambda/\Delta\lambda \sim 500$.

SATELLITE EMISSION FROM A STATIONARY PLASMA

The most intense Li-like satellites are due to the following transitions: $1s^2 2p^2 \ ^2P - 1s 2p^2 \ ^2P$, $1s^2 2p^2 \ ^2P - 1s 2p^2 \ ^2D$, $1s^2 2p^2 \ ^2P - 1s 2p^2 \ ^2S$, $1s^2 2s^2 \ ^2S - 1s 2p(1P)2s^2 \ ^2P$ and $1s^2 2s^2 \ ^2S - 1s 2p(3P)2s^2 \ ^2P$. These correspond to the abcd, jkl, mn, qr and st satellites respectively in Gabriel's notation (Gabriel 1972). Experimental resolution limitations and the proximity of many satellites allow us to distinguish two separate features only on the long wavelength side of the He-like resonance line W; the Li-like satellites (qr, abcd and jkl) and the blending of He-like intercombination line Y and st, mn Li-like satellites. A third feature of interest is the He-like satellite (excluding $1s 2s^1 \ ^1S_0 - 2s 2p^1 \ ^1P_1$ and $1s 2p^1 \ ^1P_1 - 2p^2 \ ^1S_0$) which occurs on the long wavelength side of the H-like resonance line.

We investigated the possibility to use the wavelength integrated relative intensities of these features as a density and temperature diagnostic using the RATION code (Lee *et al* 1984). This code calculates steady state ionic and excited state populations. The calculated ratio of the Li-like satellites (qr, abcd, jkl) to

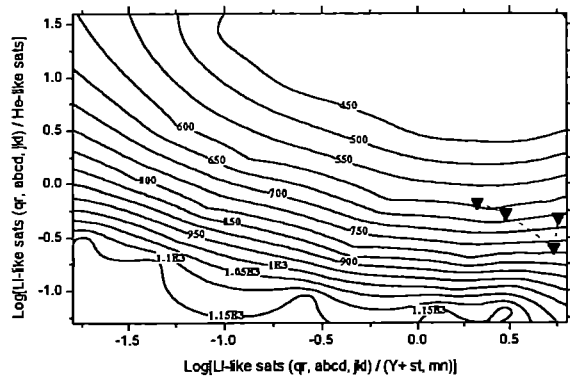


Fig. 1: Contour of the logarithm of the electron density (cm^{-3}) as a function of the logarithm of the ratio of the wavelength integrated intensity of the Li-like satellites (qr, abcd, jkl) to the He-like intercombination Y and the st, mn Li-like satellites and the ratio of the wavelength integrated intensity of the Li-like satellites (qr, abcd, jkl) to the He-like satellites (excluding $1s 2s^1 \ ^1S_0 - 2s 2p^1 \ ^1P_1$ and $1s 2p^1 \ ^1P_1 - 2p^2 \ ^1S_0$). The triangles are time resolved experimental points.

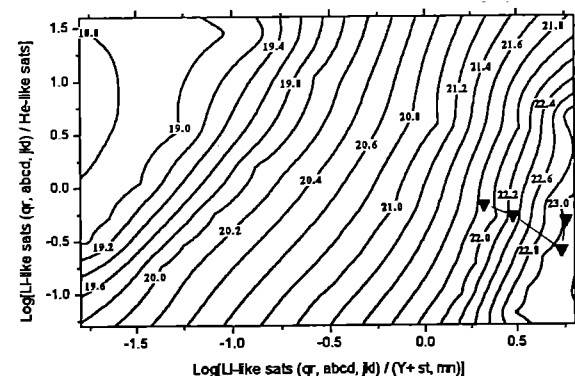


Fig. 2: Contour of the electron temperature (eV) as a function of the logarithm of the ratio of the wavelength integrated intensity of the Li-like satellites (qr, abcd, jkl) to the He-like intercombination Y and the st, mn Li-like satellites and the ratio of the wavelength integrated intensity of the Li-like satellites (qr, abcd, jkl) to the He-like satellites (excluding $1s 2s^1 \ ^1S_0 - 2s 2p^1 \ ^1P_1$ and $1s 2p^1 \ ^1P_1 - 2p^2 \ ^1S_0$). The triangles are time resolved experimental points.

the He-like satellites has a strong dependence on temperature with a weaker dependence on electron density. The calculated ratio of the Li-like satellites (qr, abcd, jkl) to intercombination line Y plus the st, mn satellites has a strong dependence on density with a weaker dependence on temperature. Each one of these ratios is a poor diagnostic when used on its own. The two ratios used simultaneously provide a more reliable diagnostic as shown in Figs 1 and 2 where we have plotted contours of density (logarithm of density) and temperature as a function of the ratios of the wavelength integrated intensities.

TIME-DEPENDENT CALCULATION OF EMISSION

The experiment was simulated with a one dimensional Lagrangian hydrodynamic code which includes time-dependent, non-LTE, average atom model ionisation physics (Djaoui and Rose, 1992). Energies involved in the atomic processes of excitation and ionisation are included in the electron energy balance equation. Radiative energy losses are also included under the assumption of an optically thin plasma. The laser energy is absorbed by inverse bremsstrahlung in the undercritical blow off plasma with a fraction of the laser energy reaching the critical density dumped in the corresponding computational cell as resonance absorption. The inclusion of hot electrons did not have any significant effect on the hydrodynamic behaviour of the target due to the small value of $I\lambda^2 < 7 \times 10^{14} \text{ W}\mu\text{m}^2/\text{cm}^2$ in these experiments. Flux limited heat conduction with a flux limiter equal to 0.1 was used. The relative magnitude of the different ions depends strongly on the irradiance conditions with higher intensities resulting in more ionisation towards the fully stripped stage. We therefore chose the laser intensity also by means of comparing the calculated H-like and He-like ion densities and the measured Ly- α and W-line intensities. Results for an incident peak intensity approximately equal to $5 \times 10^{15} \text{ W/cm}^2$ are analysed here.

Detailed information of the satellite intensities can be obtained by taking the temperature and density history of the computational cells across the original Al layer from the hydrodynamic simulation as input to FLY, the time-dependent version of the RATION code. FLY then calculates the intensities at each timestep and for each cell for transitions of interest (including the satellite intensities). Since the optical depth of the satellites is small, the volume integrated emission is obtained by summing the contribution from each cell. This simplification provides a major advantage compared to the use of resonance lines where optical depth effects on level populations and intensities make the calculation much more complicated. Figure 3 shows the ratio of the wavelength integrated intensity of the Li-like satellites (qr, abcd and jkl) to the He-like intercombination line Y plus Li-like satellites (mn and st). The hydrodynamic/FLY calculations at the two intensities shown are in overall agreement with measurement considering the large relative error of the experimental data which is estimated to be about 50 per cent. The ratio of the Li-like satellites to the He-like satellites is shown in figure 4. This ratio shows less variation than the previous one (except for the first data point at -10 ps) and is very sensitive to the laser intensity used in the hydrodynamic simulation. The experimental data is bracketted by the two calculations at 10^{15} W/cm^2 and 10^{16} W/cm^2 . This overall agreement is good considering the difficulty in measuring the experimental intensity on target accurately.

Using the experimentally measured ratios at 0, 10, 20 and 30 ps, electron density and temperature points are indicated in Figs 1 and 2 with the triangles. It is important to note here that

the plasma parameters indicated in Figs 1 and 2 are that of a stationary plasma which would have the same line ratios as the transient plasma considered. Nevertheless the tendency of the plasma to rapidly expand and cool is clearly well reproduced.

In conclusion, the behaviour of Li-like and He-like satellites emitted by a short pulse laser-produced plasma is well reproduced by time-dependent calculations of the hydrodynamics, ionisation and excitation dynamics.

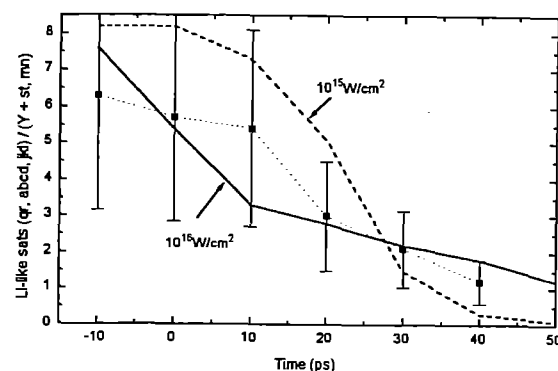


Fig. 3: Ratio of the wavelength integrated intensity of the Li-like satellites (qr, abcd, jkl) to the He-like intercombination line Y plus the Li-like satellites st and mn as a function of time with respect to the peak of the laser beam as measured experimentally (squares) and as calculated with an incident intensity of 10^{16} W/cm^2 (full curve) and 10^{15} W/cm^2 (dashed curve). The experimental error bars are also indicated.

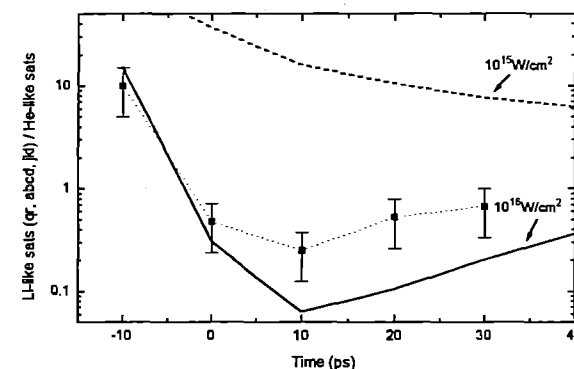


Fig. 4: Ratio of the wavelength integrated intensity of the Li-like satellites (qr, abcd, jkl) to the He-like satellites (excluding $1s2s \ ^1S_0 - 2s2p \ ^1P_1$ and $1s2p \ ^1P_1 - 2p^2 \ ^1S_0$) as a function of time with respect to the peak of the laser beam as measured experimentally (squares) and as calculated with an incident intensity of 10^{16} W/cm^2 (full curve) and 10^{15} W/cm^2 (dashed curve). The experimental error bars are also indicated.

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SHORT PULSE LASER HEATING OF NEON GAS TARGETS.

A. Djaoui(*) and A. A. Offenberger(+)

* Rutherford Appleton Laboratory, Chilton, Didcot, Oxon, OX11 0QX, U.K.

+ Department of Electrical Engineering, University of Alberta, Edmonton, Alberta, Canada T6G 2G7.

INTRODUCTION

It has been suggested that for short laser pulses (~ 100 fs) a plasma far from collisional equilibrium (with an electron thermal temperature much smaller than the ionisation temperature) can be produced [1] by optical field ionisation (OFI). Three-body recombination following the short laser pulse can result in population inversion between excited states and the ground state of the ions. This transient x-ray laser scheme imposes severe requirements on plasma conditions and is not possible if the electron temperature is much higher than about a tenth of the ionisation energy of the recombining ions. For the Li-like neon scheme [2] the plasma must be almost completely ionised to the He-like stage following the passage of the optical laser pulse and the electron temperature no more than about 30 eV. Processes which contribute to plasma heating include above threshold ionisation (ATI) energy and nonlinear inverse bremsstrahlung (IB) [1]. Under special conditions stimulated Raman scattering [2], space charge effects associated with the laser ponderomotive force [3] and other collective mechanisms may be important but are not considered here. The purpose of this work is to present a detailed time dependent simulation of the ionisation and heating of a neon plasma. Since these calculations do not include the contribution of stimulated processes to the heating, they only provide a lower limit to the electron temperature.

In the absence of focusing or defocusing effects, the interaction volume near the laser focus can be well approximated by a cylindrical plasma irradiated axially by the beam. For small focal spots and long pulses, radial thermal conduction plays an important role in determining the temperature and is accounted for in these calculations.

DESCRIPTION OF PHYSICAL MODEL

The most important ionisation process for short pulse high intensity laser interaction with gases is tunnel ionisation. This is a very nonlinear process for which the rate rises very rapidly above a threshold intensity which can be defined as the intensity for which the electric field depression of the atomic potential is equal to the binding energy of the electron [3,4]. Experiments [5] are in good agreement with the formula for the rate of tunnel ionization by Ammosov, Delone and Krainov [3,4] and this rate is used here.

The ATI heating can be attributed to the mismatch between the peak of, and the point in the laser cycle at which ionisation occurs [6]. The ATI energy is dependent on the laser wavelength (proportional to λ^2) as well as the intensity at which the ionisation occurs. For short pulse lasers ionisation occurs approximately at the threshold intensity (the exact value depends on the rise time of the pulse). The ATI energy is also strongly dependent on the polarisation of the laser field and varies from approximately $0.1 U_p$ for linearly polarisation to $2 U_p$ for circular polarisation.

The most important mechanism for heating undercritical plasmas with lasers is inverse bremsstrahlung or collisional absorption. The absorption is proportional to the electron-ion collision frequency and to the laser intensity. At high laser intensities the electron velocity distribution is severely modified as a result of the oscillatory motion in the laser electric field. Consequently the electron ion collision frequency is reduced and the absorption becomes nonlinear and decreases with intensity [7-8]. This is accounted for in our model using a high field correction term to the standard inverse bremsstrahlung absorption coefficient.

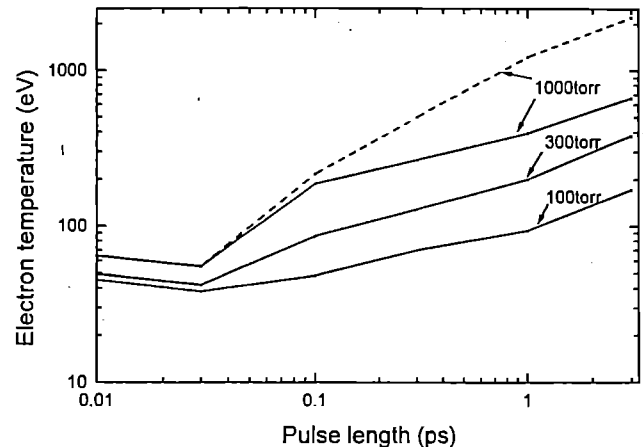


Figure 1: Electron temperature on axis of a neon cylindrical target irradiated axially with a $5 \mu\text{m}$ radius, $0.268 \mu\text{m}$ wavelength, $4 \times 10^{17} \text{ W/cm}^2$ peak intensity Gaussian (radially and temporally) laser pulse at the end of the laser pulse (taken as 3 times the FWHM past the peak of the pulse) as a function of pulse length (FWHM). Results of calculations using the non-local heat conduction model for 100, 300 and 1000 torr (full curves) are shown. A calculation for 1000 torr without any heat conduction is also shown (dashed curve).

For small laser focal spots and long pulses, thermal conduction cooling plays an important role in determining the electron temperature. When the temperature gradient is so steep that its scalelength becomes smaller than the average electron mean free path or when the electron distribution deviates significantly from a Maxwellian, the heat flux as calculated from the diffusion approximation, might exceed its physical upper limit, the free streaming limit. A common remedy is to simply impose an upper limit on the local heat flux in order to avoid such non-physical behaviour. A heat conduction formulation which does not rely on a flux limiter and which also accounts for non-local effects is the convolution integral [9-11] formula. This reduced to the diffusion approximation when temperature gradient scalelengths are much larger than the electron mean free path. We have adapted this procedure to cylindrical geometry and included it in our model.

APPLICATION TO NEON GAS TARGET

The temperature of a cylindrical gas irradiated with a laser pulse (gaussian in time and in the radial direction) can now be calculated by including the above physical phenomena in 1 D hydrodynamics code. The electron densities envisaged for transient xuv lasers are of the order of 10^{20} cm^{-3} (about 400 torr neon gas pressure). The effect of changing the density and pulse length on the electron temperature at the end of the laser pulse (defined as 3 times the FWHM past the peak) is shown in Fig. 1. As the density increases the heating contribution from IB heating increases and results in a higher temperature at the end of the laser pulse. As the pulse length decreases the contribution from IB becomes negligible and only ATI contributes to the heating. The slight increase in temperature for the shortest pulse length is due a small increase in ionisation intensity. A focal spot radius of $5 \mu\text{m}$ is used in these calculations and the effect

of heat conduction can also be seen in Fig. 1 where the results for 1000 torr neon without heat conduction are also shown. As expected, for sub-picosecond pulses heat conduction is not effective on time scales of the order of the laser pulse length.

The time dependent temperature as calculated by our model for a 200 fs laser pulse at an intensity of 10^{18} W/cm², a wavelength of 0.248 μ m, in a Ne⁸⁺ plasma at an electron density of 10^{20} cm⁻³ is shown in Fig. 2. The temperature at the end of the pulse is about 150 eV with a contribution from ATI of approximately 25 eV and the rest from inverse bremsstrahlung. This is in better agreement with Ref. [12] where a temperature of 200 eV was calculated, than with Ref [3] where inverse bremsstrahlung was found negligible and an ATI temperature of 10 eV was calculated. Our calculated temperature is too high for the transient XUV laser scheme in Li-like neon proposed in Ref. [2], which requires a temperature < 30 eV. Although this temperature might be achievable for pulse lengths < 50 fs (see Fig. 1), no such pulses with sufficient energy to ionise the plasma has yet been produced. For the longer pulses available (> 100fs) additional cooling by heat conduction can be used to achieve inversion between excited states for quasi steady state recombination schemes. The rate of cooling for a cylinder with temperature T_e , electron density N_e and radius r immersed in a cold background is approximately proportional to $T_e^{5/2}/(N_e r^2)$ and can be much higher than the adiabatic expansion cooling rate for a cylindrical plasma expanding into vacuum which is approximately proportional to $T_e^{1/2}/r$ [1]. Provided the difficulties associated with the production of long filaments of ionised plasma are resolved, it should be possible to control the rate of cooling by varying the radius of the filament. For small radii the cooling occurs on a time scale much smaller than the hydrodynamic time scale for expansion (> 10 ps). This cooling by a supersonic heat wave is favourable for high gain at high electron densities and low temperatures. The dependence of the time evolution of the electron temperature on filament radius for the 200 fs case discussed above is shown in Fig. 3. The dependence of the cooling rate on radius is very strong at early times but diminishes at later times as the heat front progresses to larger radii, in agreement with the simple scaling.

CONCLUSION AND DISCUSSION

The temperature at the focus of a short pulse intense laser beam irradiating a neon gas target is calculated using a hydrodynamic model in cylindrical geometry. Tunnel ionisation, ATI heating, inverse bremsstrahlung, radial thermal conduction and hydrodynamic expansion are included. The application of the model to short pulses and high densities relevant to the Li-like neon transient XUV scheme shows that for pulse lengths greater than 100 fs, inverse bremsstrahlung heating becomes much more important relative to ATI. A calculation for a 200 fs laser pulse at an intensity of 10^{18} W/cm², a wavelength of 0.248 μ m, in a Ne⁸⁺ plasma at an electron density of 10^{20} cm⁻³, gives a temperature of 150 eV at the end of the pulse, in reasonable agreement with Ref [12]. The ATI contribution is only 25 eV and the rest is from IB. The contribution from IB can be minimised by using shorter pulses with the shortest pulse being determined by the requirement that there is enough energy in the pulse to ionise the plasma to the required state. The ATI contribution could also be minimised by removing the outermost electrons using a low intensity prepulse to preform a plasma channel which is allowed to expand and cool and then using a short intense pulse to ionise a filament to the final desired ionisation state. At this stage it might be advantageous to use circularly polarised light so that the electrons ionised by the short pulse acquire a very high ATI energy and will not interfere with the recombination process on a transient recombination time scale (~ 1 ps).

It must be stressed that at higher densities and laser intensities heating associated with stimulated Raman scattering can become much more important than ATI or IB. The model should then be modified in order to be applicable in such situations.

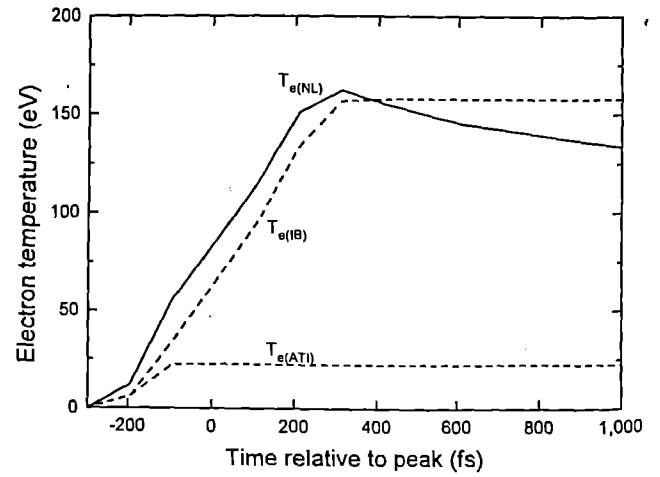


Figure 2: Contributions to electron temperature in a He-like neon cylindrical target from ATI ($T_{e(ATI)}$), from IB ($T_{e(IB)}$) and the resulting temperature given by the non-local heat conduction model ($T_{e(NL)}$) as a function of time relative to peak of intensity. The electron density is 10^{20} cm⁻³ and the target is irradiated by a 5 μ m radius, 0.248 μ m wavelength, 200 fs FWHM, 10^{18} W/cm² peak intensity Gaussian (radially and temporally) laser pulse.

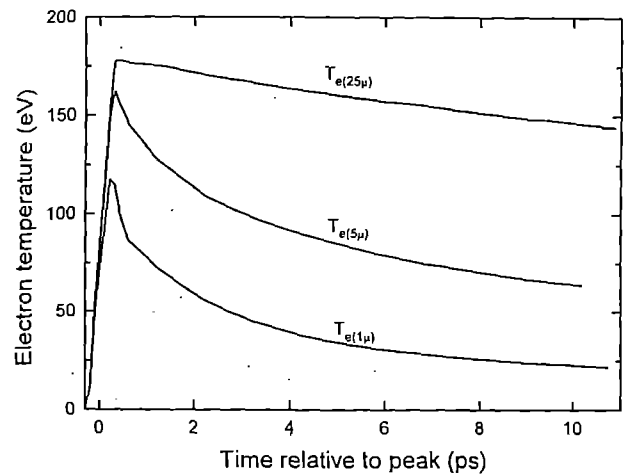


Figure 3: Electron temperature in a He-like neon cylindrical filament as given by the non-local heat conduction model as a function of time relative to peak of intensity. The electron density is 10^{20} cm⁻³ and the target is irradiated by a 0.248 μ m wavelength, 200 fs FWHM, 10^{18} W/cm² peak intensity Gaussian (radially and temporally) laser pulse. Curves corresponding to 1 μ m, 5 μ m and 25 μ m radius are shown.

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CALCULATION OF THE EFFECTS OF VELOCITY GRADIENTS AND OPACITY ON LINE TRANSFER IN LASER PRODUCED PLASMAS.

A. Djaoui¹, S. J. Rose^{1,2} and J. S. Wark³

¹Rutherford Appleton Laboratory, Chilton, Didcot, Oxon OX11 0QX, U.K.

²Department of Physics and Space Science, University of Birmingham, Birmingham B15 2TT, U.K.

³Department of Physics, Clarendon Laboratory, University of Oxford, Parks Road, Oxford, OX1 3PU, U.K.

INTRODUCTION

Recent experiments at the Rutherford Appleton Laboratory have made high resolution measurements of hydrogen-like resonance line emission from a nanosecond laser produced aluminium plasma as a function of angle¹ with respect to the normal to the target, in a quasi-1D planar situation. Typically, in these plasmas, the velocity gradients in the normal direction to the target surface exceed 10^9 s^{-1} and result in a bulk Doppler shift of about 12 mÅ which is not only larger than the Doppler width ($\sim 3 \text{ mÅ}$) of the Al $1s_{1/2}-2p_{1/2}$ and Al $1s_{1/2}-2p_{3/2}$ doublet but also their separation ($\sim 5 \text{ mÅ}$). Both components of the Ly- α doublet should therefore be considered and their interaction taken into account when calculating the emitted line shapes and intensities. In work reported here, a model which incorporates time-dependent atomic physics, opacity and velocity gradient effects is presented. A multifrequency radiative transfer algorithm is used for the calculation of detailed resonance line profiles. The inclusion of velocity gradients is found to be critical in determining the intensity and shape of emitted spectral lines.

HYDRODYNAMICS AND IONIZATION KINETICS CALCULATION

The numerical model used to simulate the experiments is based on the 1D Lagrangian hydrodynamics code MEDUSA². The model follows the excitation and ionisation in each Lagrangian cell by using the time-dependent non local thermodynamic equilibrium (non-LTE) average-atom model³. Energies involved in atomic processes are included in the free electron energy balance equation which is iterated with the rate equations to ensure self consistency. The plasma is assumed to be optically thin to continuum radiation and to most line radiation. The effect of reabsorption in optically thick resonance lines on state populations is accounted for only approximately using escape probabilities as multiplicative factors to the spontaneous radiative rates in the average atom model. In the presence of large velocity gradients significant absorption occurs only over a distance for which the Doppler shift is less than the line width. The escape probability for the case of a large velocity gradient is therefore larger than in the static case. This mechanism is an important contributor to the successful operation of recombination-pumped XUV lasers⁴ because reabsorption of resonance line photons pumps the lower laser level reducing the population inversion. In the presence of large velocity gradients escape probabilities are in good agreement with detailed line transfer calculations⁵⁻⁶.

LINE TRANSFER CALCULATION.

Given the temperatures, densities, velocities and ionic populations from the calculation described in the previous section, the emitted spectrum of a selected number of lines can be calculated. This line transfer calculation post-processes the output from the hydrodynamic/ionization simulation. The fact that the absorption profiles of ions that absorb are Doppler-shifted relative to the emission profile of the emitting ions because of the velocity difference between them, is taken into account. In this work we concentrate on the

aluminium Ly- α doublet in order to illustrate the model. The number density of state $2p_{1/2}$ and $2p_{3/2}$ in Lagrangian cell m are calculated from the time-dependent shell populations in the average-atom model using the method described in Ref. 3. This method predicts the total density in the ground state ($1s_{1/2}$) and in the $n=2$ states ($2s_{1/2}$, $2p_{1/2}$ and $2p_{3/2}$). The population in $2p_{1/2}$ and $2p_{3/2}$ is then calculated by assuming $n=2$ states are populated according to their degeneracies. Knowing the excited state populations, the intensity of photons which are born in cell m with frequency ν that emerge at the boundary is then calculated. Within a cell in which the photons originate the radiative transfer method does not allow interaction between the components of the doublet. This is a reasonable approximation within a cell where the material is moving with approximately the same velocity. The photons from cell m are transported to the edge of the plasma through successive cells l taking into account their relative velocity ($V^l - V^m$). The emerging flux at the boundary is then corrected for the Doppler shift between the observer and cell m . The observed frequency is given by

$$\nu_{\text{observe } r} = \nu \left(1 + \frac{V^m}{c} \cos \theta \right)$$

where V^m is the velocity of cell m and θ the angle of observation with respect to the laser beam and c the speed of light in vacuum. The above Galilean transformation is only valid for non relativistic flows. To compare with experimental measurements, the contributions from all cells m are finally added. Typically 100 frequency points are used and thousands of timesteps are needed in order to calculate time integrated spectra.

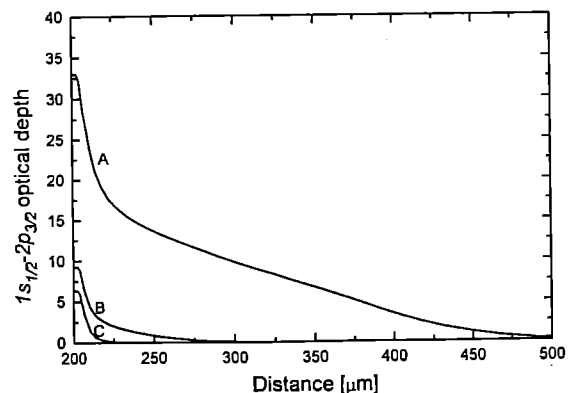


Fig. 1: Optical thickness of a photon born at the peak of the $1s_{1/2}-2p_{3/2}$ Al Ly- α component as a function of position in the plasma. Curve A is for a static plasma while curve B includes the effect of bulk Doppler shifts resulting from plasma flow. Curve C does not include the contribution from $1s_{1/2}-2p_{1/2}$ contribution while curve B does.

Ly- α EMISSION FROM NANOSECOND LASER PRODUCED PLASMA.

As an application of this model, we present calculations of the time integrated Al Ly- α emission from a plasma produced by a beam of 0.53 μm light in a 0.6 ns full width at half maximum (FWHM) having a peak intensity of $10^{14} \text{ W cm}^{-2}$. The laser pulse is incident normally onto a 10 μm thick, 500 μm diameter foil. Figure 1 shows the peak optical depth of the $1s_{1/2}-2p_{3/2}$ component in the frame of the emission cell, as seen by an escaping photon emitted at different positions in the plasma at the time of the peak of the laser beam. Curve A is obtained assuming a static plasma, while curve B includes the effect of velocity gradients. Doppler decoupling results in a reduction of opacity by at least a factor of 4. Line interaction is non negligible due to some overlap of the components but also because high energy photons from the $1s_{1/2}-2p_{3/2}$ component from the region of peak emission appear red shifted to the outer expanding plasma and are absorbed there by the $1s_{1/2}-2p_{1/2}$ component. The absorption by the $1s_{1/2}-2p_{1/2}$ is included in curve B but not in curve C.

We next investigate the effect of trapping in the hydrodynamic/excitation/ionisation calculation on line emission. The effect of including trapping in the calculation of ionic population on the emitted Ly- α line profile is shown in fig. 2. The line intensity is strongly dependent on the amount of trapping which transfers ground state populations to the $n=2$ excited state populations and results in a higher intensity. Thus absolute line intensity measurements can provide valuable information on the radiative transfer problem.

The effect of opacity, velocity gradients and their combined effect can be studied using the radiative transfer algorithm. This is illustrated in Fig.3. Curve A is the result of a calculation where the plasma is assumed static (all flow velocities set to zero) and optically thin. The line shows two peaks corresponding to the $1s_{1/2}-2p_{3/2}$ and $1s_{1/2}-2p_{1/2}$ components. Curve B assumes an optically thin plasma but includes the effect of the velocity field. The effect of velocity gradients is to blue shift and smear out the whole spectrum since the emitting plasma is moving towards the observer. Curve C assumes a static plasma but includes opacity only. In addition to a reduction in intensity, the line shape is completely different and shows a reversal at the $1s_{1/2}-2p_{3/2}$ and $1s_{1/2}-2p_{1/2}$ at the peak positions in the opacity. Curve D includes both opacity and velocity effects. Its shape is intermediate between curve A and B and bears no resemblance to curve C while its intensity is more nearly that of curve C. This shows that the line shape is mainly determined by the velocity profile while the intensity is dominated by the optical thickness between the point of emission and the plasma boundary as well as by excited level populations which depend strongly on the probability of escape (see Fig. 2).

CONCLUSION

We have developed a two stage 1D planar model for calculating line transfer in laser produced plasmas where temperature, density and velocity gradients, as well as the effect of optically thick line reabsorption on level populations, are taken into account. In the first stage the hydrodynamics and the ionic populations are solved in a self-consistent manner. The atomic physics of excitation and ionization is calculated using a time dependent non-LTE average atom model where resonance photon trapping in hydrogen and helium-like ions is treated approximately using escape probabilities. A detailed line transfer algorithm which takes the output from the hydrodynamic/ionization calculation is used in the second stage to calculate line profiles for hydrogen and helium-like resonance lines in aluminium. In its present form this is already a powerful tool for interpreting and understanding radiative transfer in laser produced plasma. Simulation of time integrated Ly- α emission as a function of angle in a long pulse (nanosecond) experiment show that velocity gradients are critical in determining the line shapes and intensities.

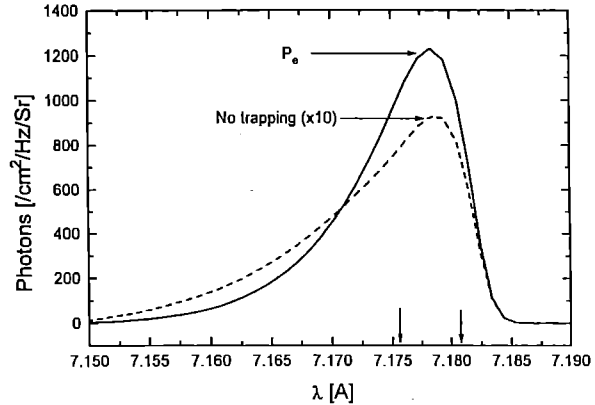


Fig. 2: Al Ly- α front emission at 30 degrees with respect to the laser beam as calculated from ion densities obtained with the use of escape probabilities and with no trapping (ie escape probability = 1). The dotted curve has been multiplied by the factors shown in parentheses. The arrows on the wavelength axis indicate the position of the peaks of the doublet for a static plasma.

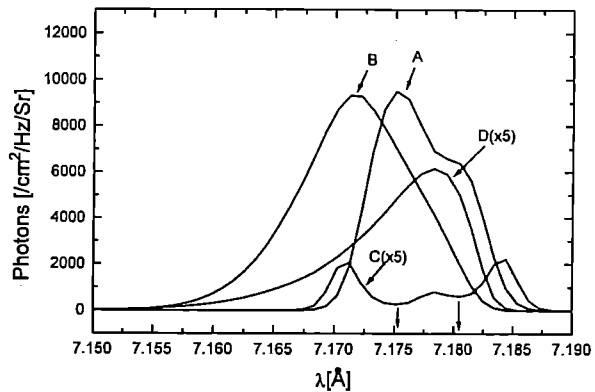


Fig. 3: Al Ly- α profile as calculated by the line transfer algorithm at 30 degrees with respect to the laser beam. Curve A assumes a static and optically thin plasma. Curve B assumes an optically thin plasma but includes velocity effects. Curve C assumes a static plasma but includes absorption. Curve D includes the effect of velocity fields as well as absorption. The last two curves have been multiplied by the factors shown in parentheses. The arrows on the wavelength axis indicate the position of the peaks of the doublet for a static plasma.

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INTRODUCTION

In a previous report¹ a time dependent model for ionisation balance and atomic populations in a laser plasma was described. Typical simulation results were presented for irradiation of aluminium targets with 12ps laser pulses from the SPRITE Raman amplified KrF laser, with incident irradiances of a few times 10^{15} Wcm⁻². In this report the model is used to compare actual experimental data to simulation.

In the experiment two types of target were used. These were solid aluminium slabs and targets consisting of a thin layer of aluminium coated onto a Mylar substrate and overcoated with a layer of CH. The laser was focused with approximately 50µm spot size and both time integrated and time resolved data were taken.

ATOMIC MODEL

The basic atomic model used is the same as described in the previous annual report¹. However, in the radiative transfer algorithm, line trapping is extended to include the Ly α (Al XIII 1s-2p), Ly β (Al XIII 1s-3p), He α (Al XII 1s²-1s2p 1P) and He β (Al XII 1s²-1s3p) lines as well as two Li-like satellite lines to the He α line which have strong radiative transitions to the Li-like ion ground state. These are the 1s(2s2p³P) 2P and 1s(2p²3P) 4P levels. In addition, a version using Sobolev escape factors was also used and found to give good agreement with the line transfer model, due to the very large velocity gradients present in the plasma.

HYDRO-DYNAMIC MODELLING

As before, the MED101² version of the MEDUSA code was used. For the data presented here, an outer layer of 1.5 µm was modelled with 50 cells for the solid targets and 16 cells were used to model the 0.1µm Al in the layered targets. Figure 1 shows the typical temperature and velocity profiles at the peak of the pulse for an irradiance of 3×10^{15} Wcm⁻².

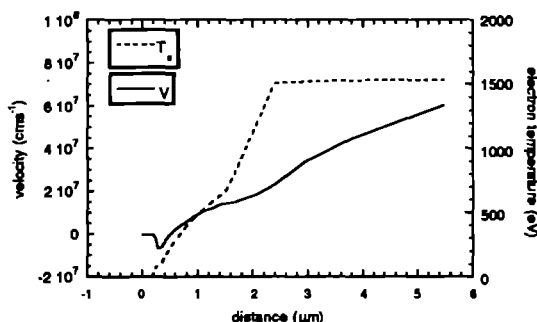


Figure 1. electron temperature and expansion velocity at the peak of the laser pulse for irradiance of 3×10^{15} Wcm⁻².

It can be seen that the velocity gradient is approximately 10^{11} s⁻¹. Figure 2 shows the predicted ratio of the He β to Ly β line for both radiative transfer and Sobolev escape factors, for two cases at irradiance of 4×10^{15} Wcm⁻². In the first case (upper set of lines) the trapping on the β lines is omitted. In the second case, trapping on these lines is included. We can see two things from this figure. Firstly, the Sobolev escape factors give very good agreement with the line transfer algorithm. Secondly, it is not clear that including

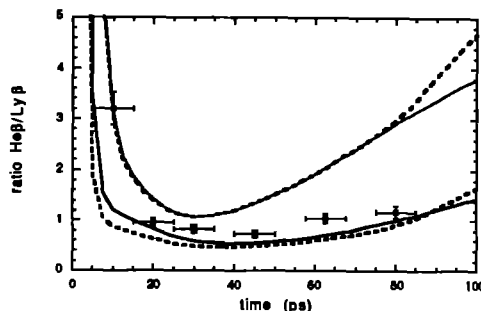


Figure 2. Solid line is radiative transfer. dotted line is Sobolev escape factors. In the upper pair of lines the trapping of the β lines is omitted.

the β lines improves the modelling as the data lies in between the cases were they are either left in or out of the calculation. At early time it seems better to not include them, but at later time the agreement is better. Presumably, this is a result of the changing width of the lines. The 1s-3p lines do not have an unshifted central Stark component and are quite broad and so a poor approximation to the Doppler shape assumed in the escape factors or the Voigt profile of the line transfer. The 1s-2p lines on the other hand have a narrow unshifted central component only subject to electron and Doppler broadening and so are likely to be a better approximation. It should be noted however, that neither method of treating line transport accounts for interaction of lines, in particular in this case the Ly α -intercombination line separation is such that Doppler shifting ought to allow photons from either one of these transitions to be absorbed by the other.

DENSITY MEASUREMENTS

The electron density was estimated from the width of the He β and Ly β emission lines. At the expected densities of about 10^{23} cm⁻³ these have considerable width above the instrument spectral resolution which varied according to the particular transition but was typically $\lambda/\Delta\lambda \sim 300$. In order to predict the line width, the hydro-dynamic data from MEDUSA was processed with the time dependent ionisation code to produce the electron density and upper state population in each cell as a function of time. By assuming that the broadened Stark profiles are optically thin (considered reasonable at least at early time), the line shape codes of R.W. Lee³ were then used to add together individual line shapes from the 50 cells, each scaled according to cell size and upper state population and including a Doppler shift due to expansion velocity. The instrument width of 10ps was also folded into the predicted line shape history, as it was for all the simulations presented. Figure 3 shows a comparison of predicted FWHM and experimental data for the He β line. We can see that, by assuming the β lines to be optically thin, good agreement is achieved. The comparison with β lines trapped is not good. It should be noted that the latter comparison is made by assuming trapping in the relative emission from each cell, but not in the actual profiles. Optical depth effects were not included in the code version used, and since trapping is local, the effect of opacity on Stark broadened profiles would not be expected to be great.

Figure 4 shows the He β /Ly β line ratio for this same shot, indicating again that at early time the assumption of no trapping is probably better than including trapping with a narrow, Doppler broadened profile for the observed lines.

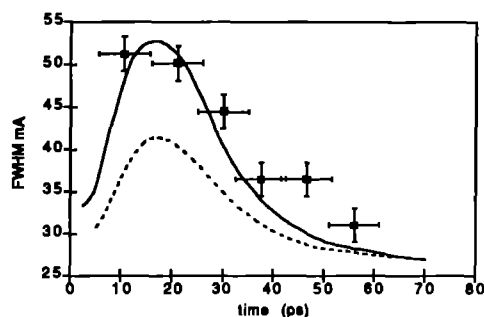


Figure 3 Comparison of predicted and measured line width for the Heβ line. Incident irradiance is $3 \times 10^{15} \text{ Wcm}^{-2}$ on a solid aluminium slab.

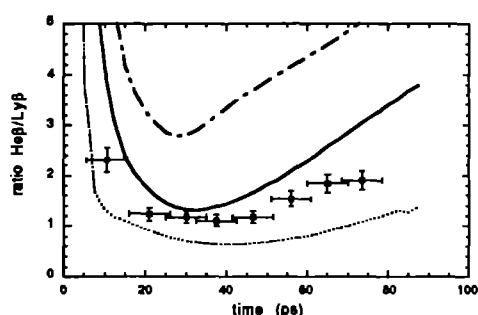


Figure 4 Heβ/Lyβ ratio for the same shot indicated in figure 3. The dash-dot line is for no line trapping, the solid line for trapping on Li-like satellites, Heα and Lyα only. The dashed line is for trapping of Lyβ and Heβ included.

The local nature of the line trapping is emphasised by considering a layered target. Figure 5 shows the predicted line ratio compared to experiment for a shot at $6 \times 10^{15} \text{ Wcm}^{-2}$ on a target consisting of a $0.1 \mu\text{m}$ Al layer coated onto a $10 \mu\text{m}$ thick

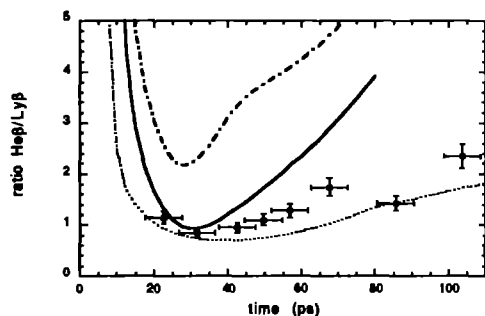


Figure 5 line ratio measured for a layer target compared to simulation for no trapping (dot-dash line), trapping on Li-like satellites, Lyα and Heα (solid line) and trapping on Lyβ and Heβ also included (dashed line)

Mylar substrate and overcoated with $0.1 \mu\text{m}$ of CH plastic. It can be clearly seen that the effects of line trapping are very similar to those for the slab targets

TIME INTEGRATED DATA

Figure 6 shows a simulated time integrated Heβ line shape for the slab target shot at $3 \times 10^{15} \text{ Wcm}^{-2}$. The simulation assumed that this line is optically thin and the spectral resolution is crystal limited to $\lambda/8\lambda \sim 500$. The width of 20.6 mÅ is similar to the experimentally observed value⁴. It should be noted that this data was taken on film rather than through the streak camera, which contributes quite a significant amount of the total instrument broadening in the time resolved data⁵. By adding the time resolved data a width of some 29 mÅ is obtained. Considering the poor response of the film at low intensity, this compares well to the predicted width of 27 mÅ assuming a resolution of about 300.

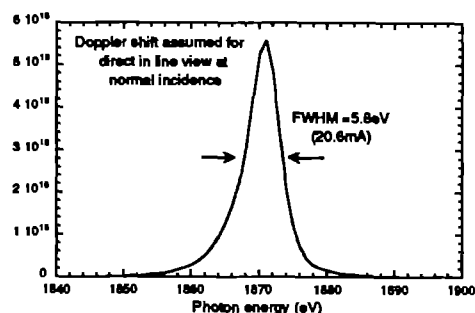


Figure 6 Simulated time integrated line shape for Heβ transition

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MODELLING OF RESONANCE LINE TRANSITIONS OBSERVED IN GERMANIUM SOFT X-RAY LASER PLASMA AMPLIFIERS

E. Robertson¹, A. E. Kingston¹, C.L.S Lewis², G Cairns², G. Pert³, S. Healy³.

¹Department of Applied Mathematics and Theoretical Physics, David Bates Building, Queens University, Belfast BT7 1NN, Northern Ireland.

²Department of Pure and Applied Physics, Queens University, Belfast BT7 1NN, Northern Ireland.

³Department of Physics, University of York, Heslington, York YO1 5DD, England.

Recent work by Pert and others in the theoretical simulation of Ne-like collisionally pumped laser schemes has resulted in the successful development of the code EHYBRID. The resulting agreement between experimentally measured values of the gain characteristics and those predicted using the code is excellent in virtually every respect. The following report describes work in which this code has been extended to study the resonance emissions which have been observed from x-ray laser plasmas.

Time resolved spectroscopic studies of the Ne-like resonance lines from germanium slab targets had shown significant measurements of F-like resonance emission. The significance of the F-like presence in the plasma was underlined by the fact that large values of measured gain were only observed in plasmas where F-like resonance emission also appeared. The ratio between the intensities of the F and Ne-like lines was used by experimentalists as an additional qualitative measurement of the plasma being studied.

Though it was possible to simulate the Ne-like emission, to obtain a measure of the F-like emission required further development to the atomic physics of the code. Using collision rates calculated by Zhang and Sampson and energy levels and oscillator strengths calculated at Belfast the code was adapted to include the atomic kinetics of the first 112 excited levels of the F-like ion with a collisional-radiative treatment. This was in addition to the 124 excited levels of the Ne-like ion already present in the model. An energy level diagram indicating the relevant transitions of interest is shown in Fig1.

The code divides the target into a series of Lagrangian cells the number and dimensions of which are defined at the start of each run. In each cell at specified time steps the populations of the excited levels of both F- and Ne-like ions are calculated. Using the populations of the upper levels of the resonance line along with their A-values the intensities of the radiative transitions can be calculated. The full effects radiative transfer is not included in the model. Opacity effect are partially included through a calculation of the Holstein trapping factor to modify the the radiative rate to account for reabsorption.

Preliminary results from the extended model provide good agreement with experiment. In Fig 2 the time resolved experimentally measured intensity profile of the Ne-like $3s(^1P_1)-2s(^1S_0)$ resonance transition and the combined line intensity measurements of the $3s-2p$ F-like array are compared with the simulated profiles for a shot taken with a driving laser pulse of 1.1ns and intensity $\sim 1.4 \times 10^{13} \text{ Wcm}^{-2}$. The experimental measurements are relative intensities and for comparison results were normalised to the peak value of the Ne-like line.

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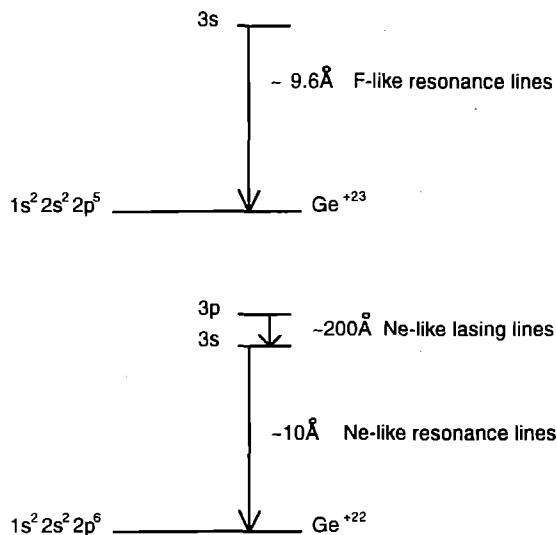


Fig 1 Schematic Energy Level Diagram of F and Ne-like Resonance Lines

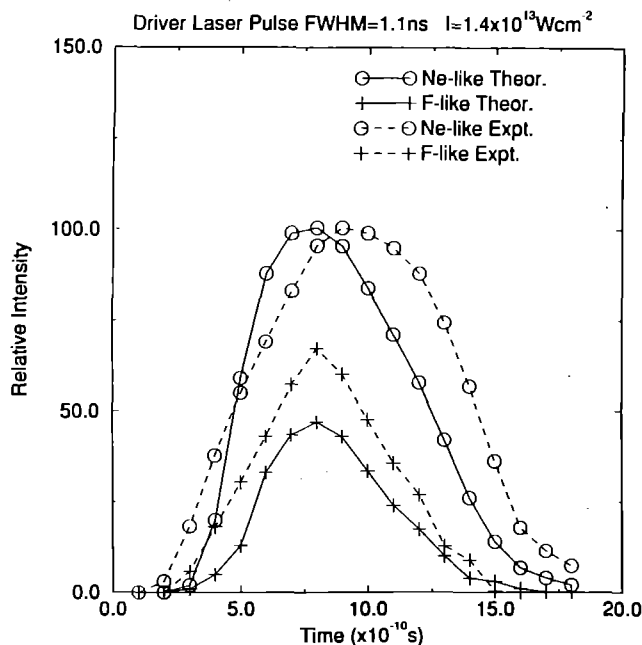


Fig 2 Time Resolved Intensity Profile

FOKKER-PLANCK CALCULATIONS WITH IONISATION DYNAMICS OF SHORT PULSE LASER PLASMA EXPERIMENTS

R P J Town¹, A R Bell¹ and S J Rose²

¹Imperial College of Science, Technology and Medicine

²Rutherford Appleton Laboratory

The availability of subpicosecond, high intensity lasers has generated considerable interest in the laser-plasma community [1,2]. An initially cold, solid density, un-ionised material is rapidly heated by the short-pulse laser to a plasma that is close to solid density at a temperature of a few hundred eV. The brevity of the laser pulse leads to density and temperature scalelengths that are typically of the same order as the mean free path of the thermal electrons. We have previously reported [3] that this scaling leads to a strongly non-Maxwellian electron distribution. We also showed that the Spitzer-Härm theory [4] of thermal conduction is inapplicable under such extreme conditions.

In a previous paper [5] we reported simulations using our one dimensional Fokker-Planck (FP) code and an ionisation dynamics model. The non-LTE ionisation material package (NIMP) [6] was used to post process the density and temperature predicted by the FP code to obtain the ionisation state as a function of time. It was found that to achieve full ionisation in the strongly heated region of the Carbon target took the rise time of the laser pulse (typically 0.5 ps). Further into the target the Carbon never reached the fully ionised state. We also examined the relative importance of the various ionisation and recombination processes for the low Z materials considered. The absence of radiative processes did not alter the time evolution of the ionisation state. Collisional excitation and de-excitation did not alter the final ionisation state achieved, but led to a faster ionisation rate. We concluded that to a good approximation the dominant mechanisms were collisional ionisation and its inverse process three body recombination.

In last year's report [7] we investigated the effects of the non-Maxwellian distribution on the collisional ionisation rates. This was performed by multiplying the collisional ionisation cross sections, taken from work performed at Belfast [8], by the actual distribution function and integrating over all velocities then comparing the resulting rates with the rates obtained by using a Maxwellian distribution function of the same density and temperature. We found that the difference between the rates calculated using the real distribution and the Maxwellian rates was quite small. We concluded that this justified the continued use of ionisation rates based on a Maxwellian distribution in simulations of experiments.

We have recently coupled a non-LTE ionisation model self consistently to a FP treatment of thermal conduction [9]. We can thus model the distortion of the electron velocity distribution by the dominant ionisation processes and examine the effect of this distortion on the heat flow.

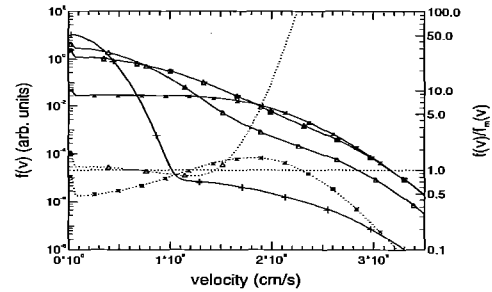


Figure 1: The electron distribution (solid line) and the ratio of the actual distribution to a Maxwellian (dotted line) at the peak of the pulse for certain Lagrangian cells in the carbon target. The initial cell positions relative to the surface on which the laser beam was incident are 0.0 μm (x), 0.05 μm (*), 0.1 μm (Δ), and 0.2 μm (+).

As already reported the Belfast group [8] has derived the collisional ionisation cross section in terms of the incident electron velocity. However, we need to know the division of energy between the ionising and newly ionised electron. We have made the approximation that the newly ionised electron is placed in the lowest velocity group. The electron that caused the ionisation just loses the ionisation energy. We also need the three body recombination cross sections. We obtain these in terms of the collisional ionisation cross sections by employing the law of detailed balance for a partially ionised plasma in equilibrium.

We shall consider the interaction of a 0.3 ps Gaussian KrF ($\lambda = 0.25 \mu\text{m}$) laser pulse on a 0.4 μm thick Carbon target that was initially at 20eV. The initial distribution of ionisation states was such that the ionisation and recombination processes balanced and corresponded to a Z of 1.49.

Figure 1 shows the electron distribution function and the ratio of the distribution function to a Maxwellian of the same density and temperature for certain Lagrangian cells in the Carbon target at the peak of the pulse (0.3ps). The cell closest to the laser, located at 0.0 μm and denoted by x, shows the effect of laser energy deposition, thermal transport and ionisation. The laser energy is taken up by the low velocity, collisional electrons and thus we see the well-known Langdon reduction of the low velocity electrons and excess at high velocities compared to a Maxwellian of the same density and temperature. At very low velocities there is an increase in the number of electrons (although this

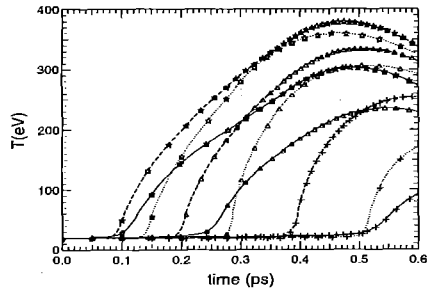


Figure 2: The time evolution of the temperature for certain Lagrangian cells in the carbon target. The initial cell positions relative to the surface on which the laser beam was incident are 0.0 μm (X), 0.05 μm (*), 0.1 μm (Δ), and 0.2 μm (+). The solid line refers to the variable ionisation case, the dashed line a fixed $z=3.75$, and the dotted line to fully ionised simulations.

is still below the Maxwellian). This blip is due to newly ionised electrons, which are placed in the lowest velocity bin.

At 0.3ps the main heat front has penetrated approximately 0.1 μm into the target. The effect of the arrival of the main heat front on the distribution function for this cell is shown in figure 1 (denoted by Δ). The distortion of the distribution function is dominated by the ionisation process. There is an increase in the electrons at low velocity. This increase is due to the thermalisation of the newly ionised electrons and a reduction in the energy of the ionising electrons. At higher velocity there is a depletion of electrons compared to a Maxwellian. This velocity region is where the bulk of the electrons ionising the plasma are taken from. At even higher velocities there is again an excess of electrons compared to a Maxwellian. These are the long free path electron penetrating from the heated region. These electrons are more clearly seen on the 0.2 μm curve, denoted by a +. At the time this snap shot of the distribution was taken the main heat front had not arrived and the distribution is dominated by the cold, almost Maxwellian distribution and the high velocity electrons that have the "temperature" of the heated region from where they have penetrated.

We have clearly seen that the inclusion of ionisation dynamics and a FP treatment of heat flow substantially modifies the distribution function. We now examine the effect that ionisation dynamic has on the penetration of the heat front into the target. Figure 2 shows the time history of the temperature for the Lagrangian cells located at 0.05 μm , 0.1 μm , and 0.2 μm .

There are two main factors that may lead to a modified heat front penetration. Firstly, the low ionisation state of the material allows electrons to penetrate further into the target. This would enhance the heat flow. Secondly the energy expended in reaching an ionisation state would inhibit the heat flow. These two competing effects lead to a time varying inhibition in the heat flow compared to fixed ionisation simulations. To isolate the effect of ionisation

dynamics on the heat front penetration figure 2 shows three different simulations with different ionisation states. The solid curve represents the evolution of the Lagrangian cells using the FP code coupled self consistently to the ionisation dynamics model. The ionisation state varies as a function of space and time throughout the simulation. The dashed curve refers to a simulation with a fixed ionisation state of 3.75 throughout the target. The dotted curve refers to a simulation with a fixed fully ionised state throughout the target. All simulations had the same energy deposited in the target to within 10%.

For the Lagrangian cell at 0.05 μm the temperature rapidly and abruptly increases for the $Z=3.75$ case at 0.09ps, while there is a more gradual increase for the variable ionisation case. The gradual increase in temperature is a signature of the penetration of the long mean free path electrons from the heated region. It is more evident in the variable ionisation simulation, because the unheated cell is initially in a lower ionisation state that enables the high velocity electrons to penetrate further. The fully ionised simulation arrives last. This slow arrival of the main heat front is due to the higher Z compared to the other simulation. Note that the variable ionisation simulation temperature increase slows at later times as energy is expended in increasing the ionisation state. At 0.1 μm into the target the same pattern emerges. First the $Z=3.75$ heat front arrives, followed by the gradual build up in temperature of the variable ionisation case. Finally the fully ionised simulation's heat front arrives. Again at later times the variable ionisation simulation has a lower temperature. Even further into the target the variable ionisation simulation's heat front arrives after both fixed ionisation simulations' heat front. Finally the energy expended in ionisation has slowed down the heat front penetration.

In conclusion, we have reported results using our FP code coupled self consistently to an ionisation dynamics model. We have shown that the ionisation process substantially alters the electron velocity distribution. This led to a time varying inhibition of the heat flow compared to fixed ionisation simulations.

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SUPER-INTENSE FIELD PHYSICS: WHAT IS CLASSICAL AND WHAT IS QUANTUM ?

J. B. Watson,¹ C. H. Keitel,² P. L. Knight² and K. Burnett¹

¹ Clarendon Laboratory, Department of Physics, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom.

² Blackett Laboratory, Optics Section, Imperial College, Prince Consort Road, London SW7 2BZ, United Kingdom.

INTRODUCTION

It is well established that the interaction of atoms with super intense electromagnetic fields gives rise to the generation of high harmonics. This was predicted quite some time ago [1] and has since been extensively studied experimentally [2], and the topic of various review articles [3]. Many aspects of this interaction have been explained satisfactorily with purely classical models while others have required a quantum mechanical treatment of the atom. In the super-intense regime a semi-classical model of a tunneling process followed by a quiver-induced “re-collision” has been successful in interpreting high harmonic generation, above-threshold ionisation and the frequency cut-off observed in the harmonic plateau. The phenomenon of stabilization, i.e. the reduction of ionisation with increasing laser field intensity for intense and high frequency laser fields, has recently been observed experimentally. There has been much discussion on whether this phenomenon can be described by purely classical models [4], or whether a full quantum mechanical treatment [5] is required. In an attempt to address this question Bestle, Akulin and Schleich [6] have employed a phase space description of the stabilization process in which they concentrated on two dynamic regimes of the system: For a sudden turn-on of the field they predict classical stabilization while for an adiabatic turn-on the origin of the process is of a purely quantum nature.

METHODS

In this work we use quantum and classical phase space distributions to show that a realistically shaped laser pulse in fact displays both quantum and classical behaviour but at different times in the interaction with the pulse. In the quantum calculation we solve the time-dependent Schrödinger equation in one dimension with a soft-core potential to obtain the wavefunction at later times, we then use these time-dependent wavefunctions to calculate the Wigner function for the system. In the classical case we start with a distribution of points in phase-space corresponding to the classical orbit (1D) and then solve the classical equations of motion for each set of initial conditions. We find three distinct regimes during the pulse. Towards the end of the pulse we find remarkable similarities between the Wigner function and the classical phase distribution.

At the beginning of the pulse we find disagreement between the quantum and classical pictures, this is to be expected as the field is relatively small, and therefore the distributions are still dominated by the (widely different) initial conditions. Close to the peak of the pulse we find that there is total disagreement between the quantum and classical results.

In order to explain this behaviour we consider the Wigner functions of the corresponding dressed eigenstates, i.e. the eigenstates of the Kramers Henneberger potential. Pulsed excitation results in a superposition of KH eigenstates which interfere; this interference generates substantial regions in which the Wigner functions are negative, something which classical dynamics cannot replicate. A coherent superposition of a few quantum states can not be modelled simply with a classical distribution and as a consequence we have seen total disagreement between the quantum and classical models in this domain.

RESULTS

In Fig. 1 (a) and (b) we have displayed the Wigner function as a contour plot (a), at the end of the laser pulse which can be compared with the classical distribution in (b). We find at a glance that both are remarkably similar at this stage of the interaction. The size of the structure is larger by roughly $\hbar$ in the quantum mechanical case due to the uncertainty relation. The fringes, which can be identified with the ionizing part of the wave function, display small negativities in the quantum case but are quite similar in length, separation and orientation.

When the peak of the pulse is reached we still find that the distributions are totally different. We note that in the quantum mechanical distribution the fringe structure is greatly reduced, this corresponds to a reduction of the ionisation since the atom is in the stabilization regime. We also find two symmetrically arranged and strongly negative areas. It has been established in quantum optics that these negative areas are associated with purely quantum mechanical behaviour, in particular arising from the coherence among the components of superpositions of eigenstates of the system. We have confirmed this idea with the evaluation of the Wigner function of superpositions of eigenstates of the

corresponding Kramers Henneberger potential. These superpositions show quite clearly the same symmetrically arranged areas of negativity, a property which is quite well known already in the field of quantum optics [7].

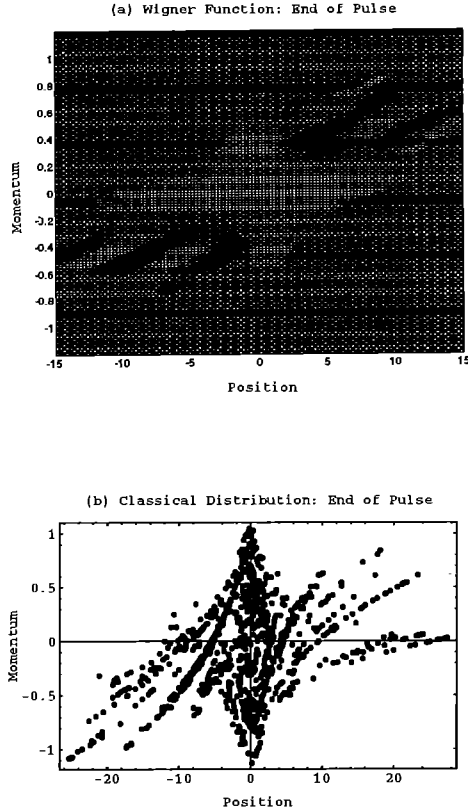


Figure 1 : (a) Contour of the Wigner function and (b) classical phase space distribution of the hydrogenic electron *immediately after* the application of a pulsed laser field with 12 turn-on cycles, 12 turn-off cycles, maximal electric field strength $E_0 = 10$ a.u. and angular frequency $\omega = 1$ a.u. The quantum mechanical calculations leading to (a) are based on the dynamics of the hydrogenic electron in the softcore potential as described by the corresponding Schrödinger equation and the distribution of dots in (b) arises from a Monte Carlo distributed set of 1000 initial conditions in space and momentum which have evolved in time via the corresponding classical Newton equation.

In conclusion we have demonstrated that early in the pulse, before the onset of stabilization, we observe fringes in the quantum distribution consistent with ionisation of the atom. At the peak of the pulse, where we expect stabilization to occur, coherent superpositions of eigenstates of the time-averaged KH potential are formed. This is a purely quantum mechanical effect and therefore it is clear that classical calculations

at this stage of the pulse will lead to incorrect results. At the end of the pulse the number of eigenstates involved in the superposition becomes increasingly large. As we leave the stabilization regime the decay between these states is no longer negligible, and as a consequence there are decoherence effects. This decoherence erases the quantum signatures. It is for this reason that the classical calculations are so successful for this part of the pulse.

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Stimulated Raman scattering from intense short-pulse laser plasmas

F.I. Gordon, H.C. Barr, T.J.M. Boyd, and S.J. Berwick

Department of Physics, University of Essex, Colchester, CO4 3SQ

I. INTRODUCTION

Stimulated Raman scattering (SRS) is a parametric instability in which an electromagnetic wave (the laser driver) (ω_0, k_0) decays into a scattered electromagnetic wave (ω_s, k_s) and an electron plasma wave (ω, k). Sub-picosecond-pulse laser development has led to interest in SRS in new regimes applicable, for example, to recombination X-ray lasers and to plasma accelerator schemes. The shortness and high intensity of the pulse give rise to new physics seen in recent experiments [Darrow et al(1992), Najmudin et al(1994)].

A model of SRS driven by short laser pulses has been developed which shows the possibility for absolute (i.e. temporal) growth of Raman scattering due to the inhomogeneities in the laser intensity and plasma density at the pulse boundary. The model is based on a pulse-frame analysis, where the boundaries of the pulse are stationary. This frame is Lorentz-transformed from the lab at the group velocity of the laser driver. The resonant SRS daughter waves for both back and forward scattering emerge from the trailing edge of the pulse; in the pulse frame their group velocities have the same direction and hence the instability is always convective according to the standard analysis [Rosenbluth(1972)]. However, the short pulse duration, high laser intensities, intensity inhomogeneities and relativistic effects all give rise to material differences in the Raman scattering process. High intensities at high density mean that Raman backscattering (SRBS) is in a strongly coupled regime where the linear dispersion characteristics of the daughter waves no longer hold. Inhomogeneities in density or intensity give rise to mode conversion within the pulse. This can be sufficient to complete a feedback loop which can sustain absolute growth in the pulse frame. Relativity places a limit on the driver strength as well as giving an effective density reduction and hence a quenching of the instability in strongly relativistic regimes. For this paper SRBS will be examined most closely.

The elements of the model are explained most simply for the case of a homogeneous preformed plasma through which a *rectangular* laser pulse propagates and within which the only inhomogeneities are confined to the pulse boundaries. Regions inside and outside the pulse are then easily analysed in terms of their (respective) dispersion relations. The scattered frequencies detected are thus distinct from those generated within the pulse [Darrow et al(1992)], but there is continuity of phase across the pulse boundary. The boundaries cause finite reflectivity of the waves (more properly a mode conversion since it is not simply a case of reversing wavenumbers). Although in most cases this is at a low level, it is sufficient to close a feedback loop between counterpropagating waves within the pulse. In the pulse frame, since the problem is a static one, all frequencies are unchanged with only the wavelengths altered. Viewed from the laboratory frame the moving pulse boundaries cause frequency upconversion for

the wave reflected from the trailing edge of the pulse and an equal downshift on conversion at the leading edge. Therefore, there is some feedback from trailing edge to leading edge due to a right propagating light wave. The amount of conversion is small but sufficient to sustain absolute growth.

II. MODEL

The relativistic 1D Raman equations for a linearly polarised driver are routinely obtained from the relativistic fluid equations for warm plasma electrons. The ions are assumed to be stationary and homogeneous. Use of a circularly polarised laser driver avoids the complication of longitudinal electrostatic effects which arise in the case of linearly polarised light. However, since most experiments are carried out using linearly polarised pulses, we take that as the polarisation in our model, but it will be seen that circular polarisation gives a similar dispersion relation. The derivation of the dispersion relation is presented elsewhere e.g. McKinstrie and Bingham(1992). The dispersion relation in the pulse frame normalised variables is:

$$1 + \lambda \left[\frac{1}{\epsilon_s(k, \omega - \omega_0)} + \frac{1}{\epsilon_s(k, \omega + \omega_0)} \right] \left[1 - \frac{(k + u\omega)^2}{\epsilon_p(k, \omega)} \right] = 0 \quad (1)$$

where

$$\epsilon_p(k, \omega) = (\omega + ku)^2 - \frac{3v_e^2}{\gamma_a} (k + u\omega)^2 - \frac{\omega_{pr0}^2}{\gamma_u^2} \quad (2)$$

$$\epsilon_s(k, \omega) = \omega^2 - k^2 - \omega_{pr0}^2 (1 - q^2/4\gamma_a^2) \quad (3)$$

and

$$\lambda = \frac{q^2 \omega_{pr0}^2}{4\gamma_a^2} = \frac{q^2 \omega_{p0}^2}{4\gamma_a^2} \quad (4)$$

where ω_{p0} is the laboratory frame plasma frequency, q the peak quiver velocity of an electron, v_e is the thermal electron velocity, γ_u is the Lorentz factor for the pulse frame. Here γ_a is the relativistic dynamical factor due to electron mass increase, defined by $\gamma_a = \sqrt{1 + q^2/2}$. Here frequencies are normalised to ω_0 , densities to n_0 and wavenumbers to c/ω_0 . Except for SRFS at low densities, the anti-Stokes term at $\omega + \omega_0$ represents a small correction. For the Raman-unstable waves the first term in the final bracket is likewise small; the dispersion relation also contains a branch corresponding to a 4-wave modulational instability when this term is significant [McKinstrie and Bingham(1992)]. Landau damping is accounted for at low densities by adapting ϵ_p to include the correct kinetic susceptibility.

In the fluid model, the effect of relativity is twofold. First the plasma frequency ω_{p0} is replaced by the reduced 'relativistic' Lorentz invariant plasma frequency $\omega_{pr0} = \omega_{p0}/\gamma_a^{1/2}$ (ω_{p0} is the laboratory frame plasma frequency). So inside the

pulse the instability occurs at an effective density lower than that which exists externally. Besides modifying the plasma wave and scattered wave dispersion relations, the coupling strength λ is also weakened by this reduction. Second, $q \rightarrow q/\gamma_a$ which simply reflects the fact that the electron quiver velocity is relativistically limited to values less than c . There is an additional relativistic correction $1 - q^2/\gamma_a^2$ in the scattered wave dispersion relation which varies between 1 and 1/2. Circular polarisation has an identical dispersion relation except for this term; the orthogonal driver fields of circular polarisation give exactly cancelling components.

III. ABSOLUTE GROWTH EIGENVALUES

In the pulse frame, the solution of (1) gives six modes. Neglecting the anti-Stokes modes for the time being, three modes travel from right to left and a single mode is right travelling in the pulse frame, which, as it reflects off the leading edge of the pulse converts partially into the three modes which are travelling to the left. The amount of mode conversion depends on the gradients at the edges but can be sufficient to allow absolute growth in the pulse frame. The immediate consequence of this is that exponentiation of growing modes then occurs not over the pulse duration but over timescales associated with the plasma length (Rayleigh length).

Eigenvalues for absolute growth are found by phase-matching the solutions of the dispersion relation inside and outside the pulse such that a time-asymptotic state is reached for given values of density, pulse duration and laser intensity. Seen from the laboratory frame modes impinging on the pulse trailing edge are upshifted in frequency and downshifted by an equal amount at the leading edge. This results in an upshifted wave for backscattering by a factor of approximately $4\gamma_a^2 = 4\omega_0^2/\omega_p^2$ which can be a large 'photon acceleration' factor in an underdense plasma. For forward scattering this upshift is by a factor $\omega_0/\omega_d \simeq 1$. A threshold for absolute growth is found, dependent on the pulse length and plasma density, but well below intensities achievable with current high power laser systems, as shown in Figure 1 for SRBS at the two densities $n = 0.1n_c$ and $n = 0.01n_c$. Here we use the variable $v_0 = q/2$ to represent the laser intensity. For backscattering the threshold intensity rises at lower densities. The results for backscattering (eigenfrequencies) are shown in Figure 2 for a density of $n = 0.1n_c$, a temperature of 100eV and a pulse length of 100fs. As expected from the discussion above, there is a blueshifted wing of unstable frequencies due to the matching between the modes at the pulse boundaries.

The relativistic increase in the electron mass effectively lowers the plasma frequency in the pulse, so an apparent density discontinuity is created at the pulse boundaries. Also the laser driving strength is self-limiting, so that increasing the laser intensity does not drive the system any more strongly. In fact it is found that peak growth occurs at around $q \simeq 1.4$ and higher intensities give weaker growth. This relativistic quenching of Raman growth [Sakharov and Kirsanov(1994)] can be seen in Figure 2 for SRBS. A comparison of a non-relativistic calculation with one which is fully relativistic for SRBS shows that relativity leads both to a reduction in growth rates and to the Raman emission being confined within a narrower spec-

tral range. The first feature is a consequence of the effective reduction in density within the pulse and the second arises due to the self-limiting of the laser driver strength- it acts to give a lower effective intensity, so that for a given pulse length, the threshold in intensity is higher. Thus SRS in the relativistic limit behaves as if it were occurring at a lower density. In particular, for SRFS the anti-Stokes (upshifted scattered light) must be included, and it is found that in the pulse frame, the scattered waves are at a frequency which is below the plasma frequency external to the pulse- so they can be trapped within the pulse. The scattered light propagates at a velocity comparable to the laser pulse group velocity.

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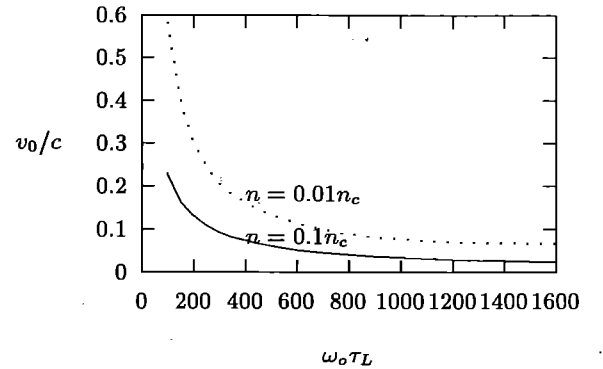


Fig. 1. SRBS absolute growth threshold intensity as function of pulse length $\omega_0\tau_L$ in fs.

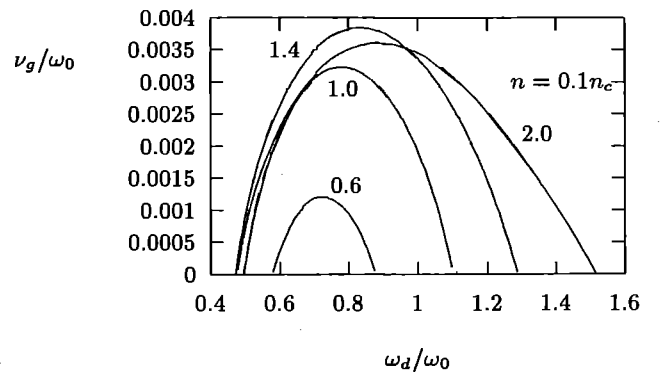


Fig. 2. SRBS lab frame growth rates ν_g/ω_0 versus detected frequency ω_d/ω_0 for various intensities, at density $n = 0.1n_c$.

WAKEFIELD AND BEATWAVE GENERATION WITH MULTIPLE AND CHIRPED PULSES

R A Cairns¹, D A Johnson¹ and R Bingham²

¹University of St Andrews

²Rutherford Appleton Laboratory

In recent years, the generation of beat waves and wakefields in a plasma has attracted a great deal of attention, because on the potential use of such waves as a means of accelerating particles to high energy. In a recent paper¹ we have shown how, in wakefield generation, it may be advantageous to split a given total energy available from a laser driver into more than one pulse. This is because of the nonlinear nature of the equation describing the generation of the plasma wave. In normalised coordinates¹, the equation for the potential, in a simple one-dimensional geometry, is

$$\frac{d^2\phi}{d\xi^2} = \frac{1}{2} \left[\frac{\gamma_a^2}{(1+\phi)^2} - 1 \right] \quad (1)$$

with

$$\gamma_a^2 = 1 + a^2$$

and a the vector potential of the electromagnetic wave. In our work, the advantage of multiple pulses was described in terms of a phase space analysis of equation (1). The essential feature is that the effectiveness of a driving term is much enhanced if it is applied near a point where ϕ is close to its minimum value of -1.

This, and other previous work along similar lines, for example that of Umstadter et al², has generally considered drive by a series of pulses of comparable amplitude. This may not be the best way to use a given total energy. If, instead, a substantial fraction of the available energy is used in the first pulse, in order to drive the system well into the nonlinear regime, then smaller pulses following behind can produce a dramatic increase in the amplitude. This is illustrated in Fig. 1, where the wakefield produced by a large initial pulse is shown to be amplified by two following pulses containing 20% and 10% of the energy of the initial pulse.

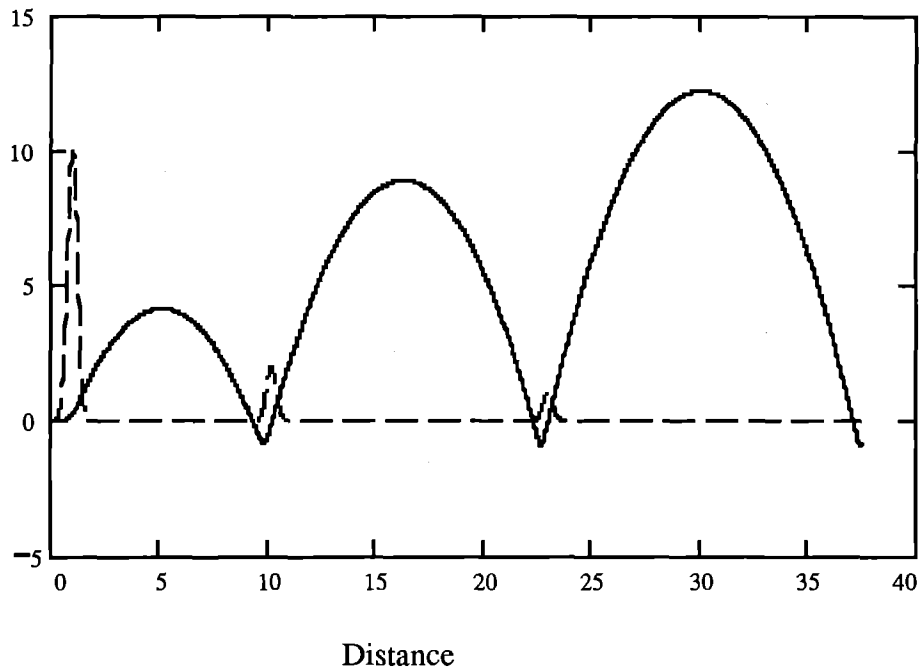


Fig 1 The wakefield potential (full line), produced by a series of three pulses (dashed line). The normalisation is as described in Ref. 1.

The width of the pulses should be of the order of half the plasma wavelength or less.

The idea of producing the electrostatic wave with a series of pulses is obviously rather closely related to the beatwave scheme, where the pulses can be thought of as being produced by the beating of two waves of slightly different frequency. Generally the

theory of saturation of beat waves has concentrated on the spatially uniform case and on the temporal evolution and saturation of the Langmuir wave. However, if we think of the overall envelope of the driving pulse for the beat wave being of finite length, then we can use Eqn. (1) to calculate the Langmuir wave generated in the steady state in the frame moving with the pulse. Fig 2 shows the way

in which a driver of constant wavelength goes out of phase, while if the frequencies are chirped in such a

way as to give a wavelength increasing through the pulse the final amplitude is enhanced.

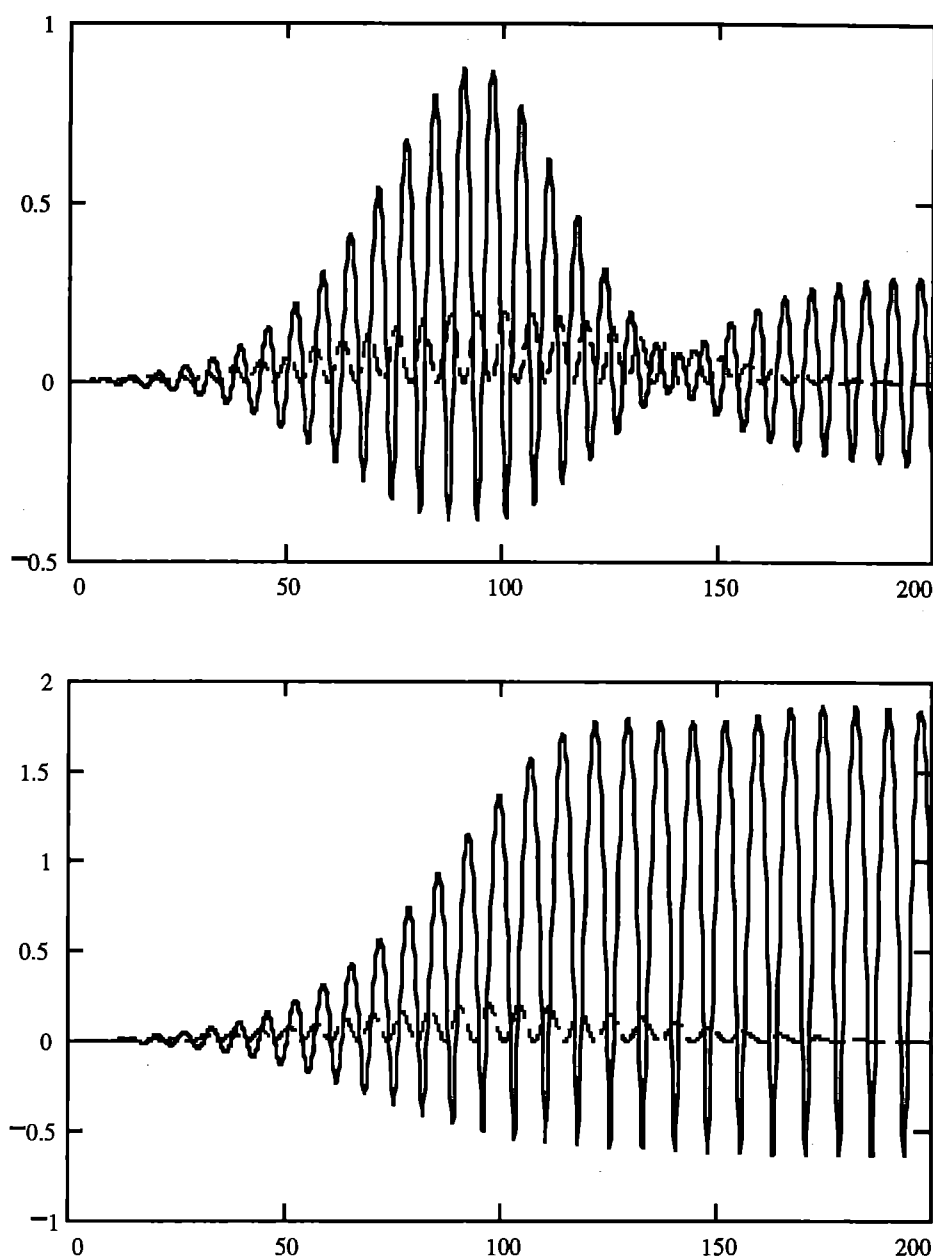


Fig 2 The top graph shows the beatwave driven by a pulse of constant wavelength and the lower graph the effect of making the wavelength of the beat increase through the pulse.

These results illustrate the fact that it is likely to be useful to give some consideration to the way in which the available laser energy is utilised to drive a wakefield or beatwave. Although the results we have presented are not optimised in any way, they illustrate some of the possibilities.

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HARMONIC GENERATION IN RARE GAS IONS

K. Burnett, A. Sanpera, X. Chen and J.B. Watson
Department of Physics, Clarendon Laboratory, Parks Road, Oxford

INTRODUCTION

The aim of this report is to show the atom/ion harmonic generation in the single atom response. In the last few years there have been several experimental studies [1-4] of harmonic generation at laser intensities between 10^{14} and a few times 10^{17} W/cm². These intensities range from close to well above the saturation intensity of any rare gas. The saturation intensity is conventionally understood as the intensity at which a sample of atoms is mostly ionized. In principle, going past the saturation intensity means that we should now have a clear experimental evidence of the role of ions in harmonic generation. Due to the larger ionization potential of rare gas ions compared to the neutral atoms, the harmonics generated by ions are expected to extend to a higher order than the ones generated by neutrals.

Interpreting the results of present experiments is not however, as simple as one might wish; going above saturation intensity does not mean we completely deplete the neutral population. Moreover, the neutrals that are left will experience very high intensities indeed, and could generate very high order harmonics. The efficiency in producing these high harmonics, however, will be much reduced due to the small amount of atoms left.

A qualitative explanation of the experiments performed to date can be obtained by combining the neutrals single atom response, with that of ions, for a full pulse. This is what we present in this report, so we can better assess the relative role played by the single atom/ion response. We shall address the case of Helium, and compare the responses of neutral He for a full pulse, with that He⁺, the latter produced along the pulse as the neutral atoms are ionised. Helium has the largest ionisation potential of all rare gases ($U_i=24.6$ eV) and can therefore survive to very high intensities in the ionisation process. The intensities we have used in the simulations range from 6×10^{14} to 2×10^{16} W/cm² for a fixed KrF wavelength. The choice of this wavelength is related to the efficiency in producing harmonics. Larger wavelengths will produce higher order harmonics but with an associated lower efficiency [5].

Finally, our results are compared with the available experimental data [6]. Quantitative comparisons should, of course, involve a calculation of the macroscopic issues of propagation with all its associated complexity. The single atom response is, nevertheless, useful in indicating at what intensities neutrals and ions are competitive.

METHOD

Our approach is based on a two-step procedure where, as a first step, the ionization and harmonic generation for the neutral Helium atom is calculated in the Single Active Electron (SAE) approximation with an absorbing boundary condition. The validity of this approximation relies on the assumption that ionization is a stepwise process. The effective potential $V_{\text{eff}}(r)$ of Helium is constructed from Hartree calculations on its ground state, using a modified version of the Hartree-Fock program. The ionization potential we obtain from the effective potential is $U_{\text{He}}=0.901$ a.u. (24.50 eV), very close to its experimental value (24.58 eV). From the decrease of the norm of the neutrals' wave function, a time-dependent ionization function, $P_{\text{ion}}(t)$ is obtained as the pulse proceeds. In the second step, we solve the time-dependent Schrödinger equation for the ion, and then, the ion wave function is weighted by the time-dependent $P_{\text{ion}}(t)$ ion-population function. In this way the problem is reduced from the time propagation of a two-electron wave function to the propagation of two one-electron wave functions. The numerical time-evolution of the one-electron wave function is performed using standard techniques.

NUMERICAL RESULTS

We calculate the harmonic generation from neutrals and ions for a number of laser intensities in the range 10^{15} - 5×10^{16} W/cm² for a sine-squared pulse of 128 cycles at different laser peak intensities,

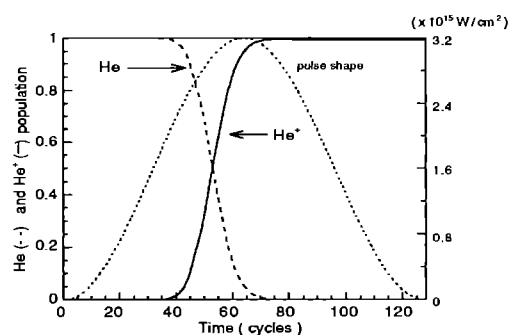


Figure 1

including the ions' response during the pulse. The populations of He and He⁺ together with the pulse profile for these parameters are

plotted in Figure 1. Finally, we compare our harmonic generation results with the experimental ones from references [6]. The best fit between experimental and numerical results occurs at intensities around $3.5 \times 10^{15} \text{ W/cm}^2$. This is shown in Figure 2 where the experimental data are plotted together with the neutrals and ions response at $I=3.2 \times 10^{15} \text{ W/cm}^2$. (The experimental data are normalized to the 13th harmonic).

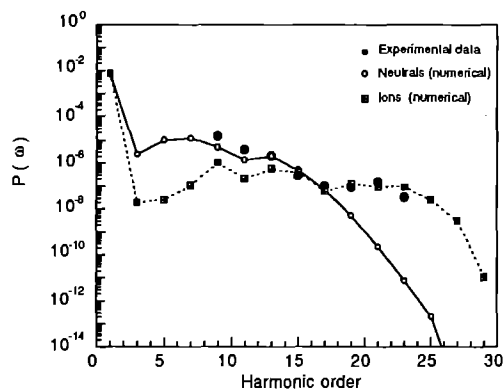


Figure 2

The agreement between the experimental and the numerical spectra is remarkably good. We can see that both, neutrals and ions, show the characteristic structure of a plateau followed by a cutoff. The plateau of ions extends to higher orders than the neutrals, and it is crossed by the neutrals cutoff around the 15th-17th harmonic. So, in fact, only the higher peaks (19-23 orders) are produced by ions exclusively.

The fact that such good agreement between numerical simulations and experiments is reproduced by using a peak intensity almost two orders of magnitude lower than the one claimed in the experiments, suggests that the effect of the defocussing of the laser beam at such intensity, gas density and experimental geometry is important. A full treatment of the defocussing inside a gas will require a complex calculation on the refractive index gradient produced by ionization. We could not assert that defocussing is solely to blame for the apparent decrease in intensity. In fact, other transverse properties of the beam - intensity variations, filamentation, poor beam quality - as well as longitudinal propagation effects, should be taken into account to have a full description of the experiments. It is quite impossible, at the present time, to combine a full treatment of single ion/atom evolution within a pulse with all the issues of beam propagation. The present study does, however, provide exceptionally strong motivation for pursuing studies of this kind [7].

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TWO COLOUR HARMONIC GENERATION

A. Sanpera ¹, K. Burnett ¹, M. Protopapas ² and P.L. Knight ²

¹ Department of Physics, Clarendon Laboratory, Parks Road, Oxford.

² Optics Section, Blackett Laboratory, Imperial College, London.

INTRODUCTION

The dynamics of an atom driven simultaneously by two frequencies displays important novel features that cannot be seen with single frequency driving [1-4]. For example, an enhancement of high order harmonic generation has been demonstrated in irradiation of rare gas atoms with a combination of the fundamental and third harmonic of a laser field [5]. Also early work on two-colour experiments using the fundamental and the second harmonic of a laser field showed that the relative phase between two fields had a significant influence on the ionization yield, the photoelectron spectra and the associated angular distributions [6]. The effects associated with the relative phase between two fields should be more evident for the cases in which the electron wave packet is strongly driven by the laser field. One should expect, therefore, to observe larger effects in the tunneling regime and in the stabilization regime. With all this premises in mind, we certainly want to know not only how two-color excitation of neutral atoms boosts overall harmonic generation but whatever one can drive the higher order harmonics more efficiently.

We shall address these questions by calculating the harmonic generation from a Hydrogen atom interacting with a two-color laser field. The two laser frequencies are chosen to be the fundamental and its third harmonic with the same intensity and a relative phase between them :

$$H_{int} = \vec{r} \cdot (\vec{E}_\omega \sin(\omega t) + \vec{E}_{3\omega} \sin(3\omega t) + \Phi)$$

where E_ω , and $E_{3\omega}$ are the fundamental and third harmonic field amplitudes, ω is the fundamental frequency and Φ , is the relative phase between the two fields (all in atomic units).

Our goal has been to study the effects of the two-colours and the influence of the relative phase in the tunneling regime. This has been done by solving the Schrödinger equation for different frequencies and values of the relative phase Φ . The method used to solve the two-color excitation is an straightforward extension (provided both fields have the same linear polarization) of the standard methods used to solve the interaction of an atom with a linearly polarised field [7].

To produce the maximum enhancement of the harmonic conversion

efficiency we choose the same field amplitude for both fields, i.e. $E_\omega = E_{3\omega}$. With these conditions the maximum time varying electric field is achieved for a relative phase difference between the two fields of $\Phi = \pi$, when the maximum field strength of the combined fields reaches the value $E = 2 \times E_\omega$ twice each cycle.

TUNNELING REGIME

The tunneling regime is conventionally characterized by the Keldysh parameter being smaller than unity, i.e. $\sqrt{U_i / 2U_p} < 1$.

(Here U_i refers to the ionisation potential and $U_p = E^2 / 4\omega^2$ refers to the ponderomotive potential). Physically that means that the laser field can be considered as a quasi-static field (high-intensity, low-frequency) that lowers the atomic potential so that the electron is able to tunnel out. The subsequent evolution in the continuum of the wave packet that has tunnelled out can then be described almost "classically" since the interaction with the electric field dominates and the influence of the Coulomb potential could therefore be neglected. Harmonic generation is understood in this context as due to the rescattering of the electron being driven back by the field to the nucleus and emitting a photon. This view is supported by the success of the classical model in explaining the origin of the harmonic cut-off [8].

In the two-color tunneling regime we expect the ionisation yield to be dependent not only of the maximum field strength of the combined fields but also on the relative phase Φ , since the potential barrier is significantly modified by this relative phase. In order to see the effect of the two-color interference we calculate ionisation yields and generation of harmonics for one single color at different intensities and compared them with the two-color results.

RESULTS

We first present results for a hydrogen atom irradiated by the combination of the Nd:YAG laser frequency ($\lambda = 1064$ nm) and its third harmonic ($\lambda_{3\omega} = 350$ nm) at $E_\omega = E_{3\omega} = 0.05$ a.u. ($I = 8.3 \times 10^{13}$ W/cm²). For the sake of comparison we also include the ionisation produced by the fundamental laser alone at E_ω and at twice the electric field amplitude $E = 2 \times E_\omega$ ($I = 3.5 \times 10^{14}$ W/cm²).

latter case we have already surpassed the saturation intensity for this frequency and pulse shape. (The saturation intensity is

conventionally understood as the intensity (for a given wavelength and pulse shape) at which a sample of atoms is mostly ionised). Since surpassing the saturation intensity diminishes the efficiency on generating harmonics one should be careful in comparing the effects of one and two colours at these high intensities.

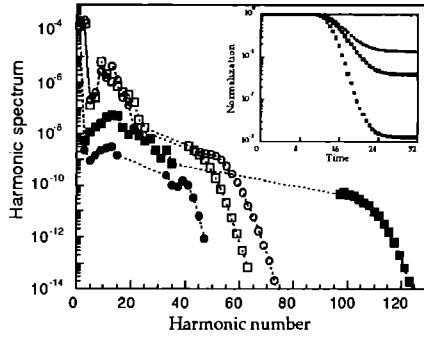


Figure 1

In Fig. 1 we show the harmonic spectra in the tunneling regime for one and two-color cases (the corresponding ionisation yields are shown in the inset) (Full symbols correspond to one colour case and open symbols to two colours at relative phases of 0 and π). All the spectra have been normalised to the first harmonic so that a better comparison of the conversion efficiencies can be made.

The two-color harmonic spectra show formally the same pattern as their one color partner: a plateau of harmonics and an abrupt cut-off where the intensity of the harmonics drop off rapidly. However a closer inspection reveals significant differences. First of all, the plateau structure appears less clearly defined than in the one color case. The most striking difference, however, is the conversion efficiency in the two-color cases compared to one color. Generally speaking the conversion efficiency is much better for the two-colours case regardless the relative phase. This is specially true for low energy harmonics, those with energies below $U_i + U_p$ which are enhanced by almost four orders of magnitude compared to the one color case at $E = E_\omega$. Furthermore, if we compare conversion efficiencies between two colours $E_\omega = E_{3\omega}$ and the fundamental alone at $E = 2 \times E_\omega$ we still find harmonics with intensities two orders of magnitude larger in the bichromatic case. Since the residual ionisation for two color is smaller than the one produced by the strong field ($E = 2 \times E_\omega$), as it is clearly shown in Fig. 1, the conversion efficiency increase seems to be partially related to the interference between the two colours. To reinforce this idea we compare the harmonics generated by one color and two colours, when both cases produce the same ionisation yield. This is

accomplished for example, with a single laser operating at $E_\omega = 0.09$ a.u. ($I = 2.8 \times 10^{14}$ W/cm²) (open symbols) and the two-color lasers at $E_\omega = E_{3\omega} = 0.05$ a.u. and $\Phi = \pi$ (full symbols). For both cases the residual ionisation at the end of the pulse is practically the same but the enhancement of the harmonics is drastically different as it can be seen in Fig. 2.

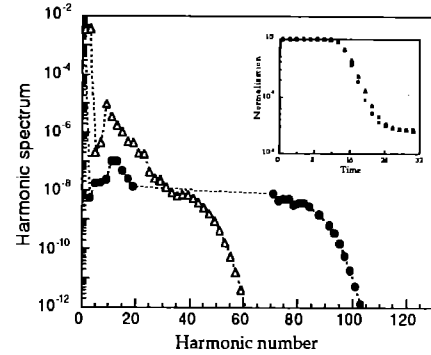


Figure 2

We have repeated our calculations for higher field intensities and the results are in agreement with what we have already presented. However the improvement on the conversion efficiency is obviously reduced if we are above saturation intensity or over barrier ionisation. Finally for lower ratios between the two fields i. e. $E_\omega / E_{3\omega} > 1$ the enhancement in the harmonics is also smaller.

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SPECTRAL LINE TRANSFER MODELLING IN COLLISIONALLY PUMPED X-RAY LASERS

KA Janulewicz, SB Healy and GJ Pert
Department of Physics, University of York.

Introduction

Demonstration of effective X-ray laser generation necessitated a resurgence of interest in ASE devices because all present X-ray lasers work as mirrorless structures. The earlier, basic model formulated in terms of a pure two-level scheme and rate equation approximation (REA) did not give good agreement with the available, limited amount of experimental data on the spectral line profiles of existing X-ray lasers and had to be re-examined. In more general treatment Pert (1994) and Koch (1994) removed many of the simplifying assumptions of the basic model. It was confirmed that even a relatively small contribution of a homogeneous component reduces markedly the rebroadening effect associated with the presence of an inhomogeneous component. No rebroadening effect was observed in the experiment (Koch 1993, 1994). However, the question of finding a full and effective descriptive method determining spectral profile modification is still open.

In the present paper, the kinetics of an active medium and the medium-radiation interaction are described in the terms of the density matrix elements. It has been assumed that the interaction of linear polarized plane wave has a quasi-resonant character. The spontaneous emission term and degeneracy are introduced phenomenologically. The output signal is analysed in the terms of Rabi frequency, which can be easily, if required, transformed into the intensity domain. Power broadening is introduced explicitly. In typical X-ray lasers schemes (especially collisionally pumped) we can expect comparable homogeneous and inhomogeneous components, hence we have decided to extend our analysis to arbitrarily broadened medium described by a Voigt profile. A generalised two-level atom model is used and connections to other levels are taken into account by effective values of the rate constants. Calculation of the complex expression for the Voigt profile is facilitated by adopting of the approximation method proposed by Kielkopf (1973).

Use of the strong collision model, which considerably simplifies calculations, is justified by the physics of collisionally pumped X-ray lasers. In collisionally pumped lasers electron energies $kT_e = 500-900$ eV are significantly larger than the energy gap between the laser levels (50-60 eV) and as result inelastic electron-ion collisions are the dominant process in the plasma and elastic, phase changing collisions can be neglected.

Model

An idealized ASE element consisting of a homogeneous, isotropically emitting, amplifying medium of length L and diameter $2a$ is considered to be interacting incoherently with a plane, linearly polarized wave. A photon flux is spontaneously emitted within whole volume of the active medium, but only radiation within a solid angle $\Delta\Omega = (a/2L)^2$ is assumed to be amplified. No refraction and nonlinear effects changing the beam parameters are included in our model.

The use of a unidirectional laser line transfer model is justified by the relation between typical gain durations (100-200 ps) and propagation time scale (velocity of light). The steady-state limit is valid because the laser intensity does

not change significantly on a time scale comparable with characteristic times of the system.

Kinetic equations describing the interaction of quasi-monochromatic radiation with a medium consisting of generalized two-level atoms have the form (Kolchenko 1968)

$$\left(\frac{\partial}{\partial t} + \vec{v} \cdot \vec{\nabla}\right) \rho_{ll} = \pm \Re(jV_{mn}^* \rho_{mn}) + S_l + q_l \quad (1)$$

$$\left(\frac{\partial}{\partial t} + \vec{v} \cdot \vec{\nabla}\right) \rho_{mn} = jV_{mn}(\rho_{mm} - \rho_{nn}) + S \quad (2)$$

Here $V_{mn} = -X e^{-j(\Omega t - k\vec{r})}$ is the matrix element of the interaction and q_l is the excitation rate of the level $l = m, n$. We will use for the sake of simplicity only one subscript l to denote parameters connected to each of the laser levels (m, n) and the corresponding diagonal density matrix elements ($\rho_{ll} = \rho_{mm}, \rho_{nn}$). $X = \rho \mathcal{E}_0 / 2\hbar$ is so called Rabi frequency; ρ denotes a matrix element of the transition dipole moment and $\mathcal{E}_0$ is the amplitude of the electric field. Ω determines the frequency detuning from the line centre ω_0 ($\Omega = \omega - \omega_0$) and $\rho_{mn} = \rho e^{-j\Omega t}$ is the nondiagonal element of the density matrix. Collision integrals S_l and S have the standard form and its kernel are specially simple for the strong collision model.

To solve integro-differential equations (1) and (2) Rautian and co-workers introduced partition of the atom lifetime at the given state into two parts (Rautian 1979). The first with the length τ_{1l} determines the lifetime of the atom in an unperturbed state (mean time between successive collisions). The second part with the length τ_{2l} corresponds to the rest of the lifetime and determines time of radiation interaction with the atom in its perturbed state. Thus solution of (1) and (2) for the amplitude of nondiagonal density matrix element has extremely simple form given by,

$$< \rho(\Omega, X, z) > = \frac{jX(\Omega, X, z)NY}{1 + 2|X_{int}(\Omega, X, z)|^2 \tau_2 Y'} \quad (3)$$

where Y is the convolution of the homogeneous and inhomogeneous spectral component. Using Kielkopf's approximation we can transform our radiative transport equation for a spectral component of the emitted line (3) after some algebra to its final form

$$\begin{aligned} \frac{d|X_\lambda(\Omega, z, X)|^2}{dz} &= g_0(0) \left(1 + \frac{\Omega}{\omega_0}\right) \\ &\frac{|X_\lambda|^2 U(x)}{(1 + \kappa) \left[1 + \frac{|X_{int}|^2 \tau_2 V_0 U(x)}{1 + \kappa}\right]} + \\ &+ g_0(0) A_{mn} \left(1 + \frac{\Omega}{\omega_0}\right) [\eta_{inv} - \\ &- \frac{|X_{int}|^2 \tau_m V_0 U(x)}{1 + \kappa}] U(x) \end{aligned} \quad (4)$$

From the last equation it is clear that $\tau_2 \neq 0$ introduces some additional saturation which appears as a homogeneous (non-selective) amplifying band in the population inversion. This additional saturating factor is the result of the finite probability that an atom removed from its initial state returns as a result of successive collisions.

Results

Expression (4) was used to model the modification of the spectral line profile propagating along an amplifying medium for Ne-like selenium and Ne-like germanium collisional lasers. We limited our interest to only one line for each laser, the $J=2-1$ transitions at $\lambda=206.38 \text{ \AA}$ and $\lambda=236 \text{ \AA}$ for selenium and germanium, respectively. For selenium the kinetic data are taken from (Koch 1994) and for germanium it were obtained from the EHYBRID-code described by Holden *et al.* (Holden 1994). In both cases the dependence of the FWHM and output signal on 1) the length of amplifying medium, 2) small signal gain coefficient and 3) the level of spontaneous emission was investigated. Additionally, having put $\tau_2 = 0$ influence of the redistribution by inelastic collisions can be derived.

The FWHM of Lorentz and Doppler broadened components for Ne-like selenium were equal to $14.1 \text{ m\AA}$ and $35.9 \text{ m\AA}$, respectively. The FWHM of the resulting Voigt profile was estimated to be $43.95 \text{ m\AA}$.

The following parameters were used in the calculations: small signal gain in the centre of the line - $g_0(0)=3.2 \text{ cm}^{-1}$ and 5.5 cm^{-1} ; ion temperature $T_i=400 \text{ eV}$; ion mass $m_i=78.9$ in atomic units; electron temperature $T_e=900 \text{ eV}$. Output signals appear only weakly dependent on the uncertainty in determining the spontaneous source emission level. The major uncertainty is associated with the estimated value of the solid angle in which radiation is directed. The larger aspect ratio of the medium results in the broader line of $0.73 \text{ m\AA}$. The calculated FWHM values have been compared with those obtained experimentally by Koch, and the FWHM dependence on medium length is shown in Fig.1 for lengths up to 65 mm . The experimental point corresponding to the length 63 mm was obtained as a mean value from two different measurements and the curves are plotted for two different aspect ratios. The agreement is satisfactory, apart from the first two points which were accompanied by reduced measurement accuracy (especially the first) because of high noise level (Koch 1993). The uncertainty in the first point was conservatively estimated as equal to $\pm 10 \text{ m\AA}$. In contrast to the theoretical calculations by Koch, our calculations do not show slight rebroadening for lengths greater than 3 cm , in accordance with experiment. Moreover, the linewidth is still decreasing with rate $\approx 0.0001 \text{ m\AA/mm}$. This small discrepancy could be explained by the contribution of power broadening in our model, in contrast to that used by Koch.

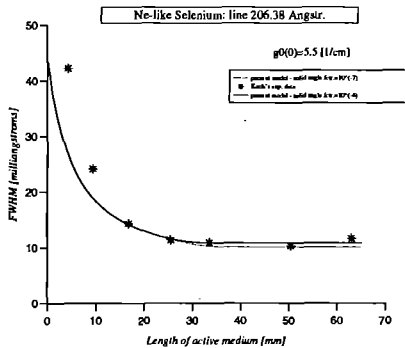


Figure 1: Laser linewidth vs. target length for Ne-like Se laser determined: - within the present model with the aspect ratio equal to 10^{-7} (dotted line); - within the present model with the increased value of the aspect ratio to 10^{-6} (more relevant to the experimental conditions) (solid line); - by Koch's experimental data (Koch 1993) (asterisks)

For the germanium laser the steady-state kinetic data was taking from calculation performed with the EHYBRID numerical code. All rate constants greater than 10^9 1/s were taken into account. The small signal gain coefficient used was $g_0(0)=3.8 \text{ cm}^{-1}$. We assumed considerably lower electron temperature $T_e=600 \text{ eV}$ than that used earlier for modelling (Holden 1994) and an ion temperature of $T_i=250 \text{ eV}$ was assumed. This data set is more relevant to the experimental situation. Changes of the absolute value of the linewidth are shown in Fig.2. The calculated linewidth of the intrinsic profile is comparable with that presented in Table.5 in (Holden 1994) where it was estimated for higher electron temperature (900 eV) to be equal to 2.1×10^{-4} . Our calculated value amounts to 1.84×10^{-4} for $T_e=600 \text{ eV}$.

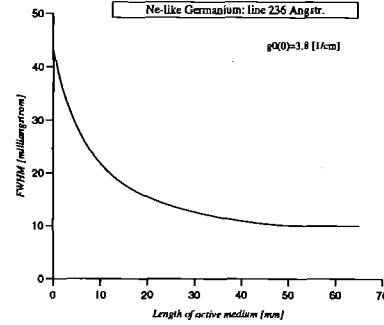


Figure 2: Absolute value of the linewidth vs. target length for Ne-like Ge

Conclusions

The solution of the general kinetic equations after some simplifications relevant to the physical processes were used to derive the final, relatively simple formula for saturated radiative transport adapting Kielkopf's Voigt profile approximation. Using Eq.(4) changes of the FWHM, output signal, and gain coefficient during the propagation in saturable amplifying medium have been determined. The results presented confirm that this model would be adequate for the purposes of predicting the laser linewidth in collisionally pumped lasers.

The proposed method possesses the additional advantage that the Voigt profile approximation implemented, reduces the time consuming calculation of the component convolution. In the unidirectional case presented and medium length of 65 mm the full run time has been reduced to less than 2 min. on the Silicon Graphics workstation used.

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Raytracing Simulations For X-ray Lasers

JA Plowes and GJ Pert

Department of Physics, University of York

Introduction

Electron density gradients cause refraction of the laser signal as it propagates through the plasma. This limits the maximum propagation distance within the gain region and so limits the maximum signal achievable using a single amplifier. Several methods of overcoming these problems have been suggested.

The benefits of using a toroidal shaped target has been suggested by Lunney[1]. This compensates for refraction both radially and transversely by curving the target to follow the signal path.

Radial refraction compensation by bending the target along its length has been demonstrated experimentally at Osaka[2]. Refraction in the radial direction is stronger than that in the transverse direction. A large increase in the signal is observed due to more efficient sampling of the target length. The effects of target curvature have been investigated using a 3D raytracing model[3].

The raytracing code is applied to a *Lunney* target to investigate the possible benefits of refraction compensation in both radially and transversely.

The effects of curvature along the target length are taken into account by altering the ray angles at each cell exit. The effects of the transverse curvature of the toroidal system on the ray path are modelled by altering the transverse density profile accordingly. The test case is a 4cm long, $100\mu\text{m}$ wide germanium slab target irradiated at $1.5 \times 10^{13} \text{ W cm}^{-2}$ at a wavelength of $1.06\mu\text{m}$. The target was curved along the lasing axis by 6 mrad/cm and the radius of transverse curvature was varied from $\infty \rightarrow 1.25\text{mm}$.

Simulation Results

Figure 1 shows the far field pattern when the target is curved only along the lasing direction. Figure 2 shows the identical pattern when the radius of transverse curvature is decreased to 1.25mm . The only noticeable improvement is a slightly better defined lobe structure. This indicates that the optimum gain length path is hardly altered even by this severe amount of curvature. Figure 3 shows the output signal versus transverse radius of curvature. A target with a radius of transverse curvature of infinity is marked on the intensity axis as a single point. Little increase in the signal is observed until the radius of transverse curvature is $\sim 0.2\text{mm}$. This curvature would be difficult to achieve experimentally by bending the target by mechanical pressure. Alternative methods for transverse refraction compensation are currently being investigated.

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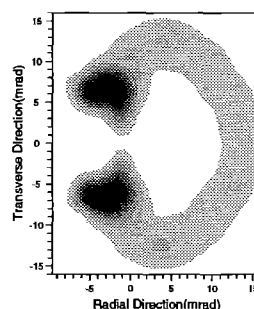


Figure 1: Far field, no transverse curvature

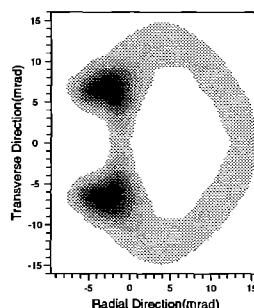


Figure 2: Far field, transverse curvature

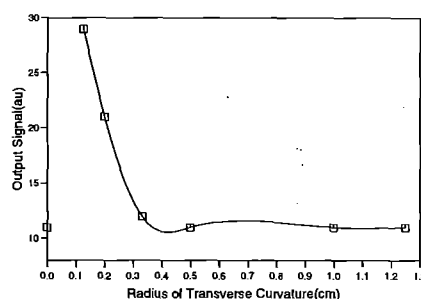


Figure 3: Intensity vs Radius of Curvature

COMPARISON OF APPROXIMATIONS FOR COLLISIONAL ABSORPTION IN HIGH FIELDS

G J Pert

INTRODUCTION

The development of high power ultra-short pulse lasers has led to generation of cold ionised plasma via multiphoton ionisation, and the related effect of high harmonic generation. The importance of these plasmas as a medium for recombination laser action at sufficiently high density, has re-opened the question of collisional, or inverse bremsstrahlung, absorption at high fields and low temperatures. In longer pulse, lower power applications the electrons will undergo several 90° scatterings during the pulse, and their 'temperature' will rapidly become comparable to the quiver energy. In consequence the perturbation of the distribution function away from Maxwellian rapidly becomes the determining factor in modifying the absorption coefficient - the Langdon effect. However with short pulses and high fields the number of 90° scatterings will be small, and the 'temperature' remains small compared to the quiver energy, ie the absorption remains in the strong field limit. In this regime the absorption is dominated by the quiver motion, rather than thermal.

The general theory of collisional absorption in arbitrary fields has not yet been developed, although a basic formalism was identified by Kroll and Watson. Rather the problem has been tackled by various approximations valid under appropriate conditions. These are identified by limits set by three characteristic energies, $\hbar\omega$, the photon energy, $\frac{1}{2}mv^2$ the thermal energy and $\frac{1}{2}mu^2$ the quiver energy. These exist in essentially three distinct approximations:

- i. *Low field* The general single photon quantum mechanical theory of Sommerfeld¹ *et al* is valid if $\frac{1}{2}mu^2 \ll \hbar\omega$. The classical description of bremsstrahlung emission is related to absorption via detailed balance and valid if $\frac{1}{2}mu^2 \ll \frac{1}{2}mv^2$. Both yield the same form with the results usually expressed in terms of a connection-Gaunt factor.
- ii. *Straight line path* The general straight line path approximation requires that the perturbation is induced by the collision including absorption be small. It appears in two forms quantum mechanical, Born⁴, or classical¹ depending on the ratio of the electron wavelength to the Debye length.
- iii. *Impact (classical) approximation* In this approximation the photon energy is small and the electron quiver motion is treated classically as a vector component of the total velocity. Only collisions occurring in a time short compared to the period of the field are considered to give rise to a change in the thermal velocity. The collision is governed by the instantaneous electron velocity which limits the impact time, and also establishes whether the collision itself can be treated by the classical or Born limits for elastic scattering.

Calculations in the high field limit have mainly been performed using one of the straight line path approximations: the Born approximation being especially popular, although the classical approach has also been used, but has the disadvantage of requiring an imposed logarithmic cut-off to take account of the close large angle scatterings with impact parameter approximately equal to the Landau parameter Ze^2/mv^2 .

This study arose from the realisation of a small discrepancy between results obtained with the Born approximation⁷ and the impact⁸ model

at high fields and small photon energy. We therefore concentrate on this limit, although more general results have been obtained and will be published elsewhere.

At high fields the energy absorbed per second by an electron in the Coulomb field of ions of density ρ may be written

$$q = 4 \frac{(Ze^2)^2}{Mu} \rho S \quad (1)$$

where Z is the ion charge, and e and m the electron charge and mass S is a parameter to be determined.

Straight line path

In both limits the final absorption rate may be expressed as a sum of rather similar terms over an index, which is the photon number in the quantum limit. Both the high field limit and the impact approximation correspond to a large range in this sum. For electrons with an isotropic distribution of uniform speed these sums can be analytically evaluated to give in the high field limit. For the Born approximation when the photon energy is small:

$$S = \ln(x) \ln(4y) + \frac{1}{4}[\ln(x)]^2 \quad (2)$$

and for the classical case

$$S = \ln(x) \ln(z') \quad (3)$$

where $x = (u/v)^2$, $y = \hbar\omega/mv^2$ and $z' = k'v/\omega$ are all large, and k' is the wave number cut-off required by classical theory to allow for the divergence imposed by the failure of the straight line path approximation at small impact parameters. For the present we assume k' is constant, but in practice it will depend on the total velocity rather than the thermal alone.

For the Born approximation we may obtain Polishchuk and Meyerter-Vehn's⁷ result by integrating over the Boltzmann distribution.

Impact approximation

The impact approximation can also be used under these same conditions to yield⁸:

$$S = \ln(4x) \ln(2y) + \frac{1}{2}[\ln(4x)]^2 \quad (4)$$

in the Born approximation. For comparison with the straight line path, the classical cross section must be calculated with a constant wave number cut-off to give:

$$S = \ln(4x) \ln(z') + \frac{1}{4}[\ln(4x)]^2 \quad (5)$$

DISCUSSION

Comparing the leading terms in these two sets of expressions we note a constant difference of $\frac{1}{4}[\ln(x)]^2$ in the leading terms. Since this appears in both cases it is clearly associated with a fundamental difference in the two approximations.

Within the impact approximation it is easily shown that the coefficient of this $[\log(x)]^2$ arises from the variation of the argument of the Coulomb logarithm with the total velocity: $w = v + u$. In the impact approximation this is imposed by the condition that the

collision be complete within the period of the wave, ie the outer impact parameter cut-off is w/ω .

The impact approximation can be treated as an asymptotic limit of the straight line, however a careful review of the asymptotic condition shows that it implies of an outer limit v/ω , ie determined by the thermal speed. This result is consistent with the results obtained directly. However returning to the general validity condition for the straight line path it can be seen that when the absorbed energy is large compared to the thermal energy, ie $x \gg 1$, the velocity is markedly changed during the collision and the approximation cannot be accurate. We therefore conclude that at high fields the additional term in (4) and (5) should be included.

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Phys. Rev. E - in press

Introduction

The collisionally pumped x-ray laser is currently considered the most robust method for generating high intensity Amplified Spontaneous Emission (ASE) in the XUV region. As the general physics of these systems is becoming progressively more understood (eg Holden et al 1994), increasing attention is being paid to improving the efficiency of these lasers. Consequently, alternatives to the original single pulse, flat target experimental configurations are being investigated. Both refraction compensation using target curvature (Kodama et al, 1995) and driver energy reduction using multiple pulses (Daido et al, 1995) have given promising results. An alternative approach, which is considered in this work, involves irradiating the target material with a low energy prepulse which precedes the main pulse by several nanoseconds. Nilsen et al (1994) have demonstrated that this can have a dramatic effect on the output of the $J = 0 - 1$ line for ions ranging from Ti^{12+} to Se^{24+} . Experiments have also been performed by groups at Garching (Fill et al, 1995) and Orsay (Klisnick et al, 1994). However, this report is motivated primarily by experiments undertaken at RAL by Cairns et al (1995), which have shown an enhancement of the 196Å and 236Å lasing lines of Ge^{22+} with prepulses of 0.02% and 2% of the main pulse.

In order to investigate how a prepulse can improve the output of an x-ray laser, two aspects of the resulting plasma have been considered in detail. Firstly, preforming the plasma may lead to increased absorption and result in conditions which are more favourable for generating large gain coefficients (gain at higher electron temperatures and densities). Secondly, it should reduce the electron density gradients and refraction of the laser beam, enabling better sampling of the gain regions.

The Computational Model

The 1.5D hydrodynamics/atomic physics code, as described by Holden et al (1994), has been modified for the purposes of this work. Bonding of the cold solid has been included by implementing the Chart D Equation of State package (Thompson et al, 1975). In addition, experimental values for the thermal conductivity (Regel et al, 1971) of Ge in the solid and molten liquid regimes upto a temperature of $\sim 0.8eV$ have been included. These modifications have been made in order to describe the plasma formed during the prepulse as accurately as possible. The electron density and gain profiles are postprocessed with the raytracing code described by Plowes et al (1995).

Results

The results that are presented refer to a Ge slab target of the length 1cm width $10^{-2}cm$ and thickness (parallel to the pumping laser) $10^{-4}cm$, irradiated by a $1.06\mu m$ optical pump. The prepulse precedes the main pulse by 5ns, both of which have the same trapezoidal shape, characterised by a rise to peak power in 200ps, 300ps flat and 200ps fall. The peak intensity of the main pulse is $1 \times 10^{13} W cm^{-2}$ which is fixed in all the simulations, giving 50J delivered on target. The prepulse level is varied and is specified as a percentage of the main pulse.

The absorption of the main pulse is significantly enhanced as the prepulse level is increased as a consequence of more in-

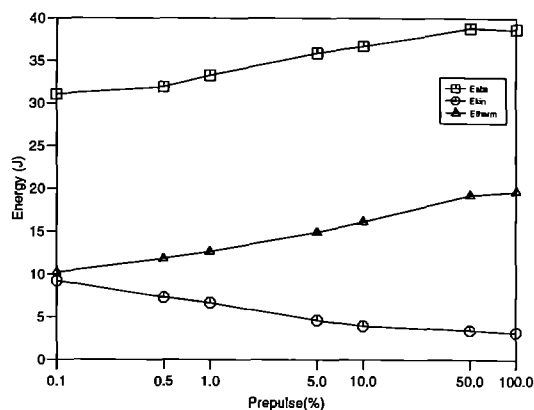


Figure 1: Variation with prepulse level of the absorbed energy (E_{abs}), the kinetic energy of expansion (E_{kin}) and random thermal energy (E_{therm}) at 500ps into the main pulse.

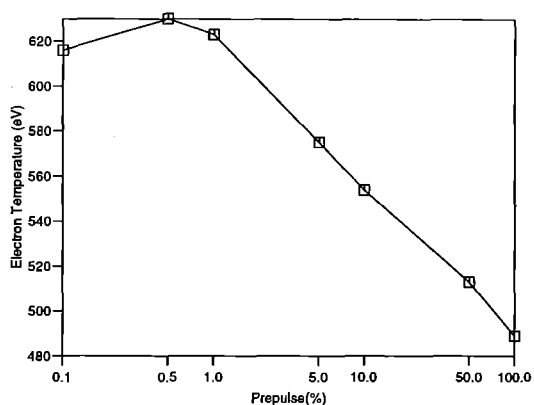


Figure 2: Variation of the peak electron temperature with prepulse level at 500ps into main pulse.

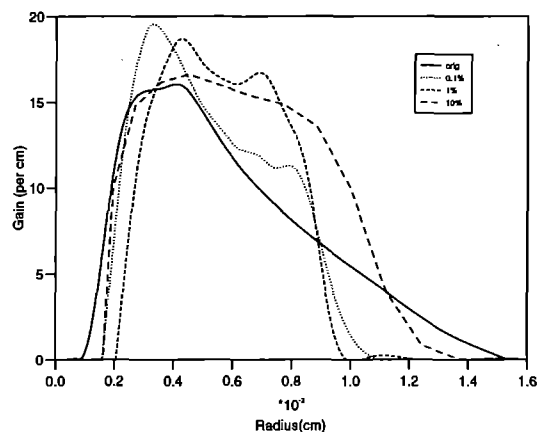


Figure 3: Variation of the 196Å radial gain profiles at 500ps with prepulse level.

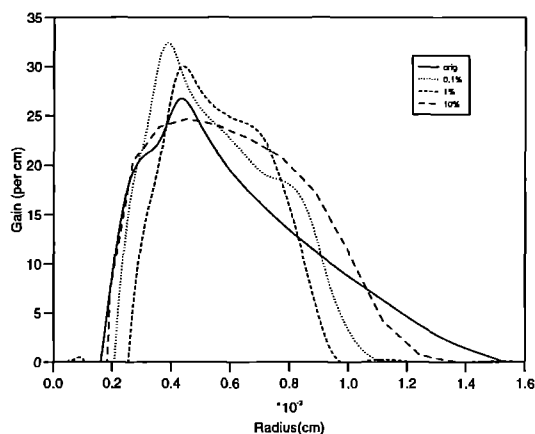


Figure 4: Variation of the 236Å radial gain profiles at 500ps with prepulse level.

verse Bremsstrahlung heating in the preformed plasma. This is apparent in figure 1, which shows the absorbed energy 500ps into the main pulse when 40J has been delivered. In addition, the expansion cooling during the main pulse ('Ekin' shown in figure 1 which gives a measure of the expansion cooling) is reduced with prepulse level, as progressively more work is done in expanding the plasma before the main pulse switches on. Consequently more of the absorbed energy is stored as random thermal energy ('Etherm' in figure 1) or is used to ionise the plasma, which indicates improved conditions for gain. However, if the prepulse is too big, too much plasma is formed and electron temperatures within the plasma will begin to fall as shown in figure 2.

Figures 3 and 4 show the radial gain profiles of the 196Å and 236Å transitions respectively, 500ps into the main pulse. The high gain regions initially broaden as the prepulse level is increased. However, at prepulse levels above 10% the gain coefficients begin to fall, reflecting the reduced temperature as too much plasma is created. In contrast, the electron density gradients become flatter as the prepulse level is increased. The combination of these factors gives rise to an optimum prepulse level: the prepulse must be large enough to produce a flatter density profile whilst not being so large as to significantly reduce the electron temperature and thereby bring down the gain coefficients within the plasma. Identifying the optimum level can only be done by raytracing profiles predicted with the hydro/atomic physics code. The results of such a calculation are shown figure 5. It should be noted that both the 196Å and 236Å lines show an enhancement over their single pulse values although it is more dramatic for the former. The 236Å line has a higher inten-

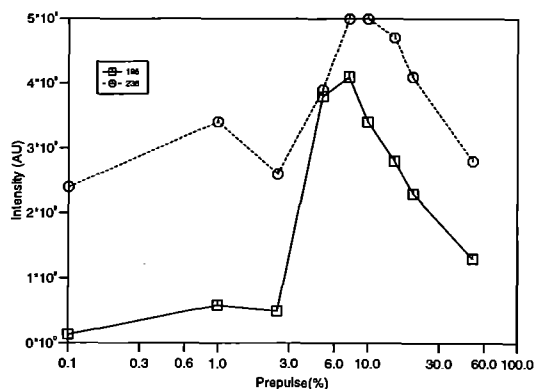


Figure 5: Variation of the output intensity with prepulse level of the 196Å and 236Å transitions

sity for all the simulations but the 196Å line is comparable which is unlike the single pulse simulations where the former dominates. It is interesting to note why the 196Å line is enhanced more than the 236Å line. Simulations to calculate the 'source' (Plowes et al 1995) of the far field pattern for both lines indicate that the 196Å beam originates in a lower density region and propagates further along the laser axis before it is refracted out of the plasma. Therefore, this line is more sensitive to better conditions within the plasma as the laser beam has a longer path length through the improved gain coefficients than the 236Å line.

Conclusion

The simulations have shown that a prepulse which precedes the main pulse by 5ns can significantly modify the state of the laser produced plasma during the main pulse. This is due to increased inverse bremsstrahlung absorption in the preformed plasma and a reduction of expansion cooling during the main pulse. Consequently, the ionisation and random thermal energy components of the overall energy balance are found to increase indicating that the plasma is more ionised and hotter. However, if the prepulse is too big, too much plasma is formed and the electron temperatures begin to fall. An optimum prepulse level arises because larger prepulses give rise to flatter density gradients, enabling better propagation of the laser beam but this is eventually countered by lower gain coefficients due to the reduced electron temperature. Raytracing indicates the optimum prepulse level is in the 5 – 10% region.

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Residual Electron Energy Simulation of Optical-Field-Ionised Nitrogen

MJ Grout and GJ Pert

Department of Physics, University of York

Introduction

A conventional thermally ionised recombination x-ray laser scheme involves irradiating a target with a high power and energy pump laser. This forms a high temperature plasma of either fully stripped or closed-shell configuration ions. If rapid cooling follows, a population inversion may result from the ensuing electron recombination. However with recent advances in ultrashort-pulse, high intensity and low energy pump lasers, optical-field-ionisation of gas targets resulting in low temperature electrons may now be possible, without the need for cooling. In this paper the case of lithium-like nitrogen is considered, which may produce lasing to the ground state. A crucial factor to the feasibility of the scheme is the electron temperature. Simulations of the dominant heating mechanisms present during the pump pulse have been performed using both particle in cell (PIC) and analytical codes.

Description of Codes

The 1D PIC code MULTIPHOTON1 considers a Gaussian profile pump beam symmetrically positioned such that the $1/e$ full width covers the centre quarter of the mesh. The beam profile has random phase modes superimposed on it. The code calculates a tunnelling probability for multiphoton ionisation by a strong linearly polarised EM field. Above threshold ionisation (ATI) heating due to phase mismatch between the instant of ionisation and the peak of the EM wave, is calculated at the time of ionisation of each electron. Strong field inverse Bremsstrahlung heating from transfer of quiver energy into thermal energy during an elastic electron-ion collision is calculated by Monte-Carlo methods for multiple small angle Coulomb scattering. A 1D external field structure and space-charge effects are included.

Two numerical codes ANALYTIC and AVERAGED were used in conjunction with MULTIPHOTON1. Both codes calculate ionisation and ATI contribution every half cycle of the incident Gaussian wave. Again the ionisation was calculated from the tunnel ionisation probability, and the strong field finite temperature approximation [1] gave the inverse Bremsstrahlung heating. MULTIPHOTON1 and AVERAGED produce an electron energy averaged over electrons found in the centre three quarters of the beam. Whilst ANALYTIC gives an energy for the electrons at the peak of the beam spatial profile.

Simulation Results

Simulation conditions were as follows. A driver pulse of 100fs (FWHM) at wavelength 250nm, was focussed over a length of 10cm to produce a $30\mu\text{m}$ (FWHM) spot, with peak intensity $3 \times 10^{16} \text{Wcm}^{-2}$. This intensity was above the threshold for producing a very high degree of helium-like nitrogen ions close to the peak of the spatial profile.

For an ion density of $1.0 \times 10^{18} \text{cm}^{-3}$ MULTIPHOTON1 produced the residual energy trace shown in figure 1. This shows that kinetic energy was dominant whilst the potential energy due to electron expulsion by the ponderomotive force was small. Most heating occurred during the first half of the

pulse and the time just after the pulse started played an important role.

Figure 2 shows strong agreement between MULTIPHOTON1 and AVERAGED indicating that for these conditions the analytical model works well. Thus ANALYTIC gives accurate energies for electrons ionised at the peak intensity to leave helium-like ions which can recombine into the lithium-like state.

Now that realistic values for electron temperatures have been found, work will continue by running the ANALYTIC code with an atomic kinetics model. Initial studies suggest that lasing on the $3d_{5/2} \rightarrow 2p_{3/2}$ (248Å) transition in nitrogen may give high transient gain, and thus this scheme may possibly result in a viable tabletop x-ray laser.

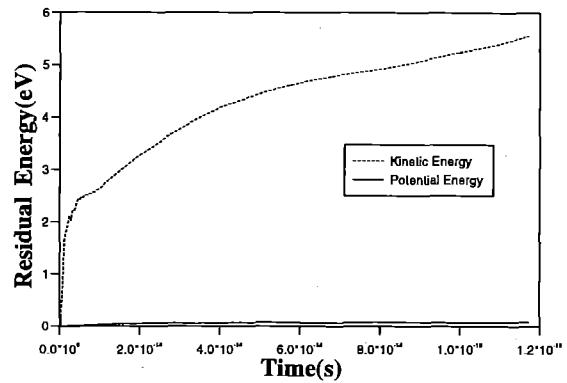


Figure 1: Residual Energy During Pulse

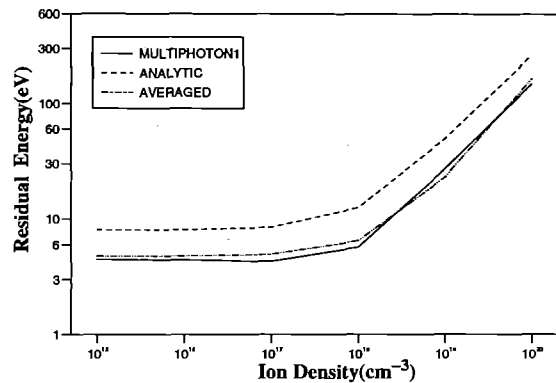


Figure 2: Residual Energy vs Ion Density

Reference

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HIGH POWER LASER DEVELOPMENT AND OPERATIONS

M J Shaw

Rutherford Appleton Laboratory, Chilton, Didcot, Oxon. OX11 0QX

INTRODUCTION

This year has been a very significant year in the development of the high power laser programme at the CLF. It has been the last year of operations for the Sprite KrF laser system. The last shot on Sprite was fired by Paul Williams on March 31 1995. The new KrF laser system Titania will be built in Sprite's place over the coming year. The work on laser development reported here reflects the urgency of the KrF programme with a healthy series of reports on R & D work necessary for the Titania upgrade. Vulcan, on the other hand, has been fully occupied supporting the high power experimental programme. New developments in plasma diagnostic instrumentation are also reported here.

KrF LASER DEVELOPMENT

Sprite has been used for two major R & D runs during this year. The first was concerned with characterising the Raman system and especially investigating the cause of the off-axis second Stokes light which had been observed previously. By imaging the Stokes far field, three distinct parametric processes were observed. It was found that all three involved interaction of scattered light with either the strong pump or strong Stokes signals. One of the processes - that giving rise to rays - has never been seen before. This work has lead to a forthcoming paper in Phys. Rev. Letts. (May 1995).

An important check on the effect of multiple pulses on the KrF pump beams was also undertaken to demonstrate the principle of Raman multiplexing. It was found that the far field beam quality was unaffected by the existence of the previous pump (and Stokes) pulse which occurred only 1.5 ns earlier in time. This result gives confidence to the design of the Titania Raman system.

The second major R & D run on Sprite was work which was jointly supported by the US Department of Energy via The Naval Research Laboratories. It involved an investigation into the origin of the "dip" in the KrF laser gain spectrum. The existence of the dip is detrimental to both the ultra-short-pulse and ultra-broad-bandwidth performance of KrF and is hence of importance to both the Titania and the Nike (NRL) laser programmes. The origin of the dip was found to be due to excited state absorption within the KrF molecules itself. So, unfortunately, nothing can be done to remove it. This result, however, is not expected to affect the performance of Titania (or Nike) in any significant way.

Two further R & D reports are both theoretical studies to do with bandwidth. The first, by Reid et al is a numerical study of third harmonic generation to 248 nm. It is concluded that β BBO is the crystal of choice and that efficiencies of 25% should be achieved in an optimum system. In the other report, McDonald et al show that soliton-like solutions exist in the transient regime of multifrequency Raman generation.

TITANIA UPGRADE

This section contains three papers reporting on the current status of the Titania system. In the first, the commissioning of the Titania module pulsed power system is reported. The target figure of 20 kJ in the gas was achieved with significant Marx voltage to spare. This was largely due to higher than expected diode-to-gas transfer efficiency which was found to be 40% rather than the (conservative) design figure of 30%. The energy deposition was found to be highly uniform across the aperture, having a peak-to-valley variation of under 10% and an rms variation of 3%.

The second report is on the laser performance of the Titania module. Problems were encountered in the oscillator measurements due to the lack of optimum cavity mirrors. The

maximum recorded output energy was 850 J. The gain measurements were more encouraging with a pulsed net small signal gain of 6000X being measured at full pump rate. Taking account of static losses should give a module gain of around 2000X in the Titania system.

The third report describes the completion of the optical system design for Titania. The design of the Raman system was considerably aided by the use of a 3D CAD package which has been used to optimise the positions of all of the mirrors in the system.

INSTRUMENTATION

The developments in plasma diagnostics this year have centred around imaging both in the visible and X-ray regions of the spectrum. The usefulness of the existing flat-field spectrographs has been greatly extended by the addition of a variable focal length X-ray imaging optic. A grazing incidence cylindrical mirror is used to relay an image of the X-ray source to the output spectral plane which is a true flat-field both spatially and spectrally. The spectral region covered is from 2 - 50 nm with a spatial resolution matched to micro-channel plate detectors. The new design also provides a considerable improvement in sensitivity over previous spectrographs by maintaining the micro-channel plate under high vacuum for extended periods.

With the increasing use of micro-channel plate detectors in the soft X-ray region there is a need for accurate determinations of spectral sensitivity. The report in this section describes measurements of the sensitivity of the channel plate/phosphor combination which will be of great benefit to future users.

The final report in this section describes a new f2.5 visible imaging system for optical diagnostics. The advantage is that long focal length lenses are used which are situated well away from the focal region and incoming laser beam. Even so, 3 μ m resolution is achieved with monochromatic images over a spectral range 0.25 to 1.1 μ m.

OPERATIONS

This has been another fruitful year for operations with Vulcan fully scheduled and Sprite hitting another all-time high in target shots. On Vulcan the CPA configuration has been very popular but long pulse operation primarily for X-ray laser work in TAE has also been productive. Increased energy for TA4 has resulted in benefits for the biological imaging work.

Sprite has been configured in the CPA mode for most of the year. The highlight of the year being the observation of the shortest wavelength ever produced by harmonic generation.

ON THE ORIGIN OF THE DIP IN THE KrF LASER GAIN SPECTRUM

E J Divall, G J Hirst, C J Hooker, J M D Lister, I N Ross and M J Shaw,
Rutherford Appleton Laboratory, Chilton, Didcot, Oxon. OX11 0QX
A Kvaran,

Science Institute, University of Iceland, 107 Reykjavik, Iceland
K Osvay,

Dept of Optics and Quantum Electronics, JATE University, Szeged, Hungary.

INTRODUCTION

The presence of a dip in the KrF laser gain spectrum at a wavelength of 248.9 nm has been known about for a long time. For narrow band laser light centred at the gain maximum (248.4 nm) the dip is of no consequence, but for the amplification of broadband light it is a major issue. The reduction in bandwidth caused by the dip seriously restricts the use of KrF laser systems for ultra-short pulse amplification, or fusion applications where incoherent light is required for beam smoothing purposes.

In the past, the dip has been attributed to impurity[1], particularly CF₂, but its ubiquitous nature (not a single laser spectrum without the dip could be found in the literature) points to a more fundamental cause. In last year's annual report Ross et al [2] using discharge lasers showed that the dip was more dependent on gain than anything else. The results reported here follow up these measurements with measurements on the Sprite e-beam-pumped module.

EXPERIMENTS

The main experiment consisted of measuring the single-pass ASE spectrum from the Sprite module as a function of gain. The experimental layout is shown in Figure 1. To record the ASE spectra, the output window of Sprite was imaged onto the slit of a high-resolution grating spectrometer. The spectrometer used in these experiments was specially constructed on an optical breadboard. It was an asymmetric Czerny-Turner configuration having an input focal length of 1768 mm and an output focal length of 1068 mm. The 100 mm diameter collimating mirrors gave an f/18 relative aperture at the input. The holographic grating had 3600 g/mm.

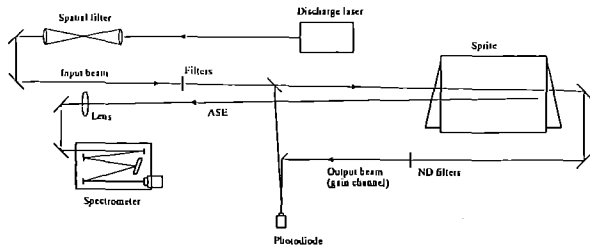


Figure 1. Experimental layout for the ASE measurements

The detector used was a Princeton Instruments intensified diode array which incorporated a Peltier cooler for low noise operation. The intensifier could be externally gated with a minimum pulse length of 10 ns. The spectrometer was calibrated using a hollow-cathode iron lamp which is rich in wavelength standard lines in the region of 248 nm. The measured dispersion was 0.00555 nm/pixel and the spectral range was 4.00 nm. The resolution was 0.0178 nm and is consistent with the 50 μ m input slit width and the magnification of 0.6. The absolute wavelength calibration was accurate to 0.005 nm, somewhat better than a single pixel.

The probe pulse used to measure the gain was generated by a KrF discharge laser (Lambda-Physik EMG 103) configured as an oscillator, and had a duration of about 15 ns. A 10 ns gate was used on the detector and the gain was measured with the probe pulse at the same time as the spectrometer gate pulse. Most of the data were obtained at the peak of the e-beam pump pulse. It was important to use a method of varying the gain which perturbed parameters other than the gain as little as possible. To this end, in the bulk of these experiments, it was decided to keep the total pressure and the electron beam current and voltage constant, and obtain the gain variation by varying the krypton fraction in the Ar/Kr/F₂ gas mix. This should keep the positive and negative ion densities constant and more importantly the vibrational relaxation of the KrF B state would be constant so that the changes on the B-X spectrum would not be due to a changing vibrational distribution. The disadvantage of this technique is that the positive ion and excited species type changes from predominantly Ar⁺, Ar₂⁺, Ar^{*}, Ar₂^{*} to Kr⁺, Kr₂⁺, Kr^{*}, Kr₂^{*} as the krypton concentration is increased. To see if these effects were important some spectra were recorded while changing the gain by varying the pressure, fluorine concentration and time in the pulse.

ANALYSIS OF THE ASE SPECTRAL PROFILES

At very low values of gain the fluorescence spectral distribution in KrF has a Gaussian distribution with a FWHM of 2.3 nm. This distribution is a very good fit to the measured curves as seen in Figure 2.

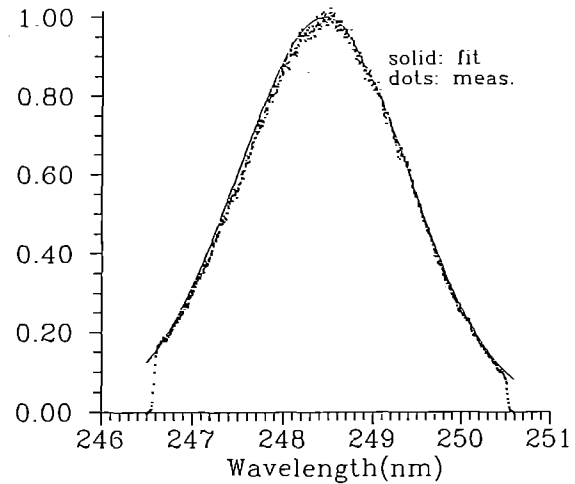


Figure 2 KrF fluorescence spectrum recorded at very low krypton concentration. The fitted Gaussian curve assumes a gL = 0.04

The ASE is modelled by amplifying the fluorescence with a gain term $g(\lambda)$ which has an identical spectral distribution to the fluorescence distribution and by attenuating it with an effective loss term $\alpha(\lambda)$. The magnitude and spectral distribution of the loss term is chosen to give the best fit between the calculated ASE and the measured spectral profile. The amplified contribution to the output power from dA into the solid angle $d\Omega$ and in a spectral element $d\lambda$ is given by:

$$\delta P(\lambda)d\lambda = \frac{NdAdz}{\tau_{rad}} \cdot \frac{d\Omega}{4\pi} \cdot h\nu \cdot \frac{g(\lambda)d\lambda}{\int g(\lambda)d\lambda} \cdot \exp[g(\lambda) - \alpha(\lambda)]z$$

The output ASE spectral brightness $B(\lambda)$ is given by:

$$B(\lambda) = \int \frac{\delta P(\lambda)}{dA d\Omega} =$$

$$\frac{Nh\nu}{4\pi\tau_{rad}} \cdot \frac{1}{\int g(\lambda)d\lambda} \cdot \frac{g(\lambda)}{[g(\lambda) - \alpha(\lambda)]} [\exp(g(\lambda) - \alpha(\lambda))L - 1]$$

Let the peak value of $g(\lambda) = g(\lambda_0) = g_0$ and similarly let $B(\lambda) = B_0$ and $\alpha(\lambda) = \alpha_0$ at $\lambda = \lambda_0$. Then:

$$\frac{B(\lambda)}{B_0} = \frac{g(\lambda)(g_0 - \alpha_0)}{g_0(g(\lambda) - \alpha(\lambda))} \cdot \frac{\exp(g(\lambda) - \alpha(\lambda))L - 1}{\exp(g_0 - \alpha_0)L - 1}$$

If the fluorescence lineshape is given by:

$$F(\lambda) = \exp\left[-4 \ln 2 \left(\frac{\lambda - \lambda_0}{\Delta\lambda_0}\right)^2\right]$$

then $g(\lambda) = g_0 F(\lambda)$.

In order to find an appropriate form for the absorption spectrum $\alpha(\lambda)$ it is useful to divide a measured ASE spectrum by the calculated spectrum without absorption (i.e. set $\alpha(\lambda) = 0$). When this was done there were found to be three primary absorption features; the "dip" at 248.9 nm, a broadband blue

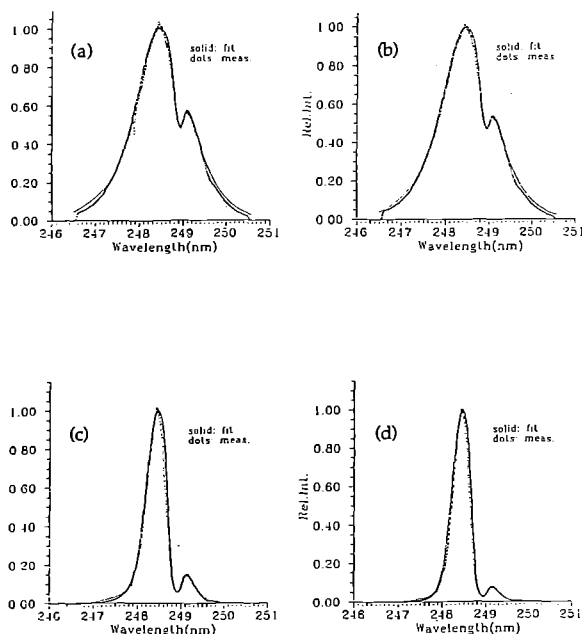


Figure 3 Examples of experimental ASE Spectra with best-fit curves. The measured gains were (a): 3.6, (b): 7.6, (c): 64, and (d): 104.

feature peaking around 247.6 nm and a smaller red peak at 249.65 nm. To analyse the absorption spectrum quantitatively, a trial absorption spectrum containing just the two main features is assumed i.e. $\alpha(\lambda) = \alpha_d(\lambda) + \alpha_b(\lambda)$ where d = "dip" and b = blue. The spectral distribution of each term was chosen to be Gaussian (Lorentz distributions were also tried but did not give quite such a good fit to the experimental curves). Thus:

$$\alpha_d(\lambda) = \alpha_{d0} \exp\left[-4 \ln 2 \left(\frac{\lambda - \lambda_d}{\Delta\lambda_d}\right)^2\right]$$

$$\alpha_b(\lambda) = \alpha_{b0} \exp\left[-4 \ln 2 \left(\frac{\lambda - \lambda_b}{\Delta\lambda_b}\right)^2\right]$$

Figure 3 shows several examples of experimental ASE spectra together with best fit curves according to the above recipe. Each fitting process yields values for the seven parameters: g_0 (peak gain coefficient), α_{d0} (peak loss coefficient for the main dip), α_{b0} (peak loss coefficient for the blue dip), $\Delta\lambda_d$ and $\Delta\lambda_b$ (FWHM of the loss curves), λ_d and λ_b (peak loss wavelengths).

The results showed that the wavelengths and widths of the two main absorption lines were essentially constant and that the absorption coefficients varied linearly with gain.

Thus:

$$\begin{aligned} \alpha_{d0} &= 0.32 g_0 \\ \alpha_{b0} &= 0.2 g_0 \\ \Delta\lambda_d &= 0.27 \text{ nm} \\ \Delta\lambda_b &= 0.7 \text{ nm} \\ \lambda_d &= 248.91 \text{ nm} \\ \lambda_b &= 247.6 \text{ nm} \end{aligned}$$

These simple relationships (in particular the linear relationship between α_0 and g_0) give the strongest possible suggestion that these structured spectral features are due to excited state absorption within the KrF molecule itself. In this case, the initial state for both the gain and the absorption is the KrF B state and the linearity follows directly.

In addition to these spectrally narrow loss features there may also be a broadband loss with a spectral profile large compared to the KrF gain bandwidth which is not directly apparent from the ASE spectra. The magnitude of such a loss can, however, be obtained by the difference between the gain as measured by the probe laser at the peak of the gain profile and the peak gain, g_0L , as calculated from the spectral narrowing. The broadband loss determined by this method as a function of calculated gain is a good straight line giving a gain-to-loss ratio of 2. This value is an overestimate since the gain measuring pulse has a finite bandwidth (0.3 nm), but the correction due to this effect, although significant, is not large.

ABSORPTION SPECTRUM

The above analysis considered the main absorption features as independent but the conclusions drawn suggest that the spectra can also be analysed on the basis of a single absorption spectrum $f_a(\lambda)$ giving an absorption:

$$A(\lambda) = \exp[-\alpha_{d0}L f_a(\lambda)]$$

where the spectrum is normalised to unity at the peak of the "dip". From the above fitting routine we have $\alpha_{d0}L/g_0L = 0.32$ and so:

$$f_a = -\frac{g_0L}{0.32} \ln \left[\frac{M(\lambda)}{B(\lambda)_{\alpha=0}} \right]$$

where $M(\lambda)$ is the measured ASE spectrum and $B(\lambda)_{\alpha=0}$ is the calculated gain narrowed fluorescence spectrum assuming no absorption.

The absorption spectrum so calculated is shown in Figure 5, which is an overlay of 7 spectra taken with widely different measured gains of between 3.6 and 122. The main dip overlays extremely well whereas the data in the wings of the line which are very sensitive to choice of fluorescence spectrum and assumed gain do so less well. The smaller peak to the red at 249.65 nm is a prominent feature on all spectra and there also appear to be small features at 249.29 nm and possibly 249.98 nm.

SIMULATION OF ABSORPTION SPECTRUM

The absorption spectrum obtained from analysis of the ASE spectra shows definite evidence of some structure other than the main dip at 248.91 nm. The striking linearity between the gain and

absorption coefficients strongly suggests that this absorption originates from within the KrF molecule itself. If this is so then the observed absorption spectrum should correspond to a plausible KrF molecular potential.

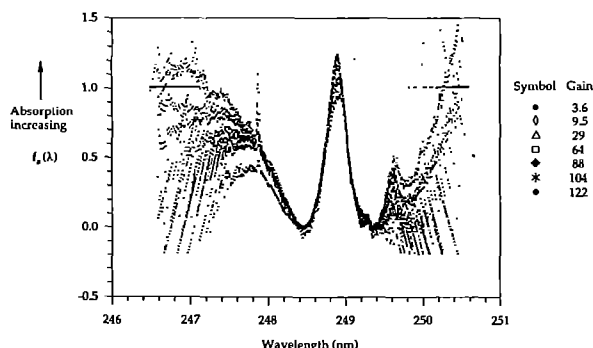


Figure 5 Composite plot of calculated absorption spectra at various gains.

To account for absorption at the observed wavelengths a molecular state at around $80,000 \text{ cm}^{-1}$ above the ground state needs to exist. The lack of fine structure in the absorption spectrum suggests that the potential in question is repulsive, but the fact that the features are much narrower than the B-X fluorescence indicates that the state is considerably less

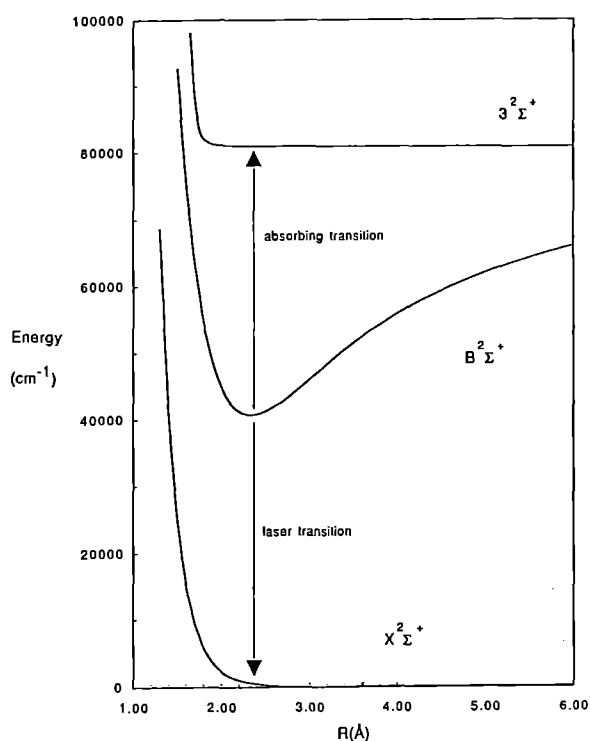


Figure 6. Proposed potential energy curves in KrF

repulsive than the KrF ground state. Thus we are looking for a situation as shown in Figure 6. There are in fact a number of candidates for the state in question which dissociate into excited atoms with energy levels in the region in question. Table 1 lists the possibilities.

Table 1. Energies of separated atoms around $80,000 \text{ cm}^{-1}$

Kr level [config]	F level	Energy
Kr(3P_2) [$4p^5 5s, J=2$]	F($^4P_{3/2}$)	79972.53
Kr(3P_2) [$4p^5 5s, J=2$]	F($^2P_{1/2}$)	80376.54
Kr(3P_1) [$4p^5 5s, J=1$]	F($^4P_{3/2}$)	80917.56
Kr(3P_1) [$4p^5 5s, J=1$]	F($^2P_{1/2}$)	81321.56

The molecular states which result from these separated atoms can be labelled if we assume weak spin-orbit coupling (this is not really a good approximation but is useful for designation purposes). The resultant states from 3P and 2P will be Σ^+ , Σ^- , Π , and Δ , with doublet or quartet multiplicities. Of these possibilities, the important state which is optically connected to the $^2\Sigma^+$ B state will also have $^2\Sigma^+$ symmetry. Following Hay and Dunning [3] we designate this state as $3^2\Sigma^+$.

Preliminary calculations of the absorption spectrum from the $B^2\Sigma^+$ to the $3^2\Sigma^+$ state have been undertaken at The

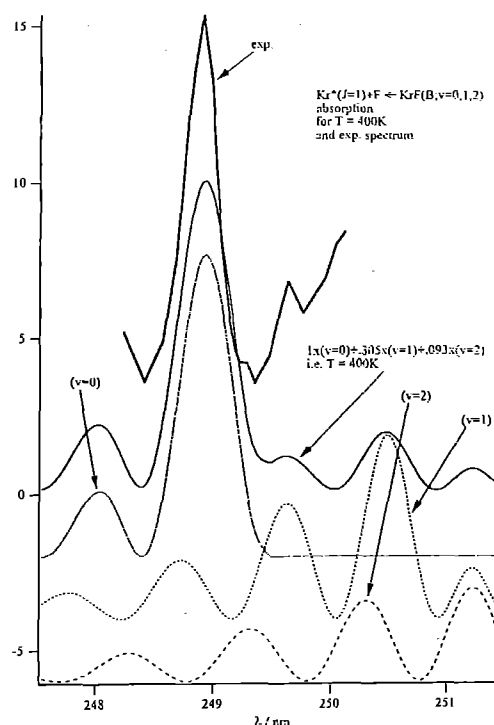


Figure 7. Comparison of simulated and experimental KrF absorption spectra.

University of Iceland. From the nature of the spectra it was clear that the bound potential calculated by Hay and Dunning could not be correct. Born-Mayer repulsive potentials of the form $U(R) = a \exp(-bR) + c$ were tried with the constant c being equal to the separated atom energies given in table 1. The problem is that only a small fragment of the absorption spectrum is available making the unique identification of the potential difficult. As a starting point it was assumed that the prominent dip was due to transitions originating from the $v=0$ level of the B state and the parameters a and b varied to fit this peak. This study is rather preliminary, but a good fit was obtained with the potential:

$$U(R) = 3 \times 10^5 \exp(-3.656R) + 80917 \text{ cm}^{-1}$$

which uses the Kr (3P_1) + F($^2P_{3/2}$) separated atom energy.

A simulated absorption spectrum using this potential is shown in Figure 7. This shows the contributions from the first three vibrational levels separately and the sum for a vibrational temperature of 400 K. The experimental spectrum is shown in bold. The various spectra are displaced vertically for ease of reading. It can be seen that the fit to the experimental absorption spectrum is remarkably good, especially considering the small spectral window over which data is available. The position and width of the main peak are well reproduced as is the zero in absorption at the KrF gain maximum 248.4 nm. The long wavelength peak would appear to be due to the $v = 1$ contribution with enhanced absorption further to the red. However, the rather narrow blue absorption peak does not fit the broad band feature observed experimentally too well and the small feature to the red side of the main peak is not observed. These features may well originate from other states in the same vicinity.

CRYOGENIC GAS CLEAN-UP

The Sprite laser cell is normally connected to two external gas flow loops. One of these has a relatively high flow rate and is used to filter particles out of the gas. The other includes a trap to remove hydrogen fluoride which would otherwise etch the laser windows. To test the effects of cryogenically cleaning the gas mix the diaphragm pump in the particle filter loop was replaced with a commercial temperature-controlled trap and circulator (Oxford Lasers Ltd, model number GP 3000). The cell was filled with a mixture of 5% fluorine in helium (40 torr), krypton (100 torr) and argon (640 torr). Over a period of 30 hours approximately 40 shots were fired with this gas fill while the gain spectrum was measured. The temperature of the trap was then set to 105 ± 1 K and the gas circulator was switched on. (At this temperature the vapour pressure of each of the constituents was higher than its partial pressure in the mix.) The flow rate through the trap was measured to be at least 5 litres/minute and the total gas volume was approximately 100 litres.

The Sprite output spectrum was measured 30 minutes before the circulator was switched on, and was measured again approximately 75 minutes later, with 5 shots having been fired in the interim. After a further 12 shots the laser was shut down. The circulator was left running for 15 hours overnight and the spectrum was measured once more using the first shot of the day.

It was found that there were no there were no significant differences between the three spectra. To within the limits of experimental uncertainty they are identical.

The aim of this part of the experiment was to remove impurities from the laser gas mix and to note any resulting effect. Since there was no observable effect it remains only to establish that any impurities were indeed removed. It was not practical to check this experimentally, but a basic theoretical analysis showed that the trap should have worked well.

PICOSECOND PUMP/PROBE MEASUREMENTS

This section of the experiment was designed to investigate the time-resolved behaviour of the absorption feature following sudden depletion of the gain. A low-intensity, non-saturating short pulse with as large a bandwidth as possible was used to probe the gain in the laser cell before, during or after the passage of a short, intense, saturating pump pulse through the same region of the active medium.

The experiment consisted of firing two or three shots at each different timing of the probe beam, and monitoring the gain and the probe spectrum. Probe spectra recorded for various relative times in the region of the gain depletion step showed several interesting features. Firstly, the amount of gain narrowing between the higher and lower gain shots is smaller than expected from the ASE measurements. Secondly, the change in the magnitude of the dip resulting from the gain depletion is quite small, even with the high-energy pump pulses. The change in the dip produced by lower-energy pump pulses is very slight. The

reasons for these effects are not clear at present. Thirdly, spectra taken at times when the pump and probe pulses overlapped exhibit some broadening, and a change of shape which suggests self-phase modulation of the probe in the presence of the intense pump pulse. If self-phase modulation is present the interpretation of these spectra is particularly difficult, as it is hard to establish to what extent the dip has been filled in by the redistribution of energy in the spectrum.

CONCLUSION

The main conclusion of this work is that the "dip" in KrF is due to internal absorption to an excited molecular state which is identifiable as that dissociating to $\text{Kr}^*(^3\text{P}_1) + \text{F}(^2\text{P}_{3/2})$. The evidence for this conclusion is strong. The lack of removal by cryogenic clean up; the different behaviour with time in the pulse compared with known impurities such as C I; the linear relationship between the absorption coefficient and gain; the constancy of this relationship no matter how the gain is produced and especially the ability to simulate the main features of the observed spectrum by absorption to a highly plausible molecular potential.

There remain, however, a number of questions which this study has not answered or may have raised for the first time. The short pulse saturation results were unexpected and as yet remain inexplicable. It would appear that the gain narrowing effects may be different for short pulses compared to long (stochastic) pulses.

The value for the gain-to-broadband-loss of 2 obtained from the gain narrowing analysis is dramatically lower than the "typical" gain-to-loss ratios which have been measured in KrF (8-12). It must be remembered, however, that these latter figures are for the gain to *non-saturable* loss. If the bulk of the loss measured here is excited state absorption then it will be saturable with the same saturation intensity as the gain. Furthermore, such excited state absorption is not strictly a loss to the KrF excited state population since the excited Kr atoms produced will react with F_2 to yield more KrF^* .

The implications for broadband laser fusion are significant. Clearly, if maximum bandwidth is to be used, some additional spectral filtration to enhance the wings at the expense of the centre will be required. Experiments at Los Alamos have already shown that this is possible. The price to be paid is lower efficiency. Thus tailoring of the spectrum needs to be done early in the chain and certainly before the final amplifier so that energy is not wasted. In typical large installations the final amplifier runs with a gain in the order of a few tens. If the input spectrum is shaped like the fluorescence spectrum then our results show that the gain narrowing will result in a FWHM bandwidth of about 200 cm^{-1} at a gain of 10 reducing to about 75 cm^{-1} for a gain of 100. In either case sufficient bandwidth for adequate temporal averaging of a fusion laser should be available.

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EXPERIMENTAL STUDIES OF A RAMAN AMPLIFIER

C J Hooker, E J Divall, G J Hirst, J M D Lister, M J Shaw and D C Wilson¹

Rutherford Appleton Laboratory

¹Blackett Laboratory, Imperial College, London

INTRODUCTION

Following the installation of the upgraded Raman system on Sprite at the beginning of 1994 [1], and the completion of two experiments using it, the oscillator was reconfigured to generate 25-picosecond pulses, and some experimental studies were carried out to obtain specific performance data to aid the design of the Titania Raman system. Two of these experiments are described here. The first was the measurement of the I/O characteristic of RA3 over an extremely wide range of input energies, with the aims of verifying the recently-developed 2-D code model of Raman amplification, and if possible measuring the small-signal gain of the amplifier directly. The second was an investigation of the principal mechanism for energy loss in the Raman system, namely the generation of second Stokes light at 291 nm.

INPUT/OUTPUT CHARACTERISTIC OF RA3

The set-up used for this experiment is shown in Figure 1. The input energy to RA3 was controlled in two ways. One or more of the pump beams to RA2 could be blocked to reduce its output, and metallized neutral density filters could be placed in the beam between RA2 and RA3, to attenuate the input to RA3. The output of RA2 was measured by an energy monitor, placed behind a turning mirror after the beam-expanding vacuum spatial filter. If used, the filters were placed in the beam after this mirror, to avoid reducing the energy reaching the monitor, and the delay of the Stokes pulse into RA3 was reduced to compensate for the optical thickness of the filters. The measured filter transmission was used to calculate the actual input energy to RA3. By these means the input to RA3 was varied between a few tens of millijoules and a minimum of less than one nanojoule.

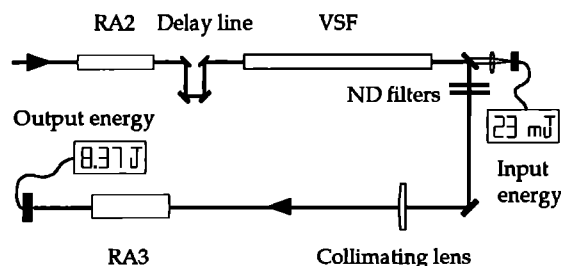


Figure 1. Experimental arrangement used for measuring the input/output characteristic of RA3.

By reducing the input to this level, we hoped to measure the small-signal gain of the amplifier. However, as can be seen from the input/output characteristic shown in Figure 2, the gain is so high that even with inputs as small as 1 nJ, the amplifier is outside the small-signal regime. Unfortunately, the measurement of the output energy was limited by noise in the detector, which did not allow us to follow the behaviour of the amplifier to still lower input energies. A theoretical estimate of the small-signal gain of RA3 under these conditions is 10^{13} , whereas a measurement of the slope of the I/O characteristic in the decade between 1 and 10 nanojoules yields a gain of 5×10^{10} . The measured characteristic is useful, however, in that it shows how the laser output can be controlled over a useful range simply by attenuating the input Stokes beam.

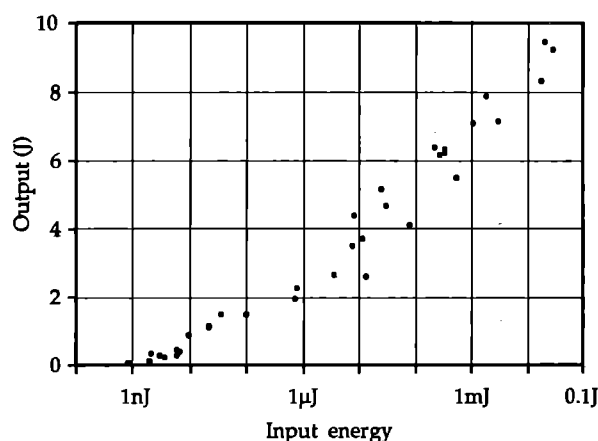


Figure 2. The measured input/output characteristic of RA3.

SECOND STOKES GENERATION

In gaseous Raman media at near-atmospheric pressure the phase-matching angles for parametric generation of high-order Stokes and anti-Stokes frequencies are typically a few milliradians [2]. Although the angles between pump and Stokes beams in the Sprite Raman system are far larger than this, between 1.5 and 5 degrees, we nevertheless found that the generation of off-axis second Stokes radiation at 291 nm was the main cause of reduced output from RA3, with as much as 10% of the output energy lost to second Stokes radiation.

To try to understand, and if possible eliminate, the loss, we investigated the second Stokes emission using the experimental

arrangement shown in Figure 3. The output beam from the amplifier was attenuated using two uncoated glass plates, M1

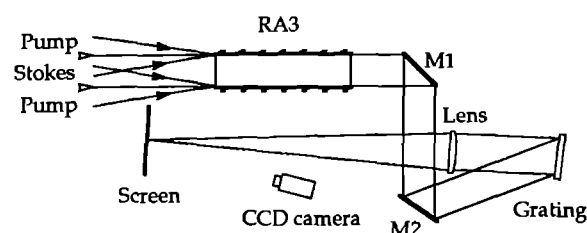


Figure 3. Experimental arrangement used for the investigation of second Stokes emission from RA3.

and M2, each reflecting approximately 10% at 45 degrees, which reduced the maximum energy in the beam to around 100 mJ. A diffraction grating with 3600 lines per mm was then used to isolate the second Stokes wavelength, which was focused onto a fluorescent screen by a lens of 4 metres focal length. Far-field images were recorded using a CCD camera and framestore. The angular scale in the far-field plane was calibrated by recording images of a grid placed in the screen position. The grating was used in an off-Littrow configuration with incidence and diffraction angles of 33 degrees and 18 degrees respectively.

Figure 4 shows two typical far-field images of the second Stokes emission. The main features are a pair of concentric rings, a number of bright spots and a set of eleven extended rays, slightly offset from the axis and oriented along similar directions to the arms of the pump mirror array. Two of these rays are weak, and are hard to see in the figure. The intensities of the spots varied unpredictably from day to day; the two parts of Figure 4, which were recorded on different days, show the degree of variation. The rays are slightly concave towards the axis, which lies at the centre of the rings. Where the rays overlap the ring pattern, they seem to disappear behind the dark ring, although the ring is not opaque but simply a region in which there is no second Stokes light. The elliptical shape of the rings in the images is due mainly to the non-Littrow configuration of the grating, which changes the angular scale in the dispersion direction by a factor $(\cos \theta_i / \cos \theta_d)$. In the geometry used, this factor had the value 0.88. The true angular radius of the dark ring, measured orthogonal to the dispersion direction, was 1.37 milliradians. The ray offset angles between the centre of the ring pattern and the closest point on each ray were also measured, and found to be 23 times smaller than the off-axis angles of the corresponding pump beams.

To study these phenomena further, we recorded a series of far-fields in which different pump beams were blocked one at a time, and the effect on the various features observed. The rings were present on all shots, apparently unchanged by the absence of any single pump beam. The spots and the rays, however, were affected when different pump beams were blocked. In particular, each ray was associated with one of the pump beams, so that

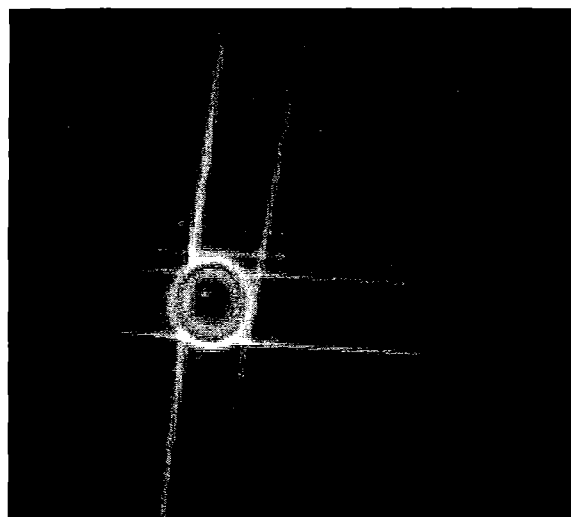


Figure 4. Far-field images of the second Stokes emission at 291 nm, showing the rays (upper image) and the bright spots (lower image), plus the on-axis ring pattern (visible in both).

when that pump beam was blocked, the ray was absent from the far-field pattern. Comparison of the different images with the pump mirror array showed that each ray is oriented at right angles to the plane containing the axis and the corresponding pump beam, and that the locations of the rays relative to the axis mimic the positions of the mirrors in the pump array. The array is not a perfectly regular cross: some of the mirrors in the horizontal axis lie above or below their ideal positions, hence the associated rays are not exactly vertical.

The behaviour of the spots was more complicated. For example, one of the spots in Figure 4 was absent when beam 1, beam 2 or beam 7 was blocked, but reappeared when all three of these beams were present. We also found that the positions of the spots changed from day to day, whereas the rays and rings remained in the same positions throughout the experiment. Each spot is thus associated with three pump beams rather than only one, and the positions of the spots are extremely sensitive to the small angular changes made during the daily alignment of the laser system.

The spread of angles in the rings and rays, and the faintness of all three phenomena relative to the output Stokes beam, suggest that they can be explained by interactions of the primary pump or Stokes beams with light scattered from the amplifier windows

or elsewhere. Weak second Stokes radiation generated by such interactions will be strongly amplified by the normal Raman gain process, with the amplified first Stokes beam now acting as the pump; indeed, without amplification the features would probably be too faint to observe. The gain will be high over a range of forward directions, allowing the amplification of angularly-extended features.

The axial symmetry of the rings implies that the primary Stokes beam is one of the interacting components, and we therefore suppose the other to be diffusely scattered KrF pump light. The wave-vector diagram in Figure 5(a) illustrates the phase-matching condition $2\mathbf{k}_s - \mathbf{k}_{p'} = \mathbf{k}_{2s}$; here and elsewhere, primed subscripts denote scattered waves. In reality the angles α and β are both around 1.5 milliradians, and are exaggerated in the figure for clarity. Phase-matching will occur for any pump

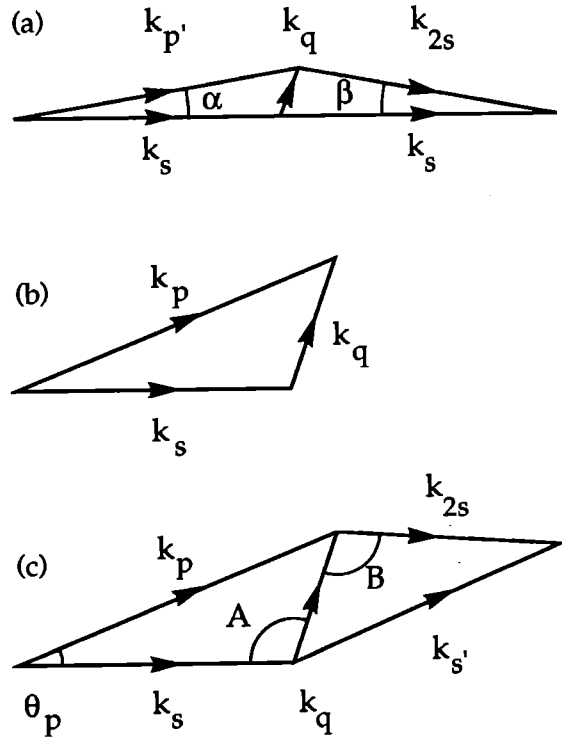


Figure 5. Phase-matching diagrams for (a) formation of the rings, (b) generation of the phonon wave in the Raman medium and (c) formation of the second Stokes rays.

radiation scattered at an angle α to the axis, and hence the second Stokes will be generated in a cone of semi-angle β around the first Stokes propagation direction. The size of β is sensitive to the composition of the gas mixture in the amplifier, which was a mixture of methane with between 25% and 40% of helium. Values of β calculated for these two compositions were 1.32 and 1.49 milliradians respectively, and the measured value of 1.37 milliradians is consistent with an intermediate percentage of helium.

The double ring structure is due to the well-known suppression of SRS gain at the exact Stokes/anti-Stokes phase-matching angle [3, 4], which corresponds in our case to second Stokes/KrF phase-matching, since the amplified first Stokes is acting as the pump. The gain will be suppressed for all second Stokes radiation propagating at an angle β to the axis, irrespective of how it was generated, which explains why there are also gaps in the inner rays where they intersect the dark ring.

The bright spots in the second Stokes far-field were found only near the axis, and they were brightest when close to one of the rings, i.e. when near the phase-matching angle. This suggested that the same four-wave mixing process as for the rings might be responsible, if pump light was being scattered in specific directions. We believe the mechanism for this is the formation of transient diffraction gratings in the entrance window of the amplifier, due to the non-linear refractive index of silica and the high intensities of the pump beams. The variation in intensity in the interference fringes between a pair of beams modulates the refractive index, causing a phase modulation across the wavefront of a third beam. With two beams of energy 1.5 joules, diameter 10 cm and pulse length 25 ps, the peak intensity in the fringes is 3.0 GW cm^{-2} . The phase change (B integral) due to this intensity in a fused silica window 0.6 mm thick is 0.021 radians, so the interference pattern will form a sinusoidal phase grating with approximately 0.3% modulation.

Where the two beams forming a grating and the beam that will be diffracted all lie on either the vertical or the horizontal arm of the pumping array, it is possible for the diffracted order to propagate nearly parallel to the axis. Calculations using the measured off-axis angles of the pump beams showed that four combinations involving beams 1, 2, 7 and 12 in the vertical arm of the cross gave diffracted beams very close to the axis, which is consistent with our observation that beams 1, 2 and 7 were all essential for the formation of one of the spots. Small changes to the pump beam angles during alignment, typically a few tenths of a milliradian, would move the diffracted beams by similar amounts; such changes are very similar to the observed day-to-day movements of the spots.

The explanation of the rays must be different from the above, in that each ray is associated with a single pump beam. By hypothesis, the observed spread of emission directions in each ray requires one of the interacting waves to be diffusely scattered, and since the pump beams have unique directions the scattered wave must be the Stokes. A single pump beam and the axial Stokes beam together drive an optical phonon in the Raman medium, with a wave vector $\mathbf{k}_q$ satisfying the relation $\mathbf{k}_p - \mathbf{k}_s = \mathbf{k}_q$, as shown in Figure 5(b). The stimulated scattering of each pump beam from its associated phonon is the mechanism of amplification of the Stokes beam. A diffusely-scattered Stokes wave can itself be scattered from the phonon to generate second Stokes, if the phase-matching condition $\mathbf{k}_{s'} - \mathbf{k}_q = \mathbf{k}_{2s}$ is satisfied: the vector diagram for this is shown in Figure 5(c). The magnitudes $k_{s'}$ and k_{2s} are such that phase-matching is possible for all pump beam angles θ_p in our

system. Although k_q is fixed for a given pump beam, the right-hand triangle in Figure 5(c) can be rotated freely around k_q , and hence the second Stokes can be emitted in a broad cone of directions, some of which lie close to the axis and appear as a slightly curved line in the far-field. The strongest emission is restricted to forward directions relatively near to the axis by both the predominance of forward scattering of the Stokes, and the need for the second Stokes to be amplified by the co-propagating Stokes in order to reach an observable intensity.

The mechanism described above accounts for the slight curvature of the rays and their orientation with respect to the pump beams. It also accounts for the offset of the rays from the axis. Knowing the magnitudes of k_p , k_s and k_{2s} in Figure 5(c), the angles A and B can be found for any pump beam angle θ_p . The calculated ray offset angle A - B was found to be 1/23 of the pump beam angle, exactly as observed; this angle is plotted against θ_p as the solid line in Figure 5, and the points are the measured offset and pump beam angles for each ray. The fact that A is slightly larger than B means that the rays will appear on the same side of the axis as the corresponding pump beams, and will be concave towards the axis, as observed.

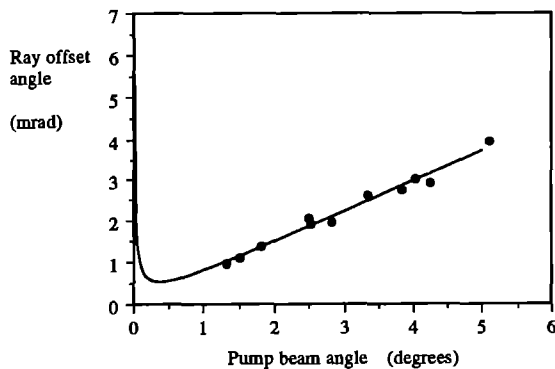


Figure 6. Comparison of the measured ray offset angles for each pump beam (points) with the prediction of the model (solid line).

We have identified three mechanisms responsible for the generation of off-axis second Stokes emission in our Raman amplifier. In each case, some form of scattering is involved, and thus the elimination of the scattering would reduce or eliminate the loss. The diffuse scattering is probably due to dust particles in the Raman gas, which could be removed by filtering. Scatter from the entrance window of the cell and Rayleigh scattering in the gas itself will together set a lower limit on the losses. There is no way to prevent the formation of gratings in the windows, but a careful choice of pump beam angles will ensure that none of the diffracted orders lie close enough to the phase-matching direction to cause significant losses. These results will be taken into account in the design of the Titania Raman system.

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RAMAN MULTIPLEXING - A PROOF OF PRINCIPLE EXPERIMENT

G J Hirst, E J Divall, K N Drodge, C J Hooker, J M D Lister and M J Shaw

Rutherford Appleton Laboratory

INTRODUCTION

The implementation on Sprite of forward Raman multiplexing, which allows efficient extraction of a large KrF laser while using only a relatively small number of transport optics, was discussed by Edwards, O'Neill and Shaw as long ago as 1983 [1]. More than ten years later, however, the concept had still not been experimentally checked. Since Raman multiplexing is central to the design of the Titania laser [2] it was decided in 1994 to carry out a proof of principle test. The aim was to see whether multiplexing would compromise the Raman system's performance, which is usually characterised by exceptional beam quality, good conversion efficiency and very low prepulse [3]. Unfortunately prepulse measurements are hard to make [4] and would have needed major changes to the laser set-up. This experiment therefore concentrated on using the existing Sprite hardware to study the beam quality and conversion efficiency of a multiplexed Raman system.

EXPERIMENTAL APPARATUS

The layout is shown in Figure 1. The KrF chain was the same as that used for normal Raman operation [3] except that a pulse-stacker was added before Goblin to put a second, delayed pulse onto each of the beamlines. The spacing between the pulses was 1.5 ns, the same as will be used in the Titania system. The Stokes beamline was more extensively altered. The output from RA2 was split into two pulses, the first of which was expanded to a diameter of 8 cm and passed through RA3 at an angle of 1.6° to the axis, synchronised with the first set of pump pulses. Its energy after amplification was expected to be good, but its non-optimal transport optics and the lack of any spatial filtering would have degraded its beam quality. The second pulse from RA2 followed the normal path into RA3, via a VSF and telescope with corrected optics. It then passed to the diagnostics suite which included calorimeters, measuring the energy in both Stokes pulses, and a CCD far-field monitor, looking only at the focal spot of the second one. This would reveal whether the presence of the first pulse led to any reduction in the beam quality of the second.

RESULTS

The first experiment involved measuring the KrF output energy from Sprite with the reduced (1.5 ns) pulse spacing. This was found to be

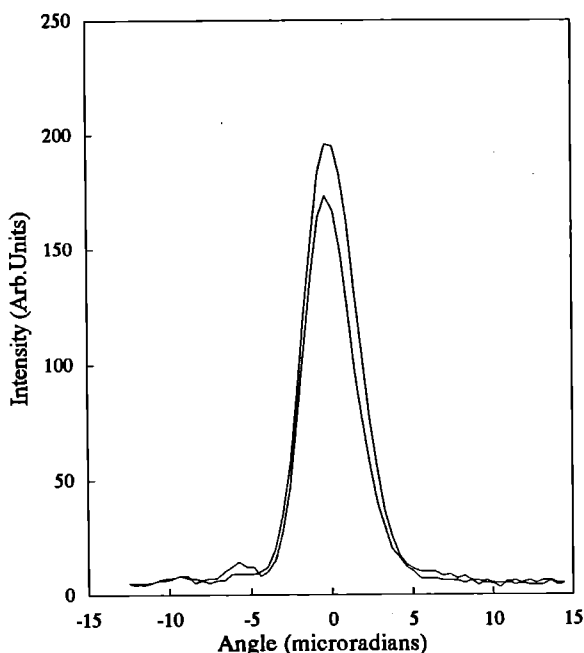


Figure 2 Profiles of the Stokes far-field with (lower curve) and without (upper curve) Raman multiplexing

0.97 J per pulse, compared with the 1.25 J per pulse when the spacing was 3 ns. Part of the fall was due to the reduced input to Sprite, the output from Goblin having dropped by about 12% per pulse. The remainder was due to the incomplete recovery in 1.5 ns of the Sprite KrF* population. This result was in good agreement with earlier model predictions.

Measurements of the Stokes energies leaving RA3 were made next. The best shot (from a small dataset) with no multiplexing and a 3 ns KrF spacing had an RA3 pump energy of 14.9 J and a Stokes output energy of 6.5 J, giving an overall conversion efficiency of 44%. The average results from the three best Raman multiplexed shots had energies of 4.9 J in the first Stokes pulse and 4.7 J in the second. With an average total pump energy of 21.9 J the efficiency was again 44%.

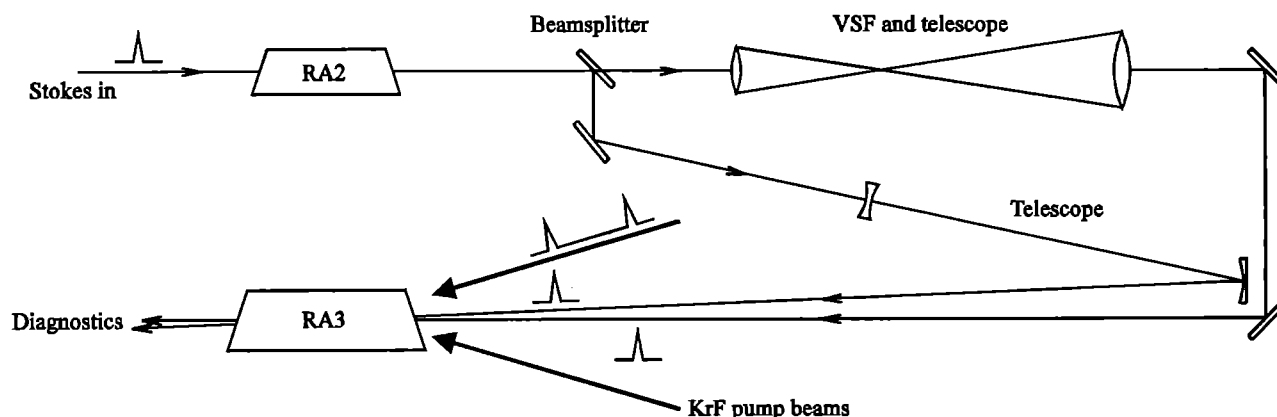


Figure 1 Layout of the experimental apparatus

It therefore appears that Raman multiplexing does not significantly affect the amplifier's energy conversion efficiency.

The effects of the first pulse on the far-field of the second can be seen in Figure 2. The profiles of the second pulse focal spots, in the presence and absence of the first pulse, have the same FWHM width ($\sim 3.8 \mu\text{rad}$) so multiplexing has caused no significant increase in the divergence of the "good-quality" part of the beam. The peak heights of the profiles (168 counts and 191 counts) reflect, fairly closely, the measured pulse energies (4.9 J and 6.5 J). The ratio of these two quantities is a measure of the fraction of the total energy which is within the good-quality focal spot, and the data suggest that this fraction is actually slightly larger when the first pulse is present. This surprising result probably arises from a combination of measurement uncertainties and the fact that the Stokes energy is somewhat lower when the amplifier is multiplexed, so nonlinear scattering effects are reduced. In any event it seems that multiplexing does not increase the fraction of the energy scattered out of the central spot.

CONCLUSIONS

The experiment has shown that the Sprite KrF amplifier can be operated with a pulse spacing of 1.5 ns and that it then delivers the expected energy per pulse. It has also shown that simple two-beam Raman multiplexing does not lead to any reduction in the Raman amplifier's energy extraction efficiency nor to any degradation in the beam's optical quality.

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COHERENCE EFFECTS AND BANDWIDTH OPTIMISATION IN MULTIFREQUENCY RAMAN GENERATION

G S McDonald¹, G H C New¹, L L Losev², A P Lutsenko² and M J Shaw³

¹Imperial College of Science, Technology and Medicine

²P.N. Lebedev Physical Institute

³Rutherford Appleton Laboratory

INTRODUCTION

Stimulated Raman scattering (SRS) is well-established as a simple and efficient method of converting laser radiation to one or more lower (Stokes) frequencies. However, parametric Raman conversion to higher frequencies, or the simultaneous generation of multiple Raman lines, has generally been found to be much less efficient. The common assumption has been that, for optimal conversion of the input energy, the phase-matching conditions for the parametric processes dictate the geometry of the interacting waves. Radiation is therefore generated over a wide range of angles, resulting in low interaction lengths and a multifrequency "beam" that cannot be focused to a single spot. We have proposed that the collinear generation of higher orders, in the presence of finite dispersive phase mismatch and with fully symmetric pumping (input pulses of matching intensity and shape), has much greater potential than phase-matched generation. Results to date predict that the generation of a single multifrequency beam consisting of nearly 50 waves of comparable energy is possible^{1,2,3}. The most immediate application of this work is in the field of inertial confinement fusion (ICF) where the use of a broadband light source may overcome several problems related to the transmission and delivery of high optical energy⁴. However, the predicted magnitude of bandwidth generated is such that our results may also find application in other areas such as plasma physics, measurement techniques, spectroscopy and sensing.

In multifrequency Raman generation, an optical beam initially containing 2 frequency components (ω_0 and ω_{-1}) drives a Raman resonance in a nonlinear medium at the difference frequency $\omega_R (= \omega_0 - \omega_{-1})$. As a result, the incident energy is distributed among the Raman orders $\omega_n = \omega_0 + n\omega_R$, situated on both the red and blue sides of the incoming radiation ($n = 0, \pm 1, \pm 2, \dots$). To model this process, the total electric field is expanded in terms of the constituent plane waves. For the propagation of the n^{th} normalised electric field envelope, A_n , and the dynamics of the polarisation wave, P , one finds

$$\begin{aligned} \frac{\partial A_n}{\partial Z} &= \frac{\omega_n}{2\omega_0} [P^* A_{n+1} e^{-i\Gamma_{n+1}Z} - P A_{n-1} e^{i\Gamma_{n-1}Z}] \\ \left(\frac{T_2}{t_p}\right) \frac{\partial P}{\partial \tau} &= -P + \sum_j A_j A_{j-1}^* e^{-i\Gamma_j Z} \end{aligned} \quad (1)$$

$Z = \gamma I_p z$ is a gain-length product, γ is the Raman gain coefficient, I_p is the peak input intensity, τ is local time (in units of input pulse width t_p), and T_2 is the medium dephasing time. Dispersion gives rise to a set of finite values of normalised mistuning, Γ_n , which can be parametrised by a single value, $\Gamma_1 = (k_1 + k_{-1} - 2k_0)/\gamma I_p$. Here, we take the input fields to be either Gaussian or square pulses and results are presented for rotational SRS in H_2 gas pumped by the second harmonic of a Nd:YAG laser.

COHERENCE EFFECTS

We have found that two distinct regimes of multifrequency generation exist - the coherent and incoherent regimes⁵. Towards the steady-state limit, $t_p/T_2 \rightarrow \infty$, distinct temporal regions become decoupled (incoherent) and are, ultimately, independent. In this limit, square input pulses lead to square output pulses. However, for pumping with Gaussian pulses one finds that the generated Raman waves are modulated with rapidly-varying envelopes which are a direct consequence of the input pulse shapes. Furthermore, the presence of finite coherence time and finite dispersion can result in highly complex patterns in the time domain - see Fig 1(a). In ICF high gain targets require collisional absorption to be the dominant process in the laser-target coupling. Experiments have shown that reducing the coherence of the incident light can suppress laser-driven plasma instabilities. On the other hand, analyses have predicted that a simple increase in the spectral bandwidth can also increase thresholds and lower growth rates for such instabilities. Thus the use of light which is both incoherent and ultra-broadband may be optimal for this application.

Considering cases where $T_2 > t_p$ (the coherent regime), we find that well-defined pulse trains dominate both the Stokes and anti-Stokes orders⁶. For square input pulses, the generated waveforms are further simplified - see Fig 1(b). These pulse trains appear for both Gaussian and square input pulses; they are intrinsic to the nonlinear dynamics of the system and are not a simple consequence of input pulse shape. Indeed, these spontaneous structures are robust Raman soliton pulse trains, which result from the self-organisation of many interacting waves⁶.

BANDWIDTH OPTIMISATION

We have undertaken a full investigation of the roles played by dispersion, gain-length product, transiency and the temporal characteristics of the incident light fields. For dispersionless propagation we find that the effect of transiency is to increase the bandwidth generated - see Fig 1(c). This follows from the fact that the polarisation wave is no longer constrained to follow its source term adiabatically and can continue to grow even while its driving term falls to zero. For sufficient Z , the bandwidth is maximised in the coherent regime where the broadband spectrum locks on to the soliton pulse train solution. In the incoherent regime, the combined effect of transiency and moderate dispersion enhances the output bandwidth since strong parametric and non-parametric processes can contribute to the frequency conversion². As can be seen in the case of $t_p/T_2 = 4$, the generated bandwidth can be increased by around 50%. However, when either transiency or dispersion is too strong we find that the net effect is to suppress broadband generation.

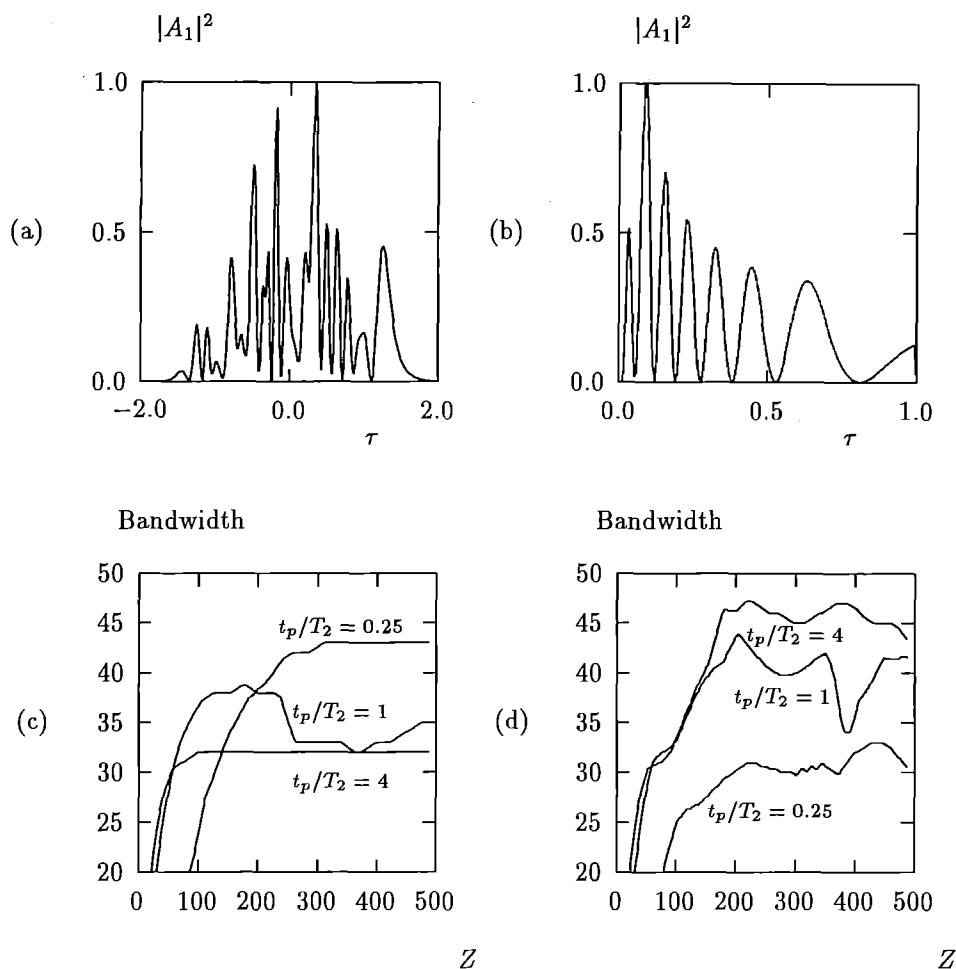


Fig 1 Characteristics of the generated Raman spectra. (a) and (b) show normalised intensity profiles at the anti-Stokes frequency ($n = +1$). (a) is for Gaussian input pulses in the incoherent regime with $t_p/T_2 = 4$, $Z = 250$ and $\Gamma_1 \approx 2 \times 10^{-3}$ ($\gamma I_p = 1 \text{ cm}^{-1}$, 1 atmosphere of H_2 gas). (b) is for square input pulses in the coherent regime with $t_p/T_2 = 0.25$, $Z = 250$ and $\Gamma_1 = 0$. (c) and (d) show the generated bandwidth (in units of ω_R) as a function of the gain-length product, Z . Results for three levels of transiency, $t_p/T_2 = 4, 1$ and 0.25 are shown. In (c) $\Gamma_1 = 0$ (the dispersionless case) while (d) is for $\Gamma_1 \approx 2 \times 10^{-3}$. Note that for low Z bandwidth is relatively independent of moderate levels of dispersion.

In multifrequency generation the key quantity is the parametric gain¹. A simple expression, which is proportional to the maximum possible value of this quantity, has been found to be a good qualitative predictor of the dependencies of the generated bandwidth in the regime of unsaturated amplification⁷. This expression is

$$P_{max}(T) = \exp(-T) \int_{-\infty}^T \exp(T') A_0(T') A_{-1}(T') dT' \quad (2)$$

where T is local time in units of the dephasing time T_2 . Simulations have been performed to examine dependencies on parameters such as the input Stokes amplitude, the input pulsewidth ratio and the relative timing of the pump pulses. The correlation of bandwidth generated with the above expression demonstrates the importance of spatiotemporal overlap of the input pulses for efficient multifrequency generation and, consequently, why symmetric pumping is optimal for maximising the output bandwidth.

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INVESTIGATION OF THIRD-HARMONIC GENERATION OF AMPLIFIED AND STRETCHED FEMTOSECOND PULSES

D T Reid, M Ebrahimzadeh and W Sibbett

The J F Allen Research Laboratories, North Haugh St Andrews University, St Andrews, Fife KY16 9SS

INTRODUCTION

Experimental and theoretical studies of second, third and fourth harmonic generation of femtosecond Ti:sapphire laser pulses have already been described in the literature [1-3]. The experimental systems [1,2], which both rely on unamplified laser sources, first derive second harmonic of the fundamental radiation by frequency-doubling in a critically-phasematched LBO crystal at room temperature. The third harmonic is then produced by sum-frequency-mixing the second harmonic and the fundamental in a critically phasematched BBO crystal. One system which uses a 1.75 W average power laser [1] has achieved a conversion efficiency of 7% from the fundamental to the third-harmonic. A scheme using BBO to generate both the second and third harmonic has been studied theoretically using a method which includes group velocity dispersion effects [3].

SPRITE FRONT-END SYSTEM

The calculations presented later assume that the fundamental radiation is derived from a 20 mJ transform-limited 100 fs pulse which is stretched to a duration of $\tau \approx 50$ ps. The centre wavelength of the resulting pulse is taken as 746 nm, implying a FWHM bandwidth of 6.0 nm. Variation of the instantaneous frequency across the stretched pulse is therefore very slow since the phase-shift due to dispersive broadening introduced by the stretcher is distributed over $\approx 20,000$ optical cycles. The beam diameter after stretching is 3.0 nm and corresponds to an intensity of 5.7 GW/cm². A summary of the parameters of the fundamental pulse is given in Table 1.

Stretched Pulse Parameter	Value
Intensity	5.7 GW/cm ²
Energy	20 mJ
Duration	50 ps
Centre wavelength	746 nm
FWHM bandwidth	6 nm
Beam diameter	3.0 nm

Table 1. Parameters of the fundamental radiation

SECOND-HARMONIC GENERATION SCHEME

Frequency-doubling from 746 nm $\rightarrow$ 373 nm can be carried out in a number of nonlinear materials but only BBO and LBO offer sufficient resistance to optical damage together with high

Crystal	Θ	Φ	o-ray	e-ray	d_{eff}	ρ_E	$\Delta\lambda_{746\text{nm}}$	$I_{MAX}^{(1\text{ ns})}$
β BBO	31.4°	90°	//y	//xz	1.9 pm/V	4.1°	4.0nm.mm	13.5 GW/cm ²
LBO	37.2°	0°	//z	//xy	0.7 pm/V	0.9°	6.3nm.mm	18.9 GW/cm ²

Table 2. Parameters for 746 $\rightarrow$ 373 nm second-harmonic generation

Crystal	Θ	Φ	o-ray	e-ray	d_{eff}	ρ_E	$\Delta\lambda_{746\text{nm}}$
β BBO	49°	90°	//y	//xz	1.4 pm/V	5.0°	2.0nm.mm

Table 3. Parameters for third-harmonic generation.

nonlinear coefficients. These crystals were compared on grounds of phase-matching, effective nonlinear coefficient, walkoff angle, acceptance bandwidth and damage threshold. Calculations were made using theory described in detail elsewhere [4,5,6] and are based on Sellmeier refractive index expansions published for BBO [7] and LBO [8]. Axes conventions used in the description of the nonlinear interactions are adopted from Roberts [4]. A summary of the results of calculations concerning second-harmonic generation is contained in Table 2.

In the plane-wave limit, conversion efficiency for second-harmonic generation is given by,

$$\frac{P_2}{P_1} = 8 \left(\frac{\mu_0}{\epsilon_0} \right)^{3/2} \frac{\omega_1^2 a_{MKS}^2 I^2 I_1}{n^3}$$

In the absence of focusing, BBO is superior to LBO because of its higher nonlinear coefficient. However, if focusing was considered to be necessary, the large walkoff angle of BBO would reduce conversion efficiency considerably. Bandwidth considerations favour LBO which could support the pulse in a crystal of length 1.0 mm compared to BBO which requires lengths shorter than 0.7 mm. Taking these considerations into account, in the plane-wave limit, BBO has a figure-of-merit 3 times greater than that of LBO.

The conversion efficiency obtained from a 1mm-thick BBO crystal was modelled by solving the coupled amplitude equations. A Gaussian pulse described by the parameters given in Table 1 was used as the fundamental radiation. Beam and crystal parameters were taken from Tables 1 and 2 and a 1 mm thick crystal was used. Results taken when the chirp on the fundamental pulse due to the stretcher is neglected are displayed as Figure 1 and show the amplitude of the original (dashed) and depleted pump pulse (a) and the second-harmonic pulse (b). Results from the model in the presence of the chirp introduced by the pulse stretcher (c, d) show no change in the pulse amplitudes. The power conversion efficiency from the fundamental to the second-harmonic predicted by the model is estimated from the ratio of the pulse intensities to be around 40%.

THIRD-HARMONIC GENERATION

Crystal details for generation of the third-harmonic by sum-frequency mixing the pump and second-harmonic are displayed in Table 3. Only BBO can be used since LBO only phasematches for fundamental wavelengths longer than 785 nm. There are no other common materials providing suitable uv-transparency, a high damage threshold and large nonlinear coefficients.

The smaller bandwidth of BBO for third-harmonic generation requires the use of a shorter crystal than was necessary for frequency-doubling. Sum-frequency generation in a 500 μm thick crystal was studied by mixing the depleted fundamental and second-harmonic outputs from the frequency doubling model to produce a third-harmonic output. The original (dashed) and depleted inputs to the mixing stage are given in Figure 2 (a, b) and the resulting third-harmonic pulse is shown in (c). The conversion efficiency from the original fundamental pulse to third harmonic is estimated from the ratio of intensities to be around 25%.

CONCLUSIONS

In the absence of focusing, the material of choice for the frequency-doubling section is βBBO and conversion efficiencies from the fundamental of 40% are predicted in a 1 mm crystal. Chirp introduced by the pulse stretcher is not expected to affect conversion efficiency. Third-harmonic generation in a 500 μm thick crystal should achieve conversion from 746 nm $\rightarrow$ 249 nm with an efficiency of 25%.

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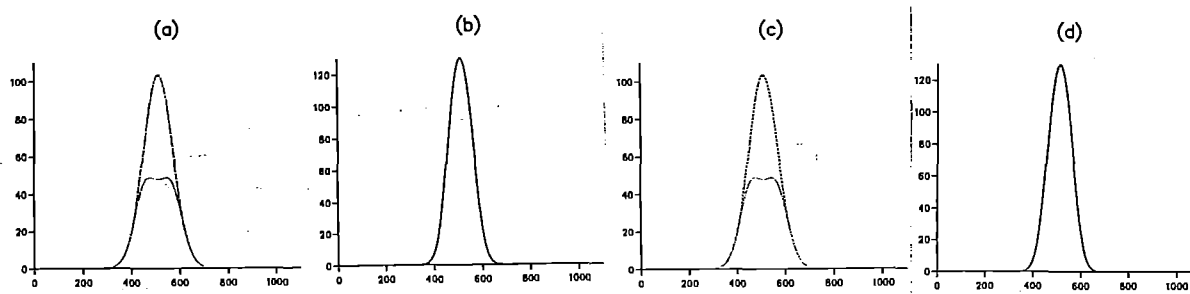


Figure 1. Amplitude of the fundamental pulse before (dashed line) and after (solid line) the frequency-doubling stage and the resulting second-harmonic pulse neglecting chirp (a, b) and including chirp (c, d). Units of amplitude of the fundamental (a, c) are 2 $\text{V}/\mu\text{m}$ and of the second-harmonic are $\text{V}/\mu\text{m}$. Units of time are 500 fs. Crystal length is 1 mm.

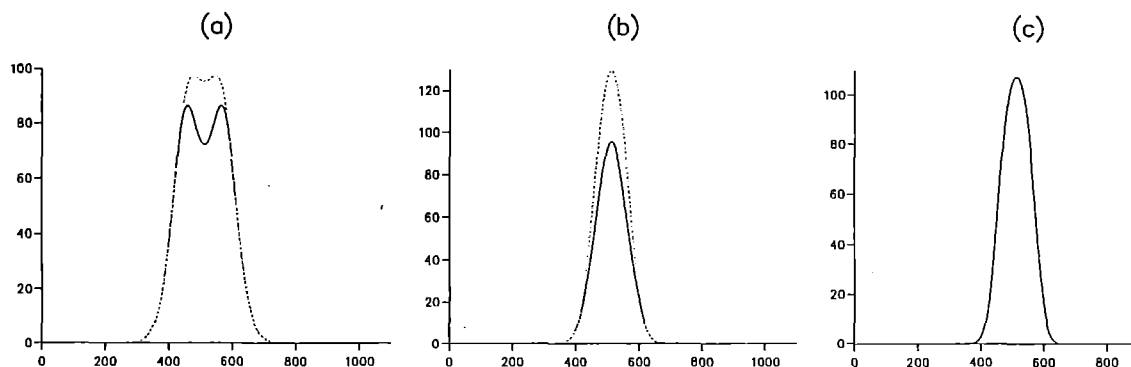


Figure 2. Amplitudes of the input pulses to the sum-frequency mixing stage before (dashed lines) and after (solid lines) the crystal (a, b) and resulting third-harmonic pulse (c). Units of amplitude are $\text{V}/\mu\text{m}$. Units of time are 500 fs. Crystal length is 0.5 mm.

COMMISSIONING OF THE TITANIA PULSED POWER SYSTEM

A K Kidd, A P Visser and M J Shaw

Rutherford Appleton Laboratory

PULSED POWER SYSTEM

This report describes the commissioning of the Titania pulsed power system and in particular the performance of the diodes and electron beam energy deposition, efficiency and uniformity.

DIODE PERFORMANCE

The design of the Titania diodes is described in detail elsewhere¹. Each of the four 4Ω output lines bifurcate into two 8Ω sections to feed through into the diode vacuum chamber. Pairs of feeds are connected in parallel to the cathode support structure which is in turn divided into two cylindrical 8Ω sections. The 8Ω diodes are octagonally arranged around the laser cell to improve e-beam energy deposition uniformity.

The Titania diodes have been operated up to the full design voltage of 600kV. Figure 1 shows a comparison between the experimental diode voltage and a simulated waveform obtained from the transmission line code, BERTHA². The FWHM of the voltage pulse is measured to be 175ns. At a diode voltage of 520kV, the average current per feed is 77kA, 3% above the design value of 75kA.

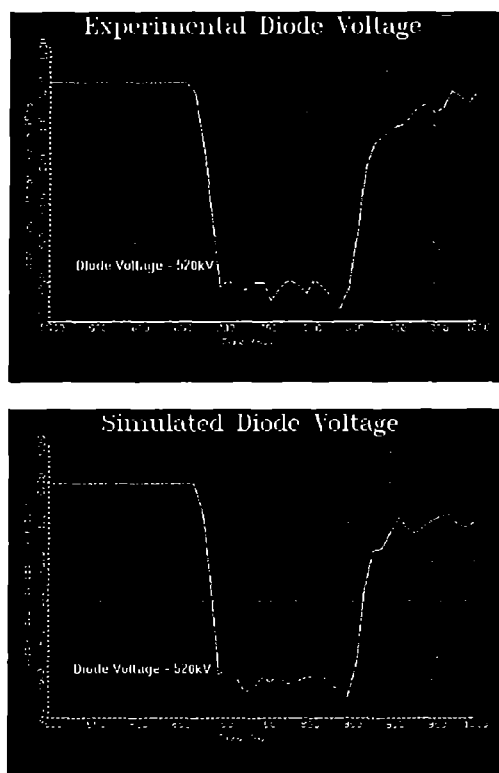


Figure 1. Experimental (upper trace) and simulated diode voltage. Vertical scale: 100kV/div; Horizontal scale: 50ns/div.

Foil lifetime is an important parameter from an operations standpoint. Figure 2 shows the diode pressure obtained over the period of two foil lifetimes. Initial diode pressures are typically 5×10^{-6} mbar.

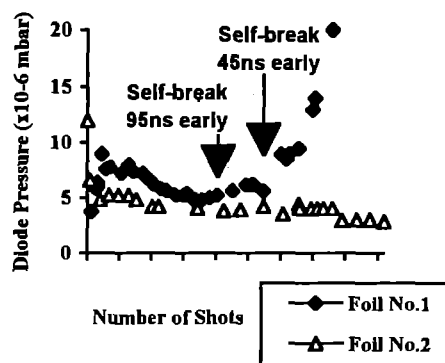


Figure 2. Diode pressure vs shot number
Horizontal scale: 50 shots/div

The first foil tested on Titania survived 400 shots. From Figure 2, it can be seen that diode pressure increases following pre-firing of one of the four output switches. Two pre-fires at 45ns and 95ns before the other three switches results in increasing diode pressure and leads rapidly to foil failure. During lifetime testing of the second foil, care was taken to keep switch jitter to a minimum - for synchronous firing of the diodes - and in particular to avoid pre-firing of the switches. To date this foil has survived 460 shots without failure or significant rise in diode pressure.

ELECTRON BEAM ENERGY DEPOSITION

The design specification for the Titania laser vessel indicates that 20kJ of e-beam energy be deposited in the Ar/Kr mixture at a diode voltage of 600kV. It is also required that the energy deposition profile be reasonably uniform without excessive hot spots or unpumped volumes.

The pumped length of the Titania amplifier is 150cm, leaving 0.3m unpumped at each end. The clear laser aperture is 42cm diameter implying a pumped volume is $0.39 \times 10^6 \text{ cm}^3$. 20kJ in 168ns into a volume of $2.08 \times 10^5 \text{ cm}^3$ corresponds to a specific pump density of 0.57 MW cm^{-3} .

In order to characterise electron beam behaviour within the Titania laser vessel, the cell is filled with various KrF gas mixes. The electron beam energy deposition, efficiency and beam uniformity are investigated as a function of:

1. Total pressure
2. Partial krypton pressure
3. Diode voltage.

Energy Deposition

The Titania e-beam enters the gas through a titanium anode foil 12 μ m thick and a pressure foil 35 μ m thick bonded to a cylindrical support structure. The Hibachi foil-support structure has an 88% geometrical transparency. The amount of energy deposited in the vessel manifests itself as a rise in pressure of the target gas. This pressure jump, ΔP , is related to the magnitude of the deposited energy, E , by:

$$E(J) = 60\Delta P(\text{torr}) \quad (1)$$

and is recorded by means of a baratron. However, the deposited energy recorded by the pressure jump monitor does not include radiative losses. Measurements performed on KrF gas mixes indicate that a correction factor must be applied to recorded energies of approximately 25%³.

The krypton partial pressure is maintained constant at 300 mbar whilst the total pressure is increased from 1050 to 1350 mbar. The Marx charge voltage was varied in 10kV steps from 60 to 80kV in each sequence of shots. The pressure jump was recorded and Equation (1) is used to convert pressure jump readings into deposited e-beam energies for specific charge voltages. Figure 3a shows the variation in deposited energy with Marx charge voltage, for cell pressures of 1050, 1150, 1250 and 1350mbar. The krypton partial pressure is maintained constant at 300mbar.

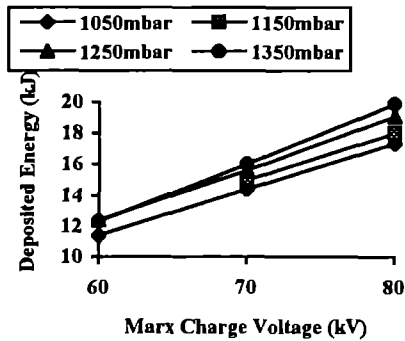


Figure 3a. Variation in E-beam energy deposition with Marx charge voltage (partial pressure of krypton = 300mbar; different cell pressures)

Figure 3b shows the variation in deposited energy with cell pressure for Marx charge voltages of 60, 70 and 80kV. Note that at no combination of cell pressure and Marx charge voltage does the deposited energy exceed 20kJ for a partial pressure of krypton of 300 mbar.

Next, the krypton partial pressure is increased in 100mbar steps from 300 to 600 mbar corresponding to a variation in total pressure from 1050 to 1350 mbar (Figure 3c). The Marx charge voltage is kept constant at 80kV in each sequence of shots.

Finally, Figure 3d shows the variation in deposited energy with krypton partial pressure over the range 300 to 600 mbar for a constant cell pressure and Marx charge voltage of 1350 mbar and 80kV.

From Figure 3d, the optimum energy deposition is 21kJ in 150/600/600 mbar (Ar:F2/Kr/Ar) at 80kV Marx charge voltage. A deposited energy in excess of 20kJ is achieved for partial pressures of krypton of 400 mbar and above at total cell

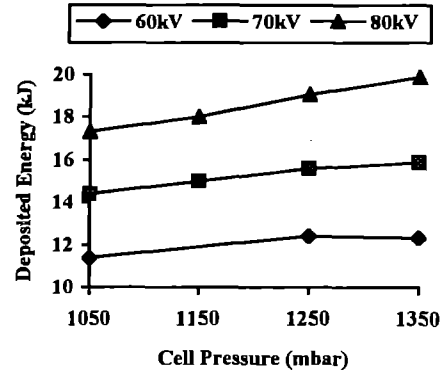


Figure 3b. Variation in E-beam energy deposition with cell pressure (partial pressure of krypton = 300mbar; different Marx charge voltages)

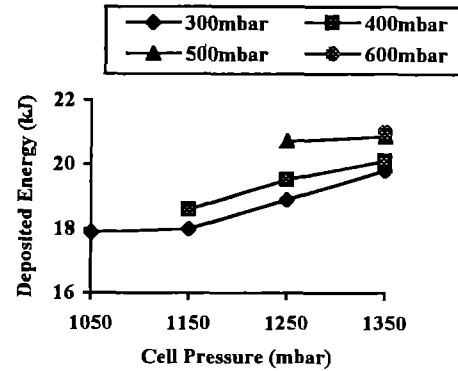


Figure 3c. Variation in E-beam energy deposition with cell pressure (different krypton partial pressures; Marx charge voltage = 80kV)

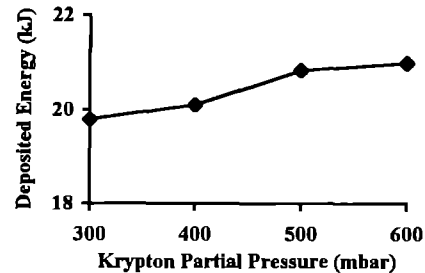


Figure 3d. Variation in E-beam energy deposition with krypton partial pressure (cell pressure = 1350mbar; Marx charge voltage = 80kV)

pressures of 1350 mbar. In order to decrease the pressure loading on the cell, the operating pressure of the cell is decreased whilst maintaining the partial pressure of krypton at 600 mbar. Extrapolating the curve of Figure 3c implies that at 1200 mbar total pressure, in excess of 20kJ will be deposited in the gas. A Marx charge voltage of 80kV results in a voltage on the diode of only 520kV. This is well below the design value of 600kV. Other key components in the system (switches, Marx capacitors) have been successfully operated over and above the design values, but are similarly derated. This will result in lower electrical stress and consequently improve component lifetimes.

Deposition Uniformity

The e-beam deposition is monitored through the output window of the cell by means reflecting light from the cell from a 50cm square piece of glass onto a CCD camera with gated framestore. Figure 4 shows a three-dimensional plot of the energy deposition within the gas for a diode voltage of 600kV.

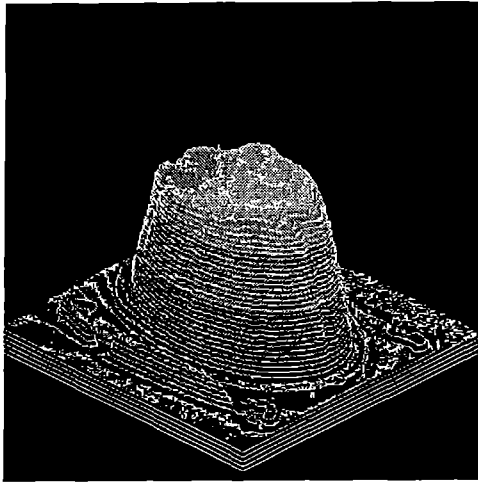


Figure 4. Electron beam deposition at a diode voltage of 600kV. 3D distribution

The krypton partial pressure and cell pressure are maintained constant at 300 mbar and 1350 mbar respectively whilst the Marx charge voltage is varied in 10kV steps from 60 to 80kV in each sequence of shots. Figure 5a shows the variation about the mean in e-beam deposition with Marx charge voltage. The uniformity is optimum at 80kV.

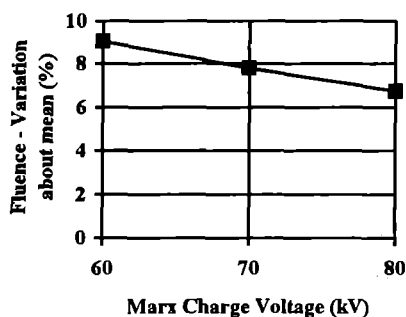


Figure 5a. Variation about the mean in average e-beam deposition with Marx charge voltage

Next, the krypton partial pressure is maintained at 300 mbar whilst the total pressure is varied in 100mbar steps from 1050 to 1350 mbar (Figure 5b). The Marx charge voltage is kept constant at 80kV.

Finally, the krypton partial pressure is increased from 300 to 600 mbar whilst maintaining the total cell pressure constant at 1350 mbar (Figure 5c). The Marx charge voltage is fixed at 80kV. Although only two points were recorded in this sequence of shots, uniformity is improved at 600 mbar compared with 300 mbar of krypton. This is due to the increased stopping power of krypton over argon and consequently less electrons are lost to the far walls of the cell.

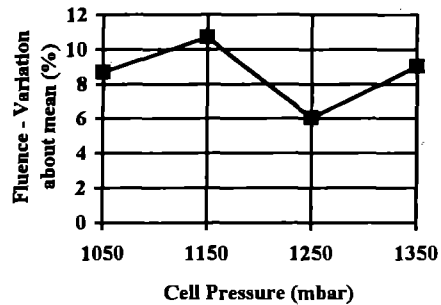


Figure 5b. Variation about the mean in average e-beam energy deposition with cell pressure

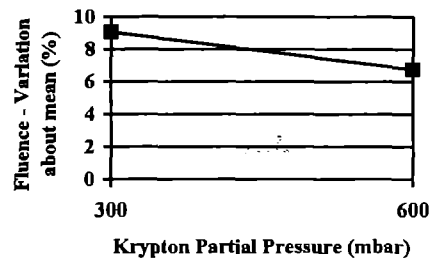


Figure 5c. Variation about the mean in average e-beam deposition with partial pressure of krypton

The optimum beam uniformity is 3.0% (σ_{n-1}) over the extracted aperture in 150/600/600 mbar (Ar:F2/Kr/Ar) at 80kV Marx charge voltage. This gas mix is the design mix and also provides the maximum e-beam energy deposition in the cell. Figure 6 shows a two-dimensional image of the deposited energy. The maxima of deposition occur at the anodes and the minima at the inter-diode spaces.

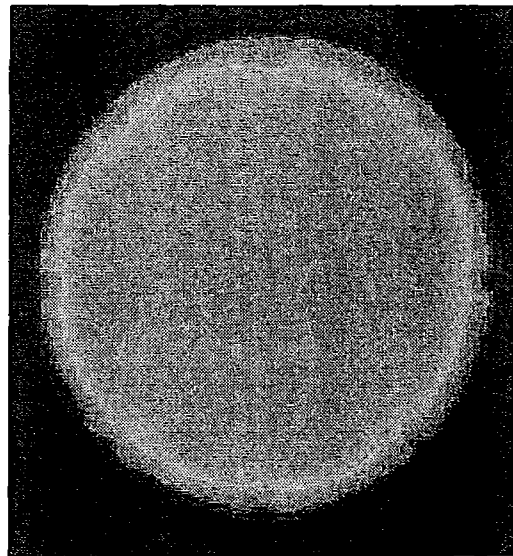


Figure 6. Cross-section of deposited energy

Figure 7 shows the cross-sections measured between (a) two geometrically opposing diodes and (b) two geometrically opposing inter-diode spaces for 150/600/600 mbar (Ar:F2/Kr/Ar) at 80kV Marx charge voltage.

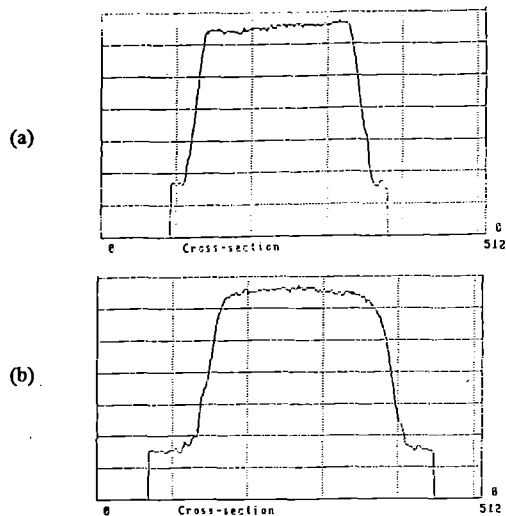


Figure 7. Cross-sections measured between
(a) two geometrically opposing diodes and
(b) two geometrically opposing inter-diode spaces
for 150/600/600 mbar (Ar:F2/Kr/Ar) at 80kV Marx
charge voltage.

Diode-to-Gas Transport Efficiency

The energy in the diode, E_{diode} , is determined by the computer code EXPTZ⁴ from the digitised traces of diode voltage and total current according to:

$$E_{diode} = \int_0^{\infty} i(t)v(t)dt. \quad (2)$$

The foil transmission efficiency, η , is defined as:

$$\eta = \frac{E_{gas}}{E_{diode}} \quad (3)$$

where E_{gas} is the energy deposited in the gas. Figure 8 shows the energy in the Titania diodes as a function of Marx charge voltage.

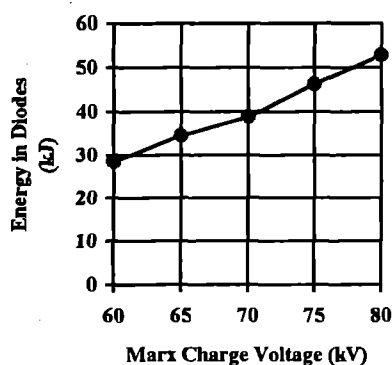


Figure 8. Energy in the Titania diodes

Figure 9 shows the foil transmission efficiency as a function of Marx charge voltages in the range 60 to 80kV. The target gas is that which will be employed in Titania operations - 150/600/450 mbar (Ar:F2/Kr/Ar) - based on the results of energy deposition and uniformity described above. The efficiency is 44% at the lowest energy and drops to 39% at higher voltage as the electrons penetrate further into the gas and

are lost to the far wall. This efficiency is well above the design value of 30%.

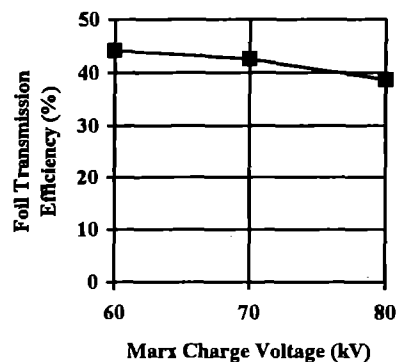


Figure 9. Efficiency vs. Marx charge voltage

Conclusions

- 20kJ of e-beam energy is deposited in the target gas for a partial pressure of krypton of 600 mbar at a Marx charge voltage of 80kV for cell pressures of 1200mbar and above. The optimum beam uniformity is 3.0% (σ_{n-1}) over the extracted aperture in 150/600/600 mbar (Ar:F2/Kr/Ar) at 80kV Marx charge voltage.
- A foil transmission efficiency of 39% has been achieved at a diode voltage of 520kV, well above the design value of 30%.
- The diode voltage of 520kV is well below the design value of 600kV, which will result in lower electrical stress on the key components in the system (cathodes, insulators, switches) thus increasing component lifetimes.

These conclusions indicate the optimum operating parameters of the Titania laser vessel to be:

Switch voltage:	1100kV
Diode voltage:	520kV
Marx charge voltage:	80kV
KrF mix:	150\600\450 mbar Ar:F2\Kr\Ar (1200mbar total pressure)

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CHARACTERISATION OF THE TITANIA LASER MODULE

A K Kidd, C J Hooker, A P Visser and M J Shaw

Rutherford Appleton Laboratory

OPERATION AS A LASER OSCILLATOR

The Titania laser is operated as an oscillator in order to test machine reliability and extract maximum energy in a 168ns pulse. The cavity is configured with an output coupling window angled at 90 degrees to the optical axis in order to provide about 10% feedback into the cell. The rear window is tilted at 2 degrees and the back mirror has a focal length of 9.6m. The first laser oscillator shot was fired on 9th November, 1994. To date (April 1995), a total of 860 laser shots have been fired on the Titania module at up to 40 shots per day.

In order to measure the energy contained in the 42cm diameter beam, a concave mirror was constructed to focus the beam onto a 20cm dia. detector. To cut costs, the mirror was designed within the CLF to be 110cm in diameter and to have a focal length of 150cm. The mirror was constructed from 10 and 5cm square pieces of glass (reflectivity measured as 3.4% at 248nm) overlain on a moulded polystyrene substrate.

The cell is filled with various KrF gas mixes. The laser output energy is measured as a function of:

1. Partial krypton pressure
2. Fluorine concentration.
3. Diode voltage

The cell pressure is maintained constant at 1100 mbar whilst the krypton partial pressure is increased from 300 to 500 mbar. The Marx charge voltage was varied in 5kV steps from 60 to 80kV in each sequence of shots. Figure 1 shows the variation in laser output energy with Marx voltage for different krypton partial pressures.

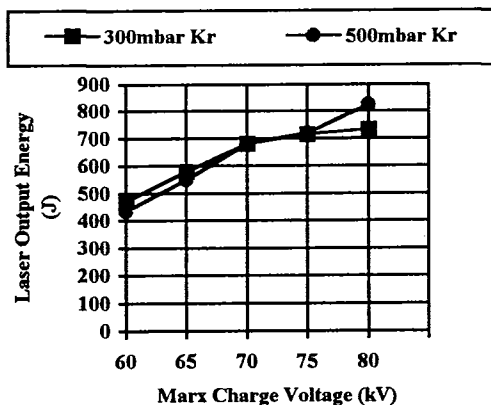


Figure 1. Variation of laser output energy with Marx charge voltage

From Figure 1, there is an increase in laser output energy as the krypton partial pressure is increased from 300 to 500mbar over the range of Marx voltages considered. Next, the fluorine partial pressure is increased in 50mbar steps from 100 to 200 mbar whilst maintaining constant total and krypton partial pressures of 1100 mbar and 500 mbar (Figure 2). The Marx

charge voltage is kept constant at 70kV in each sequence of shots.

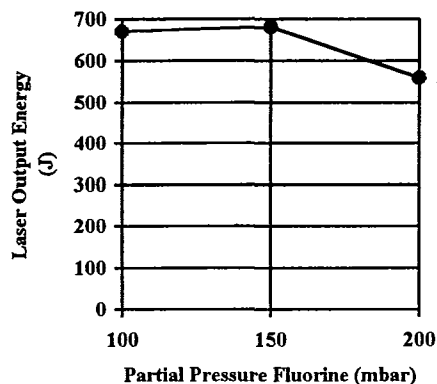


Figure 2. Variation of laser output energy with fluorine partial pressure

From Figure 2, the laser energy is reasonably constant at 100 and 150 mbar but falls off steeply at 200 mbar. Figure 3 shows the variation in laser output energy with Marx voltage for a fluorine pressure of 100mbar.

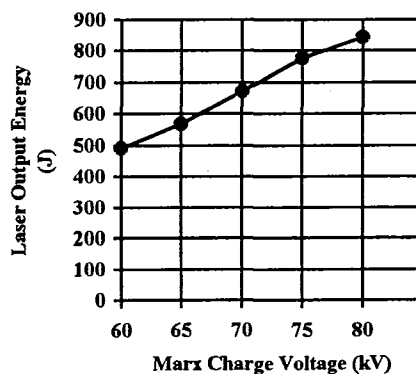


Figure 3. Variation of laser output energy with Marx charge voltage. Gas mix: 500/550/100 (Kr/Ar/He:F₂).

To date, the maximum laser energy recorded has been 845J achieved with 1150 mbar of 500/550/100 (Kr/Ar/He:F₂). This is well below the predicted 1500J. Possible reasons for the lower than expected laser energy are:

- The electrons see a thin gas target - the gas density is 20% below the design value. A 248nm probe beam was used to make measurements of the transmission of the Titania output window during the laser pulse to ensure that this effect was not due to X-rays emitted during the excitation pulse.
- Although the fluorescence follows the pump waveform very closely, the laser pulse collapses during the fluorescence. In particular, there is a greater tendency for

the on-axis laser pulse to collapse than the off-axis component (Figure 4). The FWHM of the on-axis laser pulse is 100ns compared with 140ns off-axis.

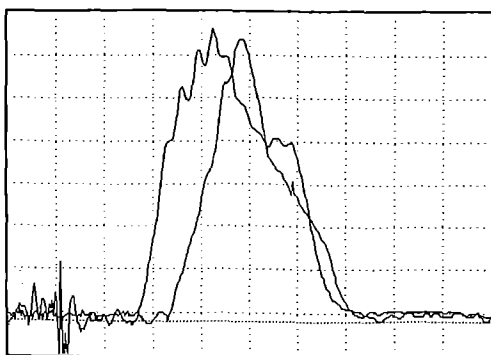


Figure 4. On-axis (RHS curve) and off-axis laser pulses.
Vertical scale: arbitrary scale.
Horizontal scale: 50ns/div.

It is proposed that the laser mode is collapsing on-axis during the pulse due to the particular oscillator arrangement used in this experiment. Since the module will not employ this configuration in the Titania laser system, this is not a problem.

SMALL-SIGNAL GAIN MEASUREMENTS

One of the most important parameters associated with the laser amplifier is the small-signal gain (SSG). Figure 5 shows the experimental arrangement used to measure the SSG of the Titania amplifier. An EMG150 discharge laser is used to trigger the Titania switches. A section of the beam is split off and sent via a multi-pass delay line through the Titania laser cell. The 25ns 248nm pulse is measured at the output by means of an rf-shielded photodiode. The input pulse is monitored by splitting off around 10% of the input beam by means of a quartz flat.

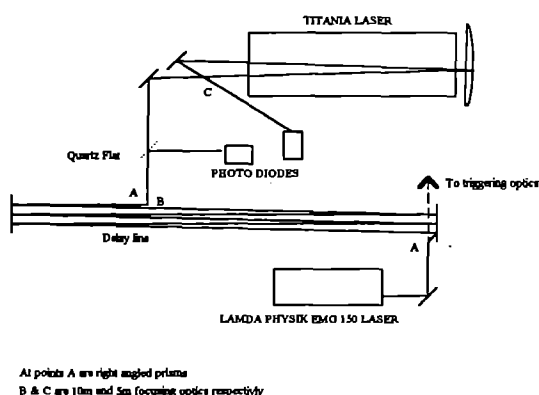


Figure 5. Experimental apparatus used to measure the small-signal gain.

The design specification¹ for the Titania laser vessel indicates that 20kJ of e-beam energy be deposited in the Ar/Kr mixture at a cell pressure of 1350mbar. Measurements of e-beam energy deposition have shown² that this is achieved at pressures of 1150mbar and above. In order to reduce the pressure loading on the module, the cell is operated with 1200 mbar of 600/450/150 (Kr/Ar/He:F₂). The small-signal gain is investigated as a function of:

1. Diode voltage (pressure jump)
2. Laser input energy.

Variation in Diode Voltage

The cell pressure is maintained constant at 1150 mbar whilst the Marx charge voltage was varied in 5kV steps from 60 to 80kV. The input and output energies are recorded on-axis. Figure 6 shows the variation of small-signal gain with Marx voltage.

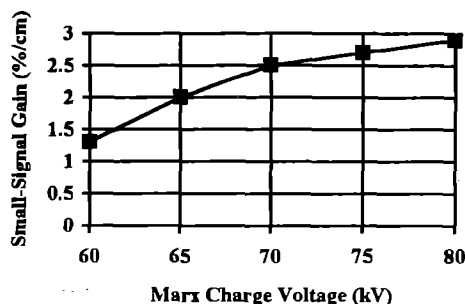


Figure 6. Variation of small-signal gain with Marx charge voltage (constant laser input energy)

At a Marx charge voltage of 60kV, the SSG is 35x, corresponding to a net ASE-depumped SSG of 1.3%/cm⁻¹. At 80kV, the figures are 6000x and 2.9%/cm⁻¹. This is slightly above the design value of 2.6%/cm⁻¹.

Variation in Laser Input Energy

The cell pressure is maintained constant at 1150 mbar whilst the laser input energy is progressively increased by two orders of magnitude to ensure that operation is within the small-signal regime. Figure 7 shows the variation of small-signal gain with laser input energy for a Marx charge voltage of 80kV. The straight horizontal line confirms that we are in the small-signal region.

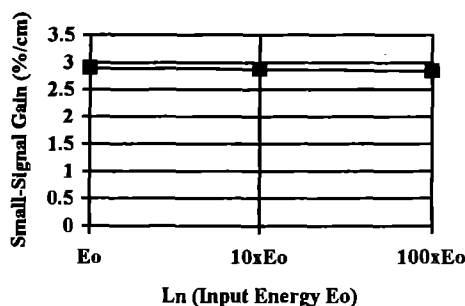


Figure 7. Variation of small-signal gain with laser input energy. Marx charge voltage 80kV. Eo is the input energy used to obtain the data in Figure 6.

A second technique to calculate the SSG is to measure the level of amplified spontaneous emission (ASE) emitted by the amplifier. The ASE intensity I_{ASE} , together with a knowledge of the static transmission of the laser cell, enables calculation of the SSG by means of the equation:

$$I_{ASE} = I_{SAT} A \exp((g_{ss} - \alpha)2L) \frac{d\Omega}{4\pi} \quad (1)$$

where A depends on linewidths and lifetimes and is typically 0.1^2 . The saturation fluence, I_{SAT} , is approximately 2.5 MWcm^{-2} under our conditions. Both single- (no back mirror) and double-pass (back mirror installed) ASE have been measured. Figure 8 shows the on-axis double-pass ASE fluences measured with a 10cm diameter calorimeter at various Marx charge voltages for a standard gas mix of 600/450/150 (Kr/Ar/He:F₂). The Marx charge voltage is varied from 60 to 80kV in 5kV steps. The calorimeter is placed at a distance of 3.3m from the Titania front window.

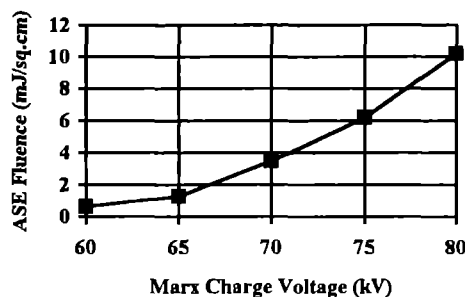


Figure 8. Variation of double-pass ASE fluence with Marx charge voltage 80kV at a distance of 3.3m from the Titania front window.

Figure 9 shows the on-axis double-pass ASE fluences measured at various distances from the Titania front window at a Marx charge voltage of 80kV.

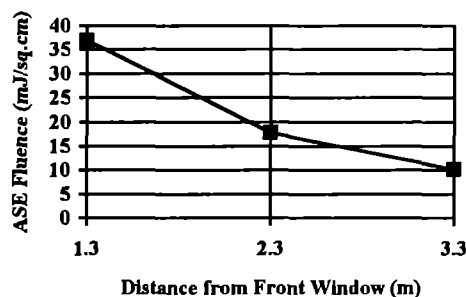


Figure 9. Variation of double-pass ASE fluence with distance from the Titania front window at a Marx charge voltage of 80kV.

The ASE fluence at a distance of 3.3m from the front window at 80kV is 10.2 mJcm^{-2} . Evacuating the cell and filling with pure argon revealed a static transmission of 28%, implying some etching of the windows has occurred. The calculated transmission of the fluorine with the standard gas mix is also 28%, indicating a static transmission of 7.8%. Using equation (1), we get an SSG of $3.0\% \text{cm}^{-1}$ at a Marx charge voltage of 80kV, which is consistent with the value reported above.

Conclusions

A maximum laser output energy of 845J has been achieved with a non-optimised oscillator cavity and gas mix. Under standard

operating conditions - with 1200 mbar of 600/450/150 (Kr/Ar/He:F₂) and a Marx charge voltage of 80kV - the net ASE-depumped SSG is measured directly to be 6000x, or $2.9\% \text{cm}^{-1}$. A second method of calculating SSG using ASE measurements gives an approximate value of $3.0\% \text{cm}^{-1}$. This exceeds the design value of $2.6\% \text{cm}^{-1}$.

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J Appl Phys **70** (10), 5238 (1991).

DESIGN OF THE TITANIA OPTICAL SYSTEM

S Hancock and C J Hooker

Rutherford Appleton Laboratory

SUMMARY OF THE OPTICAL SYSTEM

In its final configuration, the Titania system will provide four Raman beams on target, with high brightness, very low prepulse, a range of possible pulse lengths and near diffraction-limited quality. A good-quality KrF CPA beam will also be available, plus a second KrF beam for backlighting, broadband irradiation and other specialized applications. The Raman chain will consist of two new amplifiers, with lightguide apertures of 5 cm and 16 cm, which will be called RA4 and RA5 to avoid confusion with names in existing publications. Both will be angularly multiplexed, with two extracting beams in RA4 and four in RA5, and the KrF pump beams will carry trains of four pulses, rather than single pulses as in the Sprite system. The KrF chain will consist of Sprite and Titania only. Sprite will have twelve angularly multiplexed beam channels, each of which will drive two angularly multiplexed channels in Titania. A full description of the optical system can be found in Reference 1.

OPTICAL SYSTEM DESIGN

In the start-up configuration in April 1996, there will be a single KrF beam in operation, carrying a chirped pulse of around 50ps duration. This will be amplified to around 5 joules in Titania, and after attenuation and recompression by a grating pair will give a pulse of 100-200 femtoseconds duration on target.

The arrangement for multiplexing and demultiplexing the KrF beams in Titania is the most complicated part of the optical system, and imposes the severest constraints on the rest of the layout. Most of the design work on the optical system in the past year has therefore been devoted to laying out the mirrors and beam paths so that the beams are unobstructed and all have the correct optical paths. By using a 3-D CAD package in which the beams are treated as solid objects, any interception of one beam by mirrors handling other beams could be detected and appropriate corrections made. After the positions of all the Raman pump beams and mirrors were defined, the comparatively simple CPA beam line was incorporated into the design.

The input and output optics of Sprite were changed as part of the Raman upgrade last year, and do not need further modification. The CPA beam will use the beam line closest to the axis of Sprite in order to minimise aberrations, and will be timed so it is the first pulse to be amplified. The optical configuration of Sprite is shown in the following table:

Input beam:	1.8 cm diameter
Input optic:	F = 1.12 m silica lens

Back mirror:	R = 19.14 m concave
Output optic:	F = -9.59 m CaF ₂ lens
Output beam:	9 cm diameter

Following Sprite, the beam lines carrying the Raman amplifier pump beams will be split and delayed to give twenty-two equally-spaced groups of pulses into Titania. The CPA beam will not be split, and will be the first beam to be amplified in Titania, again using the beam line closest to the axis to minimise aberrations.

The beams entering Titania will be expanded using convex mirrors rather than lenses, and the output beams will be recollimated in the same way. This reduces the number of transmissive optics and 45-degree mirrors in the system, and makes the best use of the space in the KrF optics bay. To maximise the extracted volume in Titania, the beams will expand to 42 cm diameter at the back mirror, leading to a loss of around 7% of the total beam area from vignetting of off-axis beams.

The use of convex mirrors inevitably involves off-axis reflections, and will result in a few waves of aberration on the pump beams; the calculated amount of aberration will not degrade the operation of the Raman amplifiers in any way. Because the CPA beam will be used directly on target, it must be as close as possible to diffraction-limited, so a ray-tracing code was used to study the effects of the off-axis reflections. When the directions and angles of the off-axis reflections are chosen correctly, the total geometrical aberration is around 1/3 of the diffraction-limited beam divergence, so there should be only a slight degradation of the focal spot from this cause. The ray-trace also showed that the negative lenses in the Sprite beam pipe introduce significant chromatic aberration, which if uncorrected would result in poor compression of the pulses. To compensate for this aberration, a non-achromatized spatial filter was designed, to be placed in the beam before the input to Sprite. The optical configuration of Titania is shown in the following table:

Input beam:	9 cm diameter
Input optic:	R = 17.45 m convex mirror
Back mirror:	R = 45.56 m concave
Output optic:	R = 39.39 m convex mirror
Output beam:	16 cm diameter

The CPA beam is one of only two beams that suffers no vignetting in the Titania cell. However, to ensure that the edge of the beam is as clean as possible, it will be apodized at a point between Sprite and Titania; this will eliminate any diffraction

fringes that might cause localized damage to the diffraction gratings.

The layout of the Titania input/output array has been made as compact as possible to fit into the KrF optics bay, which occupies the northern side of R2. A diagram of this array is shown as an inset in Figure 1: the small beams are inputs and the large beams outputs. The KrF bay will contain the input and output beams for Sprite and Titania, the delay paths following the 12-to-24 beam split and the demultiplexing paths for the Raman pump beams. After demultiplexing, the pump beams will be steered into RA4 and RA5 by 45° mirrors opposite the entrance to the Raman optics bay. A diagram of the pump array is shown as a second inset in Figure 1, and the main figure is a 3-D schematic of the whole of the KrF system.

THE RAMAN OPTICS BAY

The Raman optics bay lies in the link building between R2 and R7, and will contain the optical delay lines for the Raman

beams, vacuum spatial filters and the two main Raman amplifiers. The pulse compressor for the CPA beam will occupy a vacuum chamber in this bay, and the recompressed pulse will propagate to the target chamber in an evacuated beam pipe. Output beams from RA5 will be propagated to the target area in beam pipes filled with either argon or helium.

The design of the optical system for the CPA beam is complete, and mechanical designs for the compression chamber and the grating mounts are nearly complete. Following the gratings, a telescope will reduce the beam diameter from 15 cm to 10 cm, the same as on Sprite, to allow the use of existing target chamber optics. The telescope will consist of one concave and one convex mirror, placed five metres apart in the beam pipe. Ray-tracing of the telescope optics has confirmed that no significant aberration will be introduced by the off-axis reflections involved.

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1. S Angood et al, 'Specification for the Titania KrF Laser System', RAL-94-014

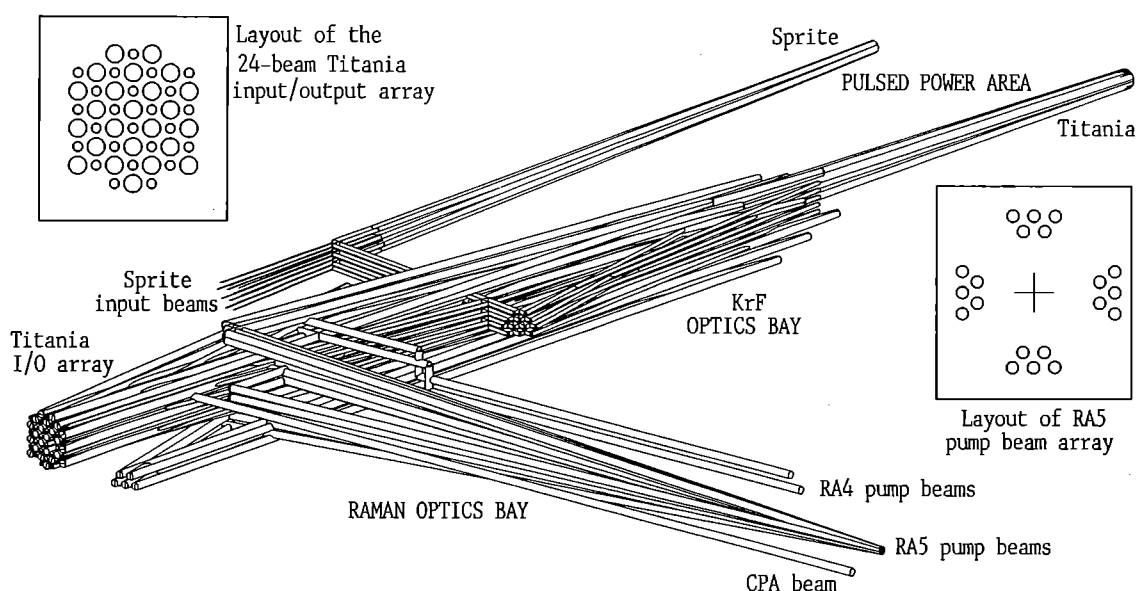


Figure 1. 3-D CAD image of the KrF beam layout in the Titania system. The Raman beams are not shown in this view, nor are any of the mirror support frameworks. The two insets show the Titania I/O array and the turning/pump mirror array for RA5 in greater detail.

A HIGH SENSITIVITY IMAGING FLAT-FIELD SPECTROMETER DESIGN.

D. Neely¹, A. Damerell¹, R. Parker¹ and M. Zepf²

- (1). Rutherford Appleton Laboratory, Chilton, Didcot, Oxon, OX11 0QX
(2). Clarendon Laboratory, Oxford University, Parks Road, Oxford, OX1 3PU

INTRODUCTION

X-ray emission spectroscopy is a widely used technique to determine the temperature and density conditions present within a laser generated plasma. It is also an essential diagnostic to measure the gain coefficient of a soft X-ray laser transition¹ or the efficiency with which harmonics of the drive laser wavelength can be produced². Typically, on experiments carried out on the VULCAN or SPRITE high power lasers, a Rutherford designed spectrometer incorporating an aperiodically ruled flat-field grating has been used to examine emission in the 20 ↔ 300 Å region. To facilitate new experimental investigations an increase in the minimum detection sensitivity and spectral range was required. This paper discusses the design and benefits of a new spectrometer constructed to fulfil these needs.

STANDARD NON-IMAGING FLAT-FIELD

The standard flat-field spectrometer design used previously at Rutherford did not use any collection optics but instead relied on placing the entrance slit close to the plasma emission to obtain sufficient photons for detection. Typically, the entrance slit was placed at 20 - 100 mm from the source but in some instances the spectrometer was operated slitless and the plasma placed at the location of the entrance slit plane. A Hitachi concave flat-field diffraction grating was located at a distance of 237 ± 0.5 mm from the entrance slit and focused the emission from the entrance slit onto the detector plane which was situated at 235 ± 1.0 mm behind the grating. A number of different options for the detector were available. For time integrated results either films such as Kodak 10101, 10402 and Ilford Q-plates were used or, a soft X-ray CsI coated fibre optic bundle vacuum face plate coupled to a Mullard 50/40 intensifier. For time resolved results a Kentech streak camera was used.

The standard glancing angle setting for the flat-field gratings were 3.0 ± 0.2 degrees for the 1200 lines/mm grating and 1.25 ± 0.1 for the 2400 lines/mm grating. To initially set-up the spectrometer a pre-aligned telescope was used to define the spatial location of all the components. Then, a collinear HeNe laser was injected through the telescope to set the grating angle. The 1200 lines/mm grating shows a slow fall off in higher order efficiency. This can greatly increase the effort required to deconvolve the spectrum. To reduce this effect a double parallel mirror soft X-ray reflection filter was installed between the entrance slit and the grating³. The filter could be set to efficiently transmit only emission longer than a cut-off wavelength which was normally in the 40 ↔ 100 Å region. The typical spectral ranges studied were 30 ↔ 300 Å using the 1200 lines/mm and 15 ↔ 100 Å with the 2400 lines/mm grating.

IMAGING FLAT-FIELD SPECTROMETER

To increase the range of experimental investigations carried out at Rutherford a spectrometer with much higher sensitivity in the 50 ↔ 500 Å region was required. The main modifications implemented for achieving this were :-

- (1) to increase the flux density at the detector plane by using an additional cylindrical mirror with a surface orthogonal to the flat-field grating to image the source in one dimension. By using a variable focal length mirror it was also possible to greatly increase the locational flexibility of the system.
- (2) to use a micro-channel plate (MCP) permanently maintained under high vacuum as the detector. MCP's are more sensitive detectors than film, particularly in the longer wavelength portion of the range.
- (3) to insert an additional limited aperture filter between the grating and the detector, which efficiently discriminates against fluorescent and scattered emission within the spectrometer

As the initial diagnostic requirement was in the 50 ↔ 500 Å region this report will only consider a spectrometer design based on the 1200 lines/mm grating which is optimally blazed for this region.

The spectral resolution and imaging properties of this grating⁴ were investigated using a geometrical ray tracing program which solved equation (1).

$$\sin^{-1} [\hat{n} \wedge \hat{z} - m\lambda\rho] = \theta_{out} \quad (1)$$

Where ρ is the groove density, λ the wavelength, m the order being examined, and θ_{out} is the output angle. The geometry is as depicted in Figure (1). Since the imaging mirror's surface is orthogonal to the grating surface the spatial imaging of the mirror and spectral imaging/dispersion of the grating can be considered separately and combined at the final design stage.

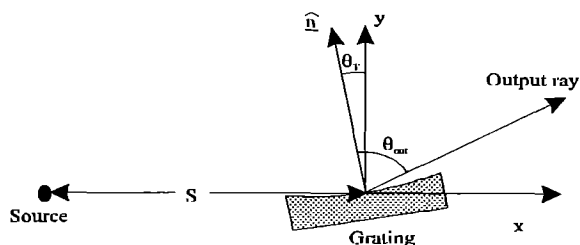


Figure 1. Schematic diagram showing geometry used to ray trace a flat-field grating.

For the 1200 lines per mm flat-field grating the groove density ρ is given as⁵

$$\rho = \rho_0 \left[1 + \frac{2b_2}{R}x + \frac{3b_3}{R^2}x^2 + \frac{4b_4}{R^3}x^3 \right]^{-1} \quad (2)$$

Where $r = 5649$ mm, $\rho_0 = 1200$ lines/mm, $b_2 = -20$, $b_3 = 455.8$ and $b_4 = -11840$ and the grating surface is 50 mm long.

The ray tracing program followed a bundle of 21 rays (set to intercept the grating across its full length, at equal intervals). It located the focal plane for each wavelength by finding the location where the distance between the centroid and the refracted rays in the output bundle was smallest. The program examined the spectral focal planes for a range of grating angles $2^\circ \leftrightarrow 6^\circ$ and typical outputs are shown in figures 2 and 3 for different source distances.

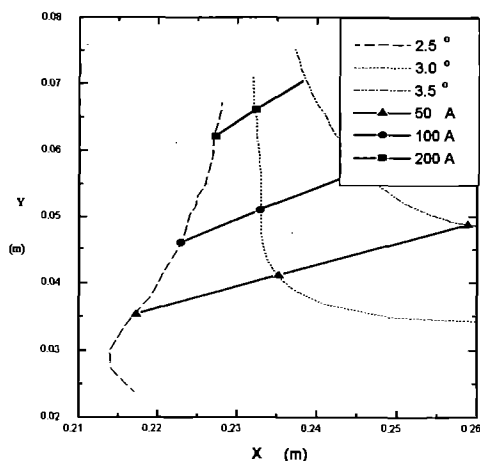


Figure 2 Focal planes for a 1200 lines/mm flat-field grating and a source to grating distance of 0.237 m.

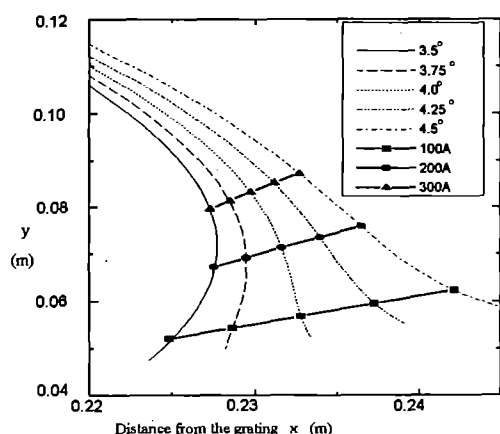


Figure 3 Focal plane for a source to grating distance of 0.62m

To obtain maximum sensitivity it is necessary to simultaneously obtain optimum spatial and spectral resolution. This condition is satisfied when both the grating and imaging mirror focal planes overlap exactly. A relaxation of this condition occurs due to the limited resolution p of the detector ($p \approx 5 \mu\text{m}$ for film and $p \approx 100 \mu\text{m}$ for a double MCP). If ϕ_i and ϕ_s are the angular convergences of the spectral and imaging ray bundles respectively, then this constraint can be expressed as

$$2\phi(\lambda)|G(\lambda)I(\lambda)| < p \text{ for } \lambda_{\min} < \lambda < \lambda_{\max} \quad (3)$$

Where $G(\lambda)$ is the grating focal plane, $I(\lambda)$ is the imaging mirror's focal plane and $\phi(\lambda)$ is the greater of the pair ϕ_i and ϕ_s for a given wavelength. The image plane $I(\lambda)$ will be circular, of radius $r = u - |mg|$ where u is the image distance and $|mg|$ is the distance between the mirror and grating centres. Since most detectors are flat then the condition expressed in equation 3 can only be satisfied over a limited wavelength range. For the case where the source to grating distance is 0.62 m as shown in figure 3, it is possible to adequately image wavelengths both spectrally and spatially between 200-400 Å by using the grating at 4.25 degrees and situating the detector appropriately.

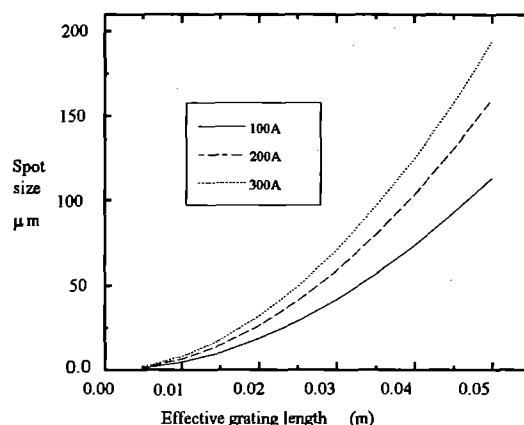


Figure 4 Plot of geometrical focal spot size vs. wavelength.

EXPERIMENTAL

The imaging mirror substrate was a flat Au coated piece of BK7 glass of size 160x10x1 mm with a surface roughness of less than 4 Å. This substrate was placed in a bending jig and adjusted to the required radius. To reduce stray light and improve image quality a limiting input aperture was situated so that only the central 80 mm were illuminated. Using the variable radius obtainable with the bending jig, the mirror could be utilised with a source distance of 0.2-1.4 m. The spectrometer (Figure 6) was tested with a source to mirror distance of 230 mm, mirror glancing angle 4° and grating angle at 3.8° giving a spatial magnification at the detector plane of 2.3. The solid subtended by the spectrometer was 1.5×10^{-4} Sr and tests demonstrated a spatial resolution of 30 μm.

CONCLUSION

The addition of a glancing angle mirror to image the source onto the detector plane in a flat-field grating spectrometer greatly enhances the detection sensitivity of the instrument. To increase dynamic range the introduction of a low dispersion transmission grating orthogonal to the flat-field is planned. Such a grating could also help in separating

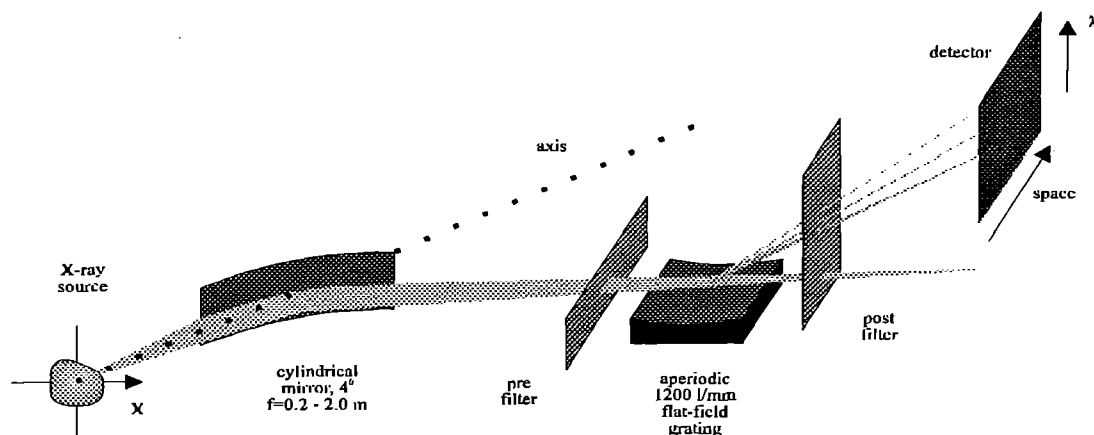


Figure 5 Schematic diagram showing the geometry of the imaging flat-field spectrometer.

The raytracing program was also used to examine the spectral resolution at the output plane as a function of grating angle and illuminated grating width. The resolution was found to be only a weak function of grating angle, with greater angles and greater wavelengths giving larger focal spots. The focal spot size was observed to be an almost linear function of grating illumination length as shown in figure 4. For the majority of this curve the resolution obtained is significantly lower than the diffraction limit considering the number of grooves illuminated. This result would tend to indicate that the spectral resolution can be easily increased by at least a factor of x10 by limiting the length of grating illuminated.

the high order spectral overlap. It is also planned to further extend the spectral range of the instrument with operation up to 500 nm envisaged by the introduction of a second grating which intercepts the zero order emission and disperses it onto a second detector.

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- 2 S. Preston et al, this report
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- 4 Part No. 001-0437 blank 30x50x10 mm Blaze angle 87 degrees.
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COMPARISON OF SOFT X-RAY DETECTORS

M.Zepf¹, A.G. MacPhee², C.L.S. Lewis², J.S.Wark¹.

¹Clarendon Laboratory, Department of Physics, University of Oxford, Parks Road, Oxford OX1 3PU, UK,

²Department of Pure and Applied Physics, Queen's University, Belfast, BT7 1NN, UK,

INTRODUCTION

Understanding the response of a detector and being able to compare different types of detectors is of crucial importance for the analysis and planning of experiments. Here two soft X-ray detectors, a Microchannel Plate assembly (MCP) with two microchannel plates in series and a CCD [1] coupled to a phosphor coated fibre optic plate are compared in the range of 40 to 200 Angstroms.

The actual microchannel plates are two 500 μm thick glass plates with 10 μm holes at a 12 μm pitch. These act like a matrix of electron multipliers, accelerating the electrons down the channels and generating secondary electrons by collisions with the glass walls. The amplified beam is then accelerated onto a phosphor screen and the image is then stored on a CCD/computer data acquisition system (Fig. 1). The MCP's tested here were uncoated.

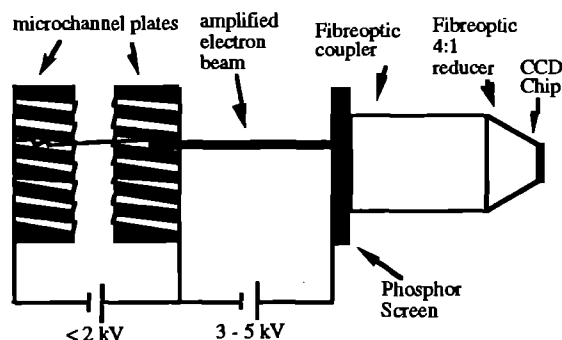


Fig 1: Schematic of MCP detector

The fibre optic system relies on the conversion of high energy photons to visible light in the phosphor coating. The light is then guided by the fibre optic plate onto the cooled CCD chip. (Fig 2)

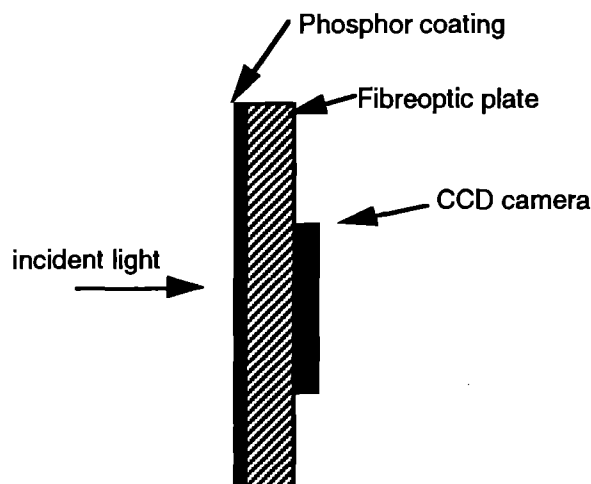


Fig. 2: Schematic of phosphor detector

The detectors were compared in terms of sensitivity, spatial resolution and dynamic range. The results presented should be valid over a far greater wavelength range as the relative response of the phosphor (linear with photon energy) and the MCP (essentially flat with a sharp cut-off at 1000 $\text{\AA}$) are known.

The MCP was also calibrated in terms of absolute photon number relative to the CCD + phosphor detector, which had previously been calibrated against film.

THE EXPERIMENT

The source used for calibration was a Carbon plasma generated from a rotating target with 1 J, 3 ns pulses from a 10 Hz, frequency doubled Nd:YAG laser. The light was collected by a flatfield spectrometer using a 1200 l/mm Hitachi grating [2]. The spectrometer was used without a slit to maximise the collection solid angle and the grating was reangled to keep the instrument in focus. The incident light levels were adjusted by using Aluminium filters in thicknesses between 0.4 μm and 4 μm . The detectors were then mounted in the same position relative to the gratings.

Three different set-ups were tested. These were the 16 bit cooled CCD + phosphor coated fibre optic, the MCP with the above CCD camera and the MCP + Photonic Science "DarkStar" intensified CCD camera. The first and the latter are the configurations which have previously been used in experiments at the RAL [4,5]. The second configuration was used to determine the dynamic range of the MCP which was thought to be limited by the intensified CCD. Also this gives a direct comparison of the spatial resolving power of the MCP compared to the phosphor assembly.

SENSITIVITY

The sensitivity curve of the MCP was taken over as wide a range of gain settings as was possible under the experimental conditions. It was put onto an absolute scale by comparing the integrated number of counts to the CCD + phosphor assembly which is known to have a linear response with photon energy. The sensitivity of the system had previously been calibrated to be 0.03 photons/ $\mu\text{m}^2/\text{count}$ @ 182 $\text{\AA}$ and 0.007 photons/ $\mu\text{m}^2/\text{count}$ @ 40 $\text{\AA}$ [1,3]. Due to the pixel area of $27 \times 27 \mu\text{m}$ this corresponds to 22 photons/pixel/count

For a voltage of 1.1 kV this results in 3.21 at 40 $\text{\AA}$ and 3.6 photons/count at 182 $\text{\AA}$, well within the observed 30% shot to shot variation of the signal level. This is in agreement with the expected flat response of the MCP.

Due to excitation of neighbouring channels one photon at high gains will produce an image of around $400 \mu\text{m}^2$ on the CCD coupled to the back of the MCP, corresponding to about 16 pixels on the Photonic Science CCD camera. This explains the high number of counts/photon on the calibration curve. At high gains, one detected photon is enough to be clearly visible above background. The quantum efficiency is quoted at around 5% to 10%.

This also allows an independent check on the consistency of calibration: If one assumes the smallest spots on an image are due to single photon events, then the integrated number of counts under these spots should be predicted by the calibration curve. Indeed the smallest spots were found to have 40 counts at 1.5 kV gain compared to the predicted 50.

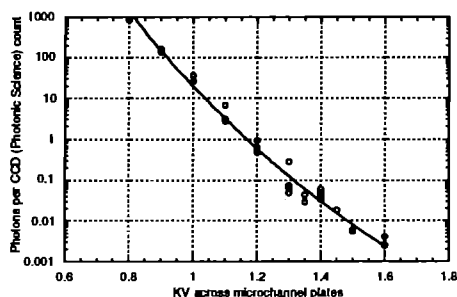


Fig 3: Calibration curve of microchannel plate

The MCP assembly proved to be far more sensitive than the phosphor system. It was possible to take single shot images with any of the gain settings used on the MCP. The phosphor system needed 50 shots to produce a well exposed spectrum. Single shot data collection was only possible through integrating all counts in one column of the CCD array.

SPATIAL RESOLUTION

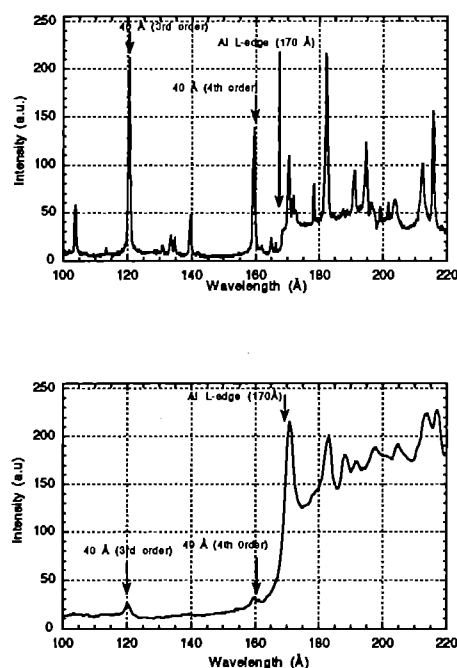


Fig. 4: Comparison of Carbon spectra taken with the MCP (bottom) and CCD + phosphor (top)

The excitation of neighbouring channels mentioned above serves to limit the MCP to about 200 μm spatial resolution. This results in comparatively poor spectral resolution compared to other media such as film or the CCD/phosphor system.

The phosphor system's spatial resolution by contrast is only limited by the combination of fibre optic and CCD. Typical line widths were around 2-3 pixels or 56 - 86 μm . Figure 4 shows a comparison of a single shot MCP spectrum and a lineout of 50 shot spectrum in the phosphor system. The difference in resolution is quite clearly visible. The different line ratios are largely due to the difference between the flat MCP response with photon energy compared to the phosphor response favouring short wavelengths.

DYNAMIC RANGE

The dynamic range of the systems compared here is defined as the ratio of the maximum number of counts and the average noise level. These are effectively the number of greyscales which the detector can reproduce.

The dynamic range of the phosphor system is determined by the performance of the CCD detector. The scale on this system stretches to 65000 counts while typical noise levels are of the order of a few counts.

This results in a dynamic range of about 4 orders of magnitude.

The dynamic range of the MCP assembly was determined using the low noise, cooled CCD from the phosphor system. In this set-up the dynamic range is limited by the saturation of the MCP's and can clearly be seen by the line ratios approaching unity. This effect can distort relative line intensities when one approaches the saturation of the MCP and affects the interpretation of the data. The dynamic range of the MCP before the onset of a significant change in the line ratios is about 1000.

The dynamic range of the MCP with the Photonic Science CCD camera was limited to about 100 by the CCD camera.

CONCLUSION

In conclusion it can be said, that the MCP's advantage in sensitivity will make it the choice for experiments where low light levels are expected. Furthermore the relatively flat response from around 1400 Å (1800 with CsI coating) to 0.1 Å makes it a very versatile detector, especially at long wavelengths. In the range from 600 Å to around 0.3 Å the phosphor system, however, will produce cleaner spectra with a higher dynamic range once the light levels are sufficiently high and therefore be the detector of choice for such experiments.

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F2.5 OPTICAL IMAGING SYSTEM.

R. Parker, K. Clifton, D. Neely, P. A. Norreys and F. N. Walsh.
Rutherford Appleton Laboratory, Chilton, Oxon., OX11 0QX, England

INTRODUCTION.

The optical emission generated when high intensity laser pulses are focussed into gas-jet targets can give important information concerning the size and location of the focal spot and any filamentary structure. An important consideration when using a short pulse high intensity CPA (Chirped Pulse Amplified) laser beam is the choice of optics used for focussing the beam onto the target. The optical path through transmissive optics must be kept to a minimum in order to conserve beam quality. At Rutherford, to minimise this problem, we use a reflective off-axis parabola to focus the laser beam. In order to reduce costs and ease construction, the off axis angle is kept to a minimum, typically $12^\circ \rightarrow 14^\circ$. The folded nature of the beam path in such a geometry precludes the use of small, short focal length lenses for diagnostic purposes, as the access to the target plane is restricted. To overcome this, an imaging system using f2.5 aspheric doublet lenses with a long focal length of 0.275 m, was designed. Using these optics allowed the lenses and support brackets to be mounted at the opposite side of the CPA beam from the target, without obscuring any of the beam. The lenses used were originally intended for focussing a drive laser beam to a near diffraction limited point at the target plane. These high collection efficiency optics were used due to the cost of new, long focal length lenses. Measurements of the imaging properties of the system were made using a ray-tracing package, Beam 4, which only included geometric effects. The system was tested experimentally and produced excellent results.⁽¹⁾

THE F2.5 DOUBLET

The f2.5 doublet consists of 2 plano-convex lenses, with one element being asphericised to give optimum focussing of a parallel beam to a point.⁽²⁾ Imaging using a single f2.5 aspheric doublet lens was investigated, to establish the geometric source resolution at different magnifications. The results showed that the resolution was almost constant at around $3\ \mu\text{m}$, at any magnification over $\times 5$. However, at magnifications lower than $\times 5$ the resolution degraded considerably to over $100\ \mu\text{m}$ at $\times 2$. To image at different wavelengths, it was necessary to optimise the separation of the elements in the doublet, which ranged from $4.88\ \text{mm}$ @ $1\ \mu\text{m}$, to $58\ \text{mm}$ @ $0.25\ \mu\text{m}$. The imaging system was ray-traced at $1\ \mu\text{m}$, $0.5\ \mu\text{m}$ and $0.25\ \mu\text{m}$. It was found that the optimum separation of the elements required to image a point to a point was the same as that required to image a parallel laser beam to a point.

The system was required to operate at very low magnifications, to image a large field of view ($\sim 8\ \text{mm}$) onto a CCD camera. To overcome the problem of low resolution at low magnifications, a system was designed using a pair of doublets as described below.

IMAGING WITH TWO F2.5 DOUBLETS.

The imaging properties of a system consisting of two doublets placed opposite each other in an infinite conjugate ratio were investigated.

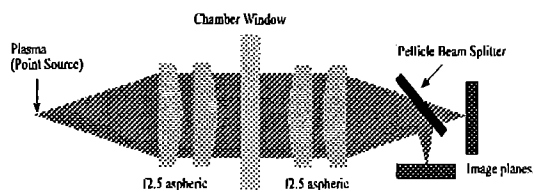


Figure 1 Schematic of the twin doublet imaging system.

The imaging system (figure 1) was initially set up for operation at the second harmonic ($\lambda=527\ \text{nm}$), and was ray-traced accordingly. When the centres of the adjacent lens surfaces are the optimised $17.9\ \text{mm}$, the calculated resolution is $3\pm 2\ \mu\text{m}$. When the beam passes through the chamber window shown in figures 1 and 2 it is parallel and no noticeable effect is made upon the calculated resolution. The imaging

system was also required to be used at infra-red wavelength ($\lambda=1054\ \text{nm}$), and the ray-tracing showed that the element separation for this wavelength was optimised at $4.88\ \text{mm}$. However, it was found that there is no significant degradation in resolution ($\sim 6\ \mu\text{m}$) when imaging at $1054\ \text{nm}$ using the doublet separation optimised for $527\ \text{nm}$, although the image plane position increases by $14\ \text{mm}$. For a given wavelength it is thus possible, using a combination of bandpass filters and pellicle beam splitters, to image multiple wavelengths simultaneously onto multiple CCD detectors. Figure 2 shows a schematic of the location of the final imaging system with respect to the laser beam.

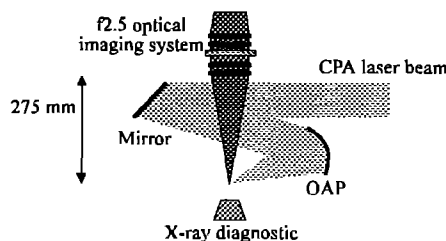


Figure 2 Schematic of imaging system position, within chamber.

As the wavelength of the light to be imaged changes, so does the focal plane of the imaging system. Figure 3 shows the change in calculated focal plane distance, relative to the rear surface of the second doublet. A kink can be seen at $400\ \text{nm}$. This is attributed to the numerical accuracy of the ray-tracing program.

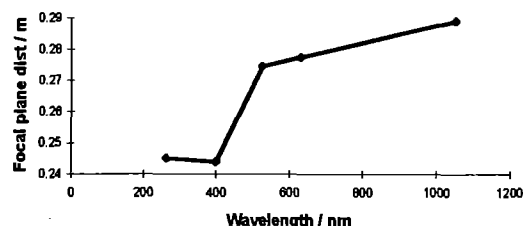


Figure 3 Plot showing the results obtained with the imaging system.

The refractive index, n , of the two fused quartz elements in the doublets varies with wavelength, from 1.50079 at $264\ \text{nm}$ to 1.44972 at $1054\ \text{nm}$. To a first order approximation, for small changes in n caused by changes in wavelength, the change of the focal plane position is proportional to $(n-1)^2$. This shows that the wavelength bandwidth (the range of wavelengths which are brought to a tight focus in the same plane) of the imaging system is narrower at shorter wavelengths, because $dn/d\lambda$ is higher. The dependence of refractive index on wavelength is not a great problem when studying harmonic emissions as the intensity is such that it normally dominates any other emissions.

CONCLUSION.

An imaging system using a pair of f2.5 doublets has been used successfully in Target Area West several times and has achieved the required resolution. There are plans to develop the system further, by introducing a grating or prism between the two doublets, thereby creating a 2D spectrometer.

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VULCAN OPERATION AND STATISTICS

DA Pepler and CN Danson

Rutherford Appleton Laboratory

Sixteen full experiments have been conducted this year, using all four target areas, with a significant number of these experiments being in the main target areas TAW and TAE. Both of these areas have been used for chirped pulse amplification (CPA) operation with TAE used in this mode for the high harmonic X-ray laser experiment early in the year. TAW has been intensively and continuously operated throughout the year with CPA with extremely varied experimental requirements.

TA4 has continued to be productive providing for several short experiments and due to the high level CPA programme TA2 was used for a single experiment at the beginning of the year.

A total of 7792 laser shots were fired 3439 of which were to the target areas. The statistics for the experimental programme are given in Table I which indicates the total number of shots, target shots and failed shots each individual experiment received. Due to the increasing complexity of the experimental setups nearly half the total number of these shots were required to diagnose the experimental conditions. A total of 1886 shots were fired to target for full data collection. The relative breakdown of these shots into the four individual target areas is indicated in Chart I.

The failure rate of target shots continues to be low at ~3.5 %.

TA4 has had an upgrade this year with installations of new high-reflectivity mirrors and an aspheric focusing lens. These improvements have increased the energy throughput to target by a factor of two and enable 20 J in a 6 ns pulse to be delivered. This has substantially improved the images obtained from the X-ray contact-microscopy work.

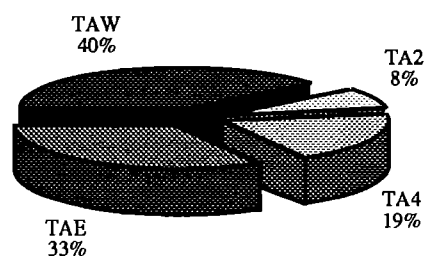


Chart I Relative number of target shots per area

EXPERIMENTAL PERIOD '94 - '95	EXPERIMENT (Total number of shots - Target shots - Failures)			
	TAE	TA2	TAW	TA4
28 Mar - 8 May		Plasma collisions (369 - 156 - 5)	Recombination X-ray (88 - 88 - 4)	Facility development (83 - 83 - 1)
16 May - 5 June	High harmonic X-ray laser (133 - 103 - 7)		Solid target CPA (122 - 82 - 3)	
6 June - 26 June				Biological imaging (153 - 135 - 1)
4 July - 31 July	Ge X-ray laser (90 - 90 - 3)		CPA gas jet (198 - 95 - 2)	
8 Aug - 28 Aug	Ni X-ray laser (424 - 61 - 3)		Plasma wakefield (253 - 53 - 2)	
29 Aug 17 Sept				Biological Imaging (58 - 58 - 0)
25 Sept - 5 Nov	Strain relaxation in brittle solids (180 - 92 - 4)		X-ray laser network (132 - 132 - 1)	
13 Nov - 24 Dec	Foil impact on foam targets (480 - 146 - 8)		Relativistic plasma wave (240 - 180 - 3)	Facility development (46 - 46 - 0)
9 Jan - 29 Jan	Foam smoothing (200 - 138 - 4)		Solid target CPA (167 - 107 - 4)	
30 Jan 19 Feb				Gas jet characterisation (23 - 23 - 0)
TOTAL NUMBER OF SHOTS	(1507 - 630 - 29)	(369 - 156 - 5)	(1200 - 737 - 19)	(363 - 363 - 1)

Table I VULCAN statistics for the year to March 1995

SPRITE OPERATIONS

E J Divall, G J Hirst, J M D Lister and R Mathumo

Rutherford Appleton Laboratory

INTRODUCTION

This section summarises the experiments and laser development work carried out using Sprite over the last year. More detailed descriptions of each can be found elsewhere in this report.

SHOT STATISTICS

Over the last year 3321 shots have been fired on the Sprite system, of which 2187 were target shots (i.e. were delivered to the Target Area during scheduled external user experiments). The remainder were used for laser testing/optimisation and development experiments. These represent a record shot rate, as Figure 1. shows. The year started with a recently upgraded laser which enabled more time to be scheduled for user-experiments compared with 1993/94 (only a minimal period being needed to convert between the Raman and CPA configurations). This explains the increase in the number of shots.

SCHEDULED OPERATIONS

The last year saw a significant increase in experimental time with 34 weeks allocated for external user experiments (an increase of 13% over the previous year). The remaining weeks were split between development experiments (8 weeks), and periods of system maintenance, reconfiguration and scheduled shutdown (10 weeks).

The external user experiments are summarised briefly in Table 1. Further details can be found in the first Chapter of this Section of the Report. The dates shown for each experiment include the standard one or two-week set-up period, in which target shots are not usually fired. The gain bandwidth experiment was slightly different in that it concentrated on basic laser research as opposed to plasma physics and was funded by the US Department of Energy via the Naval Research Laboratory. The final experiment, carried out by staff from Friedrich Schiller University in Jena (Germany), was the first Sprite experiment funded by the E.U.'s Large Facilities Access Program.

We have continued to log unplanned shutdowns, with a total of 16 lost days (9% of scheduled time) in this reporting year. Of these 7% were lost in set-up weeks (16% of time) and 8% in experimental

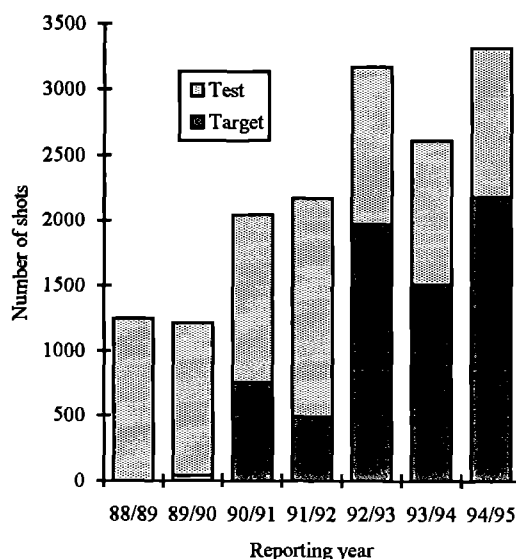


Figure 1. Annual numbers of Sprite test shots and target shots

weeks (7 % of time). Although the overall reliability has remained at about 90% the lost experimental time has been reduced somewhat this year.

HARDWARE

At the end of July the titanium foils surrounding the Sprite cell finally gave way after 9700 shots, spanning 4 years of use. This is an impressive lifetime, especially when compared to similar machines whose foils typically last for only hundreds of shots. New foils were fitted and have survived 2276 shots to date.

ACKNOWLEDGEMENTS

The hard work and dedication of CLF support staff has been crucial to the smooth running of the Sprite system over the last 12 months.

Experiment	Institution(s)	Dates (inc. Set-up)	Laser Configuration	Pulse Duration	Typical Energy	Target Shots
Water-window XUV lasers	Oxford-Belfast-RAL X-Ray Laser Consortium	1.4.94 - 15.4.94	Raman	60 ps, 25 ps	8-10 J	127
Ionisation of O VI Li-like ions.	Oxford	20.6.94 - 29.7.94	KrF-CPA	300 fs	0.2-0.3 J	385
Optical guiding in plasmas.	Imperial College	1.8.94 - 8.9.94	KrF-CPA	300 fs	0.2-0.3 J	274
CPA pulses on solid targets	Imperial College	11.9.94 - 13.10.95	KrF-CPA	300 fs	0.2-0.3 J	261
Harmonic generation	Oxford - RAL	23.10.94 - 24.11.94	KrF-CPA	300 fs	0.2-0.3 J	283
2-Axis Thompson scattering	Oxford - RAL	27.11.94 - 24.12.94	KrF-CPA	300 fs	0.2-0.3 J	378
Gain bandwidth measurements	NRL - RAL	9.1.95 - 10.2.95				375*
Short pulse absorption and X-ray emission.	Jena	20.2.95 - 31.3.95	KrF-CPA	300 fs	0.2-0.4 J	479

Table 1. Summary of external user experiments

(* These were data shots, since there was no target shooting in this experiment.)

LASER SUPPORT FACILITY PROGRAMME

A W Parker

INTRODUCTION

The pages that follow report on the many programmes of work carried out this year using LSF lasers. Scientific topics range from the use of laser plasma X-rays to the measurements of rates of chemical reactions at 13 K.

Scientific highlights include:

The high repetition rate laser plasma X-ray source operating at 1 nm wavelength has been applied to the production of 180 nm gates in transistors of high quality essential for the development of the next but one generation of computer systems. This work has been done using 100 mWatt X-rays average power. The source has recently been upgraded to 1 Watt average power.

Time-resolved Raman spectroscopy (ps and ns) has been used in studies ranging from the ps dynamics of hot electronically excited molecules to the spectroscopy of biologically relevant radicals. Vitamin E has been shown to quench aromatic carbonyl triplet states by hydrogen atom transfer. This is evidence that Vitamin E can prevent the formation of potentially damaging intermediates in

addition to its established role in oxidative damage repair. As part of the ongoing fs programme to investigate the interaction of intense ultrafast pulses with matter, a novel interferometric method has been developed in the fs laboratory which allows ultrashort pulses to be shaped prior to focusing with an achromatic $f/2$ lens which was especially designed and constructed for the project.

Gas phase investigations include measurement of unexpectedly fast reaction rates at low temperatures (13 K), measuring the depolarisation dynamics of small molecules and the first detection of dimethylgermylene. Jet spectroscopy techniques (including ablation and entrainment) have been applied to simple heavy metal halides/hydroxides, organometallic compounds and substituted aromatics.

A major reorganisation of the ps/fs laser laboratories has been necessary to house the soon to be delivered laser for the Ultrafast Spectroscopy Laboratory, the sub 50 fs laser for ultra short pulse work and the time resolved (100 ps) confocal Microscope. These reorganisations will increase the ability of each laboratory to individually develop expertise for particular programmes of work.

ANTI-STOKES RESONANCE RAMAN SPECTRUM OF PHOTOEXCITED S_1 *TRANS*-STILBENE

P Matousek¹, D L Faria², R E Hester², J N Moore², A W Parker¹, W T Toner¹, M Towrie¹

¹Central Laser Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxfordshire, OX11 0QX, UK

²Department of Chemistry, University of York, Heslington, York, YO1 5DD, UK

INTRODUCTION

Mode-specific, solvent-dependent changes in the resonance Raman spectrum of photoexcited S_1 *trans*-stilbene, tS, occurring on a *c.* 10 ps timescale following photoexcitation have recently been reported by ourselves and others¹⁻⁵. Our earlier work⁴ demonstrated that the magnitude of the bandwidth and band position changes in the Stokes spectrum on this timescale increased monotonically with the energy of the exciting photon and that similar changes in the spectra could be induced by an increase in the solvent bath temperature. Previous attempts to measure the anti-Stokes (AS) spectrum of S_1 tS have met with difficulty, although the AS Raman band of S_1 tS at *c.* 285 cm^{-1} and CARS of high lying modes have been observed^{2,4,6}. We now report detailed measurements of the AS spectrum of S_1 tS up to the band arising from the C=C olefinic stretching mode of tS at *c.* 1600 cm^{-1} . A preliminary account of this work has been presented⁷ and a full paper is currently in press⁸.

EXPERIMENTAL

The apparatus has been described earlier⁹. Pulses of 6 ps duration (FWHM), cavity-dumped from a synchronously pumped, mode-locked dye laser were amplified up to 2 μJ (probe) and 1 μJ (pump) at 4.46 kHz. All chemicals were of the highest purity available commercially and used as received. The concentration of tS in all solutions was $1 \times 10^{-3} \text{ mol dm}^{-3}$.

RESULTS

AS resonance Raman spectra at 0 and 50 ps pump-probe time delays are shown in Fig. 1 together with the Stokes spectrum for comparison. A comparison of the two AS spectra shows that several bands which are prominent at the early time delay decrease in relative intensity at the later time; the notable features are at 1570, 1240, 1180, 980 and 720 cm^{-1} . Further data (see Fig. 2) were obtained using pump and probe wavelengths of 295 and 590 nm respectively, which our earlier work⁴ showed would induce more easily measured time-dependent band position shifts in the Stokes spectrum.

It is notable that, in general, the anti-Stokes to Stokes intensity ratios of the high frequency S_1 bands are higher than predicted by the Boltzmann factor [$\exp(h\nu/kT)$]. By contrast, the solvent bands (which are not resonance enhanced) gave anti-Stokes to Stokes intensity ratios (*c.* 0.001 for high wavenumber bands), consistent with Boltzmann factor calculations.

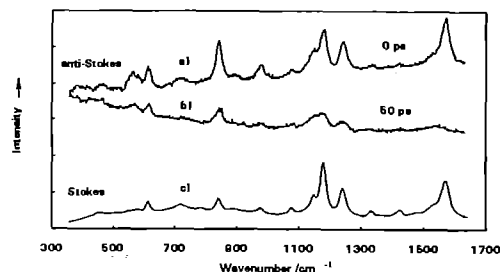


Figure 1 Anti-Stokes resonance Raman spectra of photoexcited S_1 *trans*-stilbene in *n*-hexane at (a) 0 ps and (b) 50 ps pump/probe time delay; and (c), scaled Stokes spectrum at 10 ps time delay. The pump and probe wavelengths were 305 and 610 nm, respectively, and the temperature of the sample reservoir was 16 °C.

The time dependence of the four normalised AS band intensities are shown in Fig. 2(a). The intensities are seen to decay with a characteristic timescale of *c.* 10 ps and relaxation is virtually complete by 50 ps. The small differences in shape at very early times in the data of Fig. 2(a) are not at present established, and deconvolutions are not presented in this first analysis. Fig. 2(b) compares the time dependence of the normalised intensity of the 1570 cm^{-1} AS band with the change in the position of the Stokes band at 1570 cm^{-1} relative to that at 1180 cm^{-1} . These time dependencies are clearly very similar.

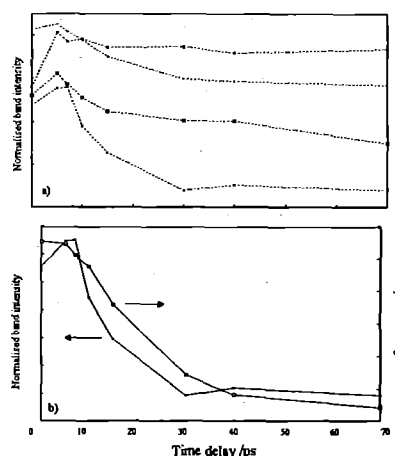


Figure 2 (a) Time dependence of selected AS band intensities ($\blacksquare$ 1570, $\square$ 1240, $\blacklozenge$ 1180 and $\circ$ 1150 cm^{-1}) normalised to the integrated area of the *c.* 1530 and *c.* 1570 cm^{-1} Stokes bands. (b) Comparison of the time dependencies of the normalised 1570 cm^{-1} AS band intensity and the position of the Stokes band at 1570 cm^{-1} , relative to the 1180 cm^{-1} band. Pump/probe wavelengths were 295/590 nm for (a) and (b).

Extensive studies of the sensitivity of our observations to pump and probe intensity were made, including measurements at 800 kHz with 1 nJ and 7 nJ energies in pump and probe pulses, respectively. A depletion of the S_1 state was demonstrated for the Stokes signal at very high probe intensities, but all of our main observations were reproduced at low intensities, i.e. the shapes of the AS spectra at both early and late time delay, their relative intensities, and the magnitudes of the AS/S intensity ratios.

DISCUSSION

The AS spectra in Figure 1 indicate the formation of vibrationally hot ($v > 0$) S_1 tS at short pump-probe delay times. Fig. 2 shows that relaxation to thermal equilibrium takes place on a c. 10 ps timescale and is complete by 50 ps. However, the populations of the excited vibrational levels at early times cannot be deduced directly from the changes in the AS/S intensity ratios between early and late times because, as also recognised by others¹⁰, the Raman excitation profiles may change during the relaxation process. This may contribute to the anomalous effect demonstrated in Fig. 1, where the 1570 cm^{-1} band is not observed in the AS spectrum at 50 ps but the neighbouring 1530 cm^{-1} band and those at 1420 and 1330 cm^{-1} are clearly seen. We estimate the AS/S intensity ratio of the 1530 cm^{-1} band at 50 ps to be at least 150 times the Boltzmann factor. A detailed analysis of the resonance enhancement factors, using the S_1 electronic absorption band profile, will be required to reveal the origin of this unexpectedly high AS/S intensity ratio.

Whatever its precise origin, the magnitude of the anti-Stokes signal and its rapid change with time requires a re-examination of the mechanism producing the changes in the Stokes spectrum reported earlier⁴. In the case of modes having significant anharmonicity, band position and bandwidth changes in the Stokes spectrum will result in part from the superposition of time-dependent Stokes $v = 1$ to $v = 2$ hot bands which might reasonably be supposed to have intensities similar to those of the AS $v = 1$ to $v = 0$ bands since both sets of bands arise from population of the $v = 1$ level. A study of the 1570 cm^{-1} region of the Stokes spectrum suggests that a substantial part of the time dependent effects seen previously may arise from this cause.

The results of a new Lorentzian analysis of the $1500\text{--}1600\text{ cm}^{-1}$ region of the Stokes spectrum for time delays ranging from -10 to $+50$ ps showed this point. A good fit is obtained at long delay times (c. 50 ps) by using three components, in accord with earlier work on S_0 tS¹¹ and on T_1 tS¹², both of which show three bands in this region. The c. 1570 cm^{-1} component is assigned to ν_7 , the olefinic $C_0=C_0$ stretch, and the 1528 cm^{-1} component to ν_9 , an aromatic ring stretching mode². The third component at 1565 cm^{-1} is attributed to ν_8 , which is another mode of mainly aromatic ring stretching character. We now find that fits at short delay times are improved by adding a fourth band at 1554 cm^{-1} , which we attribute to the $C_0=C_0$ mode in the vibrationally hot state ($v = 1$) of S_1 tS. This grows in to a maximum at c. 5 ps and then decays rapidly; in parallel with this, the 1570 cm^{-1} component, which arises from the corresponding mode in $v = 0$, grows in as the 1554 cm^{-1} band decays. The temperature-dependent changes in the Stokes spectrum of S_1 tS reported previously⁴ have not been fitted to this interpretation.

CONCLUSIONS

The anti-Stokes spectrum of photoexcited S_1 *trans*-stilbene has been observed at wavenumbers up to 1570 cm^{-1} . Mode-specific, time-dependent changes in band intensity on a c. 10 ps timescale parallel the changes in the Stokes spectrum reported previously. The anti-Stokes to Stokes intensity ratios of the high wavenumber S_1 bands are large relative to the Boltzmann factor.

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PICOSECOND TIME-RESOLVED RESONANCE RAMAN SPECTROSCOPY OF PHOTOCHROMIC SPIRO-INDOLINO-NAPHTHOXAZINES

D.R. Worrall¹, F. Wilkinson¹, S.L. Williams¹, L.M.G. Jansen¹, and P. Matousek²

¹ University of Technology, Loughborough, Leicestershire LE11 3TU

² Rutherford-Appleton laboratory, Didcot, Oxfordshire

INTRODUCTION

Naphthoxazine-spiro-indolino-(NOSI) compounds which are comprised of two heterocyclic moieties connected by a tetrahedral linkage represent an important class of photochromic compound having superior resistance to prolonged UV irradiation compared with, for example, the related spiro-pyran compounds¹. NOSI compounds undergo a photoisomerisation from the spiro- form following irradiation in the UV region of the spectrum, which causes a breaking of the carbon-oxygen bond at the spiro- linkage (figure 1).

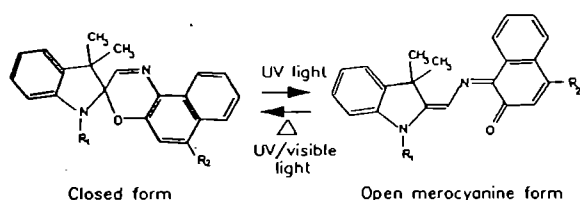


Figure 1: Photoisomerisation reaction in NOSI compounds

Subsequent rotation about the remaining carbon-carbon bond forms a more planar transoid structure resembling that of a merocyanine dye which absorbs strongly in the visible region of the spectrum.

Previously² we have reported time resolved resonance Raman spectra for one particular NOSI compound (R_1 = methyl, R_2 = piperidine) in 1-butanol and cyclohexane solution. However, in these earlier studies although evolution of the bands in the 1-butanol spectrum could clearly be seen, the rate constants associated with this evolution could not be estimated due to the conditions under which the experiments were performed. In the current series of experiments the conditions were adapted such that rate constants for spectral evolution in both 1-butanol and cyclohexane could be extracted, and compared with those determined using femtosecond time resolved transient absorption measurements³.

EXPERIMENTAL

The excitation source comprises a frequency doubled mode locked Nd:YAG laser (Spectron) synchronously pumping a cavity dumped rhodamine 6G dye laser (Spectra-Physics) to give 6ps, 580nm pulses. These are amplified by six passes through a dye cell pumped at 4.46 KHz by a copper vapour laser. Pump pulses are produced by doubling the amplified

pulses in a BBO crystal. Resulting pump and probe powers were 0.9 and 2 mW, respectively. Data acquisition in the window 1100 to 1600 cm^{-1} is with a cooled CCD camera with accumulation times of 300 to 1200 seconds. The output power of the laser system is stabilised by monitoring the output of the copper vapour laser and adjusting this power with a variable neutral density filter. Pump-probe overlap over the length of the delay used was checked using the resonance Raman emission from a solution of DMQ in dioxane.

Cyclohexane and 1-butanol were spectrophotometric grade and were used as received. Cyclohexane solutions were used at room temperature while 1-butanol solutions were chilled to approximately 0°C in an ice bath to reduce the equilibrium concentration of the merocyanine form. Solutions were made to an absorbance of approximately 0.6 at 290nm (corresponding with a solution concentration of approximately 10^{-5} molar), and were irradiated in a flowing jet.

RESULTS AND DISCUSSION

Kinetic data was obtained for the (R_1 = methyl, R_2 = piperidine) NOSI compound in both cyclohexane and 1-butanol over the timescale 0 to 1500 ps. Analysis of the data obtained in cyclohexane revealed that the absolute peak areas of the bands in the spectrum all increase over a period of 80ps, after which no spectral evolution can be detected. This time duration compares very well with a rise time of 70ps for the same compound in the same solvent determined using femtosecond transient absorption spectroscopy analysing at 436nm. Hence this provides the first evidence that the transients seen in the femtosecond transient absorption work are involved in the evolution to the photomerocyanine.

In the work in 1-butanol, the absolute Raman band intensities proved not to be diagnostic of the changes occurring in the spectra, possibly as a result of small fluctuations in temperature. However, as mentioned previously evolution of the relative intensities of the Raman bands can clearly be seen (figure 2), and consequently the ratio of peak areas was taken as the parameter which should be diagnostic of the relative rates of evolution.

Taking the relative intensities of the peaks at 1260 and 1425 cm^{-1} , a plot of the ratio of these peak areas versus time is shown in figure 3. this clearly demonstrates two time components associated with the evolution of the spectrum. Fitting of the data in figure 3 to a biexponential function yields lifetimes associated with the two time components of 36 ± 15 ps and 1.3 ± 0.2 ns, which compare favourably with those determined using femtosecond transient absorption spectroscopy analysing at 436nm, where lifetimes of 30 ± 2 ps and 1.4 ± 0.1 ns were associated with the observed transients.

This correlation again provides evidence that the transients observed at 436nm are in fact directly involved in the photoisomerisation process.

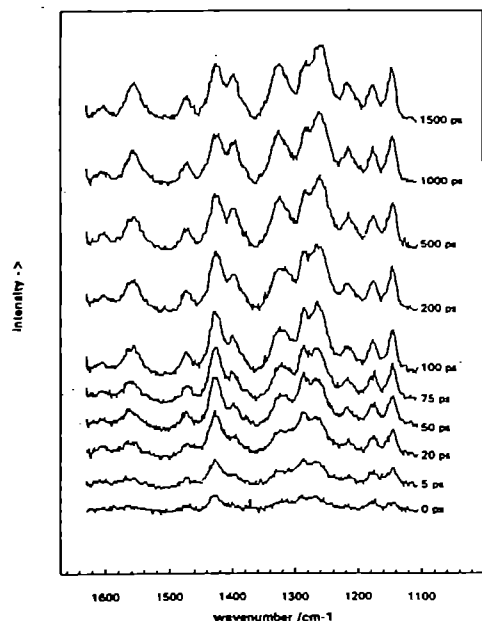


Figure 2: Change in resonance Raman spectrum with time for the (R_1 = methyl, R_2 = piperidine) substituted NOSI compound in 1-butanol

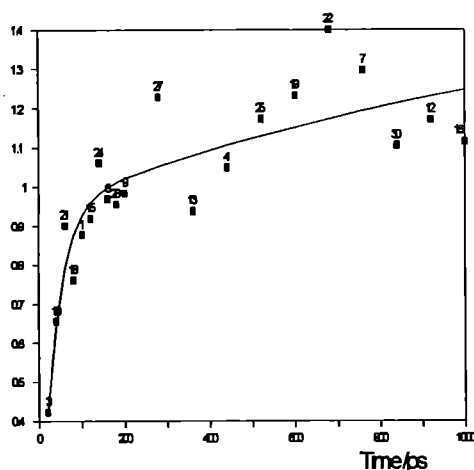


Figure 3: Variation of the ratio of the peak at 1260 cm^{-1} to that at 1425 cm^{-1} . The numbers near the points indicate the order in which they were recorded.

Hence there are at least two time components associated with the evolution to the final photomerocyanine spectrum, which is identical with that determined for this compound in 1-butanol using nanosecond resonance Raman spectroscopy.

CONCLUSIONS

The kinetic data extracted from the time resolved resonance Raman data presented here clearly demonstrate a correlation between this and data recorded analysing at 436nm using femtosecond time resolved transient absorption techniques. At least two metastable intermediates are evident in 1-butanol in going from the naphthoxazine-spiro-indoline form to the photomerocyanine. Both transient absorption and resonance Raman data suggest a simpler picture in cyclohexane.

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THE PICOSECOND TRANSIENT RAMAN SPECTRA OF S_1 EXCITED OLIGOPHENYLS, OXAZOLS AND STILBENE DERIVATIVES

M.Towrie,¹ R. E. Hester², P.Matousek¹, A.W.Parker¹, W.T.Toner¹

¹Central Laser Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxfordshire, OX11 0QX, UK

²Department of Chemistry, University of York, Heslington, York, YO1 5DD, UK

INTRODUCTION

Conjugated organic molecules have attracted much attention in recent years for several reasons: i) for their nonlinear optical properties, ii) to promote an understanding of conjugated π -electron systems, and iii) as oligomers in photoconductive polymers. To date Raman spectroscopy has been usefully employed to analyse the structure of such molecules in their electronic ground states but the time-resolved Raman technique has so far met with only limited success in studies of excited electronic states of such systems. This is mainly due to interference from strong fluorescence of the excited singlet states. The ps-TR³ system is capable of recording the resonance Raman spectra of the singlet excited states of a number of p-oligophenyls, phenyloxadiazols (commonly used as UV/blue laser dyes) and a few derivatives of stilbene. A full paper has been submitted for publication¹.

EXPERIMENTAL

The ps-TR³ pump and probe pulse energies were 0.1 - 2 μ J and accumulation times were 100 - 600 s. The compounds were purchased from Aldrich, Exciton and Lambda Physik and used without further purification. The concentration of solute in all solutions was 1×10^{-3} mol dm⁻³. The following abbreviations are used:

p-terphenyl / PTP
quaterphenyl / QP
2-methyl-5-t-butyl-p-quaterphenyl / DMQ
4,4'-bis-(2-butyloctyloxy)-p-quaterphenyl / BBQ
Exalite 389 / E389
3,5,3'',5'''-tetra-t-butyl-p-quinquephenyl / QUI
3,5,3'',5'''-tetra-t-butyl-p-sexiphenyl / TBS
Exalite 428 / E428

2-(4-biphenyl)-5-phenyl-1,3,4-oxadiazol / PBD
2-(4-biphenyl)-5-(4-t-butylphenyl)-1,3,4-oxadiazol / BPBD
2,5-bis-(4-biphenyl)-1,3,4-oxadiazol / BBD
2-(4-biphenyl)-6-phenylbenzoxazol-1,3 / PBBO

trans-stilbene / tS/Diox
4,4'-dimethoxystilbene / DMtS
trans-p,p'-diphenyl stilbene / DPtS
stilbene 1 / tS1
4,4'-diamino-stilbene-2,2'-disulfonic acid / DtSDA
1,2,3,4-tetraphenyl-1,3-cyclopentadiene / TPC

RESULTS AND DISCUSSION

Figures 1, 2 and 3 show the ps-TR³ spectra of several oligophenyl, oxazols and stilbene derivatives in the 1100 to 1600 cm⁻¹ range.

Approximate scaling factors are listed beside each spectrum and molecular structures are given. The structure of E428 is similar to that of 'Dye number 8' described by Seliskar *et al.*². The scaling

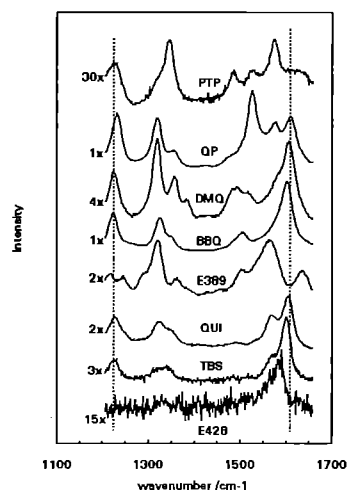


Fig. 1 Ps-TR³ spectra of photoexcited oligophenyls. The spectra were obtained at pump and probe wavelengths of 305 and 610 nm respectively, from dioxane solutions with the exception of TBS which was dissolved in THF.

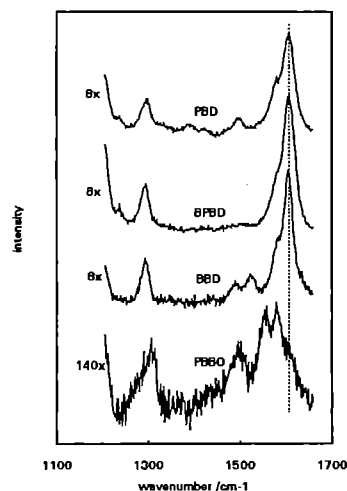


Fig. 2 Ps-TR³ spectra of photoexcited oxazols. The spectra were obtained at pump and probe wavelengths of 305 and 610 nm respectively, from dioxane solutions.

factors are derived by normalising to BBQ but are not absolute since at the pump and probe energies used the excitations were possibly in saturation for some molecules and the laser power was controlled to only $\pm 50\%$. The spectra were all taken with 20 ps delay between the pump and probe laser pulses at 305 and 610 nm, respectively.

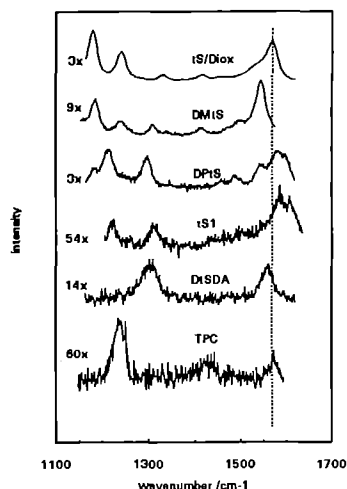


Fig. 3 Ps-TR³ spectra of photoexcited stilbene-like molecules. The spectra were obtained at probe and pump wavelengths of 305 and 610 nm respectively, from dioxane solutions with the exception of DtSDA which was dissolved in water (pH = 12).

The ps-TR³ spectra all were observed to decay on a timescale characteristic of the excited state lifetimes. However, by using the picosecond Raman technique we have observed more subtle relaxation processes taking place on shorter timescales than the excited state lifetime. We and others have observed such rapid and subtle changes in the ps-TR³ spectra of S₁tS in solution and more recently on DMtS and quinquiphenyl^{3,4,5,6,7,8}. These changes occur on a timescale of a few picoseconds and manifest themselves as shifts and broadening of Raman bands, in particular for vibrational modes associated with the olefinic portions of the molecules. For the oligophenyls and oxazols, where the ps-TR³ signal-to-noise ratio is large, similar changes are observed. Fig. 4 shows the difference between spectra taken at 5 and 50 ps for PBD. The c.1600 cm⁻¹ band is shifted to higher wavenumber and gets narrower on going from 5 to 50 ps delay. The origin of this effect is not yet known.

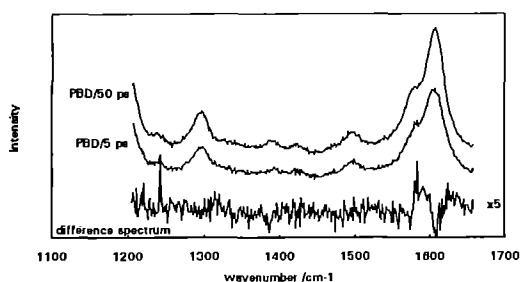


Fig. 4 Time dependence of PBD spectrum

The 5 and 50 ps difference spectra for QP (shown in Fig. 5) and TBS show very different behaviour. Cancelling the c.1520 cm⁻¹ band in the subtraction leaves structure which is associated with a short-lived excited state or twisted configuration of the molecule. QP and TBS are not unique; the quaterphenyls and QUI all show structural differences of this type. This effect will be the subject of further investigation.

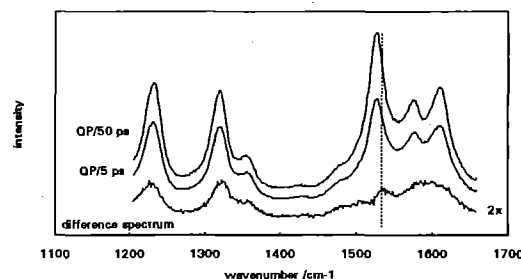


Fig. 5 Time dependence of QP spectrum

SUMMARY

We have demonstrated ps-TR³ can be used to investigate the structure and dynamics of several *p*-oligophenyls, phenyloxadiazols, and derivatives of stilbene. This work demonstrates that even highly fluorescent molecules can be studied using this technique. This preliminary study shows the presence of fast structural changes of short-lived, sub-50 ps, transients. Further work should attempt to clarify whether this impairs the efficiency of the radiative processes of the molecules. The technique has great potential for studies of electron transfer and TICT states.

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AN INVESTIGATION OF CAROTENOID RADICAL CATIONS BY TIME RESOLVED RESONANCE RAMAN

J H Tinkler¹ S M Tavender² A W Parker² D J McGarvey¹ L Mulroy¹ and T G Truscott¹

¹ Chemistry Department, Keele University, KEELE, Staffs. ST5 5BG

² Laser Support Facility, Rutherford Appleton Laboratory, Chilton, DIDCOT, Oxon. OX11 0QX

INTRODUCTION

Carotenoids are present in the chloroplasts of green plants^{1,2}, where they have at least two functions. One being that of an accessory pigment in which light energy is absorbed and transferred to chlorophyll and the other being that of a protective species in which triplet states of chlorophyll are quenched, eliminating the production of damaging singlet oxygen, or possibly by direct quenching of singlet oxygen. It is thought that the primary process of photosynthesis involves electron ejection from chlorophyll *a* and various oxidation/reduction reactions. Therefore, the properties of the radical cations of carotenoids (and chlorophylls) are of importance. Indeed, it has been reported^{3,4} that upon light excitation of chloroplasts absorption changes due to the carotenoid radical cation have been observed at reaction centres.

Carotenoid radical cations are also produced by the reaction of carotenoids with many oxy-radicals^{5,6}, such as $\text{CCl}_3\text{O}_2^\cdot$, NO_2 and $\text{NO}_3^\cdot$, yet little is known about the properties of these radical cations.

Previous work using resonance Raman spectroscopy has provided some structural information on excited singlet and triplet states^{7,8} of carotenoids. We now report the use of this technique to study the radical cations of 7,7' dihydro β -carotene (77DH) and septapreno β -carotene (SEPT), see Figure 1, along with their respective ground state spectra. To date, all work on carotenoid radical cations has been performed using the technique of pulse radiolysis. For TR³ studies an essential first step was to develop an efficient photochemical method for their generation. Our earlier attempts involving two-photon ionisation of 77DH and the addition of tetranitromethane achieved promising results but we now report on an improved method using 1-nitronaphthalene (NN).

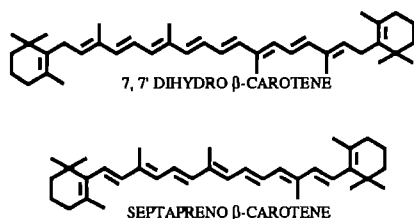
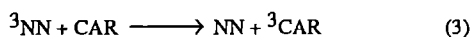
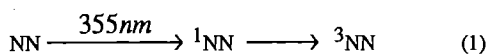


Figure 1. Carotenoid structures

RESULTS AND DISCUSSION

Carotenoids ($2 \times 10^{-5}\text{M}$) were dissolved in methanol and 1-nitronaphthalene (10^{-3}M) was added and bubbled with nitrogen. Laser flash photolysis (355nm) produces the triplet state of NN (^3NN) which then reacts with the carotenoid (CAR) via electron transfer to produce the radical cation or by energy transfer to produce the triplet carotenoid.



In methanol we obtained 65% energy transfer and 35% electron transfer. The carotenoid radical cations absorb in the near infra-red region, Figures 2 and 3 whilst the triplets absorb typically around 450nm.

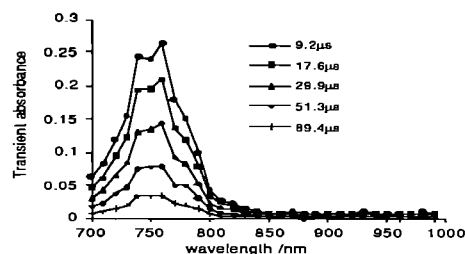


Figure 2. Transient absorbance spectrum of 7,7' dihydro β -carotene radical cation in methanol.

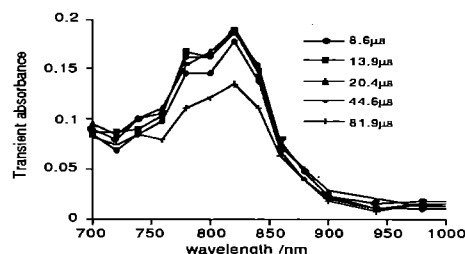


Figure 3. Transient absorbance spectrum of septapreno β -carotene radical cation in methanol.

For the radical cation Raman spectra, the laser excitation sources are described by Conn et al⁷ with the pump wavelength at 388nm (1.5mJ/pulse at sample) and the probe 760nm (0.5mJ/pulse at sample). The delay between the pump and probe was set at 2 μ s. The carotenoid solutions were flowed through a quartz capillary tube and the Raman light collected at 90°. The light detector was a back illuminated liquid nitrogen cooled CCD camera with a 1024x1024 detector chip controlled by a model ST-130 controller interfaced to CSMA software. Ground state Raman spectrum were obtained by c.w. excitation using an argon ion laser at 457.6nm

The corrected TR³ spectra of ground state and radical cation are shown in Figures 4 and 5 and the corresponding bands are tabulated in Table 1.

Both ground state carotenoid spectra show the expected similarities with ground state all-*trans* β -carotene.⁹ The most intense peaks observed at 1551cm^{-1} and 1541cm^{-1} for 77DH and SEPT respectively are thus assigned to the C=C. In the carotenoid radical cation this spectral region shows changes. For the 77DH⁺ spectrum the C=C band shifts to a lower frequency at 1541cm^{-1} and for the SEPT⁺ spectrum the C=C band shifts again to a lower frequency at 1517cm^{-1} . The strong ground state bands at 1162cm^{-1} for 77DH and 1155cm^{-1} SEPT are assigned to the C-C stretch and these shift to higher frequencies of 1188cm^{-1} and 1173cm^{-1} respectively. These frequency shifts can be explained by considering the conjugated structure of the carotenoid molecule. In the ground state the molecule shows clear and

regular bond alternation with the C-C bonds longer than the C=C bonds. To form the radical cation, an electron is removed from a π -bonding orbital to leave a positive charge and to stabilise the system the molecule becomes considerably more delocalised, lengthening the C=C bonds and shortening the C-C bonds. This is reflected in the Raman spectra with the C=C and C-C bands becoming closer together in the radical cation spectra, as the molecule heads towards a uniform bond structure.

Another spectral feature in the 77DH^+ spectra is the splitting of the C=C into three bands. A possible explanation is that within the conjugated system there are two C=C modes i) a methylated (*trans* C-CH=C(Me)-C) and ii) an unmethylated (*trans* C-CH=CH-C). The third Raman band is tentatively assigned to the C=C in the β -ionone rings. Formation of the strongly delocalised radical cation allows these modes to be resonantly enhanced.

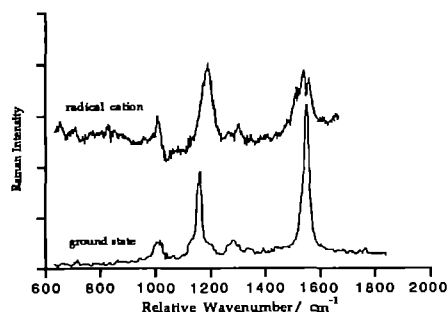


Figure 4. Resonance Raman spectra of ground state and radical cation of 7,7' dihydro β -carotene in methanol.

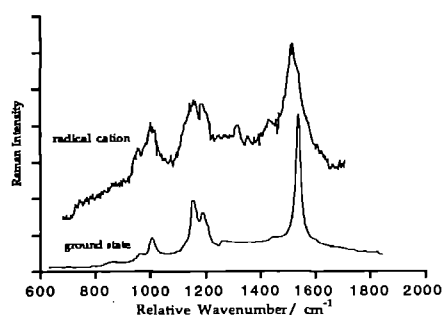


Figure 5. Resonance Raman spectra of ground state and radical cation of septapreno β -carotene in methanol.

	77DH	77DH ⁺	SEPT	SEPT ⁺
MeOH				
C-H str	1009 m	1009 m	1008 m	1008 s
C-C str	1162 s	1188 vs	1155 s 1191 s	1173 s
C=C str	1551 vs	1514 s 1541 s 1563 s	1541 s	1517 s 1536 sh
TX100				
C-H str	1010 m	1005 s	1008 m	1007 s
C-C str	1134 s 1162 w	1162 sh 1188 s	1156 s 1192 m	1132 sh 1178 s
C=C str	1548 s	1516 s 1537 s 1558 s	1539 s	1515 s 1538 sh

Table 1: TR^3 frequencies for carotenoids.

To investigate the effect of environment on the carotenoid radical cations we also obtained their TR^3 spectra, along

with the corresponding ground states, in the neutral detergent Triton X-100 micelles (TX-100). Laser flash photolysis studies leads to the radical cations of 77DH and SEPT at 770nm and 860nm respectively. Their TR^3 spectra are shown in Figure 6 and 7, with their frequency shifts in Table 1, and show very similar characteristics to those obtained in methanol with the exception of the intensity of the C=C, 1560 cm^{-1} band. This band being more intense in TX-100. This would tend to suggest that the carotenoid radical cation spectrum may be sensitive to environmental changes in contrast to triplet state carotenoids.⁷

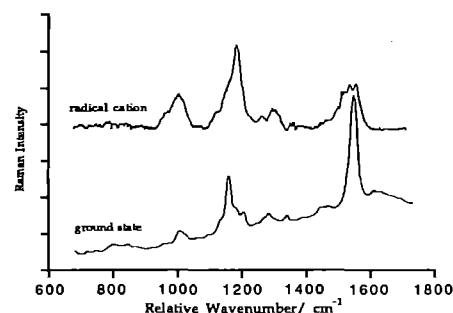


Figure 6. Resonance Raman spectra of ground state and radical cation of 7,7' dihydro β -carotene in TX-100.

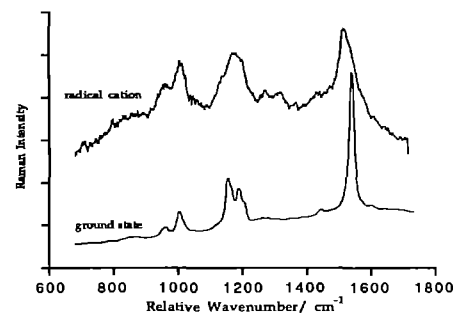


Figure 7. Resonance Raman spectra of ground state and radical cation of septapreno β -carotene in TX-100.

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A TIME RESOLVED RESONANCE RAMAN STUDY OF ENOLISED 1,3-DIKETONES USED AS UV-A SUNSCREENS

Andrew Beeby¹, Catherine Coultous¹, David J. McGarvey², Louise Mulroy², Susan Tavender³ and T. George Truscott²

¹ Department of Chemistry, University of Durham, Durham DH1 3LE

² Department of Chemistry, University of Keele, Keele, Staffs. ST5 5BG

³ Central Laser Facility, Rutherford Appleton Laboratory, Chilton, Didcot, OX11 0QX

INTRODUCTION

The aim of this study is to characterise the photoproduct distribution of a range of enolised aromatic 1,3-diketones which are currently used as UV-A sunscreens and photostabilisers of polymers. These compounds exist almost exclusively as the cyclic hydrogen bonded enol form, (I), and exhibit strong absorption bands in the UV-A region. The parent compound dibenzoylmethane, DBM, has bands extending to 370nm and this may be fine tuned by substitution of the aromatic rings; for example the sunscreen Parsol 1789, 4-*t*-butyl-4'-methoxydibenzoylmethane, absorbs up to 410nm. These enolised 1,3-diketones have facile relaxation pathways available to their excited states, allowing them to absorb the potentially damaging UV radiation and dissipate the energy harmlessly as heat to the surroundings [1]; some of these pathways are illustrated in figure 1. A range of possible reactions may occur giving rise to a number of different products and the relatively few studies reported have been somewhat contradictory. For example it has been reported that irradiation of DBM produces both an enol and keto form [2,3]. Flash photolysis studies by our groups have shown the presence of a transient species absorbing at 290nm and the data suggested that this was due to a non-H-bonded enol form [4]. However, because of similarities between the absorption spectra of model compounds locked into the keto and enol forms in the wavelength range <270nm this data is not conclusive and further evidence was needed given the ambiguity of the literature on the identity of the photoproducts. The experiments reported here were carried out in order to further characterise the transient species produced upon irradiation of DBM and Parsol 1789 in non-polar solvents using ns time-resolved resonance Raman spectroscopy, TR³.

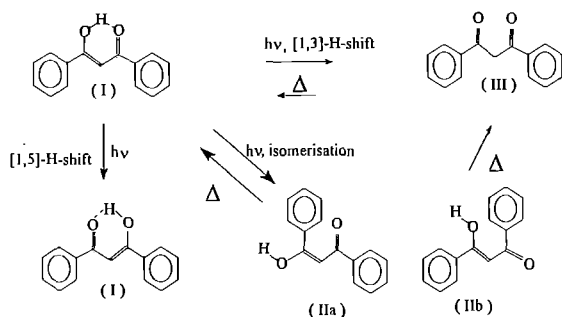


Figure 1 Possible pathways following irradiation of dibenzoylmethane

RESULTS AND DISCUSSION

A solution of dibenzoylmethane in 1,1,2-trichloro trifluoroethane was pumped at 355nm and the transient species probed at 295nm. This gave Raman spectra showing a new band at 1618cm⁻¹ with a shoulder at ca. 1583cm⁻¹, no other bands could be observed in the range 3500-1000cm⁻¹. In order to characterise this species we also obtained the resonance Raman ground state spectra of model compounds that are locked into the keto and enol forms by C and O-methylation

respectively. These clearly showed the presence of two bands at 1602 and 1698cm⁻¹ for the keto form, assigned as a aromatic C-C stretch and C=O stretch, whilst the enol has a closely spaced, double band at 1574 and 1602cm⁻¹, assigned as an aromatic C-C stretch. These spectra are illustrated in figure 2. This, in conjunction with flash photolysis data, is compelling evidence for the formation of a non-H-bonded enol isomer of DBM following excitation in this solvent. Similar results were also obtained when DBM was irradiated in cyclohexane and also from samples of Parsol 1789.

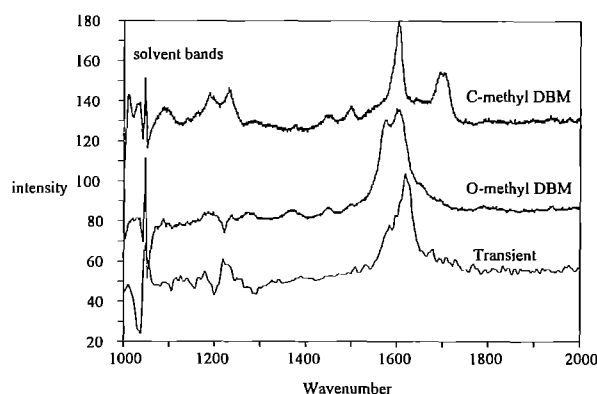


Figure 2 Resonance Raman spectra of transient species of DBM and locked keto and enol model compounds. Probe 295nm, pump 355nm, delay 1μs (transient only)

CONCLUSIONS

Irradiation of the H-bonded enolised 1,3-diketones in non-polar solvents produces a ring opened, non-H-bonded form and not the keto form as has been reported by other groups. This is a very significant finding since it is established that the keto forms of these compounds are efficient sensitisers of singlet oxygen, $\Phi_{\Delta} \approx 0.3$, whilst the enol form does not generate singlet oxygen.

However, at present it is not clear which isomer is formed, (IIa or IIb), nor the pathway, although flash photolysis experiments have demonstrated that the growth of the species is rapid, <50ns and unaffected by oxygen. It will also be necessary to investigate the photochemistry of these compounds in polar solvent systems, where it has been established by our group that long term photolysis of DBM gives rise to the formation of the diketone, (III).

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FLASH PHOTOLYSIS OF PHTHALOCYANINE SENSITISERS FOR PDT: SULPHONATED ALUMINIUM PHTHALOCYANINES IN UNILAMELLAR VESICLES

S Dhami & D Phillips

Department of Chemistry, Imperial College of Science, Technology & Medicine, South Kensington, London, SW7 2AY

Currently, there are two major groups of phthalocyanine compounds under investigation for use as photosensitisers in the Photodynamic therapy (PDT) of tumours: (i) water-soluble compounds such as di-sulphonated aluminium phthalocyanine (AlPcS₂) and (ii) lipophilic sensitisers such as zinc phthalocyanine. These sparingly water-soluble photosensitisers are incorporated into compartmentalised systems, such as liposomes, to facilitate direct injection into the bloodstream. Liposome-associated photosensitisers are accumulated by experimental tumours with a high degree of selectivity¹. This selectivity is generally thought to be governed by the association of the sensitizer and carrier with serum lipoproteins, particularly the low density (LDL) and high density (HDL) fractions² during delivery and are thought to accumulate in tumour tissue as this expresses an increased number of LDL receptors^{3,4}. On these bases, it appears important to characterise the spectroscopic and photophysical properties of photosensitisers incorporated into liposomal formulations.

There are two possible modes of photosensitisation of water-soluble sensitisers. The first is simple accumulation of the sensitizer at the cell surface which will cause damage to the cell membrane upon irradiation. The second process, known as endocytosis, allows macromolecules which are too large and too hydrophilic for simple diffusion to gain entry into the cell. In this case, the sensitizer is concentrated in lysosomes located within the cell which are damaged upon irradiation, thus releasing the sensitizer and other cell-destroying lysosomal enzymes. Liposomes or unilamellar vesicles provide good working models to mimic lysosomal membranes.

In this study, the spectroscopic and photophysical properties of aluminium phthalocyanines incorporated into unilamellar vesicles of dipalmitoylphosphatidylcholine (DPPC) have been elucidated. The cis di-sulphonated aluminium phthalocyanine (AlPcS₂) showed no aggregation when incorporated into the liposomal dispersions. Consequently, only this system will be discussed here.

The liposome dispersions were prepared by the injection method of Kremer *et al.*⁵. This procedure allows the preparation of unilamellar phospholipid vesicles of pre-determined sizes through the injection of an ethanolic solution having different phospholipid concentrations into an aqueous buffered solution (0.01 M Tris-HCl, 0.1 M NaCl; pH = 7.4). Small unilamellar vesicles (SUVs) (radius, 26 nm, as estimated by electron microscopy⁶) and large unilamellar vesicles (LUVs) (radius, 42 nm) of DPPC were prepared using 9.5 mM or 32 mM phospholipid solutions in absolute ethanol. The desired phthalocyanine concentrations in the liposomes were obtained by adding suitable volumes of a stock (1 mM) pyridine solution of the sensitizer to the ethanolic solution of DPPC. The injection was performed at 55 °C under magnetic stirring and at a speed of 1 $\mu\text{l s}^{-1}$. Following injection, the system was allowed to cool to room temperature and the solution was passed through a 450 nm Millipore filter. The organic solvents and any 'free' phthalocyanine were eliminated by dialysing the liposome aqueous dispersion at 4 °C against 250 ml of the tris buffer for 2 hours with one change of the buffer after the first hour. Dialysis tubing was sterilised in EDTA / bicarbonate solutions prior to use. After preparation, all solutions were stored in the dark at 3-4 °C. The cis-isomer of AlPcS₂ used has both sulphate groups substituted to the same side of the molecule.

Triplet state studies were carried out using the nanosecond laser flash photolysis system developed by Beeby *et al.*⁶ A pump wavelength of 670 nm was used which corresponds to the

maximum absorption of AlPcS₂ and also minimises any light scattering produced from the liposomes. The T-T absorption maximum was found to occur at 490 nm, corresponding to the value observed in homogeneous solutions⁷. Triplet quantum yields were determined by the relative method using AlPcS₂ in methanol ($\Phi_t = 0.24$, $\epsilon_t = 36\,000\text{ M}^{-1}\text{cm}^{-1}$ at 490 nm)⁷ and ZnPcS₂ in methanol ($\Phi_t = 0.46$, $\epsilon_t = 36\,000\text{ M}^{-1}\text{cm}^{-1}$ at 490 nm)⁷ as standards.

The variation of triplet quantum yield (Φ_t) with AlPcS₂ concentration in LUVs and SUVs is shown in table 1. The triplet quantum yields are seen to decrease with increasing concentration. In the case of AlPcS₂ in LUVs this decrease is linear with concentration. In both cases, the decrease in triplet quantum yield mirrors a decrease in the fluorescence quantum yield. The deviation of the plots from linearity of Φ_f and Φ_t versus concentration in SUVs indicates the possible formation of dimers which are not evident in the absorption spectra. The triplet lifetimes were found to be $\sim 1000 \pm 100\text{ }\mu\text{s}$ in SUVs and $\sim 1200 \pm 100\text{ }\mu\text{s}$ in LUVs and did not vary significantly with sensitizer concentration. The decrease in quantum yield is believed to be due to self-quenching of the phthalocyanine excited state by ground state molecules as a result of the increased endoliposomal sensitizer concentration which will encourage the close approach of two or more phthalocyanine molecules without necessarily inducing dimerisation. Similar results have been observed for AlPcS₂ in leukaemia cells where an endocytotic mechanism of cell entry was hypothesised⁸. The results obtained from this study along with other fluorescence work carried out at Imperial reinforce this mechanism.

LUVs		SUVs	
Conc / μM	$\Phi_t (\pm 0.05)$	Conc / μM	$\Phi_t (\pm 0.05)$
0.76	0.24	0.25	0.20
3.04	0.19	1.01	0.14
5.51	0.17	2.67	0.12
8.17	0.11	4.0	0.10
10.45	0.06	7.03	0.09
12.46	0.04		

Table 1 Triplet Quantum yields of cis-AlPcS₂ in DPPC LUVs and SUVs with varying sensitizer concentration.

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FLASH PHOTOLYSIS OF PHTHALOCYANINE SENSITISERS FOR PDT: THE EFFECT OF PH ON SOME PHOTOPHYSICAL MEASUREMENTS OF SULPHONATED ALUMINIUM PHTHALOCYANINES

R.Ostler & D.Phillips

Department of Chemistry, Imperial College of Science, Technology & Medicine, South Kensington, London, SW7 2AY

"Clinical" di-sulphonated aluminium phthalocyanine is a mixture of water soluble disulphonated isomers and is currently undergoing medical trials for use in photo dynamic therapy.

Although the exact mechanism of phototoxicity is not known it almost certainly involves the formation of cytotoxic singlet oxygen ($^1\Delta_g$), produced as a result of energy transfer from the excited triplet state of the photosensitiser to ground state triplet oxygen. In order to optimise the quantity of singlet oxygen produced it is necessary to maximise both the triplet quantum yield (Φ_t) and triplet state lifetime (τ_t) of the phthalocyanine.

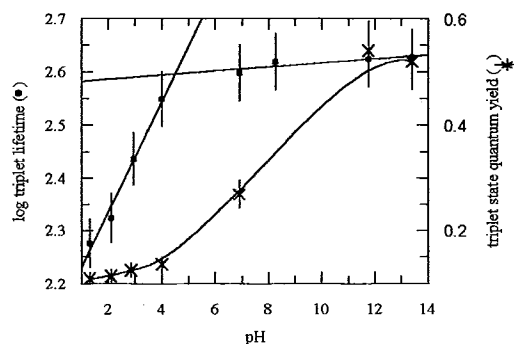
In the aqueous biological environment the photosensitiser may be exposed to a range of pH values. The pH of homogenous solutions affects the aggregation of disulphonated aluminium phthalocyanine, lower pH leading to increased aggregation. Aggregated phthalocyanine undergoes internal conversion as the major deactivation pathway *Darwent et al*¹. It is therefore of interest to equate both the triplet state quantum yield and triplet state lifetime to pH value. The aim of this study was to investigate the changes in these parameters with pH.

The solutions used in this study were of constant ionic strength (0.1) and freshly prepared using deionised water, the pH being adjusted by the addition of HCl and NaOH with NaCl being used to maintain ionic strength. The solutions were deoxygenated by argon bubbling and had absorbances less than 0.6 at the pump wavelength.

Photophysical data was collected using nanosecond flash photolysis apparatus with a pump wavelength of 670 nm corresponding to absorption in the phthalocyanine Q band and a probe beam of 490 nm corresponding to the T-T absorption maximum² ($\epsilon_t = 48\,000\text{ M}^{-1}\text{cm}^{-1}$). Triplet state quantum yields were obtained by the comparative method using chlorophyll a in diethyl ether ($\Phi_t = 0.53$,) as a standard.

pH	τ_t	Φ_t
1.3	190	0.11
2.1	211	0.115
2.85	282	0.126
4.0	343	0.136
6.9	424	0.269
8.25*	387	-
11.75	420	0.54
13.38	466	0.52

Table 1 Results obtained with a probe beam of 670 nm except
* obtained using 640 nm.



photophysical parameters of clinical disulphonated aluminium phthalocyanine as a function of pH. (Error bars are 10% of measured value)

The decrease in the triplet quantum yield, as shown above, is consistent with pH dependent aggregation. The proton dependent quenching of the monomer triplet state is under investigation at present.

The variation in the parameters elucidated emphasises the importance of environment on photosensitiser efficacy. In this study the photosensitiser, which is anionic at neutral pH, aggregates at low pH undergoing a proton dependent quenching process. This implies that the localisation of hydrophilic photosensitisers in pH variant microenvironments is of major importance in determining their efficiency as PDT agents. It may be that in order to optimise the effect of photosensitisers in PDT, a "cocktail" of hydrophilic anionic, cationic and amphiphilic (e.g. cis-disulphonated aluminium phthalocyanine) photosensitisers be administered. Tumour localisation is of course of the utmost importance in choosing an effective photosensitiser, and although tumour selectivity has been demonstrated in sulphonated phthalocyanines^{3,4}, it has not been demonstrated in cationic phthalocyanines⁵.

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CHARACTERISATION OF THE NORTH COAST SCIENTIFIC EO817-P GERMANIUM PHOTODIODE AND OPTICAL FILTERS FOR AQUEOUS PHASE SINGLET OXYGEN STUDIES

TC Oldham and D Phillips

Department of Chemistry, Imperial College of Science, Technology and Medicine, London SW7 2AY

The mechanism of photodynamic therapy (PDT) of tumors remains somewhat of a mystery. Indirect physiological studies on living cells and direct photophysical measurements on model systems have led most workers to accept the crucial intermediacy of singlet oxygen ($^1\text{O}_2$) in the PDT mechanism. Recent studies on the application of PDT sensitisers to the lethal photosensitisation of microorganisms have done little to improve our knowledge of the dark processes immediately following excitation of the sensitiser inside cells.

In order to probe these processes directly and non-invasively, spectroscopic studies of the sensitisers and likely intermediates are required. $^1\text{O}_2$ has been detected by its emission at 1270 nm in the buffer surrounding living cells in suspension and models of its diffusion³ and static quenching of triplet states² await rigorous investigation. To evaluate and extend this work, and to correlate it with physiological studies, improved $^1\text{O}_2$ detection is required. $^1\text{O}_2$ luminescence decays are characterised by an intense, short lived pulse coincident with the laser pulse. This is most intense in the case of red emitting sensitisers such as aluminium disulphonated phthalocyanine (AlS₂Pc). This pulse distorts the early part of the $^1\text{O}_2$ decay and makes measurements in aqueous solution where $\tau_{\Delta} = 3.09 \mu\text{s}$ extremely unreliable. Inside living cells, τ_{Δ} is likely to be even shorter.

Given the short τ_{Δ} in systems of interest, accurate characterisation of the response function of the EO817-P detector is needed even if this spike is eliminated, since it will almost certainly need to be deconvoluted from experimental decays. (Iu *et al* reported a faster germanium detection system² which we will also investigate.)

Figure 1 shows the response of the diode to a single 10 ns pulse at 670 nm. In "L" mode, the rise time is slightly faster than in "H" mode due to its lower sensitivity in the former mode. The "L" mode is characterised by a fall time of 330 ± 4 ns. In "H" mode the response function is characterised by $\text{FWHM} = 296 \pm 10$ ns. The "H" mode response also shows a significant undershoot which distorts the early part of the decay (up to 1 μs) when the spike is present. Indeed, when the fluorescence spike is very large, it can lead to saturation of the diode and in such cases the diode may take up to 3 ms to return to its proper zero. The result is considerable distortion to at least the early part of any decay when the spike is present. This renders studies in aqueous solutions and cell suspensions very difficult to interpret. Even if the less sensitive "L" mode is used, very high S/N ratios are needed to enable the instrument response to be deconvoluted.

In the past, the short lived pulse has been attributed to residual fluorescence from the sensitisers at 1270 nm. This seems rather unlikely given the observed steep wavelength dependence of this emission in steady state studies and its displacement from the sensitiser emission maximum. Recently, Scurlock *et al*¹ attributed this emission to fluorescence from silicon and Schott glasses used in the bandpass filters commonly used to isolate 1270 nm. This in turn is excited by the sensitiser emission. Our present work supports this theory.

AlS₂Pc (synthesised in our group and characterised by HPLC according to the method of Bishop⁴), zinc tetraphenylporphyrin (ZnTPP), rose bengal (RB) and chlorophyll a (Chla) (all ex Sigma) were used as fluorophores without further purification. Distilled water and HPLC grade methanol (MeOH), ether (Et₂O) and toluene (Tol) were used as solvents. Solutions with absorbance of 0.3-0.5 at 355 nm were prepared. The North Coast Scientific EO817-P cooled germanium photodiode was used in the arrangement previously described.⁴ It was cooled with liquid nitrogen such that the DC voltage reading was -1.495 ± 0.001 V throughout the experiment. Samples were excited using 10 ns pulses at 355 nm. Using a scattering suspension of Ludox in water and no filters it was shown that the detector was not appreciably sensitive to the laser pulse. 1270 nm light was isolated using various combinations of a 1270 ± 2 nm narrow bandpass filter (NBP), a 1135 nm longpass filter (LWP) and a 450-750 nm cold mirror from Infrared Engineering Ltd. Care was taken to ensure that light did not leak around the filters.

The height of the spike was measured at at least three different laser powers for each fluorophore and found to be reasonably linear in all cases, confirming that the diode was not saturating. The emission spectrum of each fluorophore was recorded over the range 500-900 nm for a solution optically matched ($\pm 2.4\%$) to that used for

the detector analysis using a SPEX FluoroMax spectrometer. No correction was made for the spectrometer spectral response. Although this falls away in the near IR, the correction factors available appear to overcorrect in this region. The spike height was normalised for laser power, and the normalised value averaged. This averaged value was then normalised by the integrated emission intensity. Figure 2 shows the completely normalised spike height as a function of peak emission wavelength for the NBP case. It also shows the excitation spectrum of silicon and the emission spectra for silicon and a typical Schott glass.

The excellent correlation between our experimental points and the silicon excitation spectrum tends to confirm the results of Scurlock *et al*¹. There is also a weaker correlation between the normalised spike height and a spike in the transmission of the bandpass filter at 772 nm. To eliminate the latter effect, we placed the silicon free LWP filter before the NBP. The normalised spike heights increased, confirming that the spike was due to filter emission rather than filter transmission. In the case of the LWP, the emission was attributed to the Schott glass from which the substrate is made. Further increases were observed when the cold mirror was placed in front of the LWP. Final verification of the origin of the spike requires these measurements to be repeated using a blue emitting sensitiser. We plan to repeat the experiment using 9,10-dicyanonaphthalene in the near future. It is clear that the choice of substrate for the filters used to isolate 1270 nm emission can affect critically the sensitivity of the detector, particularly for short singlet oxygen lifetimes.

We conclude that for detailed $^1\text{O}_2$ studies in aqueous phases, detector speed must be increased and non-IR emitting NBPs used. We suggest sapphire as a possible alternative substrate for the latter. We continue to work on both problems.

Figure 1 EO817-P response function.

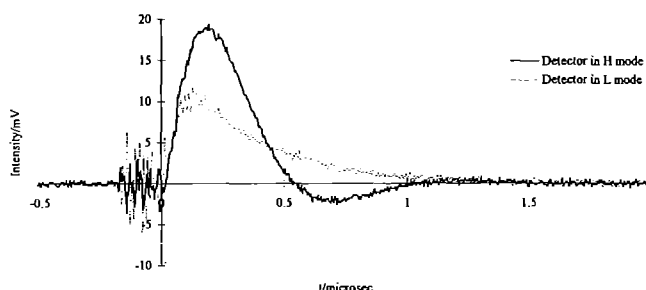
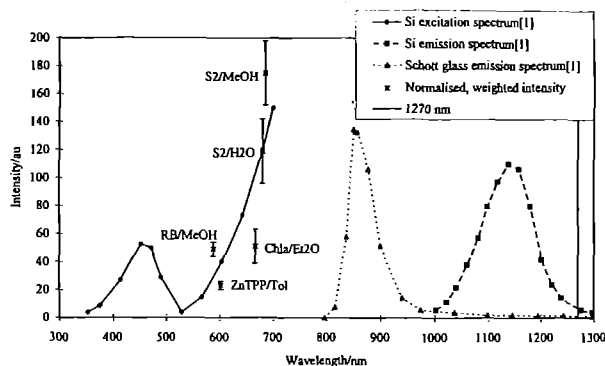


Figure 2 Correlation between completely normalised spike intensity and Si excitation spectrum.



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SOLUTION PHOTOPHYSICS OF ETHYL 4-(*N,N*-DIETHYLAMINO)BENZOATE

Robert Clark and Robert G. Brown

Chemistry Department, University of Central Lancashire

INTRODUCTION

The fluorescence properties of 4-(*N,N*-dimethylamino)- and 4-(*N,N*-diethylamino)- benzoic acid, various of their esters and their nitriles are anomalous in that at least two fluorescence bands are observed. In one interpretation, the shorter wavelength band is ascribed to "normal" fluorescence from the Franck-Condon excited state whilst the longer wavelength emission is considered to originate from a twisted intramolecular charge transfer (TICT) state¹⁻³. Alternatively, the latter fluorescence may be ascribed to exciplex formation⁴ (with the short wavelength band arising from a folded isomer of the Franck-Condon excited state) or to excimer formation⁵. We have also found evidence for exciplex formation (in mixed solvent systems⁶) but have found fluorescence properties of 4-(*N,N*-dimethylamino)-benzoic acid and its methyl ester (M4DMAB) in polar solvents which are suggestive of ground state complex formation^{7,8}. The photochemistry of these systems is clearly both complex and controversial and we have therefore continued the work reported last year⁶ by expanding the range of compounds and solvent systems under study. Here we report some of the photophysical properties of ethyl 4-(*N,N*-diethylamino)benzoate (E4DEAB).

RESULTS AND DISCUSSION

Excitation of 10 μ M solutions of E4DEAB in hexane with UV light in the wavelength region around 300 nm yields a single emission band which decays exponentially with a lifetime of 1.7 $\pm$ 0.1 ns - behaviour which is entirely analogous to that of M4DMAB in hexane⁷. We find no evidence of dual fluorescence in this solvent. In mixed solvents (hexane/toluene, hexane/acetonitrile or hexane/*N,N*-dimethylformamide) or in the pure solvents toluene, acetonitrile and *N,N*-dimethylformamide, anomalous (dual) fluorescence is observed and the fluorescence kinetics become multi-exponential, displaying both wavelength and solvent dependence. The kinetic behaviour may be exemplified by the data shown in the Table for hexane/acetonitrile.

Acetonitrile Concentration mol dm ⁻³	τ_1 ns	τ_2 ns
0.000	1.70 $\pm$ 0.1	-
0.020	2.00 $\pm$ 0.10	0.65 $\pm$ 0.25
0.038	2.20 $\pm$ 0.20	0.60 $\pm$ 0.20
0.096	2.70 $\pm$ 0.15	0.35 $\pm$ 0.10
0.192	3.10 $\pm$ 0.10	0.29 $\pm$ 0.05
0.287	3.25 $\pm$ 0.05	0.24 $\pm$ 0.05
0.383	3.40 $\pm$ 0.05	0.22 $\pm$ 0.05
0.479	3.30 $\pm$ 0.05	0.21 $\pm$ 0.04
0.574	3.45 $\pm$ 0.10	0.17 $\pm$ 0.05
Pure	3.25 $\pm$ 0.10	0.01 $\pm$ 0.01

Table: Calculated lifetime values for E4DEAB in hexane as a function of added acetonitrile.

On addition of acetonitrile, the single exponential decay of lifetime 1.7 $\pm$ 0.1 ns in pure hexane becomes essentially biexponential with the two derived lifetimes lying either side of the original value of 1.7 ns. As the concentration of acetonitrile is increased the shorter lifetime (τ_2) decreases in value whilst the longer lifetime (τ_1) increases. Additionally, a third component of very small amplitude is sometimes required to achieve acceptable kinetic fits to the experimental data. The amplitude of τ_2 decreases as the emission wavelength increases and becomes negative at wavelengths of 425 nm and above.

Analysis of this data has been undertaken on the basis of a standard exciplex model⁹ i.e.

- (E4DEAB)* $\rightarrow$ E4DEAB + $h\nu$ Fluorescence
- (E4DEAB)* $\rightarrow$ E4DEAB Non-radiative decay

- (E4DEAB)* + Acetonitrile $\rightarrow$ Exciplex Exciplex formation

- Exciplex $\rightarrow$ (E4DEAB)* + Acetonitrile Exciplex break-up

- Exciplex $\rightarrow$ E4DEAB + Acetonitrile Exciplex decay

These processes are assigned rate constants k_f , k_{nr} , k_+ , k_- and $(k'_f + k'_{nr})$ respectively.

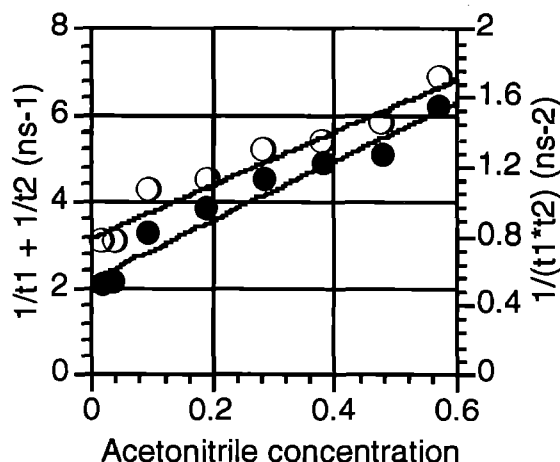


Figure: Lifetime analysis using exciplex model. Filled points refer to left hand scale.

Use of this model together with the appropriate plots of the lifetime values versus acetonitrile concentration (see Figure) yields $k_+ = 7.4 \times 10^9 \text{ dm}^3 \text{ mol}^{-1} \text{ s}^{-1}$, $k_- = 1.2 \times 10^9 \text{ s}^{-1}$ and $(k'_f + k'_{nr}) = 2.2 \times 10^8 \text{ s}^{-1}$. The latter two values indicate an exciplex lifetime of the order of 700 ps. The formation of the exciplex is close to diffusion-controlled but the reverse reaction is less efficient than in exciplexes with benzene and toluene where $k_- > k_+$. It is interesting to note, however, that the sum of the rate constants for the other exciplex decay processes is very similar to those obtained for these processes for the other exciplexes⁴. At present we are subjecting these datasets to global analysis to support the conclusions which we have drawn above. E4DEAB exhibits similar behaviour in the other solvent systems studied but the data also awaits global analysis to (hopefully) yield rate constant data similar to that above.

Our preliminary conclusions at least partially support those of de Lange *et al.*⁴ in that exciplex formation is observed but confirmation of the participation of a folded E4DEAB isomer must await further experiment and analysis.

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PICOSECOND PROCESSES IN CYANINE DYES

R.J. James¹, J.N. Chacón¹, A.J. McLean¹ and R.S. Sinclair¹

A.J. Langley², P.F. Taday² and W. Shaikh²

¹University of Paisley, Paisley, Renfrewshire, PA1 2BE

²Rutherford-Appleton Laboratory, Chilton, Oxfordshire

INTRODUCTION

Cyanine dyes belong to the polymethine class of dyestuffs and have practical applications as laser materials and mode-locking absorbers, as well as sensitisers in photography (1).

Upon continuous and microsecond pulsed irradiation, cyanines have been shown to undergo a reversible trans to cis isomerisation (2,3), which occurs principally via the singlet manifold (3). Fluorescence competes with isomerisation while the quantum yield of intersystem crossing is low. An energy model has been proposed (4) where the ground state all-trans form is irradiated to an excited singlet state which then passes through a twisted intermediate form on the upper excited surface, before returning to the original ground state configuration, or isomerising to produce a photoisomer. The return of the photoisomer to the original conformation is a thermal process which occurs on the millisecond timescale (3).

For two carbocyanine dyes we have studied the photoprocesses occurring upon excitation with 100 fs pulses in an attempt to obtain transient information regarding the precursor to the long-lived photoisomer. The compounds under study were 1,1',3,3,3',3'-hexamethyl-indotricarbocyanine iodide (HITCI) and 3,3'-diethyl-4,4',5,5'-dibenzothiatricarbocyanine iodide (DDTTCI). Both compounds possess three carbon-carbon double bonds in their methine chains.

RESULTS AND DISCUSSION

During the last year we have made use of the Tsunami femtosecond titanium-sapphire laser pumped by a Spectra Physics 171 argon-ion laser. The pump wavelength used was 740 nm (100 fs, 5 μ J) while the probe beam was focused into a 3 mm thick fused silica disc in order to generate a continuum between 400-700 nm as the analysing light.

The main ground state absorption bands of both HITCI and DDTTCI lie on the long wavelength side of the visible spectrum with a maximum wavelength of absorption (λ_{max}) at 740 nm and 800 nm respectively. Both compounds showed significant absorption at 740 nm allowing direct excitation at the fundamental of the titanium-sapphire laser.

Time resolved transient spectra, showing the decay of HITCI in acetonitrile over the picosecond timescale, are illustrated in Figure 1. The spectra were found to consist of two absorption bands at 490 nm and 550 nm, an isobestic at 580 nm, with evidence of ground state depletion from 580-700 nm. The position of the absorption bands was unchanged when using methanol and water as solvents. Transient spectra of DDTTCI in methanol showed only a single absorption band centred at 540 nm.

The decay of the transient species, measured at the maximum wavelength of absorption, followed first order kinetics. For all the data there was some residual absorption which did not decay on picosecond timescales and is assigned as the photoisomer. First order lifetimes are displayed in Table 1.

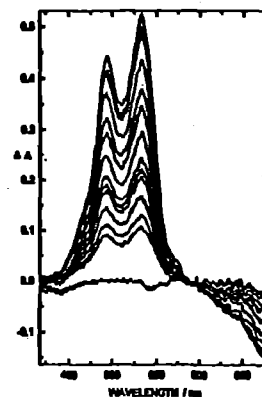


Figure 1. The decay of the transient absorption spectrum of HITCI in acetonitrile

Table 1. Transient First Order Decay Lifetimes of the Compounds Under Study.

COMPOUND	SOLVENT	LIFETIME / ps
HITCI	Methanol	579
HITCI	Acetonitrile	530
HITCI	Water	368
DDTTCI	Methanol	690

The transient absorption spectra of both HITCI and DDTTCI are proposed to be due to a twisted intermediate in the first excited singlet state, which is the precursor to the photoisomer. The transient absorption bands are blue-shifted relative to the ground state spectra due to a loss of extended conjugation, and therefore may consist of absorption which is isolated on each part of the twisted molecule. In the case of DDTTCI, the single band transient spectrum suggests that the two parts of the twisted molecule show near equivalent absorption properties. The two bands observed in the transient spectrum of HITCI suggest non-equivalent absorption from the two parts of the twisted molecule and therefore an isomerisation around a different double bond in the methine chain to that observed with DDTTCI. From this data it has been possible to propose possible structures for the photoisomer of each compound and this is in general agreement with recent work on carbocyanine dyes with two double bonds in the methine chain (5),(6).

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STUDY OF DEPOLARIZATION MECHANISMS BY TIME AND ANGLE RESOLVED PHOTOELECTRON SPECTROSCOPY

Katharine L Reid,¹ Simon P Duxon¹ and Michael Towrie²

¹Department of Chemistry, University of Nottingham, Nottingham NG7 2RD

²Laser Support Facility, Rutherford Appleton Laboratory, Chilton, Didcot OX11 0QX

INTRODUCTION

Time-resolved laser photoelectron spectroscopy of an excited molecular electronic state presents a new method of determining information on intramolecular dynamics.^{1,2} So far, researchers have concentrated on attaining high energy resolution in the photoelectron spectra, and in particular the "ZEKE" threshold method³ has been employed.^{1,2} In these studies, photoelectron intensity is measured as a function of the time delay Δt between the laser pulse that promotes the molecule to the excited electronic state, and the laser pulse that performs the ionization step. A neglected aspect of these studies is the evolution of the alignment of the levels populated in the excited electronic state as a function of Δt . This alignment can only be probed by observing the *angle-resolved* photoelectron intensity, and "ZEKE" type experiments preclude the determination of such angular information.

Here, we report the first measurements of time- and angle-resolved photoelectron flux from a molecule. We present the time evolution of photoelectron flux at a fixed angle of emission for the excitation-ionization process $\text{NO}(X^2\Pi, v_g=0, J_g=2.5) \rightarrow \text{NO}(A^2\Sigma^+, v=0, J=1.5, N=1) \xrightarrow{\Delta t} \text{NO}^+(X^1\Sigma^+, v^+=0)$ in which the ionizing beam is circularly polarized. In this process the alignment of J in the intermediate state evolves as a result of coupling to the spin of the nitrogen nucleus, and this depolarization manifests itself as oscillations in the time- and angle-resolved photoelectron signal.^{4,5}

EXPERIMENT

The overall experimental setup is very similar to that employed by Leahy et al. in their measurements of photoelectron angular distributions.⁶ The photoelectron spectrometer consists of a diffusion pumped stainless steel vacuum chamber inside which nitric oxide is released through a hypodermic needle to give an operating pressure of $\approx 10^{-6}$ torr. The excitation and ionization laser beams counterpropagate through quartz windows at the ends of two stainless steel arms, and photoelectrons are generated at the intersection of the two laser beams and the stream of nitric oxide. The photoelectrons travel 60cm vertically through a mu metal shielded, field-free region, to a 25mm diameter microchannel plate detector (Photek). The output from the microchannel plate detector passes through a fast preamplifier (EG&G Ortec) and discriminator to a computer-controlled fast oscilloscope (HP 54510A), triggered by a photodiode that monitors pulses from the ionization beam. In this experiment, every time a photoelectron arrived within ± 25 ns of the flight time of interest, a hit was registered and passed to the computer. In other words, no attempt was made to resolve the internal energy states of the NO^+ ion.

Laser systems in the Laser Support Facility at the Rutherford Appleton Laboratory were used. The excitation beam was generated by an excimer (Lumonics HE460) pumped dye laser (Lambda Physik FL3002) operating with Coumarin 47, and subsequently doubled and attenuated to produce around 1 μJ light at $\approx 226\text{nm}$. This light was tuned to be resonant with the $X^2\Pi(v=0) \rightarrow A^2\Sigma^+(v=0)$ transition $P_{12}(2.5)$, and was passed through a rotatable zero-order half waveplate (Halbo Optics) before entering the chamber. The position of the half waveplate controlled the detected angle of emission, ϕ , of the photoelectrons. The ionization beam was generated by a second excimer (Lumonics HE460) pumped dye laser (Lambda Physik FL3002) operating with Coumarin 153, and subsequently doubled and attenuated to produce around 100 μJ light at 266nm. This beam was passed through a zero-order quarter waveplate (Lambda Photometrics) with its optic axes at 45° to the electric vector of the linearly polarized incident beam in order to generate circularly polarized light, before entering the chamber. Both lasers were unfocused, giving beam diameters of around 2mm in the chamber. The degree of polarization of the laser beams following passage through the waveplates was not

measured, and will cause a small uncertainty in the photoelectron intensity. However, this uncertainty is expected to be well within that produced by the jitter in the time delay (see later). The laser powers were kept low in order to avoid the effects of saturation; this resulted in a signal level of around 1 photoelectron per 10 laser shots. Photoelectrons resulting from excitation by the 226nm beam followed by ionization with the 266nm beam have flight times of around 1.1 μs , and are well separated in time from photoelectrons resulting from one-colour ionization. The FWHM pulsewidth of both doubled laser beams was around 5ns. Both lasers were triggered by an SRS digital delay/pulse generator which was controlled by the computer. During the course of an experimental run, the half waveplate was set to a given angle, and the time delay between the laser pulses was scanned from (nominally) $\Delta t = -30\text{ns}$ to $+90\text{ns}$ in steps of 5ns, accumulating 10 laser pulses at each delay. This method was employed in order to compensate for power fluctuations, and temporal drifts. Each experimental run encompassed 65,000 shots, i.e., 2600 shots per delay, and several runs per angle were performed.

RESULTS AND DISCUSSION

The excitation beam aligns $J=1.5$ in the $A^2\Sigma^+$ state at $\Delta t=0$. For $\Delta t>0$, this alignment depolarizes and repolarizes according to the equation:⁷

$$T_Q^2(\Delta t) = \sum_{FF'} \frac{(2F'+1)(2F+1)}{(2I+1)} \begin{Bmatrix} F' & F & 2 \\ J & J & I \end{Bmatrix}^2 \cos(\omega_{FF'}\Delta t) T_Q^2(0), \quad (1)$$

where F is the total angular momentum quantum number, I is the nuclear spin quantum number ($I=1$ for the nitrogen nucleus), $\{::\}$ is a Wigner 6- j symbol,⁸ $\omega_{FF'}$ is the hyperfine splitting between the levels F and F' , and $T_Q^2(\Delta t)$ represents the alignment at time Δt , with projection Q . From Eq. (1) it is clear that in order to observe the maximum effect it is necessary both to choose a low value of J , and a transition that produces a high degree of alignment. In addition, it is important to find a transition that is well resolved. These three criteria led to the choice of the transition $P_{12}(2.5)$ for the present study.

The angle-resolved photoelectron flux can be written as

$$I(\theta = \frac{\pi}{2}, \phi; \Delta t) = \exp\left(-\frac{\Delta t}{\tau}\right) \sum_{LQ} \beta_{LQ}(\Delta t) Y_{LQ}(\theta, \phi), \quad (2)$$

where the $\beta_{LQ}(\Delta t)$ coefficients contain both the dynamical parameters controlling the photoionization step and the alignment; they are given in Ref. 4, Eq. (5). τ represents the $1/e$ lifetime of the intermediate state, and the $Y_{LQ}(\theta, \phi)$ are spherical harmonics. The use of circularly polarized ionizing radiation ensures that $I(\theta, \phi; \Delta t)$ is sensitive to intermediate state alignment even when the ion states are not resolved,⁴ a twist that is peculiar to $\text{NO } A^2\Sigma^+$.^{6,9} Figure 1 shows photoelectron counts with statistical error bars as a function of time delay, for a fixed angle $\phi=30^\circ$. There is also some error in the time measurement due to the finite pulse widths of the lasers, and some "jitter" on these pulses (order of 5ns). It is possible to either fit the data for $\Delta t>0$ to the function $a(1+b\cos\omega\Delta t)\exp(-\frac{\Delta t}{\tau})$ or to compare it with predictions. Figure 1 shows the result of the comparison with predictions (see later). Data for other angles can be found in Ref. 10. The values of the parameters a, b and ω for each angle are shown in Table I. In principle, three oscillation frequencies should appear; $\omega_{0.5,1.5}$, $\omega_{1.5,2.5}$ and $\omega_{0.5,2.5}$. However, we are able to fit the data to only one frequency because the coefficient for $\omega_{0.5,2.5}$ in Eq. (1) is very small, and $\omega_{0.5,1.5}$ produces an oscillation

with a long period on the timescale of the experiment (see calculated frequencies later). Thus we expect $\omega \approx \omega_{1.5,2.5}$.

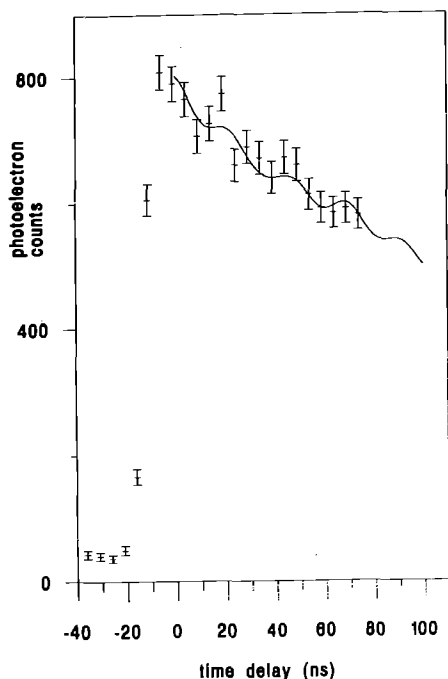


Figure 1 Time evolution of photoelectron flux

ϕ	$\omega(\text{MHz})$	a	b	$\tau(\text{ns})$
30°	42	770	2.6×10^{-2}	240
300°	42	1100	-1.8×10^{-2}	240
75°	-	745	0.0	240

Table I Values of the parameters used in the fits of the data to the function $a(1 + b \cos \omega \Delta t) \exp(-\frac{\Delta t}{\tau})$

The solid lines in Fig. 1 represent the (normalized) calculated photoelectron intensity from Eq. (2), using the dynamical parameters reported by Leahy et al. for a slightly lower photoelectron energy,⁶ hyperfine energies $\omega_{0.5,1.5} = 13.6 \text{ MHz}$ and $\omega_{1.5,2.5} = 42.9 \text{ MHz}$; initial alignments $T_0^2(0) = 0.097$ and $T_{12}^2(0) = -0.122$; and $\tau = 240 \text{ ns}$. The hyperfine energies were calculated by Western¹¹ using the nuclear and spin-rotation coupling constants reported by Bushaw and coworkers for the $v=1$, $N=3$ level in $\text{NO}(A^2\Sigma^+)$.¹² The error bars on the frequencies required to fit our data do not enable us to deduce any difference between the $v=0$ constants and the $v=1$ constants. In previous work it has been found that the nuclear quadrupole coupling constant $eQq = -7.2 \pm 0.5 \text{ MHz}$ in $v=1$,¹² and $-6.77 \pm 0.17 \text{ MHz}$ in $v=3$.¹³ Either of these values would serve to fit the data presented here, and in fact the value of eQq can only be determined to within about $\pm 5 \text{ MHz}$, due to the jitter in the laser pulses. The value of τ was chosen to best fit the data.

It is clear that the observed (and predicted) time evolutions depend on the angle of photoelectron ejection. We note that if the angle-integrated photoelectron signal were monitored no oscillations would be observed. The most pronounced oscillation is observed for $\phi=30^\circ$ as evidenced by the value of b in Table I. Photoelectrons ejected at this angle are therefore most sensitive to the intermediate state alignment. The oscillations at $\phi=300^\circ$ are smaller and oppositely signed to those at $\phi=30^\circ$, and no oscillations are observed for $\phi=75^\circ$. Thus, we can think of $\phi=75^\circ$ as being a "magic angle" for this experiment, where the value this magic angle takes depends on the β_{LQ} parameters in Eq. (2).

As indicated above, the data presented in this paper is not of sufficient quality to enable the hyperfine coupling constants of $\text{NO}(A^2\Sigma^+)$ to be deduced accurately. In order to use this method as a means of performing high resolution spectroscopy it would be necessary to employ lasers with shorter and more stable pulses. It should be noted that quantum beat spectroscopy is a good method of obtaining information on hyperfine coupling from states of molecules that fluoresce.¹⁴ However, the point of this experiment was to illustrate a new method of studying intramolecular depolarization mechanisms, which is applicable to states that do not fluoresce, and should complement time-resolved ZEKE experiments.¹⁵

In future experiments, where systems are chosen for which the cause of alignment evolution is unknown, it will be desirable to extract the values of $T_Q^2(\Delta t)$ from the data in order to gain information on the depolarization mechanism. McKoy and coworkers have shown that it is possible to do this without knowledge of the photoionization dynamics by measuring the circular dichroism in the photoelectron angular distributions.¹⁶

CONCLUSIONS

We have reported the first observation of time- and angle- resolved photoelectron flux for a molecule. The method has been applied to the study of hyperfine depolarization in $\text{NO}(A^2\Sigma^+, v=0, J=1.5, N=1)$, and the data agrees well with predictions that use coupling constants previously reported for $v=1$.¹² We plan to apply the method to other molecular depolarization mechanisms, and in particular to use it as a means of assessing the degree of rotational coupling involved in intramolecular vibrational redistribution.¹⁷

ACKNOWLEDGEMENTS

We are indebted to Ivan Powis (Nottingham) and Sue Tavender (RAL) for technical advice, and to Colin Western (Bristol) for advice on the hyperfine energies. We thank the EPSRC for the award of an Advanced Fellowship (to KLR), and for the allocation of laser time at the Rutherford Appleton Laboratory. This work was supported by equipment grants from the EPSRC (GR/H88299) and the Royal Society.

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The Photolysis of 3-Cyclopentenone Adsorbed on an MgF₂ Surface

C.J.S.M. Simpson¹, P.T. Griffiths¹, H. Wallaart¹, R.L. Lovegrove¹,
A. Weedall² and M. Towrie²

¹ Physical and Theoretical Chemistry Laboratory, Oxford University

² Rutherford Appleton Laboratory

Introduction

The object of these experiments is to determine the translational, rotational and vibrational energy distributions of CO formed from carbonyls, physisorbed on a surface, and to compare the results with those obtained in gas-phase work.

For this purpose, we employ a dielectric surface of single crystal MgF₂ which is highly transparent at the wavelength of the photolysis laser. It also has phonon modes far removed from the vibrational frequency of CO. Since a large quantum of energy has to be removed for the vibrational deactivation of CO, there is a good chance that the molecules will not be deactivated on the timescale of dissociation and desorption. Such indeed has been found for the dissociation of the alkyl nitrites and NO dimers [1,2].

We have now turned our attention to the carbonyls which dissociate more slowly than the nitrites. We have considered the molecules HNCO, acetone and finally 3-cyclopentenone. All of these molecules dissociate to give vibrationally excited CO, with 10 - 20 % population of ($v=1$).

All of these molecules produce rotationally excited CO in the gas phase photolysis. Since the rotational quanta are so much smaller than those of vibration, there is a far higher chance that the desorbed molecules will be rotationally relaxed even though they are vibrationally excited. Detection of the desorbed CO was by (2+1) resonance enhanced multi-photon ionisation spectroscopy *via* the B state.

Experimental

The Ultra-High Vacuum apparatus is described elsewhere [1]. The chamber is diffusion pumped, with additional cryopumping provided by liquid nitrogen- and helium-cooled shrouds elsewhere in the chamber. The single crystal MgF₂ surface is held on a transmissive probe which is liquid helium cooled. In conjunction with a heater circuit, the temperature of the crystal may be held between 20 and 300 K to an accuracy of 1 K.

A Lambda Physik 105i excimer is used to initiate desorption. The central, most uniform portion of the beam is selected using an elliptical mask and is then focused onto the crystal face to give a circular spot, diameter 5 mm.

The excimer-pumped dye laser system provided at the LSF was used to ionise the CO produced. The second harmonic of the laser was focused using a 30 cm lens into a region 5 cm above the surface. The ions formed were deflected onto the Gallileo micro-channel plate by a grid and repeller plate system based on the Wiley-McClaren design.

The detection of CO using REMPI is very sensitive. We could obtain spectra with good signal to noise down to partial pressures of CO of 2×10^{-10} Torr, Figure 1. The spectra consist of an unresolved Q-band. The spectra are analysed by fitting the signal to a rotational temperature [3].

Results and Discussion

Unfortunately, HNCO was found to polymerize within the metal preparation line used to admit gases to the chamber. Our investigations into acetone were problematic: very high fluences were required to give an adequate signal of CO, at which thermal pathways were dominant. We turned at last to 3-cyclopentenone which has been studied in the gas phase under collision-free conditions by Jimenez *et al.* [4].

Most importantly, the dissociation of the adsorbed 3-cyclopentenone gave CO in both ($v=0$) and ($v=1$), Figure 2. This is surprising in the light of the longer dissociation time of the carbonyl compound, and the fact that acetone dissociated to give CO entirely in ($v=0$).

A Boltzmann distribution of energies is observed with characteristic temperatures in the range 200 to 700 K. The rotational temperature has been found to depend on fluence, surface temperature and kinetic energy of the desorbed CO.

The dissociation resembles the gas phase photolysis in many respects. The rotational energy increases with

kinetic energy, and the ratio of ($v=1$) to ($v=0$) is similar, around 20 %. However, much lower rotational and translational energies are observed.

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Figure 1: B State REMPI Spectrum CO_g

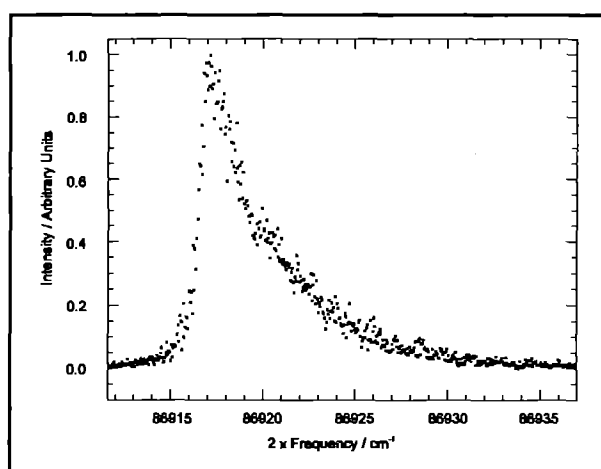
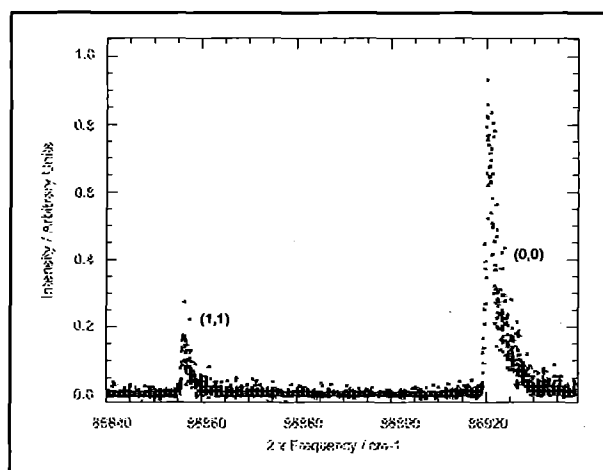


Figure 2: B State REMPI Spectrum of CO formed from photolysis of 3-cyclopentenone.



ELECTRONIC SPECTROSCOPY OF INVOLATILE, HEAVIER ELEMENT AND TRANSITION METAL COMPOUNDS FORMED IN A FREE JET EXPANSION

J.M. Brown, C.J. Whitham, N.M. Lakin, D.M. Goodridge and M.P. Barnes.

The Physical Chemistry Laboratory, Oxford University.

i. INTRODUCTION

High temperatures are required to produce sufficient vapour pressure of involatile solids. Under these conditions however, gas phase spectroscopy yields little information because the spectra are broad and featureless.

In our experiment, molecules are formed in a heated alumina nozzle and are seeded into an inert gas stream (usually argon or helium) at high pressure. The nozzle will operate up to 1000°C. The gas mixture undergoes a free jet expansion through a small orifice (250µm in diameter) into a rapidly pumped vacuum chamber. As a result, the vibrational and rotational degrees of freedom are considerably cooled, to about 100K and 10K respectively. The spectrum recorded under these conditions is greatly simplified and its analysis is normally much more straight forward.

To record the spectra, the focussed beam from a narrow band tunable dye laser is used to excite molecules in the jet and the resultant fluorescence is recorded (laser excitation spectroscopy). We have made use of the JK Hyper Nd:YAG and Spectron PDL2 dye laser combination from the LSF loan pool. Near-UV wavelengths were obtained by using the wavelength extension (WEX) accessory. The measurements on InOH, In₂O and CuCl₂ and their analysis is described briefly below.

ii. INDIUM(I) HYDROXIDE, InOH

The first recording of the $\tilde{B} \leftarrow \tilde{X}$ system of InOH was made at the RAL [1],[2]. The analogous systems of InOD and In¹⁸OH have now also been recorded [3],[4]. The spectrum consists of transitions to three excited states, denoted $\tilde{\alpha}^3A'$, $\tilde{\beta}^3A''$ and $\tilde{\gamma}^3A'$. An extensive band system, dominated by a bending progression with separations of about 800cm⁻¹, was observed for all three components. Calculations performed by C.J. Whitham suggest that these states arise from an extremely strong Renner-Teller splitting of an excited state which would be a $^3\Pi$ in the linear limit. These assignments are supported by the relative intensities of the component systems ($\tilde{\beta}$ and $\tilde{\gamma}$ bands are similar in intensity since both are derived from the $^3\Pi_1$ spin-orbit state) and lifetime measurements (which are in agreement with a triplet-singlet transition).

Several of the key bands in these systems have been recorded at rotational resolution. The individual P and R lines in each branch were observed but for the strongest bands (corresponding to the transition moment perpendicular to the a-axis) the very intense Q branch remained unresolved. A typical band in the $\tilde{\beta}$ system of InOD is shown in figure 1. The rotational constants of InOH and its isotopomers were determined from a least squares analysis of these transition wavenumbers. Figure 2 shows the geometry of InOH in its ground and the $\tilde{\beta}$ excited state deduced from these constants. The large reduction in bondlength on excitation is in agreement with the strong bending progression observed in the excitation spectrum. The In-O and O-H bondlengths in the excited states are close to those expected by comparison with similar molecules. The O-H bondlength in the ground state is however unusually short.

This results from the ground state of InOH being very floppy or *quasilinear*; the O-H bondlength represents the average over the arc of a large amplitude bending motion.

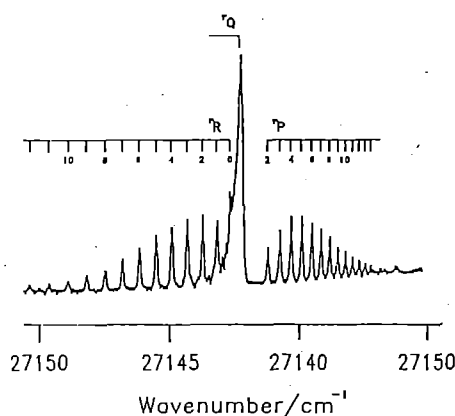


Figure 1: LIF excitation spectra of the $\tilde{\beta} 1_0^2 0_3$ $K_a = 1 \leftarrow 0$ of InOD

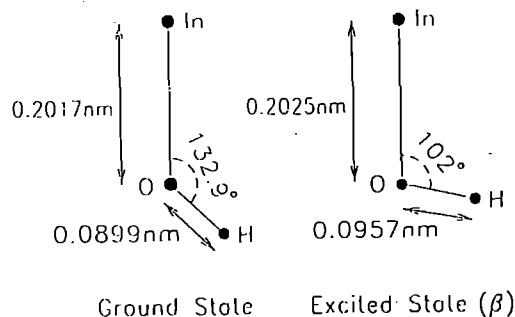


Figure 2: the geometry of InOH in its ground and the $\tilde{\beta}$ excited state

To investigate the quasilinearity of the ground state, the fluorescence from several of the bands was dispersed using a monochromator. This provides information about the vibrational intervals in the ground state. Figure 3 shows the bands recorded by dispersing the Q branch of the band shown in figure 1. The large number of bands has been explained by collision-induced transitions in the free jet expansion. The lowest bending levels resemble those in a bent molecule; however as the bending 'ladder' is ascended the separations become more similar to those in the linear limiting case. The vibrational intervals measured were used to determine an approximate bending potential surface which is shown in figure 4. This diagram shows the relative positions of the levels on this surface and demonstrates the excellent agreement between the experimentally determined bending potential and that obtained by *ab initio* calculations.

LASER-INDUCED FLUORESCENCE SPECTROSCOPY OF JET-COOLED LASER-DESORBED MOLECULES

Fiona L. Plows and Anita C. Jones

Department of Chemistry
University of Edinburgh

INTRODUCTION

The use of supersonic molecular beams in spectroscopy has revolutionised the study of the electronic spectra of large polyatomic molecules. Until recently, supersonic jet spectroscopy could only be applied to *thermally stable* polyatomic species, because of the need to heat the sample to elevated temperatures (typically 100–300°C) to produce sufficient vapour pressure to seed the expansion. However, with the development of pulsed infrared laser desorption [1], thermally labile molecules can now be vaporised as intact neutrals. Such molecules are now accessible, therefore, to investigation under jet-cooled conditions. Although laser desorption is becoming a well-established technique in mass spectrometry, there has been little work to date on the spectroscopy of laser-desorbed molecules, because of the difficulty of achieving stable desorption conditions. Of the few spectroscopic studies which have been reported, almost all have been extensions of mass spectrometric measurements, using mass-resolved resonant 2-photon ionisation (R2PI)

We report here on the use of laser-induced fluorescence (LIF) to investigate the jet spectroscopy of laser-desorbed molecules. In a previous study of the LIF of laser-desorbed dipeptides, Levy et al [2] reported considerable interference from scattered laser light which was attributed to the presence of particulate matter produced in the desorption process. They found it necessary to detect the fluorescence through a monochromator in order to discriminate against the scattered light. In our experiments, using the desorption/entrainment method described below, we have encountered no problems with scattered light.

EXPERIMENTAL

The successful use of laser desorption as a source of molecules for jet spectroscopy requires the production of a stable supply of sample vapour pulses at a reasonably high repetition rate (≥ 10 Hz) for extended periods of time (hours rather than minutes). Furthermore, in order to produce sufficient jet-cooling to give well resolved vibronic spectra, the desorbed molecules must be efficiently entrained in the high pressure throat of a pulsed supersonic expansion. These conditions can be achieved using a desorption nozzle assembly of the form shown schematically in Figure 1.

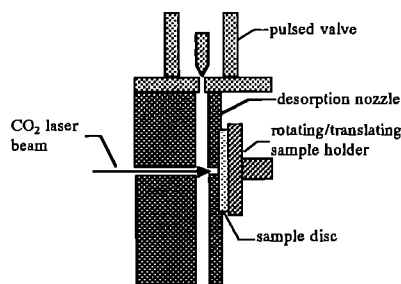


Figure 1. Schematic cross sectional diagram of the desorption nozzle assembly.

The desorption nozzle assembly consists of a pulsed supersonic valve (General Valve Corporation Series 9) fitted with an extension block into which the desorbed molecules are released

and then entrained into the supersonic expansion. The nozzle assembly is mounted on an xyz translator to enable accurate positioning relative to the desorption laser beam and excitation laser beam. The desorption sample holder consists of a stepper motor-driven rotating disc (30mm diameter) mounted on a motor-driven translation stage, enabling the sample to be rotated and translated relative to the desorption laser beam. The sample is prepared in the form of 25 mm diameter pressed solid disc (using a hydraulic press) and mounted on the sample holder with double sided tape. The sample is moved continuously relative to the desorption laser beam during the experiment; desorption can be carried out continuously from the same sample for several hours.

Desorption was achieved using a pulsed TEA CO₂ laser (Alltec 854MS) which outputs up to 300mJ of 10.6 μ m radiation in a 100ns pulse. The desorbed molecules were entrained in a supersonic beam produced by expansion of 7bar of helium through a 0.5mm diameter nozzle. Fluorescence was excited using a Nd:YAG-pumped dye laser system (JK HY750 Nd:YAG/Quanta Ray PDL2 dye laser with WEX UV accessory). The excitation laser beam crossed the expansion 5–10mm downstream of the desorption nozzle. Undispersed fluorescence was collected by a 50mm focal length f/1 lens and detected by a Hamamatsu R928 photomultiplier tube. The signal was processed by a SRS SR250 gated integrator and stored on a PC. The experiment was carried out at a repetition rate of 10Hz and the signal was averaged over 30 laser shots. The relative timing of the pulsed valve, desorption laser and excitation laser was controlled by a SRS DG535 digital delay generator.

LIF OF JET-COOLED LASER-DESORBED PARA-AMINO BENZOIC ACID AND BENZOIC ACID

Para-amino benzoic acid (PABA) is a highly involatile compound, with a vapour pressure of only 10⁻⁶ mbar at room temperature, and is thus difficult to vaporise using conventional techniques. It can, however, be easily vaporised using laser desorption. Meijer et al [3] have reported the mass-selected R2PI spectrum of jet-cooled laser-desorbed PABA and have observed both monomer and dimer species. We have investigated the laser-induced fluorescence.

Figure 2 shows the low energy region of the LIF excitation spectrum of jet-cooled laser-desorbed PABA. The spectrum consists of the superposition of two spectra, due to the presence of both PABA monomer and hydrogen-bonded dimer species in the expansion. The dimer 0–0 transition at 34214cm⁻¹ and associated vibronic transitions at 231cm⁻¹ and 251cm⁻¹ are indicated by asterisks. Low frequency vibronic transitions of the dimer, due to intermolecular hydrogen bond vibrations, are shown in the inset. The dimer 0–0 is blue-shifted some 40cm⁻¹ from that of the monomer which lies at 34173cm⁻¹. The ability to generate a high relative concentration of dimers in the expansion demonstrates that efficient cooling of the laser-desorbed molecules can be achieved.

On the basis of the critical dependence of the 2-colour photoion signal on the temporal overlap of the two laser pulses, Meijer et al [3] estimated the lifetime S₁ state of PABA to be only a few hundreds of picoseconds, due to the occurrence of efficient

intersystem crossing. However, the high intensity of the fluorescence signal in our experiments indicates that both the monomer and the dimer have rather large fluorescence quantum yields, which would not be consistent with such a short lifetime.

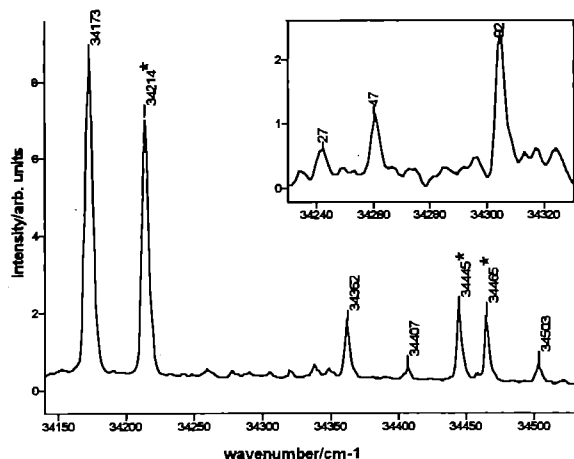


Figure 2. LIF excitation spectrum of jet-cooled laser-desorbed p-amino benzoic acid. * indicates bands due to the dimer. Low frequency transitions of the dimer are shown in the inset.

The structure of the PABA dimer is thought to be analogous to that of the well-known benzoic acid dimer. Condensed phase studies [4,5] have shown that at low temperatures, in accordance with thermodynamic predictions [6], benzoic acid exists as a dimer in which the two monomer units are doubly hydrogen-bonded through the -COOH groups.

Benzoic acid is much more volatile than PABA and has been studied using conventional LIF jet spectroscopy [7]. Under normal conditions of jet-cooling, benzoic acid exists almost exclusively as the dimer, and the monomer was not observed in the latter study. However, Meijer et al [8] have shown that laser desorption jet cooling can produce a molecular beam with a relatively large benzoic acid monomer concentration and they have recorded the mass-selected two-colour R2PI spectra of both dimer and monomer.

Figure 3 shows the LIF excitation spectrum of jet-cooled laser-desorbed benzoic acid. The spectrum corresponds to that of the dimer [8]. We observed no spectrum due to the monomer,

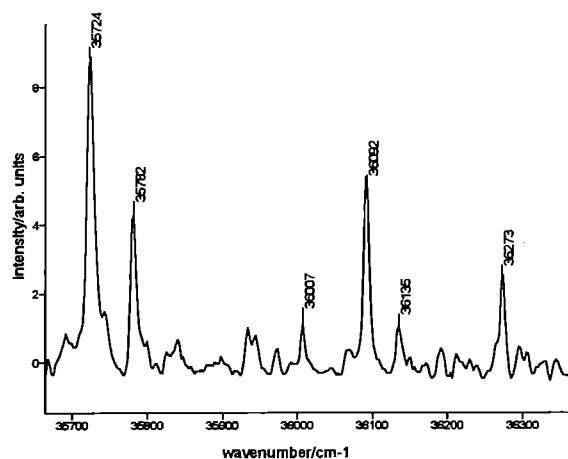


Figure 3 LIF excitation spectrum of jet-cooled laser-desorbed benzoic acid.

although there should have been a considerable monomer concentration present in the beam under our experimental conditions. This indicates that the fluorescence quantum yield of the benzoic acid monomer is much smaller than that of the dimer. The benzoic acid monomer has been predicted to have a very low fluorescence quantum yield [9] due to the near degeneracy of the S_1 state with a $^3(n,\pi^*)$ state, which results in very efficient intersystem crossing. The much higher quantum yield of the dimer can be accounted for by a decrease in the rate of intersystem crossing as a result of an increased energy separation between these two states. In contrast, in the case of PABA, we have found that both the monomer and the dimer show intense fluorescence. This suggests that in neither the PABA dimer nor the PABA monomer is there a particularly efficient route for intersystem crossing.

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ELEMENTARY REACTIONS BETWEEN NEUTRAL SPECIES AT ULTRA-LOW TEMPERATURES

Bertrand R Rowe,¹ Ian R Sims² and Ian WM Smith²

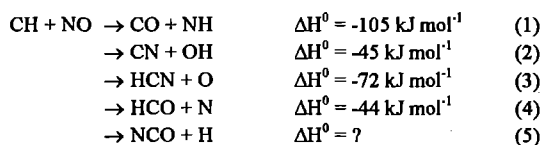
¹Université de Rennes 1
²University of Birmingham

INTRODUCTION

The highly reactive methyldene radical (CH) is found in an unusually wide variety of environments,¹ with temperatures ranging from an ultra-low 10–50 K in dense interstellar clouds to 1000 K and above in flames. The kinetics of its reactions are of interest in combustion systems,² where its reaction with N₂ is crucial in the formation of 'prompt' NO.³ Reactions of CH are also important in the chemistry of planetary atmospheres, especially in the nitrogen- and hydrocarbon-rich atmosphere of Titan,⁴ as well as in the chemistry of interstellar clouds.⁵ Knowledge of reaction rates over as wide a temperature range as possible is therefore very desirable.

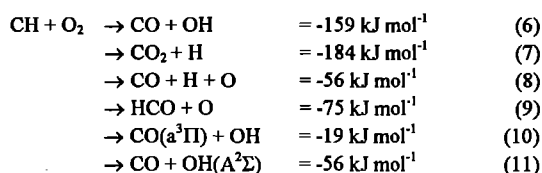
As a result of the interest in CH radical kinetics in such a wide range of environments, and in order to better understand its high reactivity, we have launched a programme of experiments to determine the rates of a number of elementary reactions of CH covering a temperature range from about 13 K to 600 K. In the experiments described here, the CRESU (Cinétique de Réaction en Ecoulement Supersonique Uniforme) technique has been used to achieve temperatures below 300 K, in conjunction with the PLP-LIF (pulsed laser photolysis - laser induced fluorescence) technique for the production and detection of radical species in the gas phase. We have already applied these techniques to a range of CN radical⁶ and OH radical⁷ reactions.

In this article we report results for reactions of the CH radical with O₂ and NO. The reaction of CH with NO has a number of energetically accessible reaction channels:⁸



The NH product of channel (1) has been observed by UV chemiluminescence,^{9,10} and on the evidence of the vibrational and rotational product state distributions,¹⁰ it is thought that this channel proceeds via a direct four-centre mechanism. However, there is considerable disagreement in the literature regarding the products of this reaction, and measurements of product branching ratios are urgently needed.

The reaction of CH with O₂ has again a number of energetically accessible channels:⁸



Calculations suggest that the CH + O₂ reaction proceeds to the formation of H + CO₂ and OH + CO via the HOCO intermediate:¹⁰ again, we await the results of product branching measurements.

EXPERIMENTAL

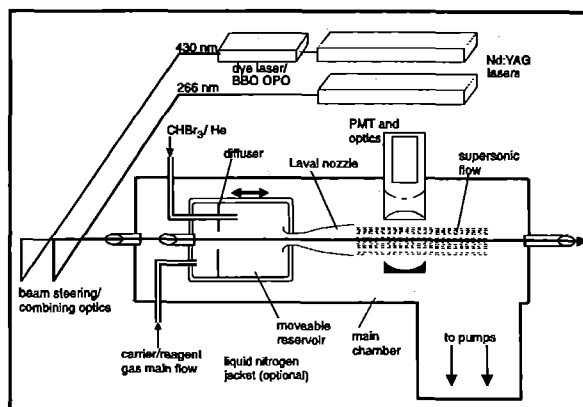


Figure 1 Schematic diagram of the combined PLP-LIF/CRESU technique for the study of CH reactions at ultra-low temperatures

The CRESU technique and its adaptation to the study of neutral-neutral reactions (in particular those of the CN radical) by means of the PLP-LIF technique has been described in detail elsewhere.^{6(b)} Here, we will give a brief summary of the method as applied to measurements at ultra-low temperatures on the reaction kinetics of CH radicals. A schematic diagram of the experimental apparatus is displayed in Figure 1.

The CRESU technique provides, via the isentropic expansion of gas through a Laval nozzle, a 'collimated' flow of ultra-cold gas which is uniform in temperature, density and velocity. The expansion and subsequent cooling are rapid enough that heavily supersaturated conditions may prevail, avoiding the major problem of condensation associated with the use of cryogenically-cooled cells. However, in contrast to free jet expansions and molecular beams where the concept of temperature is not really valid, the relatively high gas density (10^{16} – 10^{17} molecule cm⁻³) in the uniform supersonic flow ensures that frequent collisions take place during the expansion and subsequent flow, maintaining thermal equilibrium at all times. Several nozzles were employed in the present work, each providing a particular temperature and density for the selected carrier gas. Flow conditions have been verified previously by impact-pressure measurements, as well as by independent determination of the temperature via spectroscopic measurements on the (0,0) band of the ($B^2\Sigma^+ - X^2\Sigma^+$) system of CN.^{6(b)}

CH radicals were generated by the sequential multiphoton photolysis of CHBr₃ at 266 nm, and detected by off-resonance LIF, exciting the (0,0) band of the CH(A²Δ–X²Π) system at ~430 nm, and detecting off-resonance fluorescence from the (0,1) band at ~490 nm. Kinetic data were gathered using the strongest available rotational line, usually the R₂₂(1) line at 430.03 nm.

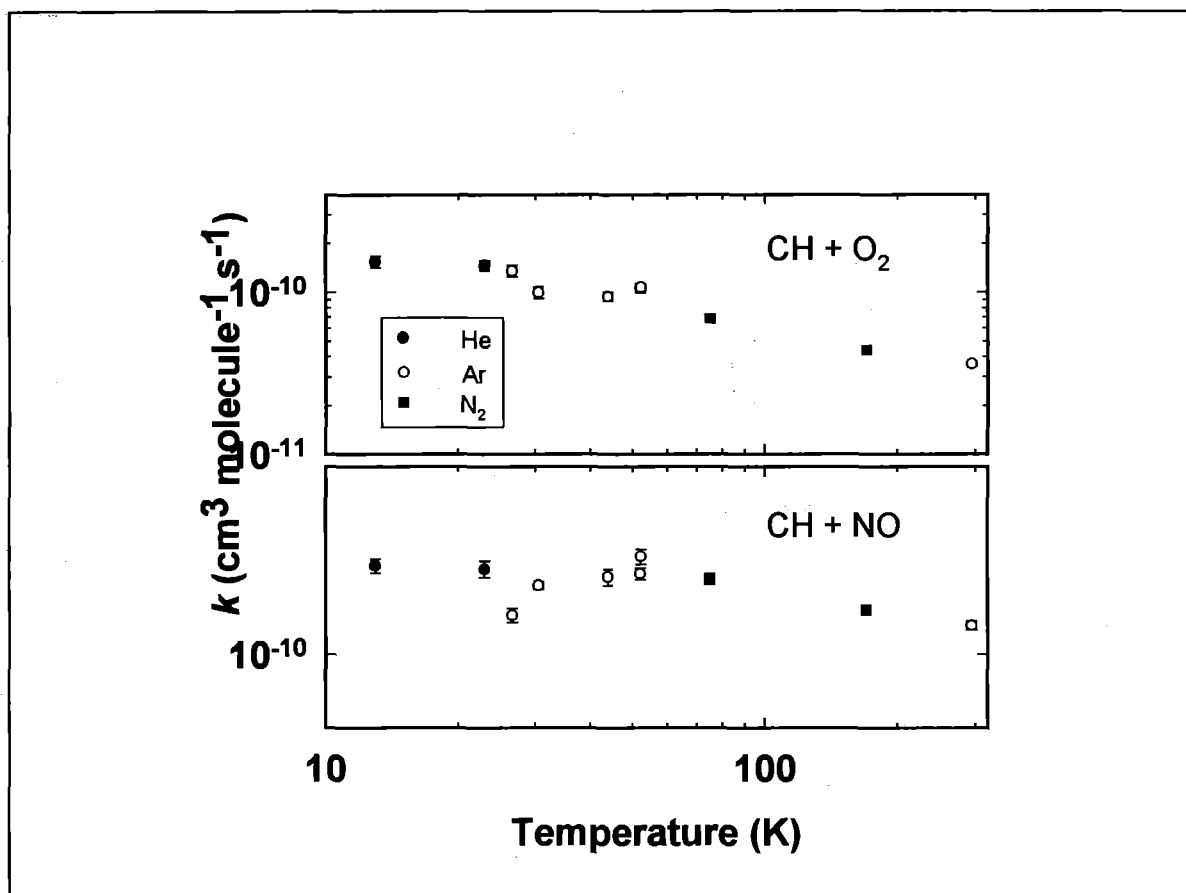


Figure 2 Rate constants obtained in the CRESU apparatus for the reactions $\text{CH} + \text{O}_2$ and $\text{CH} + \text{NO}$, in different carrier gases as indicated

The results obtained in the CRESU apparatus for the kinetics of the reactions of CH with O_2 and NO are displayed in Figure 2. As can be seen, both reactions are fast at room temperature, and are in good agreement with the literature values.⁸⁻¹⁰ Only a mild negative temperature dependence is observed, in contrast for example to the reaction of the CN radical with O_2 , which shows a strong $T^{-0.6}$ dependence of its rate constant on temperature.⁶ This could be due to what may be termed a 'saturation' effect, whereby CH already reacts on virtually every collision at room temperature, leaving it very little possibility for further increase at low temperatures.

Both reactions display extremely rapid rates of greater than $10^{-10} \text{ cm}^3 \text{ molecule}^{-1} \text{ sec}^{-1}$ at temperatures of interstellar interest (10–50 K), and these results serve to reinforce the conviction that neutral-neutral reactions play a crucial role in the chemistry of dense interstellar clouds at ultra-low temperatures.¹¹

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R. Becerra¹, S. Boganov², M. Egorov², O. Nefedov² and R. Walsh³¹Instituto Química Física "Rocasolano", C.S.I.C. C/Serrano 119, 28006 Madrid, Spain,²Zelinsky Institute of Organic Chemistry, Leninsky Prospekt 47, Moscow 117913, Russia,³Department of Chemistry, University of Reading, Whiteknights, PO Box 224, Reading RG6 2AD, UK.

INTRODUCTION

Dimethylgermylene, GeMe_2 , may be reasonably expected to play an important role in the breakdown of organogermanium compounds^{1,2}. Experimental studies, mainly in the liquid phase, indicate that germynes behave rather like silylenes in their reactions: they insert into Ge-H and O-H bonds and add to the π -bonds of dienes and alkynes, although apparently not to alkene π -bonds.

While most of the available information about germylene reactions has come from end-product analyses, recently, time-resolved kinetic studies by the groups of Mochida³ and Gaspar⁴ have supplied the first directly measured rate constants for their reactions. These studies, carried out in cyclohexane solution, suggest that while GeMe_2 has a similar pattern of reactivity to SiMe_2 , it is a much less reactive species. Our knowledge and understanding of the mechanisms of silylene reactions has been greatly increased in the last ten years by the application of such time-resolved studies, particularly in the gas-phase⁵. Accounts of some of these studies, carried out using lasers supplied by the LSF, have appeared in previous annual reports⁶. We therefore decided to extend this technique to the investigation of germynes such as GeMe_2 in the gas-phase.

EXPERIMENTAL

The apparatus and equipment for these studies have been described in detail previously⁵. Only essential and brief details are therefore included here. The target reactive transient was produced by flash photolysis of appropriate precursor molecules (see below) using an Oxford Lasers KX2 exciplex laser operating at 193nm (ArF fill). Transient absorptions were monitored in real time by means of the LSF Spectra Physics 2025 Argon Ion laser. For species characterisation all 9 available lines of the probe laser were used, but for the kinetic studies the argon ion laser was operated at 479nm. Experiments were carried out in a spectroil quartz cell with demountable windows. The photolysis beam (4cm x 1cm cross-section) entered the centre of the cell laterally, while the probe beam was multipassed along the axis of the cell up to 32 times giving a total path length of ca 1.2 m. Photolysis laser pulse energies were typically 60-70 mJ. Light signals were measured by a dual photodiode/differential amplifier combination, and signal decays were stored in a transient recorder (Datalab DL 910) interfaced to a BBC microcomputer. This was used to average the decays of 5 or 10 photolysis laser shots (at a rep rate of 1 - 2 Hz). Gas mixtures for photolysis were made up containing 100 mTorr of the transient precursor, variable pressures of reactive substrates with total pressures made up to between 1 and 100 Torr with inert diluent (SF_6). Pressures were measured with capacitance manometers (MKS Baratron). Most measurements were made at 294 ± 2 K. The organogermanium materials used in this work were synthesised by literature methods⁷. Gas chromatography was used to check for end product formation.

RESULTS

GeMe₂ precursors

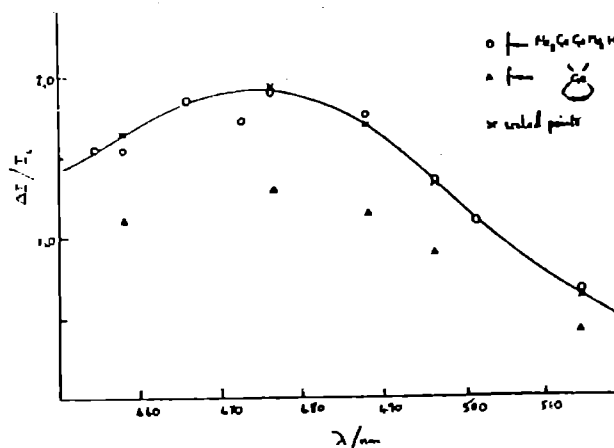
Pentamethyldigermane (PMDG), shows a strong UV absorption ($\lambda_{\text{max}} = 202$ nm, ext. coeff. = $2.5 \times 10^3 \text{ dm}^3 \text{ mol}^{-1} \text{ cm}^{-1}$) making it suited to photolysis studies at 193nm. End product analyses reveal that Me_3GeH is the major (stable) photolysis product, although other small peaks are present on the gas chromatogram.

Under time-resolved conditions strong absorptions were obtained in the 450 - 510 nm region. Although the extinction coefficient of GeMe_2 is not known, transient decay traces exhibiting good signal to noise ratios could be obtained with 100 mTorr of PMDG with averaging over 5 shots. Although as much as 10% of the transient precursor may be decomposed per laser shot, only ca 5% of the total vessel volume was irradiated. Thus the precursor pressure in the photolysed volume was replenished by diffusion between shots.

1,1-Dimethyl-1-germacyclopent-3-ene (DMGCP) also shows a strong UV absorption ($\lambda_{\text{max}} = 190$ nm, ext. coeff. = $2.1 \times 10^3 \text{ dm}^3 \text{ mol}^{-1} \text{ cm}^{-1}$). End product analyses show 1,3-butadiene to be the major (stable) decomposition product, although photolysis was often accompanied by concomitant dust formation, making DMGCP difficult to work with for time resolved studies. Nevertheless strong absorption signals were observed in the 450 - 510 nm region under these conditions. With care reasonable decay traces could be obtained. Under the same experimental conditions (precursor concentration and total pressure) transient signals were about 67% of those obtained from PMDG.

The GeMe₂ visible absorption spectrum

The transient absorption spectrum was obtained by monitoring the zero time absorbance (obtained by computer fitting of an exponential decay curve) at each of the nine available wavelengths of the argon ion probe laser. In these experiments 100 mTorr of precursor was photolysed in the presence of SF_6 at a total pressure of 10 Torr, at a photolysis laser energy of 65 ± 5 mJ/pulse. Both precursors were used for this purpose, although only five wavelengths were used for the transients from DMGCP. The results are shown in fig. 1. The values for the absorptions from DMGCP were scaled by a common factor of 1.49 to match the signal magnitude from PMDG. The satisfactory agreement between the spectral absorptions at different wavelengths from both precursors provides good support for a common species. That this should be GeMe_2 is reinforced by the comparison with the known absorption spectrum obtained in an argon matrix⁸. Although not shown in the figure there are several reports of the condensed phase spectrum with a peak maximum at 420 - 430 nm⁴. A gas phase red shift is usually expected, although the magnitude here is slightly larger than that found previously (ca 10 nm) for SiMe_2 ⁹. A point worth noting is that the decay constants (see below) for

Figure 1. Absorption spectrum of GeMe_2

signals from each precursor, although different from one another, were independent of the wavelength for a given precursor, again indicative of a common species. In sum, the wavelength location of this spectrum, the use of rational chemical precursors, and agreement between two different sources provide strong evidence that this is indeed the visible spectrum of GeMe_2 .

Gas-phase rate constants for reactions of GeMe_2

For kinetic studies, PMDG was used as the GeMe_2 precursor and the transient was monitored at 479nm, near its absorption maximum. Decay traces (5 shot averages) were found to fit exponential forms consistent with first order kinetics. Linear least squares fitting over ca 70 - 90% of each decay trace was used to obtain the first order rate constant, k_{obs} . Rate constants were found to be independent of excimer laser energy (within a range 40-80 mJ/pulse) and also the number of laser shots (up to 20 shots). At a given substrate pressure the reproducibility of k_{obs} values was within $\pm 10\%$ of the mean (maximum spread 20%), although often better than this.

Substrates were chosen to cover a selection of potential reaction types. No reactions were found with C_3H_8 (33 Torr), Me_3SiH (112 Torr) or Me_3GeH (3 Torr). Reactions were however found with oxygen, ethylene, 3,3-dimethylbutene-1 (C_6H_{12}) and 1,3-butadiene. In these studies, the partial pressure of reactive substrate was varied in a systematic way to test the dependence of k_{obs} . These experiments yielded reasonable linear plots as expected for second order kinetics. Examples of such plots are shown in fig. 2, for the reaction with C_2H_4 . As can be seen in this case, the slopes of the second order plots varied with total pressure, thus showing the reaction rate constants to be pressure dependent. Such overall pressure dependencies were not investigated in other cases except for C_6H_{12} , where rate constants were found to be the same at 10 and 30 Torr (SF_6), but a slightly lower value was found at 3 Torr. The second order rate constants, found from the slopes of these plots are collected in table 1. Apart from these, one other substrate *viz.* C_2H_2 was studied, but the scatter in the data was rather higher than for the other molecules. Reaction clearly took place rapidly (k of ca $1.3 \pm 0.3 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$) but the scatter was too great to determine whether this reaction showed any pressure dependence (3 - 30 Torr pressure range).

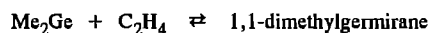
DISCUSSION

The measurements reported here represent the first experimentally determined gas-phase rate constants for GeMe_2 . Two features of the data are apparent from the table. GeMe_2 does not insert into C-H, Si-H or Ge-H bonds on the time scale of these experiments. It does however add readily to the π -bonds of alkenes, dienes and alkynes. By contrast, SiMe_2 not only adds to π -bonds with similar

Table 1. Rate constants for GeMe_2 at 294 K

Substrate	$k/\text{cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$	
O_2	4.6×10^{-14}	
C_2H_4	9.1×10^{-13}	(3 Torr SF_6)
	1.7×10^{-12}	(10 ")
	4.5×10^{-12}	(30 ")
	7.0×10^{-12}	(100 ")
$\text{H}_2\text{C}=\text{CHCMe}_3$	9.8×10^{-12}	(10,30 ")
1,3- C_4H_6	1.2×10^{-11}	
C_3H_8	no reaction	
Me_3SiH	"	
Me_3GeH	"	

facility, but also inserts readily in Si-H bonds (SiMe_2 does not however insert in C-H bonds, and its reaction with Ge-H bonds has not yet been studied). We do not speculate here on the reasons for these differences since these are likely to become more evident when we have extended these measurements to other reactive substrates and higher temperatures. The pressure dependent addition reaction to ethylene almost certainly indicates that it produces the weakly bound germirane ring product as follows:



This pressure dependence is characteristic of a third body assisted combination process and indicative of the weakly bound nature of the product. Evidence that germirane rings are highly strained comes from the fact that they have only recently been synthesised despite many efforts¹⁰.

A word of caution should be entered that these results are in some contrast to the findings of Mochida³ and Gaspar⁴ in their solution studies. Both sets of investigators find no reaction of GeMe_2 with alkenes (or alkynes), and rate constants with dienes that are ca 10^3 slower than those reported here. Only the reaction with O_2 is faster than our measurements. The only obvious difference here is the nature of the reaction medium. If the ground state germylene is complexed in solution then it might become less reactive. The solvent however is the non-polar cyclohexane. The explanation for these differences may become more evident on further investigation.

The findings here are necessarily preliminary and we hope to throw more light on the reactivity and behaviour of GeMe_2 and other germynes in future work. A more extended account of current findings is in preparation⁷.

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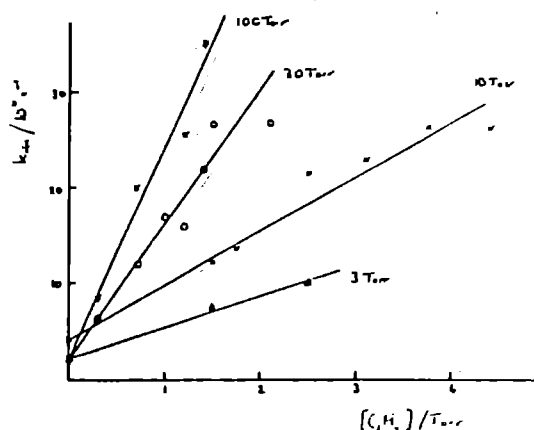


Figure 2 Second order plots for $\text{GeMe}_2 + \text{C}_2\text{H}_4$ (in SF_6)

DYNAMICS OF HETEROGENEOUS STRATOSPHERIC PROCESSES

H. Rieley, S. M. Reeman and D. P. McMurray
Department of Chemistry, University of Liverpool

INTRODUCTION

The oxides of nitrogen all feature in the chemistry of the atmosphere. All have a bearing on the NO_x cycle¹ and, recently, there has been renewed concern in the direct deposition of N_xO_y molecules in the atmosphere from increased numbers of high flying jet aircraft². There is some uncertainty about the uptake and subsequent reaction of species such as NO_2 on water-based ices and we recognise the need to study these molecules in a controlled high vacuum environment at rather lower temperatures. In addition, the photochemistry of atmospheric species in the adsorbed state has received little attention to date. This acknowledges the fact that most of the products formed heterogeneously on PSCs depart into the gas-phase to await photolysis or further reaction. However, some consideration should be given to modifications of the gas-phase chemistry and photochemistry of these molecules which may occur in the adsorbed state, not least, to establish their significance in cases where heterogeneous reactions are rate limiting (e.g. on sulphate aerosols where rate coefficients are lower²). The presence of a surface, even in weakly adsorbed systems has been shown to radically change both the dynamics and the reaction pathways which follow photodissociation^{3,4}. Finally, our recent FTIR study of N_2O_4 adsorbed on ice shows that the molecules in the overlayer are oriented on the surface⁵: a dynamics study should reveal the consequences of this orientation on the photodissociation.

EXPERIMENTAL

Experiments were conducted using a high vacuum (HV) apparatus in conjunction with a Quanta-Ray Nd:YAG laser (RAL Laser Loan Pool). The HV system was a low volume vessel (~5 litres) constructed of stainless steel and pumped by two liquid nitrogen trapped 2" diffusion pumps to an ultimate pressure of 10^{-9} torr, as measured by an ion gauge. A differentially pumped quadrupole mass-spectrometer (VG Micromass PC) permitted residual gas analysis and monitoring of the temporal evolution of neutral species from a pre-prepared sample surface. A copper single crystal mounted on tungsten wire between current carrying copper rods beneath a liquid nitrogen cooled dewar provided a means for condensing thin layers of water at the desired temperature. Unfocused and unpolarised radiation at 355 nm was incident on the centre of the sample, in the plane of the detector and sample normal.

RESULTS AND DISCUSSION

Figure 1 shows a typical single-shot time-of-flight spectrum of photodetached NO_2 (as probed at $m/z=30$, NO^+) following photolysis of N_2O_4 , deposited approximately as a monolayer film on the surface

of ice. After 4-5 shots no further signal was evident - indicating complete photodepletion of the monolayer. The TOF spectrum is well described by a Maxwell-Boltzmann with a superimposed stream velocity. The most probable energy of the nascent NO_2 is ~0.1 eV or equivalent to a Boltzmann "temperature" of ~1200 K. Considering that the surface temperature is around 100 K, the photodissociation is clearly direct in nature with little accommodation at the surface. This is consistent with, yet distinct from, the gas phase photofragmentation at this wavelength which has an available energy of ~1.10 eV⁶. There is obviously some quenching of the product energy to the ice surface. The angular distribution of the nascent NO_2 appears to be strongly peaked at the surface normal and drops beneath the limit of detection at 45° to the normal. This observation is consistent with the adsorbate geometry as determined by RAIRS⁵ and the repulsive force of the photolysis being directed along the breaking N-N bond and hence parallel to the surface normal.

In summary, the photolysis of N_2O_4 and other oxides of nitrogen is mediated by adsorption on an ice surface. In cases where the desorption temperature is low this will be of little consequence to current atmospheric models, but in cases where adsorbed species have a significant lifetime modifications to the photodissociation energies and pathways may need to be taken into account.

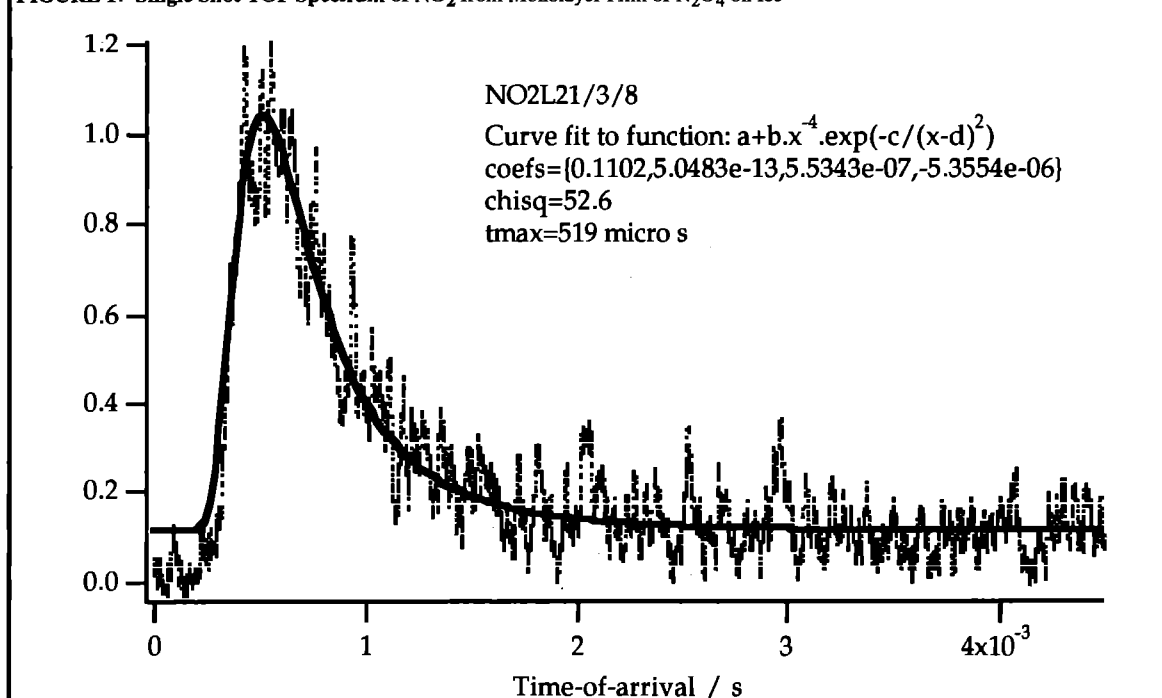
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FIGURE 1: Single Shot TOF Spectrum of NO_2 from Monolayer Film of N_2O_4 on Ice



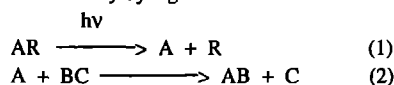
THE DYNAMICS OF ELEMENTARY OXYGEN ATOM + HYDROCARBON REACTIONS

Gillian M. Sweeney and Kenneth G. McKendrick

Department of Chemistry, The University of Edinburgh

INTRODUCTION

The technique of laser photolysis to produce 'hot atoms' for the investigation of the dynamics of elementary chemical processes is now well established. The approach may be represented schematically by a generic reaction scheme



where a precursor molecule, AR, is photolysed to produce a reactive fragment, A, which then subsequently collides and reacts with a molecular reagent, BC. The AB product of the reaction is typically observed spectroscopically using an independent laser source. We are exploiting this method with the intention of studying the dynamics of the reactions of oxygen atoms in the ground (O^3P) and first excited (O^1D) states with hydrocarbon molecules. Such reactions are not only of considerable fundamental interest but also have practical significance in combustion and atmospheric processes.¹

EXPERIMENTAL

The CLF has provided a high power excimer laser (Questek Series 2000), which acts as the photolysis source in step (1). A second tunable dye laser system produces pulses which are used to detect the products of the reaction either by laser-induced fluorescence (LIF) or by resonance-enhanced multiphoton ionisation (REMPI). The delay between photolysis and probe pulses is kept sufficiently short to ensure that the nascent product state distributions are not modified by subsequent inelastic collisions.

RESULTS

This report is being prepared approximately midway through a six month loan period. Some success has already been achieved, with further experiments planned.

O^3P Reactions

Ground state oxygen atoms have been generated by KrF excimer laser photolysis of NO_2 at 248 nm. This has allowed us to extend our own earlier studies using longer photolysis wavelengths² by providing O^3P atoms with higher translational energies.³ We have successfully detected by LIF the OH products of the reactions of these translationally hot atoms with a variety of hydrocarbons, as illustrated in Fig. 1. The results generally confirm that very little of the available energy is partitioned as product rotation, as observed at lower collision energies.^{2,4} It has been inferred that the reactions therefore exhibit a very strong preference for a collinear C-H-O geometry. Further details of the partitioning between vibrational and fine structure states of the OH are under investigation.

We also intend to extend these studies to include reaction with CH_4 , which so far has not received attention because of the relatively high activation barrier associated with the stronger C-H bond. In this case, we will also attempt to detect the nascent CH_3 coproduct of the reaction by REMPI. Preliminary one-colour experiments involving photolysis of CH_3I have demonstrated the viability of a [2+1] REMPI scheme for CH_3 with our apparatus.⁵

O^1D Reactions

We intend to complement the studies of ground state oxygen atom reactions by generating the first excited state, O^1D , by operating the excimer laser on ArF at 193 nm and photolysing N_2O . The highly internally excited OH product state distributions from O^1D + hydrocarbon reactions are well established,⁶ but only very recently has any information appeared on the CH_3 product state partitioning from the reactions with CH_4 and C_2H_6 .⁷ The contrasting dynamics of the reactions of O^1D and O^3P with the same partners provide a very interesting test of theoretical predictions of low-lying singlet and triplet potential energy surfaces for relatively simple systems involving light atoms.

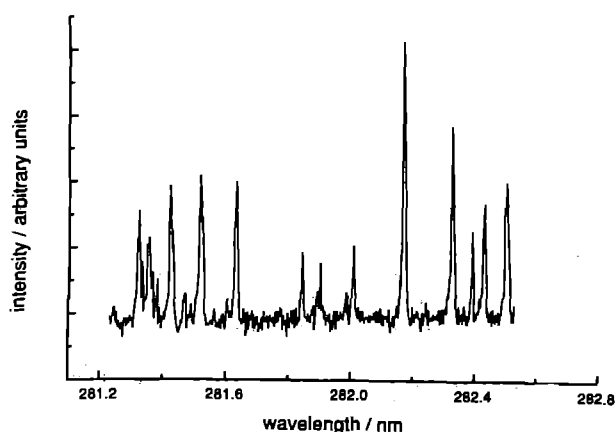


Figure 1 LIF Excitation spectrum of the A-X (1,0) band of OH produced in the reaction of O^3P with isobutane. The O^3P was generated by 248 nm photolysis of NO_2 .

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LASER SPECTROSCOPY OF ORGANOMETALLIC INTERMEDIATES

A. J. Bezant, G. K. Corlett, S. J. Pooley, and A. M. Ellis
Department of Chemistry, University of Leicester, Leicester LE1 7RH

INTRODUCTION

This work was a continuation of a project described in the 1994 Annual Report in which organometallic radicals, prepared in a supersonic jet using pulsed electrical discharge methods, were investigated via laser spectroscopy. Electrical discharge methods were employed because of their low cost relative to more expensive methods of gas-phase radical production such as laser photolysis. We have experimented with two methods for producing organometallics consisting of a single metal atom bound to a single organic ligand:

- (1) Fragmentation of a volatile organometallic precursor in a discharge.
- (2) Synthesis by metal + ligand reactions in a discharge.

Both were demonstrated to be feasible in our previous report. Since then we have extended our studies using method (1) and made further experimental modifications with respect to method (2).

EXPERIMENTAL

Our simplest discharge nozzle, employed for direct fragmentation of organometallics, is based on a design by Bondybey and co-workers [1], and provides a DC discharge between metal electrodes prior to pulsed gaseous expansion. In this simple arrangement, a voltage of up to 1500 V is applied between the electrodes and a discharge is automatically initiated when the pulsed valve is opened and gas is fed into the inter-electrode region. A more significant development is the introduction of a dual-discharge system, illustrated in Figure 1, which is employed for method (2) above. Metal atoms are produced by cathode sputtering by positive ions in the discharge, but if this is performed in the presence of an organic ligand precursor, rapid coating of the electrode surface by a carbon film soon depletes the metal atom concentration. To solve this problem, we employ two discharges, one to produce metal atoms in a clean (inert gas) environment, and the other to fragment the precursor to obtain the desired organic ligands. The gas flows are combined, reaction takes place, and the mixture is then expanded into vacuum where it is probed using laser-induced fluorescence spectroscopy.

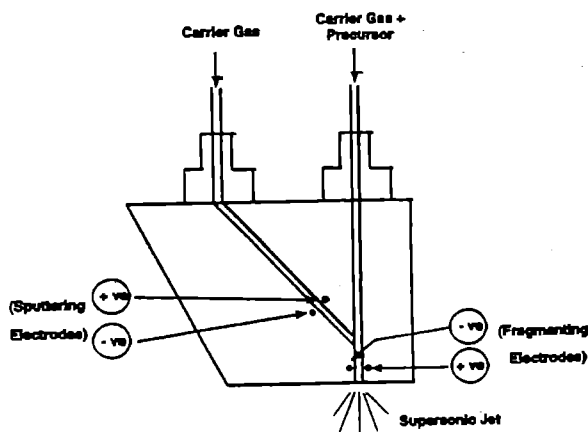


Figure 1 Schematic diagram of dual-discharge nozzle

RESULTS AND DISCUSSION

A. Dispersed fluorescence spectroscopy of zinc monoethyl

We recently recorded and published the first spectrum of the zinc monoethyl radical [2]. The laser excitation spectrum was obtained following discharge fragmentation of zinc diethyl but, apart from identifying the carrier, no firm assignments of any vibrational structure in the spectrum could be made. We have now collected dispersed fluorescence spectra of this molecule by laser exciting a number of vibronic transitions and recording the subsequent emission spectra. One such spectrum is shown in Figure 2. Using this data, we have been able to assign some of the vibrational structure arising in the laser excitation spectrum, including firm identification of the Zn-C stretching and Zn-C-C bending vibrational frequencies. There is still more work to be done, notably deuterium substitution experiments, but a full assignment is anticipated shortly.

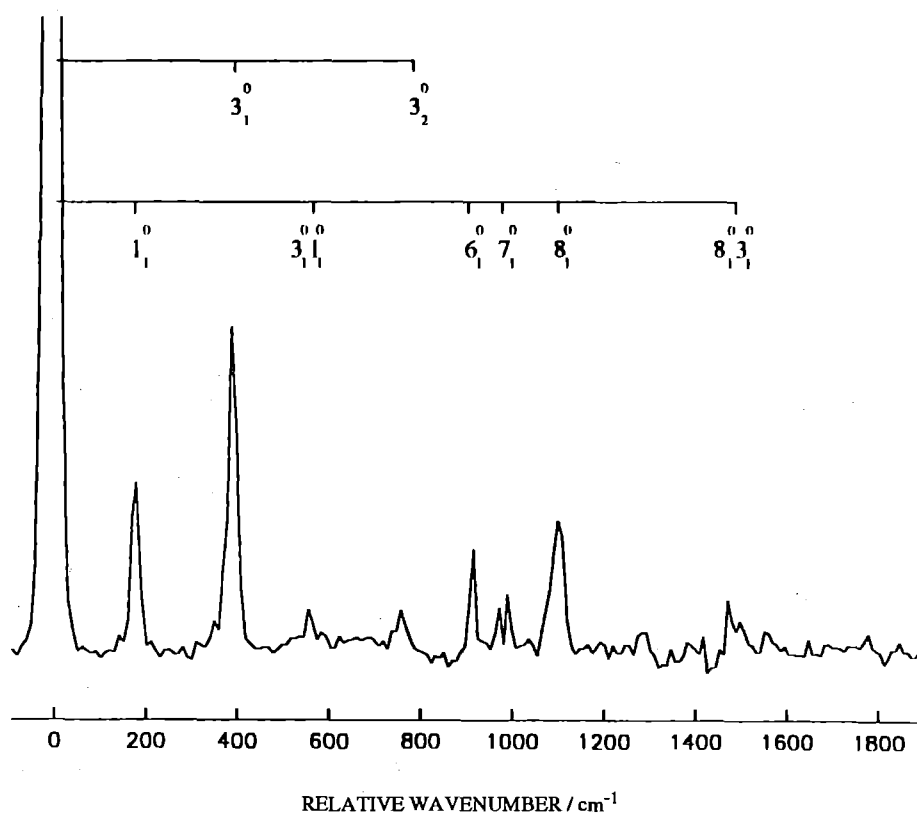


Figure 2 Dispersed fluorescence spectra of ZnC_2H_5 pumping $\tilde{A} - \tilde{X} \ 0_0^0$ transitions

B. Synthesis of ZnCH_3 using dual-discharge sputtering approach

Our new discharge method has been tested by synthesising the zinc monomethyl radical using $\text{Al}(\text{CH}_3)_3$ as the methyl precursor. ZnCH_3 has a strong LIF spectrum and so is an ideal molecule for testing the sputtering method. Figure 3 shows the excitation spectrum in the region of the $\tilde{A} - \tilde{X}$ electronic origin; a strong ZnCH_3 spectrum is obtained, much stronger than observed previously using a single discharge sputtering synthesis (1994 report). This highly successful test demonstrates that the dual-discharge method has great potential for the production of a variety of metal-containing intermediates for spectroscopic studies. In the case of ZnCH_3 , the observed radicals are vibrationally hot, as evidenced by the broadening of the origin band by sequence structure. Attempts to improve the vibrational cooling will be one target of future work.

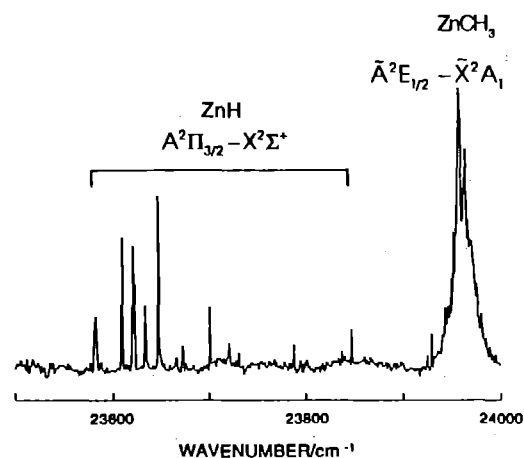


Figure 3 Laser excitation spectrum of $\tilde{A} - \tilde{X}$ origin region of ZnCH_3 obtained using dual discharge nozzle

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TIME-RESOLVED RESONANCE RAMAN SPECTROSCOPY OF INDOLYL AND RELATED RADICALS.

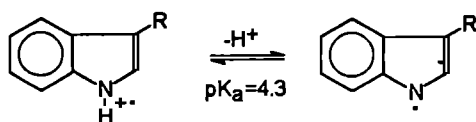
Roger H Bisby¹, Steven A Johnson¹, Anthony W Parker² and Susan M Tavender²

¹Department of Biological Sciences, University of Salford, Salford M5 4WT

²Laser Science Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxon, OX11 0QX

INTRODUCTION

Tryptophan and related radicals are important intermediates in many biochemical processes, occurring as enzyme radicals in the catalytic activities of cytochrome c peroxidase [1] and DNA photolyase [2], after photo- and radiation damage to proteins [3] and during the action of indole derivatives having antioxidant properties [4,5]. In many previous studies, the tryptophanyl radical cation has been identified by its characteristic UV-visible absorption spectrum, and shown [6] to have a pK_a of 4.3:-



The only reported resonance Raman spectrum of a tryptophanyl radical is that obtained by Johnson, Ludwig and Asher [7] using a focused 235 nm laser pulse as a simultaneous pump and probe beam. Since this is likely to create both excited states and radical species, there must be some ambiguity about the species studied by Johnson et al.. For biochemical studies the use of such a short wavelength probe pulse is impractical due to the multiplicity of other chromophores absorbing in the far UV. In the present study we have employed a probe laser pulse in the long wavelength tryptophanyl radical absorption band at 580 nm (radical cation) or 520 nm (neutral radical) and have used triplet duroquinone as a reliable method for generating the radicals by hydrogen atom abstraction.

RESULTS

The indole radicals were generated in the present experiment by sensitised photooxidation with triplet duroquinone (³DQ). The UV/visible spectra of the product radicals confirmed their identity. This is demonstrated for L-tryptophan in Figure 1, which shows the characteristic long wavelength absorptions of the radical cation (λ_{max} 580) in acidic solution and of the neutral radical (λ_{max} 520 nm) in

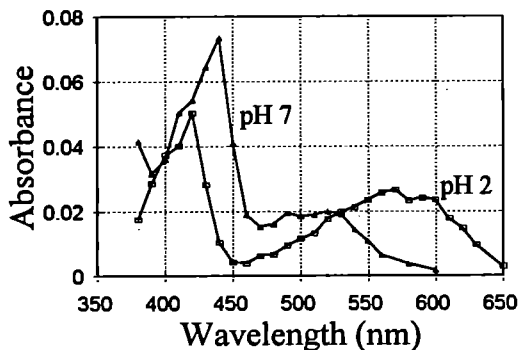


FIGURE 1 - Tryptophanyl radical spectra from laser flash photolysis (360 nm, 5 mJ) of acidic and neutral solutions of tryptophan (0.8 mM) and duroquinone (2 mM) in ethanol-water (50:50 v/v) ethanol/water (50/50 v/v) solutions.

neutral solution., superimposed on the shorter wavelength absorption bands of the duroquinone radicals (λ_{max} 410-440 nm). The rate of oxidation of tryptophan by triplet duroquinone was found to be $(1.4 \pm 0.1) \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$ at pH 7. A similar value was obtained for oxidation of acetaminophen by ³DQ.

Time-resolved resonance Raman spectra obtained in aqueous ethanol solutions at pH 7 containing both duroquinone and tryptophan shown in Figure 2 indicate the decay of the ³DQ band at 1549 cm^{-1} and the subsequent formation of a number of other weaker bands which are ascribed to the neutral tryptophanyl radical. Similar TR² spectra were obtained for a number of other indoles. Figure 3 shows the TR² spectra of the radical cation from tryptophan and the neutral radical from tryptamine. It is demonstrated that the resonance Raman spectra clearly distinguish between the neutral radical and radical cation. Figure 2 also shows the spectrum of the radical cation from 1-methyltryptophan, which cannot deprotonate. This is very similar to the spectrum obtained tryptophan under acidic conditions, and helps to confirm the assignment of the latter to the radical cation. However, in addition the 1-methyltryptophan radical spectrum contains an additional band at 1247 cm^{-1} which might be assigned to the N-C (methyl) stretching vibration.

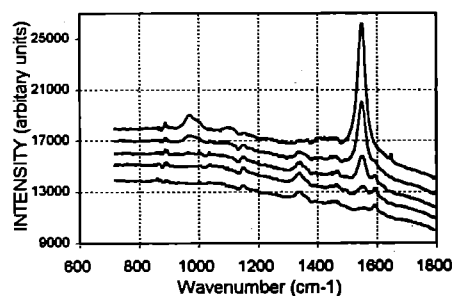


FIGURE 2 - Time-resolved resonance Raman spectra from ethanol-water (50:50 v/v) solutions of duroquinone (5 mM) and tryptophan (2 mM). The delay between pump (360 nm) and probe (520 nm) pulses was 10, 100, 250, 500 and 2000 ns from top to bottom.

Since the indole ring lacks a high degree of symmetry, vibrational assignments are difficult and have only recently been considered in detail [8]. The band at about 1590 cm^{-1} in the spectrum of the neutral indolyl radical (1609 cm^{-1} in the radical cation) appears to be related to the 1621 cm^{-1} band in the resonance Raman (260 nm excitation) spectrum of tryptophan [9]. Johnson et al [7] reported a similar band at 1593 cm^{-1} which we may assign to a ring C-C stretching mode. The lower frequencies of the bands in the radical spectra are consistent with the weaker bonds in the molecule following removal of a bonding π electron on oxidation, whilst the lower frequency of the neutral radical compared with the radical cation indicates more extensive radical site delocalisation in the former species. Although several similarities exist between the spectra of the neutral tryptophan radical reported here and by Johnson et al, the latter also report additional bands at 1522 and 1646 cm^{-1} which we have been

unable to observe. It is possible that whilst these are strongly resonantly enhanced on excitation at 235 nm, they are absent from the spectrum excited at 520 nm. This would be similar to the 1591 cm^{-1} band of the phenoxyl radical which is present in the resonance Raman spectrum probed at 245 nm [7], but not when probed at 410 nm [10]. Alternatively, these additional bands may represent another transient species, such as tryptophan singlet and triplet states with lifetimes of about 2 ns and 2 μs respectively, and which would therefore be sensed in the experiment of Johnson et al using a single 4 ns pulse as both pump and probe. It is known that the quantum yields for monophotonic ionisation of tryptophan are substantially less than unity [3].

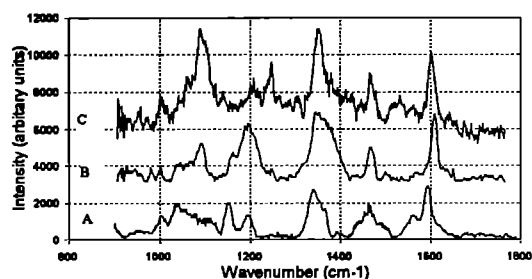


FIGURE 3 – Resonance Raman spectra of indolyl radicals. A:- neutral radical from tryptamine (2 mM) and duroquinone (5 mM) in ethanol/water (50:50 v/v) probed at 520 nm; B:- radical cation from L-tryptophan (2 mM), duroquinone (5 mM) and HCl (5 mM) in acetonitrile-water (80:20 v/v), probed at 580 nm; C:- radical cation from N-methyltryptophan (2 mM) and duroquinone (5 mM) in acetonitrile-water (80:20 v/v) probed at 580 nm.

TR^2 spectra from radicals formed on one-electron oxidation of 6-hydroxy- and 5-hydroxy-indole are shown in Figure 4. In both cases one-electron oxidation of these compounds is suggested [5, 11] to lead to indoloxyl radicals with the radical site predominantly located at the *exo* oxygen atom. Assignment of bands in these spectra requires further work using isotopically labelled compounds. The 5-indoloxyl radical undergoes deprotonation with a pK of 12.2 [5] and the spectrum A in Figure 4 measured in the presence of 50 mM sodium hydroxide should therefore correspond to the 5-indoloxyl radical anion. For comparison, TR^2 from the radicals derived from acetaminophen (N-acetyl-4-aminophenol) are shown in Figure 5. The neutral acetaminophen radical has a pK of 11.1 [12] and the spectrum

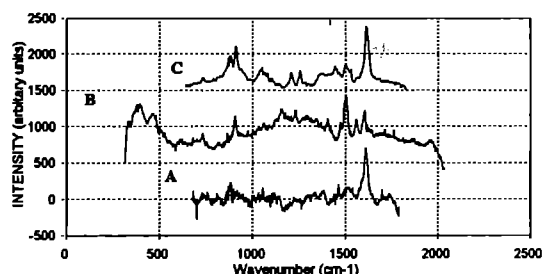


FIGURE 4 – Resonance Raman spectra of hydroxy-indolyl radicals. A:- Radical from oxidation of 5-hydroxyindole by triplet duroquinone in ethanol-water (50:50 v/v) and 50 mM NaOH, pump 355 nm, probe 520 nm delayed by 1 μs ; B:- as in A but without alkali, pump 355 nm, probe 425 nm delayed by 1 μs , after subtraction of durosemiquinone radical anion spectrum; C:- radical from oxidation of 6-hydroxyindole by triplet duroquinone in ethanol, pump 360 nm, probe 500 nm delayed by 500 ns.

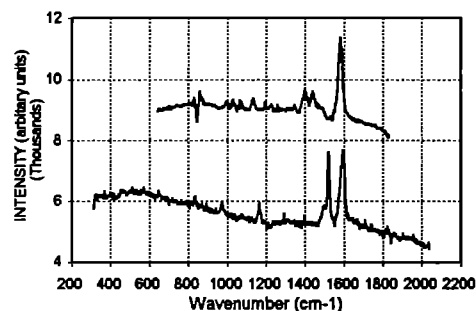


FIGURE 5 – Time-resolved resonance Raman spectra of radicals from one-electron oxidation of acetaminophen. Lower spectrum - the neutral semi-iminoquinone radical in ethanol-water (50:50 v/v), pump 355 nm, probe 425 nm (after subtraction of durosemiquinone radical anion spectrum); upper spectrum - the semi-iminoquinone radical anion in ethanol-water + 10^{-2} M NaOH, pump 360 nm,

recorded in the presence of 10^{-2} M sodium hydroxide represents the radical anion. These acetaminophen radicals may be regarded as prototypes for the 5-indoloxyl radicals, and indeed the two radical anion spectra are very similar.

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MIGRATION OF OXIDATIVE DAMAGE IN DNA.

P. O'Neill,¹ S. Botchway,¹ T. Melvin,¹ A.W. Parke,² P. Cullis.³
MRC Radiobiology Unit,¹ Rutherford Appleton Laboratory,² Leicester University.³

INTRODUCTION

The various biological effects of ionising radiation and UV light such as cellular inactivation, transformation and radiation mutagenesis are thought to be as a consequence of chemical modifications to DNA.¹ Of all the lesions, non repairable DNA double strand breaks (dsb) have been implicated in cellular inactivation by ionising radiation.² The initial chemical events occurring in DNA following ionising radiation arise by the interaction of water radicals, namely the hydroxyl radical, with DNA, and the direct deposition of energy within DNA (direct effects). The damage produced upon interaction of the OH radical with DNA and model systems has been well documented³ whereas much less is known about the chemical events following direct energy deposition in DNA.

It is difficult to study the species produced upon direct energy deposition in DNA by ionising radiation in an aqueous environment due to the overwhelming yield of water radicals. The majority of the information about the radicals formed from the direct effects of ionising radiation comes from esr studies⁴ on DNA irradiated at 77K. Two types of species are produced namely electron loss and electron gain centres which are localised upon the base moieties. With the advent of lasers which produce light at < 200nm, photolysis is an alternative approach. Ionisation of DNA and model compounds by 193nm light has been shown,^{5,6} from spectrophotometric studies of transient species, to be an important photochemical process. Migration of this oxidative damage to the most easily oxidised base, guanine, is predicted from their oxidation potentials, T→C→A→G.⁷ Although DNA acts as a good matrix for photo-induced electron transfer between intercalated donor and acceptor molecules⁸, definitive evidence for migration of the radical cations of nucleic acid bases in aqueous solutions of DNA or model systems (i.e. oligodeoxynucleotides) has not been reported.

The following studies were done to assess the role of charge migration in DNA following photolysis with 193nm light and to determine the sequence-specificity of strand breakage. Although photo-ionisation produces isolated radicals and does not produce clustered events as with ionising radiation, the intention of these studies is to identify potential precursors to DNA damage induction through direct ionisation events.

MIGRATION OF OXIDATIVE DAMAGE IN DNA.

Transient absorption spectra, obtained by laser flash photolysis by 193nm light, of a series of single stranded oligonucleotides, were assessed for the migration of oxidative damage. The oligodeoxynucleotides (1,2,3,4,5, see Table) were synthesised as well as three nonamers containing either guanine (G9), adenine (A9) or cytosine (C9). The initial distributions of photo-induced radical cations formed at each of the nucleic acid base species can be estimated from the extinction coefficients (193nm) and the quantum yields of photo-ionisation of the monomeric nucleic acid bases⁹ and are shown in the Table. Optical absorption spectra obtained 5μs after photolysis with 193nm light of single stranded oligodeoxynucleotides and the nonamers in oxygenated, aqueous solution (293K, pH=7±0.5) are displayed in Figures 1-3. The extinction coefficients shown were determined by reference to the absorbance of the hydrated electron at 650nm ($\epsilon_{650} = 15800 \text{ M}^{-1} \text{ dm}^3$) for identical solutions of these oligodeoxynucleotides, but saturated with argon.⁵ The quantum yield for photo-ionisation of the oligodeoxynucleotides studied are in the range 0.043-0.110. Evidence for migration is obtained by comparison of the transient optical spectra obtained for 1,2,3,4 or 5 with that obtained for the nonamer containing the most easily oxidised base.

The first group of oligodeoxynucleotides (1,2 & 3) contains two nucleobases, whereby the less easily oxidised base is in excess. The absorption spectra for 1 and 2, following photolysis, were remarkably similar to that obtained for G9 (figure 1). It is estimated that migration of the radical cation from either adenine or cytosine to guanine occurs by a significant degree (>80% migration with <10% contribution from the radicals of adenine and cytosine). In contrast, migration of the oxidative damage from cytosine to

adenine is not significant (<10% migration); the transient spectrum obtained from photo-ionisation of 3 (figure 2) is a composite spectrum containing components for A9 and C9 (identified by major optical absorption bands at 330 and 290nm respectively) reflecting the distribution shown in the Table. An interpretation of this result is that alternative chemical pathways for loss of the oxidising cytosine radical compete with electron transfer.

Calculated Mole Fraction of Radical Cations formed by photo-ionisation.

Oligodeoxynucleotide	G	A	C
AGAAGAAGA (1)	0.68	0.32	-
CGCCGCCGC (2)	0.74	-	0.26
CACCACCAC (3)	-	0.40	0.60
CAACGCAACGCAAC (4)	0.44	0.32	0.22
CAACGCGCAAC (5)	0.33	0.32	0.35

Figure 1. Transient UV spectra (5μs) for G9 (filled circles and solid line), 1 (filled triangles and dashed line), 2 (open circles and dashed line), by laser flash photolysis (193nm).

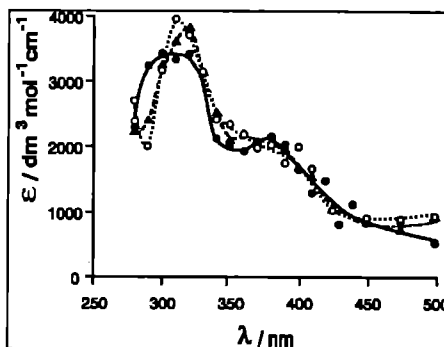
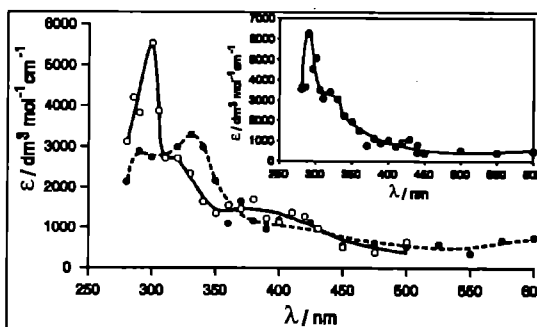
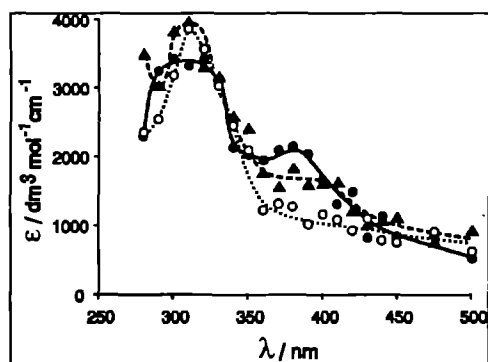


Figure 2. Transient UV spectra (5μs) for A9 (filled circles and dashed line), C9 (open circles and solid line), 3 (inset spectrum, filled circles and solid line), by laser flash photolysis (193nm).



Oligodeoxynucleotides (4 & 5) containing adenine, guanine and cytosine were used to determine whether oxidative damage formed at adenine could be transferred to guanine via a linking chain of either one or two cytosine molecules. Based on the evidence above with 1,2 and 3, radical cations of cytosine are predicted to transfer to guanine only. The most obvious spectral difference between the transient species of 4, 5 and G9 (figure 3) is in the spectral region, 350 - 420nm. The oxidation products of the guanine moiety absorb strongly in this region, whereas those of adenine and cytosine do not (see figure 2 for comparison). The dissimilarity of transient photo-ionisation spectra of 4 and 5 with that for G9 indicates that there is incomplete migration of the oxidative damage to guanine. The magnitude of the spectral absorption bands of 4 and 5 between 350 - 420nm is consistent with that predicted for insignificant migration of the oxidising radical from adenine to guanine via the cytosine moiety.

Figure 3. Transient UV spectra (5 μ s) for G9 (filled circles and solid line), 4 (filled triangles and dashed line), 5 (open circles and dashed line), by laser flash photolysis (193nm).



These results indicate that migration of oxidative damage in single stranded oligodeoxynucleotides occurs between certain neighbouring nucleic acid bases and is severely hindered when separated by a third, less easily oxidised base, e.g. cytosine as shown in these models. Migration of oxidative damage through the ordered double stranded helix of DNA, although as yet unstudied, is predicted to be less hindered.

CHARACTERISATION OF SINGLE STRAND BREAKS IN DNA CAUSED BY 193NM LIGHT

To determine whether 193 nm photolysis yields DNA strand breaks and whether the sites of cleavage are site specific, a known double stranded DNA sequence was 5' end-labelled on one strand, photolysed and resolved by denaturing sequencing gel electrophoresis. Under aerobic conditions a non-random cleavage of the DNA was observed as depicted in the autoradiograph shown in Figure 4 (lane 2). Hot alkali treatment increased the electrophoretic mobilities of single stranded fragments by ~1 nucleotide (Figure 4, lanes 2 and 3). From comparison of the fragmentation pattern observed after hot alkali treatment (Figure 4, lane 3) with that of the Maxam and Gilbert G&A chemical sequencing products (Figure 4, lanes 1 and 4), the breaks occur preferentially at guanine. From the effect of hot alkali (compare lanes 2 and 3 of Figure 4), it is inferred that the strand breaks occur 3' to the guanine and that the alkali treatment removes a residual sugar remnant to leave a fragment with an electrophoretic mobility similar to that produced by the M & G chemical sequencing reaction. The intensity of the bands in lanes 2 and 3 of Figure 4 cannot be directly compared due to differences in the treatment of the samples. However, our data do not preclude the possibility that other damaged guanine residues are converted into additional strand breaks upon treatment with hot alkali. A similar fragmentation scheme to explain the specificity of strand break following biphotonic excitation of DNA by 248nm light has been reported.⁹

From closer inspection of the sequencing gel of the 193nm photolysed DNA (Figure 4, lane 2), some breakage also occurs at non-guanine residues but at a very much reduced frequency. The efficiency of strand breakage arising from the other bases is estimated to be at least an order of magnitude less than that from guanine. More strikingly, the photo-induced cleavage associated with guanine shows a selectivity as shown on the densitometric analysis of the autoradiograph. For instance, neighbouring guanines generally result in bands of high intensity whereas the bands for guanine with a neighbouring 3'-thymine are less intense.

Photolysis of DNA in aqueous solution with 193nm light has been shown to induce 'frank' ssb in DNA with a ϕ_{ssb} of 4×10^{-4} which represents an efficiency of 2% when related to ϕ_{PI}^{ssb} . The ϕ_{ssb} for DNA with 193nm light is essentially independent of the presence of oxygen indicating a minor role for ssb induction by superoxide or singlet oxygen. From our findings, it is inferred that strand breakage induced by 193nm light in double stranded DNA occurs preferential by strand scission 3' to the guanine moiety and the hot piperidine treatment removes a residual sugar remnant on the 5'-terminus. This cleavage, predominantly at guanine, is consistent with the migration and localisation of the hole at guanine. Furthermore, it is inferred that strand breakage of the complementary strand opposite these guanine cleavage sites does not occur. Therefore, double strand

breaks would not be expected to arise from single photon events with 193nm light.

Figure 4. Base specificity of ssb induced in the DNA fragment of known sequence by 193nm laser light as resolved with denaturing sequence gel electrophoresis. Lanes 1 and 4 Maxam Gilbert (G/A) cleavage products; lane 2, laser induced cleavage products from irradiated DNA; lane 3, laser induced cleavage products from DNA following a hot piperidine treatment.



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RESONANCE RAMAN SPECTROSCOPY OF THE FREE RADICAL INTERMEDIATE IN COPPER AMINE OXIDASE

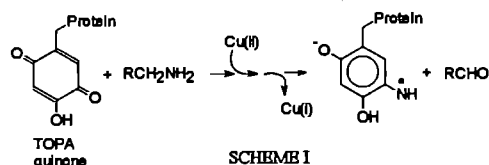
Roger H Bisby¹, Steven A Johnson¹, Anthony W Parker² and Susan Tavender²

¹Department of Biological Sciences, University of Salford, Salford M5 4WT, UK

²Laser Support Facility, Rutherford Appleton Laboratory, Chilton, OX11 0QX, UK

INTRODUCTION

Copper amine oxidases from a variety of sources, including *Arthobacter*, *E.coli*, *Pisum sativum* and pig kidney, have been shown to contain the quinone form of trihydroxyphenylalanine (TOPA quinone) in the active site^{1,2}. In the aerobic oxidation of amines to aldehydes and hydrogen peroxide catalysed by this enzyme, the TOPA quinone is transiently reduced to a free radical intermediate which has been detected and studied by electron spin resonance techniques³. The mechanism appears to involve adduct formation between the amine substrate and TOPA quinone cofactor of the enzyme, followed by reduction of the Cu(II) ion to the cupric state and formation of a semiiminoquinone radical². This mechanism of TOPA quinone is summarised in Scheme 1. However the actual substitution positions of nitrogen and oxygen atoms in the ring are unknown. Accordingly, the precise structure of the radical species is also uncertain.



The most recent investigation of the structure of the radical using electron spin resonance techniques suggest a radical dianion structure with only the nitrogen atom carrying a covalently bonded hydrogen atom⁴. The formation and decay of the radical and other intermediates have been observed by stopped flow techniques with visible absorption spectroscopy⁵. The radical site in several copper amine oxidases is stabilised in the presence of cyanide³. In the pea seedling amine oxidase (PSAO) addition of amine substrate in a closed cuvette leads to rapid anaerobiosis and formation of the radical absorption (λ_{max} 464 nm)⁶ which we have found to be stable for several hours. We have therefore been able to study the radical in PSAO by resonance Raman spectroscopy. In addition, we are developing "caged" amines which will release amine substrates on photolysis and which will permit time-resolved spectroscopic studies of the catalytic cycle.

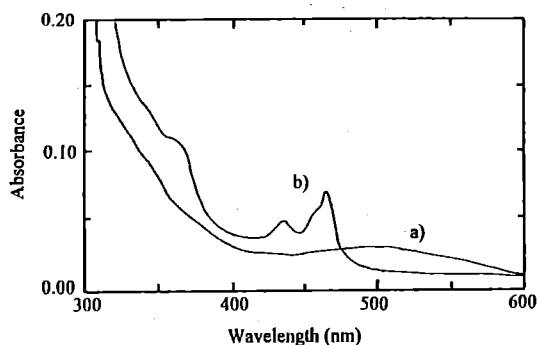
RESULTS

Resonance Raman spectrum of the free radical in PSAO

PSAO was isolated from sprouting pea seedlings using ammonium sulphate precipitation, ion exchange and gel chromatography⁶. The isolated protein was pink due to the visible absorption of the TOPA quinone cofactor (λ_{max} 500 nm, Figure 1). On addition of benzylamine the solution turned green due to the formation of the radical with λ_{max} 464 nm. The cw resonance Raman spectrum of the radical at pH 7 was probed using the 457.9 nm line of the Coherent Innova 90 Ar³ laser, and is shown in Figure 2. The spectrum may be compared with those of radicals formed on one-electron oxidation of hydroquinone, 4-aminophenol and 1,2,4-benzenetriol reported by Tripathi, Schuler and coworkers⁷. The resonance Raman spectrum of

the PSAO radical contains a strong band at 1649 cm⁻¹ assigned to the Wilson ν_{10} C=C ring stretching vibration. A medium intensity band at 1490 cm⁻¹ is similarly assigned by comparison with the

FIGURE 1 -- Absorption spectra of pea seedling amine oxidase before (a) and after (b) addition of benzylamine.



other species as being associated with the Wilson ν_{10} C-O (C-N) stretching mode. The spectrum contains several weaker bands including one at about 1220 cm⁻¹ attributed to the ν_{10} C-H bending mode. The relative intensity of these two strongest bands and the known pK_a values of the 1,2,4-benzenetriol radicals suggest that the radical species observed in PSAO is the radical monoanion. The principle bands in the 1,2,4-benzenetriol radical monoanion have been reported to occur at 1625 cm⁻¹ and 1530 cm⁻¹. These two bands are more separated in the PSAO radical spectrum with the frequency of the ν_{10} C=C stretching mode being higher also than in the p-benzosemiquinone radical (1620 cm⁻¹) and more similar to that in the p-aminophenol radical (1636 cm⁻¹). The PSAO spectrum therefore strongly supports the identity of the radical as a semi-iminoquinone rather than as a semiquinone. The wider

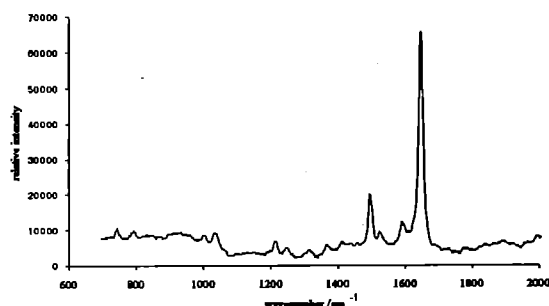
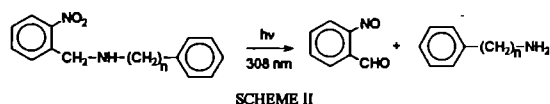


FIGURE 2 -- Resonance Raman spectrum of the radical site in pea seedling amine oxidase obtained on excitation at 457.9 nm.

separation of the two major bands is consistent with the lower electronegativity of nitrogen compared with oxygen, resulting in a weaker C-X (X=O, N) bond and higher electron density in the benzene ring of the semi-iminoquinone. Consequently in the semi-iminoquinone the weaker C-X bond has a band shifted to lower frequency whilst the stronger ring C=C bonds have a band at higher frequency.

Release of amine substrates from "caged" adducts.

"Caged" compounds have now been quite widely developed⁴ with the nitrophenyl group as one of the most commonly employed caging groups. Adducts of benzylamine and phenethylamine with 2-nitrobenzaldehyde have been synthesised. These have been shown to release the corresponding amines on photolysis with a 308 nm pulse from an excimer laser (scheme II).



Spectra from laser flash photolysis of aqueous solutions of caged phenethylamine are shown in Figure 3. The increase in absorbance between 300 - 420 nm in the spectrum measured 36 μ s after the laser pulse is ascribed to the *aci*-nitro intermediate. The spectrum at 300 μ s is identical to that of the observed permanent 2-nitrosobenzaldehyde product. Decay of the *aci*-nitro intermediate is usually taken to indicate the rate of uncaging of the substrate. At 25°C this took place with rate constants of $6 \times 10^3 \text{ s}^{-1}$ ($\tau = 160 \mu\text{s}$) and $4 \times 10^3 \text{ s}^{-1}$ ($\tau = 250 \mu\text{s}$) for caged phenethylamine and benzylamine respectively. The Arrhenius plots in Figure 4 show an activation energy of $40 \pm 2 \text{ kJ mol}^{-1}$ in each case. Further work is under way synthesise a wider range of caged amines and to measure quantum yields for photochemical uncaging.

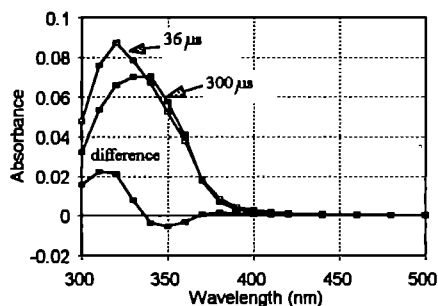


FIGURE 3 – Spectra from laser flash photolysis (308 nm) of caged phenethylamine at pH 7 and 45°C.

This work shows that photochemical uncaging of amine substrates provides superior time resolution to conventional stopped-flow mixing techniques and will permit time-resolved spectroscopy of transient intermediates in the catalytic cycle of amine oxidases using pump-probe techniques.

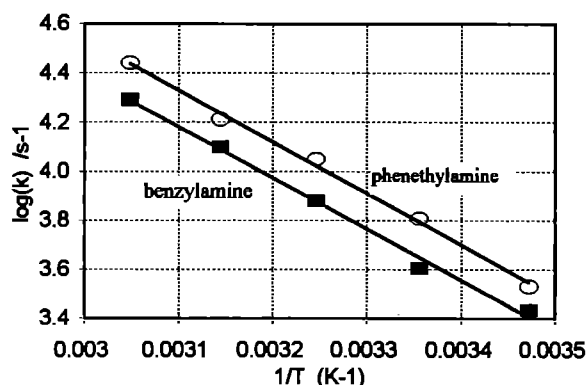


FIGURE 4 – Arrhenius plots for decay of the *aci*-nitro intermediate after laser flash photolysis of caged benzylamine and phenethylamine.

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THE KINETICS AND MECHANISMS OF REPAIR OF UV AND SOFT X-RAY INDUCED DNA DAMAGE IN MAMMALIAN CELLS

R. Meldrum,¹ S. Ward,¹ E. Turcu,² C. Wharton¹

¹School of Biochemistry, University of Birmingham, Edgbaston, Birmingham B15 2TT

²Central Laser Facility, Rutherford Appleton Laboratory, Chilton, Didcot

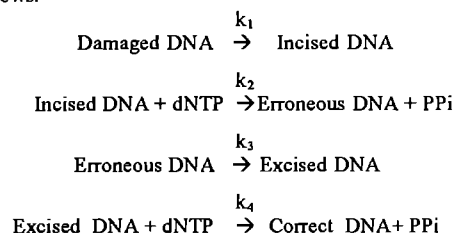
INTRODUCTION

Time resolved measurements of DNA repair synthesis following 248 nm UV laser induced DNA damage in HL60 cells showed an unexpected transient in the first 60 seconds after damage and this was followed by a more steady state of DNA synthesis (1). Use of inhibitors of DNA polymerase indicated that a polymerase with an associated nuclease was active in the first few seconds following DNA damage by UV laser irradiation and that newly incorporated bases were being rapidly excised.

VERIFICATION OF THE EXCISION OF NEWLY INCORPORATED BASES

To test for excision of the newly incorporated bases, a kinetic model of the reactions was derived using a computer modelling program.

The reaction scheme which describes the profiles of DNA synthesis in the first 120 seconds following UV damage to DNA is as follows.



Gamma 33p-TTP (Amersham, custom made) and $\alpha^{32}\text{P}$ -TTP (or ^3H -TTP) were introduced into HL60 cells and kinetic experiments carried out as described in reference 2 and previous CLF annual reports. In this case an EDTA solution in buffer was used as a quench rather than sodium hydroxide. The cells were washed and the DNA was extracted with TCA. The remaining 'cytosol' fraction was analysed by HPLC for radiolabelled PPi and TMP. This was repeated in order to obtain sets of data on several populations of cells. Figure 1 shows experimental data projected upon two computer derived models

The time profiles of the synthesis of DNA following damage are repeatable on the same cell population but if different populations are used (ie. experiments performed on different days) the magnitude and acuteness of the transient may vary. The general forms of the time profiles, however, remain consistent. Relative rate constants for the reaction scheme are estimated from the experimental data for the incorporation of radiolabel into DNA. The corresponding rate constants for the two models are shown. Both profiles show a reasonable fit of the experimental points of TMP to the predicted profiles of release and this provides evidence of excision of newly incorporated nucleotides. The radiolabelled PPi release also corresponds well to the predicted release during the transient and steady state phases of DNA synthesis.

Experiments were performed where the radiolabelled nucleotide was prevented from incorporating in the DNA by the caging group (α -1-(2-nitrophenyl)ethyl ester) for 30 seconds after damage, whence it was photoactivated (by a 351 nm UV laser pulse) and allowed to incorporate in the cellular DNA for various times thereafter. These time profiles began with a distinct lag phase followed by a steady state phase of DNA synthesis. When projected with the time profiles which begin 5 seconds after damage, the lag phase coincides with the period in which excision

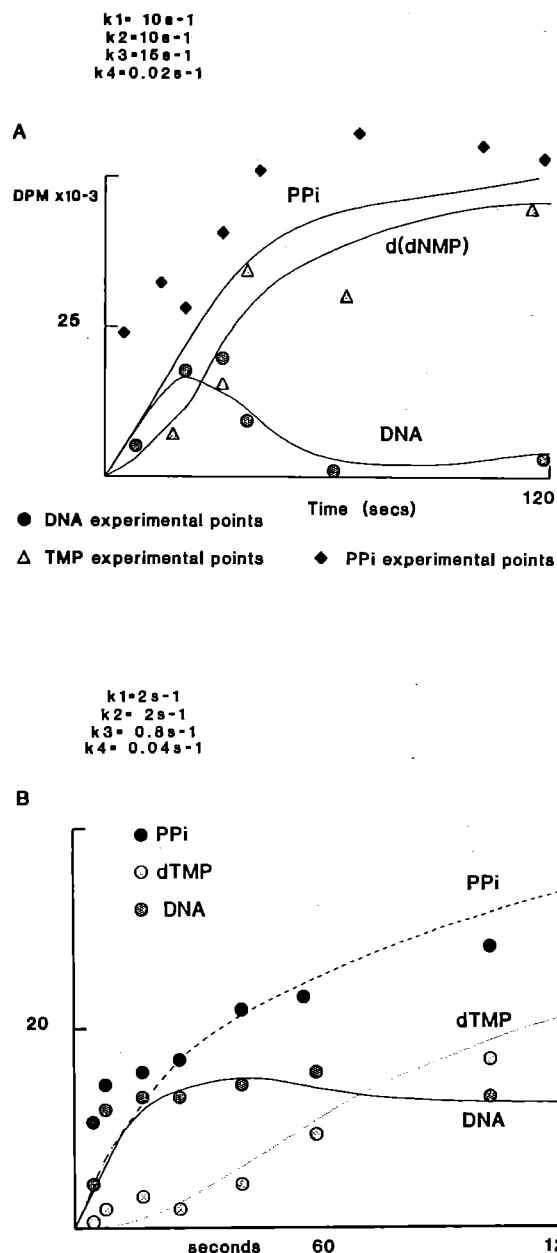


Figure 1

of nucleotides appears to take place and the onset of the steady state of DNA synthesis is also matched in the two different time courses. These 'staggered time courses' can therefore form a very useful method of verifying and extending the analysis of the nature of enzyme activity in the cells at different times.

KINETICS OF REPAIR OF X-RAY DAMAGE TO CELLULAR DNA.

Transient incorporation of radiolabelled nucleotide into the DNA of V79 fibroblast cells is also seen following damage by 1.2KeV Cu L-shell soft x-rays. The cells used in these experiments are grown as monolayers on mylar membranes and the monolayers are electroporated by a series of platinum electrodes (3). This common transient response to x-ray and 248 nm UV laser induced DNA damage suggests that it is a repair response to a common form of damage. We have found that DNA breaks may be induced by biphotonic absorption of 248 nm UV laser irradiation and these breaks are likely to be similar to a component of x-ray damage (4).

The kinetic profiles of repair of x-ray damage are generally more complex than those of 248 nm UV laser induced damage and computer modelling of these is more difficult. The time resolved studies of early repair of x-ray damage to cell DNA have, however, revealed two significant findings. The first of these is that bases which have been newly incorporated at replication sites are almost immediately excised when damage to the DNA is inflicted by x-rays (ie loss of bases incorporated by normal DNA replication is seen as early as 1 second following x-ray damage). Secondly, early DNA repair activity is greatly stimulated in cells which have received a non-lethal dose of irradiation 2 hours previous to a second non-lethal or lethal dose. The functions and kinetics of synthesis of proteins which are involved in the regulation of DNA repair are being studied in relation to these findings of the DNA repair kinetics. The synthesis of poly(ADP-ribose) from NAD is an immediate stress response in mammalian cells exposed to ionising radiation (5). The function of poly(ADP-ribose) synthesis is particularly accessible to study in relation to the fast kinetics of DNA repair since the poly(ADP-ribose) polymerase may be labelled with ³²P-NAD in synchrony with ³H-TTP labelling of the DNA and co-precipitated with TCA. Synthesis of poly(ADP-ribose) in the early stages following 248 nm UV laser induced DNA damage has been detected in this manner and this adds to the evidence that x-ray like damage may be induced by the 248 nm laser irradiation

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THE EFFECTS OF FREE RADICAL DAMAGE ON THE PHOTOACTIVATION OF RHODOPSIN AND BACTERIORHODOPSIN

R. Withnall¹, M. Goss-Sampson¹, R.W. Sparrow² and P. Matousek³.

¹School of Biological and Chemical Sciences, University of Greenwich, London SE18 6PF.

²Division of Life Sciences, Bromley College of F & HE, Bromley Kent BR2 8HE.

³Central Laser Facility, Rutherford Appleton Laboratory, Chilton Didcot, Oxford OX11 0QX.

INTRODUCTION

Rhodopsin consists of 11-cis-retinal bound to the protein opsin (41kDa) present in mammalian photoreceptors. Following photoabsorption by rhodopsin the 11-cis-retinal isomerises to the all trans form. Within ~ 10 ps further conformational changes occur eventually forming the activated state (Metarhodopsin II) where the retinal and the opsin dissociate and visual transduction is initiated.

Bacteriorhodopsin is composed of an all-trans retinal (vitamin A) attached to a 26kDa protein. The chromophore isomerises to the 13-cis state following photoactivation. This system undergoes a cyclical series of reactions returning to its initial state after several ms. The reactions of particular interest were the formation of components K, within 3 ps, KL, within 4 ns, and then L within 1.5 μ s.

Recently free radicals have been implicated in the aetiology of visual abnormalities. This preliminary study has been carried out in order to determine whether TR³ can be used to monitor the effects of free radical damage on these photochemical systems and their photoproducts.

The primary aim of this study was to determine the extent to which free radical damage affects the photochemistry of rhodopsin and to study the possible sites of damage. This approach may provide an insight into the role played by oxidative processes in abnormal visual transduction.

MATERIALS AND METHODS.

Bacteriorhodopsin (Sigma) was reconstituted in water, and regenerated bovine rhodopsin was isolated from fresh cow eyes and stored in phosphate buffer pH 7.4. All samples were kept in the dark on ice prior to analysis. In the TR³ experiments, samples (10 cm³ at 1-2 absorbance units) were spun in a rotating solution cell at a speed of approximately 100 revolutions per minute.

The TR³ spectroscopy was performed at the Laser Support Facility, Rutherford Appleton Laboratory on a picosecond TR³ set-up. The following experiments were conducted

- Pump and probe at the same wavelength covering 550-570 nm and collecting the resonance Raman scattering in the Stokes region.
- Pump/Probe at 570nm and observing resonance Raman scattering in the anti-Stokes region 520-565 nm.
- Pump/probe both at a wavelength of 511nm using the Cu vapour laser only by-passing the amplification cell, and then attenuating the beam.
- The cavity dumped laser was run at different pulse repetition rates to obtain data on transient species with lifetimes in the ns- μ s range.

Hydroxyl radicals were produced in nitrogen purged samples by irradiation with a ⁶⁰Co source at the MRC Laboratory Harwell. Exposure times were calculated to give doses of 800,1200,1800 and 3600 Gy. The extent of the damage caused was measured by recording an optical spectrum (250-800 nm) before and after irradiation.

RESULTS

For bacteriorhodopsin, following high and then low pump intensities subtraction revealed the presence of transient species having lifetimes in different time domains. Three species were detected; one with a lifetime in the order of psec, another with a lifetime of nsec and the third with a lifetime of μ sec (as shown by antistokes data in Fig. 1). These species seem to correspond with K, KL and L as previously described.

Attempts were made to obtain Raman spectra with bovine rhodopsin. However it was not possible to get a clean signal from our rhodopsin samples due to a combination of possible factors.

- Using this laser configuration, 550nm is not optimal for rhodopsin excitation.
- A high level of fluorescence was emitted from our sample.
- There was insufficient volume of sample available. Bovine rhodopsin is not self regenerating and thus was rapidly being bleached on exposure to the laser

Optical spectra are shown before and after hydroxyl free radical damage of bacteriorhodopsin and bovine rhodopsin. There were obvious differences at both the absorption maximum for each of the photopigments (550 and 490 nm respectively) and also in the protein absorbing region at around 280 nm corresponding to damage of aromatic amino acid residues.

SUMMARY

The data indicate the presence of transient species in bacteriorhodopsin, one with a lifetime in the order of psec (K), another with a lifetime of ns (KL) and a third with a long lived lifetime of μ s (L). Further experiments are to be carried out in order to determine whether the spectral changes associated with hydroxyl radical damage are related to changes in bacteriorhodopsin or its transients.

For future TR³ spectroscopic studies using bovine rhodopsin, it would be of interest to probe the protein using UV excitation in the region of 280 nm. This would provide detailed information on the effects of hydroxyl radical damage on the opsin protein structure. It would also be beneficial to be able to carry out Raman studies in conjunction with pulse radiolysis.

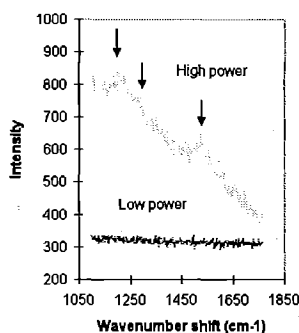


Fig 1. Antistokes Raman spectrum using an excitation wavelength of 570 nm

We would like to acknowledge the invaluable help of Dr P O'Neill of the MRC Radiobiology unit, Harwell.

STUDIES OF DAMAGE TO PLASMID DNA USING 248nm LASER PULSES

K.M. Prise¹, B.D. Michael¹, R.A. Meldrum² and C.W. Wharton²

¹CRC Gray Laboratory, PO Box 100, Mount Vernon Hospital, Northwood, Middlesex HA6 2JR,

²School of Biochemistry, University of Birmingham, Birmingham, B15 2TT

INTRODUCTION

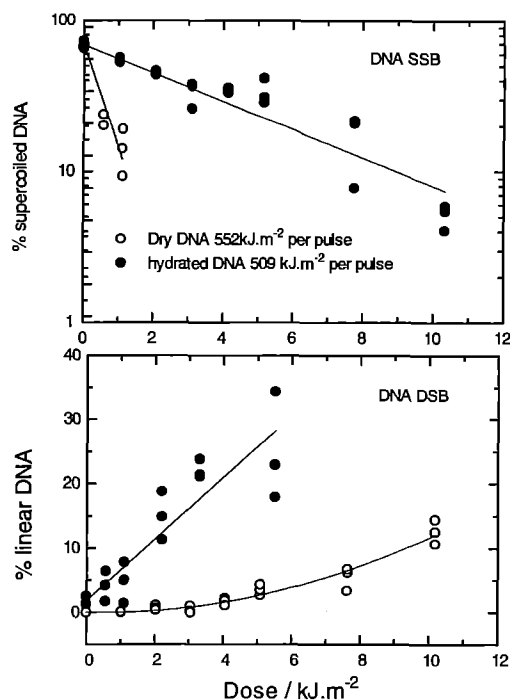
Understanding the effectiveness of radiation tracks as they cross cellular DNA requires a fundamental understanding of the distribution and size of energy events on the DNA and action spectra for lesion production. There is little good experimental data for the energy dependence of strand breakage on DNA but recent studies using VUV from the Daresbury Synchrotron have provided action spectra for dry plasmid DNA systems (1,2). These studies have shown that previous estimates of the threshold energies for DNA single-strand breaks (ssb) and double-strand breaks (dsb) production of 17.5eV and 50eV respectively need to be revised as dsb are produced efficiently down to energies of less than 10eV and ssb to energies below 7eV. Work with synchrotron light is problematic due to low photon fluxes and the need to work under vacuum for most studies to attain appreciable exposure of samples. An alternative approach at the low energy region of <7eV is to use high intensity pulsed laser sources. These allow measurement of the effectiveness of low energy photons in both hydrated DNA systems and even in cellular systems (3). In this study we have carried out preliminary measurements of the effectiveness of 248nm (5eV) laser light at inducing ssb and dsb in plasmid DNA under conditions where the DNA was dried under vacuum or fully hydrated in solution.

MATERIALS AND METHODS

Plasmid pMSG-CAT (8300bp) purified from *E.coli* HB101 was resuspended at a concentration of 0.25µg/µl in 0.068mM EDTA (pH7.0). For hydrated studies 1µl of plasmid solution was spread over a 5 x 5mm area on a glass coverslip which had been precoated with a 1:5000 dilution of Tween detergent. For evacuated samples the glass coverslip was placed in a vacuum chamber consisting of a quartz window and evacuated to 10⁻³mbar prior to irradiation. After irradiation samples were rehydrated in TE buffer and subjected to electrophoresis on 1% (w/v) agarose gels at 16 volts for 16 hours. DNA was stained with ethidium bromide and analysed using a PC-based image capture system. The fractions of relaxed linear and supercoiled plasmid DNA were quantified and DNA ssb and DNA dsb yields calculated.

RESULTS AND DISCUSSION

Figure 1 shows a comparison of DNA ssb and dsb yields for samples of plasmid DNA irradiated under fully hydrated or under vacuum with similar intensity laser pulses. For DNA ssb production, measured as a loss of intact supercoiled plasmid DNA moving from dry to hydrated DNA leads to a 6-fold reduction in the yield of ssb per pulse (upper panel). A similar reduction in DNA damage is also observed with the induction of dsb (lower panel). Also, the yield of dsb changes from a linear response with dry DNA to a quadratic response in the hydrated system. This is indicative of a change in the mechanism of dsb formation from one of direct production of dsb from 248nm laser light to dsb production via two ssb's being formed close together. This indicates an important role for the presence of water in the energy transfer mechanisms involved in damage induction at these energies. Further studies are now continuing with a view to understanding the relationship between laser intensity and damage efficiency under conditions of dry or hydrated DNA.



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TIME RESOLVED FLUORESCENCE MEASUREMENTS OF FIREFLY LUCIFERIN

N S Parkinson¹, R S Chittock², N Berovic¹, M Towrie³, P Matousek³, C W Wharton², J B Jackson², T D Beynon¹

¹ School of Physics and Space Research, University of Birmingham, Edgbaston, Birmingham B15 2TT

² School of Biochemistry, University of Birmingham, Edgbaston, Birmingham B15 2TT

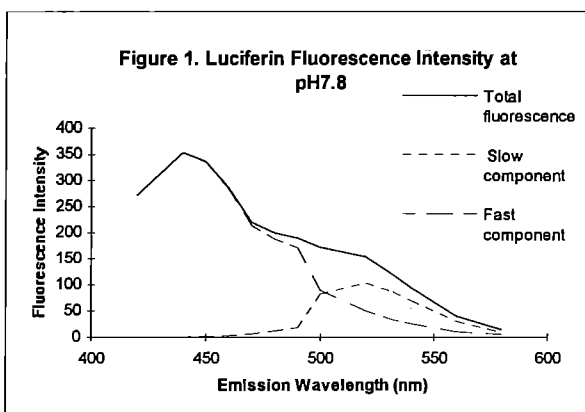
³ Rutherford Appleton Laboratory, Chilton, Didcot, OXON, OX11 0QX

INTRODUCTION

Firefly bioluminescence is produced by catalysis of the oxidation of the substrate luciferin to an excited state of the product oxyluciferin by the enzyme luciferase, in the presence of ATP and O₂. Light emission occurs as the product relaxes to its ground state by emitting a photon. To explain the bioluminescence mechanism a knowledge of the electronic states of luciferin will be necessary. The LSF time-resolved single photon counting apparatus was used to characterise the fluorescence component amplitudes and lifetimes of luciferin and several of its analogs excited at a wavelength of 330nm, across a range of emission wavelengths in a variety of conditions. This continues on from preliminary work¹ which showed several deviations from the predictions of the luciferin fluorescence model proposed by Morton².

RESULTS AND DISCUSSION

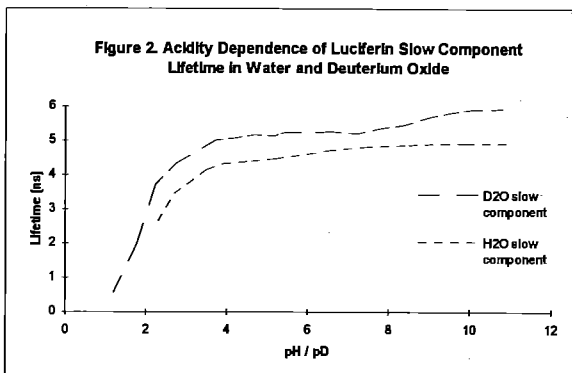
As predicted by Morton, luciferin fluorescence at pH10.4 was observed to be mono-exponential. This emission from the phenolate form of luciferin peaks at 510nm, with a characteristic lifetime of 4.9ns which is independent of emission wavelength.



Fluorescence of phenol luciferin is bi-exponential at pH7.8, with a fast component of lifetime of approximately 120 ps at its peak of 435nm, and a slower component with a lifetime of 4.8ns peaking at 510nm (Fig.1). Only the lifetime of the faster component varied with emission wavelength, increasing to an apparent value of 350ps at 510nm. A similar variation of fast component lifetime with wavelength was observed at all acidic pH values examined. The 4.8ns component is assigned to fluorescence from the excited phenolate state, as it has been estimated that the excited state phenol → phenolate pK* = -0.5, compared to pK = 8.6 for the ground state³. In addition the peak wavelength and spectral distribution of this component remains constant across the range pH 2-12, with only the magnitude of the lifetime changing (Fig.2). The faster component is attributed to the phenol form. This is in conflict with Morton's model, which predicts the phenol lifetime should be slower than the phenolate.

The fluorescence of luciferin remains bi-exponential down to approximately pH4, at which point the fluorescence starts to become inadequately described by sums of exponentials. The best fit exponential for the lifetime of the phenolate component was also observed to sharply decrease in this region (Fig.2). This is attributed to self-quenching of the phenolate form by its geminate

proton at low pH in a manner analogous to that described for 1-naphthol⁴, leading to a non-exponential decay.



The fast component lifetime, as measured at 435nm, was observed to remain constant within the limits of experimental error. Examination of luciferin fluorescence in D₂O shows similar wavelength and pD dependence as determined for H₂O (Fig.2). The lifetime of the slow component was observed to increase by a factor of 1.18.

Luciferin fluorescence in solvents of ethanol and in 98% dioxane:2% H₂O has also been investigated. In ethanol the observed lifetime was a bi-exponential and faster than in aqueous solution. Results of the dioxane solution show that, as has been implied by the theoretical calculations of Wada⁵, reducing the relative permittivity of the solvent increases the phenolate lifetime (to 5.3 ns in alkali conditions) by stabilising the excited state. In addition the fluorescence characteristics of luciferin analogs cyano 6-methoxy-benzothiazole, methoxy-luciferin and cyano 6-hydroxy-benzathiazole (CHBT) were examined in ethanol, H₂O and D₂O over a range of acidities. CHBT had very similar lifetimes, pH and solvent dependencies to luciferin, whilst the fluorescence characteristics of the other analogs was dissimilar. This implies that the fluorescence characteristics of luciferin are predominantly determined by the hydroxy-benzathiazole group.

Ongoing analysis is examining the fitting of a grow-in time for the acidic phenolate fluorescence, to account for the phenol → phenolate proton loss lifetime. It is believed this may account for the apparent variation of the fast component lifetime with wavelength. Also the non-exponential decays observed at low pH will be fitted, thus examining the self-quenching mechanism. It is planned this data will then be used to form a more complete model of luciferin fluorescence than currently exists.

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F.M.Qureshi¹, J.R.G.Thorne², D.D.C.Bradley² and H.L.Anderson³

²Dept Chemistry, Univ. Manchester, Manchester M13 9PL

³Dyson Perrins Laboratory, Oxford University

It is becoming increasingly clear that successful materials for third order optical applications must have intense near infra red absorption to enhance the response at the frequency of the preferred telecommunications window at $1.5\mu\text{m}$. Knowledge of the dynamics of the real excitations that cause NLO enhancement is vital to an understanding of the speed of device response. Understanding of the spectroscopy of excited states, is important to the minimisation of device losses.

$$\text{Me}_3\text{Si}-\equiv \left[\begin{array}{c} \text{R} \\ | \\ \diagdown \quad \diagup \\ \text{C}=\text{C} \\ | \qquad | \\ \text{N} \quad \text{N} \\ / \quad \backslash \\ \text{C}=\text{C} \\ | \qquad | \\ \text{N} \quad \text{N} \\ \diagup \quad \diagdown \\ \text{C}=\text{C} \\ | \\ \text{R} \end{array} \right]_n -\equiv \text{SiMe}_3$$

$\text{R} = \text{CH}_2\text{CH}_2\text{C}(=\text{O})\text{QCH}_2(\text{CH}_2)_6\text{CH}_3$

Femtosecond pump excitation with ~100fs pulses at 740 nm excited the Q-band of both dimer and polymer samples.(Figure 2)

Steady state fluorescence spectra indicate that in transient absorption experiments we are observing excited state dynamics within the excited singlet manifold. There is no evidence for any triplets or charge transfer states in these materials. The lifetimes are ~ 1.5 ns for the monomer, ~ 800 ps for the dimer and ~ 400 ps for the polymer in solution[3]. The small Stokes shift in the dimer and the absence of vibronic structure indicates weak electron-phonon coupling. There is little phonon sideband

phonon coupling. There is little phonon sideband structure in absorption and emission in dimer and polymer.

We observe excited state bleach lifetimes as shown in Figure 3.

The dimer has a lifetime of ~ 800 ps in agreement with the excited state lifetime obtained from time correlated single photon counting fluorescence. In contrast, the polymer displays sub picosecond excitation dynamics, which are the subject of the remainder of this discussion.

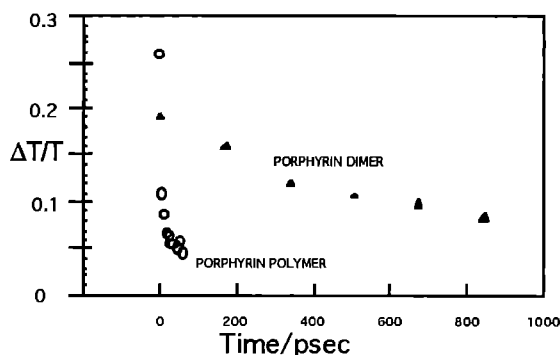


Figure 3 Transient bleaching kinetics

Photodecomposition of the polymer in solution leads to a shift of the Q band to the blue upon strong irradiance, from a maximum at ~ 850 nm to a maximum at ~ 800 nm. This photolysis did not occur to any appreciable extent during the transient experiments described here.

The bleaching maximum wavelength of the polymer in transient absorption shifts slowly to the red (by 15-20 nm) during the lifetime of the excited state. (Figure 5)

The fluorescence maximum of the polymer is Stokes shifted by 25-30 nm from the absorption maximum, whereas there is very little Stokes shift in the dimer.

All the above facts point to the polymer being inhomogeneous. Excitation of high energy states leads to a time dependent population of lower energy states which eventually emit. Strong irradiance destroys these lower energy states (perhaps by breaking long chains into shorter segments or partially destroying their conjugation).

ULTRAFAST DYNAMICS:

Figure 4 shows the femtosecond bleach recovery at 500 nm in the porphyrin polymer upon excitation at 740 nm. Upon excitation the polymer ground state is immediately bleached: this is seen both at Q and Soret absorption frequencies. On a subpicosecond timescale the excitation relaxes to emitting states near 850 nm. There is a rapid bleach recovery process at both Q and Soret bands occurring on a 500 fs timescale. This process appears laser power dependent which we believe to be evidence of exciton annihilation. Annihilation on a 500 fs timescale has been observed in phthalocyanine aggregates[4]. In our case, the timescale for the process appears to be the same in solution and thin film.

Excitation densities are large in these experiments estimated at about one photon for every three macrocycle chromophores at the highest levels used ($I=I_0$), although the transient strength was still linearly dependent upon laser power below this level.

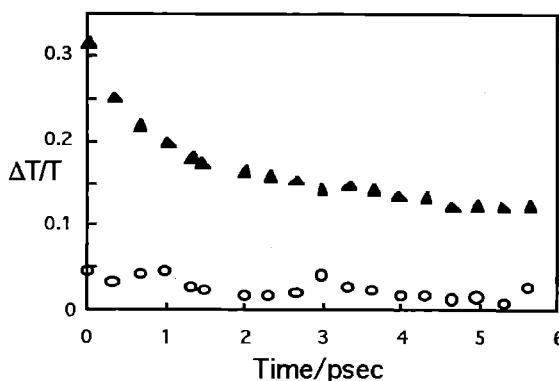


Figure 4 Laser Power dependence of transient bleach kinetics (Triangles $I=I_0$, circles $I=I_0/4$)

Figure 5 demonstrates that slower relaxation processes are also occurring in the polymer on a timescale of tens of picoseconds. This is most easily interpreted as the exciton transferring to the lowest energy states of a disordered polymer. This spectral diffusion process may well involve spatial diffusion of the excitation along the chain until it locates a chain segment of lowest energy.

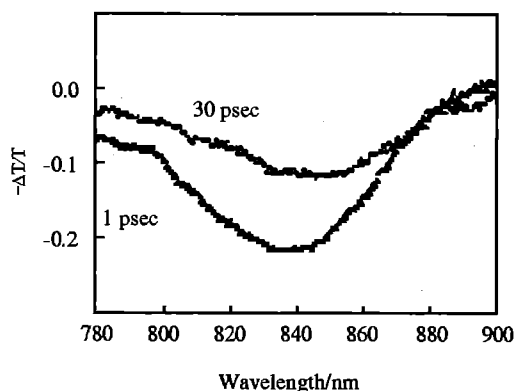


Figure 5: Polymer Spectral diffusion: bleach spectrum 1ps and 30ps after 740 nm excitation.

Excited state absorption at high laser powers must correspond to biexciton creation- that is a second excitation will almost certainly be in the immediate vicinity of the first on a single polymer chain.

Phase space filling appears to be occurring in the polymer at high laser intensities. In figure 6, we observe that excitation of the polymer at 740 nm leads to a bleach that is maximal at 850 nm after 1 ps at high intensity but is red shifted in comparison when we use lower laser power. It is clear in the former case that states absorbing near 800 nm remain bleached: in the latter case these states are able to relax to unpopulated lower energy levels.

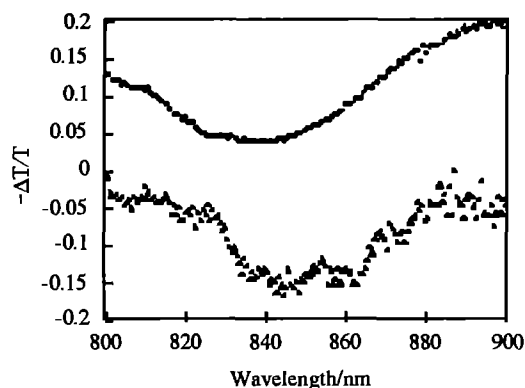


Figure 6: Phase space filling at high excitation densities. Polymer bleach spectrum at 1ps: upper trace $I=I_0$, lower trace $I=I_0/5$. Spectra displaced vertically for clarity.

CONCLUSIONS:

We have observed a wide variety of excitation dynamics in the novel edge linked porphyrin polymer in solution in the femtosecond and picosecond time regimes. Our experiments are consistent with a picture of highly mobile excitations moving along a disordered chain. Further experiments are in progress.

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ELECTRON TUNNELLING IN CdTe/CdMnTe DOUBLE BARRIER STRUCTURES

N Cain, M O'Neill, J E Nicholls, T Plorek, W E Hagston, A J Langley¹ and P F Taday¹

The University of Hull
¹ Rutherford Appleton Laboratory

INTRODUCTION

The quantum mechanical tunnelling of carriers through barriers in ultrathin semiconductor heterostructures is of fundamental interest and also of practical importance in devices. For example, the tunnelling of injected electrons from one well to another may be the principal factor which limits the switching speed of quantum well lasers. In recent years dynamical aspects of carrier tunnelling have been examined both experimentally¹⁾ and theoretically²⁾ in III-V GaAs/AlGaAs systems. We investigate the phenomenon in the II-VI CdTe/CdMnTe system using a pump-probe technique with femtosecond resolution. Our theoretical model treats the carrier transfer between wells as an intersubband relaxation process between two subbands each of which is localised in a different well.

EXPERIMENT

The CdTe/CdMnTe double barrier sample was grown by MBE on an InSb substrate. It contained a series of five double barriers of configuration 120 Å CdTe / 60 Å Cd_{0.96}Mn_{0.04}Te / 60 Å Cd_{0.89}Mn_{0.11}Te / 60 Å Cd_{0.96}Mn_{0.04}Te / 120 Å CdTe separated by 180 Å Cd_{0.89}Mn_{0.11}Te barriers. The sample was excited using amplified pulses of 300 fs duration from a mode-locked Titanium-sapphire laser. A spectral continuum generated by focusing a fraction of the pump beam onto a rotating glass slide was used as the probe beam. On transmission through the sample the probe was dispersed by a monochromator and detected using a CCD camera. The experiments were carried out at 10 K.

EXPERIMENTAL RESULTS

The sample was pumped at the energy of the exciton resonance of the Cd_{0.96}Mn_{0.04}Te well. The pump induced changes in transmission of the probe beam ($\Delta T/T$) were measured as a function of wavelength about the fundamental electron-heavy hole (e1-hh1) and electron-light hole (e1-lh1) exciton resonances of the CdTe wells and as a function of the delay between pump and probe.

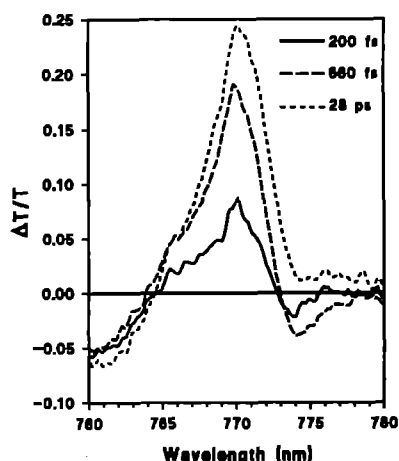


Figure 1

Spectrum of pump induced changes in transmission of probe beam for various delays. An excitation density of $1 \times 10^{13} \text{ cm}^{-2}$ was used

Figure 1 shows a set of typical differential transmission spectra obtained using an excitation density of $1 \times 10^{13} \text{ cm}^{-2}$ and for time delays of 200 fs, 660 fs and 28 ps. The spectra show exciton bleaching or a pump-induced increase of transmission about the e1-hh1 exciton transition at 770 nm and the e1-lh1 transition at 766 nm. At early times, an increase in absorption at longer wavelengths is also observed. The magnitude of exciton bleaching increases with time up to about 30 ps after the pump pulse and then decays with the characteristic life-time of the exciton. There are three different mechanisms which may contribute to the observed carrier-induced exciton nonlinearities. Phase space filling, the filling of states from which excitons are created, reduces the oscillator strength of the exciton resonance and hence increases its transmission. At high carrier densities, exchange interactions renormalise the single carrier and exciton states and so reduce the oscillator strength of the exciton. Exchange interactions may also give a carrier density dependent shift in the energy of the exciton transition. Such an energy shift is not observed in our experiments but this may be due to insufficient spectral resolution. Coulombic screening may also lead to bandgap renormalisation but it is a long range interaction with a small effect in quantum well systems³⁾. At high carrier densities the Coulombic interaction may also broaden the exciton absorption line because of a reduction in the coherence time due to carrier-carrier scattering. Calculations are in progress to verify whether this mechanism is responsible for the observed photo-induced increase of absorption at the low energy side of the exciton peak.

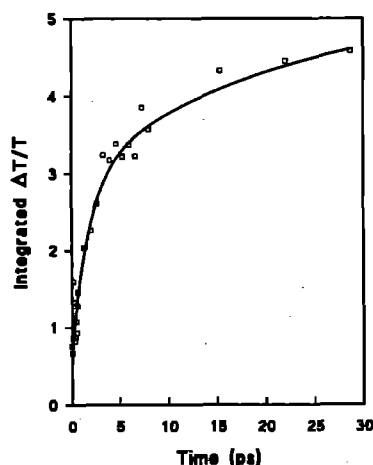


Figure 2

Time dependence of spectrally integrated pump induced changes in transmission of probe beam for a carrier density of $1 \times 10^{13} \text{ cm}^{-2}$.

The reduction of oscillator strength (ROS) of the exciton may be measured independently of exciton line broadening by integrating the photo-induced transmission changes over the spectral width of the exciton transition⁴⁾. Changes in the ROS can be used as a measure of population changes of the e1 and h1 subbands of the CdTe well since, as will be shown below, the contribution to the ROS by carrier cooling is negligible. Figure 2 shows the temporal evolution of the ROS obtained via integrated $\Delta T/T$ for a carrier density of $1 \times 10^{13} \text{ cm}^{-2}$. A large ROS immediately after the pump pulse is followed by a slower change. The sample was pumped at the energy of the exciton resonance of the Cd_{0.96}Mn_{0.04}Te well. By inspection of the linear absorption spectrum of the sample we find that 40% of carriers are

injected into the Cd_{0.96}Mn_{0.04}Te well and 60% into the CdTe wells. Taking the injected carrier distribution into account, the time dependence of the ROS is fitted to the sum of two exponentials. Rise times of 300 fs and 8 ps are found. Previous experiments show that carrier relaxation in a well occurs within a timescale of 300 fs⁵. The longer time is then a measure of the tunnelling time from the Cd_{0.96}Mn_{0.04}Te well to the CdTe wells.

THEORY

A Monte-Carlo method was used to simulate electron tunnelling between the wells and electron relaxation in the wells of the double barrier system. Figure 3 shows the energy profile of the conduction band and the envelope wavefunctions of the three lowest electron subbands. Subbands e1 and e2 are primarily localised in the CdTe wells whereas subband e3 is localised in the Cd_{0.96}Mn_{0.04}Te well.

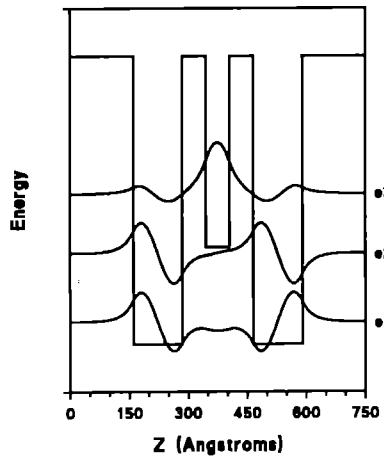


Figure 3

Energy profile of the conduction band showing the Cd_{0.96}Mn_{0.04}Te well separated by 60 Å barriers from the two CdTe wells. The envelope wavefunctions of the three lowest electron subbands are also shown.

In the model the scattering of 2-dimensional confined electrons with 3-D CdTe like longitudinal optical (LO) phonons is calculated using Fermi's golden rule. The scattering rate of an electron of energy E and momentum in the xy plane $p_{\parallel}$ into any other possible state of energy E' and momentum $p_{\parallel}'$ is given by

$$\Gamma(p_{\parallel}) = Y \sum_{\beta, p_{\parallel}'} \frac{1}{\beta^2} \delta(p_{\parallel}' - p_{\parallel} + \hbar\beta) \delta(E' - E + \hbar\omega_{\beta}) |G(\beta_z)|^2 \quad (1)$$

where β , with $\beta^2 = \beta_{\parallel}^2 + \beta_z^2$, is the wave vector of the phonon and ω_{β} its energy. The δ functions in equation (1) represent momentum and energy conservation respectively. Y is a constant describing the Frohlich interaction and the form factor

$$G(\beta_z) = \int_0^L \exp(\pm i\beta_z z) \psi \psi' dz \quad (2)$$

is an overlap integral of the phonon wavefunction and the wavefunctions ψ and ψ' of the initial and final states of the electron. Using equation (1) the probabilities of various transitions between the e3 and e1 or e2 subbands can be evaluated. Taking into account these probabilities random numbers are used to determine which route the process will take. The simulation is repeated a number of times so that the average scattering rate is a good approximation of the behaviour of the real system. We found that 80% of electrons excited

in the e3 subband transferred to the e1 or e2 subbands by means of LO phonon emission within 10 ps. Intracubband relaxation and intersubband transfer between the e1 and e2 subbands both occur by means of LO phonon emission within a time of 200 fs. These numbers compare well with the measured tunnelling time of 8 ps and relaxation time of < 300 fs.

Electron-acoustic phonon scattering was also calculated to consider the timescale of carrier cooling to the bottom of the e1 subband. the transition rate for acoustic deformation potential is given by

$$\Gamma(p_{\parallel}) = \kappa \sum_{\beta, p_{\parallel}'} \beta \delta(p_{\parallel}' - p_{\parallel} + \hbar\beta) \delta(E' - E + \hbar\omega_{\beta}) |G(\beta_z)|^2 \quad (3)$$

where κ is a constant including the acoustic deformation potential and β the phonon wavevector is related to its energy with $\omega_{\beta} = v_s \beta$ where v_s is the velocity of sound in the solid. Elastic scattering is assumed to simplify the integration and the Monte-Carlo method calculates an average intracubband relaxation time of 120 ps.

The good agreement between experimental results and the numerical simulations involving LO phonons suggests that tunnelling between the Cd_{0.96}Mn_{0.04}Te well and the CdTe wells occurs by means of electron-LO phonon scattering. The time evolution of the ROS is not affected by carrier cooling via acoustic phonon scattering. This is not surprising because of the high carrier densities used in the experiment. However other scattering mechanisms must also be accounted for. The contribution of carrier-carrier scattering and hole-LO phonon scattering to the observed tunnelling process is currently being investigated. At low carrier densities the tunnelling rate of heavy holes is significantly smaller than that of electrons because of the greater mass of the former. High carrier densities were used in the experiment so that heavy-hole light-hole mixing, which enhances the hole tunnelling rate, is expected.

CONCLUSIONS

Nonlinear changes in the absorption spectrum of a CdTe/CdMnTe double barrier system are used to investigate carrier tunnelling between the central Cd_{0.96}Mn_{0.04}Te well and the outer CdTe wells. A tunnelling time of 8 ps is measured when an injected carrier density of $1 \times 10^{13} \text{ cm}^{-2}$ is used. Monte Carlo simulations show that the timescale of tunnelling via electron-LO phonon scattering is 10 ps. Intracubband relaxation with the emission of LO phonons is calculated to occur in 200 fs in agreement with experimental results.

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State-Specific Electron Diffraction

M.A.D. Fluendy, R.B. Fender, G. Maitland and D.W.H. Rankin

Department of Chemistry, University of Edinburgh

INTRODUCTION

Electron diffraction has long been an important tool for determining the structure of relatively complex molecules containing up to approximately fifty atoms. Past studies, however, have been confined to neutral molecules with vapour pressures (> 1 Torr), where the resulting structural data is averaged over the ensemble of quantum states populated at the relatively high temperatures required to achieve the necessary vapour pressure.

A new electron diffraction apparatus has been constructed which takes full advantage of several new experimental techniques.

(a) A telefocus electron gun which enables very intense and low divergence electron beams (close to the space charge limit) to be produced.

(b) A cooled molecular target beam formed by a high Mach number ($M > 80$) supersonic expansion from a source at stagnation temperatures yielding the required vapour pressure. This technique produces very high densities in the molecule-electron collision zone (up to 10^{20} molecules m^{-3} and with a relative translational temperature < 1 K. The rotational and internal molecular modes are also very substantially cooled.

(c) A position sensitive detection system, based on hot microchannel plates, capable of counting individual electrons at a rate up to 10 MHz in each counting channel. The present detector has an active diameter of 75 mm with 127 discrete channels [1,2,3,4]

These techniques when combined should yield an improvement in sensitivity over conventional electron diffraction methods of approximately 10^4 . The apparatus constructed is shown in figure 1 and incorporates several stages of differential pumping for the electron gun (a), the nozzle expansion chamber (b), the detector (c), collision zone and flight path (d) and the beam dump and mass spectrometer monitor (e).

EDINBURGH ELECTRON DIFFRACTION APPARATUS

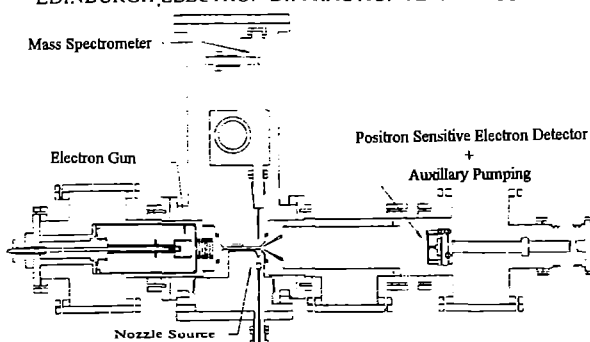


Figure 1 Schematic of electron diffraction apparatus

OBJECTIVES

The immediate objectives of this project were:

(a) To optimise and fully characterise the supersonic molecular beam with particular reference to the rotational and internal cooling and the influence of the detailed expansion conditions on the achievable distribution of quantum states.

(b) To design, construct and install suitable optics, signal collection and processing systems to allow internal state distributions in the fully expanded molecular beam to be explored and optimised. This system was also expected to provide the prototype for use (with a suitable laser system) in actual state specific electron diffraction measurements on optically pumped target beams.

(c) To explore the feasibility of studying positive ion structures formed by REMPI.

DEVELOPMENT OF OPTICAL & SIGNAL COLLECTION SYSTEMS

As indicated it is intended to develop this apparatus so as to permit transient electronically excited species to be studied. These species will be produced by laser excitation of the fully expanded and cooled beam. Several modifications to the molecular and electron beam components were required as well as the development of the necessary laser hardware and software.

Beam Line Components

The metal tube (beam tube) is the critical component in defining the beam paths and locating the collision zone. This component, which also provided both differential pumping and magnetic shielding, was modified to allow the laser beam to intersect the collision zone. This was achieved by incorporating two opposed extension arms into the beam tube immediately adjacent to the collision zone. Brewster angle windows mounted at the end of each arm then maintained the vacuum integrity but allowed entry and exit of the laser beam. A primary focussing lens was mounted off the beam tube support on a 1 dimensional translator to allow basic adjustment of the laser focussing. This was combined with a secondary of much longer focal length mounted outside the vacuum chamber on a vertical rail which allowed for much finer adjustments of the laser focus.

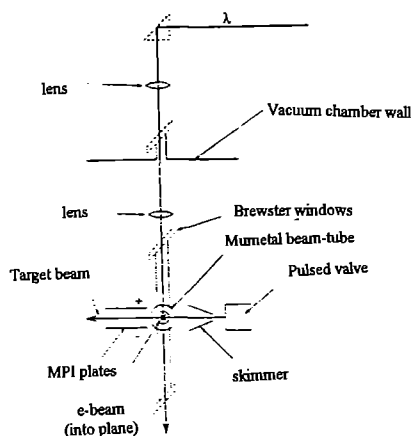


Figure 2 Schematic of modified beam tube and vacuum chamber modifications

Data Collection Hardware

The ions from the REMPI process were collected on parallel plates with an electric field of 400 cm^{-1} approximately 25 mm downstream of the collision zone and the current fed into a Grant differential amplifier. The amplified signal was then passed to a boxcar integrator (Stanford Research Systems SR250), via an optional preamp (Stanford Research Systems SR240), and on to a data acquisition board (National Instruments AT-MIO-16 housed in a PC-AT). The signal was read using one of the ADC convertors on the AT-MIO-16 with the digital output facility on the same board being used to trigger the laser flashlamps. All other timing signals were taken from the YAG laser's Q-switch synch output. A fast laser pulse monitor (Moletron J3-09) was used to measure the laser power on a shot by shot basis for the purpose of power normalisation. The signal from the Moletron was integrated using a second boxcar integrator and sampled by a second ADC channel on the AT-MIO-16.

Data Collection Software

All software was written in house using the National Instruments driver libraries to control the AT-MIO-16 board. The software allowed a prescribed number of shots to be summed at each wavelength before moving the laser to the next required data point. This allowed some control over the signal to noise ratio in the resulting spectrum. All data were stored in plain text format to ease subsequent data analysis.

TARGET BEAM CHARACTERISATION

Ultimately it is intended that this new apparatus will enable structural studies of specific, transient electronically excited, or ionised, species using laser based optical pumping. To this end a laser was obtained on loan from the Rutherford-Appleton Laboratory to enable a number of diagnostic experiments to be performed. These included evaluation of the rotational and vibrational temperatures within the molecular target beam (of great importance, in conjunction with the laser line width, in determining the efficiency of the pump process) and of the achievable ion yields from a range of paradigm species.

An appropriate technique for this diagnostic work was the Resonance-Enhanced-Multi-Photon-Ionisation spectroscopy, a well developed technique [5] [6] providing high sensitivity and probably the best route to molecular ions. The loan laser configuration used comprised a Quanta-Ray PDL3 pumped by a GCR-3 Nd:YAG laser. The dye laser output was first doubled and then mixed with the Nd:YAG fundamental to give tuneable light in the 220 nm to 250 nm region. Several problems were encountered while using this system. Firstly, due to missing optics (a rotator for the YAG fundamental) the resulting power in the uv was much lower than expected. This combined with the erratic behaviour of one of the tracking modules in the WEX mixing unit made it very difficult to maintain a steady uv output with sufficient power for experimental purposes.

It was not anticipated that the total time averaged ion yield would be sufficient to carry out electron diffraction measurements on the ionic species since the duty factor of the loan system was only 10^{-7} . Initial work centered on the $A^2\Sigma^+ \leftarrow X^2\Pi$ transition of the NO molecule as extensive data is already available [7-10]. A 1+1 REMPI scheme was employed to produce ionisation and the laser calibration and signal collection system checked by experiments using a gas cell containing a partial pressure of NO. Large amplitude signals with the expected spectrum were observed. This species was then introduced into the beam target in a 5% NO 95% He mixture at stagnation pressures from 10 - 40 bar and could be run as either a continuous or pulsed beam. Beam intensities corresponding to target densities of 10^{19} - 10^{20} molecules m^{-3} were observed using the mass spectrometer beam monitor. However, no signals corresponding to NO ionisation were seen under any conditions despite several attempts. This problem was unfortunately traced to the fitting of incorrect op-amp chips (741 instead of 3410) in the locally constructed Grant amplifier used. These two chips have similar gains at low bandwidths but the 741 gain drops rapidly at high speed. The expected gains for the two chips as a function of pulse width are shown in the figure. It can

be seen that the 1-2 us pulses observed in our gas cell are readily detectable but the much faster pulses expected in the collision free beam measurements fall below the useful gain envelope. Unfortunately by this point there was insufficient laser loan time remaining to correct the amplifier fault and continue the experiment.

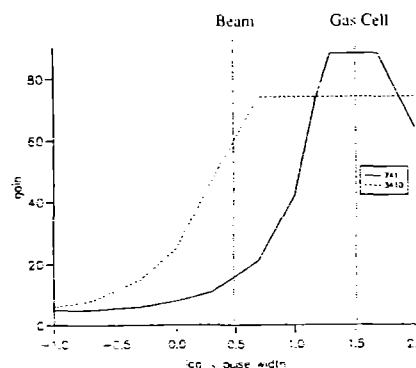


Figure 3 Plot showing gain of the 741 and 3410 op-amps as a function of $\log_{10}$ pulse / us. The expected pulse widths for the molecular beam and gas cell experiments are shown

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THIN FILM DEPOSITION OF CERAMICS USING A HIGH-POWER PICOSECOND KRF LASER

I. Zergioti¹, D. Xenakis¹ and I.C.E. Turcu²

¹ Foundation For Research and Technology-Hellas, Institute of Electronic Structure & Laser, GR-711 10 Heraklion, Greece.

² Rutherford Appleton Laboratory.

INTRODUCTION

Titanium Diboride is a refractory compound with certain unusual semimetallic ceramic properties [1] such as high hardness, chemical and thermal stability, high electrical and thermal conductivity. These characteristics make it a promising candidate as a protective coating on steels in order to increase their wear resistance under extreme chemical, thermal and mechanical conditions and as a diffusion barrier on Silicon in microelectronics applications. Until now, films of TiB_2 have been deposited by conventional techniques such as chemical vapour deposition and physical vapour deposition which require substrate temperature above 500°C. These temperatures can cause property changes and/or thermal damages in certain substrates.

Pulsed Laser Deposition (PLD) is a film deposition process in which a laser beam is incident on a target and induces explosive evaporation of the material. The high energetic particles travel under vacuum and are deposited on the parallel oriented substrate. This technique offers a number of advantages over conventional growth methods. Among these advantages are the potential for congruent target evaporation, the capability of obtaining stoichiometric films of polycomponent materials [2], the simplicity of the process and the potential for film growth at lower substrate temperatures.

The aim of this work was to investigate the use of the high power short-pulse-train excimer laser of the Laser Support Facility for thin film deposition of TiB_2 on D2 tool steel and silicon substrates.

Theoretical and experimental studies [4, 5] of the PLD process have shown that it is possible to establish a direct relation between experimental conditions (plasma density, energy particle) and some film properties (thickness, adhesion, structure, stoichiometry). The plasma parameters can be controllably varied over a wide range by varying the energetic and spectral parameters of the laser radiation. In this instance time-resolved imaging of the ejected material in a nanosecond scale would facilitate the evaluation of its time and space evolution.

This work is part of a comparative study of the deposition process of ceramic materials concerning the influence of laser parameters (energy, pulse duration) on the film quality. The motivation was to overcome problems encountered during PLD experiments using standard excimer lasers with nanosecond pulse durations such as poor adhesion of the film and presence of particulates.

EXPERIMENTAL PROCESS AND RESULTS

A schematic diagram of the experimental set-up is presented in figure 1. The system consists of a deposition chamber evacuated by means of a turbomolecular pumping system down to 4×10^{-5} mbar. TiB_2 targets are irradiated at an angle of 50° by the excimer laser beam and are rotated between each laser shot using motors. Two types of TiB_2 material were used in the experiments: commercially available pellets (GOODFELLOW $d=4.52 \text{ g/cm}^3$) and pellets prepared by hot pressing sintering at 2000°C under controlled atmosphere (CERECO $d=4.35 \text{ g/cm}^3$). The substrates were D2 tool steel specimens ground to 600 grid with SiC paper, and Silicon specimens.

The KrF laser system [3] delivers pulse trains of 30ns overall duration. Each train consists of 15 pulses of 2-10ps duration and 15mJ energy. The beam was focused to a 3mmx5mm spot size at the target using a plano-convex lens with focal length 1m. Several experimental parameters were examined as outlined below:

Energy density: 0.5-2 J/cm²

Distance Target-Substrate: 35 - 50 mm

Laser Pulse Repetition rate: 20-50 Hz

Total Irradiation time : 10-40min

The morphology of the films were examined by Scanning Electron Microscopy, whereas the microstructure and the composition by Energy Dispersive X-Ray microanalysis, X-Ray Diffractometer and Transmission Electron Microscopy. Thickness measurements were

performed by stylus profilometer. The coating characterization, performed at FORTH/IESL, is still in progress.

The SEM/EDX analysis showed the presence of coherent coating consisting of titanium, boron and oxygen. The thickness of the deposition was between 200nm and 1µm. The deposition rate varied between 0.1 Å/pulse to 0.3 Å/pulse. The adhesion of the coatings was moderate. Irregular particles with the same composition, and size between submicron to 10 microns, were detected, of which the density and the size was reduced at low energy density (figure 2). The irregular shape of the particulates means that they are produced from solid ejecta and their micron range size, indicates that they are produced directly from the target and not from the vapour species condensation. The proposed mechanism for the particulate generation, in the literature [2] is the existence of pits, craters, microcracks that exist, or are substantially formed due to the laser - induced thermal and mechanical shock.

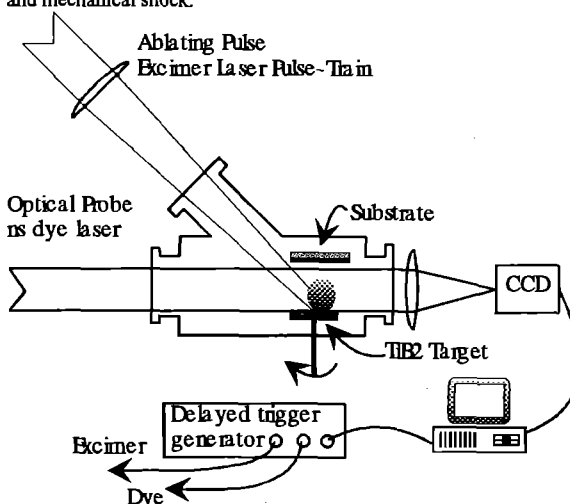


Figure 1. Experimental setup.

An excimer pumped dye laser illuminated the material ablated from the target (cf. figure 1). The image was recorded on a silicon CCD camera which was synchronized to the lasers via a microcomputer. The delay between the two lasers was adjusted from a synchronization unit/delayed trigger generator. The range of delays investigated was 0 to 80 ns with a resolution of ~10 ns. An estimation for the velocity of the ablation front of the ejected material is 10⁶cm/sec.

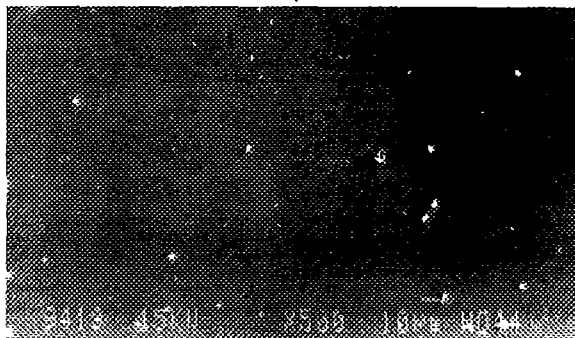


Figure 2. TiB_2 coating on Si substrate (SEM picture).

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INTERFEROMETRIC SHAPING OF FEMTOSECOND LASER PULSES TO STUDY MULTIPLE IONIZATION OF MOLECULES IN INTENSE FIELDS

K Codling¹, L J Frasinski¹, A J Giles¹, A J Langley², J H Posthumus¹, W Shaikh², P F Taday² and M R Thompson¹

¹J J Thomson Physical Laboratory, University of Reading, Whiteknights, Reading, RG6 6AF, UK

²Central Laser Facility, Rutherford Appleton Laboratory, Didcot OX11 0QX, UK

1 INTRODUCTION: THE NEED FOR PULSE SHAPING

Despite considerable theoretical and experimental effort, the behaviour of simple molecules exposed to intense laser fields (focused intensities $> 10^{14}$ W/cm²) is poorly understood. On the theoretical front, standard multiphoton theory cannot be applied because a laser field of such strength cannot be regarded as a small perturbation to the system. Nevertheless, some advances have been made by assuming that the multielectron dissociative ionization (MEDI) process can be described in terms of a sequential, field ionization, Coulomb explosion model^{1,2}.

On the experimental front, there are two interesting aspects of the MEDI process that require explanation. Firstly, the kinetic energies of the fragment atomic ions appear to be independent of laser pulse rise time over a large range of pulse lengths — from 0.1 to 2 ps in a recent study³. Secondly, for a particular molecule, the dissociation energies of all fragmentation channels ($I^+ + I^+$, $I^+ + I^{2+}$, $I^{2+} + I^{2+}$, etc) are a specific fraction (~70% for I₂) of the Coulomb energy (given by $E = Q_1 Q_2 / R_e$, where Q_1 and Q_2 are the atomic ion charges and R_e is the equilibrium internuclear separation of the neutral molecule) [3,4]. This strongly suggests that all of the Coulomb explosions occur at a molecule-specific critical distance, R_c , larger than R_e ($R_c = R_e / 70\%$ for I₂).

The Saclay group has pointed out that laser-induced stabilisation at the critical distance can provide an explanation for the above facts⁵.

Since both the stabilisation model and the inertial confinement model invoke multiple ionization close to the critical internuclear distance, they cannot easily be distinguished by measuring the kinetic energies of the fragment ions. However, the branching ratios (partial cross sections) for the various fragment channels are predicted to be independent of laser pulse rise time in the stabilisation model, whereas in the inertial confinement model there should be substantial changes. We describe here an interferometric approach to varying the laser rise time which should allow us to determine which, if either, of the two models is correct.

2 THE INTERFEROMETRIC METHOD

Varying the rise time of femtosecond pulses is quite difficult. The obvious method is to change the pulse duration and assume that the rise time changes accordingly. Unfortunately, such a change usually requires a significant modification of the laser system, giving laser pulses that are likely to be different in other respects. Spectral filtering to produce pulses with specific temporal profiles is a possibility, but this is by no means straightforward^{7,8}. The rise time, or indeed the shape of any other part of the laser pulse can be directly manipulated using a temporal approach, that is interferometry.

The principle of the interferometric method is to split each laser pulse into two pulses of unequal intensity, delay the main pulse and recombine them. In this way the low-intensity prepulse can interfere constructively or destructively with the leading edge of the main pulse, producing a decrease or increase in its rise time. The advantage

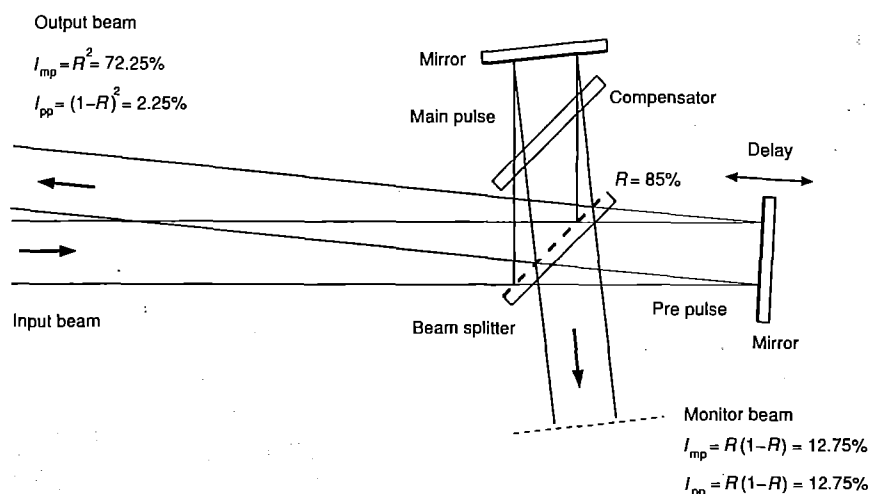


Figure 1. A schematic diagram of the pulse shaping interferometer. The main pulse, I_{mp} , and the prepulse, I_{pp} , intensities depend on the reflectivity, R , of the beam splitter. The prepulse mirror is mounted on a flexure stage, allowing a delay to be introduced between the main pulse and the prepulse.

The Reading group has recently used a phenomenological model which predicts kinetic energies and threshold intensities⁹. Here, too, laser-induced bound states (bond hardening) are invoked but it seems unlikely that such bound states can persist at high ionization stages. The approach used by the Ottawa group, on the other hand, does not require stabilisation; inertial confinement and extremely rapid sequential excitation and ionization at, or near, the critical distance provide the basis of their explanation⁶.

of this method is that it is external to the laser system and allows a rapid and controlled adjustment of the laser pulse rise time.

The modified Michelson interferometer is shown in figure 1. The laser beam is incident on the beam splitter, which has a reflectivity of 85% at a wavelength of 740 nm. In this arrangement the main pulse and the prepulse intensities (I_{mp} and I_{pp}) that pass through to the experiment are, respectively, 72.25% and 2.25% of the input

intensity. Other main pulse and prepulse intensities can be obtained by selecting a beam splitter of different reflectivity. A compensator in the main beam arm equalizes the glass thickness seen by both beams and balances their chirp. (This chirp can be cancelled by introducing an opposite chirp before the pulse enters the interferometer.) About 1 mm movement of the prepulse mirror mounted on a flexure stage provides sufficient delay range. To maintain a stable interference pattern the interferometer is made compact and rigid and identical materials and dimensions of the main pulse and prepulse arms ensure equal thermal expansion.

The desired overlap of the pulses can be achieved by observing the interference fringes in the monitor arm of the interferometer. Since both beams have the same intensity, the following adjustments are possible: zero pulse delay can be set using white light and locating the dark fringe; the time delay can be calibrated by simply counting interference fringes; wavefronts can be set parallel by producing a uniform intensity across the monitor beam.

3 EXPERIMENTAL VERIFICATION

The laser used to verify the interferometric modification of pulse rise times was a Spectra-Physics argon-ion pumped titanium-sapphire laser. This provides near-bandwidth-limited pulses of 100 fs duration at a pulse repetition rate of 82 MHz. The laser was operated at an output wavelength of 740 nm. Pulse duration measurements were made with a Femtochrome rapid-scanning autocorrelator with a temporal resolution better than 30 fs. Spectral measurements of the pulses were made with a 0.27 m Princeton Instruments spectrometer using a 1200 grooves/mm grating and with a slit width of 50 μ m that gave a spectral resolution of 0.3 nm. The output from the spectrometer was detected using a CCD camera. Although it was convenient to test the interferometer with the low energy pulses from the titanium-sapphire laser, in MEDI experiments the interferometric method will be applied using 100 fs pulses amplified to several hundred microjoules. This amplified laser system has been described in detail elsewhere⁹.

Figure 2(a) indicates how the theoretical pulse duration (FWHM) varies with time delay for constructive (solid line) and destructive (dashed line) interference, for a laser pulse duration of 108 fs. As expected, the gap between the two curves initially opens up, reaching a maximum at about 100 fs delay and then closes as the pulses are separated in time. Also shown in figure 2(a) are the experimental pulse duration data obtained with the autocorrelator. Note the excellent agreement between theory and experiment.

Figure 2(b) shows theoretical plots of rise time versus time delay for constructive and destructive interference of these same two pulses used to produce the theoretical curves of figure 2(a). Note the surprising fact that the maximum and minimum rise times (defined here as the time interval between the 5% and 90% intensities) do not occur at the same time delay but at about 100 fs and 200 fs, respectively. The reason for this difference is that at the larger delays, where the leading edge of the main pulse has a low amplitude, destructive interference is inefficient in reducing the rise time of the leading edge whereas in constructive interference the rise time can be successfully increased.

Figure 2(c) shows the theoretical spectral bandwidth (FWHM) versus time delay for constructive and destructive interference. For small and medium time delays the variation is opposite to that in figure 2(a) and the bandwidth-duration product stays approximately constant. At large time delays the spectra become multiply peaked and the peak positions depend markedly on whether the interference is constructive or destructive. The relatively large separation of the two curves, especially for large time delays, can be used to identify the positions of constructive and destructive interference. Indeed at large time delays, this spectral method is more practical than visually monitoring the interference fringes. Once again, the squares and circles are the experimental points; these agree well with theory.

The overall agreement between theory and experiment, represented by figures 2(a) and (c), gives us strong but indirect evidence that the variations in rise time represented by figure 2(b) are realized in practice. To observe the changes in pulse shape more directly we stretched the pulses and cross-correlated them with the unstretched

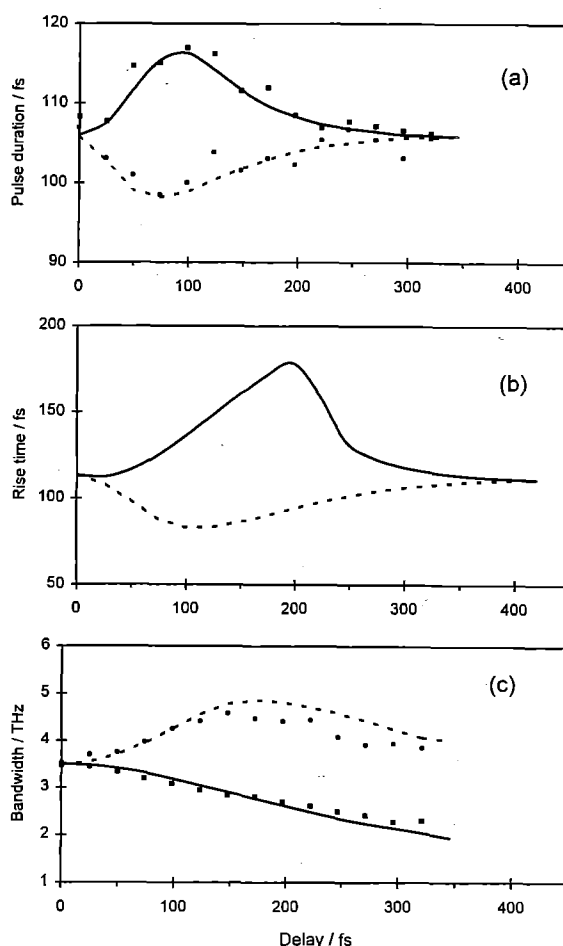


Figure 2. Plots of experimental points and theoretical curves for constructive (■ and —) and destructive (● and - - -) interference between prepulse and main pulse of 108 fs duration, (a) pulse width (FWHM) versus time delay, (b) pulse rise time versus time delay, using the 5–90% criterion, (c) spectral width (FWHM) versus time delay

ones. The stretching from 80 to 650 fs was introduced by passing the laser beam through a block of glass, 105 mm in length. The stretched pulses were directed into the interferometer (figure 1) and time delays introduced in order to produce a variety of pulse shapes. They were then cross-correlated with the original 80 fs pulse in the rapid-scanning autocorrelator. Figure 4 shows the cross-correlation signal for various time delays between the two pulses. Because of the shortness of the 80 fs pulses, these plots give a direct measure of the shape of the longer, interferometrically modified pulse.

Figure 3(a) shows cross correlation of the 80 fs pulse with the unmodified 650 fs pulse. The solid line shows the experimental shape, the dashed line the theoretical one given by the envelope of formula (1) with $\Delta t = 650$ fs. Note the good approximation to the sech^2 shape. Figure 3(b) shows the cross correlation of the chirped pulse that has been interferometrically modified by introducing time delays ranging from 319.6 to 322.0 fs between main pulse and prepulse. This range of time delays covers exactly one optical cycle, corresponding to one fringe of 740 nm radiation. As expected, the first and the last shapes are almost identical. The temporal beats visible on these shapes are due to frequency mismatch occurring when the strongly-chirped pulses are delayed with respect to one another. The excellent agreement between experiment and theory demonstrates the high reliability and reproducibility of the interferometric pulse shaping.

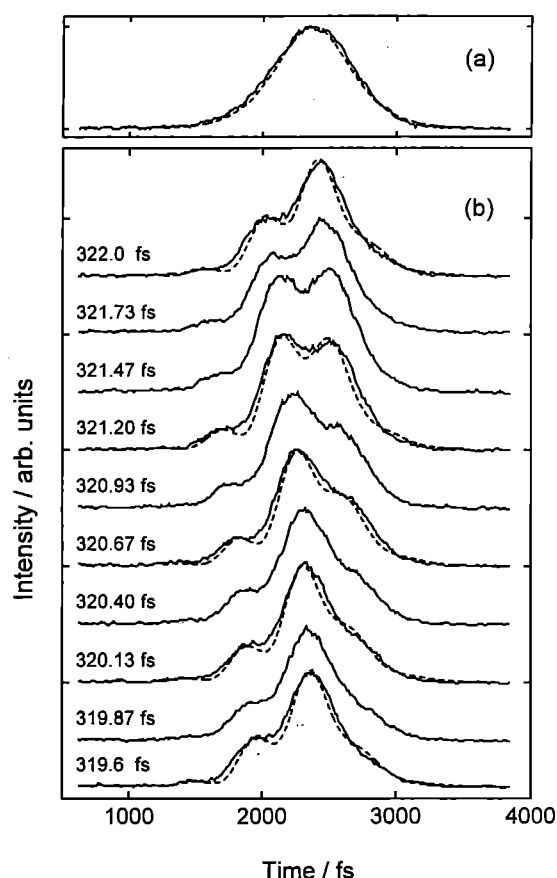


Figure 3. Experimental (—) and theoretical (---) pulse shapes produced by cross-correlating a 650 fs chirped pulse with an unchirped 80 fs pulse:
 (a) the raw 650 fs chirped pulse,
 (b) interferometrically modified 650 fs chirped pulse using time delays between the prepulse and the main pulse spanning one optical cycle (319.6–322.0 fs)

One situation of particular interest in the context of the multiple ionization of molecules is indicated in figure 4. This shows the temporal shape of two interferometrically generated laser pulses using 108 fs prepulse and main pulse, as in figure 2. The time delays between prepulse and main pulse are 100 fs (the destructive interference minimum of figure 2(b)) and 173 fs (constructive interference). There is about a factor of 2 difference in pulse rise time, using the 5%–90% criterion, see figure 2(a). However, the specific

pulse shapes at low intensities can be used to great advantage. Consider, for example, the multiple ionization of I_2 . According to the Ottawa model⁶ the threshold for producing the $(I^+ + I^+)$ channel occurs at an intensity of about $9 \times 10^{13} \text{ W/cm}^2$. This threshold is indicated in figure 4 by the horizontal dashed line. It takes approximately 80 fs for the I^+ ions to reach the critical internuclear distance when, as mentioned earlier, the rapid sequential ionization of I_2^{2+} occurs. In this time period, the laser pulse with the longer rise time will reach an intensity of about $3.7 \times 10^{14} \text{ W/cm}^2$, whereas the laser with the shorter rise time will reach an intensity of about $1.8 \times 10^{15} \text{ W/cm}^2$, a factor of almost 5 greater. In the context of the Ottawa model, this should lead to a considerable difference in observed branching ratios; in the Saclay model the branching ratios should remain unchanged. Accurate branching ratios can be obtained using covariance mapping¹⁰.

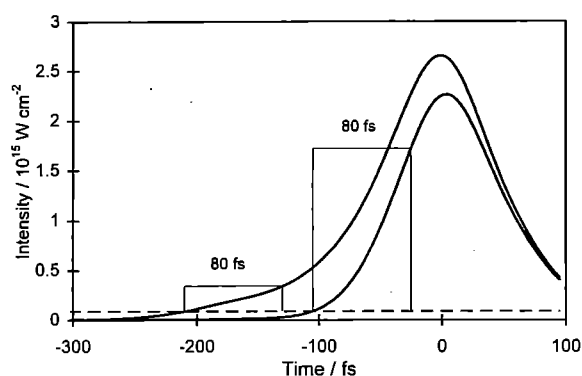


Figure 4. Intensity profile of a 108 fs pulse modified using constructive interference and a time delay of 173 fs (long rise time) and with destructive interference and a time delay of 100 fs (short rise time). The horizontal dashed line represents an intensity of $9 \times 10^{13} \text{ W cm}^{-2}$. The rectangles emphasise the fact that the intensity increase in 80 fs differs by a factor of 5.

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DIRECT MEASUREMENT OF FEMTOSECOND PULSE FRONT DISTORTION DUE TO LENSES AND ITS ELIMINATION USING AN ACHROMATIC LENS

S Ameer-Beg², A J Langley¹, I N Ross¹, W Shaikh¹ and P F Taday¹

¹Central Laser Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxon., OX11 0QX, U.K.

²Department of Physics, University of Essex, Colchester, Essex, CO4 3SQ, U.K.

INTRODUCTION

Temporal deformation of ultra-short optical pulses in lenses can significantly reduce the focused power and time resolution available from lasers. Aside from conventional aberrations, two types of temporal pulse distortion can occur in lenses¹⁻⁴ namely: pulse front distortion (PFD) and group velocity dispersion (GVD)⁵. PFD arises because of the difference between the phase and group velocities in the lens as it varies in thickness across the beam. This results in a radially dependent delay between the phase and pulse fronts. GVD on the other hand results in pulse stretch. Since the amount of GVD depends on the thickness of dispersive material, axial beams are affected most in positive lenses.

It is important to find optical designs which avoid pulse distortion. One solution to the problem is to use paraboloidal reflective optics since these do not distort pulse fronts. Such optics may not always be convenient, however, where on-axis focusing in the forward direction is required, or where engineering constraints rule out off-axis paraboloidal focusing. Such problems were encountered in the design of an $f/2$ optic for focusing intense, 100 fs pulses in multiphoton ionisation (MPI) experiments. An alternative solution is to use achromatic lenses for focusing since, as theoretical studies^{1,6} have pointed out, these lenses should produce a constant delay across the beam between the phase and pulse fronts. This being an acceptable solution for our application, an achromat was designed and tested to meet our focusing requirements.

DESCRIPTION OF EXPERIMENT AND RESULTS

Two types of 50 mm focal length lenses were compared in this investigation. The first of these comprised a fused-silica 34 mm diameter plano-convex doublet. Each component was 5.5 mm thick on axis and was separated by 2.5 mm. One component was aspherised to correct for spherical aberration. The doublet was used for many years for picosecond MPI experiments and provided good quality focal spots for up to $f/2$ focusing. Two identical doublets were manufactured.

The other lens was an achromat. This was designed using Kidger Optics ray-tracing software to provide 50 mm focal length $f/2$ focusing with minimum pulse front distortion (chromatic aberration) and spherical aberration at 740 nm. Minimum aberrations were achieved with a three component optic.

Pulse stretch due to GVD in the achromat was calculated to be 22 fs. Thus a 100 fs duration sech^2 pulse passing through the lens would be stretched to 102 fs. In principle this stretch could be removed by the introduction of an appropriate amount of negative GVD.

The pulse front distortion, Δt_{PFD} , is a function of the rate of change of focal length with wavelength $df/d\lambda$ ¹ and is given by

$$\Delta t_{\text{PFD}} = \frac{\lambda r^2}{2cf^2} \frac{df}{d\lambda} \quad (1)$$

where r is the beam radius at the lens of focal length, f , c is the speed of light and λ is the wavelength of the incident light.

The ray tracing software was used to calculate the effective focal length of the achromat as a function of wavelength and this data was used in equation 1 to calculate the pulse front distortion. Both these data sets are shown in Figure 1. The lower graph illustrates that pulse front distortion of less than 25 fs is expected over the

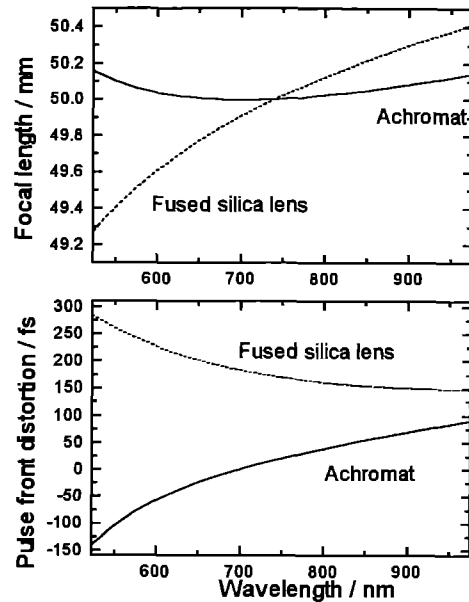


Figure 1: Focal length (upper graph) and pulse front distortion (lower graph) as a function of wavelength for the achromat described in the text and a fused-silica lens of 50 mm focal length.

wavelength range 650 - 775 nm. Also shown for comparison are the focal lengths and pulse front distortion calculated for a fused silica lens of focal length 50 mm (at 740 nm).

Two identical achromats were manufactured to the design discussed above by Optical Surfaces Ltd who also made interferometric measurements demonstrating a maximum optical path difference across the lens aperture of $< \lambda/5$ at 633 nm. Measurements of the focal spot obtained with a 740 nm, 25 mm diameter beam of uniform intensity indicated that near to diffraction limited focusing was achieved with these lenses. Each achromat was assembled in a stainless steel mount designed for operation in an ultra-high vacuum apparatus. All optical surfaces were coated for minimum reflection losses at 740 nm.

Pulse front distortion measurements were carried out using the experimental arrangement shown schematically in Figure 2. A Spectra-Physics argon-ion pumped titanium-sapphire laser provided near to transform-limited sech^2 pulse of 100 fs duration at 740 nm and at a repetition rate of 82 MHz.

The laser beam was incident upon an optical beam-splitting arrangement resembling a Michelson interferometer. The identical lens pair under investigation was placed in one arm and formed a telescope of unit magnification. The position of the mirror in the other arm was set to give a suitable delay between the plane and curved pulse fronts. After recombination at the beam splitter, different regions of the pulse fronts could be selected with a 1mm diameter aperture.

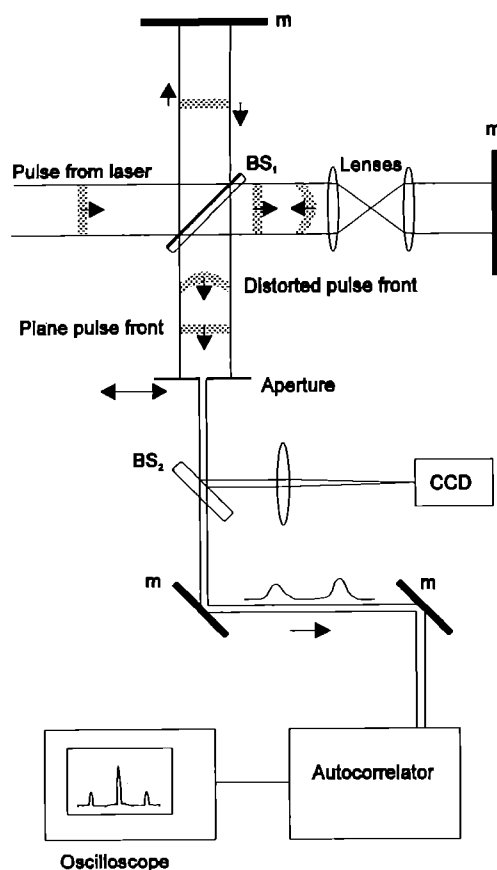


Figure 2: Arrangement used to measure the pulse front delay introduced by lenses. Key: BS beam splitter, m mirror

As an alignment aid the beams were sampled by a 15% splitter placed after the aperture and focused with a 100 mm focal length lens onto a CCD detector, the output of which was displayed on a television screen. By ensuring the images of the two focal spots were superimposed, the angle between the beams was maintained below 100 μ rad corresponding to a temporal delay of ≈ 8 fs across the 25 mm beam.

The relative delay between the two pulses emerging from the aperture was measured by directing them to a Femtochrome rapid-scanning autocorrelator. The autocorrelation measurement of the two pulses consisted of three peaks. The central one corresponded to the autocorrelation of both pulses whilst the outer peaks was a cross-correlation of one pulse with the other. The temporal separation between the central and outer peaks give a measure of the delay between the two pulses.

Scanning the aperture across the beams allowed the relative delay between the curved and reference pulse fronts to be measured. Figure 3 shows such measurements obtained with the fused silica doublets and achromatic lenses. The double pass through the lens pairs has been taken into account and the delay plotted on the vertical axis of the graph is 0.25 times that measured.

The fused silica doublet introduced a significant curvature to the pulse front. It is noted that the measured curvature was less than that calculated (dotted curve) for a 50 mm focal length fused silica singlet lens. This is due to the reduced beam radius at the second component of the doublet and aspherisation. However, the measured delay remains quadratically dependent on radius, as indicated by the close fit of a curve of the form $\Delta t_{\text{PFD}} = 0.82r^2$ (solid line). From this the radius at the second component was calculated to be 8.5 mm which is close to the value obtained by ray tracing.

Also shown in Figure 3 is the measured pulse front delay for the achromatic lens. There is evidence of a small tilt between the achromat and reference pulse fronts of about 14 fs across the 25 mm beams due to either a wedge on the lens or a slight misalignment of the instrument. However, it is clear that the quadratic chromatic pulse front delay has been eliminated.

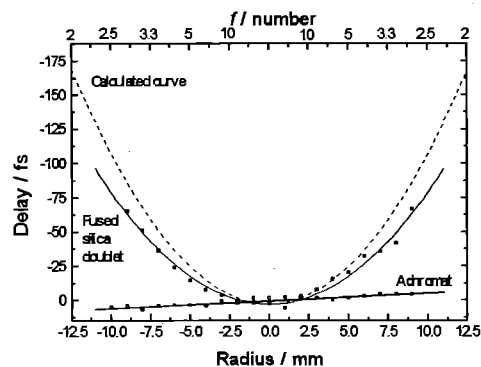


Figure 3: Measured pulse fronts (740 nm) after focusing with a fused-silica doublet and an achromatic lens.

CONCLUSION

An achromatic lens designed for high quality $f/2$ focusing has been demonstrated to introduce no significant pulse front distortion as predicted by theory.

Pulse front distortion caused by lenses has been measured directly using a scheme comprising equipment likely to be found in any femtosecond laser laboratory. The technique can be applied to any optical component and does not require apparatus designed for interferometric measurements. Unlike methods which seek to measure whole beam cross-correlation between plane and distorted pulse fronts, the technique described here measures relative temporal delay across a beam and is therefore not unduly affected by beam intensity profile, temporal pulse shape or GVD.

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FABRICATION OF 200nm FIELD EFFECT TRANSISTOR BY X-RAY LITHOGRAPHY WITH A LASER-PLASMA X-RAY SOURCE

CM Reeves¹, I C E Turcu², A M Gundlach¹, J T M Stevenson¹, A J Walton¹,
A W S Ross¹, P Prewett², R A Lawes², P Anastasi³, R Burge³ and P Mitchell⁴

¹Department of Electrical Engineering, University of Edinburgh

²Rutherford Appleton Laboratory

³Department of Physics, King's College

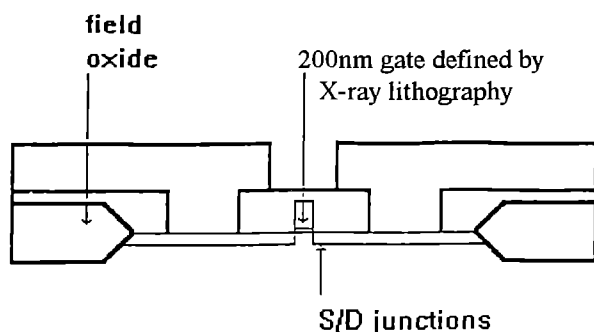
⁴Leica Cambridge Limited

INTRODUCTION

The future 1Gbit DRAM technology and its logic derivative will require devices on 180nm scalelength for both speed and device density on the microchip. The present work demonstrates the fabrication of 200nm gate length silicon Field Effect Transistors (FET) by X-ray lithography using the RAL excimer laser-plasma source generating 1nm wavelength X-rays. Only the transistor gate dimension is critically defined by X-ray lithography (Fig.1) with the other transistor elements defined by conventional lithography, the mix and match technique. The new lithography approach is outlined and electrical results are presented for devices with 5nm gate oxides. The 0.2 μ m devices exhibit transconductance of 220mS/mm and excellent quasi-long channel operation. This work provides an important first demonstration that the laser-plasma X-ray source is a credible candidate for the future 1 Gbit technology.

LITHOGRAPHY

The RAL high power excimer laser-plasma X-ray source¹ is based on commercial lasers and consists of: picosecond laser oscillator, temporal and spatial multiplexer, excimer double pass amplifier laser, and X-ray chamber filled with He gas at atmospheric pressure and containing the focusing lens and the copper tape target. The $\lambda = 248\text{nm}$ laser beam consisting of a train of picosecond pulses (8ps long) is focused onto the Cu tape target to power densities in excess of 10^{15}W/cm^2 generating a dense and hot Cu plasma. This plasma acts as an efficient high power X-ray emitter with $\lambda = 0.9\text{nm}-1.2\text{nm}$ (Cu L-shell transitions). X-ray lithography is then carried out using 1:1 printing in conjunction with X-ray mask technology as developed for synchrotron based lithography². Wafer exposures are carried out a working distance of 125mm from the foil target and mask-to-wafer spacing is set to 12 μ m. Using AZPF514 resist with sensitivity of 20mJ/cm^2 , a $20\text{mm} \times 20\text{mm}$ exposure field is exposed in 4 minutes with the the plasma source generating 100mW X-ray average power into 2π sr. Typical lithography results are shown in Fig.2 for 180nm resist features.



DEVICE INTEGRATION

This new lithography approach has been integrated into a 200nm silicon FET fabrication experiment. The fabricated FET architecture is indicated in Fig.1. The gate level is defined by X-ray lithography and pattern transferred into doped polysilicon using a SiCl_4/Ar reactive ion etch process. Etch results are indicated in Fig.3 which shows a 200nm polysilicon gate electrode, patterned over a 5nm thermal gate oxide. Threshold adjustment and punchthrough suppression are set by dual B^+ implants of 5×10^{12} atoms/ cm^2 at 20keV and 5×10^{12} atoms/ cm^2 at 60keV. Source/drain regions are formed by As^+ implantation using a dose of 2.5×10^{15} atoms/ cm^2 at 30keV. Activation is carried out using a $600^\circ\text{C}/30$ minute anneal followed by a $1000^\circ\text{C}/12$ second rapid anneal. Figure 4 shows resulting I-V characteristics for an X-ray defined 200nm gate length silicon FET. Gate voltage is stepped from 0 to 1.75V in 0.25V steps and measured transconductance is 220mS/mm. Excellent quasi-long channel operation is confirmed via the low output conductance.

SUMMARY

First electrical results are presented for 200nm silicon FETs produced by X-ray lithography using an excimer laser-plasma X-ray source. These results confirm the feasibility of using such a source as a low cost alternative to synchrotron based X-ray lithography for patterning 180nm features as required for future 1Gbit technology. Further work aimed at reduction of exposure time from 4 minutes to <10 seconds is necessary (e.g. through increased X-ray output and improved resist sensitivities) in order to meet production throughput requirements.

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Figure 1. Schematic of Field Effect Transistor Architecture used in the present work.

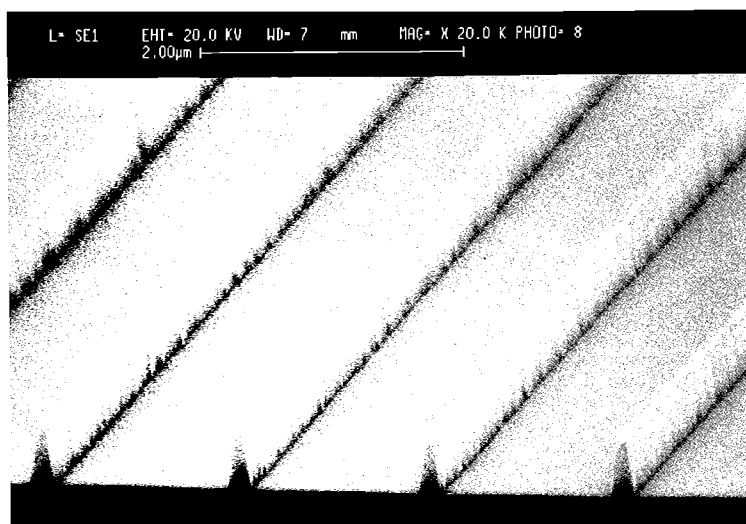


Figure 2. SEM micrograph of 180nm lines printed in AZPF514 photoresist. The rounded walls are due to the first reactive ion etch (CHF_3/He) used to pattern the features into the oxide layer.



Figure 3. Patterned 200nm doped polysilicon gate electrode over 5nm thermal gate oxide.

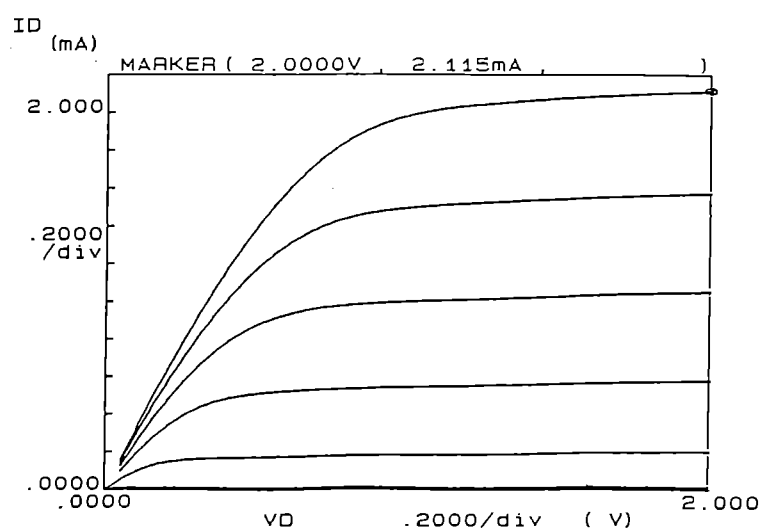


Figure 4. I-V characteristic of 200nm gate length FET defined by X-ray lithography with a laser-plasma X-ray source. Gate voltage is stepped from 0 to 1.75V in 0.25V steps.

LASER SUPPORT FACILITY: OPERATIONS AND DEVELOPMENT

A total of 48 RAL-based experiments were scheduled 153 weeks of laser time. Once again this represents an increase over last year as can be seen in figure 1 which compares figures for the past five years. Loan pool lasers were loaned for 6 years worth of scheduled time for experiments carried out in 15 university laboratories.

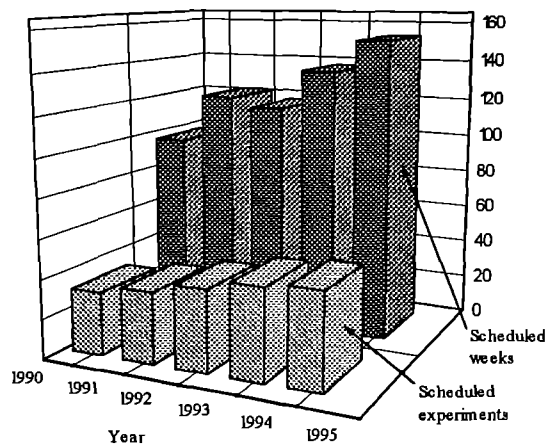


Figure 1. RAL-based experiments

There were 71 applications for time on LSF equipment; the distribution by subject is shown in table 1.

Table 1

1993/94	Physics	Chemistry	Biology
Applications %	29	47	24

Table 3 lists the lasers available for experiments at RAL or for loan.

FEMTOSECOND LASER LABORATORY

Andrew J Langley, Charles Mo, Waseem Shaikh, Phil Today.

This has been a productive and eventful year with six experiments completed despite the time taken out to extensively restructure LSF laboratories between January and April 1995. A total of 4 externally refereed papers were published and 10 conference presentations made.

Transient absorption techniques were used by O'Neill *et al.* from Hull to investigate carrier tunnelling between central CdMnTe quantum wells and outer CdTe wells. Tunnelling time was measured at 8 ps whilst intrasubband relaxation with the emission of longitudinal optical phonons was calculated to be 200 fs in agreement with theory. This and earlier work has resulted in one published paper and a conference presentation at CLEO Europe. Ultrafast relaxations in conjugated porphyrin polymers - which have possible third-order optical applications - were studied by Bradley's group from Sheffield. Another transient absorption study by Sinclair *et al.* from Paisley looked at picosecond processes in cyanine dyes. This group presented earlier work on azo dyes at the XVth IUPAC Symposium in Prague and at a Royal Society of Chemistry meeting in Edinburgh.

A novel interferometric method for modifying the rise time of femtosecond pulses was developed for experiments on the multielectron dissociative ionisation (MEDI) of simple molecules. The technique, which will be used to test theoretical models of MEDI, was presented at two IOP conferences in March and a paper has been accepted for publication. During

the year an achromatic lens allowing $f/2$ focusing of 100 fs pulses without distortion of the pulse front was designed at RAL and constructed by Optical Surfaces. This work was presented at the Applied Optics conference in York and at the Ultrafast Phenomena and Applications Conference, Trieste.

A Spectra-Physics sub-50 fs titanium sapphire oscillator (Tsunami) was recently installed as a source for the amplified laser and replaces the 100 fs Tsunami. A modified chirped pulse amplification arrangement has been built and amplification trials are underway with 50-100 fs, 300 μ J pulses the performance target. The system will continue to be used for femtosecond pulse interactions i.e. multielectron dissociative ionisation studies, and for ultra-short pulse measurements and techniques.

NANOSECOND LASER LABORATORY

Sue Tavender, Tony Parker and Andrew Weedall

Throughout the year, the nanosecond lasers were scheduled for a total of 48 weeks with 11 different user groups using the facility. Laser down-time was only 2 weeks. A total of 8 papers and 7 conference presentations have resulted from work done during the period. Once again time resolved resonance Raman (TR³) featured prominently in the programme using two thirds of the allocated time. The CCD is now used routinely for Raman detection from 320 nm to 940 nm with the diode array for detection in the UV region. The laser flash photolysis data collection has been upgraded to include automatic measurement of $I(0)$ values.

The wide range of experiments carried out using the nanosecond facility are detailed elsewhere in this report. One of the year's highlights included successful experiments by a new user of the facility, Dr Katharine Reid from Nottingham University, using the REMPI technique to study the hyperfine depolarisation of the $A^2\Sigma^+$ state in nitric oxide(NO). This complex experiment used both excimer pumped dye lasers in a frequency doubled mode, one coupled to the $X^2\Pi(v=0) \rightarrow A^2\Sigma^+(v=0)$ transition in the NO and the second to ionise the $A^2\Sigma^+$ state. The two lasers were run with the time delay between the pump and ionisation beams alternating between -30 and 90 ns. Photo-electrons were detected using an MCP as a function of the delay between the two lasers and the relative polarisation of the excitation and ionisation beams.

Dr Roger Bisby of Salford University continued his work on biologically important free radicals and has recorded TR³ spectra of tryptophan and indol radicals. This work has gone on to look at the intermediates in copper amine oxidase which catalyses the oxidation of amines to aldehydes. A 'cage' consisting of an adduct of the amine with nitrobenzaldehyde has been synthesised and tested using 308 nm laser light. The amine substrates were released which will allow the transient intermediates of the catalytic cycle to be studied. Prof T G Truscott (Keele) obtained the first TR³ spectrum of carotenoid radical cations which have been implicated in photosynthesis, vision and the prevention of cancer. The radical ions were generated photochemically using triplet nitronaphthalene as an electron acceptor and probed in the near infra-red, at 760 nm.

PICOSECOND LASER LABORATORY

Pavel Matousek, Gary Morton, Andrew Scully and Mike Towrie.

This year there have been a total of 13 scheduled experiments, 7 single photon counting and 6 resolved resonance Raman (TR³).

Success has been achieved in the development of a laser-based fluorescence microscope facility. The microscope, which uses a Nd:YAG pumped cavity dumped ps dye laser as a source, uses one of

the fastest gated image-intensifiers available to provide a time resolution of 120 ps. The instrument, which is capable of sub-micron spatial resolution, will be used for studying the distribution of a phthalocyanine photosensitizer (used in photodynamic treatment of tumours) within cells.

Following the success last year of a user backed grant application to EPSRC to develop picosecond spectroscopy at RAL, the LSF have purchased a Spectra-Physics argon-ion pumped titanium sapphire oscillator (Taunami) and regenerative amplifier (Spitfire). The new facility, which will be called the Ultrafast Spectroscopy Laboratory, will be commissioned over the summer of 1995 with the first scheduled experiments likely to begin next year.

The new laboratory will provide independently tunable UV-VIS pump and probe beams for femtosecond or picosecond time-resolved studies in the condensed or gas phase. Both picosecond time-resolved Raman and transient absorption measurements will be available - the latter using either single frequency or white light continuum as a probe source. Signal acquisition and processing apparatus for other techniques such as fluorescence or REMPI detection may be available if warranted by demand.

LASER-PLASMA X-RAY LABORATORY Edmond Turcu

The "High Average Power Laser-Plasma X-ray Source" was upgraded to the record 1watt X-ray average power output at $h\nu \sim 1\text{keV}$ ($\lambda \sim 1\text{ nm}$), into $2\pi\text{ sr}$, or $\sim 200\text{ mW/sr}$. In biological experiments a record X-ray dose rate of 10 Mrad/min was delivered to biological material. This was achieved with a new, 70W, LPX 210Fi (Nova tube), excimer laser amplifier in a new, patented, laser configuration which amplifies trains of picosecond pulses. A large part of the R&D programme was performed under the contract from JMAR Industries Inc. (of California) who are constructing a commercial laser-plasma X-ray source for X-ray lithography.

During the year, 49 weeks were scheduled for experiments with 2 weeks used for installation and testing of the new excimer laser. New applications of the X-ray source were the measurement of enzyme size (Birmingham University) and direct comparison of biological effects of α -particle and X-ray radiation (MRC-Radiobiology Unit). The investigation of X-ray DNA damage and repair (Birmingham Univ.) continued, as well as 100 nm X-ray lithography (Edinburgh Univ.). The investigation of DNA with UV laser radiation: 149 nm and 351 nm (Birmingham Univ.) and 193 nm (MRC and Leicester Univ.) continued. New groups have investigated picosecond UV laser ablation and thin film deposition: FORTH/IESL, Greece, as a Large Scale Facility Euro-experiment, and Warwick University.

The X-ray work resulted in six publications, three patent applications, seven conference presentations, four postgraduate and one degree thesis, and two publications in specialist magazines. Four publications resulted from use of the UV lasers to investigate the photochemistry of DNA with 193 nm and 248 nm light by O'Neill (MRC) and Symons (De Montford).

LSF LABORATORY RESTRUCTURING

The construction of the new laboratory for ultrafast spectroscopy mentioned above has involved considerable restructuring of the LSF laboratories. This was carried out between January and April 1995. The laboratories previously used for time-resolved picosecond Raman spectroscopy (TR^3) and single-photon counting (TCSPC) experiments have been reorganised into the Ultrafast Spectroscopy Laboratory. This rearrangement has allowed for the construction of a small laboratory dedicated to confocal microscopy. The source for this will be the Spectron mode-locked Nd:YAG pumped Spectra-Physics 3500 cavity dumped laser. The amplified femtosecond laser laboratory remains essentially unchanged.

LASER LOAN POOL Mike Towrie

In 1995 there have been a total of 72 months of laser loan time with only 5 % down-time which was predominantly due to the breakdown of the fluoride excimer.

The pulsed nanosecond lasers have been used in a wide range of gas phase experiments such as electron beam diffraction (Fluendy/Rankin, Edinburgh), jet spectroscopy of nucleic acid bases (Jones, Edinburgh) and organometallic intermediates (Ellis, Leicester) and for reaction dynamics (Rieley, Liverpool & Smith, Birmingham). The CW lasers were used to continue the work of Prof. Walsh of Reading on silylenes, for the study of C-H activation photochemistry (Lees, UoCL) and of crystal waveguides (Wilkinson, Southampton). MRC and RAL have collaborated in the polarisation of xenon gas via spin exchange with optically pumped rubidium vapour using the titanium sapphire laser. Very large NMR signal enhancement is produced by the polarisation of xenon and this holds great potential for monitoring blood perfusion in the brain or in tumours.

A breakdown of the results of last years consultation with the user community with regards to the loan pool upgrade is shown in the table below. The demand for flexible, broadly tunable pulsed lasers is clear and, in response to this, the LSF bought three pulsed lasers during this first year of the upgrade.

A Lambda-Physik Scanmate OPPO will, for the first time, provide access to a high resolution IR source. This system is a hybrid using tested dye technology combined with an optical parametric oscillator system. The OPPO was commissioned in Birmingham (Smith and Sims) in April. Using the UV-IR capabilities Dr. Sims will probe kinetic reactions and pump energy transfer processes at ultra-low temperatures using the new CRESU apparatus recently built there.

Two Spectron SL805 Nd:YAG lasers were bought to replace the existing Spectron SL800 and the JKHY750 lasers. The latter have been given to Sims (Birmingham) and Jones (Edinburgh) respectively. The PDL-2 dye laser specifications and reliability remain good and these have been retained.

Outline specifications the above systems are shown in table 3. For more detailed information please contact M.Towrie. (Tel 01235 446141, Fax 01235 44588, E-Mail MT2@RL.AC.UK).

Table 2. Summary of questionnaire returns

LASER	Percentage
nanosecond UV-IR tunable	60%
femtosecond-picosecond tunable ($\mu\text{J-mJ}$)	13%
CW tunable	11%
nanosecond VUV	8%
Nd:YAG	4%
Excimer	2%
Diode lasers	2%

VISITING RESEARCH STAFF AND STUDENT TRAINING

The LSF hosted one post doctoral RA from IC and another funded by the EU Human Capital and Mobility Programme. A total of seven students received training: two MSc students, a visiting student from Germany and four students on industrial placement.

Table 3. Lasers and techniques available at the Rutherford Appleton Laboratory's LSF

	LASERS	TECHNIQUES AVAILABLE and APPLICATIONS
R A L B A S E D L A S E R S	Two excimer pumped dye lasers (205 nm to 900 nm 10 ns pulse length/20 mJ/30Hz/ 0.4cm ⁻¹ bandwidth)	ns time-resolved resonance Raman spectroscopy, Raman, fluorescence, laser flash photolysis, REMPI. Delays from ns to μ seconds
	CW argon-Ion (525, 514, 488, 457, 364, 351 nm)	Raman spectroscopy
	Laser-plasma x-ray source (3, 1, 0.7 nm) 1 W average power (200 mW/Sr), 100 Hz, 3.5 krad/s dose rate at specimen, 10 MRad/min (specimen in chamber).	DNA damage and repair, lithography, microscopy, calibration.
	UV fluoride excimer laser (157, 193, 222, 248 nm/400 mJ/20 ns or 4 ps for 248 nm using YAG laser seed pulse 100 Hz).	DNA damage and repair, flash photolysis, singlet oxygen reactions, mutation studies.
	CW argon-ion (all-lines) pumped mode-locked titanium sapphire laser (<100 fs, 10 nJ, 750-850 nm) amplified to 300 μ J fundamental, 10 μ J SHG.	Ultra-short pulse measurements and techniques. Femtosecond pulse interactions e.g. multielectron dissociative ionisation studies. Experiments in femtosecond VUV generation for spectroscopy.
	⊕ CW argon-ion pumped mode-locked titanium sapphire laser. 50 kHz Nd:YAG pumped regenerative titanium sapphire amplifier. (125 fs, 760 - 840 nm, and 10 μ J + Tunable OPAs). Independently tunable pump and probe beams.	Ultra-fast Raman and transient-absorption spectroscopy.
	Mode-locked Nd:YAG pumped cavity dumped dye laser. 82 MHz, 1 nJ, 550-650 nm.	Confocal microscope. (<200 ps time-resolution, sub- micron spatial-resolution). Distribution of photosensitisers (for photodynamic therapy) in tumour cells.
L A S E R O A N P O L O	Quanta-Ray GCR-11 Nd:YAG (1064 nm/320 mJ, 532 nm/155 mJ, 355 nm/70 mJ, 266 nm/30 mJ, 10Hz.) With PDL-3 dye laser. (390-900 nm/40 mJ/ <0.08 cm ⁻¹)	Spectroscopy, photolysis and reaction kinetics.
	1) Spectron SL805G Nd:YAG (1064 nm/1100 mJ, 532 nm/500 mJ, 355 nm/260 mJ, 266 nm/90 mJ, 10 Hz.) With PDL-2 laser (400-900 nm/80 mJ/0.1 cm ⁻¹) WEX SHG 260-320	Spectroscopy, photolysis and reaction kinetics.
	2) Spectron SL805G Nd:YAG (1064 nm/1100 mJ, 532 nm/500 mJ, 355 nm/260 mJ, 266 nm/90 mJ, 10 Hz) With PDL-2 laser (400-900 nm/80 mJ/0.1 cm ⁻¹) WEX SHG 260-320	Spectroscopy, photolysis and reaction kinetics.
	Coherent Scanmate OPPO (Continuum 8010 pump laser) OPPO (420-2200 nm, 40 mJ max.) SHG (210-420 nm, > 5% conversion) DFG (2200-4500 nm, 5% conversion)	Spectroscopy, photolysis and reaction kinetics.
	Spectra-Physics 2025 argon-ion (5 W visible/0.4 W UV/ single line, CW)	Gas phase kinetics, particle image velocimetry and semiconductor science.
	Spectra-Physics CW 3900 titanium-sapphire laser (680-1040 nm/800 mW max, 50 GHz or <200 MHz bandwidth)	IR Raman studies, fiber amplifier and waveguide studies.
	Questek fluorine excimer laser (157 nm/5 mJ, 193 nm/200 mJ, 249 nm/300 mJ, 351 nm/200 mJ, 0-50 Hz)	Photolysis, ablation and laser ionisation source.

⊕ Becoming available for scheduled experiments in 1995/96.

1WATT PLASMA X-RAYS SOURCE AT $h\nu \sim 1\text{keV}$

ICE Turcu, IN Ross and C McCoard

Rutherford Appleton Laboratory

Soft x-rays of $\sim 1\text{nm}$ wavelength are a strong contender for the production of 1Gbit DRAM microchips which require transistor gates of $\sim 180\text{nm}$ width (Ref. 1). Compact synchrotrons, generating tens of watts of x-rays per beam into ten beams or more, will constitute the main source for large-scale production of such microchips. Nevertheless, such machines require a relatively large initial investment, occupy extensive factory space and have a relatively high running cost. Therefore, smaller, laboratory size, x-ray sources will be required to generate the power of only one synchrotron beam to be used either by large companies for R&D work or by small companies with limited production.

The use of excimer lasers for generation of such plasma x-ray sources has the following advantages:

- (1) commercial lasers deliver average powers up to 1kW,
- (2) the UV wavelength is absorbed in a high (near solid) density plasma, resulting in very efficient emission of plasma radiation,
- (3) the broad bandwidth of excimer transition allows direct amplification of picosecond pulses, therefore generating GW peak powers.
- (4) Good extraction of excimer laser energy in picosecond pulses is achieved by amplifying trains of pulses with $\sim 2\text{ns}$ separation, corresponding to the repumping time of the excimer laser. The $\sim 2\text{ns}$ separation of laser pulses is sufficient for the plasma generated by the previous pulse to dissipate, such that each picosecond laser pulse generates a picosecond x-ray pulse.
- (5) the good quality beam (near diffraction limit) and short wavelength allow tight focusing. This, together with the high peak power (3) generate very high power densities on target ($>10^{15}\text{W}/\text{cm}^2$), resulting in a very high plasma temperature ($\sim 1\text{keV}$) and therefore very high energy conversion efficiency ($\sim 10\%$) from laser to x-rays at $\sim 1\text{nm}$ wavelength. The high plasma temperature leads to very efficient radiation cooling of the x-ray target (steel or copper tape).
- (6) the high breakdown threshold of the UV laser in helium gas allows operation of the x-ray chamber at atmospheric pressure which results in: stopping of target debris, target cooling and elimination of thick x-ray windows for extracting the radiation from the x-ray chamber.

Figure 1 shows our concept (Ref. 2) for a high power excimer laser-plasma x-ray source based on commercial lasers and consisting of : picosecond oscillator, temporal and spatial multiplexed, excimer preamplifier laser, excimer final amplifiers and x-ray chamber filled with He gas at atmospheric pressure. Such a system was used with only one KrF excimer laser amplifier (Lambda 210Fi with Nova tube) to establish a new record of x-ray average power at $\sim 1\text{nm}$ wavelength from a non-synchrotron source.

With a steel tape target in the chamber, the plasma converts 10.4% ($2.5\%\text{sr}^{-1}$) of the focused laser energy into 1.4nm x-rays, and with a copper tape target the conversion efficiency is 5.4% ($1.3\%\text{sr}^{-1}$). When the laser system is operated at 80Hz repetition rate, the x-ray average power emitted by the plasma is 1.5W ($350\text{mW}/\text{sr}$) at 1.4nm wavelength and 0.9W ($220\text{mW}/\text{sr}$) at 1nm .

This work was partially supported by the Advanced Research Project Agency of the US Government.

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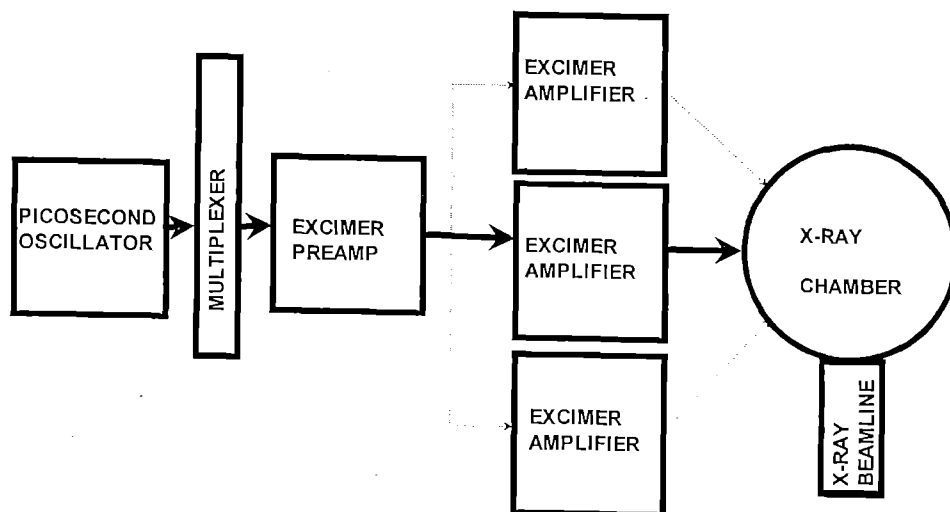


Figure 1 High power excimer laser-plasma X-ray source

DEVELOPMENT OF A LASER-BASED FLUORESCENCE MICROSCOPE WITH SUB-NANOSECOND TIME RESOLUTION

A.D. Scully^{1,3}, A.J. MacRobert², A.W. Parker¹ and D. Phillips³

¹ Central Laser Facility, Rutherford Appleton Laboratory

² National Medical Laser Centre, University College London Medical School

³ Department of Chemistry, Imperial College

INTRODUCTION

Quantitative fluorescence microscopy using new highly sensitive array detectors, and the application of laser-scanning confocal optics has made considerable impact recently on microscopic studies of biological and solid samples, and interfacial phenomena. The extension to time-resolved detection and imaging with sub-nanosecond time resolution will enhance the range and complexity of the many applications of laser microfluorimetry. This non-invasive technique offers the key practical advantages of spectroscopic, temporal and spatial selectivity which derive from the coherence of the laser source. The main objective of the development of this apparatus in the LSF is to exploit the convergence of these techniques and to combine them in studies of fluorophores in heterogeneous samples, not only biological, on a microscopic scale. Topics to be studied include the 4-dimensional mapping of intracellular fluorescence from photosensitizer molecules used in the photodynamic therapy (PDT) treatment of tumours, evanescent wave fluorescence microscopy and photophysical measurements involving polymeric substrates. This is to be achieved by constructing a laser microfluorimeter capable of sub-nanosecond time resolution and sub-micron spatial resolution using confocal optics to provide a 3-dimensional

representation of the fluorescence intensity, lifetime, spectroscopic and polarization data.

INSTRUMENT DEVELOPMENT

The work will be carried out in two stages:

1. construction of sub-nanosecond fluorescence imaging capability
2. upgrade to confocal imaging capabilities

Stage 1 Instrument Development

A schematic diagram of the apparatus developed in Stage 1 is shown in Figure 1. A mode-locked Nd³⁺/YAG laser operating at 82 MHz synchronously pumps a cavity-dumped dye laser. The dye laser pulse repetition rate is 0.8 KHz. The output from the dye laser is directed to a ≈ 5 m PMMA fibre optic delay line (ϕ 1 mm) which is required for synchronization of the arrival of the laser pulse at the object plane (OP) of the microscope and the triggering of the high voltage gating pulse to the photocathode of the gated optical imager (GOI) via the photodiode (PD). The output from the fibre is magnified and imaged at the back focal plane (BFP) of the microscope objective (MO) which is mounted in a commercially available optical microscope. The microscope is

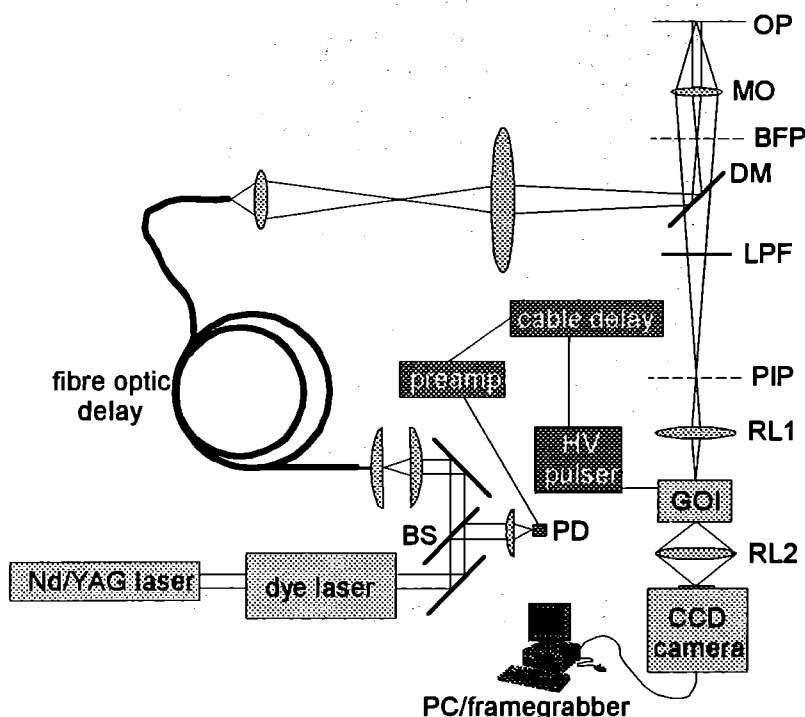
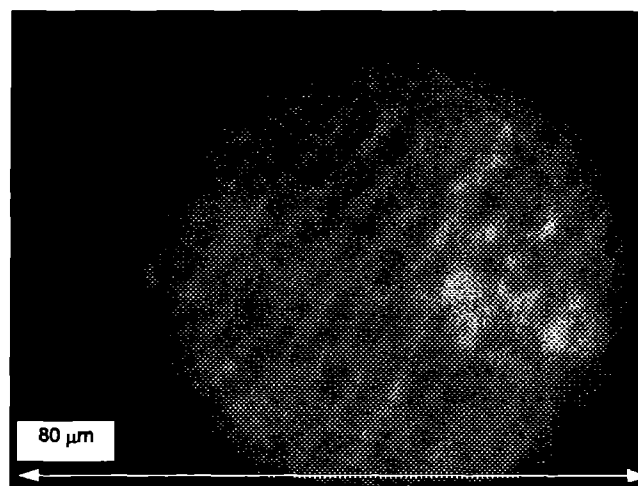
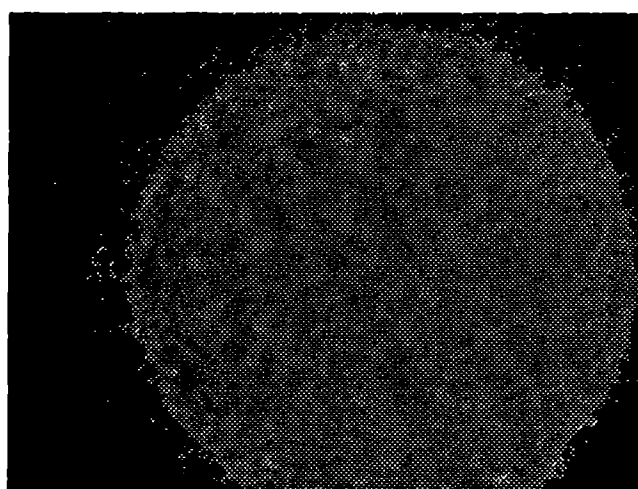
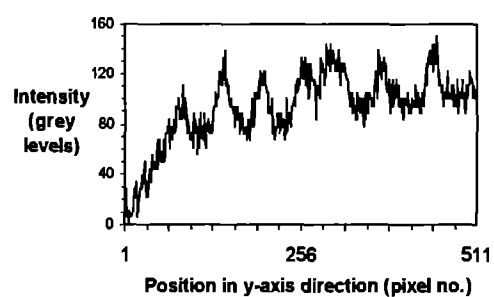


Figure 1. Schematic diagram of the time-resolved fluorescence microscope



Fluorescence intensity map



Fluorescence lifetime map

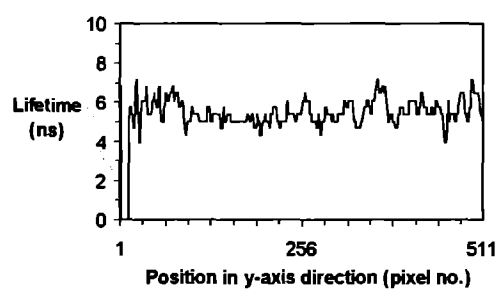


Figure 2. Fluorescence images of onion cells stained with a phthalocyanine photosensitizer

photocathode of the GOI by a relay lens (RL1). The output signal is projected from the phosphor of the GOI with a gain of $\approx 10^3$ after amplification in a single-stage microchannel plate. This image is then relayed via a second relay lens (RL2) to the CCD chip of an 8-bit scientific grade video camera capable of on-chip integration times of up to ≈ 60 seconds. Camera control and image analysis is performed using a commercially available package which includes a framegrabber card installed in the computer. Selection of delay times between the gating of the GOI and the arrival of the laser pulse at the sample in intervals of 0.5 ns is achieved using a cable delay unit built in-house. Software required for analysis of the time-resolved images and generation of fluorescence lifetime maps have been written during the course of this work.

Results from Stage 1

Results of preliminary measurements of the distribution of a phthalocyanine photosensitizer in onion cells is shown in Figure 2. This sensitizer is used in the photodynamic therapy (PDT) treatment of tumours. The wavelength of excitation was 610 nm and fluorescence emission was detected through an RG665 long-pass filter. The upper image is the fluorescence emitted from the cells during the first 1 ns after the laser excitation pulse arrives at the sample and has a diameter of approximately 700 μm . This image was obtained after on-chip integration of 750 video frames. Approximately 2.4×10^6 laser excitation pulses were incident on the sample during this time. The brighter regions in this image result from the fluorescence from the sensitizer chromophores located at the cell walls. A cross-section through roughly the centre of this image demonstrates the extent of spatial modulation of fluorescent intensity, with a lower intensity fluorescence being observed for the interior of the cells. The lower image in Figure 2 shows the spatial distribution

of fluorescence lifetime of the sensitizer in the cells. This image was generated by calculating the fluorescence decay time at each pixel from images collected at 0 ns, 4 ns, 8 ns and 12 ns after arrival of the excitation pulse at the sample. The same cross-section through this image as used above demonstrates that the fluorescence lifetime of the sensitizer is independent of its position in the cell. Therefore, the spatial modulation observed in the upper image can be attributed to a genuine modulation in sensitizer concentration, rather than an artifact arising as a result of quenching of the sensitizer fluorescence. The fluorescence lifetime of ≈ 5 ns obtained in this work compares well with values reported¹ for this molecule in various molecular environments. Our aim is to now extend these types of measurements to animal cells stained with a range of photosensitizers, as well as intracellular pH and Ca^{2+} fluorescent probes.

Stage 2 Instrument Development

The confocal upgrade to the instrument developed in Stage 1 will incorporate a slit-based "direct field" confocal optical arrangement. A line of excitation will be scanned across the sample and the fluorescence from the sample will be imaged onto the photocathode of the GOI through a confocal adjustable slit assembly. Essentially diffraction-limited spatial resolution will be achieved laterally and sub-micron resolution is anticipated in the axial direction. This will enable the generation of 2-dimensional fluorescence intensity and lifetime maps at various axial positions through the sample and, ultimately, a 3-dimensional rendering of all relevant fluorescence information. This apparatus is currently being constructed.

REFERENCES

1. M. Ambroz, A.J. MacRobert, J. Morgan, G. Rumbles, M.S.C. Foley, D. Phillips, J. Photochem. Photobiol. B: Biol. 22 (1994) 105.