

Amplified spontaneous emission from nitrogen laser pumped dye lasers

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Abstract. The nitrogen laser pumped dye laser output has been studied with emphasis on the behaviour of amplified spontaneous emission (ASE) with respect to the tuning wavelength, pump energy, dyes and their concentration and solvents. Spectral spread of ASE is narrower than fluorescence and its maximum is shifted towards the red side. However, lasing occurs beyond the ASE region. ASE is small at high gain wavelength and increases at the edges of the gain curve. Laser energy is highest at the ASE peak with minimum ASE present in the output. ASE is reduced with increased laser energy in the energy transfer dye lasers.

Keywords. Amplified spontaneous emission; grazing incidence; gain curve; laser pumped dye.

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1. Introduction

Pulsed laser pumped dye lasers have become convenient tunable lasers in many scientific and technical applications. The single pass gain in such systems is variable and can be as high as 10^7 resulting in amplified spontaneous emission (ASE) which appears as a background to the laser output and is a source of noise. The dependence of ASE on cavity parameters, intensity and pulse width of the pump laser, the concentration of the dye and the solvent was studied (Ganiel *et al* 1975; Duarte and Piper 1980, 1981; Bor 1981; McKee *et al* 1982). It was reported (Nair and Dasgupta 1985; Littman and Metcalf 1978) that the grazing incidence grating (GIG) cavity suffers considerably from ASE and that prism pre-expansion improves the situation (Duarte and Piper 1981). A comparative study (Bor 1981; McIntyre and Dunn 1984, 1985) for different cavities and detailed analysis was reported by Nair and Dasgupta (1985).

Since the GIG cavity is simple and highly efficient, we have examined ASE in this case and report its dependence on the concentration, solvent, pump energy and the dye for the entire tuning range. We also examine its behaviour in energy transfer dye lasers (ETDL), an aspect which has not received much attention.

2. Experimental

The GIG configuration of Littman and Metcalf (1978) (see figure 1) with wavelength tuning by mirror M_2 and a grazing incidence angle of 85° at the holographic grating (2100 l/mm) was used. A stainless steel cell with quartz windows containing the dye solution was transversely pumped by a home-built N_2 laser (1.5 mJ, 5 ns) operated at

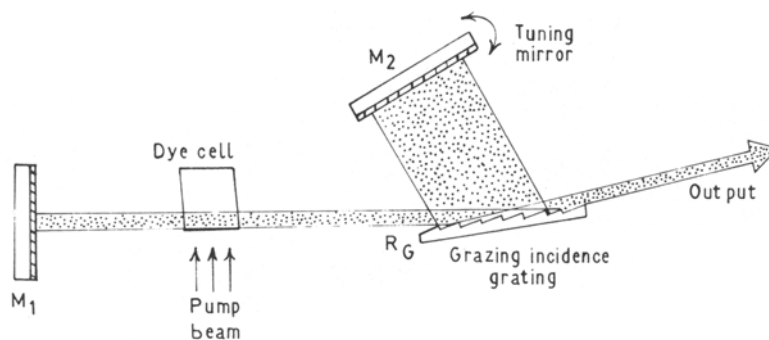


Figure 1. Schematic diagram of the dye laser cavity.

70 mbar and a pressurized spark gap triggered at 10 Hz. Dye laser wavelengths were read with a monochromator (Jobin-Yvon HRP 1/2 M) coupled to a detection system consisting of a detector (Vidicon) and an optical multichannel analyser (OMA-2 EG & G). ASE and laser output at various lasing wavelengths were stored on the OMA diskettes and later recorded using an X-Y recorder (Hewlett-Packard). Dye laser output energy was measured with a laser energy meter (RJP-735, Laser Precision Corporation).

Laser dyes [(Lambda Physik) Coumarin 1 (C.1), Coumarin 2 (C.2), Coumarin 102 (C.102), Coumarin 307 (C.307) and Rhodamine 110 (Rh.110)] and analar grade solvents (ethanol, methanol, isopropyl alcohol) were used to prepare samples with concentrations ranging from 0.5 mM to 10 mM.

3. Results and discussion

The dye laser output contains two components: (1) ASE spread over a broad spectral range and (2) laser radiation (LR). Since ASE and LR energies vary with lasing

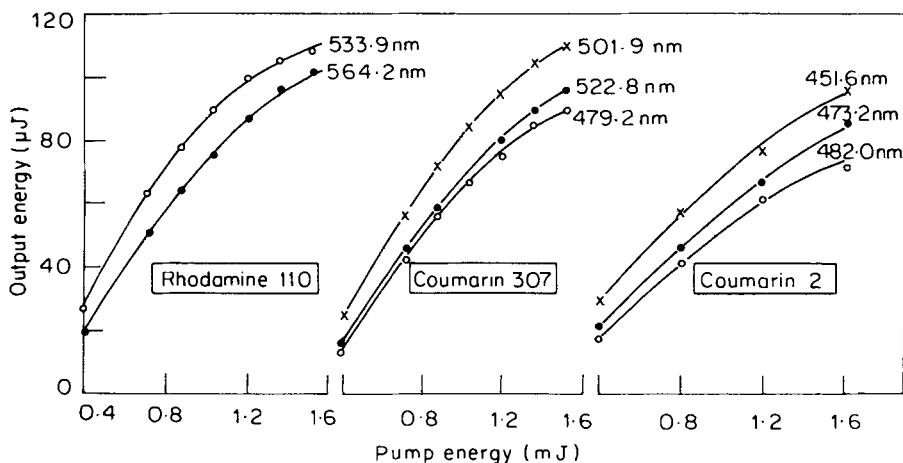


Figure 2. Output energy vs pump energy.

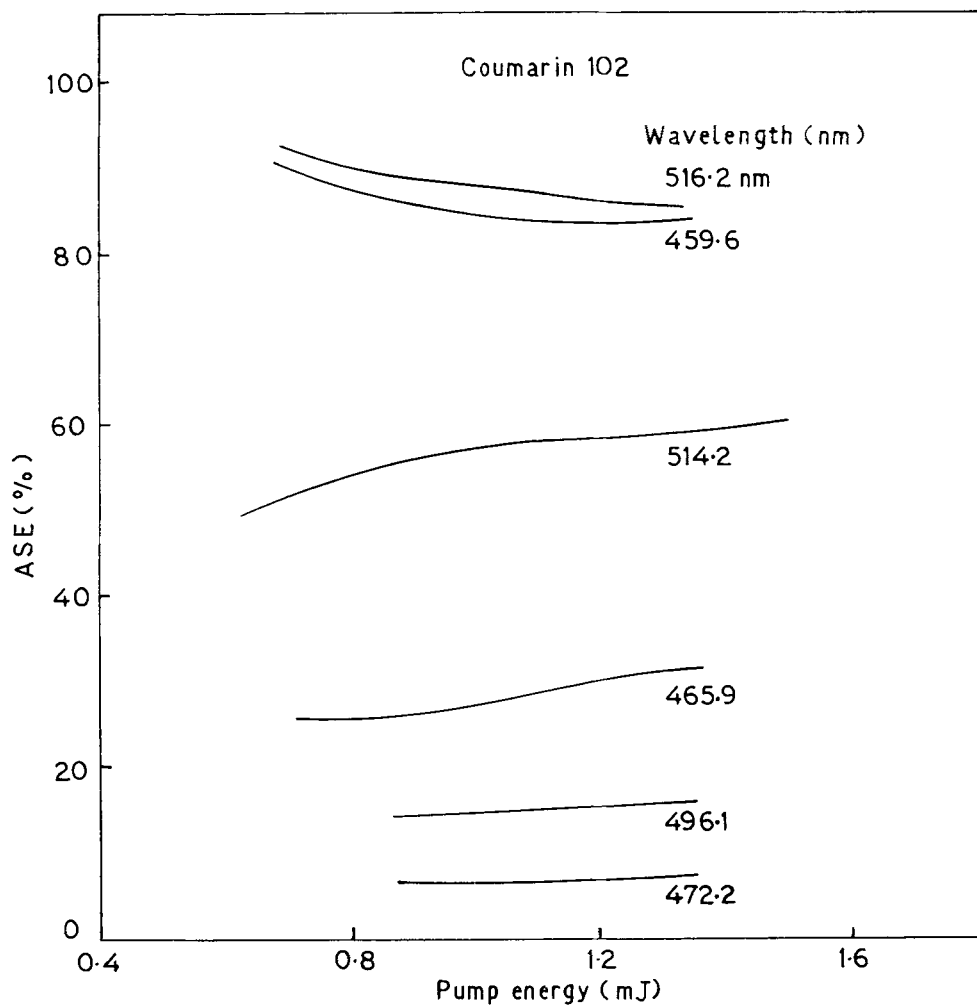


Figure 3. % ASE vs pump energy.

Table 1. Spectral range and λ at peak (λ_p) for fluorescence, ASE and laser for dyes and mixture of dyes.

Dye/mixture	Fluorescence		ASE		Laser	
	Spectral range (nm)	λ peak (nm)	Spectral range (nm)	λ peak (nm)	Spectral range (nm)	λ peak (nm)
C.2	400–520	440	435–465	450	430–478	441
C.102	420–580	472	465–495	485	458–519	487
C.307	430–600	495	490–530	510	479–538	516
Rh.110	500–600	525	540–560	550	537–569	549
C.307 + Rh.110	500–590	522	535–553	546	528–566	550
C.1 + Rh.110	—	—	536–550	543	536–562	546
C.102 + Rh.110	—	—	530–547	535	532–561	544

wavelength, concentration of the dye, solvent and pump energy, these were measured over the tuning range for all these parameters. The spectra of the laser and ASE stored in the OMA diskettes were used to derive the relative (integrated) intensities of the laser and the ASE. The total energy (laser + ASE) was measured by an energy meter. The values were then used to derive absolute values of laser energy from the ratio of relative intensities of laser and ASE as determined from the OMA data. These results are shown in figure 2 for three representative lasing wavelengths and three dyes. It is seen that in all the dyes LR first increases with pump energy at different tuning wavelengths reaching a saturation at high pump energy when the pump energy was changed from

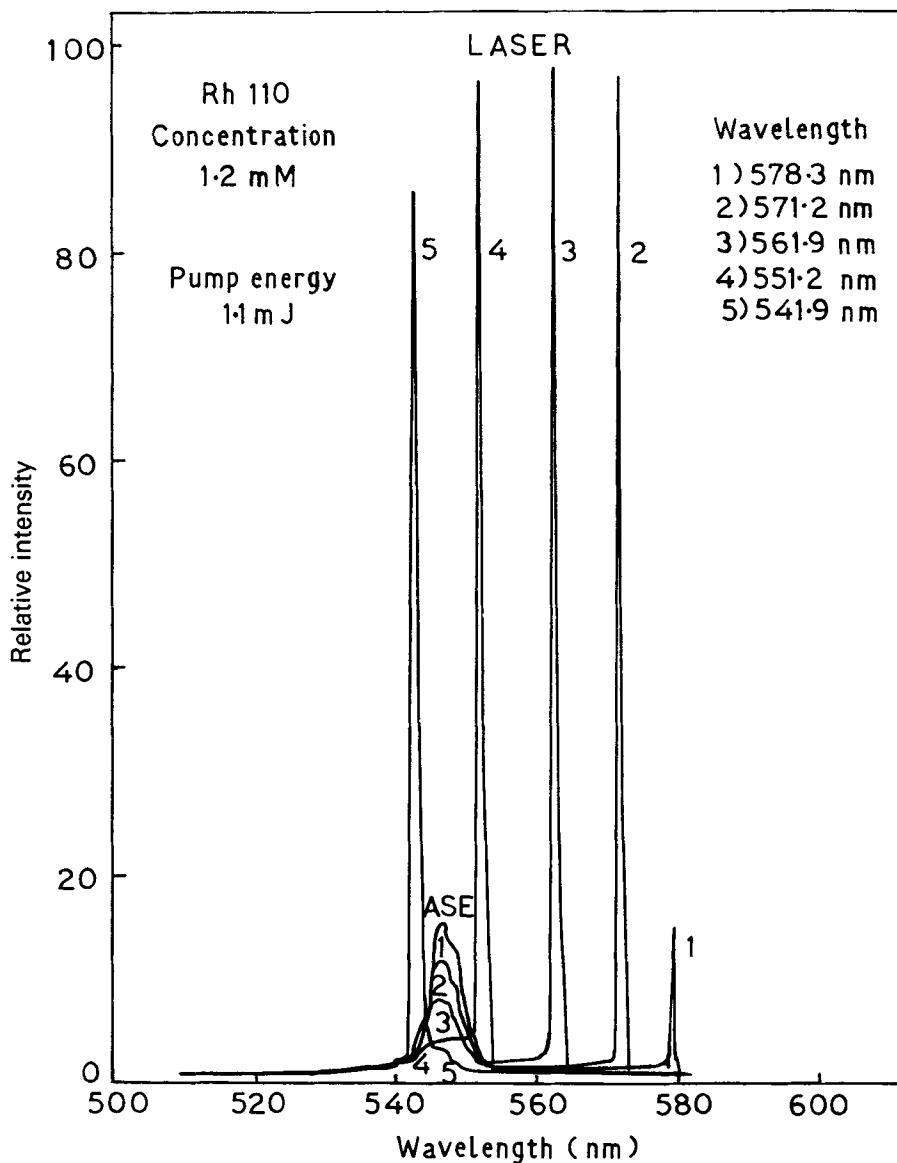


Figure 4. Spectra of laser and ASE at different lasing wavelengths.

0.4 mJ to 1.6 mJ. The energy increase was faster at a low pump energy. Figure 3 describes the typical behaviour of ASE with pump energy at different wavelengths. LR and ASE energies increase with pump energy keeping their ratio (i.e., LR/ASE) almost constant at any given wavelength. However, the proportion of ASE (say % ASE) was different at different tuning wavelengths. For example, % ASE was small at high gain wavelengths (496.1, 472.2 nm) but large at low gain wavelengths (516.2, 459.6 nm). Similar observations were reported by Nair and Dasgupta (1985).

ASE alone (with tuning mirror blocked) was measured and compared with fluorescence. Table 1 shows that ASE is much narrower in spectral range compared to fluorescence and the ASE peak was shifted (10–25 nm) on the longer wavelength side compared to the fluorescence peak. Since ASE represents essentially a single pass gain it can be considered as the gain curve for a single pass high gain laser and, therefore, it is narrower than the fluorescence spectrum. Generally ASE was observed at a high concentration (5 mM) and hence the shift in peak towards red could be a concentration effect. Lasing was observed beyond the ASE region but was within the fluorescence profile, indicating that the lasing threshold was reached even outside the ASE region because of the resonant cavity.

As tuning was varied from one end of the spectrum to the other LR increased

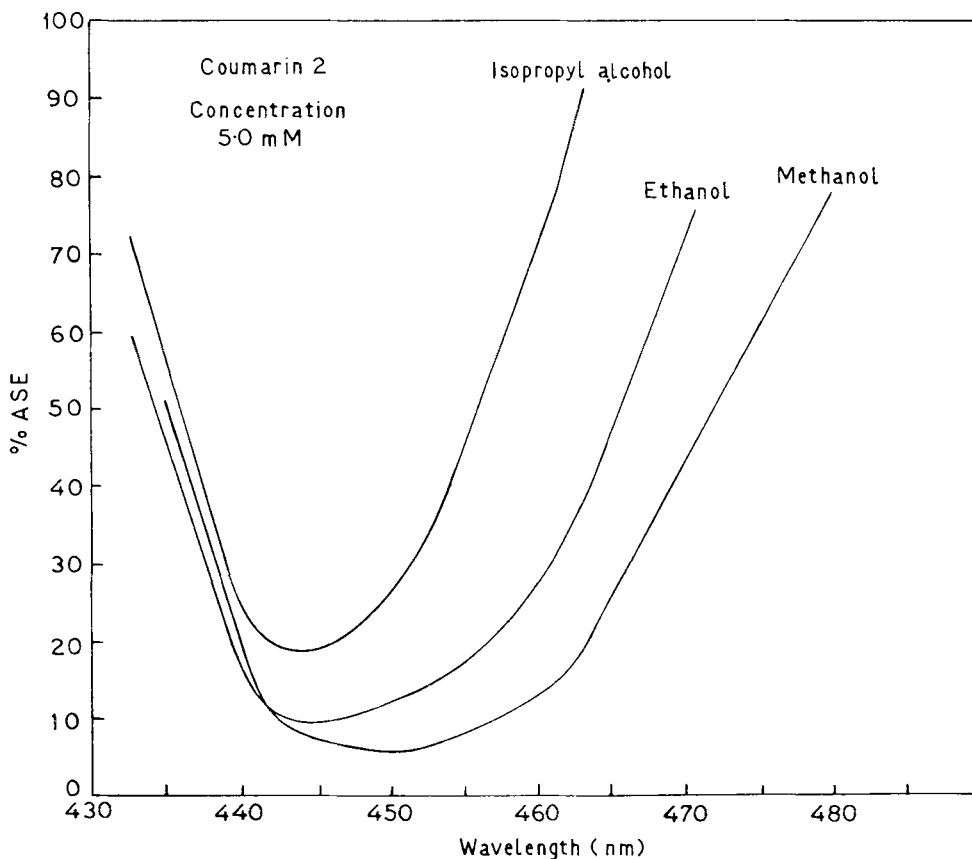


Figure 5. % ASE in output energy vs lasing wavelength.

reaching a maximum at the maximum of the ASE spectrum. At this point the % ASE showed a minimum (see figure 4) and LR decreased as the wavelength was further changed and correspondingly the ASE component increased, being the highest at both ends.

The position and general shape of the ASE remained mostly unaltered but there was a change in % ASE at different tuning wavelengths (figure 4). This is a convenient way to separate LR from ASE by using a monochromator at the output end. At the maximum LR, though the best condition for LR output, ASE cannot be separated since LR peaks on the ASE peak.

It was found that both ASE and lasing properties depended on solvents and dye concentration. The percentage of ASE associated in the laser output at different lasing wavelengths over the tuning range in different solvents (under optimum conditions of concentration) is shown in figure 5. The % ASE was smaller in methanol as compared to ethanol and isopropyl alcohol for the same concentration. The lowest value of ASE was in methanol at lower concentration (0.5 mM) at which laser action was not observed in other solvents. At a low concentration the lasing wavelength shifted towards blue, the threshold energy increased, the output energy slightly

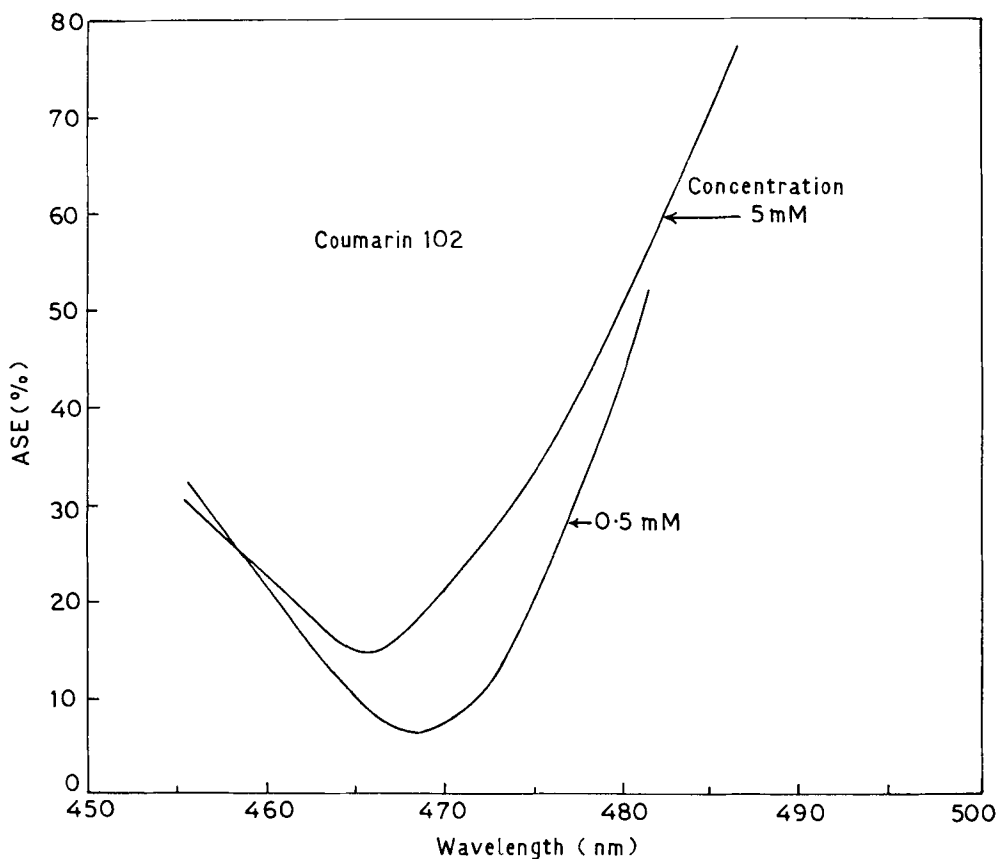


Figure 6. % ASE dependence on concentration.

decreased and the energy of the laser obtained for different lasing wavelengths close to peak energy was almost the same as the total energy (LR + ASE). At high concentrations, say, 10 mM, % ASE was high at the edges of the tuning range. The variation of % ASE with tuning wavelength is shown in figure 6 for two typical concentrations. The behaviour is similar in C.102 and C.2, the other two dyes examined.

For dye mixtures (C.1, C.102 and C.307 as donors and Rh.110 as an acceptor) operating as ETDs, the output energy increased with increase in donor concentration reaching a maximum for a certain donor concentration. A further increase in donor concentration decreased the output energy due to concentration quenching of the donor. For a given acceptor concentration there was an optimum donor concentration

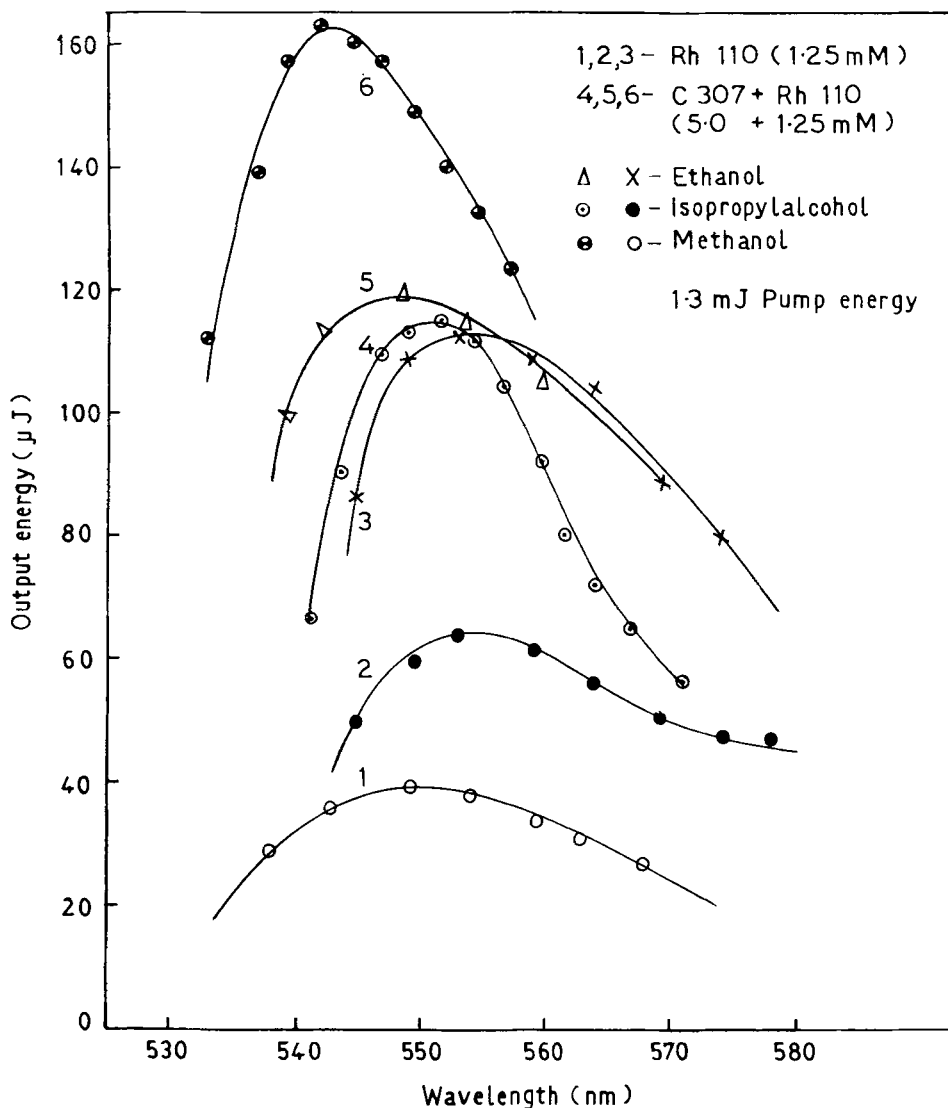


Figure 7. Output energy vs lasing wavelength.

for the highest output energy. The increase in donor concentration shifted both the laser emission peak and the upper lasing limit towards the violet side although it was small (9 nm) compared to the shift observed in peak emission wavelength (16 nm) towards red with increase in acceptor concentration in the case of C.307 + Rh.110. The output energy increased faster at a lower concentration of acceptor and approached saturation at a higher concentration for a fixed donor. In the dye mixture energy saturation was reached at a lower pump energy value compared to lasing in the single dye.

In the dye mixture acceptor alone lased at all concentrations of the donor and no lasing was observed at two wavelengths, one corresponding to the donor and the other to acceptor though there are some reports (Berlman *et al* 1973) of such behaviour at medium concentration of donor and acceptor. In dye mixture the component dyes remained intact and no complex was formed either by chemical or photochemical reaction as evidenced by our study of spectroscopic measurements of absorption and fluorescence spectrum of dyes before mixing and after mixing and also after pumping.

In solvents the behaviour of ASE and LR did not show a pattern and dye-solvent interaction was not amenable to any generalization. Figure 7 shows the output energy

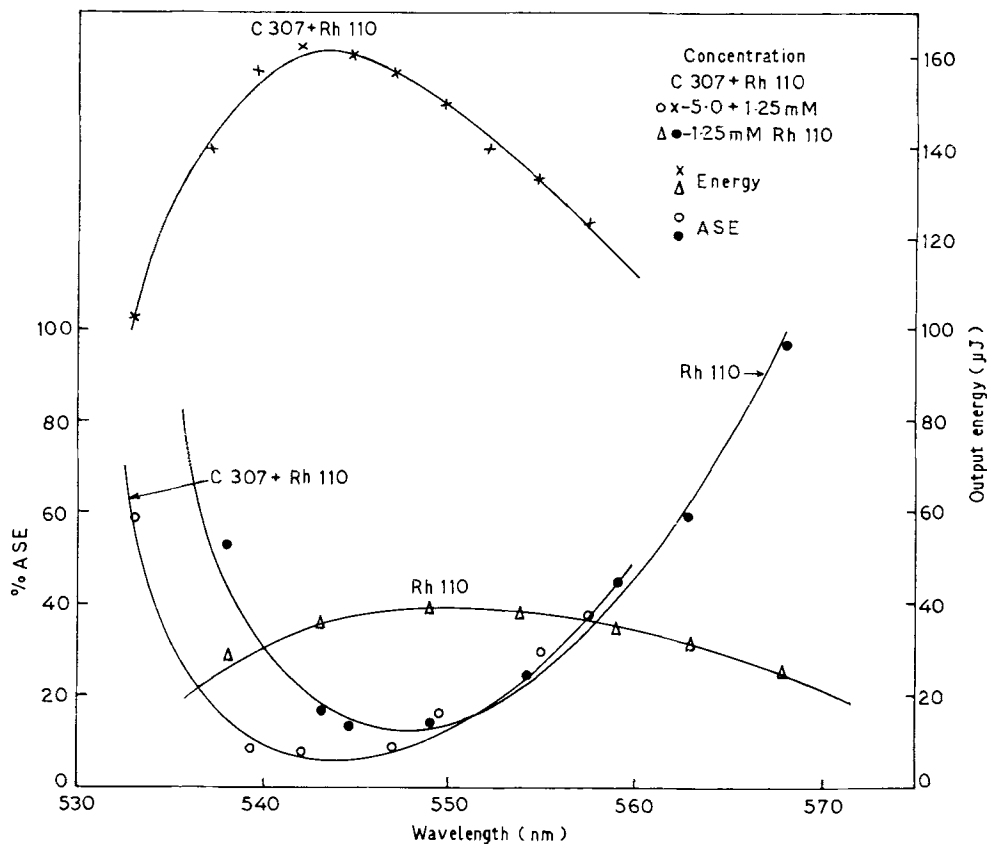


Figure 8 Comparison of % ASE and output energy as a function of lasing wavelength.

versus tuning wavelength for a dye mixture and a single dye at one pump energy in different solvents. The output energy and the tuning wavelength were different in different solvents. Not only did the energy maximum change but the energy distribution changed with the solvents. It was also seen that output energy of laser peaked at different wavelengths in different solvents. Among the solvents used methanol was more suitable because of its higher conversion efficiency (Schäfer 1977). When the mixture was tuned for lasing the tuning range was wider (ETDL: 38 nm, Rh.110: 32 nm) and the energy of the laser was enhanced by a factor of 4 to 5 (figure 8) for donor C.307 with a conversion efficiency of about 16% whereas for other donors the energy enhancement was only by a factor of 2.5 and 1.5 for C.102 and C.1

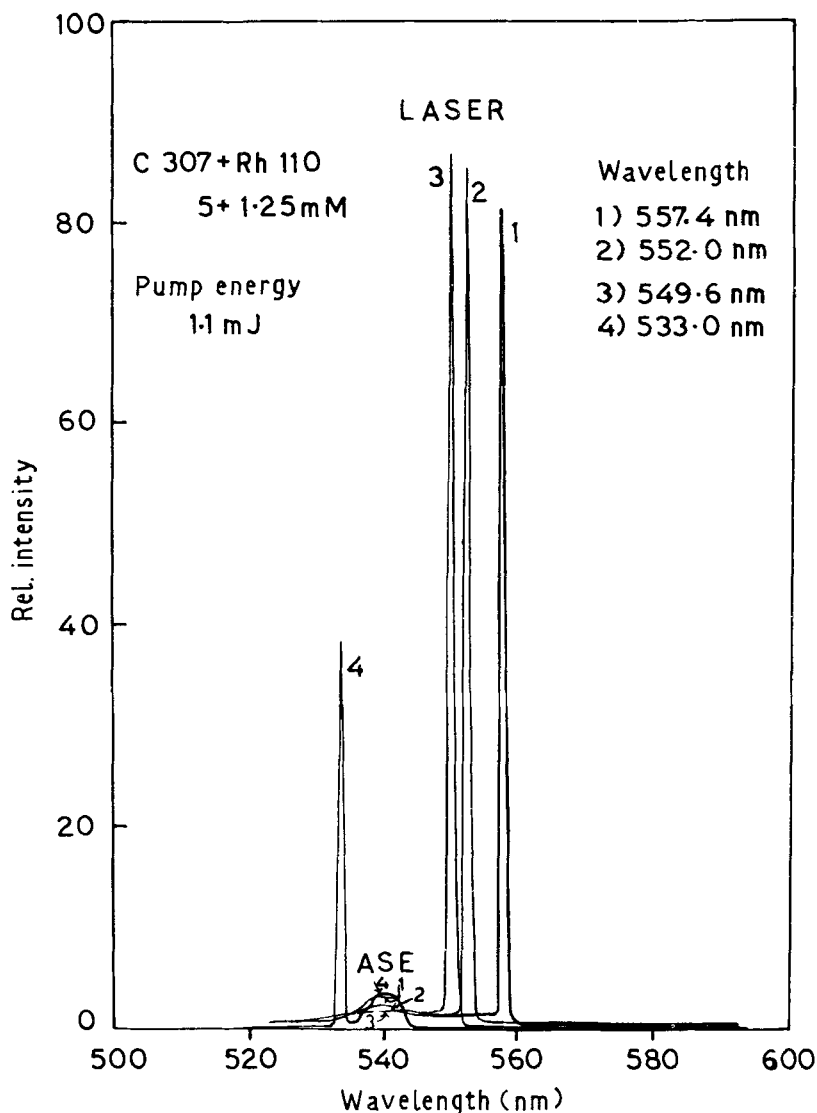


Figure 9. Spectra of laser and ASE in dye mixture at different lasing wavelengths.

respectively, even though for single dye Rh.110 the solvents ethanol and isopropyl alcohol were better than methanol. The solvent which suits a donor results in better performance with dye mixture, or in other words, a dye in which the solvent has higher absorption results in higher energy transfer leading to higher conversion efficiency in laser pumped dye laser.

ASE associated with LR was lower in dye mixture compared to single dye as can be seen from figures 4 and 9. The variation of % ASE as a function of lasing wavelength for dye mixture as well as for single dye both under optimum conditions of concentration is shown in figure 8.

The increase in LR was due to the suppression of ASE and also the supportive action of the donor. There was reduction at the threshold for lasing and further it lased at lower concentration compared to acceptor alone. It is a simple and promising method of reducing ASE, enhancing LR and expanding the tuning range.

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