DISPERSION OF INDUCED AMPLITUDE AND PHASE ANISOTROPY OF VISCOUS SOLUTIONS OF DYES

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Studied are conducted to investigate changes in the state of polarization of probing radiation that passed through anisotropically excited solutions of rhodamine 6G and 3,6-tetramethyldiamino-N-methylphthalimide in glycerin. The studies allowed the authors to find characteristics of spectral behavior of amplitude and phase anisotropy in these systems. A qualitative difference is found between the spectral behavior of differences in the refractive indices $\Delta n = n_y - n_z$ of these solutions simulated by different systems of energy levels of active molecules.

Key words: amplitude anisotropy, phase anisotropy, Stokes parameters, ellipticity of polarization, amplification (brightening) dichroism, birefringence.

As is known, interaction of linearly polarized high-intensity radiation with organic molecules in solutions changes their populations and orientation order in the ground and excited states. As a result, an excited system is characterized by amplification (brightening), dichroism, and birefringence, i.e., by the appearance of amplitude and phase anisotropy. The excited molecular system acquires the optical properties of a uniaxial amplifying crystal. Theoretical consideration of the origin of this phenomenon is based on the anisotropic optical properties of organic molecules and their orientation order induced by linearly polarized high-intensity radiation [1, 2].

Intensive studies of amplitude and phase anisotropy of excited solutions of dyes have been carried out in recent years, since such molecular systems are used as active media in lasers and amplifiers and as phototropic shutters for generation of nano-, pico-, and femtosecond pulses of radiation [3-9]. In these studies special attention is rendered to phase anisotropy. However, it should be emphasized that a serious drawback of particular works, for example, [7, 9], is that their authors consider theoretically only the case of isotropic excitation. Meanwhile, their experimental method involves polarized excitation, which produces anisotropy of the orientation distribution of dye molecules, which is neglected in theoretical considerations. Therefore, comparison of the theory and experiment may be considered not quite correct. In most of the quoted works, studies of phase anisotropy revealed only the presence of ellipticity of polarization of probing radiation that passed through an excited solution of a dye, but provided no information about the parameters of the ellipse (the orientation, the ratio of the axes, the direction of rotation). It is evident that this information can be considered rather important and useful when dyes are used in various laser systems. This information was obtained for the first time in [10, 11] for a glycerin solution of rhodamine 6G in the amplification band at the wavelength $\lambda_p = 567$ nm of the probing radiation.

The present work is devoted to a search for the main spectral relations of amplitude and phase anisotropy in systems of excited and unexcited molecules of various organic compounds in solutions, in which ellipticity of the probing light wave is possible as a result of its interaction with this medium. Investigation of this problem will result in estimation of induced optical anisotropy of the studied solutions at absorption and amplification bands, which is important for further development and perfection of the theory of polarization of radiation of dye lasers and...
amplifiers. The state of polarization of probing radiation that passed through an excited solution of a dye was studied with the use of Stokes parameters [12, 13]:

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\begin{align*}
S_0 &= I(0^\circ, 0) + I(90^\circ, 0), \\
S_1 &= I(0^\circ, 0) - I(90^\circ, 0), \\
S_2 &= I(45^\circ, 0) - I(135^\circ, 0), \\
S_3 &= I(45^\circ, \pi/2) - I(135^\circ, \pi/2).
\end{align*}
\]

These parameters carry the most complete information about the state of polarization, i.e., they allow detection of ellipticity of polarization and determination of the ratio \( b/a \) of the axes of the ellipse, its orientation, and the direction of its rotation. Since an excited solution of a dye is optically a uniaxial crystal, whose optical axis is directed along the electric vector of exciting radiation, then with known parameters of the ellipse, it is possible to determine the difference between the refractive indices of ordinary and extraordinary waves \( \Delta n \) and its sign. For this experimental geometry, the difference of the refractive indices of the ordinary and extraordinary waves is defined by the expression \( \Delta n = n_Y - n_Z \). Knowledge of the sign at \( \Delta n \) is necessary to determine if we are dealing with a negative or a positive crystal. This method is described in more detail in [10, 11]. It should be noted that this method was preliminarily tested for light sources with known polarization.

The optical scheme and geometry (in the chosen coordinate system) of the experimental setup used in the studies is shown in Fig. 1. A YAG:Nd\(^{3+}\) laser (the repetition frequency is 1 Hz) was used as an excitation light source. Radiation pulses of its second harmonic (\( \lambda = 530 \) nm, \( E = 33 \) mJ, duration is 15 nsec) were split into two beams by a divider plate. One of them was used for excitation of the dye laser, whose radiation was used for probing the studied dye solution. If necessary, both beams were attenuated by neutral light filters. The dye laser operated near the lasing threshold, which provided rather short light pulses (of about 2-3 nsec) in comparison with the pumping pulses. In the experimental studies, which were carried out in a wide spectral range, the wavelength of the probing radiation was smoothly changed by replacing the dye and using a diffraction grating with a period of 1200 line/mm as a "blank" mirror. In addition, the active medium of the dye laser was excited by radiation of both the second and third harmonic of the YAG-Nd\(^{3+}\) laser (\( \lambda_{ex} = 530 \) and 353 nm). A diaphragm provided on the path of the probing light beam decreased inhomogeneity of amplification in the cross-section and eliminated the effect of luminescence of the studied solution. The necessary orientation of the polarization plane of the probing beam was achieved by using a \( \lambda/4 \) phase plate and a Glan prism. At the inlet to the studied solution the electric vector of the light wave of the probing radiation formed a certain angle \( \phi_0 \) with the axis \( Z \) (Fig. 1), which was varied from 0 to 90° during measurements.