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Report

REPORT ON THE LIQUID LASER PROJECT 1970-76

D ANDREOU
M R GREEN
A C SELDEN

CULHAM LABORATORY
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REPORT ON THE LIQUID LASER PROJECT 1970-76

D Andreou, M R Green* and A C Selden

Culham Laboratory, Abingdon, Oxon OX14 3DB, UK
(Euratom/UKAEA Fusion Association)

ABSTRACT

The objectives of the liquid laser research project were to investigate the high power and repetition rate possibilities of the medium. The characteristics of the two devices constructed toward this end, a 10 pps circulating system and a multi-gigawatt amplifier, are described and the principal results summarised. This is followed by a review of the physics of the liquid laser, concluding with a discussion of its potential applications and prospects for future development. A set of tables giving the relevant parameters of the laser medium is appended, together with a full list of publications by the authors.

*Now at CRPP, Lausanne, Switzerland CH-1007

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1. INTRODUCTION

The liquid laser programme initiated at Culham in 1970 was intended to investigate the laser characteristics and long term potential of the new laser medium, with the aim of exploiting its fluid properties in two ways, viz:

(i) in flow systems capable of pulse repetition rates up to 10 pps;

(ii) in static systems of large volume generating multi-gigawatt pulses for plasma heating.

A representative system of each type was constructed and tested during the period, and the above objectives were met. The performance and experimental parameters of each were determined both for scaling purposes and with a view to possible applications of the liquid laser.

In the first - item (i) - it was necessary to take account of the effects of flow and cooling on beam quality and reproducibility of the output; in the second - item (ii) - both linear and non-linear effects needed to be controlled for the maximum focused power density to be reached on target.

The experience gained with circulating the active medium in a repetitively pulsed system can be applied to overcoming the very long cooling times associated with the large neodymium glass laser systems currently used in plasma heating and compression experiments. By contrast with solid state lasers, the liquid is available in large optically homogenous volumes at a nett cost in direct proportion to the total volume used, a fact which makes it increasingly worth while considering fluids - both liquid and gaseous - as the active media for scaling to the ever more powerful lasers required in fusion research.

The discovery and early history of the neodymium liquid laser is described in the literature (A Heller, Physics Today, 20 (11), 34-41, Nov. 1967); here we outline the results of our research on it and our understanding of the laser physics involved. We also give brief descriptions of the circulating system and the multi-gigawatt amplifier, concluding with a discussion of the potential performance of the liquid laser in these two areas.

Tables of the relevant laser parameters and a few selected figures are included. The chemical properties, precautions in handling, storage and use of the liquid, and experience in its laser operation, are described and carefully documented in the theses (A C Selden 1970, D Andreou 1973, M R Green 1976; Ph.D theses, University of London).

2. LIQUID LASER OSCILLATORS

Two kinds of liquid laser oscillator have been developed. Small systems with dimensions of the order of 80 mm long and 4 mm diameter which could deliver energies up to 100 mJ, and larger systems with dimensions of the order of 150 mm by 8 mm which could give Q-switched output energies of over 1 J.

The smaller systems had an overall efficiency of 0.4% but unfortunately this was reduced by a factor $\sim$ 3 when the systems were Q-switched, largely because of the extra losses introduced by the polarising prism. However, the slope efficiency remained practically the same. Such systems find application in rangefinders and hole drilling in thin metals; our results have shown that the liquid laser is ideal for such uses.

A Q-switched laser oscillator which delivered up to 1 J in multimode operation and over 100 mJ in single mode stable cavity operation has been constructed. This was used to drive a large single-stage amplifier. Q-switched outputs with intensities of up to 120 MW/cm^2 could be obtained before stimulated Raman effects were observed. The latter severely affected laser operation for powers in excess of 150 MW/cm^2.

Therefore, it is important to use the minimum output mirror reflectivity possible in order to lower the power per unit area within the resonator, and thus obtain less Raman conversion for higher power output.

The emission characteristics of neodymium liquid lasers differ from those of glass and crystal lasers in a number of ways. Free running liquid oscillators exhibit more regular relaxation oscillations than either glass or YAG lasers, and it appears that it would be easy to bring them under complete control by modulation techniques. This behaviour may be closely related to the effect of self-Q-switching (discussed in section 5 below) and it can be of great importance in applications requiring regular pulse trains with a duration of 200-300 μs, eg small hole drilling, high speed photography and multi-frame interferometry. The output of Q-switched neodymium liquid lasers consists of completely smooth Gaussian pulses in time, in contrast to the modulated pulses obtained using either glass or YAG systems. The pulse duration is also less, typically 5 to 10 ns FWHM, because of the higher gain and consequent reduction in build up time. The recorded emission spectrum was of the order of 5 cm^{-1} FWHM for multimode operation, a figure much smaller than the corresponding value in glass (approximately 20 cm^{-1}).

3. REPETITIVELY PULSED LASER

Because the active medium can be flowed through the laser cell, the inclusion of a suitable pump and heat exchanger allows a circulating system to be constructed for repetitively pulsed operation of the liquid laser. By replacing the liquid between shots with freshly cooled material, only the flow characteristics and single pulse behaviour need be examined once the system has reached thermal equilibrium. Work on a 10 pps Q-switched liquid laser was carried out under contract in the period April 1970 to May 1972 (refs. 8 and 9 in publications list) with the aim of developing 100 mJ output per pulse. The system was constructed of pyrex, nickel and PTFE in chemically clean and dry conditions, and purged with helium before filling, thus avoiding contamination by adsorbed water and the consequent formation of scattering particles (precipitates) in the laser medium. Separate Q-switching experiments were carried out with static cells of different dimensions to determine the optimum size for the required output, and the corresponding input energy to the flashlamps. Measurements of turbulence and optical scattering in the liquid as a function of flow rate were made, using a helium-neon laser. Circular symmetry of the output beam was observed with the flow cell immersed in cooling fluid, indicating sufficiently uniform excitation of the laser medium in the two lamp close-coupled arrangement used for optical pumping. At sufficient flow rates to clear the cell volume between shots and achieve fully developed turbulent flow (to avoid thermal lensing effects associated with laminar flow) good reproducibility in the pulse energy at 10 pps was also observed. The inner surface of the cell in contact with the liquid was roughened to reduce whispering modes, resulting in improved output efficiency. These results show that good optical quality can be maintained in circulating liquid lasers, with repetition rates limited only by the flow rate, and with energy scaling dependent on power supplies.

4. GW AMPLIFIER

A four-lamp assembly containing a liquid laser tube 600 mm long and 25 mm diameter, together with a liquid filter and cooling jacket, was constructed to generate multi-gigawatt pulses, and investigate the linear and non-linear processes associated with their production. The system operated as a single pass amplifier with an energy gain of 100. Input pulses of 50 MW peak power and energy 0.5 J were provided by a small Q-switched oscillator 155 mm x 7.5 mm. These were sufficient to saturate the gain of the large amplifier and therefore control its output. (This probably represents the only neodymium system capable of amplifying Q-switched pulses to multi-gigawatt power levels in a single stage.) Multimode outputs with 48 J maximum energy and 13 ns pulse duration, corresponding to peak powers of 3.5 GW and average intensities of $\sim$ 750 MW cm^{-2}, were observed for flashlamp pumping energies of 12 kJ in 700 μs. The peak intensity at the centre of the beam was estimated to be $\geqslant$ 3.75 GW cm^{-2} from the measured intensity profile.

For input energy densities of only 0.1 J cm^{-2}, giving 10 J cm^{-2} output, extraction of $\sim$ 45% of the stored energy - estimated from the measured small signal gain - was observed. This is to be compared with less than 5% for glass in a similar situation. The overall electrical input to optical output efficiency was 4.10^{-3} for 10 ns pulse width. By comparison the total efficiency increased to 1.2% when the laser was run as an oscillator in the long pulse mode (500 μs) with an output energy of over 140 J.

Observations of optical distortion induced by discharging the maximum pumping energy of 12 kJ, recorded by photographing a grid through the length of the liquid column, showed the inner 40% (diameter) to be practically undisturbed 10 seconds after firing. However, the optical distortion of the outer regions was sufficient to produce severe flaring of the output beam profile. The recovery time for complete optical homogeneity to be restored was 45 minutes for the static volume, indicating a clear need for the liquid to be circulated through a heat exchanger between shots.

Observations were made of energy conversion by stimulated Raman scattering, generation of back-scattered radiation, transient depletion of the laser pulse intensity, emission of anti-Stokes radiation, spectroscopic (line) profiles, linear optical distortion and scattering in the high-gain medium, and pulse broadening associated with gain saturation when the amplifier was operated under these conditions.

5. LASER PHYSICS

During the period of the development of liquid lasers at Culham a number of scientific observations have been made and experiments performed to investigate the physical aspects of liquid amplifying media and the amplification of light pulses in general.

Experiments carried out on small liquid laser systems have indicated the dependence of the transverse mode structure on the symmetry of thermal lensing induced in the liquid by the optical pumping radiation. Transverse modes corresponding to the TEM_{mn} patterns of rectangular symmetry were observed when the system was pumped by a pair of flashlamps and passively mode-locked. The dependence of periodic pulse generation on mode-selection in the cavity has also been demonstrated.

A mode-locked glass oscillator was used to investigate the amplifying characteristics of the liquid medium in the pico-second regime. The oscillator delivered mode-locked pulse trains with total energy of approximately 50 mJ and individual pulse duration of 5 ps, as measured by the two photon fluorescence method. The gain of the liquid laser amplifier was measured by recording the oscillator and amplifier signals as two inter-penetrating pulse trains on a fast photodiode. The observed transmission loss (of the unpumped liquid medium) was 0.5 ± 0.2%/cm, which is in close agreement with other reported values. Although there is a mismatch in the line centres of the liquid and glass lasers, gains of nine have been obtained from a liquid laser amplifier 150 mm long and 7 mm diameter. Saturation effects have also been observed for high pumping of the amplifier, which enabled us to determine the stimulated emission cross-section of the liquid laser medium. The latter was found to be $7.5 \pm 3.0 \times 10^{-20}$ cm^2, which is in good agreement with the values obtained by spectroscopic methods. The saturation curves followed the predictions of laser amplifier theory.

By measuring the gain of the amplifier at different times during the firing of the flashlamps, an estimate of the effective lifetime of the upper laser level could be made, assuming that the pumping rate was proportional to the light power output from the flashlamps, whose pulse shape was previously recorded with a suitable photodiode. The effective lifetime was found by making a computer search for the value which would make the theoretical curves expected from the rate equations match the experimental gain measurement curves. The value giving the best fit was found to be 140 μs, which is less than the fluorescence lifetime of ∿ 300 μs.

The Nd:liquid laser exhibits the rather unusual phenomenon of self-Q-switching, namely that the output of the laser consists of isolated pulses whose number decreases and whose peak power increases as the output mirror reflectivity is reduced. Experiments have been carried out in our laboratory in order to explore this behaviour of the liquid laser. From theoretical considerations a thermal mechanism has been proposed, based on the fast non-radiative decay to the ground state of the terminal level of the laser transition. This produces a spatially periodic deposition of heat, with maxima corresponding to anti-nodes of the simulated optical standing wave (because the population of the lower laser level is determined by the stimulated emission process). As a result, a phase grating is formed in the liquid, with a reflectivity proportional to the cavity gain. Owing to the much greater thermal expansivity of liquids over solids the effect of this process is significant in liquid lasers. In order to examine the relation of the gain to the self-Q-switching of the liquid laser, experiments have been carried out using combined active media, with a glass and a liquid laser placed together in the same cavity. In this way the gain could be increased by pumping the glass laser without affecting any of the liquid laser parameters in the cavity. Strong self-Q-switching occurred when the glass laser was fired at the same time as the liquid laser, indicating the dependence of self-Q-switching on the overall gain of the cavity. A single high power pulse was also generated by the 60 cm GW amplifier with a 100% R mirror set at one end.

The fundamental light amplification processes suggest that if a Gaussian (or Lorentzian) shaped narrow spectrum signal is amplified by a wide-band amplifier whose line centre does not coincide with that of the signal, frequency shifts occur during the amplification process. As the peaks of the spectra of the Nd:glass and Nd:liquid systems do not coincide, such shifts should be observable provided the pulse duration is short compared with the spectral cross-relaxation time for the amplifying medium. We have used the output from a mode-locked Nd:glass laser as the input signal to the Nd:liquid amplifier and recorded the spectra using an image converter (ref. 13 in publications list). Frequency shifts of over 20 Å towards the centre of the amplifier linewidth were observed. The effect of such frequency mismatches on the power gain of a laser amplifier has also been demonstrated by measuring the gain of the liquid amplifier in the forward and backward directions. The gain in the backward direction was found to be greater than that in the forward direction as a result of the frequency shift introduced in the spectrum of the mode-locked pulses during their first pass through the amplifier. The difference in gain was also calculated theoretically and found to be in close agreement with the experimental results.

Observations of temporal broadening and forward movement of the pulse maximum have been made on a single stage multi-gigawatt amplifier. The peak power point advanced by approximately 3 ns, consistent with the calculated value of $\sim$ 5 ns for the experimental conditions.

Simulated Raman scattering has been observed in the output of a Q-switched Nd:liquid laser oscillator when the power density within the cavity exceeded 600 MW/cm^2, and output powers amounted to more than 150 MW/cm^2. It is thus important to make sure that the input to a high gain liquid laser amplifier does not contain any Raman wavelengths, otherwise their growth will deplete the laser light during amplification. Observation of stimulated Raman scattering from a travelling wave Nd:liquid laser amplifier 60 cm long has also been made. The output Raman energy was measured to be less than 10% of the total for a multimode input beam corresponding to output powers of 660 MW/cm^2. In some of the recorded spectra the anti-Stokes emission line was just observable. Narrow intense forward travelling pulses superimposed on the smooth envelopes of the giant pulses usually accompanied Raman scattering. Investigation of these pulses showed that two types of structure can be distinguished: a narrow intense forward-travelling light pulse and a jagged "depletion" of the amplified pulse shape. Simple theoretical calculations on the growth of stimulated Raman scattering have indicated that once it starts it grows at a very fast rate during the amplification of single mode radiation, and will totally deplete the laser light within a few centimetres at gigawatt power levels. It is thus important for the construction of multi-gigawatt amplifiers, that the Raman intensity is not allowed to rise above noise level. This can be done by introducing selective rejection filters between stages in a suitably designed amplifier chain.

6. CONCLUSIONS

The work summarised in this report clearly demonstrates the potential of the liquid laser for the development of small repetitively pumped systems and establishes the limitations of multi-gigawatt amplifiers. In the first case repetitively pulsed liquid laser oscillators can be scaled to give mean powers of $\sim$ 100 watts, for pulses of a few joules generated at rates of some tens of pulses per second. The two main engineering problems associated with this kind of operation, namely those arising from the toxicity of the liquid and the construction of a circulating pump, have already been solved. The high peak powers available with Q-switching, and the regular pulse trains which can be achieved with modulation techniques, give them an advantage over lasers with comparable mean powers in applications such as hole drilling, high speed photography and rangefinding.

Optical scattering of $\sim$ 0.3% cm^{-1}, intrinsic to the laser medium, limits the output energy density to $\sim$ 100 J cm^{-2} for pulses of tens of μs duration, when the scattering loss balances the gain by stimulated emission. This is an upper limit for long pulse systems irrespective of intensity dependent effects. In addition, there is a cumulative effect caused by the build up of scattering precipitates in the liquid in a circulating system, which can degrade the performance steadily if impurities are not removed by continuous filtering.

There appear to be two major problems in the development of liquid lasers in the 10 to 100 GW range (and beyond): (i) optical distortion and (ii) Raman conversion. The first is a linear process caused by heating and turbulence generated in the liquid by the radiation from the flashlamps, and depends directly on the energy deposited as heat in the active medium. Observations on the multi-gigawatt laser have shown that turbulence develops in the liquid over times of hundreds of microseconds. Shorter pumping pulses and better spectral filtering of the pump radiation may well achieve improvements in the beam quality approaching that for laser glass.

The second problem, Raman conversion, is an intensity dependent effect generating collinear beams of forward and backscattered radiation at the Raman shifted wavelength of 1.1 μm. It is important to realise that the large predicted conversions to Raman wavelengths, resulting in depletion of the laser light at intensities greater than 1 GW/cm^2, would only occur if the Raman light starts well above noise level. Thus there appears to be no reason why these laser intensities should not be used, provided the Raman intensity is prevented from growing by inserting wavelength selective filters in the amplifier chain. Stage lengths considerably less than 60 cm will then be required.

Both the above mentioned problems may be enormously reduced by using disc amplifiers. The close match of refractive index at the liquid-solid interface will be of considerable help in preventing parasitic oscillation. Two further non-linear optical effects, self-focusing and self-phase modulation, which could limit the high intensity performance of the liquid laser, are comparable to the same processes in glass lasers. (A comparison of the relevant parameters can be found in Tables 3 and 4.)

Having pointed out the main problems in the construction of multi-gigawatt liquid lasers, we briefly describe their definitive advantages over other $\lambda = 1\ \mu m$ systems: (i) the relatively high gain of the medium and effective energy extraction on ns timescales means that a smaller number of amplifiers can be used to reach the same power densities as glass lasers; (ii) experience with circulating the liquid in the 10 pulse per second system suggests that a 10 GW liquid laser suitable for plasma heating could be fired at a rate of several pulses per minute, giving two orders of magnitude improvement over solid laser systems; (iii) liquid lasers are completely free from damage problems and their material cost is negligible compared with glass lasers of similar volume.

In summary, the liquid laser has several advantages over comparable solid lasers, some of which have been exploited in the systems described here, while others remain to be employed in future work.

Acknowledgments

We wish to take this opportunity to pay tribute to the late Dr V I Little who always took a keen interest in this work, and to record our thanks both to him and to Dr T K Allen for initiating the gigawatt liquid laser experiment. We would also like to thank Mr S Ward and Mr K R Fenemore who were involved in the development of the 10 pps system, and particularly Dr J Katzenstein who first introduced one of us to the field of liquid laser research.

TABLE 1

Physical Properties of $Nd^{+3}:POCl_3:ZrCl_4$ Solution

Density	1.8 g/cm^3
Viscosity	5 cp at 25°C
Boiling point	105°C ($POCl_3$)
Freezing point	gels at about - 30°C
Specific heat	0.32 cal/g/°C
Concentration of Nd	0.3 molar or 1.8 x $10^{20}/cm^3$
Refractive index	1.4783 at 1.06 μm
Change of refractive index with temperature (dn/dt)	- 4.5 x 10^{-4}/°C

TABLE 2

Liquid Laser Parameters

Parameter	Symbol	Value
Wavelength	λ_o	1.0526 μm
Bandwidth	$\Delta\lambda_o$	18 nm
Cross-section for stimulated emission	σ_o	8.10^{-20} cm^2
Fluorescence lifetime	τ_o	330 μs
Quantum efficiency	η	> 0.8
Lifetime of terminal level	τ_{21}	< 5 ns
Saturation energy density	E_s	2.4 J/cm^2
Passive loss	γ	$2\text{-}6.10^{-3}$ cm^{-1}
Non-linear index	n_2	7.10^{-13} cgs (esu)
SRS gain coefficient	g_R	1.7 10^{-9} cm/W
Raman shift ($POCl_3$)	$\Delta\nu_R$	488 cm^{-1}

TABLE 3

A Comparison of Laser Amplifying Media Capable of Very High Power and Proven Performance

	MEDIUM				
	CO_2	I*	Nd:Glass (silicate)	Nd:Glass Phosphate	$Nd:POCl_3$
Centre of lasing wavelength (λ) μm	10.6	1.31	1.062	1.054-1.056	1.052
Small signal gain (α_o) (τ = 10 ns; %/cm)	5	50	6	11-13	17
Saturation energy density (E_{SAT}); ($\tau \sim$ 10 ns; J cm^{-2})	0.5	0.075	6	4-4.8	2.4
Stored energy (E_{STOR}) (J/litre)	20	$\sim$ 50	400	400	400
Extraction efficiency η (τ = 10 ns; %)	5	0.4	0.2	-	0.5
Max demonstrated energy output/beam (E_{OUT}) (Joules)	500 (10 ns) 1000 (1 ns)	1000 ($\sim$ 1 ns)	500 (1 ns)	-	48(13 ns)
Beam divergence (radians)	10^{-4}	10^{-4}	10^{-4}-10^{-3}	10^{-4}-10^{-3}	10^{-2}(column)
Repetition rate at GW powers	> 1/minute		1/hr		1/hr(static)
COST (litre)	low	low	£10,000	> £10,000	£300

TABLE 4

The main advantages of the liquid laser medium compared with glass at high peak power are summarised in the following table:

	Liquid	Glass	Comments
Stimulated emission cross-section	8×10^{-20} cm^2	$3\text{-}4 \times 10^{-20}$ 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 for pulses a few nanoseconds long. This factor also contributes to higher efficiency
Damage threshold	No damage threshold	$\sim$ (3-4) GW/cm^2	
COST	$\sim$ £300/litre	$\sim$ £10,000/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 the glass are summarised below:

Pumping	Thermally induced optical inhomogeneities	Thermal birefringence	At the present level of development 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 $n_2 = 1\text{-}1.5 \times 10^{-13}$ 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/α_o, where α_o = gain coefficient of the medium (Suydam 1975). Thus the non-linear parameters for beam instability are in the ratio 3:1 for liquid:glass (phosphate).

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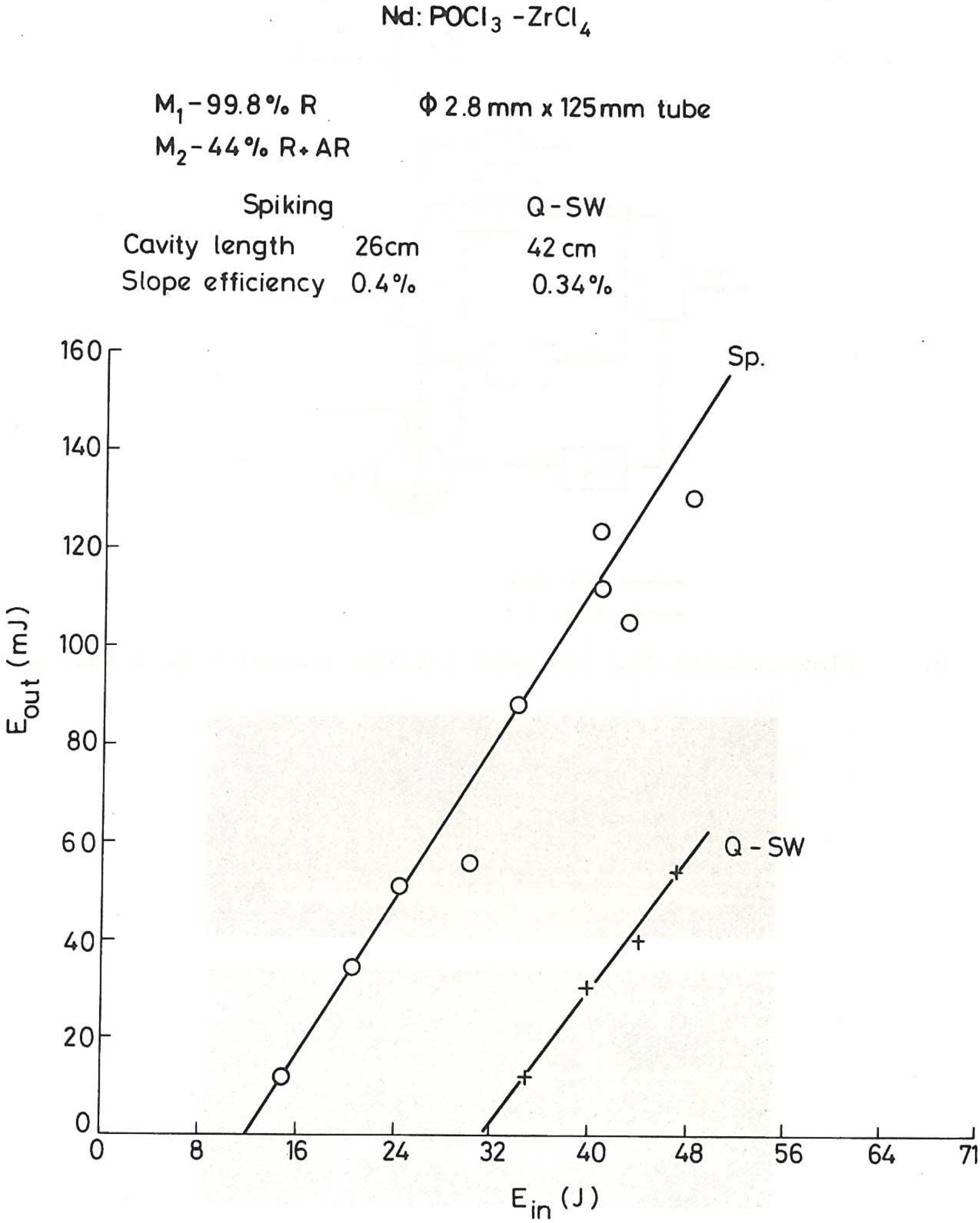
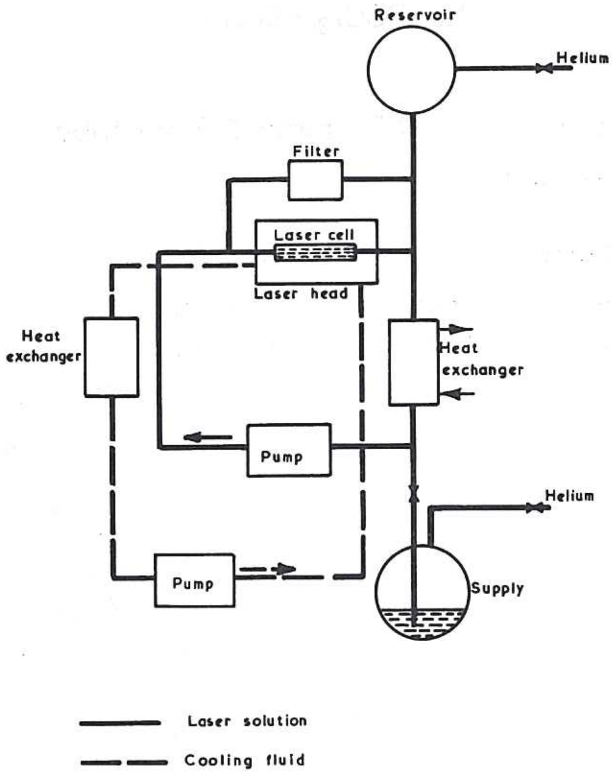
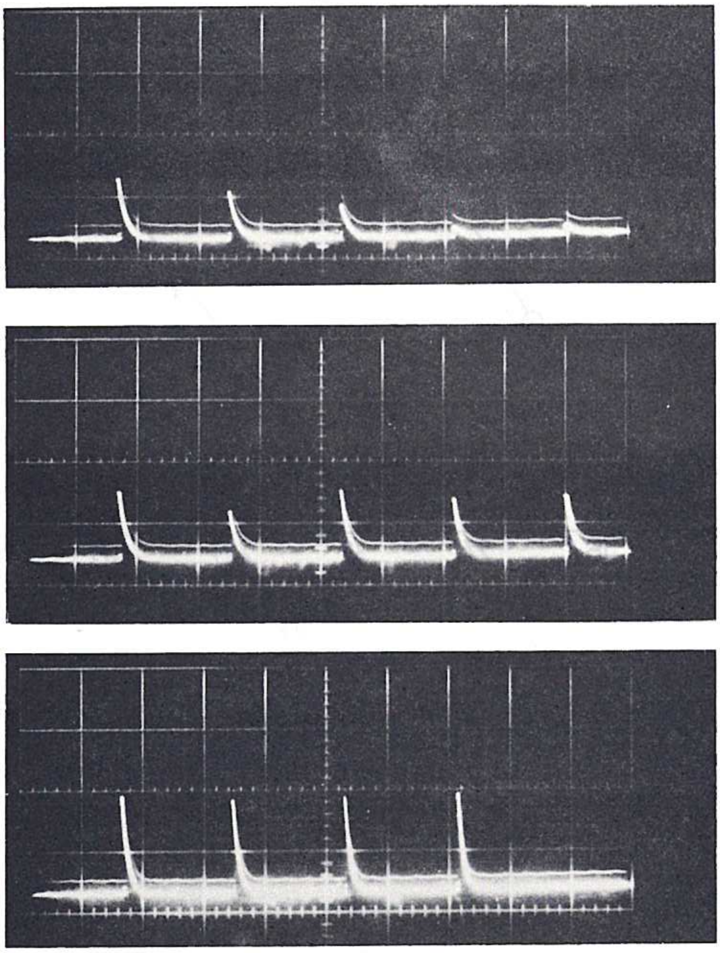


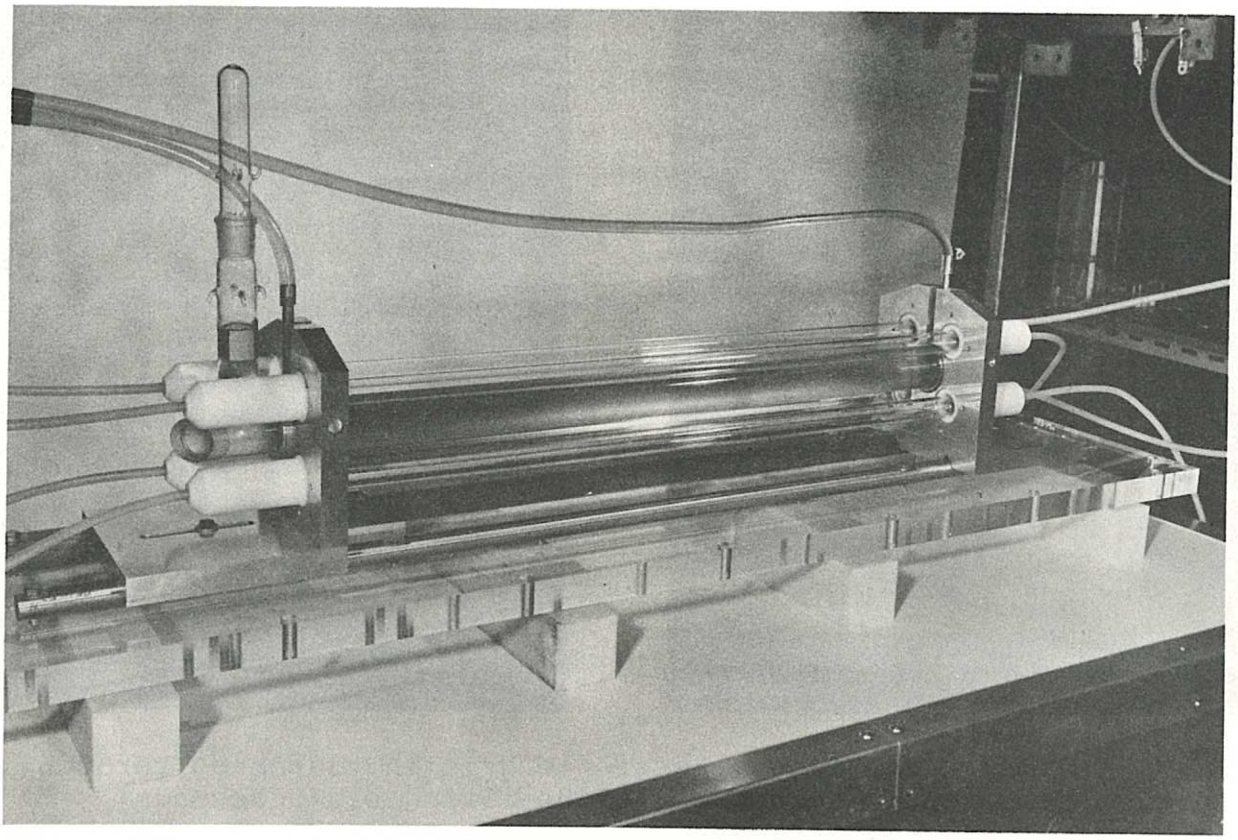
1. Performance of a small liquid laser, showing output vs. input for uncontrolled (spiking) and controlled (electro-optically Q-switched) operation.



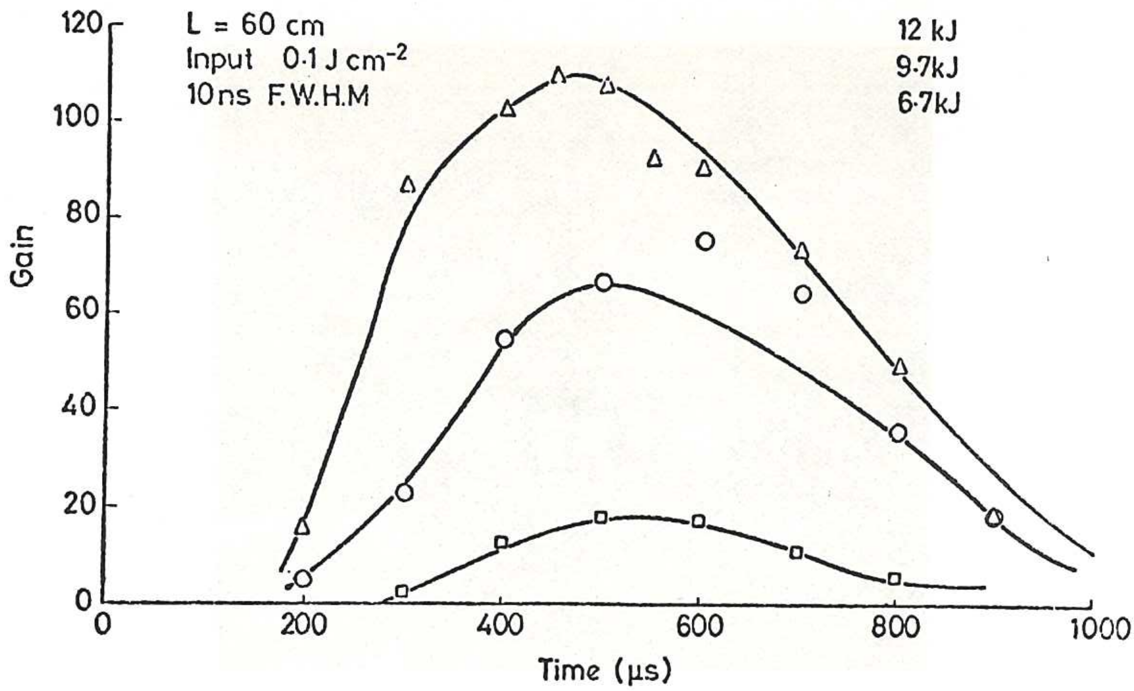
2. Flow diagram for the repetitively pulsed liquid laser.



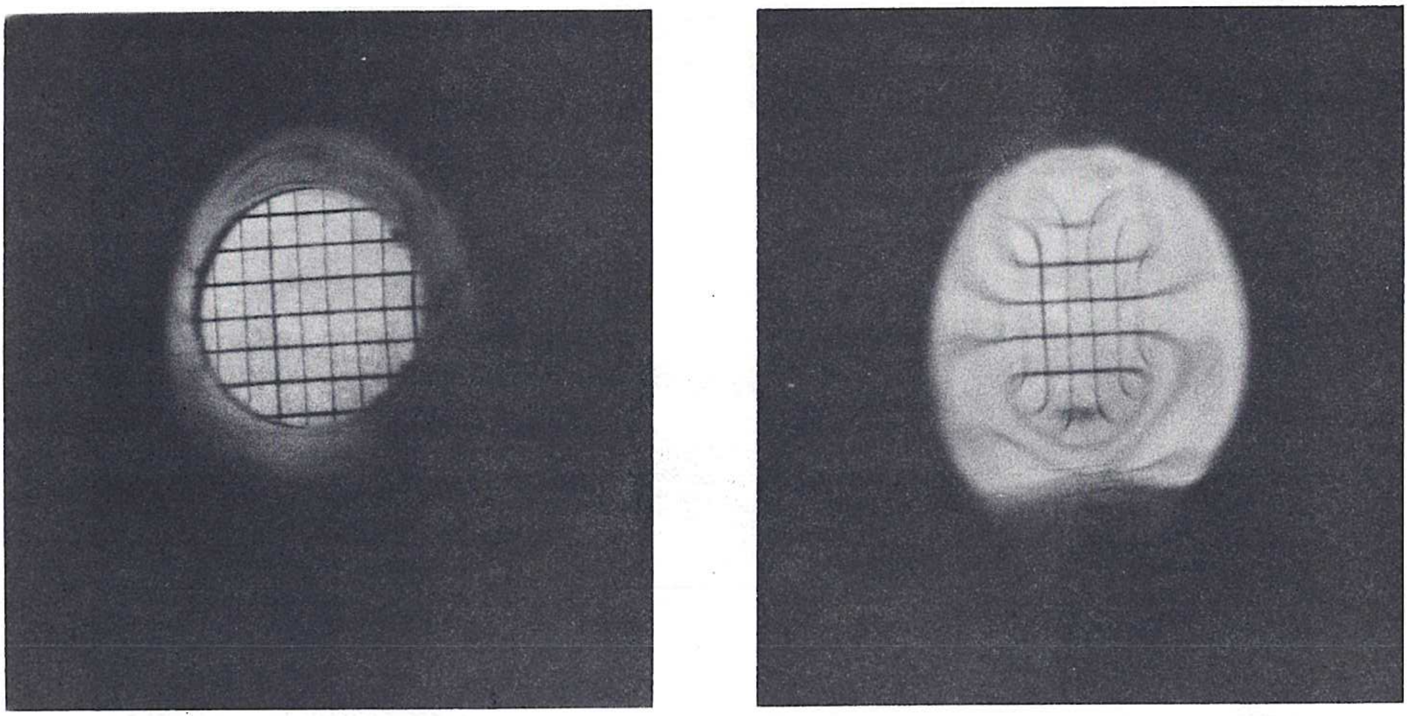
3. Q-switched laser operating at 10 pulses per second, showing improving pulse stability with increasing flow rate.



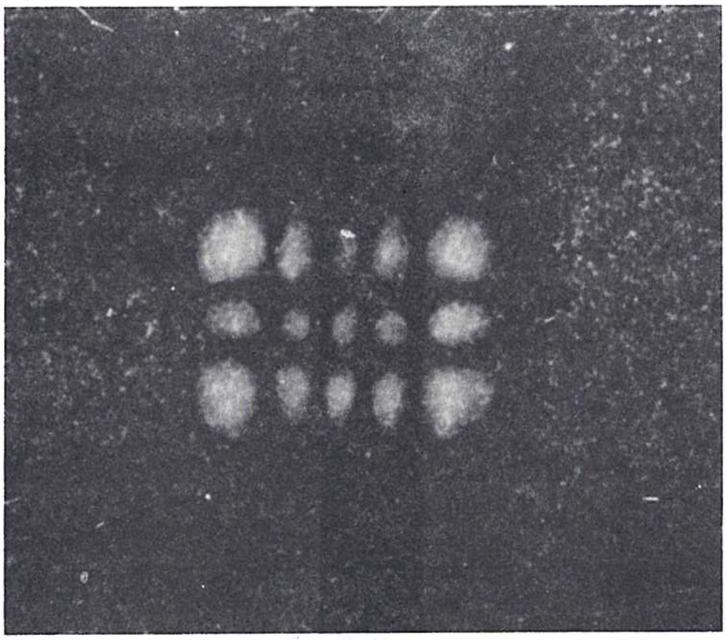
4. Gigawatt amplifier assembly with the reflector removed to show the laser cell surrounded by its four flashlamps.



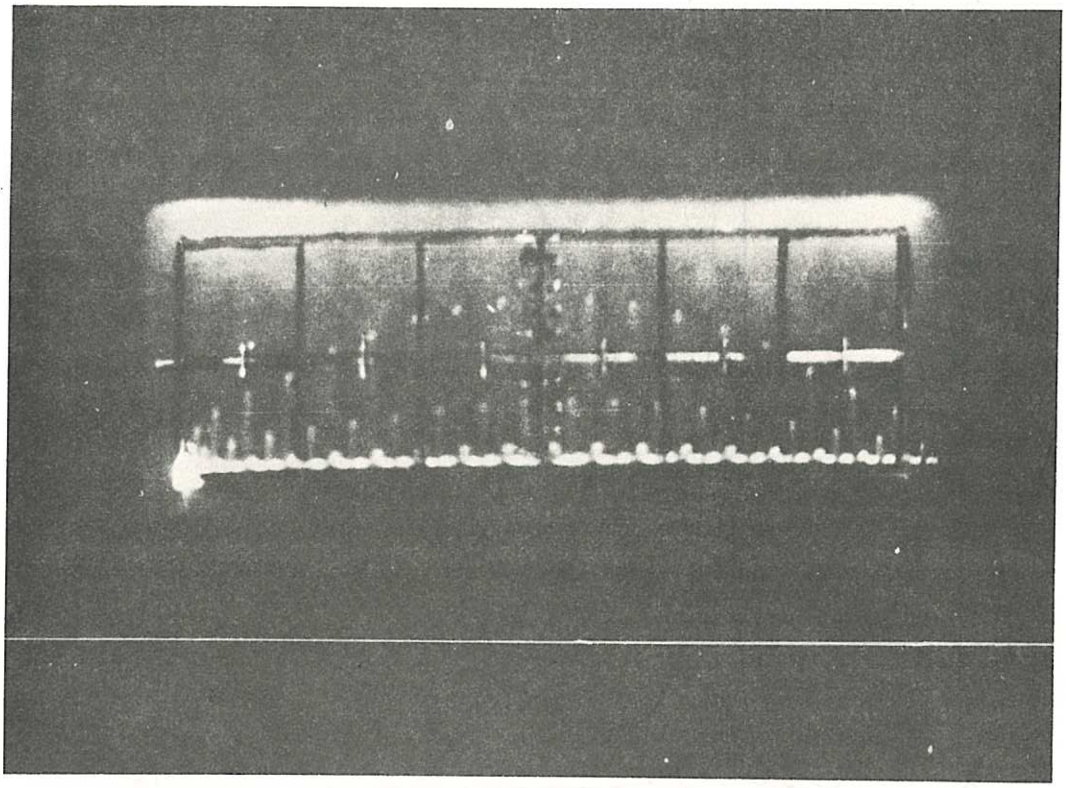
5. Peak power gain for 10ns pulse vs. time during the discharge. Curves show results for three different pumping energies.

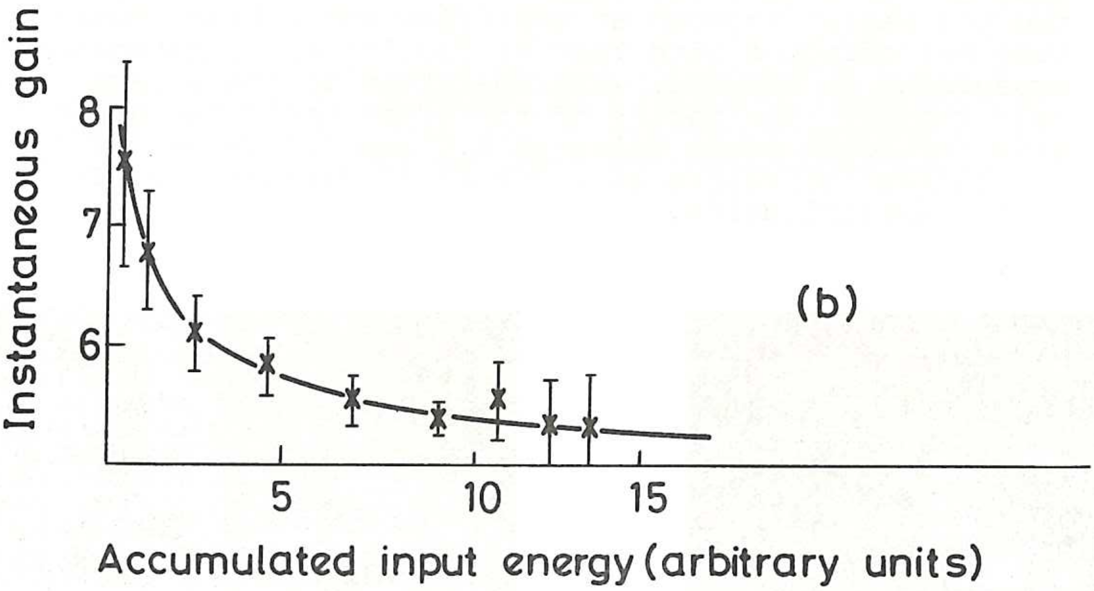


6. Thermal distortion. Reference grid photographed through 60 cm liquid column a) before firing b) 10 seconds after 12kJ discharge.

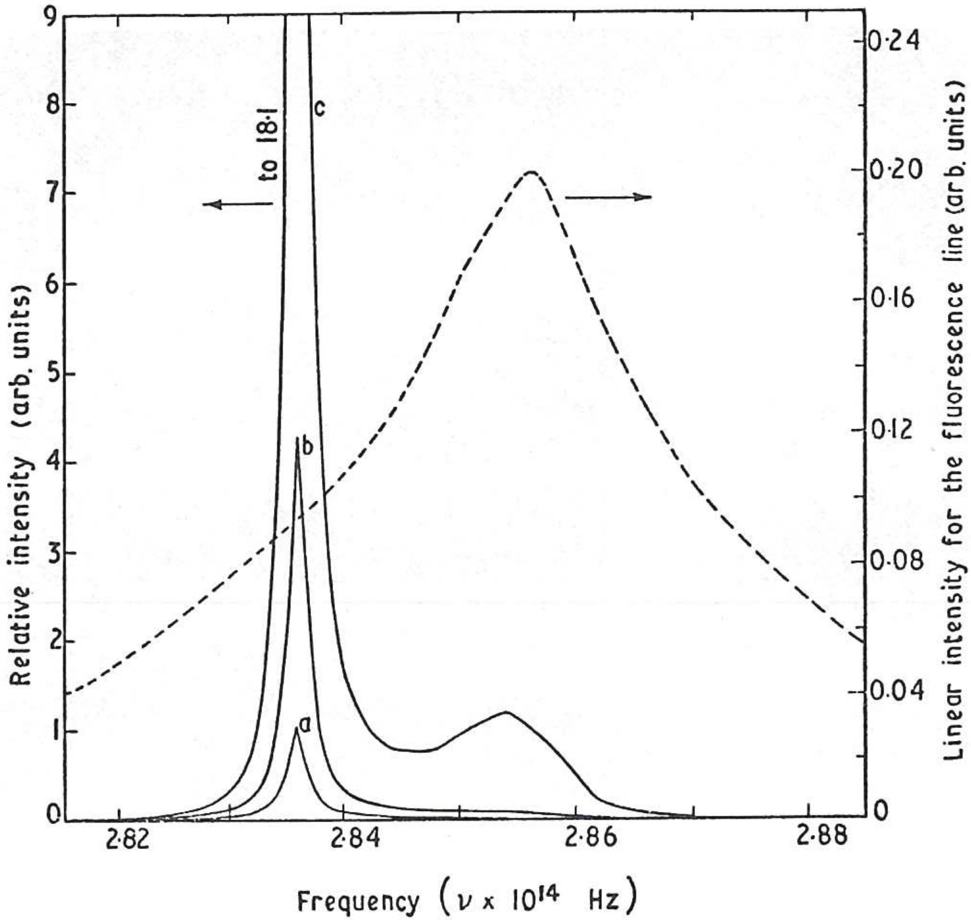


7. Transverse mode output (TEM_{42}) of a small mode-locked liquid laser.

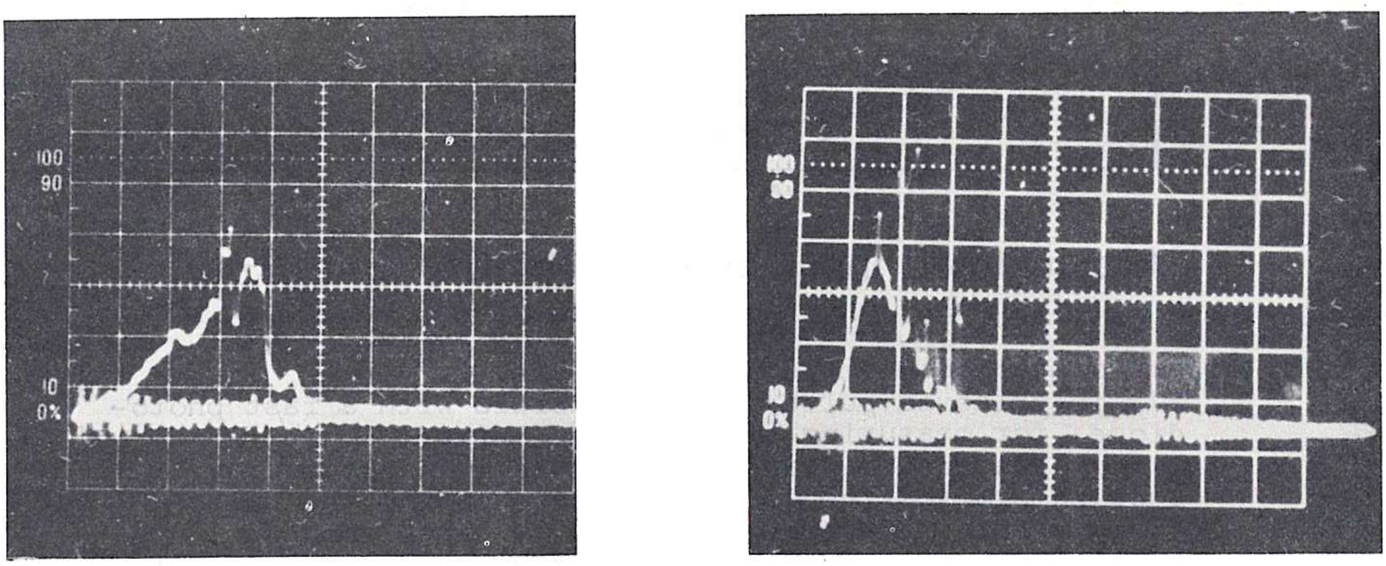




8. Amplification of mode-locked pulses.
a) interleaved pulse trains recorded with a fast photodiode, for measuring single pulse gains.
b) instantaneous gain vs. accumulated input energy, showing progressive gain saturation during the pulse train. Pulse energies in mJ can be calculated on multiplying the ordinate scale by 8 (to an accuracy of 20%).



9. Spectrum of a Gaussian signal (a) after one (b) and two (c) passes through an amplifier whose line centre does not coincide with that of the input. Inhomogeneous broadening is assumed, with operation in the unsaturated gain regime. The ratios of the areas under the graphs give the total power gains as 4.4 and 5.1 respectively, the latter increasing as a result of frequency shift during amplification.



10. Intensity structure resulting from stimulated Raman scattering in the gigawatt amplifier; a) amplified laser pulse b) back-scattered Stokes' pulse (λ 1.1 μm).

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