The developed sample tunable laser thus has a number of advantageous features and may find applications in various branches of science and engineering.

POLARIZATION DIAGRAM FOR DYE-BASED LASERS

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As is known, the radiation from dye-based lasers with laser pumping is considerably polarized. The presently accepted explanation of this polarization [1-5] rests on an analysis of the so-called pregeneration region of the process, which means that induced transitions at the generation frequency are disregarded in considering the particle balance. The amplification factor in the pregeneration region of the process with laser pumping is anisotropic with respect to pumping (in other words, the amplification factor depends on the electric vector of linearly polarized light propagating along the resonator axis). As a result of the anisotropy of the amplification, the degree of polarization of the amplified luminescence increases with time in the pre-generation period. According to the present state of the theory [1-5], this is a comprehensive qualitative account of all the processes leading to the polarization of the laser radiation. In the present work it is shown that a theoretical study of the polarization of laser radiation which confines itself to the analysis of the pre-generation region of the process, neglecting the nonlinear properties of the system, is completely inadequate.

The present work is based on a calculation of the laser kinetics, taking into account the anisotropy that is present in the distribution of dipole moments of the dye-molecule transitions. No calculations of this kind are to be found in the literature. The kinetic equations usually employed are written in terms of characteristics averaged over all the possible orientations of the molecule; this approximation is only permissible if there is intense Brownian rotational motion of molecules in the solution. In this approximation, the laser radiation should be depolarized under nanosecond pulse excitation, but such depolarization is experimentally observed only in exceptional cases, for special formulations of the experiment, i.e., under ordinary experimental conditions, the rotational motion is not so intense that the anisotropy created by the pumping is equalized.

In the present work the anisotropy that appears in the distribution of the dipole moments of molecular transitions is taken successively into account in the numerical solution (within the framework of the balance method) of problems on dye-laser kinetics, under conditions of transverse monochromatic pumping of linearly polarized radiation with arbitrary polarization angle \( \eta \) (Fig. 1). The function \( u_2(t, \phi, \psi) \) characterizes the distribution of the energy density of laser radiation over components characterized by the angle \( \phi \) between the z axis and the electric vector \( \mathbf{E}_L \) of the \( \psi \) component. The anisotropic distribution of the molecular oscillators in the ground and excited states is characterized by the functions \( n_i = n_i(t, \theta, \phi, i = 1, 2) \), where \( \theta \) and \( \phi \) are spherical coordinates specifying the orientation of the oscillator with respect to the Cartesian coordinate system chosen in Fig. 1. The kinetic equations used in the present work were formulated in [6, 7]. Here they are written in slightly modified form:

\[
\begin{align*}
\dot{n}_2 &= \frac{\nu}{4\pi} k_{\text{amp}}^2 n_1 L + \mathcal{L}, \\
\dot{n}_1 + n_2 &= \frac{N}{4\pi}, \\
\dot{n}_2 &= n_1 \left[ 3B_{21}^0 U + 3B_{21}^L \right] - n_2 \left[ 3B_{12}^0 U + 3B_{12}^L \right],
\end{align*}
\]

where the amplification factor \( k_{\text{amp}} = k_{\text{amp}}(t, \psi) \) is given by the expression

\[
k_{\text{amp}}(t, \psi) = \frac{\nu n}{U} \left[ \frac{B_{21}^0}{3B_{21}^0} \right] \int_0^{2\pi} \int_0^{\pi} \cos^2 \zeta_L \left[ n_2 - n_1 \frac{B_{21}^L}{B_{21}^0} \right] \sin \theta d\theta d\phi.
\]

the dot over the symbols denotes differentiation with respect to time. In Eqs. (1)–(4) the following notation is used:

\[
\mathcal{L} = \nu L A_{21} (\nu L) \Delta \nu \frac{A_{21}}{4\pi} \int_0^{2\pi} \int_0^{\pi} n_2 \cos^2 \zeta_L \sin \theta d\theta d\phi.
\]
In Eq. (5) the integration is taken over the semicircle formed by the intersection of the plane $\psi=\text{const}$ with the upper half-sphere. The remaining notation is conventional (see [7]).

Calculations were made for single-pulse pumping of Gaussian form with width 30 nsec at the half-height. The amplitude value of the pumping-energy density is denoted by $u_{\text{max}}$. The following parameter values were taken: $B_{12} = 2 \cdot 10^6$; $B_{21} = 0$; $B_{12}' = 6 \cdot 10^6$; $B_{21}' = 0$; $p_{21} = 2 \cdot 10^8$; $N = 5 \cdot 10^{24}$; $\mu = 0.27$; $v = 2 \cdot 10^8$; $\nu_L = 6 \cdot 10^{14}$; $\Delta \Omega_L = 10^{-6}$; $\Delta \nu_L = 10^{12}$ (all the values are in IS units).

As an example, the time dependence of a number of laser characteristics for $\eta = 60^\circ$ is shown in Fig. 2. The energy density of the laser radiation is calculated from the equation

$$u_L(t) = \int_{-\pi/2}^{\pi/2} u_L(t, \psi) \sin \psi d\psi = I_y + I_z,$$

and, as usual, the degree of polarization is

$$P = (I_z - I_y)/(I_z + I_y),$$

where $I_y$ and $I_z$ are defined in Eq. (7). As is evident from Fig. 2, the whole process of laser generation is clearly separated into three parts: the pregeneration stage, which ends with the first appearance of intense induced radiation; a transitional stage, characterized by oscillation of the amplification factor and energy density of the laser radiation; and the quasisteady stage, characterized by approximate equality of the amplification and loss factors. Each stage will be considered in turn.

In the pregeneration region, the anisotropy of the amplification is described by the expression [5]

$$k_{\text{amp}}(t, \psi) = \frac{1}{3} k_{\text{amp}}(t, \psi = 0, \eta = 0)[1 + 2 \cos^2 \eta \cos^2 \psi],$$

from which it is evident, in particular, that the generation threshold rises with increase in $\eta$. Taking into account the symmetry of $n_1$ with respect to the angular variable, it may be shown that at any moment of time

$$k_{\text{amp}} = k_y \sin^3 \psi - k_z \cos^3 \psi,$$

where $k_y = k_{\text{amp}}(t, \pi/2)$ and $k_z = k_{\text{amp}}(t, 0)$. Hence it follows that $k_y$ and $k_z$ are extremal values of the amplification factor, i.e., $k_{\text{amp}}(t, \psi)$ lies between $k_y$ and $k_z$ for values of $\psi$ in the range 0–90° at any moment of time.

As a result of the anisotropy of the amplification in the pregeneration stage, the approximately exponential rise in the intensity of the $\psi$ components* (each with its own amplification factor) is accompanied by an increase in the degree of polarization. The rate of increase in the degree of polarization is determined ultimately by the absolute difference between the maximum and minimum values of the amplification factor and does not depend on whether or not the amplification exceeds the losses. As is evident from Fig. 2, the rise in the degree of polarization occurs extremely rapidly, although the anisotropy of the amplification is relatively small at $\eta = 60^\circ$. Calculation shows that even at $\eta = 85^\circ$ the degree of polarization in the pregeneration stage rises to 0.6–0.9 (depending on the losses and the pumping energy). Thus even extremely slight anisotropy of the amplification leads to significant polarization of the amplified luminescence in the pregeneration region.

The behavior of the laser in the transitional period is of particular interest, since it shows the way in which steady — i.e., distinctively laser — characteristics of the radiation develop. As is evident from Fig. 2, intense induced radiation appearing in the resonator rapidly reduces most of the excited molecules to the ground state, which is evident in the rapid drop in amplification. At the same time, there is a significant change in the distribution of molecular orientations and in the anisotropy of the amplification. This change takes different forms for different values of $\eta$. The case of orthogonal excitation ($\eta = 0^\circ$) differs from that in Fig. 2: After the drop in amplification, the amplification factor remains a maximum for $\psi = 0^\circ$, as in the pregeneration period, and moreover it remains anisotropic for a period equal to several laser pulses. With increase in $\eta$ to

*Note that the energy density of the radiation at the laser frequency remains too small for it to be shown in Fig. 2.