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Extreme ultraviolet laser action in plasmas

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[Plate 1]

The development of a laser operating in the extreme ultraviolet or X-ray spectral regions remains an outstanding problem of current technology. Despite the many proposals, no demonstration of significant gain on a transition in this region has yet been reported. The constraints introduced by decreasing wavelength are increasingly severe, demanding high pump power in a system with a relatively short lifetime. In consequence, much attention has been paid to the use of plasma, often generated by high power laser irradiation, as the gain medium. In this paper I briefly outline the various avenues by which laser action in plasmas has been explored, with a view to identifying those which seem at present to offer promise of success. Particular attention is given to the recombination scheme with an emphasis on a recent demonstration of gain.

Introduction

The development of a working laser in the x.u.v. spectral region would represent a significant advance towards the ultimate goal of achieving X-ray laser action. The strong adverse scaling of pump power with wavelength into the X-ray region makes extreme demands on current technology. In particular pump power densities of order 1 TW/cm^2 with durations less than 1 ps are typically required. As a result interest has centred on the generation of laser action in the 10 nm spectral region, where the constraints are relaxed to within state-of-the-art systems. Of particular interest are schemes that are scalable to X-ray wavelengths.

In view of the extremely high pump power requirements, all devices that have been proposed to date use either a subsidiary laser or a particle beam as the pump. The population inversion is generated either between the inner shell levels of weakly ionized or neutral atoms, or amongst the excited state of highly stripped ions. The poor reflectivity and transmission of most materials at these wavelengths, together with the short lifetimes of excited states, place severe constraints upon cavity design. In consequence it is likely that the first demonstrations of laser action will be made by using either an amplified spontaneous emission or a travelling wave mode system.

The principal problem which has thus been addressed concerns the generation of a sufficiently large population inversion to enable laser action to be achieved. To this end many ingenious proposals have been made, although to date without any practical demonstration being successfully achieved. These schemes have been comprehensively reviewed in the paper by Waynant & Elton (1965), and I shall therefore only briefly outline some of the more promising approaches here.

Recent theoretical studies (Palumbo & Elton 1977; Vinogradov *et al.* 1977) of the collisional excitation of the 3p level by the forbidden transition 2p–3p in ions with an outer shell configuration $2p^n$ show that significant inversions of the transition 3s–3p can be attained under thermodynamic conditions typical of those generated in the plume of a laser-produced plasma from a solid target. Thus, for example, calculations of the inversions in a Ca^{XI} plasma at a temperature of about 100 eV and an electron density of about $10^{20}\ cm^{-3}$ indicate a gain coefficient of about 100 cm^{-1} at a wavelength of 563 nm (Vinogradov *et al.* 1977). Such an inversion under these conditions is clearly quasi-steady (depending on the pump laser-pulse duration) and at these wavelengths allows a straightforward cavity design to be used.

The large cross section for resonant charge exchange is extremely attractive for generating inversion by the injection of ions into a matched neutral (Louisell *et al.* 1975) or weakly ionized background (Waynant & Elton 1977; Seely & McKnight 1977; Vinogradov & Sobel'man 1973). To date direct experiments by means of this approach have been disappointing owing to difficulties in ensuring a sufficiently intense ion beam without disturbing the background gas. Observations (Dixon & Elton 1977; Dixon *et al.* 1978) of inversion generated by charge exchange have been made in the expansion of plasma from laser irradiation of a solid target into a background of cold material evaporated by a weak pre-pulse. The inversions of the Balmer series lines in C^{VI}, were, however, too weak to generate laser action.

One of the most popular approaches to this problem is via photon pumping by using filtered X-rays from a laser-produced plasma as the source. Theoretical investigations have studied both inner shell excitation of neutral atoms (Arrechi *et al.* 1974; Elton 1975) and outer shell transitions (Duguay & Rentzepis 1967; Mani *et al.* 1976; Hyman & Mani 1977) in highly stripped ions (Norton & Peacock 1975). Two major problems, however, confront the practical realization of these methods, namely the production of a sufficiently strong pump X-ray flux, and the destruction of the target itself by the absorbed X-ray energy (Axelrod 1976).

Probably the most promising proposal at present for generating laser action in the x.u.v. laser region is the recombination scheme. Population inversions have already been observed (Irons & Peacock 1974; Dewhurst *et al.* 1976; Key *et al.* 1979; Kononov *et al.* 1976; Bhagavatula & Yaakobi 1978) by several workers under different conditions in the wavelength region around 10 nm, which have been ascribed to this mechanism. Theoretical considerations (Pert 1976) have shown that by using hydrogen-like ions the scheme is scalable from the x.u.v. region into the X-ray region. We shall discuss this approach in detail in the remainder of this paper.

A direct measurement of gain at 11.74 nm has been reported by Jaegle *et al.* (1977). However, the exact mechanisms by which the inversion is created remain obscure, and subsequent experimenters (Valero 1974) have questioned the interpretation of these results.

Recombination laser

Rapid cooling of a strongly ionized plasma leads to a rapid recombination. If the electron temperature is sufficiently low, the recombination occurs preferentially into the upper excited states of the ion, which then decay by radiative and/or collisional cascade to the ground state. Within the cascade from the upper excited states, population inversions may form among the excited states depending on the relative transition probabilities. Lasing by using this method of pumping has already been observed in the visible spectral region by Silfvast *et al.* (1977) in CO_2 laser sparks in the rare gases.

The principal difficulty in generating inversion by this method is to obtain sufficiently rapid cooling. To this end the plasma is usually rapidly expanded, although direct collisional cooling has been suggested (Bhagavatula & Yaakobi 1978). In this case the characteristic expansion time must be less than the recombination time. For transitions in the x.u.v. region this requires expansion times of order 100 ps, a value characteristic of the expansion of plasma formed from small targets by laser irradiation, and indeed observations have been made of inversions generated in such expansions (Irons & Peacock 1974; Key *et al.* 1979; Kononov *et al.* 1976; Bhagavatula & Yaakobi 1978). If, however, this approach is to be used to generate sufficient gain for laser action, the target must be appropriately chosen to allow the inversion to be generated rapidly at a high density, and therefore large gain.

To this end detailed studies (Pert 1976) have been made of the generation of gain by uniformly heated cylinders. Numerical studies by means of a similarity model code showed that the maximum gain per centimetre of the Balmer-α transition of C^{VI} at 18.2 nm from a cylinder of carbon plasma of radius A (cm) and initial density N (cm^{-3}) was given by

$$G = 1.2\times 10^{6}/N^{\frac{1}{2}}A^{2}\ \mathrm{cm}^{-1}, \tag{1}$$

and occurred when the total thermal energy per ion was given by

$$\epsilon_{\mathrm{T}} = 1.4\times 10^{-10}(NA)^{\frac{2}{3}}\ \mathrm{eV}.$$

From these scaling laws, as well as physical considerations, it is immediately apparent that the initial plasma should be as small as possible, and should expand cylindrically rather than one-dimensionally, thus indicating a need to use targets of limited dimensions: fibres, ribbon, etc. Attention has therefore been concentrated on the properties of heated cylindrical fibres, either directly heated by laser irradiation (Pert 1979) or produced by imploding thin shells (Pert 1980*a*). Of these the former is more efficient and has been more actively pursued.

Experimental studies (Dewhurst *et al.* 1976) of the irradiation of thin carbon fibres by laser have shown the generation of inversion in this manner. To improve the coupling of the laser pulse to the fibre a small (10%) pre-pulse was used before the main pulse. By using an absolutely calibrated spectrograph, the absolute intensities, and spatial distributions of the Lyman series lines of C^{VI} were measured to be in accurate agreement with computational modelling if about 20% of the laser energy was absorbed into the plasma. The significance of these experiments was threefold, in that they showed

(*a*) significant gain could be generated by laser irradiation of thin carbon fibres;

(*b*) the computer model could be used to give an accurate representation of the population distribution in the plasma;

(*c*) laser–fibre coupling fractions of about 20% could be achieved.

On the basis of these results the up-rating of the experiment with the use of cylindrical focusing and larger laser energies was begun with the object of direct identification of gain. These studies will now be described.

Theoretical considerations

To construct a working laser it is necessary that the product of gain and length, Gl, exceed some minimum value; for example, for operation in a non-cavity amplified spontaneous emission mode, $Gl \gtrsim 10$. To achieve values of this order of magnitude one plasma dimension not less than about 1 cm is necessary. This immediately implies that the laser plasma must take the form of a large aspect ratio cylinder, and that the pump laser must be focused into a line.

In principle one may conceive that the laser is operated in a purely cylindrical mode. In practice, however, both the fibre and the laser focus must be terminated, and departures from the cylindrical optimum occur. It is characteristic of the gain profile of these plasmas that there is a clearly defined optimum operating condition with a relatively narrow 'window' within which significant gain occurs (Pert 1976). If the laser energy is decreased below the optimum, the plasma gain decreases, becoming absorbing and eventually relatively transparent when the ionization is incomplete. In addition, the time history of the gain is changed, the onset of gain being delayed as the laser energy decreases. Thus the illumination of a long fibre by a focus of poor uniformity will have gain significantly below the optimum value. The gain is also reduced for a finite fibre irradiated by a uniform laser focus owing to the Doppler shift associated with the axial velocity, and other related effects (Pert 1979).

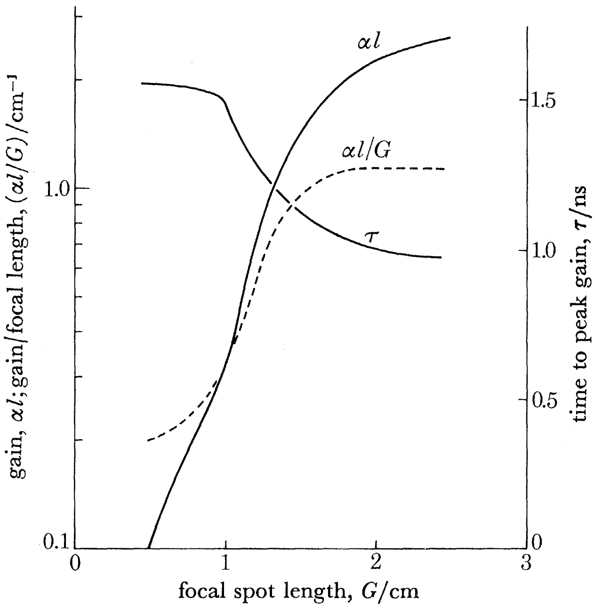

FIGURE 1. The calculated variation of the gain (αl) and time to peak gain (τ) of a carbon fibre 1 cm long heated by a laser with a Gaussian variation of intensity along the fibre length plotted as a function of the $1/e$ full width, G, of the axial focus. The laser intensity was adjusted to maintain the average intensity along the fibre at the optimum value for a uniformly heated fibre. The fibre diameter was 3 μm and the laser focus $1/e$-width 40 μm. The laser pulse consisted of a 1% pre-pulse 200ps before the main pulse of average intensity 0.3 TW/cm and width 100ps.

To study the effects of a finite fibre length and axial focusing intensity variation, a hybrid code has been constructed. The code uses a similarity expansion in the two radial dimensions, with a Lagrangian cell description along the axis. Two temperatures with axial thermal conduction are included. The complete set of collisional–radiative equations are solved in each cell (Pert 1980*b*), and the axial gain–length product calculated by taking into account Doppler shifts due to the axial velocity.

The results of a calculation for a 3 μm-diameter fibre of 1.0 cm length are shown in figure 1. The fibre was irradiated by a laser with a Gaussian focal spot geometry of $1/e$-width 40 μm radially, and variable z_0 axially. The laser power was adjusted so that the average power density on the fibre was maintained at the optimum uniform focus, infinite fibre value. The effect of absorption as the laser intensity falls at the edge of the focal spot are clearly seen once the laser focus $1/e$-length becomes less than the fibre length, corresponding to the known gain window of about ±50% in energy (Pert 1979). From these results we conclude that the laser intensity profile half-width is given by

$$z_0 \gtrsim l,$$

where l is fibre length for satisfactory operation.

In these modelling studies it is assumed that the similarity model not only gives a good representation of the hydrodynamics of the bulk, but also allows a reasonable estimate to be made of the laser–fibre coupling. To resolve these issues detailed fluid-code modelling of the laser–fibre interaction has been made. It has been shown (Pert 1980*c*) that provided that the energy is deposited relatively slowly the similarity model gives an extremely accurate representation of the expansion of uniform (isothermal) cylinders. Two problems therefore arise: Is the laser energy absorbed by the fibre? Secondly, if so, is it uniformly distributed by thermal conduction? On the basis of earlier experiments the answer to both questions would appear to be affirmative, when the focusing is spherical. However, preliminary experiments with cylindrical focusing indicated a contrary behaviour. Since this is clearly of utmost importance, a detailed computational study was made to study the hydrodynamics in detail.

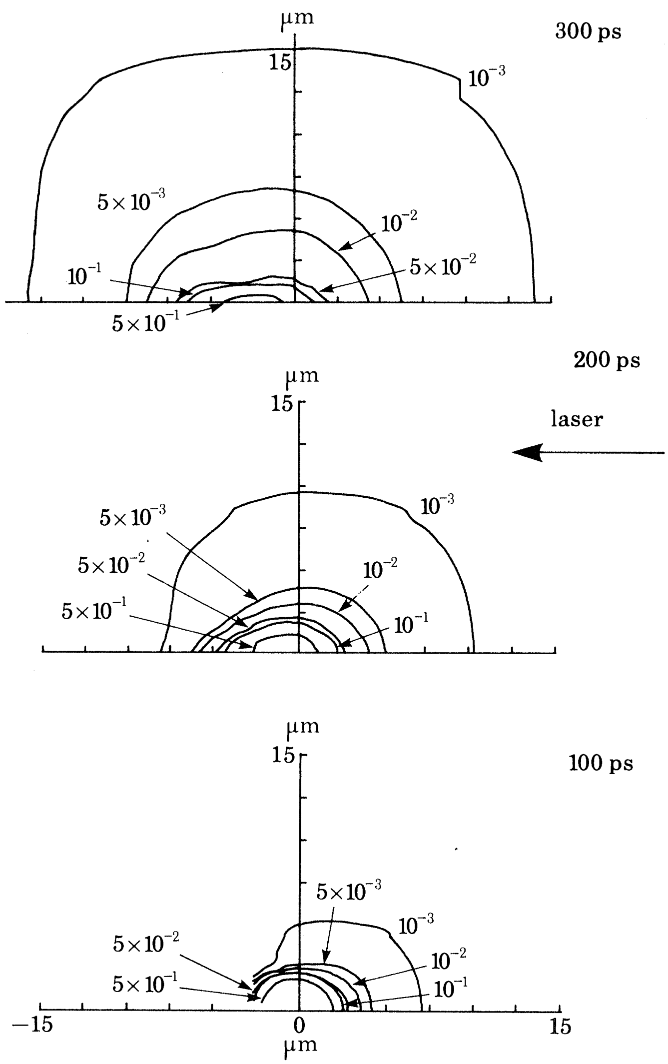

FIGURE 2. Density contours of the expansion of a uniformly heated fibre. The fibre had an initial diameter 3 μm and was irradiated by a beam of average intensity 0.3 TW/cm and width 140ps with a 10% pre-pulse 200ps before the main pulse, the focal spot width being 40 μs. The contour plots are shown 100, 200 and 300 ps after the start of the pre-pulse. The expansion of the fibre to fill the focal spot during the pre-pulse and early part of the main pulse can be clearly seen. The contours are plotted in units of the solid density.

Initial studies by means of the two-dimensional Lagrangian code LAG2 clearly revealed the importance of the pre-pulse in forming a plasma 'blanket' consisting of material evaporated off the front face of the fibre, and which flowed around the fibre. Since the mesh, in consequence, became severely distorted, further study was abandoned with this code. A rezoning scheme in the two-dimensional Eulerian code MAGT was used to extend the above modelling. As before, it was found that the plasma 'blanket' was formed by the pre-pulse totally enclosing the fibre core with a relatively low density plasma, which significantly increased the effective cross section for absorption and provided a low density medium in which heat could readily circulate around the core during the main laser pulse, greatly increasing the effective heating area thereby (figure 2). For typical optimum laser pulses (as predicted by the similarity code) it was found that complete heating of the core had occurred by about the peak of the laser pulse. Furthermore with a dump at the critical density, as with the similarity code, it was found that the energy absorption fraction was in remarkably close agreement with the similarity code. In this connection the important point should be made that the energy densities for optimum operation, *ca.* 0.2 TW/cm^2, are significantly below the thresholds for the most damaging nonlinear effects of flux limitation and fast electron generation, and that in confirmation no fast ion effects are observed in experiment.

These results confirm both the importance of the pre-pulse, and the suitability of the similarity code for describing the overall expansion found in earlier experiments.

Experimental considerations

Examination of the consequences (Pert 1979) of equation (1) applied to fibres heated by lasers shows that considerable improvement in efficiency can be obtained if the fibre diameter is decreased. In practice this conclusion is modified by the need to obtain sufficient coupling to form the 'blanket', but remains valid overall. Considerable effort has therefore been directed towards finding methods of fibre reduction. The most successful have been chemical etching by NO_2 and oxidation in air (Jacoby & Shorrock). With both these methods, fibres as thin as 1.5 μm diameter have been produced.

Computational results for fibres with diameters in the range 2–5 μm show that laser energies of about 50 J/cm are required. Thus if the laser output is about 10 J the beam must be focused to a spot about 40 μm wide and 2 mm long, of good uniformity. To achieve this from a laser output beam of about 55 mm diameter we have used an aspheric lens of focal length 125 mm in conjunction with a concave cylindrical lens of focal length 200 cm. Extreme care in alignment was found to be essential if the focus was not to be so severely aberrated as to be unusable.

The laser consists of a standard Nd–glass laser system giving a maximum energy of about 10 J in a single pulse of about 100 ps duration. A nominal 10 % pre-pulse is formed by a glass plate reflector placed immediately after the oscillator (Dewhurst *et al.* 1976). Extreme care is taken to maintain a good beam spatial profile along the chain to reduce focusing non-uniformity.

The laser pulse is focused by means of the lens configuration described above onto the fibre which is placed in the field of view of two grazing incidence vacuum spectrographs (figure 3). The systems are aligned to ensure that the entire illuminated length of fibre falls within the field of view of both instruments; one instrument is arranged to look along the line of the fibre, and the other across it. To avoid problems with non-uniform illumination at the edge of the focus, the fibres are mounted with a free end pointing into the spectrograph. The edge of the focus towards the spectrograph is usually left void, the fibre only filling one-half to three-quarters of the focus.

For reasons that will be apparent the spectrographs are used with photographic recording. Clearly careful calibration of the two instruments is necessary if quantitative results are to be obtained. To this end the density–intensity characteristics of the films used (Kodak/Pathé SC5 and SC7) in the vacuum ultraviolet have been determined by a novel technique (Shorrock & Tallents 1980), a multi-layer aluminium foil filter being used. The two spectrographs were cross-calibrated with respect to each other by using the spectra obtained by irradiating fibres with a spot focus produced by the aspheric lens alone (figure 4), the fibre direction being normal to that usually used. Since the spectrographs view the same plasma from essentially opposite sides, an accurate relative calibration is obtained, particularly if the line of interest is optically thin, as H_α clearly is.

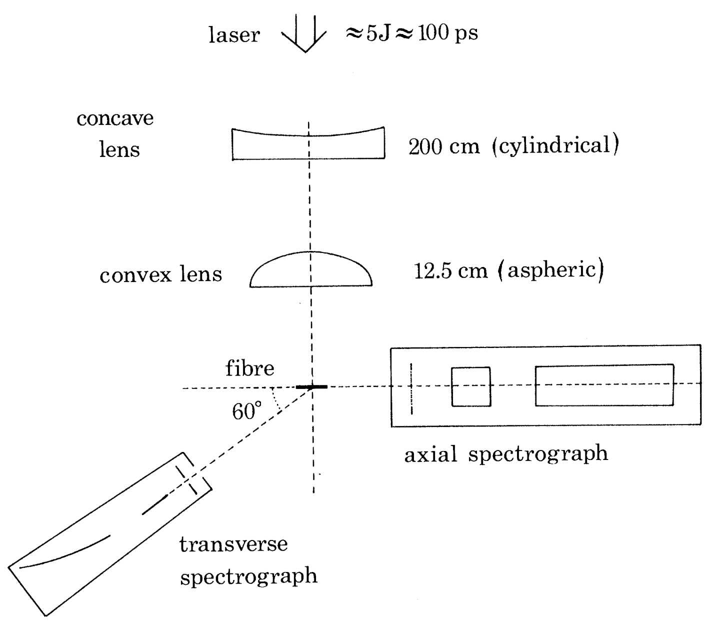

FIGURE 3. Diagram showing the arrangement of the focusing system, fibre and spectrographs used in the experiments.

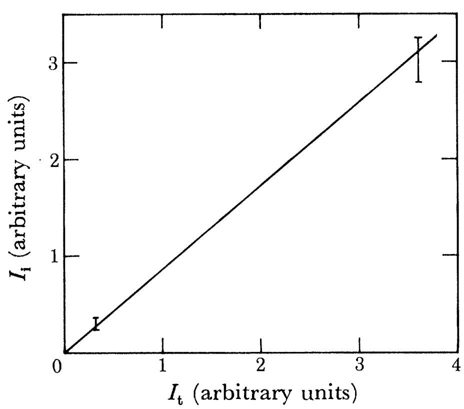

FIGURE 4. Cross-calibration plot of plate intensities between the two spectrographs, I_l and I_t being the intensities measured axially and transversely respectively. The error bars indicate the representative error at each end of the range.

Measurement of gain

Two methods have been used to measure gain. The first follows directly from the cross-calibration of the spectrographs described above. Thus, if the total (integrated over line profile) intensity of the line H_α in the spectrographs along the line and across it are respectively I_l and I_a, then the averaged gain $\bar{G}l$ over the profile along the line is given by

$$I_l/I_a = [\exp(\bar{G}l - 1)]/\bar{G}l.$$

Table 1

transition	wavenumber cm^{-1}	wavelength nm	total oscillator strength gf
$2p^2\ P_{\frac{1}{2}}$–$3d^2\ D_{\frac{3}{2}}$	549184.0	18.208833	1.3916
$2s^2\ S_{\frac{1}{2}}$–$3p^2\ P_{\frac{3}{2}}$	549158.1	18.209692	0.5798
$2p^2\ P_{\frac{1}{2}}$–$3s^2\ S_{\frac{1}{2}}$	549051.3	18.213234	0.0272
$2s^2\ S_{\frac{1}{2}}$–$3p^2\ P_{\frac{1}{2}}$	549017.4	18.214336	0.2899
$2p^2\ P_{\frac{3}{2}}$–$3d^2\ D_{\frac{5}{2}}$	548755.9	18.223039	2.5048
$2p^2\ P_{\frac{3}{2}}$–$3d^2\ D_{\frac{3}{2}}$	548709.1	18.224593	0.2783
$2p^2\ P_{\frac{3}{2}}$–$3s^2\ S_{\frac{1}{2}}$	548576.4	18.229001	0.0543

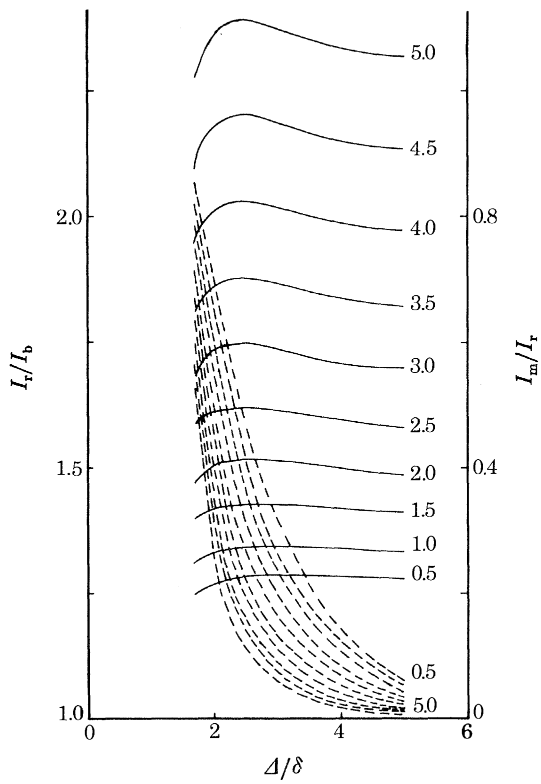

Figure 5. Calculated intensity ratios of the red (I_r) and blue (I_b) peaks, and of the red peak to the minimum (I_m) of the line H_α of C^{VI} treated as a Doppler broadened doublet of separation Δ, and $1/e$-width δ.

The alternative method has the advantage that no cross-calibration is required. As is well known, the line H_α is not a single transition, but comprises distinct transitions at slightly different wavelengths (Garcia & Mack 1965). Since each transition has a different probability, it has a different gain. Thus after passage through a gain medium the line shape profile is enhanced (line narrowing). Examination of table 1 shows that the Balmer α-transition has

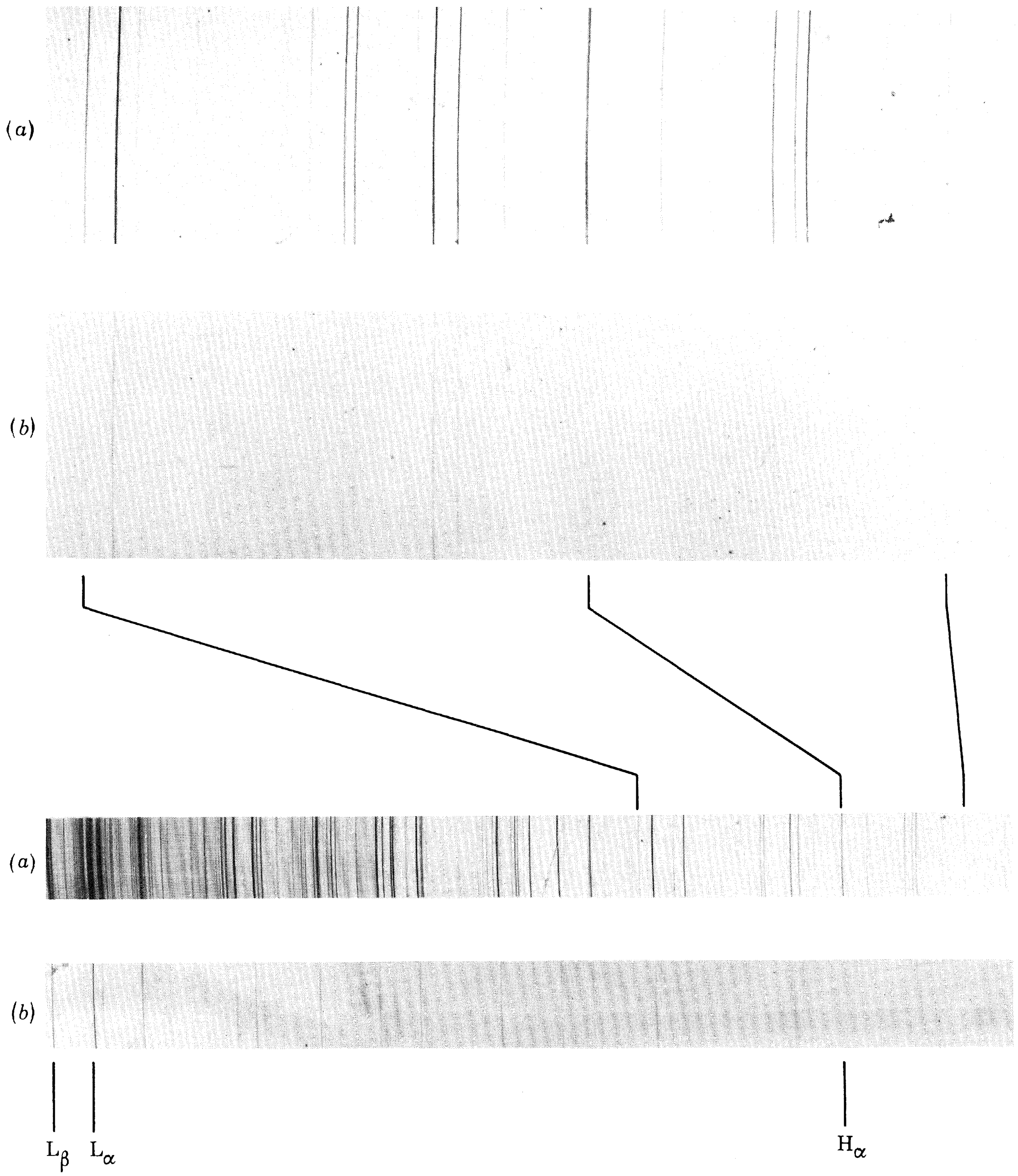

FIGURE 6. Comparison of the spectra obtained on a plate with gain (*a*) and one with zero gain (*b*). The transverse spectrograph showed approximately the same H_α-intensity for both the runs. The lower spectra show the complete plates from the axial spectrograph, and the upper an enlargement in the region of the H_α line. Plate (*a*) was obtained with ten shots of average energy 8.0 J and yielded a *Gl*-value of 4.5. Plate (*b*) was obtained with five shots of average energy 6.0 J, and gave a value of $\bar{G}l$ of 0.1.

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characteristically two groups of lines *ca.* 0.013 nm apart. Numerical calculations show that, if the peaks can be resolved, their intensity ratio is an extremely sensitive function of the gain Gl at the peak of the intense line. By assuming that the ions in the $n = 3$ state are distributed according to their statistical weights figure 5 shows a preliminary calculation of the intensity ratio of the peaks and the minimum as a function of the ratio of line separation to width (Δ/δ) for a pair of Doppler-broadened lines. If $\Delta/\delta < 0.17$ the lines are never resolved. This method is, however, extremely sensitive to departures from exact statistical equilibrium, and consequently provides only a qualitative indication of gain.

Table 2

laser energy	fibre diameter/μm	$\overline{Gl}$
8.0 ± 1.2	4.1	4.5 ± 0.5
6.7 ± 0.6	2.9	2.5 ± 0.5
6.0 ± 1.3	3.3	3.5 ± 0.5
5.9 ± 2.3	2.2	3.0 ± 1.0
6.0 ± 2.2	4.0	0.1 ± 0.5
5.3 ± 1.0	2.5	0.7 ± 0.5
5.2 ± 1.8	4.2	−0.35 ± 0.5
4.9 ± 2.2	4.3	−1.3 ± 0.5
4.9 ± 2.6	4.6	−2.5 ± 0.5

Experimental measurements of gain

Experiments to observe gain by using the above methods are currently in progress. To date ten such experiments have been made and the results are summarized in table 2. About six laser shots are usually necessary to obtain sufficient intensity of H_α on both spectrographs.

Figure 6 (plate 1) shows a spectrum with gain on H_α contrasted with one without gain. The anomalous strength of H_α in the plate with gain can be clearly seen. Furthermore the 'doublet' structure of H_α is well resolved on the plate with gain but not on the one without gain. This is a general result: if there is gain, H_α is a 'doublet', but is otherwise not resolved.

From table 2 some trends are immediately apparent. In particular the gain appears to be extremely sensitive to laser energy, but relatively independent of fibre diameter. An energy of not less than 6 J into a 2 mm line is necessary for gain. If the energy decreases below this value the absorption appears to progressively increase, in agreement with theory.

The measured gain of about two is in good agreement with computational predictions.

Discussion

These results, although rather few in number, clearly indicate the presence of gain. In view of the corroborating evidence from two independent measurements it appears that there is a clear threshold in the energy density below which significant gain is not obtained. Further work is clearly necessary to further identify the threshold behaviour and elucidate the gain scaling.

The significantly large values of gain measured ($Gl \approx 2$) for relatively small laser energy (*ca.* 7 J) indicate the extension of this system to generate laser action by amplified stimulated emission requiring a gain $Gl \approx 10$. Such a system involving an input laser energy focused along a line *ca.* 1 cm long, however, involves difficult axial focusing requirements which must be overcome. It is clear from the current experiments that to obtain a satisfactory conclusion to these experiments the experimental configuration must be carefully designed with due consideration to questions of focal spot uniformity and pre-pulse structure.

One of the attractive features of this recombination system is its scaling to shorter wavelengths by using elements of higher atomic number. Of special interest is the scaling to aluminium with a laser transition at 3.87 nm. Detailed numerical studies show that the energy requirements for foil illumination are very similar to those for a carbon fibre, the principal difference being that the laser pulse length must be shortened to about 25 ps. Fluid code calculations further show good behaviour provided that strong nonlinearities can be avoided. Since the thresholds for these effects scale as $I\lambda^2$, where I is the focused intensity and λ the laser wavelength, the success of these carbon experiments suggests that the green second harmonic at 503 nm should be used for the experiments with aluminium.

The experiments reported here were made by Dr D. Jacoby, Dr G. J. Tallents and Mr L. D. Shorrock, to whom the credit for this demonstration of gain is due.

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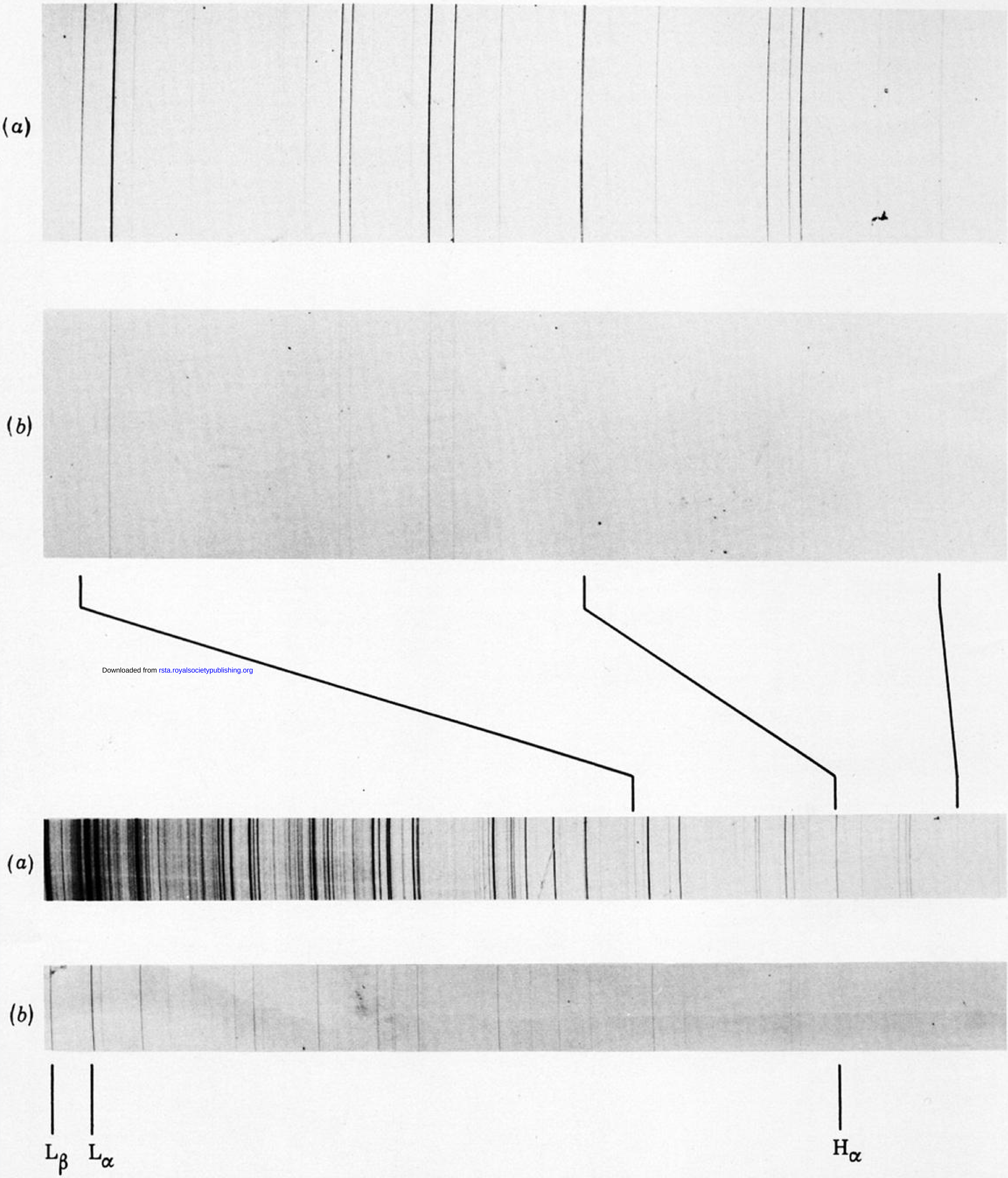

FIGURE 6. Comparison of the spectra obtained on a plate with gain (*a*) and one with zero gain (*b*). The transverse spectrograph showed approximately the same H_α-intensity for both the runs. The lower spectra show the complete plates from the axial spectrograph, and the upper an enlargement in the region of the H_α line. Plate (*a*) was obtained with ten shots of average energy 8.0 J and yielded a *Gl*-value of 4.5. Plate (*b*) was obtained with five shots of average energy 6.0 J, and gave a value of $\overline{G}l$ of 0.1.

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