INCREASED GAIN THROUGH IDENTIFICATION AND ALLEVIATION
OF DYE SELF ABSORPTION IN LASER PUMPED DYE LASERS

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The effect of overlapping absorption and emission spectra is to shift the gain curve of a dye to the red side of its fluorescence curve and reduce its gain. It is shown how simple binary mixtures of "adjacent" dyes can be used to produce a new gain maximum in the regions between the gain maximum of the single dyes.

Practically everyone who has used a dye laser has thought of improving some aspect of its performance by mixing two or more dyes together. Such mixing experiments seem to fall into several categories [1]:

a) those for allowing lasing over a broader band with a single solution,

b) those for broadening the absorption band of the solution to better match the pump energy distribution,

c) in the case of laser pumped dyes, those for introducing a good absorber of the pump wavelength which can then transfer the energy to the lasing dye (the so-called energy transfer laser).

The result of such experiments, with some notable exceptions [2], has been less than spectacular as witnessed by the fact that dye laser manufacturers tend to give lists of pure dyes together with their tuning curves in a very matter of fact way. The impression given is that it is the best you can do since no further comment or suggestion is made. It is the intention of this note to show a simple and general technique for improving the available output of laser pumped dye lasers in the regime between the gain maximum of pure dye solutions.

Fig. 1 shows the absorption and emission curves for a typical laser dye (Rhodamine G6 in ethanol). The overlap of the absorption and emission curves and the resulting "self-absorption" has long been recognised.

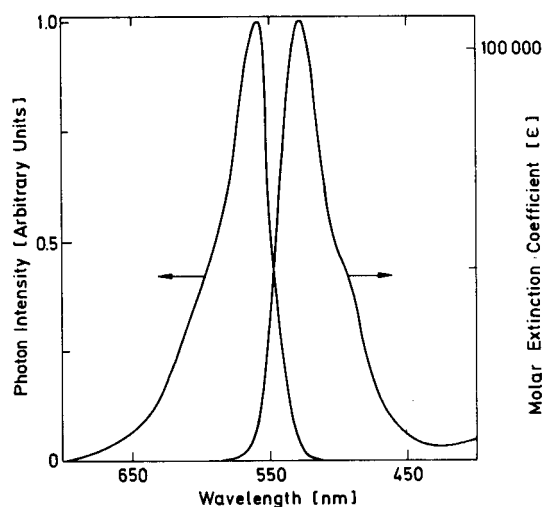* Permanent address: National Bureau of Standards, Time and Frequency Division, Boulder, Colorado 80303, USA.

Fig. 1. Absorption and emission curves for Rhodamine 6G in ethanol. The overlapped mirror image curves are common to most laser dyes.

In fact the red shift in gain maximum with increasing dye concentration produced by this process is often used to help identify the lasing species in energy transfer dye lasers [3]. However, its generally deleterious effects and alleviation by simple mixtures of dyes do not seem to be discussed in the literature.

The effect of self-absorption becomes a problem in laser pumped dye lasers that use a side pumped geometry. Such a geometry necessitates a dye concentration
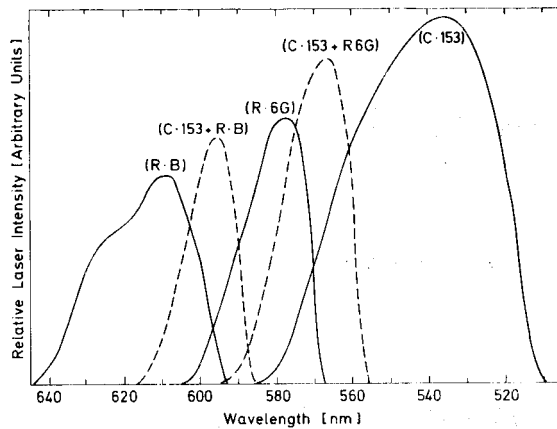
Fig. 2. Laser output versus tuned frequency for several pure dye solutions (solid curves) and for some binary mixtures of dyes (dashed curves). In the case of the mixed dyes, the more blue dye is at its normal concentration while the red (lasing) dye is 1–2 orders of magnitude less than normal. Dyes: C 153 = Coumarine 153, R 6G = Rhodamine 6G and R B = Rhodamine B.

great enough to absorb the pump energy in the typically 0.15 mm diameter gain region while at the same time requiring the stimulated emission to traverse a 1 cm or greater path in the dye. The result is that the absorption coefficient at the lasing wavelength is multiplied by a factor of 100 relative to that at the pump wavelength. This problem is further complicated by the fact that many of our best dyes do not have good absorption at the wavelengths of our best pump lasers.

The solution to this problem is suggested in a slightly modified donor-acceptor arrangement, where, in this case, the donor itself need not have a better absorption coefficient at the pump wavelength but only a lower one at the desired lasing wavelength. With the responsibility for stopping the pump energy now transfered to a different dye, the lasing dye can be used in very much lower concentration ($10^{-1} - 10^{-2}$) and still provide efficient lasing action.

The real benefit here is that the lasing action, which was restricted to the red wing of the fluorescent curve

by self-absorption, is now allowed to shift to the blue and utilize the higher gain regions of the curve. Fig. 2 shows two examples of such dye mixtures in a N_2 laser pumped dye laser of the Hänsch configuration [4]. The solid curves are for the pure dyes (Coumarine 153, Rhodamine 6G and Rhodamine B), each operated at its optimized concentration. The dashed curves were obtained by starting with the normal Coumarine 153 solution and adding a few per cent of the appropriate Rhodamine. The new lasing peaks can be continuously adjusted between the peaks of the pure dyes by adjusting the amount of Rhodamine added. This example shows the results both when using adjacent dyes (C 153 and R 6G) and with non-adjacent dyes (C 153 + RB) where none of the dyes used are particularly good absorbers at the pump frequency. Similar results have been obtained with dyes which are good absorbers of the pump wavelength (Coumarine 129 in DPS).

We believe the results of this type of dye mixing are generally applicable; whenever one needs more power on the blue side of any dye tuning curve, one just has to start with a slightly bluer dye and add a small amount of the desired lasing dye until a new peak is created where it is needed. Such results are going to be particularly valuable to the growing use of nonlinear phenomena where the observed signal is proportional to the laser power raised to the nth power, where n is 2 or greater. In the examples cited here the improvement in laser power at the cross-over point between the pure dyes is 300%.

References

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