INVESTIGATIONS ON DEGREE OF POLARIZATION OF LASER RADIATION OF ORGANIC DYE SOLUTIONS

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The effect of rotational depolarization on degree of polarization of rhodamine 6G dye laser as a function of viscosity of the solution excited by linearly polarized laser beam has been investigated. The experimental and the theoretical results agree very well for the $1 \cdot 10^{-4}$ mole/l solution, while disagreement was found for the $1 \cdot 10^{-3}$ mole/l solution.

Introduction

The polarization characteristics of laser radiation of organic dye solutions were described in [1-11]. These results show that the degree of polarization of dye laser radiation generated by a tunable dye laser which is pumped either by ruby or by neodym laser mono-impulses is nearly 100 per cent, and depends on solvent viscosity, temperature, *etc*, [1-11]. The effect of rotational depolarization has been treated recently [12-13].

Theoretical and experimental results are in striking agreement in the case of phtalimides indicating the applicability of the model used [12].

The question arises whether the polarization characteristics of other systems can be described by this model. Here we report on the polarization characteristics of rhodamine 6G dye laser using alcohol-glycerol mixture of different molar ratios as solvent pumped by the first harmonic of ruby laser monoimpulse, with regard to rotational depolarization.

Experimental arrangement

The first harmonic of ruby laser monoimpulse ($\lambda = 347.15$ nm) was used for generation, which had a pulse duration of 15—20 ns and energy of ≈ 30 mJ, so its power was 2.0—1.5 MW. The electric vector of the generating radiation was set in direction z which was perpendicular to the plane defined by the optical axis of dye laser x and by the optical axis of ruby laser y. The latter pumped the dye laser transversally. The cell of the dye laser was a fused quartz cuvette of 1×1 cm² in size; the mirrors had a reflexion coefficient of 99 and 90 per cent, respectively. The analysis of the dye laser radiation was performed by means of a Wollaston prism. The compo-

nents of the dye laser light with oscillation direction parallel (z) and perpendicular (x) to the electric vector of the exciting laser beam was observed photographically, without resolving the spectrum. Thus the degree of polarization obtained refers to the integral degree of polarization. An ORWO (Wolfen) film type NP 27 was used as a detector calibrated specially by laser pulses [14].

The block diagram of our experimental arrangement is shown in Fig. 1.



Fig. 1. The block diagram of the experimental arrangement, R_1-R_4 — mirrors, Q-Q-switch, Rb — ruby laser, E — energy measuring instrument, KDP — KDP crystal, UG-1 — ultra-violet filter, L_1, L_2 — lenses, C — dye cell, He-Ne — gas lasers, D_1-D_3 — diaphragms, W — Wollaston prism, CA — camera

Experimental results

Investigations were carried out with rhodamine 6G solutions of different viscosities varying the dye concentrations $1 \cdot 10^{-4}$ mole/l through $1 \cdot 10^{-3}$ mole/l. The viscosity of the solutions was changed by changing the glycerol content if the alcoholglycerol mixture used as solvent. The measurements were performed at room temperature, thus the error was about 2—3 per cent, smaller than the error of photographic detection. The results are shown in Fig. 2, where the values of 1/p are plotted via $K\tau$ ($K=kT/V\eta$), k is the Boltzmann's constant, T is the absolute temperature, V the molar volume of a molecule including the solvate envelop, η is the viscosity in cP, τ the decay time. With increasing viscosity the value of the integral degree of polarization tends to -1 for the $1 \cdot 10^{-4}$ mole/l solution; whereas this limiting value is lower in the case of the $1 \cdot 10^{-3}$ mole/l solution. Solutions with the same viscosity but with different concentrations exhibit different polarization degree.

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Discussion

The experimental values of degree of polarization are negative since the pumping transition is an $S_0 \rightarrow S_2$ one, whereas the lasing transition is the $S_1 \rightarrow S_0$ one.

Changes in the degree of polarization vs. the viscosity and the concentration are the result of molecular processes which take place during the excitation and the radiation, and are also affected by dye-solvent interaction. (Such processes are the Brownian rotational motion of the molecules, orientational relaxation, energy transfer with and without radiation, etc.)

The effect of rotational depolarization can be theoretically dealt with as previously described [12], taking into account that the excitation occurs into the state S_2 . Therefore, for the value of the gain (k) at a given direction ψ we can write (see Fig. 3)

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$$k(\psi) = \frac{k_{\max}}{4} \left[4 - \overline{\sin^2} \,\xi - (2 - 3 \,\overline{\sin^2} \,\xi) \cos^2 \delta \cos^2 \psi \right];$$

$$k_{\max} = k(\xi = 90^\circ, \psi = 90^\circ) = \frac{1}{5} \frac{hv}{v} \frac{\overline{B}_{21}(v_l) \overline{B}_{13}(v_g)}{\overline{A}_{21}},$$
(1)

where $\overline{\sin^2 \xi} = \frac{4K\tau}{6K\tau+1}$, here δ denotes the angle between the electric vector of the exciting light \vec{E}_g and the axis z (in our case was $\delta = 0$), ψ is the value of the angle between the electric vector of radiation of dye laser \vec{E}_l and axis z; h is Planck's constant and v is the light velocity in the active material, $\vec{B}_{21}, \vec{B}_{13}, \vec{A}_{21}$ are the Einstein's

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coefficients averaged for the vibration levels. Knowning $k(\psi)$ for the energy density $W(\psi)$ of the radiation at a given direction ψ we obtain:

$$W(\psi) = u_0(\psi)(1-R_3) \left\{ \frac{e^{k'l}-1}{k'} \left[1+R_4 e^{k'l} \right] \frac{q^{m-1}-1}{q-1} + m_c q^m (1+R_4) l \right\},$$
(2)

where R_3 and R_4 are the reflexion coefficients of the output and the broadband light reflector mirrors, respectively,

 $k' = k(\psi) - k_v$, where k_v is the loss coefficient;

 $q = R_3 R_4 e^{2k'l};$

l is the lenght of the cuvette;

 $u_0(\psi)$ is the density of spontaneous radiation in direction ψ ;

m is number of the cycles during the non-stationary generation;

m_e is the number of the cycles during the stationary generation.

The value of degree of polarization can be calculated with the aid of intensity components I_z and I_x from the equation:

$$p = \frac{I_z - I_x}{I_z + I_x} \tag{3}$$

where

$$I_{\rm Z} = \int_0^{\pi/2} W(\psi) \cos^2 \psi d\psi,$$

$$I_X = \int_0^{\pi/2} W(\psi) \sin^2 \psi d\psi.$$
(4)

Changing k_{max} in Eq. (1), and the duration of the non-stationary and stationary generation we simulated the system as a function of viscosity. Details of the mathematical simulation are given elsewhere [12]. The volume of the molecules was regarded constant. The calculations were carried out with a R—40 computer. The results obtained are plotted by dotted lines in Fig. 2. It can be seen that the experimental and theoretical results agree very well for dilute solutions (1 \cdot 10⁻⁴ mole/l), indicating that the model accounts for the molecular processes well.

Note that Eq. (3) has a limiting polarization value of -1. However, we found a value of -0.75 for the dense solution $(1 \cdot 10^{-3} \text{ mole/l})$ and, thus, we have to add a further term in Eq. (3) to take into account explicitly concentration depolarization. A quantitative treatment of this problem will be published soon.

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ИССЛЕДОВАНИЕ СТЕПЕНИ ПОЛЯРИЗАЦИИ ИЗЛУЧЕНИЯ ЛАЗЕРА НА КРАСИТЕЛЕ

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Исследовалось влияние вращательной деполяризации на степень поляризации излучения лазера на красителе родамина 6Ж, в зависимости от вязкости растворителей при лазерном возбуждении линейно поляризованным светом. Экспериментальные и теоретические результаты хорошо согласуются при концентрации активного вещества равной 1.10-4 моль/л, а в случае концентрации 1.10⁻³ моль/л наблюдается расхождение.