FIZIKA 10 (1978) 121–132 YU ISSN 0015–3206

LASER WAVELENGTH DEPENDENCE UPON THE CONCENTRATION OF THE LASING DYES

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RECEIVED 8 JULY 1977

Abstract: The paper deals with the results of the study of laser wavelength dependence of transversely pumped dye laser upon the concentration of the lasing dyes: rhodamine B and 6G and sodium fluorescein (uranin). Experimentally determined dependence of the output peak wavelength of the dye laser is compared with the results obtained by using theoretical approach of Atkinson and Pace. The theory predicts laser wavelength within 70 Å in case of rhodamin B and sodium fluoresceine and within 30 Å for rhodamine 6G for dye concentrations $5x10^{-4}$ - $2x10^{-3}$ mol/l.

1. Introduction

Most theoretical studies of dye laser oscillators follow the approach of Sorokin et al[1]. in which rate equations are written down for the single mode photon density, the population inversion and triplet state population. In this approach the quantities are all assumed to be uniform along the laser cavity. Weber and Bass[2] and Snavely[3] on the other hand considered rate equations for the population at threshold in order to calculate gain but they did not include stimulated emission in these equations. Recently Atkinson and Pace[4] have introduced simple but more complete approach by taking into account multimode laser operation and spontaneous emission. Again they assumed all quantities to be uniform along the laser cavity.

However, in most laser pumped dye laser systems gain is very high and consequently the optimum output coupling is rather large and in some cases 4% reflectivity gives the highest output. In these high-gain systems consideration of amplified spontaneous emission becomes important for understanding of their behaviour. In such systems the spontaneous emission is amplified as it propagates along the excited medium. The emitted radiation exibits "laserlike" properties such as low divergence and spectral narrowing. Therefore, for laser pumped dye laser systems it would be more appropriate to use the term "amplified spontaneous emission" than more common term "lasing".

Most complete theoretical approach where all effects are taken into account was made by Ganiel et al.[5)] Unfortunately, it is very difficult to use any of above theoretical approaches to describe conditions in transversaly pumped dye laser systems with the concentration of the dye in the range 10^{-3} - 10^{-2} mol/l where almost all neccessary data are missing. Namely, at such high concentrations where concentration quenching is very strong one can hardly find data for fluorescence lifetimes, quantum yields, apsorption coefficients etc.

Therefore, we decided in this paper to test simplest but still quite comprehensive theoretical approach[4)] at relatively low dye concentrations in order to estimate whether it can be used to predict laser pumped dye laser output with a reasonable accuracy.

2. Theory

Details of the theory are given elsewhere[4] and here we shall only mention main characteristics of the used theoretical approach.

The rate equantions 5-10 in reference 4 which describe population densities of laser energy levels and density of photons in laser cavity, are similar to the equations derived by the other authors[1,2] to obtain the time dependence of the laser output power. In addition all cavity modes are taken in account. Also, enclosed are the effects of triplet-triplet apsorption on the triplet population and the photon noise generation (spontaneous emission).

In deriving above equations following assumptions have been made:

a. the population densities are uniform throughout the dye cell and cavity;

b. all spatial inhomogenities dye to the standing-wave nature of the cavity modes are neglected as similarly done also by other authors[1-3]. This assumption reduces the complexities of solving both time and space variations of population and photon densities;

c. the time derivatives of the populations can be set equal to zero since all relaxation rates are rapid compared to the time variation of the pumping light intensity W and all the populations are in dynamic equilibrium with values depending only on the instantenous value of W.

The assumptions a. and c. may be questionable in case of laser pumped dye lasers. In particular, the assumption c. since, e.g. in the case of nitrogen laser pumped dye laser pulse length of the optical pump is of the same order of magnitude as the fluorescence lifetime of the molecule.

3. Experiment

The experimental arangement is shown in Fig.1. The dye cell is pumped transversaly with a nitrogen laser (3371 Å) focussed by a 9 cm focal length cylindrical quartz lens. The radiation from the dye cell is analyzed by means of a 0.5 meter, Ebert arrangement, Jarrell Ash spectrometer wavelength calibrated against mercury and sodium lamps.

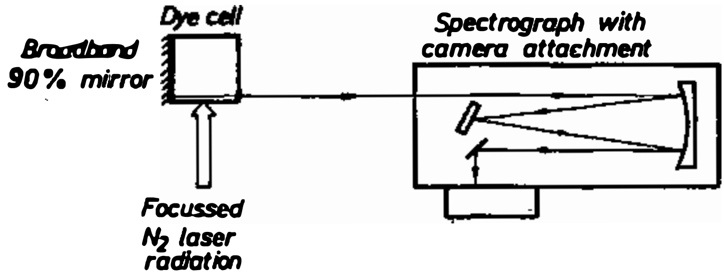

Figure 1. Schematic diagram of experimental arrangement.

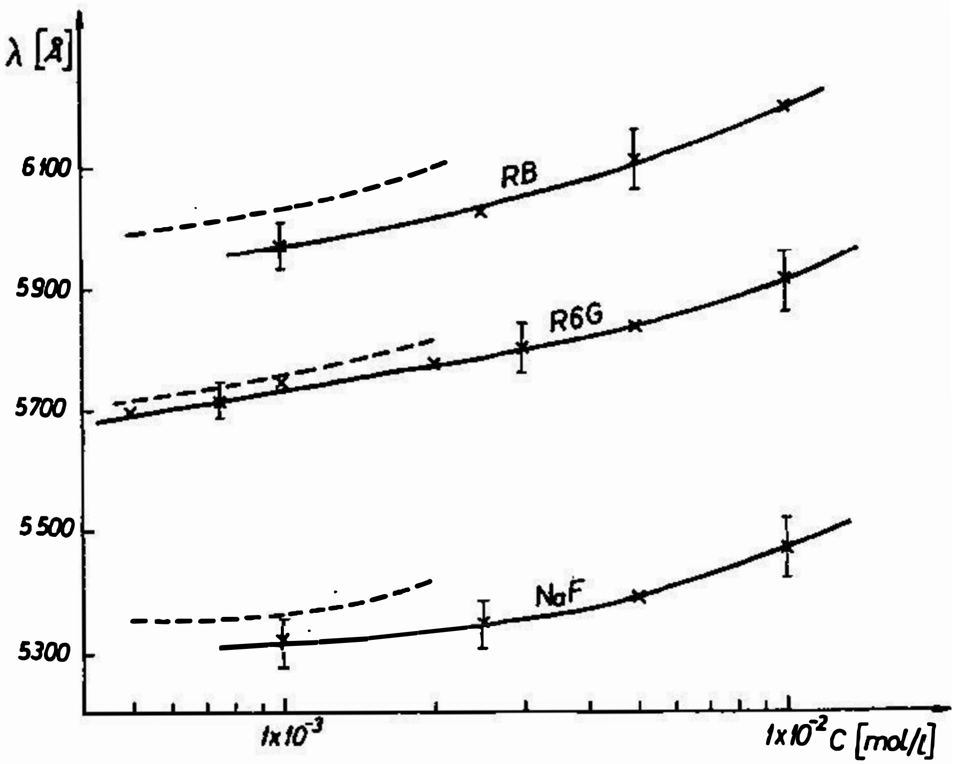

Figure 2. Concentration dependence of the peak wavelength of dye laser emission for rhodamin B and 6G in ethanol and sodium fluorescein in water.

For these experiments we have used home made nitrogen laser of the type developed by Nagata and Kimura[6). A parallel-plate Blumline transmission line consisting of barium titanate capacitors (DNL-24-YW Murata, 750 pF, TV 30kV) is used. Each of the transmission lines consists of 20 capacitors. The discharge chamber (active medium 0.25x4.0 cm cross section, 58 cm long) consisted of two Lucite plates with stainless-steel electrodes inserted between them and sealed with silicone rubber. A fuzed quartz window is atached at an angle to the axis to avoid undesirable feedback of light reflected from the window surfaces. A triggered air spark gap (after Pallaro et al.[7]) is used throughout this work.

N_2 laser pulse width is measured with planar vacuum photodiode and Tektronix oscilloscope 7704A. Time response of the whole light detection system is about 2 ns. Measured N_2 laser pulse width is around 9 ns (the halfmaximum intensity value). The energy of laser pulses are in the range 0.2-0.8 mJ (measured with Gen-Tec joule meter, Model ED200) and therefore power output ranges from 20 to 90 kW.

For all dye laser experiments dyes are obtained from Eastman Kodak and used without further purification.

4. Results and discussion

Experimentally determined wavelengths of N_2 laser pumped dye laser for various concentrations of rhodamine B and 6G and sodium fluorescein are given in Fig. 2. The experimental curves are plotted through the wavelengths of the maximum intensity of dye laser radiation. The full spectral width of the laser pulse is in the range 60 - 100 Å designated in Fig. 2 by vertical bars attached to the values of maximum laser intensity. In Fig. 2 there are also given theoretical results obtained, for the same experimental conditions, by solving numerically equations 6 - 10 from reference 4 on IBM 360/44 computer. For these computations fluorescence lifetimes and cross sections are taken from Snavely[3] for rhodamine 6G and from Peterson et al.[8] and from Mack[9] for rhodamine B and sodium fluorescein. The optical pumping term in equation 2 is assumed to be:

$$W = W_o t^2 \exp(2-0.9t),$$

where t is measured in nanoseconds and W_o is the maximum of nitrogen laser intensity. This form is in fairly good agreement with measured time shape of N_2 laser output. The following parameters are used for calculations:

- reflectivity of the mirrors of the dye laser resonator: $R_1 = 0.90$ (aluminium coating deposited directly on the wall of the quartz cuvette) and $R_2 = 0.04$;
- length of the laser resonator L = 1 cm;
- ratio of the dye cell length to the cavity length F = 1;
- excited volume of the dye V = 0.015 cm^3;
- refractive index of the solution $\eta = 1.36$ (ethanol) and $\eta = 1.33$ (water).

From the analysis of experimental and theoretical results in Fig. 2 one can draw the following conclusions:

a. The agreement between theory[4)] and experiment is within 70 Å for rhodamine B and sodium fluorescein and within 30 Å for rhodamine 6G. Theoretical results being always at the longer wavelength side than those obtained from the experiment.

b. The comparison is made only in the range of dye concentrations $0.5 - 2 \times 10^{-3}$ mol/l. For smaller concentrations it is not possible to obtain laser emission from the dye and for higher concentrations the discrepancy between theory and experiment becomes considerable.

The experimental results in Fig. 2 are reproducible within 2-3 Å and uncertainty in wavelength determination is better than ± 1 Å. Unfortunately it is very difficult to estimate the accuracy of theoretical results in Fig. 2 since the cross-sections of the dyes $\sigma(\lambda)$ and $\sigma_{SS}(\lambda)$ are taken from the drawings in reference 3.

Finally, from the comparison of theory and the experiment in Fig. 2 one can conclude that in spite of very crude approximations (see section 3 of this paper) the agreement is reasonable good. This suggests that the theory[4)] can be used to predict lasing wavelength of organic dyes under the conditions of transverse pumping and at relatively low dye concentrations.

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ZAVISNOST TALASNE DUŽINE LASERSKE EMISIJE ORGANSKOG TEČNOG LASERA OD KONCENTRACIJE BOJA

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S a d r ž a j

U radu su dati rezultati proučavanja zavisnosti talasne dužine laserske emisije iz organskih boja (rodamina 6G i B i natrijum fluoresceina) u uslovima transverzalnog pumpanja sa impulsnim azotnim laserom. Eksperimentalni rezultati zavisnosti talasne dužine laserske emisije od koncentracije boje uporedjeni su sa vrednostima dobijenim koristeći teorijski prilaz Atkinsona i Pace. Teorija i eksperiment se slažu u granicama od 70 Å u slučaju rodamina B i natrijum fluoresceina, a u slučaju rodamina 6G u granicama od 30 Å za koncentracije boja od 5×10^{-4} do 2×10^{-3} mol/l.