Optica Applicata, Vol. XIV, No. 4, 1984

Energy transfer dye laser

TOMASZ KOTOWSKI, WOJCIECH MAJEWSKI, WOJCIECH SKUBISZAK

Institute of Experimental Physics, Warsaw University, ul. Hoża 69, 00-681 Warszawa, Poland.

It is commonly known that in most cases mixing of laser dyes spoils laser action. For example, a mixture of two very efficient dyes, Rhodamine 6G and Stilbene 3 (pumped by our 100 kW nitrogen laser), does not lase at all. Only very few mixtures of dyes give an advantage of multicolour output [1], shifting the tuning range or improving the output power. The last two phenomena are based on an excitation energy transfer from a directly pumped donor to acceptor which becomes an indirectly pumped laser medium. Such a system is called the energy transfer dye laser (ETDL).

At least four types of interactions between the donor and acceptor dye are possible. They are described in the famous Förster article [2]. Two of these transfer mechanisms, i.e., reabsorption and long range dipole-dipole interaction are treated seriously in previous papers [3, 4], but most authors suggest the predominant role of the dipole-dipole transfer (known as the Förster type) [5-9].

We have found several donor-acceptor laser dye pairs: 2,7-dichlorofluorescein (DCF) \rightarrow Sulforhodamine B, DCF \rightarrow cresyl violet, Stilbene 3 \rightarrow Coumarine 30, Stilbene 3 \rightarrow Coumarine 102. Their laser properties have been investigated both in longitudinal and transversal arrangements. The pump wavelength was chosen to fit absorption peak of the donor only, to avoid a direct excitation of the acceptor. In our experimental conditions only a transverse pumping gave significant booster effect. For example, 2,7-dichlorofluoroscein with Sulforhodamine B pumped by 460-nm beam lased efficiently, whereas Sulforhodamine B alone did not lase at all.

The extensive measurements for donor-acceptor mixture of Stilbene 3 (S-3) and Coumarine 102 (C-102) were made. Nitrogen laser (home made, $\lambda = 337$ nm, 5.5 ns FWHM time, 100 kW, up to 400 Hz repetition rate) serves as a convenient pump of the dye laser oscillator in the transverse arrangement with a single rutile prism [10] as a tuning element. Although C-102 alone can be pumped by N₂-laser, absorption spectra of the dyes show that the energy transfer is possible ($\lambda_{pump} = 337$ nm). Coumarine 3 (Eastman Kodak) and Stilbene 3* were dissolved in methanol. All concentrations of dyes are given

^{*} Stilbene 3 is the commercial name of the dye commonly used as a fluorescent brightening agent, the additive to a soap powder [11]. We used Ciba-Geigy TINOPAL CBS-x.

in optical density (o.d.) units for 1-mm thick layer at absorption maxima of $S_0 \rightarrow S_1$ bands, i.e., at $\lambda = 389 \text{ nm}$ for C-102 and $\lambda = 347 \text{ nm}$ for S-3. The measurement of optical density is more convenient and in case of small quanti-

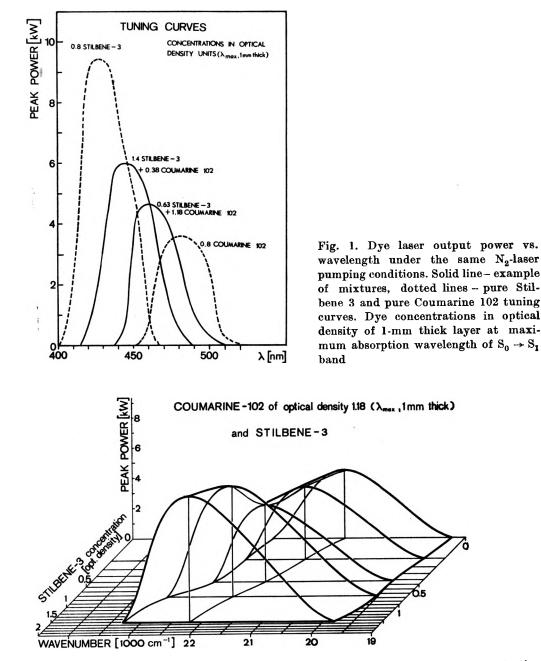


Fig. 2. Peak output power of dye mixture laser at constant Coumarine 102 concentration as a function of Stilbene 3 concentration and of laser wavenumber

ties of solutions and low concentrations – it is much more accurate than the measurement of concentration in grams (or moles) per litre by means of weighing and dissolving the dye in a given volume of solvent. Also some commercially available dyes contain optically inactive components.

Tuning curves, that is, a peak power vs. dye laser wavelength for pure S-3, C-102 and their two chosen mixtures are shown in Fig. 1. Figure 2 presents a set of tuning curves for mixture of C-102 concentration with S-3 at different concentrations. Shift of the tuning range towards shorter wavelengths with increasing S-3 concentration shown in this figure exceeds 1100 cm^{-1} for the maximal power. Similar three-dimensional plot is given by the set of tuning curves for constant S-3 concentration as a function of C-102 concentration (Fig. 3). The shift of the tuning range measured at the maximum power is

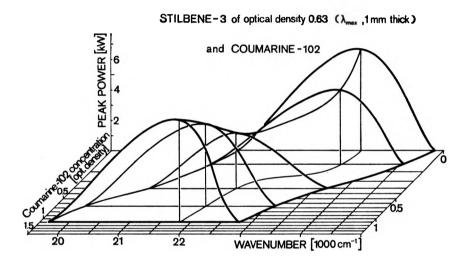


Fig. 3. Peak output power of dye mixture laser at constant Stilbene 3 concentration as a function of Coumarine 102 concentration and of laser wavenumber

almost 2000 cm⁻¹. Figure 4 presents lines of constant tuning ranges as a functions of mixture composition. Solid lines in Fig. 5 represent the maximum obtainable power for different mixture compositions. For the low concentrations a laser is below the threshold. For example, pure C-102 of concentration of 0.35 o.d. does not lase. The addition of S-3 at concentration of 0.2 o.d. enables laser action (at slightly shifted C-102 wavelengths) showing clearly energy transfer. For concentrated mixtures superradiance is overwhelming – it screens tunable laser action. Maximum obtainable power is presented in Fig. 6 as a function of two variables: mixture composition and optical density of the mixture at $\lambda = 337$ nm (a rough estimation of pumping power utilization).

In conclusion we would like to say that one possible way the transfer mechanism may be explained by applying time-resolved spectroscopy or by investi-

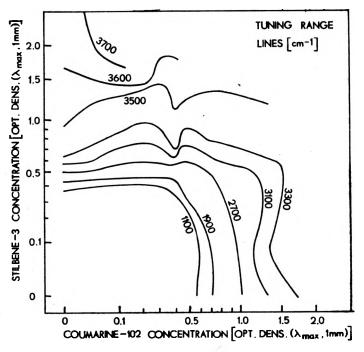


Fig. 4. Tuning range of Stilbene 3-Coumarine 102 mixture dye laser as a function of both dyes concentrations

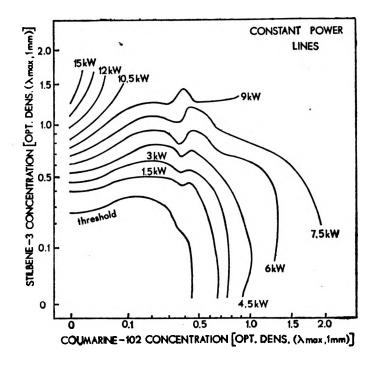


Fig. 5. Maximum output power of Stilbene 3-Coumarine 102 mixture dye laser as a function of both dyes concentrations

gating the mixtures with subsequent changes of components. We intend to examine transfer mechanism by choosing both donor and acceptor dyes with different spectral, fluorescent and lasing properties. It should be of a particular interest to use non-lasing dyes as donors.

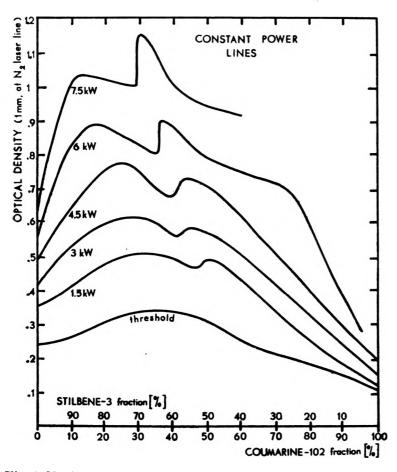


Fig. 6. Maximum output power of Stilbene 3–Coumarine 102 mixture dye laser as a function of two variables: mixture composition and optical density of 1-mm thick layer of mixture at $\lambda = 337$ nm

Acknowledgements – We would like kindly acknowledge the participation of Dr. Stoyan Dinev of Okhrydski University of Sofia, Bulgaria and of Dr. Tomasz Stacewicz in some parts of our measurements.

References

- [1] AHMED S. A., GERGELEY J. S., INFANTE D., J. Chem. Phys. 61 (1974), 1584.
- [2] FORSTER Th., Discussions Faraday Soc. 27 (1959), 7.
- [3] URISU T., KAJIYAMA K., J. Appl. Phys. 47 (1976), 3563.

- [4] KUSUMOTO Y., SATO H., MEANO K., YAHIRO S., Chem. Phys. Lett. 53 (1978), 388.
- [5] BURLAMACCHI P., CUTTER D., Opt. Commun. 22 (1977), 283.
- [6] CHINLON LIN, DIENES A., J. Appl. Phys. 44 (1973), 5050.
- [7] KATRARO R., RON A., SPEISER S., Chem. Phys. Lett. 52 (1977), 16.
- [8] BERLMAN L. B., ROKNI M., GOLDSCHMIDT C. R., Chem. Phys. Lett. 22 (1973), 458.
- [9] PORTER G., TREDWELL C. J., Chem. Phys. Lett. 56 (1978), 278.
- [10] KRASIŃSKI J., MAJEWSKI W., Rev. Sci. Instrum. 47 (1976), 1293.
- [11] MAJEWSKI W., KRASIŃSKI J., Opt. Commun. 18 (1976), 255.

Received April 10, 1984