

RAL-88-042

Science and Engineering Research Council
Rutherford Appleton Laboratory
CHILTON, DIDCOT, OXON, OX11 0QX

Science and Engineering
Research Council
Central Laser Facility

Annual Report to the Laser
Facility Committee 1988

RAL-88-042

Central Laser Facility
Rutherford Appleton Laboratory

Science and Engineering

Research Council

The Science and Engineering Research Council does
accept any responsibility for loss or damage arising from
the use of information contained in any of its reports
in any communication about its tests or investigations

PREFACE

The Central Laser Facility of the SERC provides advanced lasers for use by UK polytechnic and University research workers. It is funded by the Science Board of SERC and operated by the SERC's Rutherford Appleton Laboratory. Access to the lasers is through a competitive peer review system which is open to any programme of research in the Science Board area. Uses in other areas are catered for on a contract basis.

Work carried out in association with the CLF in the year to 31 March 1988 is reported here at a scientific and technical level. A more condensed description is available in the RAL annual report.

Two high power laser installations Vulcan (Nd glass) and Sprite (KrF) comprise the major provision of the CLF and their continuing development and operation has produced exciting results in the last year. Notable among these have been a significant improvement in thermonuclear yield in laser driven implosions using a novel irradiation pattern; the first direct observation of coalescence of laser driven shocks, which is an essential feature of schemes for laser fusion; continuing work at the forefront of XUV laser research, with demonstration of laser action at the shortest ever wavelength (44.5Å) in a collaborative experiment using a Japanese laser facility; novel observations of crystal lattice deformation under high pressure impulses; elegant Thomson scattering data from plasmas in beat wave acceleration studies and new laser plasma interaction studies with ultra short pulses on Vulcan and Sprite.

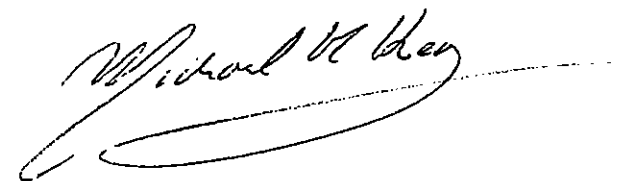
Development of the lasers has been vigorous and a major upgrade of Vulcan has been carried out, while a notable world first was achieved with Sprite by amplifying picosecond pulses to the highest ever energy 2.5J.

The Laser Support Facility has attracted a great diversity of chemists, biologists and physicists using both its RAL based picosecond and nanosecond dye laser laboratories and its loan lasers. Picosecond measurements in biology have been particularly interesting with observation of transient states in photosynthesis and studies of the conformational relaxation of proteins. Laser based methods of investigating DNA repair mechanisms have produced novel results. A

wealth of high quality work in photochemistry and interesting new physics, including trials of high repetition rate X-ray sources for scanning X-ray microscopy, completes a picture of vigorous activity.

The preparation of proposals for the future development of the CLF has been an important task shared by both facility users and staff. A major proposal is to replace the Vulcan Nd glass laser in the longer term with a unique KrF laser facility SuperSprite, details of which are included in this report. The Phase II plans are currently being assessed by a Review Panel reporting to the Science Board, where a decision will be made to take effect in the financial year 1989/90. The decision will have a major impact on the future of the CLF and will no doubt be influenced by the determination and enthusiasm of the facility users and staff reflected in their many achievements of the last year.

I am hopeful that these efforts will be rewarded by a bright future and I am pleased to record here my own appreciation of the dedication of facility users and staff alike.



Dr M H Key

CONTENTS

PREFACE

Introduction

A HIGH POWER LASERS SCIENTIFIC PROGRAMME

Introduction

A1 XUV AND X-RAY LASERS AND OTHER APPLICATIONS OF
LASER PRODUCED PLASMA SOURCES

A2 LASER PLASMA INTERACTION AND ENERGY TRANSPORT

A3 LASER COMPRESSION WITH DENSE PLASMAS

A4 THEORY AND COMPUTATION

A5 SPRITE OPERATION AND DEVELOPMENT

A6 VULCAN OPERATION AND DEVELOPMENT

A7 INSTRUMENTATION

B LASER SUPPORT FACILITY

Introduction

B1 CHEMISTRY

B2 BIOLOGY

B3 PHYSICS

B4 LASER DEVELOPMENT AND NON LINEAR OPTICS

B5 LASER SUPPORT FACILITY OPERATIONS AND DEVELOPMENT

C CENTRAL LASER FACILITY PUBLICATIONS

Introduction

C1 JOURNALS AND BOOKS

C2 PUBLISHED CONFERENCE PROCEEDINGS

ACKNOWLEDGEMENTS

INTRODUCTION

P T Rumsby (RAL)

The Central Laser Facility (CLF) is funded by the Science Board of the Science and Engineering Research Council and in the year covered by this report, April 1987 to April 1988, its allocation was £2.94M.

The programme is managed by the Laser Facility Committee (LFC) whose membership is given in Table 1. The LFC delegates responsibility to the Director of the Rutherford Appleton Laboratory for the operation of the Facility.

During the year the CLF used 59 man years of effort. Staff costs and overheads used 49% of the allocation while facility operations and minor development costs used another 34%. The remaining 17% of the total allocation was spent on the Phase 1 programme of capital facility development.

The major facilities provided by the CLF and an indication of their principal scientific applications is given in Table 2.

The Nd glass laser VULCAN is the major facility and about 45% of resources was used to support its programme of research by UK Universities.

The high power UV gas laser SPRITE has now been developed to the point that at least 50% of its time is used to provide UV and X-ray source facilities for University work. About 20% of total operating resources were committed to this work.

Multidisciplinary scientific applications of excimer lasers and excimer pumped dye lasers by UK Universities has been catered for by the Laser Support Facility using about 25% of resources. This also operates a picosecond laser system for scientific applications in physics, chemistry and biology at the CLF and a laser loan scheme for University based experiments.

A target preparation service and a small group giving theoretical support by maintaining computer codes support all CLF activities.

Fig 1 shows the CLF's present Divisional and group structure.

Use of the CLF by UK Universities is free of charge to those successful in a peer review process. Research grants of typically three years duration covering expenditure in University Laboratories and research assistants' salaries may be obtained from the LFC by application to SERC. LFC Grant holders are entitled to bid for time on the CLF's lasers through a procedure described below.

A few percent of the time may also be allocated to bids for short trial experiments from University researchers working in the Science Board area who do not hold LFC grants.

The SERC encourages use of the CLF by industry or other organisations on the basis of payment for the full cost of laser time. Such use is limited to not more than 10% of available laser time.

International collaboration in the research programme is extensive through both collaborative participation in approved experiments of UK University groups and through agreements negotiated with the CLF and SERC.

Selection and scheduling of University experiments using the VULCAN glass laser and SPRITE UV laser is via proposals to one of 4 informal specialist scientific groups which anyone may join. Each has a University Chairman and an RAL secretary (Table 3). Each group discusses the experimental proposals and makes modifications, improvements and collaborative suggestions. More formal proposals based on these discussions are then submitted to an expert panel which places scientific priorities on the proposals. This panel is chaired by the Scientific Programme Chairman. Its members include the five Scientific Group Chairmen, the Chairman of the High Power Laser Scheduling Committee (HPLSC), a representative of the LFC and co-opted experts. A draft schedule which includes as many of the high priority proposals as possible (typically $\leq 50\%$ of proposals), is then prepared, discussed and modified at a meeting of the more formal High Power Laser Scientific Programme and Scheduling Committee (HPLSPSC). All University staff members involved in the programme have voting rights, together with the group secretaries and programme coordinator. The Committee is chaired by the Head of the CLF and is formally advisory to him.

Scheduling of Laser Support Facility lasers is performed in a different way. The LFC has delegated LSF Management duties to a panel whose membership is given in Table 5. Selection of experiments requiring loan pool lasers or use of the lasers based at the CLF is carried out by this panel and scheduling is performed by an ad hoc committee of users chaired by the CLF Unit Head.

Scheduled experiments are supported by the operating budget of the CLF which covers travel and subsistence costs for University users and minor engineering work and other costs of experiments.

The experimental programme is linked to theoretical work in Universities which is supported by grants from the LFC and coordinated by the Theory and Computation Scientific group.

There is a large and diverse community of University researchers participating in the programme. They number about 150 staff and research students. Most are physicists and chemists, but recently there has been a rapid growth of participation from biologists.

TABLE 1

Membership of the Laser Facility Committee 1987/88

Professor G J Pert (Chairman) Dept of Physics University of York	* Dr C L S Lewis Dept of Pure and Applied Physics Queens University Belfast
Professor R M Dixon FRS Dept of Theoretical Chemistry University of Bristol	Professor D Phillips Dept of Chemistry Royal Institution of Great Britain
+ Professor R J Donovan FRSE Dept of Chemistry University of Edinburgh	Dr D C Robinson UKAEA Research Group Culham Laboratory
Professor M G Haines The Blackett Laboratory Imperial College of Science and Technology	Professor W Sibbett Dept of Physics St Andrews University
* Dr T A Hall Dept of Physics University of Essex	* Professor J P Simons Dept of Chemistry University of Nottingham
Dr T A King Atomic Molecular and Polymer Phys Group Schuster Laboratory University of Manchester	Dr M Vaughan Procurement Executive Royal Signals & Radar Est., MOD Great Malvern
+ Dr M J Lamb Dept of Pure and Applied Physics Queens University Belfast	Dr C W Wharton Dept of Biochemistry University of Birmingham

* until 31 August 1987

+ from 1 September 1987

TABLE 2

Major Facilities and Programmes at the CLF

Nd glass Laser facility	Laser Support Facility	High power KrF laser facility
VULCAN	LSF	SPRITE
Laser plasma Interactions and energy transport	Photo chemistry Photo biology Non linear optics	Ultra high intensity pico-second laser source
Laser compression of matter and dense plasmas	High field atomic physics Laser loan pool Raman Facility	Applications of high brightness laser produced plasma X-ray sources
XUV lasers and applications of laser produced plasma sources	Picosecond laser	Short pulse laser plasma energy transport processes
Target Preparation Service		
Theoretical support service		

TABLE 3

High Power Laser Scientific Groups

Groups of the HPLSPSC	Chairman	Secretary
Laser plasma Interaction and energy transport	Dr O Willi	G Tallents E Turcu
Laser Compression and dense plasmas	Dr T A Hall	A Ridgeley
XUV lasers and applications of laser produced plasma sources	Dr C L S Lewis	M Grande
Theory and Computation	Dr R A Cairns	Dr S J Rose
Facility Development	Dr P T Rumsby	Mr J E Boon
Scientific programme coordinator	Prof. S Ramsden	

TABLE 4

Laser Support Facility Panel 87/88

Professor R M Dixon - Chairman
Bristol University

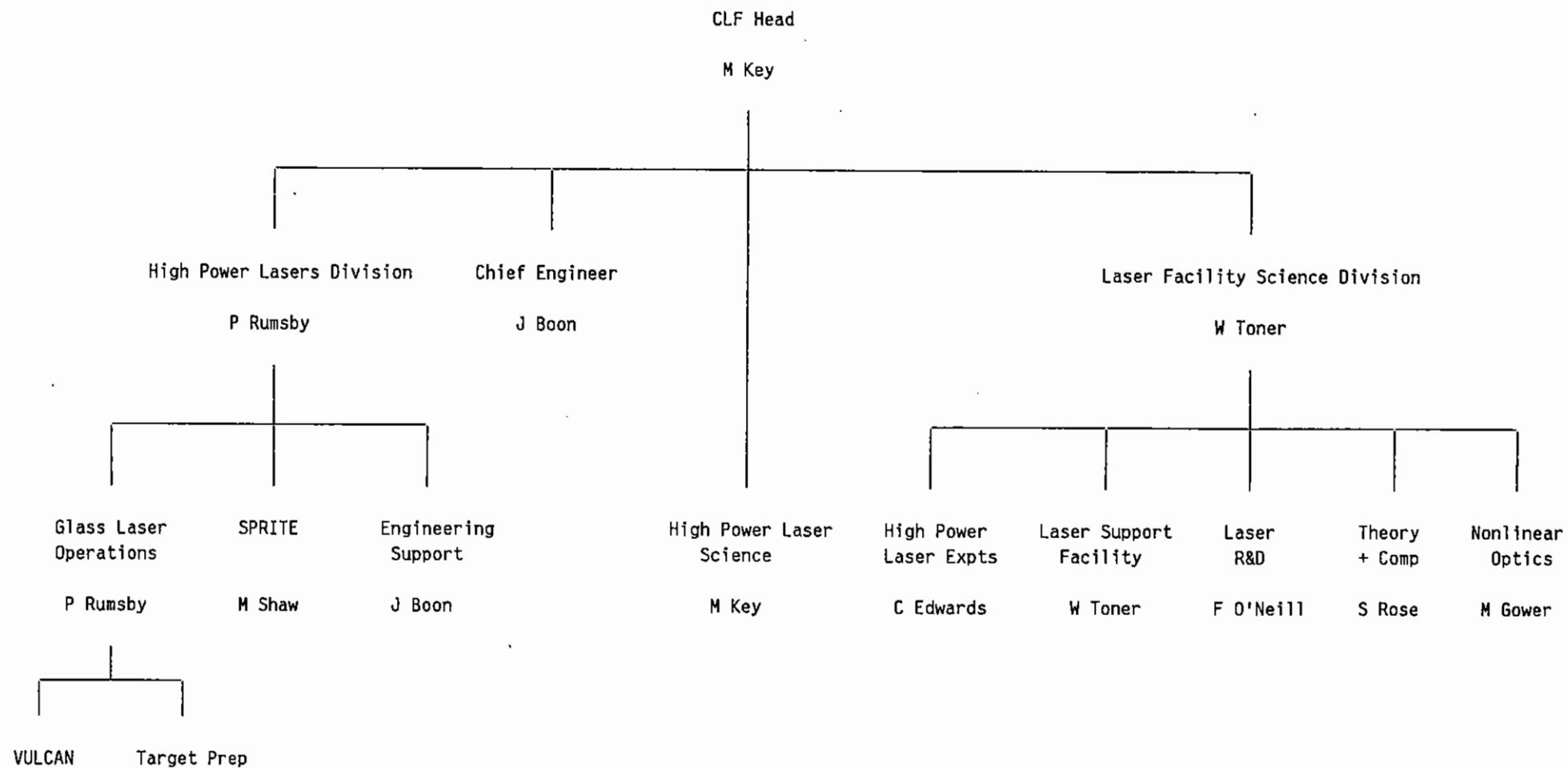
Professor R E Hester for Chemistry Committee
York University

Dr C W Wharton for Biological Sciences Committee
Birmingham University

Dr K J Burnett
Clarendon Laboratory, Oxford University

Prof. M C W Evans
University College, London

Prof. I W M Smith
Birmingham University



Central Laser Facility Structure

Fig 1

A	HIGH POWER LASERS SCIENTIFIC PROGRAMME	Pages
A1	XUV AND X-RAY LASERS AND OTHER APPLICATIONS OF LASER PRODUCED PLASMA SOURCES	5 - 43
A2	LASER PLASMA INTERACTION AND ENERGY TRANSPORT	44 - 76
A3	LASER COMPRESSION AND DENSE PLASMAS	77 - 92
A4	THEORY AND COMPUTATION	93 - 116
A5	SPRITE OPERATION AND DEVELOPMENT	117 - 134
A6	VULCAN OPERATION AND DEVELOPMENT	135 - 141
A7	INSTRUMENTATION	143 - 154

SECTION A

INTRODUCTION

The period from April 1987 until the shutdown for the VULCAN upgrade at the end of February 1988 has seen steady exploitation of the flexibility of the target areas. TAE has run scheduled experiments for 22 weeks, TAW for 19 weeks and TA2 for 21 weeks. The even spreading of experiments between the target areas in this way makes for easier changeover between experiments and increases the time available for setting up more complex systems.

There has recently been much international interest in implosions of large high aspect ratio shells filled with deuterium and tritium, producing high neutron yields. In work on VULCAN the scaling laws have been investigated and, in addition, a twenty fold increase in yield observed with a novel type of non-uniform irradiation. Also, a direct experimental observation has been made of the coalescence of two shocks generated by a shaped laser pulse - essential in laser fusion schemes for the adiabatic compression of the fuel pellet.

Scaling of the XUV laser recombination scheme to shorter wavelengths has been studied in a collaborative experiment at the Institute of Laser Engineering, Osaka University, and gain observed on Mg XIII H α at 45.5Å which is only 2Å away from the "water window". Improvements in the modelling and analysis have brought theory and experiment into better agreement in some of the earlier work and there is now a formal collaborative programme with France on XUV laser research.

There is currently much interest in obtaining experimental data on the radiation opacity of stellar materials because recent theoretical work has suggested that earlier calculations, upon which almost all theories of stellar structure and evolution are based, may be low by a factor of two. This very important problem is being investigated using point projection absorption spectroscopy of dense laser produced plasmas.

Diffraction of X-rays from laser produced plasmas has been used in a novel method of measuring crystal structure and lattice spacing in laser-shocked crystals. This new technique has great potential for dynamic studies of material properties such as phase change and plasticity.

Building upon earlier success in producing exceptionally uniform plasmas using multiphoton ionisation very elegant Thomson scattering results have been obtained in the beat wave experiment but no significant plasma wave has been observed as yet.

Some preliminary experiments have been carried out with the new short (< 30 psec) pulse available on VULCAN. These include studies of short pulse energy transport and ablation to produce heating at higher density for shorter wavelength XUV laser schemes.

A new, small, target area (TA4) has been brought into operation on VULCAN using only part of the amplifier chain but at higher repetition rate. It is intended primarily for biological work, feasibility studies and instrument development. An interesting example of the latter is work on the use of CCD detectors instead of X-ray film.

This has been a fruitful year for SPRITE. Nearly 2000 laser shots were fired - about 50% on experiments and over half of these on contract work. A notable 'world first' has been the amplification of 3.5 psec pulses to 2.5 J and the use of these in the production of hot plasma at near solid density. The operation of short pulse Raman amplifiers has been demonstrated and SUPER-SPRITE has been adopted as the preferred choice for the future high power laser system being considered for Phase II of the CLF development programme.

S A Ramsden

A1	XUV AND X-RAY LASER AND OTHER APPLICATIONS OF LASER PRODUCED PLASMAS	Pages
A1-1	Further Analysis of the F IX X-ray Laser Experiment	5 - 9
A1-2	ILE/UK Joint Experiment on X-ray Lasers	9 - 15
A1-3	Li-Like Recombination XUV Laser Schemes	15 - 19
A1-4	Na-Like Cu Recombination Laser Experiment	19
A1-5	Mn/F Recombination/Photopumping Experiment	19 - 21
A1-6	Al Strip Targets	21 - 22
A1-7	Coronal Plasma Absorption Spectroscopy	23 - 25
A1-8	Absorption Spectroscopy of a Dense Aluminium Plasma	25 - 27
A1-9	Spectroscopy of Short Pulse X-Ray Sources	27 - 29
A1-10	Investigation of Short Pulse, High Brightness X-Ray Sources	29 - 32
A1-11	Experimental Study on Blow-off Plasma from X-ray Laser Targets	32 - 34
A1-12	X-Ray Diffraction from Shocked Crystals	34 - 36
A1-13	X-Ray Sources Generated using 30ns KrF Laser Pulses at Energies up to ~ 30 J	36
A1-14	X-Ray Sources for Small Angle Scattering	37
A1-15	Soft X-ray Contact Microscopy of Biological Specimens: Aluminium Coated Silica Nitride Windows used as U/V Filters	37 - 41
A1-16	Soft X-ray Contact Microscopy using TA4	41 - 43

A1-1 FURTHER ANALYSIS OF THE F IX X-RAY LASER EXPERIMENT

R A Smith, G Kiehn and O Willi (Imperial College).

C Chenaïs-Popovics, M Grande, R Hawkins, C Hooker, M Key, S Rose, W Shaikh and T Tomie (Rutherford Appleton Laboratory).

R Corbett, C Lewis and C Regan (Queen's University, Belfast).

G Pert (York University).

S Ramsden (Hull University).

A more detailed analysis of the data from the fluorine x-ray laser experiment⁽¹⁾ carried out in July 1986 has been undertaken. This analysis confirmed earlier work which showed that gain had been observed at 81 Å in a freely expanding laser produced hydrogenic fluorine plasma. This was the first example of sub 100 Å laser action, and at the time was the shortest wavelength at which amplification had been observed.

The complete analysis however indicated the existence of three distinct gain regimes characterised by different plasma energies. The three regimes showed absorption ($\sim 0.5 \pm 0.5 \text{ cm}^{-1}$), high gain ($5.5 \pm 1.0 \text{ cm}^{-1}$) and low gain ($3.0 \pm 1.0 \text{ cm}^{-1}$) respectively on the fluorine Balmer H_α transition. This is the first observation of multiple gain regimes in an x-ray laser, and is well fitted by numerical simulations⁽²⁾.

The fluorine x-ray laser is a short wavelength scaling of the previously successful hydrogenic carbon fibre recombination scheme^(3,4). Using fluorine rather than carbon as a lasing ion gives an isoelectronic Z^{-2} scaling of laser wavelength from 182 Å to 81 Å. The corresponding pump laser intensity scales as Z^4 , but this must be accompanied by the correct scaling of plasma energy, mass and expansion rate. These parameters are controlled by the initial target dimensions, pump laser intensity and pulse length, and are critical for producing maximum peak gain. The theoretical matching of these parameters for fibre targets has been covered in previous reports⁽⁵⁾.

The experimental method has already been described in detail in earlier reports⁽¹⁾. In brief, 7 µm carbon fibres coated with 0.5 µm of LiF were irradiated in line focus geometry with 70 ps pulses of green 0.53 µm light from the VULCAN glass laser. Three opposing pairs

of 7 mm x 25 µm line foci were superimposed on target to give the required energy absorption and uniformity of irradiance.

A pair of time resolved flat field XUV spectrometers were used to observe line emission over a 40 - 140 Å spectral window. One of the instruments was aligned along the fibre axis, while the other viewed the full length of the plasma at 35° to the axis. The spectral resolution of both instruments was 0.5 Å, with temporal resolution of < 100 ps over a 3 ns window. The use of two such spectrometers allowed shot to shot variations in pump laser energy to be taken into account, and single shot measurements of gain as a function of time to be made. Plasma length was recorded with active and passive x-ray pinhole cameras, filtered to observe $\sim 500 \text{ eV}$ photons. Coupled energy was measured with an array of plasma calorimeters and plasma uniformity along the axis of the fibre was investigated using an active temperature monitor comprising a space resolving slit, beryllium filter pack, x-ray phosphor and image intensifier. Fibre targets were aligned to $\pm 5 \text{ µm}$ in position and 10^{-3} rad in angle using a split field microscope system. The six line foci were individually aligned on target; part of each beam could be masked to produce line foci shorter than 7 mm. A comprehensive data set was collected for a range of target lengths from 1 to 5.5 mm, and a range of coupled energies from 4 to 20 J cm⁻¹.

The novel diagnostic feature of this experiment as compared to our earlier work on carbon fibres was the use of simultaneous axial and transverse time resolved spectroscopy. Evidence for multiple plasma energy regimes in the data set was first seen in the transverse spectra⁽⁶⁾, and was found to be supported by results from all the other diagnostics.

Figure A1.1 shows a plot of transverse H_α line intensity against plasma length at 250 ps after the laser pulse (the time at which peak axial H_α emission occurs in spectra from long fibre shots). Clearly the data points fall onto two distinct straight lines, representing "medium energy/unit length" ($9 \pm 2 \text{ J cm}^{-1}$) and "high energy/unit length" ($16 \pm 1 \text{ J cm}^{-1}$) shots. A third set of "low energy/unit length" shots ($4 \pm 2 \text{ J cm}^{-1}$) are not represented on this plot. Similar and consistent results are found on a shot to shot basis for transverse H_α intensity at a range of later times, and also for H_β and He like line intensity variation with length. Both the plasma calorimeter (Figure A1.2) and integrated pinhole camera brightness

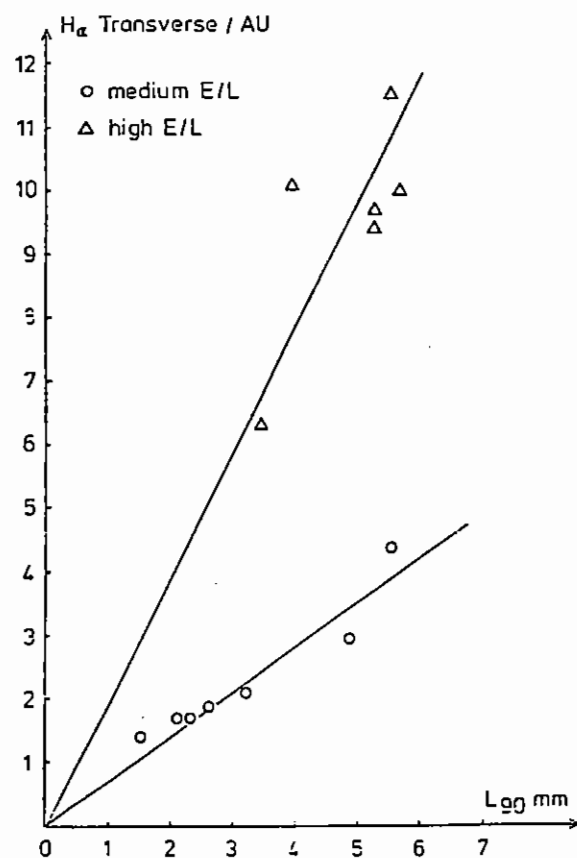


Fig. A1.1 Transverse H_α intensity versus length at 250 ps.

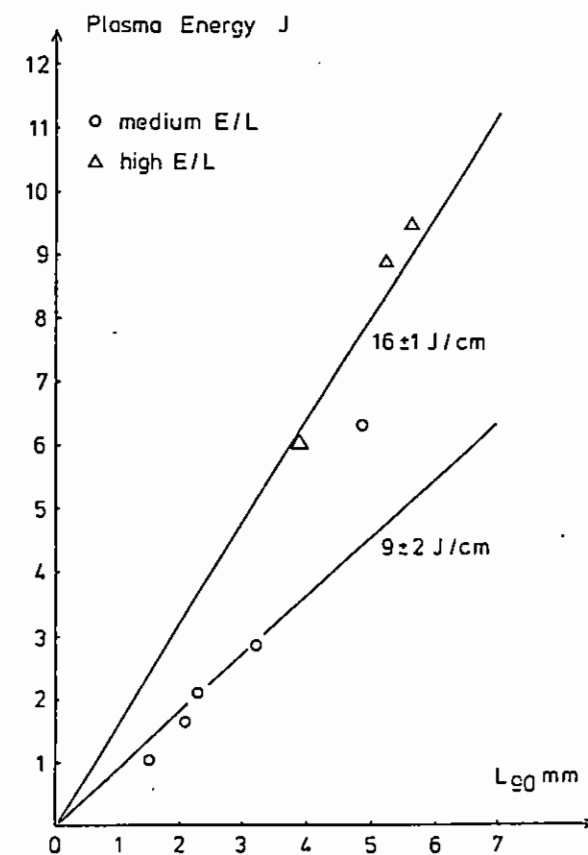


Fig. A1.2 Plasma calorimeter energy (J) versus length.

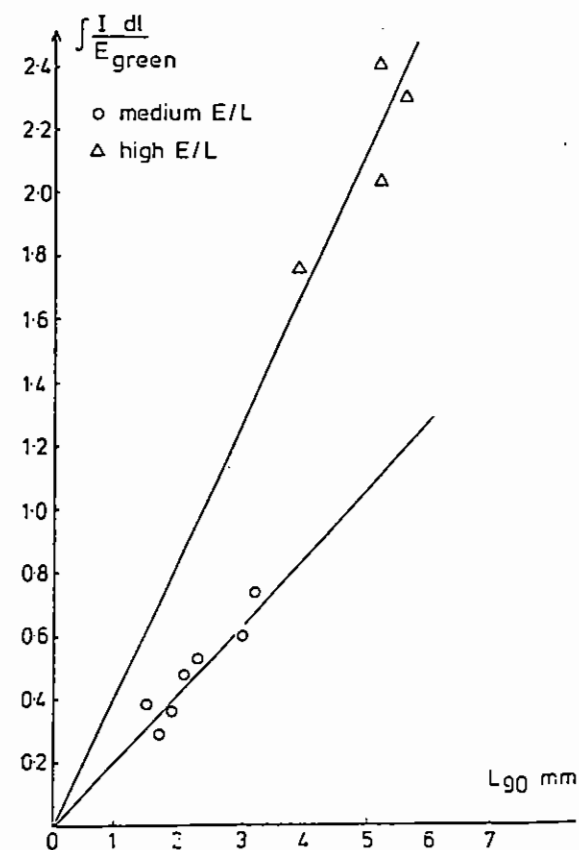


Fig. A1.3 Integrated pinhole camera brightness normalised to green energy versus length.

results (Figure A1.3) show identical splitting into two discrete regimes for these shots.

The two regimes from Figure A1.1 which we shall call medium E/L and high E/L have several distinct and characteristic features. Medium E/L transverse spectra are dominated by He like lines, notably $3d^3D-2p^3P$, $3d^1D-2p^1P$ and $3p^3P-2s^2S$. High E/L transverse spectra have He like lines and hydrogenic lines, (in particular H_α and H_β) of comparable intensity. Comparing the time evolution of transverse H_α emission, medium E/L shots show peak emission at ≈ 450 ps after the pump laser pulse, while high E/L shots show peak emission at ≈ 300 ps, with a brightness a factor of 2 higher than for medium E/L shots of comparable length. These results strongly suggest that these two regimes are characterised by different temperatures, ion populations and cooling rates. For this reason we treat them as separate entities when looking for evidence of XUV amplification.

Figure A1.4 shows a plot of axial/transverse H_α intensity against length at 250 ps after the laser pulse. The length of plasma (L_{90}) was taken to be that from which 90% of the integrated brightness in the pinhole camera image was emitted. The medium and high E/L shots fall onto two distinct growth curves. Fitting with a least squares method gives gain coefficients of $5.5 \pm 1 \text{ cm}^{-1}$ for the medium E/L regime and $3 \pm 1 \text{ cm}^{-1}$ for the high E/L regime.

The lower gain observed in the high E/L shots (even though they have a higher H like ion population) suggests that in this case the plasma is either too hot, inhibiting the population of the $n = 3$ upper laser level at early times, or the H like population density increases Ly_α trapping more than it increases the gain. A third regime is found in shots with low coupled energies ($4 \pm 2 \text{ J cm}^{-1}$). Here, the plasma is too cold, and while there is a significant population of H like ions, the high H like ground state population and lower radial expansion velocity lead to Ly_α trapping, preventing the inversion of the $n = 3-2$ levels. In this low E/L regime, the plasma becomes optically thick for H_α and these shots lie on a downwards curve in Figure A1.1 which saturates, and is fitted by an attenuation coefficient of $-0.5 \pm 0.5 \text{ cm}^{-1}$. Plots of axial H_α intensity against length give similar and consistent values for gain coefficients in the three regimes.

A similar analysis was attempted for the H_β and H_γ lines for which we

expect no significant gain. Plots of axial line intensity against plasma length were linear for these two emission lines, suggesting that there was no gain. A full axial/transverse analysis was however impossible due to the signal to noise ratio for these lines, and no clear conclusion about axial/transverse behaviour could be made. Figure A1.5 shows calculated peak gain as a function of coupled energy for the hydrogenic fluorine system over the range examined⁽²⁾. The code predicts the three gain regimes that we observed experimentally. Below 4 J cm^{-1} there is no significant gain, and this corresponds to the absorption regime. From 10 to 16 J cm^{-1} , gain is constant at $\approx 4 \text{ cm}^{-1}$ (the high gain, medium E/L regime), while for energies greater than 16 J cm^{-1} gain begins to fall off (the low gain, high E/L regime).

Figure A1.6 compares the calculated gain/time behaviour with that found experimentally from the comparison of a short and long shot in the medium E/L, high gain regime at an absorbed energy of $\approx 9 \text{ J cm}^{-1}$. The two are in very good agreement on the magnitude of the peak gain, but the experiment shows peak gain at a later time than the code predicts, and with a broadened temporal profile. The code is however in much closer agreement with experiment than previous work on carbon fibres⁽³⁾. This is due to the inclusion of Stark broadening and p level splitting in the code, both of which reduce the calculated peak gain. It is unclear whether the disagreement on the time behaviour is due to incomplete modelling of the expansion, or to small scale laser plasma instabilities which have been seen in such targets⁽⁷⁾ effects the uniformity of expansion at early times.

In conclusion, the hydrogenic recombination x-ray laser scheme was successfully scaled to shorter wavelength by changing to a higher Z lasing ion. X-ray laser action at $81 \text{ \AA}$ was observed in recombining fluorine plasmas. As predicted by improved code simulations, the system showed three gain regimes as the plasma energy was varied. These regimes were characterised by differing ion populations and expansion rates, and showed absorption, "high gain" and "low gain" on the Balmer H_α transition.

References

1. C Chenais-Popovics et al, RAL 87-041 A 1.2 Annual Report, pp 6-12, (1987).

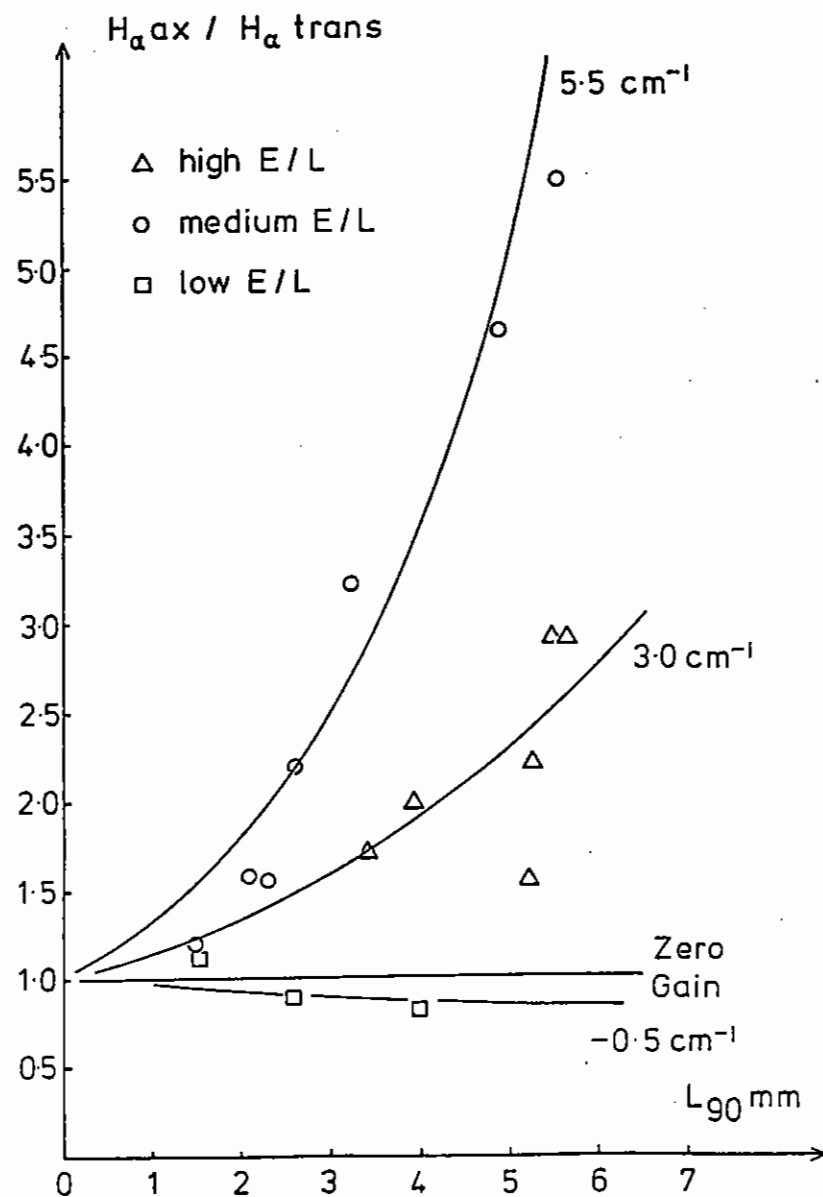


Fig. A1.4 Axial H_α /Transverse H_α versus length at 250 ps.

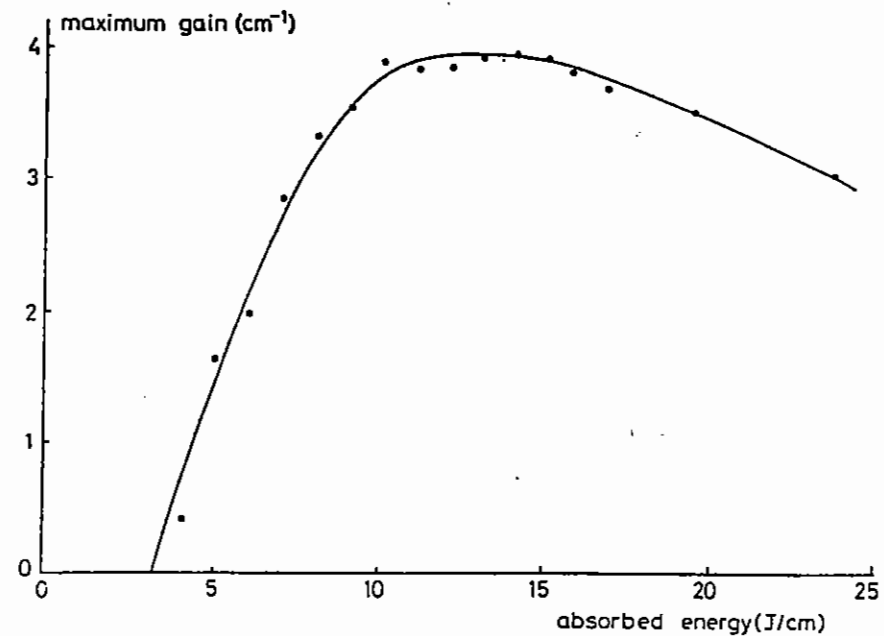


Fig. A1.5 Calculated peak gain versus plasma energy (J cm^{-1}).

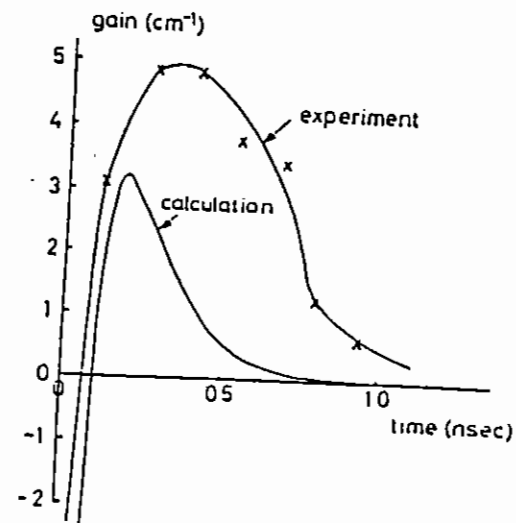


Fig. A1.6 Observed and calculated gain as a function of time.

2. S Rose, Laser Division, RAL, Private communication.
3. C Chenaïs-Popovics et al, Phys. Rev. Lett, **59**, 19 (1987).
4. G J Pert et al, RAL 86-046 A 4.5 A4.22-A4.30, (1986).
5. G J Pert, J. Phys. B, Molec. Phys. **2** No. 12, (1979). M H Key and G J Pert, RAL 86-046, A5.38 (1986). M H Key, RAL 87-041, 100 (1987).
6. G Kiehn, PhD. Thesis, Oxford University, (1987).
7. D Bassett et al, RAL 87-041, Annual Report, pp 17-18 (1987).

A1-2 ILE/UK JOINT EXPERIMENT ON X-RAY LASERS

H Daido, P R Herman, T Jitsuno, Y Kato, E Miura, S Nakai, H Nishimura, H Shiraga, T Tachi, H Takabe, M Takagi, C Yamanaka, M Yamanaka (ILE Osaka),

G J Pert (University of York)

M H Key, P T Rumsby, S J Rose, G J Tallents (Rutherford Appleton Laboratory)

1. Introduction

A new venture in 1988 has been an X-ray laser experiment in collaboration with the Institute of Laser Engineering (ILE at Osaka University) in Japan. The experiment was carried out on the neodymium glass laser facility GEKKO XII. ILE have recently made a successful start to a programme of X-ray laser research using their smaller two beam GEKKO MII neodymium glass laser producing 70 J, 100 psec pulses at 0.53 μm . They had already planned further experiments with GEKKO XII, initially with two of its 12 beams operating at the 0.35 μm third harmonic. The output available in two beams is 300 J in 100 psec at 0.35 μm (compared to 150 J in 100 psec at 0.53 μm available with the six beam VULCAN facility).

A convenient basis for a collaborative experiment arose through the grant by the Japan Society for the Promotion of Science of a visiting fellowship to one of us (MHK) and arising from this, it was proposed to ILE to use the forthcoming opportunity of an X-ray laser experiment on GEKKO XII for a collaboration. The theme of the collaborative

experiment was chosen to be the scaling to shorter wavelength of XUV laser action based on recombination. Emphasis was placed on the potential for obtaining shorter wavelength laser action with GEKKO XII through the advantages of its own shorter wavelength (0.35 μm cf 0.53 μm) and its higher power. With previous experience of laser action in F IX at 81 Å the most promising progression was to sodium XI with its H α transition at 54 Å.

2. Line focus and target alignment

This experiment was the first to use line focus irradiation on GEKKO XII and one of the first experiments at ILE using third harmonic operation. Technical problems required solution to enable sufficiently precise irradiation of X-ray laser targets such as fibres, foils and ribbons where the alignment tolerance is less than 1 mrad angle and less than 10 μm in space. Cylindrical lenses were procured at ILE to produce 7 mm line foci in conjunction with the main f/3.1 metre focal length aspheric focussing lenses. A theoretically generated ray trace of the line focus showed negligible aberrations. The finite beam divergence of GEKKO XII produced a 30 μm wide focus as predicted. The optical configuration and target chamber layout is shown in Figure A1.7. Two beams were directly opposed each with 4 element segmented crystals for third harmonic operation mounted at the target chamber. Dichroic mirrors were added to protect the main infrared turning mirrors from damage by transmitted UV light when targets of low absorption such as fibres were used.

A new XYZ θ target positioning mechanism was constructed at ILE for the experiment. The approach used differed from that of the CLF. The target positioner/injector is a large structure which transports the target on a rigid mount from outside the chamber into the focal position via an airlock. It has servomotor XY drives moving the whole structure and a 1 m travel of Z motion to inject the target from the airlock.

The ILE target chamber 2 is designed for operation in 2 opposed six beam clusters and has an imaging facility in which the focal plane in each beam is imaged in transmitted light via the optics of the opposing beam. With the cylindrical lenses in place, and using the line focus produced at the focal plane of the main lenses, this RMAP device imaged a 3 mm section of the line focus when used with the CW 1.05 μm alignment laser of GEKKO XII. A 2-axis TV microscope system gave 2

views of the target and was used to define the centre of the target chamber for experimental purposes. A duplicate of the split field microscope system developed at the CLF was adapted for use at ILE with "dog leg" mirrors to feed beams through ports off the axis of the microscope. It was constructed at RAL and shipped to ILE where it was put into operation by ILE staff and used as the primary reference for target positioning of X-ray laser targets.

Target alignment was accomplished by first positioning a 30 μm slit using the split field microscope. The RMAP system was then used to place the IR beams of GEKKO XII at the centre of the slit. Observation of the inverse Foucault pattern transmitted through the slit was used to fine tune the angle of the line focus to the slit. Finally the lenses were moved back a calculated 6 cm and low energy (200 mJ) shots at 3 ω were fired at the 10 μm thick copper slit target. Examination of the burn marks with an optical microscope gave accurate information on the positioning of the line focus relative to the slit and accuracy of $\pm 7 \mu\text{m}$ and $\pm 1 \text{ mrad}$ was obtained by correcting the systematic error of the 6 cm motion of the lenses, the random errors of repeating the process being less than 10 μm .

3. Diagnostics

Spectroscopic diagnostics for the experiment included two streak camera time-resolved XUV spectrometers developed at ILE. One was aligned to the target axis and had a 1200 lines/mm variable periodicity flat field diffraction grating operated at 3° grazing angle. It was fed by a 10:1 reduced image from a grazing incidence spherical mirror. With film recording it gave spectra from 30 Å to 370 Å over a 5 cm field with a resolution of 0.2 Å at wavelengths of interest around 50 Å. With a Hamamatsu XUV streak camera a 25 mm field was recorded with 0.5 Å resolution. A second similar instrument positioned 60° off-axis had a 2,400 line/mm grating blazed for shorter wavelength and operated at 1.2° grazing angle. It gave photographic spectra in a 5 cm field from 6 Å to 144 Å with 0.1 Å resolution. A Kentech streak camera from the CLF was used with this instrument and recorded over a 2.5 cm field with 0.3 Å resolution. Another spectrometer positioned 45° off axis gave a wide range XUV spectrum from a conventional 5 m radius grating and was used in the photographic mode for survey purposes.

An array of 12 plasma calorimeters from the CLF was positioned to record the narrow angular distribution of plasma energy perpendicular

to the target axis. A slit camera produced images in 1.5 keV X-ray emission with high magnification perpendicular to the line focus and low magnification along the line focus.

4. Target Design

A brief summary of conclusions reached on target design for the experiment is given here. Details will be presented elsewhere.

(i) Fibre targets

A scaling analysis and numerical modelling were used to assess the scaling of optimum gain in Na XI and other element fibre targets, assuming that the laser pulse duration would not be less than 70 psec and the fibre radius would not be greater than 3.5 μm . It was found that the plasma element producing most gain reaches its peak temperature just after the peak of the laser pulse and is at a greater depth in the target for higher Z. With absorbed energy scaled as Z^4 , the temperature scaled as Z^2 and the residual fraction of H-like ions scaled as Z^4 , most ions are bare nuclei at the temperature peak for expansions where optimum gain conditions later develop. For Na XI this temperature was 750 eV and gave 80% bare nuclei.

The cooling characteristic shows an interesting feature, being more rapid than adiabatic, due probably to conduction cooling to the cold core which is more effective at the higher temperature used for higher Z. In the initial phase of cooling $T_e \sim N_{e0}^{-1}$. This is in contrast to the behaviour of C VI and F IX at lower temperature where cooling is slower than adiabatic due to recombination heating.

A scaling analysis from C VI and F IX recombination experiments in the CLF indicates that the density at peak temperature N_{e0} would scale as $N_{e0} \sim Z\lambda^{-1}$, ie be higher both because of the shorter laser wavelength (0.35 μm) in this work and the higher Z.

The initial density and the cooling characteristic are important features determining the gain. A contour representation of potentially available gain for negligible ground state population was developed as shown in Figure A1.8, based on general results for excited state populations in H-like ions in terms of reduced variables n_e/Z^2 , T_e/Z^2 . Cooling adiabats where $T_{e0} \sim Z^2$ and $n_{e0} \sim Z$ are shown relative to optimised conditions for C VI.

Figure A1.8 shows that for Na XI the plasma is produced initially under conditions of potential gain, whereas for C VI it is not and also that faster than adiabatic cooling gives greater potential gain as does a higher initial density. For Na XI gain cannot occur until cooling reduces the contribution to $n = 2$ population due to collisions from the ground state. This requires only a circa 2-fold reduction in temperature (independent of density). This transition point in the cooling characteristic is indicated in Figure A1.8.

Gain is not produced at the point where collisional quenching ceases unless reabsorption of resonance radiation is also negligible (ie the opacity of the resonance transition is less than 1). The velocity gradient, fraction of population in the ground state and size and density of the plasma determine this and the onset of gain in the expansion is delayed significantly in numerical simulations as indicated in Figure A1.8.

The eventual magnitude of the gain is the potentially available gain shown in Figure A1.8 reduced in proportion to the fraction of ions remaining as bare nuclei. This depends on the rate of recombination and timescale of expansion and cooling.

Dilution of the concentration of the laser ion can give greater gain by allowing expansion in a cooler adiabat without quenching due to trapping of resonance radiation.

The numerical modelling suggested that the gain coefficient could exceed 1 cm^{-1} for Na XI at 100% concentration with 40 J cm^{-1} absorbed by the target. Dilution of the Na concentration was estimated to increase the gain up to 2-fold with a peak at about 3:1 dilution.

(ii) Foil Targets

Foil targets were also of interest having been used successfully in earlier experiments at ILE. An evaporated coating on a thin (1000 Å) CH polymer substrate was considered with a view to producing similar initial conditions to those in the fibre.

In this case the expansion is complex with a transition from planar to quasi cylindrical which requires 2D hydro modelling, but can be approximated by the slower expansion from an 'equivalent' larger fibre.

Both approaches were used to assess the behaviour, the former without and the latter including the atomic physics.

Useful insight was obtained from the analysis of exploding foil targets by London and Rosen⁽¹⁾ which shows how two-sided irradiation initially produces a plasma outflow at the 'ablation' temperature. At the onset of transparency there is a sudden temperature rise and a drop in density which is too fast for the ionisation to follow in equilibrium. This effect is highly deleterious for a recombination laser where the density at peak temperature should be as high as possible and the initial degree of ionisation should be as high as possible for the given temperature.

Because of this consideration targets were designed to produce the necessary initial temperature in the ablation outflow using a similar absorbed intensity to that in the fibres and the layer thickness was chosen to be just greater than that for burn through. One sided irradiation was chosen because there was no need to reproduce the process on both sides of the target.

For an estimated $50 \text{ }\mu\text{m}$ width for the energy deposition (including lateral transport effects) the required energy for Na XI and 120 psec pulses (scaling $\sim Z^4$) was 130 J cm^{-1} and the target thickness for NaF and $\text{Na}_2\text{B}_4\text{O}_{10}$ layers was $0.2 \text{ }\mu\text{m}$.

(iii) Stripe targets

The foil targets have the disadvantage of unlimited lateral extent so that lateral energy transport can broaden the region of energy deposition and produce unwanted plasma which slows the expansion. A narrow stripe on a thin substrate overcomes this limitation and stripes $30 \text{ }\mu\text{m}$ wide were considered to be well matched to the optical focal width. The thickness was chosen to be large relative to the ablation depth to leave a cold core which would give the advantageous conduction cooling discussed earlier. Irradiation requirements were otherwise the same as for foils.

5. Experimental results

Time and space integrated measurements of axial and transverse spectra were obtained using the three spectrometers with Kodak 101 film recording. The data covered a wide spectral range with relatively

high resolution. Film density D was related to intensity I using the Henke⁽²⁾ calibration of 101 film response, viz,

$$I = 3.12 \log_e \left(\frac{1.96}{1.96-D} \right)$$

Line intensities I_l were obtained in preliminary densitometric analysis by determining the peak intensity I_p and the background continuum intensity I_b to give $I_l = I_p - I_b$.

(i) Cross calibration

Intensity ratios for axial and transverse emission were obtained by cross calibrating the axial and transverse instruments using targets producing identical emission into the axial and transverse instrument. These cross calibration targets were narrow (1 mm wide) thin film targets placed so that the target normal was in the plane of the axial (0°) and transverse (60°) instrument and along the 30° bisector between them presenting an equivalent view of the plasma to each instrument. The laser radiation was then incident at 60° from the target normal and the line focus was perfectly focussed only at the centre of the target, widening to 140 μm at each extremity.

Targets were 0.1 μm CH polymer substrate coated with 0.3 μm NaF or 0.3 μm $\text{Na}_2\text{B}_4\text{O}_7$. Lines used for cross calibration in the spectral range of interest were:

Cross Calibration Lines

Line	λ	Target
Na IX 4 ² D-2 ² P	58.24 Å	NaF
Na IX 3 ² P-2 ² S	70.83 Å	NaF
C V 2 ¹ P-1 ¹ S	40.27 Å	NaF and $\text{Na}_2\text{B}_4\text{O}_7$
B IV 2 ¹ P-1 ¹ S	40.31	$\text{Na}_2\text{B}_4\text{O}_7$
B V 2 ¹ P-1 ² S	48.627	"
C VI 2 ¹ P-1 ¹ S	33.77	"

These lines were strong enough to be recorded with the circa 7% fraction of the energy in the 7 mm line which was intercepted by the

angled target. The lines originated from ionisation stages which were easily excited at the lower absorbed intensity of the angled and slightly defocussed line focus. Three cross calibration shots (2 NaF, 1 $\text{Na}_2\text{B}_4\text{O}_7$) were recorded with 60 to 80 J, 3w energy in the line focus at the start mid point and end of an initial series in which 9 XUV laser target shots were also recorded. In a second series of 9 XUV laser target shots, 2 further cross calibration spectra were recorded ($\text{Na}_2\text{B}_4\text{O}_7$ at 150 J 3w energy). Minor alignments were made to the spectrometers in between the two series.

In all shots the targets were irradiated with a single beam either from the rear or front side as viewed from the spectrometers. A significant spread of exposure levels on film was obtained with the various shots, but the cross calibration ratios were consistent as shown in the preliminary analysis of the data presented in Figure A1.9. The consistency for different exposure levels suggests that the sensitivity calibration of the 101 film was adequately described by the measurements of Henke⁽²⁾.

Two separate cross-calibration characteristics are fitted to the two sets of data in Figure A1.9, allowing for possible changes resulting from the adjustment made between the two data series, though a single fit would make little difference to the cross calibration ratios [axial to transverse sensitivity, $S(a/t) = 5.1$ for Na XI H_α at 54.2 Å and 2.4 for Mg XII H_α at 45.5 Å]. Figure A1.9 shows how the sensitivity of the axial instrument falls rapidly relative to the transverse instrument for shorter wavelengths, reflecting the different blaze and operating angles of the two gratings.

(ii) XUV Laser Data Na XI

The first series of XUV laser target shots were used to test the Na XI target design discussed earlier, ie 6 mm long coated foil targets with one sided irradiation irradiated at just less than the burn through level, 6 mm stripe targets with one sided irradiation and thickness well above the burn through level, and coated fibre targets supported at one end and irradiated with two beams. Both NaF and $\text{Na}_2\text{B}_4\text{O}_7$ coatings were used to give dilution of the Na ions of 0.55 and 0.22 respectively.

NaF foil targets (0.2 μm on a 0.1 μm CH substrate) were used first, with 90 J, 3w irradiation. The transverse spectra showed the NaF and C

resonance spectra as well as the Balmer series spectral region. The ionisation ratio in Na could be assessed from the ratio of the Lyman series intensities to the intensities of the corresponding He-like resonance lines, eg $I(3^2P-1^2S)/I(3^1P-1^1S)$. Approximately equal intensity of the two lines was recorded (as in the previous experiments at ILE with F IX). (Equal instantaneous intensity indicates equal fractions of bare nuclei and H-like ions in a recombining plasma).

The Na XI H_α doublet (with its stronger component at 54.2 Å and weaker at 54.04) was a relatively weak feature in the transverse spectra, almost blended with the Na IX $4p^2P-2s^2S$ transition of the Li-like ion at 53.87 Å, which from the symmetry of the broad line blend had a similar intensity. In the axial spectrum the 54 Å emission was more intense than the related Na IX $4d^2D-2p^2P$ transition at 58.24 Å. By contrast in the transverse spectrum the Na IX $4d^2D-2p^2P$ line was 3x more intense than Na XI H_α feature. Figure A1.10(a) presents relevant parts of the two spectra and shows the axial/transverse ratios. These are either substantially less than 1 indicating strong absorption eg C V $1s^1S-2p^1P$ or very close to 1 indicating small or negligible loss eg Na IX lines, or substantially greater than 1 for the Na XI H_α line, for which $I_a/I_t = 1.6$ indicating gain.

Similar observations were made for the stripe targets 30 µm wide and 1 µm thick on 0.1 µm CH substrates. The axial/transverse ratio for an 80 J shot was 1.5, similar to that for the foil. Increasing the laser energy to 150 J raised the ratio to 2.0.

Substituting $Na_2B_4O_7$ for the NaF in a stripe target irradiated at 80 J gave an axial/transverse ratio of 1.7 but an $Na_2B_4O_7$ foil target at a similar energy gave an axial/transverse ratio of only 0.9.

The slit camera images of the 6 mm long foil and stripe targets all showed fairly uniform emission of 1.5 keV X-rays from approximately 40 µm width over that length.

NaF coated fibre targets of 8 µm diameter were irradiated with two beams at 150 J/beam, positioning the unsupported end of a 10 mm long fibre nearest to the spectrometer and 1 mm from the end of the line focus. These targets gave less uniform slit camera images with brightness falling away to one end and length of intense emission of only 3-4 mm, although the plasma calorimeters indicated that 30-40 J was absorbed by the target as expected in the target design. The transverse spectra showed a poor ionisation ratio with the Lyman series

at less than half the intensity of the Helium-like resonance lines. The axial H_α intensity was weak relative to the Na IX lines and the axial/transverse ratio was about 1:1 though the relatively low absorbed energy gave rather weak exposure levels in the transverse spectra which were difficult to measure accurately.

(iii) Na XI H_α (54.2 Å) laser gain

The axial/transverse intensity ratio for any transition can be related to the gain x length for small g_l by

$$\exp(g_l) - 1/g_l = I_a/I_t$$

and the previously discussed ratios for 6 mm plasmas of 1.5 to 2 would imply g_l from 0.8 to 1.3 or g from 1.3 cm^{-1} to 2.2 cm^{-1} .

A preliminary conclusion from the Na XI results is that time and space integrated spectra show axial to transverse ratios for Na XI H_α at 54.2 Å which suggest gain was obtained in thin foil and stripe targets using NaF and in stripe targets with $Na_2B_4O_7$. Fibre targets were not successful, possibly due to insufficiently precise alignment, and neither were $Na_2B_4O_7$ foils. A proper estimate of the magnitude of the gain requires further analysis. Time and space integration mask the gain in the best time/space zone and, for small g_l values numerical calculations suggest that this is an important effect. Streak time resolved spectra obtained in the final phase of the experiment have yet to be analysed but should give additional information on this point. Line blending of the Na XI H_α line complicate the interpretation and needs further consideration.

(iv) XUV laser data Mg XIII H_α 45.5 Å

The second phase of time and space integrated spectroscopy was used to study stripe targets coated with MgF_2 (Mg dilution ratio 0.4). The laser energy could be increased relative to the Na XI by the 40% required for the theoretical Z^4 scaling. The same width (30 µm), length (6 mm) and thickness (1 µm) was used for the stripes which were on a 0.1 µm CH polymer substrate and for foils the coating thickness was 0.2 µm. One beam irradiation was used since the targets were thicker than the burn depth and the energy was at the maximum level of about 150 J in the 7 mm line focus.

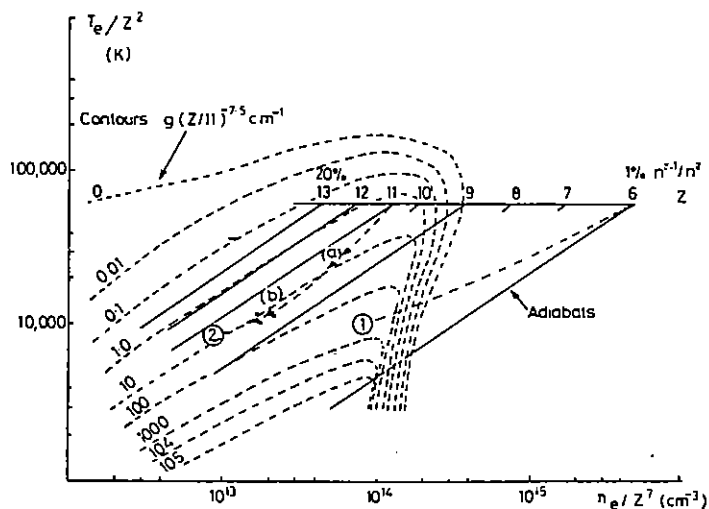
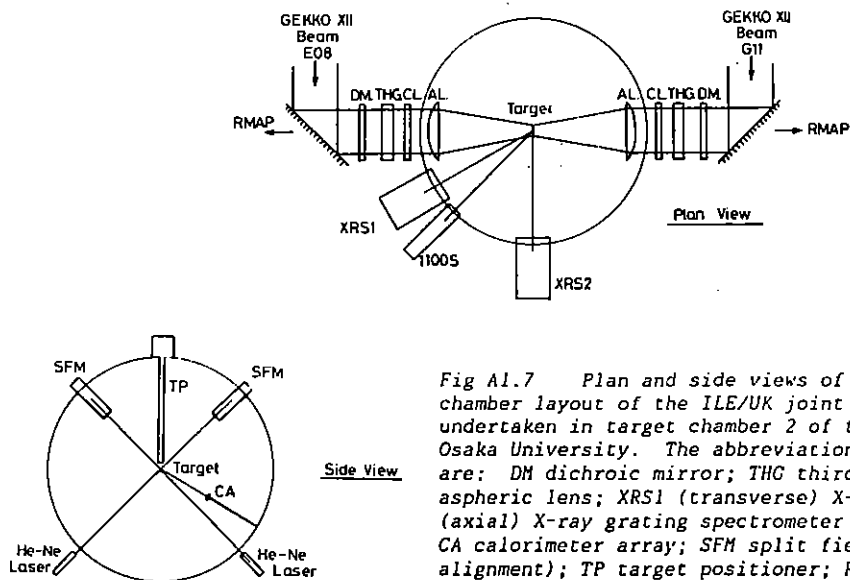


Fig. A1.8 Normalised gain contours and cooling adiabats for hydrogenic ions of different Z . (1) and (2) show typical numerically computed cooling characteristics for C VI and Na XI. a) and b) show where quenching of gain by collisions and trapping cease in the numerical calculation.

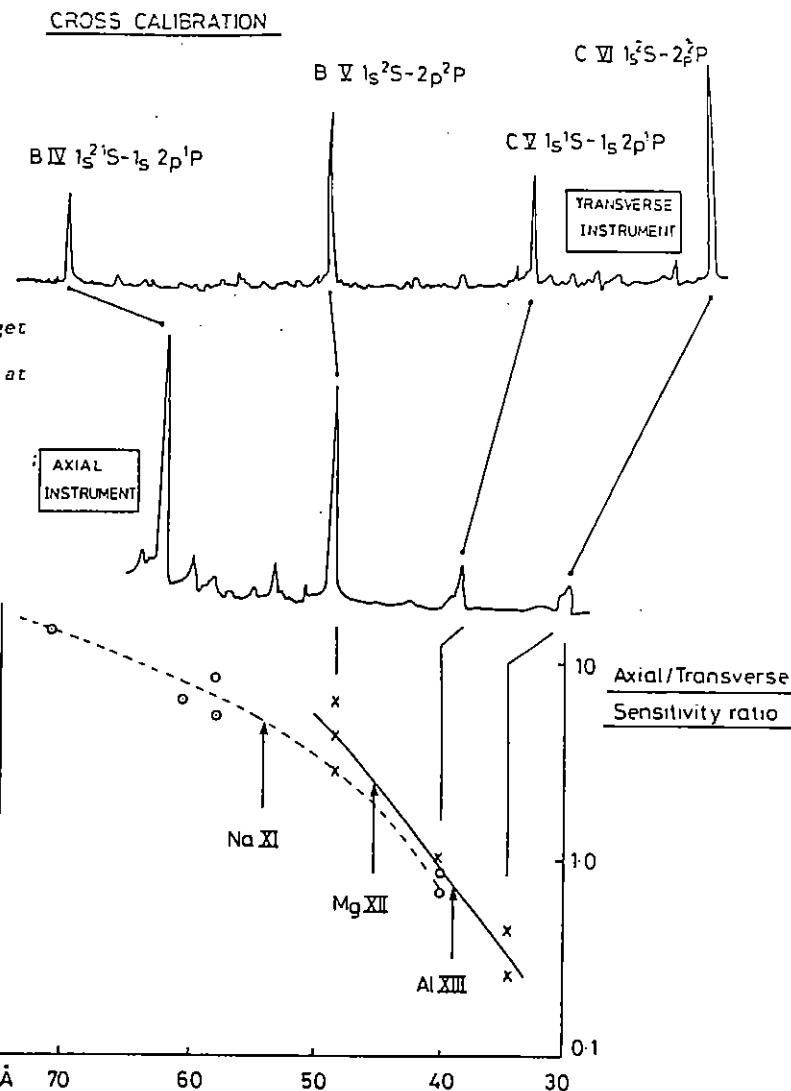


Fig A1.9 The axial to transverse spectrometer sensitivity ratio as a function of wavelength determined from cross-calibration shots. A sample cross-calibration spectrum is also shown for both of the spectrometers.

The transverse spectra confirmed that the ionisation ratio was as expected with Mg XII Lyman and Mg XI Helium like resonance spectra of equal intensity.

The H_{α} transition at 45.5 Å is not blended with other lines in the axial or transverse spectra, making interpretation simpler than for Na XI, though it is close to Mg X 2p-4d at 47.25 and Mg X 2s-4p at 44.05 and the C V Helium-like resonance line at 40.2 Å and its axial/transverse ratio can be compared with these other lines.

Similar results were obtained with several stripe targets. The Mg XII H_{α} transition was more prominent relative to neighbouring lines on the axial spectrum as shown in Figure A1.10(b). Its axial/transverse intensity was measured at 1.7 suggesting $g_{\parallel} \sim 1$ and $g_{\perp} \sim 1.6 \text{ cm}^{-1}$.

By contrast the axial/transverse ratio for the C V resonance line and for the Mg X 2s-4p transitions were substantially less than unity as shown in Figure A1.10(b).

This result for Mg XII H_{α} is particularly interesting as the wavelength is the shortest at which evidence of laser gain has been obtained to date. Here again care is needed in inferring the magnitude of the gain from the data because of the masking effect of space/time integration, though the absence of complication due to line blending make interpretation simpler than for Na XI.

Further analysis of this data and additional streak time resolved spectra is in progress.

(v) Al XIII H_{α} 39 Å

Although it was clear that the laser energy could not be increased by the required Z^4 scaling, some target shots with Al F_{β} and Al stripe targets were recorded using 150 J because of the great interest in obtaining laser action in the water window at wavelengths below the C K edge. The ionisation, as evidenced by the circa 1-2 hydrogenic to helium-like resonance line intensity ratio, was lower than required. The axial spectrometer showed very low sensitivity at 39 Å. The H_{α} line was almost blended with second order O VIII $\text{Ly}\alpha$ in the axial spectra making measurement difficult as its intensity was weak. The axial/transverse ratio appears to be significantly less than 1 however, so that though the H_{α} transition was observed, there was no indication of gain. This was not unexpected because the laser energy was

insufficient to produce the required ionisation.

(vi) Time-resolved spectra

Streak time resolved spectra of axial and transverse emission, together with cross calibration data were recorded for several Na XI and Mg XIII laser targets. These data have not yet been analysed.

1 2 . General Conclusion

The joint experiment was successful on several levels and has strengthened the base for future cooperative work in a wider field than x-ray laser experiments. The x-ray laser data are of considerable interest and represent a significant step towards our longer term objective of laser action in the water window, the 45.5 Å wavelength of Mg XIII being only 2 Å from this objective and (provided the results are confirmed by detailed analysis) the shortest laser wavelength to date.

References

1. R A London and M D Rosen, Phys. Fluids, **29**, 3813 (1986).
2. B L Henke, J Y Uejio, G F Stone, C H Dittmore and F G Fujiwara, J. Opt. Soc. Am., **B3**, 1540 (1986).

A1-3 LI-LIKE RECOMBINATION XUV LASER SCHEMES

C Regan, D O'Neill, C L S Lewis (QUB)
A Klisnick, G Jamelot, P Jaegle (Orsay)
O Willi, R Smith, J Edwards (ICL)
M H Key, M Grande (RAL).

1. Introduction

We have carried out recombination XUV laser experiments based on lithium-like ions of aluminium (November 1986) and chlorine (June 1987). Targets were 8 µm diameter carbon fibres coated with 0.35 µm Al and 0.2 µm NaCl and these were irradiated in the 7 mm line foci of up to six 0.53 µm, 100 ps laser beams. The potential lasing lines are the 4 - 3 and 5 - 3 transitions at wavelengths of $\approx 15.2 \text{ nm}$ and $\approx 10.5 \text{ nm}$

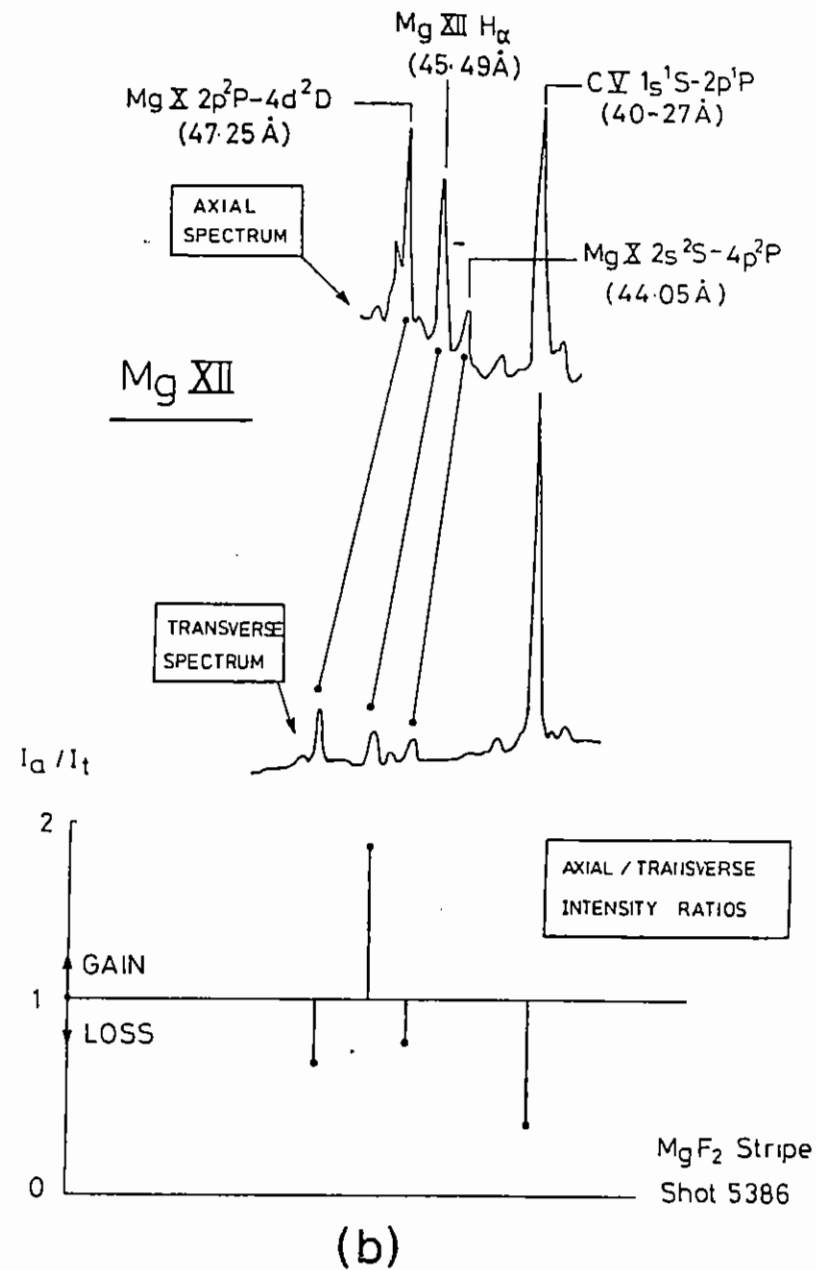
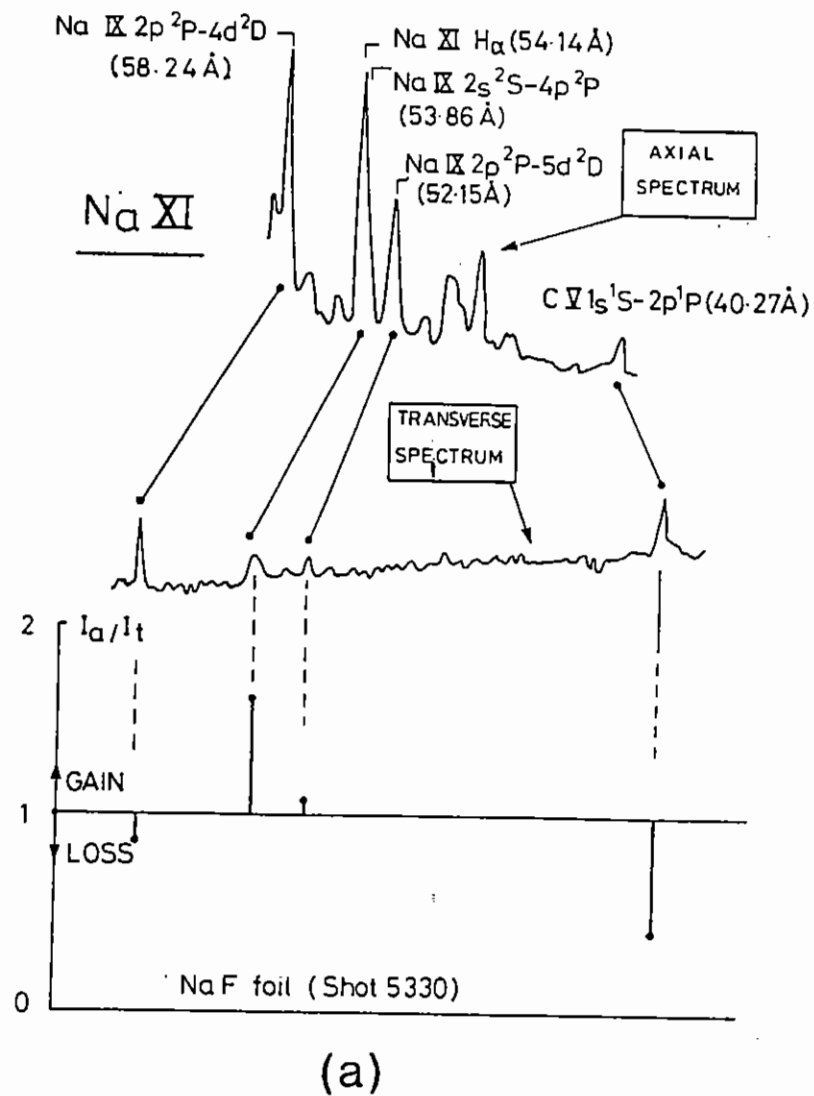


Fig A1.10 Axial and transverse spectra for (a) an NaF foil shot and (b) an MgF₂ stripe shot and the plotted axial to transverse intensity ratios showing gain ($I_a/I_t > 1$) or absorption ($I_a/I_t < 1$) for various emitted spectral lines.

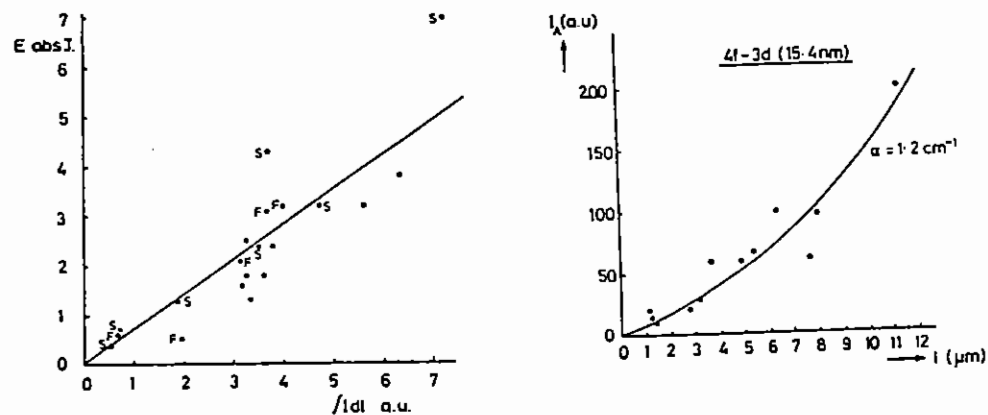


Fig. A1.11 Selection of a data set of Fig. A1.12 Variation of intensity of consistent energy per unit length. lasing transition with plasma length.

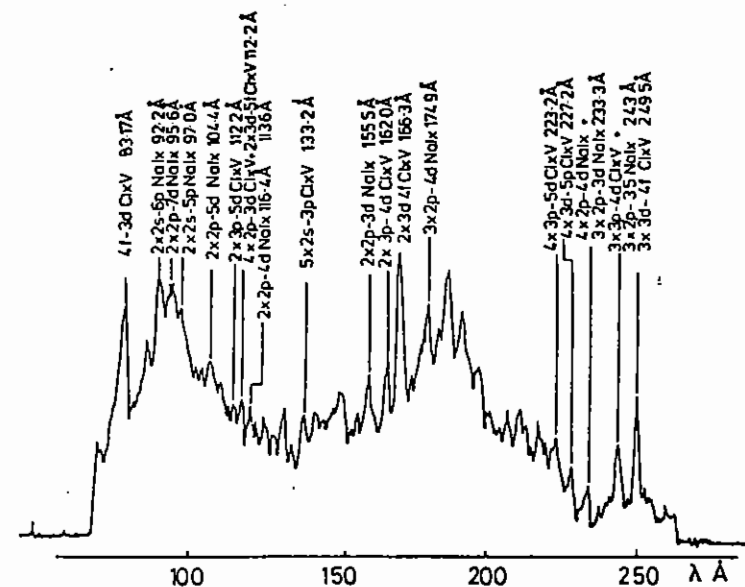


Fig. A1.14 Spectrum obtained on a NaCl target.

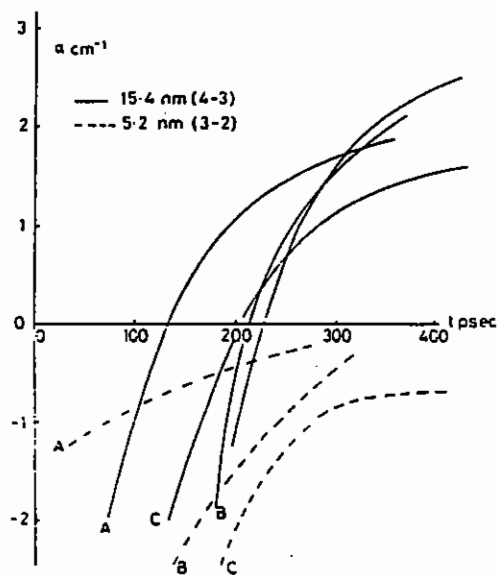


Fig. A1.13 Time dependence of gain for four separate shots note the optical thinning of the resonance line as the gain switches on.

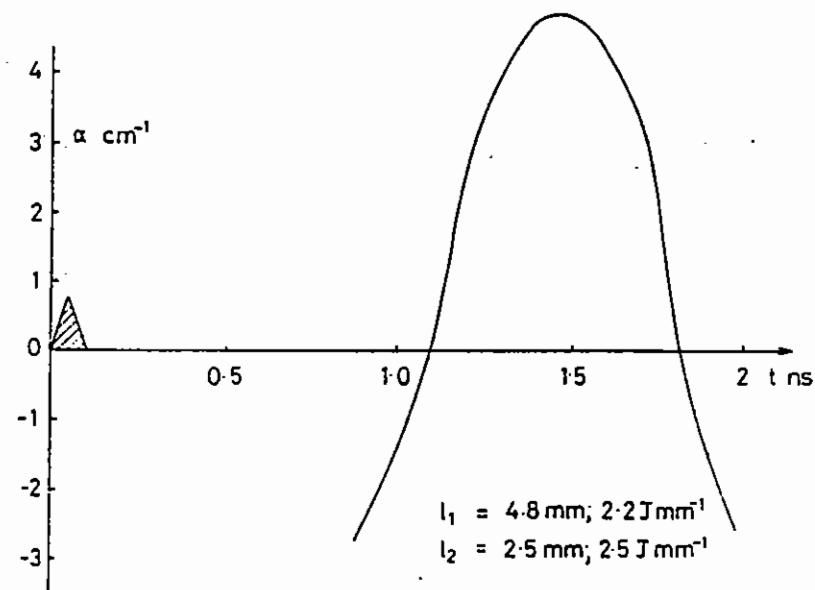


Fig. A1.15 Time dependence of gain on a NaCl target.

for Al and ≈ 8.2 nm and 5.6 nm for Cl. A preliminary analysis of some of the Al data was presented in last year's annual report and those conclusions are now modified in the light of more extensive data analysis.

2. Non-Spectroscopic Diagnostics

The heated plasma column length was monitored using active and passive pinhole cameras with phosphor screen-image intensifier and x-ray DEF film recording respectively. Absorbed plasma energy was monitored using a ten element tantalum foil array and integration over 4π assuming azimuthal symmetry about the fibre axis. Since data from each of these three diagnostics was not always available on each shot we used the following method to compensate in some cases. For the standard passive pinhole camera configuration (90 μm pinhole, 100 nm Al filter and $\times 0.9$ magnification) we plotted the 2-D integrated image intensity against measured plasma energy for all available shots and found the best linear least squares fit. For a standard active pinhole camera configuration (100 μm pinhole, 200 nm Al filter $\times 1.2$ magnification and intensifier gain setting of -3) we repeated the procedure. There were eleven shots common to both best fits. We then normalised the best fit lines to the same arbitrary units of integrated image intensity and plotted the geometric means of the common data set points against absorbed energy. Since the first best fits were quite good we also included in the final best fitting shots which had only information on one pinhole camera. In these cases the integrated intensity was normalised to the same arbitrary units and plotted as if it was a geometric mean. The complete data set is shown in Figure 1 where the common data points are solid. Since plasma calorimetry was believed to be the least reliable diagnostic we subsequently scaled all data points in terms of their absorbed energy to the best fit line and these are the values used later in determining the absorbed energy per unit length for shots which have also got spectroscopic diagnostic data. The procedure allows us to make a good estimate of this parameter for example in a shot where only one pinhole image is available. On a few shots where, for example, only an active pinhole camera image is available, at a non-standard gain setting, then raw calorimetry data is used.

To determine a plasma length from a pinhole image we adopted the following procedure. An individual record is densitometered and the intensity integrated along the length defined by background fog levels.

An average intensity is determined and a new length defined between the 10% points of this average intensity. A new average intensity is determined by integration over only the 10% limit length and this value is averaged over all data shots in the same set to yield a value I_{av} . The plasma length in individual images is then taken as the distance between $I_{av}/2$ intensity points. This cumbersome procedure has been developed to mitigate against unrepresentative lengths being assigned to shots which are non-uniformly heated along their length. A statistical analysis of intensity variations along the length of an image has also been applied to reject shots from the data set when intensity variations from the mean is considered excessive. In cases where two images were available final lengths were always in good agreement within error limits and in these cases a geometric mean length was adopted.

3. Spectroscopic Diagnostics

We used two flat-field time-resolving VUV spectrometers, one observing axial emission from the free end of the fibres and the other observing transverse emission at 30° to the axis. Axial and transverse spectrometers has 100 ps/mm and 80 ps/mm sweep speeds and 80 ps and 120 ps temporal resolutions respectively. Attenuating filters were typically in units of 100 nm of formvar and their transmission properties were allowed for when making relative spectral intensity measurements. The relative wavelength response of the two instruments was determined by cross-calibration using a short scale-length plasma as a common emission source. All data was recorded on individually calibrated HP5 film.

4. Aluminium Experiment

Two modes of fibre irradiation were used both using two pairs of opposing beams. In one case all four beams coincided axially along the fibre and in the other case one pair was axially displaced to allow longer heated plasma lengths. We refer to these as four-beam (F) and split-beam (S) shots. The acceptable data sets contain 5 and 7 shots respectively and are identified in Figure A1.11. These shots were chosen on the basis of a limited spread in the absorbed energy per unit length which is 4.7 ± 1.4 J/cm.

Figure A1.12 illustrates the axial intensity of the 4f - 3d line at 15.4 nm at 300 psec after fibre irradiation for all twelve shots as a function of plasma length. A least square fit to the growth curve of the form $I = S(e^{\alpha l} - 1)$ yields a gain coefficient $\alpha = 1.2 \pm 0.15 \text{ cm}^{-1}$.

This is somewhat less than the value 3 cm^{-1} reported previously for the four beam shots only and is mainly a consequence of eliminating shots which had an absorbed energy per unit length outside the current limited range.

We have also been able to obtain gain estimates using the ratio of axial and transverse spectrometer intensities. On a given shot the ratio at any time I_A/I_T is assumed equal to $(e^{\alpha l} - 1)/\alpha l$ and this is solved to yield the time dependence of α . The shots which yielded positive values of α are illustrated in Figure A1.3. It is interesting to note that in each of the four cases early opacity on the 15.4 nm line switches to gain at $t \approx 200 \text{ psec}$. The time dependence could not be carried further due to the rapidly decaying brightness on the transverse spectrometer. These four shots all belonged to the split beam set and were the only ones in this set which had good transverse data. Only two shots in the four beam set had any transverse data and this was of poor quality leading to inconclusive estimates of any gain. However, from Figure A1.13 it can be seen that at the corresponding time $t \approx 300 \text{ ps}$ the gain coefficient is comparable to that estimated above from axial growth only in Figure A1.12. Figure A1.13 also shows the α -value associated with the resonant 3d - 2p transition at 52.4 nm on three of the shots and it is noteworthy that the resonant line becomes relatively optically thin at about the time gain switches on. A similar analysis of the 5f - 3d line at 10.6 nm also yields $\alpha \approx 1.5 \text{ cm}^{-1}$ although the axial/transverse data set is further reduced due to lower spectral brightness from the higher series member.

5. Chlorine Experiment

Due to the higher z all six beams were focused along a common 7 mm maximum fibre length with an absorbed plasma energy per unit length of $20 \pm 5 \text{ J/cm}$. Data analysis is still in progress but Figure A1.14 illustrates an identified spectrum showing in particular the 4f - 3d line at 8.3 nm in second, third and fourth order. Figure A1.15 shows an estimate of gain development in time from the axial intensity ratio on this line from a long (4.8 mm) and short (2.5 mm) plasma length at the same emission time. We must point out that caution is required in interpreting the apparent gain at late times as this may be an artefact

of the different plasma expansion characteristics of long and short plasmas.

A1-4 Na-LIKE Cu RECOMBINATION LASER EXPERIMENT

C L S Lewis, D O'Neill, C Regan (Queens University Belfast), S A Ramsden (Hull University), G P Kiehn, R A Smith, O Willi (Imperial College), M C Richardson (LLE), J Seely (NRL), M Grande, M H Key, W Shaikh (RAL)

Following earlier measurements on H-like and Li-like ions this experiment was an attempt to observe gain by recombination from the next highest closed shell, viz recombination from Ne-like to Na-like ions. Cu XIX 5g-4f at 111Å was chosen largely because copper could be readily coated onto the carbon fibre substrate.

7µ carbon fibres were used with coating thicknesses varying from 1500 - 3000Å. All six laser beams were focussed onto a line focus which was varied up to 10mm in length. Pulse widths of 70 psec and 140 psec were used with energies up to 150J and 300J respectively. The usual diagnostics were available - namely, axial and transverse time resolved grating spectrometers, crystal spectrometer, temperature monitor, pinhole cameras, and calorimeters.

The spectra obtained were crowded and complex and are still being analysed. The 5g-4f transition of CuXIX at 111Å was, however, weak and showed no evidence of gain. Another line at 94Å appeared more promising but this has not yet been identified.

A1.5 Mn/F RECOMBINATION/PHOTOPUMPING EXPERIMENT

C L S Lewis, D Neely, D O'Neill (Queens University Belfast); S A Ramsden (Hull University); O Willi (Imperial College); M Grande, M H Key, S J Rose (RAL)

This experiment relies on the near coincidence of the 2p-3d (12.643Å) transition in Be-like Mn, as measured by Burkhalter et al (1), with the 1s-3p (12.641Å) L_{β} transition of H-like FIX. The aim is to enhance the gain in the FIX H_{α} recombination scheme using a Mn photopump as shown in Fig A1.16.

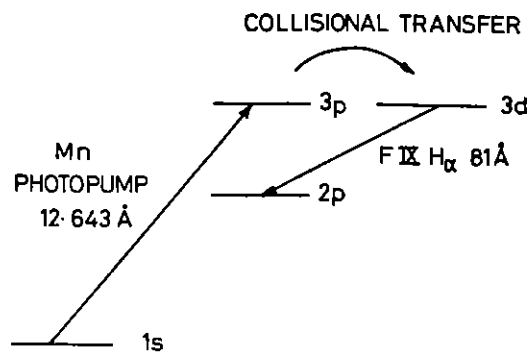


Fig. A1.16 Term diagram.

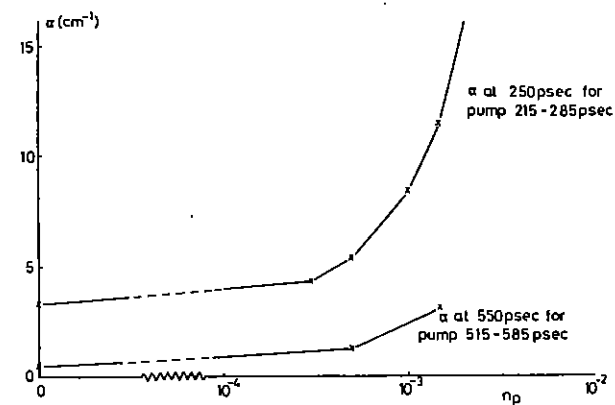


Fig. A1.18 Calculated gain, α , as a function of modal photon density, n_p , for pump delays of 250 psec and 550 psec.

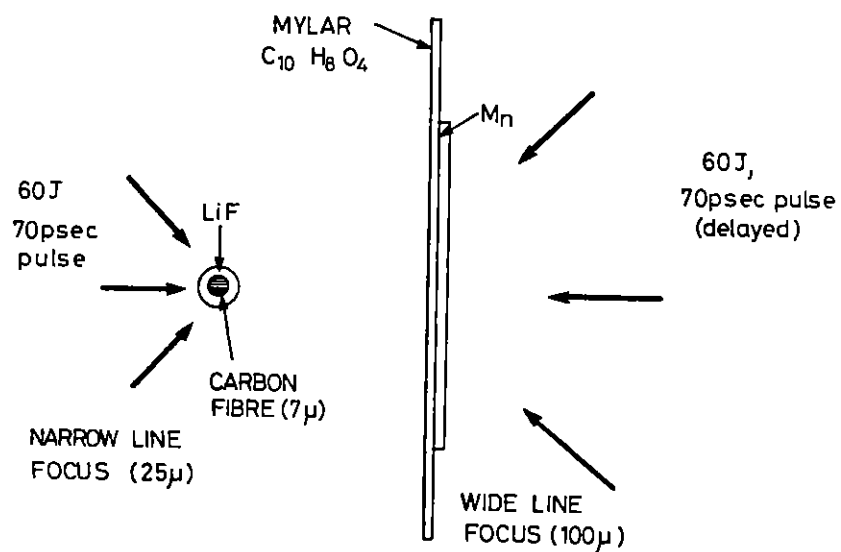


Fig. A1.17 Schematic of target configuration.

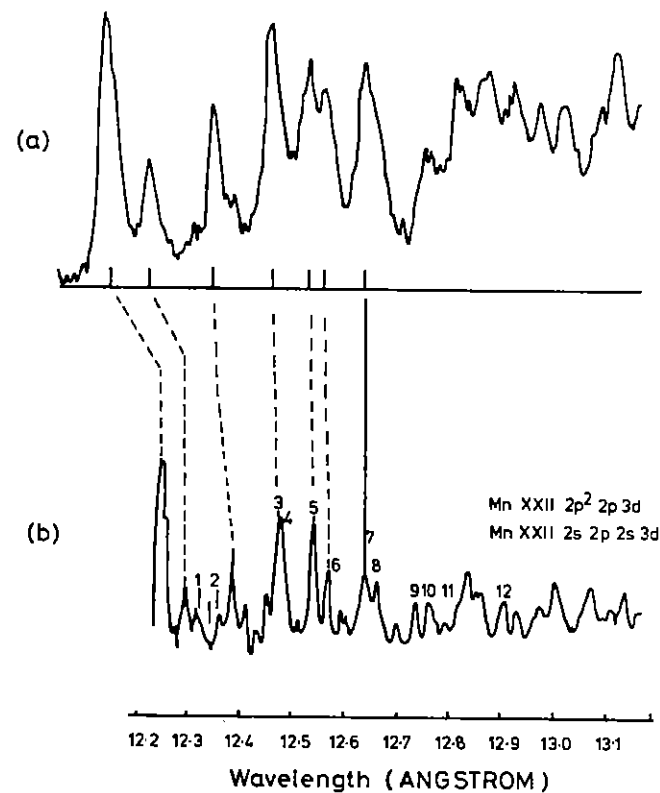


Fig. A1.19 (a) Mn flashlamp spectrum
(b) Spectrum from Burkhalter et al.

The experimental arrangement used is shown in Figure A1.17. Three beams delivering 60J in 70psec are brought to a line focus onto a LiF coated carbon fibre. The other three, delayed in time, are focussed onto a 100-200 μ wide Mn flashlamp approximately 100-200 μ away from the fibre. The flashlamp consists of a thin 1000-1500 $\text{\AA}$ layer of Mn on a 2 μ plastic foil. The foil thickness is chosen so as not to burn through during either pulse and yet allow the pump radiation to pass through with relatively little attenuation. The plastic also acts as a filter for the pump radiation in the vicinity of the FIX L_{α} line.

The effectiveness of the photopump in creating a population inversion depends on the modal photon density, n_p , where

$$n_p = \frac{1}{\exp [h\nu_{\text{pump}}/kT_{\text{brightness}}] - 1}$$

Here $h\nu_{\text{pump}}$ is the pump photon energy and $kT_{\text{brightness}}$ is the brightness temperature. This was included in the RAL average-atom X-ray laser code and Figure A1.18 shows the calculated gain as a function of modal photon density for two different delays in the flashlamp pump. From the measurements of Burkhalter et al the value of n_p is expected to lie within the range 5×10^{-4} to 1.5×10^{-3} photons/mode producing a readily observable increase in gain over that obtained by recombination alone.

In the actual experiment a crystal mini-spectrometer was used to determine the X-ray spectrum of the Mn flashlamp and a time resolved flat field grating spectrometer was used to record the emission along the axis of the fibre in the region of FIX H_{α} 81 $\text{\AA}$. A small block $\sim 100\mu$ high attached to the end of the foil reduced the amount of flashlamp radiation seen by the grating spectrometer. The X-ray emission from both the flashlamp and the fibre was monitored using an active pinhole camera.

The results have still to be analysed and only a few, preliminary, observations can be made at this stage. The spectrum of the flashlamp (Figure A1.19) closely resembles that published by Burkhalter et al, which is encouraging. It was, however, more difficult than anticipated to accurately position and align both flashlamp and fibre and very few

satisfactory shots were obtained. In some of these there was an indication of photopump enhancement of FIX H_{α} intensity but this needs to be confirmed by further detailed analysis. Problems were experienced with the plasma created by the laser energy which is not absorbed by the fibre but falls on the face of the foil and these would have to be addressed in any future experiment.

References

1. P G Burkhalter et al, J. Opt. Soc. Am. B., 12, 1894 (1985).

A1-6 A1 STRIP TARGETS

C L S Lewis, D O'Neill, C Regan (Queens University Belfast),
S A Ramsden (Hull University)
G P Kiehn, R A Smith, O Willi (Imperial College)
M Grande, M H Key, S J Rose, W Shaikh (RAL)

One drawback of fibre targets is their low coupling efficiency - typically only 10% of the incident laser energy. It was hoped that this could be improved by the use of stripe targets⁽¹⁾ but these did not behave as expected because of the interaction of the expanding plasma with the plastic substrate. A logical next step was to try to use free standing strip targets, as shown in Figure A1.20, made by etching away the substrate. These targets were 2 cm long, 100 μ wide, 3000-4000 $\text{\AA}$ thick. The support 'legs' were 3 mm apart, which is large compared with the scalelength of the instabilities observed earlier. The line focus could be varied up to 1.4 cm in length. Both long (450 psec) and short (70 psec) 0.53 μ laser pulses were used with total energy (6 beams) up to 200 J. Although the results have still to be analysed in detail there was no clear evidence of gain on either of the

Li-like Al XI lines at 105 $\text{\AA}$ and 154 $\text{\AA}$. This is probably because the strips, although state of the art, were too wide and too thick and the operating conditions were more appropriate in the ablative rather than the exploding foil mode. Further progress is therefore likely to be dependent on advances in target fabrication.

References

1. D Bassett et al, RAL Report RAL-87-041 (1987).

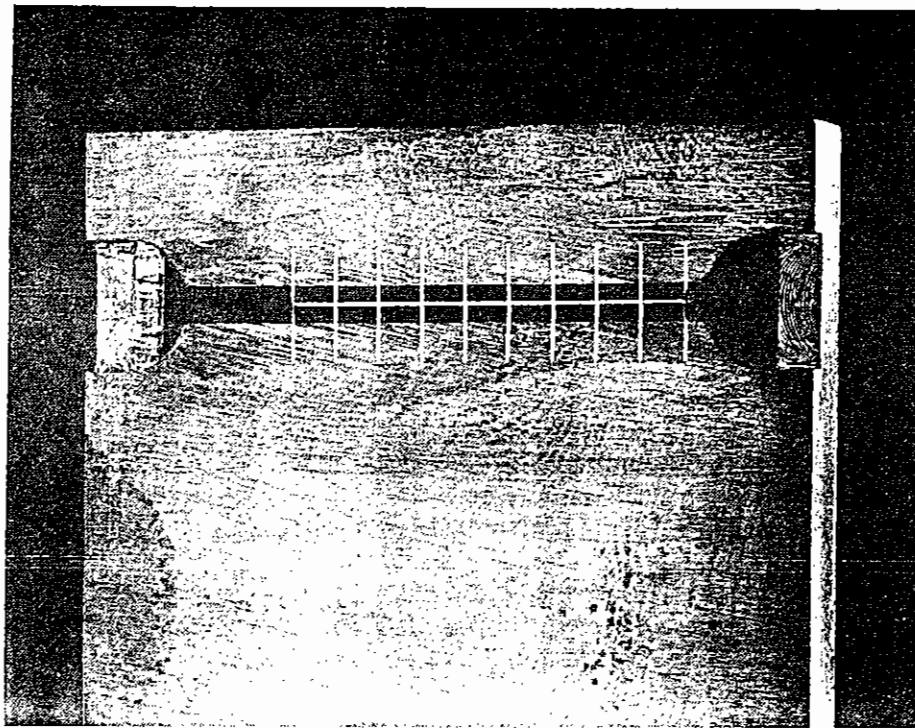


Fig. A1.20 Free-standing aluminium strip target.

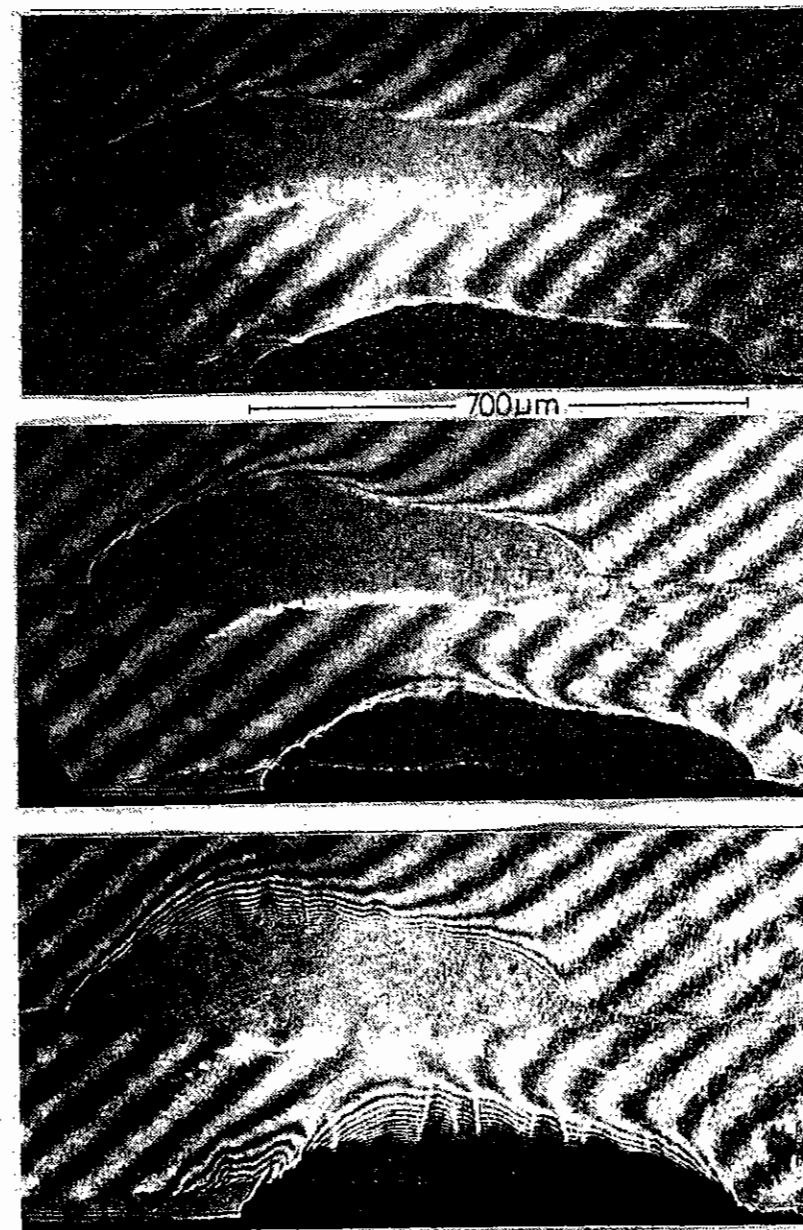


Fig. A1.21 Sequence of interferograms of expanding foil plasma.

A1-7 CORONAL PLASMA ABSORPTION SPECTROSCOPY

C L Lewis, E Robertson, D O'Neill (QUB), T Afshar-rad (RAL)

Introduction

In last year's annual report we described a technique which can potentially monitor ground state ion population distributions at a specified time in the expansion history of a coronal plasma. This is the technique of point-projection absorption spectroscopy⁽¹⁾. Although spatial resolution of $\lesssim 10 \mu\text{m}$ and time-freezing at the 100 ps level were demonstrated absolute ion densities were not accurately determined for two reasons. (a) The scale length of the absorbing plasma was typically $\approx 1 \text{ mm}$ and opacities were too high to measure accurately within the dynamic range of the DEF film recording medium. (b) The absorbing plasma itself was not homogenous either in the fixed direction of a particular probing x-ray photon energy or to the range of directions required for the Bragg matching condition imposed by the PET crystal for a range of photon energies spanning say the resonance series transitions of a He-like absorbing ion. Nevertheless absorption features, without accurate opacity determination, were useful for line classifications particularly for satellite line transitions to many He-like and Si resonance transitions.

In the experiment reported here we unsuccessfully attempted to overcome the plasma homogeneity problem and also attempted to extend absorption measurements from K-shell to L-shell transitions.

Experimental details

The basic set-up was as described in last year's annual report where a 1 ns, 2w pulse irradiates a 250 μm thick and 3 mm wide foil target to create an expanding coronal plasma of highly ionised ions. This plasma is back-lit by a secondary delayed, quasi-point, high brightness and quasi-continuous plasma source generated by irradiating the tip of a wire (or coated fibre) with a $\approx 100 \text{ ps}$, 15 J, 2w pulse focused tightly by an $f 1.5$ lens. In the projected image of the absorbing plasma 1-D spatial information in the expansion directed is retained with the other spatial direction transformed to spectral information due to the dispersing x-ray crystal situated between the plasmas and recording

film. To mitigate against the coronal plasma inhomogeneous previously observed by pinhole camera imaging, the main 1 ns beam was replaced by two separate beams superimposed spatially and temporally on the foil target surface with incident angles of 0° and 20° . The absorbing ion species was spatially confined in the absorption path length direction by coating 100 μm or 200 μm wide stripes of the material on 3 mm wide 50 μm thick mylar foils. With irradiation spot sizes of $\sim 1 \text{ mm}$ the expanding low Z foil plasma should confine the higher Z absorbing plasma and introduce negligible opacity at the energies of interest. Unfortunately pinhole camera imaging of the rectangular shaped high Z plasma still showed marked local heating. A Nomarski type interferometer set up to monitor electron density profiles normal to the original foil surface showed the resulting inhomogeneities in the plasma flow as expected. This is illustrated by a sequence of 20 ps time-frame interferograms at 0.5 ns intervals from the peak of the main heating pulse for a 700 μm focal spot at $10^{12} \text{ W cm}^{-2}$ irradiance on a simple mylar foil (Figure A1.21).

Conclusions

Quantitative data on population densities were not obtained due to the defects in the irradiation profiles mentioned above. This problem will be tackled again possibly using induced spatial incoherence, random phasing or multiply segmented lens techniques. Qualitatively we observed that only Li-like satellite lines were observed in Al stripe targets backlit by Yb coated 20 μm diameter Cu wires, and even these were barely detectable for 100 μm absorption pathlengths. This is to be contrasted with earlier observations of Li-, Be- and B-like satellites when Al and Si foils wider than the 1 mm focal spot were irradiated. This is probably due to the lack of edge effects and colder plasma conditions in the present geometry.

An attempt to observe L-shell absorption features in Sr when back-lit by Yb is illustrated in Figure A1.22. Mylar foils were coated with 200 nm thick SrF_2 stripes of 200 μm width and overcoated with 150 nm of CH and irradiated at $10^{12} \text{ W cm}^{-2}$ in an 1 mm spot. The strontium emission spectrum in the region of interest ($\lesssim 2 \text{ keV}$) is shown in Figure A1.22(a) and was recorded on the PET crystal spectrometer when the SrF_2 target was irradiated under tight focus conditions at $10^{15} \text{ W cm}^{-2}$. The principal features are $3-2$ transitions in the Ne-like ion. Figure A1.22(d) and (c) respectively show the emission spectrum of the Yb

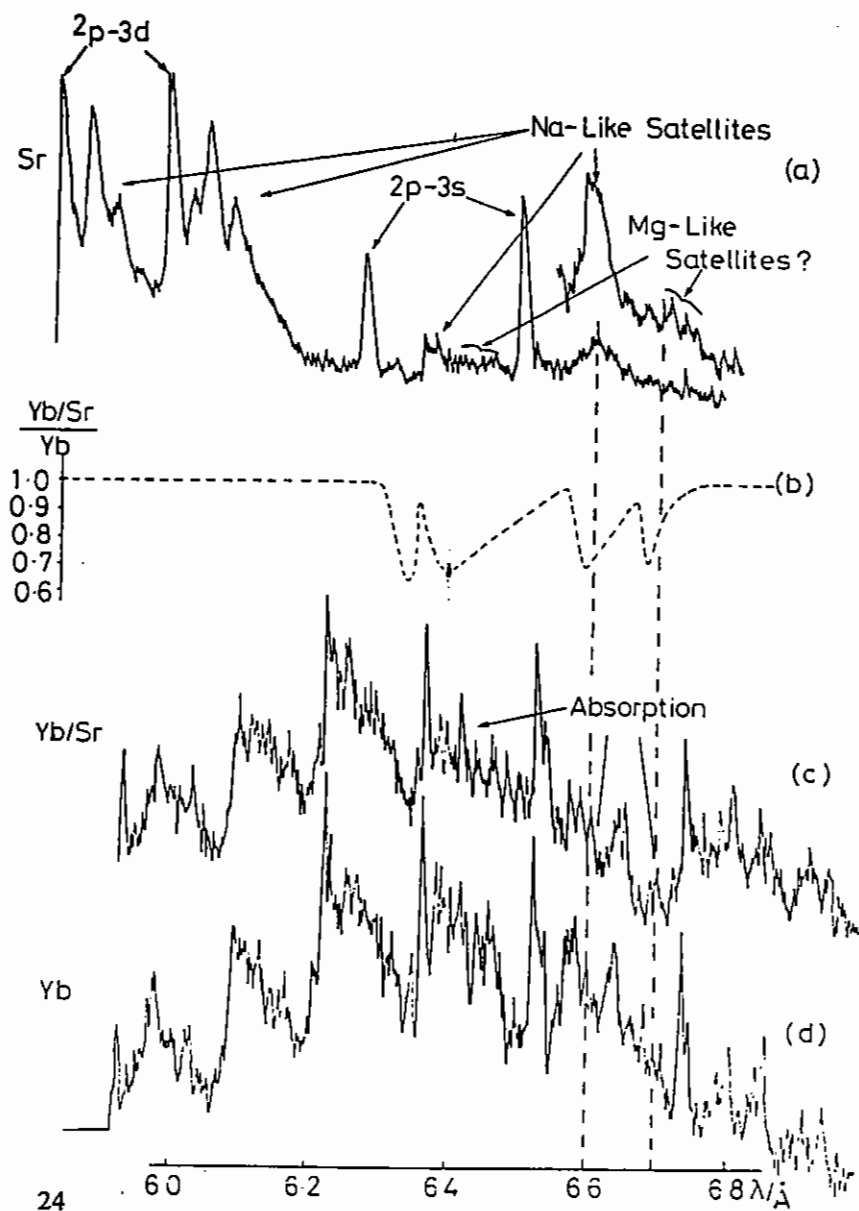


Fig. A1.22 L-shell absorption features in Sr back-lit by Yb.

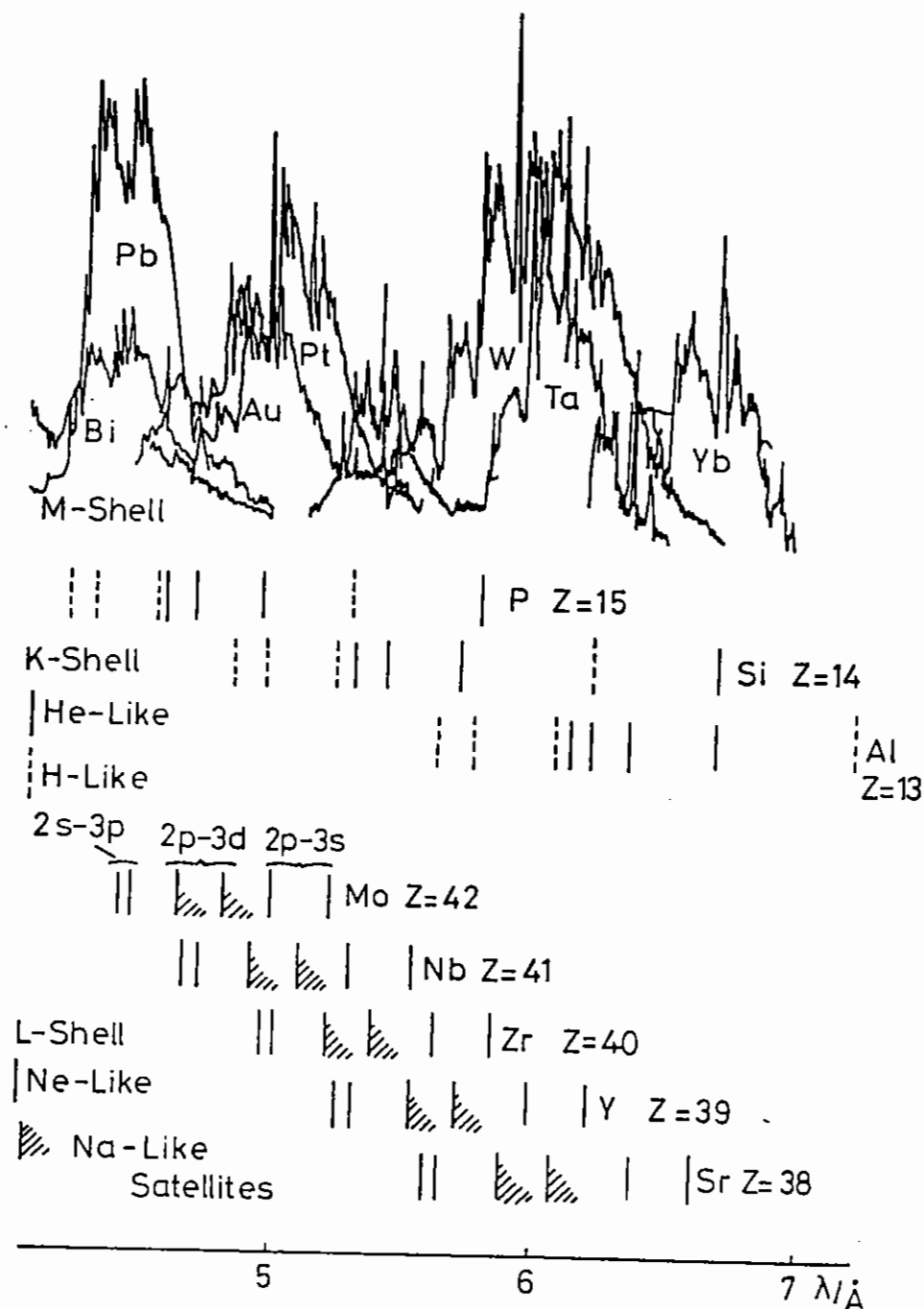


Fig. A1.23 Possible M-shell backlighters.

backlight recorded behind the mylar foil and the same spectrum in front of the foil with some absorption features due to the presence of Sr ions. In this case the backlight was synchronised to the main pulse peak and the low irradiance on the foil (10^{12} W cm⁻²) generated no Sr self-emission. Division of the two spectra yield the plasma transmission profile in Figure A1.22(b) showing opacity peaks to the low energy side of the Ne-like resonance lines. We tentatively attribute these absorption features to Na- and Mg-like satellite arrays. We intend to continue coronal absorption spectroscopic studies and Figure A1.23 illustrates the overlaps of M-shell backlighter for various K- and L-shell absorbers.

A1-8 ABSORPTION SPECTROSCOPY OF A DENSE ALUMINIUM PLASMA

S Davidson, C L S Lewis and D O'Neil (Queens University, Belfast)
S Rose, W S Shaikh and M Grande (RAL)

1. Introduction

The technique of point projection spectroscopy⁽¹⁾ has been used to measure the K-shell absorption of an aluminium plasma from which laser light is excluded. The higher density, lower temperature plasma (typically 0.02 g/cc and 50 eV) produced in this experiment should produce absorption structure characteristic of equilibrium conditions. Comparison of the data obtained with an ab initio calculation of the ion populations and the consequent absorption spectra will be reported at a later date.

2. Experimental description

A schematic diagram of the layout of the experiment is shown in Figure A1.24. The absorption sample used was a planar aluminium foil tamped on both sides with 1 μ Parlene 'N'. This was mounted behind a second foil consisting of 1500 Å gold on a 1 μ Parylene 'N' substrate. The gold was directly heated by a 20-50 J 2w 600 ps laser pulse defocused to provide an irradiance of 4×10^{13} W/cm². The foil remained above critical density throughout the duration of the laser pulse so that x-rays emerged from the rear of the foil to heat the sample but laser light did not.

The heating source size was spatially defined by mounting the gold foil

on a 500 μ hole in a copper shield which also prevented direct emission from the gold coronal plasma reaching the spectrometer.

A second 10 J 2w laser beam of shorter duration (100 ps) was delayed with respect to the first beam and focused onto a 6 μ diameter fibre built up to 10 μ with Parylene 'N' and coated with 1 μ of Samarium. This produced a quasi-continuous M-shell emission of similar duration to the laser pulse which was used to backlight the heated sample.

Direct backlighter emission that passed to the rear of the sample and backlighter emission attenuated as it passed through the sample were both recorded on 'C' type film in the same spectrometer. The target to film distance was typically 90 mm resulting in an effective magnification of 75 for a 1.2 mm source to sample distance. A PET crystal was used for most of the measurements (the advantage of higher resolving power available with ADP was offset by the decrease in signal to noise caused by the poor reflectivity).

Several diagnostics were used to provide measurements of the emission from the heating foil needed to simulate target conditions at the time of the experiment. The absolute x-ray flux was measured with better than 100 ps temporal resolution using a flat response detector⁽²⁾. The spatial distribution of the emitted x-rays were monitored with pinhole cameras and the spectral emission with a time integrated 2000 Å soft x-ray grating⁽³⁾. A pinhole camera with an internal grazing incidence mirror was used to measure the spatial distribution of soft x-ray emission from the rear of the foil.

It was pointed out in the previous report⁽⁴⁾ that instrumentally broadening unresolved saturated structure can produce absorption features that appear unsaturated. Measurements were therefore made at three sample thicknesses, (50 Å, 100 Å and 500 Å mounted both perpendicular and at 15° to the heating foil) to determine when saturated absorption was occurring.

Absorption spectra were obtained at several different times during and after the heating pulse. Duration and relative timing of x-rays produced by the two laser beams was measured using a streaked crystal spectrometer mounted directly above the target.

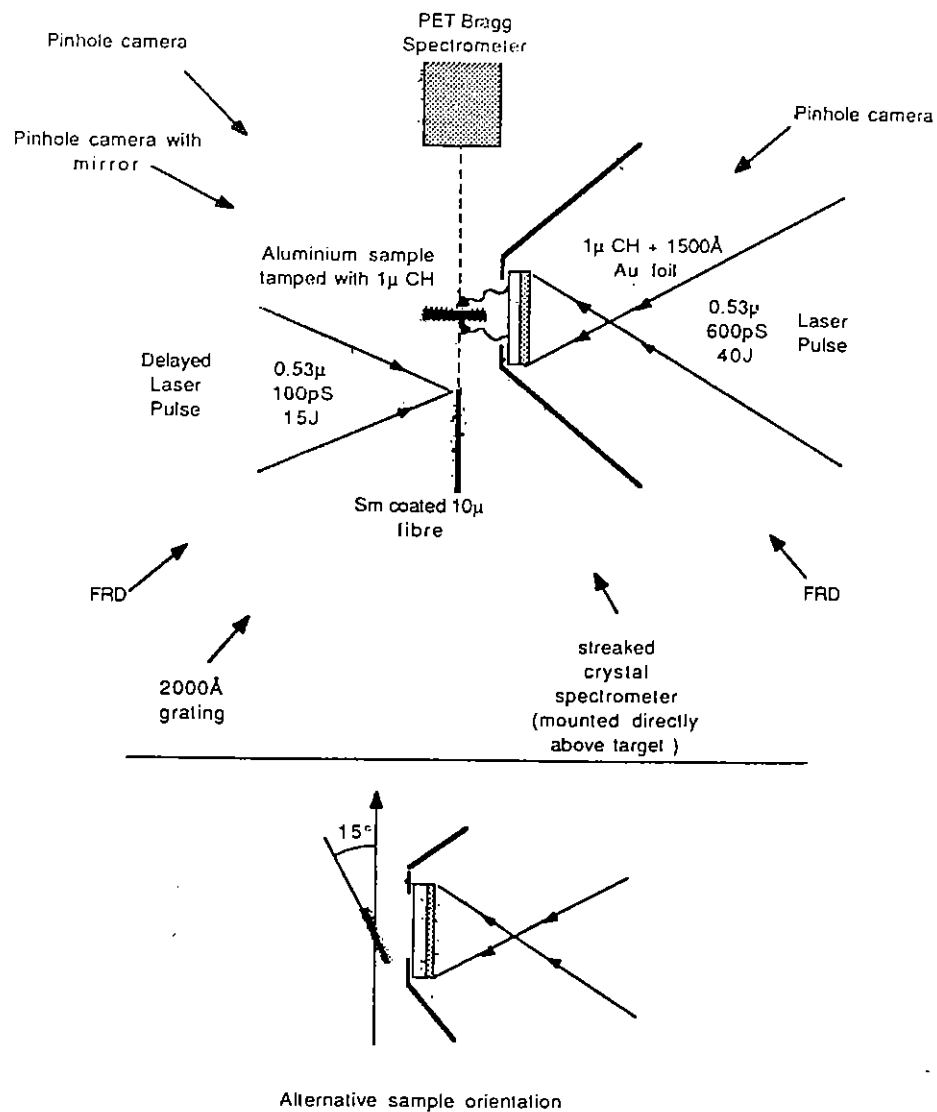


Fig. A1.24 Experimental arrangement.

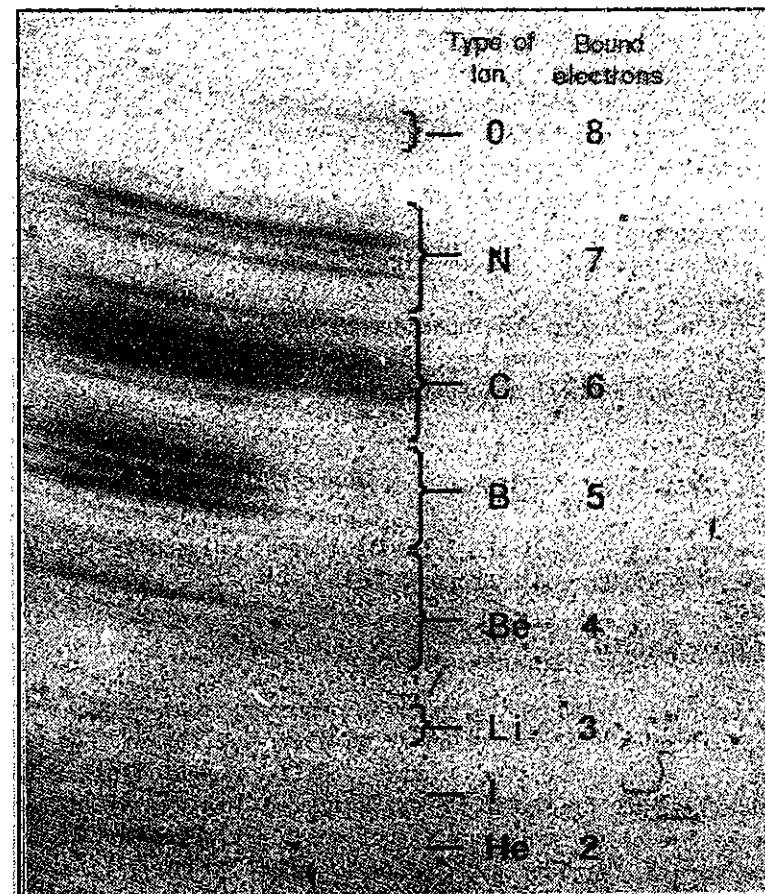
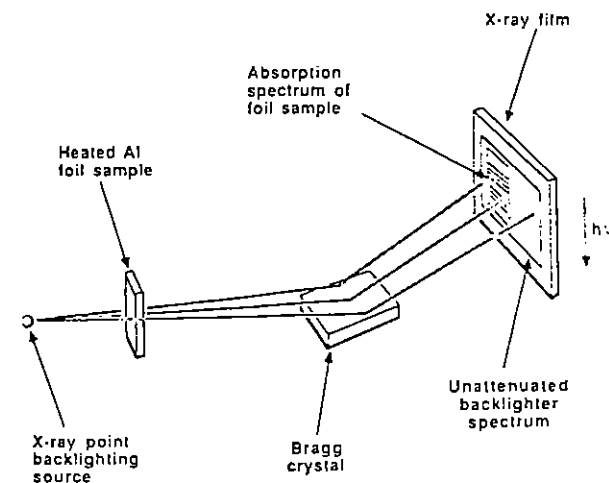


Fig. A1.25 Typical absorption measurement (shot # 6240288).



3. Experimental Results

Delaying the backlighting beam by 600 ps to obtain an absorption spectrum near the end of the heating pulse produced the spectrum in Figure A1.25. The clusters of 1s - 2p absorption lines corresponding to the various ion stages present in the sample at the time of the measurement can clearly be seen.

The spectrum, digitised with 20 μ resolution, was converted to intensity using a standard response curve and numerically post-processed to remove distortions introduced by the spectrometer geometry. It was then divided by a region of unattenuated spectrum to produce the spatial distribution of absolute transmission shown in Figure A1.26.

Scans of transmission taken through differing regions show an ionisation gradient across the sample. This is caused by the 15° sample geometry which results in scans 'A' and 'C' corresponding to regions of sample which are 160 μ and 250 μ respectively from the heating foil.

In order to identify the detailed structure in the spectrum, the multiconfigurational Dirac-Fock (MCDF) program of Grant et al⁽⁵⁾ was used to calculate the absorption lines of the various relevant configurations. A histogram of the position and strength of the absorption lines due to transitions between different terms of the configuration is shown in Figure A1.27. The absorption along scan 'B' is plotted on the same scale for comparison.

The structure seen in the experiment is in broad agreement with the calculated line position; however the detailed line positions only agree to within a few eV. This is not unreasonable since the MCDF calculations were made using the same orbitals for both the initial and final states of the transition. It is known that calculations using separately optimised orbitals can shift the position of the lines of a particular configuration by several eV.

4. Acknowledgements

We would like to thank J C V Hansom for his valuable assistance with the image processing.

References

1. C L Lewis and J McGlinchey, Opt. Comm., 53, 179 (1985).
2. D R Kania, A E Iverson, D L Smith, R S Wagner, R B Hammond and K A Stetler, J. App. Phys., 60, (7) 2596 (1986).
3. O C Barr, D E Campbell, H N Kornblum, V M Slivinski, C E Thompson and J D Wiedwald 5-17 "Laser Program Annual Report" LLNL (1981).
4. C L S Lewis, R E Corbett, E Robertson, D O'Neill, M J Lamb, S Saadat, T Afshar, S Rose, J P Mosnier and E Kenedy, A1.5 Annual Report to the Laser Facility 1987.
5. I P Grant, B J McKenzie, P H Norrington, D F Mayers and N C Pyper, Comput. Phys. Comm., 21, 207 (1980).

A1.9 SPECTROSCOPY OF SHORT PULSE X-RAY SOURCES

C L S Lewis, M J Lamb, D O'Neill (Queen's University of Belfast)
J Edwards (Imperial College, London)
M Grande (RAL)

Introduction

Intense, short lived backlighter emission generated by the irradiation, near the tip, of 10-25 μ m diameter fibres has previously been used to obtain the temporal and spatial opacity of an expanding plasma (Ref 1). Until recently the temporal resolution has been limited by the $\sim$ 100ps short pulse available from VULCAN. An ultra-short pulse ($\leq$ 30ps) oscillator development on VULCAN is in progress and this report describes some preliminary spectroscopy using such short pulses with a view to achieving shorter time-frame backlighters in future experiments.

A study of typical backlighters used previously (W, Yt, Al) was undertaken to observe the effects of the shorter time scale particularly with regard to intensity.

Unfortunately integration of the new oscillator into the system was not straightforward and problems with the pulse selection resulted in a second pulse being delivered on target approximately 8ns after the

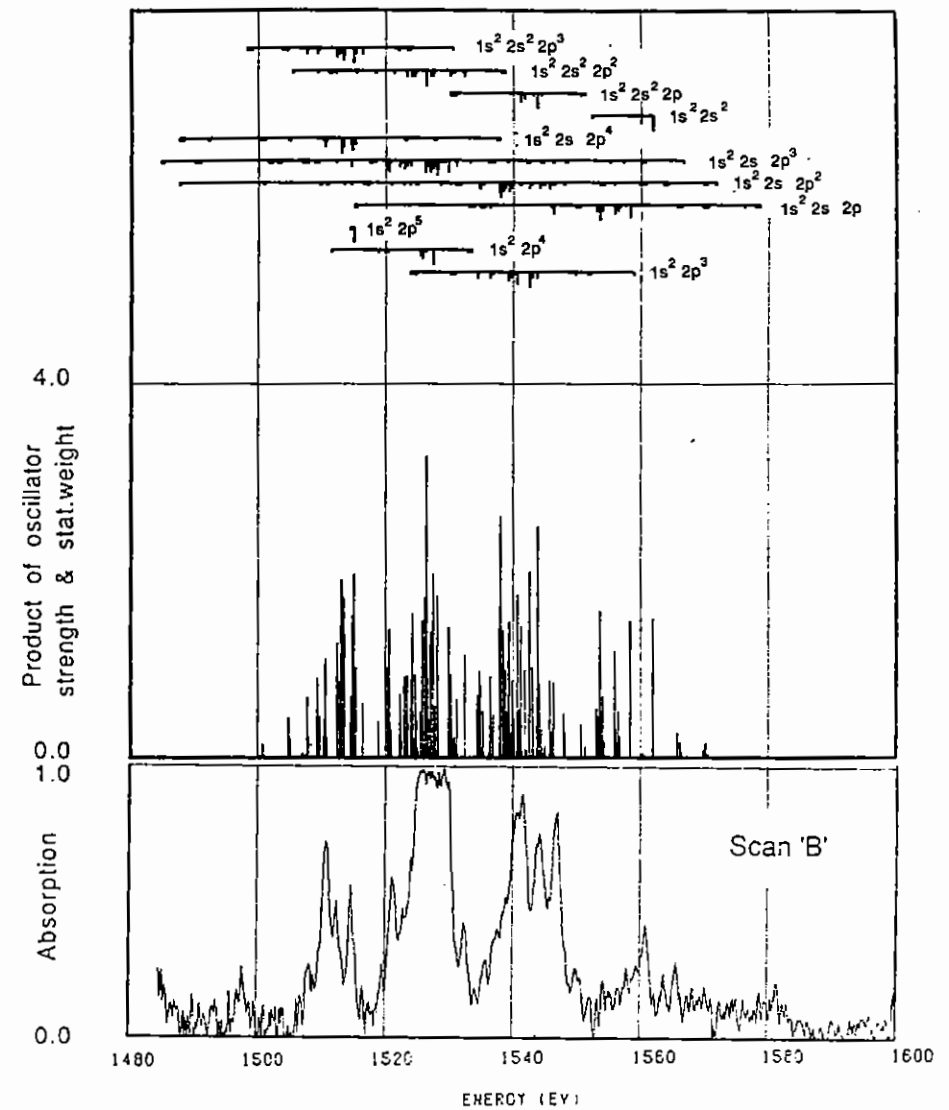
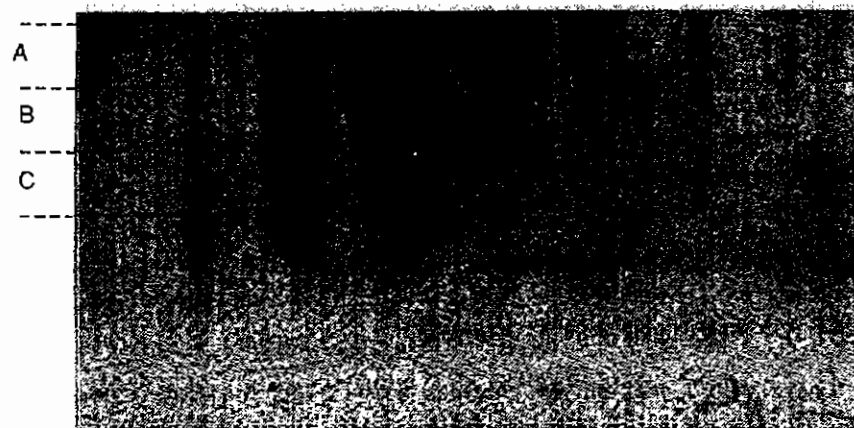
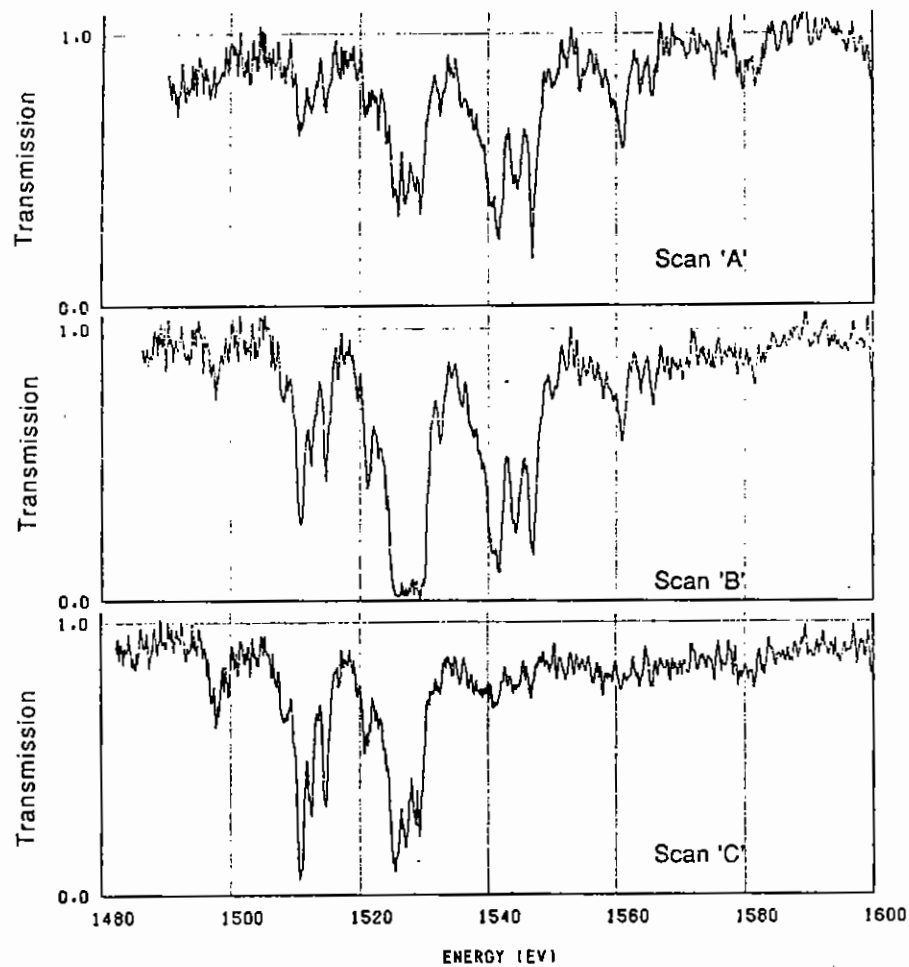


Fig. A1.27 Comparison of HEDF line positions with experimental absorption measured on shot # 6240288.

first. Neither pulse was monitored in the second harmonic and the relative intensities of the two were not constant.

Experimental set-up

Spectra were recorded on Kodak DEF film using a PET crystal spectrometer viewing axially down the fibre with a source to film distance of $\sim 125\text{mm}$. A resolving power of ~ 2000 was achieved in the 1.5 KeV region.

Total 2w energies on target were typically 5-10J tightly focussed by an f/1.5 lens.

Results

Density on film was typically 1.5 - 2 times that obtained from similar energy in 100ps single pulses as seen in a 25 μm Al wire spectra in Fig A1.28 but it is believed that the second pulse is largely responsible for this. The double pulses gave rise to other interesting effects. A grid (20 μm wire, 125 μm period) placed between an 18 μW source and crystal gave rise to two displaced shadows indicating an average velocity of the fibre of $3 - 4 \times 10^5 \text{ cm s}^{-1}$ over the 8ns period.

Also when a tungsten wire was overcoated by 200nm of Al (much greater than the expected burnthrough depth) a weak Al spectrum was observed superimposed on a very intense W spectrum as seen in Figure A1.29.

A more thorough study of ultra-short pulse backlighters will be carried out in the future under more controlled conditions.

References

1. RAL '87 Section A1.5.

A1-10 INVESTIGATION OF SHORT PULSE, HIGH BRIGHTNESS X-RAY SOURCES

C Lewis, D O'Neill (Queens University Belfast)
S Davidson, J Freeman, D Perkins (AWE, Aldermaston)
M H Key, A Ridgeley, G Tallents (RAL)

Plasmas produced by focussing laser radiation onto solid targets in vacuum are bright laboratory sources of X-ray emission. The emission is on a timescale comparable with that of the laser pulse duration (down to ps) and consequently can be employed for time-resolved measurements. Such X-ray pulses have been used for some time for radiography of laser compressed plasmas^(1,2) and also for extended X-ray absorption fine structure (EXAFS) spectroscopy^(3,5), pulsed X-ray diffraction⁽⁶⁾ and X-ray microlithography^(7,8). A joint experiment in the laser unit with the Atomic Weapons Establishment (AWE), Aldermaston was undertaken to find bright laser-produced plasmas suitable as point source backlighters for radiography of the laser-compressed plasmas produced in both laboratories. The use of a laser-produced plasma as a point source backlighter for diagnosing implosion experiments places some constraints on parameters for RAL and AWE viz:

- (i) The x-rays need to have photon energies λ 4 keV with the bandwidth of x-ray emission $\lesssim 10\%$.
- (ii) The x-ray emission needs to be for times $\lesssim 100 \text{ ps}$.
- (iii) The emission should come from an area with dimension $\lesssim 15 \mu\text{m}$.
- (iv) A single laser beam should be used with laser energy $\lesssim 50 \text{ joules}$.

In an experiment of 2 weeks duration, the following parameters were varied in order to maximise x-ray production within the above four constraints;

- (i) The length of plasma along the line-of-sight. Line focus and spot focus illuminations of long target geometries were investigated.

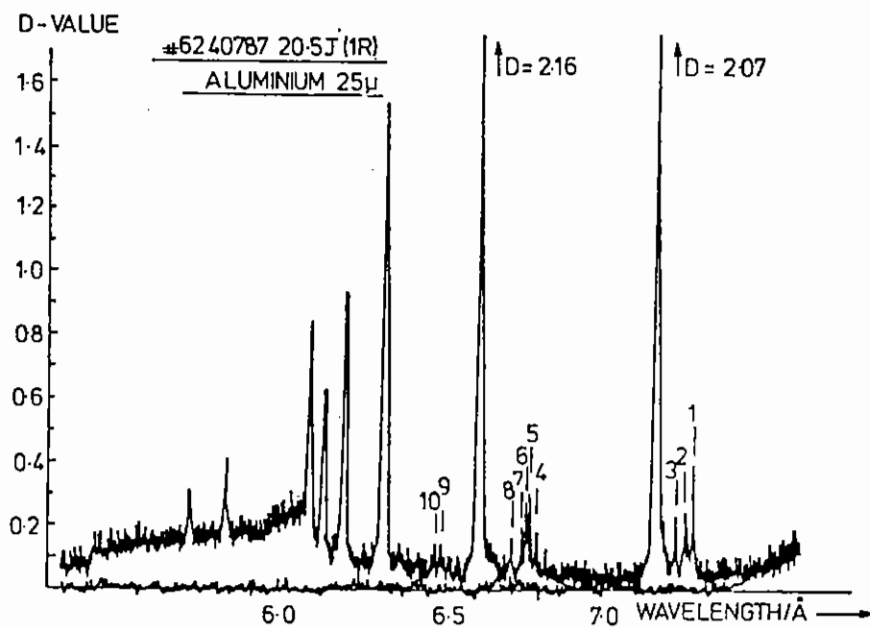


Fig. A1.28 Al spectrum.

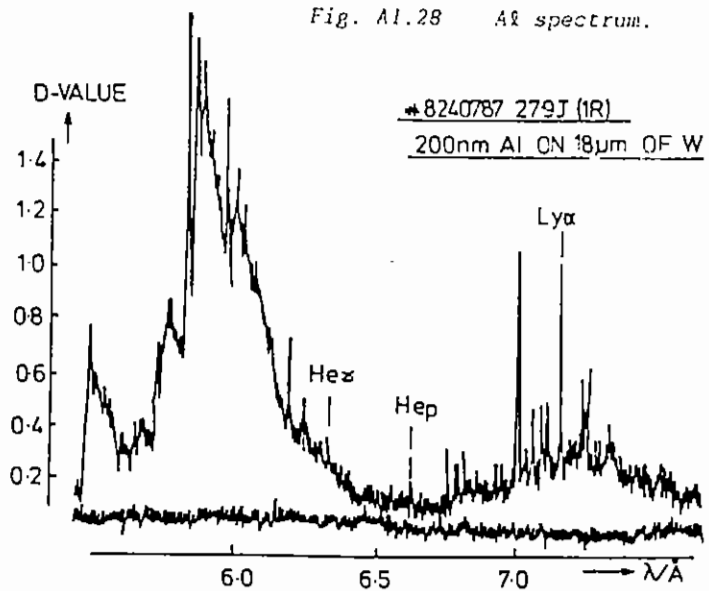


Fig. A1.29 Al overcoated tungsten wire.

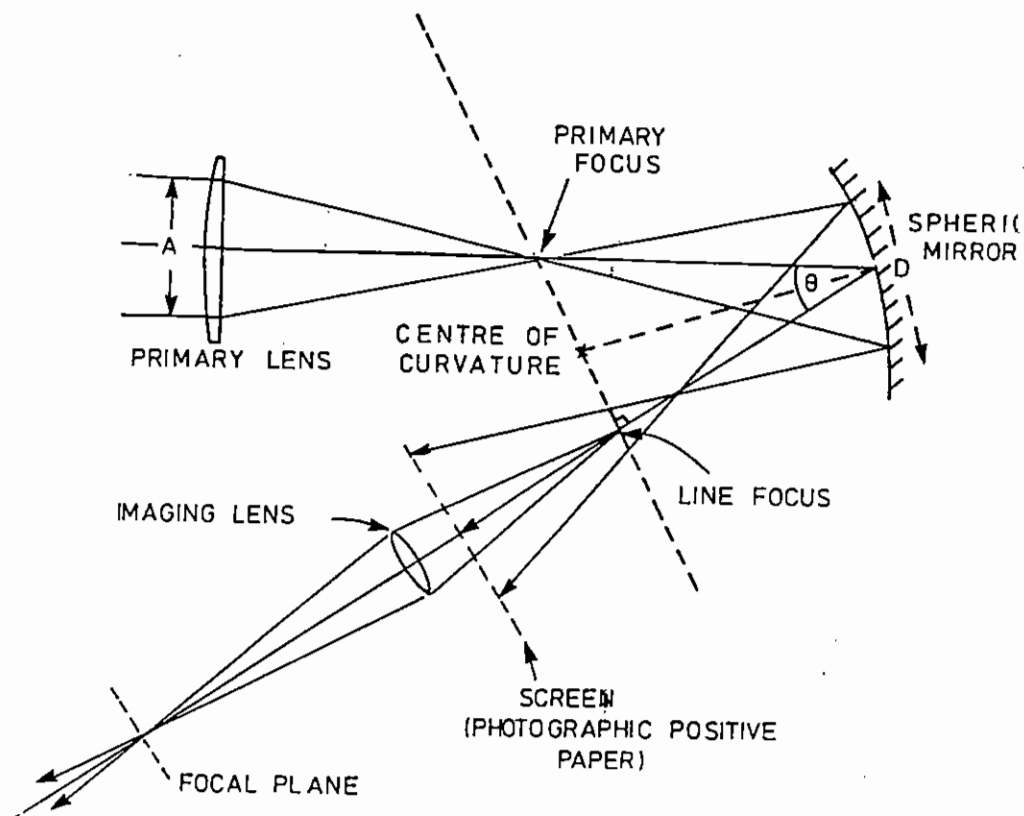


Fig. A1.30 Schematic of experiment.

(ii) The target material. Targets of Ti and Ca (K shell emitters in the 3-5 keV photon energy range) and Sn, Te and CsI (L shell emitters in the 3-5 keV energy range) were investigated.

(iii) The geometry of the target. Fibre, stripe, dot and stripes and dot targets in depressed geometries (at the bottom of channels or holes in plastic) as well as slab targets (for spectral identifications) were investigated.

Diagnostics employed in the investigations were:

- (i) A Germanium crystal spectrometer (photon energy range 3-5 keV).
- (ii) A Germanium crystal minispectrometer (photon energy range 3-5 keV).
- (iii) A Titanium filtered X-ray streak camera (photon energy range 3-5 keV).
- (iv) Pinhole camera (Be filter).
- (v) Silicon photodiode (Ti filter).
- (vi) Film pack (Ti filter).

These instruments are of varying sensitivities to X-rays which enabled photon flux measurements over a range of more than three orders-of-magnitude to be made. Laser energies at .53 μm wavelength were typically 20-30 joules.

A comparison between photodiode, film pack and spectrometer exposures has shown that the emission intensities are amply sufficient for radiography if the spectra can be recorded by the spectrometers. The magnitudes of the target emissions were broadly comparable to those recorded by Phillion and Hailey⁽⁹⁾ (eg $\sim 10^{12}$ photons/joule/4 π steradian for a Ti slab target). Shots with a 0.6 mm line focus on titanium slab targets produced measurable emission from titanium around 4.75 keV, but the relative intensities of the spectral lines (helium-like titanium and lithium-like satellites) indicated that the plasma was relatively cool in comparison to spot focus shots. The emission intensities in the line focus geometry were also less than for the spot focus.

Tin, calcium, tellurium and cesium iodide spectra were recorded with spot focus shots. Tin and calcium gave more emission for comparable conditions than titanium, but at lower photon energies. The tin spectrum consists of many lines in the 3-5 keV spectral range, with the more intense lines being of lower energy. The cesium/iodine and tellurium spectra were recorded with a slab target only, and were not very intense. The duration of emission in the 3-5 keV range was typically the same as the laser pulse (= 100 ps) for all targets.

The effect of target geometry on the emission was studied for tin and titanium targets. Stripe targets produced more emission than fibre targets, but the depressed stripe targets did not produce sufficient emission to record with the spectrometers. The depressed stripe targets consisted of an overlay of 50 μm thick plastic on titanium or tin through which a channel = 75 μm on the laser side narrowing to $\sim 15 \mu\text{m}$ at the metal was cut. The imaging of a grid onto a film pack indicated that the spatial dimension of emission when looking down the fibres or stripes was $\leq 20 \mu\text{m}$. Dot targets produced less emission than fibre targets, but the spectra could still be recorded with the spectrometers.

Conclusions of the investigation can be summarised as:

- (i) A spot focus gives more x-ray flux than a line focus.
- (ii) Tin targets give the most X-ray emission for a given laser energy in the 3-5 keV range, but titanium targets produce harder X-rays (at 4.75 keV).
- (iii) Stripe targets produce more x-rays than fibre targets and have comparable spatial and temporal emission characteristics.

References

1. M H Key, P T Rumsby, R G Evans, C L S Lewis, J M Ward and R L Cooke 45 1801 (1980).
2. D L Matthews et al, J Appl Phys 54 4260 (1983).

3. P J Mallozi, R E Schwerzel, H M Epstein and B E Campbell, Phys Rev A23 824 (1981).
4. R W Eason, D K Bradley, J D Kilkenny and G N Greaves, J Phys C17 5067 (1984).
5. G J Tallents, B Luther-Davies and M A Horsburgh, Aust J Phys 39 253 (1986).
6. R D Frankef and J M Forsyth, Science 204 622 (1979).
7. D J Nagel, C M Brown, M C Peckeran, M L Ginter, J A Robinson, T J McIlrath and P K Carroll, Appl Opt 23 1428 (1984).
8. F O'Neill, M C Gower, I C E Turcu and Y Owadano, Appl Opt 25 464 (1986).
9. D W Phillion and C J Hailey, Phys Rev A34 4886 (1986).

A1-11 UNIFORMITY OF LINE FOCUS ILLUMINATION FOR X-RAY LASER TARGETS

M Yamanaka, M Grande, W Shaikh and A Qamar (RAL)

An experimental study has been made to clarify the origin of blow-off plasma structures⁽¹⁾ observed from the 7 μm diameter carbon fibre targets used in x-ray laser experiments, when irradiated by intense 0.53 μm laser light (about 20 J at 100 ps) using the TAE line focus geometry⁽²⁾.

Figure A1.30 shows the experimental arrangement used. A photographic positive paper was set at the position of screen and exposed to the intense 0.53 μm laser light scattered from the carbon fibre. A typical example is shown in Figure A1.31(a). The structures may be explained as a coherent superposition of Fresnel diffraction patterns⁽³⁾ from fine filaments of blow-off plasma from the carbon fibre surface (generated during the laser pulse of 100 ps). To simulate this effect, a fibre was seeded with dust particles, and the resulting transmitted beam photographed in CW Argon ion light (see Figure A1.32). The dust particles (a few microns diameter) obstructed the escaping argon-ion light like thin wires. Since the line focus is transversely focused to $< 10 \mu\text{m}$ which is completely obstructed by the dust particles

on the fibre, and the distance between line focus and screen is not great, about 30 cm, a typical Fresnel diffraction pattern is produced. Figure A1.31(b) shows a burn pattern from a seeded fibre. From Figure A1.31, we can estimate the width of the blow-off plasma filaments to be on the order of a few 10 μm . This is in good correlation with observations by the Schlieren technique⁽¹⁾.

We used a (Zeiss) lens to observe how green argon-ion light was line-focused onto the 7 μm carbon fibre (Figure A1.30). At the focal plane, without a fibre in place, the line focus image was observed to be strongly affected by the aberration of the imaging lens itself (Figure A1.33(a)). The one-dimensional Fourier transform of the input beam due to the mirror is observed, as illustrated by putting a one dimensional 8 mm width grid before the primary lens and then observing a typical image of multiple-slit Fraunhofer diffraction at the line focus (Figure A1.33(b)).

We next observed the scattered green argon-ion laser light from the carbon fibre. Initially, we again obtained an aberrated image due to the lens used (Figure A1.33(c)). This was improved by aperturing the lens, in order to reduce flare (Figure A1.33(d)). It appears that the variations in intensity along the fibre correspond to fibre inhomogeneities which may provide the seeds of the plasma structure observed in the blow-off.

In conclusion following experiments in CW light in TAE it is clear that while the line focus does not suffer from non-uniformities due to coherent optical effects, the fibres themselves are subject to irregularities which certainly lead to large inhomogeneities in the CW light scattered from a fibre in the line focus, and are quite possibly the seeds for non-uniformities observed in the blow-off plasma and broad band x-ray emission. The structure in the burn pictures is due to the superposition of Fresnel diffraction patterns from these filaments. It would be interesting to investigate this connection further by taking well characterised fibres, observing them first in CW light, then in two minute shots and finally with main shots. The main shots could be diagnosed using x-ray pinhole cameras axially and transversely, and the same optical imaging system using filters to alternative pass and reject scattered 2w light, with the aim to see whether structures on the fibre are correlated with structures in the plasma emission and light scattered from the plasma, and whether fibre uniformity is a parameter requiring further optimisation in x-ray laser experiments.



Fig. A1.31 Comparison of burn papers take on fibres, without (A) and with (B) dust seeding.



Fig. A1.32 Scatter from dust on fibre.

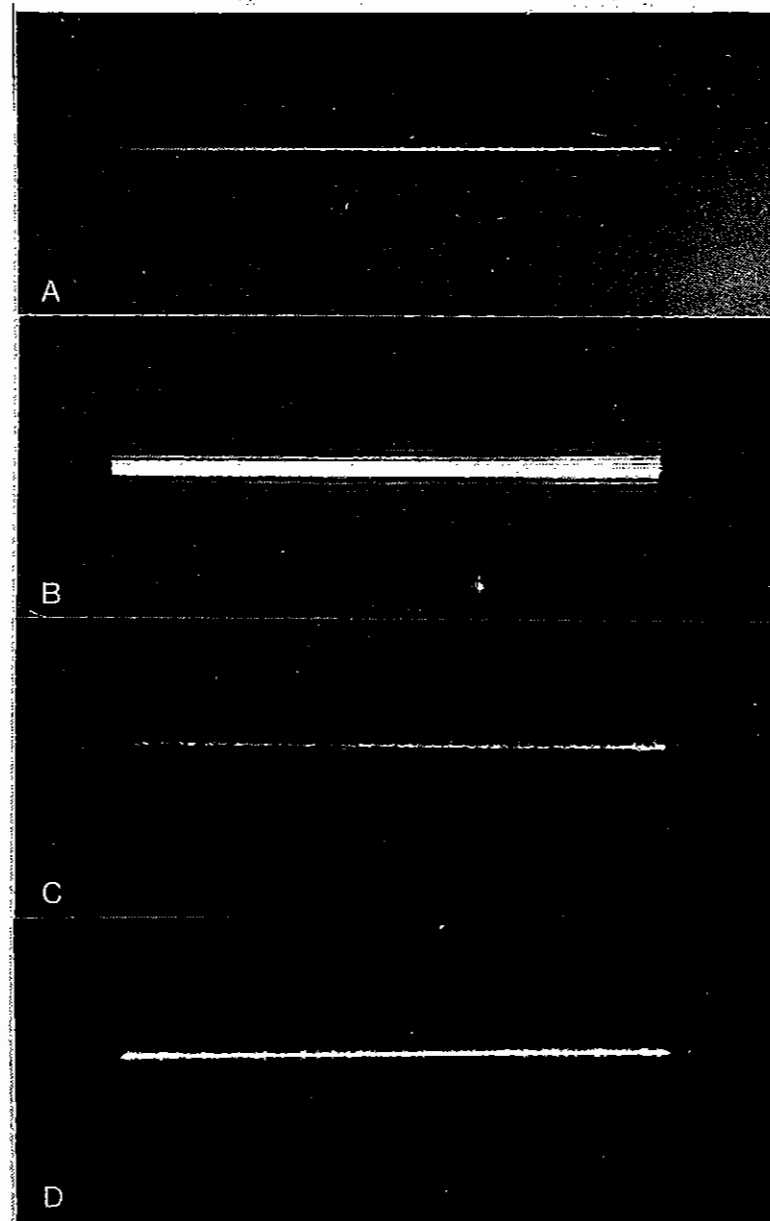


Fig. A1.33 (A) Image of line focus.
(B) Image of line focus with modulated beam.
(C) Scatter of beam from fibre.
(D) As (C) but with imaging lens appertured.

References

1. D Bassett, O Willi, T Afshar-Rad, M Grande and W Shaikh, Annual Report to the Laser Facility Committee 1987, Rutherford Appleton Laboratory RAL-87-041, p17.
2. I N Ross, J Boon, R Corbett, A Damerell, P Gottfeldt, C Hooker, M H Key, G Kiehn, C Lewis and O Willi, Appl. Opt., 26, 1584 (1987).
3. See, for example, F L Pedrotti and L S Pedrotti, "Introduction to Optics" Prentice-Hall, Inc, New-Jersey, 451 (1987).

A1-12 X-RAY DIFFRACTION FROM SHOCKED CRYSTALS

J S Wark, R R Whitlock, A Hauer

It has recently been shown that sub-nanosecond bursts of laser-plasma produced x-rays can be a novel and useful method of measuring crystal structure and lattice spacings in laser-shocked crystals¹. The basic concept of the technique is as follows. When a crystal is shocked the interatomic spacing of a given crystal plane is reduced, and thus the Bragg angle of diffraction for x-rays of a given wavelength is increased according to a differentiation of Bragg's law

$$\frac{\Delta(2d)}{2d} = - \cot \theta_b \Delta\theta$$

Thus a measurement of the shift in Bragg angle directly yields information on the compression.

The original experiment described in reference 1 measured the transient (111) lattice spacing of a shocked Silicon (111) crystal, i.e. the elastic component of the strain. In the experiment described here, both the (111) plane and (113) spacings were measured simultaneously.

A measurement of both planes was made for two main reasons. First, from the simultaneous measurement of two planes it should be possible to extract the transient unit-cell shape, and thus gain information on the plasticity (i.e. strain perpendicular to the shock front).

Secondly, because of the smaller lattice spacing of the (113) plane, the technique should be sensitive to smaller values of strain.

The experimental set-up is shown schematically in Figure A1.34. The Silicon crystal is coated with 1000 Å of Al to act as an abrupt absorber to the 1.05 µm drive beam. A further coating of 25 µm CH allowed transmission of the drive beam at the irradiances of interest ($\sim 5 \times 10^9$ W cm⁻²) but confined the Aluminium plasma to the crystal surface, thus increasing the amplitude and duration of the compression. The crystal was irradiated by a 1 ns pulse of 1.05 µm light, containing $\sim 10 \rightarrow 50$ J, and with a focal spot on target of ~ 4 cm. A beam block prevented part of the crystal from being irradiated, diffraction from this part of the crystal acted as a reference for the measurements of shift in Bragg angle.

During the shock compression a second laser beam, synchronous and delayed with respect to the first, was focused to a small (~ 40 µm) spot on a calcium - containing target. This beam contained ~ 15 J of 0.53 µm light in a pulse of ~ 100 ps FWHM. The resultant plasma was a prolific source of Helium-like calcium line radiation. The He_α resonance line was Bragg diffracted from both the irradiated and unirradiated portions of the crystal, and the diffraction from the (111) and (113) planes was recorded on x-ray film. Figure A1.34 shows diffraction data; the shifts in Bragg angle corresponding to a peak compression of the (111) planes of a few %.

The Hugoniot Elastic limit of silicon (i.e. the point where the crystal starts to respond plastically) occurs at a compression of $\sim 2.6\%$ ⁽²⁾. Unfortunately, for compressions of this order the dispersion of the (113) plane was such that the x-ray signal could not be recorded on film; thus we were unable to make a unit cell shape measurement in the compression region of interest. However, this dispersion did allow us to observe small values of strain. Figure A1.35 shows simultaneous diffraction data for the two planes with a low shock-driver energy. There is another interesting feature which occurs at even lower irradiances. Although no shift is observable in the x-rays diffracted from the shocked (111) plane, there is a significant increase in integrated reflectivity ($\sim 50\%$), as can be seen from the densitometer tracings (A1.36).

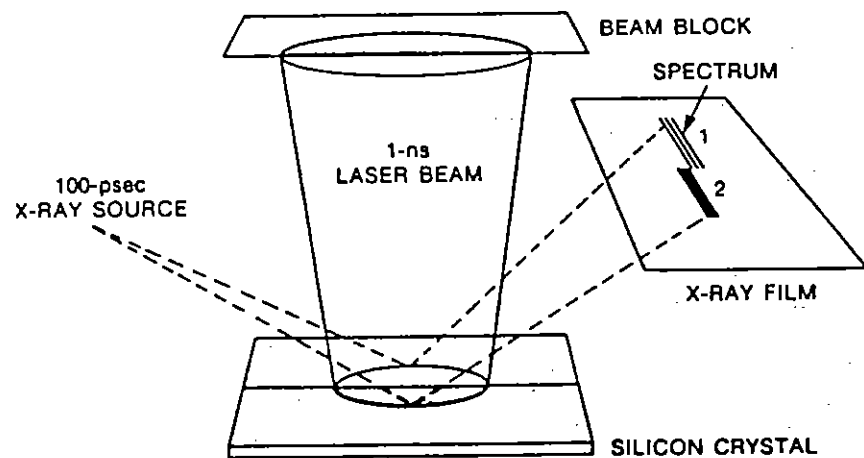


Fig. A1.34 Schematic experimental layout.

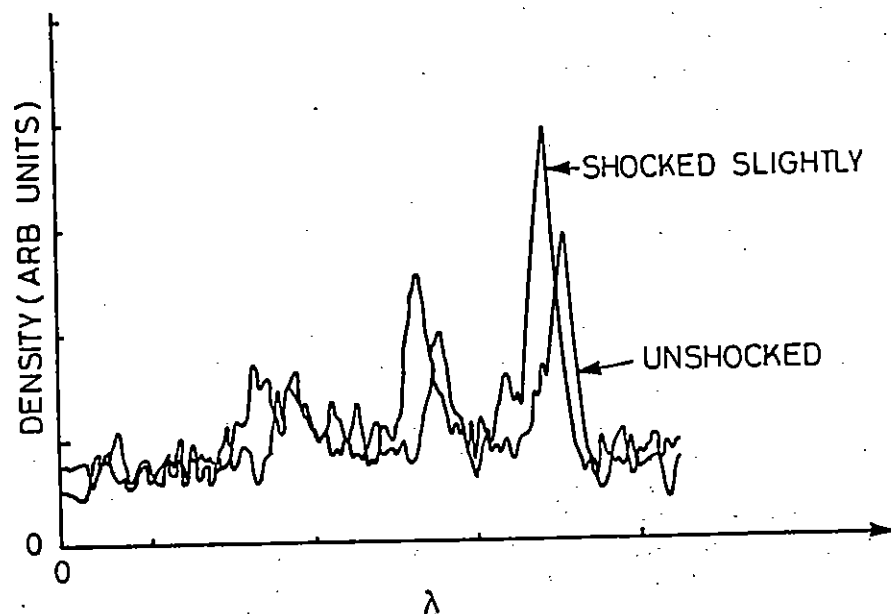
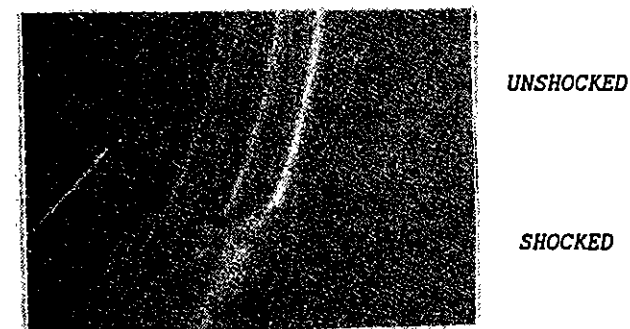


Fig. A1.36 Spectrum of He-like Ca diffracted from perturbed and unperturbed Si crystals, showing an increase in reflectivity for a slight perturbation of the crystal lattice.



(a)



(b)

Fig A1.35 Diffraction from a shocked crystal, (a) From the (111) plane and (b) From the (113) plane.

Work currently being performed by the authors using an x-ray diffraction code shows that large changes in integrated reflectivity can occur for crystals with small strain gradients. This change is associated with a deviation in the diffraction away from the dynamical regime, and towards the kinematic. Such strains can occur in x-ray crystal spectrometers for the brief time they are subject to the high radiation flux environment in high-power laser-plasma experiments. Such strains would alter the reflectivity of the instrument on the time scale of the data collection, and may invalidate the crystal calibration. These effects could be particularly important in time resolving crystal spectrometers, where the reflectivity will change throughout the recording of the streaked data. Thus the data obtained in this experiment will act as a useful comparison with the x-ray diffraction code, and will help in understanding crystal x-ray reflectivity response in extreme radiation environments.

Reference

1. J S Wark, R R Whitcock, A Hauer, J E Swain and P J Solone, Phys. Rev. B, Vol 35, 9391 (1987).

A1-13 X-RAY SOURCES GENERATED USING 30 ns KrF LASER PULSES AT ENERGIES UP TO ~ 30 J.

F O'Neill, G M Davis, M C Gower, I C E Turcu (RAL)
C W Wharton (Birmingham).

High rep. rate laser-plasma x-ray sources are being used (or are being considered) for a number of applications at the CLF such as x-ray microscopy (see sect. B3-4 of this report), x-ray lithography (sect. B4-3) and for the study of DNA damage repair (sect. B2-2). This work to date has concentrated on using commercial, discharge-excited, repetitive KrF excimer lasers for generating and heating the laser-plasma source. The KrF laser has a number of advantages over Nd:YAG lasers for this application for example:-

- (1) high overall laser efficiency (= 2%)
- (2) UV (249 nm) laser is converted very efficiently to plasma x-rays

- (3) gaseous medium gives low divergence beams under high rep. rate operation.

The main disadvantage is that in normal operation the commercial KrF laser emits pulses of 25 ns duration and < 1 J energy/pulse. There is therefore some difficulty with such relatively low power pulses in achieving a high enough irradiance on target to efficiently make x-rays.

For applications requiring radiation at $h\nu < 1$ keV (e.g. x-ray microscopy of biological cells) it appears that existing commercial KrF lasers are capable of generating sufficient quantities of x-rays (see sect. B3-4). For applications requiring x-rays at $h\nu > 1$ keV we have shown (sect. B4-3) that it would be beneficial to use laser pulses of higher energy than are presently available. For example we have shown that ~ 1 keV x-rays are only produced at ~ 0.2% efficiency when 0.3 J KrF laser pulses are focused onto Cu targets at an irradiance of $I \sim 2 \times 10^{13}$ W/cm². The x-ray conversion efficiency is however increasing in proportion to I^2 at this level of irradiance and it is of interest to study the scaling to higher energy pulses, focused to higher irradiance. It is expected that such a scaling will give a much higher plasma temperature and improved conversion to x-rays at $h\nu > 1$ keV.

In this reporting period we have therefore carried out an experiment to study the scaling of x-ray conversion efficiency for high energy KrF laser pulses. We have measured x-ray conversion efficiency as a function of target irradiance using the Sprite laser working in an oscillator-amplifier mode. The laser delivered on-target energies up to ~ 30 J in 30 ns pulses at irradiances up to 1.5×10^{14} W/cm² onto Cu targets. This pulse length and maximum energy has been chosen to simulate (and exceed) the highest performance likely to be achieved from a scaled-up discharge-excited module.

The results from this experiment are still undergoing analysis and cannot yet be discussed in detail. It has been found however that when using Cu targets to generate ~ 1 keV L-shell emission the x-ray conversion efficiency continues to scale with I^2 up to irradiances $\approx 5 - 10 \times 10^{13}$ W/cm² at which level the efficiency saturates at a few %.

A1-14 X-RAY SOURCES FOR SMALL ANGLE SCATTERING

A R Faruqi (MRC Cambridge)
A J Cole, M Grande and M H Key (RAL).

Laser produced plasmas offer an attractive source for time resolved studies of transient phenomena such as muscle diffraction, since their high brightness relative to synchrotron radiation single pulses makes single pulse recording practical in principle with a sufficient signal to noise ratio to give useful data.

Muscles are built up from regular helical arrays of protein molecules which gives rise to x-ray diffraction patterns having basic periodicities of the order of 400 - 500 Å. These patterns can give information about the structural changes which produce muscular contraction, an important phenomenon whose detailed mechanism is not yet understood. However, with conventional sources, the patterns are relatively weak, and even with synchrotron radiation, intensity is a limiting factor in recording time-resolved data, especially on small specimens such as single muscle fibres.

A trial experiment was carried out in order to investigate the suitability of a laser produced plasma x-ray source for small angle diffraction experiments with a view to investigating muscles. The experimental layout is shown in Figure A1.37; the diffraction pattern on the phosphor is viewed via an image intensifier.

The major problem encountered in the experiment is the very low (10^{-5} - 10^{-7}) diffraction efficiency of the specimens involved. This means that very efficient zero order suppression is necessary. Experiments were carried out with trial specimens consisting of bundles of collagen fibres which have a well established axial periodicity of ~ 660 Å. Using the 3.7 keV He-like resonance line of Ca as our x-ray source (driven by typically 200 J green focused onto a 100 μ m pellet) produces first order diffraction peaks 10 mR apart. The beam collimation, defined by the source and slit is ≈ 2 mR. However, it was found that a significantly larger zero order stop was required, on the order of 6 mR. This led to a zero order suppression estimated at $\sim 10^4:1$. This width of the zero order is probably explained by an extended plasma plume forming in front of the target pellet, and would result in the diffracted orders being smeared beyond recognition, both from low energy wings on the zero order, and also due to broadening of the first order; certainly no first order peaks could be identified

with confidence. In future experiments great care must be taken in the collimation of the input beam of x-rays, probably by adding an extra slit.

A1-15 SOFT X-RAY CONTACT MICROSCOPY OF BIOLOGICAL SPECIMENS: ALUMINIUM COATED SILICA NITRIDE WINDOWS USED AS U/V FILTERS

A D Stead, T W Ford and J A Catcheside (Dept. Biology, RHBNC),
C P Hills (Dept. Physics, Kings College), A Ridgeley (RAL)

Soft x-ray contact microscopy of biological specimens using laser-produced plasmas as a source of x-rays at RAL has been reported previously (Rosser et al., 1985 Eason et al., 1986; Michette et al., 1986, Stead et al., 1987). In these experiments, however, it has been assumed that the Si_3N_4 windows, used to maintain the samples under atmospheric conditions, acted as a filter against u/v radiation. Closer examination reveals that this cannot be assumed, and thus images produced using laser-plasmas may have been the result of exposure to ultra-violet radiation, or indeed harder x-rays. This is of considerable importance since it is only x-rays in the so-called "water window" (wavelength 2.2 - 4.4 nm) which will give maximum contrast between carbon and oxygen containing components of the cell. The longer u/v wavelengths would severely limit the resolution of the technique, and significantly lower the contrast between protein and water.

The spectral output of the laser-produced plasma has only been fully investigated in very few instances. Rosser et al. (1985) reported that using VULCAN in conjunction with graphite targets, the line spectrum emitted very strongly at 3.37 and 4.03 nm, both within the "water window", but radiation was also produced at 13.00 nm - outside the "water window". Using mylar targets and a KrF laser (SPRITE), it has been reported that there are similar peaks of emission in the "water window" region, however, there is also a background continuum radiation extending either side of this critical region (Eason et al., 1986). Similarly, data from a 0.53 μ m wavelength laser produces a spectrum with a peak emission in the "water window", but again, significant amounts of energy are produced outside this critical region (Mochizuki et al., 1986). The emission in the lower energy u/v region is of particular concern as the photosensitive recording medium that is used in these studies (PMMA) absorbs strongly in the u/v region, and thus it

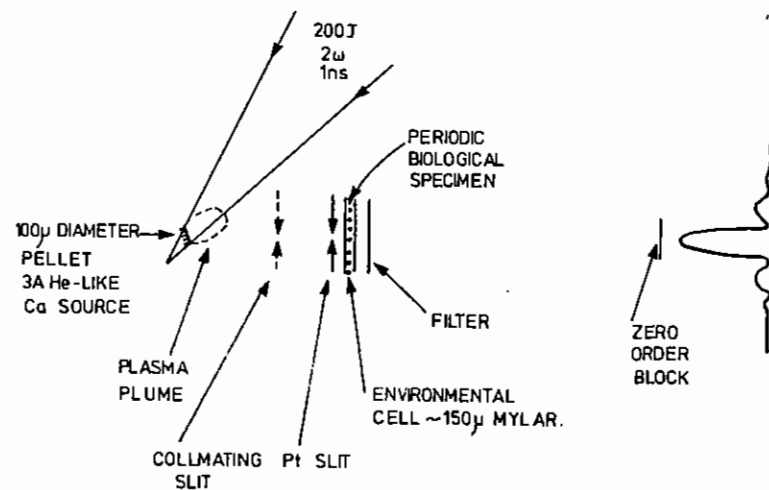


Fig. A1.37 Schematic of experimental set-up.

Fig. A1.38 Relative response vs wavelength to a gold plasma spectrum of a PMMA resist through a Si_3N_4 window with (solid) and without (dashed) a 625 nm Al filter.

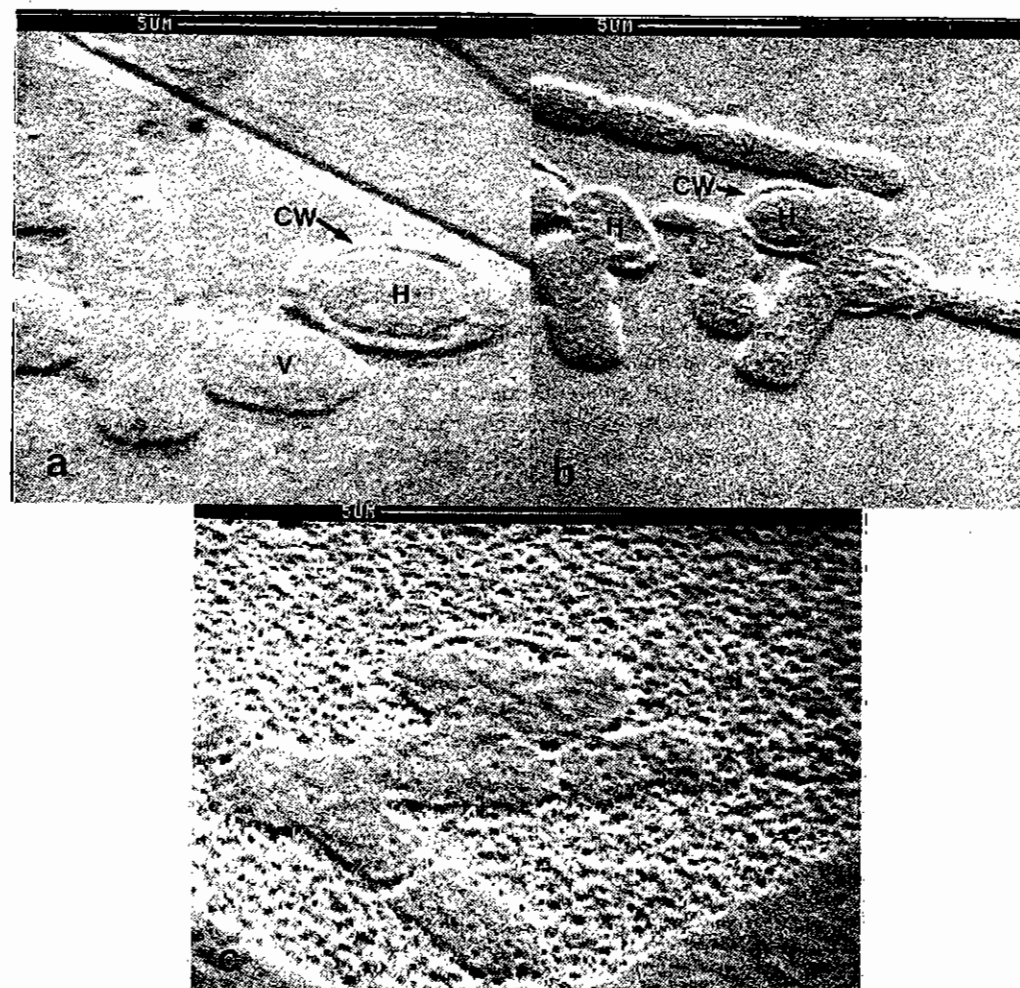
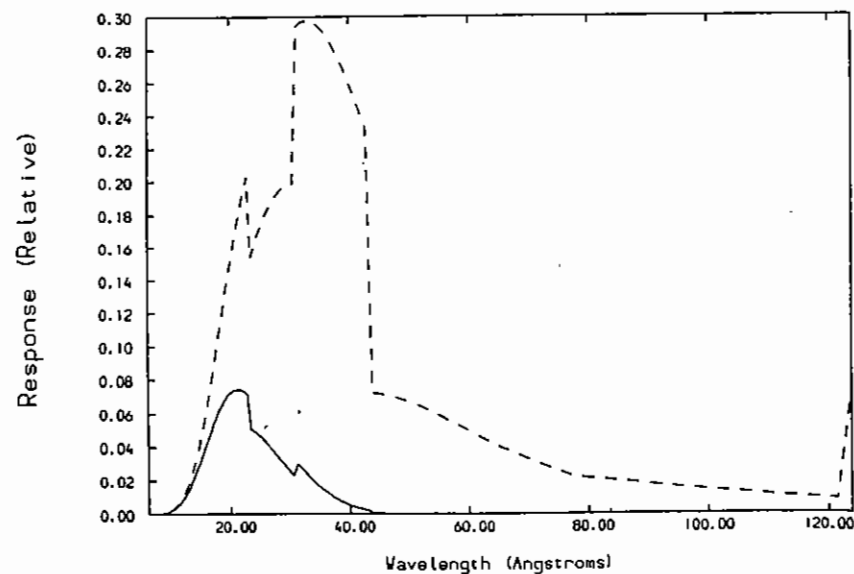


Fig. A1.39 Scanning electron micrograph of resist images of critical point dried filaments of *Anabaena* following exposure at 57 J (I.R.). Silica nitride windows received no aluminium coating (a), 100 nm aluminium (b) or 625 nm aluminium (c).
H - Heterocyst V - Vegetative cell CW - Cell Wall

is very sensitive to u/v irradiation. The problem of u/v irradiation of the sample and resist is partially overcome, in theory at least, by the use of the Si_3N_4 windows as these have been previously claimed to absorb much of the u/v (Chen et al., 1986; 1987). More recent data, however, shows that this may not be the case; therefore aluminium-coated windows were used in this study to eliminate any possibility of imaging due to u/v radiation whilst allowing a significant flux of soft x-rays to pass through and expose the specimen and resist (see Figure A1.38). Furthermore the windows rarely survive the imaging process, and therefore it has also been impossible to ascertain if samples remained hydrated at the time of imaging as evaporation of water does occur through the delicate Si_3N_4 window. The survival of the window during imaging and the discovery of wet specimens would demonstrate conclusively that the specimens do remain hydrated at the time of imaging.

Materials and Methods

In a series of experiments using TA2, biological specimens were imaged using the previously described method (Stead et al., 1987), however, the Si_3N_4 windows were coated with aluminium prior to their use. The windows were placed on a cleaned glass microscope slide and placed in a vacuum evaporating chamber (Edwards E306) with their smooth side (the side destined to be next to the specimen) uppermost. Small, preweighed quantities of aluminium wire were either hung on the tungsten filament, or when larger quantities were used, placed in a tungsten wire basket, the filament was heated, and the aluminium allowed to evaporate omnidirectionally. After each coating, one slide and associated window were removed and the depth of aluminium determined by measuring the thickness of aluminium deposited onto the slide using a Michelson interferometer (Leitz Instruments). It was therefore possible to determine the depth of aluminium to within approximately 25 nm. In all, Si_3N_4 windows were produced with a coating of either 625, 125, 25, or less than 10 nm aluminium. Each thickness of aluminium was then used at a variety of different laser energies and with a number of different biological specimens. The material was chosen for its convenience of handling and included fresh and fixed unicellular and filamentous algae, fungal hyphae and spores and epidermal floral hairs, all of which had been imaged in previous experiments so that the use of coated windows could be fully evaluated. The developed resists were examined by interference light microscopy (Leitz Dialux), and after sputter coating with Au:Pd, by scanning electron microscopy (Cambridge

S100). In addition to comparing the quality of the contact images, the survival or destruction of the Si_3N_4 windows was also carefully monitored after each shot.

Results and Discussion

In previous experiments, few Si_3N_4 windows survived the laser shot; indeed concern has previously been expressed about whether the windows were able to maintain the pressure difference across them during the pump down of the chamber. The survival of the windows during pump down and up until the laser shot is fired, is of course critical if the specimens are to remain hydrated at the time of imaging. It has also been suggested that the windows offer some protection from the longer u/v wavelengths and the ablated debris from the target (Cheng et al., 1986; 1987). The only indications that most do survive intact has come from tests in which the mounted specimen has been placed in the chamber and then pumped down as usual, followed by bringing the chamber up to atmospheric pressure without firing the laser. In such tests, most windows remained intact, but it was found that some did break, especially if the return to atmospheric pressure was too rapid. The use of windows with a superimposed grid pattern which is often found to be imaged, also suggests that the windows survive up until the imaging, but this still does not indicate whether the specimen remained hydrated at the time of imaging. Only very rarely have windows been found to survive the imaging process such that the specimen remained wet when the specimen chamber was disassembled, and in these cases it has usually proved impossible to produce an image of the specimen after development of the resist, suggesting that the alignment of the specimen holder, or that of the target, was not performed correctly. The fact that most of the windows break during the imaging process, but not if unexposed, presumably means that the debris ablated from the target is responsible for the breakage. Once broken, the specimen will dehydrate rapidly, and the image formed might be due to exposure to the longer-lived u/v irradiation, although since the u/v will only persist for approximately 40 nsec, this is likely to be less than the time taken for the window to disintegrate.

In this study many of the aluminium-coated windows survived the imaging process and therefore those specimens that were loaded in a hydrated state were found still to be wet when the specimen holders were disassembled. This proves conclusively that the images obtained on these resists must be that of the hydrated specimen. The survival of

the windows was found, not surprisingly, to be related to the thickness of aluminium deposited onto the window, thus those without aluminium failed to survive any of the laser shots irrespective of the laser energy used. Those with only very thin coatings survived only the lowest laser energies used and those with thicker coatings survived greater laser energies. This is particularly significant since the manufacture of these windows is difficult at present and therefore the ability to reuse the windows is of great importance. Furthermore, of course, it will allow users to ascertain if their specimen was hydrated at the time of imaging and to be sure that the image produced is an x-ray, not u/v image, whilst if used in conjunction with a toroidal mirror hard x-rays could be eliminated also.

The quality of the images obtained was, as expected, dependant upon the duration of development as well as type of developer and its strength. With increased thickness of aluminium, longer or more concentrated development was required to obtain a satisfactory image, that is to say an image with sufficient depth discrimination between the imaged area and the surrounding background. Similarly, the lower the laser energy, the greater the development needed, indeed, with the lowest laser energies used coupled with the thickest aluminium window coatings, it was not always possible to produce satisfactorily developed resists.

All of the developed resists were examined by light microscopy, using incident interference techniques, and scanning electron microscopy after sputter coating the resist with Au:Pd. Using critical point dried *Anabaena* cells and following minimal resist development and exposure, the images, when observed by light microscopy, showed greatest detail in those that were imaged behind Si_3N_4 windows which had no, or a very thin, aluminium coating, with less detail from those with thicker coatings. However, increasing either the strength or duration of development, enabled better images to be obtained from those samples which had been mounted behind windows coated with greater amounts of aluminium, in fact it was possible to obtain images of the cells even when the thickness of aluminium was 625 nm and this is well in excess of that required to eliminate u/v irradiation problems.

Under the scanning electron microscope, internal cell structure was visible in the heterocysts and in some cases in the vegetative cells too (Figure A1.39), this detail appeared similar to that seen in soft x-ray contact images of *Anabaena* cells as seen in resists exposed to synchrotron radiation (Stead et al., 1988). However, resists from

low laser energy shots required greater development and, especially when coupled with the thicker aluminium coatings, the resists had a badly pitted appearance (Figure A1.39(c)).

The use of aluminium coated windows has undoubtedly eliminated any possibility of the exposure of the resists to u/v irradiation, and the survival of these windows further eliminates any possibility that the images were of dehydrated cells. Therefore, these images are without doubt the first images produced as the result of exposure to laser plasma x-rays without any interference from the inevitable u/v radiation which is produced from these plasmas. By comparison, many synchrotron radiation sources also eliminate u/v radiation but as yet synchrotron-produced images require exposures that are too long to image living, hydrated biological specimens.

In conclusion the quality of these images is considerably better than that produced previously using TA2 (Stead et al., 1987), and unlike images obtained in earlier experiments, it has been possible to develop the resists to a depth which is great enough to permit examination by scanning electron microscopy, thus allowing greater detail to be seen in the resist images. The most significant fact is, however, that the technique can be used to image wet material without any interference from u/v radiation.

References

- P C Cheng, R Feder, R W Eason, A G Michette and R J Rosser, RAL Report A4.2.4, (1986).
- P C Cheng, D M Shinozaki and K H Tan, In: X-ray microscopy: instrumentation and biological applications. Eds. P C Cheng and G J Jan, Springer - Verlag.
- R W Eason, RAL Report A4.2.1, (1986).
- R W Eason, P C Cheng, R Feder, A G Michette, R J Rosser, F O'Neill, Y Owadano, P T Rumsby, M J Shaw and I C E Turco, Optica Acta, 33, 501-516, (1986).
- A G Michette, P C Cheng, R W Eason, R Feder, F O'Neill, Y Owadano, R J Rosser, P T Rumsby and M J Shaw, J. Physics, D., 19, 363-372, (1986).

T Mochizuki, T Yabe, K Okada, M Hamada, N Ikeda, S Kiyokawa and C Yamanaka, Phys. Rev. A., 33, 525-539, (1986).

R J Rosser, K G Baldwin, R Feder, D Bassett, A Cole and R W Eason, RAL Report A4.2.1, (1985).

A D Stead, T W Ford, R J Rosser, A Ridgely and C P H Hills, RAL Report A1.9, (1987).

A D Stead, T W Ford, W J Myring and D T Clarke, J. Microsc., in press, (1988).

A1-16 SOFT X-RAY CONTACT MICROSCOPY USING TA4

A D Stead, T W Ford (Dept. Biology, RHBNC), C P Hills (Dept. Physics, Kings College), N Rizvi (RAL)

If soft x-ray contact microscopy, using VULCAN, is to become a useful and routine technique available to a larger number of biologists, it is clear that there is a need for a facility that could be operated with a minimum of disturbance to other users of the same laser. This facility would also need to be made available in a manner hitherto not envisaged in the scheduling procedure of the other target areas. Such a facility is TA4; this area has been developed with the biologist first in mind and the area will remain available at relatively short notice for biological applications; there is the capability of having many laser shots in one day, and the target chamber will have an airlock through which the specimen holder can be inserted so that the time between specimen mounting and the laser shot are reduced to an absolute minimum. In addition the chamber will have a full range of diagnostics so that both the flux and spectrum of the x-rays produced are recorded. During initial tests, in December, 1987, a borrowed target chamber was in place and using this a number of high resolution contact images were obtained; these images were of a better quality than those obtained during previous experiments conducted in TA2 and have further demonstrated the feasibility of imaging biological specimens with only 2 - 3J of "green-light" laser energy.

Material and methods

The layout of the target chamber is described elsewhere in this report (A6.5). The specimen holders were redesigned from those used

previously so that the specimen could, if necessary, be placed within 10 mm of the target whilst still retaining the ease of mounting of hydrated biological specimens which was so much a feature of a previous design. In addition to the new holders, the original holders, described by Eason et al., (1986) were also used as they permitted placing the specimens that were not hydrated even closer to the target.

In order to make comparisons of the performance of this new set up, the material imaged was similar to that used previously in experiments in TA2 (Stead et al., 1986; 1988) and in those using the Daresbury Synchrotron (Stead et al., 1988), in particular fragments of a diatomaceous earth and of critical point dried algae (*Trebouxia* and *Anabaena*). The specimens were placed behind Si_3N_4 windows when appropriate. The resist material was either PMMA (250 nm thick) or PMMA-MAA (1000 nm thick) and development was in mixtures of MIBK and IPA. The laser wavelength was frequency doubled (0.526 μm) as in previous studies and the target material was gold foil (25 μm thick).

Results and discussion

Initial exposures of the diatomaceous earth were made at a distance of only 4 mm away from the target as it was uncertain if the laser energy available would be sufficient to produce an adequately exposed resist. In fact these exposures required very little development prior to examination by either light, or scanning electron microscopy. Under the light microscope details of the structure of some of the frustule fragments can be seen, whilst little detail is apparent in the remainder (Figure A1.40). Further detail can be revealed by extended development (Figure A1.40), however some of the features seen initially may be lost as the image itself is developed away (Cheng et al., 1986). After sputter coating and examination by scanning electron microscopy much greater detail can be seen. Some of the frustules appear to be almost complete and in the centric diatom (Figure A1.41(a)) the small circular fenestrations can be easily identified and measured, in this particular case they appear to be 0.2 μm in diameter and this accords well with the actual size as seen when similar specimens are examined directly by scanning electron microscopy (Figure A1.41(b)). In some other images both complete frustules and fragments of frustules can be seen (Figure A1.42), and again these are excellent images for

demonstrating the resolution of the technique. The complete frustule is from *Navicula* but a limited amount of detail is visible and this image would probably benefit from further development. However, the fragment alongside requires little further development and measurements of the graticule-like structure along the centre shows that there are 16 or 17 dark and light bands (the former are images of the siliceous structure, the latter the spaces between), within a distance of approximately 1 μm . This image has a resolved structure of less than 70 nm, is one of the best images produced at RAL, and indeed probably better than those produced elsewhere as well.

In other specimens the detail was comparable to that seen previously, thus the images of critical point dried *Trebouxia* (Figure A1.43) show the cell wall distinct from the cytoplasmic contents, but little detail can be distinguished within the cytoplasmic contents. This is similar to that reported previously (Stead et al., 1988) when synchrotron radiation was used. Preliminary attempts at imaging hydrated cells of *Anabaena* and *Trebouxia* using the new specimen holders and laser energies of 6 - 7 J (I.R.), have so far met with limited success. Although images of both organisms have been produced, the small development depth has precluded the use of SEM and therefore little internal detail could be resolved. However, this is an area which will be actively pursued in future experiments.

The images produced show conclusively that laser energies less than those used previously are sufficient to give adequate exposure of those resist materials currently available when the specimen to target distance is reduced. In the future, as more sensitive resists are developed, this distance may be increased thus facilitating the imaging of more than one specimen at a time. Experiments reported elsewhere in this report (Stead et al., 1988) have demonstrated that whilst u/v radiation is produced when using laser-produced plasmas this need not be of concern since the use of aluminium-coated Si_3N_4 can eliminate the u/v radiation. We have already demonstrated the feasibility of using a toroidal mirror (Hills et al., 1987), which will eliminate the harder x-rays, thus routine imaging of hydrated biological specimens using only soft x-rays in the "water window" will be possible at RAL, using the new TA4 target area.

References

1. P C Cheng, R Feder, D M Shinozaki, K H Tan, R W Eason, A G Michette and R J Rosser, Nucl. Instrum. Methods Phys. Res. A, 246, 668-674, (1986).
2. R W Eason, RAL Report, A4.2.1, (1986).
3. C P Hills, T W Ford, A D Stead and R J Rosser, 31st Annual SPIE Symposium, San Diego, U.S.A., (1987).
4. A D Stead, T W Ford, R J Rosser, A Ridgeley and C P H Hills, RAL Report A1.9., (1987).
5. A D Stead, T W Ford, W J Myring and D T Clarke, J. of Microscop. In press, (1988).
6. A D Stead, T W Ford, C P H Hills, J A Catcheside and A Ridgeley, This report, (1988).

A2	LASER PLASMA INTERACTION AND ENERGY TRANSPORT	Pages
A2-1	Beat-wave Experiments	45 - 49
A2-2	Measurements of the Heat Flux, Inverse Bremsstrahlung Absorption and Equilibrium in an Under-dense Laser Heated Plasma	49 - 58
A2-3	Short Pulse Energy Transport by Thermal and Superthermal Electrons	58 - 60
A2-4	Picosecond High Brightness KrF Laser Plasma Interactions	60 - 61
A2-5	Observations of Instabilities in Long Scalelength Preformed Plasmas	61 - 65
A2-6	Radiation Preheat and Radiation Transport Observations in Thin Foil Targets	65
A2-7	Energy Transport Uniformity on Spherical Targets - Further Analysis and Results	65 - 74
A2-8	Thomson Scattering in Long Scalength Preformed Plasmas	74 - 76

Editors: G Tallents/I C E Turcu

A2-1 BEAT-WAVE EXPERIMENTS

A Dyson, A E Dangor, A K L Dymoke-Bradshaw
College)

F Amiranoff (Ecole Polytechnique)

T Garvey, B Montague (CERN)

T Afshar-Rad, A J Cole, C N Danson, C B Edwards, R G Evans (RAL)

1. Introduction

The need to probe smaller and smaller distances in matter has led to the requirement for huge increases in particle accelerator energy. To accelerate a charged particle to relativistic velocities requires a longitudinal electric field and a phase velocity close to c . A plasma being fully ionised does not suffer from breakdown which limits the maximum fields achievable in a conventional accelerator. It has the major advantage that the maximum longitudinal electric field of a relativistic plasma wave is $n_e^{1/2} (\text{cm}^{-3}) \text{ Vcm}^{-1}$, so that for number densities $n_e \sim 10^{20} \text{ cm}^{-3}$ the maximum field is $\sim 10^{10} \text{ Vcm}^{-1}$ orders of magnitude greater than a conventional structure. The major disadvantage is that plasmas are inherently unstable. The beat-wave mechanism⁽¹⁾ is an interesting method of producing a relativistic plasma wave and requires that two parallel laser beams of slightly different frequencies are focused onto a plasma of resonant frequency $\omega_p = \omega_1 - \omega_0$. Since this is a resonant process the plasma density is required to be reasonably uniform, perhaps for as much as 30 m for an accelerator. Using a neodymium glass laser ($\lambda \sim 1 \mu\text{m}$) requires the plasma number density to be within about 0.5% of the resonance value, compared to the much less stringent condition of a few percent for the CO_2 lasers ($\lambda \sim 10 \mu\text{m}$) used by others in this field⁽²⁾⁽³⁾. However, the phase velocity v_{ph} of the plasma wave, characterised by the Lorentz factor $\gamma = (1 - (v_{ph}/c)^2)^{-1/2} = \omega_0/\omega_p$, is much larger ($\gamma = 100$) than for a CO_2 laser ($\gamma = 10$) and so is more relevant to particle acceleration. Preliminary experiments in this field have used DC arc discharges, Z or theta pinches, or air breakdown plasmas. These are recognised as having inadequate homogeneity and reproducibility for accelerator applications. Multiphoton ionisation of hydrogen gas produces exceptionally uniform plasmas with appropriate density and temperature and the technique is scaleable to plasmas of arbitrary size⁽⁴⁾⁽⁵⁾.

2. Experimental Arrangement

The experimental arrangement is shown in Figure A2.1. The pump beams each delivering $\sim 50\text{J}$ and of 200 ps f.w.h.m. were focused by a 2 m lens, f/20 optics, to give a 400 μm diameter spot size. The plasma was preformed by a frequency doubled YLF beam delivering $\sim 10\text{J}$ and of 200 ps f.w.h.m., focused using the same 2 m lens as for the I.R. beams and a negative correcting lens to give a focal spot at the same location as the I.R. beams of size 400 μm . The target chamber was filled with hydrogen gas at pressures from 1-2 torr. The two pump beams were individually amplified, followed separate air paths and were orthogonally polarised, finally being mixed together under vacuum and their polarisations matched. A 0.1% transmitting I.R. mirror was placed at the output of the target chamber, the emerging light further attenuated by wedged uncoated mirrors before being focused onto a Bentham spectrometer and then an S1 streak camera. These precautions were taken in order to avoid Raman scattering by atmospheric nitrogen⁽⁶⁾. Thomson scattered light was collected from the preionising green beam and from a separate green probe beam whose relative timing could be varied. The scattered light was collected at four scattering angles for both the preionising beam (40, 58, 122, 140 degrees) and the probe beam (18, 64, 116, 162 degrees). Using this arrangement the plasma conditions could be determined both before and after the pump beams had passed through the plasma. A $\lambda = 0.622 \mu\text{m}$ probe was set up to interact with the plasma wave by Raman-Nath scattering. However, time restrictions meant that the probe was not used as a diagnostic.

3. Experimental Results

The planned use of the cascade channel to detect the plasma wave was impossible due to the CARS effect in the optical components where the pump beams were together. This resulted in sidebands several times larger than those expected due to a plasma wave. Figure A2.2 shows a typical spectrum. Four simultaneous Thomson scattered signals were obtained for both the preionising green beam and the green probe beam, and by varying the timing of the probe beam it was possible to study the time evolution of the plasma. Typical spectra obtained are shown in Figure A2.3. By comparing the scattered light profiles of the 140 degree (preioniser) and 162 degree (probe) channels the heating of the plasma by the pump beams can be determined. Typical profiles are given in Figure A2.4. and show a temperature increase from $\sim 12 \text{ eV}$ to

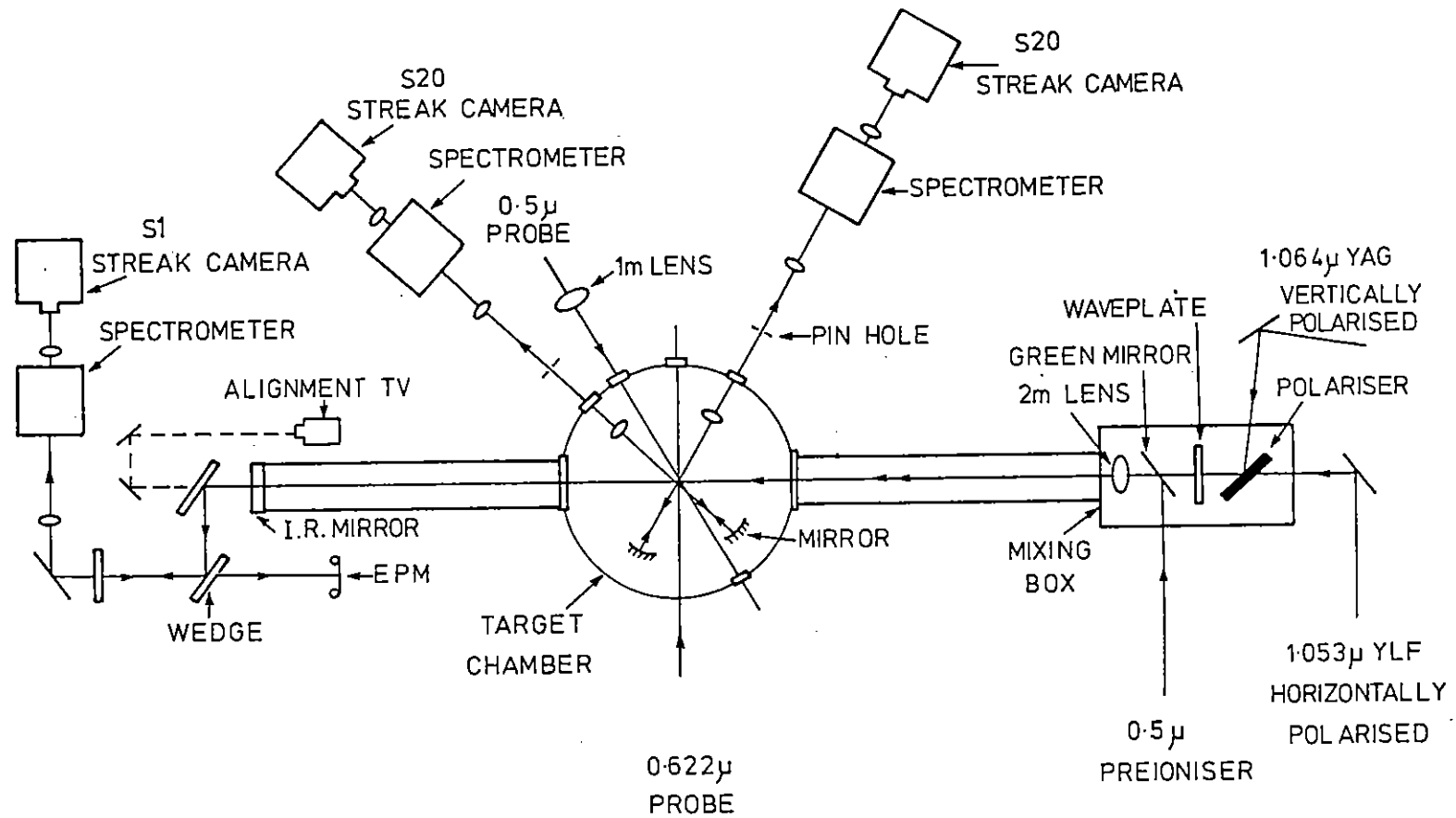


Fig. A2.1 Schematic of experimental arrangement for the beat-wave experiments.

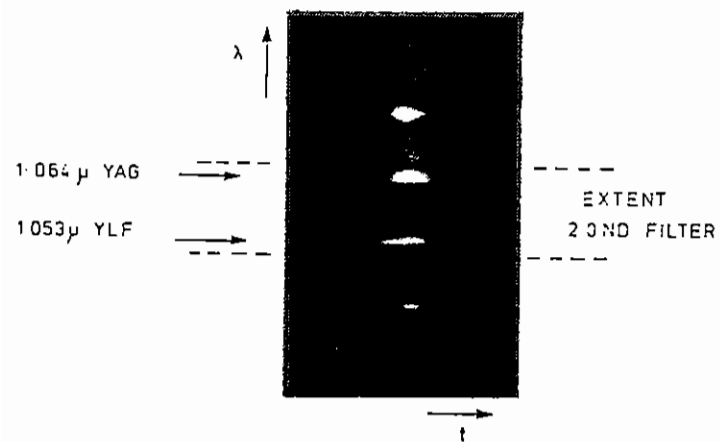


Fig. A2.2 Spectrum of pump beams due to CARS effect.

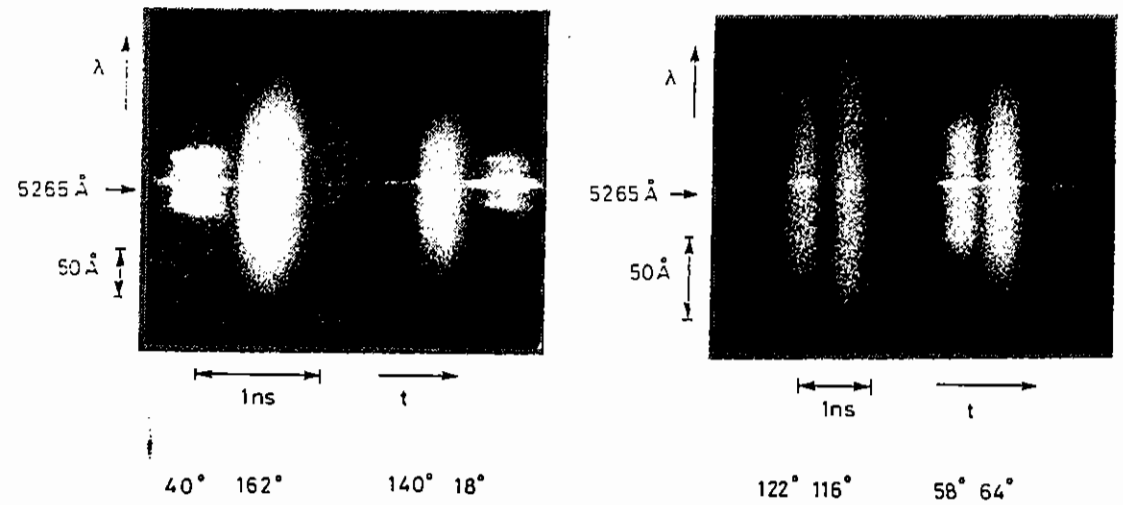


Fig. A2.3 Typical Thomson scattering spectra.

~ 25 eV. In this example the probe was delayed ~ 400 ps with respect to the pump beams. The theoretical profile, which assumes a Maxwellian distribution, does not fit the scattered signal from the heated plasma as well as the fit obtained with the signal from the performed plasma. This suggests that the heated plasma is non-Maxwellian or turbulent. The measured increases in temperature due to the pump beams show only a slight dependence on the performed plasma density, there being no indication of large increases corresponding to a particular resonant density which would be expected if a large amplitude plasma wave were driven and then decayed into thermal energy. The scattered light profiles of the 40 degree (preioniser) and 18 degree (probe) channels are used to give the plasma density to an accuracy of $\pm 10\%$.

4. Discussion

We have modelled the plasma heating with a single one dimensional (cylindrically symmetric) computer code, modelling inverse bremsstrahlung absorption, thermal conduction, ion-electron equilibration and hydrodynamic expansion. The classical absorption cross-section is modified to allow for the reduction in collision frequency⁽⁷⁾⁽⁸⁾ when the electron oscillation velocity in the light wave $v_{osc} = eE/m\omega$ exceeds the thermal speed $v_e = (k_B T_e/m)^{1/2}$. The electron thermal conductivity is limited by the electron free streaming limit. The flux limit is taken as $f = 0.1$ in agreement with calculations based on the Fokker-Plank equation⁽⁹⁾. Good agreement is found with the measured temperature increases due to heating by the pump beams. This and the fact that there is no indication of large temperature increases at any particular density suggest that no significant plasma wave was present.

5. Conclusions

Multiphoton ionisation of hydrogen gas of pressures from 1 to 2 torr was used to produce uniform plasmas, the density corresponding to 100% ionisation. However, the temperature of the hydrogen gas in the focal region was not measured very accurately. Small fluctuations in temperature of a few degrees are sufficient to move the density off resonance. Unfortunately, scattering was not performed at small enough angles and so the density could not be measured to the required accuracy of about 1%. Detection of the plasma wave by monitoring the pump light was made impossible due to the CARS effect in the mixing

optics. No additional heating due to the decay of the plasma wave was observed. This is perhaps due to the temperature being measured too late or that a resonant density was not obtained.

6. References

1. T Tajima, J M Dawson, Phys. Rev. Lett., **43**, 267 (1979).
2. C E Clayton, C Joshi, C Darrow, D Umstadter, Phys. Rev. Lett., **54**, 21, 2343 (1985).
3. N A Ebrahim, F Martin, P Brodeur, E A Heighway, J P Matte, H Pepin, P Lavigne, Proceedings 1986 Linear Accelerator Conference Stanford Ca.
4. Dangor et al., IEEE Trans. on Plasma Sci., **2**, 161 (1987).
5. Dangor et al., RAL Annual Report RAL-87-041, A2.1 (1987).
6. Dangor et al., RAL Annual Report RAL-86-046, A1.1 (1986).
7. G J Pert, J. Phys. A., **5**, 506 (1972).
8. S Rand, Phys. Rev. B., **136**, 231 (1964).
9. A R Bell, R G Evans, D J Nicholas, Phys. Rev. Lett., **46**, 243 (1981).

A2-2 MEASUREMENTS OF THE HEAT FLUX, INVERSE BREMSSTRAHLUNG ABSORPTION AND EQUILIBRATION IN AN UNDER-DENSE LASER HEATED PLASMA

A Dyson, A E Dangor, A K L Dymoke-Bradshaw (Imperial College)
R G Evans (RAL)

1. Introduction

It is well known that theoretical treatments of heat flow based on a

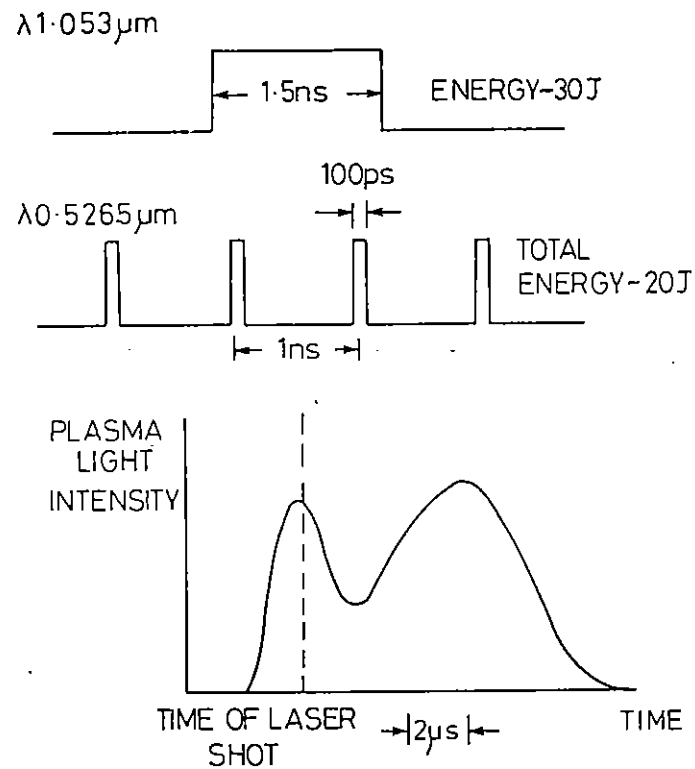


Fig. A2.6 Relative timing of the Z- pinch, the heating beam and the four probe pulses.

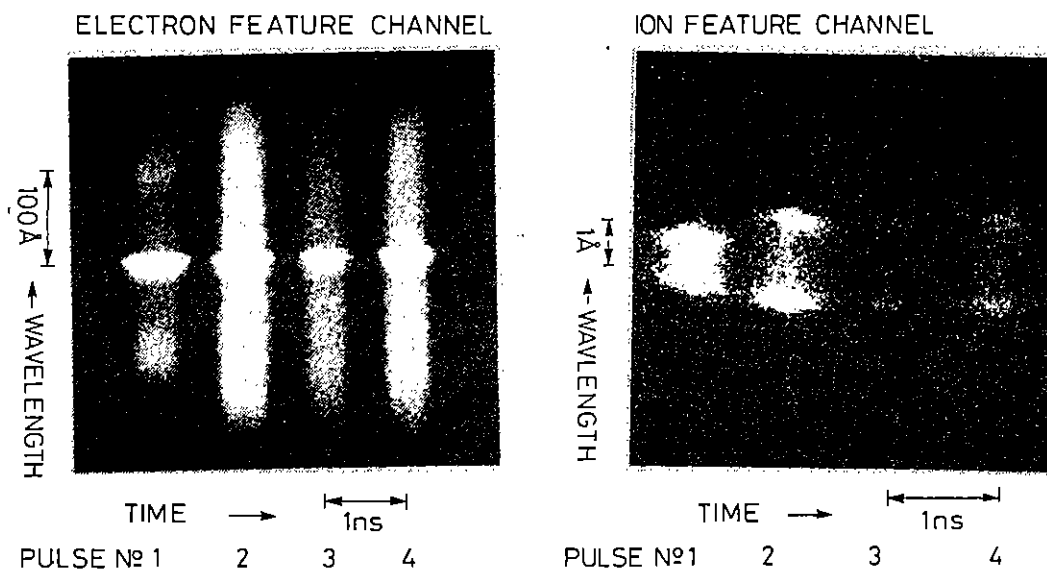


Fig. A2.7 Thomson scattering data from a typical shot.

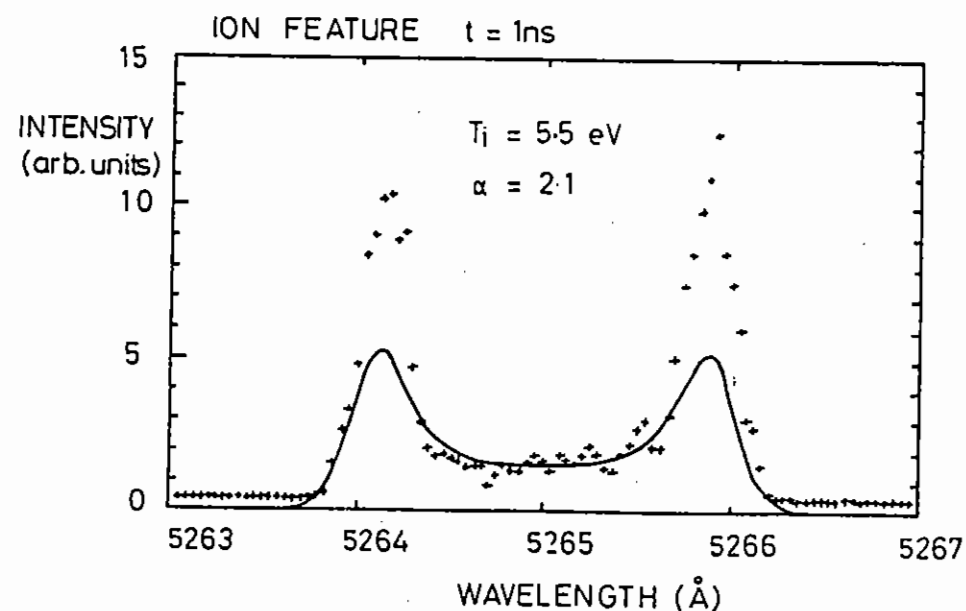
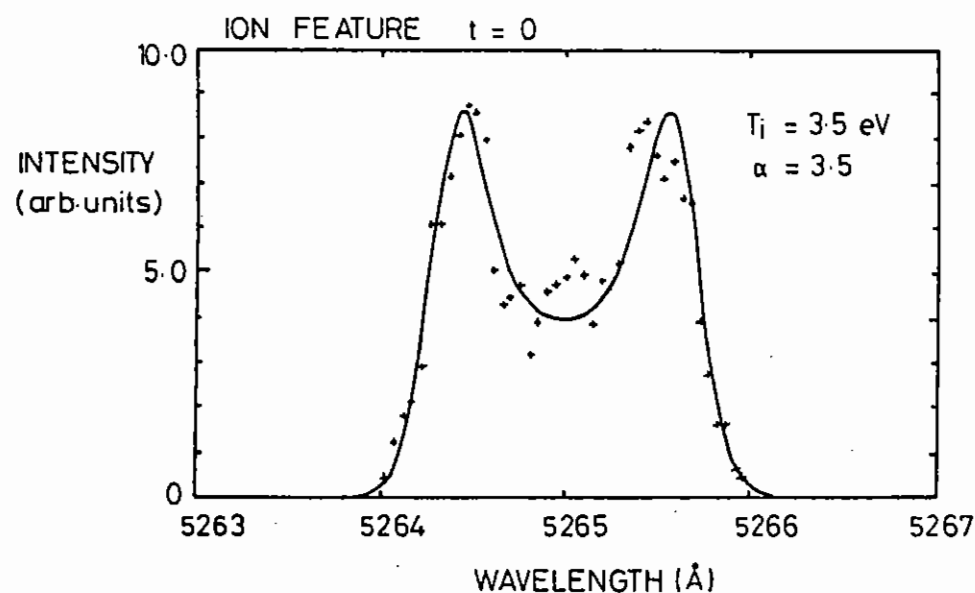
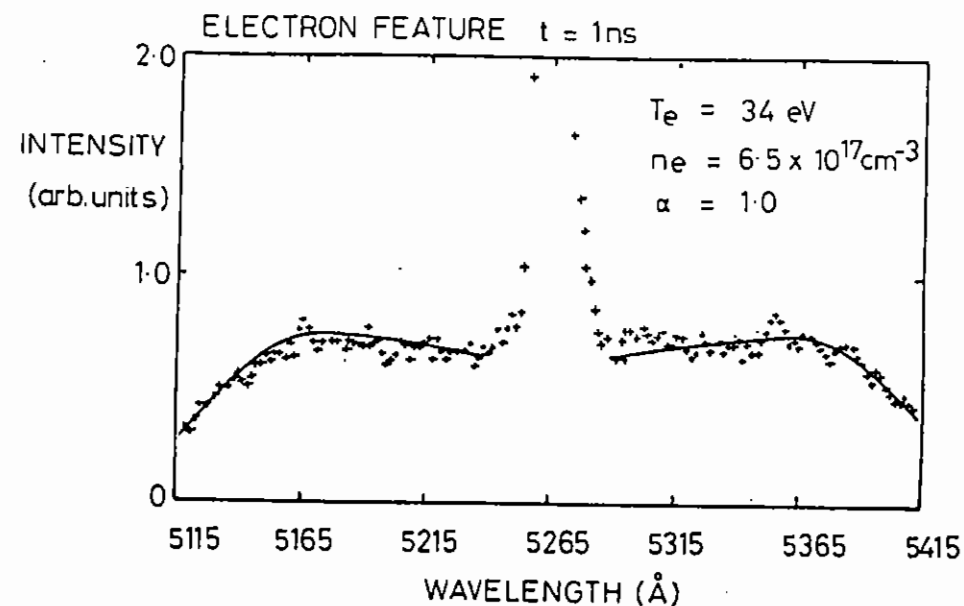
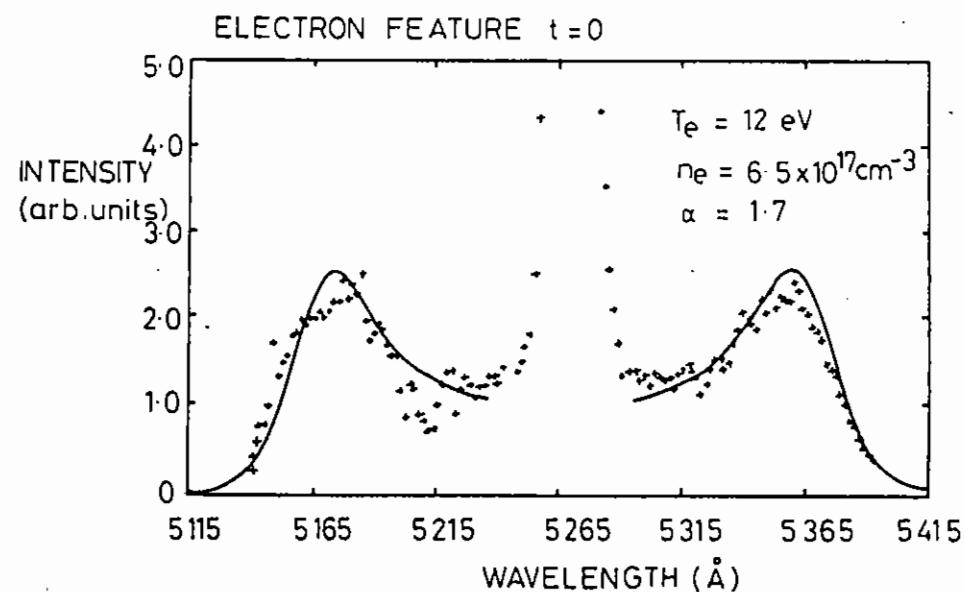


Fig. A2.8 Typical scattering profiles with the computer generated best fit. The ion feature spectrum for the second probe pulse at $t = 1 \text{ ns}$ indicates turbulence.

first order perturbation theory⁽¹⁾ breakdown for large value of λ/L , where λ is the electron mean free path and L is the temperature scale length defined by $\nabla T_e = T_e/L$. Calculations based on the Fokker-Planck equation⁽²⁾ predict that there is a limit to the maximum heat flux of about 0.1 of the free streaming limit. A maximum heat flux of 0.03 of the free streaming limit was inferred in early experimental work on laser produced plasmas based on measurements of the ion emission, the x-ray emission, or the mass ablation rate^(3,4). However, recent work⁽⁵⁾ using computer simulations which include thermal transport by fast electrons indicate that the maximum heat flux is about 0.1 of the free streaming limit.

A more direct measurement of the heat flow was reported by⁽⁶⁾ using a CO_2 laser to heat an under-dense hydrogen plasma. The maximum value for λ/L was 0.3 and the maximum heat flux was found to be limited to 0.03 of the free streaming limit, this low value being attributed to a large level of ion acoustic turbulence. In a similar experiment by⁽⁷⁾ a CO_2 laser beam of much longer pulse length was used. The maximum heat flux was found to be 0.1 of the free streaming limit at λ/L of 0.07. No turbulence was observed. The definitions for the free streaming limit and the electron mean free path used in this paper follow⁽²⁾, $Q_{fs} = nk_B T_e (k_B T_e / m_e)^{1/2}$ and λ/L such that the Spitzer heat flow for $Z = 1$ is $Q_{sp} = 3.2(\lambda/L)Q_{fs}$.

We report further measurements of the heat flux, using a neodymium laser to heat a small volume of hydrogen plasma. The laser is focused to give a much larger irradiance of $5 \times 10^{13} \text{ W cm}^{-2}$ with a pulse length of 1.5 ns, shorter than in the experiments mentioned above. In our experiment Thomson scattered spectra are obtained at two angles to give simultaneous measurements of the electron and ion temperatures and the level of ion acoustic turbulence. The spectra are obtained at four discrete times allowing the plasma evolution to be monitored in a single shot. Comparison with a 1D hydrodynamic code enables the heat flux, inverse bremsstrahlung absorption and ion-electron equilibration to be determined.

In Section 2 of this paper the experimental details are presented. The results are given in Section 3 and the numerical model is described in Section 4. Comparison between the experiment and the model is discussed in Sections 5 and 6.

2. Experimental Arrangement

The experiment was conducted at the SERC Central Laser Facility using the Vulcan neodymium glass laser. The experimental arrangement is shown in Figure A2.5. A slow Z-pinch in hydrogen was used to produce a fully ionised plasma of 30 cm length and about 3 cm diameter. A small volume of the plasma was heated with a $1.053 \mu\text{m}$ laser beam of 35 J, 1.5 ns fwhm. The beam was focused transversely into the pinch by a 1 m lens (f/10) to a spot of about $200 \mu\text{m}$ diameter, giving a peak irradiance of $5 \times 10^{13} \text{ W cm}^{-2}$. The plasma conditions were determined by Thomson scattering of a $5265 \text{ \AA}$ copropagating probe. The probe consisted of four pulses of 100 ps fwhm separated by 1 ns and focused by a 1 m lens (f/10) with the total energy in the pulses being about 20 J. The relative timing of the Z-pinch plasma, the heating beam and the green probe beams is shown in Figure A2.6.

The scattered light from each pulse was monitored in two channels each consisting of a spectrometer and streak camera with the output recorded on HP5 film. The spectrometer slits in both channels were aligned to be in the same plane as the probe beam to maximise the amount of scattered light collected. The scattering volume was a cylinder = $100 \mu\text{m}$ in diameter and = 1 mm long, coaxial with the probe beam. The temporal resolution was determined by the probe pulse duration of about 100 ps. The channel collecting light at 100 degrees had a resolution of $5 \text{ \AA}$ which was sufficient to resolve the electron feature but not the central ion feature. The spectrally resolved electron feature was used to determine the electron density and temperature. The ion feature was resolved in the second channel which collected light scattered at 22, 44 or 66 degrees with a resolution of $0.1 \text{ \AA}$. The resolved ion feature was used to determine the ion temperature and the level of ion acoustic turbulence. This was possible as the amount of stray light was insignificant. The spatial variation of the plasma temperature and density was obtained by moving the focal spot of the heating beam by distances of 0, 200, 400 and $600 \mu\text{m}$ vertically or horizontally with respect to the fixed scattering volume. This allowed the effect of the heat flux on the velocity distribution to be investigated. For most shots the fill pressure of the Z-pinch was set to give an initial number density of $2.5 \times 10^{17} \text{ cm}^{-3}$ or $5 \times 10^{17} \text{ cm}^{-3}$ and a temperature of 8 eV or 12 eV respectively. Very little variation of the initial density and temperature was observed, indicating the high reproducibility of the pinch. The magnetic field of the pinch was too

small to effect the thermal conduction ($\omega_{ce}\tau_e \ll 1$). In the absence of the heating beam the plasma conditions were found to be the same for all the probe pulses indicating that there is little heating by the probe.

3. Experimental Results

The photographic data of the scattered signals for a typical shot is shown in Figure A2.7. The data is digitised using a microdensitometer and then corrected for film response and the spectrometer and streak camera spectral sensitivity. The spectra are then fitted to theoretical Thomson scattering profiles for a thermal plasma⁽⁸⁾ corrected for instrument response. The electron feature spectra are such that both the electron density and temperature are determined using these quantities as the fitting parameters. The ion temperature is the fitting parameter for the ion feature spectra, using the electron density and the temperature obtained from the corresponding electron feature. Full details of the data analysis are to be published elsewhere.

Examples of the fitted spectra are shown in Figure A2.8. Extremely good fits are found for the spectra from all four probe pulses, except for the ion feature spectra obtained from the second probe pulse at the focal spot of the heating beam. In these spectra the wings are greater than the expected thermal level indicating the presence of turbulence. This pulse is during the rising edge of the heating beam and here coincides in both time and space with the strongest plasma heating. For these spectra the ion temperature is obtained from the positions of the wings which are shifted by the ion acoustic frequency

$$\omega_{ac} = k \left[\frac{a^2}{1+\alpha^2} \frac{k_B T_e}{m_i} + \frac{3k_B T_i}{m_i} \right]^{1/2} \quad (1)$$

where k is the differential scattering vector and $\alpha = 1/(k\lambda_D)$ is the scattering parameter⁽⁹⁾.

To determine the amount of turbulence the ratio of the integrated ion feature from the second channel to the integrated electron feature is compared with the theoretical ratio assuming a thermal plasma. Good agreement is found except for the spectra obtained from the second probe pulse at the focal spot of the heating beam where the measured

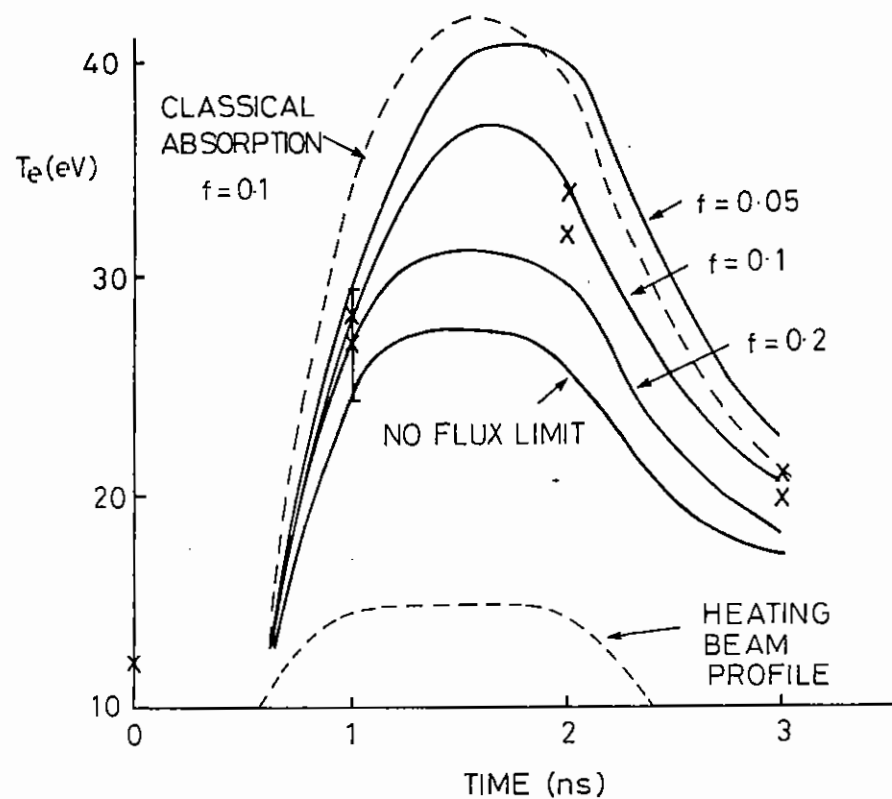
ratio is between 1 and 2 times the thermal value. For the scattering at 44 degrees, where most of the data was collected, the integrated ion feature is 1.5 times the thermal level. The measured value of T_e/T_i is about 5 and at this angle $k\lambda_D$ is about 0.5. The maximum of the energy spectrum of ion acoustic turbulence is close to this value of $k\lambda_D$ ⁽¹⁰⁾. Thus the small enhancement observed indicates that only a low level of turbulence is excited.

The results show that the heating is confined to the focal region of the heating beam with only small temperature increases observed at a distance of 400 μm and none at 600 μm . The measured electron temperatures for an initial density of $5 \times 10^{17} \text{ cm}^{-3}$ at the focal spot of the heating beam and at 200 μm are shown in Figures A2.9 a) and b) respectively. The observed heating increases with density as shown in Figure A2.10. The measured ion temperatures for an initial density of $5 \times 10^{17} \text{ cm}^{-3}$ at the focal spot of the heating beam are shown in Figure A2.12. The measured plasma conditions show only a slight dependence on irradiance of the heating beam. No change in the density is observed indicating that over the duration of the observations there is no expansion of the heated plasma. The scattered spectra do not show any variation with the direction of displacement from the scattering volume confirming that there is no turbulence other than in the focal region. Any skewness in the electron feature due to the heat flow would not be detected. This is because most of the energy is carried by electrons with velocities between 2 and 3 times the thermal velocity and this is outside the range of the measured spectra.

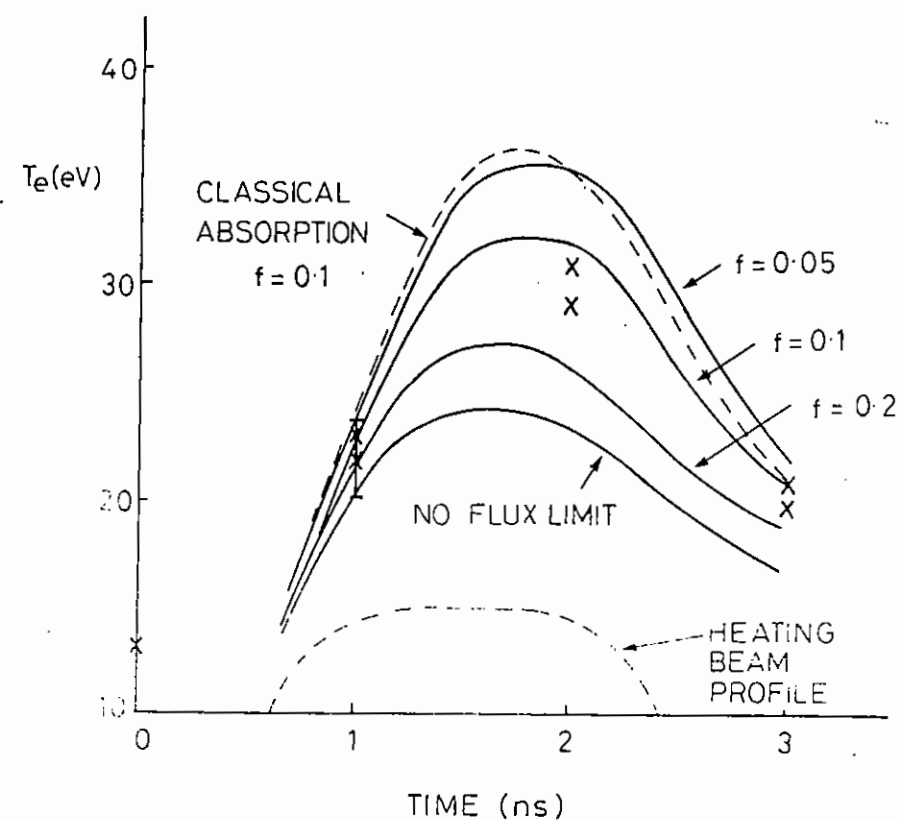
4. Modelling

The plasma heating is modelled with a 1-D two fluid hydrodynamic simulation based on the MEDUSA code⁽¹¹⁾. Cylindrical symmetry is assumed and inverse bremsstrahlung absorption, thermal conduction and ion-electron equilibrium are included. The laser is incident along the axis and has a Gaussian radial intensity profile with width equal to the measured spot size. The model assumes that the plasma is fully ionised with the initial plasma conditions set to the experimentally measured values.

The classical inverse bremsstrahlung absorption cross section is modified by the strong field correction^(12,13) to allow for the reduction in collision frequency when the electron oscillating velocity



(a)



(b)

Fig. A2.9 Measured electron temperatures in the heating beam for an electron density of $5 \times 10^{17} \text{ cm}^{-3}$. The solid lines show the predicted temperatures with strong field corrected absorption for various values of the flux limit f .
 a) at the focal spot.
 b) at 200 μm from the focal spot.

in the light wave, $v_{osc} = eE/m_e\omega$, exceeds the thermal speed, $v_e = (k_B T_e/m_e)^{1/2}$. The effective collision is taken to be

$$v_{eff} = v_{ei} \frac{v_e^3}{[v_{osc}^2 + v_e^2]^{3/2}} \quad (2)$$

Here v_{ei} is the classical electron-ion collision frequency in which the coulomb logarithm $\ln\Lambda = \ln(r_{max}/r_{min})$ has $r_{max} = v_e/\omega$, where ω is the heating beam laser frequency, and r_{min} is the minimum of $e^2/4\pi\epsilon_0 k_B T_e$ and $h/(m_e k_B T_e)^{1/2}$.

The heat flux is restricted to the electron free streaming limit by taking it to be sharply limited by

$$Q = \text{MIN} (Q_{sp}, fQ_{fs}) \quad (3)$$

or harmonically limited by

$$\frac{1}{Q} = \frac{1}{Q_{sp}} + \frac{1}{fQ_{fs}} \quad (4)$$

Here Q_{sp} is the Spitzer heat flow

$$Q_{sp} = 3.2 (\lambda/L) n k_B T_e (k_B T_e/m_e)^{1/2} \quad (5)$$

and Q_{fs} is the free streaming limit, defined by

$$Q_{fs} = n k_B T_e (k_B T_e/m_e)^{1/2} \quad (6)$$

where the flux limit f is an adjustable constant. The ratio of the electron mean free path λ to the temperature scale length $L = T_e/\nabla T_e$ is

$$\lambda/L = 1.44 \times 10^{13} T_e(\text{eV}) |\nabla T_e(\text{eV cm}^{-1})| / n(\text{cm}^{-3}) \ln\Lambda \quad (7)$$

where the coulomb logarithm is given by the usual expression.

The ion-electron equilibrium rate is taken to be a multiple of the classical expression so that the effective rate is

$$v_{eff}^e = s v_{ei}^e \quad (8)$$

where

$$v_{ei}^e = 3.2 \times 10^{-9} n(\text{cm}^{-3}) \ln\Lambda / T_e(\text{eV})^{3/2} \text{ s}^{-1} \quad (9)$$

and the equilibrium multiplier s is an adjustable constant.

5. Comparison with experiment

Figures A2.9 and A2.11 show how the calculated temperatures depend on the flux limit f , classical and strong field corrected absorption and the equilibration multiplier s .

The temperatures predicted at the time of the second probe pulse, about 400 ps after the start of the heating beam, are only slightly dependent on the flux limit f . This can be seen in Figures A2.9 a) and b) where with classical absorption too much heating results but with strong field corrected absorption good agreement is obtained. At the peak of the heating beam the strong field correction reduces the absorption coefficient by more than 50%. This is further illustrated in Figure A2.10 which shows that the predicted temperatures with strong field corrected absorption are in agreement with the measured temperatures for various densities.

The temperatures predicted at the time of the third probe pulse, close to the end of the heating beam, and at the time of the fourth probe pulse, about 1 ns after the heating beam, depend on the value of the flux limit. Figures A2.9 a) and b) indicate that the results are consistent with $f = 0.12 \pm 0.05$ in the case of harmonically limited heat flow (Equation 4). For sharply limited heat flow (Equation 3) it is found that $f = 0.09 \pm 0.02$. The maximum value for λ/L , determined from the spatial profiles of the predicted electron temperature, is about 0.1. Thus if no heat flux is imposed the Spitzer heat flow would give a maximum heat flux of about 0.3 of the free streaming limit (Equation 5). The simulations show that if the classical absorption coefficient is used with the Spitzer heat flow then there is too little heating at the focal spot and too much heating outside.

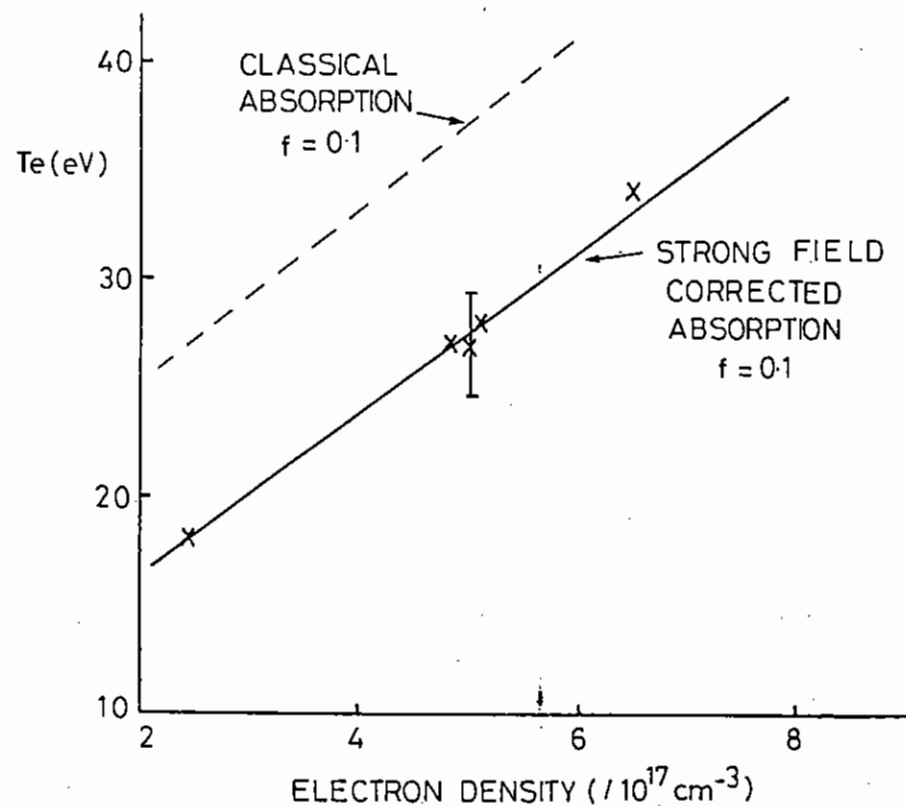


Fig. A2.10 Measured electron temperatures at the focal spot of the heating beam for various electron densities. The solid line shows the predicted temperatures for strong field corrected absorption and the broken line for classical absorption, with the flux limit $f = 0.1$.

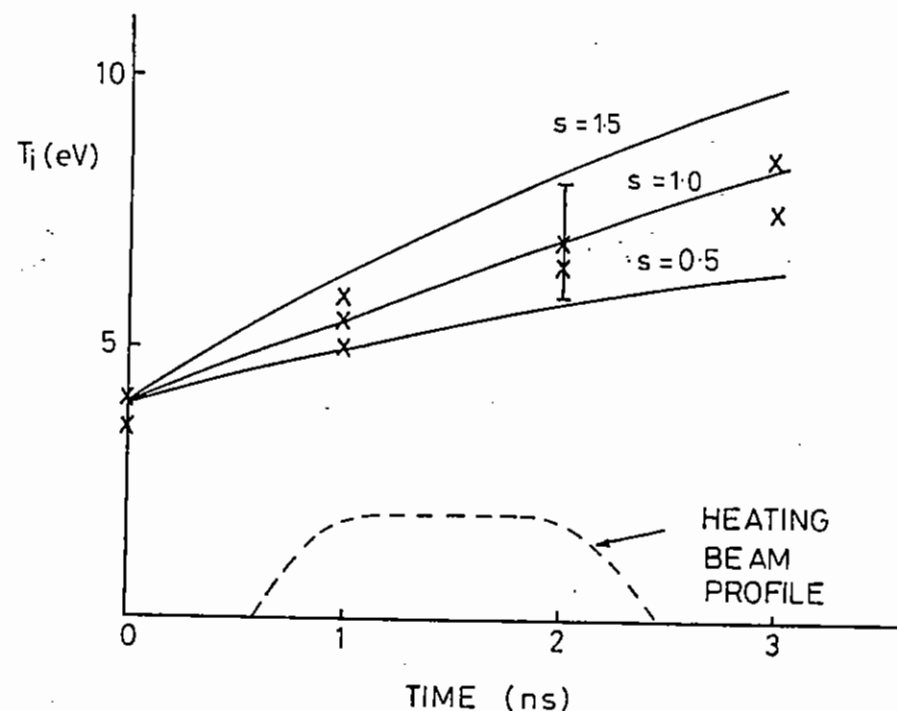


Fig. A2.11 Measured ion temperatures at the focal spot of the heating beam for an electron density of $5 \cdot 10^{17} \text{ cm}^{-3}$. The solid lines show the predicted temperatures for different values of the equilibration multiplier s .

Figure A2.11 shows that the measured ion temperatures are best modelled by the classical electron-ion equilibrium rate to within 50%. The predicted ion temperatures are only slightly dependent on the value chosen for the flux limit. The increase in ion temperature is small, consistent with the equilibrium time being very much longer than the duration of the heating beam.

The predicted decrease in electron density due to hydrodynamic expansion is small in agreement with the observation of no measurable change in density. It is found that changing the laser irradiance by 50% results in changes in the predicted temperatures of about 1 eV. This is a result of the saturation of the inverse bremsstrahlung absorption due to the strong field correction.

6. Discussion and Conclusions

The maximum heat flux is observed to be limited to about 0.1 of the free streaming limit. Temperature gradients with λ/L of about 0.1 are generated and T_e/T_i is about 5. Only a very low level of ion acoustic turbulence is observed. The strong field correction should be included in the inverse bremsstrahlung absorption coefficient when the laser irradiance is large. The ion-electron equilibrium rate is found to be within 50% of the classical value.

Our simulation code for the experimental conditions⁽¹⁴⁾ gives a poor fit to the quoted measured temperatures for all values of the flux limit. The best fit is for a flux limit of $f = 0.06$ with $\lambda/L = 0.2$ and should be compared with the published values of $f = 0.03$ and $\lambda/L = 0.3$. The low value of the flux limit was attributed to ion acoustic turbulence. A surprisingly large value of $\delta n/n = 0.09$ was inferred from a 10 times observed enhancement of the unresolved scattered signal close to the incident probe frequency at $k\lambda_D = 2$. The ion acoustic energy spectrum is proportional to $k^4 S_{i0}(k)$, where $S_{i0}(k)$ is the integrated ion feature⁽¹⁴⁾. Using this and the ion acoustic energy spectrum⁽¹⁰⁾ it can be shown that the observed 10 times enhancement corresponds to a greater than 150 times enhancement at $k\lambda_D = 0.5$, close to where the energy spectrum peaks. This should be compared with the 1.5 times enhancement observed in our experiment at $k\lambda_D = 0.5$. A higher level of turbulence in the experiment⁽¹⁴⁾ may possibly be due to the electron mean free path being comparable to the focal spot size. This results in a large return current of electrons

giving rise to ion acoustic turbulence. In our experiment the mean free path was only about 0.15 of the focal spot size.

The simulation is in good agreement with the results⁽⁷⁾ for a maximum heat flux of 0.1 of the free streaming limit with $\lambda/L = 0.07$. This result and our value for the flux limit agree with calculations based on a Fokker-Plank model⁽²⁾ which limit the maximum heat flux to about 0.1 of the free streaming limit.

Acknowledgements

We particularly wish to thank M Lewis for her considerable contribution to the experiment. We would also like to thank A Cole and W Toner for their participation and interest.

References

1. L Spitzer and R Harm, Phys. Rev., **89**, 977 (1953).
2. A R Bell, R G Evans and D J Nicholas, Phys. Rev. Lett., **46**, 247 (1981).
3. R C Malone et al, Phys. Rev. Lett., **34**, 721 (1975).
4. F C Young et al, Appl. Phys. Lett., **30**, 45 (1977).
5. J D Kilkenny et al, Phys. Rev. Lett., **53**, 2563 (1984).
6. D R Gray, J D Kilkenny, M S White, P Blythe and D Hull, Phys. Rev. Lett., **39**, 1270 (1977).
7. E S Wyndham, J D Kilkenny, H H Chuaqui and A K L Dymoke-Bradshaw, J. Phys. D, **15**, 1683 (1982).
8. D E Evans and J Katzenstein, Rep. Prog. Phys., **32** 207, (1969).
9. J Sheffield, Plasma Scattering of Electromagnetic Radiation, Academic, New York (1975).
10. C T Dum, R Chodura and D Biskamp, Phys. Rev. Lett., **32**, 1231 (1974).

11. J P Christiansen, D Ashby, K V Roberts, Comput. Phys. Commun., 7, 271 (1974).
12. G J Pert, J. Phys. A., 5, 506 (1972).
13. S Rand, Phys. Rev., 136B, 231 (1964).
14. D R Gray and J D Kilkenny, Plasma Physics, 22, 81 (1980).

A2-3 SHORT PULSE ENERGY TRANSPORT BY THERMAL AND SUPERHERMAL ELECTRONS

D Brown, P Norreys, E Wooding (RHBC)
 J Dunn, R Barnsley (Univ Leicester)
 M Key, G Tallents (RAL)
 H Baldis (NRC Canada)

Introduction

The process of energy transport and ablation when laser radiation is absorbed at a surface to form a plasma has been studied extensively for conditions where the hydrodynamic flow pattern approaches a steady state, but is less well understood for short pulse duration where the behaviour is transient.

When the laser pulse duration is less than the hydrodynamic response time, electron thermal conduction heats matter at close to solid density in contrast to steady state ablation where the main heating occurs at densities closer to the critical density.

The physics of energy transport in laser-produced plasmas is of intrinsic interest because it involves temperature gradients which are steep relative to the mean free path of the electrons responsible for the energy transport. Fokker Planck models of heat flow in the steady state have shown the nature of heat flow under such conditions⁽¹⁾ and transient Fokker Planck models are now being developed⁽²⁾. Some earlier work at the CLF has studied heat flow with 100 ps pulses⁽³⁾. There has been little evidence to date of heating at high density because more strongly transient conditions have not been studied in detail.

Heating of matter at high density is of particular interest in the context of developing recombination based XUV lasers operating at wavelengths in the 'water window' below 43 Å as discussed in Section A1.2 and the present work is motivated by this consideration.

In addition to energy transport by thermal electrons there can also be heating by superthermal electrons produced by resonance absorption. It is well known that this produces a preheating of solid matter ahead of the ablation front, but at a relatively low temperature in situations studied to date. A novel possibility is to use hot electron preheating due to very short pulses of short wavelength at high intensity to heat solid matter to high temperatures of interest for XUV lasers.

The condition for strong resonance absorption is that the sub critical corona be optically transparent. Its opacity can be expressed as

$$\kappa\lambda \sim 1.4 \times W^{-4} (\lambda/1 \mu\text{m})^{-10/3} (I/10^{14} \text{ Wcm}^{-2})^{-2/3} (t/1 \text{ ns})Z$$

and can be less than unity for high intensity, short pulse duration t and low Z even for short λ .

The use of short λ is advantageous because the hot electron range $\rho\lambda_h \sim (I\lambda^2)^{0.9}$ and it is easily seen that, with a constant fraction of the intensity converted to hot electrons, the preheat temperature scales as $T_p \sim \lambda^{-2}t$ with the inertial lifetime of the heated layer scaling as $\tau_c \sim I^{2/3}\lambda^2$ giving a peak temperature $T_{pm} \sim I^{2/3}$. Thus a short wavelength at high intensity can heat a thin layer to high temperature. A quantitative model due to Harrach and Kidder⁽⁴⁾ suggests the possibility of heating 0.5 to 1 μm depth of solid to over 500 eV in this way with 10 ps UV pulses, which would be of considerable interest for an A2 XIII XUV laser at 39 Å.

Experiment

An experiment was carried out with 20 ps pulses from the VULCAN facility at 0.53 μm wavelength in a single beam at energies of about 10 J focussed to a 50 μm focal spot giving intensities of over 10^{16} Wcm^{-2} .

The energy transport was recorded using layered targets designed to show

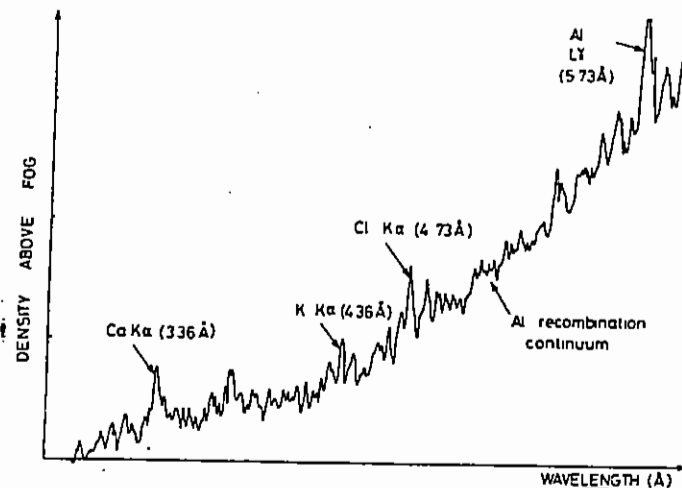


Fig. A2.12 K-emission from Cl, K and Ca and He-like/H-like lines of aluminium recorded by a minispectrometer.

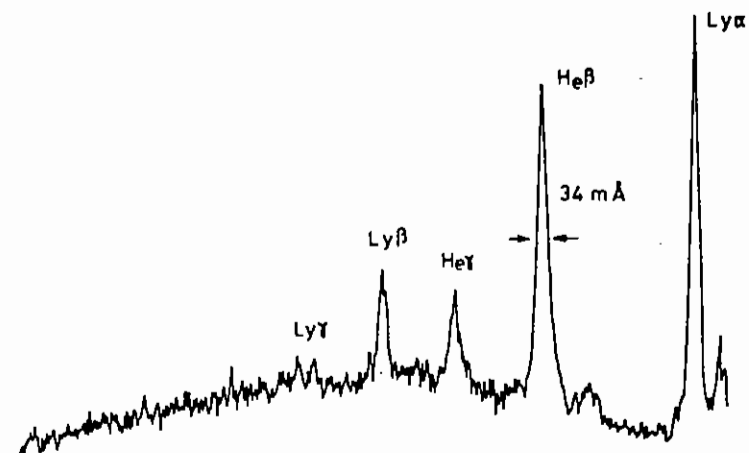


Fig. A2.13 He-like and H-like lines of aluminium recorded with a minispectrometer. The width of the Al XII $1s^2 \ ^1S_0 - 1s3p \ ^1P_1$ (He β) transition is marked. The aluminium layer for this shot was buried $0.5 \mu\text{m}$ below a CH ablator.

(a) the penetration of heating of the solid from the emission spectrum of an Al layer buried below a CH layer. The layer was thin enough to permit measurement of a time averaged n_e and T_e from its H-like and He-like resonance spectra, which were optically thin for high series members

(b) The penetration of hot electrons and their preheating effect from K_α fluorescence in deeper buried layers of K_α fluors KCl and CaF_2 .

The target layer composition was: CH ablator, 0 to 1 μm , Al 0.2 μm , CH 2.5 to 5 μm , KCl 2 μm , CH 5 μm , CaF_2 2 μm , substrate 10 μm mylar. The targets were discs of 300 μm diameter.

Several x-ray measuring instruments were used to diagnose the energy deposition of the hot electrons and the production of plasma in the layered targets. A minispectrometer with moderate spectral resolution (~ 500) using a diffracting crystal of PET ($2d = 8.742 \text{ \AA}$) was used to monitor K-emission from Cl, K and Ca and the He-like/H-like lines of aluminium as shown in Figures A2.12 and A2.13. A Johann geometry spectrometer with high resolution ($\sim 10^4$) using a diffracting crystal of PET bent to a radius of curvature of 0.5 m was employed to study the resonance and satellite lines of helium-like aluminium as shown in Figure A2.14. This instrument is described in more detail in Section A7.3.

A pinhole camera and a filtered x-ray diode array were also employed in the experiment. The pinhole camera monitored the degree of uniformity of plasma production by recording x-rays transmitted through a 25 μm Be filter. The x-ray diode array measured broad band continuum emission passed through x-ray filter foils of different atomic number and enabled plasma temperatures to be deduced.

Results and Discussion

The data from the experiment are still being analysed but some preliminary conclusions can be made.

The most interesting observation is that the emission from the buried Al layer recorded for typically 7 J 0.53 μm 20 μm irradiation showed broad lines characteristic of densities $\sim 10^{23}$ electrons cm^{-3} at a depth of 0.5 μm in contrast to the normal 10^{22} cm^{-3} of an ablating plasma produced by long pulse irradiation (see Figure A2.13). The width of the Al XII 1s - 1s 3p transition is consistent with a density

of 10^{23} cm^{-3} using the line width analysis of Kilkenny et al⁽⁶⁾ with appropriate scaling. Detailed analysis of the spectra is presently underway. This result is the most direct observation to date of thermal transport to densities close to solid.

The aluminium satellite feature are also being analysed and will give further information on the plasma densities.

In future experiments this method will be applied to the study of 10 J, 10 ps pulse irradiation at 0.25 μm wavelength using the KrF laser discussed in Section A5.

References

1. A Bell, R Evans and D Nicholas, Phys. Rev. Lett., **46**, 243 (1981).
2. Current research supported by SERC at Univ. of St Andrew (A Cairns, J Sanderson and S Jorna) and Imperial College (A Bell).
3. S D Tabatabaci, D Riley, O Willi and C Chenais-Popovics, Central Laser Facility Ann. Rep. (1987).
4. R Harrach and R Kidder, Phys. Rev., **A23**, 887 (1981).
5. A H Gabriel, Mon. Not. R Aston. Soc., **160**, 99 (1972).
6. J D Kilkenny, R W Lee, M H Key, J Lunney, Phys. Rev. A., **22**, 2746, (1980).

A2-4 PICOSECOND HIGH BRIGHTNESS KrF LASER PLASMA INTERACTIONS

V. Barrow, J Edwards, G Kiehn, R Smith, J Wark and O Willi (Imperial College)

E Turcu (RAL)

The recent development of high power short pulse laser systems now allows laser plasma interactions to be studied in new regimes. The deposition of laser energy occurs at high plasma densities near to the solid density material, since no large hydrodynamic motion can occur on

picosecond time scales. The production of hot dense plasma by picosecond lasers is of interest in basic plasma physics studies and, in addition, short wavelength, short pulse lasers may be required to extend the operational range of recombination x-ray lasers to shorter wavelengths due to the strong density scaling with atomic number of the lasing ion⁽¹⁾. The short pulse capabilities of the SPRITE laser were used to investigate ionization balance and x-ray production in ps KrF laser produced plasmas.

Planar targets were irradiated with 2.5 J KrF laser pulses 3.5 ps in duration. Using an f/2.5 lens, irradiances of up to 10^{17} W cm⁻² on target were obtained. The plasma was primarily diagnosed using XUV and x-ray spectroscopy. A flat field spectrograph was used to record time resolved XUV spectra in the 30-130 Å spectral range. Flat PET and Si crystals were used in microcrystal spectrographs to record time integrated spectra on DEF film and also coupled to an x-ray streak camera to record time resolved spectra. The plasma size was measured using an x-ray pinhole camera. The short KrF laser pulse energy and the amplified spontaneous emission level were recorded with an optical diode.

A variety of targets were irradiated to investigate the state of ionization in the plasma as a function of atomic number of the target. In particular, the possibility of using ps KrF laser pulses to produce fully ionized Al plasmas for use as x-ray laser gain media was examined. Targets from F to Ca were irradiated and the H-like and He-like x-ray emission was recorded. Figure A2.15(a) shows a time integrated Al x-ray spectrum in the 6-8 spectral Å region. Aluminium Ly_α and Ly_β are visible along with the He-like transitions. The Lyman alpha transition was observed in targets with increasing atomic number up to sulphur, Z = 16. However, only He-like transitions were observed with chlorine, potassium and calcium targets. In addition to the K-shell spectroscopy, the intensity and duration of L-shell and M-shell x-ray emission was examined for high Z ps laser produced plasmas. Figure A2.15(b) shows a time resolved spectrum of the M band of tantalum. The x-ray emission is seen to turn on sharply and last for approximately 20-50 ps.

These x-ray spectra, both time integrated and time resolved, are the first single shot spectra to be recorded from ps laser solid interactions. The preliminary observations are currently being analysed to provide details of the ionization balance in low Z plasmas and to

describe the temporal behaviour of the x-ray emission from the K, L and M shells of a variety of atomic number targets.

References

1. G Pert, Plasma Phys. and Contr. Fusion, 27, 1427 (1987).
2. J R M Barr, N J Everall, C J Hooker, I N Ross, M J Shaw and W T Toner, to be published in Opt. Comm.

A2-5 OBSERVATIONS OF INSTABILITIES IN LONG SCALELENGTH PREFORMED PLASMAS

D Bassett, S Coe, D Riley and O Willi (Imperial College)
A Giuliotti (Pisa)
A Hauer and C Gomez (Los Alamos)
T Afshar-Rad (RAL)

A series of experiments have been carried out to study non-linear laser-beam-plasma interactions in long scalelength preformed plasmas. These experiments involved groups from Pisa, Los Alamos, Imperial College and RAL.

A large hot dense cylindrical plasma up to 2 mm in length and 200 μm in diameter was produced using a modified short line focus arrangement. Two pairs of opposing green laser beams were focused onto thin film targets. All four laser beams were superimposed to give an irradiance of 10^{14} W cm⁻² in a 70 ps laser pulse. The targets consisted either of a 100 nm thick formvar substrate overcoated with a 200 μm by 350 nm Al stripe or a 200 μm by 350 nm Al ribbon. A green high power laser beam 800 ps in duration was focused into the preformed plasma with a 100 μm focal spot to interact axially with the plasma.

A number of diagnostics were used to study the plasma conditions before and during interaction as well as the non-linear interaction processes taking place such as stimulated Raman scattering, stimulated Brillouin scattering and laser beam filamentation. The temperature of the plasma was obtained from time resolved x-ray spectroscopy. Multiframe optical probing photography was used to study the plasma uniformity. Time resolved optical spectroscopy was used to investigate Stimulated Raman scatter and stimulated Brillouin scatter. The break up of the laser

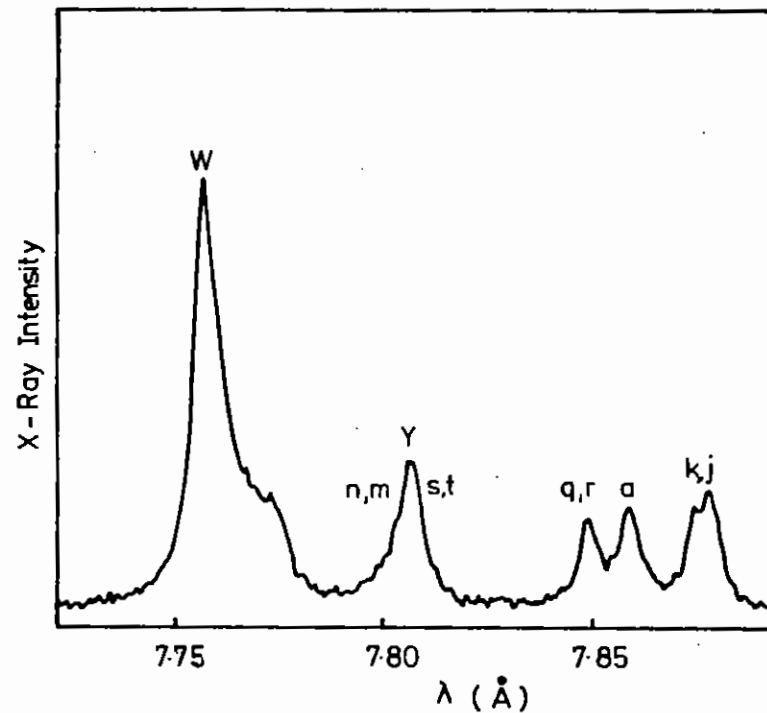


Fig. A2.14 Al XII spectrum recorded in a single shot by a high resolution Johann spectrometer. The $1s^2 \ ^1S_0 - 1s2p \ ^1P_1$ resonance line (labelled w) is shown with long wavelength satellite structure labelled according to Gabriel [5]. The aluminium was not overcoated for this shot.

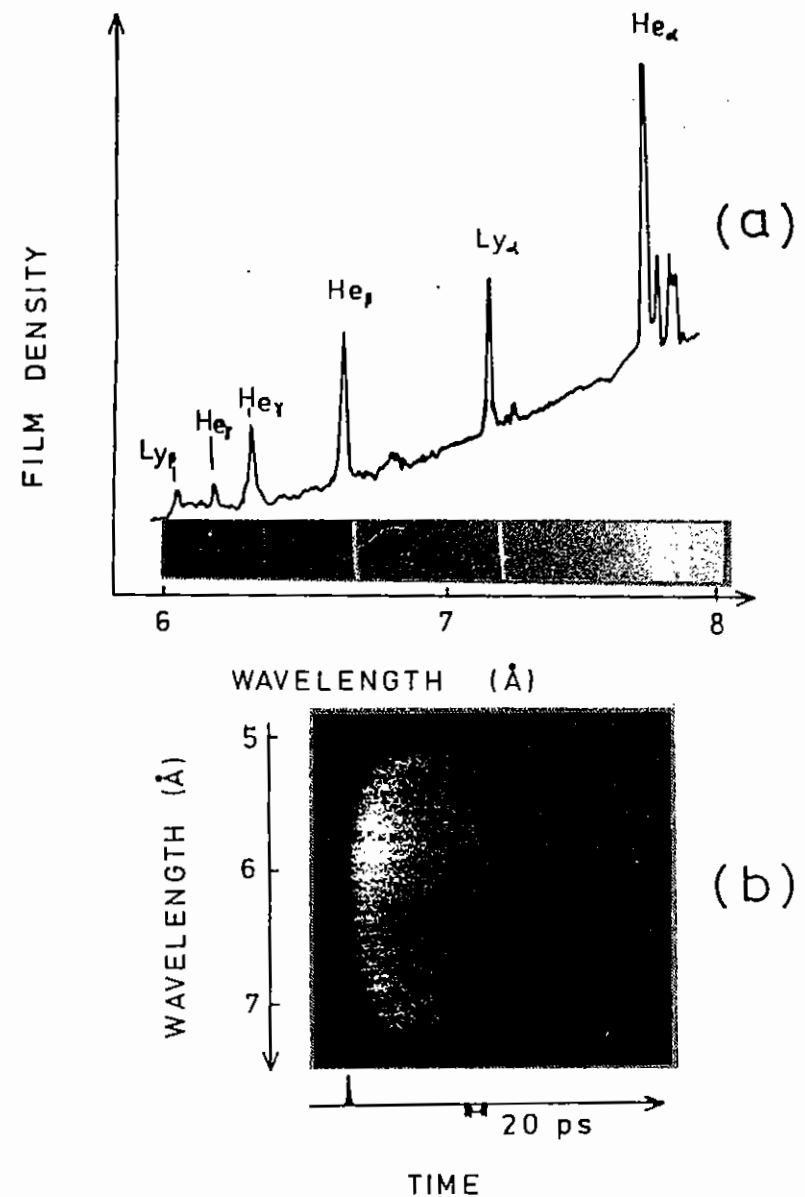
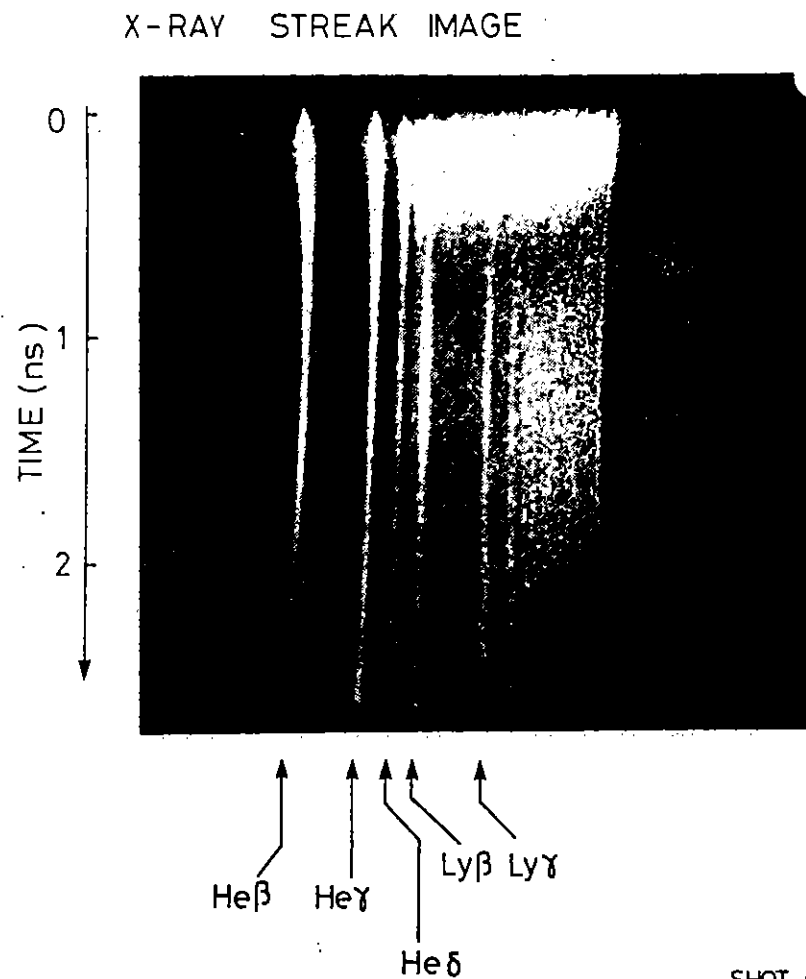


Fig. A2.15 Examples of spectra obtained in the picosecond high brightness KrF laser plasma interaction experiments. (a) Time integrated Al X-ray spectrum (b) Time resolved Ta X-ray spectrum.



SHOT 02 200887

Fig. A2.16 Spectrally resolved x-ray streak image of an Al ribbon target.

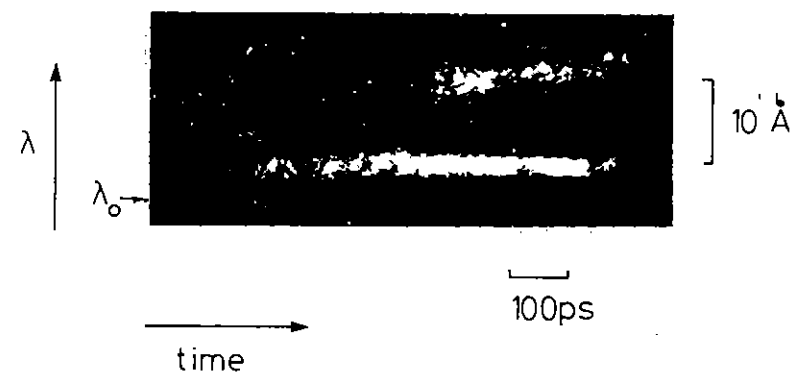


Fig. A2.17 Time resolution of stimulated Brillouin backscatter in a long scalelength plasma. The interaction laser wavelength is marked λ_0 .

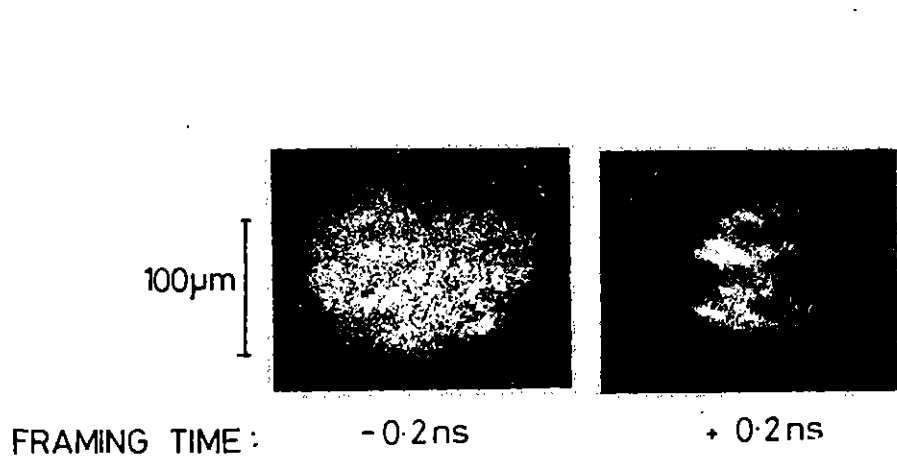


Fig. A2.18 Framed images of the transmitted interaction beam. Times are relative to the peak of the laser pulse.

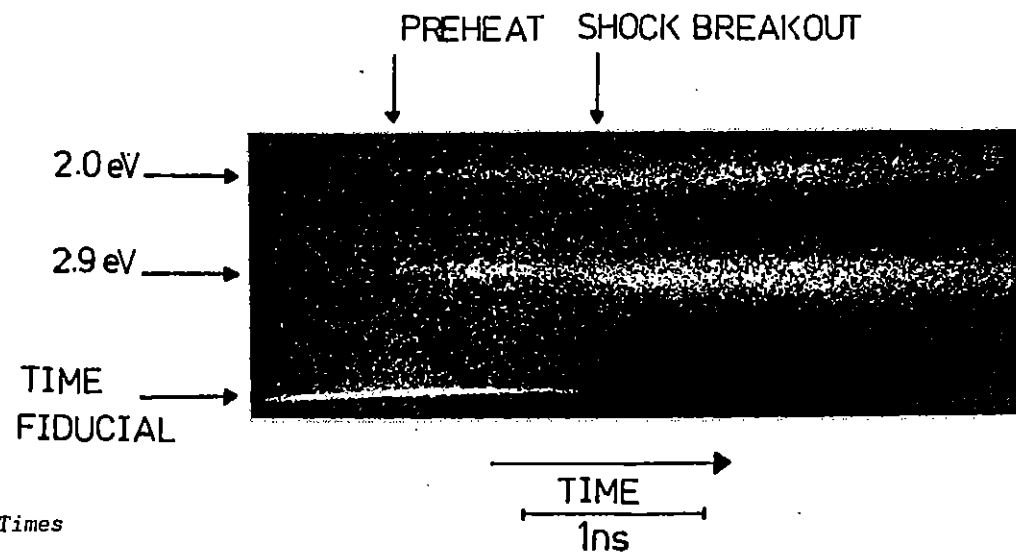


Fig. A2.19 Optical streak image of 2 and 2.9 eV photons from the rear surface of a 5 μm Al target coated on the front surface with 0.1 μm of gold.

beam tunnelling through the plasma was observed with an optical gated intensifier with a 100 ps framing time.

Figure A2.15 shows an x-ray streak image taken on an Al ribbon target. As can be seen the H-like transitions gain in intensity when the interaction beam is incident (around 1.2 ns). Initial analysis using line ratio measurements indicate temperatures of about 400 eV before the interaction takes place and about 3 keV during the interaction. Large shifts with a sudden turn on were observed in stimulated Brillouin backscattered spectra. Figure A2.17 shows a typical example, a shift of 1.3 nm is measured resulting in a temperature of about 3 keV. This sudden turn-on late in the laser pulse may be connected with localised heating caused by laser beam filamentation.

A break up of the laser beam is clearly observed after the peak of the laser pulse with the gated optical intensifier. Images were taken at different times during the interaction beam pulse on a series of laser shots. Two typical images obtained are shown in Figure A2.18, the times given are relative to the peak of the interaction pulse. The target was an Al stripe 350 nm thick and 200 μm wide. It can be seen that the image taken early in the interaction pulse shows a uniform beam profile compared with that taken later, which shows the break up of the beam profile into intense hot spots. Hence this break up of the laser beam profile takes of the order of a few hundred ps to develop, which is consistent with this effect being due to laser beam filamentation.

A2-6 RADIATION PREHEAT AND RADIATION TRANSPORT OBSERVATIONS IN THIN FOIL TARGETS

J Edwards, V Barrow, D Riley and O Willi (Imperial College)
T Afshar-Rad, S Rose (RAL)
R Benattar, V Malka (Ecole Polytechnique)

Experiments have recently been carried out to study the level of preheat at the rear side of thin foil targets caused by hard x-ray radiation which is generated in laser produced plasmas. This work was performed as part of an ongoing collaboration with R Benattar of Ecole Polytechnique.

Aluminium targets of various thickness (5 μm - 15 μm) and plastic targets (0.1 μm - 40 μm thick) coated with 0.1 μm of gold on the front side were used. The temperature behaviour of the rear side was observed using optical and VUV streaked photography. The transport processes of soft x-ray radiation through the target was investigated using time resolved XUV spectroscopy. The focal spot size was monitored with an x-ray pinhole camera, filtered with 0.8 μm of aluminium. The M-band of gold was observed with a time resolved x-ray spectrometer.

The targets were irradiated with a green laser beam 800 ps in duration. Figure A2.19 shows an optical streak image of 2 eV and 2.9 eV photons taken on a 5 μm Al target. The time fiducial can also be seen. Weak emission is observed before the shock breaks through indicating that the rear side was preheated by radiation. At the relatively low irradiances used ($I\lambda^2 \sim 5 \cdot 10^{13} \text{ W cm}^{-2} \mu\text{m}^2$), preheating due to suprathermal electrons is minimal. Similar images were obtained at 10 eV with a Schwarzschild microscope.

Figure A2.20 shows time resolved spectra taken on a very thin 0.1 μm formvar foil and on a 6.5 μm CH foil. Both were overcoated with 0.1 μm of gold on the front side. When the substrate foil is very thin, the soft x-rays are not significantly affected by the foil. However, a clear time delay, particularly at longer wavelengths, is observed when a 3 μm (or thicker) substrate foil is used. These observations indicate that the radiation has been absorbed and re-emitted in the foil.

A detailed analysis of the data obtained is presently being undertaken and the results will be presented in the near future.

A2-7 ENERGY TRANSPORT UNIFORMITY ON SPHERICAL TARGETS - FURTHER ANALYSIS AND RESULTS

P Apte, A J Cole, M H Key (RAL)
D Brown, P Norreys, E R Wooding (RHBC)

Introduction

Laser driven implosions are of major importance in ICF research and the

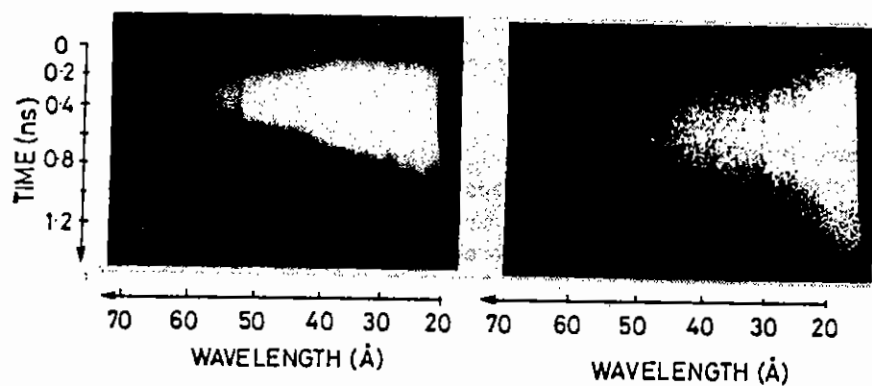


Fig. A2.20 Time resolved spectra from the rear surface of a thin ($0.1 \mu\text{m}$) formvar foil (LHS) and thicker ($6.5 \mu\text{m}$) plastic foil (RHS), both overcoated with $0.1 \mu\text{m}$ of gold on the front side.

X - RAY MONOCHROMATIC PINHOLE - STREAK CAMERA

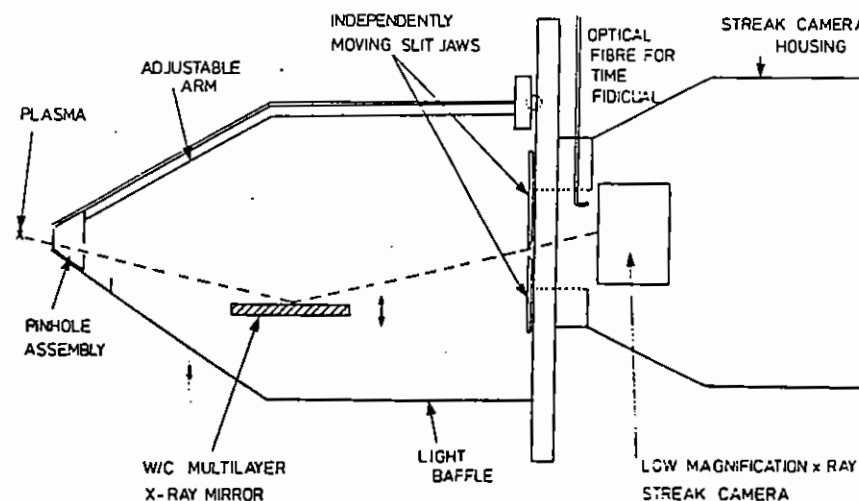


Fig. A2.21 Schematic of a time resolving monochromatic x-ray pinhole camera.

study of dense plasmas. They require highly uniform pressure produced by laser irradiation, where the degree of uniformity is a function of both the irradiation pattern and electron thermal transport.

Laser energy is deposited in the plasma by inverse Bremsstrahlung absorption in the underdense corona and energy is transported inwards to high density by electron thermal conduction. An axial separation is established between the absorption front, where the plasma outflow is sonic, and the high density ablation front, where velocity is zero and the ablation pressure is maximised. The axial separation of the absorption and ablation fronts increases rapidly for higher intensity and longer wavelength of laser irradiation⁽¹⁾.

Thermal diffusion smooths variations in pressure resulting from transverse variations in laser intensity and the smoothing effect is large when the axial separation exceeds the transverse scale length of the variations⁽¹⁾. In irradiation of spherical targets the sources of non uniformity include hot spots in the focussed laser beams and non uniform patterns resulting from beam overlap (including the effects of imbalance of power and errors of pointing in multi beam irradiation⁽²⁾).

The consequences of non uniform irradiation are both practical (the degradation of implosions), and scientific (causing errors of measurement, for example in the study of energy transport and ablation using observations of the depth or rate of penetration of the ablation).

Knowledge of the actual pattern of ablation rate is therefore important and can be sought by observations of X-ray emission from the ablation front, longer wavelengths being unsuitable because of the opacity of the plasma. Previous measurements have used a selectively filtered X-ray pinhole camera to record the burn through to a buried layer⁽³⁾. These results were inconclusive because they lacked time resolution and gave only a 'map' of regions where more than a certain burn depth was achieved. They also suffered from background emission from the blow-off plasma. Monochromatic X-ray imaging offers a solution to the background problem by imaging in a narrow spectral band matched to a bright emission feature in the spectrum of the indicator layer⁽⁴⁾.

In the present work we have used time resolved monochromatic imaging to obtain the 1D variation of the mass ablation rate in a spherical geometry for the first time. By systematically varying the focussing parameter D/R (D is the distance from the centre of the pellet to focus and R is the radius of the pellet) the non-uniformities due to beam overlap in 12 beam dodecahedral geometry have been observed and are reported in Section 2. A simple computer model has been used to derive the pattern of intensity modulations due to beam overlap on the surface of the sphere including the effect of variable energy balance between the beams. A comparison is made with the results in Section A2.2.

1. Experimental Technique

In a previous experiment an X-ray pinhole camera was combined with a multilayer X-ray mirror to produce monochromatic images⁽⁴⁾. A time and space resolving instrument has now been constructed by adding streak camera detection as illustrated in Figure A2.21.

The multilayer X-ray mirror (or LSM) had 75 layer pairs of tungsten and carbon with an effective 2d spacing of 43.8 Å and a spectral resolution $\Delta\lambda/\lambda \leq 50$ ⁽⁵⁾. The low spectral resolution is advantageous in that it 'smooths out' fine structure in the spectrum of the indicator layer giving greater spatial uniformity to the image in the dispersion direction. Selenium was selected as the X-ray emitter using its intense Se XXV L-shell emission lines at 7.8-8.1 Å giving a Bragg angle of 10° at the LSM mirror, which is sufficient to eliminate specular reflection.

The magnification of the image at the streak camera photocathode was x20 and the images were recorded on Ilford HP5 film at x17 magnification via the streak camera, image intensifier system. The size of the pinhole was 17 µm giving a spatial resolution of 18µm on the sphere. The sweep rate was 107 psec/mm in the output image and with a slit width of 0.47 mm at the photocathode, gave temporal resolution of 50psec. The slit also defined a 27 µm wide zone on the target for the 1D image. The image was filtered by 25 µ Be and 6 µ mylar.

A timing fiducial on the streak images was generated with Vulcan's synchronous 70 psec pulse oscillator converted to the fourth harmonic at $\lambda = 0.26 \mu\text{m}$.

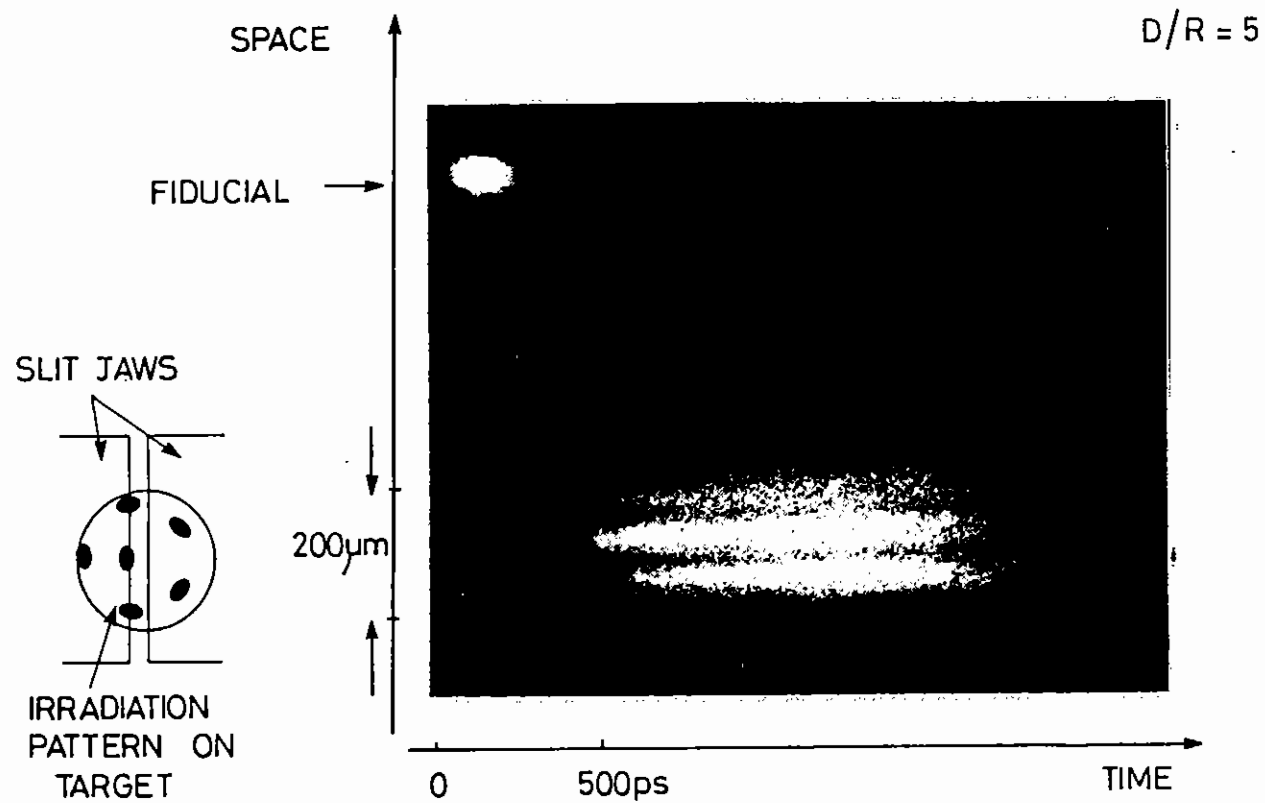


Fig. A2.22 Record with $D/R = +5$ for a $3.5 \mu\text{m}$ polymer coating on a solid polymer microsphere of diameter $\varnothing = 201 \mu\text{m}$ at an average absorbed irradiance of $6 \times 10^{13} \text{ W cm}^{-2}$.

2. Experimental Results

The targets used in the experiment were solid plastic microspheres (of diameter circa 200 μm) overcoated with 0.5 μm of Selenium and a polystyrene ablator layer which varied from 2.5 μm to 3.5 μm .

The size of the targets implies about 8 resolution elements across the surface of the sphere with a transverse width defined by the 1D slit of about 1/8 of the diameter. The targets were irradiated with 0.53 μm 0.8 nsec pulse of up to 400 J from the Vulcan laser facility with the 12 beams focussed by f/2.5 aspheric lenses. The focussing parameter D/R was varied from 5 to 15 and the laser energy from 100 to 400 J giving incident irradiance on the targets of about $2 \times 10^{14} \text{ Wcm}^{-2}$.

Figure A2.22 shows a typical result for D/R = 5. The picture is space resolved in one dimension and time resolved in the other. The time delay to burn through is seen across the diameter relative to the fiducial. Three regions of localised ablation are clearly visible, a feature that was reproducible in all D/R = 5 shots. This has been interpreted as being due to the beam overlap of the three nearest beams to the camera observation axis which was at a vertex in the dodecathral symmetry and is illustrated on the left of Figure 2.

Improvements in the uniformity were observed when the D/R parameter was increased. Figures A2.23(a) and (b) show the uniformity of ablation for D/R = 8 and 10 respectively.

The mass ablation rate $\dot{m}$ is derived directly from the observed burn through time and known ablator thickness⁽⁶⁾. The spatial variation of $\dot{m}$ changes with D/R as seen in Figure A2.24 which shows the maximum and minimum mass ablation rates for four shots with an average absorbed irradiance of $7 \times 10^{13} \text{ Wcm}^{-2}$. It can be seen that there is an improvement in the ratio of $\dot{m}_{\text{max}}/\dot{m}_{\text{min}}$ up to D/R = 10, but little improvement after this point.

To quantify the results and to allow a comparison of the spatial variation of $\dot{m}$ for different shots for the same values of D/R, the spatial uniformity is defined by $\eta = 1 - (\dot{m}_{\text{max}} - \dot{m}_{\text{min}})/\dot{m}_{\text{max}}$. If η is plotted against D/R, as in Figure A2.25, the improvement in uniformity with increasing D/R up to D/R = +10 is again apparent with little increase in uniformity occurring beyond this point. As the

uniformity of ablation increases the fraction of energy absorbed on the target decreases offering a compromise in practice between uniformity and coupling. The coupling factor was measured in a previous experiment for similar irradiation conditions and is known to peak at about 40% for D/R = 5 and fall to 30% for D/R = 10⁽⁷⁾.

The large variation in $\dot{m}$ shown in Figure A2.24 highlights the importance of spatially non-uniform ablation in measurements of ablation rate from laser burn through by time resolved X-ray spectroscopy with no spatial resolution⁽⁶⁾. Comparing data from this work for $6 \leq D/R \leq 12$ with measurements of $\dot{m}$ based on ion emission, (which are not sensitive to local intensity peaks) it is seen in Figure A2.26 that the peak values of $\dot{m}$ follow the same irradiance scaling as the ion data but are higher by a factor of 2. This shows that mass ablation rate derived solely from spatially integrated but time resolved spectroscopic measurements must be treated with some caution, a conclusion which has already been reached in other work⁽⁸⁾⁽⁹⁾.

Discussion

A computer program has been developed that maps the intensity profile of each beam over the surface of a sphere⁽¹⁰⁾. The program includes experimental parameters such as D/R and the differing energies of each beam, but neglects effects of the plasma atmosphere such as absorption and refraction⁽²⁾.

The intensity at each point on the sphere due to a single lens is given by

$$I(\theta) = (1 + \Delta)^2 \frac{1 + \Delta \cos \theta_p}{(\cos \theta_p + \Delta)^3} \times I(R)$$

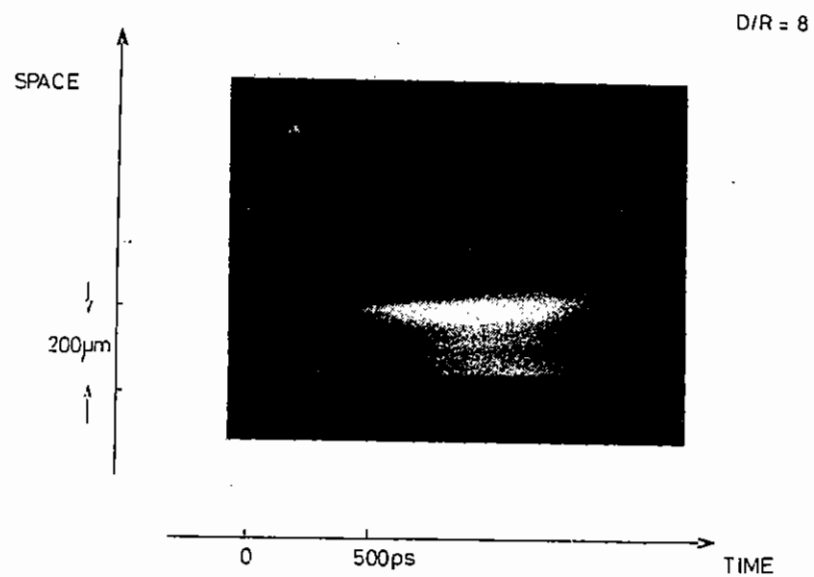
where $\Delta = D/R$

$$\cos \theta_p = \cos \theta \cos \theta_k + \sin \theta \sin \theta_k \cos(\phi - \phi_k)$$

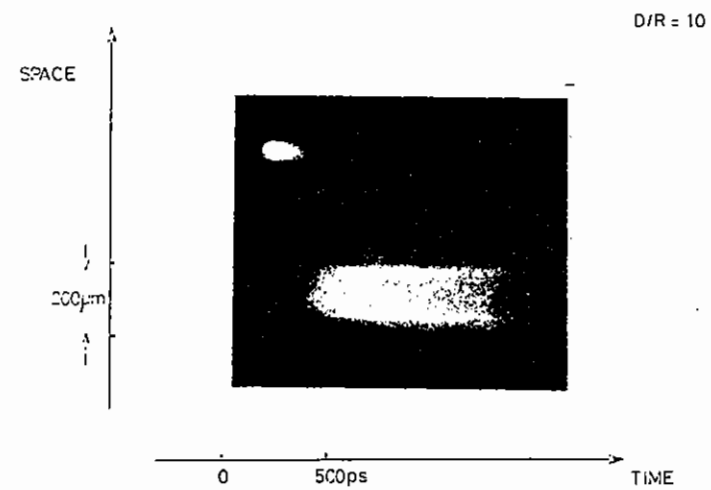
and θ is the latitudinal offset on a great sphere

ϕ is the longitudinal offset on a great sphere.

The positions of each beam (ie θ_k and ϕ_k) can be found from a schlegel diagram of a dodecahedron and are tabulated in reference (10).



(a)



(b)

Fig. A2.23 Streak record of ablation with (a) $D/R = + 8$ (b) $D/R = + 10$.

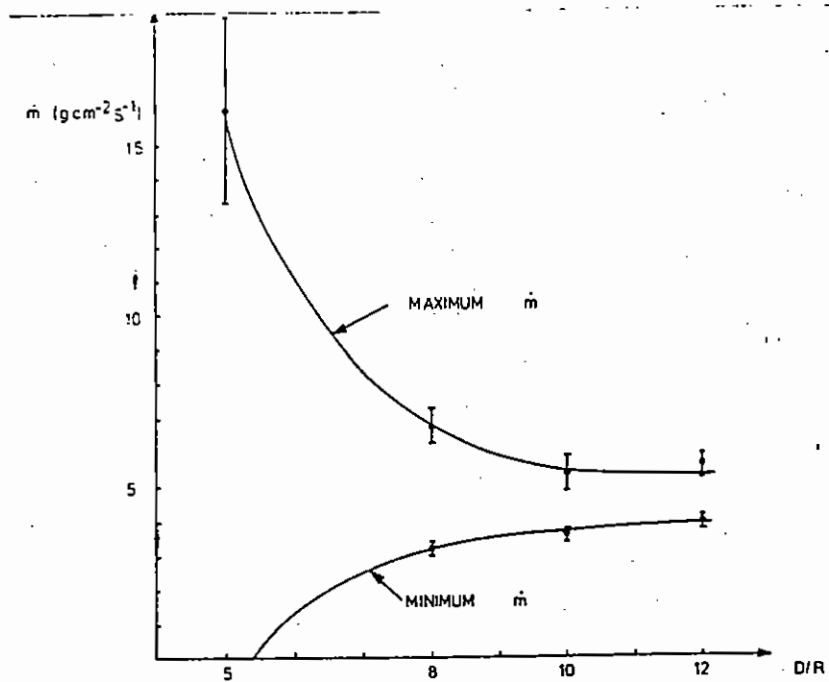


Fig. A2.24 Maximum and minimum mass ablation rates against D/R for four shots with an average absorbed irradiance of $7 \times 10^{13} \text{ W cm}^{-2}$.

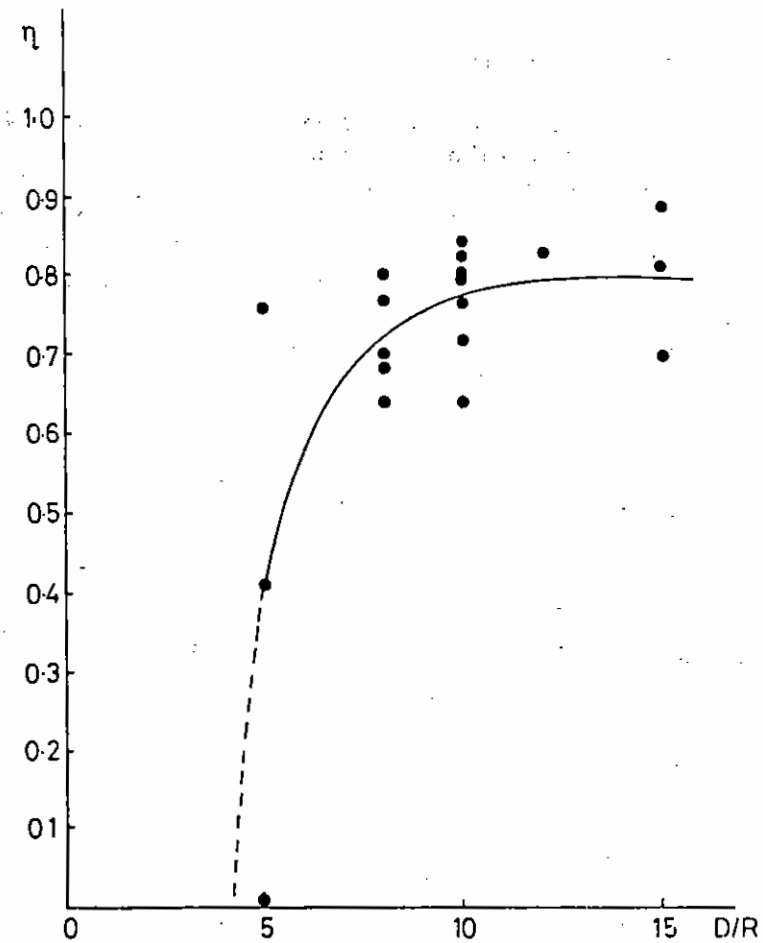


Fig. A2.25 Spatial uniformity η against D/R .

$I(R)$ in the above equation was modelled using a super-gaussian that approached a top hat function in profile as this was found to give reasonable agreement with the measured beam profile.

The target in shot 18 25/3/87 had a diameter of 204 μm and was overcoated with 0.1 μm of selenium and 3.5 μm of polymer. 299 J were incident on this target with a beam balance of $\pm 17\%$. Figure A2.27(a) shows the computed intensity profile within the slit over the hemisphere that the instrument was viewing. As such it is an angular plot, whereas the observed profile corresponds to a chord across the surface of the sphere. This has the effect of contracting the two edges of the computed profile. The D/R value was +12 in this case. Excellent agreement can be found with Figure A2.27(b).

Figure A2.28(a) shows the intensity profile for a D/R = +5 shot. 269J were incident on the target which had a diameter of 201 μm overcoated with 0.1 μm of Se and 3.5 μm of polymer. The beam balance was $\pm 20\%$ in this shot. Reasonable agreement is again seen in Figure A2.28(a) although the peak intensity to the right in Figure A2.28(a) is larger than the simulation suggests.

In all 50% of the shots could be reproduced in the modelling with explicit account being taken of D/R and differing beam energies. The remaining data showed patterns which we attribute to other factors such as pointing errors⁽²⁾.

Mass ablation rates derived from spatially integrated, time-resolved spectroscopic measurements of burn through to buried marker layers for $\lambda = 1.06\mu\text{m}$ laser irradiation broadly agree with ion velocity measurements⁽⁶⁾ which are not affected by non-uniformities in laser energy deposition at the critical surface. Thus hot spots and the effects of beam overlap and balance are smoothed out due to the large axial separation between the ablation and absorption fronts.

The present results for $\lambda = 0.53\mu\text{m}$ laser irradiation indicate that beam overlap and energy balance start to significantly affect the spatial uniformity of ablation. This is probably due to the reduced axial separation between the critical surface and the ablation front. These effects can be modelled and are on a spatial scale of $> 20 \mu\text{m}$.

When the incident laser wavelength is reduced still further to third harmonic $\lambda = 0.35 \mu\text{m}$ there is a significant divergence between mass

ablation rates derived from ion velocity measurements and those derived from spatially integrated time resolved spectroscopy⁽⁸⁾. This has been attributed to laser beam hot spots which occur on a spatial scale of less than 20 μm .

The intensity of the image could be increased in a number of ways to allow the pinhole size to be reduced and the spatial resolution to be increased to observe the smaller scale non-uniformities for 0.35 μm laser irradiation. Firstly, the filter protecting the multilayer mirror and the beryllium photo-cathode could be reduced in thickness to allow a much greater transmission of X-rays to the CSI. Secondly, the 2d spacing of the multilayer mirror could be increased to increase the intensity of the image. Thirdly, a longer wavelength marker layer could be chosen so as to increase the intensity of the X-ray emission at the ablation front.

All of these improvements will allow the pinhole to be reduced to the diffraction limit with no loss of intensity at the photo-cathode plane. Non-uniform ablation on this scale could then be detected.

Conclusion

We have measured the spatial uniformity of the mass ablation rate in spherical geometry for the first time. The best uniformity of $77 \pm 6\%$ was achieved for D/R = +10 for an irradiance of around 10^{14} Wcm^{-2} on a 200 μm microballoon. The energy coupling was 30%. This measured uniformity is significantly worse than the 97% uniformity of drive pressure needed for direct drive ICF. The results indicate that the major cause of non-uniform ablation was the poor energy balance between the beams. Significant effort should go into improving the beam balance to achieve implosions with a high density core.

New methods of smoothing the beam profiles such as random phasing⁽¹¹⁾ and induced spatial incoherence⁽¹²⁾ should significantly improve the uniformity of ablation. This experimental method could be applied to planar and spherical targets to determine if this is indeed the case.

Thermal smoothing of non-uniform energy deposition has been measured in planar geometry by a number of techniques. No direct measurements of thermal smoothing in spherical geometry have been reported. With the improved spatial resolution discussed earlier, this technique could be used to directly measure the effect of thermal smoothing in spherical

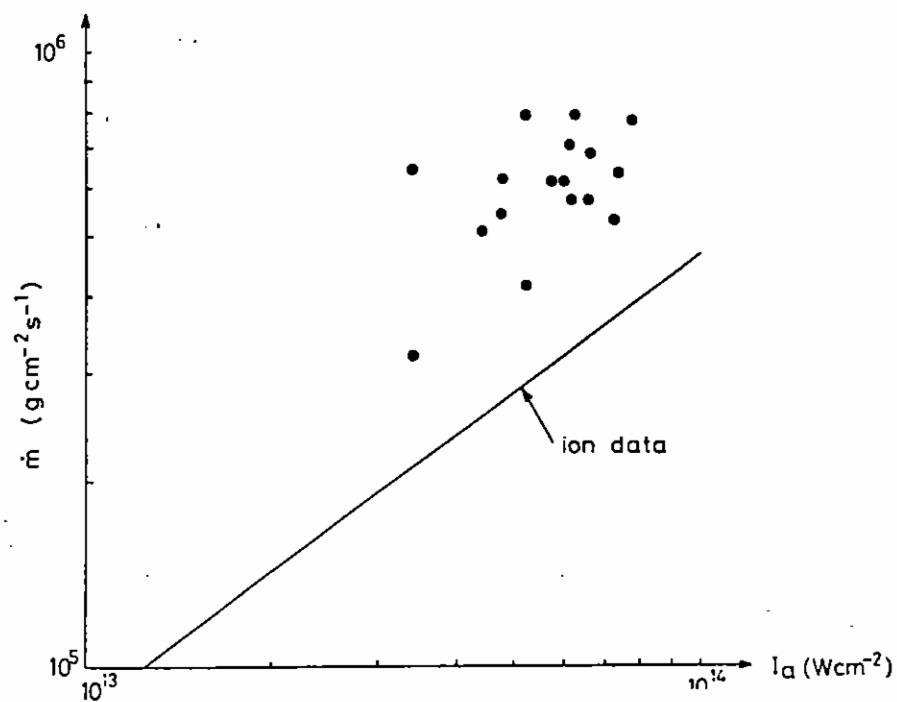


Fig. A2.26 Maximum mass ablation rates plotted against average absorbed irradiance. Ion data is from reference 6.

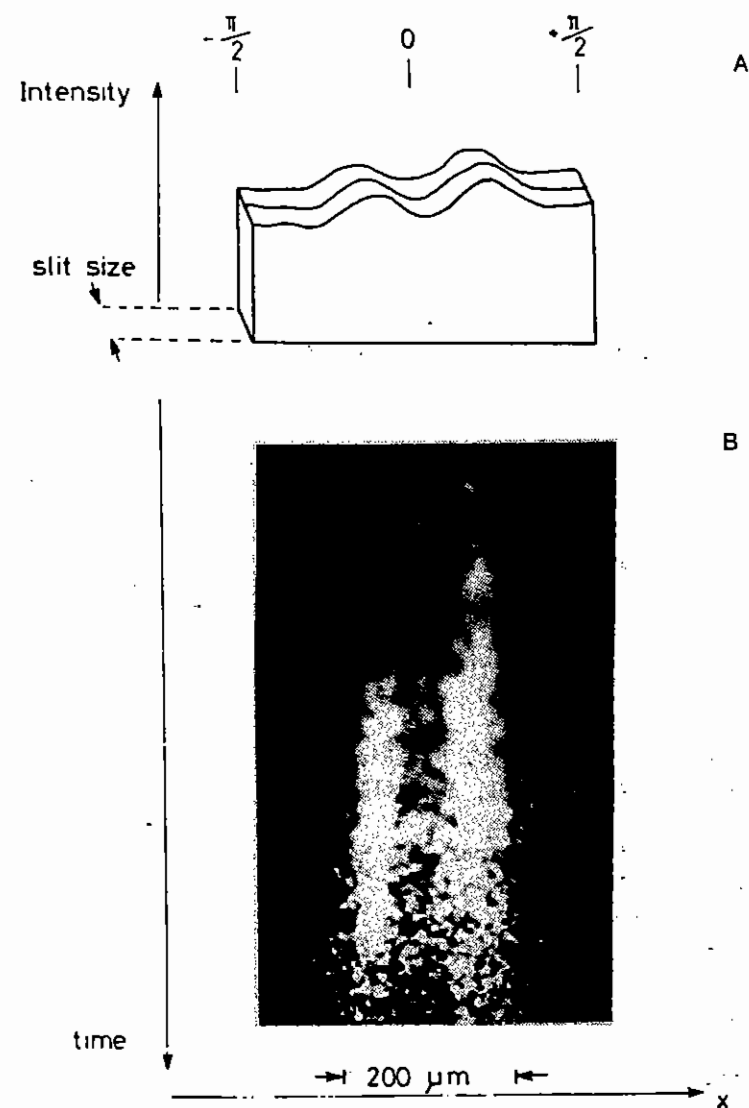


Fig. A2.27 (a) Completed irradiation pattern for $D/R = +12$ and beam balance of $\pm 17\%$.
(b) Observed ablation pattern.

geometry for both the transient and steady state build up of the plasma atmosphere⁽¹¹⁾ for various wavelengths and irradiances.

References

1. S E Gardner, S E Bodener, Phys. Rev. Lett., 47, 1137 (1981).
2. A R Bell, D J Nicholas, W T Toner, J. Phys. D., 19, 1869 (1986).
3. J Wark, J D Kilkenny, B J MacGowan, Ann. Rep. to LFC, RAL-84-049, A2.8 (1984).
4. A J Cole, M H Key, A Ridgeley, D A Brown, P A Norreys, E R Wooding, Optics Comm., 42 (1), 1 (1987).
5. K Evans, A J Cole, Ann. Rep. to LFC RAL Report RAL-85-047, 46.49 (1985).
6. T J Goldsack, J D Kilkenny, B J MacGowan, P F Cunningham, C L S Lewis, M H Key, P T Rumsby, Phys. Fluids 25(9), 1634 (1982).
7. J Corbett, C L S Lewis, A J Cole, M H Key, Ann. Rept. to LFC RAL-86-046, A2.1 (1986).
8. J Delettrez, R Epstein, M C Richardson, P A Jaanimagi, B L Henke, Phys. Rev. A., 36, 181, 3926 (1987).
9. P A Janimagi, J Delettrez, B L Henke, M C Richardson, Phys. Rev. A. 34, 1322 (1986)
10. J J Howard, Appl. Optics., 16, 2764 (1977).
11. Y Kato et al, Phys. Rev. Lett., 53, 1057 (1984).
12. R H Lehmborg, S P Obenschain, Optics Comm., 46, 27 (1983).

A2-8 THOMSON SCATTERING IN LONG SCALELENGTH PREFORMED PLASMAS

S E Coe and O Willi (Imperial College)
T Afshar-Rad (RAL)

A series of experiments have recently been carried out to study the interaction of a high power laser beam with a long scalelength preformed plasma. High levels of various instabilities such as stimulated Raman scatter, stimulated Brillouin scatter and filamentation have been observed. As a follow up to this an experiment has been carried out using Thomson scattering as a diagnostic to determine the plasma conditions (ie density and temperature) under which these instabilities occur.

The long scalelength plasmas are generated by exploding thin foil stripe targets using a line focus. The experimental set up used for Thomson scattering is shown in Figure A2.29. The probe beam was generated by Raman shifting a second harmonic (70ps) main beam to 622 nm. It was found that the largest conversion efficiency was obtained by decreasing the beam diameter to about 3 cm to increase its power density and then passing it directly through the Raman cell. The conversion efficiency was found to be highly dependent on the length of the cell and by optimization of this energies of up to 500 mJ were obtained (efficiency ~ 5%). Such a probe will appear much brighter than the background emission of the plasma.

The scattered light was detected by a spectrometer coupled to an optical streak camera, thus giving both the spectral and temporal resolution required. Both the probe beam and scattering channel were aligned on a scattering volume of 100 μm diameter. The scattering channel was initially set up at an angle of 145° and for typical plasma parameters of $N_e = 10 \text{ cm}^{-3}$, $T_e = 300 \text{ eV}$, this gives the value of the scattering parameter as:-

$$\alpha = \frac{1}{k\lambda_D} = 4$$

The 145° scattering channel can be used to determine the density of the plasma by fitting the spectra obtained to theoretical curves.

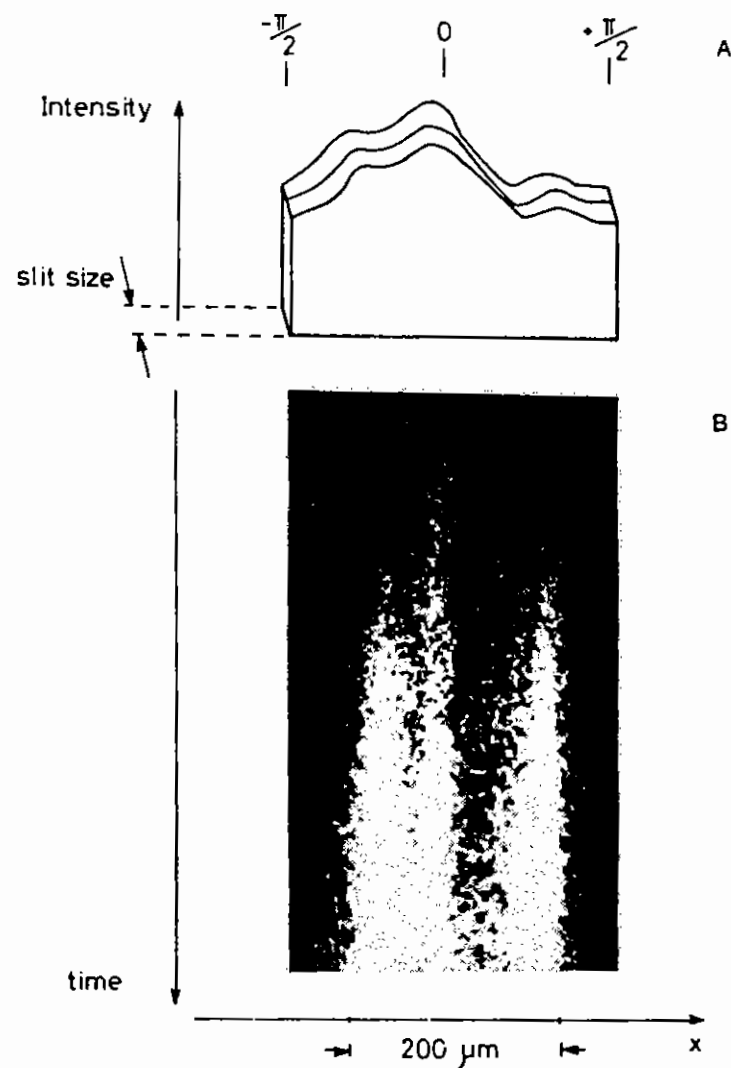


Fig. A2.28 (a) Computed irradiation pattern for $D/R = +5$ and beam balance of $\pm 20\%$.
(b) Observed ablation pattern.

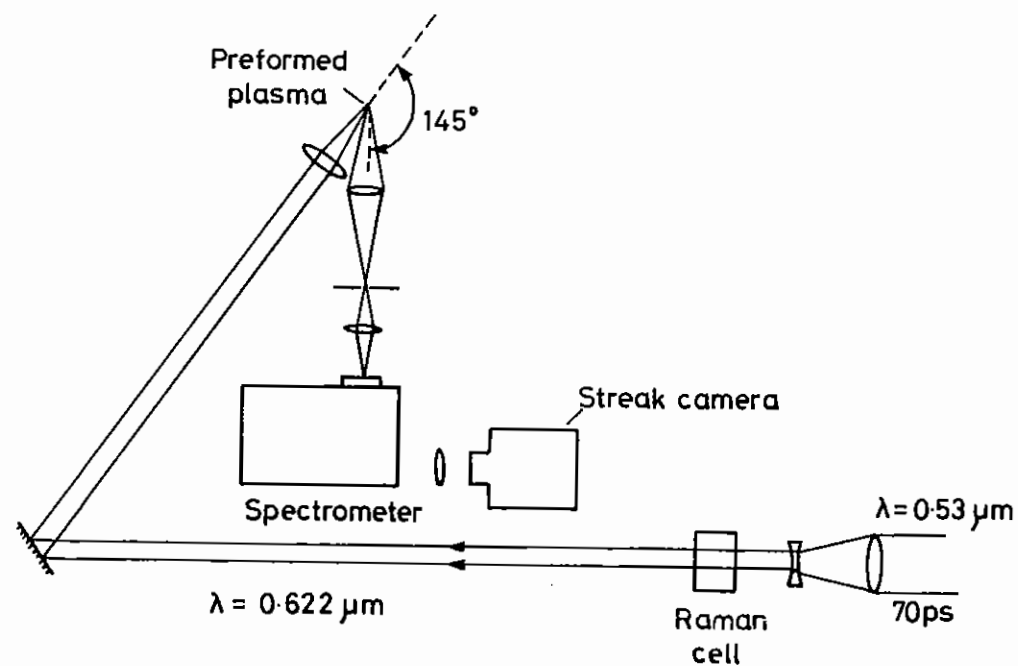


Fig. A2.29 Experimental layout for Thomson scatter in long scalelength preformed plasmas.

As this experiment has only just been completed (end of February 1988) no analysis of the data obtained has yet been carried out. This analysis will be started shortly and a full account of the results obtained given in the future.

A3	LASER COMPRESSION WITH DENSE PLASMAS	Pages
A3-1	Improvements to Ion Correlation Experiment in High Density Plasmas	77 - 78
A3-2	Twenty-Fold Increase in Thermonuclear Reaction Yield in Laser Driven Compression	78 - 90
A3-3	Production and Diagnosis of Coalescing Shocks	90 - 91
A3-4	Study of Coalescing Shocks	92

Editor: A Ridgeley

A3-1 IMPROVEMENTS TO ION CORRELATION EXPERIMENT IN HIGH DENSITY PLASMAS

B A Shiwai, T A Hall, A Djaoui, D Goodman, R W Eason, (University of Essex)

G Tallents and S J Rose (RAL)

Introduction

In last years annual report⁽¹⁾ the use of EXAFS (Extended X-ray Absorption Fine Structure) to measure the ion correlation effects in a dense shock compressed plasma was described. Although these experiments were very successful⁽²⁾, it was noticed that, in certain circumstances, the EXAFS modulation depth was not as great as would be expected. Even in laser shots where there were no targets compression it was observed that when the aluminium targets were covered by N-Parylene the EXAFS modulation was reduced. The reason for this is not understood but it appears as though the x-rays undergo small angle scattering in the N-Parylene. Since the early experiments a new type of plastic (E-Parylene) has become available and it was believed that this has a more homogeneous structure and at the same time being free from higher Z elements.

A short series of experiments was carried out to test this type of plastic coating and also to test more carefully for the level of radiative heating of the aluminium due to the backlighting source.

On the basis of the results from these initial experiments improvements were made to the technique and last years experiments on shock compressed plasmas were repeated.

Initial Experiment

In these initial experiments the targets were 3 mm square planar sandwich of 2 μm of aluminium covered on both sides by 4 μm of either N or E-Parylene. A frequency doubled laser light of 1 nsec pulse duration and 10-30 J energy was focused to a uranium backlighting target of 200 μm diameter. The x-rays from the backlighting source

which pass through the foil target are dispersed by a crystal mini-spectrometer placed about 40 mm from the backlighting target.

The single slit of the spectrometer is covered by a 25 μm Be foil to provide protection against visible light fogging.

The spacing between the backlighting and sandwich target was also varied in the range of 0.4 cm to 4 cm. This variation, together with variations in backlighting pulse energy, enable the x-ray flux absorbed by the sandwich target to be varied over a range of about 1000 times.

Results

A comparison between EXAFS spectra taken at a distance 2 cm away from the x-ray source for a 2 μm Al foil coated with 4 μm E-Parylene on both side and a similar target coated with the same thickness by N-Parylene Figure A3.1, shows that the modulation depth of the first peak in the EXAFS spectra for Al foil coated with E-Parylene is better than the one coated with N-Parylene by a factor of x2. The EXAFS modulation depth obtained from the E-Parylene coated target was insignificantly different from that with aluminium foil alone.

The EXAFS spectra taken at a distance 4 mm from the backlighting source for an E-Parylene coated target is shown in Figure A3.2. The modulation amplitude shown in Figure A3.2 should be compared with the E-Parylene target result shown in Figure A3.1. Some reduction in the modulation depth is observable. This reduction is interpreted as being the result of heating which is produced by the soft x-rays from the backlighting source. This effect will reduce the amplitude of the EXAFS modulation, with the higher energies from the K-edge being affected most.

A series of results similar to those given in Figure A3.2 show that, even at the highest backlighting beam energy, there was no noticeable heating when the separation between the backlighting and main target was greater or equal to about 2 cm. This should be contrasted with the separation of 3 mm used in last years ion correlation experiments. Clearly, some small degree of heating could have occurred in these experiments.

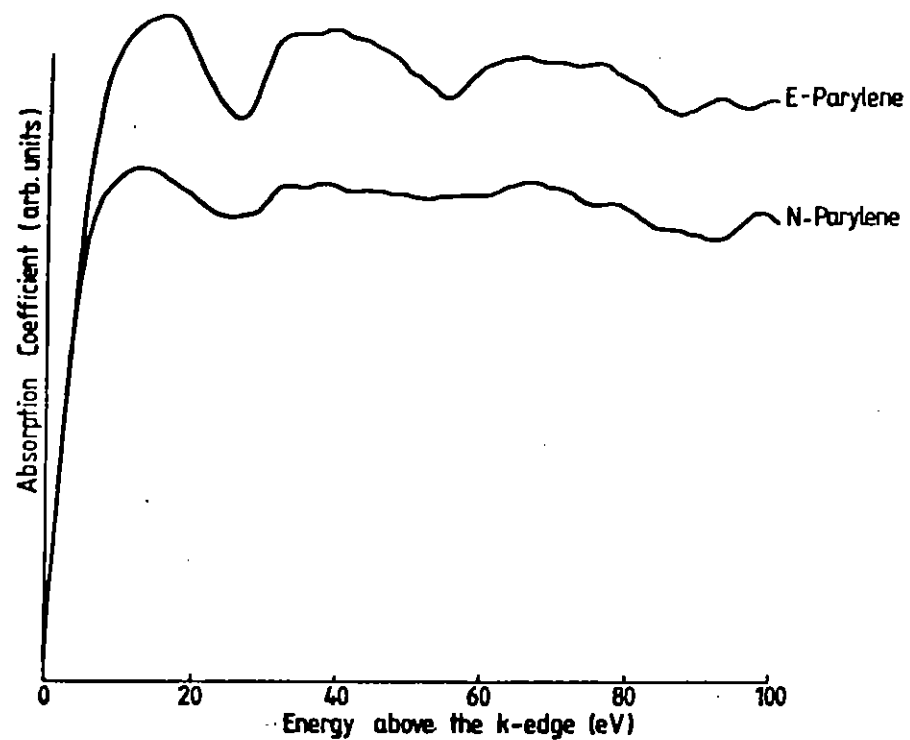


Fig. A3.1 A comparison between EXAFS spectra taken at a distance 2 cm from the x-ray source for N-Parylene (lower) and E-Parylene (upper).

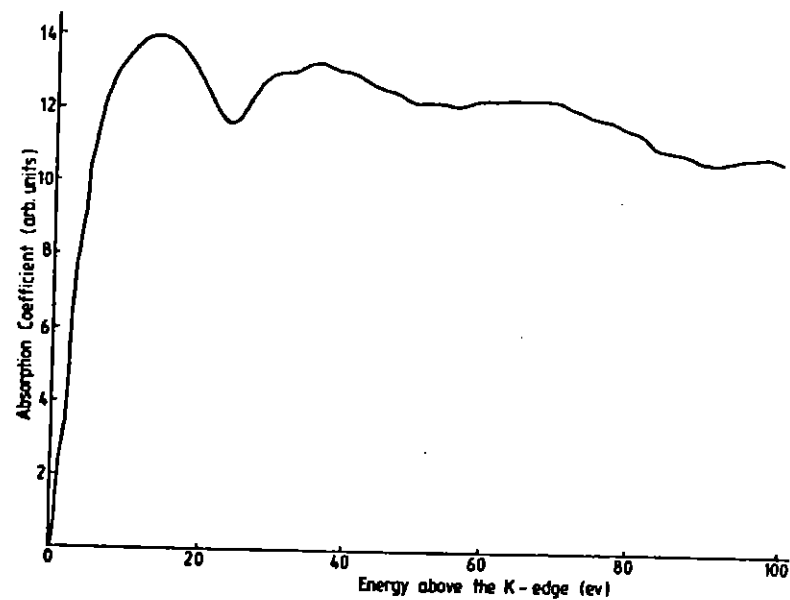


Fig. A3.2 An EXAFS spectrum for E-Parylene taken at a distance 4 mm from the x-ray source.

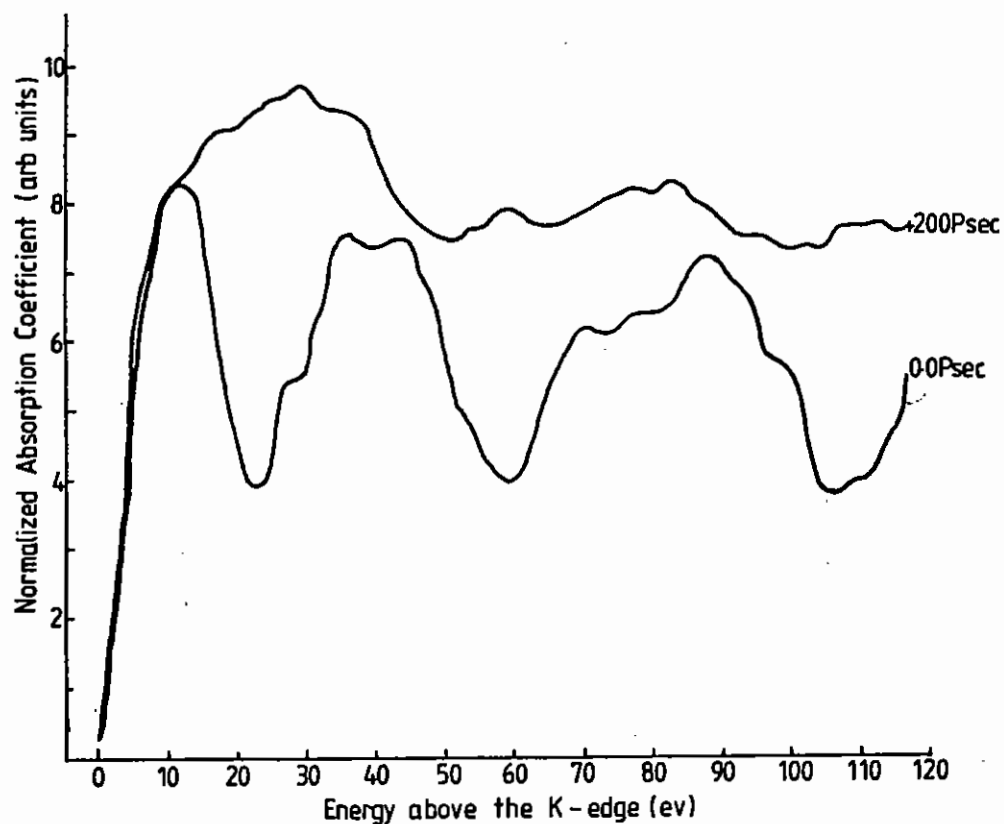


Fig. A3.3 Experimental aluminium absorption coefficient of EXAFS spectra as a function of energy above the K-edge for the uncompressed aluminium (0.0 psecs) and the compressed aluminium (+ 200 psec).

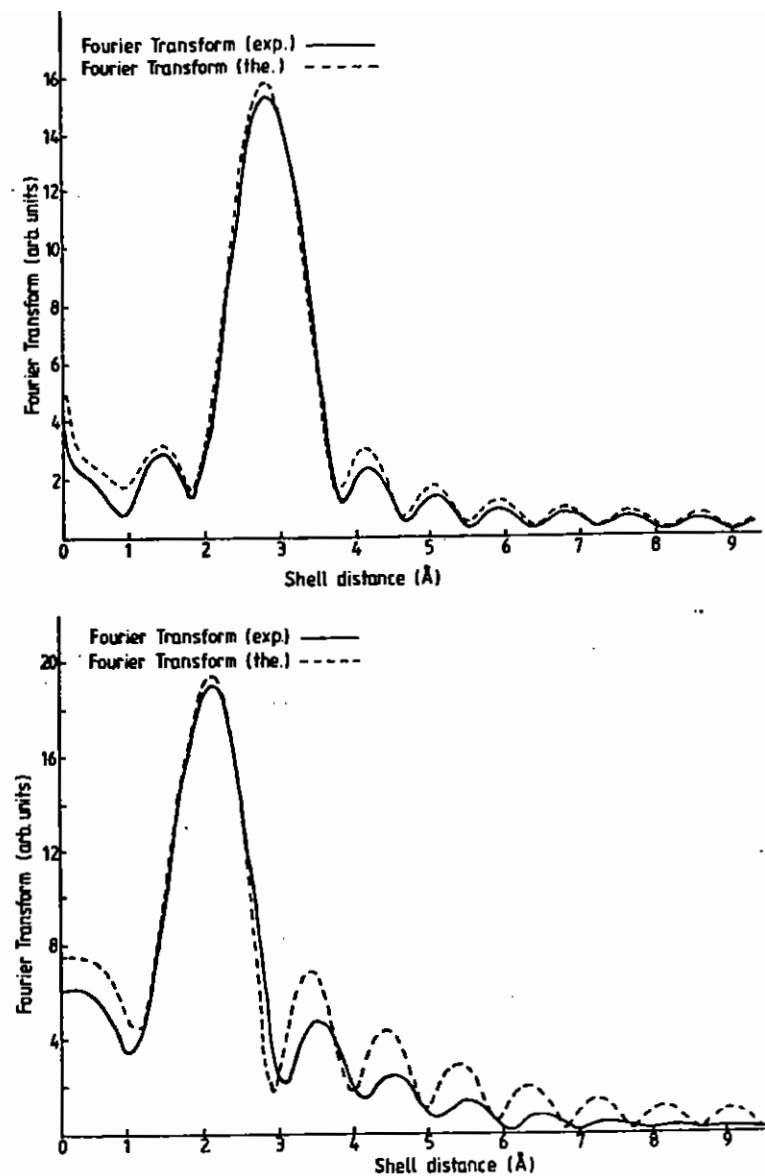


Fig. A3.4 The first shell fit of the fourier transforms (FT) for the timing 0.0 psec and + 200 psec respectively.

Ion Correlation Experiments

The experiments have been carried out using a similar geometry as the previous year [1], except that the targets were a planar sandwich of 1.8 μm of aluminium foil covered on both sides by 4 μm of E-Parylene placed about 8 mm from the uranium backlighting x-ray source. This separation is not sufficient to completely eliminate the x-ray heating effects at high backlighting energies, but was determined by mechanical constants within the target chamber. The planar colliding shock is generated in the sandwich from both sides by two frequency doubled 600 psec duration laser pulses from the VULCAN laser with energies approximately 35-45 J in each beam.

The shock driven beams were focused on a uniform spot of 1 mm diameter and the resulting irradiance is up to about $7 \times 10^{12} \text{ W cm}^{-2}$ on each side of the target.

The irradiated uranium (the backlighting target) produces a pulsed x-ray continuum in the spectral region of the aluminium K-absorption edge. The x-rays which pass through the shocked compressed region of the sandwich target are recorded on DEF x-ray film after being spectrally dispersed by a TIAP crystal mini-spectrometer placed about 40 mm from the backlighting source. As before, the second order was recorded on each laser shot separately from the first order spectrum by filtering out the first order on the same film. We successfully acquired EXAFS spectra on a total of 106 shots at various delay times between the main driving pulse (600 psec) and the backlighting pulse (70-100 psec). The quality of this data is considerably better than the results obtained last year.

Analysis of the Data

Figure 3 shows the measured values of the aluminium absorption coefficient as a function of energy at 0.0 psec (uncompressed aluminium) and at + 200 psec (compressed aluminium). Interpretation of the other data is still in progress and this will be carried out using the same way of analysis as been presented in ref⁽³⁾. Figure A3.4 shows the first shell fit of the Fourier transforms (FT) for the measured values of the aluminium absorption coefficient for the corresponding timing 0.0 psec and + 200 psec respectively. The application of a Fourier transform (FT) shows that the uncompressed aluminium (0.0 psec) peaks at $R_0 = 2.86 \text{ \AA}$ which is the interatomic

spacing of uncompressed aluminium whereas the (FT) application for the compressed aluminium (+ 200 psec) peaks at $R = 2.13 \text{ \AA}$.

From these values (R_0 , R), we estimated the value of compression as (2.42X) with the corresponding value of density to be as (6.53 g cm^{-3}). The calculated compression and density in these experiments are higher than the values previously obtained in the experiments carried out last year. The broken line in Figure A3.4 represented the theoretical first shell fitting of the Fourier transforms from the standard EXAFS code at the Daresbury Laboratory.

References

1. T A Hall et al, Section A3 Annual Report to the Laser Facility Committee, RAL-87-041.
2. T A Hall et al, (Submitted to Phys. Rev. Lett), also B A Shiwai et al SPIE - Los Angeles, Vol. 913, 10-17 JAN, 1988.
3. A Djaoui et al, (Submitted to Plasma Physics and Controlled Fusion).

A3-2 TWENTY-FOLD INCREASE IN THERMONUCLEAR REACTION YIELD IN LASER DRIVEN COMPRESSION

A J Cole, M Grande, M H Key, P T Rumsby, M Yamanaka* (RAL)
M J Lamb, E Robertson, S Saadat (Queens University Belfast)
A P Fews (University of Bristol)
P A Norreys (Royal Holloway and Bedford New College)
Y Kato, T Norimatsu, S Nakai, H Takabe, C Yamanaka (Institute of Laser Engineering, Osaka University, Japan)

*on leave from Osaka University.

1. Introduction

Research into Inertial Confinement Fusion has recently been concentrated on implosions of Large High Aspect Ratio Targets (LHART), the so-called stagnation-free mode⁽¹⁾⁻⁽³⁾. Record neutron yields of around 10^{13} have been achieved at Osaka University, Japan and at the Lawrence Livermore National Laboratory, USA. The LHART scheme

involves the spherical illumination of DT filled microballoons at high intensity ($3 \times 10^{14} \text{ Wcm}^{-2}$) by typically 12 beams or more.

A new experiment is reported that demonstrates the scaling behaviour of the fusion yield of LHART implosions with incident irradiance. In addition, a novel implosion mode (the Non-Uniformly irradiated High Aspect Ratio Target or NUHART mode) is presented. Here a non-uniform irradiation pattern was imposed on the shell where energy was concentrated in 12 dodecahedrally located focal spots.

It is shown that for the same average intensity on the target, a 20-fold increase in yield is obtained for the NUHART scheme. An explanation is provided for this increase in yield. By comparing results from various laboratories, previous experiments conducted with VULCAN's 12 beam irradiation system are understood.

2. Experimental Technique

In this experiment, the glass shells had aspect ratios of around 400. They were fabricated at the Institute of Laser Engineering, Osaka University, Japan. The gas fill was 5.5 atmospheres of deuterium which has a fusion reaction cross section which is two orders of magnitude smaller than deuterium-tritium reaction for the same ion temperature⁽⁴⁾.

The diagnostics included

- (a) three orthogonal pinhole cameras to observe the implosion symmetry
- (b) a time resolved x-ray pinhole camera to observe the implosion trajectory
- (c) CR39 nuclear track detectors to give measurements of the D-D reaction proton yield and, from the proton energy, the shell ρR and deuterium temperature⁽⁵⁾
- (d) a space resolving PET crystal spectrometer to give information on the silicon recombination continuum and fuel-shell mixing during the implosion

- (e) plasma calorimeters to measure the coupling of the laser energy to the target

- (f) a scintillator detector to monitor the yield of thermonuclear neutrons.

The x-ray pinhole cameras had four channels each with a filter of $10 \mu\text{m}$ of copper having a bandpass from 7 to 9 keV.

The magnifications of the pinhole cameras were $\times 1.6$, $\times 2.0$, and $\times 2.0$. With pinholes averaging $10 \mu\text{m}$, $17.5 \mu\text{m}$ and $20 \mu\text{m}$ for each camera, the cameras had spatial resolutions of 16, 25 and $30 \mu\text{m}$ respectively.

3. Experimental Results

Figure A3.5 illustrates the stagnation free mode. In low aspect ratio, ablatively driven targets neutrons are produced in two stages - from the initial shock wave convergence at the centre and from the final adiabatic compression. The Rayleigh-Taylor instability with a high mode number may occur during the deceleration phase of the implosion and mix the pusher and DT fuel, thus quenching the reaction in the adiabatic phase. To avoid this, neutron production can be concentrated in the initial stage. This is the stagnation - free mode.

In this stagnation free mode, the thin walled shell (or pusher) is driven by the intense laser irradiation. Most of the shell is ablated during the implosion and, as the shell is accelerated over a large distance, it consequently achieves a high implosion velocity of $\approx 10^8 \text{ cm}^{-1}$. By matching the leading edge of the gaussian laser pulse to the maximum acceleration phase of the shell, the shock waves generated at different times by the accelerating shell collapse simultaneously at the centre, generating the required temperatures for thermonuclear fusion. The neutrons are produced as the reflected shock wave hits the shell at point Y_N^* in Figure A3.5.

Figure A3.6 shows four shots where the D/R value was set at + 5, + 6.5, + 8 and + 10. The best uniformity was found to be at + 10, as was expected with the results in section A2.7. However, the D-D proton fusion yield was small ($Y_p = 5 \times 10^3$) for D/R = + 8, indicating poor

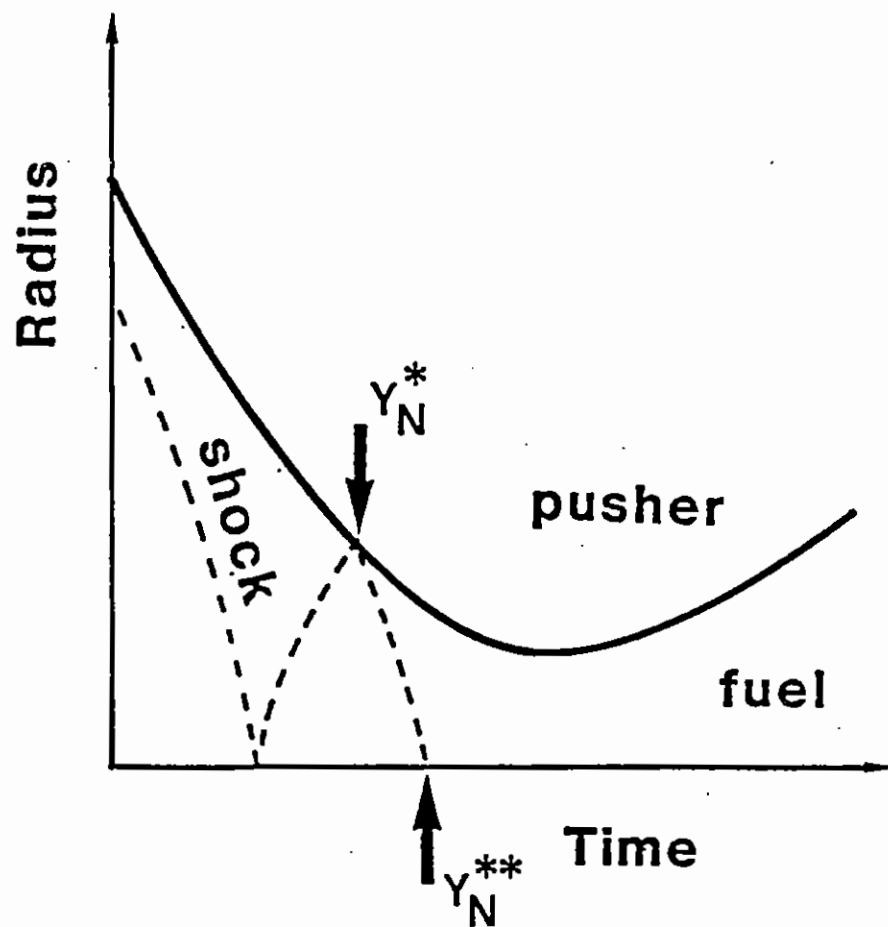


Fig. A3.5 Schematic of LHART implosion.

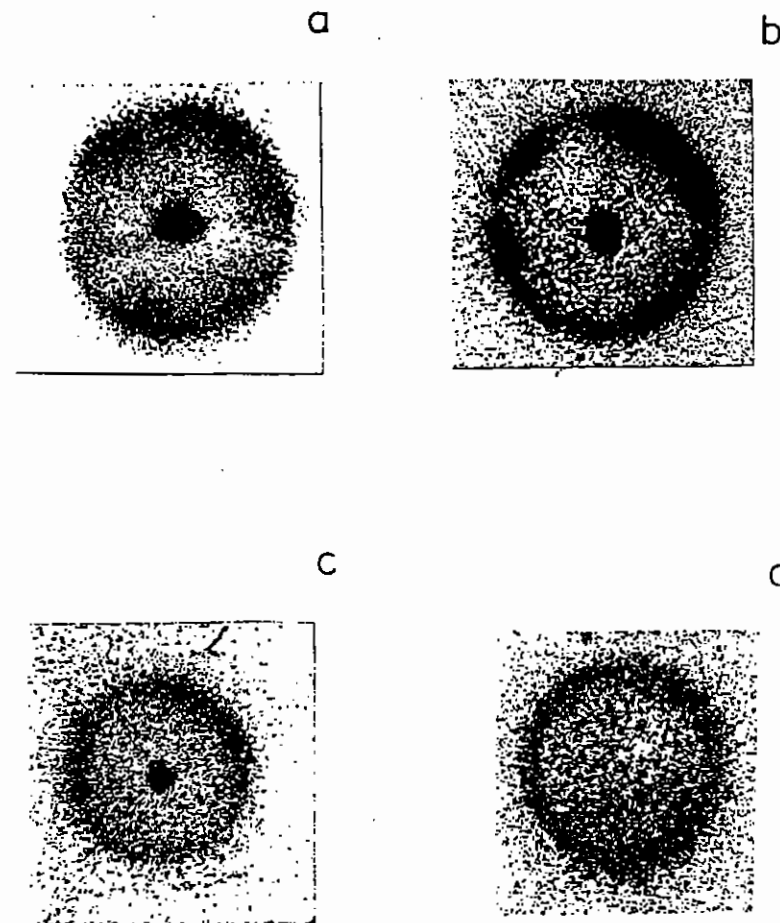


Fig. A3.6 X-ray pinhole picture of imploded glass microballoons with D/R set at
 (a) + 5
 (b) + 6.5
 (c) + 8
 (d) + 10.

energy - target coupling. The $D/R = +5$ case showed obvious non-uniformities and so $D/R = 6.5$ was chosen as the best compromise value.

Table 1 summarises the LHART shots that were observed. The highest proton yield shot was 1.5×10^7 corresponding to over 1×10^9 for an equivalent D-T filled target. The target had a diameter of $349 \mu\text{m}$ and a wall thickness of $0.45 \mu\text{m}$ ($R/\Delta R = 388$) and was irradiated with 330 Joules in 600 psecs.

Figure A3.7 shows a one dimensional computer simulation of this shot from the ILESTA-BG code⁽⁶⁾. Figure A3.7(b) shows the peak compression phase in some detail. The code predicts a proton yield of 3.0×10^7 at Y_N^* , which is only a factor of 2 higher than the observed yield.

For the NUHART mode, the D/R value was reduced to $D/R = +1$, giving a dodecahedral pattern of locally intense non-uniform illumination as illustrated in Figure A3.8(a). Figure A3.8(b) shows an x-ray pinhole camera image for a target of $649 \mu\text{m}$ diameter and $0.87 \mu\text{m}$ wall thickness. The incident laser energy was 371 J in 600 psecs. The locally intense irradiation of the glass shell is evidenced by circa $100 \mu\text{m}$ diameter spots of intense 7 - 9 keV x-ray emission in the expected pattern. A small region of low brightness can be seen in the centre of the x-ray image. The low brightness region is attributed to low brightness of the deuterium relative to the glass shell. The proton yield was 2.5×10^7 .

Figure A3.9 shows proton energy loss measurements taken from the CR39 detector. They indicate an ion temperature of 2.5 keV and $p\Delta R$ of the pusher at the time of neutron emission of $5.8 \times 10^{-4} \text{ g cm}^{-2}$.

Figure A3.10 shows a streaked pinhole picture of a NUHART implosion in 1.5 keV X-ray emission. It shows the initial X-ray emission when laser irradiation begins and the radial collapse of the shell to a brightly emitting compressed core.

Table II summarises the NUHART implosion results. The fusion yields from the LHART and NUHART schemes can be compared by normalising the yield by volume and confinement time, i.e. by R^4 to eliminate the first order dependence of yield on the target size⁽⁷⁾.

The same procedure allows us to compare in Figure A3.11 the LHART implosions observed at RAL, X, with those obtained at ILE, Osaka⁽⁸⁾ provided the factor of 100 for the DD relative to DT reaction rates are taken into account. NUHART results are marked by ∇ . Figure A3.11 shows the normalised proton yield plotted against incident irradiance averaged over the target surface. $\bar{I} = P/4\pi r^2$. This shows clearly that there is a 20-fold increase in fusion yield when the intensity is non-uniform and locally higher and in addition emphasises the irradiance dependence of the yield.

4 Discussion

4.1 Normalised Yield scaling with Incident Irradiance.

The normalised yield scales with incident irradiance but is also dependent on the initial aspect ratio of the shell. In Figure A3.11 the normalised yield is plotted against incident irradiance for 460, 360 and 220 aspect ratio targets⁽⁹⁾.

For interest, we have also plotted the normalised yields obtained in a previous experiment at RAL⁽¹⁰⁾ where the aspect ratio of the targets was 60. An experiment conducted at ILE, Osaka where random phase illumination was used to implode LHART shells of 200 aspect ratio is also included⁽¹¹⁾.

The normalised yields for 3w irradiation of LHART shells obtained at LLE, Rochester are also plotted in Figure A3.11⁽¹²⁾. Finally 4w implosions from Palaiseau, France are included⁽¹³⁾. Each yield was taken to be 100 times lower for comparison with DD reactions.

An interesting picture emerges from these results.

1. The normalised yield dependences for LHART implosions from various labs have the same scaling with incident irradiance, $Y \propto I_{\text{inc}}^{4.8}$. This is true for different laser wavelengths and for targets with different aspect ratios.
2. Increasing the aspect ratio of the targets sharply increases the fusion yield. This means that lower yields obtained from previous experiments at RAL were not anomalous or due to a pre-pulse in the laser system or to errors in beam timing⁽¹¹⁾.

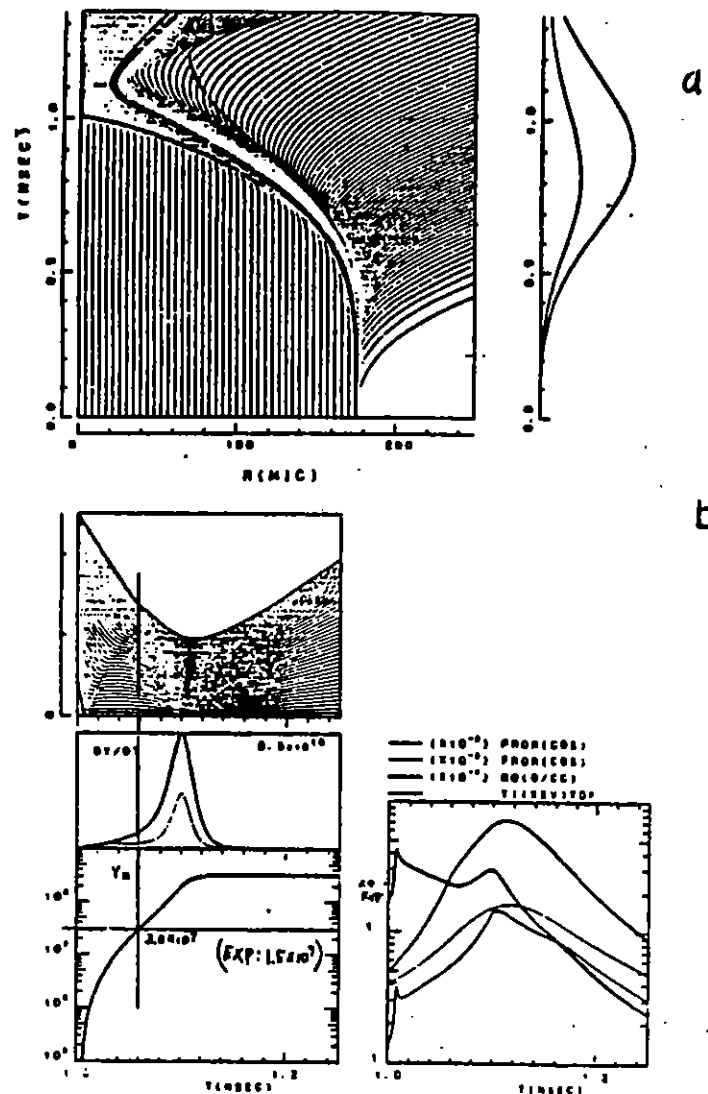


Fig. A3.7 (a) ILESTA-GB simulation of highest yield LHART shot.
(b) Detailed study of peak compression.

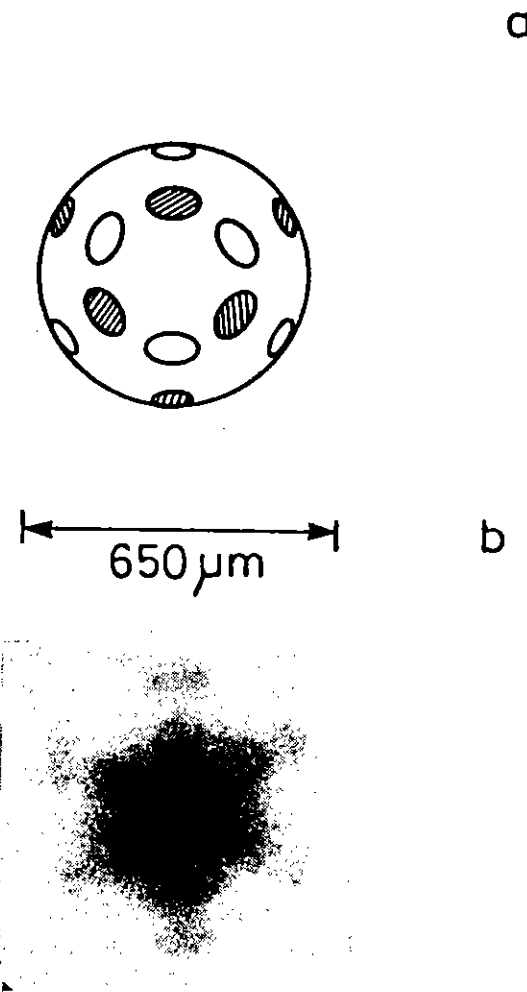


Fig. A3.8 (a) NUHART method of illumination.
(b) X-ray pinhole camera image obtained from NUHART mode.

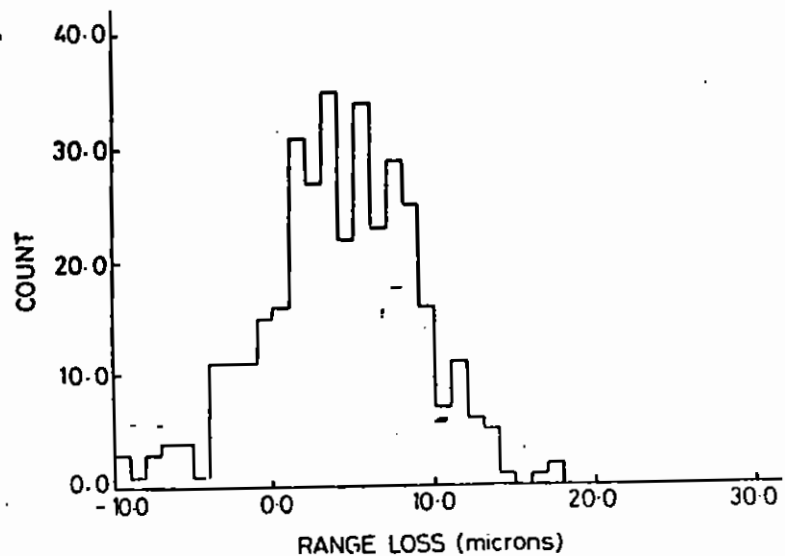


Fig. A3.9 Proton energy loss measurements taken from the CR39 detector.

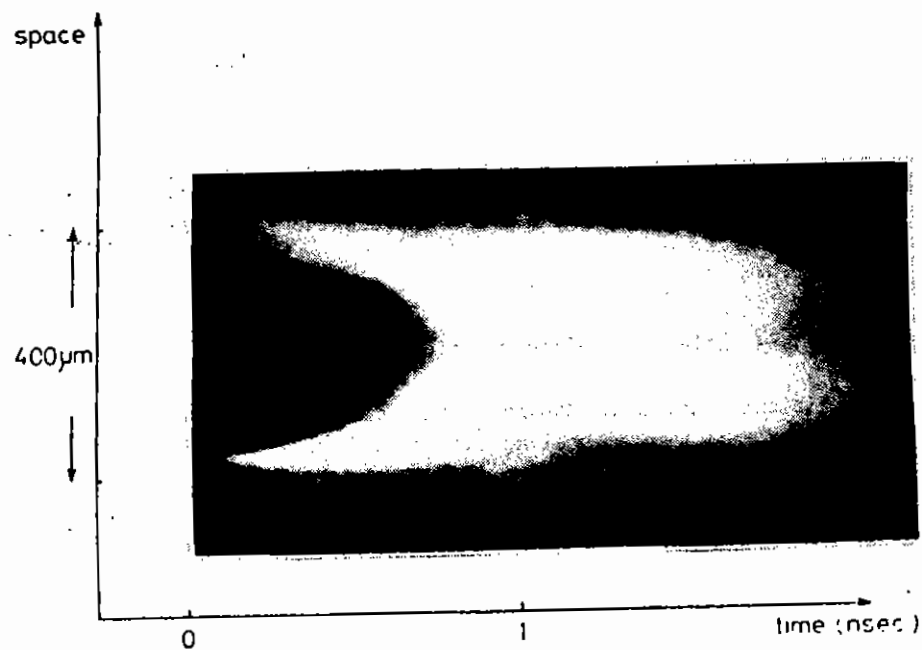


Fig. A3.10 Streaked pinhole picture of a NUHART implosion in 1.5 keV x-ray emission.

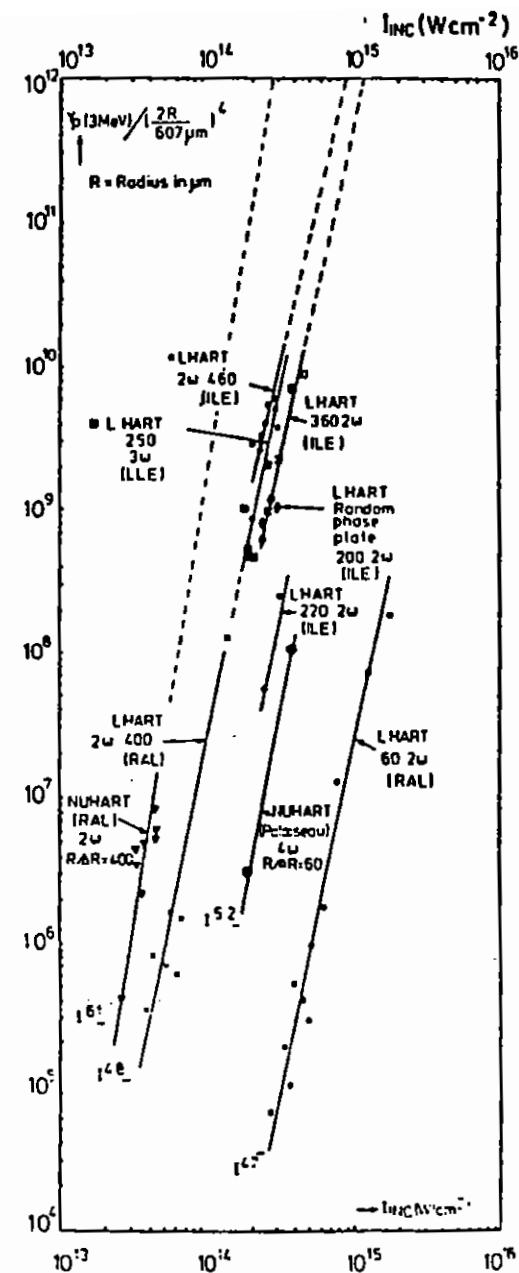


Fig. A3.11 Normalised proton yields against incident irradiance from data from various laboratories.

3. When random phase illumination is applied to 250 aspect ratio LHART shells, the fusion yield is increased by an order of magnitude. This has been attributed better symmetry of implosion allowing the reflected shock wave to reconverge to the point Y_N^{**} in Figure A3.5. It has to be verified experimentally whether increasing the aspect ratio of the targets to 460 will further increase the yield, but this result is very encouraging and is of major significance to ICF.
4. There is a two order of magnitude improvement in yield when the incident laser wavelength is reduced from green ($0.53 \mu\text{m}$) to blue ($0.35 \mu\text{m}$) for 250 aspect ratio LHART implosions. This could be due to either the increased drive pressure of the shorter laser wavelength, the increased absorption or due to the improved symmetry of the 24-beam Omega laser system. In any case, an improvement in yield could be expected for higher aspect ratio target implosions.
5. Figure A3.12 shows that there is a significant improvement in yield with when aspect ratio targets of 60 are driven by 4w light in the NUHART mode even with the 6-beam symmetry of the Palaiseau laser.
6. There is a distinct dependence of yield on the target radius and this can be further illustrated with reference to Figures A3.12 and A3.13. Here, the yield has been normalised to a particular laser power (Gekko XII and VULCAN) and plotted against the initial radius of the targets for results from ILE, Osaka and from the present experiment. The laser power normalisation for the RAL data is an average of the NUHART and LHART results. Both graphs indicate that there is a yield dependence on target radius $Y_p \propto R^{-4}$.

4.2 A Model for NUHART Implosions

Discussion of results

In Figure A3.11 the results for different aspect ratio $r/\Delta r$ scale similarity with irradiance and all the LHART data can be fitted by the scaling relationship

$$Y \sim I^{4.9 \pm 0.3} r^{4.5} \left(\frac{R}{\Delta R}\right)^{5.5} \quad (1)$$

There is also an increase of 20 times for the yield of NUHART implosions relative to yield for the same target and laser power with uniform irradiation which is shown by plotting the NUHART data at their irradiance $P_L/4\pi r^2$.

Insight into these facts can be obtained with a simple model. Fusion reactions in ablatively driven targets are known to occur mainly in heating and compression due to a spherical shock wave, before any deceleration of the imploding shell.

For a given density of gas fill the shock produces a constant compressed density irrespective of its strength. The fusion reaction yield therefore depends only on the temperature, reacting mass, $M \sim r^3$, and the confinement time $\tau \sim r$

$$\text{i.e. } Y \sim r^4 f(T) \quad (2)$$

The temperature due to shock heating in a spherical shock has a well known self similar solution for its time and space variation, but for the present purpose we note only that $T \sim v_s^2$ where v_s is the shock velocity for a given convergence ratio. The shock is driven by the imploding shell of velocity v_p . We therefore have $v_s \sim P_a r / \Delta r$ for ablation pressure P_a on the shell. The irradiance determines the ablation pressure and a suitable approximation is $P_a \sim I^{0.8}$ at $0.53 \mu\text{m}$.

The dependence of reaction rate on temperature for $1 \leq kT \leq 5 \text{ keV}$ is roughly of the form $f(T) \sim T^5$ so that we can express the yield as

$$Y \sim (r/\Delta r)^5 I^4 r^4 \quad (3)$$

using the previously explained scaling relationships.

There is sufficient agreement between the scaling exponents in equation(2) and the experimental scaling exponents from Figure A3.11 to suggest that this model is useful.

It can be extended to treat the NUHART implosions by assuming that only those conical sections of the shell and gas fill beneath the irradiated spots participate in creating the fusion reaction. The irradiance in those spots is then $I' = \epsilon I$ where ϵ is the enhancement factor. The

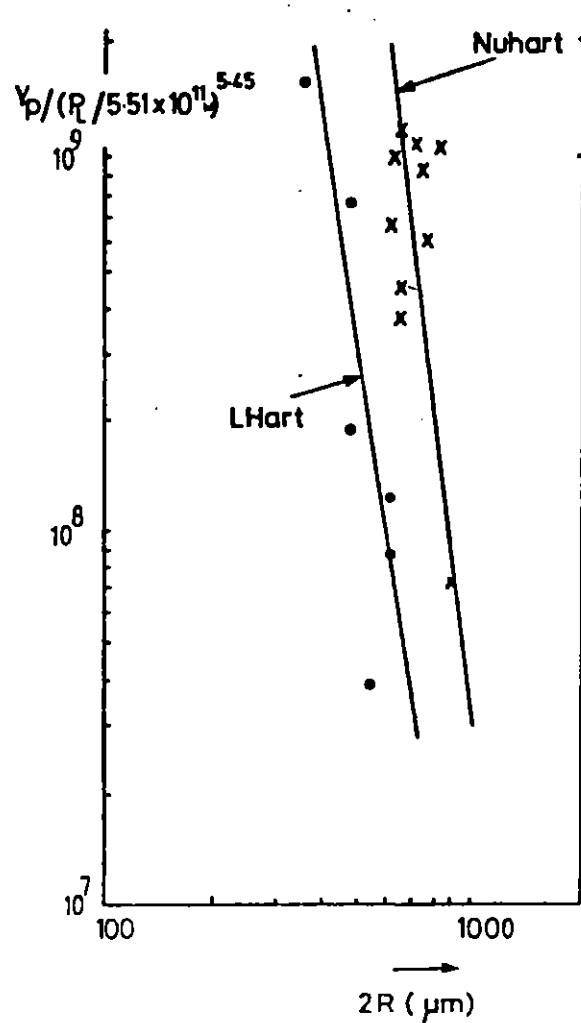


Fig. A3.12 D-D proton yield normalised by laser power plotted against target radius for NUHART and LHART results.

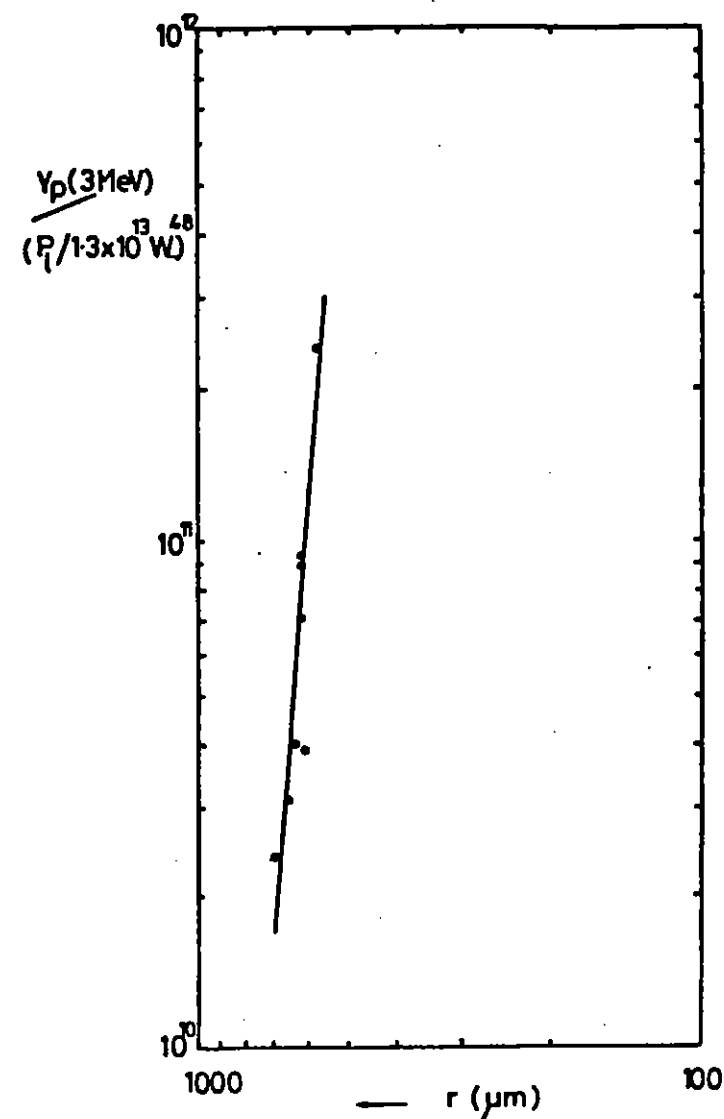


Fig. A3.13 D-D proton yield normalised by laser power plotted against target radius for ILE Osaka data.

effective mass of the gas is $M' \sim e^{-1} M \sim e^{-1} r^3$ and the effective inertial confinement time scales as $M'^{-1/3}$.

With these assumptions

$$Y \sim e^{2/3} (r/\Delta r)^5 I^4 r^4 \quad (4)$$

In assessing the enhancement ratio e it is relevant to consider lateral spreading of absorbed energy which gives a larger effective focal spot. If the area of the regions of intense X-ray emission associated with a spot are estimated from the data, such as Figure A3.8, we conclude that $e \sim 3$.

Equation (4) then suggests a yield enhancement of 19 times for targets operating at low temperatures with T^5 scaling of $f(T)$, in good agreement with the experimental results in Figure A3.11.

There is an implicit assumption that the heated spots will each launch shock waves which will successfully combine to converge on part of the mass. A recent discussion of polyhedral shock waves which suggests that the final convergence can achieve better sphericity than the initial launch is of some interest in this context⁽¹⁴⁾. This is shown in Figure A3.14.

The model is helpful in attempting to draw conclusions on the usefulness of NUHART implosions. It is clear that neither LHART or NUHART implosions represent a way to achieve high gain fusion reactions for ICF, since high compressed density is not obtainable in either way, but is essential for high gain.

The processes are both only of interest in maximising neutron and fusion yield where a powerful fusion source is required. Applications of such sources include diagnostic developments in ICF research and use of pulsed sources of fusion products for time resolved measurements on transient events⁽¹⁵⁾.

One practical advantage of the NUHART scheme arises in maximising neutron yield with lasers of sub terawatt and sub kilojoule output. The highest yield is obtained with a target aspect ratio of 400:1⁽¹⁾ but shells with a wall thickness less than about 0.6 μm have proved unobtainable so far, making LHART targets for small lasers unavailable.

Larger targets of aspect ratio 400 can however be manufactured and used in the NUHART mode to maximise yield with smaller lasers.

It is also interesting to consider whether an increase in total neutron yield could be obtained with very large laser systems which have so far produced 'record' yields with implosions of the LHART type. In this context we note that maximum yield has been obtained for optimised $I_0 \sim 3 \times 10^{14} \text{ Wcm}^2$ at an aspect ratio $r/\Delta r \sim 400$ close to shell burnthrough. If we note that the mass ablation rate scales as $I^{0.8}$, it is easy to see that the burn fraction in the implosion time scales as $(r/\Delta r)^{0.5} I^{0.1}$ and is therefore constant for constant I and $r/\Delta r$. An optimised NUHART implosion should therefore have $I' \sim eI = I_0$ and $r/\Delta r \sim 400$ with a larger target radius $r_n \sim e^{1/2} r_l$. The yield ratio $Y_n:Y_l$ for the two types of implosions is then

$$\frac{Y_n}{Y_l} = \frac{M_n^{4/3}}{M_l^{4/3}} \frac{f(T_n)}{f(T_l)} = e^{2/3} \quad (5)$$

noting that $T_n = T_l$ under the specified optimised conditions.

A two fold increase in yield is therefore apparently possible for a given laser power assuming $e \sim 3$ from the previous discussion.

The origin of the predicted increase is in the longer implosion time $t_n/t_l \sim e^{1/2}$ giving more energy at the same power and in the increased reacting mass, $M_n/M_l \sim e^{1/2}$ while operating at optimised I and $r/\Delta r$. We have not tested exponentially the validity of this last prediction but plan to do so in the future.

Acknowledgments

Our thanks are due to the staff of the SERC Central Laser Facility and of the target fabrication laboratory at Osaka University for their assistance with this work.

References

- (1) C Yamanaka et al, Phys. Rev. Lett. 56 (15), 1575 (1986).

TABLE 1

Date	#	Diameter (μm)	Wall Thickness (μm)	$\frac{R}{\Delta R}$	$E_{\text{inc}}(2\omega)$ (J)	I psec	I_{inc} ($\times 10^{12} \text{Wcm}^{-2}$)	D/R	Y_p (3 MeV)	CR39		
										R μm	pR mg cm^{-2}	T_i (keV)
15/9/87	3	605	0.85	356	322	1000	2.8	+ 8	5×10^4	YIELD TOO LOW		
15/9/87	6	607	0.92	330	360	800	3.9	+ 5	2.9×10^4	32.0	6.83	-
15/9/87	9	479	0.69	347	182	600	4.2	+ 5	3.0×10^4	30.0	8.73	1.8
15/9/87	14	481	0.77	312	246	600	5.6	+ 6.5	5.8×10^4	27.6	10.98	2.0
16/9/87	2	539	0.87	310	272.3	800	3.73	+ 6.5	2.3×10^4	YIELD TOO LOW		
16/9/87	5	587	0.93	316	233	600	3.59	+ 6.5	1.7×10^4	YIELD TOO LOW		
16/9/87	7	597	0.75	398	239	600	5.05	+ 6.5	4.9×10^4	31.0	7.36	5.73
16/9/87	13	349	0.45	388	330	600	14.4	+ 6.5	1.5×10^4	30.5	8.04	2.41

TABLE 2

Date	#	Diameter (μm)	Wall Thickness (μm)	$\frac{R}{\Delta R}$	$E_{\text{inc}}(2\omega)$ (J)	I (psec)	I_{inc} ($\times 10^{12} \text{Wcm}^{-2}$)	Y_p (3 MeV)	Shell implosion velocity ($\times 10^8 \text{cm s}^{-1}$)	CR39		
										R (μm)	pR _{fuel} (mg cm^{-2})	fuel ion temp (keV)
17/9/87	10	619	1.0	310	327	600	4.4	9.2×10^4	2.46	32.0	6.75	2.07
17/9/87	14	631	0.75	421	268	600	3.57	2.1×10^4	-	31.0	7.55	1.73
18/9/87	4	645	0.84	384	370	600	4.62	8.4×10^4	3.3	31.0	7.81	1.82
18/9/87	6	639	0.87	367	375	600	4.7	2.5×10^4	2.2	32.0	6.60	3.86
18/9/87	10	652	0.85	383	375	600	4.55	7.4×10^4	-	32.0	6.90	1.93
18/9/87	12	695	1.21	365	398	600	2.6	2.1×10^4	-	31.0	7.79	1.82
20/9/87	1	697	1.10	317	298	600	3.15	6.3×10^4	1.8	-	-	-
20/9/87	2	649	0.94	345	405	600	4.97	2.8×10^4	2.7	34.0	4.80	2.56
20/9/87	3	720	0.9	400	321.0	600	3.22	9.2×10^4	-	33.0	5.46	3.87
20/9/87	4	659	0.94	350	332.0	600	3.92	6.2×10^4	-	34.0	4.30	4.74

- (2) M C Richardson et al, Phys. Rev. Lett. 56 (19), 2048 (1986).
- (3) E M Campbell et al, Rev. Sci. Instr. 2. 57, 2101 (1986).
- (4) J J Duderstadt, G A Moses, Inertial Confinement Fusion, John Wiley, New York (1982).
- (5) A P Fews, D L Henshaw, Nucl. Instr. Methods., 197, 517 (1982).
- (6) H Takabe et al, Phys. Fluids (in press) (1988).
- (7) C Yamanaka et al, Nature 319, 757 (1986).
- (8) C Yamanaka et al, IAEA Conf Proc IAEA-CN-47/B-I-4, Kyoto, Japan (1986).
- (9) C Yamanaka et al, Nucl Fusion 27(1), 19 (1987).
- (10) P M Evans, A P Fews, W T Toner, A J Cole, Annual Report to the LFC, Rutherford Appleton Laboratory Report RAL-86-046, A2-5 (1986).
- (11) Y Kato, H Takabe, H Nishimura, T Jitsuno, H Shiraga, M Nakai, K Kondo, K Shimada, Y Hirose, S Urushihara, Institute of Laser Engineering Quarterly Report, ILE-QPR-87-21, 6 (1987).
- (12) J Wark (private communication).
- (13) J Briand, P M Evans, E Fabre, P Fews, A Michard, A Poauesse, J Virmont, Groupement de Recherches Coordonnees Interaction Laser Matiere, Ecole Polytechnique - 91128 Palaiseau, Rapport d'activite 1986, 103.
- (14) D W Schwendeman, G B Whitman, Proc. of the Royal Soc A 413, 297 (1987).
- (15) P M Evans, A P Fews, A J Cole, C Edwards, C J Hooker, D Pepler, W T Toner, J Wark, Ann. Rep. to the LFC, Rutherford Appleton Laboratory Report RAL-85-047, A2.21 (1985).

A3-3 PRODUCTION AND DIAGNOSIS OF COALESCING SHOCKS.

D Riley, M Givon, O Willi (Imperial College, London).

The production and diagnosis of coalescing shocks has two motivations. Firstly the feasibility of laser fusion schemes depends on the use of shaped laser pulses to achieve a high adiabatic compression of the target. Secondly the production of plasmas at pressures of 10's of Mbars and temperatures of only a few electronvolts is of theoretical interest, as interesting correlation and degeneracy effects take place. These effects can be studied by observing the K-edge of a particular element within the target.

The technique of K-absorption spectroscopy has been used previously for experiments with single and colliding shocks. During these experiments, Chlorinated targets were illuminated with X-rays from a thin layer of Bismuth which was deposited on the target and heated by the shock driving beams. Preliminary experiments with coalescing shocks have been performed in Target Area East. The cluster beam facility was used to produce shaped laser pulses with which to drive two coalescing shocks into target. The targets consisted of a central layer of Chlorinated Parylene sandwiched between two outer layers of N-Parylene. A thin layer of Bismuth, (150 - 600 nm), was coated onto the rear surface, and was irradiated by a separate short pulse beam of 70 ps duration. The backlighter beam could be timed so as to sample the compression at different times relative to the shock driving beams. This arrangement avoided the problem of radiative pre-heating by Bi X-rays, and so allowed a higher compression. The K-absorption edge of the Chlorine was observed by using a Germanium (III) Bragg crystal which was coupled to an X-ray streak camera, used in static mode. The main experimental difficulties encountered were due to a poor signal to noise ratio. However, preliminary data was obtained, in which a red-shift of the K-edge of approximately 5 eV was observed. Computer modelling of the shock compression with the code, MEDUSA indicated that this shift corresponded to a compression of the target to three times the initial density. A further experiment is scheduled to take place in the summer of 1988, in which several improvements will be made, including the use of a gated X-ray intensifier. This instrument will be used to obtain data with a greatly enhanced signal to noise ratio.

EXPERIMENTAL LAYOUT

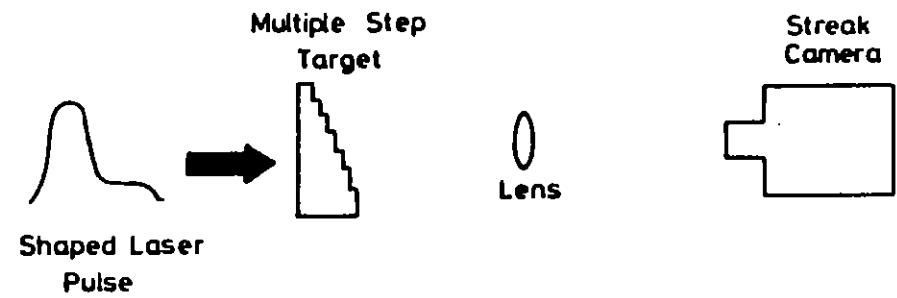


Fig. A3.15 Experimental layout.

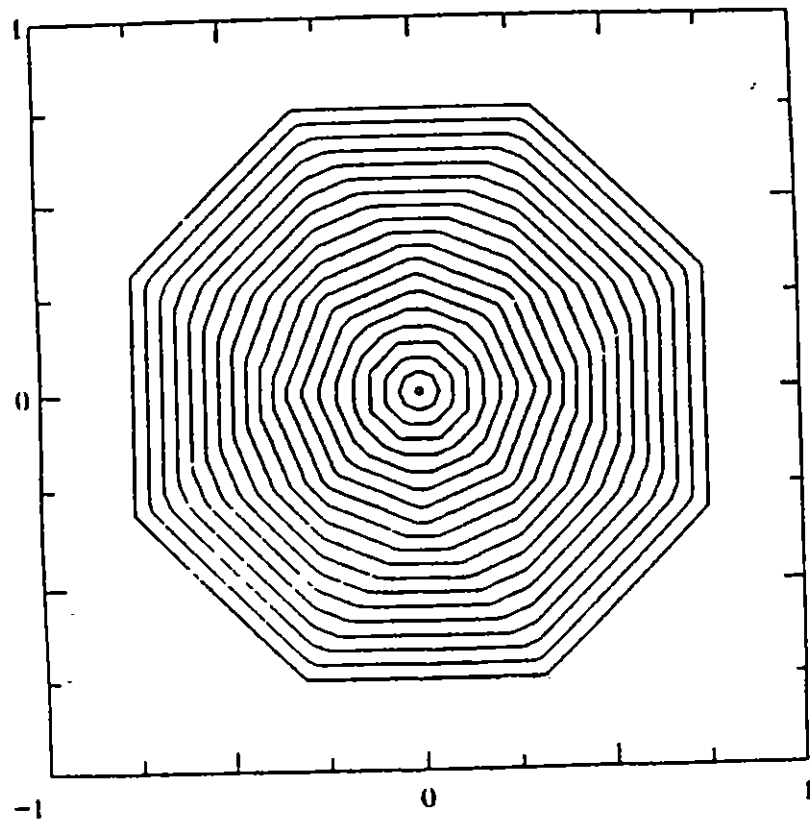


Fig. A3.14 Computer simulation of planar shock waves launched in a pentagonal symmetry after ref (15).

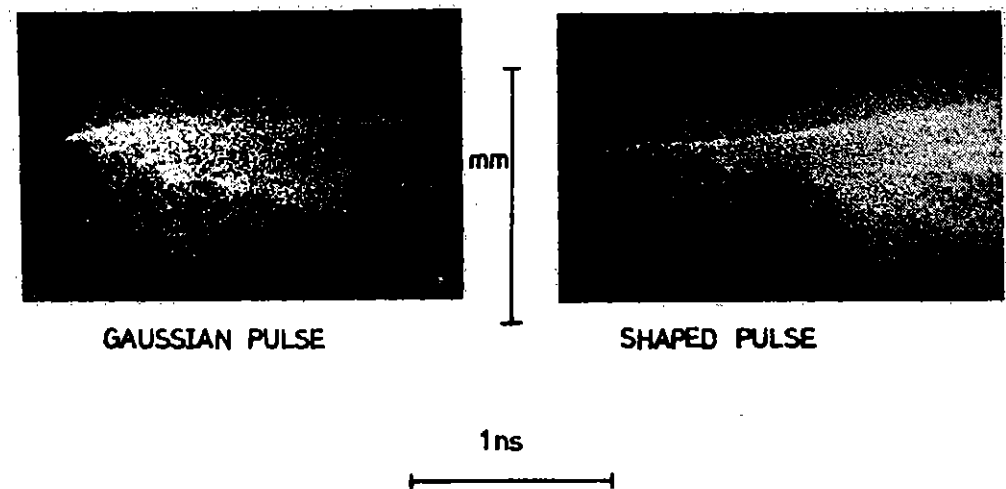


Fig. A3.16 Shock breakout for gaussian and shaped laser pulse.

A3-4 STUDY OF COALESCING SHOCKS

S E Coe, O Willi (Imperial College), T Afshar-Rad (RAL)

Introduction

Shock coalescence is an essential of laser fusion schemes for the adiabatic compression of the fuel pellet⁽¹⁾. An experiment has recently been carried out in TAE giving the first direct experimental demonstration of the coalescence of two shocks generated by a shaped laser pulse.

When a laser induced shock reaches the rear surface of a planar target it causes material to be spalled out. This heated material then emits light as a black body and the turn on of this emission indicates when the shock reaches the rear surface of the target. Hence optical streak photography of the rear surface of the target can be used to measure the transit time of the shock through the target. By using a target with a step (of known thickness) on it, it is possible to determine the shock velocity by the delay of the shock propagation through the step. This technique has previously been demonstrated to be successful with a single step⁽²⁻³⁾. Here we have extended this technique by using multiple⁽⁶⁻⁷⁾ step targets. Although this allows a much better determination of the velocity of a single shock to be made the main purpose of this experiment was to study the coalescence of a smaller shock with a delayed larger shock in the target.

Experimental set up

The experimental set up used is shown in Figure A3.15. The shaped laser pulse was generated by using six beams of the Vulcan glass laser system brought together to an overlapping focus by the cluster beam facility in TAE. By bringing one of the beams early in time by 1200 ps relative to the other beams an intensity ratio of 1:6 was produced between the front and main laser pulse. Second harmonic, 800 ps laser pulses were used.

The multiple step targets used were made of Aluminium and typically had 7 steps about 5 μm thick and 50 μm wide.

The emission from the rear surface of the target was imaged via two f/2 microscope objectives onto the slit of the optical streak camera (S20 photocathode) with an overall magnification of about 15 times. Optical coloured glass filters giving a transmission in the blue region of the spectrum were used to prevent any scattered green laser light from reaching the streak camera slit.

Experimental Results

The optical emission observed for a single shock (generated by a gaussian laser pulse) is shown in Figure A3.16(a). Shock breakout can clearly be seen from six consecutive steps. The delay between the steps gives a shock velocity of $4.0 \times 10^6 \text{ cm s}^{-1}$ which for an irradiance of $4.4 \times 10^{14} \text{ w cm}^{-2}$ is in good agreement with MEDUSA simulations. This corresponds to a shock pressure of 24 Mbar.

A typical breakout observed for a shaped laser pulse is shown in Figure A3.16(b). The breakout from the first two steps is due to the first shock and from the remaining four steps due to the second shock after coalescence has occurred. Coalescence occurs at point (a) and corresponds to a target depth of 40 μm . The sharper slope (corresponding to a larger velocity) for the breakout of the second stronger can clearly be seen, when compared to the shallower slope for the breakout of the first shock.

Conclusion

An experiment has been recently carried out to study the coalescence of two laser induced shocks. Initial analysis of the results obtained has clearly demonstrated the occurrence of shock coalescence. A full analysis, including computer simulations, of the data obtained is currently in progress.

References

1. Nuckolls, Wood, Thiessen and Zimmerman, Nature, 239, 139 (1972).
2. Vesser and Solem, Phys Rev Lett, 40, 1391 (1978).
3. Trainor et al, Phys Rev Lett, 42, 1154 (1979).

A4	THEORY AND COMPUTATION	Pages
A4-1	Nonlocal Effects and the Raman Instability	93 - 94
A4-2	Modelling the RAL Beat-Wave Experiment with Nemesis	94 - 98
A4-3	Fokker-Planck Simulations of Electron Heat Flow	98 - 101
A4-4	Developing a 3D Hydrocode: Choosing a Numerical Scheme	101 - 104
A4-5	Thermodynamic Fluctuations in the Average Atom Model in Dense Plasma	100 - 104
A4-6	Spherical Band Structure Calculation at Arbitrary Density and Moderate Temperatures	104 - 109
A4-7	Fusion Neutron and Alpha Particle Transport in Inertial Confinement Fusion Targets	109 - 110
A4-8	Improvements in the Modelling of Recombination X-ray Lasers	110 - 112
A4-9	Modelling of Li-Like and Na-Like Recombination and Ni-Like Collisionally-Pumped X-ray Lasers	112 - 116

Editor: S J Rose

A4-1 NONLOCAL EFFECTS AND THE RAMAN INSTABILITY

H C Barr, T J M Boyd and G A Coutts (University of Wales, Bangor)

Given the persistence of both threshold and spectral anomalies in Raman data⁽¹⁾, it seems appropriate to re-examine some of the assumptions underlying the standard theory of stimulated Raman scattering. Here we examine a nonlocal Raman model which allows for coupling between back- and forward-scatter and shows that an absolute instability can occur for all scattered frequencies. In particular in a monotonic density profile these assumptions have categorized Raman scattering as occurring in two distinct regimes. Below the quarter-critical density surface $n_c/4$, where n_c is the critical density for the incident laser light, a WKB analysis shows that for a linear density profile, only convective amplification is possible for both back- and forward-scatter which are regarded as decoupled. This is the SRS-C regime. The conventional "threshold" is usually taken to be $(v_0/c)^2 k_0 L > 1$ where $2v_0$ is the quiver velocity of the electrons in the laser electric field, k_0 is the wavenumber of the incident light and L the density scale length. This so-called threshold is of course arbitrary to a degree in that it is chosen to produce an amplification of $\exp(2\pi)$ over noise levels. This arbitrariness has led to suggestions that the amplification factor could be relaxed to diminish the discrepancy between theory and experiment.

Near the quarter-critical density, back- and forward-scattering resonances become degenerate. At the scattered wave cut-off, WKB analysis breaks down but by neglecting plasma wave propagation ($v_e \rightarrow 0$), a second-order is retrieved from which absolutely unstable eigenstates are obtained with a threshold⁽²⁾ $(v_0/c)^2 (k_0 L)^{4/3} > 1$. This is the SRS-A regime.

It has been common practice to regard back- and forward-scatter as independent processes when in reality they are nothing of the kind, as Koch and Williams emphasized some years ago.⁽³⁾ For laser light propagating in a plasma with a monotonically increasing density, the plasma wave amplified in generating backscattered light will propagate up the density gradient to the forward-scatter resonance. The separation in density of the back- and forward-scatter resonances is very small, being of the order of $(v_e/c)^2$ where v_e is the electron

thermal velocity. Thus unless the temperature is high enough and consequently the plasma wave damping strong enough for the wave to decay between the two resonances, the forward propagating plasma wave will act as an enhanced noise source for generating forward scattered light. If in turn the plasma is overdense to this light, it will reflect at some higher density and propagate back to the backscatter resonance point in the density profile where it will again be subject to amplification. These nonlocal effects create a feedback loop leading to temporal growth of the coupled back and forward scatter instabilities in the manner described by Koch and Williams⁽³⁾ in 1984. Their approach is essentially an amalgam of local analyses of the two resonance regions with a WKB treatment of waves propagating between them, resulting in a global description.

By contrast the model used in this work is global in its entirety. We solve the full coupled wave equations for both plasma and scattered light waves, encompassing back and forward scatter resonances with wave reflection. We write the one-dimensional equations corresponding to the back and forward scatter of laser light normally incident on an arbitrary density profile $n(x)$, corresponding to a plasma frequency $\omega_p(x)$, as

$$\left[\frac{\partial^2}{\partial t^2} + \omega_p^2(x) - \frac{\partial^2}{\partial x^2} \right] v_s = v_0 \frac{\partial E_p}{\partial x} \quad (1)$$

$$\left[\frac{\partial^2}{\partial t^2} + \omega_p^2(x) - 3v_e^2 \frac{\partial^2}{\partial x^2} \right] E_p = -\omega_p^2(x) \frac{\partial}{\partial x} (v_0 v_s) \quad (2)$$

where v_s , v_0 define the electron quiver velocity in the field of the scattered and incident light waves respectively. E_p is the plasma wave electric field and all quantities are normalized to c and ω_0 . In this model, damping terms are not included. We solve (1) and (2) by means of a code which treats the global SRS problem as opposed to resorting to a WKB treatment, except for the laser field. The equations are Laplace-transformed in time and only resonant terms retained to yield

- (a) temporally growing (i.e. absolutely unstable) eigenstates of the homogeneous SRS equations if these exist or, if this is not the case,
- (b) reflectivities (convective amplifications) from the nonhomogeneous equations with a noise source retained.

The Raman equations are solved in an inhomogeneous plasma slab, $0 \leq x \leq l$ with boundary conditions chosen to continuously match fields in the homogeneous plasma extending for $x < 0$ and $x > l$. The interaction region is assumed infinite, though only resonant locally within the region $0 < x < l$. The driven equations are solved analytically within the homogeneous plasma $x < 0$, $x > l$ to provide four independent solutions. Of these in (a) above, only the two representing outward propagating waves are chosen in each region. In the case of convective amplification a variety of options for incoming waves is available to represent noise sources.

The code has been run to obtain temporally growing eigenstates in the linear density profile $n(x) = n(l/2) + (x-l/2)n_c/L$ as a function of scalelength or maximum density, with reflections of forward propagating waves included or excluded as the case may be. In the first instance, to test the dependence on the feedback mechanism, the code was run with variable maximum density but with the scalelength held constant. This effectively controls the reflectivity of the forward scattered light. Figure A4.1 plots the growth rates of several eigenstates vs. maximum density for a plasma for which $L = 80\lambda_0$, $T_e = 1.7$ keV and $v_0 = 0.1c$. To the right of the dashed line $\omega_s = \omega_{p \text{ max}}$, growth is a maximum when forward scattered light is totally reflected, the addition of overdense plasma at the right-hand boundary does not affect the physics. If we reduce the reflectivity of forward scattered light, the growth rate diminishes so that ultimately temporal growth is quenched and a convective instability ensues.

The code has been used to determine Raman characteristics for a range of plasma parameters. In particular we have allowed the density scale length to range from $L = 40\lambda_0$ to $L = 320\lambda_0$. Figure A4.2 shows the growth rate plotted against the scattered frequency for a range of scalelengths up to $240\lambda_0$. This shows that the growth rate appears to be, if not independent, at least insensitive to scalelength well above threshold. The line spacings are approximately proportional to L^{-1} for ω_s away from $\omega_0/2$. We have also examined the growth rate as a function of v_0 . For the first eigenvalue corresponding to emission near $\omega_0/2$, growth scales linearly with v_0 . At higher scattered frequencies a nonlinear dependence, approximately proportional to v^2 , is observed.

At these frequencies $\omega_s > \omega_0/2$, the analysis of Koch and Williams⁽³⁾ is valid and agrees with our results. All of these results correspond to cases in which total reflection of forward scattered light is assumed.

In Figure A4.3 we plot the minimum thresholds (occurring when feedback is a maximum, i.e. total reflection) versus scattered wave frequency. We see from this that the SRS-A threshold is reproduced correctly, confirming that plasma wave propagation is unimportant in this case. More significantly we see that for $\omega_s > \omega_0/2$ the absolute instability threshold is lower by a factor of about 4 than the SRS-C threshold, determined for an amplification of $e^{2\pi}$. Allowing transmission of forward scattered light quickly raises thresholds for absolute instability.

The relative thresholds are more weakly dependent on ω_s than previously believed to be the case. This effect is even more marked for an exponential density profile when thresholds are almost constant as a function of frequency. We have compared our results with thresholds evaluated on the basis of the Koch-Williams analysis and found good agreement.

References

1. K Tanaka, et al. Phys. Rev. Lett. **48**, 1179 (1982); R E Turner, et al. Phys. Fluids **26**, 579 (1983); H Figueroa, et al. Phys. Fluids **27**, 1887 (1984), C L Shephard, et al. Phys. Fluids **29**, 583, (1986).
2. C S Liu, M N Rosenbluth and R B White, Phys. Fluids **17**, 1211 (1974).
3. P Koch and E A Williams, Phys. Fluids **27**, 2346 (1984).

A4-2 MODELLING THE RAL BEAT-WAVE EXPERIMENT WITH NEMESIS

P Gibbon and A R Bell (Imperial College)

Introduction

Previous theoretical work on the beat-wave accelerator concept⁽¹⁾ has concentrated on determining the saturation amplitude of the plasma wave excited by the beating of two laser pumps^(2,3). To model an

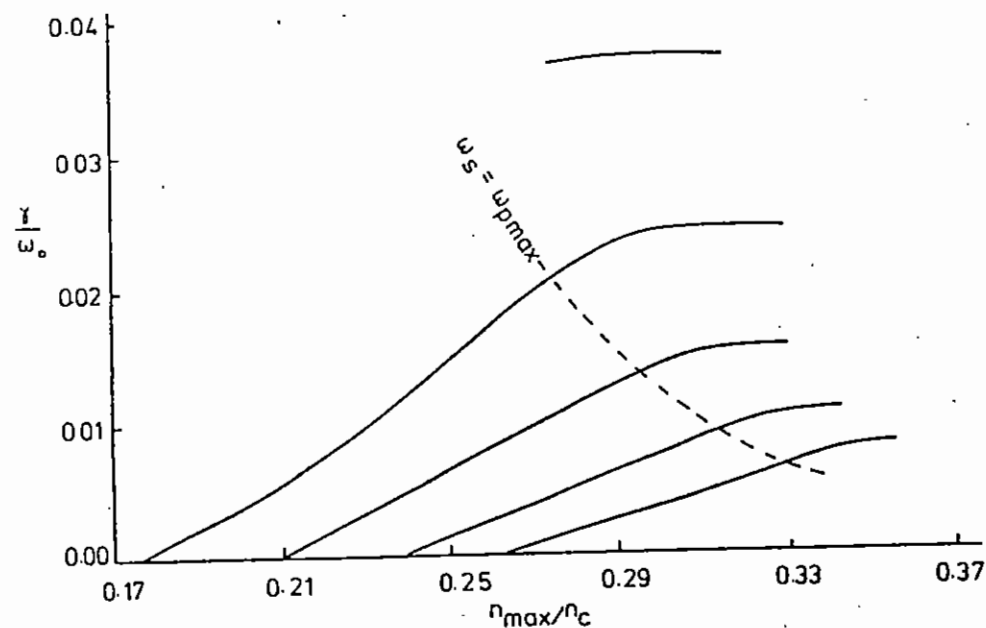


Fig. A4.1 Growth rates for several eigenstates plotted against maximum plasma density. Forward scattered light is totally reflected to the right of the dashed line; away from it on the left, forward scattered light is increasingly transmitted ($L = 80 \lambda_0$, $v_e = 0.01 c$, $v_0 = 0.1c$).

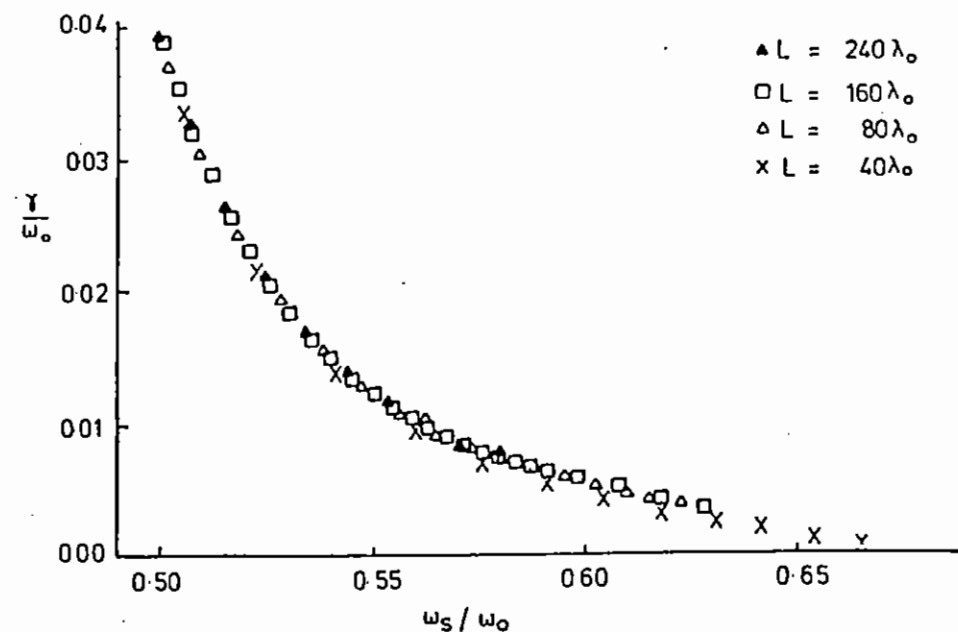


Fig. A4.2 Growth rates versus scattered frequency for scale lengths $L = 40 \lambda_0 - 240 \lambda_0$. It shows the growth rate to be independent of scale length ($v_e = 0.01c$, $v_0 = 0.1c$).

experiment realistically however, we must take into account electromagnetic cascading and the finite spatial extent of the laser pulse. Particle simulations⁽⁴⁾ have been useful in investigating qualitative aspects of these effects, but are limited to modelling plasmas with size $100 c/\omega_p \times 100 c/\omega_p$, or 2 mm x 2 mm in the case of the RAL experiment. Since the plasma to be used is roughly 1 cm long, and the pulse many times longer still, a particle simulation of the latter would clearly be beyond the limit of present-day computing power.

At Imperial College we have developed a 2D wave-envelope code (NEMESIS - Nonlinear ElectroMagnetic ElectroStatic Initial value envelope Solver) to study the beat-wave equations over a full acceleration stage length. For typical RAL parameters, we can model plasmas with dimensions $3 \times 10^4 c/\omega_p$ by $200 c/\omega_p$, or 0.5 m x 3 mm. This is made possible by formulating the envelope equations in a window moving with the laser pulse, so that the effective length of plasma on the computational grid is only a few times the pulse length.

We consider two collinear electromagnetic waves $A(\omega_0, k_0)$ and $A(\omega_1, k_1)$ propagating through a plasma slab and which satisfy the matching conditions $\omega = \omega_1 - \omega_0 = \omega_p$ and $k_1 - k_0 = k_p$. Starting from the relativistic Lorentz-Maxwell equations for a cold plasma in planar (z,x) geometry, and using the wave envelopes:

$$A(z, x, t) = \frac{1}{2} \sum_m [a_m(z, x, \tau) \exp\{i(k_m z - \omega_m t)\} + c.c.]$$

for the electromagnetic vector potential, and

$$E(z, x, \tau) = \frac{1}{2} [e(z, x, \tau) \exp\{i(k_p z - \omega t)\} + c.c.]$$

for the plasma electric field (where $\tau \gg \omega^{-1}$), we obtain the following equations for the electromagnetic cascade and plasmon respectively⁽⁵⁾:

$$\omega_m \frac{\partial a_m}{\partial \tau} - \frac{i}{2} \frac{\partial^2 a_m}{\partial x^2} = g^* a_{m+1} - g a_{m-1} \quad (1)$$

$$\frac{\partial e}{\partial z} + i \delta e - \frac{3i}{16} |e|^2 e + \frac{v_{eff}}{2} e = f \sum_m a_m a_{m-1}^* \quad (2)$$

We have used the normalisations $t \rightarrow \omega t$, $z \rightarrow \omega z/c$, $\omega_m \rightarrow \omega_m/\omega$, $v \rightarrow v/\omega$, $a = qA/mc$, $e = qE/m\omega c$, and we have defined the density detuning parameter $\delta \equiv (\omega_p^2/\omega^2 - 1)/2 = \frac{1}{2} \Delta n/n$ and $g = \frac{1}{2} \delta n/n = \frac{1}{2} (e_z - i \nabla_z e)$. The effective collision frequency v_{eff} is computed self-consistently within the plasma wave integration (equation 2), according to⁽⁶⁾:

$$v_{eff} = F v_{ei} \quad (3)$$

$$\text{where } F = [1 + \frac{v_{osc}^2}{6v_{th}^2}]^{-3/2}$$

The parameters to be used in the proposed RAL experiment are:

Laser wavelength	$\lambda_0 = 1.064 \mu\text{m}$, $\lambda_1 = 1.053 \mu\text{m}$
Intensity	$I_0 = 2 \times 10^{14} \text{ W cm}^{-2}$ (50 J per pump)
Pulse length	$\tau(\text{fwhm}) = 200 \text{ ps}$
Pulse width	$X(\text{fwhm}) = 330 \mu\text{m}$ ($3\lambda_0$)
	$\omega_p = 1.85 \times 10^{13} \text{ s}^{-1}$

The actual plasma length in normalised units is $600 c/\omega_p$, which is 25 times smaller than the minimum grid length required for the laser pulse. The chosen formulation of the equations requires that the plasma slab be propagated backwards at speed $= c$ through the grid, starting at the front of the pulse. This poses a problem in solving for the plasma wave at different times, since our method assumes that the laser pulse is always moving into fresh plasma. We avoid this problem by integrating equation (2) as if the plasma were present over the whole grid length, but then set $e = 0$ outside the plasma boundaries - see Figure A4.4

In the experiment, it is proposed to use of a probe beam at twice the pump frequency as a plasma wave diagnostic. The probe will scatter off the plasmon, generating sidebands according to equation (1). The numerical scheme for the probe beam in NEMESIS is the same as that used for the pump sidebands, but with frequencies $\omega_{probe(m)} = 2\omega_0 + m\omega$. The probe may also be initialised with a time delay relative to the centre of the pumps.

In this report we present some results for various plasma parameters. In particular, we investigate the effects of density detuning,

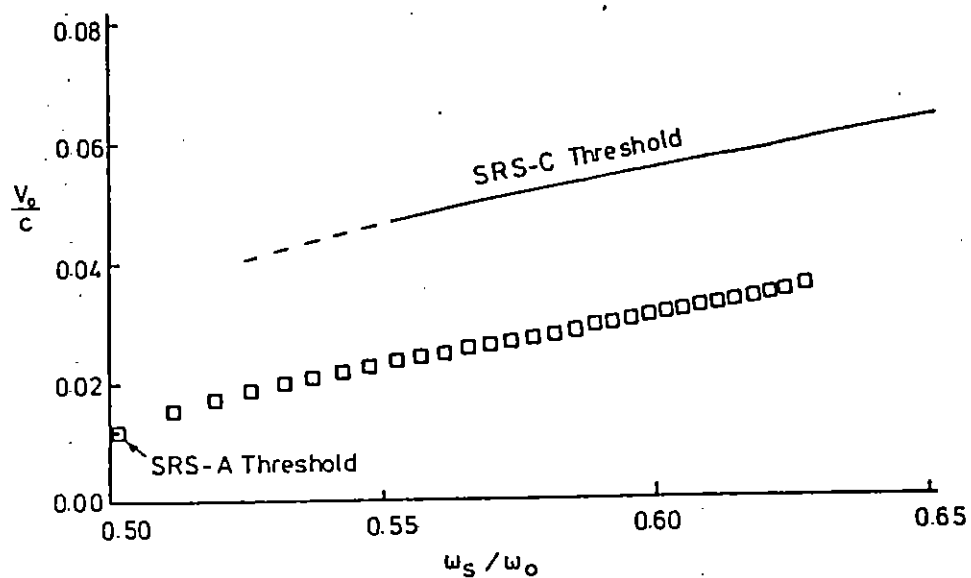


Fig. A4.3 Threshold v_0 versus scattered frequency are shown as squares. The first agrees with SRS-A theory which is valid near $\omega_0/2$. The solid line, valid away from $\omega_0/2$ is the SRS-C theory threshold ($L = 160 \lambda_0$, $v_e = 0.01c$).

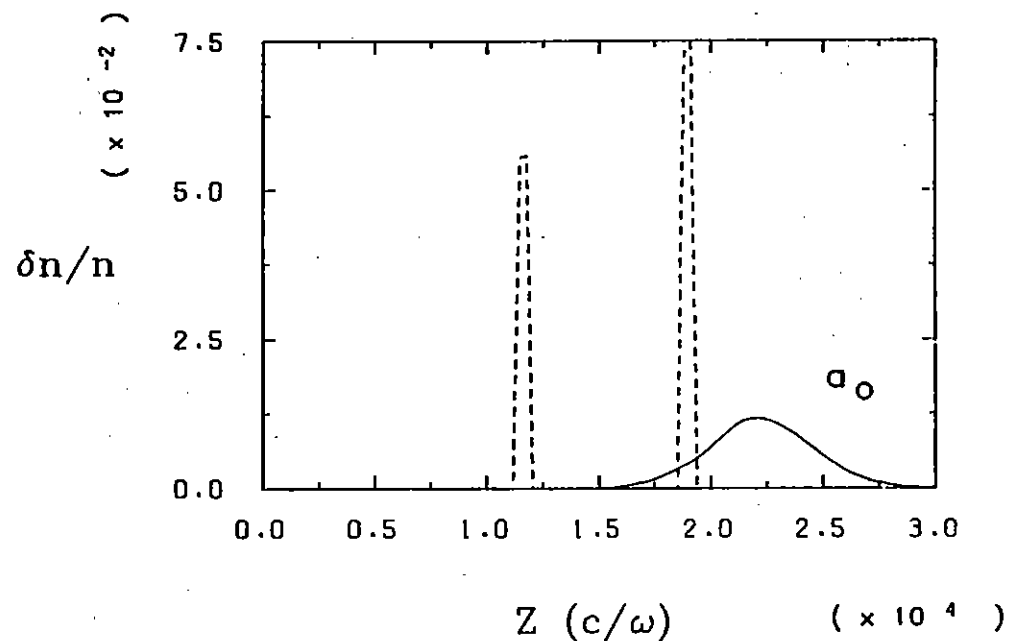


Fig. A4.4 Snapshots of plasma slab on axis ($x = 0$) moving from right to left in the laser pulse frame, at times 600 ps and 1000 ps respectively.

thermal damping, and the dependence of probe sideband intensity on the relative time delay.

Results

1. Sensitivity to detuning

Figure A4.5(a) shows the maximum amplitude on axis of a resonantly driven plasmon ($\Delta n/n = 0$). Likewise, the time-dependence of the total plasma wave energy normalised to the thermal energy $U = 3/2nkT$ is depicted in Figure A4.3(b). The sharp cutoff in the plasmon wake (Figure A4.5a) is characteristic of the form used for the damping coefficient ν_{eff} (equation 3). This effect is not as apparent in Figure A4.5(b) due to the averaging over the width of the plasma.

Figures A4.6(a) and A4.6(b) show the analogous plasmon amplitude and energy for a density mismatch $\Delta n/n = 1\%$. In this case the maximum plasmon amplitude $\delta n/n = 0.75$ (compared to 9% in Figure A4.5). This results in a much reduced plasma energy since the latter depends on $|e|^2$. There is also a noticeable reduction in the wake, which is almost entirely caused by driving the plasmon off-resonance. Increasing the temperature T_e to reduce damping has virtually no effect in this case. However, if the pulse were truncated at $t = 300$ ps, the plasmon amplitude would remain at the maximum wake value.

2. Effect of temperature on damping

Figure A4.7(a) shows the maximum plasmon amplitude on axis for different electron temperatures. The density mismatch in this case is 0.5% and the pulse length is 100 ps, which results in a sufficiently large wake for damping to be significant. The damping time increases with temperature, but is also sensitive to the initial wake amplitude. Thus, it is possible in some cases to reverse this trend - as seen in Figure A4.7(b), which is for a resonant plasmon and pulse length of 200 ps.

3. Probe delay

The relative intensities of the first probe sideband I_{-1}/I_0 are shown in Figure A4.8 for the resonant case ($\Delta n/n = 0$); and for $\Delta n/n = 1\%$. In the latter case, the plasmon amplitude starts to fall rapidly after 300 ps (Figure A4.6(a)), which is reflected in the decrease in

I_{-1} with increasing probe delay. Thus, if a significant density mismatch exists, the probe should be synchronous with the pump in order to coincide with the plasmon build-up period.

In the resonant case, the sideband intensity initially increases as the delay is increased, even though the probe 'sees' less of the plasma wave. This phenomenon can be attributed to the relativistic phase shift of the plasmon in the build-up period (~ 200 ps to 200 ps in Figure A4.5), which can reduce the rate of energy transfer to the sidebands. A delayed probe will miss the build-up period and scatter only off the plasmon wake, where the energy transfer rate is unaffected by the detuning.

References

1. T Tajima and J M Dawson, Phys. Rev. Lett. 43, 267 (1979).
2. C J McKinstrie and D W Forslund, Phys. Fluids, 30, 904 (1987).
3. S J Karttunen and R R E Salomaa, Phys. Rev. Lett. 56, 604 (1986).
4. D W Forslund et al, Phys. Rev. Lett, 54, 558 (1985).
5. P Gibbon and A R Bell, submitted to Phys. Rev. Lett., (1988).
6. See for example, R J Faehl and N F Roderick, Phys. Fluids, 21, 793 (1978).

A4-3 FOKKER-PLANCK SIMULATIONS OF ELECTRON HEAT FLOW

I R G Williams, G J Rickard, E M Epperlein and A R Bell (Imperial College).

Improvements have been made to the ID code SPARK described in the 1987 RAL Report⁽¹⁾. A Lagrangian hydrodynamic scheme has been added to model cold ion flow, and the laser energy is absorbed via inverse bremsstrahlung in all cells below critical density. The formulation of Albritton⁽²⁾ has been used to calculate the energy deposition.

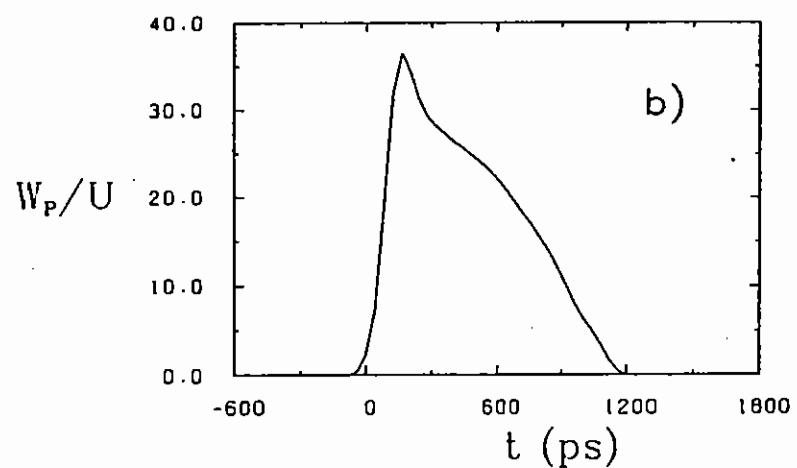
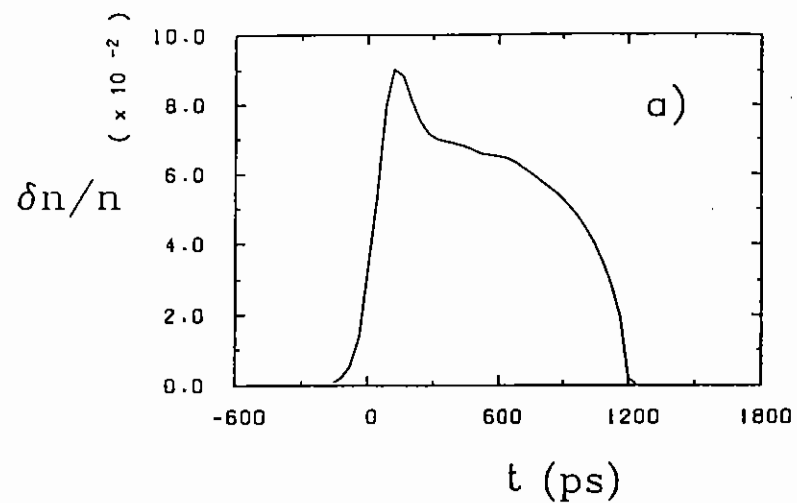


Fig. A4.5 a) Maximum plasmon amplitude and
b) total plasma wave energy as a function of
time relative to the centre of the laser
pumps for a resonantly driven plasma wave
($\Delta n/n = 0$).

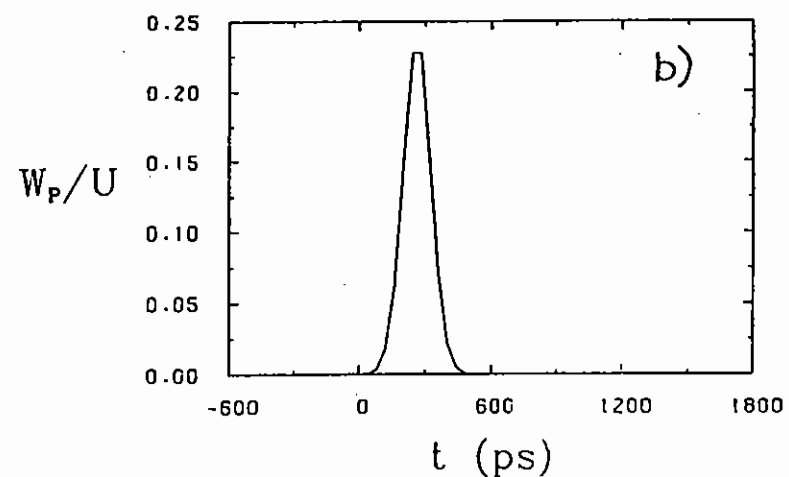
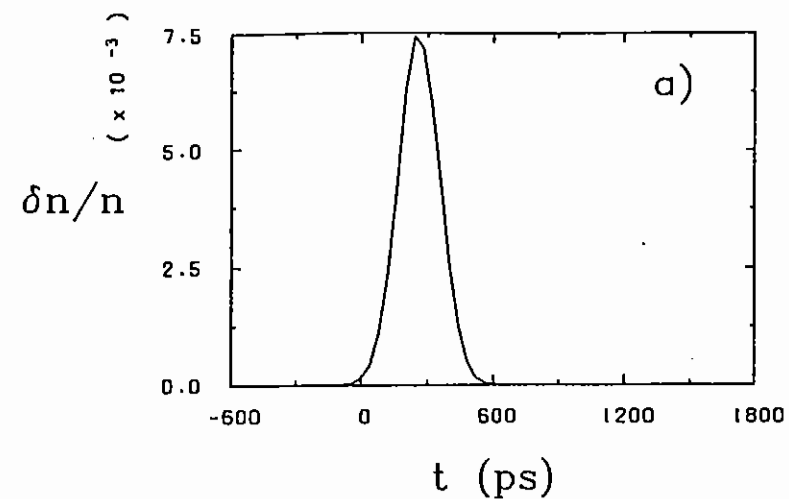


Fig. A4.6 As Figure A4.5, but with a density mismatch
 $\Delta n/n = 1\%$.

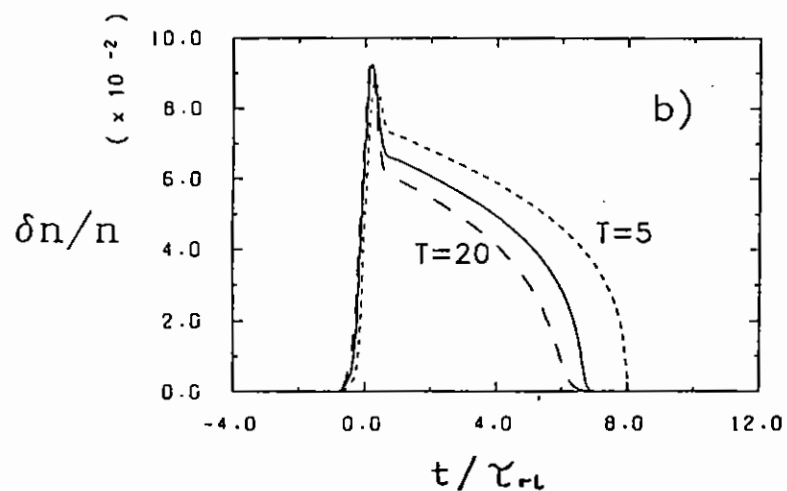
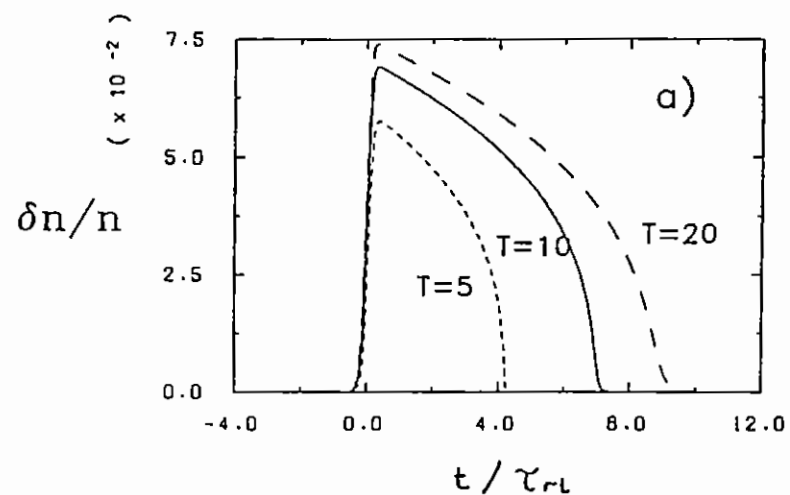


Fig. A4.7 Effect of temperature on damping time of plasmon wake for
a) $\tau_{fwhm} = 100$ ps, $\Delta n/n = 0.5\%$ and
b) $\tau_{fwhm} = 200$ ps, $\Delta n/n = 0$.
The time axis is normalised to the Rosenbluth-Liu saturation period, $\tau_{RL} = 300$ ps.

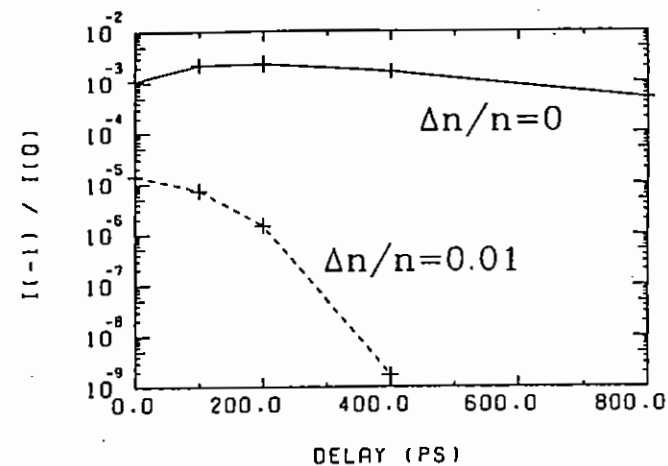


Fig. A4.8

Relative probe sideband intensity as a function of probe delay for mismatches $\Delta n/n = 0$ and $\Delta n/n = 1\%$. The curves correspond to the plasmon amplitude plots in Figures A4.5 and A4.6.

The code also has the ability to model multi-component targets, i.e. varying atomic number and density.

SPARK has been used to model a laser-target interaction with experimental parameters. A Gaussian pulse, with maximum intensity $1 \times 10^{15} \text{ W cm}^{-2}$ and full-width-half-maximum 70 psec, was incident on a cold plasma, temperature 100 eV, density $2.32 \times 10^{22} \text{ cm}^{-3}$, $Z = 4$. It was found when using the inverse bremsstrahlung heating that a large maximum velocity was necessary to avoid instabilities in f_z . We used 60 spatial and 60 velocity groups. Running time was approximately 6 CPU seconds for every 5 picoseconds of simulation on the CRAY-1S, with a timestep of 5×10^{-2} psec. The simulation was run for 55 psecs.

Figures A4.9 - A4.11 show the results of the simulation. A pressure wave is building up as the laser power peaks. There is a small temperature foot into the target. The Fokker-Planck heat flow is reduced well below the Spitzer value calculated for the same temperature profile, except in the dense material where the thermal foot penetrates.

References

1. E M Epperlein and A R Bell, Rutherford Laboratory Report RAL-87-041, 90 (1987).
2. J R Albritton, Phys. Rev. Lett., 50, 2078 (1983).

A4-4 DEVELOPING A 3D HYDROCODE: CHOOSING A NUMERICAL SCHEME

R P J Town and A R Bell (Imperial College)

Introduction

The Rayleigh-Taylor (RT) instability is a major obstacle to laser fusion. ICF targets are susceptible to the RT instability in both the acceleration and deceleration phases.

The major theoretical effort has been in the modelling of the acceleration phase using 2D hydrocodes⁽¹⁾. These have consistently shown a reduction in the growth rates below the classical value. The deceleration phase has been modelled by Roberts et al⁽²⁾, who have shown that even a small penetration of the shell into the DT fuel will severely inhibit the burn phase.

At Imperial College we are currently developing a 3D hydrocode in order to study the RT instability in the deceleration phase. Over the past year we have compared several schemes which solve the fluid equations.

Schemes Considered

FCT⁽³⁾ This consists of two difference schemes of different order blended together. One of the schemes is a very accurate high order scheme which is badly behaved near discontinuities; the other is a low order scheme which introduces large dispersion to ensure that the solution is singular at discontinuities.

Godunov⁽⁴⁾ The hydrodynamic flow is discretised into a series of constant states. These are allowed to interact (the time development being a solution of the Riemann problem). Finally the results are averaged conservatively.

FLIC⁽⁵⁾ This is a 1st order scheme. The algorithm is divided into two stages: a Lagrangian phase where the effect of pressure on the velocity and energy is calculated, and an advection phase where the fluid is transported using the Donor Cell method. An artificial viscous pressure is added near discontinuities to damp out oscillations in the solution.

Van Leer⁽⁶⁾ Using the basic FLIC algorithm the advection phase is modified to take into account the gradients of the transported quantities. This gives the scheme higher order accuracy.

Numerical Test

The simplest non-linear flow problem is the Riemann Shock Tube Problem of Sod⁽⁷⁾. This consists of two constant states separated by a discontinuity. This discontinuity resolves itself into 2 shock waves; 2 rarefaction waves, or a shock and rarefaction wave, one

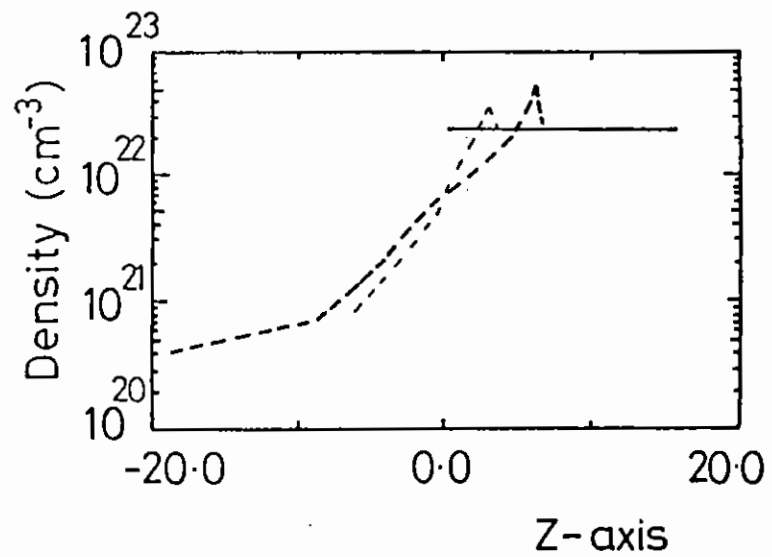


Fig. A4.9 Electron density against radius (in μm), at times $t = 55$ psec. (— · — · —), 27 psec (-----), and 0 (————).

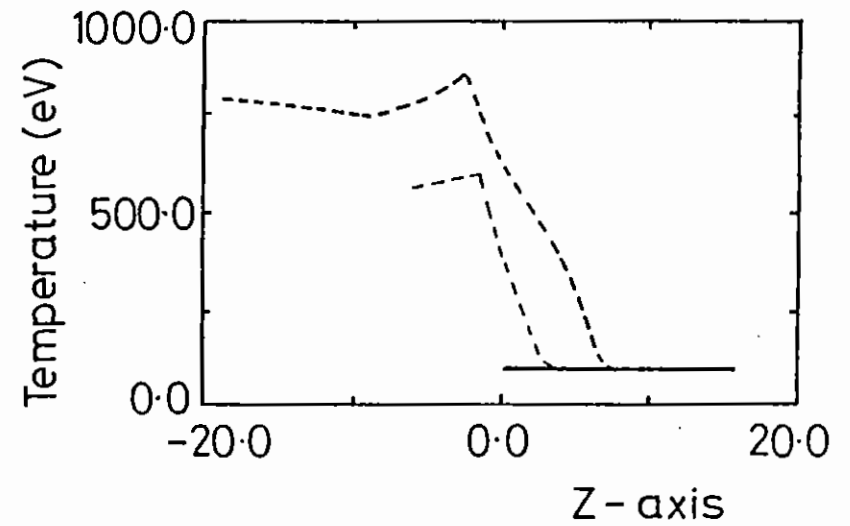


Fig. A4.10 Electron temperature against radius.

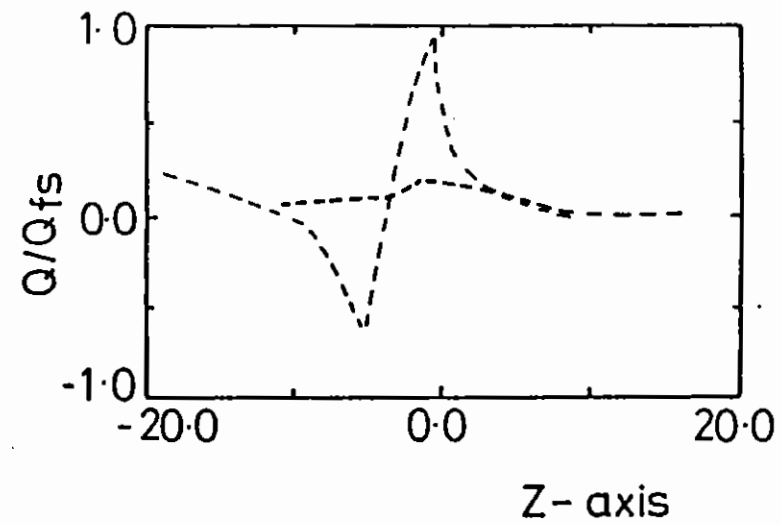
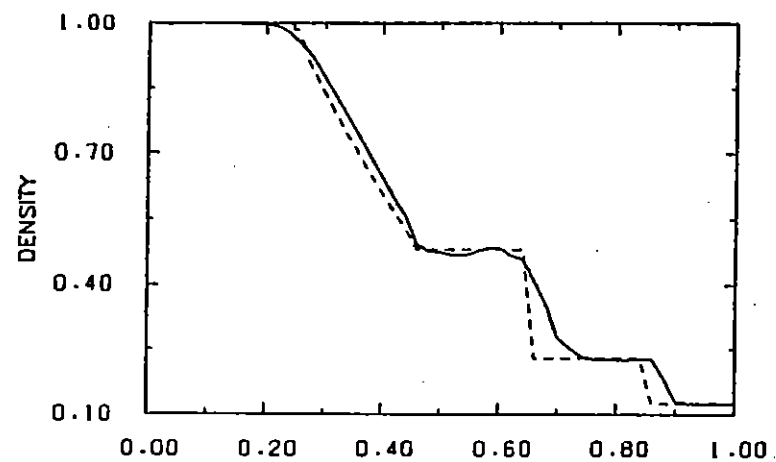
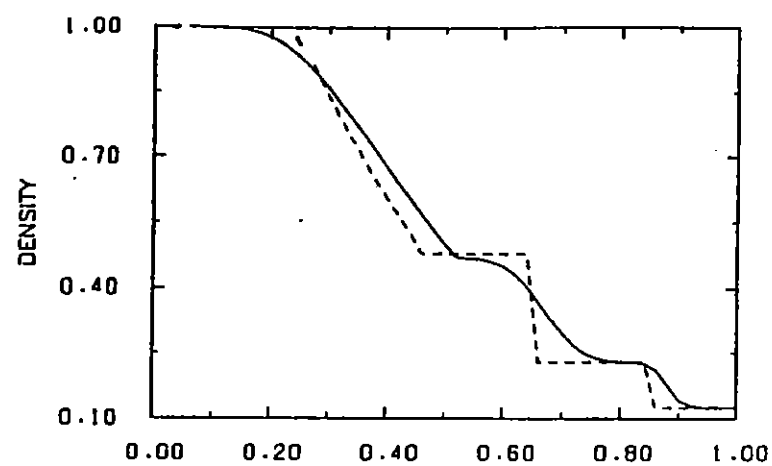


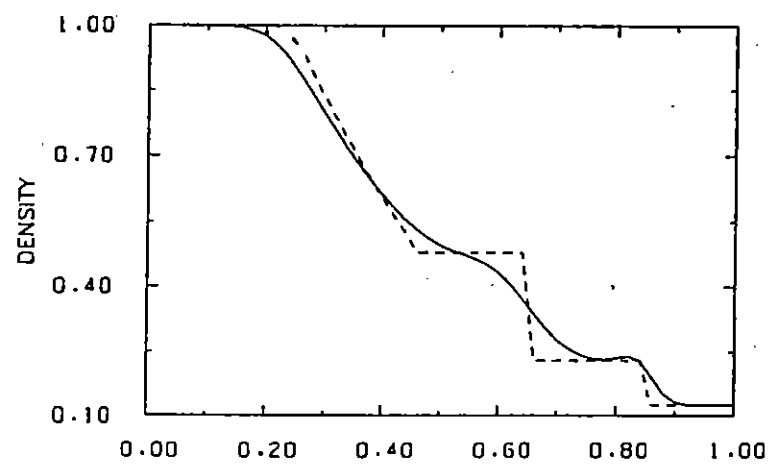
Fig. A4.11 Heat flows (normalised to free-streaming limit) against radius, at $t = 55$ psec.
 Fokker-Planck heat flow: ————
 Spitzer-Harm heat flow: - - - - -



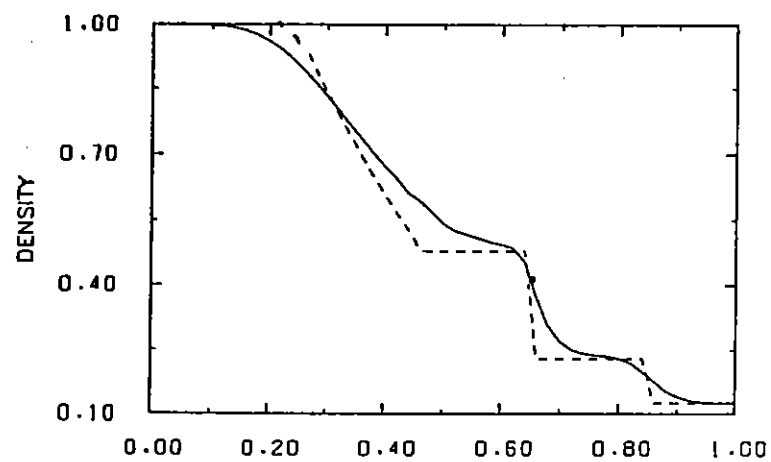
A) FCT.



B) GODUNOV.



C) FLIC.



D) VAN LEER.

Fig. A4.12 The density profiles at time = 0.2 for
a) FCT,
b) Godunov,
c) FLIC and
d) Van Leer.

travelling to the right the other to the left. Between these two waves is a middle state which in general contains a contact discontinuity. The problem is hence capable of testing the algorithms' ability to simulate the formation and development of:-

- (i) Shock Waves.
- (ii) Rarefaction Waves.
- (iii) Contact Discontinuities.

The initial configuration of the test problem is shown in Table A4.1.

Results and Conclusions

The density profiles at time = 0.2 is shown in Figure A4.12.

Godunov's scheme is complicated so is slow in executing. The benefit of higher accuracy for more computational effort is not apparent in this test.

FCT gives very good treatment of shocks and contact discontinuities. However in higher dimensions we and other authors⁽⁸⁾ have observed the solution taking on a staircase structure. Additionally we consider the unphysical movement of mass to be inappropriate for the intended application.

FLIC was fast executing, but tends to smear out the shock front and contact discontinuities too much. The Van Leer scheme sharpens the discontinuities with little additional effort. We are therefore currently using the Van Leer scheme in our 3D hydrocode.

Table A4.1 The Initial Conditions of the Riemann Shock Tube Problem of Sod⁽⁷⁾

<u>Left State S_l</u>	<u>Right State S_r</u>
Density $\rho_l = 1.0$	Density $\rho_r = 0.125$
Pressure $P_l = 1.0$	Pressure $P_r = 0.1$
Velocity $v_l = 0.0$	Velocity $v_r = 0.0$

References

1. C P Verdon et al, Phys. Fluids 25, 1653 (1982).
M H Emery et al, Phys. Rev. Lett. 57, 703 (1986).
R G Evans et al, Phys. Rev. Lett. 49, 1639 (1982).
M Henshaw et al, Plasma Phys. Cont. Fusion. 29, 405 (1986).
2. P D Roberts et al, J. Phys. D 13, 1957 (1980).
3. J P Boris and D L Book, J. Comp. Phys. 11, 38 (1973).
4. S K Godunov, Mat. Sb. 47, 271 (1959).
5. R A Gentry et al, J. Comp. Phys. 1, 87 (1966).
6. B Van Leer, J. Comput. Phys. 23, 276 (1977).
7. G A Sod, J. Comput. Phys. 27, 1 (1978).
8. P Woodward and P Colella, J. Comput. Phys. 54, 115 (1984).

A4-5 SPHERICAL BAND STRUCTURE CALCULATION AT ARBITRARY DENSITY AND MODERATE TEMPERATURES

A Djaoui, T A Hall, (University of Essex)
R C Albers, (Los Alamos National Laboratory)
S J Rose, (RAL)

Introduction

Progress in inertial confinement fusion is directly linked to our understanding of the physics of matter under compression and over a wide range of temperatures. It is well known that, under extreme conditions of temperature and pressure the Thomas-Fermi-Dirac⁽¹⁾ (TFD) statistical model provides a reasonably accurate electron potential. This is usually used in the single electron wave equation for the determination of electron states. At zero or very low temperatures,

band structure methods⁽²⁾ provide accurate electron potentials and density of states. There is, in addition to these two methods, a variety of models which have proved successful in different density-temperature regimes, depending on the physical quantity of interest (Lieberman⁽³⁾, Rozsnyai⁽⁴⁾). We present here another model which describes matter at arbitrary density and temperatures varying from zero to a few Rydbergs. This is intended for use in the calculation of electron potentials in laser shock-compressed plasmas⁽⁵⁾. This model can be seen as a bridge between zero temperature band-structure calculations and statistical models such as the ones of reference 1, 3 and 4.

The main simplifications of this model are as follows. First the polyhedral atomic cell is replaced with a spherical cell with the same volume. Finite matter density is taken into account by the splitting of upper electronic levels into bands but the density of states within each band is taken as constant. Hybridization of orbitals is also neglected. Semi-relativistic effects are taken into account by solving Dirac's equation but spin orbit coupling is neglected.

A closely related model is that of Rozsnyai⁽⁴⁾, but he treats free electrons in the TFD approximation. Here all electrons are treated in a band mode and Fermi statistics are used. Up to $n = 6$ (n = principal quantum number) are used and are sufficient for Al¹³ up to a temperature of about 50eV.

Description of the model

The set of Dirac equations to be solved is

$$[c\vec{\alpha}\cdot\vec{p} + \beta mc^2 + V(r)] \psi_i(\vec{r}) = E_i \psi_i(\vec{r}) \quad (1)$$

where atomic units are used. $\vec{\alpha}$ and β are the usual 4×4 Dirac-matrices and $\vec{p} = -i\vec{\nabla}$. $\psi_i(\vec{r})$ is a four component Dirac spinor.

Inside the Wigner-Seitz radius the potential is spherically symmetric and the solutions may be written in the form

$$\psi_k(r) = \begin{bmatrix} g_k(r) \chi^k \\ i f_k(r) \chi^{-k} \end{bmatrix}$$

where the radial functions satisfy the coupled differential equations

$$\frac{d}{dr}cf = (\kappa - 1) cf/r - (E_i - 1 - V)g \quad (2)$$

$$\frac{d}{dr}g = ((E_i - V)/c^2 + 1) cf - (\kappa + 1)g/r \quad (3)$$

and are spin angular functions. The relationship between various quantum numbers are

$$\kappa = -s(j + \frac{1}{2}), \quad j = l + \frac{1}{2}s \text{ and } s = \pm 1$$

We are mainly concerned with the solution of equations 2 and 3. The procedure is well described in reference 6 and consists of transforming to the logarithmic derivative $x = \log r$ and using a combination of Runge-Kutta and Milne methods.

If one fixes the energy E and numerically integrates equations 2 and 3 from the origin $r = 0$ to the Wigner-Seitz radius $r = S$, one can calculate the logarithmic derivative of the wavefunction at $r = S$

$$D_1(E) = \frac{cf}{g} - \frac{\kappa+1}{S}$$

Repeating this calculation for all values of the energy E one obtains a monotonically decreasing function of energy. Each nl value give a separate branch. This function plays the role of the energy and angular momentum boundary condition.

In the absence of hybridization between orbitals with different angular momentum, it is found that there are no bands for $D_1 \geq 0$. The successive nl bands for a given n are found to extend only over those energy intervals where D_1 is negative, between two energies B_{nl} and A_{nl} . The case $E = B_{nl}$ describes the bonding state (the wave function has a maximum at the Wigner-Seitz radius), which corresponds to the bottom of the nl band, while $E = A_{nl}$ describes the antibonding state (the wavefunction has a node at the Wigner-Seitz radius) which corresponds to the top of the nl -band. These conditions for the extrema of the bands are the so-called Wigner-Seitz rules.

For this model to be practical there is a need for the rapid solution of eq 2 and 3 as well as a rapid calculation of the band edges.

Andersen⁽⁷⁾ has developed a scheme for the calculation of the Wigner-Seitz band edges and this is adopted here.

The electron density $\rho(r)$ obtained from a product of the wavefunction and its adjoint. Here we distinguish between core electron density $\rho_c(r)$ and band electron = density $\rho_b(r)$.

$$\rho(r) = \rho_c(r) + \rho_b(r)$$

$\rho_c(r)$ and $\rho_b(r)$ are given by

$$\rho_c(r) = \sum_{nl} \frac{2(2l+1)}{e^{(E-\mu)/kT} + 1} |R_{nl}(r)|^2 \quad (4)$$

$$\rho_b(r) = \sum_{nl} \left(\int_0^{\Delta E_{nl}} \frac{d_{nl}}{e^{(E-\mu)/kT} + 1} |R_{nl}(E, r)|^2 dE \right) \quad (5)$$

$$\text{where } |R_{nl}(E, r)|^2 = g_{nl}^2(E, r) + f_{nl}^2(E, r)$$

μ is the chemical potential and is given by

$$4\pi \int_0^S \rho(r) r^2 dr = Z \quad (6)$$

d_{nl} in equation 5 is the density of states, assumed to be constant across each nl band and is given by

$$d_{nl} = 2(2l+1)/\Delta E_{nl},$$

where ΔE_{nl} is the bandwidth. The wavefunction is normalised to unity within the Wigner Seitz sphere.

The potential energy $V(r)$ can be written as

$$V(r) = -\frac{Ze^2}{r} + V_c(r) + V_{ex}(r) + V_{cor}(r) \quad (7)$$

The first term is due to the nuclear charge. The second is the classical electron interaction which satisfies the Poisson equation

$$\nabla^2 V_c(r) = -4\pi e^2 \quad (8)$$

The third and fourth terms correspond to the exchange and correlation respectively. Here the Hedin-Lindqvist exchange-correlation potential⁽⁸⁾ is used.

Equations 2 to 8 are solved iteratively until self consistency for every core state and for a set of energy points in each band. The number of energy points in each band was increased until the integration error in equation 5 was minimal. Since this resulted in a small increase of the computational time on the Cray, no attempt has yet been made to optimise the calculation of the band wavefunction.

Results

Calculations were performed for Al^{13} and compared with a full bandstructure calculation. Twenty one energy levels and bands were used (up to $n = 6$). The electron densities were calculated according to equations 4 and 5 and the iterative procedure was carried out until the maximum deviation in the potential in two successive iterations was less than 10^{-5} . The chemical potential μ was also adjusted at each iteration according to equation 6.

Table A4.2 shows the energy levels for Al^{13} at zero temperature as given by this model (SBS) and by the bandstructure APW⁽⁹⁾ method. The Fermi levels compare very well (0.678 Ry from SBS and 0.651 Ry from APW).

Calculations were also performed for Al^{13} at a compression $\eta = V_0/V$ of 2, 5 and 10. Under pressure the bands broaden and move up in energy. This results from the growing importance of kinetic energy versus potential energy as volume decreases due to different scaling $V^{-2/3}$ and $V^{-1/3}$ respectively⁽¹⁰⁾.

Table A4.3 shows some results. The last column of Table A4.3 contains the K-edge shift as calculated from

$$\Delta E_K(\eta) = [E_F(\eta) - E_{1s}(\eta)] - [E_F(1) - E_{1s}(1)]$$

The expression above neglects interatomic relaxation processes that shift the edge position slightly. Since it is not clear how to treat these effects under compression we have assumed that these relaxation

processes are pressure independent and therefore cancel in the expression of the edge shift.

Table A4.2: Energy levels in Al^{13} at $kT = 0$ as given by spherical band structure (SBS) and by the APW bandstructure method (energies are in Rydbergs)

nl	SBS bands		APW bands	
	bottom	top	bottom	top
1s	-109.383	-109.383	-109.552	-109.552
2s	-6.796	-6.796	-6.904	-6.904
2p	-4.018	-7.017	-4.127	-4.126
3s	-0.185	1.086	-0.175	1.078
3p	0.260	1.784	0.271	1.776
3d	0.955	2.615	0.968	2.609
4s	2.950	14.107	2.931	14.082
4p	3.614	6.715	3.592	6.692
4d	4.168	6.808	4.143	6.780
4f	2.088	4.785	2.105	4.796
5s	9.543	24.600		
5p	9.919	14.256		
5d	9.555	13.370		
5f	7.138	10.584		
5g	3.420	14.164		
6s	18.786	24.600		
6p	18.691	24.208		
6d	17.296	22.268		
6f	14.058	18.589		
6g	10.008	14.164		
6h	4.985	17.872		

Table A4.3: K edge shift of Al^{13} as a function of compression η , at $kT=0$ (Energies are in Ry)

η	E_{1s}	E_p	ΔE_K
1	-109.387	0.678	0
2	-108.432	1.605	-0.028
5	-107.093	3.577	+0.605
10	-104.093	9.071	+3.543

The effect of temperature is to increase ionization and therefore reduce the screening of the nuclear charge by core electrons. Potential energy increases more than kinetic energy. As a result the bands move down in energy while getting narrower. As the temperature increases, higher bands start to fill. This can be seen in Table A4.4. K shell ionization starts at about 100eV. Even at this temperature the occupation of the highest bands is about 1% or less. At these high temperatures one ought to include at least the nodeless 7i, 8k, 9l bands etc which have a low 'kinetic' energy compared to other state and will have a sizeable occupation (for example, the nodeless 6k has an occupation of .039 at 100eV). By increasing the number of bands one can extend the model to higher temperatures. At the highest temperature for which the number of bands becomes excessive one ought to use an alternative model such as the one in reference 1, 3 or 4.

This model can also be used for the calculation of equation of state. We are currently using the self consistent electron potential for the calculation of laser shock-compressed EXAFS.

Table A4.4: Occupation of energy levels and bands of Al^{13} at a compression of two and for different temperatures

nl	kT=0	kT=2eV	kT=10eV	kT=50eV	kT=100eV
1s	1.0	1.0	1.0	1.0	0.999
2s	1.0	1.0	0.999	0.780	0.471
2p	1.0	1.0	0.996	0.638	0.394
3s	.552	.527	0.369	0.198	0.147
3p	.301	.278	0.212	0.170	0.137
3d	.008	.027	0.091	0.138	0.122
4s	0		0.00003	0.014	0.035
4p			0.00004	0.034	0.064
4d			0.00005	0.036	0.067
4f			0.006	0.071	0.084
5s			0	0.0005	0.006
5p			0	0.001	0.015
5d				0.003	0.020
5f			0	0.008	0.031
5g			0.0002	0.035	0.058
6s				0.0002	0.0017
6p				0.0003	0.002
6d				0.00009	0.003
6f				0.0003	0.006
6g				0.002	0.015
6h				0.016	0.039

References

1. R D Cowan and J Ashkin, Phys. Rev., **105**, 144 (1957).
2. D D Koeling, Rep. Prog. Phys., **44**, (1981).
3. D A Liberman, Phys. Rev., **B20**, 12, (1979).
4. B F Rozsnyai, Phys. Rev., **A5**, 3, (1972).
5. T A Hall et al (submitted to Phys Rev Lett).

6. T L Loucks, "Augmented Plane Wave Method", New York: Benjamin, (1967).
7. O K Andersen, Phys. Rev., **B12**, 3060 (1975).
8. L Hedin and B I Lundqvist, J. Phys. C., **4**, 2064 (1971).
9. M Ross and K W Johnson, Phys. Rev., **B2**, 4709 (1970).
A K McMahan and M Ross, Phys. Rev., **15**, 718 (1977).
10. R C Albers, A K McMahan and J E Muller, Phys. Rev., **B31**, 3435-3450 (1985).

A4-6 FUSION NEUTRON AND ALPHA PARTICLE TRANSPORT IN INERTIAL CONFINEMENT FUSION TARGETS

T D Beynon and D P Edwards, (University of Birmingham).

A discrete-ordinates one-dimensional Boltzmann transport code ANISN⁽¹⁾ has been used in conjunction with the 1D hydrodynamics code MEDUSA⁽²⁾ to investigate the transport and energy loss of neutrons and alpha particles produced by D-D and D-T fusion in ICF targets.

As a neutron transport code, ANISN is used with spatial mesh boundaries to match those in MEDUSA and is run every hundred or so MEDUSA time steps. The heating rates are fed back into the MEDUSA cells to provide an explicit self-consistent approach. The neutrons interact during the thermonuclear burn period with D, T and ^4He nuclei, interactions with H and ^3He being neglected because of their low concentrations.

Alpha particle transport is treated in a similar fashion, except that the discrete ordinates description must be modified to include an extra source term $\frac{d}{dE} [S(\underline{r}, E) \Phi(\underline{r}, E, \underline{Q})]$ where Φ is the phase-space flux at energy E and transport vector $\underline{Q}$, and $S(\underline{r}, E) = -dE/dx$ is the slowing down power. Both small angle (i.e. continuous slowing down) and wide angle Coulomb interactions are included⁽³⁾. Phase space trajectories for charged particles are curved, unlike the case for neutral particles (e.g. neutrons) which are linear. It is found that each phase space cell $(\Delta E, \Delta x, \Delta \mu)$ must be characterised by a Courant-Friedrichs-Levy

stability condition, $\theta = \frac{\Delta E}{S} \frac{|\mu|}{\Delta x} = 1$, where $|\mu|$ is the modulus of the average value of the cosine of the angular flux. The condition $\theta = 1$ is necessary to ensure a full removal of particles from one phase space cell to the next. If $\theta > 1$ the particles slow down too fast, and too slowly if $\theta < 1$.

Detailed conclusions of this work depend very much on the details of the target considered but the modelling has allowed the following general properties to be established.

- i) Neutron energy deposition increases with ion number density, independently of temperature, and is therefore a maximum when the compression is greatest at the time of maximum burn.
- ii) The alpha particle-energy loss is dependent on both electron density and temperature, and decreases as the electron temperature increases. The alphas therefore deposit nearly all their energy in the fuel up to the time of maximum compression, when the increased temperature then allows a high fraction of the alphas to escape.
- iii) Alpha particles therefore will successfully self-heat the bulk fuel during the early stages, raising the temperatures to those needed for optimum burn. At maximum burn the neutrons provide comparable heating rates (typically 2 MeV per D T fusion reaction) and the two mechanisms should be regarded as equally important.

References

1. W W Engles Jr, 'A Users Guide to ANISN', Computer Technology Center, Union Carbide Corp. Rep. No. K-1693, (1967).
2. J P Christiansen, D E T Ashby, K V Roberts, Comp. Phys. Comm., 7, 271, (1974).
3. A P Thompson, Ph. D Thesis, University of Birmingham, (1982).

A4-7 IMPROVEMENTS IN THE MODELLING OF RECOMBINATION X-RAY LASERS.

S J Rose (R.A.L.)

At R.A.L. a one-dimensional hydrodynamics code, MEDUSA, has been adapted to include a time-dependent screened-hydrogenic average-atom model of the atomic physics. This model is based, in part, on the XSN package currently used in the L.L.N.L. code LASNEX. However, as was reported in last year's Annual Report, several modifications have been made in order to study recombination schemes. These include an escape factor approach to the calculation of radiation transport radially, and the explicit solution of the radiation transport equation along the axis of the amplifier to give lasing line intensities for different fibre lengths from which an integrated value of the gain, which can be compared directly with experiment, can be obtained. This code was used to produce the theoretical results on carbon-fibre experiments presented by Chénais-Popovics et al⁽¹⁾. The calculated value of the gain is found to be too large at early times, yet at the time of peak gain (over 4 cm^{-1}) in the experiment, the calculated value of the gain is less than 1 cm^{-1} .

Over the last year several improvements have been included in the modelling. First, the upper level ($n = 6$) is constrained to be the LTE value. This improves the description of the recombination. Secondly, account is taken of the fine-structure of the H_α line: the different components are separated in energy sufficiently that for regions of the plasma which show gain, amplification on one component does not, to a good approximation, interact with another component. We take the simplest approximation of only considering gain on the $2p_{3/2} - 3d_{5/2}$ component of H_α , which for a distribution of population amongst the fine-structure level according to degeneracy, shows a gain which is approximately twice as large as the next largest component $2p_{1/2} - 3d_{3/2}$. Lastly the effect of both static and dynamic Stark broadening of the H_α line is included using the calculated values of Oza, Kelleher and Greene⁽²⁾. They show that for CVI, at temperatures and densities at which significant gain occurs. Stark broadening can be comparable to, or larger than, the Doppler width. Figure A4.17 shows that inclusion of these effects brings the maximum calculated value of the gain down into better agreement with experiment. However the gain still occurs too early in the simulation and does not last for

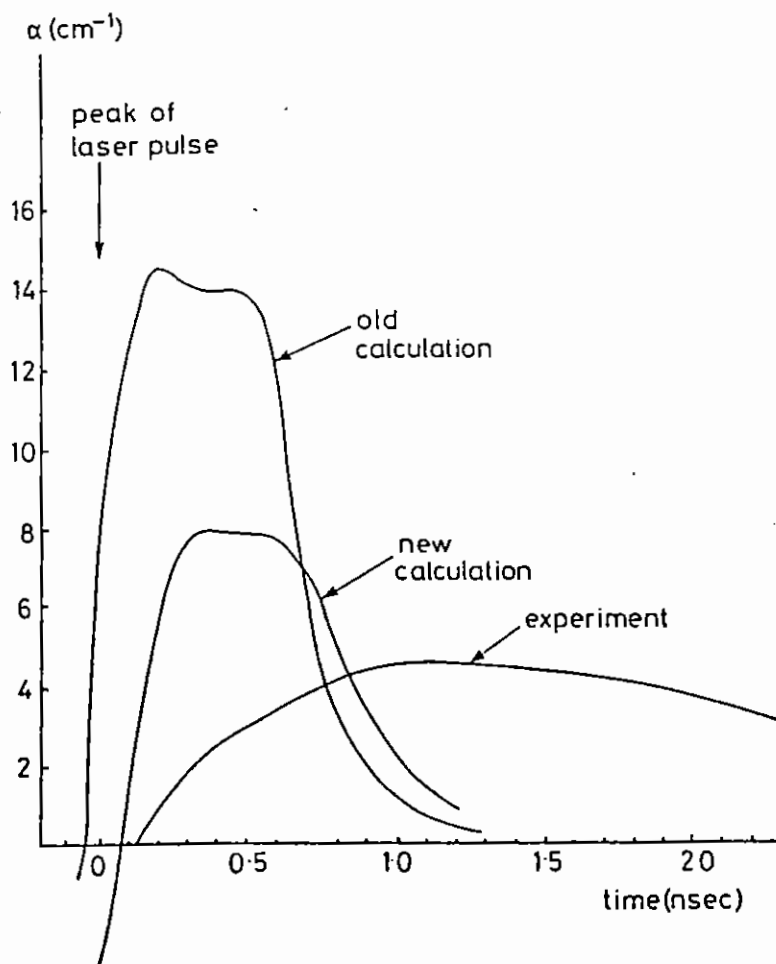


Fig. A4.17 Time dependence of the gain on H_{α} taken from two fibre lengths (2.2 mm and 8.0 mm) with 2.5 J/cm absorbed energy showing the effect of the changes to the modelling described in the text. The values shown are calculated with a convolution of the theoretical intensities in the 400 psec "top-hat" convolution function.

as long as is seen experimentally. We are still investigating the reasons for this remaining discrepancy.

References

1. C Chenais-Popovics et al, Phys. Rev. Letts. **59**, 2161 (1987).
2. D H Oza, R L Greene and D E Kelleher, Phys. Rev. **A34**, 4519 (1986).

A4-8 MODELLING OF Li-LIKE AND Na-LIKE RECOMBINATION AND Ni-LIKE COLLISIONALLY-PUMPED X-RAY LASERS

G J Pert and M Henshaw (York University)

Lithium-like Ions

Experimental observations of gains have been made on lithium-like aluminium on the transitions 3d-4f, 3d-5f, 3p-4d, 3p-5d. Simulation of the behaviour of coated carbon fibres have studied a number of elements, but have concentrated on aluminium to compare with experiment. The model has been successively refined to improve the predictions of gain.

Initial simulations indicated gain on the transitions d-f but not on p-d. The line 3d-4f had a very large peak gain ($\sim 11/\text{cm}$) but on 3d-5f only small ($\sim 0.1 \text{ cm}^{-1}$). This calculation included only levels up to $n = 5$ with l (but not j) splitting, and hydrogen-like $n = 6$ and $n = 7$ states, for the lithium-like ion, the other ions being treated by an averaged scheme.

In order to improve the representation the atomic model has been considerably extended. The helium-like ion (Al XII) is now represented by four levels, and the berythium-like ion (Al X) by eight. The lithium-like ion (Al XI) includes full j splitting up to $n = 5$, l splitting for $n = 6$ and hydrogenic states for $n = 7$ to $n = 14$ - a total of 37 levels. The oscillator strengths are taken as follows.

$$n_1, n_2 < 7 \Delta n \neq 0 \text{ Guennon and Sureau}^{(1)}$$

$$\Delta n = 0 \text{ Grant}^{(2)}$$

$$7 < n_2 < 14 \quad \text{Bates and Dangard}^{(3)}$$

Collision rates for transitions from the ground states (2s) to s, p or d states are taken from Mewe's polynomial⁽⁴⁾, the remainder by Van Regemorter's formula⁽⁵⁾.

Using this scheme the peak gain on the transition 3d-4f is reduced to $\sim 3/\text{cm}$, but increased to $\sim 0.9/\text{cm}$ for 3d-5f. In addition gain is now predicted on the lines 3d-4d and 3p-5d but only within a limited region of plasma, see Table A4.5, and Figures A4.18-A4.21.

Table A4.5

Transition	Predicted gain (cm^{-1})	Time of peak (ps)	Experimental gains (cm^{-1})
3d-4f 154 Å	3	550	2.5-3.0
3d-5f 105 Å	0.9	900	2-2.5
3p-4d 150 Å	1.8	900	2
3p-5d 104 Å	0.4	900	1.5

The model was extended to include dielectronic recombination. This, however, did not change the gain. This is due to the fact that since the lowest doubly excited state of lithium is about 2000 eV above the ground state compared to an ionisation potential of 441.9 eV, the doubly excited state is only very weakly populated from the helium-like ground state.

The calculations have been performed for solid aluminium fibre of 7 μm diameter, instead of an aluminium coated carbon fibre as in the experiment. The fibre is heated by a 70 ps laser pulse of energy 43 J of which about 20% is absorbed, values somewhat in excess of those used experimentally. This deviation is not expected to lead to major disagreements, and will be removed by further calculation. The most serious approximation in the present calculation is believed to be the

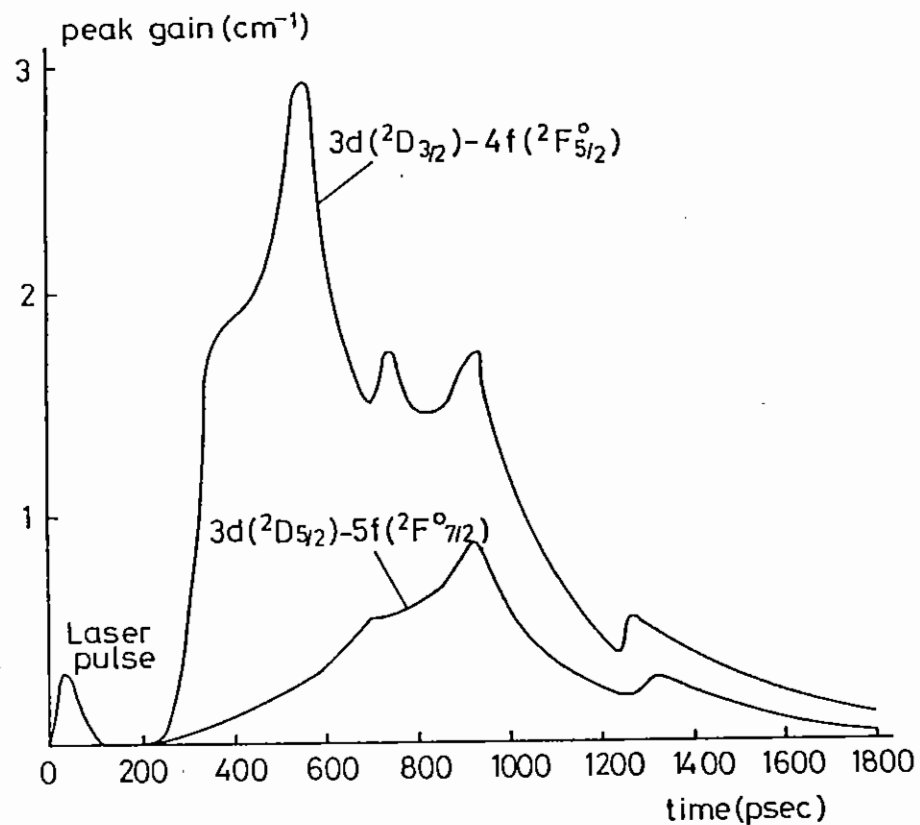


Fig. A4.18 Peak gain versus time for d to f transitions in aluminium XI.

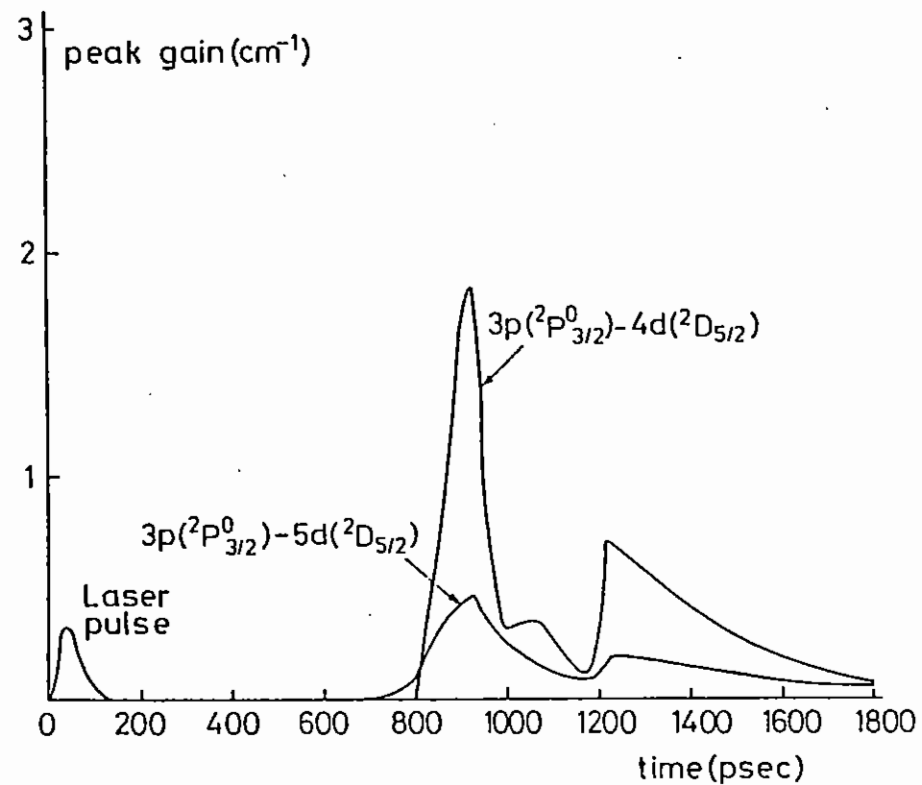


Fig. A4.19 Peak gain versus time for the p to d transitions in aluminium XI.

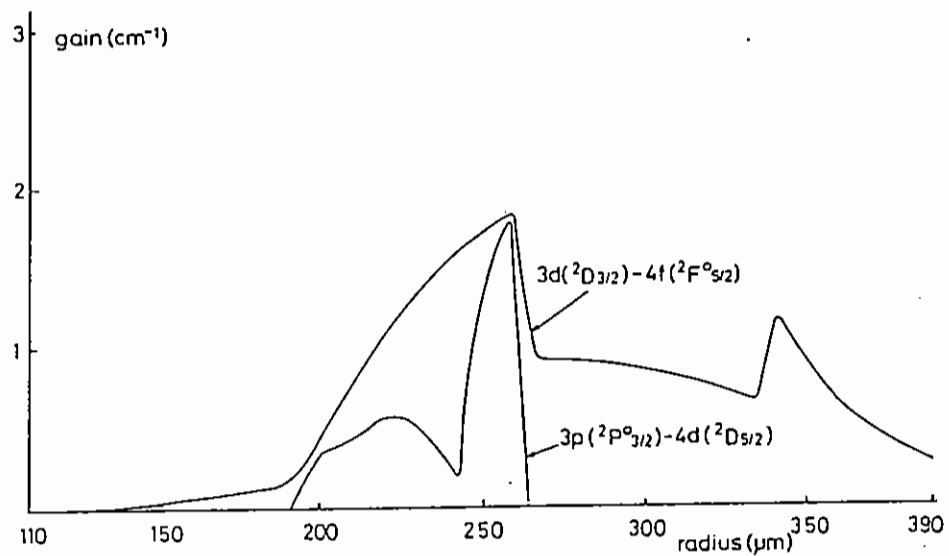


Fig. A4.20 Gain versus radius for the 3 - 4 transitions in aluminium XI at 935 psec (the maximum of 3p - 4d gain).

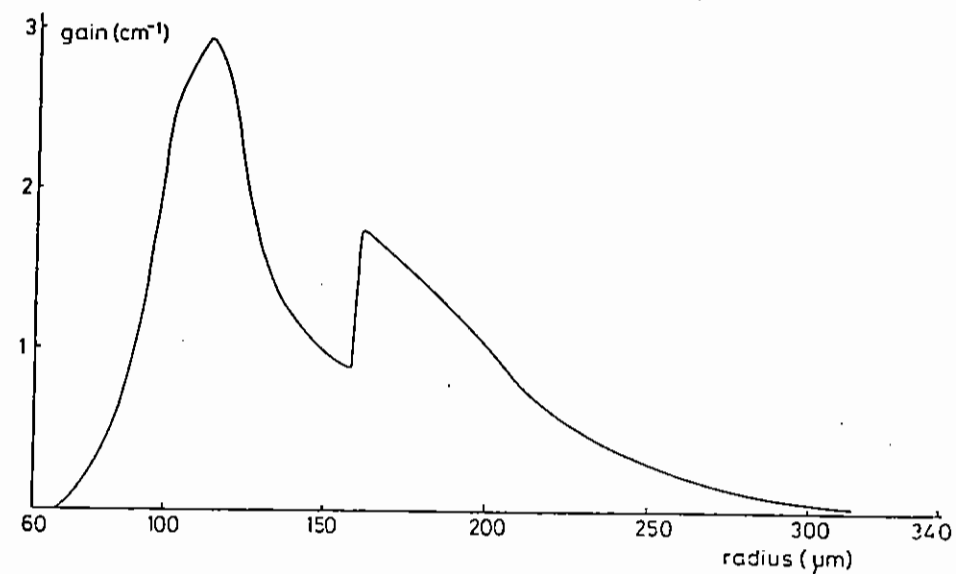


Fig. A4.21 Gain versus radius for $3d(^2D_{3/2}) - 4f(^2F_{3/2})$ transitions in aluminium XI at 560 psec (the maximum of 3d - 4f gain).

treatment of the Stark Broadening of the lines in gain, and will be improved in the near future.

Other members of the isoelectronic sequence are now being investigated. Chlorine (Cl XV) in a sodium chloride plasma shows a peak gain of $\sim 1/\text{cm}$ on the 3d-4f line. Similar values are obtained in preliminary calculations for sulphur (S XIV) and calcium (Ca XVIII), although a trend of decreasing gain with increasing atomic number is seen.

Sodium-like Ions

Limited data sets for sodium-like copper and iron have been created. No gain on any lines in these systems has been predicted.

Neon-like Selenium

Gains on a number of transitions in neon-like ions, particularly selenium, has been generated in exploding foil plasmas by the Livermore group⁽⁶⁾. The transitions are between the $2p^5 3p$ and $2p^5 3s$ levels. Gains of order $\sim 10\text{cm}^{-1}$ have been predicted, and values $2\text{--}4\text{cm}^{-1}$ measured.

In order to re-examine the question of the 182.4 Å line anomaly we have calculated gains using atomic data supplied by the Belfast group. The similarity code was used to model the hydrodynamics, and an atomic-scheme of initially 27 levels, but subsequently 37. Dielectronic recombination was not included in these calculations. The results are shown in table A4.6.

Table A4.6

Line Å	Predicted gain (cm^{-1})		Measured gain (cm^{-1})
	27 levels	37 levels	
169.2	10.9	6.9	
182.4	2.8	1.4	
206.2	34.0	21.4	4.0
209.6	15.8	9.9	3.8
220.2	0.6	0.4	2.3
262.9	7.6	4.3	3.5

In addition gain is predicted on the line at 70.4 Å^{*} over a limited region of plasma.

The poor agreement with experiment is probably due to the neglect of dielectronic recombination, as shown by Hagelstein et al⁽⁷⁾.

* $(1s^2 2s^2 2p_{1/2} 2p_{3/2}^4 3s - 1s^2 2s 2p_{1/2}^2 2p_{3/2}^4 3s)$

References

1. H Guennou, and A Sureau, J. Phys. B., **20**, 919 (1987).
2. I P Grant, B T McKenzie, P H Norrington, D F Mayers, N C Pyper, Comp. Phys. Comm., **21**, 207 (1980).
3. D R Bates, and A Damgaard, Phil. Trans. Roy. Soc. London, Ser. A, **242**, 101 (1949).
4. R Mewe, Astron and Astrophys., **20**, 215 (1972).
5. H Van Regemorter, Astrophys. J., **136**, 906 (1962).

6. D L Matthews, et al, Phys. Rev. Letts., **54**, 110 (1985).
7. P L Hagelstein, M D Rosen, and V L Jacobs, Phys. Rev., **A34**, 1931 (1986).

A5	SPRITE OPERATION AND DEVELOPMENT	Pages
A5-1	Sprite Operations	117 - 118
A5-2	High-Energy Picosecond Pulse Amplification in KrF	118 - 124
A5-3	Picosecond Raman Amplification	124 - 128
A5-4	New Diode for the E-Beam Pumped Pre-Amplifier Goblin	129
A5-5	Super-Sprite Design	129 - 134

Editor: M J Shaw

A5-1 SPRITE OPERATIONS

M J Shaw (RAL)

This has been a very fruitful year for the Sprite laser operations group. A total of nearly 2000 laser shots were fired. The breakdown is shown in Table 1. Of these, over 50% were concerned with target shooting.

The major development this year has been the amplification of picosecond pulses to energies in excess of 1J and the immediate application of such pulses to target experiments. This is a "world first" for RAL since there exists no other comparable high energy picosecond KrF irradiation facility. The laser scheme employed is described briefly below (A5.2) and in more detail in a recent publication⁽¹⁾.

One problem encountered in these experiments was the generation of a large ASE (amplified spontaneous emission) pulse which was associated with the picosecond pulse. The contrast ratio between the short and long pulses proved difficult to control in practice and gave rise to difficulties in experimental interpretation.

To overcome this problem we have been developing a picosecond Raman system which will be capable of acting as a beam combiner and so be able to increase the available energy on target. Its additional advantages are that it will decouple the target from the ASE and also improve the laser beam quality. As part of this programme the operation of picosecond Raman generators has been investigated experimentally and the results published⁽²⁾. More recently the first Raman amplifier stage has been operated and the results are described in section A5.3.

In the final period covered by this report we have been able to make some very interesting measurements on the picosecond gain dynamics of the KrF B&C states. We have found that the B-C mixing rate is extremely slow under typical laser conditions and so the C state plays no important role in the laser kinetic scheme. This is in disagreement with previous assumptions^(3,4). These results, which also give the

first direct measurements of the KrF* B state lifetime are described more fully below (A5.2).

Finally, this year has seen the fruition of the CLF gas laser development programme in the adoption of KrF as the preferred choice for a future high power laser system. This new (as yet unfunded) system is called "Super-Sprite" and some of the design considerations are discussed in section A5.5.

Table 1

Sprite Shot Record 1987-88

Date	Activity	No of laser shots
Mar 24 - Apr 24	Picosecond development	220
Jul 1 - Jul 24	British Aerospace contract 2 beams 40J/20ns	510
Aug 12 - Oct 10	Picosecond development	310
Oct 20 - Nov 6	Imperial College ps target shooting 1 beam 1J/3.5ps	230
Nov 17 - Dec 4	BA contract 2 beams 40J/20ns	200
Dec 7 - Dec 24	RAL X-ray source 1 beam 40J/20ns	200
Jan 4 - Feb 26	KrF gain dynamics	310
Total laser shots		1910
Total target shots		1070

References

1. J R M Barr, N J Everall, C J Hooker, I N Ross, M J Shaw and W T Toner, Optics Comm **66**, 127-132 (1988).

2. N J Overall, J P Partanen, J R M Barr and M J Shaw, Optics Comm, 64 393-397 (1987).
3. F Kannari, M Obara and T Fujioka, J Appl Phys 57 4309-4322 (1985).
4. S Szatmari and F P Schaefer, Appl Phys B 33 219-223 (1984).

A5-2 HIGH-ENERGY PICOSECOND PULSE AMPLIFICATION IN KrF

C Hooker (RAi.)

Introduction

During the last year the majority of Sprite operating time has been devoted to amplifying pulses of a few picoseconds to energies of several Joules. These pulses were ultimately delivered to the target area for an experiment on ionisation balance in materials of different $Z^{(1)}$. During the development of the system we characterised the behaviour of Goblin and Sprite while amplifying very short pulses, and investigated several phenomena relevant to the design of the next generation of large e-beam-pumped KrF modules. The most recent work has focused on measurements of the quenched and radiative lifetimes of the $\text{KrF}^* \text{B}$ state, and on the rate of mixing between the B and C states, which was found to be very small.

Picosecond source

The scheme for picosecond pulse generation and amplification is shown in Figure A5.1. The picosecond source is a dye oscillator tuned to 746nm, which is synchronously pumped by a frequency-doubled, mode-locked Nd:YAG laser. The dye output consists of a train of pulses at 12ns intervals having a FWHM of approximately 3ps. A three-stage excimer-pumped dye amplifier, which has a gain lifetime of 6-8ns, selectively amplifies a single pulse from the train at a repetition rate of 8Hz. The amplified pulse is frequency-tripled in a pair of KDP crystals, to give a single pulse at 248.6nm. The tripling process effectively eliminates any ASE originating in the dye amplifier, so the picosecond pulse is extremely clean.

It is spatially filtered before being amplified in a KrF discharge-pumped pre-amplifier at 8Hz, to a final energy of about 100μJ per pulse. This seed beam can be propagated around the rest of the system for alignment purposes, and is visible even when expanded from 5mm to the 27cm diameter of Sprite. The pulse length of the seed pulse has been measured by cross-correlating it with the residual 746nm light emerging from the mixing crystal and observing the difference frequency. The measured pulse length was $3.5 \pm 0.6\text{ps}^{(2)}$.

High-Power Amplification

As shown in Figure A5.1, the seed pulse was further amplified first in Goblin, then in Sprite, to a final energy of up to 2.5 Joules, although such energies were achieved only after careful optimisation of the system. Two main problems were encountered, both related to the amount of ASE generated by the KrF amplifiers.

Both Goblin and Sprite were run in double pass, as illustrated, and the presence of the mirror close to the amplifier module can lead to extensive self-depumping by ASE reflected back into the gain region. To minimise this effect, the back mirrors were moved away as far as possible from the amplifiers: 2 metres in the case of Sprite, 1 metre for Goblin. This procedure also allows the gain time to recover before the returning pulse re-enters the gain region.

Although the 248nm pulse generated by the frequency-tripling has essentially no background, the 8Hz seed pulse is superimposed on an ASE background of 20ns duration originating in the discharge amplifier. If the gain in Goblin or Sprite is too large, the picosecond pulse will readily saturate since its intensity is very high, while the ASE background will continue to experience small-signal gain. In extreme cases, the energy in the background pulse can grow to several times that in the short pulse. If the necessary gain is exceeded in any stage, the slight increase in picosecond pulse energy is outweighed by the increased amount of ASE injected into the subsequent stages. As an example, a shot with good contrast had 2.15J in the short pulse and 50mJ in the background, with an input of only 12mJ from Goblin. A shot with 67mJ from Goblin gave 1J in the short pulse and 3.4J in the background. To minimise this problem, the three KrF amplifiers were run at gains as low as possible, consistent with amplifying the picosecond pulse sufficiently.

The operating voltages were kept low, and in the case of Goblin and Sprite a low-pressure, low gain gas mix containing 3 torr F_2 , 100 torr Kr and 500 torr Ar was used.

A computer model of short pulse amplification, which includes effects of ASE, predicted output energies of between 2.8 and 3.4J, which were not achieved in practice. The difference may be due to effects of

beam profile leading to a reduced fill-factor in Goblin and Sprite. The pulse length was not measured at the output of Sprite, but other workers^(3,4) have reported no significant pulse-stretching effects with pulses of femtosecond duration, so we conclude that the output pulse would have the same length as the input pulse, namely 3.5ps. The beam profile was smooth, and the measured spot diameter at the focus of a 250mm focal length lens was 25 μ m, indicating a beam divergence of 100 μ rad. Over 200 shots were delivered on target, with focused energies up to 1J.

Sprite Gain Characterisation

As a preliminary to optimising the operating conditions of Sprite and Goblin, a series of gain measurements was made on Sprite using a long ASE pulse from the preamplifier. A fraction of the energy was split off and sent through the Sprite cell in single pass, on a path that would sample a region of average gain, and the remainder was sent to a photodiode. The amplified beam was focused through a small aperture to isolate it from the ASE as far as possible, then collected on a second diode.

The rear mirror was positioned at a distance of 2m from the cell (as required for amplifying picosecond pulses), and gain measurements were made with the mirror blocked and unblocked at a variety of pump rates.

The results are plotted in Figure A5.2, and show several interesting features. At low pump rates, the back mirror makes only a small difference to the gain, indicating that ASE feedback does not seriously depump Sprite when the mirror is 2m away. However, the gain does not increase exponentially with pump rate, as would be expected, so there is clearly some clamping mechanism at work. This could be either single pass ASE or a parasitic oscillation, although no attempt was made to track it down.

At a pump rate of 0.6 MWcm⁻³, the data shows a definite break, with higher pump rates causing a reduction in gain and a clear distinction between the presence or absence of the rear mirror. We attribute this to a parasitic oscillation involving the rear mirror, which turns on at a threshold pump rate, and is sufficiently powerful to depump the gain. To investigate further, a few shots were fired with Dyelux film near the output window to record the emission profile. No significant difference was seen between profiles with the rear mirror blocked and those with it unblocked, but no hot spots were detected that could be attributed to parasitic modes.

For the purposes of picosecond amplification, the operating point was fixed at a pump rate of 0.4 MWcm⁻³, which gave a single-pass gain of 150.

The same experimental set-up was used to investigate the behaviour of Sprite when amplifying picosecond pulses. The results are shown graphically in Figure A5.3, together with theoretical curves based on Gaussian and 'top-hat' beam profiles. The saturation fluence was determined as 2.0 ± 0.4 mJ cm⁻², which agrees with previously-measured values^(5,6,7). A full discussion of these results is contained in⁽¹⁾.

B and C State Mixing

The availability of a high-power picosecond pulse at 248 nm enabled us to investigate the mixing between the B and C states of KrF*, and to settle the hitherto unresolved question of whether such mixing contributes to the gain recovery in a KrF amplifier. On the passage of a saturating picosecond pulse through an amplifier, the depletion of the upper (B) state causes a corresponding drop in the fluorescence on the B $\rightarrow$ X transition at 248 nm. If the B and C states are strongly coupled, the transfer of energy from the undepleted C state will cause the fluorescence on the C $\rightarrow$ A transition to drop as well. Comparison of the temporal behaviour of the depletion on these two transitions will yield information on the degree of mixing between the two states.

In these experiments, Goblin was used as the test amplifier, since its cell was fitted with windows for observing the sidelight fluorescence. The saturating pulse was obtained by amplifying the oscillator output in Sprite; this yielded picosecond pulse energies of between 30 and 60 mJ in a beam of 6 cm diameter, corresponding to

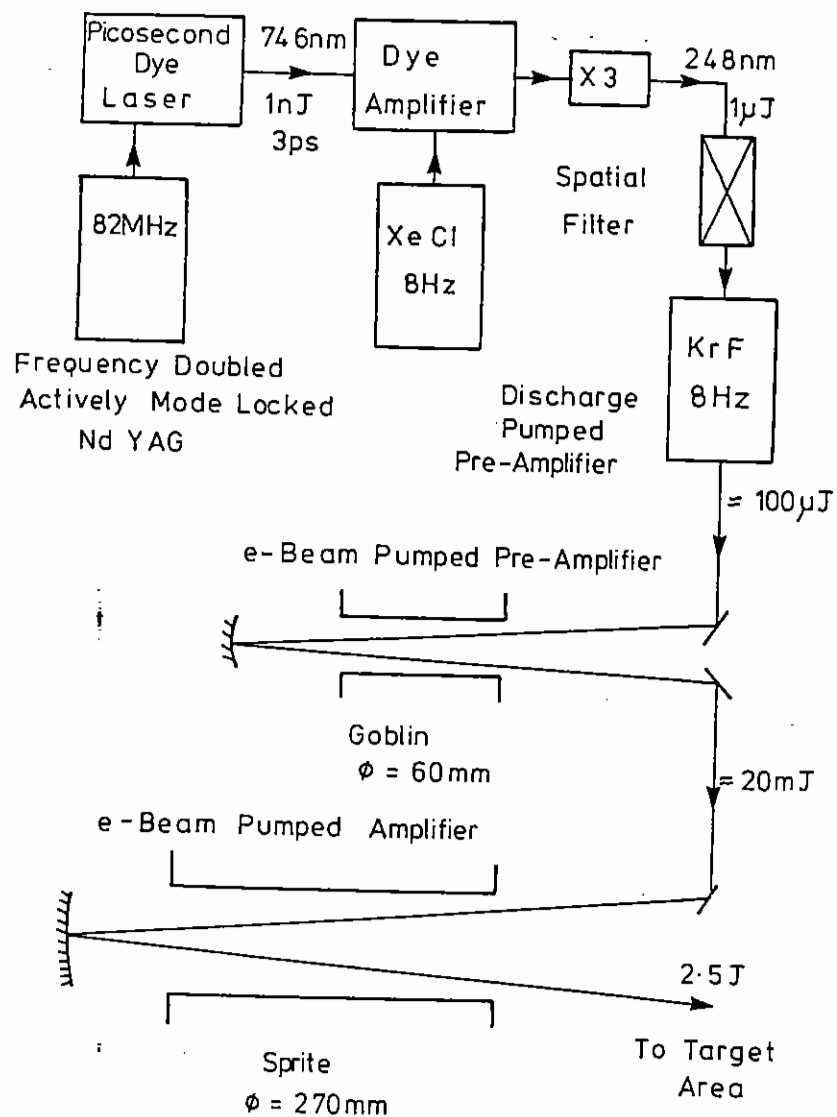


Fig. A5.1 Experimental setup for the generation and amplification of picosecond pulses at 248 nm .

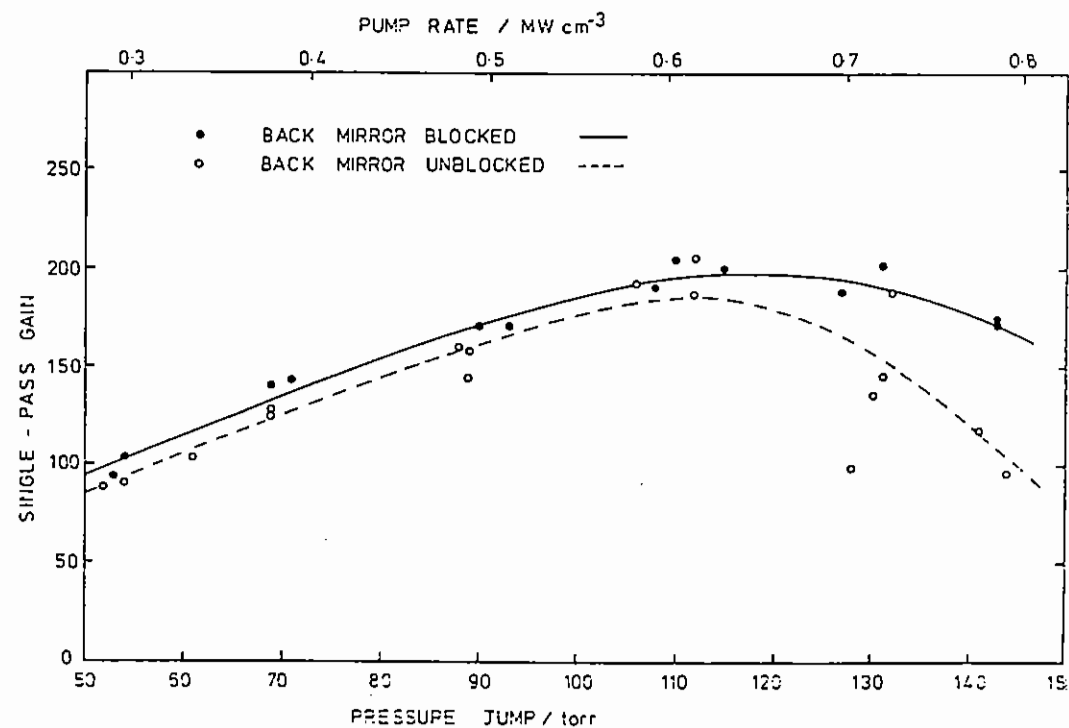


Fig A5.2 Measured single-pass small signal gain as a function of pump rate in the Sprite amplifier.

an average fluence of between 1 and 2 mJ cm⁻² at the input to Goblin. The layout is shown in Figure A5.4, which also includes some features used in the subsequent experiments on gain recovery.

As shown in the figure, the sidelight fluorescence was imaged onto a photodiode, PD1, with a lens of 100 mm focal length working at unit magnification. An iris before the diode restricted the transverse field of view to a circle of 5 mm diameter, and a 248 nm laser line filter was used to isolate the emission from the B → X transition. In the centre of the Goblin cell a metal mask (not shown) was installed to limit the height of the electron beam to 10 mm along the axis of the lens. The optical beam was masked by M1 and M2, which were square apertures 16 mm on a side. The combination of masks and apertures ensured that the entire volume monitored by the photodiode was extracted by the saturating pulse.

Typical B → X sidelight traces are shown in Figure A5.5. In Figure A5.5(a) the entire envelope of emission is shown, corresponding to the 110 ns run time of Goblin; Figure A5.5(b) shows an expanded version from another shot. The recovery part of the trace has the form $(1 - k \exp(-t/\tau))$, with a characteristic time t of about 3 ns. The maximum depletion k that was observed in any shot was 30%, which suggested that the short pulse was not energetic enough to deplete the population further. The energies measured at the output of Goblin were typically 30 mJ in a 16 mm² beam, corresponding to nearly 6 E_{sat} , so a mirror was installed to reflect the amplified pulse back through the amplifier. The result, shown in Figure A5.5(c), indicates that the maximum depletion is definitely limited to about 30%. The most likely explanation is that some of the B state vibrational levels are inaccessible to the extracting pulse, although their spontaneous emission still lies within the pass band of the 248 nm filter.

The C → A transition radiates over a broad band lying between roughly 270 nm and 350 nm with no significant peaks. An XeCl line filter (passband centred at 308 nm) was used to isolate a part of the emission lying well away from both the B → X transition and the Kr₂F band centred at 420 nm. A mirror with 98% reflectivity at 248 nm was used to further reject the B → X emission. The 308 nm filter passband overlaps the emission band from Ar₂F, a species which would normally be present in the Ar-buffered gas mix: to avoid this, the gas mixes used contained only F₂, He and Kr, with total pressures varying between 300

and 800 torr. As a final check that the emission seen was due to the C → A transition, the ratio of sidelight signals on the two diodes was measured, and found to be constant over the range of pressures employed.

When the C → A sidelight was monitored during the passage of a short pulse through Goblin, no depletion was detected (Figure A5.6(a)), although the energy at the output was measured at 30 mJ, enough to give 30% depletion at 248 nm. To determine whether any mixing of the states was occurring, a 20 ns pulse split from the output of the EMG 150 trigger laser was substituted for the picosecond pulse, and the double-pass mirror replaced. The results are shown in Figure A5.6(b) and (c), for a Goblin fill pressure of 500 torr. The B → X sidelight is strongly depleted, to a maximum depth of 35%, and the C → A sidelight trace, although noisy, exhibits a corresponding but weak depletion. Further traces were recorded at pressures of 300 and 800 torr, which, although too noisy for detailed analysis, show a trend towards increased depth of depletion of the C → A sidelight with increasing pressure, as would be expected for a collisional process in the excited medium.

The main conclusion to be drawn from these results is that there is no rapid mixing between the B and C states of KrF*. The evidence for this, the absence of a rapid depletion of the C → A sidelight, is fairly conclusive given that depletion was observed on the same channel using the 20 ns pulse. Other workers⁽⁶⁾ have reported very rapid mixing between these states; however, their conclusion is not based on a direct measurement but on an interpretation of other data. Our results point to a very slow mixing, with a characteristic time of the order of 10–20 ns.

References

1. J R M Barr, N J Everall, C J Hooker, I N Ross, M J Shaw and W T Toner, *Optics Comm* 66 127 (1988).
2. I N Ross, D Karadice and J R M Barr, Section B4.5, Annual Report 1988.
3. A P Schwarzenbach et al, *Opt. Lett.*, 11, 499 (1986).
4. S Szatmari et al, *Opt Comm*, 63, 305 (1987).

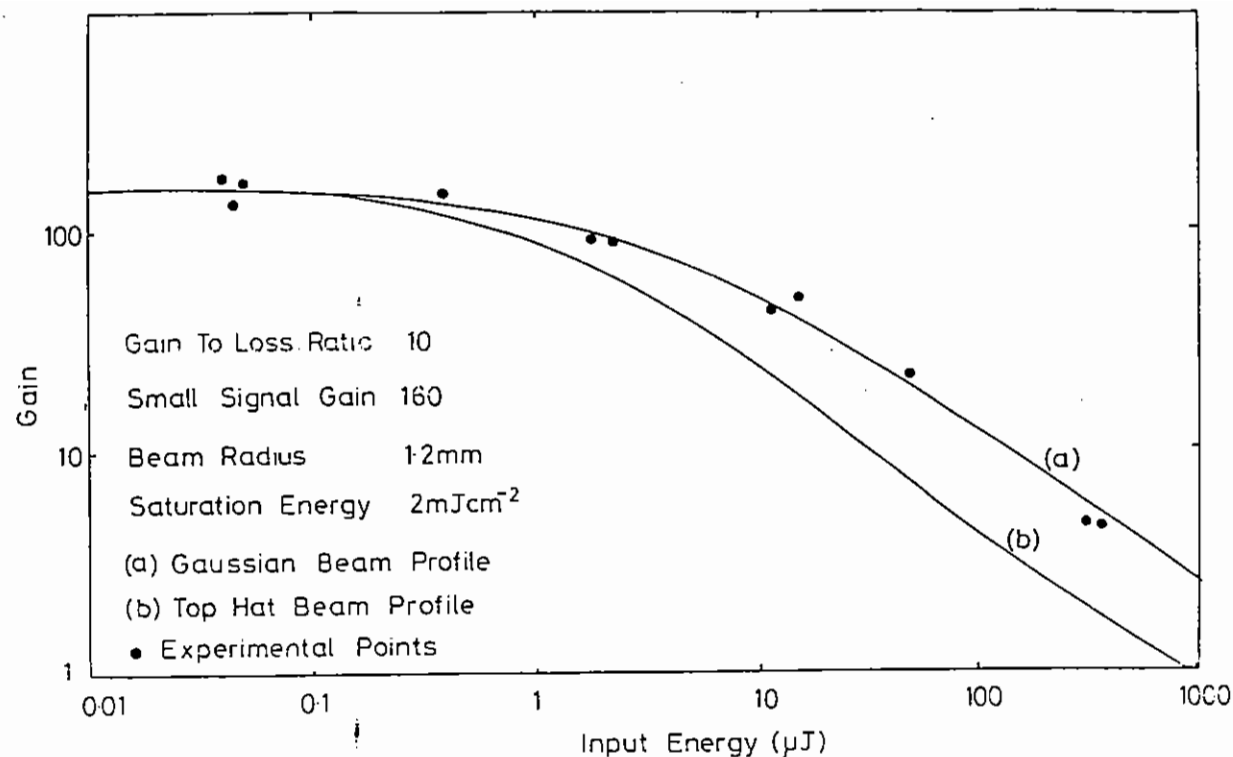


Fig A5.3 Gain saturation characteristic for picosecond pulses for an e-beam pumped KrF gain medium (Sprite module).

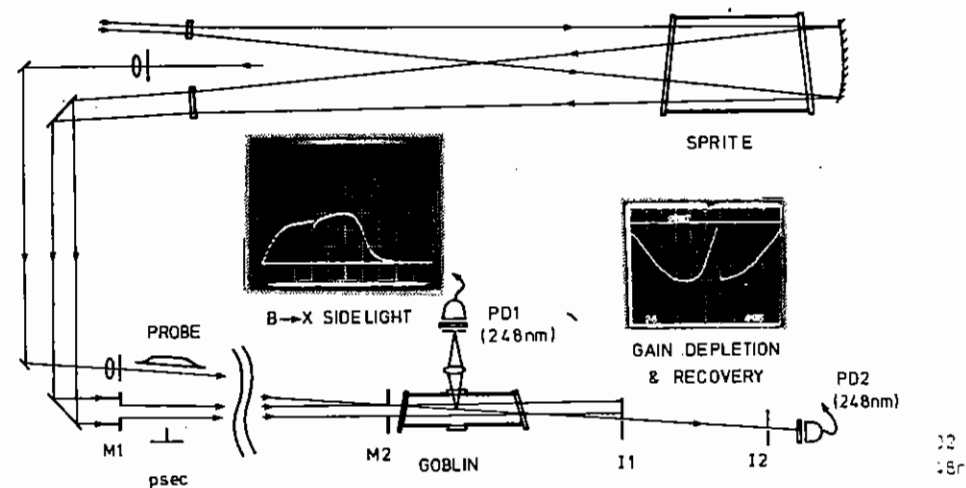
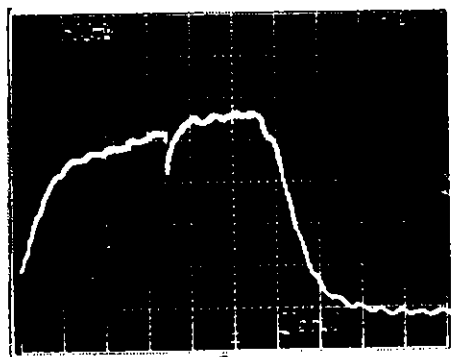


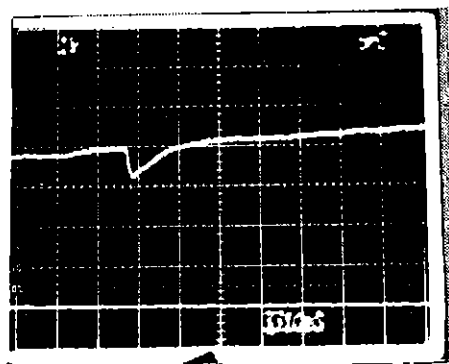
Fig A5.4 Experimental setup used to measure gain and sidelight depletion by picosecond pulses in the Goblin amplifier.

Fig A5.4 experimental setup used to measure gain and sidelight depletion by picosecond pulses in the Goblin amplifier.

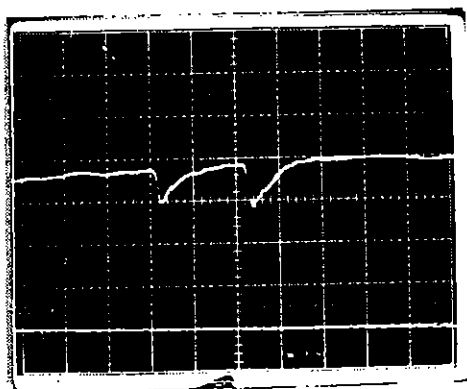


KrF * B → X
Sidelight (248nm)
depleted by psec
pulse.

(a) Single pass
20 ns / div

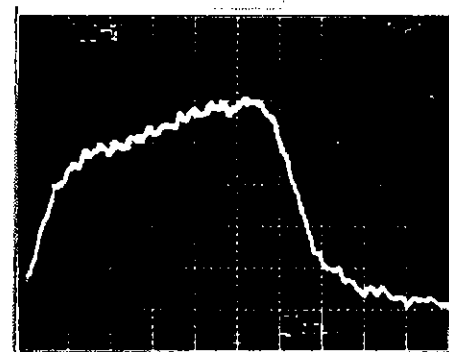


(b) Single pass
5 ns / div



(c) Double pass
5 ns / div

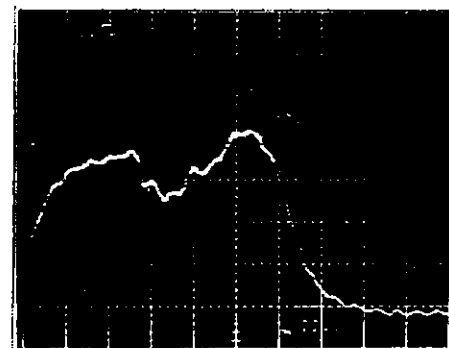
Fig A5.5 Depletion of sidelight fluorescence at 248 nm by picosecond pulses in single and double pass.



a) KrF * C → A
Sidelight at 308nm
Showing no depletion
by psec pulse

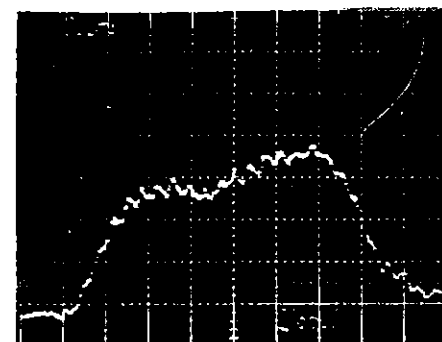
20 ns / div

↑ Position of psec pulse



(b) KrF * B → X Sidelight
depleted by 20ns
pulse in double pass

20 ns / div



(c) KrF * C → A Sidelight
depleted by 20ns
pulse in double pass

20 ns / div

Fig A5.6 Sidelight measurements on the KrF C → A transition at 308 nm showing a) no depletion with a picosecond pulse, c) very small depletion with a 20 ns pulse compared with large depletion at 248 nm b).

5. P H Bucksbaum et al, Opt. Lett, 9, 399 (1982).
6. S Szatman, F P Schafer, Appl. Phys., B33, 219 (1984).
7. A Endoh, M Watanabe and S Watanabe, Opt. Lett., 12, 906 (1987).

A5-3 PICOSECOND RAMAN AMPLIFICATION

N J Everall (RAL), E Fujiwara (Osaka)

1. Introduction

Raman amplifiers can be utilised to increase the peak power of high power excimer lasers, using beam combining and pulse compression in forward Raman light guides. Indeed, we believe that the application of Raman techniques is essential in order to obtain very high powers ($\sim 10^{15}$ W) for KrF laser systems at reasonable cost (see section A5.5⁽¹⁾).

Work at RAL over the last year has been aimed at demonstrating Raman generation and amplification in a variety of gases using picosecond duration KrF laser pulses of ~ 100 μ J energy. Simple theoretical expressions have been derived which predict threshold pump energies for stimulated Raman generation which are in good agreement with experimentally determined thresholds. More recently, Raman amplification has been demonstrated in CH₄ at ~ 3 atm pressure using ~ 4 psec laser pulses (ca 70 μ J/pulse) in which $\sim 70\%$ pump $\rightarrow$ Stokes conversion has been achieved. This is the first demonstration of transient Raman amplification using KrF pulses of this duration and is extremely encouraging as a basis for further development of the front end of the "Super Sprite" laser system (Section A5.5).

2. Transient Raman Generation

Transient stimulated Raman scattering occurs when the dephasing time T_2 of a scattering medium (defined as the inverse of the spontaneous

Raman bandwidth, $1/\pi\Delta\nu_R$) is longer than the duration of the pump and Stokes pulses. Under these conditions, growth and dephasing of the optical phonon in the medium occurs on a timescale comparable to (or longer) than the pump and Stokes pulses. We have recently demonstrated⁽²⁾ stimulated Raman generation using 20 psec (FWHM) KrF pulses in gases where the T_2 time ranged from 1000 psec (H₂, highly transient) through 30 psec (CH₄, transient) to ~ 3 psec (NH₃, near steady state). We showed that a simple theoretical expression could be used to estimate the threshold pump energy E_T required for Raman generation, which was in good agreement with the experimentally measured values:-

$$E_T = (4.2t_p + 22.3T_2) \lambda_p / \gamma \quad (1)$$

In equation (1) above, t_p is the width (FWHM) of the pump pulse (wavelength λ_p), and γ is the Raman gain coefficient, which may be determined from spontaneous Raman scattering data⁽²⁾. It was found that pump energies of ~ 30 μ J and 150 μ J were required to generate Stokes radiation, with a conversion efficiency $> 1\%$, in CH₄ and H₂ respectively at 5 atm pressure. The threshold for CH₄ has recently been remeasured using much shorter ($\sim 3 - 4$ ps pulses) and was still ~ 30 μ J. This is to be expected from equation (1) since $T_2 > t_p$ for both the 20 ps and the 4 ps pump pulses, i.e. the scattering is transient in both cases and so little variation in generation thresholds is expected or observed as the pump pulse is shortened.

It is likely that the proposed Super-Sprite system will be required to operate in two distinct regimes, namely with pulses of ~ 10 ps or ~ 1 ns duration. While CH₄ is the automatic choice for a Raman generator in the psec regime owing to its low threshold, with nsec pulses CH₄ is no longer a transient scatterer. However, H₂ is still a transient scatterer ($T_2 \sim 1$ ns) and has a lower threshold for generation (Table 1). Moreover, decomposition and carbon deposition has been observed previously in CH₄ using $\sim$ ns duration pulses⁽³⁾ whereas no such problem arises using H₂. Thus it seems likely that H₂ would be the preferred Raman medium for nsec pulse work.

Table 1

Raman generation thresholds in H₂ and Methane

Pressure/ atm	T ₂ /ps		Threshold/ μ J			
			tp = 10 ps		tp = 1 ns	
	CH ₄	H ₂	CH ₄	H ₂	CH ₄	H ₂
1	32	915	73	298	476	359
2	31	1268	36	150	242	172
3	30	1258	23	100	150	115
4	29	1140	18	75	130	87
5	28	1008	15	59	105	71

NB All thresholds are calculated energies for 1% pump depletion. Measured thresholds are likely to be a factor of 2 higher.

3. Transient Raman Amplification

While Raman generators typically produce several orders of Stokes radiation from a pump beam, it is possible to selectively amplify 1st Stokes radiation in a transient Raman amplifier with high ($\sim$ 80%) efficiency and minimal generation of higher orders. Figure A5.7 illustrates the experimental layout which we have used to demonstrate transient Raman amplification in CH₄.

The generation of psec KrF pulses has been previously described⁽²⁾ and was unchanged save that the pulses are now usually 3 - 4 psec duration rather than the previous 20 psec. This is a result of the removal of the optical fibre which was previously used to transport pulses from a sync-pumped dye oscillator (746 nm, $\sim$ 3 psec) to the dye amplifier, thereby yielding a stretched (20 psec) chirped pulse. The 746 nm pulses are now transform limited and are not significantly

stretched by subsequent amplification and frequency-tripling to 248 nm.

Light from the tripling crystals was spatially filtered, split (1:2 ratio) and both beams were amplified in a KrF discharge (Lambda-Physik EMG 103) to yield $\sim$ 100 μ J per pulse in the more intense beam.

The weaker beam was focused into the Raman generator (CH₄ at a few atm pressure) and recollimated. The output was dispersed and the 1st Stokes radiation isolated, spatially filtered and routed to the entrance of the Raman amplifier. The more intense pump beam was delayed through a suitable optical path and combined with the Stokes pulse on a dichroic reflector. Both beams (ca 4 mm diameter) were then directed into the amplifier (containing CH₄ at a similar pressure to the generator) and propagated collinearly with no focusing. The amplifier output was dispersed and the pump and Stokes pulses separated.

In practice the gain of the Raman amplifier was adjusted by lowering the gas pressure until self generation of 1st Stokes radiation from the pump beam ceased. Typical operating conditions would be 40 psi in the amplifier and 60 psi in the generator. It is important to note that the pump beam quality was quite poor at the entrance to the amplifier, having a structured profile and hot spots in the beam. Self generation occurred specifically at the hot spots rather than over the whole beam area. This effect is discussed in more detail below.

Stokes gain was measured using PD1 and PD2 (Figure A5.7) while pump depletion was recorded with diodes PD3 and PD4. A zero gain/calibration factor was determined by recording the PD2/PD1 ratio with the pump beam to the amplifier blocked, and then the PD2/PD1 ratio with the pump present could be related to the Stokes gain. A similar procedure was used to calibrate the pump depletion measurements. Also, each diode was calibrated for an absolute energy measurement so that the actual Stokes and pump energies (input and output) could be determined. This also allowed a measurement of the optical loss of the system.

It is also important to note that each diode was covered with a sheet of paper which fluoresced over nsec timescales and so avoided the

problem of diode saturation which is prevalent with psec pulses. The diode outputs were recorded with 1 M Ω input storage scopes.

The optical delay of the pump pulse was adjusted using prism P1 (Figure A5.7) in order to maximise the Stokes gain. In practice, since a 3 psec pulse is only 1 mm "long" in space, careful matching of optical paths is required to optimise amplification! Gain measurements and Stokes output energies were recorded as a function of optical delay of the pump, and also pump depletion measurements were made. In general, the pump depletion measurements were consistent with the measured Stokes output energies when allowance for the optical loss of the amplifier/diagnostics was made. Small signal energy gain measurements were made by attenuating the Stokes input using calibrated neutral density filters. In this manner the Stokes input to the amplifier could be varied from $\sim 0.003 \mu\text{J}$ to $\sim 5 \mu\text{J}$, while maintaining pump inputs at $\sim 70 \mu\text{J}$.

4. Results

Figure A5.8 illustrates the observed variation of Stokes gain with pump pulse delay relative to the Stokes. Note that the time "zero" is arbitrarily defined here since the temporal resolution of our diagnostics is low compared to the pulse durations. The important point is that the measured gain profile has a FWHM which agrees well with the width of the KrF pump pulse (~ 7 ps in this case). This is in good agreement with the measurements of Duncan et al⁽⁴⁾ who found a similar result for Raman amplification in H_2 using ~ 40 psec pulses).

Perhaps the more interesting results are the Stokes gains and output energies which were recorded (Table 2). It is clear that with high Stokes inputs ($\sim 5 \mu\text{J}$) the amplifier was significantly saturated, as indicated by the low gains ($\sim \times 10$) and high depletions (60 - 70%).

It is apparent that the small signal gain of the amplifier was high, several thousand, as would be expected with a device operating near the self-generation threshold. In practice, Stokes inputs as low as $0.04 \mu\text{J}$ led to significant pump depletion, and so a diffraction-limiting spatial filter may be used in the Stokes beam to guarantee high beam quality without significantly reducing pump depletion.

Table 2

Typical gain performance of transient Raman amplifier. Generator contained 60 psi CH_4 , amplifier 40 psi CH_4 . Pump pulse length ~ 4 psec.

Stokes Input/ μJ	Pump Input/ μJ	Stokes Gain	Pump Depletion/%
5	68	11.9	70
3	77	21.6	74
$0.35 \mu\text{J}$	59	112	66
$0.52 \mu\text{J}$	68	97	74
0.04	68	900	53
0.035	62	1207	68
0.0026	68	1834	7%
0.0024	54	1975	9%

NB The Stokes inputs were not reduced sufficiently to realise true small signal conditions ($< 1\%$ pump depletion) but the gains of ~ 2000 will be taken as approximations of the true small signal gain.

5. Theory

It is possible to calculate the small signal gain expected from the amplifier given the pump fluence. The steady state, small-signal power gain coefficient G_{ss} is given by equation 2.

$$G_{ss} = \gamma I_p l \quad (2)$$

where γ is the Raman gain coefficient, I_p is the pump intensity and l is the interaction length. The total power gain is given by $e^{G_{ss}}$. In contrast, in a transient Raman amplifier the optical phonon (which

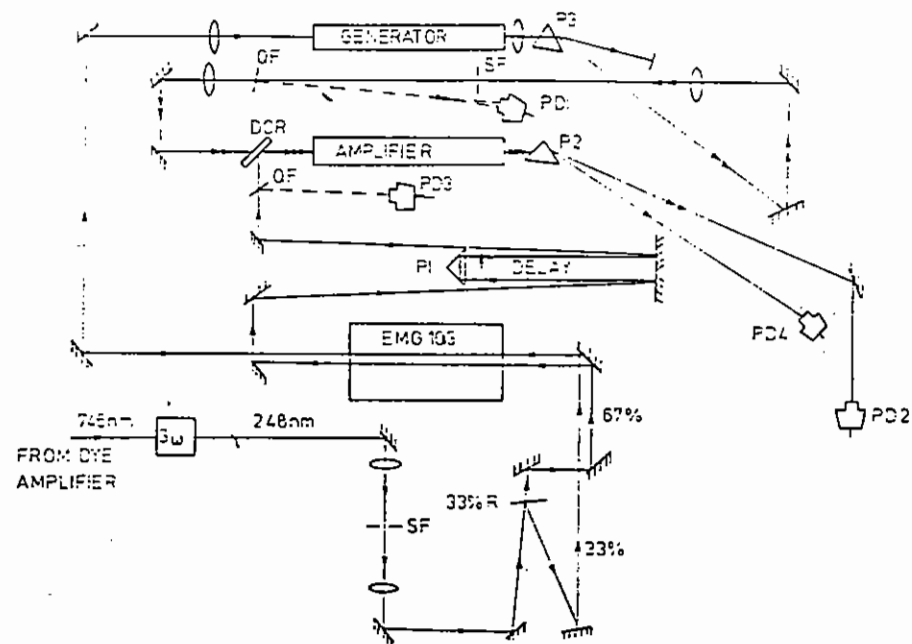


Fig. A5.7 Layout of transient Raman generator/amplifier system.

Key PD - represents a vacuum photodiode used in integrating mode:-
 PD1 - Stokes input to amplifier, PD2 - Stokes output from amplifier, PD3 - pump input, PD4 - depleted pump output, DCR - dichroic reflector, QF - quartz flat, SF - spatial filter, PI - retroprism for pump delay, P2, P3 60° dispersive prisms.
 →: pump (248 nm) beam, ⇄: Stokes shifted beam.

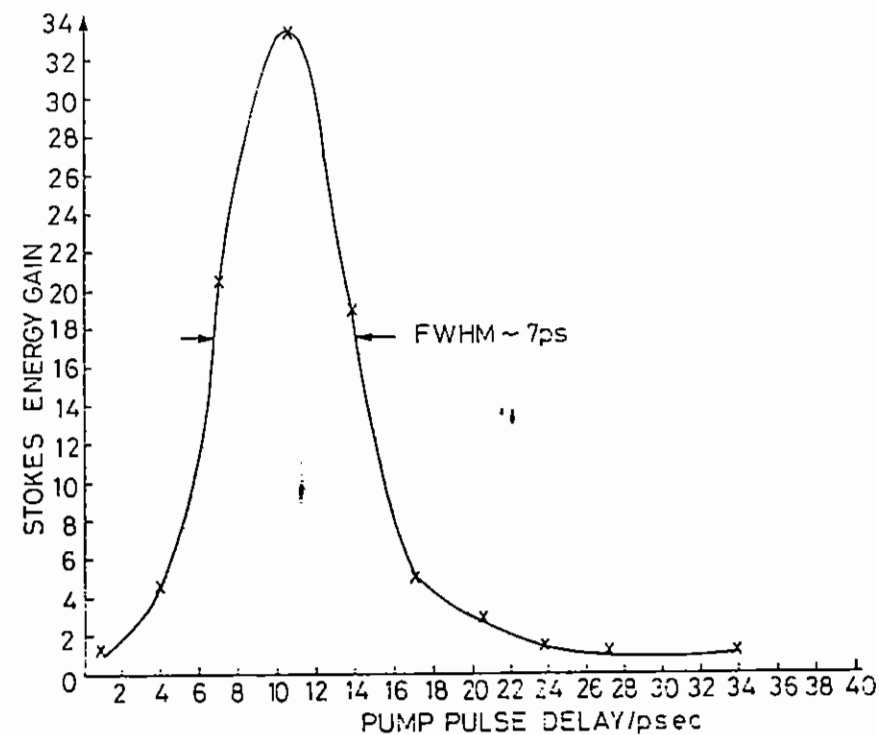


Fig. A5.8 Variation of Stokes energy gain versus pump delay (arbitrary $t' = 0$). Each gain point is an average of 10 measurements with similar pump/Stokes energies. Note that width of gain profile is same as width of pulse from sync-pumped dye oscillator (7 psec in this case).

couples energy from pump-stokes fields) requires a time of the order T_2 to grow to a steady state, and T_2 is long compared to the optical pulse durations. Thus the **transient** gain coefficient G_T is typically reduced by 1 - 2 orders of magnitude compared to $G_{ss}^{(5)}$. Nonetheless, we may estimate G_T from G_{ss} , given the t_p/T_2 ratio, using the results of Carman et al⁽⁵⁾.

For the experiments above, with a methane pressure of ~ 40 psi in the amplifier, γ may be calculated from spectroscopic data⁽²⁾ to be $\sim 6.5 \times 10^{-10}$ cm W⁻¹. Assuming pulse energies of 75 μ J (~ 4 psec duration), and that the beam intensity is **uniform** over the whole profile (see below), then $I_p \sim 1.5 \times 10^9$ W cm⁻² and $G_{ss} \sim 27$ (amplifier length $l = 280$ cm). In the small signal limit the transient energy gain is given by $e^{G_T/2}$, i.e. ~ 3.5 , or **3 orders of magnitude** lower than the measured values. If we assume that the gain measurements are correct, then clearly our initial estimate of pump intensity I_p is in error.

The actual pump intensity may easily be estimated, from the observed small signal gains, using $e^{G_T/2} \sim 2000$. The factor $G_T \sim 15$ would imply a steady state gain coefficient $G_{ss} \sim 500^{(5)}$, which in turn would require an intensity of $\sim 2.7 \times 10^9$ W cm⁻², i.e. an order of magnitude greater than that obtained if a spatially uniform beam is assumed. In fact, the pump beam was not of good quality and Stokes amplification was observed to occur at 'hot spots' in the pump beam rather than uniformly over the whole diameter, which confirms that regions of higher intensity (and hence higher gain) were present.

The intensity of the hot spots in the beam may also be estimated independently by considering the intensity required to reach self generation in the amplifier. The required intensity is given⁽²⁾ approximately (in a plane-wave amplifier) by equation 3.

$$I_p \sim \frac{1}{\gamma l} (25 + 131.5 T_2/t_p) \quad (3)$$

Self generation was observed in the amplifier at ~ 50 psi CH₄, when $\gamma \sim 8 \times 10^{-10}$ cm W⁻¹, from which we estimate $I_p \sim 4.5 \times 10^9$ W cm⁻².

This is not inconsistent with the value estimated from the observed small signal gain (2.7×10^9 W cm⁻²). Thus two independent measurements indicate that the actual intensity was an order of magnitude higher than that expected for a spatially uniform pump beam.

6. Future work and conclusions

It is clearly essential to improve the pump beam quality and to contract the beam to raise the overall intensity and maximise gain. This may be achieved by using a spatial filter/telescope combination. Measurements of pump/amplified Stokes beam quality will be carried out in the near future, and also measurements of amplification in H₂ will be made. Nonetheless we have successfully demonstrated the operation of the shortest-pulse Raman amplifier at these wavelengths, and shown that high intrinsic efficiency is attainable. This work provides an essential foundation for the development of a front end system for Super-Sprite.

References

1. Paper presented at 'O. E. Lase 88' conference, Los Angeles, 10-17th January 1988, Conference proceedings to be published by SPIE, Paper No. 874-07.
2. N J Everall, J P Partanen, J R M Barr and M J Shaw, Opt. Comm., **64** (4), 393 (1987).
3. M J Shaw, J P Partanen, Y Owadano, I N Ross, E Hodgson, C B Edwards and F O'Neill, JOSAB, **3** (10), 1466 (1986).
4. M D Duncan, R Mahon, L L Tankersley and J Reintjes, JOSAB, **5** (1), 37 (1988).
5. R L Carman, F Shimizu, C S Wang and N Bloembergen, Phys. Rev. A, **2** (1), 60 (1970).

A5-4 NEW DIODE FOR THE E-BEAM PUMPED PRE-AMPLIFIER GOBLIN

E Madraszek (RAL)

During this reporting year the front end of the e-beam pre-amplifier Goblin has been re-designed. This work was undertaken in order to improve the shot life of the Goblin foil assembly which had a MTBF of a hundred or so shots compared with Sprite's MTBF of a few thousand shots. Work is almost complete and operation is aimed for the middle of April.

The redesign can be considered as a smaller version of the Sprite system but with the gas cell pumped from two sides instead of four. Where possible the technology and experience gained in the development the Sprite system has been used. This is in keeping with the design philosophy of having a standard set of building blocks which can be used in the development of future configurations.

Goblin remains unchanged up to the SF_6 laser triggered switch. After the switch the pulse forming line splits into two water filled transmission lines. These lines are of the same design as those used on Sprite. Each transmission line passes through a vacuum feed-through into the vacuum vessel. The inner conductor of each transmission line terminates at the cathode of the diode assembly.

Figure A5.9 shows a cross section of the Goblin e-beam vessel and laser cell. Again technology gained from the development of Sprite has been used in the design of the diodes. Likewise the laser cell is of a similar construction to that the Sprite but on a much smaller scale.

Figure A5.10 shows how the laser cell, anode plate and current return vanes when assembled from a separate sub-assembly, which is slotted into the vacuum vessel. This allows easy access to the anode foils and the foil of the laser cell which may require replacing after continued periods of operation.

A5-5 SUPER-SPRITE DESIGN

M J Shaw (RAL)

During the past year the various options for the second phase of high power laser development at the CLF have been reviewed and it has been decided that the exploitation of KrF technology based on the Sprite prototype represents the most attractive alternative. The specifications for the new laser (at present known as "Super-Sprite") are shown in Table 1.

Table 1

Super Sprite Specifications

1. Energy on Target (ns pulse)	> 7 KJ
2. Wavelength (Raman shifted KrF)	268 - 277 nm
3. Pulse Duration	3 ps - 12 ns
4. Peak Power on Target	> 100 TW
5. No. of beams	12/24
6. Beam diameter	200 mm
7. Shot Rate	up to 10 per hour

The unique feature of the laser, compared with other large KrF installations such as the Aurora System at LANL⁽¹⁾ is the proposed use of Raman amplifiers as a means of staging the multiplexing. This technique, which was pioneered at RAL⁽²⁾ is shown in principle in Figure A5.11. Energy is extracted from the KrF amplifiers by a series of pulse trains, the separation between the pulses being less than the storage lifetime of the KrF molecule. Under these conditions it has been shown⁽³⁾ that short duration pulses can be amplified to several times the saturation energy, which in KrF is 2 mJ cm^{-2} . In order to obtain an efficient energy loading on the KrF aperture ($\sim 1 \text{ J cm}^{-2}$) several hundred pulses are required, for example in the final stage of Super Sprite 24 pulse trains of 12 pulses per train are amplified

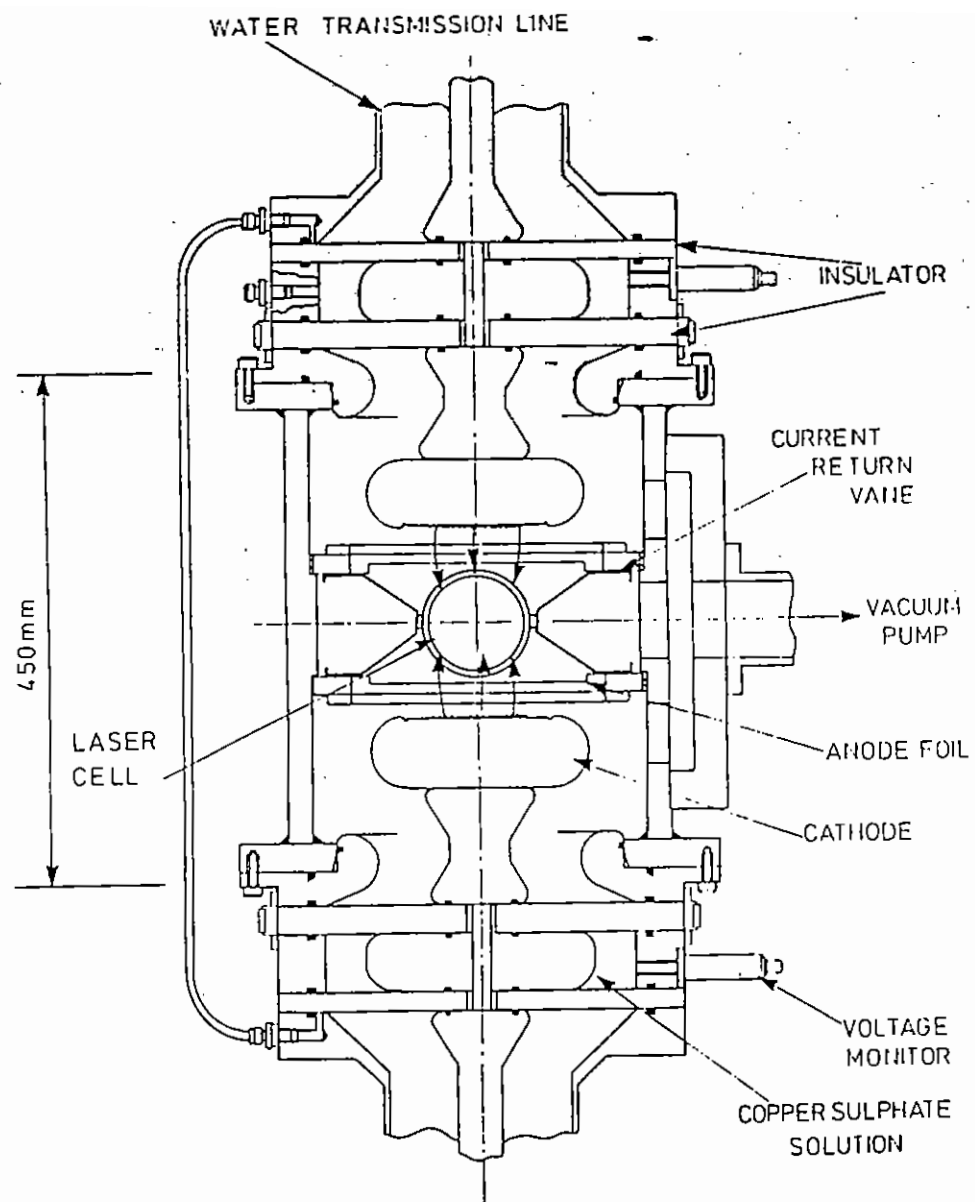


Fig. A5.9 Cross section of the new dual electron-beam-pumped Goblin laser.

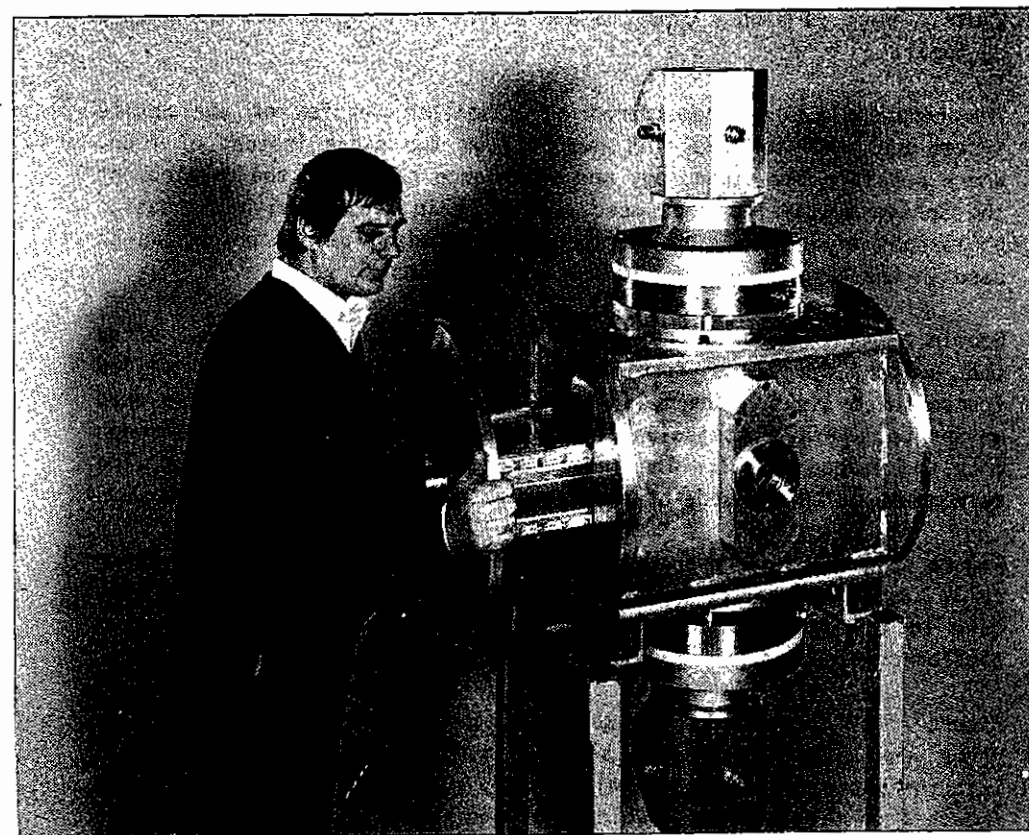


Fig. A5.10 Goblin being assembled showing easy removal of foils and laser cell.

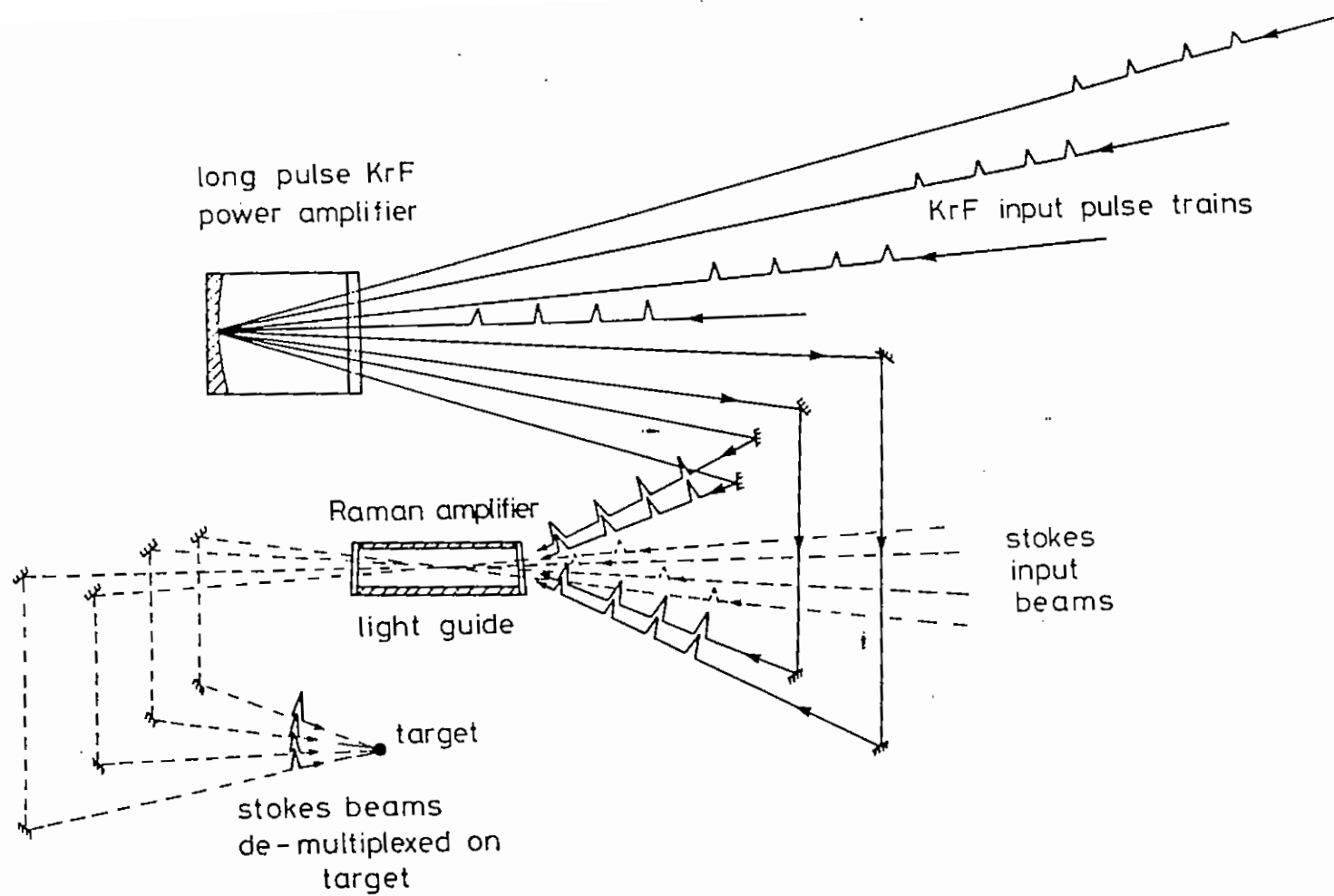


Fig A5.11 Conceptual diagram of a staged, multiplexed laser system amplifying a series of pulse trains and combining pulses from different pulse trains in a forward Raman amplifier with pump beams confined in a light guide.

giving a total of 288 output pulses. The pulse trains, however, may be de-multiplexed and used to periodically pump a Raman amplifier. This in turn can be extracted by a small number (12 for Super Sprite) of single-pulse Stokes beams which can then be de-multiplexed on target.

The use of Raman staging thus provides a means of vastly reducing the complexity of the proposed KrF system. However, it also confers a number of extremely useful additional advantages. Firstly, and most importantly, the Raman stage provides a means of isolating the target from amplified spontaneous emission from the KrF amplifiers. Secondly, by using a Raman medium contained in a light guide, the pump intensity may be averaged and the beam quality of the Stokes beams are decoupled from the pump beam quality and can be near diffraction limited. Additionally, the Raman stage also provides time averaging over the KrF pump duration and should thus lead to automatic beam balance, pulse shaping may be undertaken and in the picosecond region pulse compression due to group velocity dispersion in the light guide is obtained.

The overall system layout shown in Figure A5.12 thus comprises a number of KrF amplifier stages which essentially act as "flashlamps" to pump a number of Raman amplifiers. The gain staging is approximately x10 per stage between the KrF modules and x100 per stage between the Raman modules (high gain and high efficiency being simultaneously obtainable in Raman amplifiers⁽²⁾). The first two KrF stages comprise the existing "new Goblin" and Sprite which each amplify 6 trains of 12 pulses = 72 pulses in 72 ns. Two new KrF amplifier modules are required, "Titania", a 4-sided e-beam pumped system with a run time of 144 ns and an aperture of 580 mm giving an output energy of 1.5 kJ and the final stage, "Oberon" pumped by 12 electron beams with a run time of 288 ns, a segmented aperture of 1.5 m and an output energy of > 15 kJ. When complete, Oberon will be the World's largest KrF laser in aperture, energy and power. The energy supply is obtained from 6 Marx generators each giving over 100 kJ at 2 MV. The generators are based on a Sandia design - (Hermes III) and Sandia Labs have kindly donated sufficient capacitors to build two Marxes to this pattern. Design of the Marx generators is at an advanced stage which will allow a prototype to be built so that pulse forming lines, switches and diodes may be tested. Figure A5.13 shows an impression of how Oberon may look when completed.

Due to the "beam clean-up" effects of the Raman amplifier, the quality of the KrF beams is not so important and this fact may be exploited to save costs of the KrF optics. Some topics under active consideration at present include:- window pane segmented windows for Oberon, plastic film windows, float glass flat mirrors, lightweight composite mirrors and spin coatings for high reflectors.

The basic idea of Raman beam combining has been tested previously at RAL for pulses of several nanoseconds duration⁽²⁾. The multiplexed amplification of nanosecond KrF pulses has also been demonstrated⁽⁴⁾. Over the past year we have shown that single picosecond pulses can be amplified to high energy in KrF Section A5.2 and that picosecond Raman pulses can be readily generated and amplified Section A5.3. In the coming year we plan to demonstrate Raman beam combining of multiplexed picosecond KrF pulses and to use the resulting output for short pulse target interaction experiments. When this is achieved all the important elements of the Super Sprite system which have already been demonstrated singly will have been demonstrated in a completely integrated system which is a scaled down version of the final laser system.

References

1. L A Rosocha et al, Fusion Tech., **11**, 497-531 (1987).
2. J P Partanen and M J Shaw, J. Opt. Soc. Am., **B3**, 1374-1389 (1986).
M J Shaw et al, J. Opt. Soc. Am., **B3**, 1466-1475 (1986).
3. C J Hooker et al, Annual Report to Laser Facility Committee RAL-87-041 Sect. C2.2, 226 (1987).
4. C J Hooker et al, Optics Comm., **65**, 269-274 (1988).

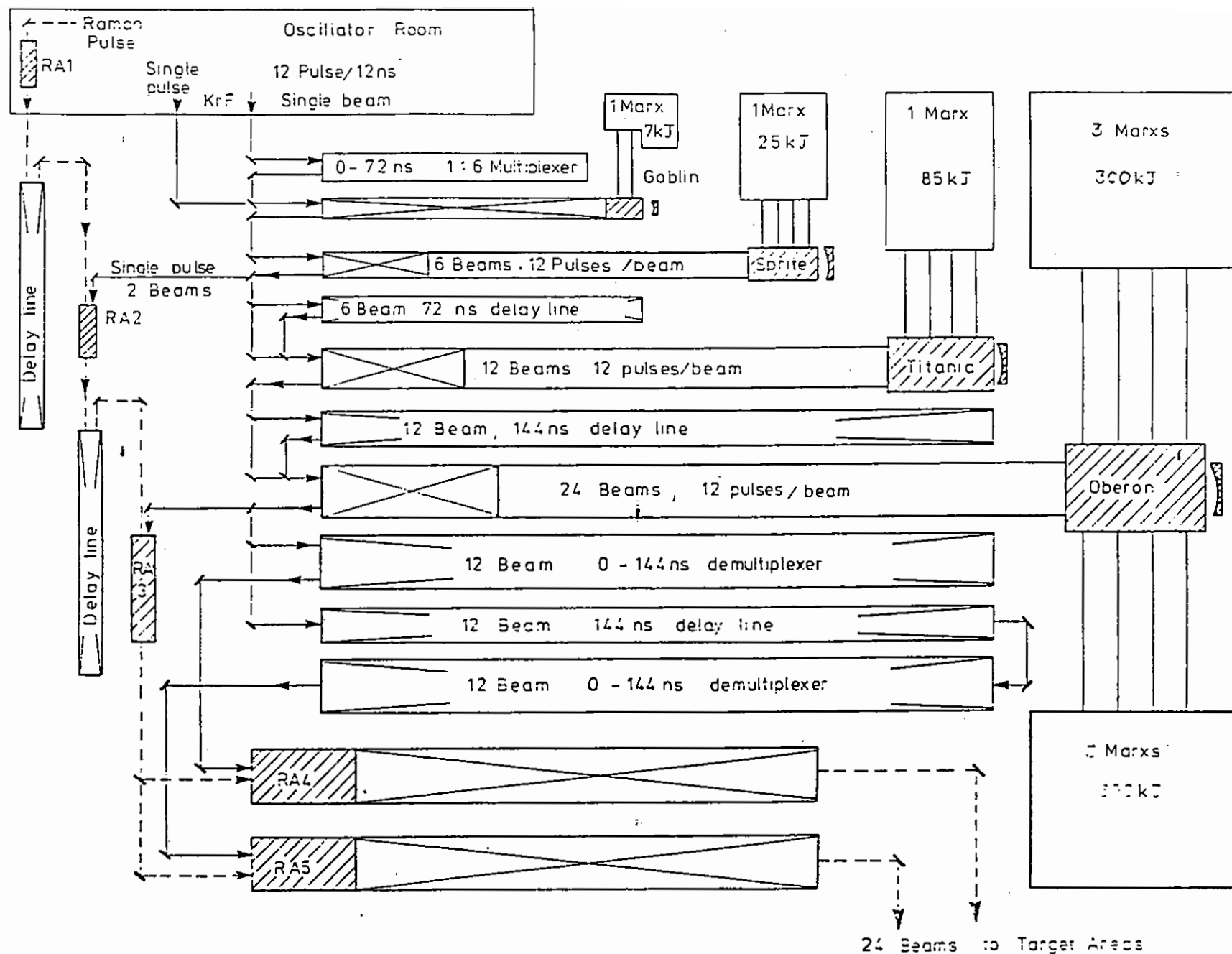


Fig A5.12 Optical layout of the proposed Super-Sprite laser system.

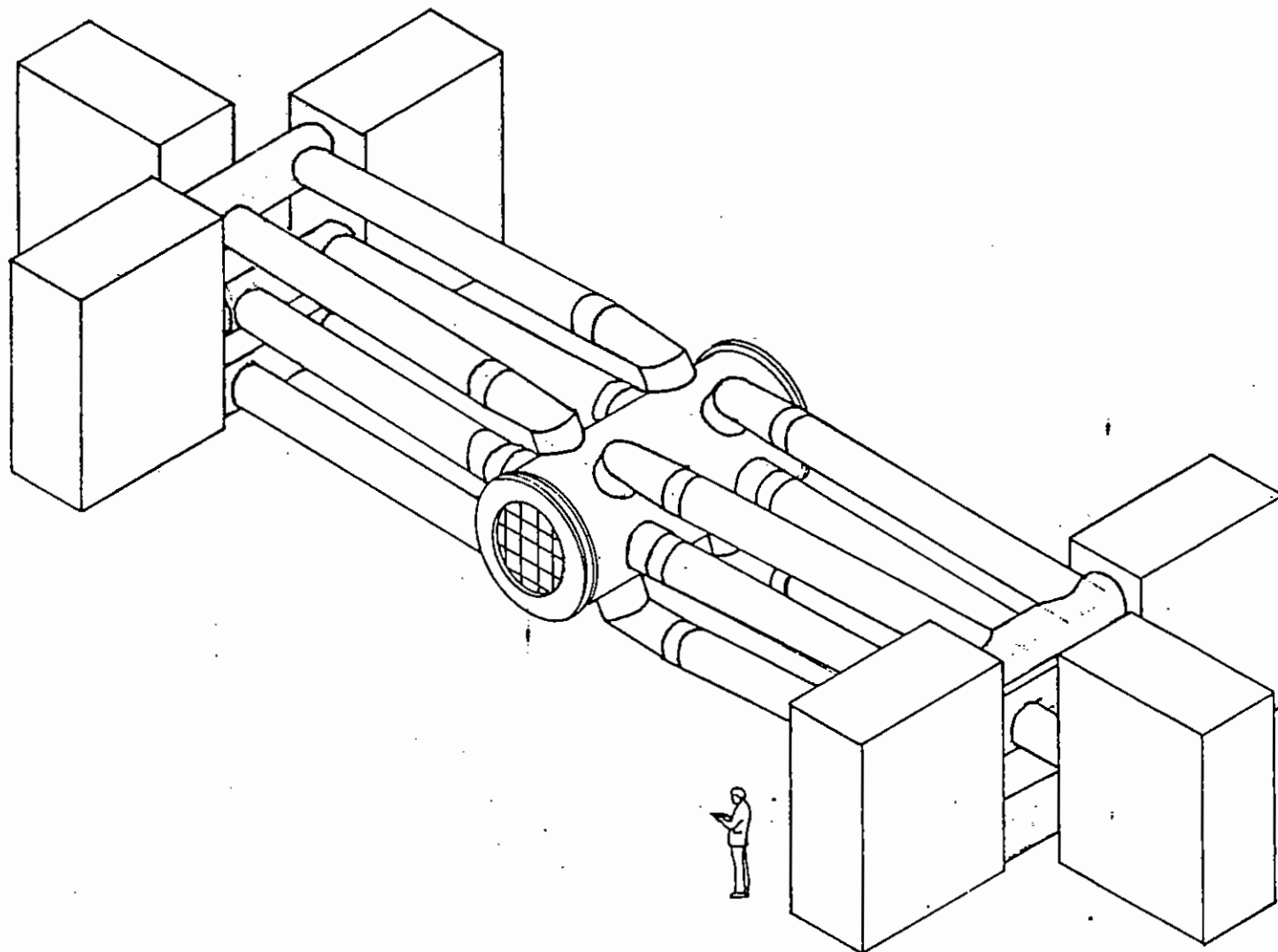


Fig A5.13 Artist's impression of the final amplifier for Super-Sprite
- Oberon pumped by 12 electron beams powered by 6 separate
Marx generators.

A6	VULCAN OPERATION AND DEVELOPMENT	Pages
A6-1	Introduction and Operational Statistics	135 - 136
A6-2	Glass Laser Development	136 - 137
A6-3	Computer Control and Data Acquisition	137 - 139
A6-4	Target Fabrication and Development	139 - 140
A6-5	Target Areas	140 - 141

Editor: P T Rumsby/J E Boon

A6-1 INTRODUCTION AND OPERATIONAL STATISTICS

P Rumsby (RAL)

Vulcan operated intensively for a full programme of experiments up to the end of February 1988 at which point operations ceased and restaging and upgrade work commenced. With 3 main target areas available a pattern of operation into 2 whilst preparing the 3rd has now become standard practice leading to more effective use of laser time. During the year a 4th target irradiation chamber (TA4) offering low energy single beam laser shots for x-ray source work and diagnostic testing and development was commissioned. This has led to more efficient use of shots into the main target areas. Details are given below.

Shot statistics for the 11 month period reported here show that total shot rate has increased significantly due in part to the additional target area. 1979 high power shots were delivered to the 3 main target areas in roughly equal numbers. Over 6000 additional low power laser shots were fired for test purposes, target area alignment and timing and TA4 operations. Failure rate for high power shots was 4%.

A 4 or 5 week experimental run cycle has been maintained leading to a total of 35 weeks of operation of VULCAN for experiments and a total of 62 target area experimental weeks (in 11 months). TA4, using only low power laser shots, has operated in parallel with the main target areas without significant disturbance to the experimental programmes.

Considerable design, development and procurement of components has taken place during the year in preparation for the restaging and upgrading of VULCAN planned to be completed by mid May 1988. When complete this major development will permit operation of VULCAN at twice its present level with over 1 kJ of green energy in 6 beams available on target in all target areas. Complete details of the new scheme will be reported next year. Details of completed development work on rod and disc amplifiers is discussed below.

A6-2 GLASS LASER DEVELOPMENT

A6-2.1 Ø 108 mm DISC AMPLIFIER DEVELOPMENT

I Ross, J E Boon and P Gottfeldt (RAL)

A new design of disc amplifier has been discussed (Ref. RAL Annual Report 85-067) for a beam size of Ø 150 mm, and has shown excellent performance. This 'box' design has been extended to the existing aperture size of Ø 108 mm and a prototype designed and built. Figure A6.1 shows details of this amplifier which has eight Ø 25 mm bore flashlamps as compared to the twenty Ø 15 mm bore lamps in the existing cylindrical amplifiers. Gain measurements were made and are presented in Figure A6.2 and show consistently higher gains than the current amplifiers at similar voltages but at reduced total energy storage (180 KJ is compared to 200 KJ). Also shown is the gain measured on the same amplifier after over 800 full energy shots without disturbance. This showed a small decrease in gain which returned to close to its initial value following a standard cleaning process on the amplifier.

This Ø 108 Box amplifier design will be used in the Vulcan upgrade planned for Spring 1988.

A6-2.2 NEW ROD AMPLIFIER DESIGN AND TESTING

D Pepler, S Hancock and J Boon (RAL)

A new design of Ø 45 mm rod amplifier capable of producing a gain of 10 is required for installation during the Vulcan Laser upgrade starting Spring 1988. This gave the opportunity to design out a number of the problems that have been found with the present rod amplifiers during 10 years of continuous service. These problems involved, the assembly and orientation of the rods in the amplifier, difficulties in inspecting and repair of faults, water leaks and resulting electrical faults at the termination of the flashlamps. It was considered desirable to provide a pre-adjusted kinematic mount to allow quick replacement of spare amplifiers, and improved water circulation to prevent trapped air pockets in the amplifier.

All these features are overcome in the new design, see Figure A6.3, by having a quickly removable lid, split ceramic reflector and a sealed

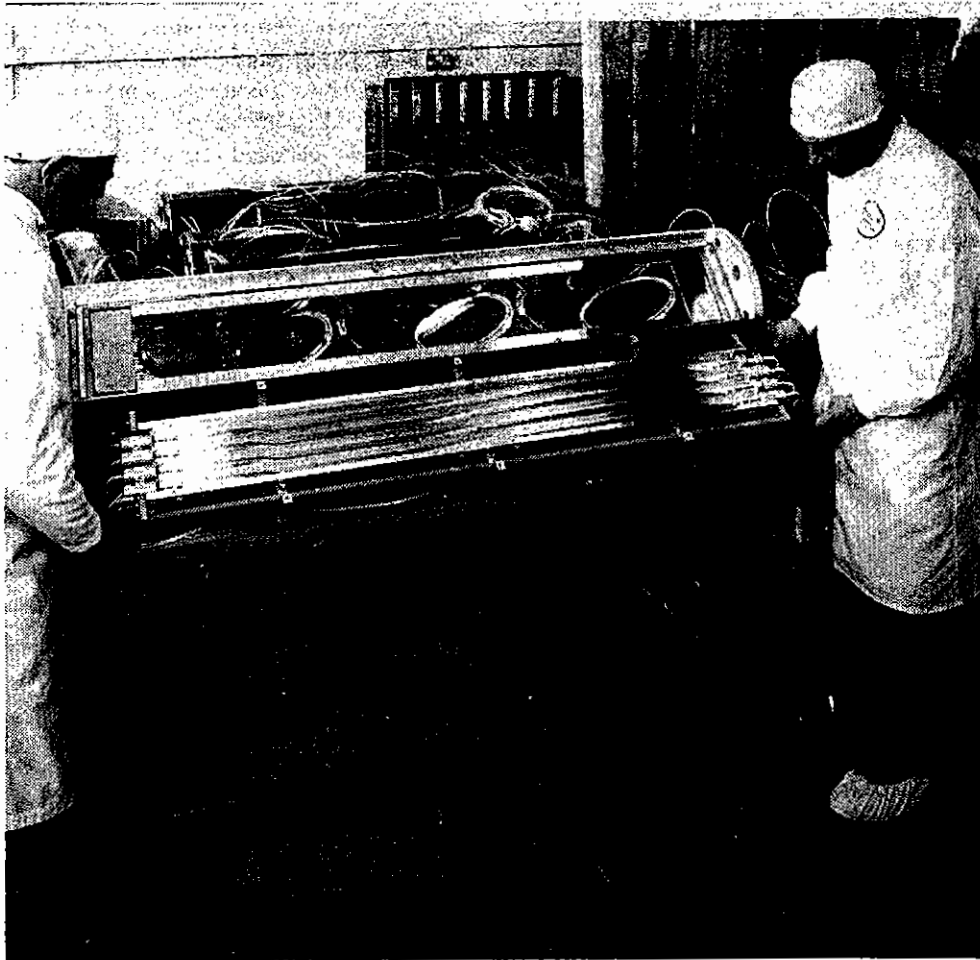


Fig A6.1 New 108 mm ϕ disc amplifier "box" design with lamp assembly removed.

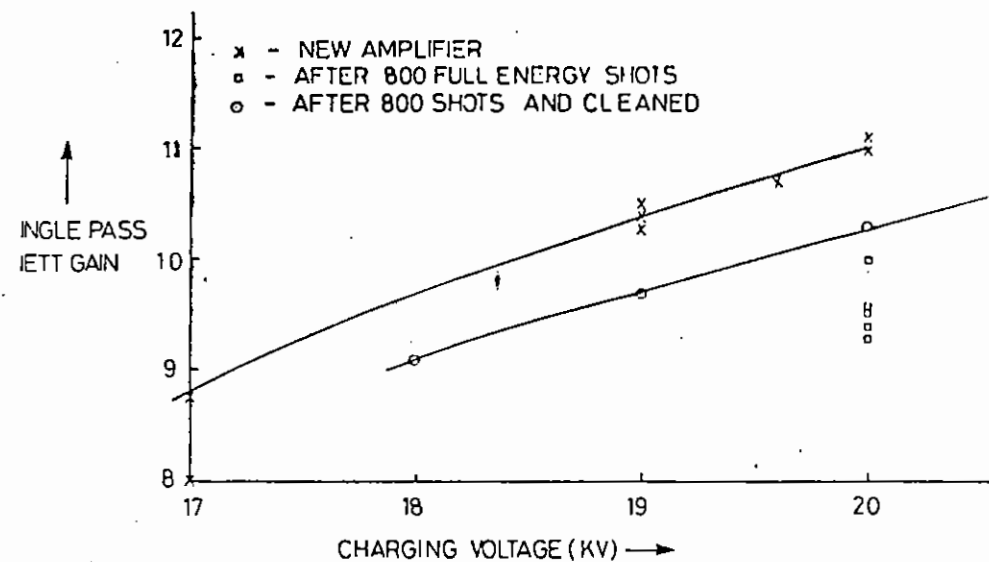


Fig A6.2 Gain measurements obtained from new ϕ 108 mm disc amplifier.

end termination to flexible lead to each end of the flashlamp similar to that used at present on the flashlamps of the disc amplifiers.

Gain measurements were made using a CW YLF laser optically chopped at a frequency of 6 KHz, passed through the rod, and focused onto a photodiode. The output from the diode is shown in Figure A6.4.

Trace 1 shows the diode signal at 2 mV/div and trace 2 the same signal at 10 mV/div, as the amplifier is fired, the fluorescence produced a large background envelope superimposed on which is the amplified pulse train.

The gain measured by this method is simply an ON/OFF gain, the real OUTPUT/INPUT gain must take into account the passive losses which in this particular rod were of the order of 10%.

With adjustment for this loss, a real active gain of 11 was obtained, running the amplifier at 2.4 kV, thus fulfilling the above requirements.

A6-2.3 VACUUM SPATIAL FILTER PUMPING TESTS

C N Danson, E Madraszek and A R Damerell (RAL)

To enable Vulcan to operate at the highest fluences it is necessary to minimise laser damage to critical components in the system. In the current upgrade restaging of the gain through the system will eliminate many of these critical points, but problems have been encountered with laser induced damage to the final vacuum spatial filter (VSF) lenses. The damage has only been occurring on the output surface of the input VSF lenses (the vacuum side), even though both surfaces are subject to approximately the same flux.

It was believed that the coating must be subject to some form of contamination. The vacuum is generated using an oil-free ion-pump system which should not produce contaminants and the VSF vessels have been under vacuum for about 2 years so that out-gassing should not be a problem. One possible theory, and a problem encountered on Shiva at LLNL, was metallic contamination produced from plasma formation at the pinhole. The VSF's were operated at $\approx 10^{-7}$ mbar, giving very little resistance to particles ejected from the pinhole towards the VSF lenses. The VSF's could be operated at a higher pressure, to reduce

the atomic mean free path, and hence reduce the amount of possible contamination reaching the lens.

For a long term test the six output spatial filters were operated using three different pumping schemes so that comparisons between them could be made. The three schemes were: oil-free ion-pumps operating at $\approx 10^{-7}$ mbar; an oil-free Turbo-pump backed by a Rotary-pump with a fore-line trap operating at $< 10^{-4}$ mbar and a two stage Rotary-pump with two fore-line traps operating at 10^{-3} mbar. The VSF's were operated in this mode for two experimental periods. There was no significant difference observed between the schemes in damage to the lenses, although there was no further contamination observed in using a Rotary-pump instead of the ion pump. These results meant that for the upgrade where extra VSF's are to be installed it will be possible to operate with the much cheaper and simpler fore-line trapped Rotary-pumps.

Tests were also carried out to find the highest possible vacuum pressure which still maintain good beam quality. On a full energy shot at 1 mbar beam break-down occurred scattering the beam and even causing significant damage to components further down the chain. The pressure in the VSF was then reduced. The VSF's had to be operated at pressures of below 10^{-2} mbar before a good quality beam was observed in transmission.

A6-3 COMPUTER CONTROL AND DATA ACQUISITION

C J Reason, D A Pepler (RAL)

The GEC 4080 computer, DEC computers and their associated CAMAC system have given a reliable service during the year. There have been changes to the software and hardware to allow for changes in the laser and the target areas. In particular the CAMAC system has been reorganised to gain more isolation using fibre optics and the microdensitometer has been removed from the GEC 4080.

The two CAMAC crates in the laser room were converted to serial fibre optic and the old capacitor room and microdensitometer room were removed from the system. The new capacitor room was added also via a fibre optic link. The current CAMAC system now consists of pairs of crates in the target areas, the laser room and the control room, with a single crate in the new capacitor room. Crates in each location are

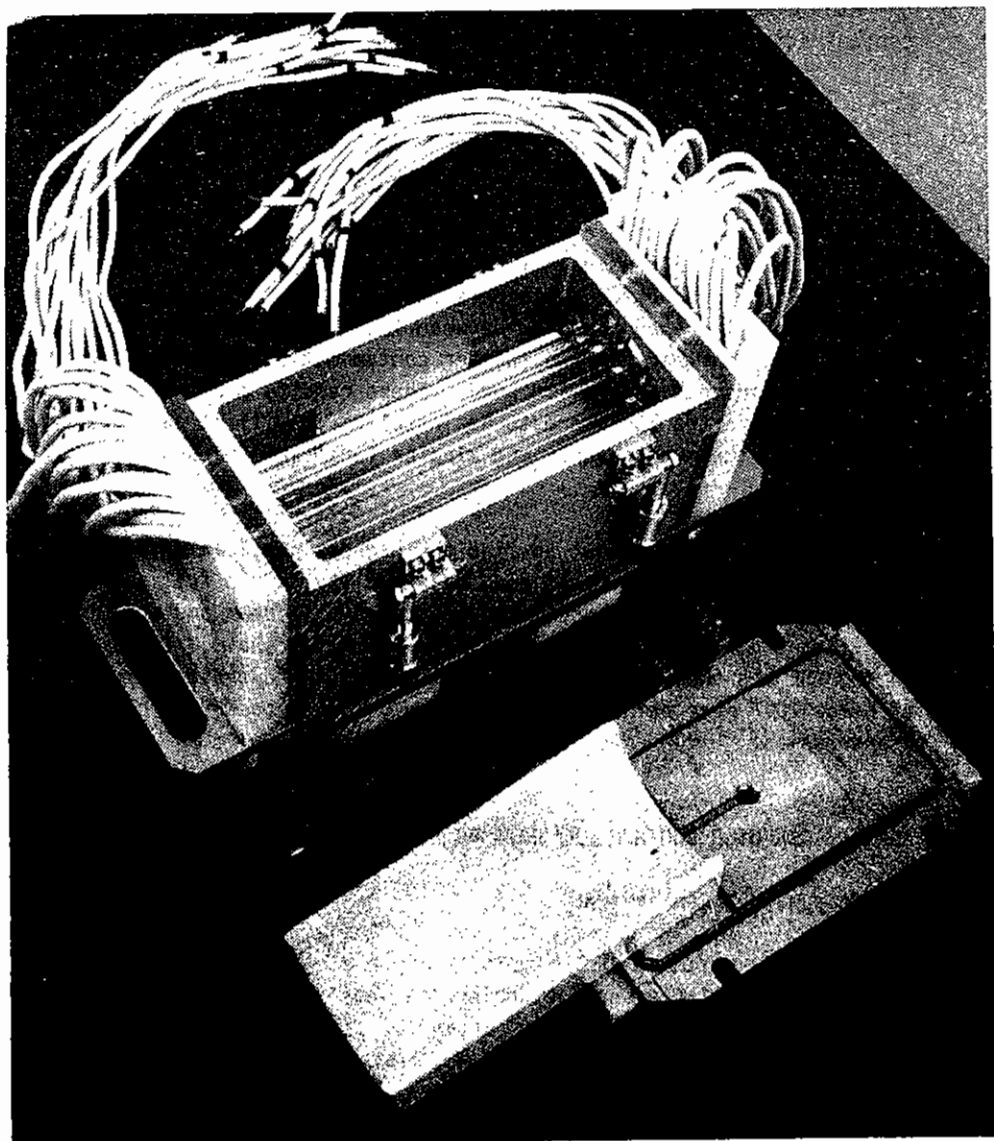


Fig A6.3 New 45 mm ϕ rod amplifier design with lid and upper half reflector removed revealing the flash tubes and rod.

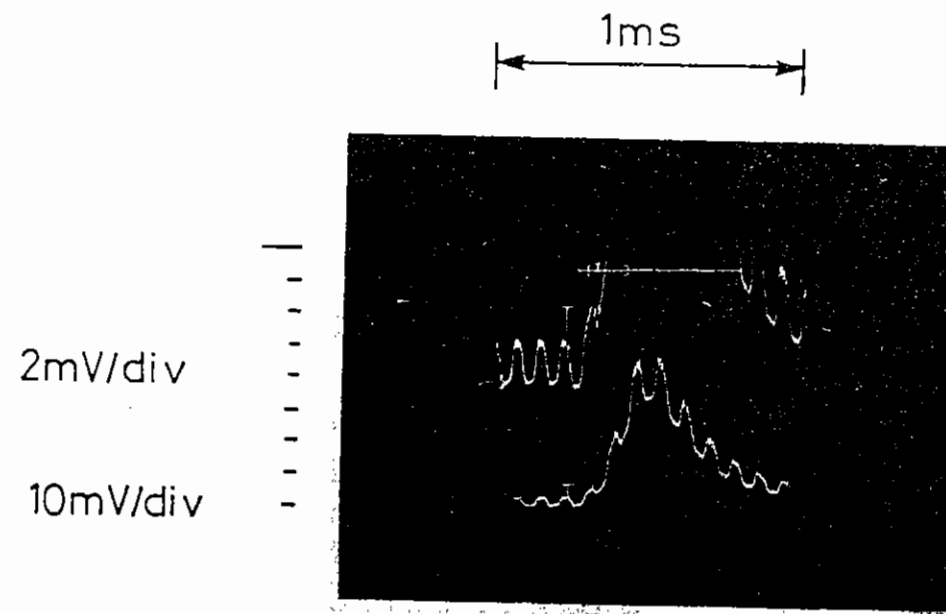


Fig A6.4 Photo diode output with combined fluorescence and pulse train.

linked electrically and locations are linked by fibres. The only parallel link (128 way electrical link) is between adjacent crates in the control room.

A CAMAC crate has been installed in the new capacitor room and contains a DEC falcon computer. This will be used to monitor the capacitor banks during charging and if any fault is detected, dump the system. The overall control will be carried out by the GEC 4080 computer. This system has already been used to test half shell assemblies under the control of the operator in the control room.

The touch sensitive screen, which is part of the operators console, has been brought back into use. The software has been removed from the main laser control program and put in an synchronous process, giving the operator a faster response and better checking that the action has been carried out. The new capacitor bank, mirrors etc will be controlled through this screen.

The microdensitometer and its CAMAC system has been removed from the control of the GEC 4080 and is now controlled from an IBM PC clone (Tandon). The computer has its own dot matrix printer and contains a framestore and colour monitor to provide graphics display that matches the scanned input image. This allows 512 x 512 resolution but will only display 256 (8 bits) grey level out of the 1024 (10 bits) that can be scanned. This however is not a problem as it more nearly matches the response of the film. The data can be stored on the hard disk, sent to the main frame or the user can take it back to his laboratory on a floppy disk. There are a number of software and hardware upgrades planned for this system in the next year.

A number of IBP PC clones (Tandon PCA 20) have been installed at the CLF, in particular in the LSF, to control specific experiments or equipment. These include a streak camera and a photon counting system. The use of IBM PC clones for such tasks is providing an increasingly attractive and cost effective solution to control and data acquisition tasks and the numbers are likely to increase in the next year.

A6-4 TARGET FABRICATION AND DEVELOPMENT

C Brown, B Child, A Hussein, S Mitchell, N Prior, P Rumsby (RAL)

This year has been a particularly busy one for the target preparation group, as in addition to supporting up to 3 simultaneous experiments on VULCAN and SPRITE, the CLF high power laser programme, targets have also been prepared for various other programmes and laboratories. Numerous wires, fibres and spheres have been supplied for x-ray source and microscopy experiments on LSF laser systems, while coated and mounted fibres, spheres and other devices have been fabricated for collaborative XUV laser experiments at ILE Osaka, x-ray generation experiments at Australia National University and transport experiments at Ecole Polytechnique.

Work has been undertaken to develop novel stripe target schemes suitable for both the XUV laser programme and long scale length interaction studies. Experimental measurements have shown that with stripes mounted on thin (0.1 μm) polymer foils lateral plasma expansion is impeded by interaction with the tenuous plasma from the support foil. An ideal stripe target would be 25 - 50 μm wide, about 100 nm thick and up to 20 mm long with ideally no supporting structure of any sort. Figure A6.5 shows results of initial attempts to fabricate a stripe target approximating to this specification. Material is evaporated through a slotted mask onto a 0.2 μm polymer film placed across a suitable holder. After coating, the polymer is removed by R. F. oxygen plasma etching leaving a free standing stripe supported by legs at 3 mm intervals, as shown. Aluminium stripe widths of 100 μm with thickness of 300 nm have been achieved to date. Work is in hand to reduce both these dimensions by factors of 2 to 3.

The Institute of Laser Engineering at Osaka has supplied empty glass shells for collaborative LHART type implosion experiments at the CLF. A considerable number of these fragile ultrahigh aspect ratio (~ 400) shells have been processed successfully. Following a prescription supplied by ILE such shells have been filled with D_2 to pressures of 5 Bar and mounted for experiments with yields greater than 90%.

Various new coatings have been developed and used in targets during the year. Using sputtering techniques 1 μm thick depleted Uranium layers

have been successfully applied to rotated carbon fibre backlighter targets for absorption spectroscopy experiments. Sputtering of multi-component alloys has been investigated further with work concentrating on using synthesized sputter targets made from compressed powders of both Nickel and Copper and Nickel and Chromium in order to produce thin film thermocouple devices for plasma calorimeters.

For high quality CH coatings on shells and foils Parylene E is now used routinely. This material is significantly less crystalline and gives much better surface quality than Parylene N for all applications.

A6-5 TARGET AREAS

A6-5.1 TARGET AREA 4 COMMISSIONING

A Damerell, E Madraszek, N Rizvi (RAL)

Following recent demands from the biological user community for a high repetition rate source for x-ray microscopy studies, a new target area, TA4, has been designed, installed and commissioned. This facility not only aids biological work but is also useful for the testing and calibration of diagnostics such as streak cameras, CCD arrays etc.

One of the problems encountered in previous microscopy work in TA2 was the long time interval between loading of a live sample and its exposure to x-rays, this being mainly caused by the long pumping down time of the target chamber and also a possible wait for a main laser shot. Since it is desirable to make an exposure as soon as possible after loading a living specimen, one of the main aims of TA4 was a quick shot rate and fast pumping time. These and other features were incorporated into the design of the area following liaison with the user groups.

The area was constructed in May 1987 in one corner of the laser room, LA3. This allowed access to TA4 from the main control room. While the target chamber was being detailed and put out to manufacture, work began on completing the optics set-up such that experiments could start using a borrowed chamber from SPRITE. Since initial work was to be done in the green, an air-cooled argon ion laser was installed and target alignment could be performed using this and a retro TV alignment system.

To facilitate a two minute shot cycle, the beam was picked off after the Pockels cell at the output of the long pulse rod amplifier chain. After collimation, the beam was sent directly to TA4, where after reduction in size, a 25 mm diameter beam was focused onto target using a f/10 or f/6.8 spherical lens.

Initial experiments began in late August. These were performed with the borrowed chamber and used the frequency-doubled beam to produce x-rays. The new target chamber (see Figure A6.6) arrived in December and was commissioned. It was found that conversion to green was limited by the divergence of the beam in the target area and so TA4 is currently being run in the infra-red. Successful experiments have been performed with this arrangement both in x-ray microscopy (Section A1.15) and diagnostics development (Section A7).

There are plans to use a larger diameter beam with lower f/number lenses to achieve higher irradiances on target, and the possibility of using short pulses is also being investigated.

A6-5.2 VULCAN TARGET AREA OPERATIONS

M Grande (RAL)

The period from April 1987 until Vulcan shut down for upgrade at the end of February 1988 has seen the steady exploitation of the flexibility of the target areas. In all TAE has run scheduled experiments for 22 weeks over this period, TAW for 19 weeks and TA2 for 21 weeks. The even spreading of experiments between the target areas in this way makes for easier changeovers between experiments and increases the time available for setting up more complex systems. Of note in this respect has been the modification of the TAE line focus configuration to produce foci of lengths 2, 7 and 14 mm at the centre of the target chamber. This considerably eases the alignment of diagnostics. In addition TAE has also been used in the cluster beam configuration. TAW has been used in a variety of setups ranging from a single hot beam to the full 12 beam arrangement. With the exceptions of the beat wave experiment, TA2 has been used for simple one and two beam experiments, for which it is particularly well suited.

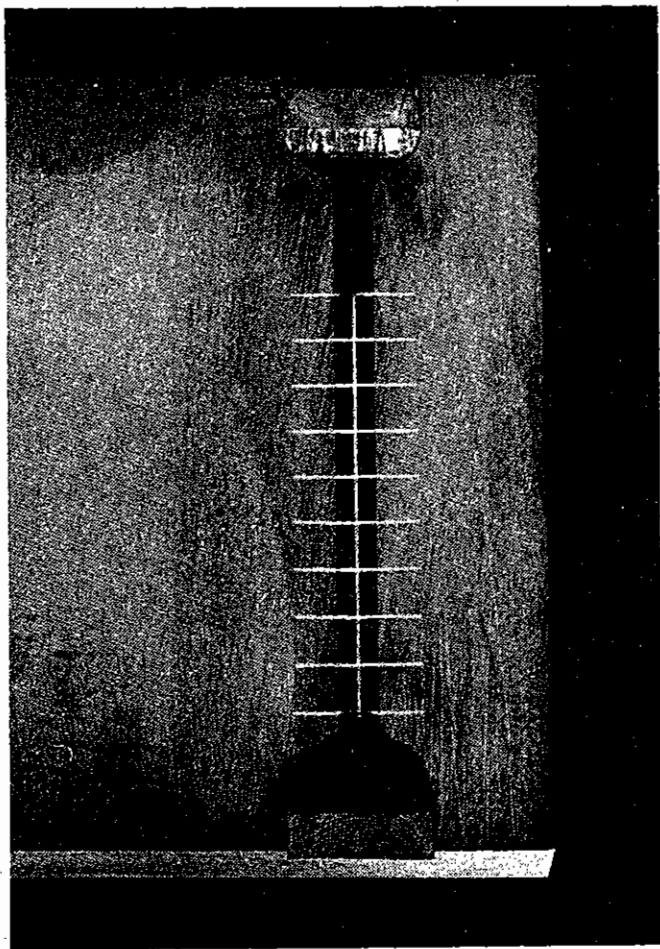


Fig A6.5 Supported stripe target for XUV laser experiments.

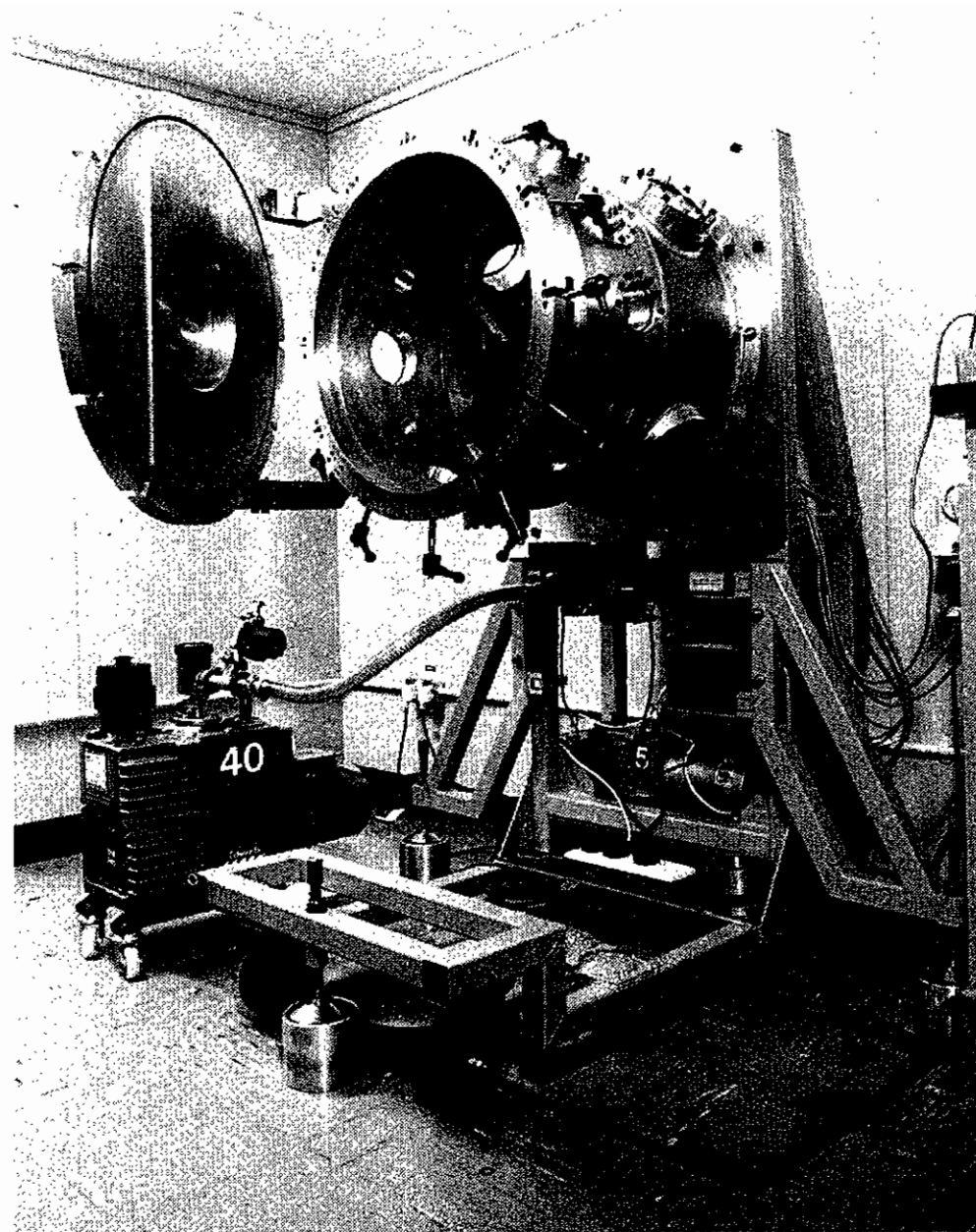


Fig A6.6 Target Area 4 target vessel.

A7	INSTRUMENTATION	Pages
A7-1	An Active Crystal Spectrometer using a Reticon Detector	143 - 145
A7-2	Active Pinhole Camera using a CCD	145
A7-3	High Resolution X-ray Spectroscopy using a Johann Spectrometer	145 - 149
A7-4	Spherically Bent Crystals for X-ray Diagnostics	149 - 154

Editor: A Ridgeley

A7-1 AN ACTIVE CRYSTAL SPECTROMETER USING A RETICON DETECTOR

N M Allinson, B Magorrian (York University)
M Grande, A Ridgeley, N Rizvi (RAL)
R W Eason, D Goodman, B Shiwei (Essex University)

Introduction

An experiment was performed in TA4 to investigate the relative merits of a Reticon photodiode array (PDA) and X-ray film as the detecting element in a crystal spectrometer. A charged couple device was also tested at the same time. The idea behind the experiment was to demonstrate whether or not an active crystal spectrometer using a PDA would have advantages over a passive crystal spectrometer using film. Likewise an active pinhole camera using a CCD might have advantages over pinhole cameras using film.

Experimental

The experimental configuration is given in Figure A7.1. Infra-red laser energies of up to 4 J in 1 ns were focused down to $120 \pm 17 \mu\text{m}$ spot size on a plane target. A TlAP crystal was mounted on the streak camera crystal spectrometer attachment and the detector system to be evaluated was placed equidistant with the target from the crystal. The source detector- distance was 170 mm.

The two detecting systems to be compared were DEF X-ray film developed for 4 mins in D19 developer at 20°C and an oxide-thinned Reticon PDA incorporated in a commercial OMA detecting system. The PDA was a 1024 channel device, each element of the array having dimensions $25 \mu\text{m} \times 2.5 \text{ mm}$. The DEF film was contained in a light-tight film pack using $5 \mu\text{m}$ of aluminised Melinex as a window which would transmit X-rays but not visible light. A $20 \mu\text{m}$ thick beryllium window was used for the same purpose for the PDA. It is calculated that these two windows have transmittance characteristics identical within 20% in the wavelength range of interest.

Results

Spectra were obtained from the PDA using aluminium, magnesium and copper targets. The copper targets gave multi-line spectra throughout the wavelength span and therefore provided the best comparison with film. Figure A7.2 shows a typical comparison of film and PDA spectra. The laser energies were similar for these two spectra being 2.2 J for film and 2.4 J for the PDA corresponding to $\sim 2.6 \times 10^{13} \text{ W cm}^{-2}$. Figure A7.3 is an enlarged view of part of the same spectra.

There is an apparent long wavelength cut-off at $9.5 \text{ \AA}$ on the PDA spectrum. This cut-off is not real but caused by vignetting in the image plane. The Si k-edge at $6.8 \text{ \AA}$ is clearly seen as a cut-off in the PDA spectrum.

Figure A7.3 demonstrates how the PDA is picking lines out of background more effectively than film. The PDA resolution element size was matched when the film was densitometered. It was not possible to exactly match the $2.5 \text{ mm} \times 25 \mu\text{m}$ dimensions because the maximum length of the densitometer slit is 1.25 mm. Therefore a slitwidth of $50 \mu\text{m}$ was employed to equalise the areas. This did not cause a loss in resolution as the line width was greater than $50 \mu\text{m}$. The background fog level was also densitometered using a $1.25 \text{ mm} \times 50 \mu\text{m}$ slit from which the standard deviation σ_d in density level at this level was measured to be 0.06. The noise level of the PDA at zero illumination was measured at ± 1 count. Since 100 counts can be seen from the figures to correspond to about density 0.1 this noise level corresponds to about 0.005 film density, which is one order of magnitude better than film. The PDA noise level can be further reduced by cooling. The superior background performance of the Reticon is demonstrated by the fact that it is picking up the continuum background near the short wave-length end of the crystal, whereas this is hardly discernable from background fog level using film.

The saturation level of the PDA is determined by the 14-bit ADC and is therefore in excess of 16,000 counts. This count level was recorded in the aluminium spectrum. This would appear to indicate that the PDA is usable up to much higher flux levels than x-ray film which will saturate at about density 4.

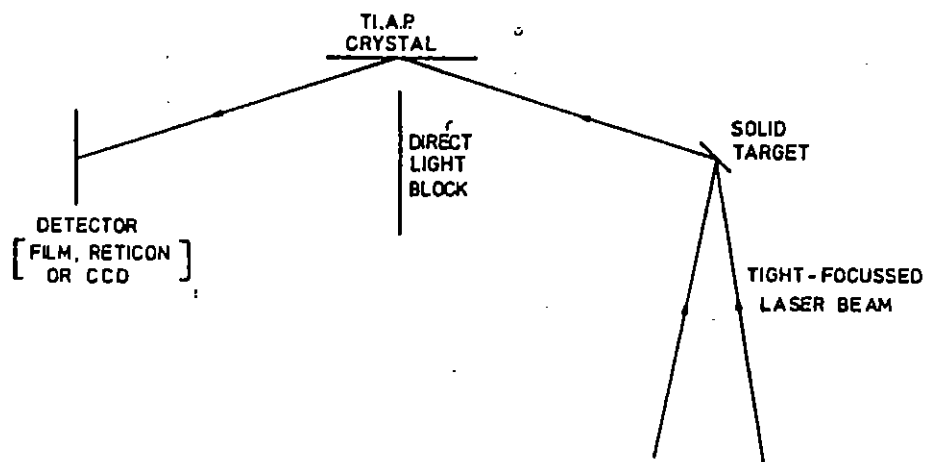


Fig. A7.1 Experimental arrangement.

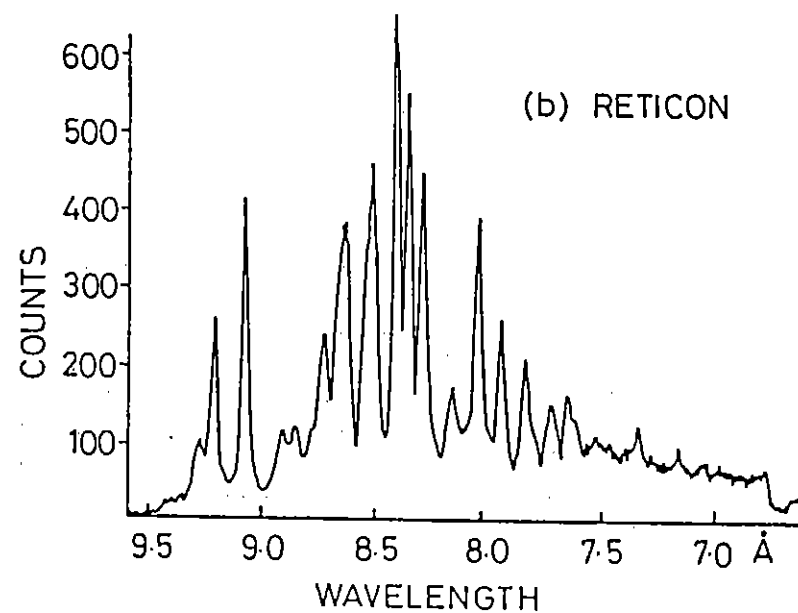
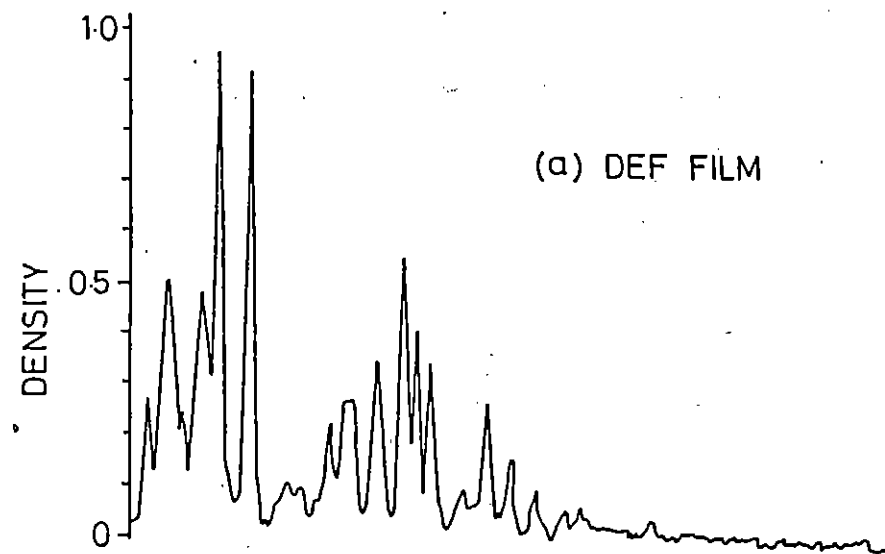


Fig. A7.2 Comparison of reticon and film responses.

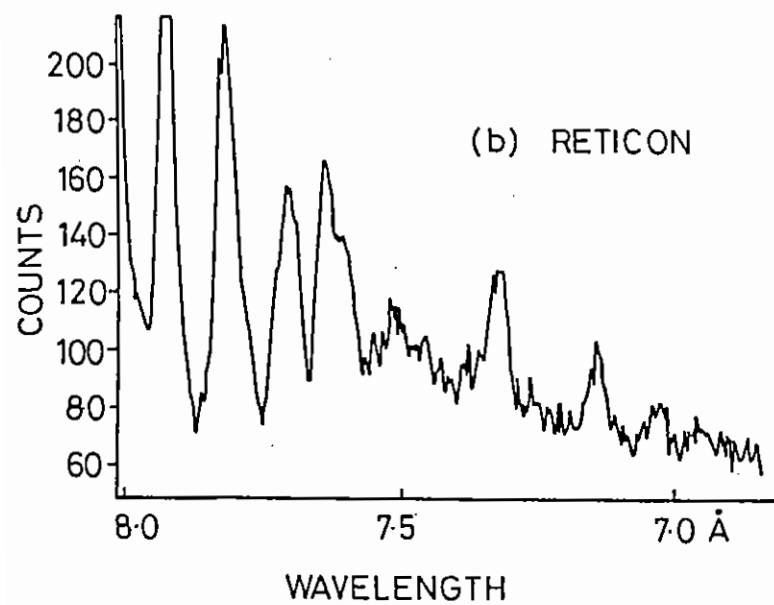
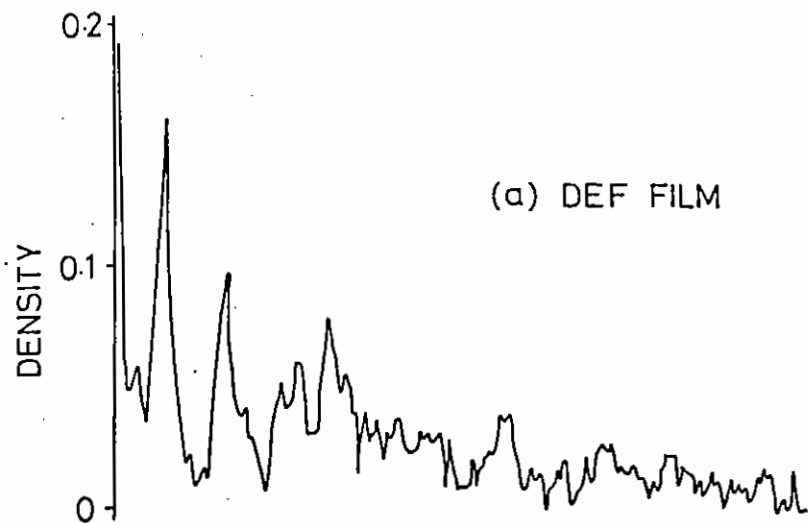


Fig. A7.3 Comparison of reticon and film responses at low flux levels.

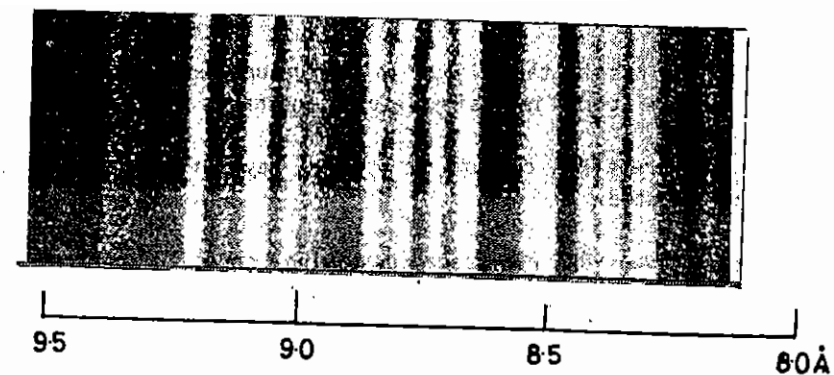


Fig. A7.4 Copper spectrum using a CCD detector.

Conclusion

1. The PDA has better signal to background and noise discrimination than film at low intensity levels, and has a better dynamic range.
2. The apparent PDA cut-off at 8.3-9 Å may not be real but has to be investigated.
3. An active PDA non-space resolving crystal spectrometer would certainly out-perform its equivalent film spectrometer. The next stage is to develop a reasonably inexpensive electronic system for the device.

A7-2 ACTIVE PINHOLE CAMERA USING A CCD

N M Allinson and B Magarrian (York University)
M Grande and A Ridgeley (RAL)

The concept of replacing the film in a pinhole camera with a charge-amplified device is an attractive one. The CCD is an ideal device for recording of two-dimensional images over a small field and enables the possibility of having instant access to the pinhole camera image through a PC.

The experimental arrangement shown in Figure A2.1 was used to demonstrate the feasibility of using a CCD in this way. The device used was a conventional front-illuminated CCD (EEV type P8600): 3852576 element array on a 22 µm pitch.

Figure A7.4 shows a copper spectrum recorded using this device which demonstrates its ability to record an x-ray image. Time did not permit a full quantitative assessment of the device but qualitatively its performance in recording spectra was broadly comparable with the Reticon device described in A7.1.

A7-3 HIGH RESOLUTION X-RAY SPECTROSCOPY USING A JOHANN SPECTROMETER

J Dunn, R Barnsley, K D Evans (Dept. of Physics, Leicester University)
N J Peacock (Culham Laboratory, Abingdon)

A description is given of a high resolution curved crystal x-ray spectrometer, using Johann geometry⁽¹⁾, developed at Leicester for the 1Å - 25Å band. This spectrometer has been used mainly to observe DITE Tokamak⁽²⁾ at UKAEA Culham Laboratory but also x-ray emission spectra from foil-excited heavy ion beams⁽³⁾ generated by the Oxford University Folded Tandem accelerator. More recently, the spectrometer geometry has been optimised for laser produced plasmas during the "Short Pulse, Hot Electron Transport" experiment⁽⁴⁾ on VULCAN in December 1987. A comparison is made between a Johann spectrometer and a flat crystal "minispectrometer"⁽⁵⁾ in the context of laser plasma x-ray sources.

Instrument Description

The instrument consists of a four bar crystal bending jig and a film holder, each mounted via precision rotation and translation stages to a steel optical bench. The crystal lamina is focused and aligned by optical methods⁽⁶⁾, prior to mounting the spectrometer inside the target chamber. The crystal curvature R can be varied in the range 0.3 m to 5 m depending on the experiment, to optimise various factors including resolving power, dispersion, sensitivity and bandpass, as discussed later. High resolving power $\lambda/\Delta\lambda > 6000$ is possible in the waveband 1Å - 13Å depending on the choice of crystal.

For a crystal curvature greater than 0.5 m the optical bench is replaced by a rigid steel tube, which forms part of the vacuum system, and the spectrometer can be bolted externally to the target chamber via an access port.

The spectrometer can be adjusted on a shot to shot basis to cover different wavebands.

Kodak DEF 392 x-ray film is used, protected from scattered visible and UV light by a filter of 2 µm Mylar coated with 1000 Å Al. Removal of the back emulsion coating plus the use of low magnification densitometer optics significantly increases the signal to noise ratio. Further improvement is possible by digitising the spectrum on the RAL computer controlled Joyce-Loebl microdensitometer, and integrating along the line height. The measured film density is converted into incident x-ray photon density using the densitometer optics and film calibration data described in Henke et al⁽⁷⁾.

Experimental Setup

Figure A7.5 shows a schematic diagram of the spectrometer relative to the source, a flat "lollipop" target illuminated with a focused spot size of $50\text{ }\mu\text{m} - 60\text{ }\mu\text{m}$, viewed at an angle of 70° to the input beam axis. The spectrometer was supported onto its own stand in the target chamber and adjusted so that the source was in the field of view and in the same horizontal plane. The crystal radius R was 300 mm and the source was placed inside the Rowland circle at about 90 mm from the crystal to increase the bandpass. A $2\text{ }\mu\text{m}$ Mylar window coated with 85Å of Al was placed in front of the crystal to protect it from reflected laser light and target debris.

The 1s - 2p resonance lines of H-like and He-like Al were studied, emitted in a single beam 7J green energy ($\lambda = 0.52\text{ }\mu\text{m}$) shot by a buried Al layer, and recorded with a PET (002) crystal, $2d = 8.74\text{ }\text{\AA}$. Figure A7.6 shows a spectrum of H-like Al Lyman- α and satellite lines at $7.17\text{ }\text{\AA} - 7.27\text{ }\text{\AA}$.

Low laser energies of 0.4 J of green light incident on the target would still register, in the He-like Al resonance and intercombination lines, as detectable film density.

Discussion

The Johann spectrometer described here has several important advantages for the observation of laser-produced x-ray sources:

- (a) **High resolving power** can be achieved independently of the dimension of the source, or its distance from the crystal. By suitable choice of crystal aperture and R , the contributions to the instrumental line broadening from Johann aberrations and crystal curvature errors can be held below the crystal diffraction width. The resolving power is then limited mainly by the crystal diffraction width.
- (b) **The large sagittal angle** subtended from the source (in the plane of dispersion) gives a high photon collection and a sensitivity advantage. However, the sagittal collection angle will decrease once the source is no longer on the Rowland circle; this effect is currently being investigated. The photon collection increases as the plasma dimensions increase, the sensitivity

advantage over the flat crystal spectrometer with a slit is proportional to the source length in the plane of dispersion. It is worth noting that the Johansson spectrometer⁽⁸⁾ has a larger sagittal collection angle than the Johann case, due to the perfectly focusing geometry, but although generally not used due to the difficulties of grinding and bending crystals to the desired tolerances, it merits further interest. Absolute source emission measurements can be made with knowledge of the crystal reflectivity, filter transmission and detector efficiency data.

- (c) **The dispersion** can be varied to suit a streak camera, for time-resolved spectra, (or CCD detector) because the crystal curvature R is a free parameter. When free from other constraints, a small radius R is used to increase the photon density at the plane of detection thus improving the signal to noise ratio over the detection area. The dispersion, inversely proportional to R , is generally much higher than the flat crystal "mini-spectrometer" e.g. for the spectrum in Figure A7.6 using a PET (002) crystal, $R = 300\text{ mm}$, observing the Al Lyman- α and satellites at $7.17\text{\AA} - 7.27\text{\AA}$, the dispersion at the film plane is $16\text{ m\AA/mm}$.
- (d) **Bandpass** $\Delta\lambda/\lambda = 0.1$ was available, allowing the simultaneous observation of the H-like and He-like 1s-2p resonance lines of Aluminium in this experiment. The bandpass considerations for sources smaller than 1 mm dictate the positioning of the source inside the Rowland circle and close to the crystal. Using a crystal with a larger lattice spacing e.g. KAP (002) $2d = 13.3\text{\AA}$, and positioning the source about 50 mm from the crystal $R = 300\text{ mm}$ will increase the bandpass to $\Delta\lambda/\lambda \approx 0.6$.
- (e) **Spatial imaging** of the plasma can be achieved in one direction, perpendicular to the wavelength dispersion, by placing a slit in the plane of dispersion, between the source and the crystal. Spatial resolution of about $10\text{ }\mu\text{m}$ is possible by optimising the Johann geometry to increase the magnification (50:1).
- (f) **Absolute wavelength calibration**, as derived from the alignment procedure, the Bragg angle θ set to 1 arcminute accuracy, and the crystal calibration, is typically 1 part in 2000. A relative calibration of about 1 part in 50000 is possible if known wavelengths are present in the spectrum.

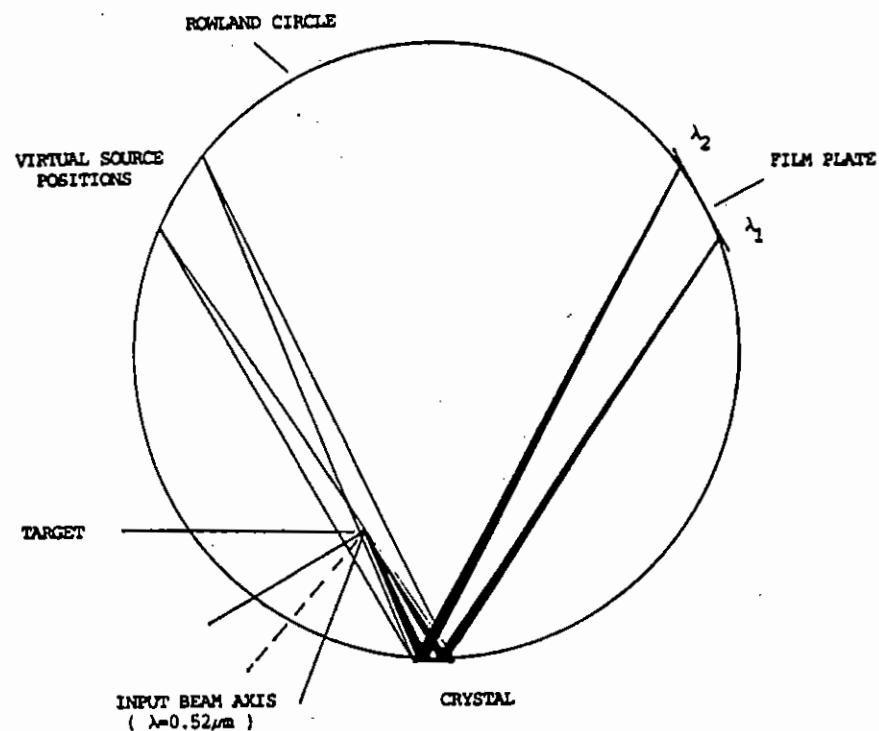


Fig. A7.5 Schematic diagram of the experimental arrangement.

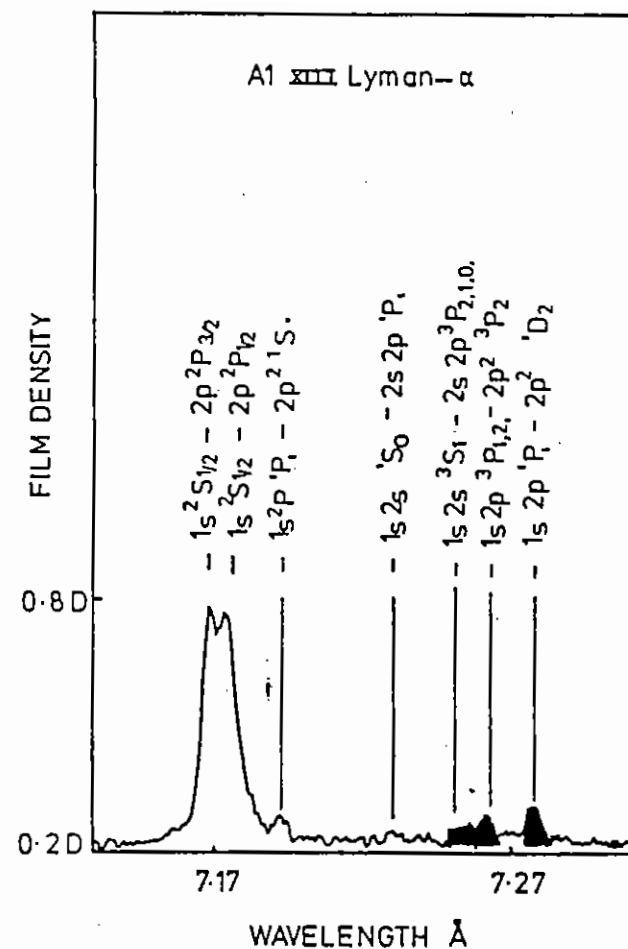


Fig. A7.6 Al XIII spectrum recorded using PET (002) crystal curved to a radius $R = 300$ mm. The x-ray source is a buried Al layer (under $0.1 \mu\text{m}$ plastic top coat) irradiated with a 20 psec "Short pulse" single green 7J energy beam generated by the VULCAN laser. The Al Lyman- α doublet is resolved along with satellite structure to the long wavelength side. Strongest satellites have been labelled and density sensitive lines (10) have been indicated by shading.

(g) Shot integration can be used for observing sources with low x-ray emission since the spectrometer dispersion is independent of the source position. This facility would be useful for selecting wavelength coincidences and observing gain in resonant photoexcitation x-ray laser scheme experiments⁽⁹⁾.

Conclusions

In summary, it has been shown that high resolution x-ray spectroscopy is possible with a Johann spectrometer and that it can be successfully optimised for various experiments involving laser plasma sources.

References

1. H H Johann, Z. Phys., **69**, 185 (1931).
2. J Dunn, R Barnsley, K D Evans, N J Peacock, in: Proc. IAU Colloq. No. 102 on "UV and X-ray Spectroscopy of Astrophysical and Laboratory Plasmas", J. Phys. (Paris) C1.88 (1988).
3. J M Laming, J D Silver, R Barnsley, J Dunn, K D Evans, N J Peacock, Phys. Lett. A., **126**, 253-257 (1988).
4. M H Key et al, Laser Division Annual Report, section A3-2 (1988).
5. J D Kilkenny, R W Lee, M H Key, J G Lunney, Phys. Rev. A., **22**(6), 2746-2760 (1980).
6. J Dunn, Ph. D. Thesis, University of Leicester (1988).
7. B L Henke, J Y Uejio, G F Stone, C H Dittmore, F G Fujiwara, J. Opt. Soc. Am. B., **3**(11), 1540-1550 (1986).
8. T Johansson, Z. Phys., **82**, 507-528 (1933).
9. P G Burkhalter, D A Newman, C J Hailey, P D Rockett, G Charatis, B J MacGowan, D L Matthews, J. Opt. Soc. Am. B., **2**(12), 1894-1905 (1985).

10. N J Peacock, "Laser Plasma Interactions", (Cairns and Sanderson, editors) Proc. of 20th Scottish Universities Summer School in Physics, 711-806 (1979).

A7-4 SPHERICALLY BENT CRYSTALS FOR X-RAY DIAGNOSTICS

N G Basov, Yu A Mikhailov, A V Rode, G V Sklizkov (Lebedev Physical Institute)

A high luminosity x-ray microscope and focusing spectrometer with spherically-curved crystals have been developed for diagnostics of laser plasmas.

The x-ray microscope based on crystals curved to surfaces of the second order (spherical, ellipsoidal, toroidal) allows plasma imaging in x-ray radiation with high space resolution ($\sim 5-10 \mu\text{m}$) and a high degree of radiation monochromatization - $\lambda/\Delta\lambda \sim 10^2-10^4(1-3)$. The plasma image in x-ray radiation obtained by a spherically-curved crystal is formed in accordance with geometrical optics combined with spectral selection due to x-ray diffraction in crystal planes with the Bragg condition $2d \sin \theta = n\lambda$, where θ - is the Bragg angle, λ - is the wave length of reflected radiation, n - is the order of reflection. The spatial resolution of this scheme is determined by the geometrical parameters and ideally to obtain the image from point to point it is necessary to use crystal reflection planes curved over an ellipsoidal surface.

Relatively small aberrations of spherically-curved crystals can be realized at Bragg angles near $\theta = \pi/2$, which means $\lambda_0 \lesssim 2d$. For changing the wavelength range to include the spectral lines different from $\lambda_0 = 2d$ in the case of the spherical crystal it is necessary to place the plasma x-ray source off the spherical mirror axis. In this case the spatial resolution is determined by astigmatism of such an optical scheme and may be evaluated as the minimum caustic cross-section diameter DC between saggital and meridional images of a point source

$$D_f = D_c \frac{1}{K} \frac{a_m^1 - a_s^1}{a_m^1 + a_s^1} \quad (1)$$

where, D_c - is the crystal diameter, K - is the magnification factor, a_m^1 and a_s^1 - are the distances from x-ray mirror to the meridional or

sagittal images. Figure A7.7 presents the results of space resolution calculations for crystal mirrors with radius of curvature $R_c = 500$ mm and crystal diameters $D_c = 5$ and 16 mm in the scheme with magnification factor $K = 1:20$.

The spectral range of crystal reflected radiation is determined by the difference between $\theta_{\max}$ and $\theta_{\min}$ (for incident radiation) and may be written as

$$\frac{\lambda}{\Delta\lambda} = \frac{K+1}{K-1} \frac{R_c}{D_c} \operatorname{tg} \theta \quad (2)$$

Figure A7.8 gives values of reflected radiation spectral ranges versus Bragg angle θ for silicon and quartz crystals and different crystal mirror diameters. The positions of wavelengths for hydrogen-like ions of Mg XII and the corresponding dielectric satellites (for quartz[10 $\bar{1}$ 0] - crystal) hydrogen-like ions of Cl XVII (for quartz[20 $\bar{2}$ 0]) and hydrogen-like ions of Si XIV (for Si [111]) are shown. Crystal diameters are 24 mm and 10 mm. As seen from these figures, the decrease in crystal aperture is accompanied by an increase of refracted radiation monochromaticity, and by changing θ_0 it is possible both to obtain the plasma x-ray image in radiation of an individual spectral line and to investigate the line shape.

For laser plasma diagnostics by x-ray microscopy it is necessary to analyse spectral intensities in chosen spectral ranges, to take into account reflection coefficients of curved crystals for different orders of reflection and special selection of absorbing filters for excluding some orders of crystal reflection. When a spectral line is within the working spectral range the image intensity becomes considerably higher, enabling one to observe low-intensity details in plasma images. The x-ray image of the compressed core of a Cl-filled shell target⁽⁴⁾ obtained in the second order of a quartz[10 $\bar{1}$ 0] crystal with Bragg angle $\nu = 82^\circ$, serves as a good example. The spectral range includes lines of Ly_α Cl XVII and corresponding dielectronic satellites (Figure A7.9). Such images enable one to make deductions about laser compression symmetry.

The application of spherically-curved crystals (using in laser plasma diagnostics) in focusing x-ray spectrographs^(2,5) is also of great importance. This allows a significant increase in the luminosity of x-ray spectrographs and to produce x-ray spectrum records with spatial

resolution without using the traditional space-resolving slits, thus producing further increase of spectrograph luminosity.

Spectrograph luminosity S can be written as follows:-

$$S = R_i \frac{\eta_v}{a+b} \frac{D_\theta}{D_\lambda} \frac{d\lambda}{d\theta} \quad (3)$$

where, R_i - is the integral reflectivity of a crystal, a and b are the distances between the plasma source and the crystal and between the crystal and x-ray detector (for example, x-ray film), D_θ and D_λ are the angular and linear dispersion, η_v is the luminosity gain coefficient due to focusing in the direction perpendicular to the dispersion direction.

Figure A7.10 presents the scheme of the spherically-curved crystal application in the focusing spectrograph. In this case luminosity increases along dispersion direction, as it does in the traditional focusing Johann scheme with a cylindrical crystal. For the last scheme the increase in luminosity (in comparison with the flat crystal scheme) due to focusing in the dispersion direction may be derived from the equation

$$\frac{d\lambda}{d\theta} = \eta_h = \frac{R_c \sin \theta + a}{R_c \sin \theta - a} \quad (4)$$

η_h increases as the source approaches the Rowland circle. The use of a spherical crystal enables one to realize focusing in a vertical direction with the focal length depending on the investigated line wavelength

$$f = R_c (2 \sin \theta)^{-1} = R_c / 2 \sin \theta \quad (5)$$

Point 5 (Figure A7.10) corresponds to the position of source 1 image in the sagittal plane (full vertical focusing); $a^1 = af(a-f)^{-1}$. For the spectral (horizontal) focusing plane 4, which is on the Rowland circle at the distance $b = R_c \sin \theta$ from the crystal the additional increase of luminosity (in comparison with Johann scheme) is

$$\eta_v = \frac{R_c \sin \theta + a}{R_c \sin \theta - 2a \sin^2 \theta + a} \quad (6)$$

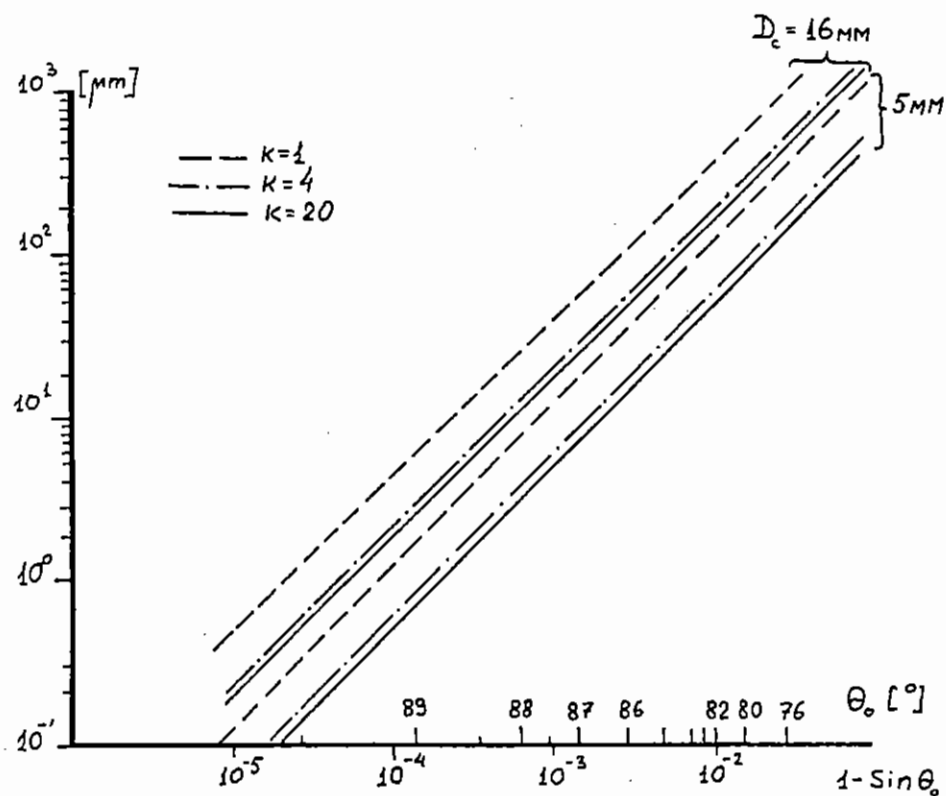


Fig. A7.7 Space resolution for a spherical mirror with the curvature radius of 500 mm. Crystal diameters are 16, 10 and 5 mm. Magnification factor $K = 1, 4$ and 20 .

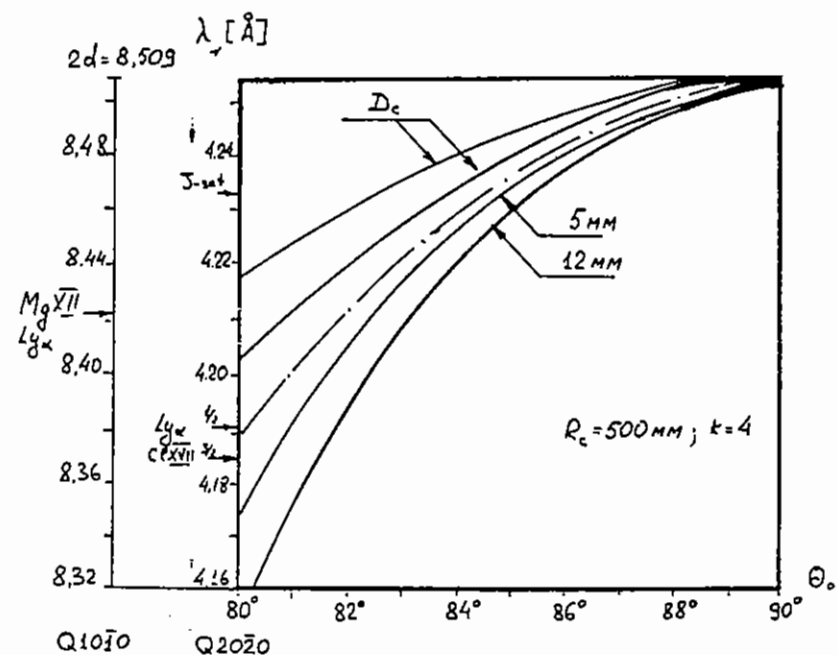
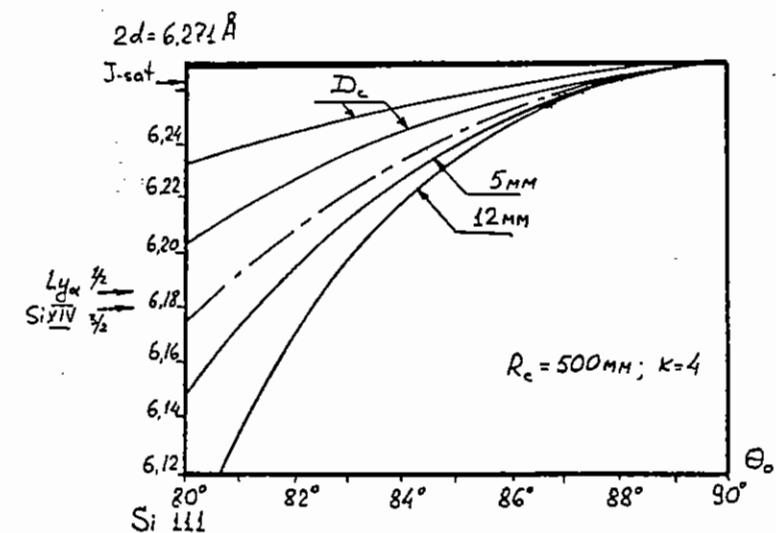


Fig. A7.8 Spectral ranges for radiation reflected from spherically-curved crystals. Radius of curvature is 500 mm. Magnification factor $K = 4$.

Q 1070

85 Å

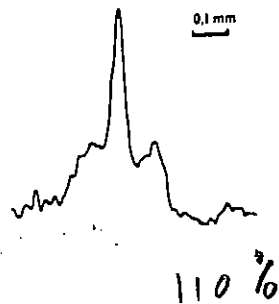
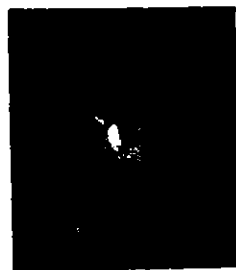


Fig. A7.9 X-ray image of compressed core for polystyrene shell target filled with Cl.

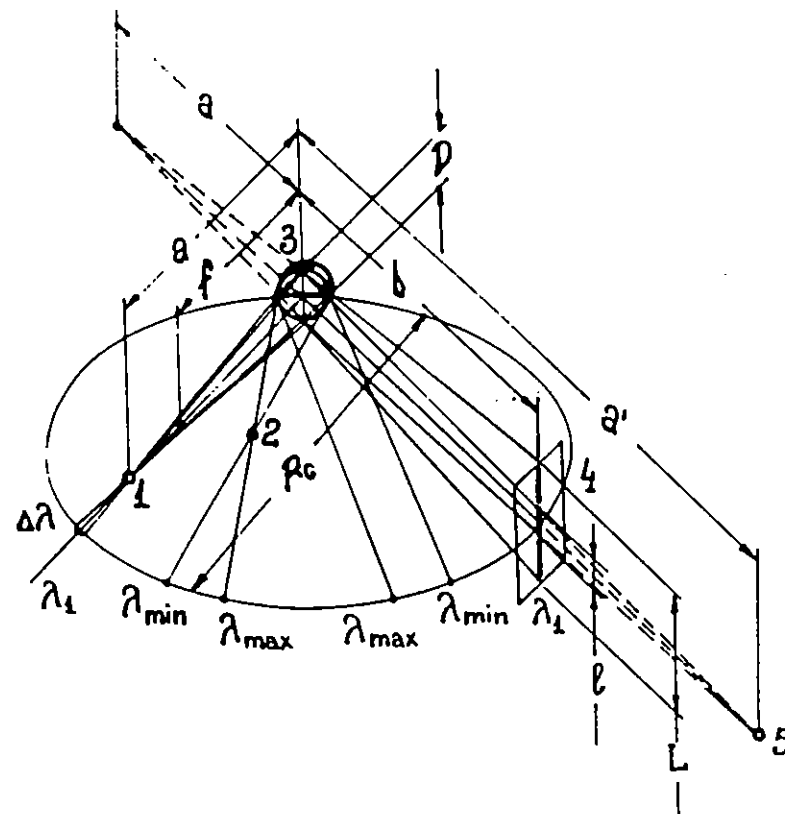


Fig. A7.10 Scheme of the focusing x-ray spectrograph with a spherical mirror. 1; 2 are plasma source positions relative to different spectral ranges; 3 is a spherical crystal; 4 is an x-ray detector; 5 is source (1) image position in the sagittal plane; L and l is vertical dimension of spectral line for spectrograph schemes with cylindrical and spherical crystals.

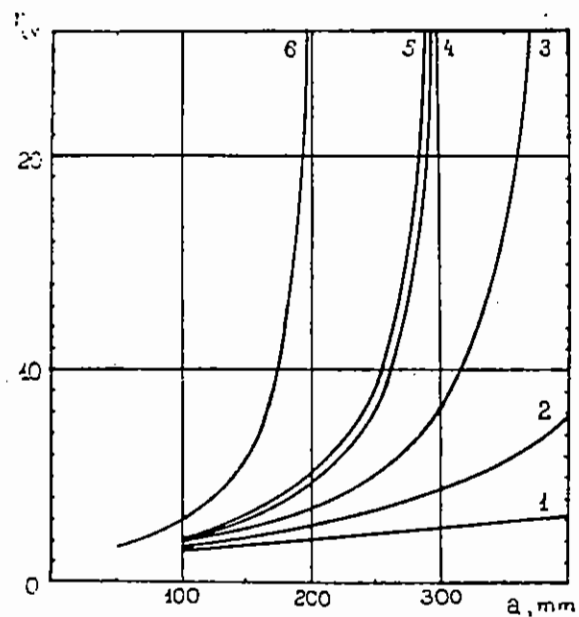


Fig. A7.11 Luminosity enhancement factor due to vertical focusing under different experimental conditions. The crystal is quartz $[10\bar{1}0]$: $2d = 8.509 \text{ \AA}$, $R_c = 300 \text{ mm}$.

1. $\eta_v(a)$ for $(1s^2 - 1s5p)$ transition of /He/-like ions of Al ($\lambda = 6.175$).
2. $\eta_v(a)$ for $L_{\gamma\alpha 1,2}$ of /H/-like ions of Al ($\lambda = 7.1749 \text{ \AA}$).
3. $\eta_v(a)$ for intercombination line of /He/-like ions of Al ($\lambda = 7.8057 \text{ \AA}$).
4. $\eta_v(a)$ for line $L_{\gamma\alpha 1,2}$ of /H/-like ions of Cl ($\lambda = 4.1897 \text{ \AA}$). The second order of reflection.
5. $\eta_v(a)$ for line K_α of Al ($\lambda = 8.339 \text{ \AA}$).
6. $\eta_v(a)$ for line K_α of Al when $R_c = 200 \text{ mm}$.

Thus, the increase of luminosity of the spectrometer due to the spherical mirror depends on the radiation wavelength and the source displacement from Rowland circle $R_c \sin \theta - a$. Practically the selection of the value a depends on the diagnostic purpose (spectral range) and possible intensity of spectral lines.

For plasma diagnostics at the "Vulcan" installation an x-ray spectrograph with a spherical crystal was prepared. A quartz crystal with orientation $[10\bar{1}0]$ ($2d = 8.509 \text{ \AA}$) spherically curved with radius of curvature $R_c = 300 \text{ mm}$, and diameter $D = 16 \text{ mm}$ was used. The operating diameter of the crystal along the dispersion direction is limited by the geometry of the experiment and diameter $2r$: $D_0 = 2rR_c(R_c \sin \theta - a)$. The values of luminosity gain coefficient η_v for some spectral lines of multicharged ions are given in Figure A7.11. For example, for the K_α line of aluminium ($\lambda = 8.339 \text{ \AA}$) the increase of luminosity due to the vertical focusing is about 5 when $a = 200 \text{ mm}$. This increases sharply up to ~ 20 for $a = 290 \text{ mm}$. However, in the last case the spectral range is very narrow, and the plasma diagnostics becomes possible only on the base of one line analysis, (e.g. line broadening measurements or absolute intensity measurements). Almost the same conditions apply for the recording of $Ly_{\alpha 1/2, 3/2}$ for Cl: $\lambda_{1/2} = 4.1897 \text{ \AA}$, $\lambda_{3/2} = 4.1843 \text{ \AA}$ (the second order of reflection is used). For electron density diagnostics in the laser plasma using relative intensities of resonance ($\lambda_w = 7.7566 \text{ \AA}$) and intercombination $\Phi \lambda_\gamma = 7.8057 \text{ \AA}$) lines of helium-like ions of Al it is necessary to provide $\Delta\lambda > 0.05 \text{ \AA}$ and $a < 250 \text{ mm}$. As seen from dependence 3 (Figure A7.11), the value of η_v is limited to ~ 5 . Dependence 6 corresponds to the same quartz crystal but with radius curvature $R_c = 200 \text{ mm}$. Low efficiency of spectrographs with spherically curved crystals in the first order of reflection for shorter-wavelength x-ray radiation is illustrated by curves 1 and 2 ($\lambda_1 = 6.175 \text{ \AA}$ and $\lambda_2 = 7.1749 \text{ \AA}$).

References

1. L M Beliaev, A B Gilvarg, Yu A Mikhailov, S A Pikuz, G V Sklizhov, A Ya Faenov, S I Fedotov, Kvantovaya Elektronika, 3, 9, 2057 (1976).

2. V A Boiko, A V Vinogradov, S A Pikuz, I Yu Skobelev, A Ya Faenov, "X-ray spectroscopy of laser plasmas", Radiotekhnika, 27, VINITI (1980).
3. N G Basov, Yu A Mikhailov, G V Sklizkov, S I Fedotov, "Laser Thermonuclear Installations", Radiotekhnika, vol. 25, VINITI (1983).
4. M J Lamb, C L S Lewis, E Robertson, J Corbett, P McCavana, P Norreys, C Popovich, A Rode, B Fraenkel, Annual Report to the LFC C5.4 RAL-87-041 (1987).
5. L M Beliaev, A B Gilvarg, Yu A Mikhailov, S A Pikuz, G V Sklizhov, A Ya Faenov, S I Fedotov, Kvantovaya Elektronika, 4, 129 (1977).

B	LASER SUPPORT FACILITY	Pages
B1	CHEMISTRY	155 - 195
B2	BIOLOGY	197 - 220
B3	PHYSICS	221 - 248
B4	LASER DEVELOPMENT AND NON LINEAR OPTICS	249 - 262
B5	LASER SUPPORT FACILITY OPERATIONS AND DEVELOPMENT	263 - 275

SECTION B

Introduction

The contributions to this section report work done in the third year of operation of the Laser Support Facility. They cover many fields of research in Chemistry (B1), Biology (B2), Physics (B3) and Laser Applications (B4). Operational aspects are reported in B5. The reports are condensed accounts of research in progress. More extended accounts have appeared in the literature, or soon will. Users published more than 30 papers in 1987 on their work using LSF facilities. The community continues to grow with roughly a fifth of the contributions coming from first time users. The loan pool accounts for a third of the reports. In contrast to previous years there were no major new facilities installed and the work reported here was done with the three RAL based laser systems and the seven loan pool lasers.

W T Toner

B1	CHEMISTRY	Pages		Pages
B1-1	Picosecond Single Vibronic Level Fluorescence Spectroscopy	155 - 157	B1-15	Decay of Transient Absorption Following Picosecond Laser Excitation of Microcrystalline 1, 2 Pyrazolines
B1-2	UV Resonance Raman Studies of Inclusion Compounds	157 - 159	B1-16	Laser Induced Desorption and Energy Transfer at Surfaces
B1-3	Resonance Raman and Time-Resolved Resonance Raman Spectral Studies of Photosensitisers and Colloidal Semiconductors	160 - 162		
B1-4	Accelerated Laser Ageing of Plastics	162 - 163		Editors: S Tavender/A Langley
B1-5	Resonance Raman Spectroscopy of Nitrobenzene	163 - 164		
B1-6	Laser Excitation Spectroscopy of Transition Element Halides in Supersonic Beams	164 - 166		
B1-7	Do Higher Excited States of the Uranyl Ion Luminescence?	166 - 169		
B1-8	Measurement of the CF A ² Σ ⁺ Radiative Lifetime	169 - 171		
B1-9	Laser Flash Photolysis/Laser Absorption Studies of SiMe ₂ Radical Kinetics	172 - 173		
B1-10	Photodissociation Dynamics of HONO ₂ at 280 NM	173 - 176		
B1-11	Photodissociation Dynamics of Jet-Cooled Methyl and t-Butyl Thionitrite	176 - 178		
B1-12	Chemiluminescence and Energy Transfer in Mn + N ₂ O Collisions at High Translational Energy	179 - 182		
B1-13	Rotational State Dependence of Electronic Energy Transfer in Na ₂	182 - 186		
B1-14	The Laser Photoacoustic Spectra of Ozone and Oxygen in the Visible Region	186 - 190		

B1-1 PICOSECOND SINGLE VIBRONIC LEVEL FLUORESCENCE SPECTROSCOPY.

J M Hollas, P F Taday, (University of Reading)
A J Langley, W T Toner, (RAL)

Introduction

If we excite a large polyatomic molecule, such as styrene $C_6H_5-CH=CH_2$, by tuning a dye laser into a vibronic band of that molecule, under collision free conditions, we may observe resulting dispersed fluorescence from the vibronic level to which the molecule has been excited. This method is known as single vibronic level fluorescence (SVLF) spectroscopy.

With our present system for observing laser induced fluorescence at Reading University we are able to observe discrete fluorescence only from the zero point level (case (a) Figure B1.1) and fairly low-lying vibronic levels of the first excited singlet electronic state S_1 . Fluorescence from higher-lying vibronic levels (case (b) Figure B1.1) is broad and featureless. This diffuseness is due to an internal energy relaxation process known as intramolecular vibrational relaxation (IVR). In this work we hope to observe the time evolution of the effects of IVR. Since IVR occurs on the picosecond time scale we require time resolution of this order. This is provided by the picosecond laser, with a pulse length of 2ps, and the Delli-Delti S-20 streak camera, with a time resolution of about 5 ps.

Theory

A brief account of the complex subject of IVR is given here: for more detail see references 1-4.

First let us consider two excited vibronic states, in S_1 , such as $|a\rangle$ and $|b\rangle$ shown in Figure B1.2. We assume that $|a\rangle$ is an optically light and $|b\rangle$ an optically dark state. At a time $t=0$, the state $|a\rangle$ is excited by a pump laser from $|g\rangle$, the ground electronic state. If they are similar in energy and have the same symmetry there is a coupling between the levels $|a\rangle$ and $|b\rangle$, which takes the form of a Fermi resonance, producing two new states $|1\rangle$ and $|2\rangle$ where

$$|1\rangle = \alpha|a\rangle + \beta|b\rangle$$

$$|2\rangle = \beta|a\rangle - \alpha|b\rangle$$

where α , β are constants that depend upon the strength of the coupling. This coupling can be interpreted as an energy flow from the state $|a\rangle$ to $|b\rangle$ and back to $|a\rangle$. The time for this oscillation is less than the lifetime of the states and, therefore, we can observe only states $|1\rangle$ and $|2\rangle$, and not $|a\rangle$ and $|b\rangle$. This is known as static IVR, with no time dependence, since this is a result of normal Fermi resonance.

Now consider when $|a\rangle$ is contained within a large number of states $|s\rangle$, as in Figure B1.1. Again, there is a coupling between $|a\rangle$ and the states of the same symmetry in $|s\rangle$ giving a resultant distribution of $|a\rangle$ and $|s\rangle$ which is approximately Lorentzian. There is an energy oscillation between $|a\rangle$ and $|s\rangle$ but, if the density of states of $|s\rangle$ is sufficiently high, there will be a damping effect on $|a\rangle$ and the time for oscillation from $|s\rangle$ back to $|a\rangle$ is longer than the lifetime of the state $|a\rangle$. This coupling is known as dynamic IVR or super Fermi resonance (also referred to as a Fermi haemorrhage (1)). Due to the Franck-Condon principle there is a fluorescence decay from this quasi-continuum of states $|s\rangle$, in the excited state, to a similar quasi-continuum in the ground state, $|g_s\rangle$. But, if we observe on a short enough time scale (i.e. picosecond), we may observe a decay in $|a\rangle$ and build up of intensity in $|s\rangle$, which exhibits itself as a broadening of the observed dispersed fluorescence spectrum. Before this build up we hope to see discrete fluorescence from $|a\rangle$.

Experiment

Using the picosecond laser in the LSF we plan to excite high lying S_1 vibronic levels of styrene and observe the time evolution of IVR by monitoring the time evolution of the dispersed fluorescence from being sharp, after a short time of the order of 10 ps, to being diffuse after longer times. Figure B1.3 shows the experimental arrangement. There are three irises (I_1 - I_3) in the system to reduce scattered light. A beam splitter (BS) is used to supply light to a vacuum photodiode (PD) which is running at saturation. The photodiode signal is attenuated, inverted and passed into a constant fraction discriminator (CFD). This produces a consistent trigger

pulse which is preamplified (PA) and delayed to match the streak speed. The streak camera (SC) then triggers the image intensifier (II) and the CCD. Meanwhile the light pulse has been optically delayed by being reflected across the laboratory by mirrors M_1 , M_2 and M_3 , and finally focussed by lenses L_1 and L_2 into a glass T-shaped cell (S) of length 10 cm and with quartz windows containing the sample of styrene at its vapour pressure, 10 Torr (20°C). The fluorescence resulting from the exciting radiation is observed perpendicular to the beam, and is focussed into a Spex Minimate monochromator (MC), with a 1200 lines/mm grating blazed at 250 nm and used in the first order. The dispersed fluorescence is then focussed, by lens L_4 , onto the Delli-Delti S-20 streak camera (SC). The streaked image is intensified and detected by a Hadlad Photonics charge-coupled device (CCD), which is controlled by a 2-D image analysis computer system (C).

Results

The experiments carried out so far have been mainly concerned with the characterisation of the streak camera and CCD. Testing the sensitivity of the system is important since the dispersed fluorescence is likely to be weak. Results have shown that the system is sufficiently sensitive in the 290 nm region where the styrene fluorescence occurs.

A weak undispersed fluorescence has been observed when exciting the pure electronic transition at 288 nm.

References

1. R A Coveleskie, D A Dolson and C S Parameter, J Phys Chem 89 645, 655 (1985).
2. P M Felker, Laser Applications in Chemistry and Biophysics 620 2 (1986).
3. P M Felker and A H Zewail, J Chem Phys 82 2961, 2975, 2994 (1985).

4. A H Zewail, Ber Bunsenges Phys Chem 89 264 (1985).

B1-2 UV RESONANCE RAMAN STUDIES OF INCLUSION COMPOUNDS

J E D Davies and V A Tabner (University of Lancaster)

1. Introduction

Inclusion compounds are two component systems wherein a 'guest molecule' is enclosed in a void formed by the 'host lattice' ⁽¹⁾. The shape and dimension of the void, the nature and the extent of the interaction between the host and the guest, and the degree of mobility of the guest are all dependent on the host lattice used for a given guest molecule.

Vibrational spectroscopy, using both infrared and Raman techniques, has been used extensively to give information about host-guest interaction and the degree of mobility of the guest ⁽²⁾. Previous Raman studies have been of the non resonant type, thus the extent of the host-guest interaction was obtained simply from the magnitude of the difference between the gaseous state and guest wavenumber values for the vibrational mode involving the bond assumed to be involved in the interaction.

2. Objectives of the Research

We wish to investigate whether UV excited resonance Raman spectra, combined with pre-resonant Raman spectra obtained at Lancaster, can yield detailed information about the nature of the interaction between the host and guest components.

Our current interest is with inclusion compounds formed between quinol, $C_6H_4(OH)_2$, acting as the host lattice and a variety of small guest molecules such as H_2S , SO_2 , CH_3OH , CH_3CN and HCO_2H . The compound formed with SO_2 is unique in that it is the only coloured (pale yellow) inclusion compound formed by quinol. This could indicate a different host-guest interaction, most probably charge transfer (CT) in origin, in the quinol/ SO_2 compound.

We have already carried out a pre-resonant Raman study of this compound using the visible region lines ranging from 610 to 456 nm obtainable from the argon ion and dye lasers available in Lancaster. This study revealed a selective intensity enhancement of the band arising from the $\nu_1(A_1)$ mode of the SO_2 guest molecule and of some bands arising from the quinol host lattice. We wished to extend the study into the UV region to see where these and any other bands are further enhanced.

We are currently carrying out a normal coordinate analysis of the quinol molecule in order to identify the vibrational modes giving rise to the enhanced bands. This analysis should thus enable us to deduce which portion of the host lattice is interacting with the SO_2 guest molecule, and thus give detailed information about the nature of the host-guest interaction.

The inclusion compounds of quinol are stable in the solid state only. During our visits to the Laser Support Facility we have concentrated on evaluating the suitability of the solid sample holders provided by Vantacon.

As a result of these studies modifications were suggested to the designs and we now have available two designs - one for solids in the form of a loose powder packed under a silica optical flat, the other for more stable solids which can be pressed under pressure to a compacted solid.

Using both of these holders we have to date obtained non resonant and pre resonant Raman spectra of a variety of compounds which can act as host lattices using 360 nm excitation (see Figure B1.4).

5. Future Work and Application

The quinol/ SO_2 compound gives an absorption band at 360 nm which is not given by other quinol inclusion compounds and thus arises from the CT band of the compound. We intend to record the Raman spectra of a variety of quinol inclusion compounds using 360 nm excitation to explore the influence of the 360 nm absorption band on the observed spectra. All quinol inclusion compounds also give an absorption band at 290 nm and we also intend to record Raman spectra using this exciting wavelength.

The immediate aim of the project is to record the Raman spectra of a variety of solid quinol inclusion compounds using 360 and 290 nm excitations to investigate whether such spectra can reveal details of the nature of the interaction between the host and guest components. Many of the quinol inclusion compounds have been studied crystallographically and it will thus be possible to compare the nature of the host guest interaction deduced from the resonance Raman study with that deduced from the crystallographic studies.

A variety of compounds, such as urea, thiourea, cyclodextrins and crown ethers, can act as host lattices giving inclusion compounds which are stable in the solid state only (urea and this urea) or stable both in the solution and solid phases (cyclodextrins and crown ethers). Since many of these compounds have not been characterised crystallographically the obvious extension of this work would be to examine the resonance Raman spectra of a variety of inclusion compounds in both the solid and solution phases and thus reveal the details of the host-guest interactions in these compounds.

References

1. J L Atwood, J E D Davies and D D MacNichol (Editors), 'Inclusion Compounds', Academic Press, London, 1984.
 - (a) Volume 1 - 'Structural Aspects of Inclusion Compounds formed by Inorganic and Organometallic Host Lattices'.
 - (b) Volume 2 - 'Structural Aspects of Inclusion Compounds formed by Organic Host Lattices'.
 - (c) Volume 3 - 'Physical Properties and Applications'.
2. J E D Davies, Chapter 2 in Ref. 1c.

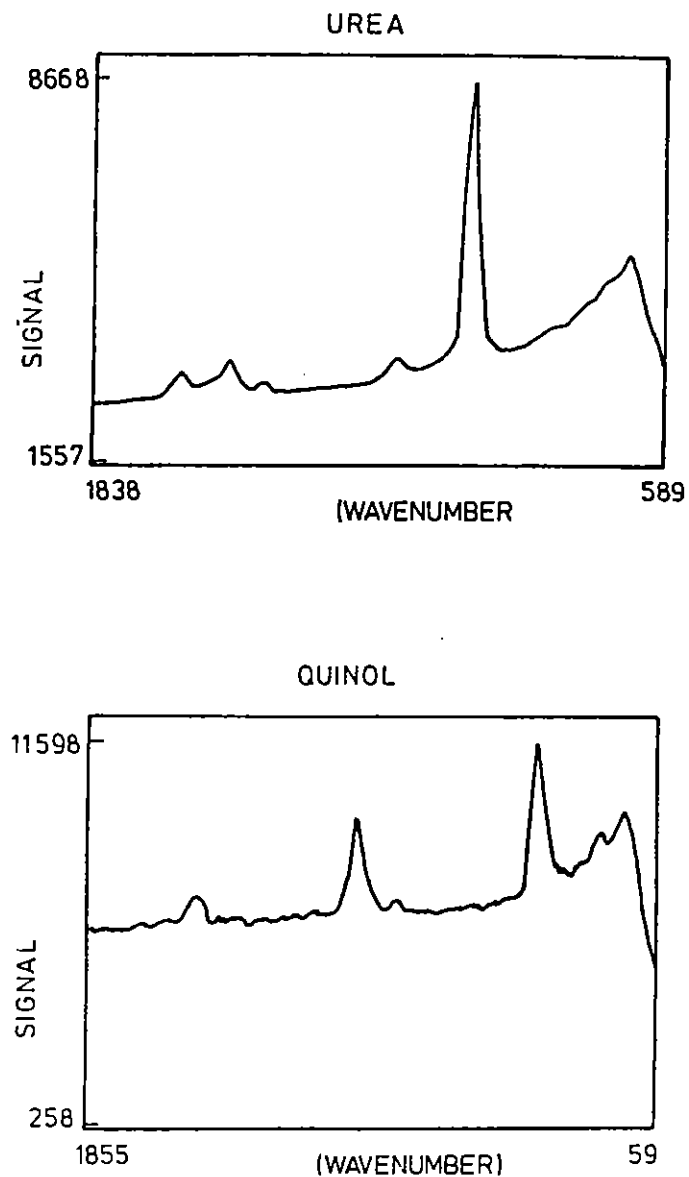


Fig. B1.4 Examples of the Raman spectra of solids using 360 nm excitation. Top - the non resonance Raman spectrum of urea. Bottom - the pre resonance Raman spectrum of quinol.

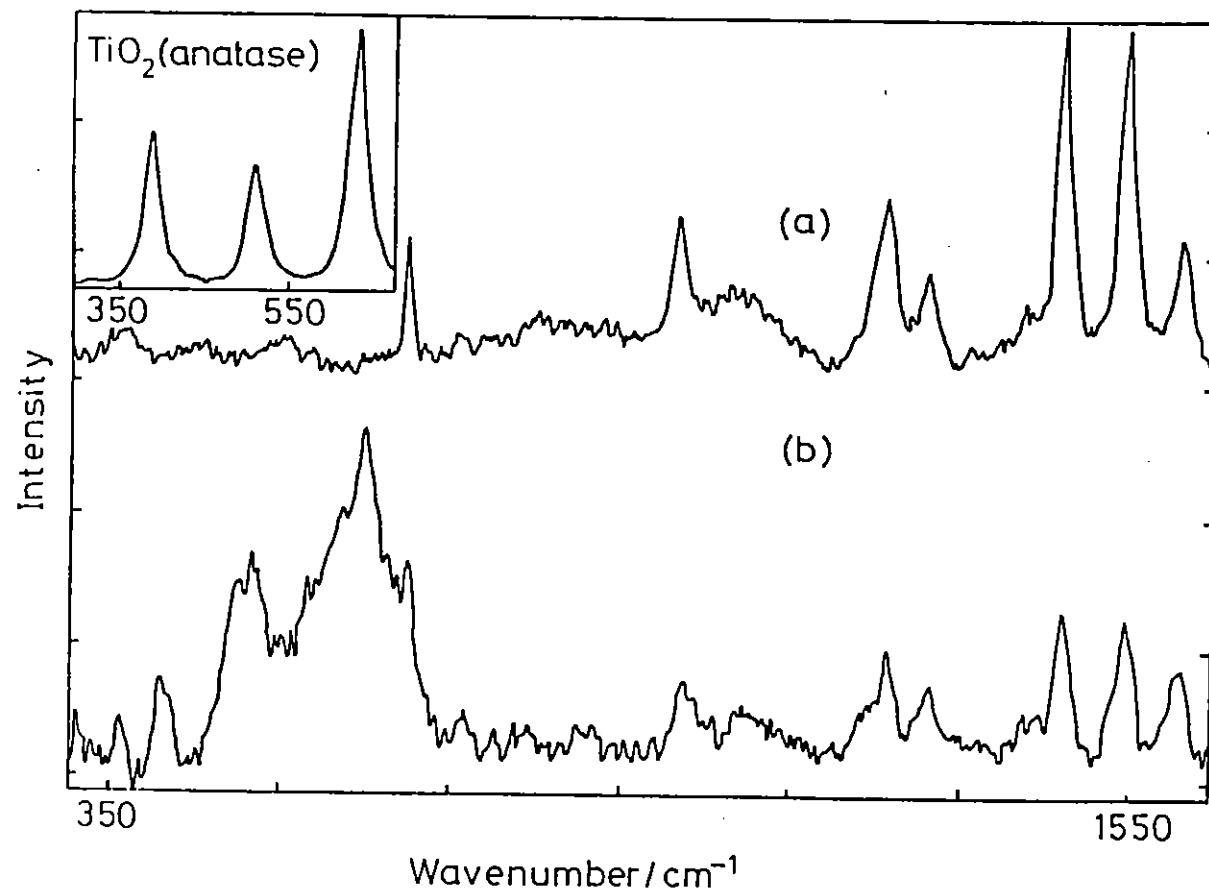


Fig. B1.5 Resonance Raman spectra of solid
(a) ruthenium(II)
tris(2,2'-bipyridyl-4,4'-diethylcarboxylate) and
(b) surface-derivatized TiO₂.

Inset: TiO₂ (anatase). λ_{exc} , 488 nm; spectral resolution 5-6 cm⁻¹.

B1-3 RESONANCE RAMAN AND TIME-RESOLVED RESONANCE RAMAN SPECTRAL STUDIES OF PHOTOSENSITISERS AND COLLOIDAL SEMICONDUCTORS.

R E Hester, S Umapathy, G Lee-Son and A M Cartner (Chemistry Dept., University of York)

1. Introduction

Bipyridyl complexes of ruthenium(II) have attracted considerable attention due to their use as sensitizers in heterogeneous redox catalysis. In particular, ruthenium(II) tris (2,2'-bipyridyl-4,4'-diethyl carboxylate) (RB4ET) and ruthenium(II) tris (2,2' bipyridyl-5,5'-diethyl carboxylate) (RB5ET) both have been used in solar energy conversion, where the water splitting redox process is catalysed by metal loaded colloidal semiconductors, such as TiO_2 ⁽¹⁾. These complexes act as antennae to harvest solar energy, following which the so-formed excited states of the complexes inject electrons into the conduction bands of the colloidal particles. Subsequently these particles mediate the redox process. Raman spectroscopy is used to study the vibrational modes of the species at the surface of the semiconductor, in order to determine the molecular structure and bonding changes that occur during the sensitisation process.

2. Results and Discussion

Efficient sensitisation has been achieved by chemisorption of the complex onto the semiconductor or by surface-derivatization of the semiconductor using the complex. In the case of RB4ET, surface-derivatization of TiO_2 by photolysing the mixture at 100°C was found to result in very efficient sensitization⁽²⁾. This efficiency was at first attributed to the formation of Ru-O-Ti surface bonds, but was later contradicted⁽³⁾. We have prepared the surface-derivatized TiO_2 (TRB4ET) and recorded its resonance Raman spectrum. The spectrum contained very intense bands in the low wavenumber region where Metal-Oxygen (M-O) bond vibrational bands are expected. Not forgetting the possible presence of bulk TiO_2 bands (see inset in Figure B1.5),

the TiO_2 (anatase) spectrum was subtracted from the TRB4ET spectrum by normalising to the band at 396 cm^{-1} . Figure B1.5(a) and B1.5(b) show the spectra of RB4ET and TRB4ET, respectively. It can be seen that in the TRB4ET spectrum, new bands are seen at 649, 626, 516, 498 and 409 cm^{-1} . By analogy with M-O bond vibrational data in the literature, we have assigned these bands to $\nu(\text{Ti-O})$ and $\nu(\text{Ru-O})$ vibrations, thus providing positive new evidence for the presence of Ru-O-Ti covalent bonding between the ruthenium complex and the semiconductor^(4,5).

In their role as photosensitizers, the electronically excited states of RB4ET and RB5ET are of importance. It has been established that simple bipyridyl complexes of ruthenium(II) form structurally asymmetric excited states, with odd electron localization on one of the three otherwise equivalent bipyridyl ligands. This well established feature could not be proved in the cases of electrochemically reduced forms of RB4ET and RB5ET, for which Raman and ESR spectroscopic studies have been reported⁽⁶⁾. By analogy, these results indicated that the excited electron may be shared by all the three ligands (delocalised) in these complexes. We have used time-resolved resonance Raman spectroscopy (TR3S) to study directly the excited state structure of these complexes.

In our experiments, we have used both one and two laser arrangements for excited state studies. In the two lasers experiment, a pump laser pulse is used to excite the complex from its ground state to its excited state and another laser (different wavelength, time delayed) is used to probe the RR spectrum of the excited state species. In the single laser experiment, the leading part of the laser pulse (pulse width 15-20 ns) is used to pump the complex to its excited state and the trailing part of the pulse is used to probe this state. This single laser experiment depends on both the ground and excited state complexes having strong absorption at 390 nm. However, this common absorption also results in interference in the two-laser experiments where 390 nm is used as the probe. We have found that saturation pumping of the ground to excited state species is achieved with only ca. 0.3 mJ/pulse. Two-laser experiments are being used to study systems where different species formed at the surface can be discriminated by their life time (*vide infra*).

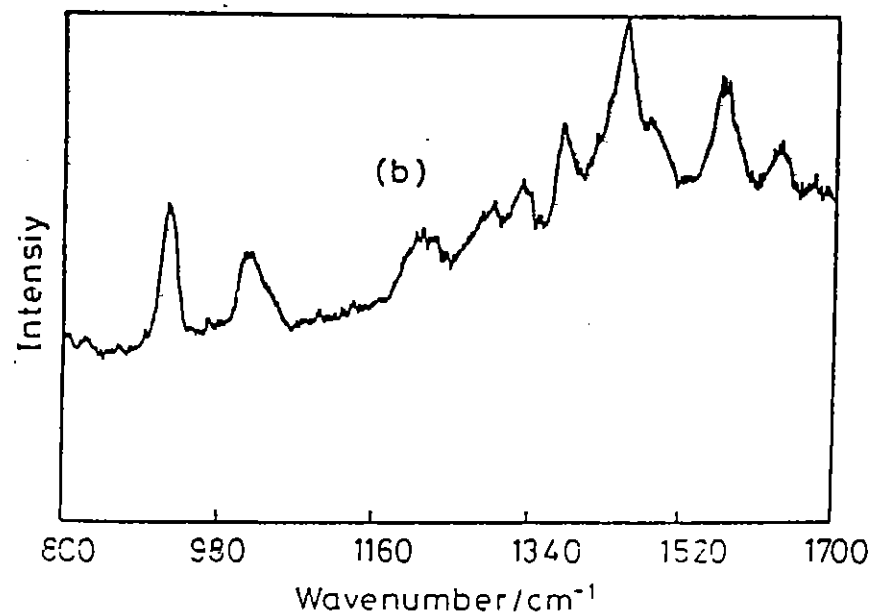
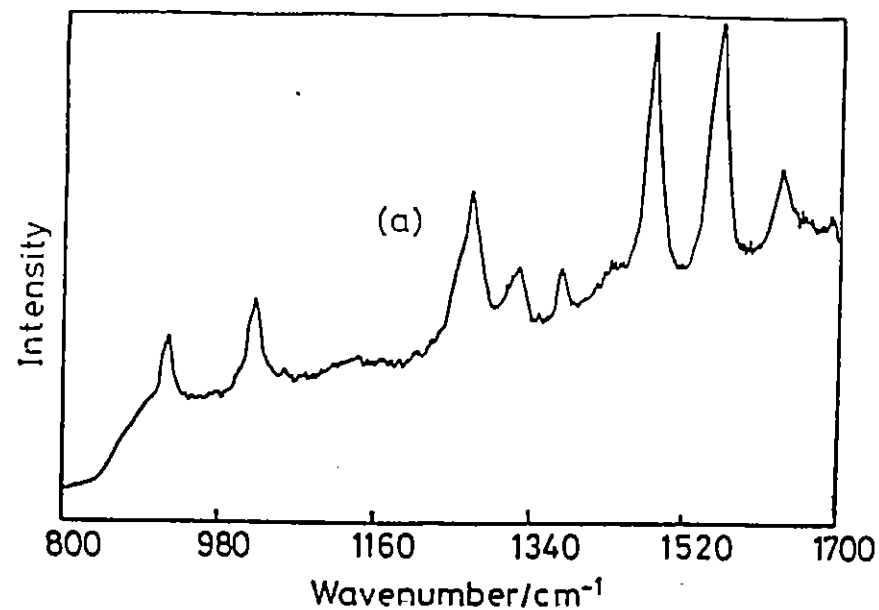


Fig. B1.6 Resonance Raman spectra of ruthenium(II) tris(2,2'-bipyridyl-4,4'-diethylcarboxylate) in acetonitrile/water mixture.
(a) Ground state, λ_{exc} , 488 nm; spectral resolution, 5-6 cm^{-1} ; and
(b) excited state, λ_{exc} , 390 nm; spectral resolution, 5-6 cm^{-1} .

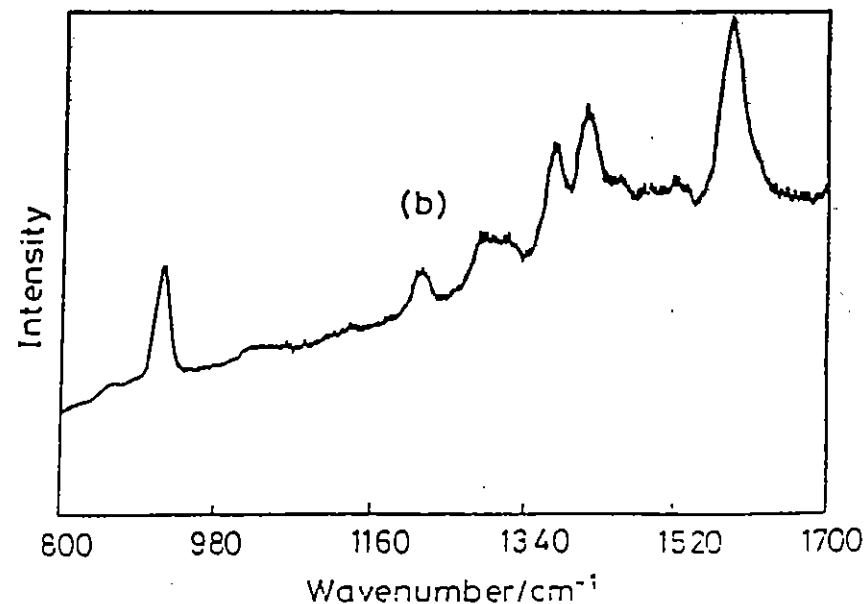
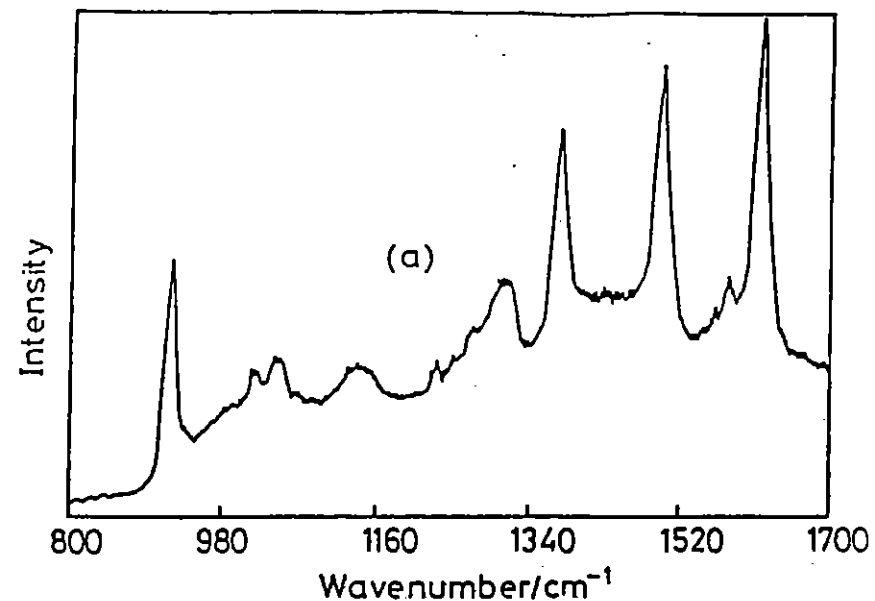


Fig. B1.7 Resonance Raman spectra of ruthenium(II) tris(2,2'-bipyridyl-5,5'-diethylcarboxylate).
(a) Ground state, λ_{exc} , 488 nm; spectral resolution, 5-6 cm^{-1} ; and
(b) excited state, λ_{exc} , 390 nm; spectral resolution, 5-6 cm^{-1} .

In Figures B1.6 and B1.7, the ground and excited states of RB4ET and RB5ET, respectively, are presented. A careful study of these spectra shown the presence of bands attributable to an electron localised ligand in their excited state⁽⁷⁾. Thus, we have established that, unlike the electro-reduced species but similar to photoexcited states of other ruthenium(II) bipyridyl complexes, in the cases of RB4ET and RB5ET excited states the odd electron is localised on one of the three ligands.

Fluorescence quenching measurements have indicated that when RB4ET is adsorbed on TiO₂, visible light excitation leads to injection of the excited state π^* electron into the conduction band of the semiconductor^(1a). From kinetic studies, the time taken for electron injection is calculated as < 10-20 ns, whereas the recombination of conduction band electron and the oxidised complex occurs in ca. 1 μ s. Since we have found that the lifetime of the RB4ET excited state is only 350 ns, we should be able to distinguish the products after electron injection from the excited state. At present, we are using two-laser experiments to study the resonance Raman spectra of such photo-products. We hope that these studies may provide new insights into the mechanisms of photosensitization of colloidal semiconductors.

3. Acknowledgements

We thank Dr A W Parker and Miss S Tavender for their valuable technical assistance at RAL and the SERC for financial support.

References

1. (a) N Vlachopoulos, P Liska, J Augustynski, and M Grätzel, personal communication - paper submitted to J. Am. Chem. Soc.

(b) F P Rotzinger, S Munaralli, P Comte, J K Hurst, M Grätzel, F Penn and A J Frank, J. Am. Chem. Soc., **109**, 6619 (1987).
2. D Duonghong, N Serpone, and M Grätzel, *Helv. Chim. Acta.*, **67**, 1012 (1984).
3. D N Furlong, D Wells, and W H F Sasse, J. Phys. Chem., **90**, 1107 (1986).

4. S Umapathy, G Lee-Son, and R E Hester, J. Chem. Soc. Chem. Comm., **24**, 1841 (1987).
5. S Umapathy, G Lee-Son and R E Hester, submitted to J. Mol. Structure.
6. R Donohoe, C D Tait, M K DeArmond, and D W Wertz, J. Phys. Chem., **90**, 3927 (1986) and reference therein.
7. S Umapathy, A M Cartner and R E Hester, manuscript in preparation.

B1-4 ACCELERATED LASER AGEING OF PLASTICS

Dr K G Snowden STC

STC Technology

This work is being carried out under the European Communities BRITE programme. The BRITE programme (Basic Research in Industrial Technologies for Europe) aims to stimulate the development of a solid foundation of advanced technologies to support traditional "smokestack" industries. In this research (BRITE Contract No. 1522) STC Technology, Standard Elektrik Lorenz (Project leader) and several European car makers (Daimler Benz, Renault, Peugeot, Fiat, Volkswagen and Austin Rover) are co-operating to evaluate a novel test procedure to accelerate the ageing/weathering of plastics. The novel test procedure devised by STC Technology uses pulsed excimer lasers to rapidly age various polymeric materials.

Background

Polymeric materials operating indoors and outdoors are subject to ageing. Natural forces such as sunlight, temperature, oxygen, water, mechanical stress etc. either singly or in combination, can cause catastrophic failure of certain materials in a matter of months or several years in others. Since the required lifetimes of many components often exceed 10 years, there is a continuing need for accelerated ageing tests. Presently available artificial ageing equipment such as weatherometers provide typical acceleration rates of only 2:1 to 5:1. Thus there is an urgent need for artificial weathering tests with increased rates of acceleration.

One of the most potent natural ageing forces is the ultraviolet (uv) portion of sunlight. Ultraviolet radiation on its own has sufficient energy to break many of the molecular bonds found in polymeric materials. This project evaluates the use of a pulsed ultraviolet excimer laser in accelerated ageing. In principle massive amounts of ultraviolet radiation can be delivered by the excimer laser in very short times without significant overheating of the sample. Laser ageing is being compared to natural and weatherometer ageing to ensure correlation and that acceleration factors can be assessed.

Experimental

Excimer laser ageing is being carried out using 308 nm and 276 nm on 17 plastics and 3 paint samples. During the past year samples have been exposed at the Laser Support Facility to 308 nm XeCl laser radiation. Energy densities of 3, 6 and 9mJ/cm² have been used for exposure periods of up to 2 hours at repetition rates of 20, 50 and 100 Hz. 276 nm radiation has been generated by the use of a KrF pumped, hydrogen filled, Raman shifter. The 1st Stokes shift of hydrogen was used to deliver about 20-30mJ of 276 nm radiation. Energy densities of 3, 6 or 9mJ/cm² were again used, but a lower repetition rate of 10Hz was dictated by the excimer pump laser output.

Results

The primary measure of ageing in the materials is a surface discolouration parameter known as ΔE^* (Total Colour Difference). A target of a minimum $\Delta E^*=1.5$ was set for the laser and other ageing treatments.

Acceleration Factors

For many of the polymers, insufficient time has elapsed (~ 1 year) during natural ageing to produce $\Delta E^* = 1.5$. However, the following limited results are available for ABS (Acrylonitrile-Butadiene-Styrene) and modified PPO (Polphenylene Oxide) polymers;-

Polymers	Laser v Natural Ageing	Laser v Weatherometer Ageing
ABS	20,000 - 300,000 : 1	500 - 1500 : 1
Mod. PPO	5000 - 30,000 : 1	300 - 1500 : 1

Other parameters such as changes in surface roughness, gloss, tensile strength, elongation at break and impact strength are currently being evaluated.

B1-5 RESONANCE RAMAN SPECTROSCOPY OF NITROBENZENE

V Foster and S R Ahmad (RMCS, Shrivenham, Swindon)

Nitrobenzene (NB) in gas phase has a prominent absorption band which peaks at about 240 nm. This band has been identified⁽¹⁾ as an intramolecular charge-transfer state caused by the resonance interaction between the benzene ring and nitro-group. Excitation to this state has been found to produce copious photodissociation.

The present investigation is part of a long-term project to study the geometries and photodissociative pathways of such excited states in NB and other nitro-aromatic molecules. Initially the studies are being carried out on NB in liquid phase using ethanol as a solvent. This choice of solvent results in a shift of the 240 nm absorption band to around 258 nm. Ideally the solvent would be used as an internal standard for normalization enabling experimental parameter variations from one spectrum to another to be eliminated, but no suitable solvents have as yet been characterized in this wavelength region. It is hoped that a Raman excitation profile (REP) will provide details of the vibronic structure of this absorption band enabling prediction of the excited state geometries with greater facility and accuracy than would be possible using absorption spectroscopy.

To obtain a REP over the relevant wavelength region (240 nm-280 nm), frequency-doubled output from the excimer-pumped FL3002 dye laser was used. A beta-barium borate crystal was used for the frequency-doubling and outputs of up to 4mJ were achieved giving a conversion efficiency of over 20%. The frequency-doubled component of the laser beam was separated from the fundamental using a 90° quartz prism. This necessitated total realignment of the detection optics at each excitation wavelength due to the variation in optical path of the beam. Use of a filter to eliminate the fundamental together with a compensating crystal to correct beam "walk-off" would have eased this situation. The SPEX Triplemate Spectrometer and OSMA-SI diode array system were used for detection and analysis of the Raman Spectra. A capillary flow system was designed to enable fresh sample to be presented at each laser shot, thereby avoiding effects of photodissociation.

Raman spectra of 2×10^{-3} mol dm⁻³ NB in ethanol were obtained at five discrete excitation wavelengths in the range 240 nm - 280 nm. Examples of these are shown in Figure B1.8. The fluorescence background increased drastically at the longer excitation wavelengths making the Raman peaks difficult to retrieve. This problem was overcome by performing pedestal subtraction on the data as shown in Figure B1.9, using software developed by Dr R S Chittock (RAL). The signal-to-noise ratio was not improved by this manipulation and so there was a high uncertainty in the Raman Intensities (RI) measured at these excitation wavelengths.

The RI of the NB peak at 1350 cm⁻¹, due to symmetrical stretch in the nitro-group, was normalized with respect to the RI of the ethanol peak at 1454 cm⁻¹ for each excitation wavelength, having first corrected the RIs of the ethanol for NB absorption of the excitation intensity, and plotted against excitation wavelength to give the REP shown in Figure B1.10. The errors shown in the figure are primarily due to those in the correction of ethanol for NB absorption and to the observed low signal-to-noise ratio at the longer excitation wavelengths. The latter could be improved upon by using longer accumulation times.

Further investigation into the nature of the REP of ethanol over this wavelength range is necessary before the results can be interpreted.

The only conclusion that may be drawn from the non-linearity of the REP shown in Figure B1-10 is that the REPs of NB and ethanol differ significantly in this region.

We intend to repeat the experiment in greater detail using more uv excitation wavelengths if a longer period for use of the equipment at the LSF becomes available. We are, meanwhile, investigating the suitability of the chosen solvent and are characterizing the REP of ethanol in the near uv region.

References

1. Nagakura, Kojima and Maruyama, J. Mol. Spectrosc., 13, 174 (1964).

B1-6 LASER EXCITATION SPECTROSCOPY OF TRANSITION ELEMENT HALIDES IN SUPERSONIC BEAMS

L R Zink, J M Brown (Oxford), T R Gilson and I R Beattie (Southampton)

Many first row transition element chlorides can be vaporized to give a few torr pressure at temperatures below 1000°C. These molecules are seeded into a high pressure (several atmospheres) of an inert gas, followed by expansion through a jet into a rapidly pumped vacuum system. In this way the rotational and, to a lesser extent, vibrational energy is converted into translational energy. This isentropic expansion results in rotationally cold molecules (~ 10K). The jet is irradiated with the beam of a narrow band tuneable dye laser and the resultant fluorescence recorded (laser excitation spectroscopy). The excimer pumped dye laser at the LSF has been used in this way to record survey spectra of several species. It has even been possible to resolve rotational structure by the use of the intra-cavity etalon.

In earlier experiments at the LSF, a spectrum was obtained with a stainless steel nozzle which we thought might be due to FeCl₂ (the same spectrum could be obtained with a whole variety of chlorine containing compounds). Since then we have developed quartz and, very recently, alumina nozzles which are not attacked by the hot chemicals. Using

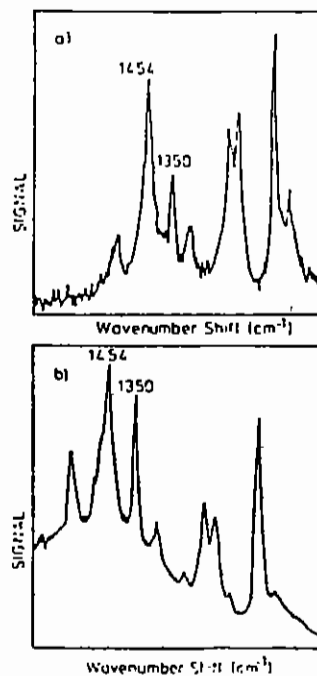


Fig B1.8 Raman spectra taken at excitation wavelengths
(a) 240nm and
(b) 260nm, showing 1350 cm^{-1} NB peak and 1454 cm^{-1} ethanol peak. Each spectrum is averaged over four data accumulations each of approximately 2600 laser shots.

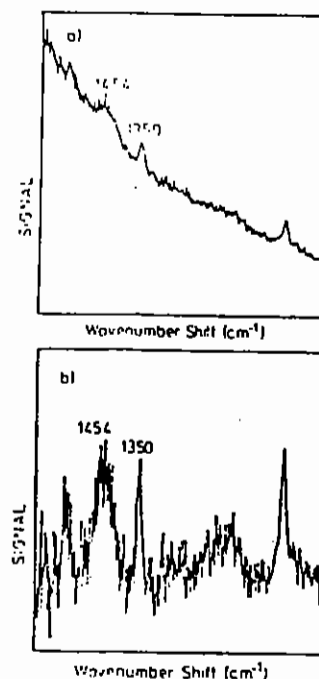


Fig B1.9 Raman spectrum at excitation wavelength 280nm
(a) before and
(b) after pedestal subtraction.

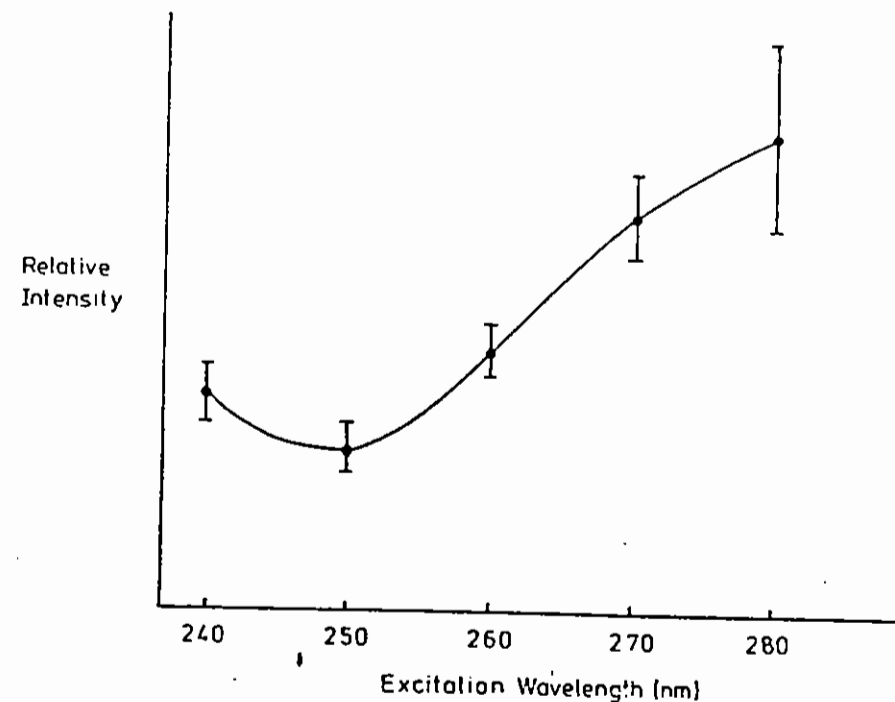


Fig B1.10 Raman excitation profile of NB with respect to ethanol.

these nozzles, we have been able to establish that the carrier of our spectrum is in fact NiCl_2 . We have recorded extensive vibrational structure between 350 and 375 nm and investigated the dependence of individual bands on stagnation pressure, to see how they are affected by temperature. The vibrational sequences in the excitation spectrum have spacings of $146(2) \text{ cm}^{-1}$ and $370(3) \text{ cm}^{-1}$. Assuming a linear molecule, these intervals can be assigned to twice the bending frequency and the symmetric stretching frequency respectively of NiCl_2 in an excited electronic state. We have also dispersed the fluorescence from a few selected bands with a small monochromator. Extensive progressions of 357 cm^{-1} at spacings of 177, 401 and 1026 cm^{-1} were observed. There have been assigned to the symmetric stretching frequency ($\nu_1 = 357(7) \text{ cm}^{-1}$, twice the bending frequency ($2\nu_3 = 1026(6) \text{ cm}^{-1}$). We do not yet have a definite assignment of the 401 cm^{-1} interval; it could possibly be a spin-orbit splitting.

We have also recorded bands in electronic transitions of CuCl between 420 and 470 nm. Rotational structure is easily resolved with the intra-cavity etalon in place in the dye laser. The experiments performed at the LSF permitted us to identify the carrier and improve our methods of production. The best signals were obtained by reacting CCl_4 with hot copper wire in the nozzle.

We have also made tentative observations of three other transition element dichlorides, FeCl_2 , CoCl_2 and CuCl_2 . These observations need to be confirmed and extended.

B1-7 DO HIGHER EXCITED STATES OF THE URANYL ION LUMINESCENCE?

I D Morrison and R G Denning*, (Inorganic Chemistry Laboratory, South Parks Road, Oxford, OX1 3QR)

1. Introduction

Because of our long and extensive interest in the optical properties of actinyl ions we were particularly interested in a report by Marcantonatos et al⁽¹⁾ that describes emission at shorter wavelengths than the familiar green emission, in crystals of $\text{CsUO}_2(\text{NO}_3)_3$, $\text{UO}_2(\text{NO}_3)_2 \cdot 6 \text{ H}_2\text{O}$ and $\text{UO}_2\text{SO}_4 \cdot 3.5 \text{ H}_2\text{O}$ and also in borosilicate glass doped with the uranyl ion; it can be observed at 77 K and at room temperature. The report concentrates on the short-wavelength emission of $\text{CsUO}_2(\text{NO}_3)_3$, a compound whose polarised absorption spectrum was

measured some years ago⁽²⁾. It is suggested in⁽¹⁾ that the spectral structure of this new 'violet' emission, which shows evidence of vibrational progressions typical of the uranyl ion, may occur from at least some of the six excited states known to occur⁽²⁾ at energies above the first excited state. The most puzzling aspect of this novel emission is its time evolution, which was measured by excitation at 343 nm using an excimer pumped dye laser. The luminescence decay appears to be compatible with two independent exponential processes⁽¹⁾, whose decay times are only a slowly varying function of the emission wavelength, varying by less than 40% over the whole 7000 cm^{-1} range of the violet emission. The slower of the two decay times is actually longer than that of the conventional luminescence, for which the rate is 494 s^{-1} at 77 K. When excited at 260 nm in a spectrofluorimeter the integrated 'violet' emission has about 3% of the intensity of the green emission.

We have tried to establish the authenticity of this emission.

2. Experimental

A single crystal of $\text{CsUO}_2(\text{NO}_3)_3$, grown from aqueous solution [2,3,4] with prismatic habit, and approximate dimensions $3 \times 4 \times 2 \text{ mm}$, was mounted on a thin brass plate supported on the central axis of an Oxford Instruments CF 204, continuous flow cryostat. By rotating the mounting rod the brass plate may be reversed, so that its back is exposed to the incident light and the sample is not directly illuminated. The normal to the mounting plate bisects the right angle formed by the entrance window axis and the collection window axis. The excitation pulse was derived from a Lambda Physik Excimer Laser, Type EMG 150, lasing on the KrF transition at 249 nm. The beam was attenuated, collimated and loosely focused to cover the area of the sample. Measured pulse energies at the sample were approximately 1 mJ, with a repetition rate of 5 Hz. The emission was collected with f/5 quartz optics and dispersed in a Hilger and Watts Monospek 600 mm focal length monochromator. A Princeton Applied Research OMA-3 optical multichannel analyser was placed in the focal plane of the monochromator. The array can be gated to follow the time dependence of the emission, with a time resolution of 1 ns.

3. Results and Discussion

Figure B1.11(a) shows the light detected by the diode array in the spectral range 450–475 nm from a single crystal of $\text{CsUO}_2(\text{NO}_3)_3$ at 4.2 K under 249 nm irradiation. In this experiment the diode array integrates quasi-continuously over 2000 laser shots and detects all emitted light regardless of its time evolution. The origin of the conventional luminescence is seen at 473 nm as expected⁽²⁾. Light detected in the spectral range 450–470 nm is approximately 100 times weaker and shows some spectral structure. In this region the total number of counts is about 300 per array element after the accumulation of 2000 laser pulses. The efficiency of the array is approximately 1 count per 8 photons, giving a flux of 1.2 photons per pulse per pixel, each pixel covering a spectral range of 0.3 Å. By gating the array on at various delay times after the laser pulse it was established that the weak 'violet' source is undetectable one microsecond after excitation and that its lifetime is about 200 ns. To test whether the source is intrinsic to the crystal the sample mount was rotated by 180° to expose its rear surface to the beam. The result is shown in Figure B1.11(b). Light is again observed in this spectral region with comparable intensity (~200 counts over 2000 shots) but with no spectral features. Gating shows this source to have approximately the same lifetime as the light detected in this region when the crystal is irradiated. A small amount of light, scattered within the cryostat, reaches the crystal on the backside of the mount exciting detectable luminescence at 473 nm, but this is now of comparable intensity to the violet source. It follows that the violet source in our experiment is not intrinsic to the crystal and we suggest that it may represent broadband emission either from low-level impurities in the cryostat windows, from involatile organic material coating those windows, or from traces of organic residues on the sample mount itself.

The spectral structure between 452 and 470 nm in Figure B1.11(a) is readily explained if this light passes through the crystal and its intensity is determined by the crystal transmittance. The sharp decrease in the intensity at wavelengths shorter than 461 nm is expected from the onset of strong absorption at that wavelength⁽²⁾.

Faced with this result we thought that the spectral structure of the violet emission reported in⁽¹⁾ might have a similar explanation. Accordingly we recorded the absorption spectrum of a thin single crystal of $\text{CsUO}_2(\text{NO}_3)_3$, using unpolarised light⁽³⁾, at 77 K with a

spectral bandpass of 0.8 nm, conditions chosen to be comparable to those employed in⁽¹⁾. Figure B1.12 shows the transmittance of this crystal and compares it with a reproduction of the emission spectrum presented in⁽¹⁾. In the region from 470 to 500 nm where the conventional luminescence occurs the spectra are naturally very different but to shorter wavelength they are almost identical in every detail; small differences are attributable to (i) the crystal thickness (no crystal dimensions are given in⁽¹⁾), and (ii) partial polarisation of the source⁽³⁾ combined with strong linear dichroism in the absorption spectrum⁽²⁾ (which ensures some transmission of unpolarised light even in rather thick crystals). It is clear that the spectral structure of the violet 'emission' in reference⁽¹⁾ is no more than a manifestation of the transmission of the sample from a source with fairly flat spectral density. An analysis of the apparatus used in⁽¹⁾ suggests that this source is probably the stray light passed through the excitation monochromator from the xenon arc source.

4. Summary

There is no evidence for spectrally structured violet emission in $\text{CsUO}_2(\text{NO}_3)_3$. Structure observed in this region is a consequence of the transmission of the crystal. This structure was, in our view, the most persuasive reason for identifying the violet emission with the higher excited states of the uranyl ion and led the authors of⁽¹⁾ to assign the spectrum to emission from specific excited states.

At 4.2 K, under 249 nm pulsed excitation, we can set an upper limit on the intensity of the violet emission, at times longer than 1.0 microsecond, at 0.1% of the green emission.

In our opinion it remains to be proven that violet emission is intrinsic to the sample. We suggest that, as yet, it has not been established that the higher excited states of the uranyl ion luminescence.

Acknowledgements

We thank the Science and Engineering Research Council for the support of I.D.M., for a research grant and for access to its Laser Support Facility. We are particularly indebted to Dr A J Langley and Dr A W Parker for their assistance.

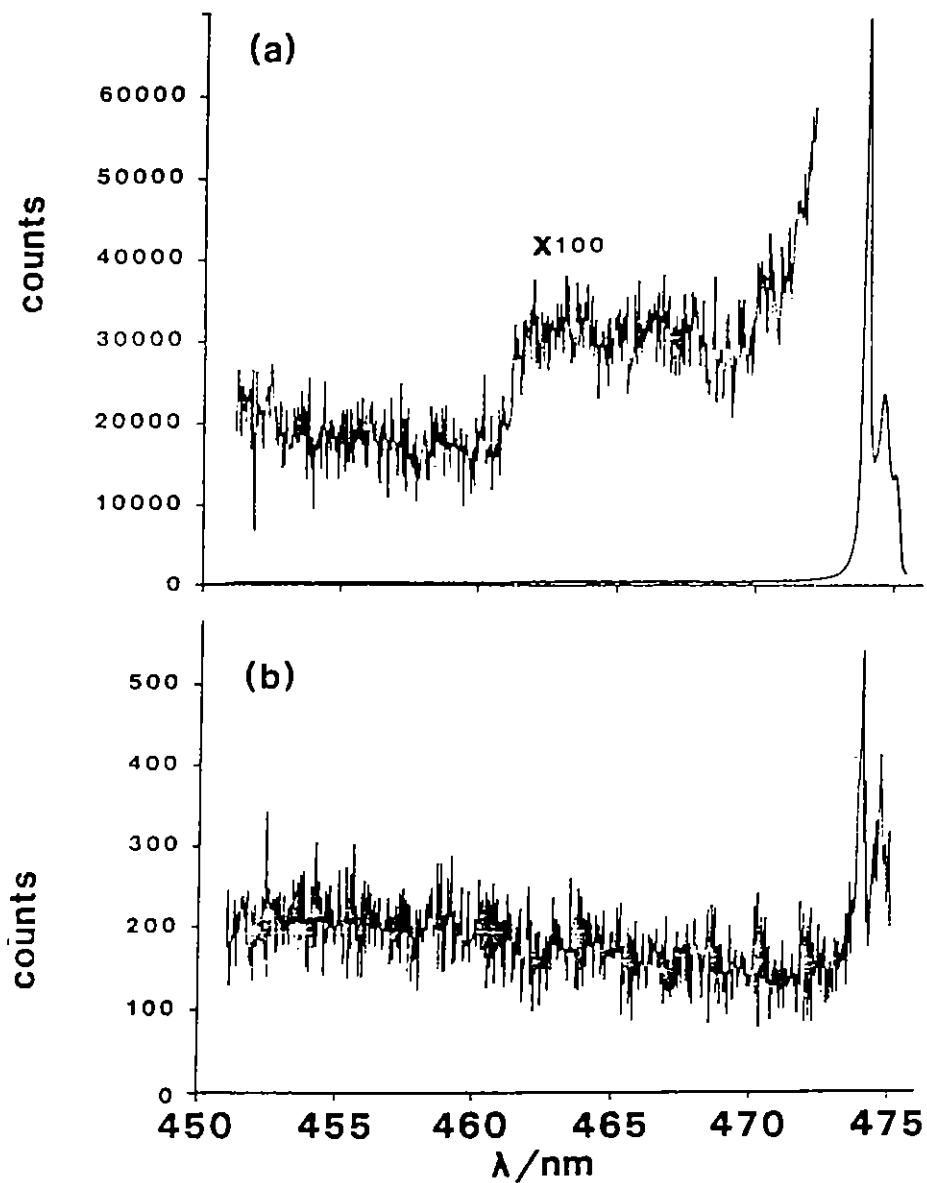


Fig. B1.11 (a) Spectrum of light detected when $\text{CsUO}_2(\text{NO}_3)_3$, at 4.2 K, is irradiated at 249 nm.

(b) The spectrum with the sample mount reversed (see text) under the same conditions as (a).

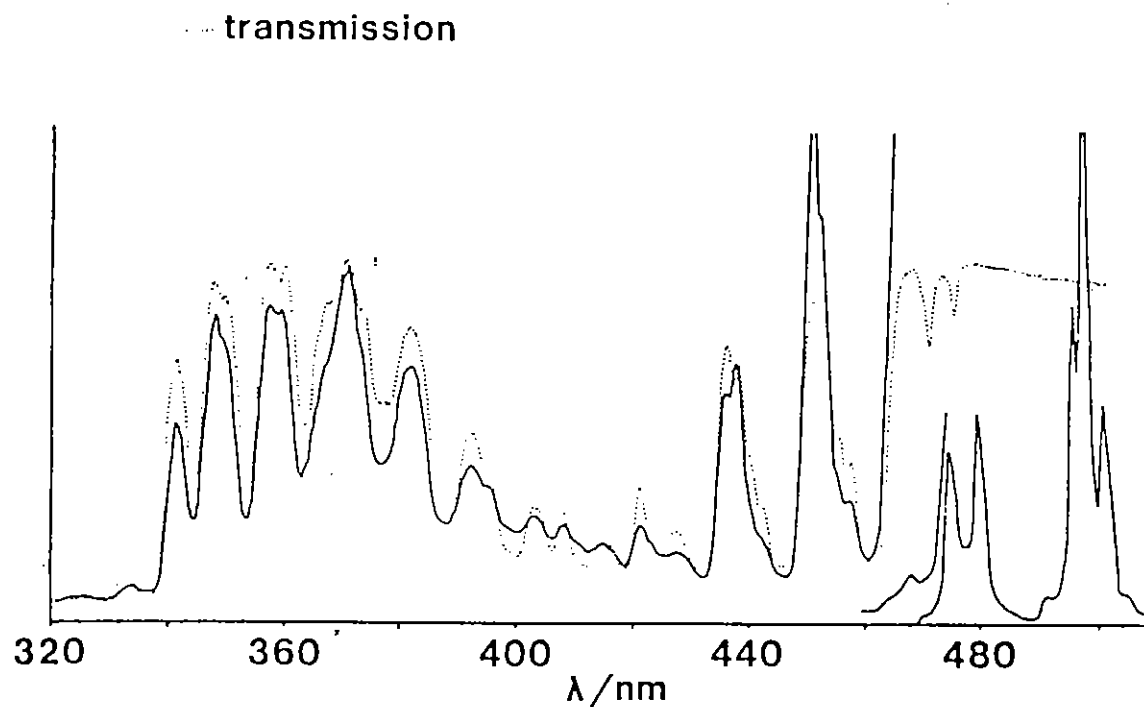


Fig. B1.12 — the spectrum of the violet emission from $\text{CsUO}_2(\text{NO}_3)_3$, at 77 K, reproduced, with permission, from reference [1]; the transmission spectrum of a single crystal of $\text{CsUO}_2(\text{NO}_3)_3$, at 77 K.

References

1. M D Marcantonatos, C Altheer, R Reisfeld, and C K Jorgensen, Chem. Phys. Lett. 132 (1986) 247.
2. R G Denning, D N P Foster, T R Snellgrove and D R Woodwark, Mol. Phys., 37, (1979) 1089.
3. Unpolarised in this context means the absence of a polariser. Light from the grating monochromator is partially polarised to a degree which is a function of wavelength.

B1-8 MEASUREMENT OF THE CF $A^2\Sigma^+$ RADIATIVE LIFETIME

J P Booth & G Hancock (Physical Chemistry Laboratory, South Parks Road, Oxford OX1 3QZ)

Introduction

An accurate measurement of the fluorescence lifetime of the $A^2\Sigma^+$ state of CF is needed in order to calculate the Einstein A and B coefficients for the A-X transition. These quantities are necessary for the determination of the absolute concentration of groundstate CF generated by a radio-frequency discharge in CF_4 in order to compare the experimentally observed radical concentration with those predicted from kinetic models of the plasma processes. Such models are of importance in efforts to understand the mechanism for plasma etching of semiconductors, for example in the etching of Si and SiO_2 surfaces by CF_4 and other fluorinated hydrocarbons. There is also some interest in the lifetimes of various vibrational levels of the A state as theoretical calculations^(1,2) have indicated that predissociation should occur for $v'=2$ and above. Emission spectra from $v'=2$ are reported to show evidence of predissociation⁽³⁾. The only reported measurement of the CF A state lifetime was carried out by Hesser and Dressler in 1966⁽⁴⁾. They produced electronically excited CF by electron impact upon $CF_2=CF-CF=CF_2$ and observed a lifetime of 19 ± 2 ns for fluorescence at 231 nm which was attributed to (1,1) emission. The lifetime of the $v'=0$ level was not reported.

A preliminary measurement in this laboratory of the $v'=0$ lifetime by LIF in an rf plasma in CF_4 indicated a value closer to 30 ns⁽⁵⁾. Although care was needed in interpreting this result, the possibility that predissociation was shortening the lifetime of the $v'=1$ level warranted a more careful measurement of the lifetime of both the $v'=0$ and the $v'=1$ levels.

Experimental

The reaction vessel used was a commercial plasma etching chamber (Plasma Technology RIE80). The CF radical was generated by a 100W 13.56 MHz discharge in pure CF_4 at pressures between 50 and 500 mTorr. Radiation at 220-240nm was generated by an excimer pumped dye laser (Lambda Physik EMG201MSC and FL2002), using coumarin 47 dye, and frequency doubled with a Potassium Pentaborate (KPB) crystal. This system could produce a peak of 2 mJ output, but the excimer pump beam was usually attenuated such that the final output was around 100 μ J, so that the transition would not be saturated, the operating lifetime of the dye would be extended, and a repetition frequency of 50Hz could be used without damage to the KPB crystal.

Fluorescence emitted perpendicular to the laser beam was collected and focussed into a monochromator (Edinburgh Instruments 121S15, f4, resolution 1.5nm with .25 mm slits) with two spectroil B lenses, and detected with an EMI 9816QKB photomultiplier. A fast gated sampler (Stanford Research systems SR 255, 1ns gate) was used to monitor the time behaviour of the electrical signal from the photomultiplier. This device was triggered by a signal from a photodiode illuminated by scatter from the excimer pump laser and amplified with a x100 radio frequency amplifier. The sampling gate delay was scanned from 0 to 100ns in 0.5ns intervals, incrementing after each shot, and repeating the scan 500 times to allow averaging and estimation of the uncertainty of the measurement at each point, whilst eliminating systematic errors due to slowly varying fluctuations in conditions. Scanning delays and signal acquisition were controlled by an IBM PCXT computer.

The combination of the laser pulse shape and the time response function of the measuring system was estimated by filling the chamber with atmospheric pressure air and tuning the monochromator to resonance with the laser frequency to observe scattered radiation.

Results and discussion

Figure B1.13 shows the laser pulse shape and LIF signal following excitation of the CF (A-X) (1,0) band at 223.86nm, corresponding to the strong P_{11} bandhead, in a plasma of 50mTorr CF_4 at an RF power of 200W. The electrical noise due to the excimer laser discharge and laser scatter signal has been subtracted from these traces by performing an identical run with the rf excitation turned off. If the data after the peak in each LIF trace is fitted to a simple single exponential decay, a poor fit is obtained, and an erroneous value for the fluorescence lifetime is obtained as the laser pulse lifetime is of comparable magnitude. Shortening of the laser pulse duration would be the best method of obtaining the true lifetime, but this was not possible with the equipment available. It was therefore necessary to analyse the data in a more sophisticated way, and the method chosen was to use a non-linear least-squares fitting program to fit the best convolution of an exponential decay with the experimentally measured laser pulse shape (which is a convolution of the true laser pulse shape and the instrumental response function) to the measured fluorescence signal shape.

The value obtained for $v'=1$, 25.9 ± 4 ns, was found to be invariant with RF power and pressure over the ranges studied (25-250W, 50-500mTorr): with a gas kinetic quenching rate constant only 2% decrease of the lifetime from its zero pressure value would be expected at the highest pressure used in this study. A similar measurement for $v'=0$ (whose excitation was at an isolated feature at 232.22nm) yielded a value of 26.7 ± 4 ns. As a check on the time response of the system, the radiative lifetime of the (0,2,0) level of CF_2 $\tilde{A}(^1B_1)$ excited at 260.1nm was measured and is shown in Figure B1.14: the value, 51.1 ± 3 ns is in good agreement with the most recently published value of 50 ± 5 ns⁶.

No evidence for predissociation in $v'=1$ of CF was observed up to $J'=23.5$ from observations of the laser induced fluorescence excitation spectrum. Our value of 25.9ns for $v'=1$ thus represents the true radiative lifetime, and the difference between this and the previously reported value of 19ns, although small, lies outside the combined error limits. Contributions to fluorescence from predissociated CF levels could account for the smaller value, but this would require electron impact excitation of high ($J' > 23.5$) rotational levels of CF ($v'=1$).

The estimation of the absolute concentration of a species by the magnitude of a Laser Induced Fluorescence signal is possible in theory if the transition probability is known, but an accurate estimate of the efficiency of the detection system is needed (including the solid angle of detection, the transmission efficiency of the monochromator and the quantum efficiency of the photomultiplier). A much better method is to compare the intensity of the signal obtained with that obtained from LIF of a known concentration of a stable molecule. This is the strategy adopted here, taking advantage of the fact that the NO molecule, isoelectronic with CF, has its A-X transition at a very similar wavelength. For this experiment the NO (0,1) transition at 236.1nm was excited and (0,2) fluorescence at 247.4nm was observed, and this signal was compared to that obtained by excitation of the CF (0,0) band at 232.9nm with observation of the (0,2) fluorescence at 247.6nm.

With knowledge of the radiative lifetimes and Franck-Condon factors for both NO and CF, the relative concentrations and hence the absolute concentration was obtained. At 50mTorr CF_4 in a 100W rf plasma the CF concentration of the CF radical was determined to be $3.4 \pm 0.5 \times 10^{11} \text{ cm}^{-3}$, corresponding to 0.02% of the initial CF_4 pressure. Although this concentration is low (of the order of 10% of the most abundant non-atomic free radical species, CF_2), it acts as a branching chain centre for polymerisation at electrode (and substrate) surfaces, and can thus influence the stability of polymers produced and deposited during the etching process.

References

1. J A Hall and W G Richards, *Molecular Physics* 23, 331, (1972)
2. W P White, R M Pitzer, C W Mathews and T H Dunning, *Journal of Molecular Spectroscopy* 75, 318, (1979).
3. W P White, Dissertation, Ohio State University, (1971).
4. J E Hesser and K Dressler, *Journal of Physical Chemistry*, 45, 3149, (1966), and J E Hesser, *Journal of Chemical Physics*, 48, 2518, (1968).
5. J Wild, Thesis, St Hugh's College, Oxford, (1986).

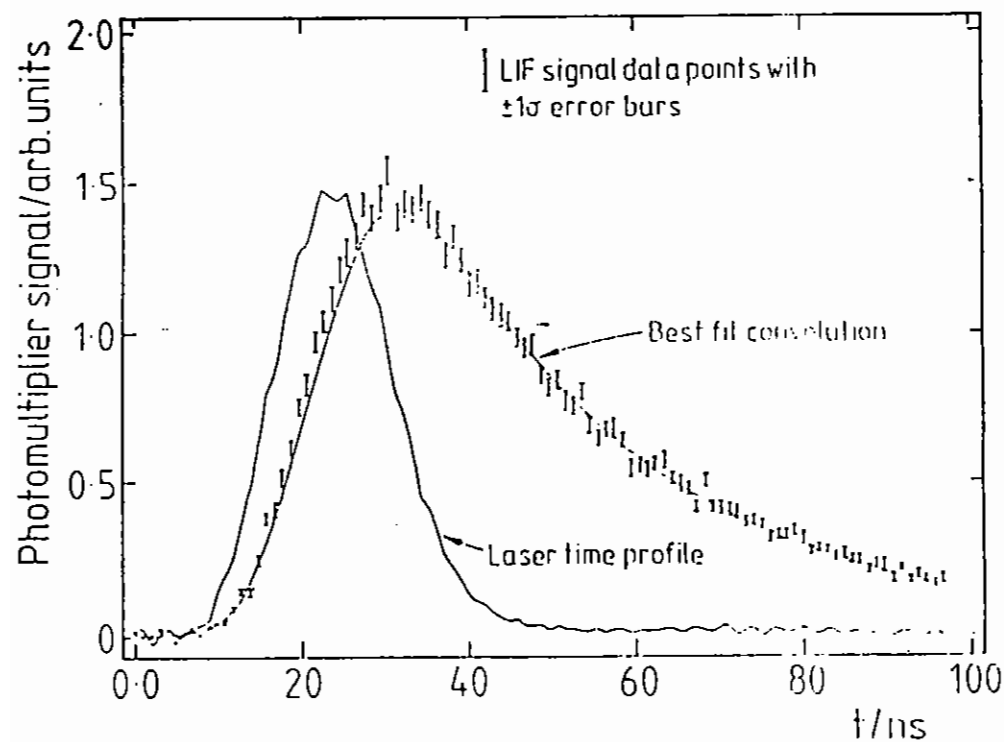


Fig. B1.13 Time dependence of the fluorescence of CF ($A^2\Sigma^+ v'=1$) excited at 223.86nm. The laser pulse time profile and the best fit convolution to the LIF signal are also shown. CF was produced in an RF plasma of 200mTorr CF_4 at a power of 200W.

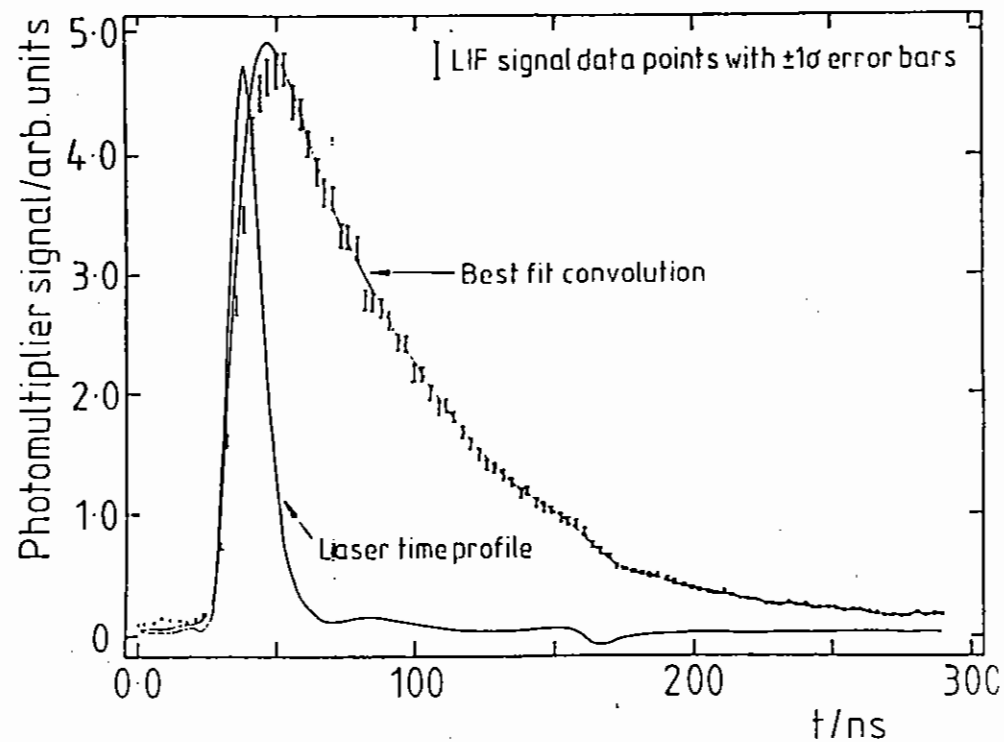


Fig. B1.14 Time dependence of the fluorescence from CF_2 ($X^1B_1, 0,2,0$) excited at 260.1nm. Conditions as in Fig. B1.13.

B1-9 LASER FLASH PHOTOLYSIS/LASER ABSORPTION STUDIES OF SiMe_2 RADICAL KINETICS

J E Baggott, M A Blitz, H M Frey, P D Lightfoot and R Walsh (Dept. of Chemistry, University of Reading)

1. Introduction

There is currently considerable interest in the chemistry of silylenes, reactive divalent silicon intermediates of general formula SiX_2 which are the silicon analogues of the carbenes. We recently published⁽¹⁾ the results of the first direct time-resolved studies of the kinetics of the SiMe_2 radical in the gas phase, reporting new room temperature rate coefficients for the insertion reactions of SiMe_2 into the Si-H bonds of a variety of methyl substituted silanes. In this work, we have studied the addition reactions of SiMe_2 with a variety of unsaturated hydrocarbon compounds. Our experimental results have provided considerable insight into the nature of the addition process, and a full account may be found elsewhere⁽²⁾.

A comparison of the gas-phase reactivities of SiMe_2 and singlet CMe_2 cannot be made, since the latter undergoes rapid isomerisation to give propene making studies of its bimolecular reactions impossible. Consequently, we feel that our studies of the kinetics of SiMe_2 radical reactions provide the only sources of experimental information on the insertion and addition processes of reactive species which possess this characteristic type of electronic structure. Singlet methylene, CH_2^1A_1 , can be studied by methods similar to the ones used here,⁽³⁾ but this species is rather indiscriminate and tends to react in the gas phase at near collision rates. There is a further complication in interpreting the results obtained with singlet methylene owing to the occurrence of collisionally induced singlet-triplet conversion. Since silylenes possess singlet ground states such problems do not arise in the interpretation of silylene removal rates.

2. Experimental

SiMe_2 radicals were generated in a static reaction cell by the ArF

(193.3 nm) exciplex laser flash photolysis of pentamethyldisilane. The Lumonics HE 460 exciplex laser was used, providing pulse energies of typically 70 mJ. The SiMe_2 radical exhibits a broad, unstructured absorption feature with a maximum at ca. 450 nm, which can be assigned as the $\tilde{\text{X}}^1\text{B}_1 - \tilde{\text{X}}^1\text{A}_1$ transition. We monitored the concentration of SiMe_2 radicals produced by flash photolysis using the Spectra Physics model 171 Ar⁺ laser operating on the 457.9 nm line in a multipass absorption arrangement. The probe laser beam was multipassed at right angles to the photolysis radiation, 40 passes giving a total path length for radical detection of 3.2 m. Radical concentration-time profiles were recorded using a transient recorder interfaced to a BBC microcomputer. The latter was used as a signal averager.

Bimolecular rate coefficients were determined by measuring the pseudo first-order SiMe_2 decay rate coefficients as a function of the concentration of added reactant. The rate coefficients measured in this study are collected in Table 1.

3. Discussion

The data of Table 1 support a general mechanism for the addition process involving attack of the π -electron system in an initial "electrophilic" phase, followed by a "nucleophilic" phase involving the lone-pair electrons of the radical. This is identical to the mechanism of addition of singlet methylenes to unsaturated compounds proposed by Hoffmann⁽⁴⁾.

Methyl groups are known inductive donors to π -systems, and so the approximate rate doublings observed for SiMe_2 + ethene, propene and isobutene are in accordance with a mechanism which attaches significance to the nucleophilicity of the reactant molecule. Similar behaviour is observed in the series SiMe_2 + ethyne, propyne and but-2-yne. However, the more highly methyl-substituted alkenes do not show the rate increases expected on the basis of increased π -electron availability. We attribute this to a steric effect, in which attack by the incoming SiMe_2 radical is hindered by the presence of the methyl groups. The unsymmetrically methyl-substituted ethenes such as isobutene do not present the incoming SiMe_2 radical with these kinds of steric barriers. This point is illustrated in Figure B1.15, in which

we show SiMe_2 approaching isobutene from the least-hindered end.

Halogen substituents reduce the π -electron availability and hence the bimolecular rate coefficients for SiMe_2 addition. The rate reduction observed on going from chloroethene to fluoroethene are in accord with the electronegativities of the halogen substituents. In trans-1,2-difluoroethene a combination of further reduced π -electron availability and steric effects would seem to be responsible for the dramatic reduction in the addition rate coefficient for this reactant. On the basis of our measurements, we would predict the rate coefficient for the addition of SiMe_2 to 1,1-difluoroethene to be greater than that for trans-1,2-difluoroethene. We hope to test this prediction experimentally in the near future with the aid of a loan laser.

4. References

1. J E Baggott, M A Blitz, H M Frey, P D Lightfoot and R Walsh, Chem. Phys. Lett., 135, 39 (1987).
2. J E Baggott, M A Blitz, H M Frey, P D Lightfoot and R Walsh, Faraday Trans., 2, in press.
3. A O Langford, H Petek and C B Moore, J. Chem. Phys., **78**, 6650 (1983).
4. R Hoffmann, J. Amer. Chem. Soc., **90**, 1475 (1968).

B1-10 PHOTODISSOCIATION DYNAMICS OF HONO_2 AT 280 NM

J August, M Brouard and J P Simons (Dept. of Chemistry, University of Nottingham).

The dynamics of molecules which photodissociate rapidly, relative to their vibrational or rotational period, is at present most readily revealed experimentally via the measurement of both the energy (a scalar quantity) and the momentum (a vector) disposal in the daughter fragments. A technique which is sensitive to these fragment properties, and one which is particularly attractive when applied to relatively complex systems (where photofragment probing may be the only

practical method of unravelling the intricate, dissociation mechanism) has emerged recently in the guise of photofragment Doppler-resolved LIF spectroscopy⁽¹⁾. The technique provides information on both the energy disposal and the mutual orientations and/or alignments between the vectors μ , the parent molecule transition dipole moment, v and j , the fragment velocity and angular momentum vectors. If linearly polarized light is used for photolysis and probing it can be shown⁽²⁾ that Doppler resolved LIF spectra of individual fragment rotational transitions are sensitive primarily to four moments $A_{0,e}^{(2)} (\equiv \frac{4}{5} \beta_0^2 (02))$, $\beta (\equiv 2 \beta_0^2 (20))$, $\beta_0^0 (22)$ and $\beta_0^2 (22)$ which describe respectively the correlations between the vectors μ and j (the rotational alignment), μ and v (the translational anisotropy), v and j and finally the triple vector correlation between μ , v and j . The presence of these correlations reveals itself in the dependence of the Doppler resolved line shape on both the pump-probe geometry and the nature of the probed transition (i.e. $Q\uparrow$ vs P or $R\uparrow$), and their determination can provide new insights into the origin of the fragment linear and angular momenta.

This report describes the measurements of the scalar and vector OH fragment properties arising from photolysis of HONO_2 near the maximum in its first absorption continuum at 280 nm. For this system, analysis of the Doppler resolved profiles is complicated by the spread of OH recoil velocities, which reflect the range of internal energies possessed by the sister NO_2 fragment. However, despite this added complexity much valuable semiquantitative dynamical information can still be obtained⁽³⁾.

Experimental

Details of the experimental and data analysis procedures have been described elsewhere⁽³⁾. Briefly, three pump-probe geometries have been employed, two coaxial geometries (A and B) in which $k_d \parallel k_a$ (a = analysis, d = photolysis laser beams) and $e_a \parallel$ or $\perp e_d$, and a third orthogonal geometry in which $k_a \parallel e_p$ (Case D) (the detector was always placed along e_a). Flowing HONO_2 at 20 mtorr pressure was photolysed at

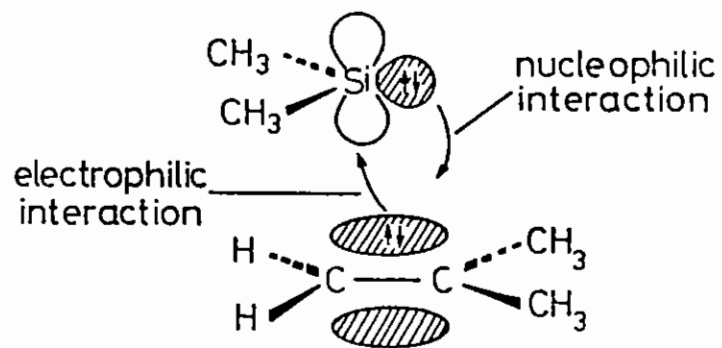


Fig. B1.15 Attack of the SiMe_3 radical by isobutene.

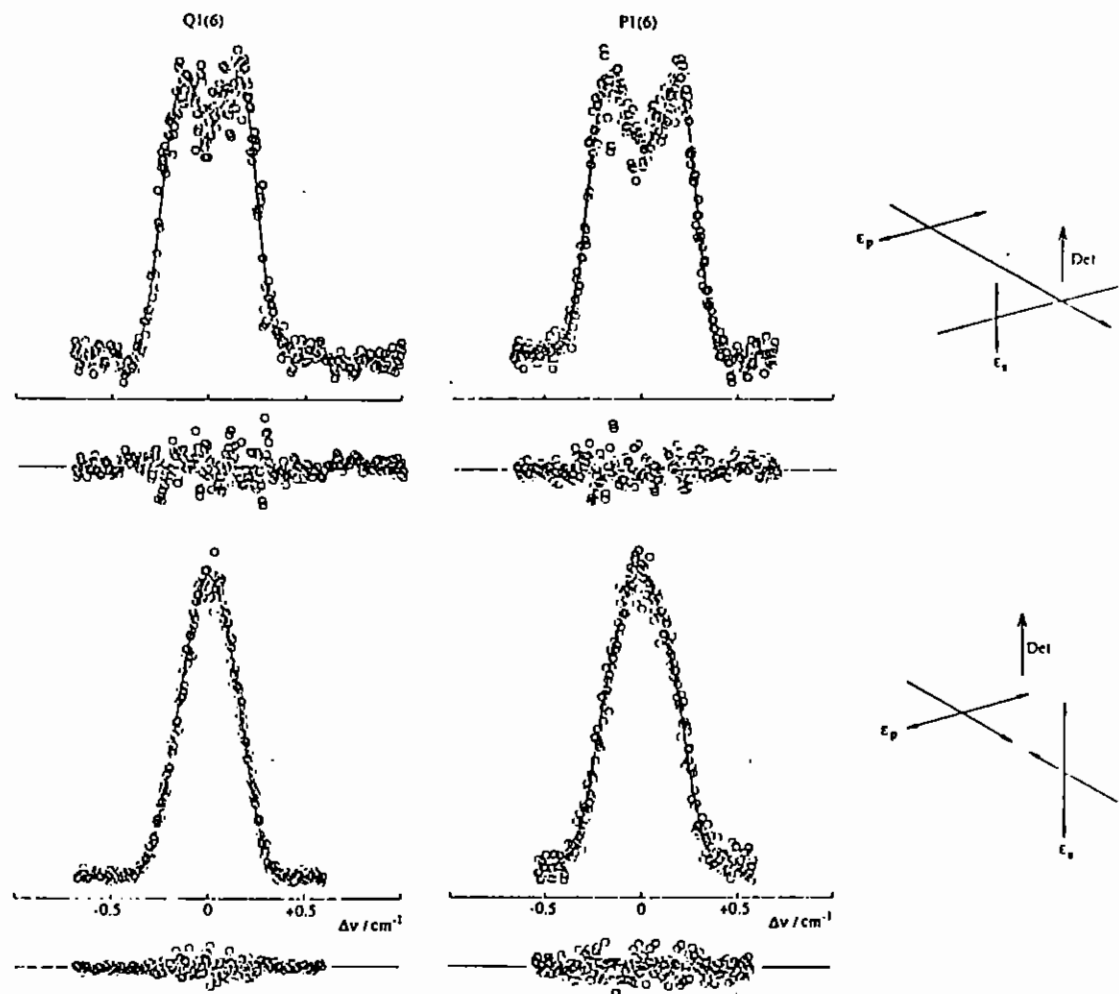


Fig. B1.16 Doppler-resolved profiles for $N''_{\text{OH}} = 6$ in the two pump-probe geometries shown. The fits to the data, which yield the momenta discussed in the text are described in reference 3.

280 nm by frequency doubled Yag or excimer pumped dye laser radiation (10 Hz, ~ 1 mJ pulse $^{-1}$) whilst the OH products were probed after a 10 - 30 ns delay via the 0 - 0 band of the (A - X) transition (tunable, frequency doubled radiation in the 310 - 320 nm region was provided by an excimer pumped dye laser with a linewidth of ~ 0.25 cm $^{-1}$ broadband or ~ 0.08 cm $^{-1}$ etalon narrowed). Both laser beams were expanded and the probe beam attenuated to avoid saturation effects. Fluorescence was detected by a photomultiplier and boxcar system, and transferred to a microcomputer on a shot by shot basis for normalisation to the probe power and averaging.

Results and Discussion

(a) Energy Disposal

The OH fragments were found to be vibrationally cold ($\langle f_v \rangle_{OH} \leq 0.01$, where the excess energy, $E_{av} = 19500$ cm $^{-1}$) and in $v'' = 0$ the rotational distribution was approximately Boltzmann in form ($T \sim 1300$ K, $\langle f_R \rangle_{OH} = 0.046$). The two spin-orbit states were near equally populated (${}^2\Pi_{3/2}; {}^2\Pi_{1/2} = 1.22 \pm 0.15$). From an analysis of the Doppler resolved spectral features, most of the excess energy resides in fragment translation ($\langle f_T \rangle = 0.29$) and internal excitation of the NO $_2$ fragment ($\langle f_{INT}^{NO_2} \rangle = 0.65$), the latter being very broadly distributed ($E_{INT} = 12590$ cm $^{-1}$, ΔE_{INT} (FWHM of assumed Gaussian distribution)

$= 6570$ cm $^{-1}$). The above energy partitioning is in general agreement with the predictions of an impulsive model in which the OH bond is assumed to be infinitely rigid. From such a model, however, one anticipates that the vectors $\vec{v}$ and $\vec{j}$ should be orthogonal ($\vec{v} \perp \vec{j}$), a conclusion which is at variance with the measurements summarised below.

(b) Vector Properties

The OH photofragment alignment, as measured from the relative integrated intensities of Q and P lines in geometries A and B, was

found to be small, positive and N'' independent ($\langle A_{0,e}^{(2)} \rangle N'' = 0.14 \pm 0.05$) indicating a preference for $\vec{j} \parallel \vec{\mu}$. The above intensity ratios also provide information on the relative Λ -doublet populations, which show a preference for the impaired electron density to lie perpendicular to the plane of OH rotation ($\pi(A'')/\pi(A') = 2$ for $N'' = 10$). Finally, from an analysis of the dependence of the Doppler resolved profiles on the nature of the probed transition and the pump-probe geometry (see Figure B1.16) the following information has been obtained.

- (i) β , the translational anisotropy, is small, positive and N'' dependent, varying from $\sim +0.3$ ($N'' = 1$) to ~ 0.8 ($N'' = 6, 8$ - the highest levels probed). This information reveals an increasing preference for $\vec{\mu}$ to lie parallel to $\vec{v}$ (and hence to the breaking N-OH bond) as N'' increases. The positive sign would be consistent with an A' excited state symmetry, if it were planar.
- (ii) $\beta_0^{(22)}$, the $v - j$ correlation, is positive and rises to a value of $+0.5$ for $N'' = 8$ (implying $\vec{v} \parallel \vec{j}$) whilst the triple vector correlation is small and negative. The interpretation of the former moment is that out-of-plane (i.e. torsional) rather than in-plane (bending or impulsive) forces, probably generated in the excited state, are the dominant source of OH rotation, at least at high N'' .

Excitations to the lowest lying singlet states of HONO $_2$ are anticipated⁽⁴⁾ to involve $n \rightarrow \pi^*$ ($A' \rightarrow A''$) transitions, primarily localized on the NO $_2$ moiety. These transitions, which are locally symmetry forbidden, are expected to be weak, as indeed is the first absorption continuum in HONO $_2$ ($\sigma = 1 \times 10^{-20}$ cm ^{2}). A more intense ($A' \rightarrow A'$) band is predicted to lie at much shorter wavelengths ($\lambda \sim 185$ nm), and it seems unlikely that this state is accessed in the present experiments ($\lambda = 280$ nm). An alternative explanation involves a vibronic transition to the above A'' state(s) mediated via the a'' out-of-plane ONO' bending motion. The A'' excited electronic state(s)

is expected to have a preferred pyramidal geometry, and activity of the a'' bending vibration would be reflected by a small, positive value of β , as found experimentally. The existence of torsional forces in the excited state may reflect the presence of π^* anti-bonding interactions between the N and OH groups which would be a maximum for planar or near planar HONO_2 molecules. In view of this possibility, it is perhaps not surprising that those OH fragments which have the largest degree of rotational excitations also possess the most positive values of β and the strongest preference for $\vec{v}$ to lie parallel to $\vec{j}$.

Acknowledgments

We gratefully acknowledge the help of the LSF in providing a YAG pumped tunable dye laser system on loan which was used in the initial stages of this research project.

References

1. J P Simons, J. Phys. Chem., **91**, 5378 and references therein, (1987).
2. R N Dixon, J. Chem. Phys., **85**, 1866, (1986).
3. J August, M Brouard, J P Simons, J. Chem. Soc. Farad. Trans. II in press.
4. L E Harris, J. Chem. Phys., **58**, 5615, (1973).

B1-11 PHOTODISSOCIATION DYNAMICS OF JET-COOLED METHYL AND t-BUTYL THIONITRITE

P A Giovanacci, M R S McCoustra and J Pfab, (Department of Chemistry, Heriot-Watt University, Edinburgh)

The photodissociation dynamics of molecules containing NO has recently become the focus of much excitement in laser photochemistry and spectroscopy⁽¹⁾. The ground state NO molecule ($^2\pi_1$) is an ideal

fragment for probing by the powerful laser-induced fluorescence (LIF) technique, and much microscopic detail of the dynamics and stereochemistry of the dissociation process can be inferred from fine-structure (spin-orbit and Λ -doublet), vibrational and rotational population distributions of this nascent fragment. In addition, vectorial properties (alignment and the correlation between recoil and angular momentum vectors) may be deduced from Doppler profiles and polarisation spectroscopy⁽²⁾.

Previous support by the LSF laser loan scheme has enabled us to perform state-of-the-art experiments on the state-selected predissociation of C-nitroso compounds in supersonic jets⁽³⁾ and much of this work is published or in print⁽⁴⁻⁷⁾. In the work described here, we have studied the direct dissociation of methyl and t-butylthionitrite in a free-jet expansion. The work forms part of an SERC supported project concerned with the laser photochemistry of polyatomic sulphur compounds.

Mixtures of between 5 and 10% by volume of the alkyl thionitrite vapour and helium are expanded from 500 torr through a commercial pulsed valve (Lasertechnics) with 0.5 mm orifice into high vacuum ($< 10^{-3}$ torr). The expansion is crossed 5-10 mm downstream of the orifice by the two collinearly aligned counter-propagating and focused dissociation (JK HyperYAG 750, 355 nm, 60 mJ per pulse) and probe (Lambda Physik EMG101MSC/FL3002, 10-15 mJ per pulse) lasers. Two-photon laser excited fluorescence of nascent NO was observed as described previously^(3,4). Population distributions were obtained by scanning the probe laser at an effective resolution of 0.16 cm^{-1} , and Doppler profiles could be recorded by insertion of an intracavity etalon, reducing the bandwidth to ca 0.04 cm^{-1} .

Rotational distributions of nascent NO ($v''=0$) and NO ($v''=1$) were obtained for the 355 nm photodissociation of both parent molecules. Figure B1.17 shows the NO ($v''=0$) distribution obtained from CH_3SNO .

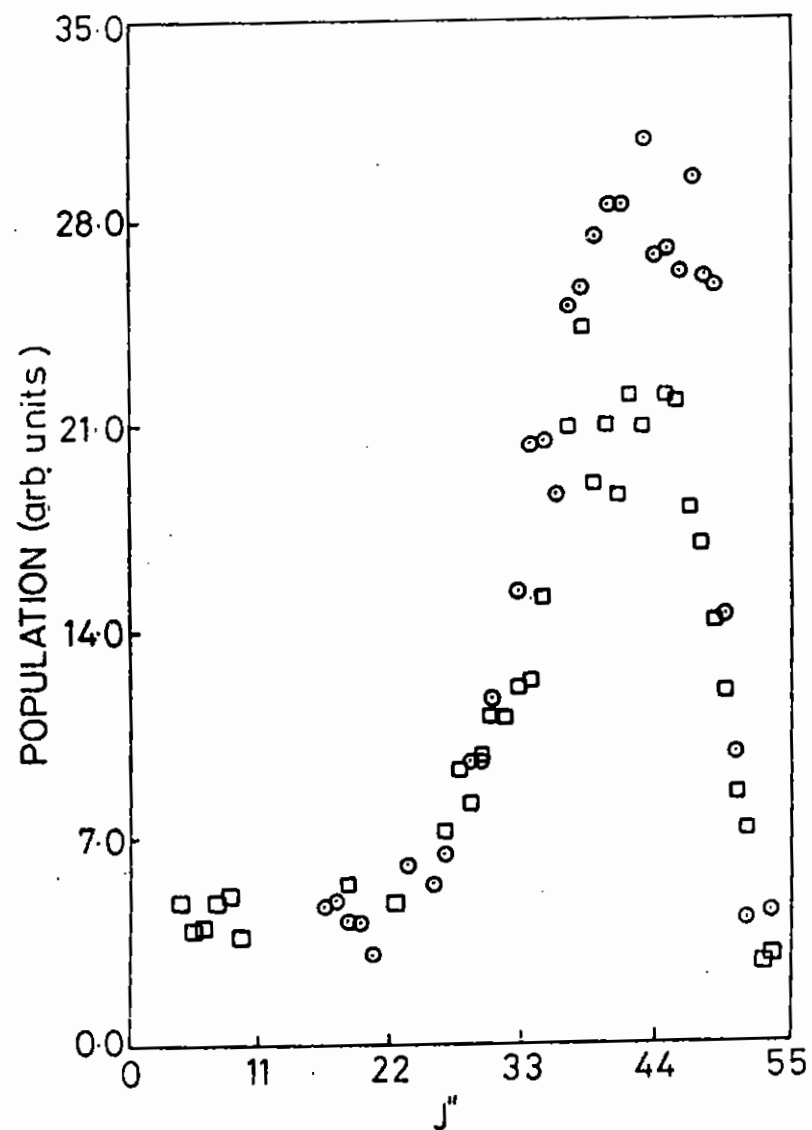


Fig. B1.17 Rotational population distributions of nascent NO ($^1\pi_{1/2}$, $^1\pi_{1/2}$ o) from the 355 nm photodissociation of CH_3SNO cooled by expansion with helium. The $J'' < 22.5$ part of the distribution arises from nascent, but translationally cold NO from a small fraction of complexed parent.

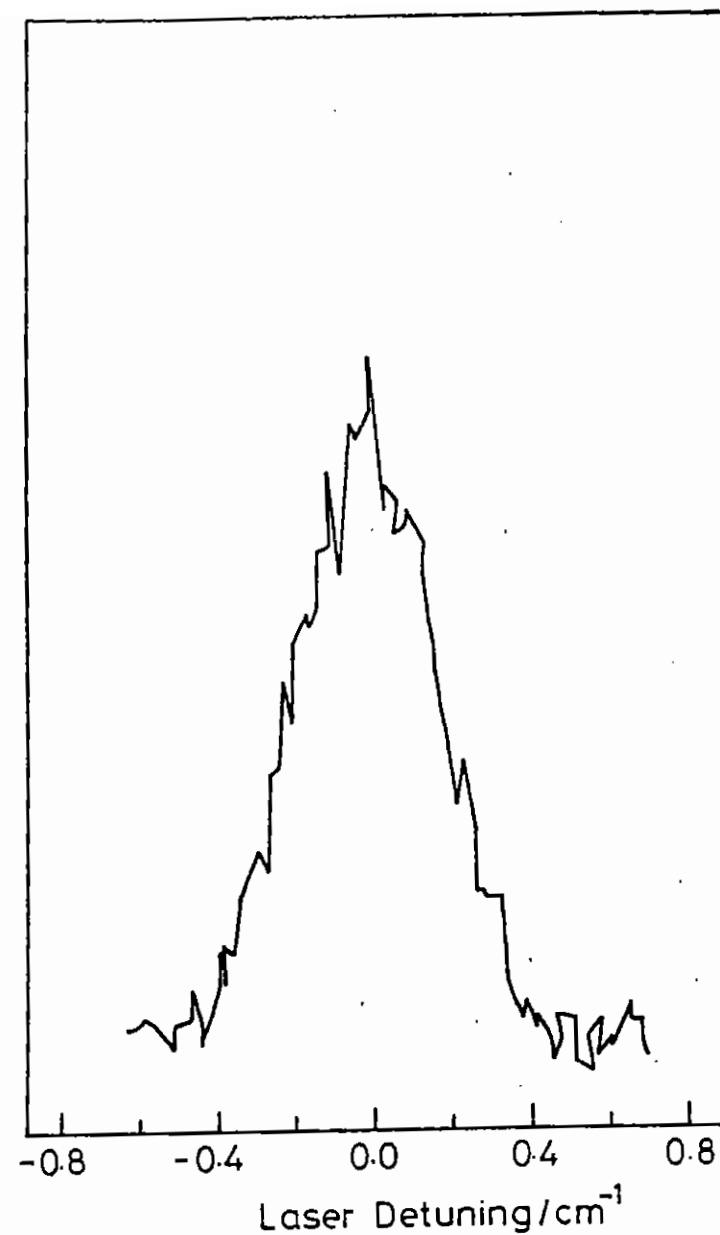


Fig. B1.18 Doppler profile of the $J''=57/2$ line in the O_1 branch of the $A^1\Sigma^+ \leftarrow X^1\pi_{3/2}$ (0,0) two-photon LIF excitation spectrum of nascent NO from the photodissociation of jet-cooled CH_3SNO .

The $J'' < 22$ part of the distribution is affected by artefacts. The NO is rotationally very hot, and the spin-orbit states of NO are not equilibrated. Analysis of the Λ -doublet populations revealed that the anti-symmetric π^- component is preferred, indicating that the dissociation is a planar process, and that the near-UV continuum of the thioalkyl nitrites is due to a transition of the same symmetry ($^1A'' \leftarrow ^1A'$ or n, π^*) as the vibrationally structured near-UV transition of N-nitrosamines and O-alkyl nitrites⁽¹⁾.

Figure B1.18 shows a sub-Doppler resolution scan of the $J'' = 57/2$ two-photon LIF line of the $O_{1,2}$ branch. Such profiles have been evaluated for several transitions and provide a linewidth of $\Delta \bar{\nu}_D = 0.5 \text{ cm}^{-1}$, from which the translational energy of the NO fragment could be calculated. A summary of the overall energy disposal for the methyl compound is collected in Table 1.

Clearly, most of the available energy in the 355 nm dissociation of alkyl thionitrites is channeled into relative translation. A substantial fraction of the available energy appears in the rotation of NO, whilst the internal energy of the CH_3S radical and the vibration of the NO fragment remain cold.

Photodissociation of $(\text{CH}_3)_3\text{CSNO}$ in a continuous jet on the S_1 transition near 600 nm was briefly examined at the beginning of the loan period. Nascent NO could be identified successfully by single-photon LIF near 226 nm, but problems with our doubling crystal forced us to change to the 355 nm dissociation, the use of the less sensitive two-photon LIF probe technique and photodissociation close to the nozzle orifice. Our use of the JK HyperYAG loan laser was severely limited by recurring problems with the firing of the Q-switch. More refined experiments are planned to study the dynamics of the S_2 and S_1 photodissociation of thionitrites in detail.

TABLE 1

Fragment	Degree of Freedom	Energy/ cm^{-1}	%/ E_{avl}
NO	Electronic	57	0.35
	Vibrational	<202	<1.25
	Rotational	2375	15
	Translational	7800	48
MeS	Internal	>725	>4.5
	Translational	4980	31

References

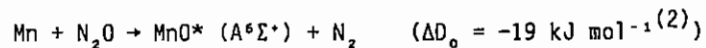
1. H Reisler, M Noble and C Wittig, *Molecular Photodissociation Dynamics*, edited by M N R Ashfold and J E Baggott, Royal Society of Chemistry, London, 139 (1988).
2. M P Docker, A Hodgson and J P Simons, *Molecular Photodissociation Dynamics*, edited by M N R Ashfold and J E Baggott, Royal Society of Chemistry, London, 115 (1988).
3. J A Dyet, M R S McCoustra and J Pfab, *Annual Report to the Laser Facility Committee B1.19*, 159 (1987).
4. M R S McCoustra, J A Dyet and J Pfab, *Chem Phys Lett* 136, 231 (1987).
5. J A Dyet, M R S McCoustra and J Pfab, *Chem Phys Lett* 135, 534 and 137, 398 (1987).
6. J A Dyet, M R S McCoustra and J Pfab, *J Chem Soc Faraday Trans II* (in print).
7. M R S McCoustra, J A Dyet and J Pfab, *Laser Chemistry*, submitted.

B1-12 CHEMILUMINESCENCE AND ENERGY TRANSFER IN Mn + N₂O COLLISIONS AT HIGH TRANSLATIONAL ENERGY

M R Levy (Dept. of Chemical and Life Sciences, Newcastle-upon-Tyne Polytechnic)

1. Introduction

In a previous report⁽¹⁾, it was shown that pulsed laser ablation could be used to generate beams of refractory metal atoms, and thereby to study their chemiluminescent reactions with oxygen-containing molecules. The technique has now been refined, allowing detailed measurements on the chemiluminescence excitation function of the reaction



This represents a suitable place to start on studies of transition metal atom reaction dynamics as the complexity is at a minimum; the Mn electronic structure (Figure B1.19) is relatively simple⁽³⁾; and the MnO (A⁶Σ⁺ - x⁶Σ⁺) spectrum from ~450 to ~750 nm is well-known, with regular vibrational structure⁽⁴⁾.

Experimental

The beam-gas configuration described in the previous report⁽¹⁾ has again been employed. The pulsed metal atom beam was generated by ablation of a solid metal target, using the JK2000 Nd:YAG laser at 1.06 μm; and monitored, at a point 283 mm along the beam axis, by means of the long-lived emission from the Mn*(⁶P_j) states. Emission from the reaction with N₂O was detected at the same point. Data, which were digitised and averaged by the Biomation/Tracor Northern system, were time-of-flight spectra of the beam number density, and the corresponding time-dependent emissions produced when N₂O was admitted to the scattering chamber.

Results

Figure B1.20 shows a typical time profile of the Mn*(⁶P_j) emission at ~540 nm, P(t), together with the derived atomic flux distribution, N'(t). The pulsed beam velocity ranges from <20000 to >2000 ms⁻¹, the precise form depending on the laser focusing. These velocities are so high that, for reaction with N₂O, the metal atom beam velocity determines the mean collision energy; and, for the bulk of the velocity range, the N₂O velocity is negligible. The shape of N'(t) given in Figure B1.20 corresponds to a Maxwell-Boltzmann distribution with temperature ~77000 K, and therefore it is not surprising that the beam contains metastable states, many others apart from ⁶P_j are expected.

Emission from MnO* (A⁶Σ⁺) is observed when N₂O is admitted to the scattering chamber. The pressure dependence is linear up to ~0.020 Pa (~1.5 x 10⁻⁴ torr), indicating first-order kinetics. A low resolution scan of the spectrum suggests that several vibrational levels are populated. Figure B1.21 shows a typical time profile, I(t), of the emission (λ > 550 nm); the peak signal, at ~50 μs, occurs well after the peak in the beam flux. The maximum time at which MnO* emission occurs cannot be precisely determined, because of the noise on the data. However, it does appear to be over by ~115 μs, while, in the analogous Mn + NO₂ reaction⁽⁵⁾, emission is detected beyond 180 μs.

Emission is also observed from Mn*(⁶P_j) at ~403 nm, and a typical time profile is likewise shown in Figure B1.21. Again, the maximum time cannot be determined precisely, but it is ~58 μs. The fall-off in MnO* signal, as t decreases, appears to be matched by the rise in Mn*(⁶P_j) production.

Since both N'(t) and I(t) are flux measurements, they have the same Jacobian factor for transformation to translational energy, E_T. The emission excitation functions, σ(E_T) (in arbitrary units), can therefore be obtained directly by dividing I(t) by N'(t). These results are shown in Figure B1.22, confirming the fall-off in MnO* production as Mn* appears. The arrows on the E-axis indicate the thresholds for the two processes^(3,4) if Mn(⁶S) is the reactant atom.

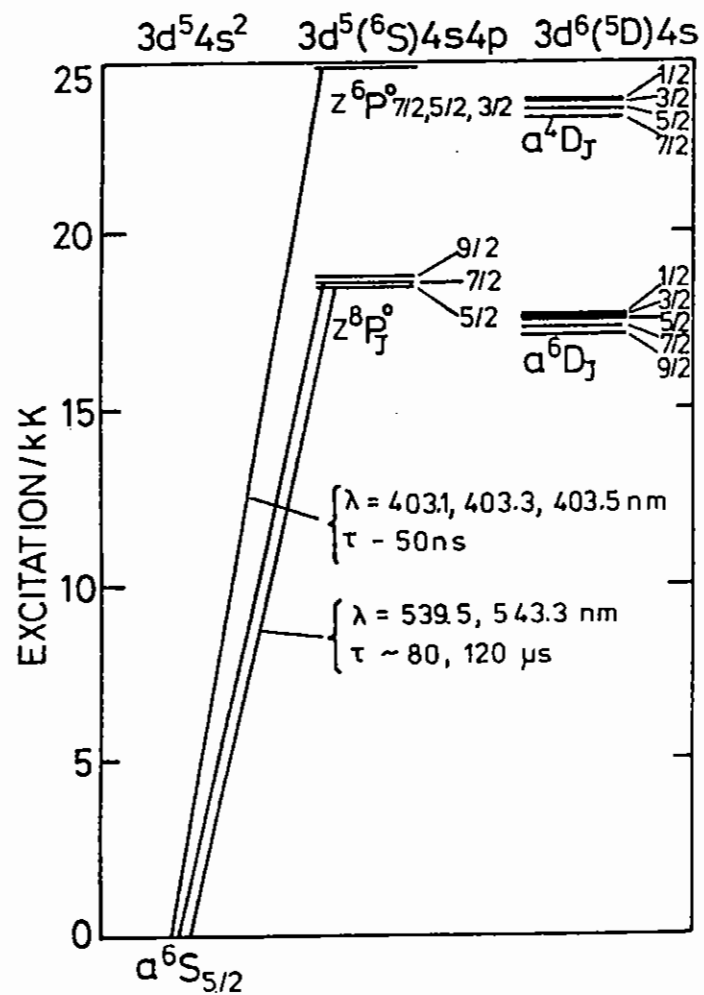


Fig. B1.19 The low lying states of Mn.

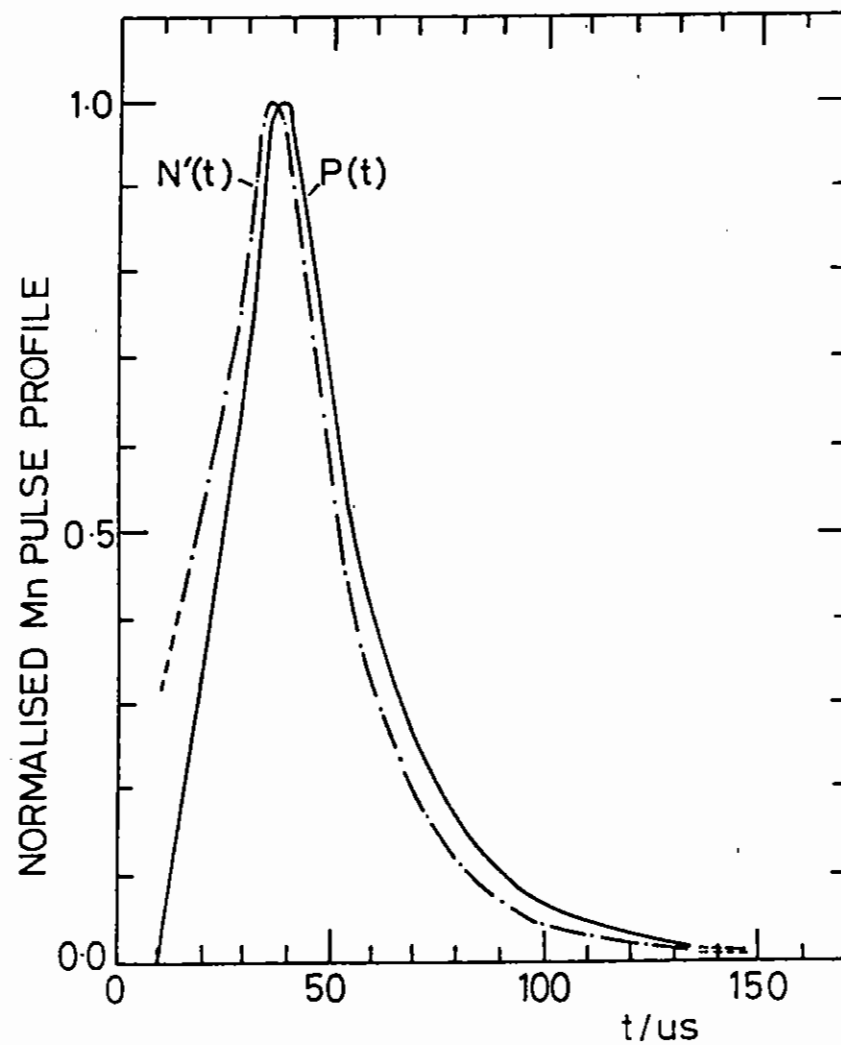


Fig. B1.20 Typical time-profiles of $\text{Mn}^*({}^6P_j)$ emission, $P(t)$, and the corresponding beam flux, $N'(t)$.

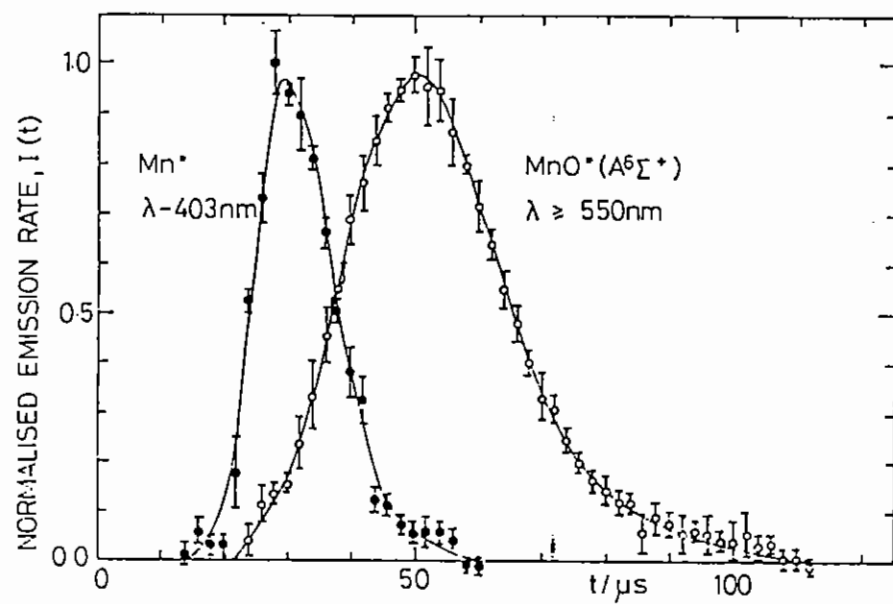


Fig. B1.21 Emission time-profiles from $\text{Mn} + \text{N}_2\text{O} \rightarrow \text{MnO}^*(\text{A}), \text{Mn}^*(^6\text{P}_j)$.

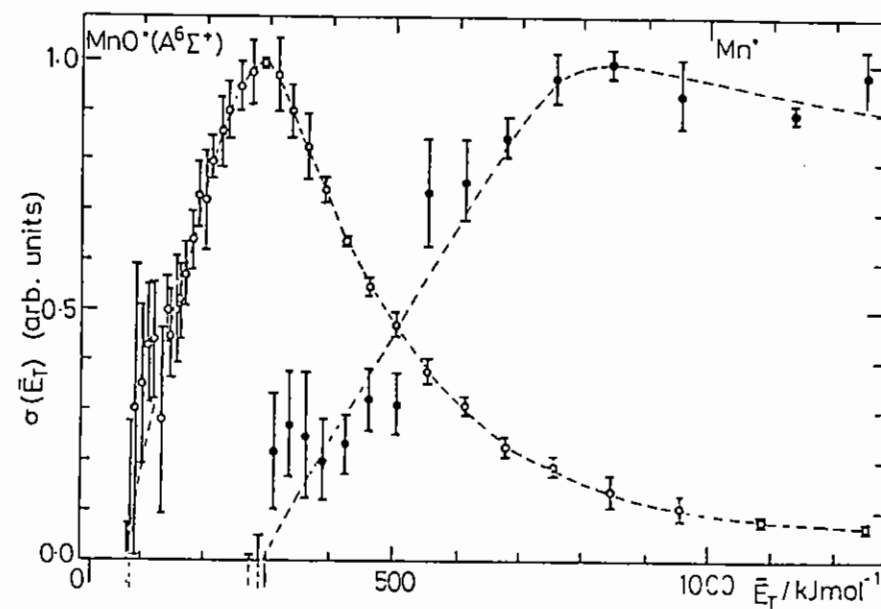


Fig. B1.22 Excitation functions derived from the data of Fig B1.21.

Discussion and Conclusions

Although ^6D and ^6P atoms, at least, are undoubtedly present in the Mn beam, Figure B1.22 suggests that the dominant contribution to both chemiluminescent reaction and $\text{Mn}^*(^6\text{P})$ excitation is from ground-state atoms. $\text{Mn}^*(^6\text{P})$ appears exactly at the energetic threshold for $\text{Mn}(^6\text{S}_0)$ precursors; and the concomitant removal of >90% of the $\text{MnO}^*(\text{A})$ emission as $\text{Mn}^*(^6\text{P})$ increases to its maximum indicates that a single precursor is responsible for both. In fact, the barrier for $^6\text{S}_0$ atom reaction may be due to crossing with the $^6\text{P}_j$ surface; a similar mechanism has been proposed to explain the high threshold energy for $\text{Mg}(^1\text{S}_0) + \text{N}_2\text{O}^{(6)}$. The lack of any "break" in the MnO^* excitation function, at the $\text{Mn } ^6\text{S}_0$ - $^6\text{D}_j$ thermodynamic threshold, indicates that ^6D atoms have a completely repulsive interaction with N_2O , presumably because the d^6s^1 configuration does not facilitate bonding overlap.

This situation contrasts with $\text{Mn} + \text{O}_2$, CO_2 , $\text{NO}_2^{(5)}$, where both $\text{MnO}^*(\text{A})$ and $\text{Mn}^*(^6\text{P}_j)$ derive, at least in part, from excited ^6D atoms.

Acknowledgements

Equipment loans from Professor I R Beattie (Southampton University), Professor R Hill (Newcastle Polytechnic) and Dr E Lewis (Newcastle University) are gratefully acknowledged.

References

1. M R Levy, Paper B.1.15, Laser Facility Committee Report 1987.
2. K Huber and G Herzberg, Constants of Diatomic Molecules (Van Nostrand, New York, 1979).
3. J Sugar and C Corliss, J Phys Chem Ref Data **14**, Suppl. 2 (1985) p 338 ff;
S M Younger, J R Fuhr, G A Martin and W L Wiese, J Phys Chem Ref Data **7** (1978) p 591 ff.
4. J M Das Sarma, Z Physik **145** (1959) 98; K C Joshi, Spectrochim Acta **18** (1962) 625.

5. M R Levy, to be published.

6. D R Yarkony, J Chem Phys **78** 6763 (1983).

B1-13 ROTATIONAL STATE DEPENDENCE OF ELECTRONIC ENERGY TRANSFER IN Na_2 .

S C Taylor, M J Wynn and B J Whitaker (University of Sussex)

Introduction

Until recently very few precise data were available on collisionally induced electronic transfer at the state-to-state level. Particularly noteworthy, however, are the rotationally resolved studies of Dagdigian and co-workers on transfer between the $^2\Pi$ and $^2\Sigma$ surfaces of $\text{CN}^{(1)}$ and of Katayama on the surface crossing between the same states of the N_2 positive ion⁽²⁾. In the alkali diatomics energy transfer between electronic surfaces is also known to occur from the observation of A - X band fluorescence in both Li_2 and Na_2 following excitation into the $\text{B}^1\Pi_u$ state. The emission is thought to be the result of energy transfer from the B state to the $(2)^1\Sigma_g^+$ state followed by radiative transfer to the $\text{A}^1\Sigma_u^+$ state (Figure B1.23). This model is supported by the work of Effantin et al. using fourier transform infra-red spectroscopy in which they observe the $(2)^1\Sigma_g^+ - \text{A}^1\Sigma_u^+$ fluorescence.

It is important to stress that the energy transfer in Na_2 cannot be the result of non-adiabatic coupling (e.g. spin-orbit, spin-rotation) between the two surfaces since the two electronic states are of different inversion symmetry. The favoured mechanism rests on the realisation that the approach of the perturber will lift the cylindrical symmetry of the molecule, inducing "optical-like" dipolar transitions between the rovibrational levels of the two surfaces through electrostatic coupling. This model may be critically tested against experiment if one can uncover the rovibrational propensity (selection) rules underlying the state-to-state transfer.

Previous work^(3,4,5,6) has relied upon the initial excitation being provided by the frequency coincidence of the Ar⁺ laser emission lines with the Na₂ absorption spectrum. Since each of the laser lines excite several levels in the B¹Π_u state it is difficult to accurately identify any systematic trends in the B¹Π_u → (2)¹Σ_g⁺ transfer cross-sections.

Experimental and Results

The proposed research program detailed the possible two colour experiment out-lined in Figure B1.23. The aim was to selectively excite a single rovibrational level in the B¹Π_u state and then to probe the (2)¹Σ_g⁺ state with a second tunable laser by excitation to the (3s,5s)¹Σ_u⁺ surface. The rovibrational population distribution of the (2)¹Σ_g⁺ state would be obtained by monitoring the UV emission ((3s,5s)¹Σ_u⁺ - X¹Σ_g⁺) as a function of probe laser frequency.

Initially it was necessary to locate some possible resonant levels in the B state. An earlier study, using a single mode Ar⁺ laser⁽⁶⁾, allowed us to quantify the relative rates of transfer out of several individual rovibrational levels in the B¹Π_u state by making simultaneous measurement of the relative quantum yields of the two bands (A¹Σ_u⁺ - X¹Σ_g⁺ and B¹Π_u - X¹Σ_g⁺). However it was impossible to unravel any general trends from such a limited data set, and so a systematic study was undertaken using the tunable output from an excimer pumped dye laser (EMG101/FL2002) to excite the B¹Π_u levels.

The experimental arrangement is shown in Figure B1.24. The output from the excimer pumped dye laser (using C102 dye solution) was focussed into the sodium vapour contained in a stainless steel vessel. The cell was based upon a commercially available six-way vacuum cross with one arm modified to accept two 600W cartridge heaters (Hedin model CP 3229) in a bored stainless steel block. A chromel/alumel thermocouple provided a means of temperature measurement and was also utilised to drive a thermostatic heater controller. This maintained the cell temperature at typically 750 K (with 5 torr of He buffer gas). At this temperature the dimer is present in about 1% abundance.

The fluorescence from the excited Na₂ molecules was collected perpendicularly to the direction of laser propagation by a cooled photomultiplier tube (EMI 9658B; S20 response). The signal was amplified using a fast amplifier (Elscint TFA-N-1) and then passed into a gated integrator (Stanford Research Systems SR250) to be subsequently output to a chart recorder. Figure 3 illustrates the time dependence of the signal, as recorded by a digital storage oscilloscope (LeCroy 9400). The early peak is due to B¹Π_u - X¹Σ_g⁺ emission and laser pulse scatter, while the delayed component is due to A¹Σ_u⁺ - X¹Σ_g⁺ band emission. We confirmed the assignment of the delayed signal by dispersing the fluorescence in a 0.25 m grating monochromator (Spex minimate) and amplifying this component using the gated integrator. In the figure the red A¹Σ_u⁺ - X¹Σ_g⁺ band emission has been enhanced in comparison to the blue laser scatter and green B¹Π_u - X¹Σ_g⁺ emission by the use of a suitable edge filter.

Attempts have been made to model the evolution of the A¹Σ_u⁺ - X¹Σ_g⁺ signal using simple rate equations, these yield a (2)¹Σ_g⁺ lifetime of around 190ns, which is consistent with an LCAO calculation of the transition moment. However, they cannot explain the time delayed nature of this signal, and whilst we have been unable to calibrate the equipment's rise time through the lack of a suitably fast detector, we must conclude that this is an artifact of the detection chain.

By suitable selection of the time delay of the integration gate we were able to observe solely the A¹Σ_u⁺ - X¹Σ_g⁺ emission resulting from the curve crossing (B¹Π_u → (2)¹Σ_g⁺) and subsequent relaxation ((2)¹Σ_g⁺ - A¹Σ_u⁺). We were able to scan the dye laser over a total of five vibrational bands in the B state using C102, and utilised the intracavity etalon in the FL2002 dye laser to study at high resolution the A¹Σ_u⁺ - X¹Σ_g⁺ emission resulting primarily from excitation of the v=2 band of the B state. A section of the spectrum is shown in Figure B1.26. This spectrum represents an X¹Σ_g⁺ - B¹Π_u excitation spectrum "modulated" by the B¹Π_u → (2)¹Σ_g⁺ transfer probability. For comparison a simulation of the X¹Σ_g⁺ - B¹Π_u LIE spectrum is shown in Figure B1.27. It is clear that there are significant differences between the two

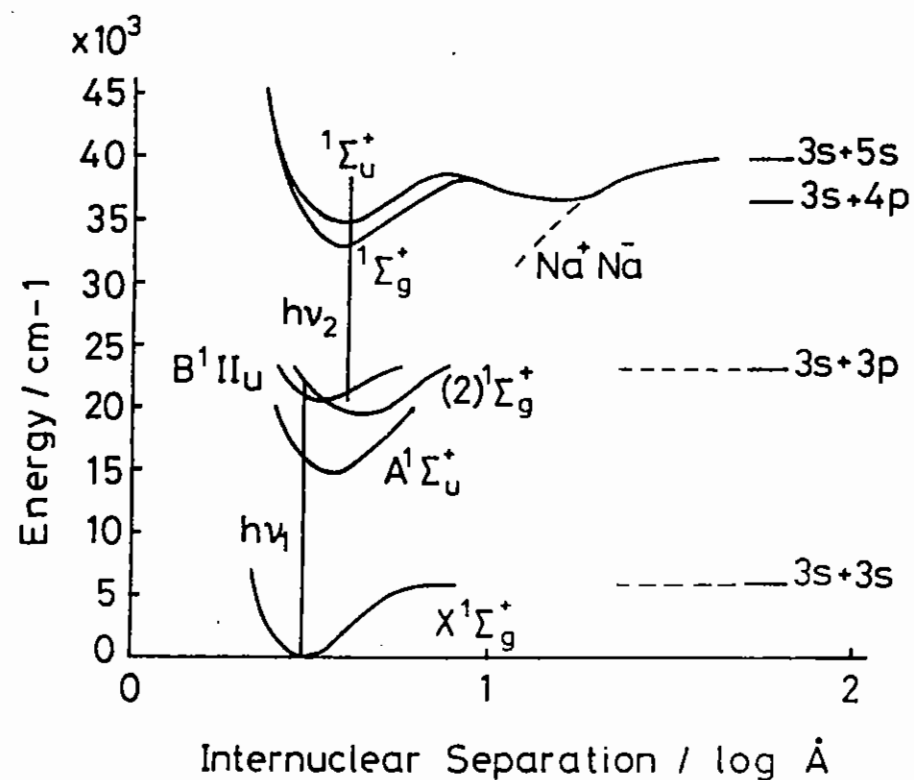


Fig. B1.23 Adiabatic potential energy surfaces of Na_2 of relevance to this work.

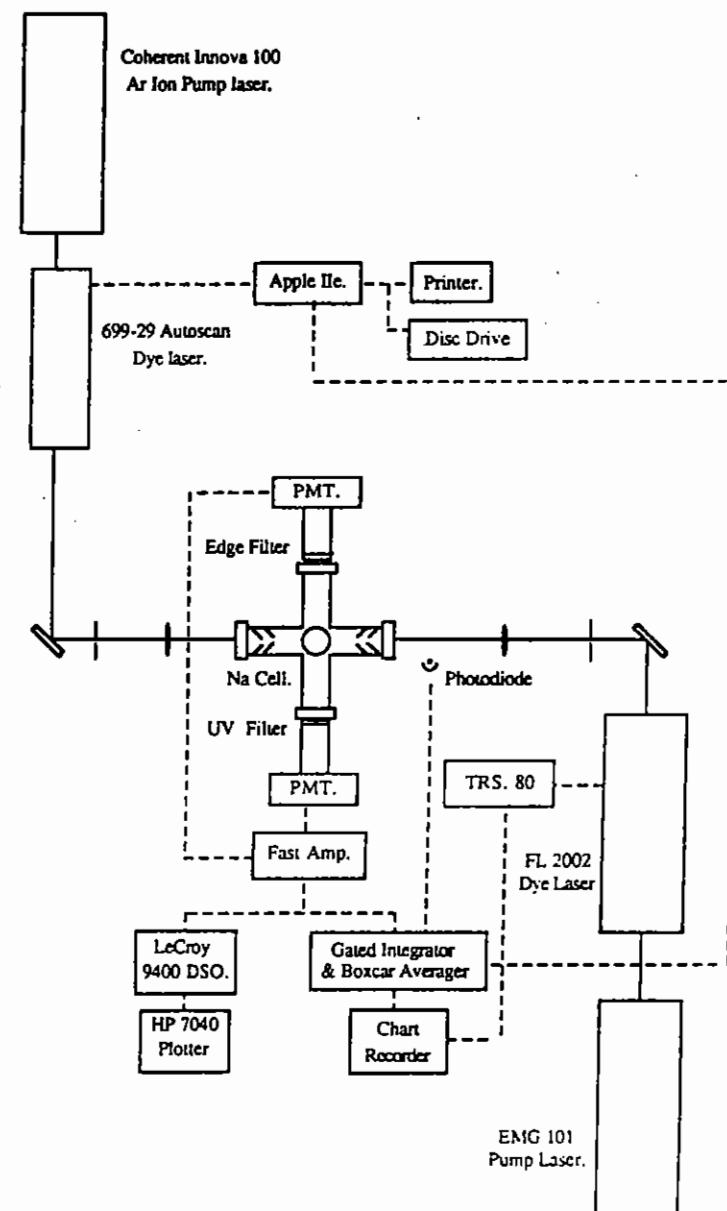


Fig. B1.24 Experimental arrangement for OODR study of state-to-state electronic energy transfer, and for $A^1\Sigma_u^+ - X^1\Sigma_g^+$ emission study.

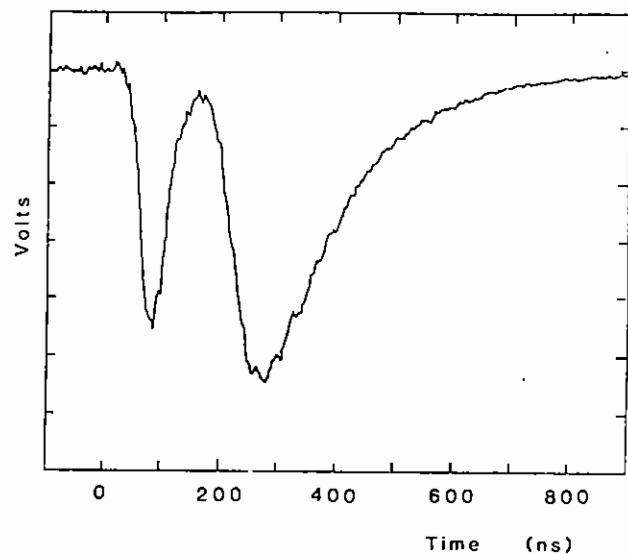


Fig. B1.25 Time dependence of the visible fluorescence from Na_2 following excitation into the $B^1\Pi_u$ state.

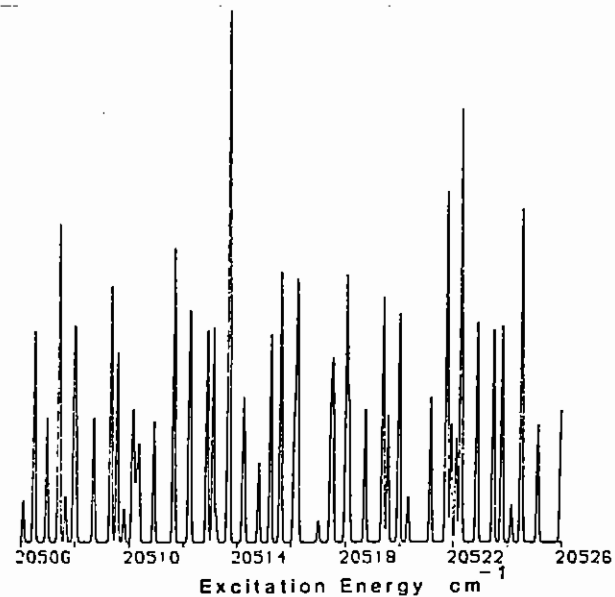


Fig. B1.27 Simulation of the $X^1\Sigma_g^+ - B^1\Pi_u$ excitation spectrum.

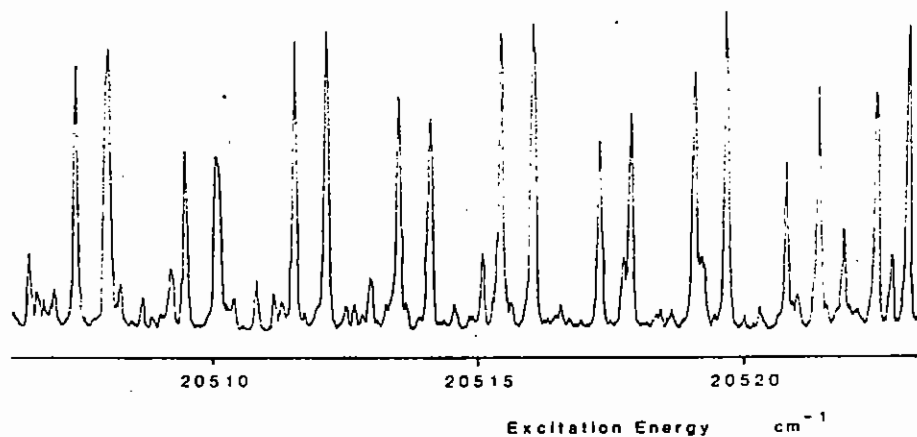


Fig. B1.26 $X^1\Sigma_g^+ - B^1\Pi_u$ excitation spectrum observed in the $A^1\Sigma_u^+ - X^1\Sigma_g^+$ band.

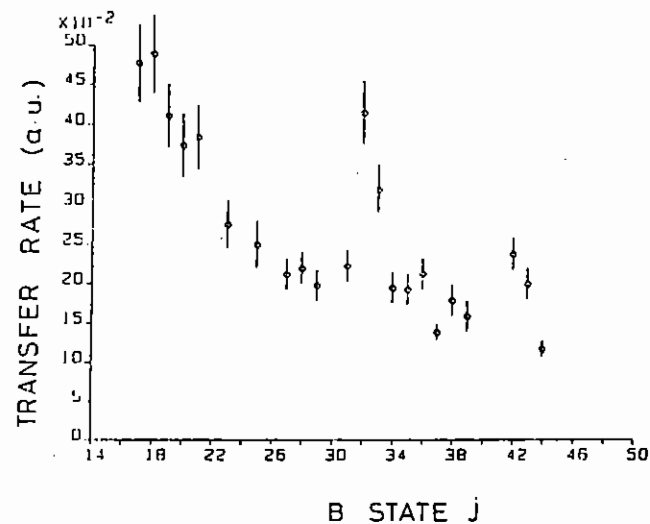


Fig. B1.28 Relative transfer rates out of the $B^1\Pi_u$ $v=2$ rotational manifold as a function of J .

spectra which reflect the rovibrational state dependence of transfer out of the B state. Figure B1.28 shows the relative transfer rates out of the $B^1\Pi_u/v=2$ rotational manifold plotted against J. Clear resonances are apparent around $J=32$ and $J=42$.

In the proposal it was suggested that one could differentiate between the excitation mechanisms $X^1\Sigma_g^- - B^1\Pi_u - (3s,5s)^1\Sigma_g^-$ and $X^1\Sigma_g^- - B^1\Pi_u \rightarrow (2)^1\Sigma_g^- - (3s,5s)^1\Sigma_g^-$ by the time dependence of the resulting UV emission. The lifetime of the $B^1\Pi_u$ state is ~ 8 ns, much shorter than the radiative lifetime of the $(2)^1\Sigma_g^-$ state. The probe beam in this experiment is provided by a cw ring dye laser (Figure B1.24). An unforeseen experimental problem was that the pump pulse caused significant atomic emission ($4p - 4s$), presumably via two photon dissociation, which interfered with the double resonance signal. In principle the time gating would overcome this problem were it not for the fact that the atomic emission is strongly radiatively trapped. This forced us to work with a highly attenuated excitation pulse. Under these conditions we eventually observed an OODR signal with the correct time delay. Sadly however we were unable to accumulate any reliable state-to-state data before the end of the loan period.

Conclusion

We had proposed a pilot experiment to obtain rotationally resolved state-to-state cross-sections for electronic energy transfer between two of the low lying surfaces of Na_2 , and we have succeeded in showing the feasibility of such a study. The interpretation of previous observations of A-X band emission following excitation of the B state as being due to $B^1\Pi_u \rightarrow (2)^1\Sigma_g^-$ energy transfer followed by radiative relaxation to the A state have been confirmed from the time dependence of the emission, and we have obtained a value for the radiative lifetime of the $(2)^1\Sigma_g^-$ state. We have also obtained data on the rotational state dependence of the transfer rates out of the $B^1\Pi_u$ state.

Acknowledgements

We thank the SERC for two research studentships (SCT and MJW) and an Advanced Research Fellowship (BJW). We are particularly grateful to the LSF and to Prof. A J McCaffery (Sussex) for their contribution to this work.

References

1. A Ali, Guo Jihua and P J Dagdigian, J. Chem. Phys. **87**, 2045 (1987).
2. D H Katayama, J. Chem. Phys. **54**, 657 (1985).
3. A G Astill, A J McCaffery and B J Whitaker, Chem. Phys. Lett. **125** 33 (1986).
4. C Effantin, O Babaky, K Hussein, J d'Incan and R F Barrow, J. Phys. B:At. Mol. Phys. **18**, 4077 (1985).
5. A G Astill, A J McCaffery and B J Whitaker, Chem. Phys. Lett. **142**, 1 (1987).
6. A G Astill, A J McCaffery, S C Taylor, B J Whitaker and M J Wynn, J. Chem. Phys. (in press).

B1-14 THE LASER PHOTOACOUSTIC SPECTRA OF OZONE AND OXYGEN IN THE VISIBLE REGION.

P M Borrell, P Borrell and D C Lainé (Depts. of Chemistry and Physics, University of Keele)

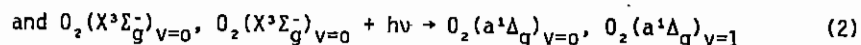
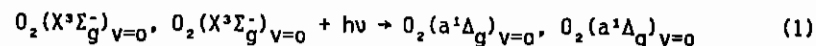
Summary

An apparatus has been developed to study the photoacoustic spectroscopy of gases in the visible region of the spectrum. Results are presented for ozone, which was used as a test gas and oxygen for which two dimol transitions were observed. Prospects for future measurements and low temperature work are discussed.

Introduction

A glance at a light across a room is sufficient to indicate that the absorption of oxygen in the visible region of the spectrum is very weak. Nevertheless in the red spectral region there are two types of transitions which lead to absorption; the weaker members of the strongly forbidden 'atmospheric bands' $O_2(b^1\Sigma_g^- \leftarrow X^3\Sigma_g^-)$, and several bands

of the cooperative 'dimol' transition which involve, in absorption, the simultaneous excitation of two molecules by a single quantum of radiation; two of these are;



They are responsible for the blue colour of liquid oxygen⁽¹⁾ but are very weak in the gas phase and have only been studied previously at elevated pressures and very long pathlengths. The spectrum of each band is a broad peak⁽²⁻⁴⁾ with a slight asymmetry which is characteristic of a collision induced transition. McKellar, Rich and Welsh⁽⁴⁾ studied the temperature dependence of the absorption coefficient and found that below room temperature it was constant down to ~150 K, below which it began to increase.

Our own interest in these transitions stems from studies of the emissions, which were first identified by Arnold, Ogryzlo and Witzke⁽⁵⁾ in flows of singlet molecular oxygen. We used the emission to measure the concentration of $O_2(a^1\Delta_g)$ at high temperatures in our discharge flow/shock tube and measured the change in emissivity which increases at higher temperatures^(6,7). Also we were able to recommend a value for the rate constant for the emission by comparing previous results from emission and absorption⁽⁸⁾. Recently we studied the emissivity at lower temperatures⁽⁹⁾ and found that, within experimental error, it remained constant down to 100 K.

Thus there is a discrepancy in the behaviour at low temperatures between the emission and absorption measurements. In view of importance of oxygen, the possibility of $O_2(a^1\Delta_g)$ providing a low temperature laser system and the difficulties experienced in both previous emission and absorption work, it is worthwhile to try to remeasure the temperature dependence of absorption and emission coefficients. Photoacoustic spectroscopy offers a potential method for measuring very weak absorption in the gas phase⁽¹⁰⁾ and the exploration

of oxygen by photoacoustic spectroscopy is important in itself since oxygen would necessarily interfere in the photoacoustic analysis of, say, true impurities in air if the absorption is appreciable.

Thus we have built an initial apparatus, tested it with ozone and measured the absorption of oxygen at room temperature.

Experimental

The apparatus is shown in Figure B1.29; it is of the conventional pattern⁽¹⁰⁾ in which a beam of light is pulsed with a rotating chopper and passed into a photoacoustic cell where the pulsed energy released into the gas by absorption is detected as sound.

The argon-ion laser (Spectra Physics 2025) is operated at 4W and pumps a dye laser (Spectra Physics 375) which had a maximum output of 400 mW. A synchronous motor with a gearbox was connected to the wavelength control to enable spectroscopic scans of variable scan rate to be made so that with Rhodamine 6-G dye, the laser could be tuned to wavelengths between 565 and 635 nm. The rotating chopper is a Brookdeal 9479 model operated at 509 Hz, chosen for its low phase jitter.

The photoacoustic cell was built to similar specifications as that of Bicanic⁽¹¹⁾. It consists of an internal 'organ-pipe' resonance tube, made of polished stainless steel, 20 cm long and 6 mm inside diameter with a 1.5 mm wall. Four electret microphones (Knowles BT 1754) are situated around the centre and sealed into close-fitting holes with silicone elastic compound. The resonant tube sits inside an outer brass jacket, 5 cm in diameter and 35 cm long, equipped with internal supporting rings of brass and teflon which also serve as noise baffles. The outer tube is terminated with brass screw caps each having a 1 cm central aperture from which protrudes an 'O' ring sealed glass tube terminated in a 1 cm glass window set at the Brewster angle.

The signals from the amplifying microphones are taken in parallel and fed to a lock-in amplifier (E G and G Princeton 501); the reference frequency is provided directly from the chopper.

The output is fed to a chart recorder and also to a precision analogue multiplier (Analog Devices AD 534 JH) connected as an A/B divider. The

denominator is provided by the signal power monitor (Photon Control, Model 11) and so a signal, normalised for power fluctuations, is sent to recorder together with the power signal.

Oxygen was used at 1 atmosphere (typically 740–750 torr pressure). Ozone was produced by passing O_2 slowly through a high frequency discharge. The concentration was estimated by passing the flow of O_2 containing O_3 into an acidified solution of potassium iodide for a known period and measuring the iodine formed by spectrophotometry at 400 and 440 nm.

The experimental conditions, obtained after much effort and development to reduce noise, were; power into the cell 200 mW; noise 0.1 μV ; prefilter 10 or 30s; postfilter, 1s; scan time 60 mins between 565 and 620 nm.

The Photoacoustic Spectrum of Ozone

Ozone, which has a known weak absorption in the visible region, was used to test the apparatus. It was passed in a flow of O_2 through the cell for between 10 and 30 minutes before the valves were shut and the spectrum taken in the stationary gas. The ozone lasted in the cell for several hours without appreciable loss and with no effect on the microphones or wiring.

A photoacoustic spectrum is shown in Figure B1.30. The central part of the Chappuis band is shown^(12,13) with the peaks at 574 and 602 nm and the trough at ~ 585 nm. The broad wings of the absorption were not accessible because of the limited range of the laser dye used.

An indication of the sensitivity of the apparatus can be obtained from the spectrum. The height of the peak was 1 mV on the amplifier and this corresponded to an ozone partial pressure of $\sim 1.00 \times 10^{-3}$ atmospheres. The noise for zero signal was 0.1 μV so the ultimate sensitivity of the present apparatus is about 2×10^{-7} atmospheres of O_3 .

This can be compared with the results of Bicanic⁽¹¹⁾ who report that, with a CO_2 laser in the infrared, O_3 can be estimated at the sub-parts

per billion level. For the 9P(14) transition of the CO_2 laser, O_3 has an absorption cross section⁽¹¹⁾ of $12.00 \text{ atm}^{-1} \text{ cm}^{-1}$, while at 578 nm in the visible, the cross section⁽¹³⁾ is only $0.13 \text{ atm}^{-1} \text{ cm}^{-1}$. Also the CO_2 laser used had an output of ~ 6 W whereas that of our dye laser is ~ 400 mW. Thus the sensitivity of our newly-built apparatus appears to be at least as good as that of the experienced Wageningen and Nijmegen groups.

The Photoacoustic Spectrum of Oxygen

The absorption of oxygen was expected to be much weaker than that of ozone and this is indeed proved to be true. However, after much attention to noise reduction, weak signals were obtained (Figure B1.31) in the region of both 579 nm and 630 nm corresponding to the two transitions (1) and (2). The peak shapes are similar to those obtained in long-pathlength absorption experiments and we have no doubt that the assignments are correct.

The signals themselves are quite strong considering that neither is at the wavelength of maximum power for Rhodamine 6-G. McKellar, Rich and Welsh⁽⁴⁾ give the absorption coefficient, A , at 579 nm for transition (2), to be 4×10^{-11} Amagat⁽²⁾ where A is defined by

$$A = (\ell/p^2/\bar{\nu}) \ln(I_0/I) \quad (3)$$

where p is the density of the gas (in Amagat), ℓ is the path length and $\bar{\nu}$ the wavenumber of the peak. For our conditions, (292 K, 740 torr, 20 cm path length), the expected absorption $(1-I/I_0)$ is 1.15×10^{-6} . For O_3 , mentioned above, with a 20 cm path length, the 1 mV peak which corresponded to a pressure of $\sim 1.00 \times 10^{-3}$ atm. gives an absorption of $\sim 3 \times 10^{-3}$. Thus the expected peak height corresponding to the O_2 absorption of 1×10^{-6} is $\sim 0.3 \mu V$ which is in good agreement with the value actually found.

Future work

The present results, obtained during a single loan period of the

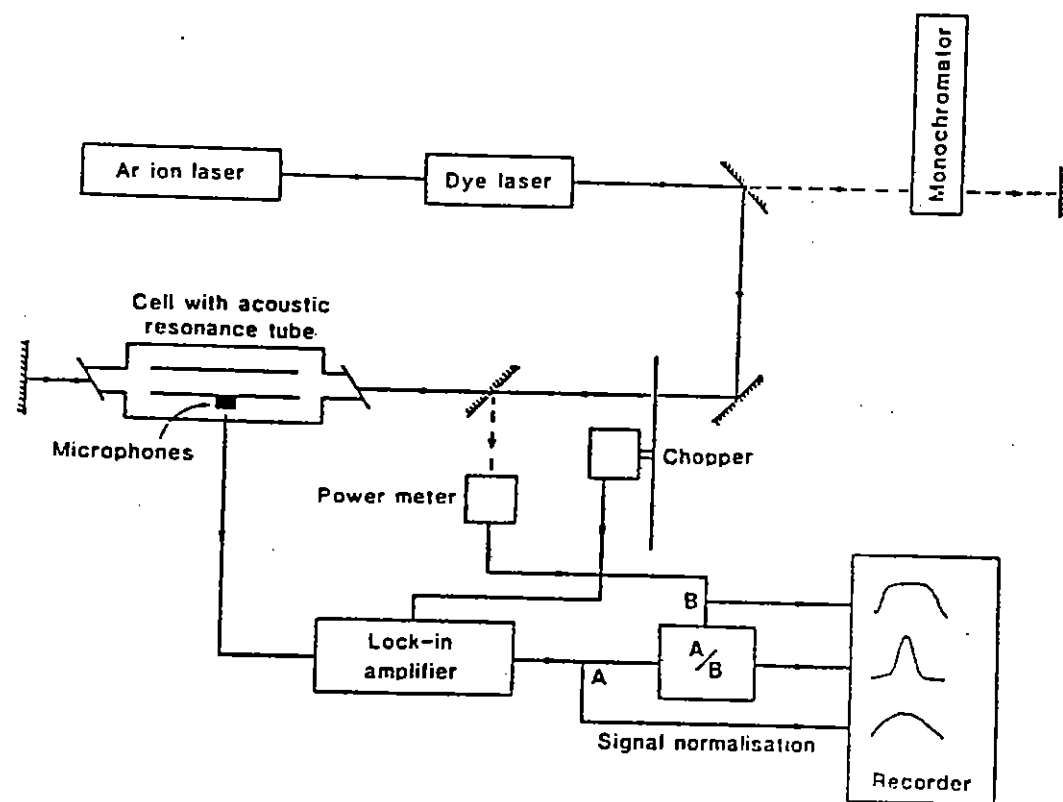


Fig. B1.29 Laser photoacoustic apparatus.

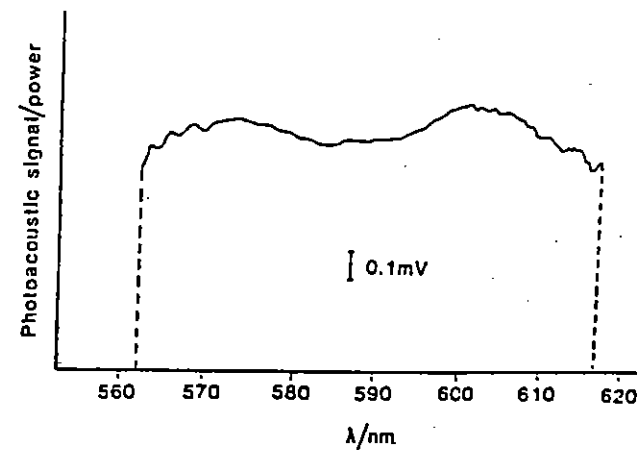


Fig. B1.30 A normalised photoacoustic spectrum showing the Chappuis band of ozone. Pressure of $O_3 = 10^{-3}$ atmospheres.

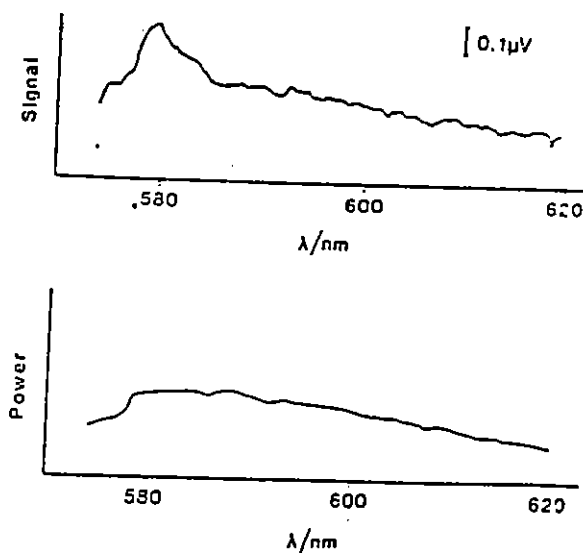


Fig. B1.31 A photoacoustic spectrum of oxygen showing at 579 nm the band due to the 'dimol' transition. The lower trace shows the power output of the laser. Pressure of $O_2 = 1$ atmosphere.

lasers, have demonstrated that it is possible to observe the dimol transitions of oxygen photoacoustically. It is now necessary to reduce the noise still further to obtain better signal/noise ratio. This will be accomplished by removing the photoacoustic cell to another room and shielding it further from external noise sources. Then we shall enclose it in a thermally insulated container to enable measurements at low temperatures to be made, which will be the primary object of future study. This may necessitate some testing of microphone performance at low temperatures.

Acknowledgements

We would like to thank Messrs M C Davies and R Goodwin for assistance with electronics and analytical work, Mr D James (University Workshop) for construction of the Photoacoustic cell, Dr P Wilson (Communications and Neuroscience) for helpful advice and Dr A Langley (Rutherford-Appleton Laboratory) for his skill and advice when installing the lasers. The provision of the laser system through the laser loan facility of the Rutherford-Appleton Laboratory is gratefully acknowledged.

References

1. J W Ellis and H O Kneser, Z. Physik., 1970, 92, 3293.
2. A U Khan and M Kasha, J. Amer. Chem. Soc., 1970, 92, 3293.
3. O L J Gijzeman, E A Ogryzlo and R P H Rettschnick, J. Chem. Phys., 1970, 52, 3718.
4. A R W McKellar, N A Rich and H L Welsh, Canad. J. Phys., 1972, 50, 1.
5. S J Arnold, E A Ogryzlo and H Witzke, J. Chem. Phys., 1964, 40, 1769.
6. P Borrell, P M Borrell and K R Grant, J. Chem. Soc., Faraday Trans. 2, 1980, 76, 1442.
7. R Boodaghians, P M Borrell, P Borrell and K R Grant, J. Chem. Soc., Faraday Trans. 2, 1982, 78, 1195.

8. P Borrell and N H Rich, Chem. Phys. Letters, 1983, 99, 144.
9. A P Billington, P Borrell and N H Rich, J. Chem. Soc., Faraday Trans. 2, 1987, in press.
10. A Rosencwaig, Photoacoustics and Photoacoustic Spectroscopy, J. Wiley, New York, 1980.
11. D Bicanic, Some Applications of Photoacoustic and Related Sensing Methods Relevant to Agriculture, Agriculture College, Wageningen, Netherlands, 1987.
12. M J McEwan and L F Phillips, The Chemistry of the Atmosphere, Arnold, London, 1975.
13. M Griggs, J. Chem. Phys., 49 (1968) 87.

B1-15 DECAY OF TRANSIENT ABSORPTION FOLLOWING PICOSECOND LASER EXCITATION OF MICROCRYSTALLINE 1, 2 PYRAZOLINES.

G P Kelly, P A Leicester and F Wilkinson (Department of Chemistry, Loughborough University of Technology)
L F V Ferreira (Centro de Química Física Molecular da Universidade Tecnica de Lisboa).

Last year we reported (1) on the first ever observation within microcrystals of a laser induced transient which decays on subnanosecond timescales using the technique of picosecond diffuse reflectance flash photolysis developed at RAL (2-4) for the case of 1, 5 diphenyl-3-styryl-2-pyrazoline. The ability to observe transients on picosecond timescales in microcrystalline materials or materials on opaque substrates is a significant advance (5). Japanese workers (6) have followed up our lead and have built an apparatus, using diffuse reflectance with diode arrays, capable of giving sub-nanosecond time-resolved spectra following picosecond laser excitation. In this report we describe work done in a three week allocation of time on the LSF picosecond laser in 1987.

We have developed software to allow for more efficient collection of data and to allow for laser pump variations, to correct for emission from samples and for non-linear saturation effects. This has led to improvements in both the quality and quantity of the data which can be obtained.

Further experiments on pyrazolines using pump and probe wavelengths of 295 and 590nm respectively showed that the transient previously observed could be strongly suppressed by the addition of an energy acceptor, disulphone magenta. In studies of the fast transient, attempts to fit the decay with the superimposition of two exponential decays i.e. with equations of the form

$$\frac{\Delta R}{R}(t) = \Delta I'_0 \exp(-k_1 t) + B \exp(-k'_1 t) \quad (1)$$

were unsuccessful (see Figure B1.32). However, superimposition of a first order and a second order decay fitted the data within experimental error (see Figure B1.33) i.e.

$$\frac{\Delta R}{R}(t) = \Delta I'_0 \exp(-k_1 t) + \frac{B}{1 + k_2 C_0 t} \quad (2)$$

This is consistent with the short-lived exponential component being absorption due to a higher excited singlet state which decays rapidly to give the first excited singlet state S_1 , (7), also absorbing at this wavelength, which then undergoes mainly bimolecular decay due to exciton-exciton annihilation.

It is of course necessary to observe the spectral shape of the transients in order to identify the mechanism involved and so attempts were made to use the picosecond continuum as a probe. Low intensity and large shot to shot fluctuations in the continuum made it difficult to extract meaningful data on this occasion although some evidence was obtained that more than one transient species was absorbing in the region between 500 and 680nm.

References

1. F Wilkinson, G P Kelly, P A Leicester, A R Anstee, J R M Barr and A J Langley, Annual Report to the Laser Facility Committee, 1987.
2. F Wilkinson, C J Willsher, P A Leicester, J R M Barr and M J C Smith, J Chem Soc Chem Comm 1216 (1986).
3. F Wilkinson, C J Willsher, P A Leicester and M J C Smith, 'Picosecond Laser Flash Photolysis of Opaque Materials'. Proc. XI IUPAC Symp. on Photochem., 155, (1986).
4. F Wilkinson, 'Diffuse Reflectance Flash Photolysis'. J Chem Soc Faraday II, 82, 2073, (1986).
5. G Porter, J Chem Soc Faraday II, 82, 2445, (1986).
6. F Wilkinson, G P Kelly, P A Leicester, J R M Barr and A R Anstee. Submitted for publication.

B1-16 LASER INDUCED DESORPTION AND ENERGY TRANSFER AT SURFACES

C J S M Simpson (Oxford University)

1. Thermal laser induced desorption of physisorbed atoms and molecules

The object of these experiments was to discover the factors which control laser induced desorption of atoms and of molecules.

We had discovered already that the velocity distribution of desorbed species depends greatly upon surface coverage.

(1) 3D layers

We investigated the desorption of 3D layers of Ar and of CH₄ on a Cu (III) crystal between 8 and 25 K. We found that the distributions are quite close to that for a single Boltzmann designed fitted to the peak maximum. The temperature of the desorbed 3D layers does not depend upon that of the crystal.

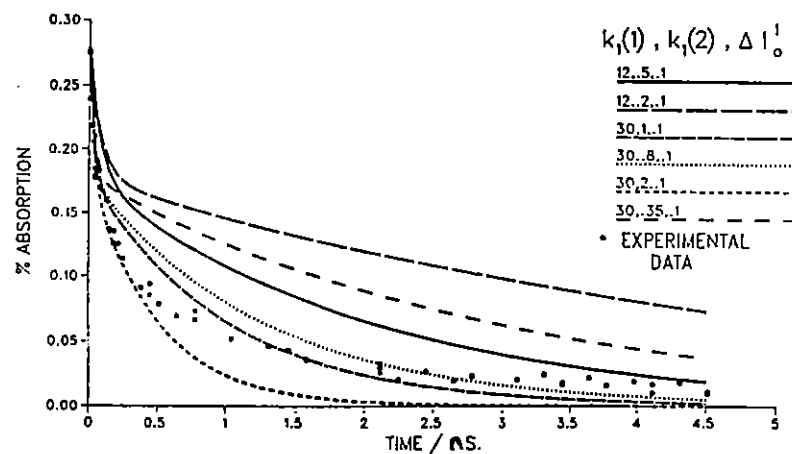


Fig. B1.32 Experimental transient absorption decay from a sample of microcrystalline 1, 5 diphenyl-3-styryl-2-pyrazoline, fitted with various data sets created from a double exponential mechanism (i.e. two superimposed transient species both decaying by first order processes).

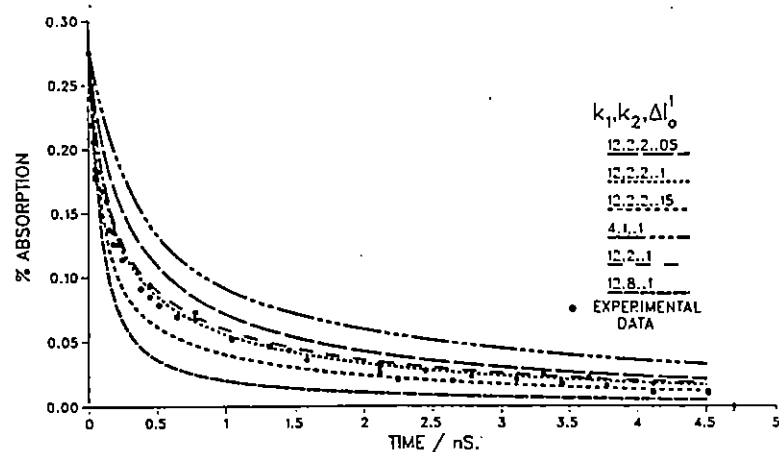


Fig. B1.33 Experimental transient absorption decay from a microcrystalline sample of 1, 5 diphenyl-3-styryl-2-pyrazoline, fitted with various data sets created from a mechanism involving the superimposition of two transient species, one decaying by a first order process and the other by a second order process.

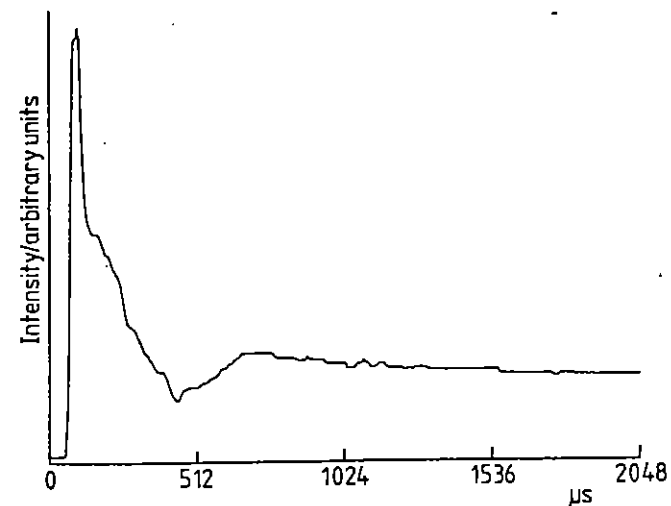


Fig. B1.34 Initial trace of the desorption of multilayers.

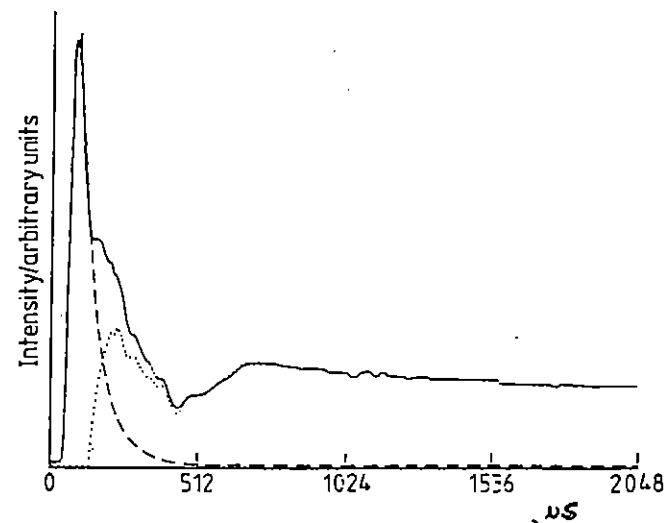


Fig. B1.35 Deducing the binding energies of successive layers. When the fit of the first peak is subtracted the second peak is seen clearly. This process is repeated.

It has been shown by Weitz and coworkers that large deviations from a single Boltzman distribution can be caused by a poorly focused mass spectrometer system. Clearly we are not afflicted by these troubles. The fairly small excess population present at longer times we ascribe to desorption of species which are bound more firmly than in the 3D multilayer.

(2) Multilayer

The thickness of the adsorbed layers for a given dosing pressure is controlled by the temperature of the crystal which governs the rate of evaporation of the adsorbed molecules, or atoms. This in itself implies a range of binding energies for the adsorbed layers dependent on coverage. We have found very broad distributions of desorbed species. The signal to noise is not nearly as good as for 3D layers and we averaged up to 120 runs. An extreme example is shown in Figure B1.34. The distributions of desorbed species are very broad. We suggested that this is due to the superposition of several different distributions corresponding with different binding energies,

multilayers being adsorbed with much lower temperatures than are 3D layers.

(3) The dependence of heat of adsorption on coverage

We have measured the change in pressure of sample gas when the crystal is cooled to different temperatures. This shows that layers are built up in a series of steps corresponding to different heats of adsorption. Our results closely resemble those found by others for the adsorption Xe on graphite.

We conclude that the broad velocity distributions which we have found are due to the superposition of different Boltzmann distributions from layers with different binding energies. For 3D layers the majority of the species are bound with comparable energies giving an overall distribution close to that for a single Boltzmann distribution.

(4) Rotation - translation equilibrium

From our results for the desorption of CH_4 and of Ar we conclude that

the rotational and translational degrees of freedom of molecules are equilibrated when they are desorbed. Exceptions to this may be H_2 and D_2 which have exceptionally small rate constants for rotation - translation equilibrium, particularly at low temperatures.

Publication

This work has been written up for publication.

(5) Extension to very low temperatures

Thermal desorption of H_2 , D_2 and HD from graphite material in the temperature range 5 to 20 K is considered to be very important in the chemistry of interstellar dark clouds. We have improved our apparatus so that we can work down to 5 K and have obtained the first results on the desorption of D_2 at this temperature. The way is now open for us to work on the desorption of n and p H_2 , O and n D_2 and HD given adequate laser time.

At present no other group studying laser induced desorption has worked under these conditions. Further experimentation is much required to conclude this work.

2. Single quantum laser desorption

In thermal laser induced desorption the surface is heated very rapidly ($> 10^{10} \text{ K s}^{-1}$) by the ns pulsed laser. This does not occur if the molecule is adsorbed onto a surface which is transparent to the laser radiation. In fact if the adsorbed molecule does not itself absorb the radiation it will not be desorbed by the laser pulse. However if the molecule does absorb the laser radiation it may be desorbed provided that (1) sufficient energy is available to break the molecule to surface bonds and (2) the process of bond rupture is fast compared with that of energy transfer from the excited molecule to the surface.

We have been able to meet these conditions working in the near u.v. (265-270 nm) with 1,3 cyclohexadiene adsorbed on MgF_2 .

(1) Neither 1,3 cyclohexadiene, nor cyclohexane is desorbed by a laser pulse at 540 nm though this would have sufficient energy to desorb these molecules from a Cu surface. This is because the MgF_2 surface remains cold.

(2) Cyclohexane is not desorbed from MgF_2 by a laser pulse at 270 nm. This is because both the cyclohexane and the MgF_2 crystal are transparent at this wavelength.

(3) 1,3, cyclohexadiene is desorbed from MgF_2 by a 270 nm laser pulse with the same energy as that which does not desorb cyclohexane. This is because the laser pulse absorbed provides sufficient energy to break the molecule to surface bands and the rate of energy transfer to the surface is slower than the bond rupture process.

The electronic state of the desorbed molecule

A quantum of energy at 270 nm provides 442 kJ mole^{-1} of energy which is far in excess of the heat of adsorption which lies in the range $30\text{--}120 \text{ kJ mole}^{-1}$. However clearly all of this energy is not available for the rupture of the molecule to surface bonds. If the process of bond rupture is faster than decay of the electronically excited state then the desorption involves electronically and vibrationally excited molecules. If the process of bond rupture is slower than that of fluorescence decay then the desorption involves vibrationally excited molecules in their ground electronic state. It should be possible to settle this question by looking for the fluorescence or phosphorescence in the case of triplets emitted. However it is likely that deactivation of the electronically excited state to the ground state occurs before bond rupture.

Again, more laser time is needed to study the luminescent processes which occur

Energy transfer between adsorbed molecules

Near resonant energy transfer is expected to be very rapid between adsorbed molecules and may be very much faster than the process of bond rupture. If this is the case energy will be transferred to a molecule which does not itself absorb the u.v. radiation and this may be sufficient to desorb this second molecule.

We have selected two molecules namely 1,3 cyclohexadiene and cyclohexane for which there is a small energy mismatch between the lowest vibration frequencies of their ground electronic states.

Both molecules are desorbed which proves that the energy transfer process is faster than the desorption of the molecule initially excited and is evidence that the ground electronic state of each molecule is involved.

If a pseudo equilibrium is established for sharing the energy between these molecules before either is desorbed then the ratio in which the molecules are desorbed will be governed by the energy mismatch the degeneracies of the molecules and the temperature of the surface. This ratio will change with temperature and can be calculated from the energy mismatch. We have proved that such a pseudo equilibrium is indeed established.

Further experiments are needed in order to determine the relative rates of energy transfer and desorption.

The effect of the angle of polarization of the laser beam.

Monolayers of the adsorbed molecules lie flat on the surface though this ordering may be lost for 3D layers. It is therefore to be expected that at least for some multilayer coverage whether molecules are desorbed will depend upon the angle of polarization of the light relative to that of the surface. This has been found to be the case.

The kinetic energy of the desorbed species

Since the energy is supplied as a single quantum the kinetic energy of the desorbed molecule depends upon the fluorescence energy, the binding energy, the vibrational energy in the desorbed molecule and that transferred to the surface.

$$\text{KE} = h\nu_{\text{abs}} - h\nu_{\text{fl}} - \Delta E_{\text{B}} - \Delta E_{\text{mol}} - \Delta E_{\text{surf}} \quad (1)$$

where $h\nu_{\text{abs}}$ is the energy of the quantum desorbed, $h\nu_{\text{fl}}$ that given out as luminescence ΔE_{B} the binding energy, ΔE_{mol} the vibrational energy in the desorbed molecule and ΔE_{surf} that transferred to the surface.

We assume that no energy is transferred to the surface and that the vibrational energy in the molecule is taken up either in breaking the

molecule to surface bond, or is released as kinetic energy. If the aluminescence occurs from the lowest vibrational state of the electronically excited state then its emission spectrum will not depend upon the magnitude of the quantum absorbed though the quantity of vibrational energy transferred to the ground electronic state will do so.

In this case the change in the K.E. released for the desorption of species with the same binding energy will be given by the difference in the size of the quanta of radiation absorbed. This is being tested using our results for 3D multilayers. We have at least a hundred results needing careful analysis.

The variation of binding energy with coverage

We have found that for the desorption of multilayers the histogram of the number of desorbed species against time gives a curve which has definite steps in it. An example is shown in Figure B1.34. The first peak can be fitted well by a Boltzmann distribution. When the values for this calculated distribution are subtracted from the experimental curve a second peak is clearly visible (see Figure B1.35). This is then fitted by a second Boltzmann distribution corresponding to a higher binding energy. By repeating this process we have been able to determine the binding energies of successive layers.

We have also determined the binding energies using layers of different thicknesses formed by dosing the crystal for different times.

We have found a change in binding energy of 70 kJ mole^{-1} between the monolayer and the 3D layer.

Because of this large variation of binding energy with coverage it is possible to employ a laser pulse with a frequency such that the quantum is large enough to desorb 3D multilayers but not monolayers. For those layers for which there is only just sufficient energy to break the molecule to surface bonds the kinetic energy released is very small and these molecules move away from the surface very slowly. This can give a dramatic change in shape in the desorption histogramme. This is illustrated in Figure B1.35.

A wave length cut off for the desorption process

It is clear from equation (1) that there must be a long wavelength cut off beyond which desorption will not occur. This can be calculated from the K.E. released for a given coverage and laser frequency. In our case the cut off frequency has been calculated to occur at a frequency at which the molecule still absorbs strongly. We have verified that this cut off frequency exists.

Publication

This work is being written up and is being submitted for publication.

B2	BIOLOGY	Pages
B2-1	UV Resonance Raman Studies of the Enzyme Glyceraldehyde-3-Phosphate Dehydrogenase	197 - 200
B2-2	Laser-Induced Trapping of Breaks in UV- Damaged Mammalian Cell DNA	200 - 204
B2-3	Investigation of Fast Absorption Changes in Photosystem 1	204 - 208
B2-4	Laser Flash Absorption Studies of Photosynthetic Reaction Centres	208 - 212
B2-5	Thermodynamic and Kinetic Basis of Globin of Gene Expression	212 - 216
B2-6	Measurement of Internal Motion in Proteins; Time-Resolved Fluorescence Studies of Lactate Dehydrogenase using a Streak Camera	216 - 220

Editors: A Parker/A J Langley/S Tavender

B2-1 UV RESONANCE RAMAN STUDIES OF THE ENZYME GLYCERALDEHYDE-3-PHOSPHATE DEHYDROGENASE.

J C Austin, R E Hester and C W Wharton*

Chemistry Department, University of York, and *Biochemistry Department, University of Birmingham.

Introduction

The enzyme glyceraldehyde-3-phosphate dehydrogenase (GAPDH), catalyses the conversion of glyceraldehyde-3-phosphate (GAP) to 1,3-diphosphoglycerate with a concomitant reduction of the coenzyme nicotinamide adenine dinucleotide, NAD^+ , to NADH. The reaction mechanism involves the formation of an acylenzyme intermediate which is fairly stable in the absence of phosphate or other nucleophiles. The acylenzyme intermediate contains a thioester group at the active site which absorbs at ca. 240 nm. Aromatic amino acid residues of the protein and NAD^+ also absorb strongly in the wavelength range 200–280 nm; see Table 1. Thus, by varying the excitation wavelength, resonance Raman (RR) spectra should, in principle, show features due to NAD^+ , the amino acid residues, and the thioester intermediate.

We have investigated (a) the RR spectra of the acylenzyme intermediate excited at two wavelengths close to the thioester absorption maximum, and (b) the effects of coenzyme binding on the RR spectra of the enzyme and coenzyme.

Acylenzyme study

Two wavelengths have been employed in attempts to observe the thioester intermediate; 240 nm, the thioester absorption maximum, and 248 nm, a wavelength where the protein absorption is at a minimum. 240 nm laser radiation was produced by frequency doubling the output from an excimer-pumped dye laser, using a β -barium borate crystal. 248 nm radiation was also obtained from this system, and alternatively from a line-narrowed KrF excimer laser. Spectra were collected using a Spex Triplemate spectrometer with an OSMA intensified diode array detector. Typically, enzyme spectra were collected for 600–1200s, operating the dye laser at ca. 1mJ/pulse at 30Hz, giving RR band intensities at ca.

1 count/s. The enzyme solutions (ca. 10^{-5}M) were continuously flowed through a quartz capillary tube so as to present a new sample to each pulse of the laser. The acylenzyme was continuously formed by mixing enzyme and substrate solutions in the presence of excess NAD^+ .

In Figure B2.1 the RR spectrum of GAPDH in the presence of excess NAD^+ is compared with the corresponding spectrum from the acylated enzyme/coenzyme system. The spectra show features due to tyrosine, tryptophan and NAD^+ vibrations, but no strong bands appear that might be attributable to the thioester. There are notable differences between the spectra; for example, a new band appears in the acylenzyme spectrum at 979 cm^{-1} , whereas bands at 1034 and $1070\text{--}80\text{ cm}^{-1}$ disappear. Bands due to the aromatic amino acid residues are significantly different in their relative intensities and show small wavenumber shifts with respect to the spectra of the corresponding free amino acids. For example, the 1510 cm^{-1} band assigned to a tyrosine vibration is found at 1516 cm^{-1} in the tyrosine solution spectrum. Such changes are attributed to the non-polar environment experienced by the side chains when buried in the bulk of the protein.

Spectra obtained with 248 nm excitation were of lower intensity, and show mainly bands due to the excess NAD^+ in the system. Attempts were also made to obtain protein spectra with 220 nm excitation but, due to limitations of the Triplemate spectrometer, satisfactory spectra were not obtained. We hope to correct this in the near future.

Coenzyme binding

Using 260 nm excitation from the frequency-doubled dye laser, excellent RR spectra of free NAD^+ were obtained, mainly showing enhancement of the adenine bands, although one nicotinamide band at 1030 cm^{-1} is just visible. Spectra of the holo- and apo- enzyme are compared in Figure B2.2. Certain bands that are common to both spectra are clearly assignable to tryptophan and tyrosine modes (980 , 1060 , 1580 and 1625 cm^{-1}). The bands due to bound NAD^+ in the holoenzyme spectrum show marked intensity changes as compared with the spectrum of free NAD^+ , and also small wavenumber shifts. The 1337 cm^{-1} band of the NAD^+ 'triplet' at 1378 , 1337 and 1310 cm^{-1} in the holoenzyme spectrum is of much lower relative intensity than in the free NAD^+ spectrum. Similarly the 1482 cm^{-1} NAD^+ peak appears to be of lower relative intensity.

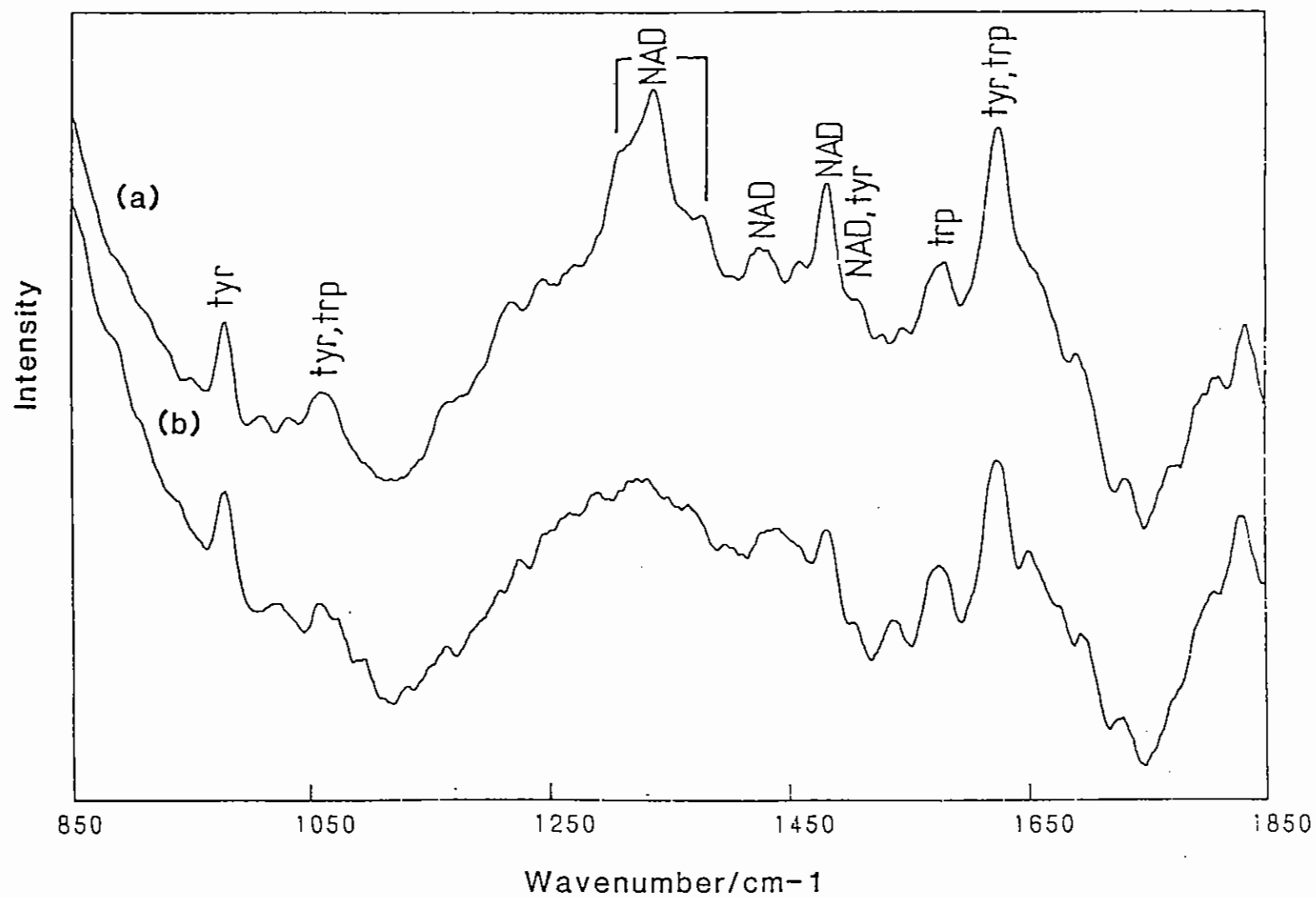


Fig. B2.1 RR spectra of (a) GAPDH and (b) GAPDH with substrate, GAP.

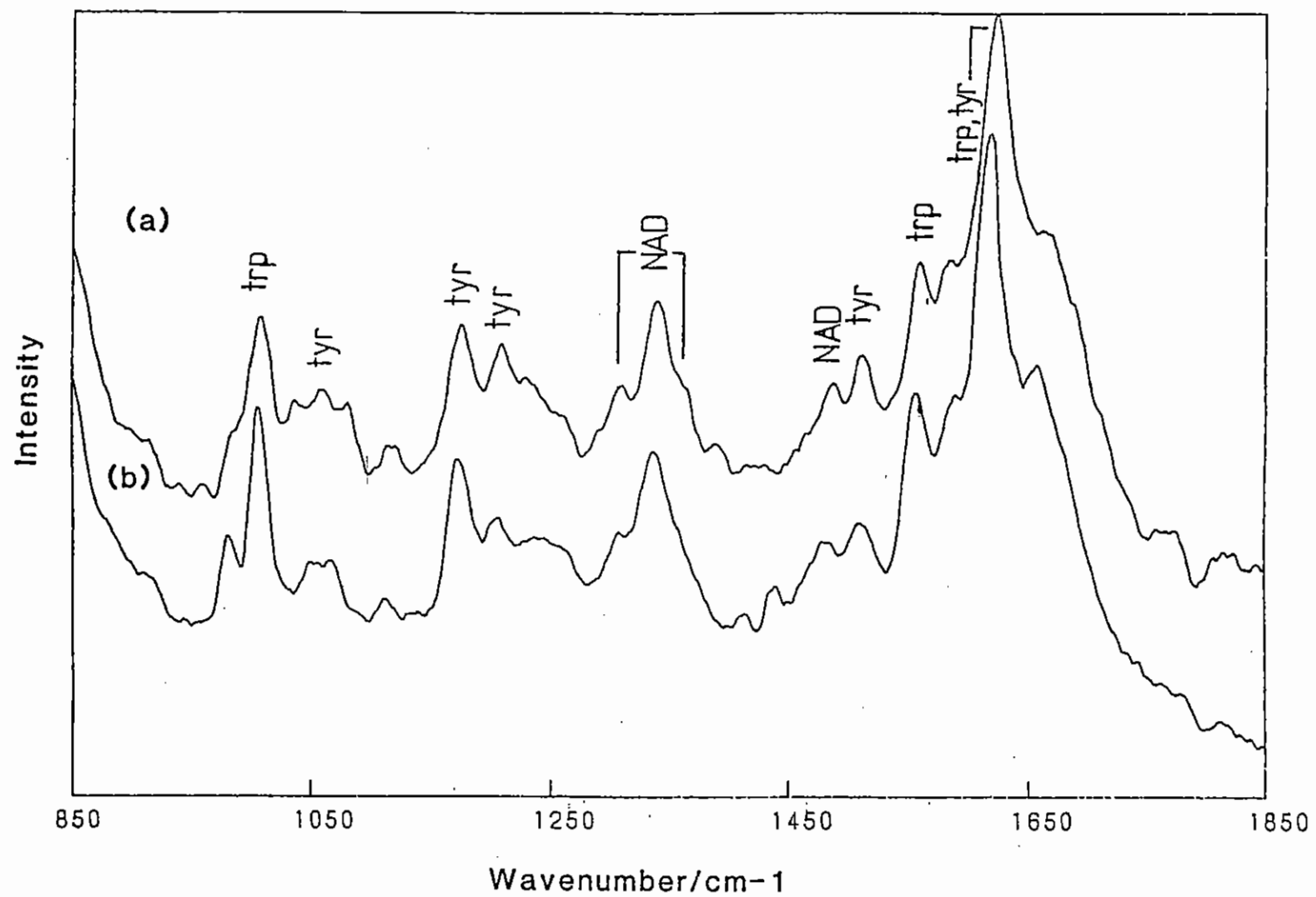


Fig. B2.2 RR spectra of (a) holoenzyme and (b) apoenzyme.

Concluding comment

We can now selectively probe, by UVRR, different moieties within the GAPDH system using the tunable frequency-doubled dye laser. The wavelength stability of this laser is superior to that of the line-narrowed excimer laser, resulting in higher resolution Raman spectra. A detailed interpretation of the spectra and changes reported here, in terms of structure and environment of the coenzyme and enzyme, is in hand.

Table 1; UV absorption maxima

	λ max/nm
NAD ⁺	260, 210
NADH	340, 260, 210
Tryptophan	280, 217, 194
Tyrosine	275, 222, 193
Phenylalanine	260, 207, 180
Thioesters	ca. 240

82-2 LASER-INDUCED TRAPPING OF BREAKS IN UV-DAMAGED MAMMALIAN CELL DNA

R A Meldrum, P Ffitch, S Armstrong and C W Wharton

The efficient repair of damage in DNA of mammalian cells by radiation or chemicals determines their ability to survive or maintain the integrity of their genetic identity and hence their resistance to mutation. The sensitivity to such damage and the ability of the cells to overcome this may be measured by the appearance and disappearance of damage-induced breaks in the DNA over a time course.

An experimental procedure for the study of early and rapid events in DNA repair of breaks induced by radiation damage was described in the 1987 report, where damage of the DNA is caused by a 248nm laser pulse and a 351nm laser pulse photoactivates a "caged" alpha-³²P-labelled dideoxynucleotide which then traps DNA breaks.

Certain modifications of the procedure which was used to test the validity of the method were required to enable measurements of DNA breaks to be made immediately after damage and in short time courses thereafter.

Electropermeabilisation of Cells

The "caged" nucleotide which is required to be in situ before damage or photoactivation takes place, is introduced into the cells by making the cell membranes permeable. Formerly this was done by putting the cells in hypotonic solution then in isotonic solution to reseal them. By this method, however it takes several minutes (ie 5-15 minutes) to permeabilise the cells.

Recently, cells have been shown to be made permeable by electric shock (Chu et al 1987) which is effective in a few milliseconds. Platinum electrodes (0.9cm x 0.017cm) 3.8mm apart are inserted into a cuvette (4.5cm x 0.5cm) containing 0.8ml of cell suspension in buffer with the caged nucleotide. An electric pulse is then passed through the suspension. We have monitored the effectiveness of this method of permeabilisation by the inclusion of trypan blue dye by the cells or by uptake of ³H-TTP in the DNA of the cells after the electric pulse. The optimum permeabilisation of all cell types tested occurs at about 300 volts in a 7ms pulse.

When cells are permeabilised their endogenous nucleotide pool is depleted and so exogenous nucleotides must be present in the buffer during permeabilisation. It is important to establish the optimum concentration (in terms of conservation of expensive radiolabelled material) of exogenous nucleotides to add, such that the optimum rate of DNA synthesis is not limited by lack of any substrates. Experiments are in progress to calculate the K_m (the Michaelis constant) of the nucleotide substrates for the DNA-polymerase-catalysed reaction i.e. DNA-synthesis) for the different cell types used. The K_m is equal to the substrate concentration at which the reaction rate (incorporation of radiolabelled nucleotide) is half of its maximum

HL 60 cells at 5×10^5 / ml

Incorporation of ^{32}P ddATP after 248 nm laser irradiation

(80 joules J m^{-2})

○ without ara C / HU

10^{-5}M 10^{-2}M

● with ara C / HU

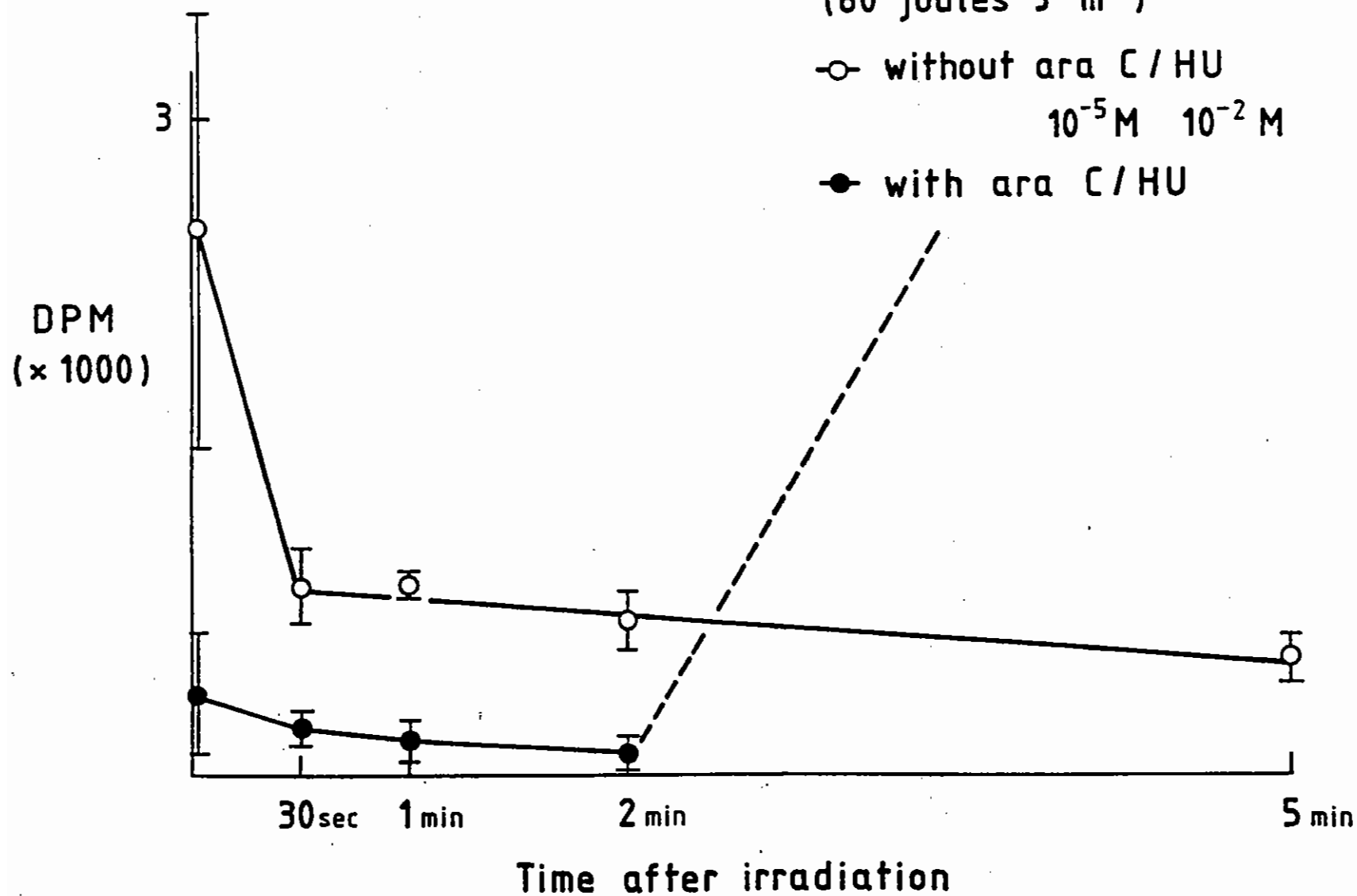


Fig. B2.3 Incorporation of ^{32}P ddATP after 248 nm laser irradiation.

value. A K_m of ca. $35\mu\text{M}$ of added nucleotides for some of the cell types used has been calculated, which indicates that a concentration of $60\mu\text{M}$ (twice K_m) would maintain an adequate rate of DNA synthesis.

Irradiation of cells in cuvettes

Automation of our break trapping method requires that cells be irradiated in quartz cuvettes so that they may be electropermeabilised. Trial experiments revealed some discrepancy between the effects of 248nm of UV irradiation on cells irradiated in cuvettes and cells irradiated in 35mm dishes. It is well established that radiation effects on cells may be modified in anoxic conditions. Cells irradiated in cuvettes experience less oxygenated conditions than cells irradiated in dishes which have a larger surface area exposed to the atmosphere. Since there is a reduced effect of radiation damage in cells irradiated in cuvettes, some adjustment of radiation doses will have to be made to take account of this.

Initial time course of DNA synthesis and repair after damage by 248nm UV irradiation

Progress has been made such that events may now be studied on a timescale extending to a few seconds after damage. The first event seen is the shutoff of replication synthesis which occurs within 30 seconds of damage (see Figure B2-3). Repair synthesis progresses rapidly from 2 min after damage at which time DNA replication synthesis shutoff is complete. Examination of Figure B2.3 reveals that initiation of repair synthesis occurs at two minutes if DNA replication inhibitors are present. It would appear from this experiment that the shutoff of DNA replication is not complete before 5 minutes when replication inhibitors are absent and repair does not begin until after this time. However in other experiments on different celltypes where no inhibitors are present replication synthesis shutoff has been shown to be complete before 2 minutes and repair synthesis initiated in this time (data not shown). We do not attempt to explain these experiments further until more work has been carried out.

Use of photolysed "caged" dideoxythymidine-triphosphate to inhibit incorporation of ^3H -thymidine-triphosphate

In the experiments which tested the validity of the DNA break trapping technique, "caged" ^{32}P -dideoxyadenosine triphosphate (^{32}P -ddATP) was

used as the break trapping agent. This was a compromise on the design of the experimental technique. Dideoxythymidine was the nucleotide analogue of choice as the nick trapping agent since deoxythymidine is specifically incorporated in DNA. Adenosine nucleotides are components of three major coenzymes, NAD⁺, FAD and CoA. ATP is also the universal currency of energy in biological systems. Therefore the incorporation of a dideoxyadenosine analogue may not be solely into DNA and background, non-specific incorporation may be not insignificant. A labelled form of dideoxythymidine is not as yet commercially available; indeed ^{32}P -ddATP is the only labelled dideoxynucleotide analogue available at the present time.

We modified our strategy to overcome this complication of possible non-DNA uptake of ^{32}P -ddATP. This was done by making "caged"-dideoxythymidine triphosphate and by measuring the inhibition by the photoactivated molecule on the incorporation of ^{32}P -thymidine triphosphate into DNA. Both these molecules are specifically incorporated into DNA. Thus the kinetics of the DNA repair process can be measured by the inhibition of ^{32}P -TTP uptake by photolysed "caged" dideoxythymidine triphosphate.

Measurement of incorporation by DNA synthesis of ^{32}P -TTP is not affected by non-specific uptake and results of experiments using this compound revealed that 351nm irradiation alone induced an immediate stimulation of DNA synthesis. Since 351nm UV irradiation has been shown not to damage DNA at the dose applied (RAL-CLF Report, 1985), this effect would appear to be a stimulation of DNA replication synthesis.

Action spectrum of DNA synthesis stimulated by UV light

In view of this unexpected finding we proceeded to determine the action spectrum of this effect by measuring DNA synthesis following irradiation at a level of $1\text{KJ}/\text{m}^2$ over the wavelength range 300-400nm. The different wavelengths were obtained as follows - 308nm from the Xenon Chloride HX460 laser, 320, 330, 340 from a FL 3002 dye laser pumped by an EMG 150 at 248nm (KrF). 350, 360, 370, 380, 390 and 400 from a FL 3002 dye laser pumped by the Lumonics HX460 Krypton Fluoride laser. Cell suspensions were placed in a 2cm^2 quartz vial and irradiated at $2\text{KJ}/\text{m}^2$. The quartz vial was turned through 180 degrees after receiving $1\text{KJ}/\text{m}^2$ radiation in order to even out light absorption.

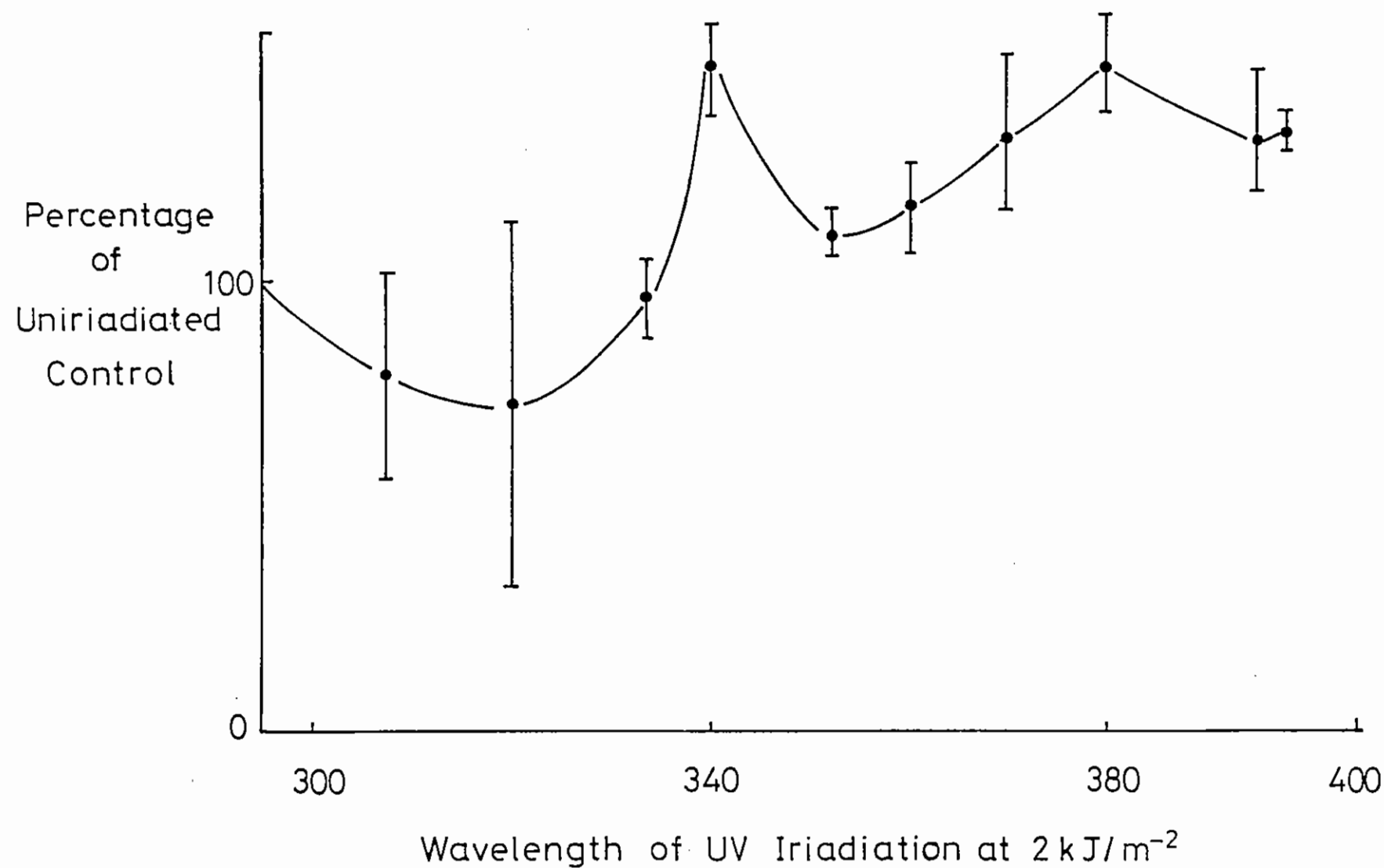


Fig. B2.4 Action Spectrum of ³H-TdR Uptake in HL60 cells.

Two peaks of stimulated DNA synthesis were obtained in the action spectrum. One peak was centred at 340nm and the other centred at 380nm (Figure B2.4).

At 340nm, we suspect that the target of radiation is NADH, since it shows an absorbance maximum at this wavelength. NADH is an important coenzyme involved in biosynthesis of nucleotides. NADPH which also absorbs at this wavelength is involved in the complex reaction of reduction of ribonucleotides to form the precursors of DNA. Absorption of 340nm UV by NADPH may affect the cell's nucleotide pool and hence the incorporation of radiolabelled thymidine which is a measure of DNA synthesis. NAD⁺ which arises by oxidation of NADH is an energy source for DNA ligase, the enzyme that seals gaps in the DNA. During DNA replication gaps in DNA exist on the "lagging" strand of the replication fork since the DNA is synthesised in small fragments on this strand. Absorption by NADH may lead to interference of the ligase activity and so appear to enhance DNA synthesis. There are however several possibilities which may explain this result and these will require extensive investigation in the future.

A less pronounced peak in the action spectrum occurs at 380nm. We do not venture to explain this, at present, and more evidence is required to confirm its authenticity.

We have also shown in our home lab that although similar trends occur in an action spectrum obtained by irradiating cells with a non-coherent light source (ie fluorimeter) the peaks are not as clearly distinguishable indicating that narrow bandwidth light is required for these experiments.

Immediate plans are to investigate DNA damage and repair induced by laser generated soft X-rays. Much information exists about the effect of X-ray pulses of the duration of seconds or longer but none on very rapid pulses such as those generated by lasers. It should therefore be possible for the first time to study the effect of photon hits received almost simultaneously.

Reference

1. Chu G, Hayakawa H, and Berg P, 1987, Nucleic Acids Research, 15, 3, 1311-1326.

B2-3 INVESTIGATION OF FAST ABSORPTION CHANGES IN PHOTOSYSTEM 1

R Sparrow, E G Brown and E H Evans (Lancashire Polytechnic, Preston, Lancs. U.K.)

R S Chittock and W T Toner (RAL)

M C W Evans (UCL, U.K.).

Introduction

We have continued to characterise fast absorbance changes in Photosystem 1 (PS1) with the aim of discriminating between absorption changes due to the light-harvesting chlorophylls, the P700 reaction centre chlorophyll and a proposed chlorophyll acceptor (1). Our original experiments, described in last year's report, had involved the pump-probe technique, whereby the sample was excited with the pump at 585 nm, the beam being split to provide a probe at 705 nm by Raman shifting that part of the beam through ethanol. During this last year we have attempted to produce 'tunability' to the system by exciting a continuum with the probe beam and monochromating this beam with interference filters. Considerable problems were experienced with this technique, so we have now returned to Raman shifting the probe beam and intend to acquire 'tunability' by alteration of the Raman shifting solvents. Using compensated beam splitting/balancing optics (see section B5.6) and an improved difference detector, data were routinely obtained with a precision as good as the best previously achieved on rare occasions.

Results and Discussion

All experiments were performed using a PS1 preparation from the cyanobacteria *Chloroglea fritschii* as previously described. Figure B2.5 shows an absorbance transient measured at 705 nm following excitation by a 6 ps. flash. This data show the presence of two-component bleaching transients which saturate with an effective excitation cross-section of $5 \times 10^{-15} \text{ cm}^2$. Time dependence for various samples is shown in Figure B2.5 and pump intensity dependence in Figure B2.6. The rise time is in all cases consistent with a prompt excitation but since the pump-probe overlap function is estimated to be 8 psec FWHM an excitation rise time of up to 5 psec is possible. The data are not consistent with a single exponential decay. The simplest model which fits all the data is that the observed transients are of

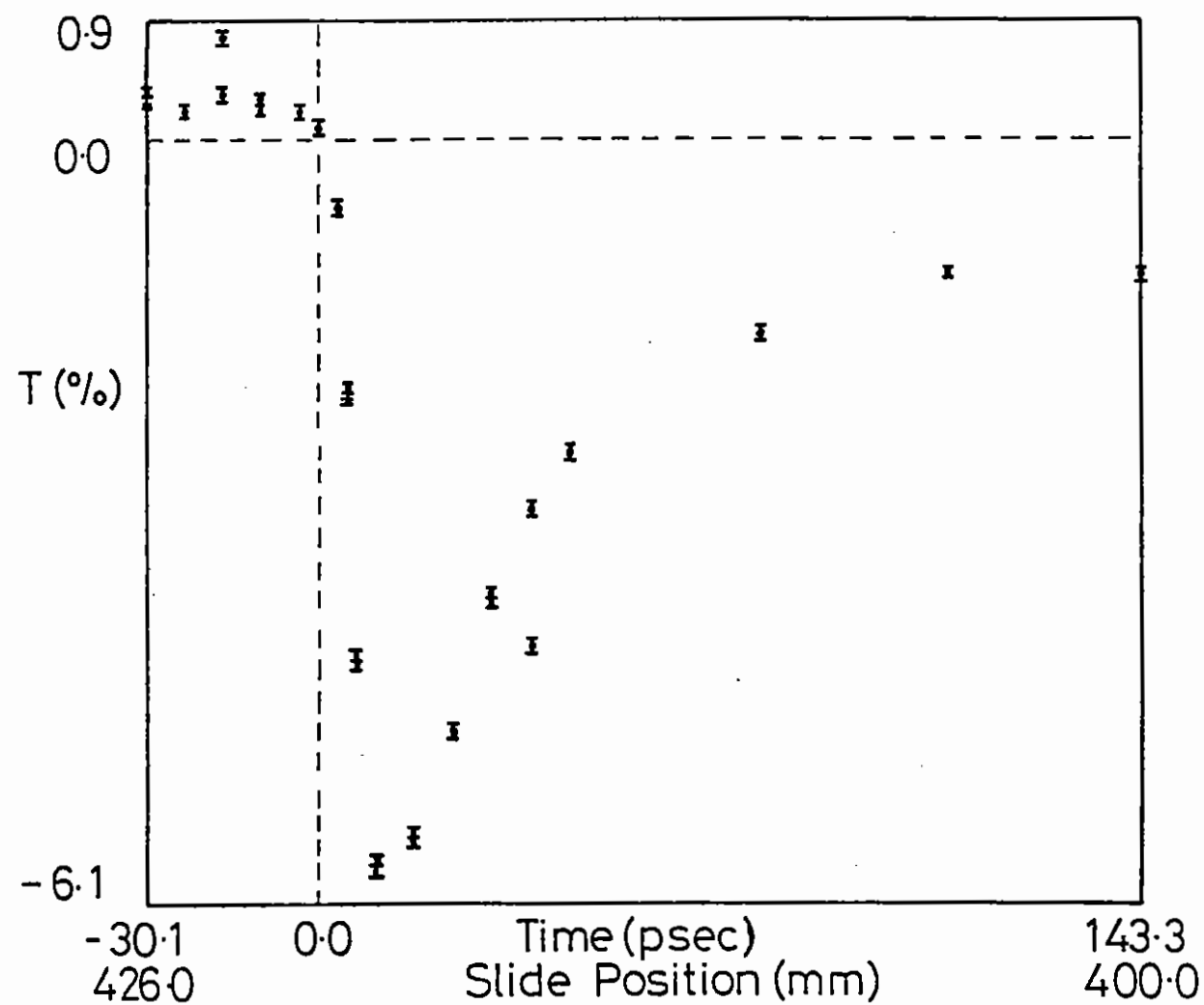


Fig. B2.5 Transient absorption change of PS1 at 705 nm in the presence of DCPIP (50 μ M) + Ascorbate (100 μ M). PS1 (22 μ g/ml SH1) in 0.06 M Tris-HCl, 0.03 M EDTA pH 7.8.

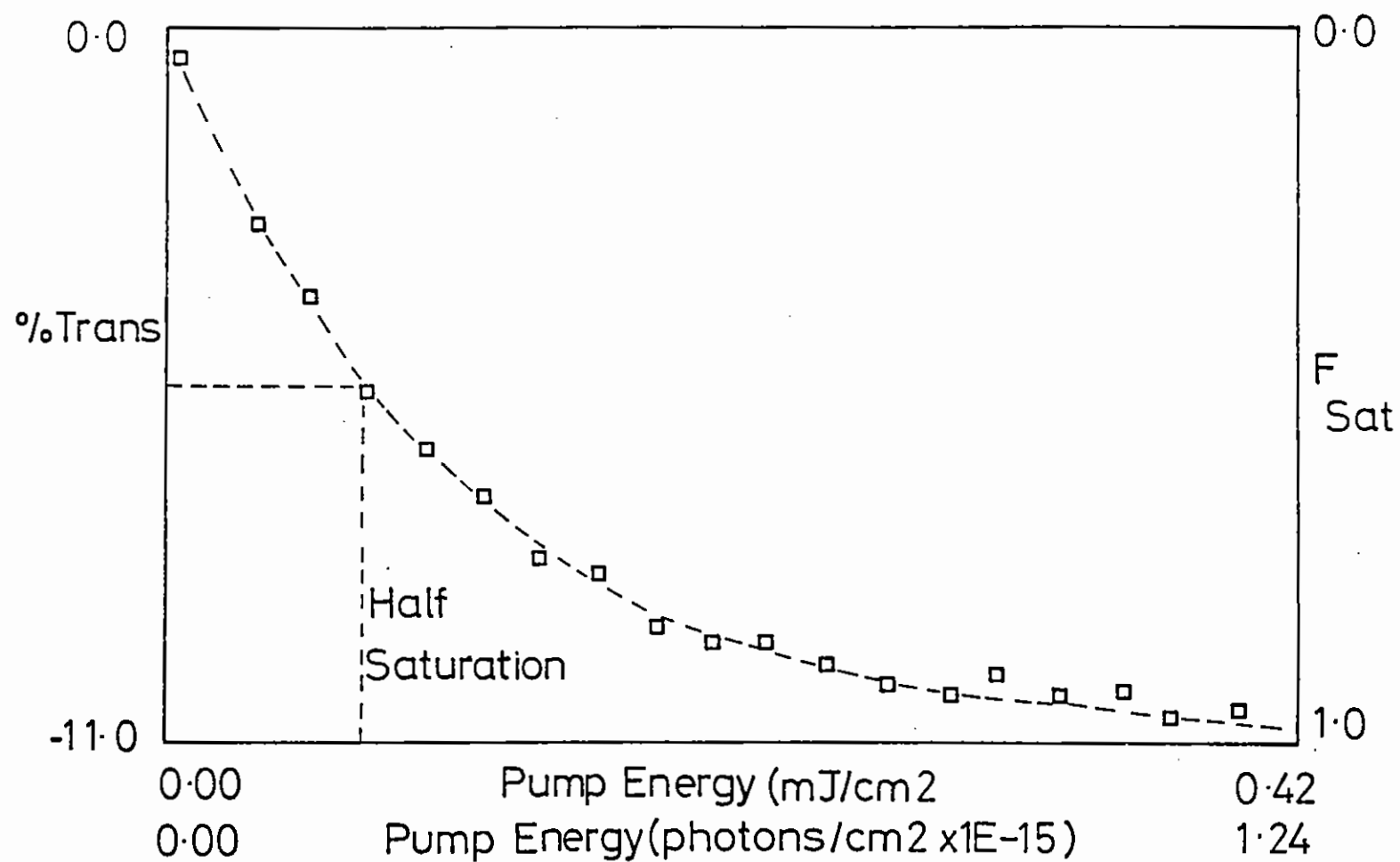


Fig. B2.6 PS1 absorption change dependency upon excitation intensity measured at 30 ± 7 ps after excitation at 705 nm. PS1 (22 $\mu\text{g/ml}$ CH1) in the presence of DCPIP (50 μM) + Ascorbate (100 μM) + Dithionite (excess) in a 0.06 M Tris-HCl, 0.03 M EDTA pH 7.8 buffer.

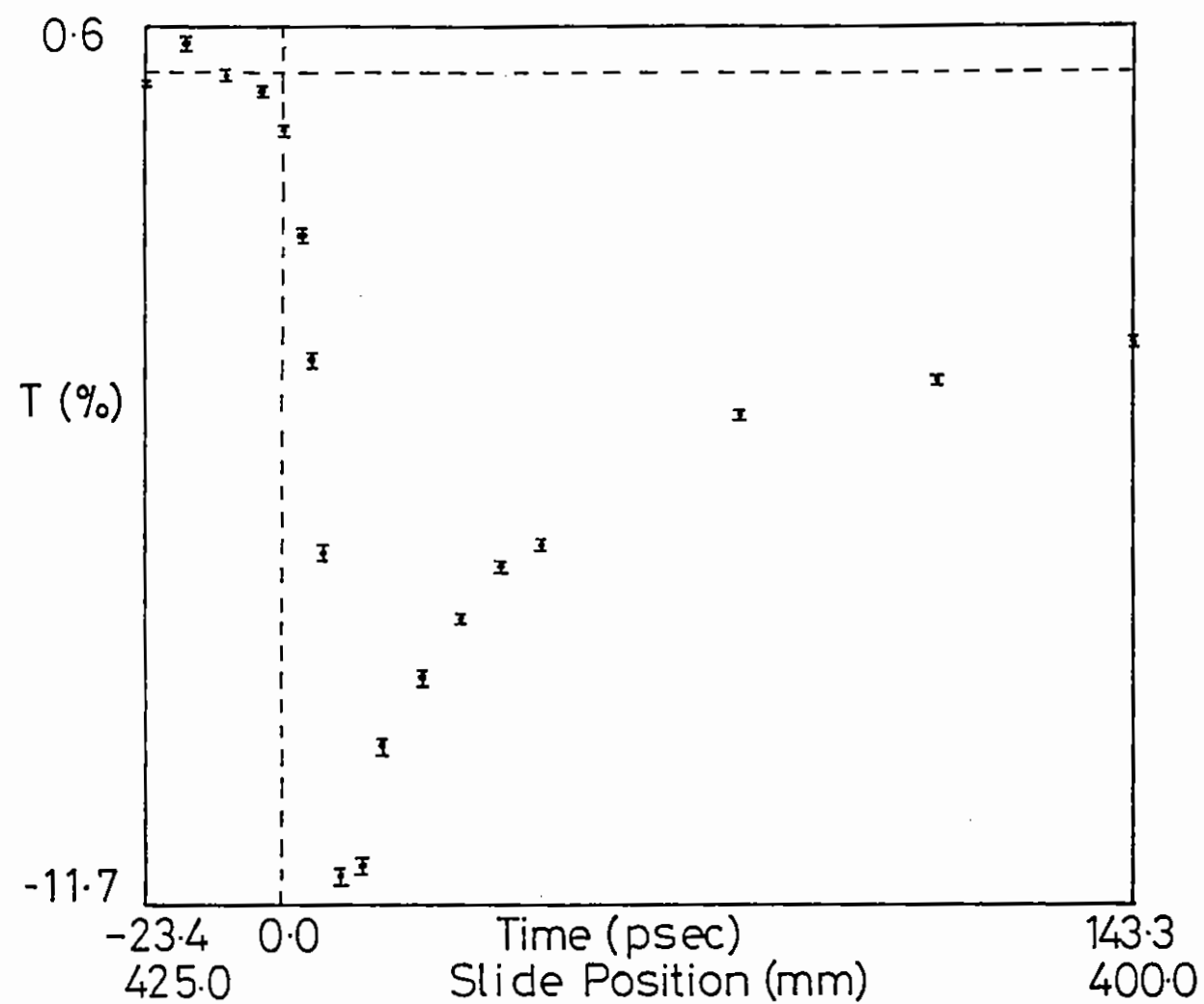


Fig. B2.7 Transient absorption change of PS1 at 705 nm in the presence of DCPIP (50 μ M) + Ascorbate (100 μ M) + Dithionite (excess) PS1 (22 μ g/ml CH1) in 0.06 M Tris-HCl, 0.03 M EDTA pH 7.8.

the form

$$a \exp - t/\tau_1 + b \exp - t/\tau_2$$

with $\tau_1 = 25 \pm 5$ psec and $\tau_2 \geq 11$ nsec,

the values of a and b varying from sample to sample and both transients having rise times of < 5 psec.

All the pump saturation data shown in Figure B2.6 can be fitted by a model with a single effective excitation cross-section of $5 \times 10^{-15} \text{cm}^2$.

As this is much too large for a single photon receptor, we interpret the observed bleachings as due to an effect in the reaction centre (loosely defined) of the PS1 consequent on the absorption of a photon in an antenna molecule. The saturation fits do not call for any appreciable linear component such as might be produced by antennae transients although contributions at the few percent level cannot be excluded. This is unlike data obtained by other workers (2), and may be a consequence of improved sample preparation and instrumentation.

We note that there is no evidence for any change in the character of the data at high flux where each PS1 suffers several hits. Most of the time dependent data were taken in conditions where single hits predominate.

We have also commenced a series of experiments to investigate the effect of varying redox potential on P700 kinetics. Our earlier experiments showed that the 705 nm transient exhibited a biphasic decay in the absence of externally added electron donor. Addition of DCPIP (2,6 dichlorophenolindophenol) and ascorbate had no effect on the recovery kinetics, unlike the observed situation in lettuce PS1 where the transient is irreversible in the absence of external electron donor (Linda Giorgi, personal communication). Addition of sodium dithionite to the sample at concentrations which would partially reduce the acceptor complex (Figure B2.7) caused a two fold increase in the extent of the long-lived absorbance change but had little effects on the short lived component. On some occasions enhanced long-lived components were recorded without the addition of reducing agents. This puzzling behaviour was noted in previous runs, indicating the presence of an uncontrolled variable in sample preparation, such as sample exposure to ambient light of probe wavelength. Our interpretation of this increased absorption and alteration in kinetics awaits spectral

resolution to establish whether the alteration is solely due to P700, or the presence of some additional component.

It is tempting to speculate that the transient is a mixture of both P700 bleaching and the reduction of a second component with overlapping absorption bands. The fast recovery could be due to the reoxidation of the latter and the long-lived component due to the slow reduction of P700.

References

1. J Bonnerjee and M C W Evans, FEBS Letts., **148**, 313-316, (1982).
2. M D Il'ina, V V Krasauskas, R R Rotoskis and Y Y Borisov, Biochim. Biophys. Acta **767**, 501-506, (1984).

B2-4 LASER FLASH ABSORPTION STUDIES OF PHOTOSYNTHETIC REACTION CENTRES

J A M Hubbard and M C W Evans

It is well established that the photosynthetic electron transfer chain of chloroplasts consists of two types of reaction centre operating in series. Photosystem I catalyses the oxidation of plastocyanin and reduction of ferredoxin, and photosystem II catalyses the oxidation of water and reduction of plastoquinone. Several aspects of the nature of these two photosystems that have been studied using laser-flash induced absorption changes are described below:-

1. Function of electron acceptor A_1 in photosystem I

Several electron acceptors have been identified in photosystem I, that may function between the excited donor, P700 and soluble ferredoxin. The nature and function of one of these electron acceptors known as A_1 has recently been the subject of much interest. Recent reports suggest A_1 is a quinone (vitamin K1), although the requirement for vitamin K1 for electron transport is controversial, so the function of A_1 still remains to be described. Some of our experiments directed to study this are described.

(a) Extraction of electron acceptor A_1 from pea photosystem I

Photosystem I particles were washed twice in either 0, 50 or 100% water-saturated diethyl ether. The characteristic electron spin resonance signal associated with electron acceptor A_1 was progressively lost with increasing percentage saturation of the ether. Optical measurements of $P700^+$ reduction at 820 nm were followed using a laser-flash spectrophotometer so that effects of removal of electron acceptor A_1 on electron transfer in PSI could be examined at room temperature (Figure B2.8). Control samples reduced with ascorbate showed decay kinetics with a half time of 30 ms characteristic of a back reaction from the iron-sulphur centres. This signal was decreased or absent in the samples in which A_1 had been removed, but it was replaced with a fast phase ($t_{1/2}$ 25 μ s) indicative of the back reaction from the $P700$ triplet to the ground state. Thus treatment of photosystem I with water-saturated diethylether which removes electron acceptor A_1 blocks electron flow to the terminal iron-sulphur centres (Mansfield et al., 1987).

(b) Extraction and reconstitution of electron acceptor A_1 using hexane or hexane plus methanol

Electron flow to the terminal iron-sulphur centres was not reconstituted by addition of vitamin K_1 (A_1) to ether-extracted particles so that we could not be certain that the effects seen could be attributed to removal of A_1 . Therefore another method was used to extract vitamin K1 from PSI particles of *Phormidium laminosum* and *Synechococcus leopoliensis*. Vitamin K1 was added back to the sample and in some cases incubated overnight at 4°. The optical absorbance changes were examined at 820 nm as before. The addition of vitamin K1 had no effect on control samples which showed a slow back reaction characteristic of electron transfer to the iron-sulphur centres.

However addition of vitamin K1 to the extracted samples resulted in an increase in the size of the initially small absorbance changes at 820 nm, back to values similar to those obtained with the controls. However the time course of this reconstitution and decay kinetics observed differed between the hexane and hexane plus methanol extracted samples. Further experiments are required to clarify these results. As A_0 and A_1 cannot be reduced by chemical methods, sodium dithionite

was added to reduce any exogenous vitamin K1 that may act as an electron acceptor. 5 mM but not 1 mM sodium dithionite completely removed the optical signals in the hexane extracted sample indicating that reconstitution of A_1 by vitamin K1 was incomplete.

2. Redox potential dependence of P680 reduction Kinetics

Several components have been identified on the acceptor side of PSII including I (pheophytin), Q_A and Q_B (plastoquinones). However a number of experiments including fluorescence spectroscopy and E.P.R. suggest that the acceptor side is very complex and that additional electron acceptors may exist. We have used absorption spectroscopy to study the acceptor side of photosystem II by following the reduction of the P680 cation formed by a laser flash. In samples where oxygen evolution is active almost all $P680^+$ is reduced by various components on the donor side, this takes place in < 50 ns, faster than the response time of our spectrophotometer. Treatment with hydroxylamine plus illumination inhibits oxygen evolution and electron donation from the donor D. $P680^+$ is now reduced more slowly (μ s and ms) by back reactions from reduced electron acceptors formed by the reaction $P680 + h\nu \rightarrow P680^+ + e^-$.

The reduction of $P680^+$ show at least 2 - 3 phases with different kinetics. The presence of the different phases were found to depend upon the ambient redox potential (Figure B2.9). At potentials above about -110 mV (at pH 10.0) the decay consists of 2 phases with half times of 300 - 400 μ s and several ms. Below -110 mV and above -300 mV 2 phases are still observed with half times of 100 - 200 μ s and several ms. At potentials below -300 mV but above -430 mV the decay is monophasic with half times of several ms. Below -430 mV absorbance changes due to $P680$ cannot be observed in the response time of our instrument.

The redox potential changes occur at similar potentials to the steps seen in the titration of the electron acceptors of Photosystem II studied by EPR. These results indicated that multiple electron acceptors may exist on the acceptor side. Thus although the molecular nature of the components giving rise to the different kinetics remains to be described, the laser-flash optical studies of photosystem II appear to confirm the EPR studies.

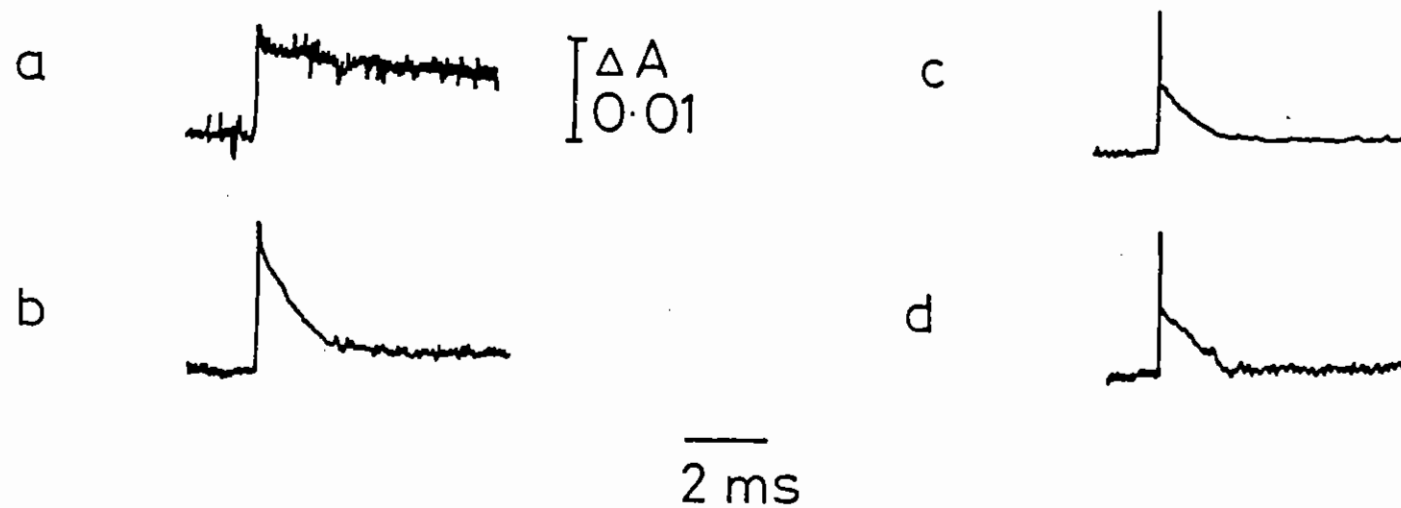


Fig. B2.8 Flash-induced absorbance changes at 820 nm in PSI following ether washing. (a) Freeze-dried control, (b) 0% water-saturated ether-extracted, (c) 50% water-saturated ether-extracted, (d) 100% water-saturated ether-extracted. Samples were flashed at 5 Hz and the following number of measurements were averaged: (a) 512; (b,c,d) 1024.

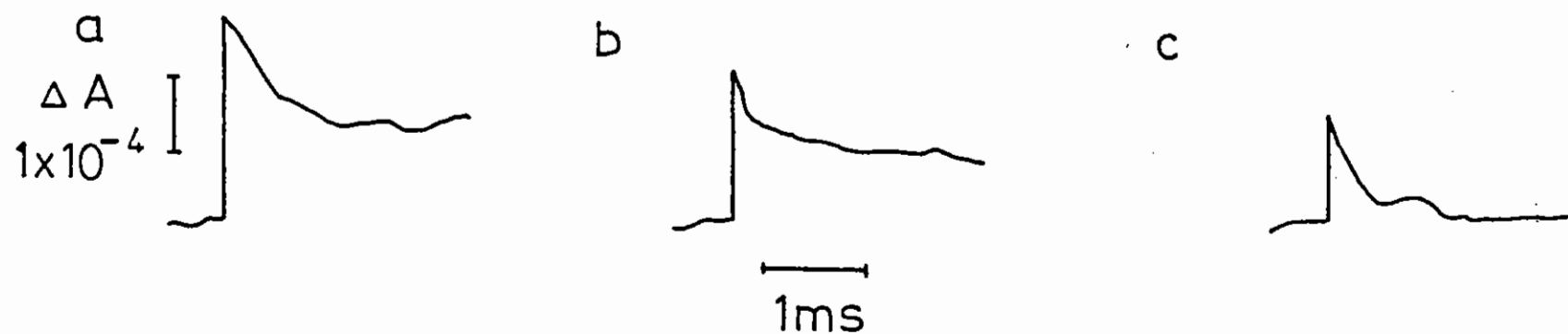


Fig. B2.9 Flash-induced absorbance changes at 820 nm in hydroxylamine-inhibited PSII at pH 10 at several different redox potentials: (a) + 110 mV; (b) - 110 mV; (c) - 395 mV. In each case 64 measurements were averaged at a flash rate of 2 Hz.

References

1. R W Mansfield, J A M Hubbard, J H A Nugent and M C W Evans, FEBS Lett., 220, 74-78, (1987).

B2-5 THERMODYNAMIC AND KINETIC BASIS OF GLOBIN GENE EXPRESSION

F Watanabe, E R Pike* and H J Gould

Department of Biophysics and *Department of Physics
King's College London

Summary

We have tested the feasibility of a direct measurement of migration of proteins on DNA. Using the fluorescence photobleaching recovery (FPR) technique, we were able to obtain the diffusion constant of histone H1 in the complex with λ phage DNA. This diffusion constant, D , is $\sim 1.8 \times 10^{-8} \text{ cm}^2/\text{s}$ at 20°C , about 50% larger than the diffusion constant of the DNA itself. The results will be used in support of a new SERC grant application to examine the motion of linker histones on DNA in greater detail, and also a range of other DNA binding proteins of different structure and function.

Introduction

In this report we summarize results of experiments between 1 August - 16 November, 1987, carried out with an argon ion laser from the Laser Support Facility Loan Pool. The purpose of the experiments was to test the feasibility of a direct measurement of "facilitated diffusion" in ligand-substrate systems. During this time we were able to demonstrate the operation of facilitated diffusion in the interaction between the linker histone, H1, and λ phage DNA.

Method and Measurement

(a) Instrument and Measurement

Figure B2.10 shows a schematic diagram of the apparatus. The laser beam (Spectra Physics 2020) is transmitted through beam splitters, BS1 and BS2. A chopper, CH, switches the laser intensity between bleaching level and monitor level. With CH closed the monitoring beam passes

through BS2 and is weaker by a factor of $\approx 10^4$ than the bleaching beam with CH open. The beams are directed by mirror M into a dichroic mirror DM. Before DM a thin glass plate GP and a lens L1 are added. The DM reflects the beam through a microscope objective L2 (N.A. = 1.25, $\times 100$), but allows only the sample fluorescence to strike a photomultiplier (PM). A barrier filter BF is used along with DM to block the back-scattered and reflected light.

The fluorescence photobleaching recovery (FPR) experiment was performed with a laser wavelength of 488 nm. The temperature of the solution was controlled by a metal sample holder through which water runs from a thermostat. The photomultiplier output is connected to a Malvern photon correlator with 24 channels, operated in its multi-scaling mode. A photodiode PD produces a trigger for the multiscaler from the bleaching beam, reflected by GP.

Fluorescein isothiocyanate (FITC)-labelled calf thymus histone H1 and FITC-labelled bovine serum albumin (BSA) were used for measurements of diffusion constant by the use of the FPR technique. The fluorescence recovery signal of the H1 - λ DNA complex was obtained by multiscaling of 2 - 10 shots.

(b) Analysis of FPR signals

The degree of recovery of the fluorescence intensity was examined by measuring the number of photons per second from sample solutions before and after bleaching. We observed usually more than 90% recovery of fluorescence.

The time course of the fluorescence recovery was non-exponential, as described by Axelrod et al (1976). Following these authors, the relaxation time τ of the diffusion process was estimated at the point where 50% of fluorescence is recovered.

The diffusion constant D was evaluated from the relation

$$D = w^2 / (4\tau) \quad (1)$$

where w is the effective radius of the focused laser beam in the sample solution.

(c) Estimation of "effective" radius of the focused beam.

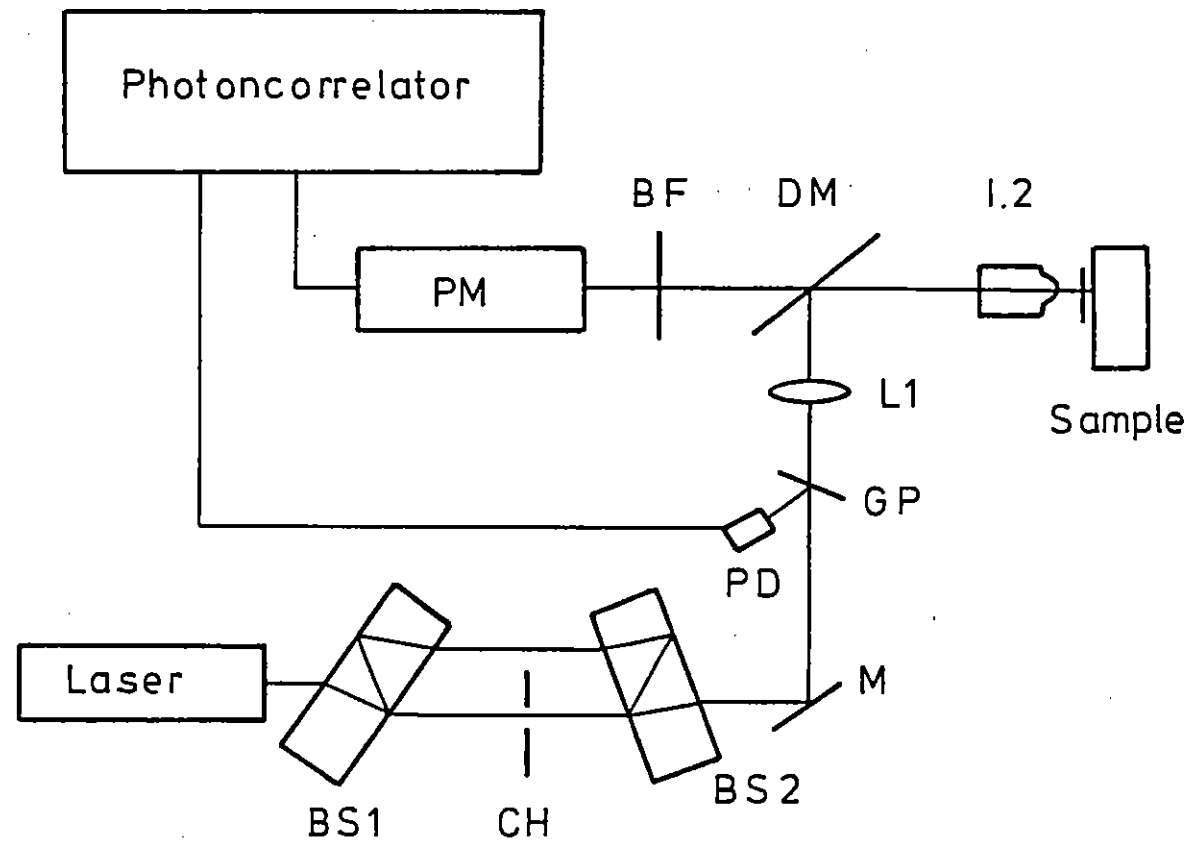


Fig. B2.10 Schematic diagram of the apparatus. The symbols used are
 BF: barrier filter, BS: beam splitter, CH: chopper,
 DM: dichroic mirror, GP: glass plate, L: lens, M: mirror,
 PD: photo diode, PM: photomultiplier.

Since there are some technical difficulties with respect to the FPR calculation due, to the divergence from a sharp waist of the laser beam focused in the sample solution by the use of a microscope lens with high numerical aperture, an "effective radius" of the beam traversing the sample has been evaluated in the following way. Using fluoresce isothiocyanate (FITC) and FITC-labelled BSA, more than ten FPR measurements were performed so as to obtain an accurate relaxation time τ . Then the calculated value τ was inserted into eq. (1) and the value of w was determined using the known values of D for both FITC and BSA. This procedure posed no problems and proved to give a reliable calibration for our data.

RESULTS

(a) Diffusion constants

The translational diffusion constant D for solutions of samples of FITC, FITC-labelled BSA and FITC-labelled histone H1 was measured by the FPR technique at 20°C. An example of the time course of the fluorescence recovery for H1 is shown in Figure B2.11. The results are summarized in Table 1. The result for H1 is in agreement with that expected from its molecular weight

Table 1

Diffusion Constant, D , of the Sample at 20°C in 40 mM NaCl

Sample	Molecular weight	D (cm ² /s)
FITC	~ 400	~ 1.0×10^{-6}
Histne H1	~ 21,000	~ 9.0×10^{-7}
BSA	~ 66,000	~ 6.5×10^{-7}

(b) Temperature dependence of D of H1

The dependence of D of H1 on temperature was examined at four different temperatures. The value of D increased from ~ 5.0×10^{-7} cm²/s at 10°

to ~ 9.0×10^{-7} cm²/s at 25°C. The corresponding energy barrier of the diffusion was ~ 17 kJ/mol. This value is in the expected range for aqueous systems. Therefore, the FPR signals can be ascribed to the diffusion process, and not to any convectional flow which might have been caused by the energy input of the laser beam (~ 30 mW). Thus, this result indicates that we can indeed measure diffusion constants with our FPR apparatus.

(c) DNA concentration dependence of D of the H1-DNA complex

A small amount of the labelled H1 was mixed with a large excess of lambda DNA so as to fulfill the condition that less than 7% of the DNA surfaces should be covered by bound H1. This should provide enough space for H1 to slide along the DNA chain. The FPR measurement was carried out at 20°C and in 40 mM NaCl. An example of the FPR signal is shown in Figure B2.12. The value of $D = 1.8 \times 10^{-8}$ cm²/s is about 50% larger than the diffusion constant of the DNA itself (~ 1.3×10^{-8} cm²/s). This result suggests that the movement of H1 along the DNA surface is largely governed by a hydrodynamic frictional barrier (see Discussion).

The dependence of D on DNA concentration was measured over the range of 5-40 µg/ml DNA. The value of D was practically independent of the DNA concentration. This result suggests that transfer of H1 between two DNA molecules may be neglected, or at least, a significant facilitation of H1 migration by an intermolecular ping-pong type transfer mechanism can be excluded. The results, moreover, suggest that the intersegmental transfer (within a single DNA molecule) of H1 can also be neglected, because the persistence length of DNA at 40 mM NaCl is quite large.

Discussion

The binding of H1 to DNA is essentially electrostatic and driven by displacement of 5 - 6 counterions (Watanabe, 1986). The positive charge of H1 is substantially delocalized relative to the DNA phosphates. In addition, no net charge alteration takes place, while H1 is sliding with a steady velocity v . As observed in the present work, there is no DNA concentration dependence of D for the H1-DNA complex, implying that the contribution of intersegmental transfer to D is negligible. Therefore, H1 should slide on an isopotential surface of the DNA cylinder with the hydrodynamically-limited one-dimensional

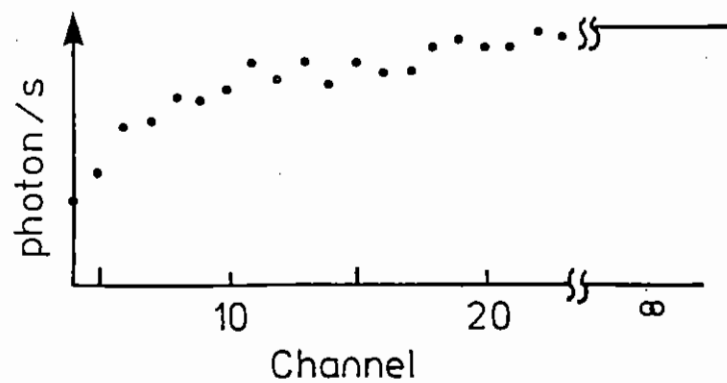


Fig. B2.11 The FPR signal for H1 of 30 ug/ml in 20 mM Tris-HCl of pH 7.0 at 20°C. 40 msec/bin.

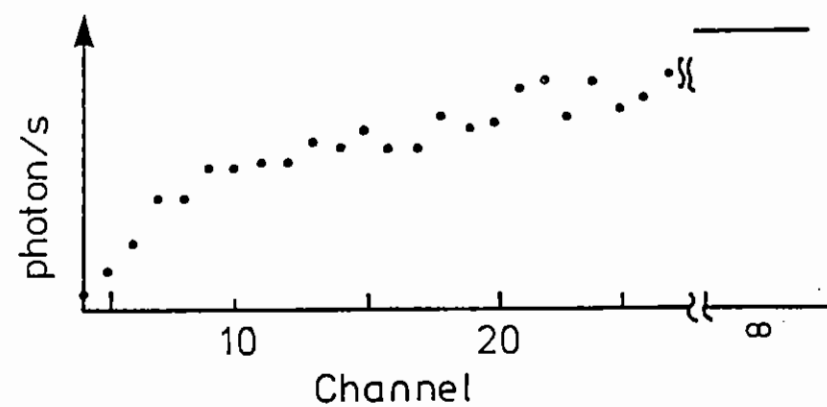


Fig. B2.12 The FPR signal for H1-DNA complex: 2 ug/ml H1 and 20 ug/ml DNA in 40 mM NaCl and 20 mM Tris-HCl of pH 7.0 at 20°C. 1 sec/bin.

diffusion constant D_1 .

The theoretical value of D_1 was calculated assuming that H1 moves along the spiral array of phosphate charges of the DNA helix of 3.4 nm per turn (Watanabe, 1986). Then the rate of energy dissipation for the translational movement and the rotational movement was calculated using Stokes Law. The effective frictional coefficient f for the sliding is given by the relation

$$f = 6\pi\eta r[1 + (4/3)(2\pi)^2(r/(3.4 \times 10^{-7}))^2] \quad (2)$$

where η and r are the solvent viscosity and the Stokes radius of H1 ($r \approx 2.4$ nm), respectively. Since the one- and three- dimensional diffusion constant D_1 and D_3 are given by $D_1 = kT/f$ and $D_3 = kT/(6\pi\eta r)$, respectively, the theoretical one-dimensional diffusion constant is $D_1 = D_3(6\pi\eta r)/f \approx 3 \times 10^{-8}$ cm²/s. The present result, i.e. $D_1 = 1.8 \times 10^{-8}$ cm²/s, is satisfactorily close to this theoretical calculation.

Although a series of control experiments will be needed to confirm the results, they appear at least highly promising.

Conclusion

This study has demonstrated that a direct measurement of one-dimensional diffusion is possible by the FPR technique. Moreover, for biological applications, it should be possible to operate with a minute amount of protein, as low as ≈ 10 ng. Therefore, using our apparatus, even as it stands, we can examine the velocity of migration of protein over the surface of DNA or nucleoprotein substrates such as chromatin.

References

1. D Axelrod, D E Koppel, J Schjlessinger, E Elson and W W Webb, Biophys. J. 16, 1055, (1976).
2. F Watanabe, Nucl. Acids Res. 14, 3573, (1986).

B2-6 MEASUREMENT OF INTERNAL MOTION IN PROTEINS; TIME-RESOLVED FLUORESCENCE STUDIES OF LACTATE DEHYDROGENASE USING A STREAK CAMERA

A Waldman, A R Clarke, J J Holbrook - Bristol University
A J Langley, M Coulter, D Sansom, W T Toner - RAL

Introduction

Internal mobility is known to be important in the mediation of biological specificity and catalysis by the enzyme lactate dehydrogenase (LDH) which undergoes a number of conformational rearrangements during turnover (1,2).

The intrinsic fluorescence from tryptophan residues provides a good probe of local internal mobility in proteins. By use of genetic engineering techniques, we are now able to insert tryptophans at any locus in the polypeptide chain and can hence measure mobility and environment at selected positions in the three dimensional structure of the enzyme subunit (3). This allows the study of both local conformational changes which take place in the enzyme during catalytic turnover and the pathways by which the polypeptide chain folds to form an active enzyme in vivo.

Method and results

A streak camera is used to measure the decay in fluorescence emission anisotropy following a single pulse of laser excitation radiation of 5ps duration.

Figure B2.13 shows the fluorescence decay from PPO measured following a single pulse of exciting light at 295nm. The observed excited-state lifetime is in good agreement with the published value.

Figure B2.14 shows fluorescence decays of tryptophan fluorescence in the enzyme Lactate Dehydrogenase (LDH) from the thermophilic bacterium, *B. stearothermophilus*. This is compared with a mutant LDH derived from this in which two of the three naturally occurring tryptophans have

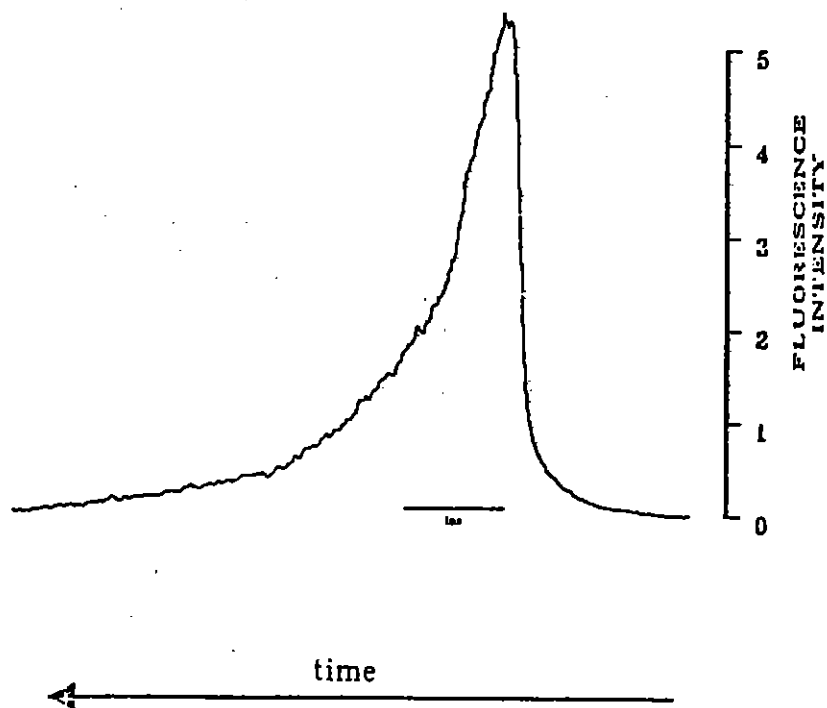


Fig B2.13 Single-shot Fluorescence Decay of PPO.

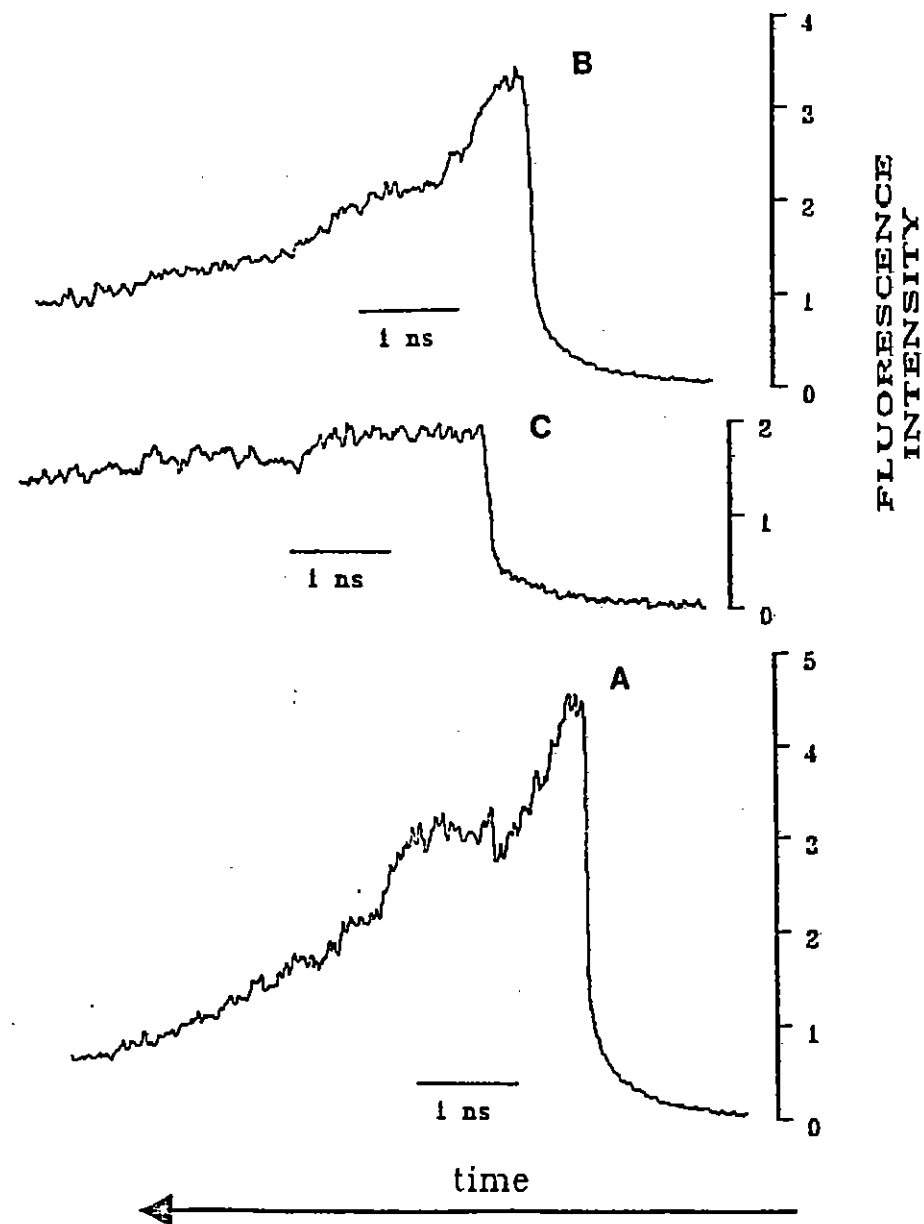


Fig B2.14 Single-shot Measurements of Tryptophan Lifetimes
(A) *N*-acetyl tryptophanamide ($30\mu\text{M}$), (B) *B. stearothermophilus* LDH ($30\mu\text{M}$ sites), (C) Mutant LDH containing a single tryptophan residue at position 203 ($30\mu\text{M}$ sites)

All measurements were made at pH 6 in 100mM triethanolamine HCl buffer. 247
amplitudes of the decay curves are not comparable. Note: the systematic distortions in the decay curves are due to spatial variations in sensitivity in the streak camera and can be corrected using calibration data.

been removed by genetic engineering and with a small analogue, N-acetyl tryptophanamide (NATA). The fluorescence characteristics of the LDH's have been the subject of previous study (4) and hence provide a suitable biological test system.

NATA shows a single exponential decay with a lifetime of 3ns which is in agreement with the published data.

The wild-type LDH (which contains three tryptophans/subunit) shows a bi-exponential decay with one component of similar lifetime to that seen in NATA and a long-lived component which has decayed little by the end of the streak. This is consistent with previous single photon counting studies which showed two major lifetime classes of 3ns and 7.5ns (4).

In the case of the single tryptophan (TRP-203) mutant, the observed fluorescence intensity decreases very little over the duration of the streak which is consistent with the unusually long lifetime of TRP-203 (7.4ns).

Figure B2.15 shows the decay from the enzyme yeast Phosphoglycerate Kinase (PGK) which contains two tryptophans. This is a weak emitter whose major lifetime component is 0.5ns but also has a small amplitude emission with a lifetime of 1.5ns. The short lifetime is readily observed even in a single-shot experiment but, although there is evidence of a longer one, this part of the signal is too noisy to measure with certainty.

In the case of PPO, averaging just 10 shots will yield signal to noise ratios comparable with those obtained from time-correlated single photon counting measurements but with data collection times of less than 5s. By virtue of having much lower quantum efficiencies (typically 5-10%), the biological samples show far noisier decays than those seen with PPO. However, even in the case of the weakest emitter studied here (yeast PGK), the signal to noise is better than 10:1 for a single-shot measurement. Signal averaging (10-100 shots) will yield data of sufficient quality to allow the accurate determination of lifetimes in less than one minute.

Figure B2.16 shows the decays in the parallel and perpendicular

components of emitted fluorescence for PPO, NATA and the single tryptophan LDH mutant described above. The polarised components were measured simultaneously by positioning orthogonal polarising filters so that each covered half of the width of the slit onto which the fluorescent image was projected.

PPO and NATA both show a rapid loss in emission anisotropy as evidenced by the coalescence of the parallel and perpendicular fluorescence components. This is consistent with the rapid isotropic rotational diffusion of a small molecule in solution. The single tryptophan-203 in the mutant LDH enzyme, however, shows no appreciable loss in emission anisotropy over the timescale of the streak; this is consistent with the tryptophan being held rigidly within the protein matrix and only showing the same tumbling rate as the whole enzyme tetramer (τ_c 90ns).

Figure B2.17 shows the decay from PPO measured using a faster streak speed; here, the rate of the decay in anisotropy is more easily observed. This rate is consistent with the very short rotational correlation time reported for this compound (40-60ps).

The data shown here are uncorrected for system distortions. Programmes and calibration data for this purpose have been developed and a more complete analysis is in progress.

Conclusion

The detection system has now been developed and characterised to a level at which corrected, sub-nanosecond anisotropy decay measurements can be made on biological samples in 1-500 shots (ie 0.1-50 seconds).

This will allow measurements of local mobilities in intermediates which exist only transiently in catalytic turnover and protein folding pathways.

References

1. Clarke A R, Waldman A D B, Hart K W, and Holbrook J J (1985) *Biochem Biophys Acta* **829**, 397-407.

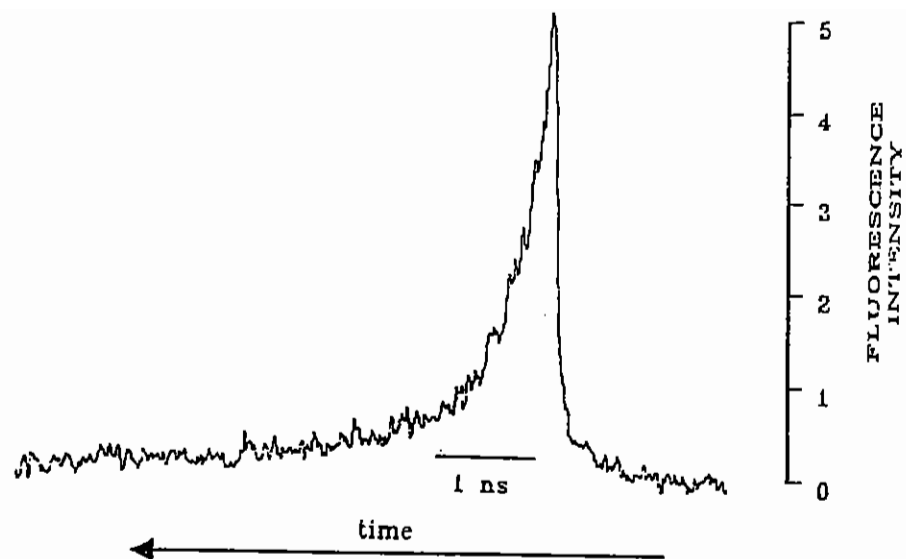


Fig B2.15 Fluorescence Decay of Yeast Phosphoglycerate Kinase. PGK (50 μ M) in 10mM Tris HCl buffer, pH 7.

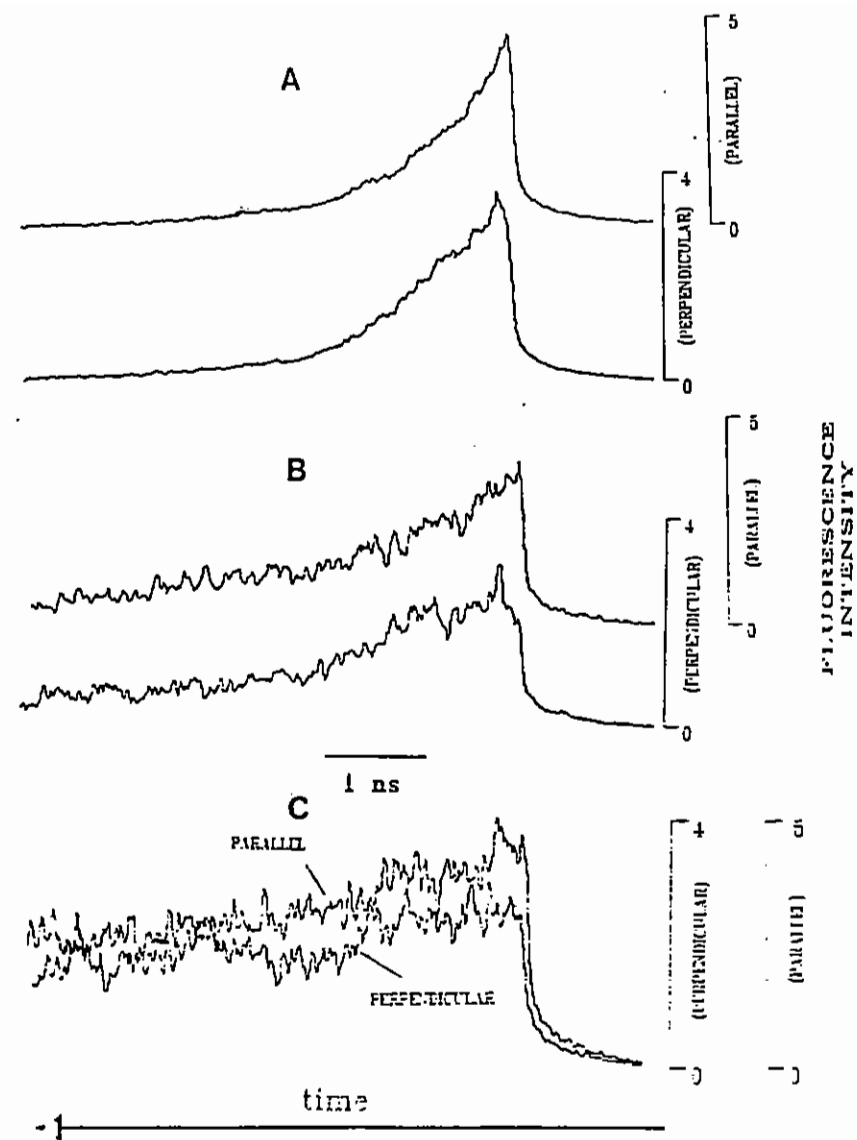


Fig B2.16 Parallel and Perpendicular Components of Fluorescence Showing Emission Anisotropy (A) PPO, (B) NATA and (C) TRP-203. Note that the parallel and perpendicular intensities are not normalised.

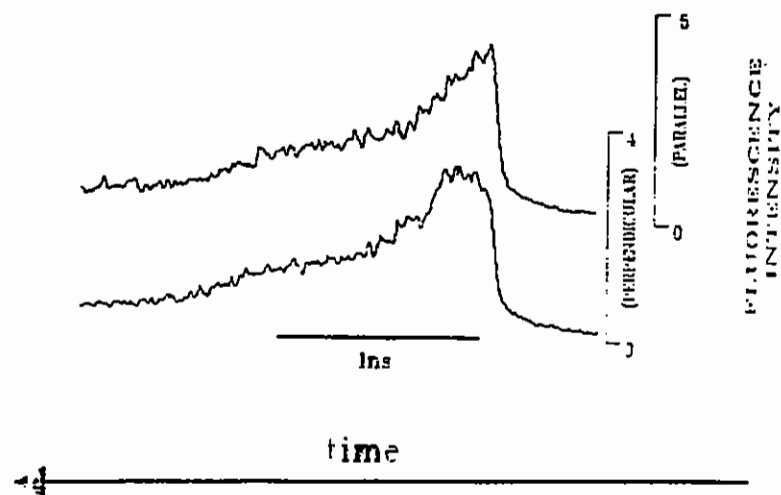


Fig B2.17 Parallel and Perpendicular Emission Components of PPO Fluorescence on Short Timescale.

2. Waldman A D B, Birdsall B, Roberts G C K and Holbrook J J (1986) Biochem Biophys Acta, 870, 102-111.
3. Waldman A D B, Hart K W, Clarke A R, Wigley D B, Barstow D A, Atkinson T, Chia W N and Holbrook J J (1988) Biochem Biophys Res Comm 150, 752-759.
4. Waldman A D B, Clarke A R, Wigley D B, Hart K W, Chia W N, Barstow D A, Atkinson T, Munro I and Holbrook J J (1987) Biochem Biophys Acta 913 66-71.

B3	PHYSICS	Pages
B3-1	Resonant Excitation of the GR Centre in Diamond	221 - 222
B3-2	Investigation of Thallium-Xenon Exciplex Emission	222 - 226
B3-3	Optoacoustic Investigation of Cr ³⁺ Doped Laser Materials using a PVdF Piezoelectric Detector	226 - 233
B3-4	The Use of KrF Laser-Generated as X-ray Sources for Scanning Microscopy: A Preliminary Study	233
B3-5	Multiphoton Multiple Ionisation of Aliphatic Hydrocarbons	234 - 236
B3-6	Degenerate Four Wave Mixing in Semiconductor-Doped Glass	236 - 239
B3-7	He II Balmer Line Shifts in a Dense Z-pinch Plasma	239 - 243
B3-8	In-situ Studies of MOVPE Processes using Optical Second Harmonic Generation	244 - 248

Editors: A Parker/A J Langley/A Tavender

B3-1 RESONANT EXCITATION OF THE GR CENTRE IN DIAMOND

A T Collins*, J Szechi* and S Tavender*

* Wheatstone Physics Laboratory, King's College, Strand, London
WC2R 2LS

* Laser Support Facility, Rutherford Appleton Laboratory, Chilton,
Didcot, Oxfordshire OX11 0QX

Photoluminescence in the GR1 zero-phonon region has been examined in detail, using a tunable dye laser resonant with the higher excited states GR2 to GR8. We conclude that the temperature-dependent structure observed when electron-beam excitation is used is not due to perturbation of the centre resulting from the fast non-radiative relaxation from the higher levels to the first excited state.

This work has been carried out to test the proposal (Collins 1987) that temperature-dependent structure observed in the zero-phonon region of the GR+ cathodoluminescence spectrum results from perturbation of the GR centre following the rapid non-radiative relaxation from higher excited states.

Vacancies in the neutral and negative charged state are produced in diamond by primary radiation damage. The neutral vacancy predominates in natural semiconducting diamond (type IIB diamond) because of the proximity of the Fermi level to the valence band (Davies 1977). The neutral vacancy gives rise to the GR1 vibronic band with a major zero-phonon line at 1.673 eV and a weaker line at 1.665 eV. The zero-phonon lines are particularly sharp in natural type IIB diamonds because of the very low impurity concentration, and consequently very small amount of strain broadening. In addition to the GR1 lines, a number of zero-phonon lines due to transitions to higher energy states (GR2 to GR8) are observed in the absorption spectrum at energies between 2.88 and 3.01 eV (Walker et al 1974).

Absorption of light in the GR1 vibronic band produces GR1 luminescence. However, no luminescence is observed which can be identified with transitions from the higher excited states to the ground state.

Instead, following the absorption of light in the GR2 to GR8 region, the centre relaxes non-radiatively to the first excited level, then decays, producing GR1 luminescence (Collins 1981). GR1 luminescence may also be generated using electron beam excitation. In this case, however, there is additional temperature-dependent structure in the zero-phonon region which is not observed when the centre is excited by the absorption of light in the GR1 vibronic band (Collins 1978). Furthermore the photoluminescence decay time of 2.55 ns at low temperature (Davies et al 1987) is somewhat shorter than the 3.1 ns determined for the decay of the cathodoluminescence (Collins 1987). The latter value is for the main zero-phonon line; the additional structure present in the zero-phonon region of the cathodoluminescence spectrum has a complex luminescence decay behaviour (Collins 1987).

The precise mechanism by which the generation of electron-hole pairs leads to excitation of vibronic centres is not known. It has been suggested (Collins 1987) that the additional structure observed in the cathodoluminescence spectrum may arise because the centre decays from the highest excited levels. In qualitative terms we envisage that the diffuse orbitals of the GR2 to GR8 levels interact with the exciton, and the excited centre then decays via the GR1 level, as indicated above. The rapid non-radiative relaxations from the higher excited states produce (in this model) some perturbed centres, leading to a splitting of the first excited state into a thermalising triplet (Collins 1978). To test this hypothesis we have measured the luminescence in the GR1 zero-phonon region following photoexcitation of the diamond in the GR2 to GR8 region.

In order to study the shape of the zero-phonon emission it is important that the diamond has relatively weak GR1 absorption, to avoid distortions caused by self-absorption. Absorption by the GR2 to GR8 lines is then very weak indeed, and to generate sufficient GR1 luminescence by the absorption of monochromatic light in this region the only practicable approach is to use an intense laser. We have used a Lambda Physik FL3002 tunable dye laser pumped by a Lumonics Hyperex-400 excimer laser. The gas fill in the excimer laser was XeCl, and Stilbene 3 was used in the dye laser.

The diamond, a polished plate of natural semiconducting diamond measuring 2 x 1 x 0.5 mm³, was mounted in indium on the end of a liquid cold finger. A small concentration of GR1 centres had been produced in this diamond by irradiation with 10²¹m⁻² 2 MeV electrons, using the van

de Graaff accelerator at Reading University. This irradiation was followed by a 12 hour anneal at 500°C to ensure that the zero-phonon lines were as sharp as possible. Luminescence generated by the laser was collected by a lens and focused through a deep red filter on to the entrance slit of a Spex 1700 monochromator, fitted with a 1200 groove/mm grating at 500 nm and an RCA 31034A photomultiplier.

The laser was operated at a pulse repetition frequency of 99 Hz, and the signal from the photomultiplier was fed to a DC amplifier with an integrating capacitor at the input.

Figure B3.1 shows the intensity of the luminescence at 1.673 eV (the main GR1 zero-phonon line) as the laser was scanned from 410 to 435 nm. All of the GR2 to GR8 lines are clearly seen in this excitation spectrum, superimposed on the envelope of the Stilbene dye. It is straightforward, therefore, to set the laser, in turn, at the wavelength corresponding to each of the excited states and to measure the luminescence produced in the GR1 zero-phonon region. A typical spectrum for excitation into the GR2 level is shown in Figure B3.2. There is no difference between this spectrum and the photoluminescence produced by excitation at 632.8 nm near the peak of the GR1 absorption band. This measurement was repeated for all of the excited states, and also for excitation at a number of points in the underlying continuum. The shapes of the spectra were the same in all cases, and none of the additional structure observed in cathodoluminescence was detected.

The work reported in this study therefore shows that the additional temperature-dependent structure observed in the zero-phonon region of the GR1 cathodoluminescence spectrum is not simply due to the non-radiative decay from a number of higher excited states to the first excited level. This negative result does make a positive contribution to the study of the vacancy in diamond in that it rules out the possible explanation put forward earlier (Collins 1987) to account for the differences in the GR1 luminescence spectra when using direct or indirect excitation. The origin of these differences remains unresolved, however.

References

1. A T Collins, J. Phys. C: Solid State Phys. 11 2453-63 (1978)
2. A T Collins, J. Phys. C: Solid State Phys. 14 289-94 (1981)

3. A T Collins, J. Phys. C: Solid State Phys. 20 2027-33 (1987)
4. G Davies, Nature 269 498-500 (1977)
5. G Davies, M F Thomaz, M H Nazaré, M H Martin and D Shaw, J. Phys. C: Solid State Phys. 20 L13-17
6. J Walker, L A Vermeulen and C D Clark, Proc. R. Soc. A 341 253-66

B3-2 INVESTIGATION OF THALLIUM-XENON EXCIPLEX EMISSION

H F J Cormican, M J McAllister, C S Madden, R G C Thomson, Dr I Duncan, Dr T Morrow (QUB).

In previous work at the Laser Support Facility, the 7S_{1/2} quasi-molecular or exciplex state of Thallium-Xenon (Tl-Xe) was studied⁽¹⁾. Viable exciplex laser action from this state is made difficult by the extreme temperatures and pressures needed. The stringent physical conditions could be relaxed if a more strongly bound exciplex system could be found. According to Drummond et al⁽²⁾, the 6D states of Thallium with Xenon were expected to form such a system, and therefore these states were the subject of this experimental investigation.

The experiment consisted of two parts. First, a fluorescence study of the 6D exciplex system was carried out to determine the shape of the potential energy curve and the binding energy. Secondly, the laser potential of this state was explored. The various wavelengths and energies of the Tl-Xe system are shown below in Table 1.

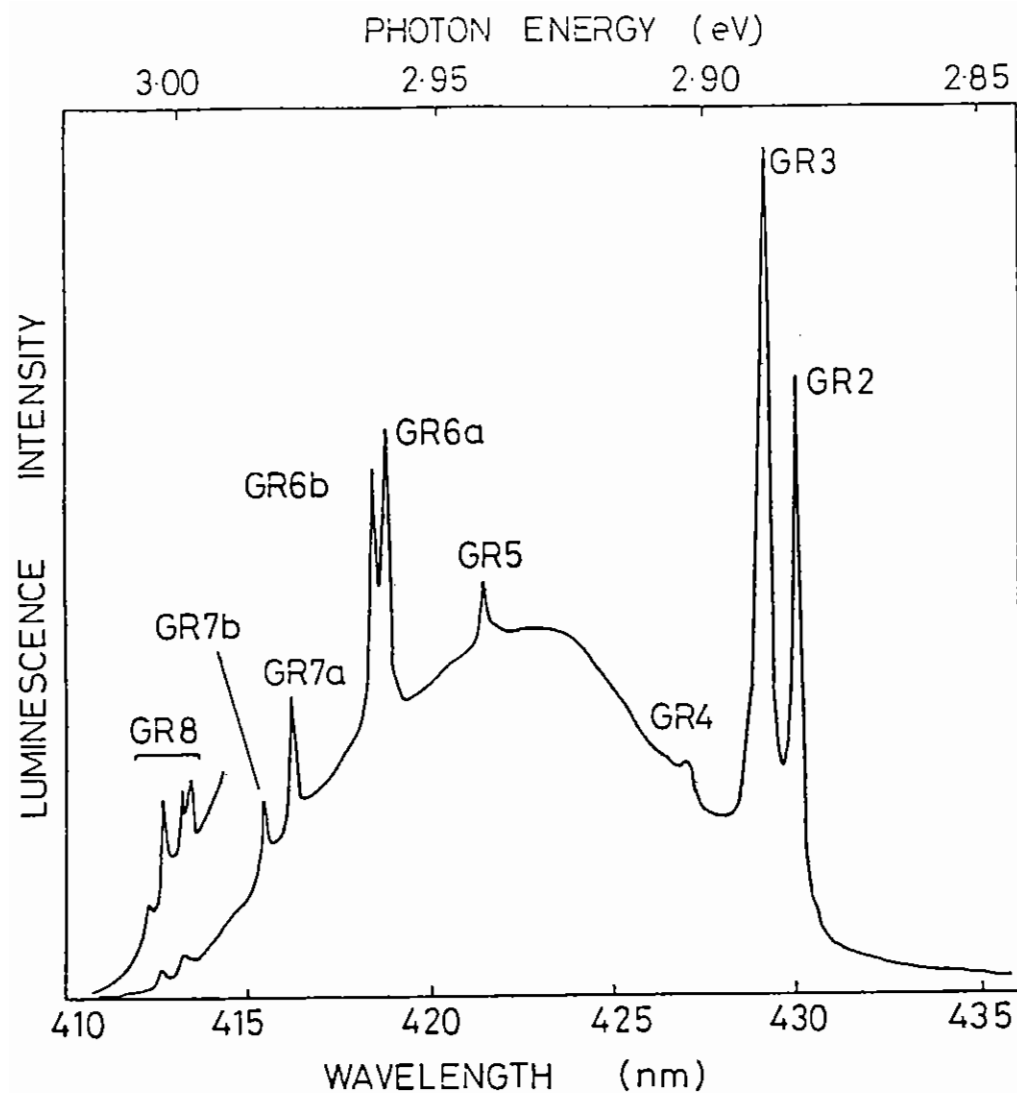


Fig. B3.1 Luminescence intensity of the GR1 zero-phonon line (1.673 eV) shown as a function of the wavelength of the tunable dye laser. The sample temperature was 77 K. All of the GR2 to GR8 excited states are clearly seen, superimposed on the envelope of the Stilbene dye. The full spectrum was recorded with one attenuator between the pump laser and the dye laser. Measurements in the GR8 region were repeated with this attenuator removed.

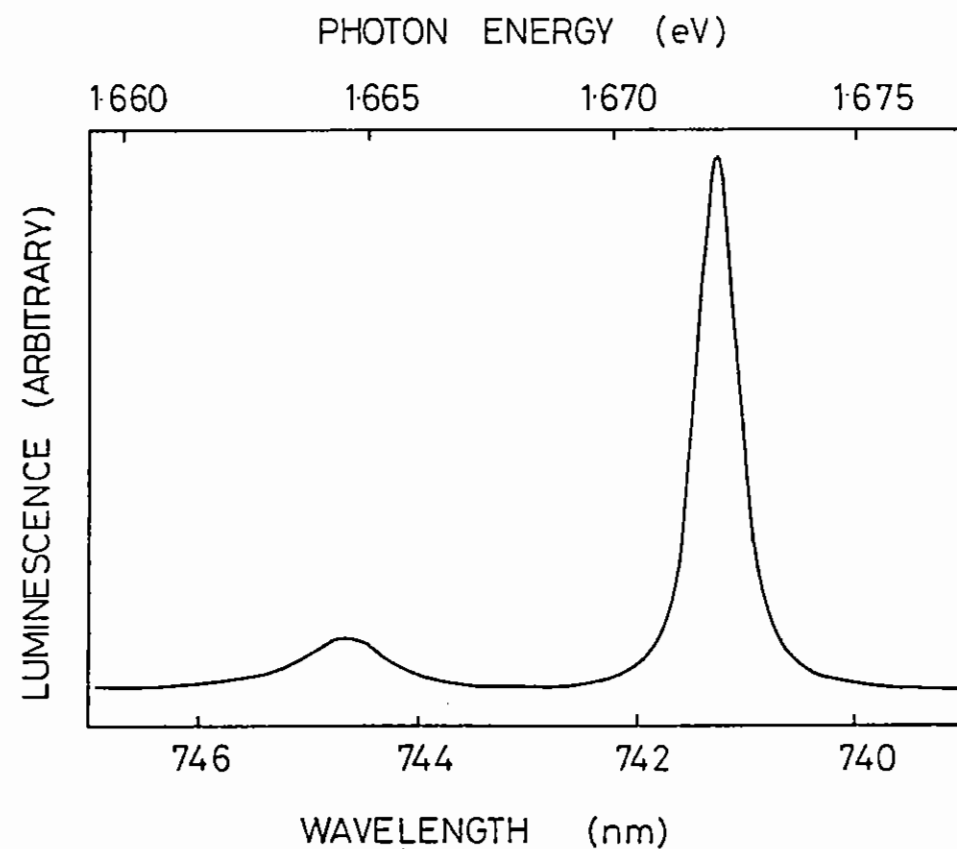


Fig. B3.2 Photoluminescence spectrum in the GR1 zero-phonon region produced by excitation into the GR2 excited state. The sample temperature was 77 K.

TABLE 1

Thallium state	Exciplex state	Transition	Wavelength/nm
$6P_{1/2}$	$X_{1/2}$	$6P_{1/2}-7S_{1/2}$	377.6
		$6P_{1/2}-6D_{3/2}$	276.8
$6P_{3/2}$	$X_{3/2}$	$6P_{3/2}-7S_{1/2}$	535
		$6P_{3/2}-6D_{3/2}$	353
	$A_{1/2}$	$6P_{3/2}-6D_{5/2}$	351
$7S_{1/2}$	$B\Sigma_{1/2}$		
$6D_{3/2,5/2}$	unknown		

Section One

The fluorescence experiments supplemented studies carried out at Queens using a similar experimental layout (Figure B3.3). The change in fluorescence or fluorescence excitation as a function of temperature can be used to derive the potential energy (P.E.) curves of the excited states⁽³⁾ and, from the potential energy curves, the gain and absorption of the exciplex bands can be calculated. The Tl-Xe was formed by photodissociation of a Thallium halide vapour in an excess of Xenon. Using a suitable combination of Thallium halide and photodissociation laser wavelength, atomic Thallium can be formed in large number densities in the $6P_{1/2}$, $6P_{3/2}$ or $7S_{1/2}$ state. Interaction with Xenon results in the formation of the respective exciplex state (Table 1). The vapour can then be probed with a suitable dye laser to produce higher excited Tl and Tl-Xe states.

The Tl halide was present in small (1 cm square) sealed quartz cells with a previously determined concentration of Xenon atoms. The cells were placed in an oven, controlled by a variac, and left for a suitable period to reach equilibrium. The temperature was measured using a

calibrated thermocouple attached to the cell and fluorescence excitation used to study the exciplexes. In this technique, the integrated emission from the exciplex is measured as the dye laser is tuned over the region of absorption of the exciplex. Examples of the excitation runs are shown below (Figure B3.4). The Apple computer, shown in the experimental layout, controlled the tuning of the dye laser as well as integrating the signal from the diode array. The diode array was attached to a low resolution spectrometer which was set to study exciplex emission well away from any laser emissions. A further detector, a pyroelectric, was used to normalise for any dye laser energy fluctuations.

Although the analysis of the P.E. curves is not completed, the preliminary results show that the $6D$ Tl-Xe exciplex is only weakly bound. Fitting the results to a Morse potential, we find the equilibrium separation, R_e , is approximately 4.0Å and the binding energy is only 800 (± 200) cm^{-1} . A more serious complication also became apparent in the fluorescence study. At higher pressures of Xenon, the $6D$ state of Thallium is quenched into the $7S_{1/2}$ state and this severely limits the effectiveness of the exciplex as a laser.

Section Two

A laser study of the exciplex system used a similar experimental layout except that the Tl was contained in long (14 cm) cells and oven to provide more suitable conditions for lasing. Two sealed cells were used containing Tl chloride and Xenon at pressures of 100 torr and 500 torr respectively. The oven was heated to provide a suitable Tl concentration (0.1 torr) and the cells were arranged to provide for a single pass of the pump dye laser, which was carefully overlapped with the photodissociation laser. The pump laser was focused with a 20 cm lens to obtain a power density of the order of 500 MW/cm^2 . The photodissociation laser was of sufficient energy (typically 50 mJ) to dissociate all the Tl halide present and this was verified by measuring the input and output energy and comparing these against bleaching calculations. The pump laser energy and the resulting laser emission energy, from the Tl, were measured using pyroelectrics. The laser emission was also examined spectrally by the diode array attached to a high resolution spectrometer, the output first being suitably attenuated using a reflection from a flat and a quartz diffuser.

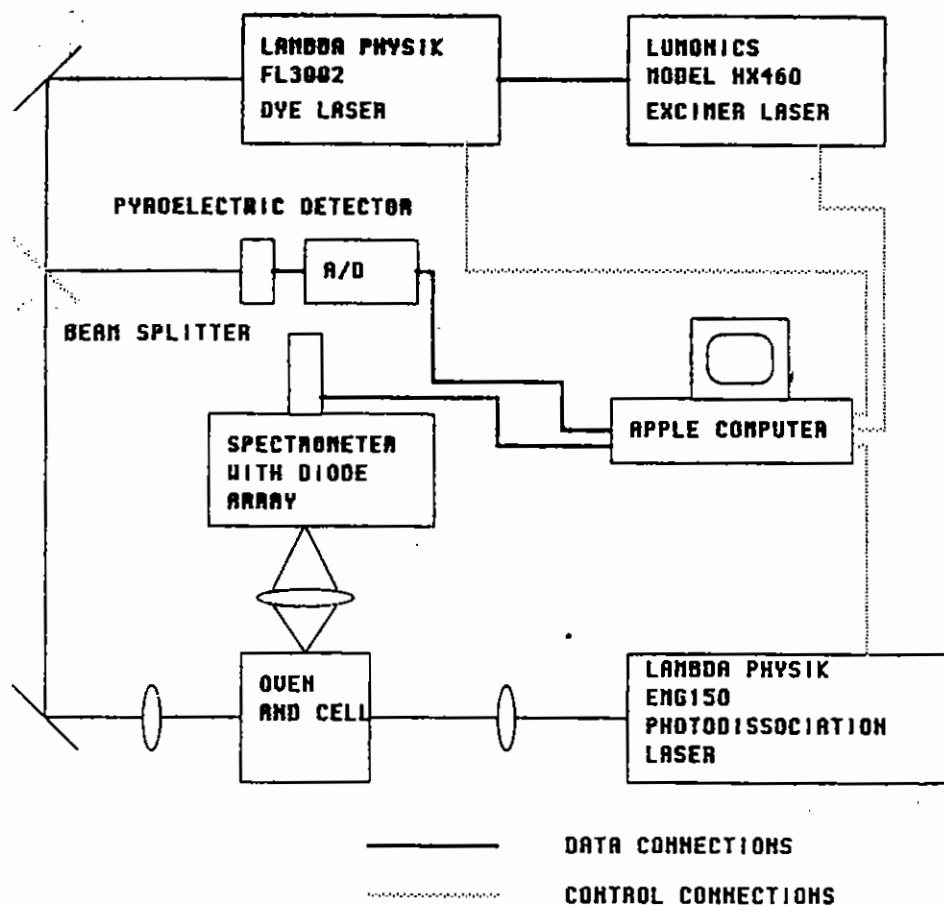


Fig. B3.3 Schematic diagram of experimental layout.

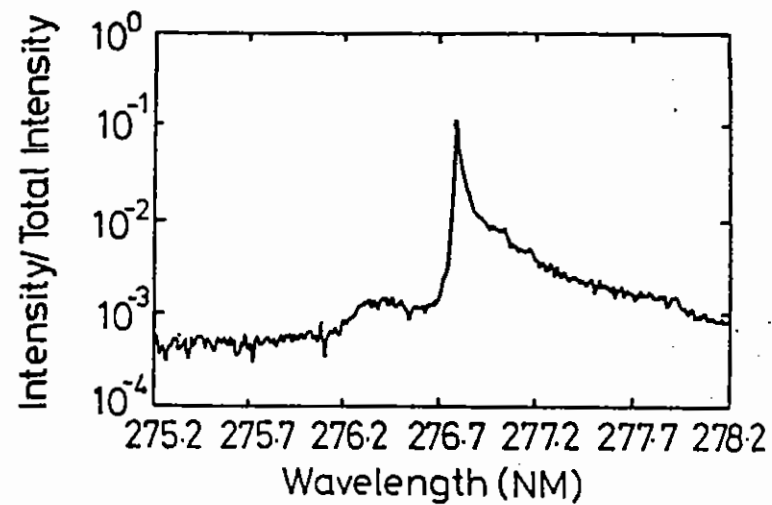


Fig. B3.4(a) 450 torr Xe, 0.02 torr Tl at 580 Kelvin.

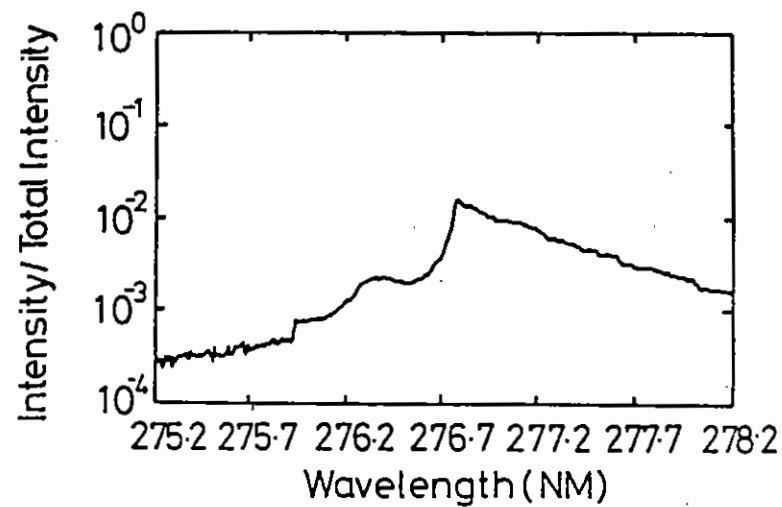


Fig. B3.4(b) 450 torr Xe, 0.1 torr Tl at 780 Kelvin.

Using the low pressure Xe cell, no exciplex emission was observed but, as expected, laser action was obtained on the Stokes and anti-Stokes collisional induced laser lines. The Stokes collisional induced laser was formed by photodissociation at 248 nm which gives mostly $6P_{1/2}$ Tl. The medium was then pumped around the $6D-6P_{1/2}$ resonance transition at 276.8 nm and laser emission from the $6D$ to $6P_{3/2}$ (353 nm) was observed, even when the pump laser was detuned from resonance by several nanometres. The gain from the $6D$ to $6P_{3/2}$ laser, shown in Figure B3.5 is consistent with theoretical expectations. At this low pressure there was already evidence of quenching from the $6D$ state and, at higher pump intensities, laser emission from the $7S_{1/2}$ state to the $6P_{1/2}$ and $6P_{3/2}$ states was also observed.

The anti-Stokes laser was formed by photodissociation at 193 nm, which gives mostly $6P_{3/2}$ Tl, and pumping on the $6D-6P_{3/2}$ (351 and 353 nm) transitions. Output then occurred on the $6D_{3/2}-6P_{1/2}$ and $7S_{1/2}-6P_{1/2}$ transitions, which reveals that by pumping to the higher $6D_{5/2}$ state, Thallium is quenched into the lower $6D_{3/2}$ and $7S_{1/2}$ states. This is probably due to the formation of a common exciplex state for both $6D$ states. Like the Stokes case, lasing occurred after pumping several nanometres either side of both resonance lines.

Experiments using the high pressure cell revealed no exciplex emission and no emission from the $6D$ states. Instead, at higher pump powers, lasing occurred only on the green $7S_{1/2}$ to $6P_{3/2}$ transition. Strong lasing was achieved when pumping around the $6D-6P_{3/2}$ transition (351-353 nm). It was during a quick check of the laser gain of these conditions that an anomalous result became apparent. As exemplified by Figure B3.6, the laser emission output energy has a peculiar dependence on pump energy input. At certain regions on the graph a small increase in pump energy input can result in a considerable laser emission decrease and this happens repeatedly in a cyclic manner. The points are the mean of many experimental determinations and the corresponding error bars represent twice the standard deviation. The anomalous behaviour was repeatable and found to occur both detuned from, and on, resonance, the onset of this behaviour always occurs when the normal linear exponential gain reaches saturation. Attempts are currently under way to explain this phenomena in terms of a combination of amplified spontaneous and superfluorescent emission.

In conclusion, we have shown that, despite earlier reports, the

Thallium $6D$ states do not form a strongly bound exciplex with Xenon. Instead experimental determination of the binding shows it to be only of the order of 800 cm^{-1} and this makes the states much less attractive as a source of exciplex laser emission. Collisional induced lasers were however observed for the first time on these states in the low pressure regime. The results obtained also show some anomalous behaviour in the quenching of the $6D$ state and in the unusual gain curve of green 535 nm emission in the high pressure limit and these results are currently under further investigation.

References

1. D G Cunningham, D Denvir, I Duncan and T Morrow (QUB), Rutherford Appleton Laboratory Laser Division Annual Report RAL-85-047, Section B3-3, (1985).
2. L A Schlie, L E Jusinski, R D Rathge, R A Hamil, and D L Drummond, The Journal of Chemical Physics, 72, No. 8, p 4529, (1980).
3. B Cheron, R Scheps and A Gallagher, The Journal of Chemical Physics, 65, No. 1 p 326, (1976).

B3-3 OPTOACOUSTIC INVESTIGATION OF Cr^{3+} DOPED LASER MATERIALS USING A PVdF PIEZOELECTRIC DETECTOR.

M J McAllister, H F J Cormican, C S Madden, R G C Thomson, I Duncan, T Morrow (Queens University Belfast)

Non-radiative losses in Cr^{3+} doped laser materials have been examined using pulsed optoacoustic spectroscopy⁽¹⁾. This technique involves illuminating the sample with an intense short pulse of monochromatic light. Heat is released into the lattice through non-radiative de-excitation on a very short time scale, resulting in an elastic pressure pulse propagating through the crystal. This pulse can be detected using a piezoelectric transducer placed in contact with the sample surface. The amplitude of the piezoelectric signal is proportional to the amount of non-radiative de-excitation.

In the experiment carried out at the Laser Support Facility we first set out to determine whether the pressure pulse propagating in the crystal could be detected using our home produced transducer design.

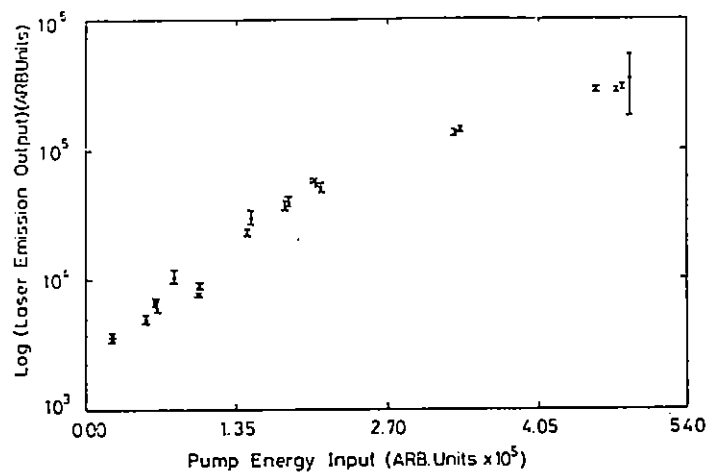


Fig. B3.5 353 nm Laser output vs 276.8 nm pump energy.

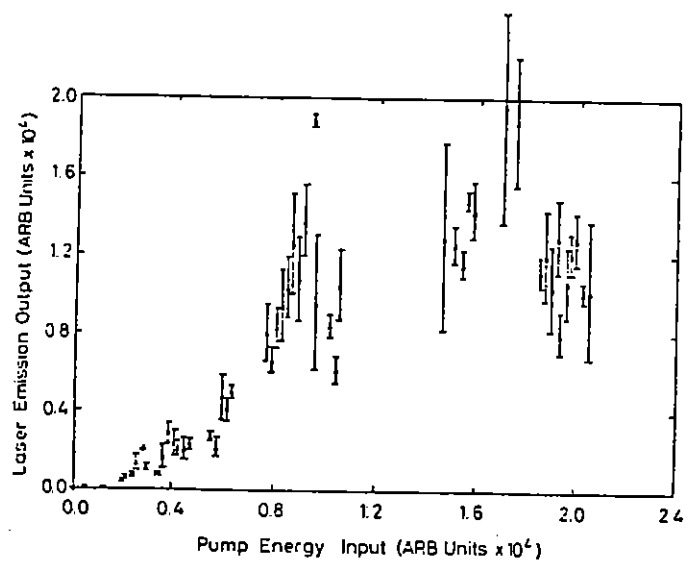


Fig. B3.6 535 nm Laser output vs input pump energy.

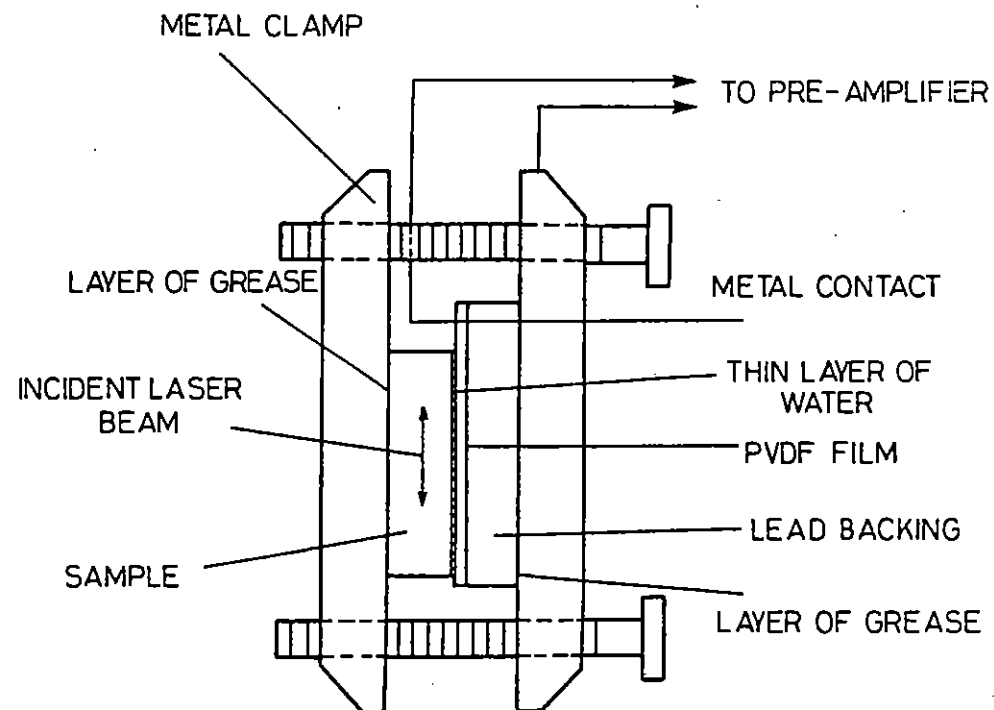


Fig. B3.7 Sample and piezoelectric detector system

We then proceeded to investigate the signals obtained from the Cr^{3+} doped materials.

A blown up view of the transducer and sample combination is shown in Figure B3.7. The excitation source was the excimer pumped dye laser combination (Lumonics HX460 and Lambda-Physik FL3002), which produced excitation wavelengths in the range 420-720 nm by using a number of different dyes. The beam passed through two optical irises placed 1 metre apart with the apertures closed to about 2 mm diameter. These ensured the beam trajectory was maintained for each dye used and were not moved during the experiment. The beam was then focussed to give a vertical line focus using a 10 cm focal length quartz cylindrical lens. The focal plane was positioned about 1 cm from the front surface of the sample. Both the front and back faces of the sample were polished to minimise any scattering and enable an accurate transmission value to be obtained. The transmission was obtained by monitoring the incident and transmitted intensities using two pre-calibrated pyroelectric detectors. To ensure both detectors were operated in the linear response regime, small fractions of the incident and transmitted beams were monitored by using the reflections from a number of quartz flats and the full energies were calculated.

Figure B3.7 shows the construction of the homemade piezoelectric transducer used in these experiments. The heart of the transducer is a 100 micron thick polyvinylidene difluoride (PVdF) film, which has an aluminium coating on each side to which electrical contacts can be made. The PVdF film has a remarkably short risetime and exhibits very little ringing, unlike materials like PZT etc., and, due to recent design advances, it now exhibits a very high sensitivity. The film is glued, using a conducting epoxy adhesive, to a piece of polished lead approximately 3 mm thick. The lead acts as an acoustic 'damper', attenuating any unwanted reflections at the transducer surface. The combination of the PVdF film and the lead backing is then clamped onto the polished surface of the sample using a simple laboratory clamp. Before clamping a small drop of deionised water was placed on the exposed aluminium surface of the film. The water acts as an acoustic coupler, filling the gaps (due to sample surface roughness of approximately 1 micron) at the contact interface. The electrical contact to one side of the transducer was made by a piece of wire glued with a conducting adhesive to the aluminium surface. The contact to the other side was made, via the lead backing and the metal clamp, by attaching a wire to the clamp.

Since the piezoelectric transducer is essentially a capacitor with very high impedance and the signals being measured are usually of the order of microvolts or less, suitable low noise amplification is necessary. This was achieved using a high bandwidth impedance buffer, which was positioned very close to the transducer assembly to minimise electro-magnetic pickup. The signal was further amplified by two wide bandwidth operational amplifiers, the whole assembly being placed inside a small metal box to minimise the effects of interference. The signal was then fed to a boxcar detector and, by suitable adjustment of the width and position of the boxcar gate, the averaged signal values were obtained. The output of the boxcar detector and the two pyroelectric detectors were monitored by an Apple IIe microcomputer, which saved the data to disc for subsequent analysis.

The materials examined using the pulsed optoacoustic technique were aluminium garnet doped with Cr^{3+} (Ruby), with a concentration of approximately 1.7×10^{19} centres per cc, and a material we have been investigating in collaboration with RSRE Malvern, gadolinium scandium gallium garnet doped with Cr^{3+} (GSGG:Cr $^{3+}$). Two samples of this material were investigated, one with a concentration of approximately 1.0×10^{20} centres per cc (GSGG5), and the other with almost twice this concentration (GSGG7). Both these materials exhibit two broad band absorptions in the visible (see Figures B3.10 and B3.11), corresponding to absorption into ${}^4\text{T}_1$ and ${}^4\text{T}_2$ states from the ${}^4\text{A}_2$ ground state. The excitation schemes for both crystals are shown schematically in Figures B3.8 and B3.9.

From these schemes we can see that in the case of Ruby, excitation into either of the excited states is followed by rapid non-radiative de-excitation (approx. 10^{-11} sec.) to the bottom of the ${}^2\text{E}$ level, where there is a mixture of radiative and non-radiative deexcitation to the ground state. We have found the lifetime of this level to be approximately 2.7 milliseconds, therefore any non-radiative energy release from this state will be over a long period of time compared to the length of the laser pulse and as a result not contribute significantly to any fast acoustic wave generated in the crystal. From the argument above, we can see that, provided the laser pulse length is short compared to the lifetime of the radiative state, we should be able to investigate the amount of non-radiative energy released during

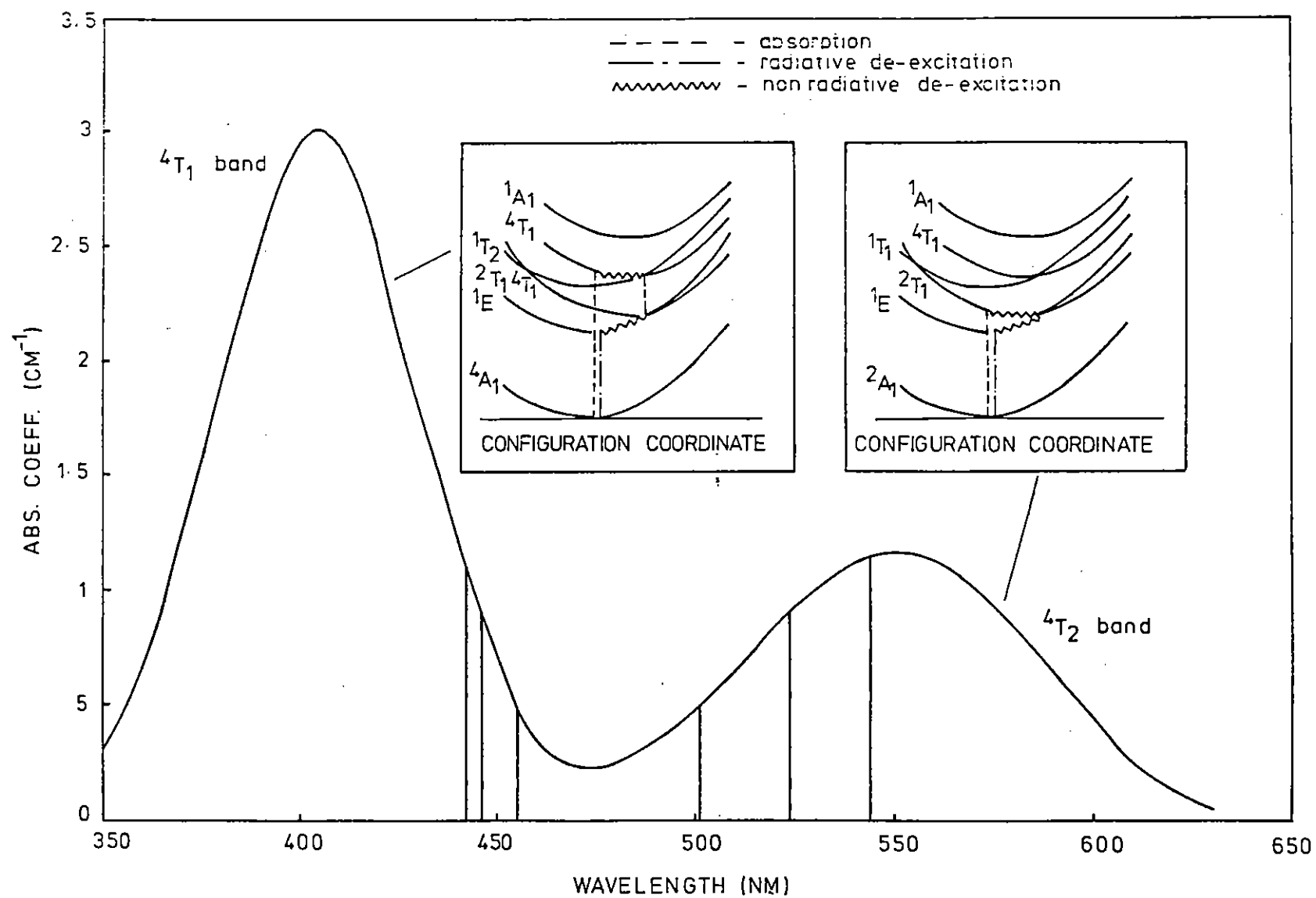


Fig. B3.8 Absorption spectrum and excitation scheme for ruby.

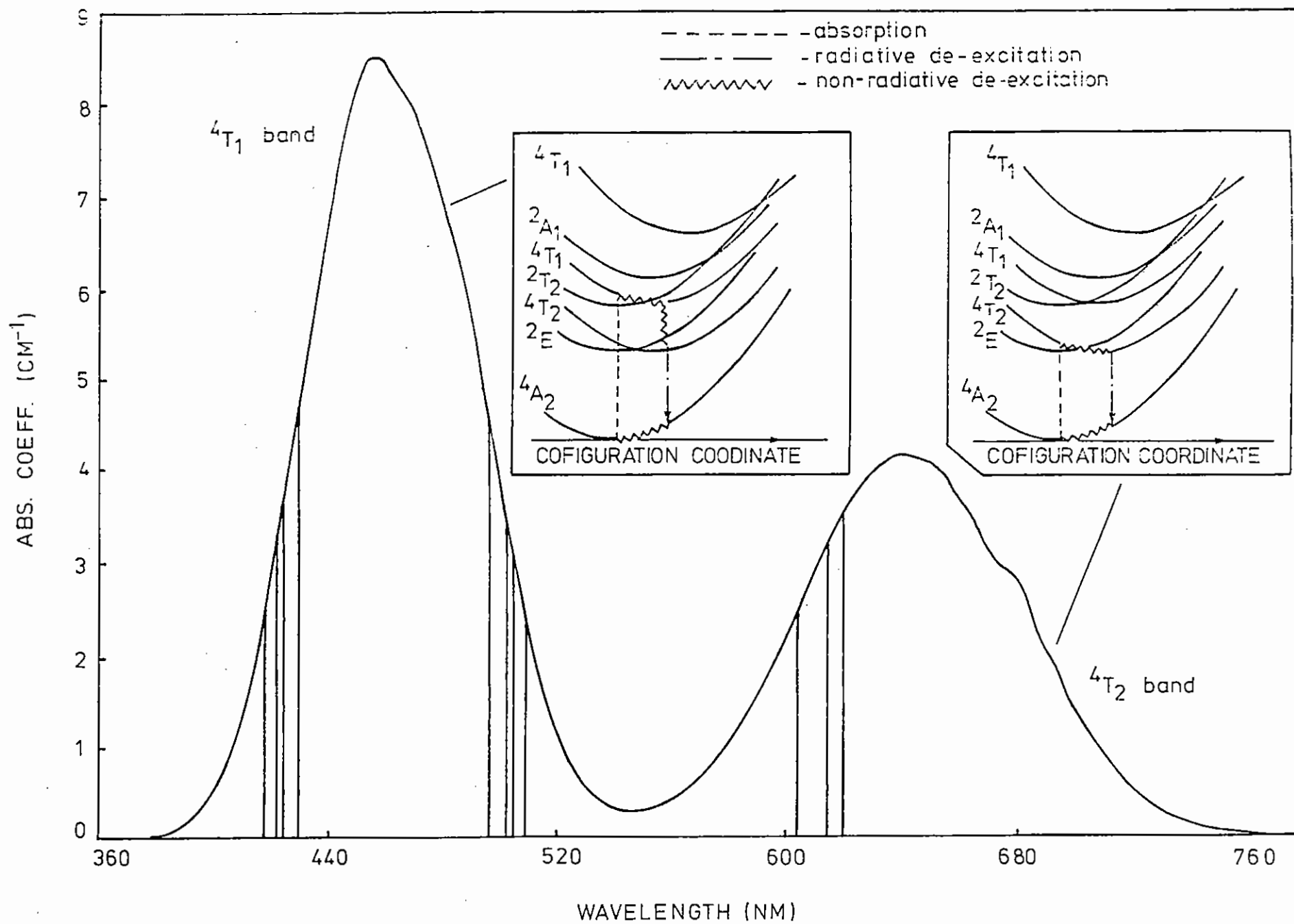


Fig. B3.9 Absorption spectrum and excitation scheme for GSGG:Cr³⁺.

de-excitation to the radiative meta-stable state. By exciting at different wavelengths within an absorption band we can gain some insight into the non-radiative decay processes, and compare this behaviour with that following excitation into a different band.

One problem encountered using this technique is that the amplitude of the pressure wave is dependent on the shape of the absorption profile within the sample. As a result, although the ruby sample was excited at a number of different wavelengths corresponding to different positions within each band, comparisons were only made at those points where the absorption due to the Cr^{3+} ion is the same. The signals obtained at these equi-absorption points were normalised for the incident number of photons at each excitation wavelength, and then the ratios compared with those predicted by the de-excitation scheme described previously. The amount of 'fast' non-radiative energy expected for each photon absorbed is obtained by subtracting the mean fluorescence energy, in this case 1.79 eV, from the incident photon energy. The results are given for three pairs of equi-absorption points in Table 1.

From Table 1, we can see that the agreement between the experimental ratios and those predicted theoretically is quite good, and in all cases lies within the experimental error. In addition, we can also see that as the excitation wavelengths get closer to each other, the difference in fast non-radiative energy decreases accordingly. These results are a good indication of the validity of the assumption that excitation through either band in ruby does result in fast non-radiative transitions to the meta-stable level. More information could be obtained by looking at the other two equi-absorption points on the high energy side of the ${}^4\text{T}_2$ band and the low energy side of the ${}^4\text{T}_2$ band, however time did not permit this to be carried out. Another factor which must be considered when determining the validity of the results is the contribution, if any, of excited state absorption, which would give rise to an enhanced non-radiative heat release. Ruby exhibits quite strong excited state absorption bands⁽²⁾. For instance, the excited state absorption cross-section at 455 nm is almost five times that for ground state absorption. By modelling the absorption process on a computer, the contribution of excited state absorption was found to be only about one per cent of that for ground state absorption

TABLE 1. Piezoelectric (PZE) signals obtained at groups of equi-absorption points in ruby compared with theoretical predictions

Wavelength (nm)	Photon Energy (eV)	Abs. Coeff. (cm^{-1})	PZE Signal (per photon)	Theor. Energy Released (per photon)
442	2.811	1.08	148 ± 15	1.021
544	2.284	1.08	67 ± 7	0.494
446	2.786	0.90	117 ± 12	0.996
524	2.371	0.90	59 ± 6	0.581
455	2.731	0.47	82 ± 8	0.941
501	2.480	0.47	51 ± 5	0.690

for the energies and focussing used in the experiment. However, as a precaution the transmission and signal variation with incident energy were checked at a number of wavelengths, and again no indication of excited state absorption or ground state bleaching was observed.

In the case of GSGG:Cr^{3+} , it is generally thought that excitation into either of the excited states is followed by rapid non-radiative de-excitation, as above, to the bottom of the ${}^2\text{E}$ level. However, due to the smaller crystal field in GSGG, there is a very small energy gap, of the order of a few cm^{-1} , between the ${}^4\text{T}_2$ level and the ${}^2\text{E}$ level. This allows thermal repopulation of the ${}^4\text{T}_2$ level from the ${}^2\text{E}$ level, resulting in both radiative and non-radiative transitions from the ${}^4\text{T}_2$ band. We have found the combined radiative lifetime of these levels to be approximately 120 microseconds. However, as this is still quite long compared to the laser pulse length, any energy released as a result of transitions from these levels should not contribute significantly to any pressure pulse in the first few microseconds. For the reasons described previously, the sample was excited at equi-absorption points and the signals obtained normalised for the

incident number of photons at each excitation wavelength. The results are given for a number of groups of excitation wavelengths in Table 2.

From the table, we can see that the energy released for excitation via the high energy side of the ${}^4T_2$ band is quite similar to that released for excitation at a corresponding point on the high energy side of the ${}^4T_1$ band, suggesting that the energy resulting from the transition from the bottom of the ${}^4T_1$ state to the bottom of the ${}^4T_2$ state is not being given out non-radiatively. However, if this were the case, we would expect to see a large difference in the energy released as a result of excitation at equi-absorption points on opposite sides of the same band. Under these circumstances, the expected ratio of non-radiative energies for excitation at 418 nm and 509 nm would be approximately 6.0. This of course is nothing like the ratio observed of 1.24. Due to experimental circumstances it was not possible to investigate the behaviour for excitation at two equi-absorption points within the ${}^4T_2$ band. This result, though quite puzzling agrees in some respects with the results obtained at QUB using a conventional modulated photoacoustic and microphone detection technique, where we have observed a shortfall in the amount of non-radiative energy release after excitation via the ${}^4T_1$.

As in the case of ruby, GSGG:Cr³⁺ exhibits quite strong excited state absorption (ESA) bands in the vicinity of the ground state absorption bands. Using the ESA cross-sections reported by Andrews⁽³⁾, and the correction factors for these reported by Seelert and Strauss⁽⁴⁾, the contribution of ESA was modelled using a computer program as before, and was found to be less than 2 per cent for all the results given above. This was checked experimentally by monitoring the dependence of the signal and the transmission on the incident laser energy at a number of different wavelengths.

An investigation of the heavier doped sample of GSGG:Cr³⁺, at the same set of wavelengths as used for the GSGG5 sample, yielded the same results within experimental error.

Another application of the technique is to determine the velocity of sound in the crystal lattice under investigation. This can be done by generating an acoustic pulse at a discrete distance from the

TABLE 2. Piezoelectric (PZE) signals obtained at groups of equi-absorption points in GSGG:Cr³⁺ compared with theoretical predictions.

Wavelength (nm)	Photon Energy (ev)	Abs. Coeff. (cm ⁻¹)	PZE Signal (per photon)	Theor. Energy Released (per photon)
418	2.972	2.41	485 ± 50	1.399
509	2.441	2.41	392 ± 40	0.868
604	2.057	2.41	463 ± 46	0.484
422	2.944	3.06	505 ± 50	1.371
505	2.460	3.06	432 ± 43	0.887
614	2.023	3.06	497 ± 49	0.450
424	2.930	3.47	527 ± 53	1.357
502.5	2.472	3.47	445 ± 45	0.899
620.5	2.002	3.47	574 ± 57	0.429
430	2.889	4.71	600 ± 60	1.316
496	2.505	4.71	512 ± 51	0.932

transducer, thus causing a delay between the incident laser pulse and the detection of a pressure transient at the transducer. Then, by moving the laser beam a known distance from the transducer and noting the change in delay time, an accurate value of the velocity of sound can be obtained. A crude investigation of this type was carried out using the GSGG5 sample and the velocity was found to be;

$$V = 3750 \pm 500 \text{ ms}^{-1}$$

which is good agreement with the value given by Kitaeva et al⁽⁵⁾, of 3508 m^{-1} for a transverse shear wave.

In conclusion, we have shown that a thin PVdF film can be used as an effective piezoelectric detector for monitoring fast non-radiative decay processes in crystals. The results obtained for ruby are consistent, within the experimental error, with the accepted de-excitation scheme for that material. However, in the case of GSGG:Cr^{3+} , there appears to be a considerable shortfall in the amount of non-radiative energy released following excitation via the ${}^4\text{T}_1$ band as opposed to that obtained following excitation via the ${}^4\text{T}_2$ band. This is in agreement with results we have obtained using a modulated photoacoustic and microphone detection technique, in which all the non-radiative energy released following excitation is detected, where again a shortfall in energy following ${}^4\text{T}_1$ excitation was observed. Unfortunately, the results are inconsistent with the accepted de-excitation schemes and further experimental investigation is required.

References

1. C K Patel and A C Tam, Rev. Mod. Phys., 53, No. 3, P517 (1981).
2. T Kushida, J. Quant. Elect., 2, No. 9, P524 (1966).
3. Andrews et al, J. Chem. Phys., 84, No. 10, P5229 (1986).
4. W Seelert and E Strauss, Opt. Letts., 12, No. 10, P798 (1987).
5. V F Kitaeva et al, Phys. Stat. Sol., 92, P475 (1985).

B3-4 THE USE OF KrF LASER-GENERATED AS X-RAY SOURCES FOR SCANNING MICROSCOPY: A PRELIMINARY STUDY.

A M Rogoyski and A G Michette, (Physics Dept., King's College, London)
 I C E Turcu and F O'Neill, (Rutherford Appleton Laboratory)
 F Best, (Physics Dept., Portsmouth Polytechnic)

In the optimization of a laser-generated plasma source for scanning

soft x-ray microscopy several features have to be borne in mind. These include total x-ray flux in the required spectral region, pulse to pulse repeatability, and angular distribution of the radiation. The LSF Lambda Physik EMG150 KrF excimer laser has been used in some preliminary work for such an optimization. Absolute fluxes were measured using two silicon PIN diodes with $0.25 \mu\text{m}$ thick silicon windows; one, with a filter of $0.5 \mu\text{m}$ thick pure vanadium supported on a 90% transmitting $24 \mu\text{m}$ thick nickel grid, was sensitive to soft (280 - 510 eV) and hard ($> 800 \text{ eV}$) x rays, and the other, with a $25 \mu\text{m}$ thick beryllium filter, was sensitive only to hard x rays. The soft + hard filter had an effective area of $1.23 \times 10^{-5} \text{ m}^2$ and that of the hard filter was $1.96 \times 10^{-5} \text{ m}^2$. The filters were positioned at a fixed angle of 45° to the laser beam axis, 250 mm from the target. The averaged sensitivities of the (soft + hard) diode were $1.2 \times 10^{-2} \text{ C J}^{-1}$ for soft x rays and $3.3 \times 10^{-2} \text{ C J}^{-1}$ for hard x rays, while that of the hard diode was $1.5 \times 10^{-2} \text{ C J}^{-1}$. The soft x ray flux was obtained from a comparison of the signals from the two diodes.

The laser beam was focused to a minimum spot diameter of $25 \mu\text{m}$ on tungsten wire targets and the irradiance was varied by stepping the target through focus, thereby changing the spot size. The focal position was determined by occlusion of a $25 \mu\text{m}$ wire mounted at the top of the target.

The beam energy was attenuated by about 40% by windows, lenses and a protective pellicle before the target and so the focused beam energy was 215 mJ for the data presented here, corresponding to a maximum irradiance of $1.75 \times 10^{12} \text{ W cm}^{-2}$.

The soft x ray flux was insensitive to the spot diameter in the range 25 - $40 \mu\text{m}$, and corresponded to a total conversion efficiency of $\approx 5\%$ over 2π steradians in the energy range 280 - 510 eV. For the type of zone plate optic typically used in scanning x ray microscopy, with radius $\approx 35 \mu\text{m}$ and accepted bandpass $\lambda/\Delta\lambda \approx 350$, and assuming a 10% efficient monochromator, $\approx 1.2 \times 10^4$ photons of energy 380 eV are incident per pulse on the zone plate at 200 mm from the source. This flux would be comparable to that currently obtained at synchrotrons for a laser with a repetition rate of a few hundred Hz or greater.

Data was also taken for lower focused beam energies, by using an NO-filled absorbing cell. Analysis of this data is in progress.

B3-5 MULTIPHOTON MULTIPLE IONISATION OF ALIPHATIC HYDROCARBONS

L J Frasinski, K Codling and P A Hatherly (University of Reading)
J R M Barr (Rutherford Appleton Laboratory)

Experiments were recently performed on the multiphoton multiple ionisation (MPMI) of $N_2^{(1)}$ using an intense sub-picosecond laser (wavelength 600nm, pulse length 0.6ps, 200μJ per pulse, repetition rate 10Hz). An f/3 doublet lens focussed the laser radiation to produce a light intensity of about $10^{16}W/cm^2$ in the focal spot. The resulting ions were energy-analysed using time-of-flight (TOF) mass spectrometry and the spectra recorded on a LeCroy transient digitiser. An example of the TOF spectra obtained is shown in Figure B3.10. The ions of immediate interest, however, are the H^+ ions in the range 0.4–1.2μs, with energies up to 100eV. These ions were thought to result from the MPMI of residual hydrocarbon impurities (ie pump oil) in the experimental chamber, base pressure 1×10^{-8} Torr. The simultaneous observation of C^+ ions tended to support this view.

In order to confirm this hypothesis, a series of linear-chain hydrocarbons have been systematically studied, from butane (C_4H_{10}) to dodecane ($C_{12}H_{26}$) using an identical experimental arrangement. Examples of these spectra are shown in Figure B3.11 for butane, octane and dodecane. One observes in butane in particular a large number of peaks associated with fragment ions H^+ , C^+ , CH^+ , C_2H^+ etc. These sharp 'backward' peaks are associated with ions ejected initially in a direction away from the drift tube. These ions reverse their direction in response to the applied electric field across the interaction region ($\sim 50V/cm$) and arrive at the microchannel plate detector almost simultaneously, regardless of their initial kinetic energy. On the other hand, the 'forward' peaks involve ions whose TOF varies almost linearly with velocity and so ion energies can be measured. For example, one can easily determine the 'peak' H^+ energies and also the C^+ and C^{2+} ion energies.

The H^+ , C^+ and C^{2+} peak energies were determined for all 9 alkanes and plots of peak energy versus chain length (n —the number of C atoms) are shown in Figure B3.12 for both H^+ and C^+ . One notes that the two 600nm

plots are linear, with almost identical slopes. This suggests a similar mechanism may be responsible for the production of these energetic ions. In earlier experiments on $N_2^{(1)}$ and HI (2), the existence of energetic atomic ions was explained in terms of a rapid multiple ionisation process followed by a "Coulomb explosion". In an attempt to explain the present data, a similar mechanism was invoked. The application of the enormous laser E-field (approximately 20V/Å for a focussed intensity of $10^{16}W/cm^2$) is imagined to strip one electron from each of the C atoms in a very short time, possibly less than 100fs. The H^+ atom sitting at the end of the chain is now expelled by the line of C^+ ions and most of the Coulomb energy is taken by this H^+ ion.

The result of this simple Coulomb model, Figure B3.12, is seen to be at variance with the experimental data in terms of absolute energy and trend with increasing n . If one further assumes that the C^+ ions now disperse in response to their mutual repulsion, the energies obtained theoretically do not agree with the result of Figure B3.12. An alternative scenario is that of sequential fragmentation, where a fragment such as CH_2^+ or CH_3^+ is first created, but subsequent fragmentation into CH^+ and H^+ or CH_2 and H^+ could not explain the increase in kinetic energy of the H^+ ions with chain length. The maximum H^+ kinetic energy was determined for each alkane and once again there is a linear relationship with chain length. Moreover, extrapolation to an H^+ energy of 100eV (observed in Figure B3.10(b)) strongly suggests that the parent hydrocarbon responsible should have a chain length of 24 (± 2) carbon atoms, equivalent to a molecular mass of about 338amu. This is typical of rotary pump oil and appears to confirm our initial hypothesis.

The MPMI of these alkanes has recently been studied with a KrF excimer laser (wavelength 248nm, pulse length 5ps, 1mJ per pulse, repetition rate 5Hz) focussed to produce about $10^{16}W/cm^2$. The results for butane, octane and dodecane are shown in Figure B3.13. They differ considerably from those using the 600nm wavelength, Figure B3.11. The peak H^+ energies are very low, approximately 1eV, and the variation with chain-length is almost insignificant (1.0 to 1.4eV from butane to dodecane). The general shape of the H^+ energy spectrum hardly changes, implying a similar fragmentation mechanism is responsible. Presumably a particular fragment is created which subsequently fragments further to create the low energy protons. One also notes that the C^{2+} ions are hardly visible and the C^+ ions have their peak close to zero energy.

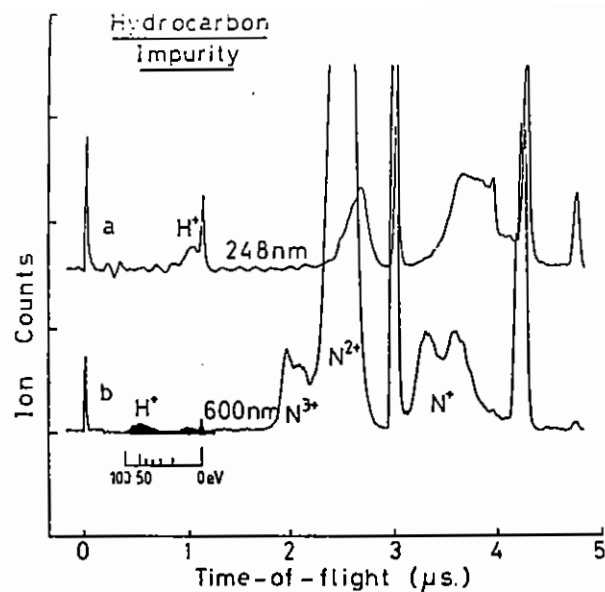


Fig. B3.10 Time of flight spectra obtained from a 0.6 ps, 200 μJ , 600 nm pulse focused to $\sim 10^{14}$ W/cm² in N_2 .

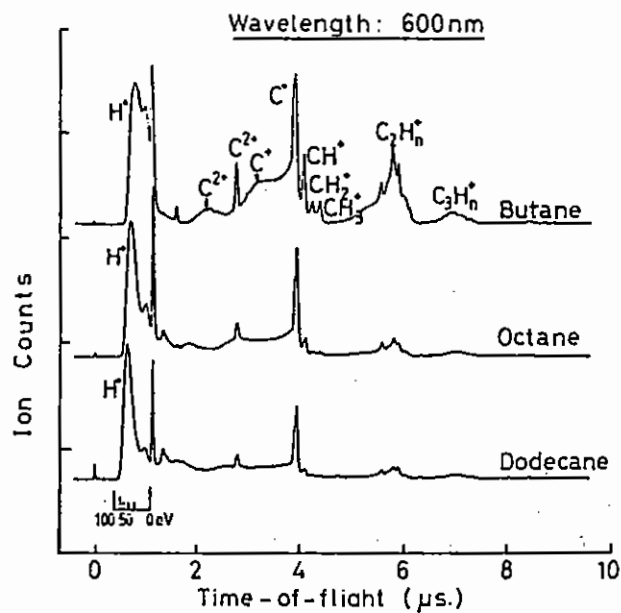


Fig. B3.11 Time of flight spectra of butane, octane and dodecane.

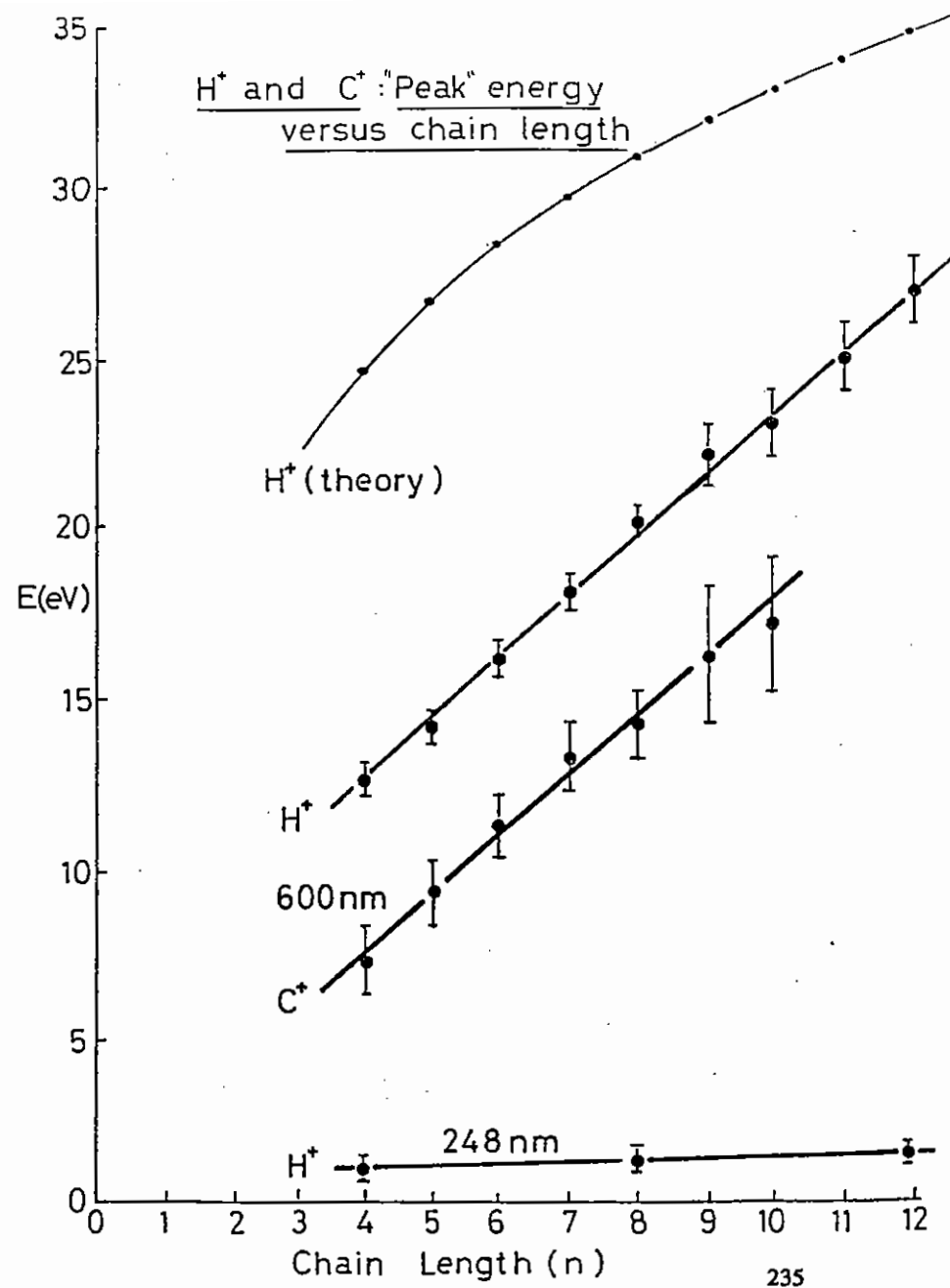


Fig. B3.12 H^+ and C^+ peak energies versus n the number of c atoms in the hydrocarbon chain.

On the basis of previous experience with the diatomic molecules N_2 , CO , O_2 one might be tempted to explain this different behaviour in terms of the different pulse widths involved:- 5ps for the 248nm laser compared with 0.6ps for the 600nm laser. (In fact it is the pulse rise-time that is important). In the case of diatomics, the ionisation process is thought to be caused by a field ionisation process, and the wavelength of the laser radiation is of secondary importance⁽¹⁾. However, earlier experiments at 600nm with a 3ps pulse length produced energetic protons (up to 50eV) from the hydrocarbons, the wavelength is important. This is confirmed by the considerable increase in cross section for the MPMI process at 248nm. In butane, for example, the gas pressure used to obtain the spectrum in Figure 83.13 (248nm) was a factor of 30 lower than that used for the Figure 83.11 (600nm) TOF spectrum. This behaviour is reflected in the large contribution of H^+ ions from hydrocarbon impurities seen in Figure 83.10(a), even though the base pressure here is improved to 4×10^{-9} Torr.

A general feature of all spectra, whether taken at 600nm or 248nm, is the absence of fragments containing more than four C atoms. This is not surprising when one considers the intensity used in these experiments ($\sim 10^{16}$ W/cm²). One is clearly far beyond the soft ionisation regime (3) of MPMI ($\sim 10^6$ W/cm²) and well into the region where hard fragmentation is dominant ($\geq 10^9$ W/cm²).

References

1. L J Frasinski, K Codling, P Hatherly, J Barr, I N Ross and W T Toner, Phys Rev Lett 58, 2424 (1987).
2. K Codling, L J Frasinski, P Hatherly and J R M Barr, J Phys B: At Mol Phys 20, L525 (1987).
3. H J Neusser, Int J Mass Spect Ion Proc 79, 141 (1987).

B3-6 DEGENERATE FOUR WAVE MIXING IN SEMICONDUCTOR-DOPED GLASS

R B Rodger, I S Ruddock and R Illingworth, Department of Physics and Applied Physics, University of Strathclyde, Glasgow

Semiconductor-doped glasses (SDGs) are commercially available as sharp cut low-pass filters. They have received a considerable degree of attention recently as nonlinear optical media because of their large effective $\chi^{(3)}$ and relatively low cost compared to the bulk semiconductor. Optical phase conjugation⁽¹⁾ and chaos⁽²⁾ have been observed in SDGs and there are potential applications of the materials to optical signal processing in waveguide and fibre devices^(3,4). SDGs usually consist of CdS_xSe_{1-x} microcrystals of $\sim 100\text{\AA}$ in size embedded in a glass host. The absorption band-edge is determined by the S/Se ratio, the microcrystal size which is fixed during the heat treatment stage manufacture when they grow and the temperature at which the filter is used. The nonlinearity is associated with the generation of electron-hole plasmas within the isolated microcrystals, with measured recombination occurring on timescales ranging from picoseconds to hundreds of nanoseconds, although there is much debate as to the dominant processes and the effect of microcrystal size. In this work we were particularly interested in the behaviour of these nonlinear optical materials under high intensity illumination and in the semi-permanent darkening which has previously been reported⁽⁵⁾ and attributed to carrier trapping on impurity or defect sites.

The Laser Support Facility Quanta Ray PDL dye laser pumped by the frequency doubled JK Hyperyag 750 Nd:YAG laser was used to investigate degenerate four wave mixing in 2mm thick samples of Hoya 0-58 and R-60 glasses. The input laser beam was divided at a 50:50 beamsplitter to form two counterpropagating beams of equal intensity. The object or probe beam was produced by reflecting a portion of the input beam from an uncoated beamsplitter while the phase conjugate reflection was detected after reflection from another uncoated beamsplitter. The input and output pulses were monitored by photodiodes and analysed by means of the LSF Stanford Research gated integrator. Rhodamine 640 was used throughout the experiments, yielding up to 35mJ per 10ns pulse at 604nm and tunable between 600nm and 620nm.

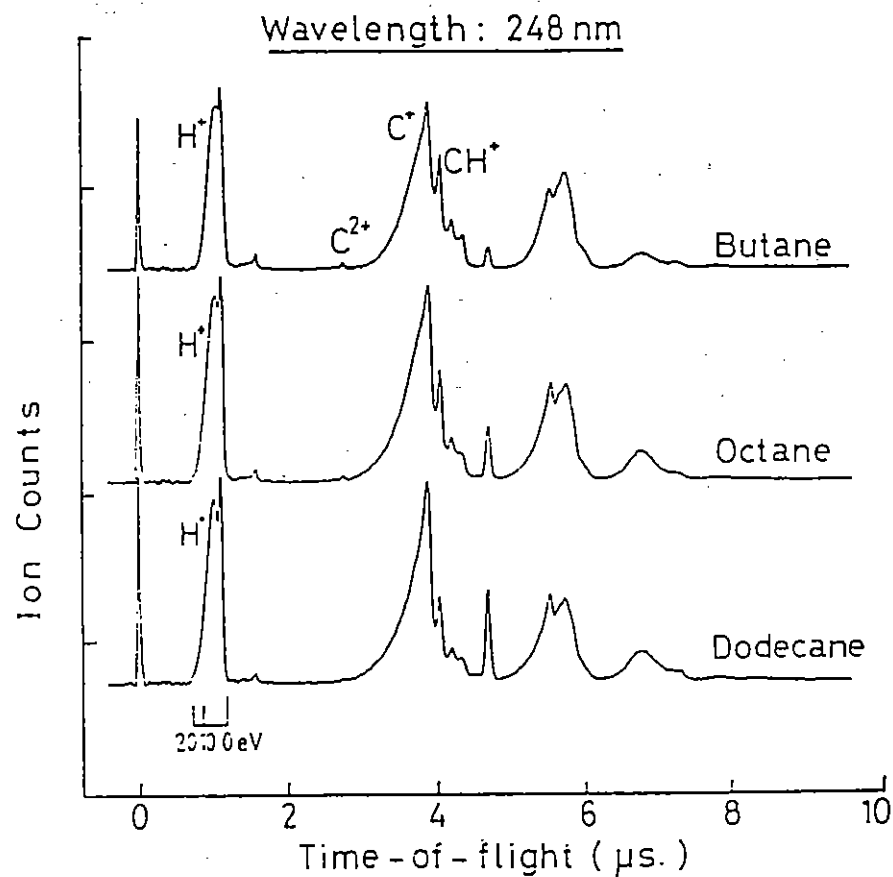


Fig. B3.13 Time of flight spectra of butane, octane and dodecane obtained using 248 nm, 5 ps, 1 mJ pulses focused to $\sim 10^{16}$ W/cm².

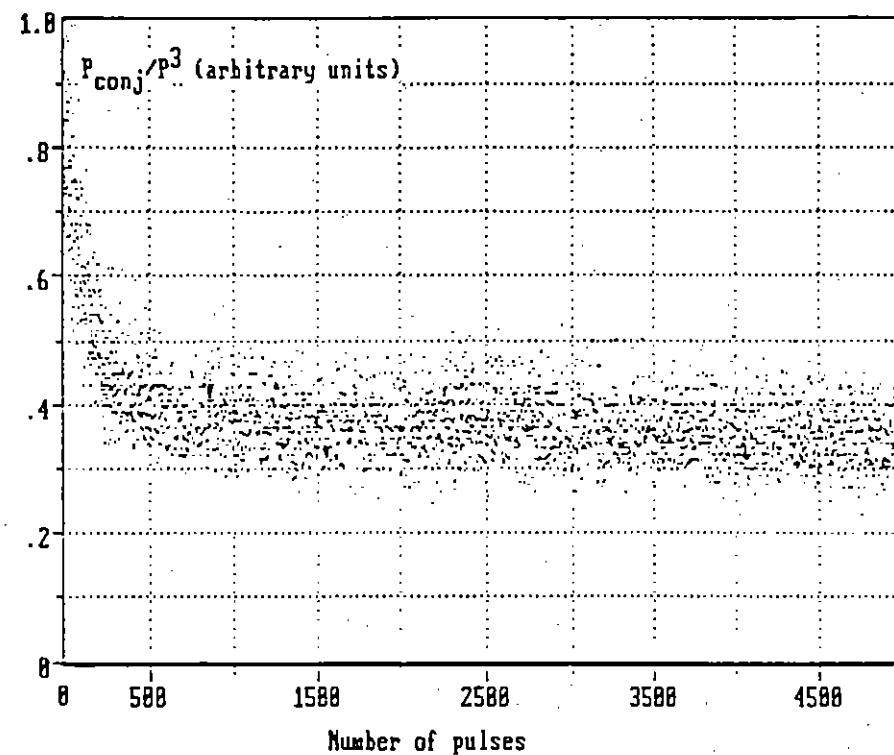


Fig. B3.14 The dependence of the normalised output pulse energy on the number of pulses incident on the sample for Hoya 0.58 glass at 608nm. Maximum input pulse energy = 25mJ.

An example of the effect of laser induced darkening is given in Figure 83-14 in which the normalised conjugate output energy per pulse is shown for 5000 successive pulses in O-58 glass at 698nm. The pulse energy was 25mJ and the normal glass transmission was 0.9. After exposure to the first 500 pulses the reflected signal decreased to ~ 0.4 of that for a fresh sample of the glass. This rapid decrease was superimposed on a slow decline in the response of the SDG of ~ 0.04 for the 5000 pulses. Direct measurement of darkening in O-58 glass with 25mJ pulses at 608nm showed that the decrease in transmission was proportional to $[\text{number of pulses}]^n$ where $n = 0.6$ in this case. After $\sim 10^4$ pulses when the fractional decrease in transmission had reached 0.09, darkening began to saturate. Spectrophotometry of the darkened spot showed that the transmission was uniformly reduced but that the position of the band-edge remained unaffected. Within the loan period it was not possible to determine the complete dynamics of darkening but subsequent experiments with an argon-ion laser have failed to create a comparable effect with up to two orders of magnitude more photons being absorbed indicating either the existence of a threshold intensity or the deleterious effect of heating arising from the use of a cw laser. These darkening results showed that although the response of the SDG decreased sharply during the first 500 pulses, this was before an appreciable change in transmission was discernible. By this stage, however, the luminescence spectrum had changed considerably, both in strength and distribution. Conversely, the slow background decrease in the SDG response was approximately equal to the decrease in transmission of the glass caused by the pump beams during the remaining 4500 pulses. It is not thought that the background decrease in the SDG response was due to the erasure of a permanent grating arising from a spatial modulation of the darkening as reported elsewhere⁽⁵⁾. No evidence for one was observed and in addition the experimental set-up was not interferometrically stable which prevented the creation of a stationary modulation in the darkened spot in the active region of the SDG during illumination by thousands of pulses.

The dispersion of $\chi^{(3)}$ in the R-60 glass was investigated for the wavelength range 600-608nm in which the transmission varied from 0.4-0.7. Results reported by other workers have shown that for fresh glass $\chi^{(3)}$ is proportional to the absorption coefficient within the exponential tail of the absorption edge^(5,6). We observed that as the

laser was tuned to shorter wavelengths into the absorption edge, the wavelength dependence of $\chi^{(3)}$ deviated considerably from this expected relationship by decreasing rapidly and then remaining approximately constant. We interpret the initial decrease as being due to the onset of darkening in the glass followed by a relatively flat dispersion of $\chi^{(3)}$ for this affected region.

The use of the LSF dye laser enabled us to make some basic measurements on the behaviour of these media under high intensity conditions. Understanding the origin and consequences of the laser induced darkening is essential if such nonlinear optical media are to be used in miniature devices where the power density is high. Experiments carried out on SDGs subsequent to the loan have concentrated on studying thermo-optical effects and the properties of the darkened glass. Work presently in progress will use glass produced under controlled conditions in which the microcrystal size and composition is predictable. The aim is to make time-resolved measurements of the recombination processes involved in grating formation and decay and to extend previous work by us which concerned the slow (ms) nonlinearities and their role in the generation of modulation subharmonics and chaos^(2,7).

References

1. R K Jain and R C Lind, J Opt Soc Am, 73, 647 (1983).
2. N I Zheludev, I S Ruddock and R Illingworth, Modern Optics, 34, 1257 (1987).
3. B J Ainslie, H P Girdlestone and D Cotter, Elect Lett, 23, 405 (1987).
4. T J Cullen, C N Ironside, C T Seaton and G L Stegemen, Appl Phys Lett, 49, 1403 (1986).
5. P Roussignol, D Ricard, J Lukasik and C Flytzanis, J Opt Soc Am, B4 5 (1987).

6. P Horan and W Blau, *Semicon Sci Technol*, 2, 383 (1987).
7. N I Zheludev, I S Ruddock and R Illingworth, *Opt Quant Elec*, in press (1988).

B3-7 He II BALMER LINE SHIFTS IN A DENSE Z-PINCH PLASMA

J P Marangos and D D Burgess (Blackett Laboratory, Imperial College)

Introduction

The magnitude of the shifts of hydrogenic ion lines in a dense plasma is a long-standing problem in the field of plasma spectral line shapes. The issue of the size of the contribution from the plasma polarisation shift to the total shift of the lines is yet to be resolved. The other important contributions to the shift are the ion quadrupole effect and the electron impact contribution for which there is also some uncertainty. Knowledge of the size of the total shift is required in the calculation of plasma opacities and in some x-ray laser scheme gain calculations.

Work has been carried out in the Blackett Laboratory over a number of years on this problem (see Goto and Burgess (1974), Baker and Burgess (1979)). A dense, cool, z-pinch plasma source has been built in order to study line shapes and shifts in dense and non-Debye plasmas (Baldwin (1983)). Due to the difficulties encountered in emission studies, from cool boundary zones in the plasma and the limits set by the resolving power of monochromators, a programme of absorption line shape/shift studies has been pursued. This relies upon the generation of narrow-bandwidth coherent tunable VUV light by up-conversion of intense dye laser light by either degenerate fwm or Raman up-conversion.

Earlier work in this laboratory has demonstrated the successful application of tunable VUV light to the measurement of the Lyman- α line profile in a dense z-pinch plasma (Baldwin et al (1984)) and a recombining hydrogen z-pinch. A problem often encountered in these measurements is the large magnitude in the fluctuation on the background plasma emission compared to the transmitted tunable VUV signal. In some cases the transmitted tunable VUV is completely lost

in these fluctuations. Under these conditions it is not possible to perform absorption measurements, however, it is still possible to use the narrow-bandwidth tunable VUV light to calibrate the monochromator to a high wavelength accuracy. An emission measurement can then be performed with an accurate wavelength calibration which is essential if a shift measurement is to be made.

Theoretical Background

In the absence of a single unified theoretical treatment of the plasma line shape/shift problem it is usual to tackle calculations of the shift by considering separately contributions from various mechanisms, even though these mechanisms are interconnected. The major causes of contributions to ionised helium line shifts are believed to be:

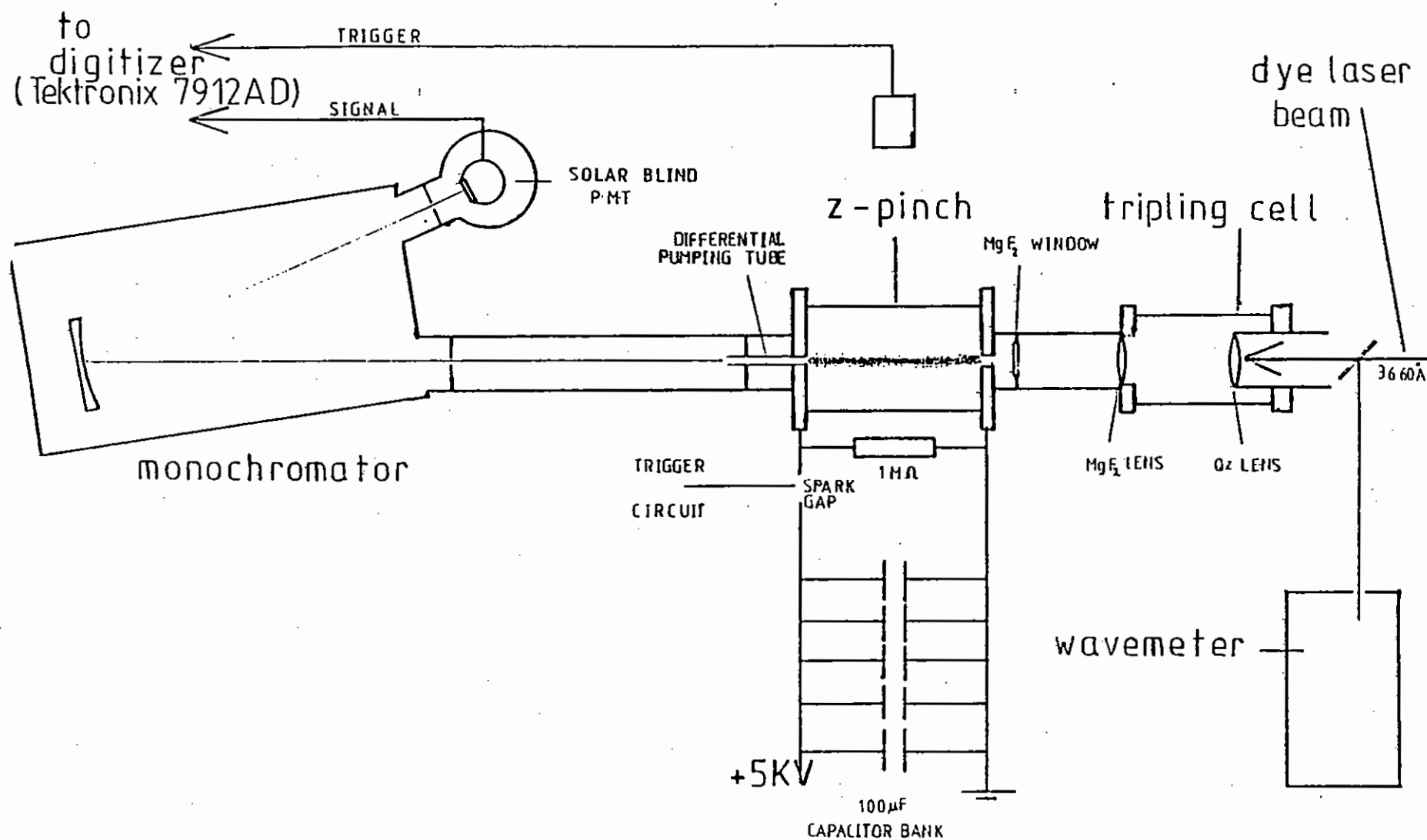
- (a) Ion quadrupole effect, due to the field gradients across the emitting ion.
- (b) Electron impact shift contribution, arising from the effects of predominately distant electron collisions with the emitter.
- (c) Plasma polarisation shift, due to initial correlations between the ion emitter and the plasma. When the time averaged charge distribution is calculated an excess of negative charge is found in the vicinity of the ion (due to coulomb attraction). As a result polarisation charge (free plasma electrons) may penetrate the ionic wavefunctions causing shielding of the nuclear charge from the bound electrons and thus leading to energy level shifts.

Calculations have been made of the magnitude of each of these for instance Griem has calculated the size of the shifts due to (a) and (b) (Griem (1983)) and Blaha and Davis have calculated the shift due to plasma polarisation shift (Blaha and Davis (1983)). The results of these calculations for the first two members of the Balmer series of He II in a plasma with electron density 10^{18} cm^{-3} are shown in Table 1, the shifts scale linearly with electron density.

Experimental Techniques

Z-Pinch

The high peak densities available in this plasma are achieved by



constructing the capacitor bank from low inductance capacitors, and configuring the z-pinch discharge vessel and current feeding lines so that there is a low total inductance in the discharge system. This leads to a large peak current in the discharge with an associated large compression force in the pinching phase.

Plasma Diagnostics

Diagnosis of the plasma electron density, for the firing conditions selected for this work, was achieved using several techniques. Line width studies were made on the He I 5876A, He II 4686A and He II 1640A lines. The results for both the He II 1640A and the He I 5876A lines indicated that the electron density was $1.0 - 2.0 \times 10^{16} \text{ cm}^{-3}$ (using tables from Griem (1976)). He II 4686A line width however implied a density of only $3.0 \times 10^{17} \text{ cm}^{-3}$. These line width measurements were however made without good spatial resolution, thus this discrepant result cannot be viewed necessarily as an indication of any disagreement of the measured He II 4686A line width with theory. Another density diagnostic was employed in order to confirm this value of electron density in the plasma. Interferometry using He-Ne and cw ring dye laser light was performed, this gave a density of $2.0 \pm 0.5 \times 10^{16} \text{ cm}^{-3}$ for the core of the discharge column at peak compression. Further confirmation of this was obtained by analysis of the results of continuum opacity measurements at 1650A yielding a density of $2.4 \pm 0.5 \times 10^{16} \text{ cm}^{-3}$.

The plasma temperature was determined using the He I 5876A and He II 4686A line intensity ratio and assuming LTE (a good assumption at these plasma densities). A temperature of $3.3 \pm 0.2 \text{ eV}$ was deduced by this technique.

Coherent Tunable VUV Generation

To study the first two members of the He II Balmer series it was necessary to generate light tunable around 1640A (3-2 transition, Balmer- α) and 1215A (4-2 transition, Balmer- β).

Light tunable around 1640A was generated by Raman up-conversion, in H_2 , of excimer pumped dye laser light at 3600A. The 8th anti-Stokes order of this laser light corresponds to 1640A. Intensities of around $2 \times$

10^8 photons per pulse were generated. Frequency tripling in Krypton, of excimer pumped dye laser radiation at around 3660A, was used to generate coherent VUV light tunable from 1203-1234A. Intensities of around 3×10^7 photons per pulse were generated. This light source was used previously by the authors for the measurement of Lyman- α line profiles in a dense z-pinch plasma and a recombining z-pinch plasma.

Emission and Absorption Measurements

The optical system employed in these measurements is shown in Figure B3.15. Measurements were performed along the axis of the plasma. Initially, to simplify alignment, MgF_2 end windows were used, these were adequate for the absorption measurements as the transmission of the optical system was checked after each shot. For emission measurements however, the deterioration of MgF_2 transmission each time the plasma was fired distorted the emission profiles. A differential pumping device, consisting of a 200 mm long tube of 2 mm bore, was used to replace the window. This cured the problem of signal deterioration entirely and also gave excellent spatial resolution in the measurements.

During absorption measurements at each point in the profile the laser wavelength was measured using a laser wavemeter (Candela Corp LS-1). This resulted in an accuracy of better than 0.05A in the VUV wavelength determination. For emission measurements the tunable VUV was used to carefully calibrate the monochromator wavelength, this wavelength calibration was demonstrated to be reproducible to an accuracy of around $\pm 0.2\text{A}$ providing the scan was made in the same direction. Also during each emission profile scan the tunable VUV was used to re-check the calibration at 1212.8A, these re-checks were always found to be consistent with the calibration curve.

Results and Discussion

The results of the He II Balmer- α absorption measurement are shown in Figure B3.16. There was insufficient VUV signal to measure the opacity closer than 0.8A from line centre. The error bars result from the low VUV signal level and the shot to shot fluctuation in the signal. A curve is drawn to aid the eye, it is drawn on the basis of a visual "best fit" superimposing the two wings of the experimental profile.

The data are consistent with a red shift of between 0.1Å and 0.4Å. They do not support a shift of greater than 0.4Å.

Emission results for the He II Balmer-β line are shown in Figure B3.17(a). The tunable VUV signal level was too small to allow any absorption measurements to be made of this profile. An accurate wavelength calibration of the monochromator was obtained by using the tunable VUV.

The profile in Figure B3.17(a) shows the two Stark components split by 3.5Å. An absorption dip, believed to be due to hydrogen Lyman-α in cooler end zones of the plasma, is apparent from the red peak to the centre of the profile. A visual "best-fit" curve has been drawn onto the data in Figure B3.17(b). This indicates a full width at half-maximum of 8.8Å. The line centre occurs at 1215.9 + 0.2Å, the unshifted line centre of this line should be at 1215.2Å. Further confirmation of this shift is obtained from the relative positions of Lyman-α (1215.7Å) and the centre of the measured He II Balmer-β profile.

The width of the He II Balmer-β line corresponds to an electron density of $1.4 \times 10^{18} \text{ cm}^{-3}$ if Griems tables are used. In contrast the separation of the two Stark components of the line corresponds to an electron density of $1.9 \times 10^{18} \text{ cm}^{-3}$ again using these tables, more consistent with other diagnostics of this plasma. This indicates our line profile is significantly narrower (by around 20%) than Griems theoretical prediction.

A comparison of our shift result for the Balmer-β line with the predictions in Table 1 indicate that at our diagnosed plasma density of $2 \times 10^{18} \text{ cm}^{-3}$ the measured value of 0.7Å is consistent with the predicted value of 0.8Å. The measured value is not consistent with the prediction if the plasma polarisation shift contribution is ignored. For the Balmer-α line the experimental upper limit on the shift of 0.4Å is also consistent with the prediction, although more accurate data is needed for a detailed comparison to be carried out.

Conclusion

The high wavelength accuracy provided by narrow-bandwidth tunable coherent VUV sources has been exploited to make measurements of He II

Balmer series line shifts at plasma densities nearly an order of magnitude higher than in any previous work known to the authors. For the He II Balmer-β line a red shift of $0.7 \pm 0.2\text{Å}$ has been measured in a plasma of electron density $2 \times 10^{18} \text{ cm}^{-3}$ and temperatures 3.3 eV.

Acknowledgements

This work was supported by the S.E.R.C., the lasers used were made available by the Laser Support Facility of the Rutherford Appleton Laboratory. We are indebted to Dr Gillian Nave and Mr David Pierce for assistance given during these experiments.

References

1. K G H Baldwin, University of London PhD Thesis, (1983).
2. K G H Baldwin, J P Marangos and D D Burgess, J. Phys. D, 17, L169, (1984).
3. E A M Baker and D D Burgess, J. Phys. B, 12, 2097, (1979).
4. M Blaha and J Davis, N.R.L. Memorandum Report 5155, (1983).
5. T Goto and D D Burgess, J. Phys. B, 7, No. 7, 857, (1974).
6. H R Griem, Phys. Rev. A, 27, No. 5, 2566, (1983).
7. H R Griem, "Spectral Line Broadening in Plasmas", Academic Press, New York, (1976).

Table 1

He II Transition	Ion Quadrupole	Electron Collision	P.P.S. (Blaha & Davis)	Total
n = 2 - 3 (1640 Å)	- 0.02 Å	+ 0.08 Å	+ 0.12 Å	+ 0.18 Å
n = 2 - 4 (1215 Å)		+ 0.15 Å	+ 0.25 Å	+ 0.40 Å

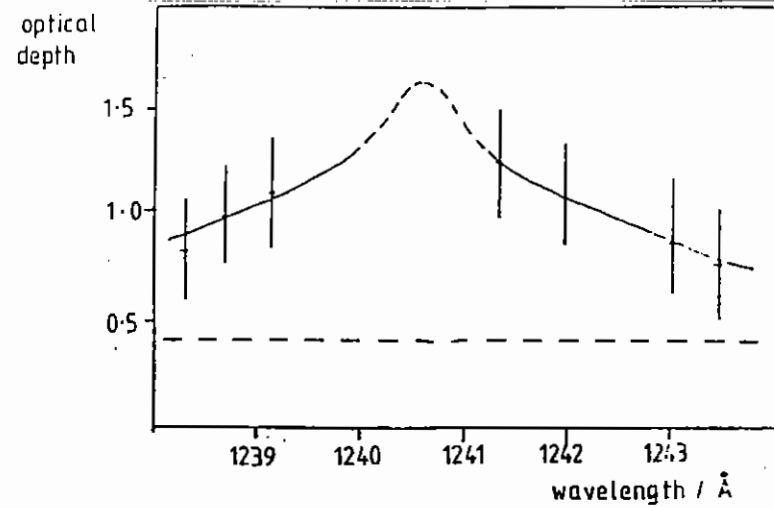


Fig. B3.16 He II Balmer- α absorption profile.

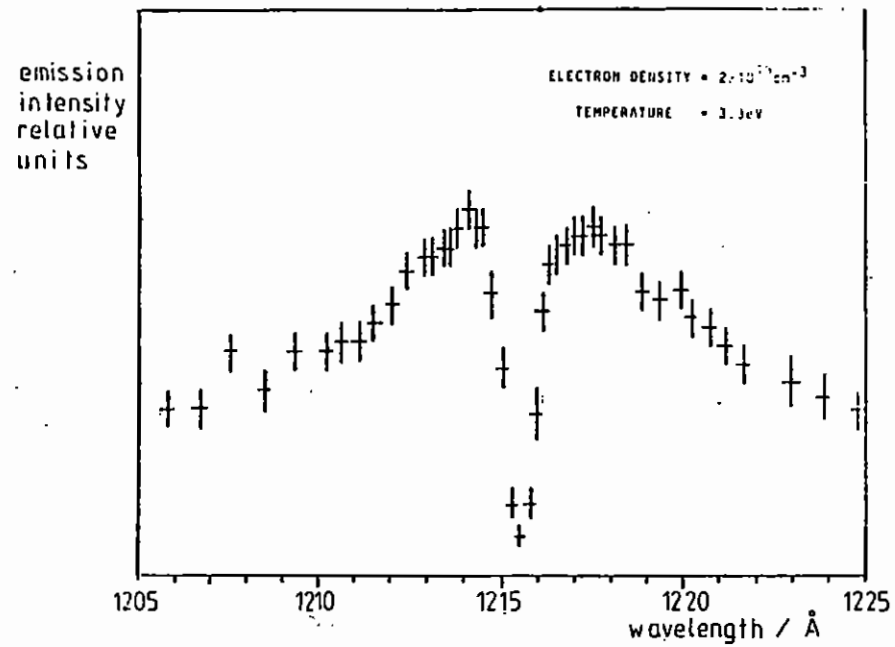


Fig. B3.17(a) He II Balmer- β absorption profile.

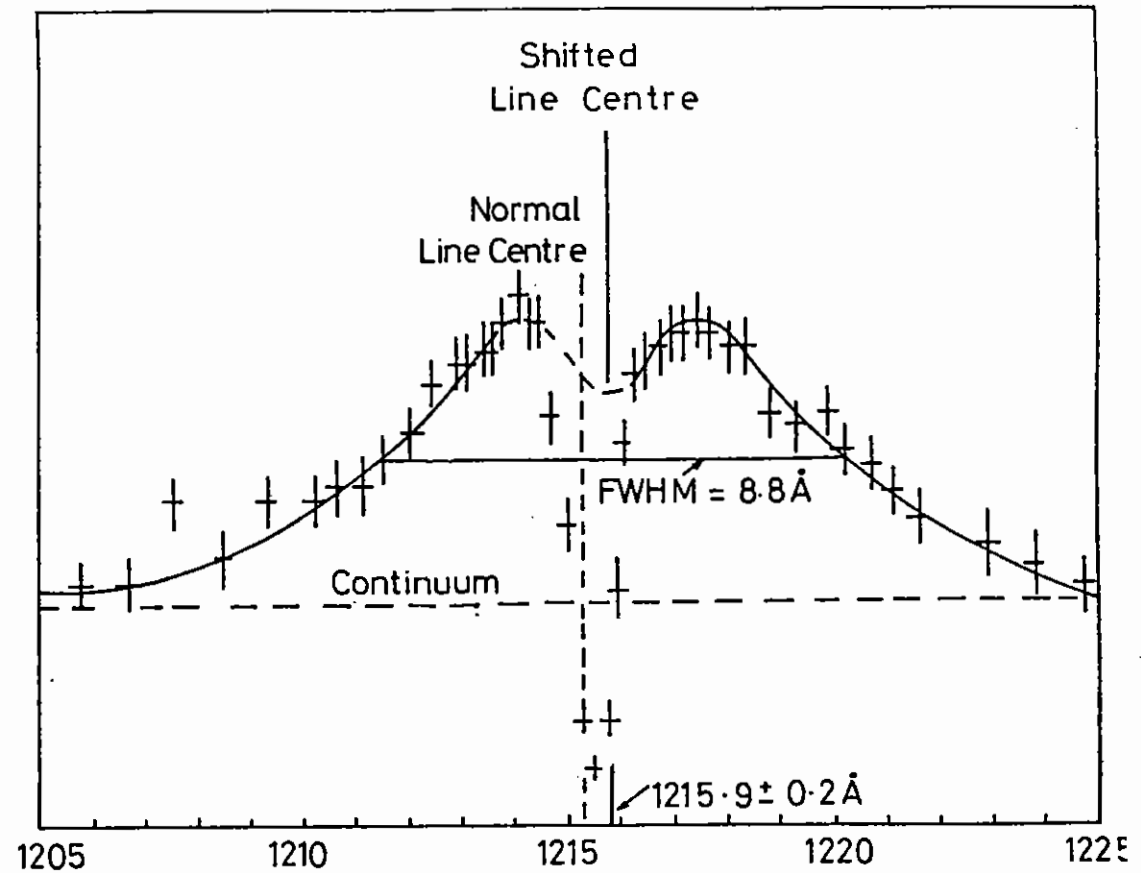


Fig. B3.17(b) He II Balmer- β profile showing shift and width.

B3-8 IN-SITU STUDIES OF MOVPE PROCESSES USING OPTICAL SECOND HARMONIC GENERATION

D S Buhaenko, S M Francis, P A Goulding and M E Pemble*, (Department of Chemistry, University of Manchester, Institute of Science and Technology, PO Box 88, Sackville Street, Manchester M60 1QD).

Introduction

Both molecular beam epitaxy (MBE)^(1,2) and metal-organic vapour phase epitaxy (MOVPE)^(3,4) are routinely employed in the growth of thin epitaxial layers of compound semiconductors such as GaAs, GaAlAs and ZnSe and improvements in both processes are constantly being sought. At UMIST we are interested in both understanding the chemistry of MOVPE processes and the development of new growth procedures such as photo-assisted MOVPE⁽⁵⁾. The approach that we have adopted is based on the premise that since semiconductor growth is a surface phenomenon, the chemistry may be studied using surface science techniques. In addition to the normal surface science techniques such as Auger electron spectroscopy and low energy electron diffraction, we are involved with the development of novel optical techniques for the study of MOVPE processes. Such techniques employing photons rather than say electrons would not be inherently restricted to high vacuum operation and thus may be applied in-situ, during a growth reaction. With this in mind we have recently been able to evaluate the feasibility of using optical second harmonic generation to monitor MOVPE processes using a Nd:YAG laser on loan from the Rutherford Appleton Laboratory.

We report here the results of a study of the 'real time' monitoring of the interaction of a typical MOVPE precursor, triethyl gallium (TEG) with a GaAs (100) surface.

Theory

It is known that when a beam of monochromatic radiation of frequency ω reflects from the surface of a conductor, the non-linear surface susceptibility gives rise to a frequency doubling effect such that the reflected beam contains radiation at a frequency 2ω ⁽⁶⁾. This

phenomenon, known as second harmonic generation (SHG) is allowed under the dipole approximation for a medium that does not possess inversion symmetry. A surface by definition falls into this category whereas a gas does not and thus the technique is surface sensitive but independent of overpressure. SHG has been extensively applied to monitor processes occurring on metal surfaces where it has been shown that adsorbates alter the surface susceptibility and hence the intensity of the SHG signal⁽⁷⁻¹²⁾. In addition, studies of the indirect band gap materials Si and Ge have concluded that SHG from semiconductor surfaces is allowed only when the energy of the primary beam is greater than or equal to the band gap of the material since only under these circumstances is the dipole approximation valid⁽¹³⁾.

Experimental

Figure B3.18 depicts the ultra-high vacuum system in which the SHG experiments were performed. This system is equipped for Auger electron spectroscopy, low energy electron diffraction, mass spectrometry and atmospheric pressure studies^(14,15). The typical base pressure of the system is $< 1 \times 10^{-10}$ mbar. The 1064 nm line of the Nd:YAG laser was used to stimulate SHG being incident on the GaAs (100) sample at an angle of 45° at a power of approximately 20 mJ per pulse and a repetition rate of 20 Hz, and the signal at 532 nm was detected after filtering from the 1064 nm line via dichroic filters using a photomultiplier (EMI 9217 QB) operated at 1200 V. This tube was selected since it has high sensitivity in the uv and visible and very low sensitivity in the IR. No particular polarisation of the laser was selected in these experiments. The signal from the photomultiplier tube was fed into an electrometer which provided a voltage output to an A/D converter. This output was recorded using an Amstrad 1512 microcomputer using software developed in our laboratory. The GaAs (100) sample was cleaned via a process of chemical etching and treatment in H_2 which is described in more detail elsewhere⁽¹⁵⁾.

Results

The reproducibility of the background SHG signal was checked over a period of time, Figure B3.19(a) and the sample exposed to 5×10^{-9} mbar

TEG while monitoring the SHG signal Figure B3.19(b). It can be seen that in the absence of TEG the SHG signal is constant. In order to achieve this great care had to be taken to minimise stray light fluctuations. While dosing with TEG the SHG signal increased to a maximum and then levelled off after an exposure of approximately 0.5L (1L = 1×10^{-6} torr sec). Figure B3.20 depicts Auger electron spectra of Ga, As and C before and after exposure to TEG. From Figure B3.20 it is clear that exposure to TEG results in an increase in Ga signal, a slight decrease in As signal and no change in the C signal.

Discussion

In contrast to the results of previous studies⁽¹³⁾, it would appear that we have been able to observe an SHG signal from GaAs (100) using an incident source of energy below the band gap energy ($E_g = 1.4$ eV)! We suggest that this occurs due to either laser-induced heating resulting in the promotion of a significant number of electrons to the conduction band or a multiphoton electron-hole pair excitation mechanism possible since the transition between the valence band maximum and the conduction band minimum is direct for GaAs. At present this latter argument seems more plausible since it has been shown that up to the pulse power required to melt a GaAs (110) surface, heating does not significantly alter the intensity of the SHG signal⁽¹⁶⁾. The structure of GaAs is centroasymmetric and thus it is likely that the observed SHG signal contains components due to both surface and bulk contributions. The Auger electron spectra in Figure 3 suggest that the Ga adsorbs without any residual carbon fragment. If this is the case then surface heating may have induced decomposition of the TEG. Alternatively the laser may have decomposed the TEG above the surface. The variation in the SHG signal with exposure time suggests that the surface process follows Langmuir kinetics in which the rate of adsorption of Ga is proportional to the number of available surface sites. Correlations of SHG signals with adsorption kinetics have been made previously⁽⁹⁾ and we intend to repeat these calculations for this system.

Conclusions

We believe that this short study has demonstrated the potential of the

SHG technique for the monitoring of dynamic semiconductor growth processes in real time. With simple optimisation of the optical arrangement, it is felt that surface heating can be overcome (i.e. use lower incident power) and sensitivity can be dramatically improved.

Acknowledgements

We gratefully acknowledge the support of the GEC Company for a CASE studentship (DSB) and substrate materials, the SERC for equipment grants and an IT studentship (PAG) and MOD/DCVD for a research assistantship (SMF). Finally we would like to thank Dr Andrew Langley of the Rutherford Laser Support Facility for useful discussions.

References

1. E Tokumitsu, Y Kudou, M Konagi and K Takashashi, J. Appl. Phys., **55**, 3163 (1984).
2. S Horiguthi, K Kimura, K Kamon, M Mashita, M Shimazu, M Mihara and M Ishii, Jap. J. Appl. Phys., **25**, L979 (1986).
3. M J Ludowise, J. Appl. Phys., **58**, L21 (1985).
4. M W Jones, D R Daniel, J Pooladdej and R M Gibb, Semicond. Sci Technol., **2**, 167 (1987).
5. S J C Irvine, J B Mullin and J Tunnicliffe, J. Crystal Growth, **68**, 188 (1984).
6. N Bloembergen and P S Pershan, Phys. Rev., **128**, 6399 (1962).
7. C K Chen, T F Heinz, D Ricard and Y R Shen, Phys. Rev. Lett., **46**, 1010 (1981).
8. T F Heinz, C K Chen, D Ricard and Y R Shen, Phys. Rev. Lett., **48**, 478 (1982).
9. H W K Tom, C M Mate, X D Zhu, J E Crowell, T F Heinz, G A Somorjai and Y R Shen, Phys. Rev. Lett., **52**, 348 (1984).

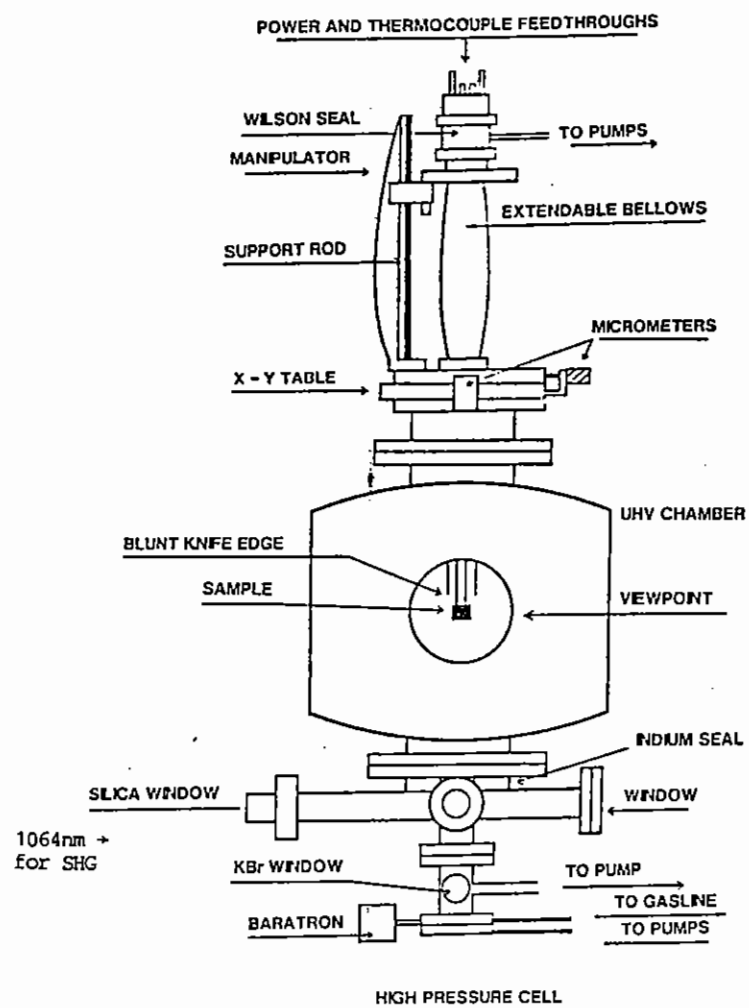
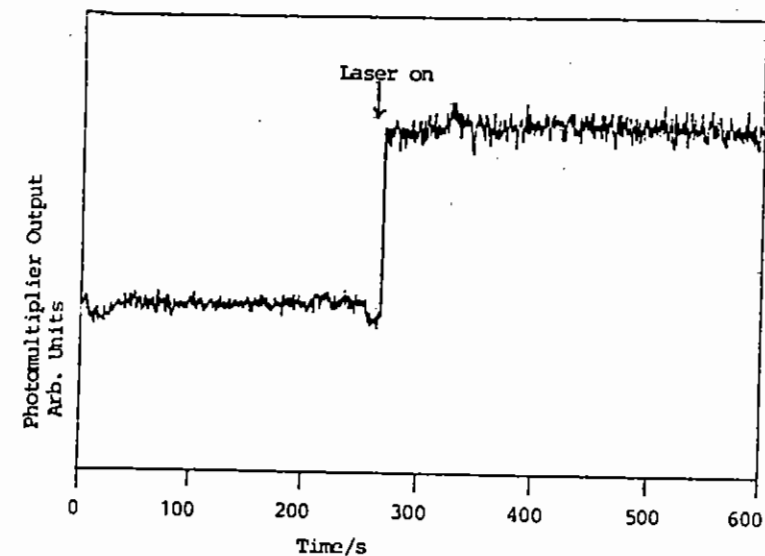


Fig. B3.18 The Ultra-high Vacuum System.

(a)



(b)

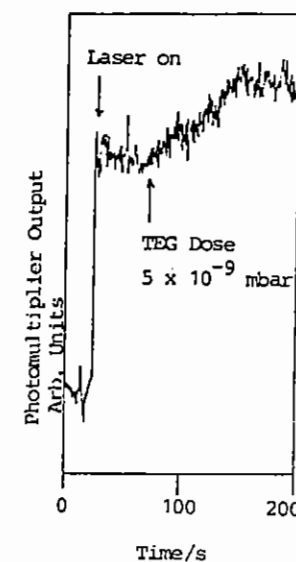


Fig. B3.19 SHG Measurements from GaAs (100)

(a) Background SHG

(b) TEG Dynamic Exposure SHG.

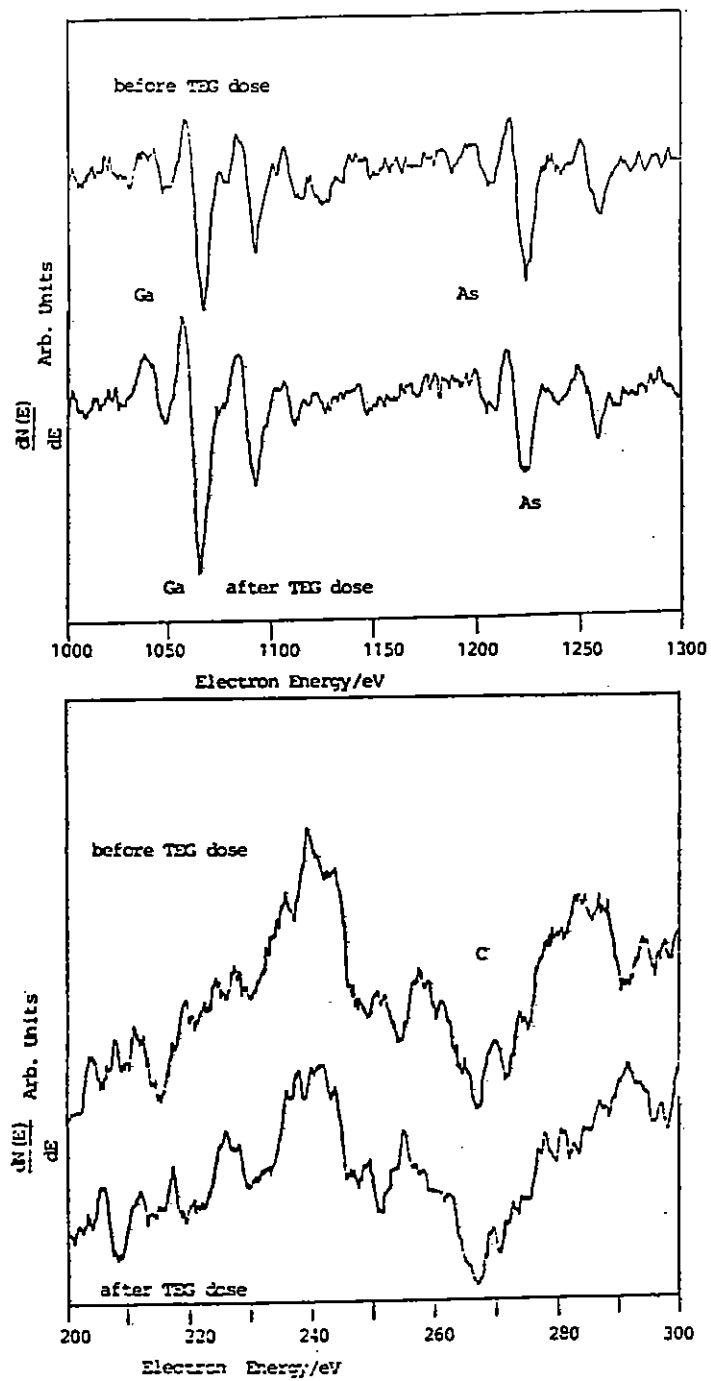


Fig. B3.20 Auger Electron Spectra from GaAs (100).

10. Y R Shen, J. Vac. Sci, Tech. B, 3, 1464 (1985).
11. D Heskitt, K-J Song, L Urbach and E W Plummer, J. Vac. Sci. Tech. A, 5, 690 (1987).
12. J F McGilp, J. Vac. Sci. Tech. A, 5, 1442 (1987).
13. D Guidotti, T A Driscoll and H J Gerritsen, Solid State Comm., 46, 337 (1983).
14. M E Pemble, Chemtronics, 2, 13 (1987).
15. D S Buhaenko, S M Francis, P A Goulding and M E Pemble, submitted to J. Vac. Sci. Tech., Jan. 88.
16. A M Malvezzi, J M Liu and N Bloembergen, Appl. Phys. Lett., 45, 1019 (1984).

B4	LASER DEVELOPMENT AND NON-LINEAR OPTICS	Pages
B4-1	Line Narrowing of An ArF Laser	249 - 257
B4-2	CW Stimulated Brillouin Scattering in Optical Fibres	251 - 152
B4-3	X-ray Lithography using a KrF Laser-Plasma Source	253 - 256
B4-4	Plasma X-ray Source Generation using mJ-level 100 psec XeCl Laser Pulses	256 - 258
B4-5	A Novel Single Shot Cross-Correlator for Measuring Pulse Lengths at 249nm	258 - 261
B4-6	Ultra-violet Generation by Frequency Doubling using β -Barium Borate	261 - 262

Editor: W T Toner

B4-1 LINE NARROWING OF AN ArF LASER

G M Davis and M C Gower (RAL)

This experiment was designed to optimise the performance of a Lambda Physik Model EMG150 excimer laser for operation as a source of narrow linewidth ($\sim$ few wavenumbers) radiation at the ultraviolet wavelength of 193 nm (ArF). This commercial excimer laser employs a standard two-module oscillator-amplifier configuration. The laser linewidth and divergence are controlled by placing dispersive optical elements and small apertures in the oscillator cavity. The low power output radiation from the oscillator module is subsequently amplified by the second laser module.

In order to optimise the oscillator energy output while simultaneously providing a reasonable amount of line-narrowing ($\sim 10 \text{ cm}^{-1}$) we have studied several oscillator cavities formed using different combinations of prisms, gratings and output couplers. These simultaneous energy and linewidth requirements were found to be best satisfied by the arrangement shown in Figure B4.1. A fairly small angle of incidence (55°) at the grating was used and two fused silica prisms expanded the laser beam and thereby increased the length of illumination of the grating. For a given angle of incidence at the grating the amount of line narrowing achieved increases as the length of illumination of the grating increases. In addition, since the grating efficiency decreases at higher angles of incidence, we wished to use the minimum angle of incidence compatible with achieving the required linewidth of operation.

Of the two available dielectric ArF reflective end mirrors of 50% and 100% the highest oscillator energy output was achieved by employing the 100% reflector and using the zero-order reflection from the grating as the oscillator output.

Using the optimised oscillator cavity shown in Figure B4.1 studies were undertaken to try and improve the energy and shot to shot variability of the oscillator. These studies included:

- (a) Varying the relative amounts of F_2 :Ar in the laser gas fill.
- (b) Varying the overall gas pressure by altering the amount of He in the gas fill.

- (c) Replacing up to 50% of the He by Ne.

The most satisfactory (energy and stability) operation was achieved using:

He/ F_2 : 5%	150 mbar
Ar	350 mbar
He	1300 mbar

The maximum energy output was 9 μJ , typical output was $\sim 1 \mu\text{J}$. We note that employing a Ne/He mixture rather than pure He as the buffer gas did not noticeably affect the operation of the line-narrowed ArF oscillator.

A detailed study of the spectral properties of the laser output from the arrangement shown in Figure B4.1 was undertaken. This study employed an EG&G PARC 1420, 1024 UV-sensitised diode array detector and a Rank Hilger Monospek 600 spectrometer equipped with a 2400 l/mm grating which was used in 3rd order. The line-narrowed spectral output from the oscillator was studied for 100 consecutive pulses using an ~ 1 hr-old laser gas fill. Figure B4.2 shows a typical line-narrowed output pulse (a), one of the most spectrally pure laser pulses (b) and the broad bandwidth output (c) which was obtained by blocking mirror M_1 in Figure B4.1.

From this study of spectral properties we found that:

- (a) The FWHM of the broad bandwidth output (Figure B4.2c) was 175 cm^{-1} .
- (b) The FWHM of the typical line-narrowed pulse (Figure B4.2a) was about 13 cm^{-1} , the narrowest linewidths observed (e.g. Figure B4.2b) were about 8 cm^{-1} .
- (c) The shot to shot shift in the position of the peak of the line-narrowed output was less than 3 cm^{-1} ($\sim$ the resolution of the detector arrangement).

It was observed that firing of the EMG150 amplifier in addition to the EMG150 oscillator prevented the oscillator from lasing on the majority of shots. We attribute this poorer oscillator performance to a

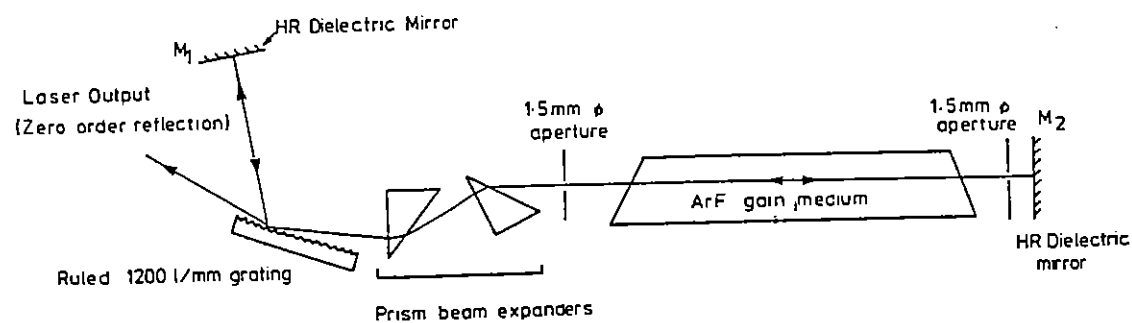


Fig. B4.1. ArF oscillator cavity.

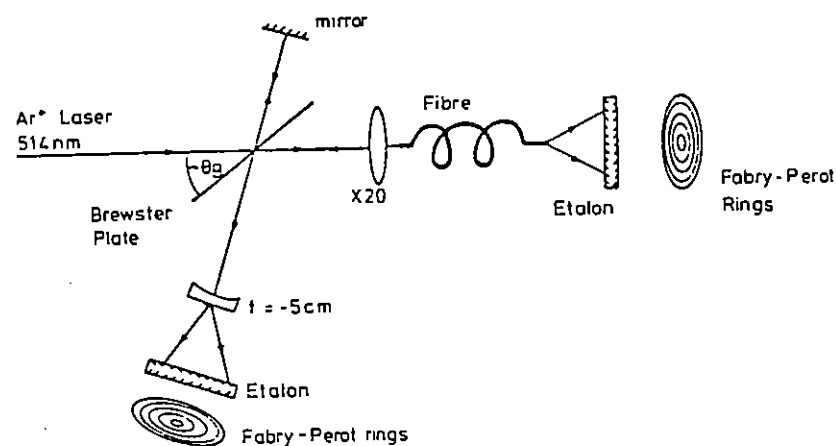


Fig. B4.3 Experimental arrangement.

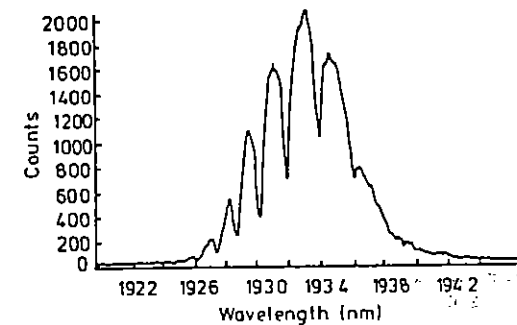
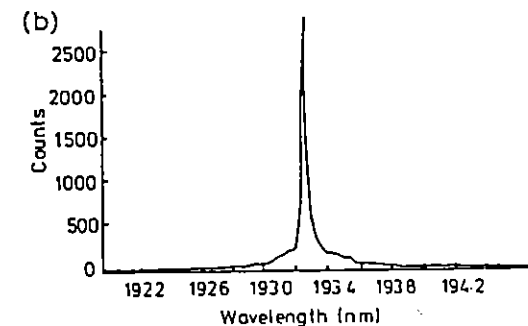
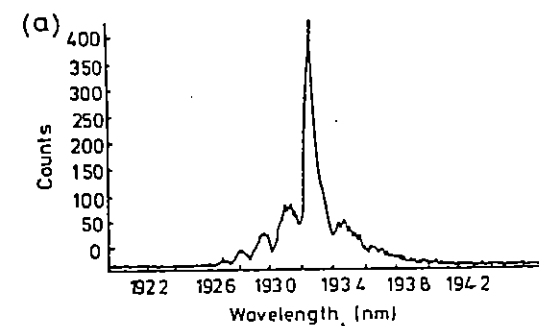


Fig. B4.2. Spectral profile of the oscillator radiation

- (a) typical line-narrowed output pulse
- (b) one of the most spectrally pure laser pulses
- (c) broad bandwidth output pulse.

reduction in the voltage applied to the oscillator when it is operated in conjunction with the amplifier compared to the voltage available when the amplifier is electrically disconnected as was the case for the experiments reported above.

From this work we conclude that the EMG150 laser is not suitable for reliable operation as a line-narrowed ArF oscillator-amplifier system.

B4-2 CW STIMULATED BRILLOUIN SCATTERING IN OPTICAL FIBRES

G M Davis and M C Gower (RAL)

Stimulated Brillouin Scattering (SBS) is a nonlinear optical phenomenon which usually requires extremely high laser beam intensities to be observable. However, since an optical fibre can maintain a high light intensity over a long distance, relatively small incident laser powers are required for the observation of SBS in a fibre. Hence optical fibres are an attractive medium for investigating SBS with relatively low power CW lasers.

We have performed a preliminary experiment to study the properties of CW backward stimulated Brillouin scattering in a monomode optical fibre.

The experimental arrangement employed in this initial investigation is shown in Figure B4.3. A single transverse and longitudinal mode Ar⁺ laser (Coherent Innova 90) operating at a wavelength of 514 nm was focused into a 250m length of monomode silica optical fibre (YORK VSOP) using a X20 microscope objective.

To observe the backscattered Stokes beam a fused silica plate was placed in the horizontally polarised laser beam and orientated close to Brewster's angle for this beam. For this orientation of the plate the vertically polarised component of the Stokes beam could be observed as a partially reflected signal from the Brewster plate while the reflection from the front surface of the fibre was primarily transmitted through the plate. To optimise the power of the backscattered Stokes beam careful launching of the pump beam into the fibre was required.

A Fabry-Perot (FP) interferometer was used to study the spectral content of both the light transmitted by the fibre and that reflected from the Brewster plate. The FP fringes were recorded on a 1-D diode array. Figure B4.4a shows the frequency components of the transmitted beam. In addition to a set of FP fringes corresponding to the transmitted pump beam a set of weaker fringes at a frequency corresponding to that of the backward Stokes beam is observed. We attribute this signal to the fraction of the backward travelling Stokes beam which has been reflected from the output coupler of the Ar⁺ laser and subsequently been coupled back into and transmitted back through the fibre.

To enable the backscattered signal reflected from the Brewster plate to be identified as the Brillouin Stokes shifted signal a mirror was used to supply a weak calibration beam at the pump frequency which was copropagating with the backscattered beam reflected from the Brewster plate. As shown in Figure B4.4b two sets of Fabry-Perot fringes were observed and the backscattered beam was shifted from the pump light by the backward Stokes Brillouin frequency shift of about 34 GHz.

The set of fringes at the pump frequency contains a small contribution due to reflection of the pump beam from the front surface of the fibre.

Observation of a backscattered signal required a threshold CW pump power incident on the fibre of 60 mW. For higher pump powers of about 200 mW we observed a maximum reflection of ~ 2% for backscattered light polarised in the vertical plane.

This preliminary experiment has shown that a CW single longitudinal and transverse mode Ar⁺ laser operating at a wavelength of 514 nm can generate SBS in a 250m length of monomode fibre for incident power levels greater than about 60 mW. Further experiments are planned which will study the generation of SBS in multimode fibres and enable the phase conjugate properties of the backwards SBS beam to be analysed.

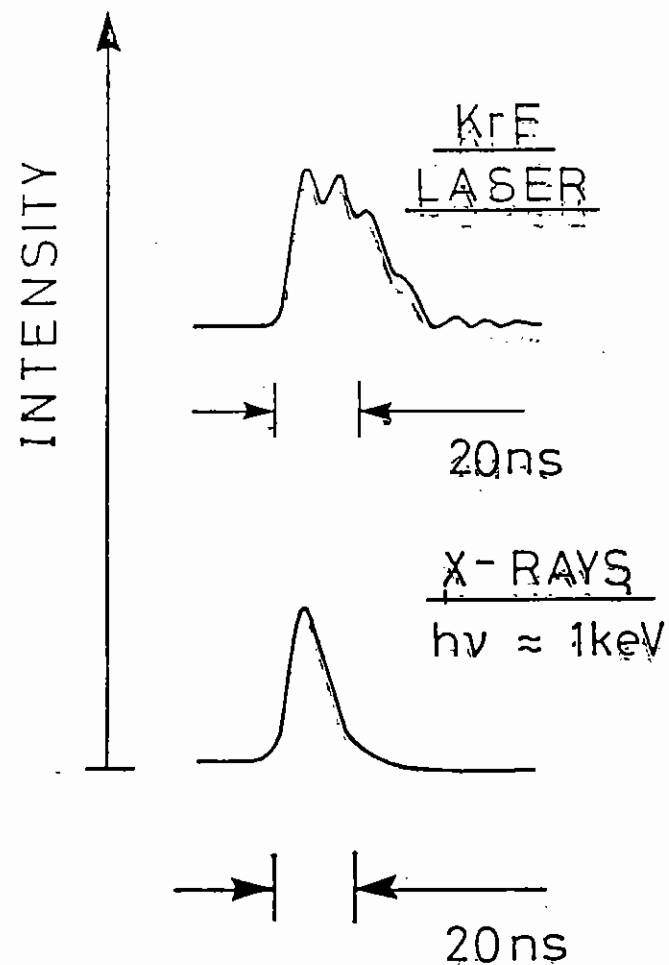
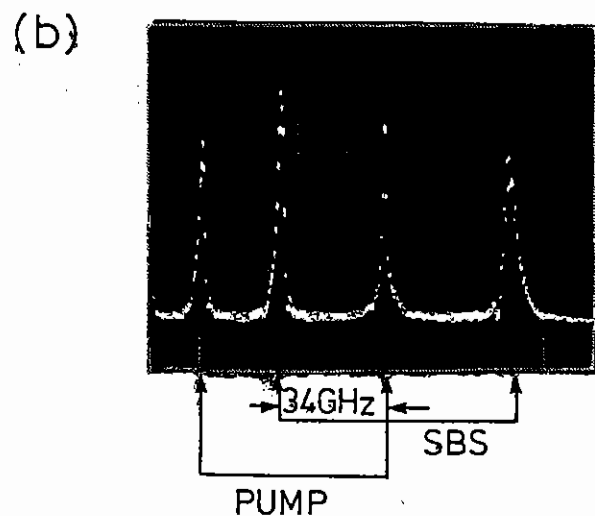
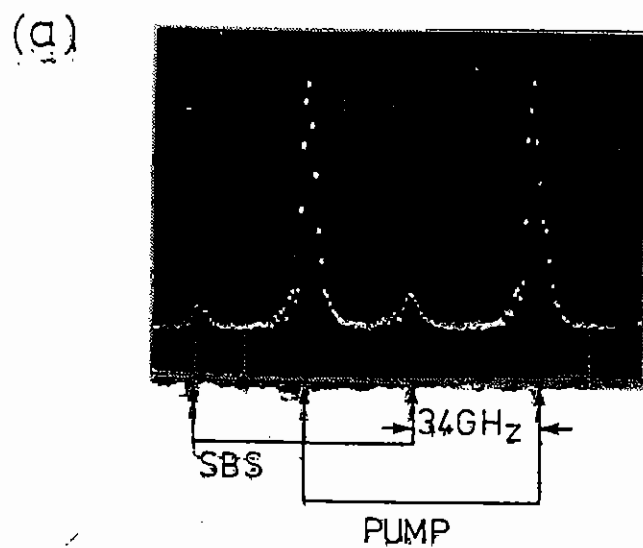


Fig. B4.5 Laser pulse and x-ray pulse. Oscilloscope traces not synchronised.

Fig. B4.4 Fabry-Perot fringes recorded on a 1-D diode array for (a) the transmitted and (b) the backscattered light.

B4-3 X-RAY LITHOGRAPHY USING A KrF LASER-PLASMA SOURCE

F O'Neill, G M Davis, M C Gower, and I C E Turcu (RAL)

In section C6-1 of the 1987 Annual Report we described the first experiments using a low energy (< 0.5 J) KrF laser to generate ≈ 1 keV x-rays from a laser plasma source for x-ray lithography. In those experiments the KrF laser irradiance on target was a maximum of 1.2×10^{12} W/cm² for the 25 ns laser pulse. The best conversion efficiency from laser light to 1 keV x-rays from a Cu target-plasma was 0.03%. While it was therefore shown possible to use a commercial excimer laser (λ -Physik EMG150) to generate 1 keV photons from a laser-plasma source the conversion efficiency was too low to be of commercial interest for x-ray lithography. 1 keV x-ray photons are required for this application in order to achieve efficient radiation transfer through the x-ray mask membrane (typically 1 - 2 μ m thick Si) onto the resist coated Si wafer.

We have performed some new experiments on this topic in present reporting period^(1, 2). The experimental arrangement was essentially the same as that shown in the 1987 Annual Report (see Figure C6-1, p271). The main improvement was to the laser performance i.e. by improving the resonator optics the EMG150 laser now produces a beam of divergence close to diffraction limit (≈ 30 μ Rad full angle). Thus the laser beam can now be focused to a smaller spot to give a higher irradiance of 2×10^{13} W/cm² on target. We have performed new measurements of conversion efficiency (KrF laser light $\rightarrow$ 1 keV x-rays) using Si pin x-ray diode detectors filtered by 10 μ m Be. A typical KrF laser pulse and plasma x-ray pulse are shown in Figure B4.5. It should be noted that the x-ray pulse is considerably shorter than the laser pulse an effect which places a major limitation on the conversion efficiency. Si pin diode measurements have also been used to measure the angular distribution of the emitted x-rays showing a $(\cos \theta)^{0.6}$ distribution. By carrying out a time integration and an angular distribution we can calculate the total amount of x-rays from the target. Dividing this number by the laser energy on target gives the conversion efficiency.

Figure B4.6 shows the new measurements of conversion efficiency at variable irradiances up to 2×10^{13} W/cm². With a Cu target the best

conversion efficiency is 0.2% and one measurement with a Ni target gives 0.3%. It should be noted that these values are approximately 50 times less than has been achieved in other experiments using a 0.5 ns, 15 J, 266 nm Nd:Glass laser focused to similar irradiances⁽³⁾. It is clear that the long duration KrF pulses (≈ 25 ns) are not optimum for x-ray generation mainly because the x-ray production terminates early (see Figure B4.5). Some more experiments are needed to fully explain the reason for this premature termination of x-rays and to see if techniques can be developed for preventing it.

The plasma x-ray source described above is clearly not as efficient as the sources generated using high energy short pulse Nd:Glass lasers. The source generated by the KrF laser is interesting however because it can be operated repetitively by using a rotary target system such as shown in Figure B4.7.

In order to demonstrate that the KrF laser-plasma x-ray source described above is practically useful for lithography we have made trial exposures using the positive e-beam resist EBR-9. These exposures were made by operating the laser at 2Hz repetition rate and focusing it onto a rotating target made from a silver coating on a steel rod. For this repetitive target it was necessary to place a quartz protection plate between the $f = 9$ cm lens and the target to prevent the accumulation of target debris on the lens surface. Even though this plate was made as thin as possible ($t \approx 1.5$ mm) it still introduced a severe aberration on the focused laser beam thus reducing the x-ray emission from the target by a factor of 5 from that obtained without the plate. This effectively made the resist exposure time a factor of 5 longer than necessary.

A fine line exposure into resist was made using the experimental arrangement shown in Figure B4.8. A 6.5 μ m thick Be foil (e^{-1} transmission at $h\nu \approx 0.9$ keV) and a free-standing gold (Au) diffraction grating⁽⁴⁾ are used to simulate a 2 μ m thick Si-substrate x-ray mask structure and the x-ray exposure is made in a 0.9 μ m thick layer of EBR-9 resist. The resist was spun onto a glass plate substrate and baked for 30 mins at 200°C. After exposure to x-rays the resist was developed for 5 mins in MIBK. Because of the uncertain flatness of the Au grating we chose to employ a contact exposure technique. An SEM

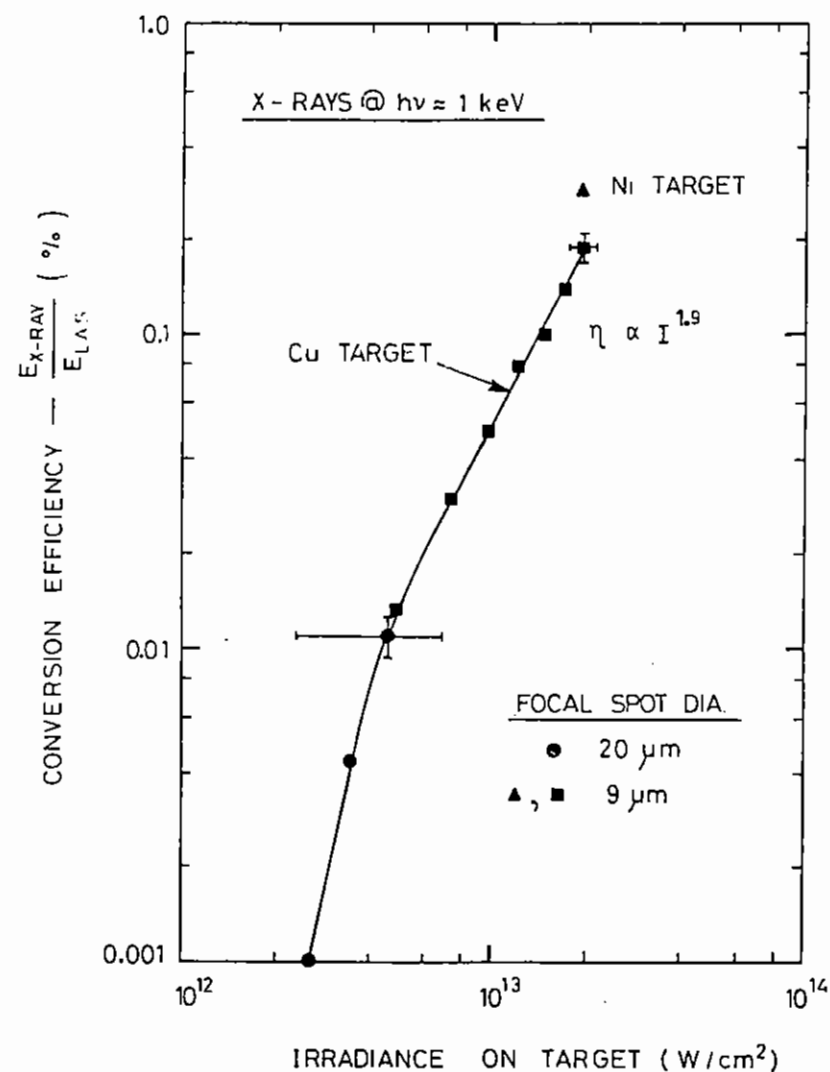


Fig. B4.6 Conversion efficiency to keV x-rays as a function of KrF target irradiance. Circles are data from 1987 Annual Report. Squares and triangle are new data with improved laser beam focussed to a smaller spot on target.

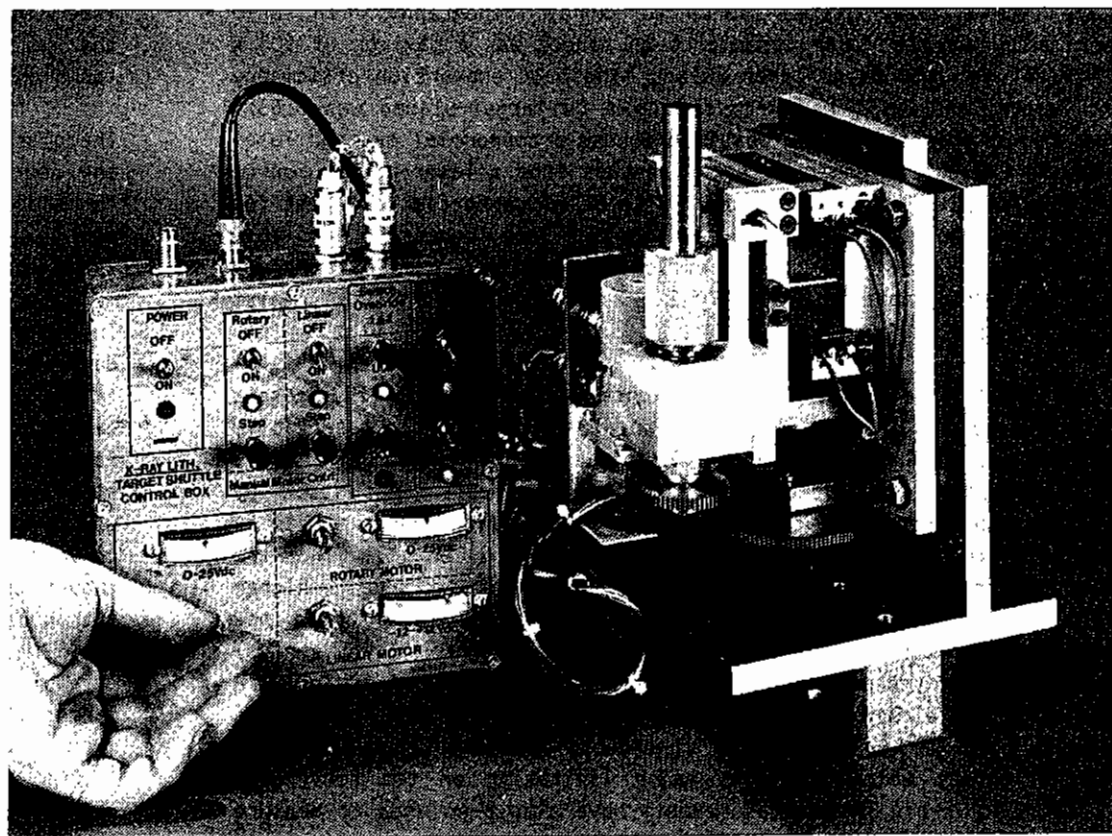


Fig. B4.7 Rotary Cu target system used for x-ray exposure of resist.

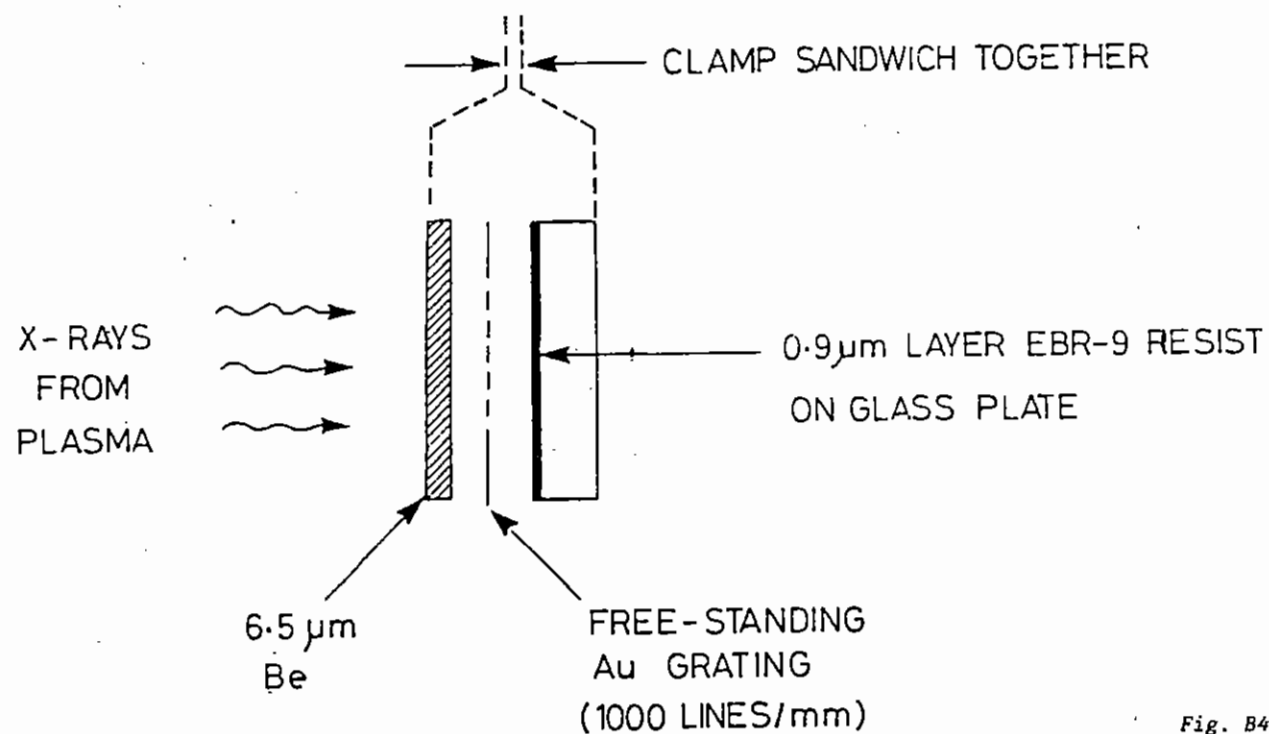


Fig. B4.8 Experimental arrangement used to make a fine-line contact exposure into x-ray sensitive resist.

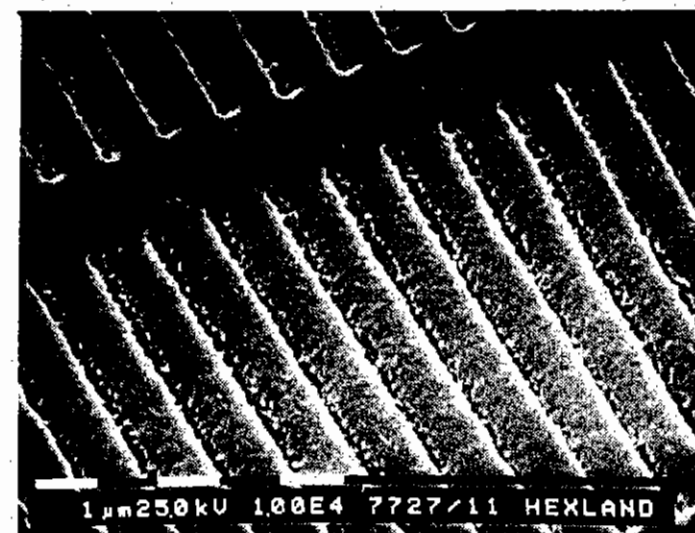


Fig. B4.9 SEM picture of Au grating lines replicated into EBR-9 resist. Image recorded using SEM beam at $\approx 30^\circ$ angle of incidence.

picture of an x-ray exposure of the grating into the resist is shown in Figure B4.9. To make this image the resist was placed a distance of 1 cm from the x-ray source and the exposure required 3000 laser shots (25 mins running at 2Hz repetition rate) which delivered an estimated x-ray flux of 130 mJ/cm² onto the resist. The lines of the Au diffraction grating are etched steeply into the developed resist and the exposure has gone the full way through the 0.9 μ m thick layer. The SEM picture in Figure B4.9 displays all the attractive features of x-ray lithography even for the present non-optimum set-up. In particular straight, high-aspect-ratio, sub-micron lines have been replicated into a single layer resist of $\approx$ 1 μ m thickness under non-clean-room conditions.

In conclusion we can say that the above results have demonstrated that commercially available injection-locked low energy KrF lasers can be used to generate 1 keV plasma x-ray sources suitable for x-ray lithography applications using Si masks with 1 - 2 μ m thick substrates. However the best conversion efficiency to keV x-rays is only $\approx$ 0.3% from Ni targets as compared to the 5-10% values obtained using high power 0.5 ns Nd:Glass lasers. The data displayed in Figure B4.6 shows however that the x-ray conversion efficiency is increasing in proportion to the square of the incident laser irradiance. It will be interesting therefore to carry out conversion efficiency measurements using higher energy KrF laser pulses and some data of this nature has been recently recorded using the high energy Sprite laser (see Section A1-13 of this Annual Report).

References

1. F O'Neill, G M Davis, M C Gower, I C E Turcu, M Lawless, and M Williams, Proc SPIE, 831 (to be published).
2. E Turcu, G Davis, M Gower, F O'Neill, and M Lawless, Microelectron. Eng., 6, 287 (1987).
3. M Chaker, H Pépin, V Bareau, S Boily, B Lafontaine, R Fabbro, I Toubhans, B Faral, J F Currie, D Nagel, and M Peckerar, Proc SPIE, 733, 58 (1986).
4. Au transmission diffraction grating supplied by Dr K Eidmann, Max-Planck-Institut für Quantenoptik, D-8046, Garching, W Germany.

B4-4 PLASMA X-RAY SOURCE GENERATION USING μ J-LEVEL 100 psec XeCl LASER PULSES

F O'Neill, I C E Turcu, C Hawker (RAL)
M H R Hutchinson, D Xenakis (Imperial College).

Plasma x-ray source generation using sub-nanosecond laser pulses is of interest for a number of reasons. For example by using pulses of duration < 100 psec it is possible to achieve sufficient irradiance on target ($> 10^{13}$ W/cm²) for x-ray generation by using tightly focused beams of only 1 - 10 mJ energy/pulse. Also the plasma x-ray pulses produced by such a laser will be of short duration and will be useful for measuring the response time of x-ray detectors. Finally the use of sub-nanosecond duration excimer laser pulses (instead of the normal 25 ns pulses) might give improved x-ray generation at photon energies > 1 keV which is of interest for a number of applications.

In section C6-2 of the 1987 Annual Report we described the first work on a project at Imperial College to study laser-plasma x-ray source generation using low energy 100 psec XeCl laser pulses. In that report we described the construction of a 100 psec, 308 nm laser oscillator capable of generating 10 - 50 μ J pulses at 0.5 Hz repetition rate. The experiments in the present reporting period have been concerned with amplifying these low energy pulses to ≈ 5 mJ energy in an XeCl laser amplifier. Also these amplified pulses have been used with an RAL transportable target chamber for some preliminary target experiments to make plasma x-ray sources.

The short pulse amplification experiments have been performed using the experimental arrangement shown in Figure B4.10. The μ J-level, 100 psec pulse from a frequency doubled excimer-pumped distributed feed-back dye laser (see Fig. C6-3, p273, 1987 Annual Report for details) is amplified to a level ≈ 5 mJ by triple passing it through an XeCl laser amplifier (Lumonics Model TE-860). Considerable care has been taken to achieve a good beam quality and the amplified beam is of 7 mm dia. with a beam divergence $\approx 3 \times$ times diffraction limited.

For target experiments these pulses are focused at normal incidence onto 0.5 mm dia. copper wires to excite L-shell emission at $h\nu \approx 1$ keV using the arrangements shown in Figure B4.11. The focusing lens is an aspheric fused silica singlet of 9 cm focal length and the laser spot size on target is $\approx 10 \mu$ m.

A typical laser pulse energy on target is $\approx 2 - 4$ mJ resulting in an irradiance $\approx 2 - 4 \times 10^{13}$ W/cm². The low rep. rate of the laser (0.5 Hz) means that a number of pulses can be fired sequentially onto the same spot on target and x-rays can be measured from each individual laser pulse. X-rays from the laser-produced plasma are measured using a Si pin diode detector placed at approximately 45° to the incident laser beam and 4 cm distance from the target. The pin diode is filtered by a 6.2 μ m Be filter (e^{-1} transmission at $h\nu \approx 0.9$ keV) of aperture 5 mm dia. This filtered detector is therefore mainly sensitive to x-rays at photon energies ≈ 1 keV from the Cu plasma.

The most obvious effect in measuring the x-ray pulses has been the change in x-ray signal for sequential shots incident onto the same spot on target. Figure B4.12 shows a set of oscilloscope displays of laser pulses (first on each trace) and x-ray pulses (second on each trace) to demonstrate this effect. The time width of the displayed laser pulses are limited by the response time of the detector/oscilloscope combination the time pulse-width being ≈ 100 psec. Also the time-width of the x-ray pulses are limited by the response of the pin diode. The x-ray pulses are expected to be ≈ 100 psec duration.

Figure B4.12(a) shows the typically low plasma x-ray signal at $h\nu \approx 1$ keV for the first laser pulse incident on a fresh Cu target surface. Figure B4.12(b) shows the much larger x-ray yield obtained from the second laser pulse on the same target spot. In this case the second laser pulse is incident into the small (≈ 50 μ m deep) target crater made by the first pulse. The crater is probably causing some plasma confinement effect thus increasing the temperature and enhancing the x-ray output. For the third (Figure 3(c)) and subsequent laser shots on the same spot the x-ray output drops off again. The difference between x-ray signals was not always as pronounced as shown in Figure 3 but the second laser shot consistently produced the most x-rays.

The x-ray signal displayed in Figure B4.12(b) has been used along with pin diode sensitivity data to calculate the conversion efficiency to ≈ 1 keV x-rays. We estimate the conversion efficiency to be $\approx 0.1\%$ sr⁻¹. These experiments are continuing in order to study x-ray conversion efficiency using higher energy 100 psec laser pulses and also to study the effect of prepulses.

B4-5 A NOVEL SINGLE SHOT CROSS-CORRELATOR FOR MEASURING PULSE LENGTHS AT 249nm

I N Ross, D Karadia and J M Barr

B4.5.1 Introduction

There has been much interest in the generation of ultra-high power picosecond pulses in the ultraviolet using excimer laser amplifiers (B4.5.1-B4.5.9). A necessary part of these programmes is to make measurements on pulse duration and pulse temporal profiles, where possible, and excellent techniques are now available for quasi-cw pulse trains and single pulses at visible wavelengths (B4.5.10-B4.5.16). The absence of appropriate second harmonic crystals has prevented the direct extension of these techniques into the ultraviolet. A two photon fluorescence technique (B4.5.17) has been demonstrated successfully but there is still room for a more sensitive, and background free device.

B4.5.2 Cross-correlator

The proposed technique is one way of extending the correlator-type technique into the ultraviolet while still making use of standard crystals such as KDP. The technique is possible because the source of pulses for most high power ultraviolet systems operates at a visible wavelength which is then converted to the desired ultraviolet wavelength by a suitable nonlinear process. We make use of these synchronous visible pulses (which can be measured using a standard autocorrelator) by mixing them with the ultraviolet pulse in a parametric crystal and generating the difference frequency (B4.5.7). This difference frequency signal as a function of time difference at the crystal represents a cross-correlation between the two input pulses and provides information on the pulse duration of the ultraviolet pulse.

The difference frequency signal under conditions of low efficiency, plane waves and exact phase matching is given by (B4.5.18):-

$$I_{\omega_3} = \text{Const } I_{\omega_1} I_{\omega_2} l^2$$

where I represents the intensity of the signals

l is the interaction length

$$\omega_3 = \omega_1 - \omega_2$$

Thus the signal $S(\tau)$ corresponding to a time difference τ is given by:

$$S(\tau) = S(0) \frac{\int I_{\omega_1}(t) I_{\omega_2}(t-\tau) dt}{\int I_{\omega_1}(t) I_{\omega_2}(t) dt}$$

There is an interesting feature of the cross-correlator, if the visible (ω_2) pulse is made short compared to the pulse to be measured, for then the cross-correlation signal is given to a good approximation by:

$$S(\tau) = \text{const} \times I_{\omega_1}(\tau)$$

and we have a direct measurement of intensity distribution in the ultraviolet pulse.

The intensities of input beam required to generate sufficient signal to drive a diode array are of the order of 10 to 100 MW/cm². This corresponds to 0.1 to 1 μJ in 1 ps on a focal line of 10 μm by 10 mm and the technique can therefore be regarded as having a good sensitivity. The method for generating the full pulse profile on a single shot is to introduce a linearly variable delay across one beam with respect to the other at the crystal where both beams are brought to a line focus. A linear diode array samples the intensity variation along the generated difference frequency line this intensity being the cross-correlation between the two input beams. The technique is a 'background free' technique whether we use collinear or noncollinear geometry, since there is of course no difference frequency signal unless the two input beams are synchronous and there is no difficulty in discriminating

between the output beams using appropriate spectral or spatial filters. It is desirable to separate the three wavelengths after the crystal and measure the linear intensity distribution on each using three diode arrays. By so doing it is possible to normalise out the spatial variations of intensity on the input beams.

Two techniques are available to generate the linearly variable delay across one beam. A noncollinear geometry automatically does this but is restricted to shorter pulses since there is a limit to the angle between beams while still retaining reasonable conversion efficiencies. A dispersive element in one beam is the second technique. This introduces the delay across the beam through group velocity dispersion. We have chosen to use a grating since it can give large delays across modest size beams. For example a grating of 3600 lines/mm operating in first order at 249 nm gives a variable delay of 3 ps per millimetre across the beam. When using a grating it is important to minimise the spectral dispersion effects to prevent a significant blurring of the delay across the beam. This implies that the mixing crystal must be close to the grating, or to an image plane of the grating.

B4.5.3 Experimental demonstration

A diagram of the correlator is shown in Figure B4.13. Input beams of diameter about 6 mm are focussed into coincident line foci using two cylindrical lenses. Both the focussing and noncollinear angles were in the non critical phase matching direction of a type I KDP crystal cut at 80° to the crystal axis. A focussed line width of approximately 10 μm with a noncollinear angle of 3 degrees implied an interaction length in the crystal of about 200 μm. With this geometry the effects of double refraction and dispersive pulse stretching were negligible. A 3600 lines/mm grating tilted about a horizontal axis to operate in first order was used to generate a variable time delay across the ultraviolet beam. A typical diode array trace is shown in Figure B4.14(a) recorded using about 10 μJ pulses in each beam. The time scale along the diode array was calibrated by noting the change in position of the pulse peak for a known movement of the variable delay prism. Figure B4.14(b) was recorded by then tilting the grating back to operate as a simple mirror (in zero order) thereby generating a normalising trace with no relative time delays. An autocorrelator monitored the average pulse profile of the long wavelength at 746 nm and, for the example presented, gave a

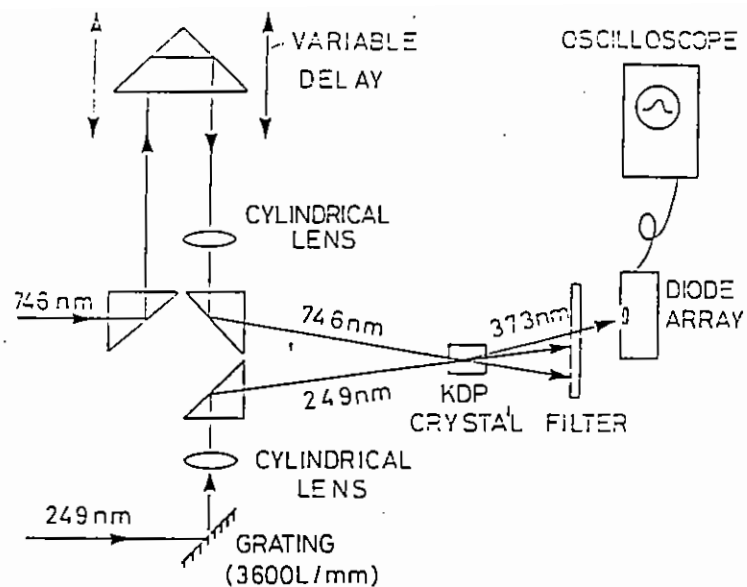


Fig. B4.13 Schematic of single pulse correlator.

(a) With Dispersion (Grating First Order)
in 249nm Beam



(b) Without Dispersion (Grating Zero Order)

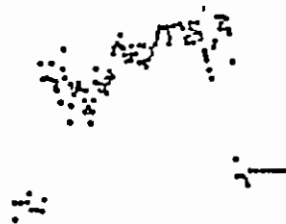
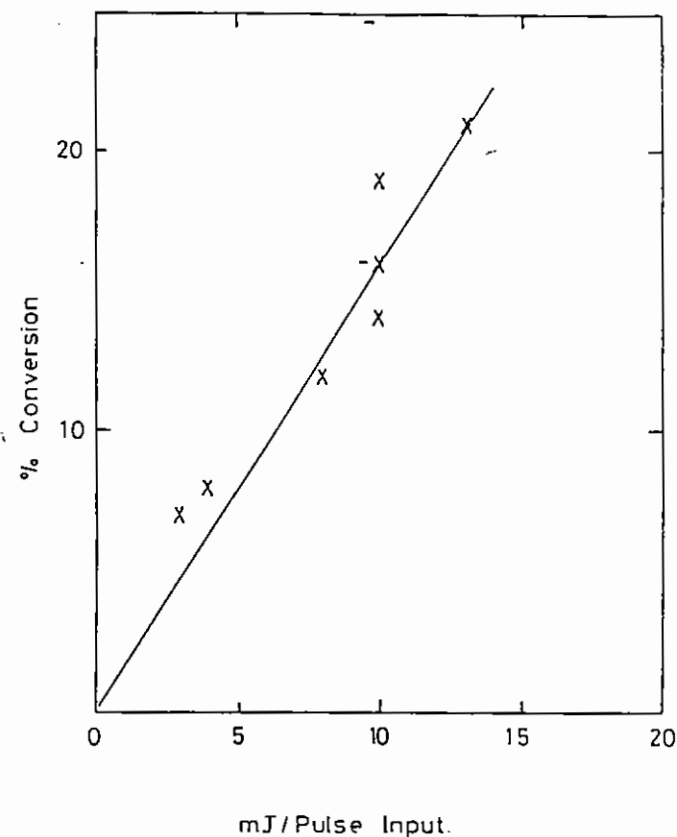


Fig. B4.14 Diode array traces from single pulse correlator

- a) with dispersion giving a linear time sweep along the array
b) without dispersion and no time change along the array



B4.15 Figure illustrating the measured BBO conversion efficiency for various input energies.

FWHM trace of 3.8ps with a close to transform limited pulse shape. A number of traces recorded as for Figure B4.14(a) were measured giving a FWHM of 4.9 ± 0.6 ps. These numbers would suggest, for gaussian shaped pulses, a width of 2.7ps for the 746nm beam and a width of 4.1ps for the 249nm beams.

References

- B4.5.1. P B Corkum, R S Taylor, IEEE J Qu Elect, **QE-18**, 1962 (1982).
- B4.5.2. P H Bucksbaum, J Baker, R H Storz, J C White, Opt Lett, **9**, 399 (1982).
- B4.5.3. H Egger, T S Luk, K Boyer, D F Muller, H Pummer, T Srinivasan, C K Rhodes, Appl Phys Lett, **41**, 1032 (1982).
- B4.5.4. S Szatmari, F P Shafer, Opt Comm, **48**, 279 (1983).
- B4.5.5. S Szatmari, F P Shafer, Appl Phys, **B33**, 219 (1984).
- B4.5.6. J H Glowina, G Arjavalingam, P P Soirokin, J E Rothenberg, Opt Lett, **11**, 79 (1986).
- B4.5.7. A P Schwarzenbach, T S Luk, I A McIntyre, U Johann, A McPherson, K Boyer, C K Rhodes, Opt Lett, **11**, 499 (1986).
- B4.5.8. S Szatmari, F P Shafer, E Muller-Horsche, W Muchenheim, Opt Comm, **63**, 305 (1987).
- B4.5.9. A Endoh, M Watanabe, S Watanabe, Opt Lett, **12**, 906 (1987).
- B4.5.10. E P Ippen and C V Schank, Ultrashot Light Pulses, Springer-Verlag, Berlin, **83**, (1977).
- B4.5.11. J M Diels, J J Fontaine, I C McMichael and F Simoni, Appl Opt, **24**, 1270, (1985).
- B4.5.12. J Jansky, G Corradi and R N Gyuzalian, Opt Comm, **23**, 293, (1977).
- B4.5.13. R N Gyuzalian, S B Sogomonian and Z Gy Horwarth, Opt Comm,
- B4.5.14. R Wyatt and E E Marinero, Appl Phys, **25**, 297 (1981).
- B4.5.15. J Jansky, G Corradi and R N Gyuzalian, Appl Phys B, **33**, 79, (1984).
- B4.5.16. Y Ishida, T Yajuna and A Watanabe, Opt Comm, **56**, 57, (1985).
- B4.5.17. M H R Hutchinson, I A McIntyre, G N Gibson and C K Rhodes, Opt Lett, **12**, 102, (1987).
- B4.5.18. R G Smith, Laser Handbook, North-Holland, Amsterdam, **837**, (1972).

B4-6 ULTRA-VIOLET GENERATION BY FREQUENCY DOUBLING USING β -BARIUM BORATE

A W Parker and I N Ross

The new non-linear optical material beta-barium borate, β -BaB₂O₄ (BBO) was tested at the RAL during this last year. BBO has several properties which make it very suitable for harmonic generation of laser light, particularly at ultra-violet wavelengths between 205 and 300 nm. Its special properties include;

- (i) High damage threshold, ~ 13.5 GW/cm²
- (ii) Large temperature bandwidth of 55°C, this means change in tuning angle with temperature is extremely small (c.f. KDP 6.7°C). Thus laser induced heating does not effect the conversion efficiency.
- (iii) Large transmission range - 189-3500 nm
- (iv) Very high conversion efficiency

The BBO crystal studied at RAL has a phase match angle of 62° with a tuning range specification of 440-480 nm (ie doubling to 220-240 nm). Figure B4.6.1 below illustrates the variation of conversion efficiency with energy input. The results were obtained using a Lumonics 460X

laser (308 nm, 10 ns pulse width) pumping a Lambda Physik FL3001/2 dye laser. The output laser beam area was $\sim 3 \text{ mm}^2$ and maximum input energy of 13 mJ/pulse was used giving a maximum intensity of 110 MW/cm^2 .

The shortest and longest obtainable wavelength from the 62° crystal were 215 nm and 290 nm respectively. Beyond this the necessary tuning angle was too great to allow the laser beam to emerge from the back surface of the crystal.

A direct comparison for the conversion efficiencies between KDP and BBO was made using the dye-amplified mode-locked Spectra Physics Nd:YAG-pumped dye laser system. The system produced 581 nm laser light, the longest wavelength doubled. The conversion values for KDP and BBO respectively were 40 μJ (13%) and 60 μJ (20%) for a power density of $\sim 4 \text{ GW/cm}^2$ from 4 ps pulses and using a 6 mm long BBO crystal and a 22 mm long KDP crystal.

Uses of BBO

One of the main areas of use for BBO at RAL so far has been to produce tunable UV-laser light suitable for Resonance Raman spectroscopy. Prior UV-sources were line narrowed 248 nm pulses from the EMG-150 excimer laser. A small amount of wavelength selectivity could be made using a H_2/D_2 Raman 8 shifter. Whilst the present crystal mounting does not allow continuous scanning of wavelengths, resonance Raman excitation profile experiments have been performed albeit needing intensity correction and a minor optical realignment for the various wavelengths selected (see section B1.1). A custom built BBO crystal and housing to fit the FL3001/2 dye laser with beam compensator has now been ordered and this will allow continuous scanning.

The Raman spectra obtained using the doubled laser light are both more intense and have improved resolution. The latter being a consequence of the narrower, more stable, laser line width ($\sim 0.4 \text{ cm}^{-1}$).

Further possibilities

Attempts have been made at RAL to line narrow 193 nm pulses from an EMG150 (ArF) laser. This has met with some difficulty, see section B4.1, and we believe it possible to produce this wavelength by frequency mixing within a BBO crystal. The 193 nm output could be used

to pump an ArF laser. Recently the literature reports frequency mixing of two pulsed dye lasers using BBO to produce 189 nm pulses (B4.6.1). The wavelengths mixed were 788 and 248.5 nm.

Conclusion

The wide tuning range of BBO combined with its efficient harmonic generation will be of great use as a tunable UV laser light source. For Raman spectroscopy this UV-light has been found to be more useful to that obtained using the EMG-150 line narrowed KrF pulses.

The typical working energy produced from the Lumonics/dye laser system between 215 and 270 nm is $\sim 4 \text{ mJ/10 ns pulse}$.

References

B4.6.1 Lasers and Optonics 47, December (1987).

B5	LASER SUPPORT FACILITY OPERATIONS AND DEVELOPMENT	Pages
B5-1	RAL Based Operations	263
B5-2	Loan Pool Operations	263 - 264
B5-3	Charge Coupled Device Detectors for Raman Spectroscopy	264 - 268
B5-4	Single Photon Counting	268 - 270
B5-5	Developments in Pump-Probe Experiments	270
B5-6	Streak Camera Developments	270 - 275

Editor: W T Toner

B5.1 RAL BASED OPERATIONS

a. Nanosecond Programme

A full programme of experiments was run in laboratories A and B using the (HX460) excimer pumped (FL3002) dye laser, the EMG150 excimer laser and the Raman spectrometer system. The HX460/FL3002 had no significant down-time. The annual overhaul of the EMG150 was carried out successfully but subsequent problems with a cracked insulating body and eroded pre-ioniser pins necessitated a second rebuild, carried out in "Division Head's" time. The laser is now working well. The time required to strip and rebuild the laser is now down to one week. Line narrowed performance was investigated by Dr Gower and improved optics have been installed for use at 248nm. The performance of this laser on line narrowed ArF (193nm) is, however, marginal and its electrical/mechanical design precludes switching oscillator and amplifier functions to obtain better injection locking.

Equipment purchases for this year were:

i) An OMA III (optical multichannel analyser) console to replace/update the OMA II system which was beginning to show signs of age. The system may be used to drive the videcon detector or the 1420 gatable intensified diode array. The console has two notable extras, Hemingway BASIC software for user routines and an I/O-B module consisting of an 8-bit programmable bidirectional I/O port. The latter comes with a centronics and an RS232 serial port and provides an additional 512K bytes of memory. The 1420 detector is having a new intensifier fitted to replace one damaged by over exposure to light. The repaired detector is to be returned in early 1988.

ii) A number of the new β -barium borate (BBO) crystals (see section B4.6) have been ordered. The crystals give very efficient second harmonic generation of laser light and have been shown, at RAL, to have great potential as tunable UV (200-300nm) Raman excitation sources. A total of 5 crystals have been ordered (due February 1988) allowing the doubling of 400-1300nm wavelengths.

There were no funds for the purchase of new lasers.

Development of the DSMA system has continued. Last years annual report gave details of problems encountered when spectral intensities could only be assigned interger values giving noisy spectra. The fault has now been rectified with the introduction of "real number" spectral accumulation. To further improve S/N we now recommend the use of gated mode when using pulsed lasers.

Five of the seven mirrors in the Triplemate spectrometer were re-coated at York with "UV enhanced" overcoated aluminium. Throughput in the 230nm region has been improved by a factor of five.

b. Picosecond Programme

The picosecond laser operated throughout the year with no breakdowns and with the CW modelocked YAG pumping the scheduled laser in lab D and the development laser in lab E in parallel. The laser has now operated for nearly 8000 hours since its delivery. Most experiments used the 10Hz YAG-pumped amplifier and nominal 3-nsec pulses but 500fsec pulses and also excimer amplified pulses were used for some experiments (B3.5, B5.5). Improvements to the pump and probe system are reported in B5.5 and to the streak cameras in B5.6. Streak camera and readout reliability is still a problem. A single photon counting system is now being commissioned (B5.4).

B5-2 LOAN POOL OPERATIONS

A J Langley (RAL)

The LSF Laser Loan Pool has now been operating for three years providing laser systems and diagnostic equipment to users in their own laboratories. Since the scheme began 26 user groups in 18 academic institutions, from as far apart as Edinburgh to Southampton, have borrowed lasers from the pool. During the past year alone, 20 laser and 5 diagnostic loans were made to 17 institutions. Table 1 shows the distribution of loans (one loan = 13 weeks) between chemistry, physics and biology departments and the total number of loans per laser in the past 12 months. The Table clearly shows that more than half the users were from chemistry departments. Interestingly, for the first time, more physicists than biologists have used the loan scheme. Demand remained high for high-energy, pulsed Nd:YAG pumped dye lasers, with

Table 1.

Laser and diagnostic loans between March 1987 and March 1988

Laser System	Academic Department			Total No of Loans
	Chemistry	Physics	Biology	
LL1 JK2000 Nd:YAG + Dye laser 400-700nm	1		3	4
LL2 Quantel Nd:YAG + Dye laser 220-2000 nm	3	1		4
LL3 JK HY750 Nd:YAG + Dye laser 220-2000 nm	3	1		4
LL4 λ -Physic Cl excimer + & FL2002 Dye Laser LL5 220-700 nm	2	1		3
LL6 Spectra-Physics CW argon-ion + dye laser 500-700 nm	1	2	1	4
LL7 Lumonics Fluoride excimer 193, 248	1			1
Diagnostic systems LD1 Imacon 500 streak camera (used at RAL)				0
LD2 S.R & LD3 Gated Integrators	4	1		5
Total	15	6	4	25

tunable UV and IR options, and for the CW argon-ion/dye laser system. In total 12 papers were published by loan pool users in 1987 and their scientific results are presented in this annual report in sections B1.11, B1.12, B1.14-B1.22, B2.6-B2.7.

During 1987 there were some additions to the loan pool. The Lumonics HX460 fluoride laser was commissioned and despite some initial problems with laser powers and poor passivation, which still need to be resolved, the laser was considered to be working sufficiently well to be loaned to Bristol University during the Autumn. This laser weighs nearly half a ton and the considerable transportation problems were overcome by the construction of a hydraulically height adjusted trolley. Also commissioned in 1987 was a Quanta-Ray PDL-2 dye laser to be pumped by the JK HY750 Nd:YAG. These lasers have proved to be a reliable combination giving users access to ~ 50 mJ (8 ns)/pulse in the red visible region and $\sim 1-5$ mJ tunable radiation in the UV and near IR. Another development was the installation of a line narrowing etalon on the Quantel Datachrome 5000 Nd:YAG. This has reduced the linewidth of the 1064 nm fundamental from 0.7 cm^{-1} to $\leq 0.04 \text{ cm}^{-1}$. Linewidths of the order of 0.15 cm^{-1} rather than 0.75 cm^{-1} can now be achieved in the UV when mixing the 1064 nm with doubled dye laser output.

B5-3 CHARGE COUPLED DEVICE DETECTORS FOR RAMAN SPECTROSCOPY

K P J Williams (BP Research Centre)

Abstract

The Charge Coupled Device (CCD) has been used for low light measurements in astronomy for the past decade. Recently, it has been applied to Raman spectroscopy. This paper details the performance of one such detector cooled to liquid nitrogen temperature. A comparison between this CCD and a widely used intensified diode array detector (Spectroscopy Instrument) located at the RAL is made. The data indicate that the CCD is superior both for sensitivity and detector noise levels.

Introduction

Detector technology for Raman Spectroscopy has improved slowly over the past decade. Good photomultiplier tubes have been available with relatively low (~ 12 pcs) dark currents for more than a decade. Multichannel intensified diode array detectors have become more reliable with some advance being made in double intensified diode arrays. Some of the main aims of a Raman spectroscopist when selecting his perfect detector is for 100% quantum efficiency, no dark current or readout noise and for an infinite dynamic range. The detectors mentioned above fall somewhat short of this but are nevertheless adequate.

The CCD detector is a relatively new advance for the Raman spectroscopist. The work of Murray and Dierker⁽¹⁾ first really made people aware of its potential. An advance, of interest to us, was to compare the performance of the intensified diode array with that of a cooled CCD. This report details the data obtained at RAL from which a comparison between a cooled CCD system, supplied by Wright Instruments Ltd, and an intensified diode array system (Spectroscopy Instruments) can be made.

Experimental

The theory of operation of the CCD detector has been dealt with in a comprehensive review elsewhere⁽²⁾. The cooled (77 K) CCD detector supplied by Wright Instruments comprised the following specification;

Resolution - 600 x 400 pixels

Pixel size - 25 μ m

Quantum efficiency - Greater than 15% from 400 - 900 nm (40% at peak)

Readout noise - 7 electrons RMS

Readout time - 100 μ sec/pixel

Saturation level - 3×10^5 electrons/pixel

The CCD was controlled from an IBM AT computer the software for which was adequate, although appeared rather awkward to use. The specifications can be contrasted with those from a typical intensified diode array system. In brief the quantum efficiency is $\sim 15\%$ (peak), readout noise ~ 1000 electrons, saturation level $\sim 5 \times 10^3$, pixel size 25 μ m x 2 mm, resolution upto 1000 pixels in linear array.

Raman spectra were recorded using 514.5 nm laser excitation from a Coherent Innova 90 argon laser and a Spex Triplemate spectrometer. All instrumental parameters, data acquisition time, laser power and sample alignment remained unchanged during the assessment of the two detectors.

Results and Discussion

Spectra from a medium strength Raman scatterer were obtained. Figure B5.1 illustrates the spectrum recorded from a polyethylene sample, using ~ 100 mW of laser power and an exposure time of 100 ms, with the cooled CCD system. Figure B5.2, on the other hand, is the spectrum recorded under identical conditions using the intensified diode array system. The difference in spectral width seen by changing over detectors reflects the different physical size of the detection elements (8 mm for the CCD and ~ 17 mm for the intensified portion of the diode array). A comparison of the signal to noise ratio provides an estimate of the superior sensitivity of the CCD system. The S/N ratio from Figure B5.1 is $\sim 14:1$ whereas from Figure B5.2 is only 6:1, as measured from the most intense peak. Accordingly, CCD appears to produce data that is approximately twice the quality of a diode array under the same conditions.

An experiment, designed to test the capability of the systems for detecting very weak signals, was arranged whereby a mercury lamp was positioned below the entrance slit of the spectrometer. The entrance slit was closed to an extent that only minimal amount of light (at 546 nm) passed into the system. The result obtained using the CCD detector is shown in Figure B5.3. This was obtained after ~ 2 minutes accumulation time. The data obtained from the diode array is shown in Figure B5.4(a) and essentially is only noise with no peak detected. Figure B5.4(b) was recorded with the first slit of the spectrometer open wider and is shown for reference.

This test is conclusive and is in favour of the CCD for ultimate sensitivity. As one can see the mercury line recorded on the CCD detector is distinct with a S/N $\sim 2:1$. However, with the same light level on the diode array detector there is no discernable signal.

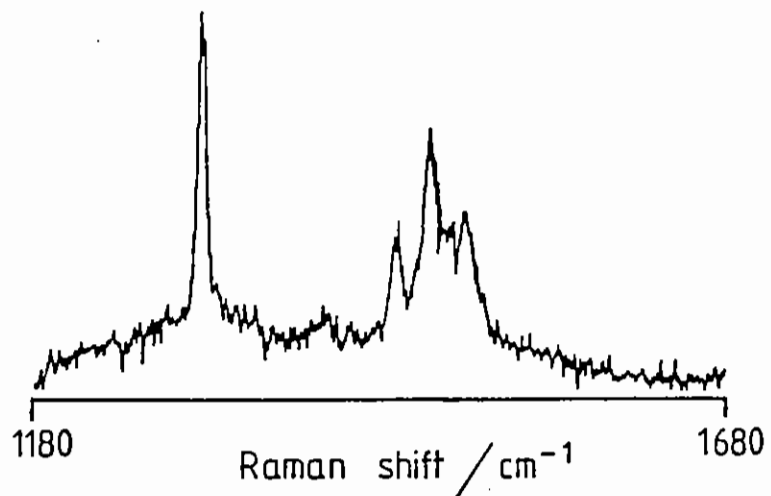


Fig. B5.1 Raman spectrum of polyethylene with CCD detector in 100 m sec.

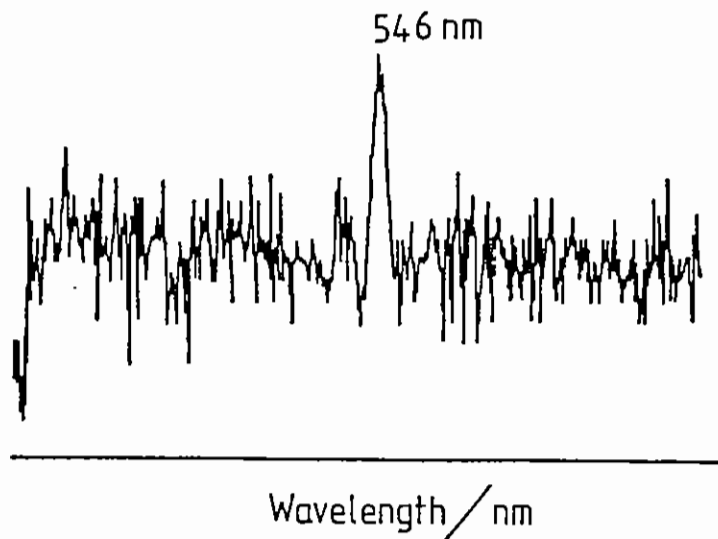


Fig. B5.3 Emission line of Hg lamp at 546 nm with CCD detector.

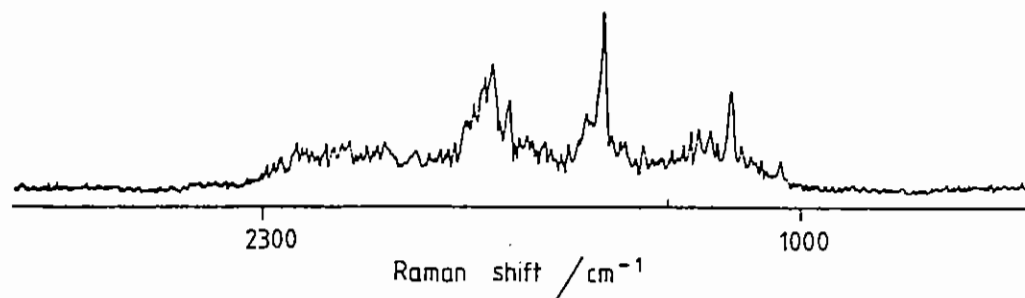


Fig. B5.2 Raman spectrum of polyethylene recorded with a diode array system in 100 m sec.

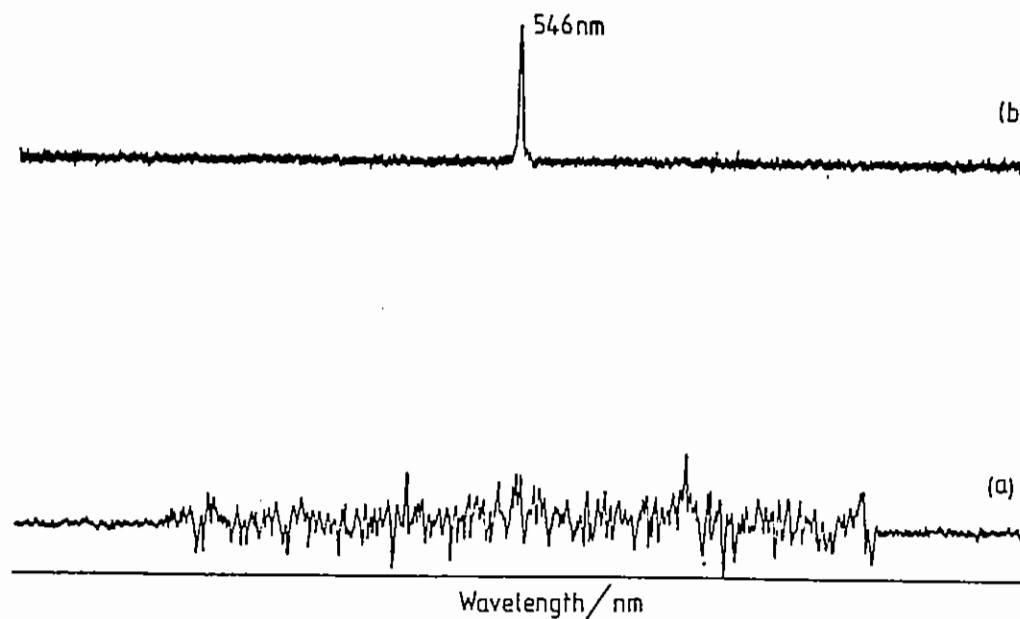


Fig. B5.4 Emission line from a Hg lamp at 546 nm recorded with a diode array.
(a) For comparison with Figure B5.3.
(b) As reference.

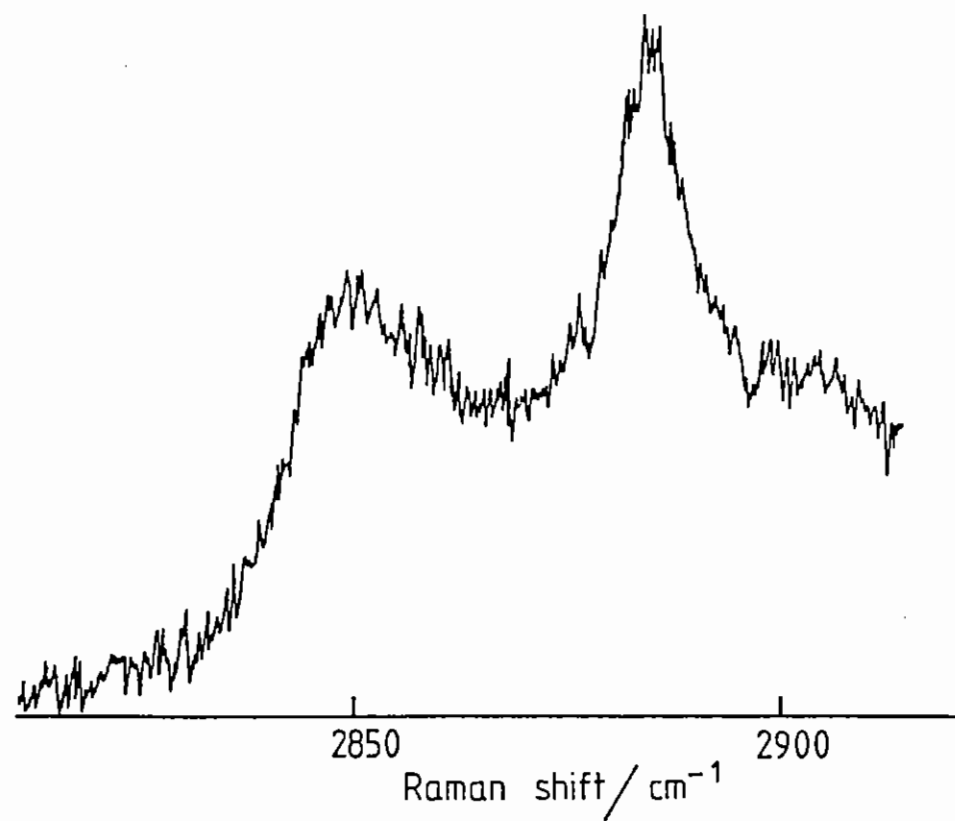


Fig. B5.5 Raman spectrum of 7 layers of Cd stearate recorded with a CCD detector.

A further example of the cooled CCD's superior performance for detecting low light levels is given in Figure B5.5. The spectrum shown was recorded at Queen Mary College, London using a modified Spex 1401 spectrometer. The sample is a Langmuir-Blodgett film, 7 monolayers of cadmium stearate. The laser power was only 40 mw at 514.5 nm and the accumulation time was 10 minutes. The total number of scattered photons detected was 1000, this represents only 1.7 photon counts sec^{-1} . This experiment was not possible using either diode array or photomultiplier tube detectors.

Conclusion

The cooled CCD is an extremely useful tool for Raman Spectroscopy particularly when working at very low light levels. The sensitivity and the noise characteristics are superior to the intensified diode array. The drawbacks of no gating facility and very poor (if not non existent) U.V. response make it an inappropriate detector for some Raman applications. The physical size of the active element means that it does not completely match with the dimensions of the exit plane of commercial spectrometers. However, different gratings or even condensing lenses may overcome this minor problem.

Acknowledgements

The author would like to thank Wright Instruments for the use of a cooled CCD system. Permission to publish has been given by British Petroleum Plc.

References

1. C A Murray and S A Dierker, J. Opt. Soc. Am. A, **3**, 2151 (1986).
2. J Janesick and M Blouke, Sky and Telescope, 239, (1987).

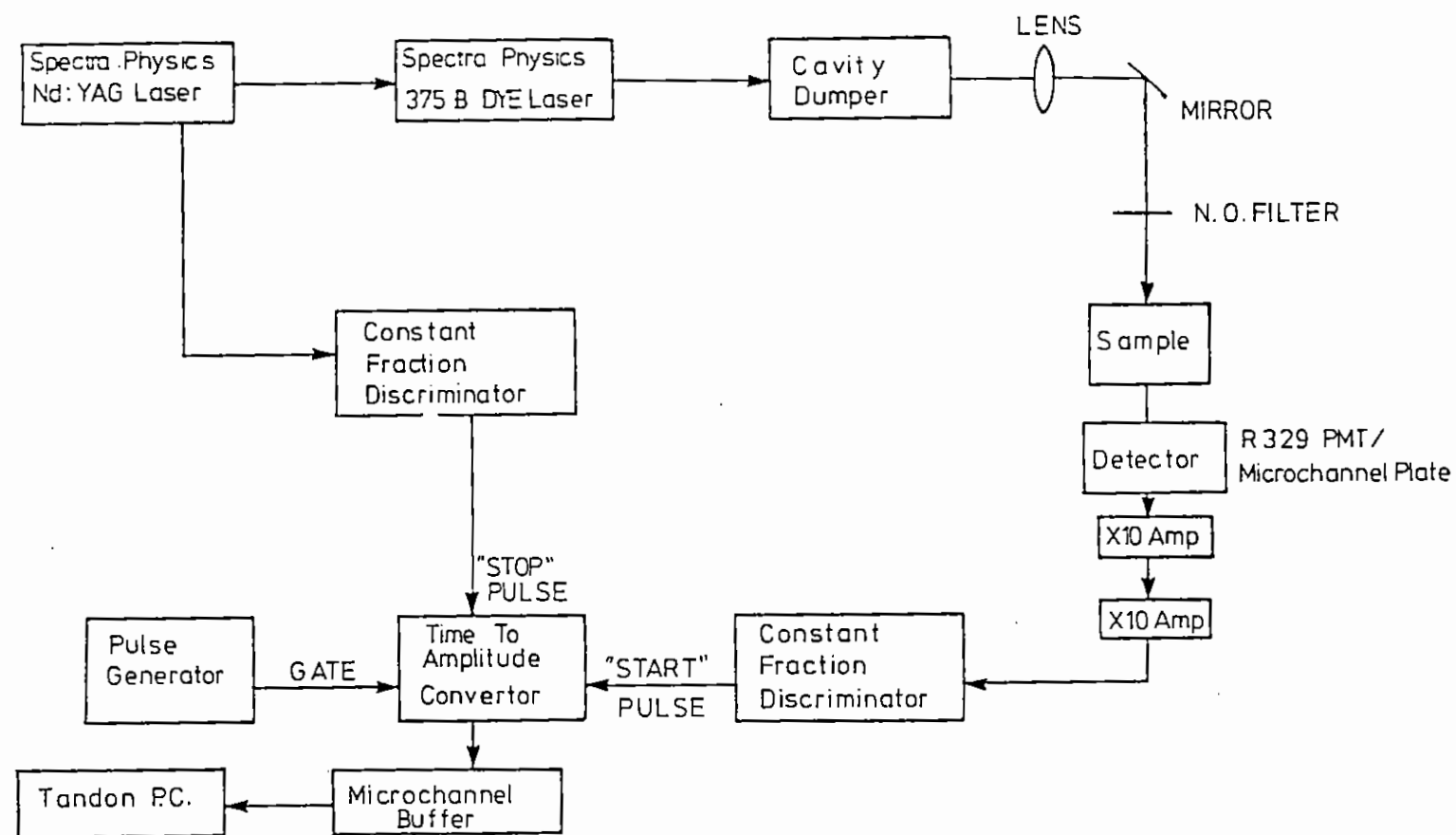
B5-4 SINGLE PHOTON COUNTING

B Flintham, Queens University Belfast
P F Taday, University of Reading
S M Tavender and W T Toner, RAL.

The LSF picosecond laser is being used to develop a time resolved single photon counting (SPC) system to allow short-lived fluorescence lifetimes to be measured (ref. 1) The frequency doubled Nd:YAG laser is used to pump a Spectra physics 375B dye laser to give a tunable excitation wavelength. As SPC requires a pulse repetition rate less than the 82 MHz produced by the laser, the beam is opto-acoustically cavity diffracted using a cavity dumper which means the repetition (and hence excitation) rate can be selected. The basic SPC experimental arrangement is shown in Figure B5.6.

Initially photons were detected by a Hamamatsu R329 photomultiplier tube (PMT). This is a dynode-type 12 stage PMT for which a typical anode to cathode supply voltage of 1.5 kV gives a tube current gain of 1.1×10^6 or about 1.76×10^{-13} C at the anode for a single photoelectron generated at the photocathode. The transit time of a photoelectron between the photocathode and the anode is about 48 ns which limits the resolution of a SPC system incorporating this type of PMT to about 1 ns. The R329 PMT was later replaced by a microchannel plate (MCP) PMT, Hamamatsu R28094U-07, which consists of a closely packed 2-D array of 6 micron diameter, short channel electron multipliers. Due to the much shorter transit time (0.5 ns) of the photoelectrons, a superior time resolution is possible. The best reported value is 43 ps FWHM (ref. 2).

As in conventional SPC the "START" pulse is provided from the fluorescence decay. The signal from the PMT is amplified by a fast preamplifier, ORTEC 9301, and is discriminated by a constant fraction discriminator (CDF), TENNELEC TC 454. The CFD provides a low jitter logic pulse from a varying anode signal, which triggers the "START" input of a time to amplitude converter (TAC), ORTEC 457. The "STOP" pulse is provided by laser light incident on a vacuum photodiode. This is discriminated by another channel in the CFD, and is delayed by several meters of co-axial cable before stopping the TAC. A gate pulse is obtained from the radio frequency driver of the cavity dumper. The



BASIC SPC EXPERIMENTAL SET-UP.

Fig. B5.6

raw data from the TAC is processed by use of a multichannel buffer, ADCAM 918A, and fed into a TANDOM-pc, where data analysis can occur, with either an ORTEC ADCAM 100 multichannel analyzer (MCA) or by a user program.

A time resolution of about 90 ps has been obtained and it is hoped that in the near future to reduce this to 40 ps, by providing the "STOP" pulse from the 41 MHz radio frequency driver of the picosecond laser.

The system at present is being characterised by observing the fluorescence decay of materials of known lifetime with excitation from the frequency doubled output of the dye laser.

References

1. B Flintham, MSc. Thesis, Queens University Belfast, September 1987.
2. R Rigler, F Claesens, and G Lomakka, in *Ultrafast Phenomena 4*, edited by D H Auston and K B Eisenthal (Springer, New York, 1984), p472.

B5-5 DEVELOPMENTS IN PUMP-PROBE EXPERIMENTS

R Chittock and W T Toner

The accuracy of the pump-probe experiments conducted by Lancashire Polytechnic (see Biology Section of this report) has been limited in previous runs by the accuracy of the complicated optical set-up required for splitting and balancing the probe beam. Therefore a beam-splitting device shown schematically in Figure B5.7 has been constructed to simplify the alignment and reduce errors caused by fluctuations in beam pointing and spectral distribution. Transmission follows reflection to give one beam, reflection followed by transmission gives the other. Three polarisers are used to balance the beams while allowing the polarisation of the beam incident on the pumped sample to be controlled independently. In addition, the

analogue difference detector has been modified to give outputs proportional to each beam as well as the difference so that separate normalisation is no longer needed.

Typical results (Figure B5.8) comparable to the peak performance of beam splitting with separate components were obtained routinely and the time required to align the apparatus was greatly reduced. This increased simplicity and convenience represents a significant improvement in the experimental method.

B5-6 STREAK CAMERA DEVELOPMENTS

A J Langley, D N Sansom and W T Toner (RAL)

Hardware and software development of the S25-UV Delli Delti streak camera and Hadlands Photonics two-dimensional CCD readout system has continued during the past year. The equipment has been used by several research groups to obtain fluorescence decay-time and anisotropy data of styrene (B1-3), protein (B2-6) and semiconductor materials.

The off-axis front optic' of the streak camera built last year has now been mounted, enclosed, and a slit assembly constructed with optional orthogonally oriented uv/visible polarizers for anisotropy measurements. One of the two channel plate intensifiers in the streak camera has been replaced by a proximity intensifier. This has resulted in a modest though desirable improvement in the signal to noise of about 20%.

Image analysis software has been developed at Hadlands Photonics and at RAL. Consecutive streak images derived from individual laser shots can be acquired and stored at a rate of ≤ 30 Hz by summing in real-time full frame images into smaller framelets. Typically 200 framelets or single shot images can be acquired in 20 seconds. Figure B5.9(a) shows 16 of a total of 27 single streak images of the fluorescence from 2, 5-diphenyloxazole excited at 295 nm. The timing marker observed at the top of each framelet is used by the software to correct for timing jitter and thence average all the framelets together as shown in Figure B4.9(b). Variations in streak camera and CCD sensitivity across the streak "window" can be corrected by measuring the fluorescence from a long lived species. Figure B5.9(c) shows 6 of 27 single shot images of

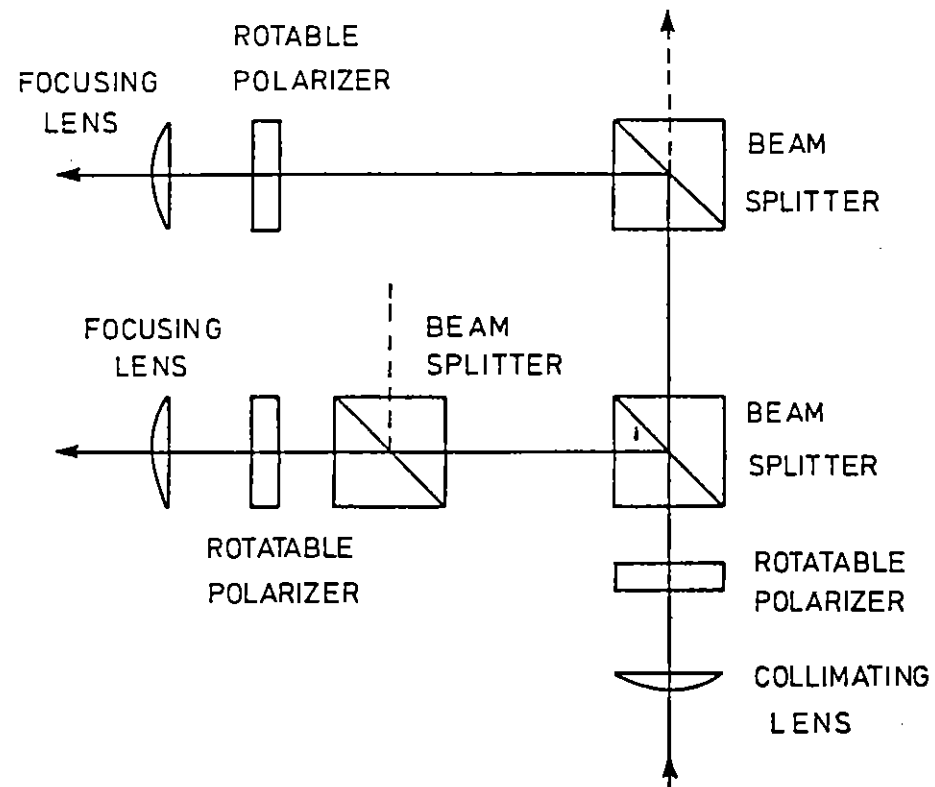


Fig. B5.7 Arrangement of Beam Splitters. Each split beam is reflected once and transmitted once. This corrects for variations in wavelength and angle automatically.

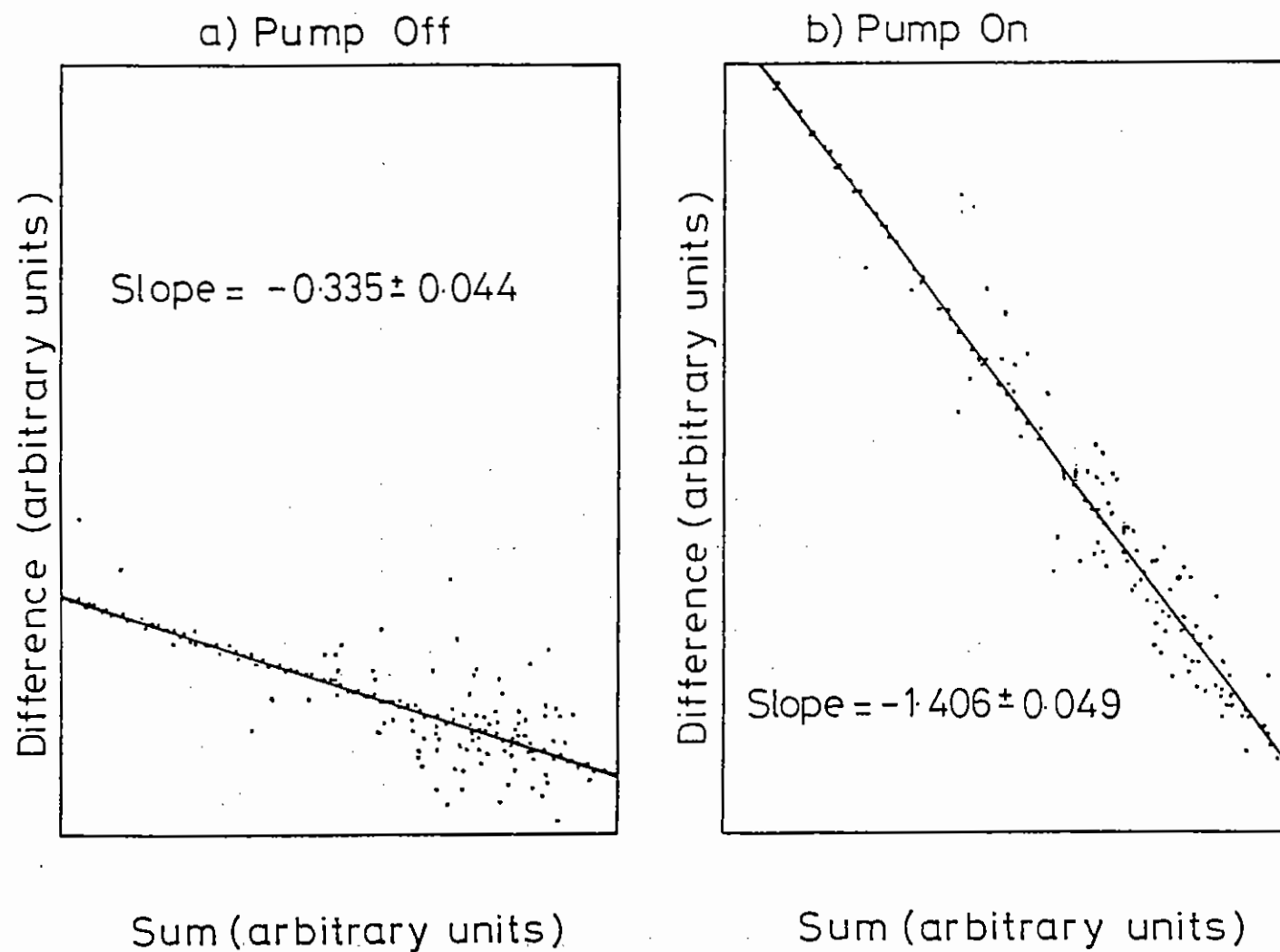


Fig. B5.8 Typical results from the Beam Splitter and Sum-Difference Detector combination, showing the effect of the pump on the difference signal from a PSI preparation. It corresponds to a 10% increase in the light reaching the detector.

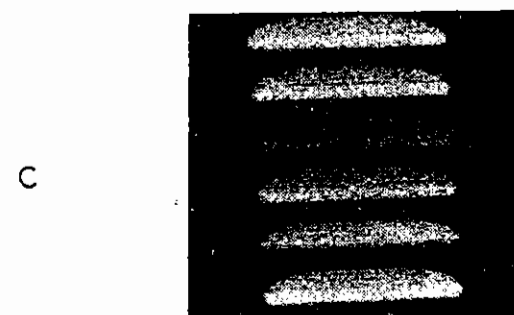
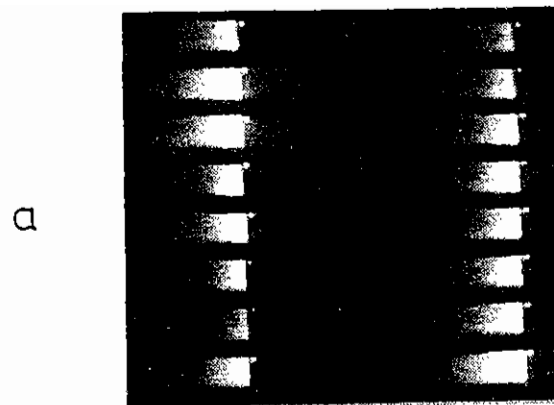


Fig. B5.9 Photographs of CCD display screen

- (a) Sixteen of 27 single shot framelets of rapid fluorescence decay
- (b) Sum of 27 single shot framelets, corrected for jitter before summing
- (c) Six of 27 single shot framelets of long lived fluorescence decay, used for correcting non-linearities.

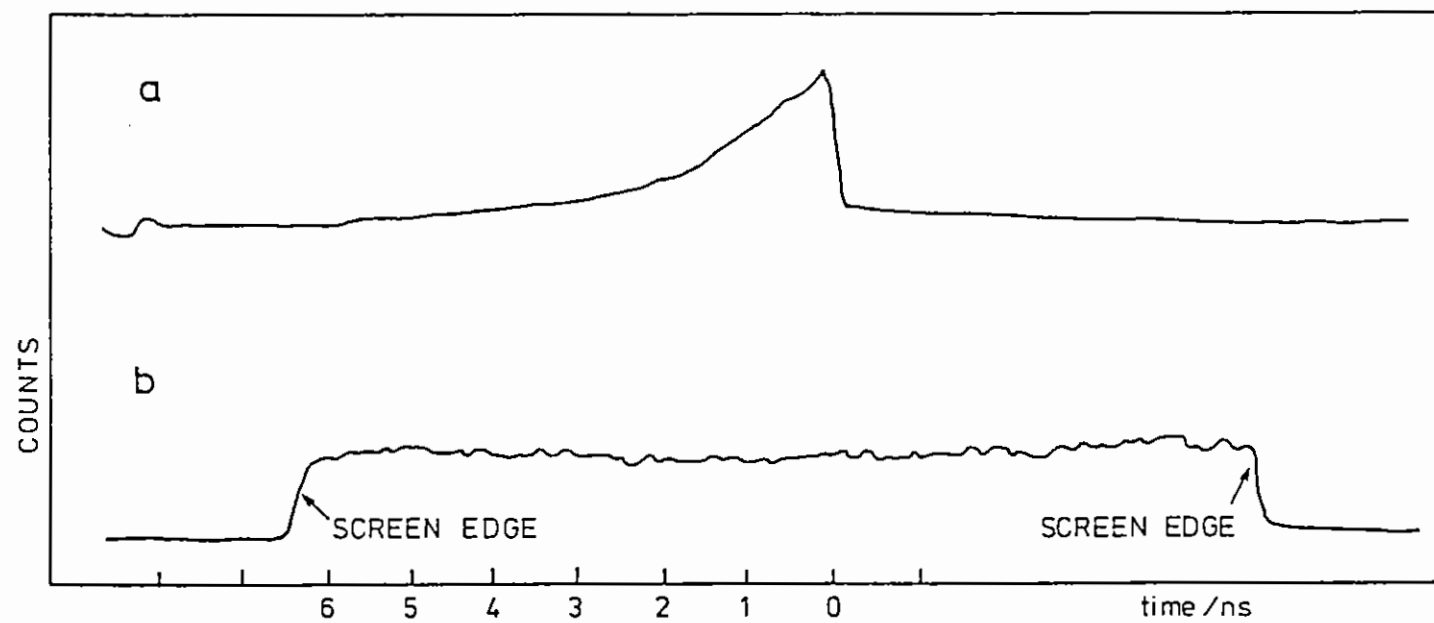


Fig. B5.10 (a) Trace of rapid fluorescence decay
(b) Calibration trace (slow decay)

fluorescence from fluoranthene which has a decay of 56 ns. The start of each decay is parked "off-screen" to the right. In Figure B5.10(a) is shown the decay of 2, 5-dephenyloxazole derived from the data displayed in Figure B5.9b) together with a characterization trace from fluoranthene (Figure B5.10(a)).

Further software refinements include correcting for non-linear sweep and background subtraction. Data can now be transferred between the Hadlands Photonics 2d analysis computer and a Tandon PC and off-line software has been developed to perform arithmetic manipulation of data, calculate anisotropies and plot decays. Users can also remove data for analysis at their own laboratories and write software for use at RAL.

C	CENTRAL LASER FACILITY PUBLICATIONS	pages
C1	JOURNALS AND BOOKS	277-280
C2	PUBLISHED CONFERENCE PROCEEDINGS	281-283

Editor: P T Rumsby

C1 JOURNALS AND BOOKS

P Apte, D Brown, A Cole, M Key, P Norreys and E Wooding.
Measurement of the uniformity of ablation for 0.53 μm laser irradiation of spherical targets.
Optics Comms. (1988) in press.

J C Austin and R E Hester.
Probing enzyme active sites with Raman and time-resolved resonance Raman spectroscopy.
Spectroscopic and Structural Studies of Biomedical Materials, ed J Twardowski, Sigma Press (1987).

J August, M Brouard, C Milne and J Simons.
Vector correlations and molecular photodissociation H_2O_2 and t-butyl hydroperoxide.
Ber. Buns. fur Phys. Chem. (1987) in press.

K Bajdar, W Peticolas, C Wharton and R Hester.
Ultraviolet resonance Raman spectra of an enzyme-substrate intermediate of papain.
J Raman Spectrosc. 18 211 (1987).

T J Barker, I R Denning and J Thorne.
Applications of 2-photon spectroscopy to inorganic ions.
Inorg. Chem. 26 1721 (1987).

H C Barr and F F Chen.
Raman scatter in a nearly resonant density ripple.
Physics of Fluids 30 1180 (1987).

H C Barr, T J M Boyd and G A Coutts.
The Raman Instability: nonlocal effects and the transition between absolute and convective regimes.
Bull. Am. Phys. Soc. 32 1715 (1987).

H C Barr, T J M Boyd and G A Coutts.
Studies of stimulated Raman scattering in a plasma filament.
Physics of Fluids (1988) in press.

J R M Barr, N J Everall, C J Hooker, J P Partanen and M J Shaw.
"Picosecond Raman pulse combining and compression in light guides for ultra high power laser systems".
SPIE Proceedings, Vol XXX - "Phase conjugation, Raman beam combining and adaptive optics" (1988) in press.

J R M Barr, N J Everall, C J Hooker, I N Ross, M J Shaw and W T Toner.
"High energy amplification of picosecond pulses at 248 nm".
Optics Comm. (1988) in press.

F Bombarda, R Giannella, E Kallne, G J Tallents, F Bely-Dubar, P Faucher, M Cornille, J Dubar and A H Gabriel.
"Observations and comparisons with theory of the helium like and hydrogen like resonance lines and satellites of nickel from the JET tokamak".
Phys. Rev. 31A 504-22 (1988).

D K Bradley, J Kilbenny, S Rose and J Hares.
Time-resolved continuum edge shift measurements in laser-shocked solids.
Phys. Rev. Lett. 59 2995 (1987).

C Chénais-Popovics, R Corbett, C Hooker, M Key, G Kiehn, C Lewis, D Pepler, G Pert, C Regan, S Rose, S Sadaat, R Smith, T Tomie and O Willi.
Strong XUV laser amplification at 18.2 nm (CVI Balmer alpha) in recombining plasma from a laser irradiated carbon fibre.
Phys. Rev. Lett. 59 2167 (1987).

A Cole, M Key, A Ridgeley, D Brown, P Norreys, E Wooding and T Barbee.
A pinhole camera for monochromatic x-ray imaging.
Opt. Comm. 621 (1987).

A Collins, J Szechi and S Tavender.
Resonant excitation of the GR centre in diamond.
J. Phys. C (1988) in press.

- R S Chittock, C Wharton and R Hester.
UV resonance Raman spectroscopy of glycinaldehyde-3-phosphate dehydrogenase.
Biochem. Soc. Trans. 16 49 (1988).
- K Codling, L Frasninski, P Hatherley, J Barr and J Thomson.
On the major mode of multiphoton multiple ionisation.
J. Phys. B. 20 L525 (1987).
- F Cross, A Langley, A MacRobert, T Bowler, D Phillips and S Bown.
Eximer laser angioplasty: analysis of photoproducts of ablation at 193 and 249 nm by emission spectroscopy.
Laser Med. Sci. 1 314 (1986).
- A Dangor, A Dymoke-Bradshaw, A Dyson, T Garvey, I Mitchell, A Cole, C Danson, C Edwards and R Evans.
Generation of uniform plasmas by multiphoton ionisation.
IEEE Trans. Plas. Sci. PS-15 161 (1987).
- G M Davis and M C Gower.
Time-resolved transmission studies of PMMA films during UV laser ablative photodecomposition.
J. Appl. Phys. 61 2090 (1987).
- G M Davis and M Gower.
Wavelength dependence of the excimer etching characteristics of polymeric resists.
App. Phys. Letts. 50 1286 (1987).
- G M Davis, D Thomas and M Gower.
Excimer laser direct etching of GaAs.
J. Phys. D. Appl. Phys. (1988) in press.
- A Djaoui, B Shiwei, T Hall, R Eason, C Jackson and S Rose.
EXAFS analysis of laser shocked, compressed aluminium plasmas.
Plasma Phys. and Controlled Fusion (1988) in press.
- J de Groot, R E Hester.
Surface-enhanced resonance Raman spectra of oxyhaemoglobin adsorbed on to colloidal silver.
J. Phys. Chem. 91 1693 (1987).
- J A Dyet, M McCoustra and J Pfab.
The visible spectrum of jet-cooled CF_3NO .
Chem. Phys. Lett. 135 534 (1987), 137, 398 (1987).
- N J Overall, J P Partanen, J R M Barr and M J Shaw.
Threshold measurements of stimulated Raman scattering in gases using picosecond KrF laser pulses.
Optics. Comm. 64 393-397 (1987).
- L J Frasninski, K Codling, P Hatherley, J Barr, I Ross and W Toner.
Femtosecond dynamics of multielectron dissociative ionisation using a picosecond laser.
Phys. Rev. Lett. 58 2424 (1987).
- M C Gower.
Phase conjugation.
J. Mod. Opt. (1987) in press.
- M C Gower.
Le fil de rasoir du laser excimere.
La Revue Polytechnique 5 614 (1987).
- M C Gower.
Phase conjugate mirrors.
Phys. Bull. 38 220 (1987).
- T Hall, R W Eason, C Jackson, B Shiwei, A Djaoui, S Rose, A Cole and P Apte.
Experimental observation of ion correlation in a strongly coupled laser produced plasma.
Phys. Rev. Letts (1988) in press.
- M C Gower and P Hribek.
Mechanism for internally self-pumped phase conjugate emission from BaTiO_3 crystals.
J. Opt. Soc. Am. B. (1988) in press.
- R J Harrison, B Pearce, G Beddard, J Cowan and J Sanders.
Photo-induced electron transfer in pyromellitimide-bridged porphyrins.
Chem. Phys. (1987) in press.

- R E Hester
Raman spectroscopic studies of species in situ at electrode surfaces.
Comp. Chem. Kin. 29 (1987).
- C J Hooker, F Kannari and M J Shaw.
Multiplexed short-pulse amplification in a KrF laser.
Optics Comm. 65 269-274 (1987).
- F Kannari, M Shaw and F O'Neill.
Parametric studies of an electron beam pumped krypton rich KrF laser.
J. Appl. Phys. 61 476 (1987).
- A Kvaran, J P Simons and M J Shaw.
Vibrational relaxation in KrF* and XeCl* by rare gases.
Appl. Phys. B45 (1987) in press.
- C L S Lewis, R Corbett, D O'Neill, C Regan, S Sadaat,
C Chenais-Popovics, T Tomie, J Edwards, G P Kiehn, R Smith, O Willi,
A Carillon, H Guennou, P Jaegle, G Jamelot, A Klisnick, A Sureau,
M Grande, C Hooker, M H Key, S J Rose, I N Ross, P T Rumsby, G J Pert
and S A Ramsden.
Status of soft x-ray laser research at the Rutherford-Appleton
laboratory.
Plasma Physics and Controlled Fusion 30 35 (1988).
- M R S McCoustra and J Pfab.
Photodissociation of methylthionitrites near 450 nm.
Chem. Phys. Lett. 137 534 (1987).
- M R S McCoustra, J Dyet and J Pfab.
Laser photofragment spectroscopy of jet-cooled CCIF₂NO.
Chem. Phys. Lett. 136 231 (1987).
- I Morrison and R Denning.
Do higher excited states of the uranyl ion luminesce ?
Chem. Phys. Letts. 143 409 (1988).
- D J Nicholas and S G Sajjadi.
Numerical simulation of laser filamentation experiments.
Las. Part. Beams 5 154 (1987).
- D J Nicholas and S G Sajjadi.
An analytic treatment of the Fokker Planck equation for non-Spitzer
heat flow.
Phys. Rev. Lett. (1988) in press.
- D J Nicholas and S G Sajjadi.
Non-Spitzer heat flow in an unsteady ablating plasma.
Phys. Fluids (1988) in press.
- B A Omar, S E Clark, D C Emmony, D B Hollis and M J Shaw.
The application of doped glass fluorescers to the recording of pulsed
U V laser beam profiles.
Optics and Laser Tech. (1988).
- J P Partanen and M J Shaw.
A single-mode KrF laser.
Appl. Phys. B43 231-237 (1987).
- R T Phillips, Z Sobierierski, W Toner, J Barr and A Langley.
Initial photoluminescence decay rates in amorphous phosphorus.
Solid State Comm. 63 481 (1987).
- R T Phillips and Z Sobiesierski.
Recombination in amorphous red phosphorus.
J. Phys. C., Solid State (1987) in press.
- D Raybone, T Watkinson and J Whitehead.
On the role of iodine atoms in the production of IF(B²pi) in fluorine
atom/iodine flames.
Chem. Phys. Lett. 135 170 (1987).
- I N Ross, G M Davis and D Klemetz.
High resolution holographic image project.
Appl. Optics. 27 967 (1988).
- I Ross, J Boon, R Corbett, A Damereil, P Gottfeldt, C Hooker, M Key,
G Kiehn, C Lewis and O Willi.
Design and performance of a new line focus geometry for x-ray laser
experiments.
Appl. Optics. 26 1584 (1987).

B Stewart and R Hester.

Protein structure from Raman and ultraviolet resonance Raman spectroscopy.

Laser Scattering of Biological Systems: Studies in Physical and Theoretical Chemistry, Elsevier, ed J Stepanek et al 45 229 (1987).

E Turcu, G Davis, M Gower, F O'Neill and M Lawless.

X-ray lithography using a KrF laser plasma source at $h\nu = 1$ keV.

Microelectronic engineering 6 287 (1987).

J Wark, R Witlock, A Hauer, J Swain and P Solone.

Shock launching in silicon studied with use of pulsed x-ray diffractions.

Phys. Rev. B. 35 9391 (1987).

C2 PUBLISHED CONFERENCE PROCEEDINGS

T Barker, R Denning and J Thorne.

Vibronic coupling in two-photon absorption.

Proc. Int. Conf. on Dynamical Processes in the excited state of solids.

T Sukuba, Japan, August (1987).

H C Barr, T J M Boyd and G A Coutts.

The Raman Instability: Non-local effects and the transition between absolute and convective regimes.

29th Annual Meeting of the American Physical Society (1987).

H C Barr, T J M Boyd and G A Coutts.

Stimulated Raman scattering in the presence of filamentation.

Proc. 14th Conf. on Controlled Fusion and Plasma Physics, Madrid, 11D, 620 (1987).

H C Barr, T J M Boyd and G A Coutts.

The Raman instability: non-local effects.

Proc. 14th Conf. on Controlled Fusion and Plasma Physics, Madrid, 11D, 637 (1987).

R Benattar, J Goddard, V Malka, J Edwards and O Willi.

Time resolved XUV imaging of the rear side of laser heated foils.

SPIE, 831, 171 (1987).

A Dangor et al.

Plasma beatwave experiments at RAL.

Proc. New Dev. in particle acceleration techniques, Orsay Workshop, 2, 375 (1987).

C N Danson and N H Rizvi.

Short pulse active/passive mode locked Nd YLF oscillator for use on the VULCAN glass laser system.

8th Nat. Quant. Elect. Conf., St Andrews, J. Mod. Opt., to be published (1987).

C N Danson et al.

Oscillator and system development on the VULCAN laser system for the European beatwave experiment.

8th Nat. Quant. Elect. Conf., St Andrews, J. Mod. Opt., to be published (1987).

R W Eason, T A Hall, A Djaoui, C Jackson, B Shiwal, D K Bradley, A J Rankin, S D Tabatabaei, G N Greaves and S J Rose.

EXAFS/RefLEXAFS experiments using laser-produced plasma x-ray sources. SPIE Vol. 831 (1987).

R G Evans and T J Katsouleas.

Summary of plasmas for particle acceleration.

Proc. Workshop on New Developments in Particle Acceleration Techniques, Orsay (June 1987).

R G Evans.

Noise in plasma lenses for high energy particle beams.

Proc. Workshop on New Developments in Particle Acceleration Techniques, Orsay (June 1987).

R G Evans.

Plasma-based accelerators.

AIP Conf. Proc., 156, 3 (1987).

M C Gower.

Phase conjugation.

8th Nat. Quant. Elect. Conf., St Andrews, J. Mod. Opt., to be published (1987).

M C Gower and P Hribek.

Mechanisms for self-pumped phase conjugate emission from BaTiO₃ crystal/OSA Topical meeting on Photorefractive Material, Effects and Devices, Los Angeles (1987).

M C Gower.

Excimer laser lithography.

Conf. Lasers and Electro-Optics, Baltimore (1987).

- M C Gower and P Hribek.
Mechanisms for self-pumped phase conjugate emission from BaTiO₃ crystal.
Conf. Lasers and Electro-Optics, Baltimore (1987).
- C J Hooker et al.
Multiple pulse amplification in a KrF laser.
Conf. Lasers and Electro-Optics, Baltimore (1987).
- M H Key et al
Studies of XUV laser action in recombination to hydrogenic and lithium-like ions.
Int. Conf. Lasers, Xiamen, China, to be published (1987).
- M H Key.
XUV Lasers - a survey of progress.
IAU Colloq. on UV and x-ray spectroscopy of astrophysical and lab plasmas. J de Physique Colloque U88 (in press).
- M H Key.
XUV lasers.
8th Nat. Quant. Elect. Conf., St Andrews, J. Mod. Opt., 35, 575, 1988.
- M H Key.
XUV lasers.
XUV and x-ray spectroscopy of laser produced plasmas.
Proc. Scottish Universities Summer School in Astrophysical and Laboratory Plasma Spectroscopy, Edinburgh.
- M H Key et al.
Review of progress of the UKCLF in developing XUV lasers based on recombining laser produced plasma.
SPIE Vol. 831 (1987) in press.
- G Kiehn et al.
X-ray laser research at the Rutherford Appleton Laboratory.
SPIE, 831, 270 (1987).
- G Kiehn, T Garrey, R Smith, O Willi, A Damerell and J Westlake.
Absolute calibration of XUV time resolving spectrograph.
SPIE, 831, 150 (1987).
- E Madraszek et al.
Laser plasma soft x-ray source: a small dedicated source at the CLF.
Proc. Int. Symp. X-ray Microscopy, Brookhaven (1987).
- I Morrison, J Thorne and R Denning.
The 2-photon excitation spectra of Manganese II compounds.
Proc. 1987 Int. Conf. on Luminescence, Beijing (1987).
- M R S McCoustra et al.
The spectroscopy and photophysics of jet-cooled C-nitroso compounds.
11th Int. Symp. Molecular Beams, Edinburgh (1987)
- J Pfab, M McCousta and J Dyet.
Photodissociation of jet-cooled C-nitroso compounds.
11th Int. Symp. Molecular Beams, Edinburgh (1987):
- F O'Neill, G Davis, M Gower and E Turcu.
KrF laser plasma x-ray sources for lithography.
Proc. Inst. Phys. Conf. on Advanced Lithography, Didcot (1988).
- F O'Neill, G Davis, M Gower, E Turcu, M Lawless and M Williams.
Plasma x-ray sources for lithography generated by 0.5 J KrF laser.
SPIE, Vol. 831, 230 (1987).
- S A Ramsden et al.
Soft x-ray laser research in the UK.
International symposium on short wavelength lasers and their applications.
Institute of Laser Engineering, Osaka, Japan, 11-13 November 1987.
- B Shiwei, T Hall, A Djaoui, R Eason, C Jackson, S Rose, A Cole and P Apte.
Experimental observation of ion correlation in high density plasmas.
SPIE, 913 (1988).

J Simons.

Product alignment in reactive, inelastic and half collisions.

Selectivity in chemical reactions.

NATO Adv. Res. Wksp., Reidel, Boston (1987).

J Thorne, A Harrison, I Morrison and R Denning.

Flourescence line narrowing in uranyl glasses.

Proc. Int. Conf. on Dynamical Process in the excited states of solids.

Tsukuba, Japan (1987).

E Turcu, F O'Neill, U Zammit, Y Al-Hadithi, R Eason, A Rogoyski, C Hills and A Michette.

Calibration of a KrF laser plasma source for x-ray microscopy applications.

SPIE, Vol. 831, 211 (1987).

P T Rumsby.

Recent developments to the high power glass and gas lasers at the UK Central Laser Facility.

Int. Conf. Lasers, Xiamen, China, to be published (1987).

A D Stead, T W Ford, R W Eason, A G Michette, W Myring and R Rosser.

Soft x-ray contact microscopy of botanical material using laser-produced plasmas or synchrotron radiation.

Proc. Int. Syn. X-ray Microscopy, Brookhaven (1987) (Springer).

I C E Turcu, G Davis, M Gower, F O'Neill and M Lawless.

X-ray lithography using a KrF laser-plasma source at $h\nu \sim 1$ keV.

Proc. Int. Conf. Microlithography, Paris (1987).

J Wark, R Witlock, A Hauer, J Swain and P Solone.

Proc. 1987 APS topical conf. on shockwave in condensed matter.

Mortery, Calif. July 1987, North Holland, p781, Ed Schmidt and Holmes.

O Willi, D Bassett, S Coe, J Edwards, M Grande, P Jaegle, G Jamelot, M Key, G Kiehn, A Klisnick, C Lewis, D O'Neill, G Pert, S Ramsden, C Regan, S Rose and R Smith.

XUV amplification in cylindrically expanding laser plasmas.

APS meeting on Radiation Transport and Atomic Physics, Santa Fe (1987) (in press).

O Willi, V Barrow, D Bassett, S Coe, J Edwards, M Grande, M Key, G Kiehn, C Lewis, D O'Neill, G Pert, S Ramsden, C Regan, S Rose, R Smith and J Wark.

XUV amplification in cylindrically expanding laser plasmas.

Laser 1987. Lake Tahoe, USA (in press).

O Willi.

Studies of laser plasma interaction and energy transport.

Int. Conf. Lasers, Xiamen, China, to be published (1987).

C Wharton, R Chittock and R Hester.

Vibrational spectroscopy of enzyme-substrate intermediates.

Proc. 2nd European Conf. on the spectroscopy of biological molecules, Freiberg (1987).

Acknowledgements

The production of the report has been a team effort and as usual most of the work has been done by Chris Ayling and Clare Schroder, assisted by Jo Burgess for the typing, Jan Smith for the illustrations, and Nic Allen for the production plan. RAL reprographics section have carried out the printing and binding.

Considerable editorial effort has been put in by the section editors, while facility users have contributed vital reports on their work.

Thanks are due to all for a job well done.