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## ABSTRACT

'Q'-switched pulses of duration 10 ns and energy 0.5 joules have been amplified to multi-gigawatt powers using a large, single stage $Nd^{+3}:POCl_3:ZrCl_4$ liquid laser. The general design of the amplifier is described together with the gain characteristics and derived laser parameters. Stored energies of 300 J/litre have been achieved of which more than 50% could be extracted with 10 ns pulses at an overall amplifier efficiency of 0.4%.

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## 1. Introduction

The construction of large, high power liquid laser amplifiers has always been of prime importance due mainly to their inherent advantages over other kinds of laser, namely that the active medium can be circulated and is free of damage problems. The development of the $Nd^{+3}:POCl_3:ZrCl_4$ liquid laser system with its high intrinsic gain, has stimulated great interest in the field. Its properties have been extensively investigated in the past few years using small oscillator-amplifier systems (Samelson et al 1970, Andreou 1974). It was the purpose of the present investigation to examine the characteristics of large liquid amplifiers under conditions of high power excitation and the amplification of light pulses of nanosecond duration to multi-gigawatt powers. Many of the problems encountered, particularly those related to the heating of the active medium by the flashlamps, are vital in evaluating the potential of liquid media in general as high power amplifiers. The recent construction of large dye laser systems (~ 400 J) in the Soviet Union has indicated that thermal distortion of the active medium through heating by the pumping radiation still remains a major problem (Baltakov et al 1974).

The laser parameters of the $Nd^{+3}:POCl_3:ZrCl_4$ system are well known (Samelson and Kocher 1974). In addition to its inherent advantages mentioned above it has a larger stimulated emission cross-section ($8 \times 10^{-20}$ $cm^2$) and higher efficiency than Nd-glass; it is also much cheaper and available in arbitrarily large volumes with perfect homogeneity. The problems encountered when constructing an inorganic liquid laser are however considerable. These arise partly from the chemical properties of the medium - its corrosive and volatile nature and the fact that it is a powerful dessicant - and partly from non-linear effects arising from the interaction of the pulse undergoing amplification with the active medium (Alfano et al 1971). In addition to these the thermally induced optical inhomogeneities in the laser liquid due to heating by the pumping radiation is a serious problem (Andreou et al 1972).

In the present work, the construction of a large single stage $Nd^{+3}:POCl_3:ZrCl_4$ liquid amplifier, (700 mm long and 25 mm diameter) is described. Giant pulses of duration 10 ns and energy 0.5 joule have been amplified to a peak power of approximately 3 GW at an extraction efficiency of ~ 50%. Peak power and energy gains of 100 have been obtained with input pulses of ~ 10 ns duration. Patterns showing the beam distortion at different times during the pumping pulse due to heating by the pumping radiation are presented. Non-linear effects occurring in the amplifier have been observed. Total energy conversion to the Raman shifted wavelength - first observed in a travelling wave amplifier in the present system (Green et al 1975) - never exceeded 8%. The spectral and temporal characteristics of the amplified pulses are examined. An assessment of the results together with suggestions for the improvement of the performance of liquid systems is given at the end of the paper.

## 2. Experimental Arrangement

Figure 1 shows a photograph of the $Nd^{+3}:POCl_3:ZrCl_4$ system. The laser solution was contained in a 700 mm long by 25 mm diameter Pyrex cell. The end windows of fused silica were set at the Brewster angle and secured by a chemically resistant adhesive (Gupalon 20 transparent) (Green and Mason 1973). The active medium has a concentration of $1.8 \times 10^{-20}$ ions/$cm^3$ and fluorescence lifetime 330 $\mu s$. Four linear flashlamps 600 mm long and 19 mm bore with a xenon filling pressure of 450 torr were arranged symmetrically around the cell. The light from the flashlamps was concentrated onto the laser cell by an electro-formed silver-plated reflector of a clover leaf cross-section. In order to reduce the distortion of the liquid due to heating, the UV component of the pumping radiation was filtered out by a solution of $K_2 Cr_2 O_7$ in water, contained in a glass jacket surrounding the cell. The filter solution, which was kept at constant temperature through contact with a large external reservoir, was also used to cool the laser liquid by circulating it around the cell and through a heat exchanger. The whole of the laser head was placed in a perspex box for safety purposes against accidental breakage of the laser cell.

The power supply consisted of two banks of 10 μF, 20 kV rapid-discharge capacitors and air-cored 100 μH inductances in a transmission line arrangement. The maximum energy used to pump the laser cell was 15 kJ, with a pumping pulse of 750 μs duration. The laser was fired at intervals of 45 minutes to ensure that the liquid had returned to thermal equlibrium and hence optical homogeneity.

The oscillator-amplifier experimental arrangement is shown schematically in figure 2. The Q-switched oscillator was similar to that described by Andreou et al (1972) for generating 0.5 joule pulses of 10 ns width (FWHM). The width could be adjusted in the range 7 to 15 ns by changing the oscillator cavity length. A 2 mm thick cell containing undiluted Kodak Q-switch solution type A9740 was placed between the oscillator output mirror and the amplifier to prevent coupling between them, and the onset of self-oscillation in the high gain system. The dye cell was also used as a discriminator, to suppress spurious, low intensity spikes occasionally generated in the oscillator. The whole arrangement was aligned with a He-Ne laser placed behind the oscillator.

The amplifier input and output signals were monitored by a pair of ITTF 4000 (S1) photodiodes connected to a Tektronix 519 oscilloscope via power dividers in the manner described by Andreou (1974). Diffusing discs of sintered PTFE were mounted in front of the diodes to avoid any transmission non-linearity caused by fine structure in the laser beam profile. Neutral density filters were placed in the transmitted light to ensure operation in the linear regime. Energy measurements were made with a Gen Tec Joulemeter (ED 500 (1)) connected to a Tektronix 555 oscilloscope.

The spectra of the amplified pulses were recorded on Kodak I-Z plates, using a Hilger and Watts 3-metre prism spectrograph. Spectra of the output were also taken with a Monospek 1000 grating spectrometer, and detected on a Mullard 6929-1 (S1) image converter placed in the plane of the exit slit (Andreou and Little 1973). In both cases the exit of the amplifier was imaged at unit magnification onto the slit of the spectrometer with a 35 cm focal length lens. Glass disc attenuators were used to protect the spectrometers from damage by the high laser intensities.

## 3. Amplification of Q-Switched Pulses

In order to complete the comparison between the earlier results obtained with small liquid lasers, and the present, much larger version, the laser was first operated in the free running mode. Figure 3 shows the results obtained for the output energy of the oscillator as a function of the pumping energy for a variety of output mirror reflectivities. The maximum output of nearly 150 joules was recorded with a 6% mirror reflectivity for a pumping energy of

12 kJ. Figure 3 shows the laser slope efficiency of 2%. The results are similar to those reported by Samelson et al (1970) but on a much larger scale.

Before describing the performance of the amplifier it will be useful to outline briefly the temporal and spectral characteristics of the input pulses. Q-switched pulses from Nd-liquid oscillators have been shown to be much shorter than those obtained from glass oscillators (Andreou et al 1972). The reason for this is that the lower laser level relaxation time to the ground state is very short (less than 5 ns (Hongyo et al 1972)) and the liquid laser behaves as a four level system even in the Q-switched mode of operation. The short lifetime of the lower laser level is of great importance in the amplifier operation also, for it means that the amplifier behaves as a four-level system even during the amplification of light pulses a few nanoseconds long. This partly accounts for the much higher efficiency of liquid amplifiers compared to glass amplifiers in the nanosecond region. Another characteristic of interest is the narrow spectral linewidth of the Q-switched pulses, from Nd-liquid lasers approximately 7 - 8 Å (Andreou 1974), compared to those from glass lasers (approximately 25 - 40 Å, Voronich et al 1969)). This means that the spectral cross-relaxation in liquids (which has not yet been measured) is much faster than in glass (of order of microseconds, Mak et al 1972)).

Figure 4 shows the results obtained when the laser was operated as an amplifier. The oscilloscope trace displays the 'Q'-switched pulse before and after amplification. The output pulse has been attenuated approximately 100 times with respect to the input pulse.

The hump after the amplified signal is a reflection of the first signal at the end of the cable connecting the photodiodes, which could not be eliminated.

At high power levels a temporal pulse broadening occurs as shown in Figure 4c. The distortion of light pulses in high power amplifiers has been studied extensively and shown to be strongly dependent on the shape of the input pulse. For pulses with exponential rise time pulse broadening occurs, whilst Gaussian shaped pulses experience pulse shortening (Kryukov and Letokhov 1970) (in both cases compact support was assumed) (Icsevgi and Lamb 1969). In the present experiments the Q-switched pulses from the liquid oscillator had an exponential rise with a time constant $\tau_o \sim 1.5 \times 10^{-9}$ s. It has been shown that during the non-linear amplification of a pulse with exponential leading front, a shift of the maximum takes place causing pulse broadening to occur (Kryukov and Letokhov 1970). This shift is approximately equal to $\Delta\tau \approx \alpha_o L_{nonl} \tau_o$, where $\alpha_o$ is the small signal gain coefficient and $L_{nonl}$ is the distance traversed by the pulse with energy $E > E_s$ i.e. the length of the non-linear gain. In the present case the values $L_{nonl} \sim 30$ cm and $\alpha_o = 0.12$ cm$^{-1}$ have been calculated from the amplifier equation and the experimental results, giving a shift of the maximum $\Delta\tau \sim 5$ ns. The shift observed in the present system is approximately (2.5 - 3) ns The main error lies in the determination of the length $L_{nonl}$ due to the attenuation of the energy density as a result of divergence.

Figure 5 shows the energy output of the amplifier for various pumping energies and at different times during the pumping pulse.

Figure 6 shows the power gain of the peak of the pulse. This was found by comparing the input and output signal heights recorded by the diodes, Figs 4b, c. These were normalized with respect to the signals obtained when the amplifier was unpumped (Fig. 4a). The true value for the gain was found by multiplying by the attenuation of the filter used in the diode monitoring the amplifier output. The maximum laser pulse energy recorded by the Joulemeter was 48 Joules for a pulse width of 13 ns, the average intensity being approximately 750 MW/cm$^2$. The output beam profile is sharply peaked at the centre. In order to discover how the energy is distributed in the beam the output was sampled by five beam splitters, each one progressively attenuated. By allowing the five beams to burn unexposed polariod film (which has a distinct energy density threshold) a 'contour' map of energy density can be plotted (Figure 7(a)).

Figure 7(b) shows the relative intensity of the beam in space, where each circle represents the average value of each contour line obtained in the experiment. In this way a lower estimate of the peak power density that occurs at the centre of the beam can be made. This is not less than 3.3 GW/cm$^2$.

Figure 8 shows the burns made by the beam when it falls on to unexposed polariod film for different times during the pumping pulse for a pumping energy of 7 kJ. Clearly the beam suffers considerable distortion which increases with time. At higher pumping energies the distortion was more serious. The burns show in a qualitative way the distortion that arises in the liquid during the pumping pulse.

The spectrum of the laser output was measured using a 5 element neutral density step filter placed in front of the spectrometer slit to enable the line shape to be found. In this way fig. 9 was plotted showing the approximate spectral linewidth of the amplified pulse to be 6 Å FWHM.

Finally we must point out that conversion to Raman wavelengths did not exceed 8% of the total output. This is mainly attributed to the fact that the input pulse was distributed over many transverse and longitudinal modes, of which no one reached an intensity high enough to cause large conversions by itself (we recall here that stimulated Raman scattering grows as $dI_{Raman}/dx = K\, I_{Raman}\, I_{Laser}$ for a single mode of radiation). The growth of stimulated Raman scattering (SRS) in high power liquid amplifiers is at present under more detailed investigation. Initial results, including transient effects, have been reported (Green et al 1975).

## 4. Discussion and Conclusions

In this paper we have reported the performance of a single stage $Nd^{+3}:POCl_3:ZrCl_4$ liquid laser amplifier at power levels near the damage threshold of glasses. The laser has been operated for a range of pumping energies up to 12 kJ for more than 2500 firings, with no apparent deterioration in performance. The results are very encouraging from the point of view of efficiency, damage threshold and lifetime of the system. The only serious problem appears to be the poor beam quality resulting from thermal distortion induced in the medium by optical pumping, and the relatively large divergence (~ 20 mrad) associated with it. A similar result was obtained with a large co-axially pumped dye laser, for which a divergence of ~ 30 mrad was reported (Baltakov et al 1974). The situation could be much improved with better matching of the lamps to the liquid (~ 300 μs discharge) and an alternative geometry for more uniform pumping e.g. disc laser. The main advantages of the liquid laser medium compared with glass at high peak power are summarized in the following table:

| | Liquid | Glass | Comments |
|---|---|---|---|
| Stimulated emission cross-section | $8 \times 10^{-20}$ cm$^2$ | $\sim(0.5-3)10^{-20}$cm$^2$ | The liquid has higher gain coefficient for the same inversion. |
| Lower laser level lifetime | < 5 ns | > 12 ns | The liquid laser amplifier behaves as a four level system even for pulses a few nanoseconds long. This factor also contributes to higher efficiency. |
| Damage threshold | No damage threshold | ~(3-4)GW/cm$^2$ | |
| Cost | ~ £300/litre | ~£10000/litre | |
| Deterioration in performance | No deterioration after ~ 2500 firings | Deterioration begins at ~ 2000 firings | In both cases high power amplification is assumed (over 1 GW/cm$^2$). |

The two main disadvantages of the liquid laser compared with glass are summarized below.

| | Liquid | Glass | Comments |
|---|---|---|---|
| Pumping | thermally induced optical inhomogenieties | thermal bi-refringence | The liquid system gives low beam quality compared with glass and also has bigger divergence. |
| Non-linear effects | Conversions to Raman wavelengths. Non-linear refractive index $n_2 = 7 \times 10^{-13}$ cgs (esu) | Self focusing can cause damage. Non-linear refractive index $n_2 = 1.52 \times 10^{-13}$ cgs(esu) | The difference in value of the non-linear refractive indices is not as detrimental as it first appears to be because the perturbation parameter for self-focusing in an amplifying medium is proportional to $n_2/\alpha_0$, where $\alpha_0$ = gain coefficient of the medium (Suydam 1975), and $\alpha_0$ is higher in the liquid laser. Thus the non-linear parameters for beam instability are roughly comparable (ratio 1.7:1 for liquid:glass (ED-2)). |

The results presented here show that the liquid laser can be a very useful tool for the production of cheap high power radiation energy at 1.052 μm wavelength from fairly simple systems. In addition the high extraction efficiency obtained from a straight forward oscillator-amplifier combination (more than 50% of the stored energy at an overall amplifier efficiency of 0.4%) can be used to advantage in the amplification of powerful nanosecond pulses (Figure 10). The observed extraction efficiency for the liquid laser is in close agreement with the theoretical predictions of Fill and Finckenstein (1972), and is much greater than that for glass under similar conditions (~ 10%).

## 5. Acknowledgements

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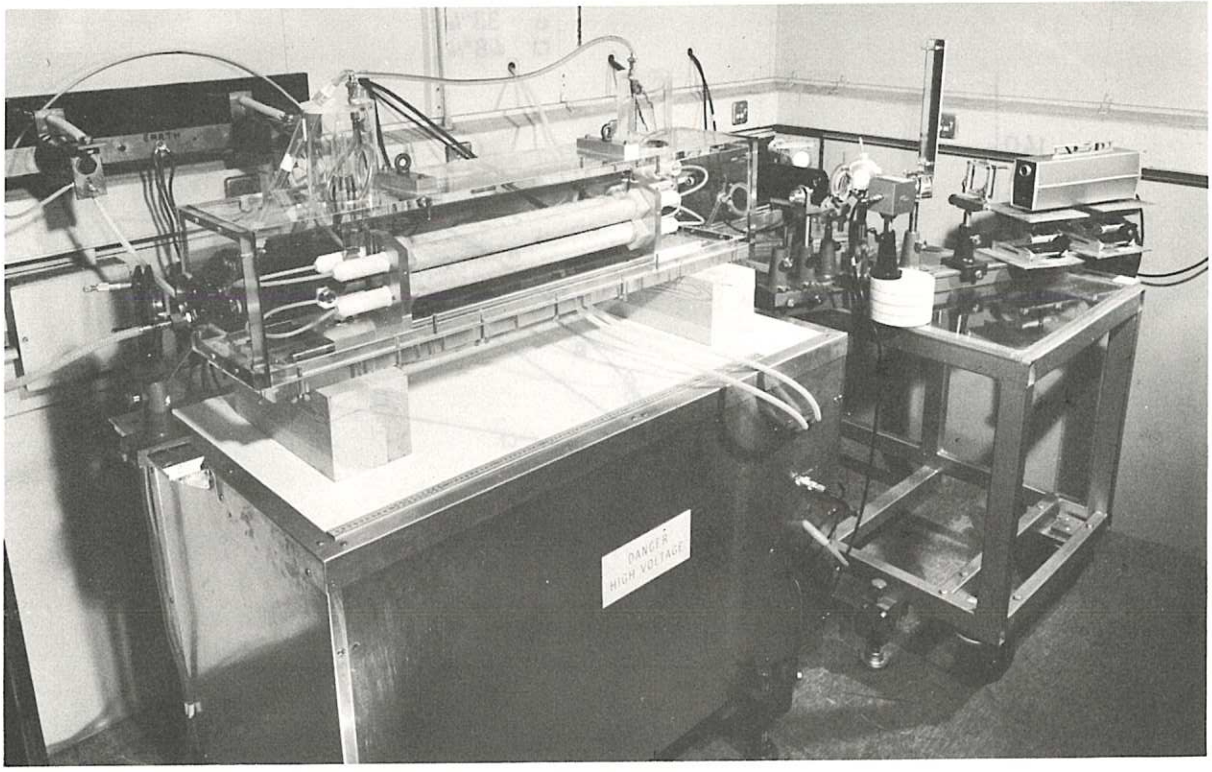


Fig.1 Photograph of the 700 mm long, 25.4 mm diameter liquid laser amplifier system Nd$^{+3}$:POCl$_3$:ZrCl$_4$. The laser head is placed in a large perspex box for safety purposes against accidental breakage.

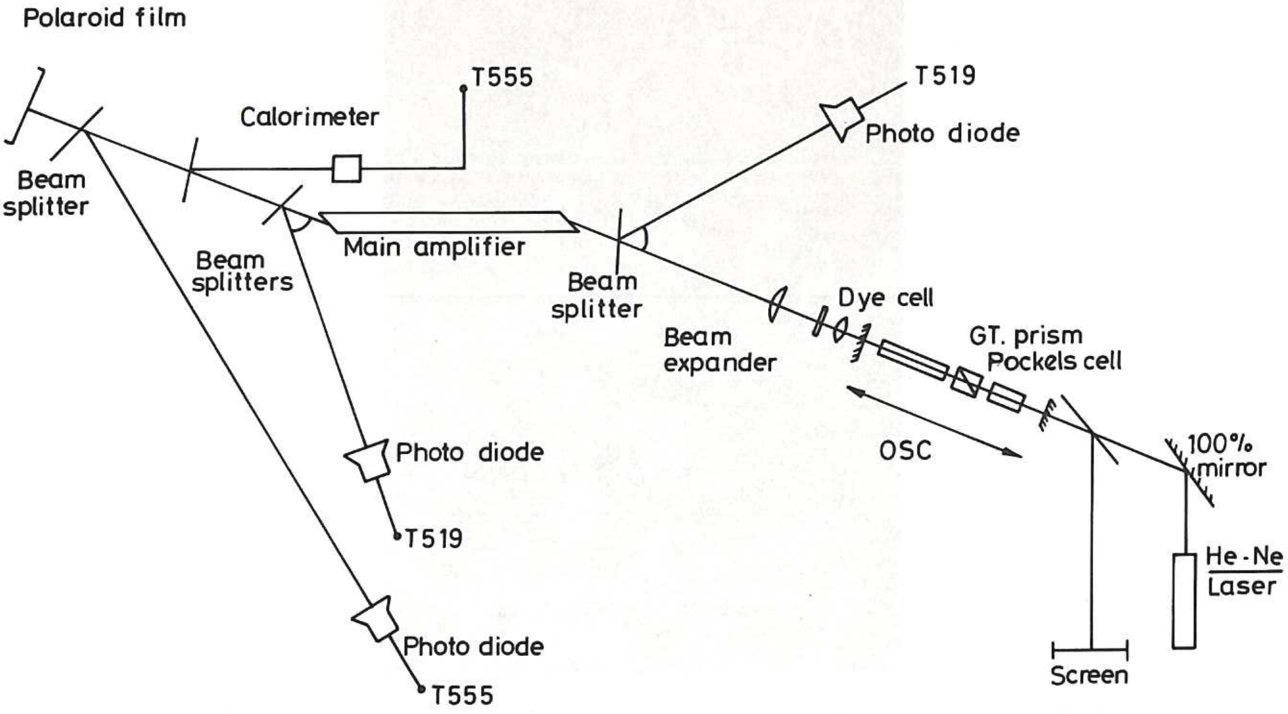


Fig.2 Experimental arrangement for the amplification of giant pulses.

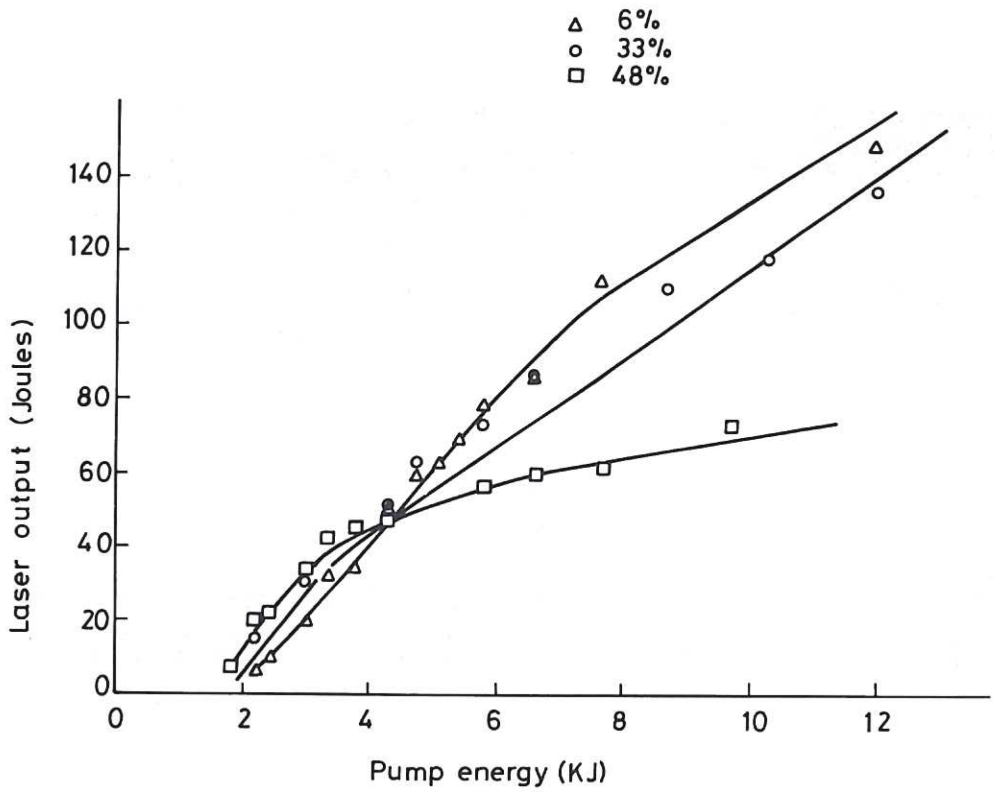


Fig.3 Experimental results showing the input-output energy curves for three different output mirror reflectivities when the system was operated as an oscillator.

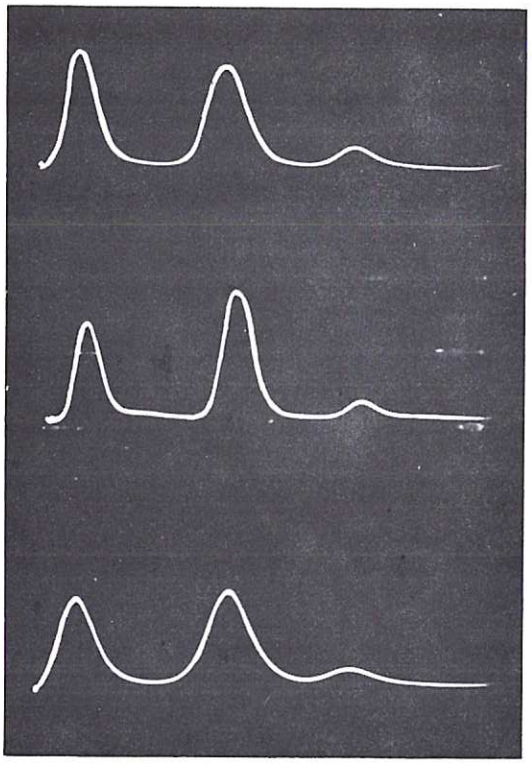


Fig.4 (a) Oscillogram showing the input-output signals for the unpumped amplifier. Time scale 13.6 ns/cm.
(b) Oscillogram showing the input-output signals when the amplifier was pumped with 7 kJ energy. Time scale 13.6 ns/cm. Time after firing the flashlamps 500 μs. The output pulse was attenuated 60 times with respect to (a).
(c) Oscillogram showing the input-output signals when the amplifier was pumped with 12 kJ energy. Time scale 13.6 ns/cm. Time after firing the flashlamps 450 μs. The output pulse was attenuated 100 times with respect to (a). By measuring the separation of the maxima in (a) and (c) it is easily seen that the maximum of the pulse in (c) has moved forward by approximately 2.5 ns. It can also be seen in (c) that pulse broadening has occurred during the amplification. The pictures have been retraced for clarity.

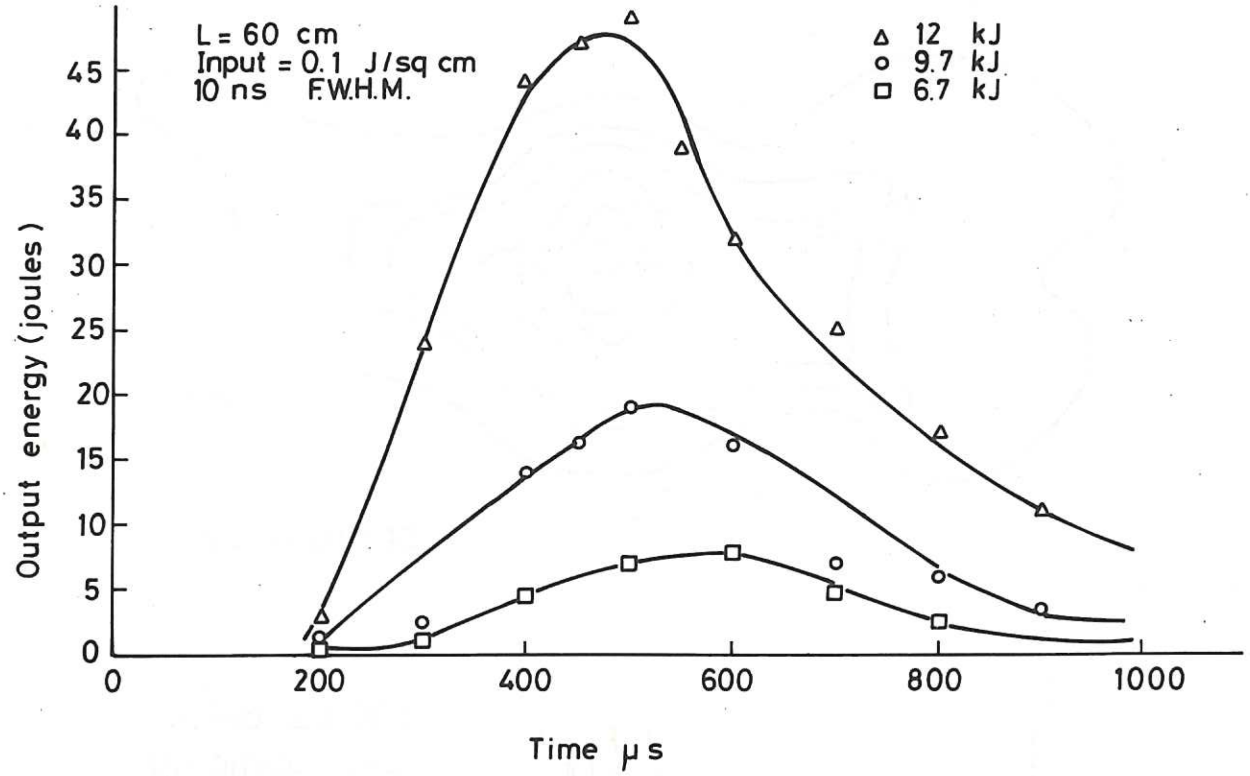


Fig.5 Graphs showing the output energy of the amplifier as a function of time delay after firing the flashlamps for three different pumping energies.

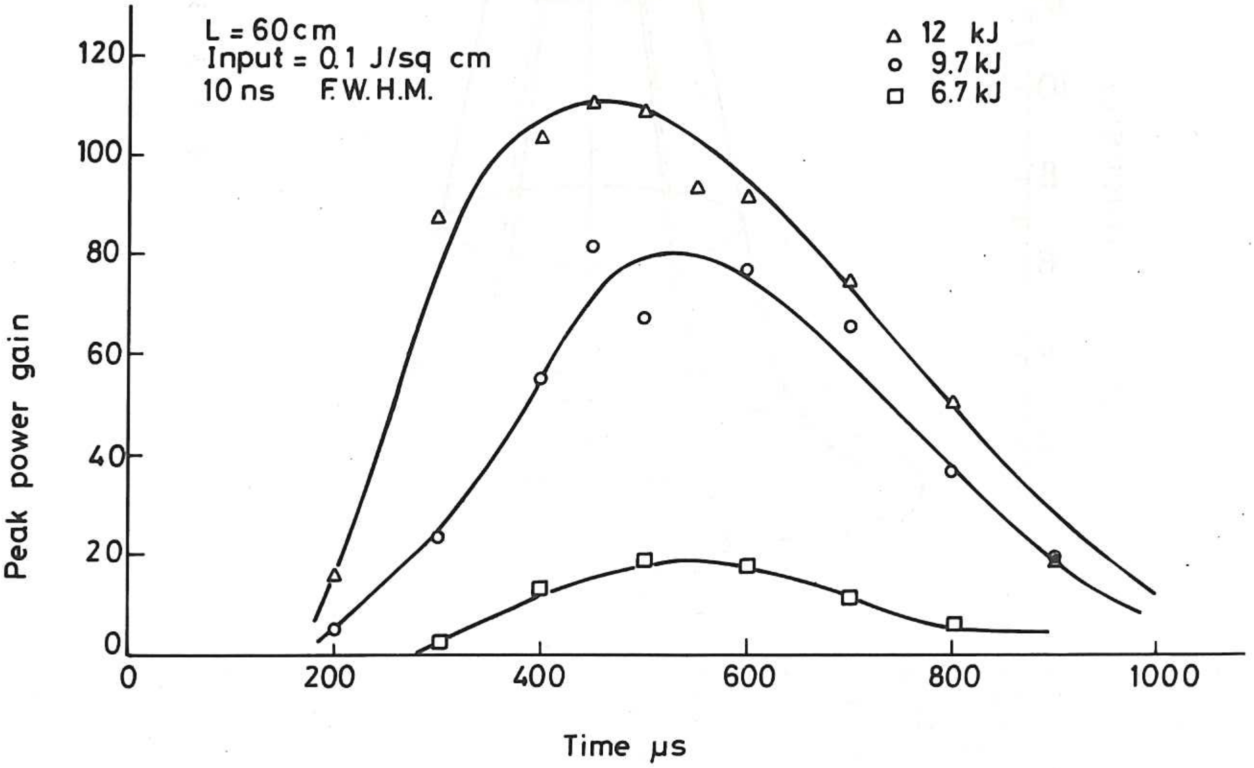


Fig.6 Graphs showing the peak power gain as a function of time delay after firing the flashlamps for three different pumping energies.

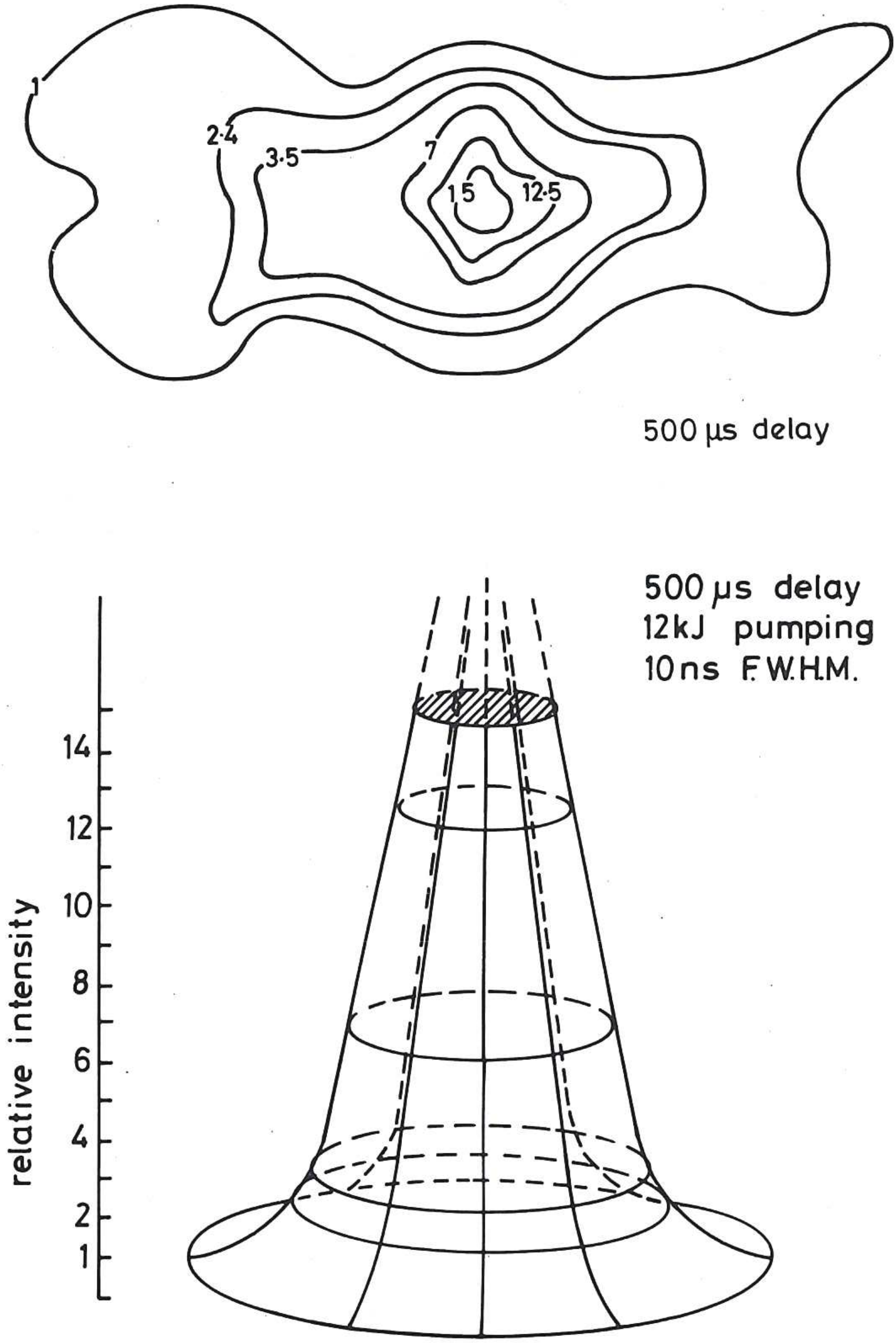
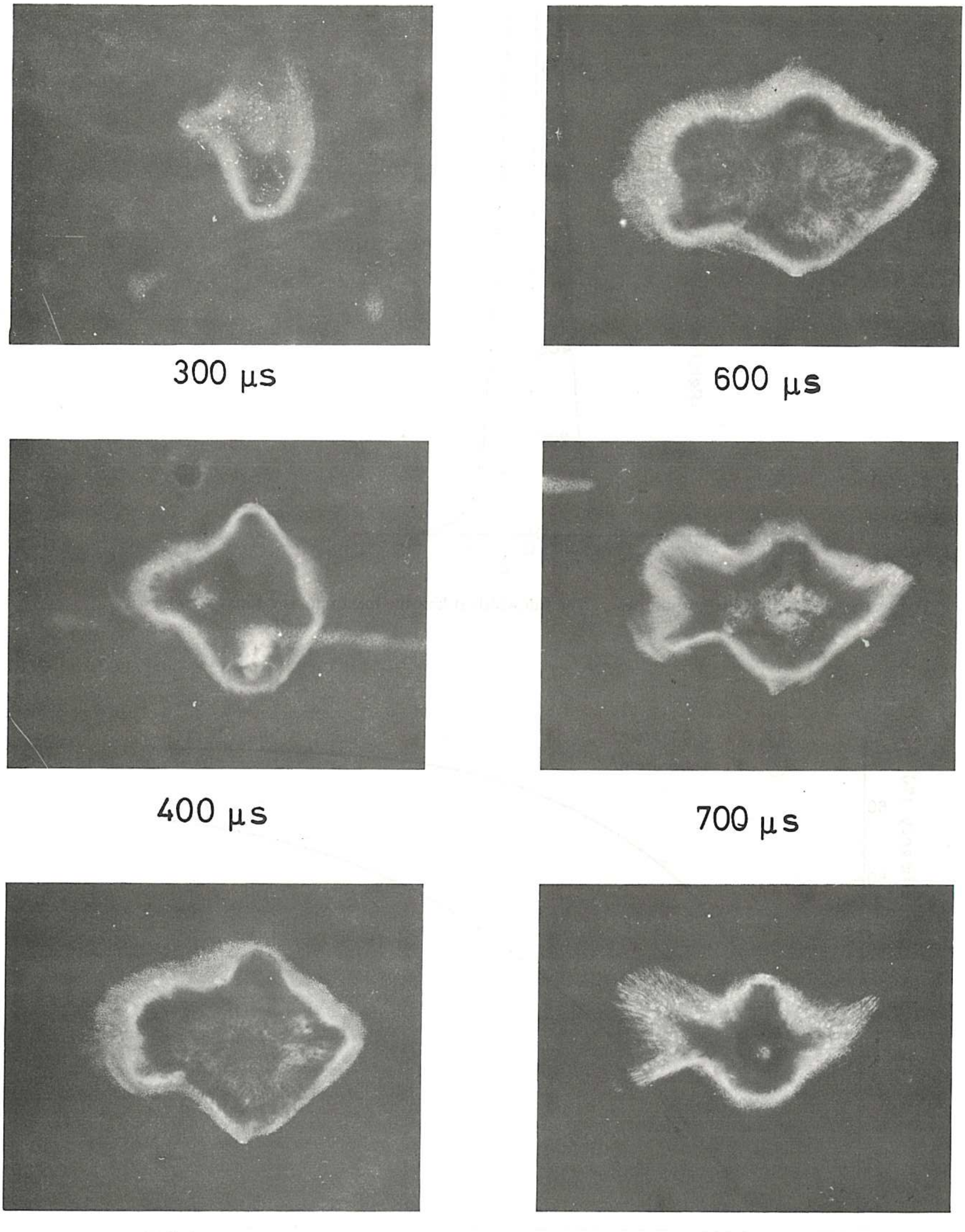


Fig.7 (a) Contour map showing the distribution of relative energy density in the near field. Numbers refer to the level of attenuation for the corresponding beam. (b) The relative intensity in space of the amplified pulse. Each horizontal circle represents the average of each contour line (Fig.7(a)) obtained from the experiment described in the text. In actuality the beam is less uniform (cf. Fig.8) but this figure served as a guide to estimate the peak power at the centre of the beam.



300 µs

600 µs

400 µs

700 µs

500 µs

800 µs

Fig.8 Beam patterns obtained on developed unexposed polaroid film at a distance of 150cm from the output of the amplifier for a pumping energy of 7kJ. Time intervals measured from the beginning of the discharge.

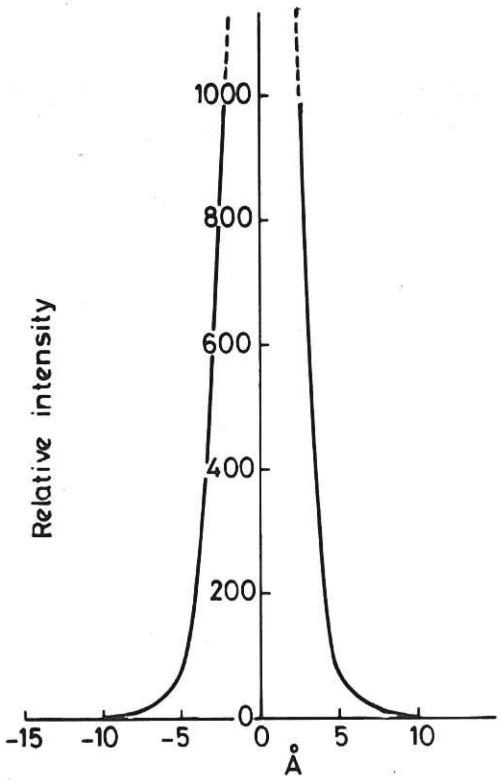


Fig.9 The spectrum of the output pulse. The full width at half the maximum is estimated to be 6 Å.

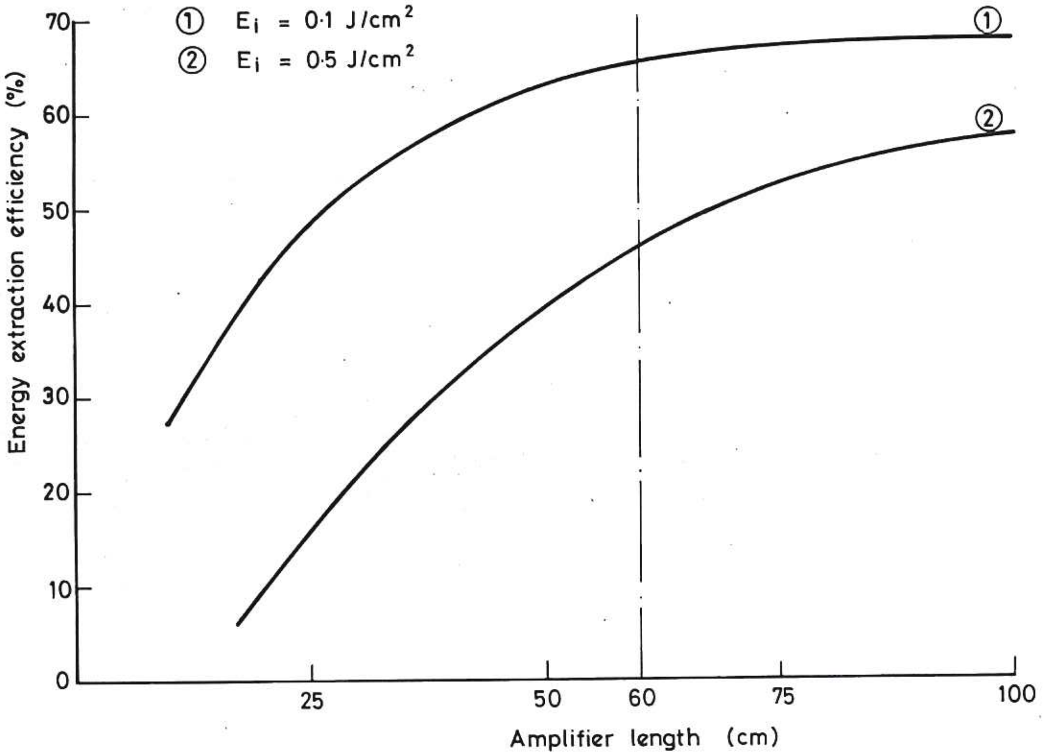


Fig.10 Energy extraction vs. amplifier length, calculated for 10 ns pulse width. Curves adapted from computer solutions of Fill and Finckenstein (1972), with saturation parameter $E_s = 2.4\,J\,cm^{-2}$, small signal gain coefficient $a_o = 0.12\,cm^{-1}$, loss coefficient $\gamma = 0.006\,cm^{-1}$.