DICHROISM OF THE GAIN OF AN ACTIVE MEDIUM OF DYE LASERS GENERATING POLARIZED RADIATION

S.V. Nikolaev and V.V. Pozhar

Institute of Radio Physics and Electronics,
Ukrainian Academy of Sciences, Khar'kov
Received October 9, 19975

The behavior of the gain of dye lasers with polarization anisotropic cavities is theoretically investigated. In the stationary approximation, the spectral and polarization characteristics of the gain are calculated for two regimes of laser operation: with broadband and narrow-band cavities. It is shown that gain anisotropy of linearly polarized radiation is frequency dependent, that is, it exhibits dichroism. It is established that this effect is caused by the overlap of the dye emission and absorption bands and depends on the laser parameters: pumping rate and Q-factor of the cavity. In addition, for a narrow-band tunable laser the dependence of dichroism of the gain on the tuning frequency has been established.

Polarization of the pulsed radiation of dye lasers (DLs) with flash-lamp pumping is most often used for minimization of losses on reflection from edges of various intracavity elements used to control over a generation spectrum. This circumstance connecting in practice the polarization and spectral characteristics of DL calls for their joint examination when studying the generation processes in these lasers. However, in the investigations of the DL polarization characteristics carried out previously, only a single-frequency approximation was used, which cannot give a comprehensive idea of the specific features of the gain of their active media with wide overlapping bands of absorption and emission. Therefore, this paper is devoted to a theoretical analysis of spectral properties of the DL gain with polarization-anisotropic cavity.

To solve the formulated problem, we proceeded from the fact that in general, when modeling the dye molecule as a linear oscillator, common for the absorption and emission, the gain can be described by the formula[5]

$$K_g(t,\psi,\nu) = 3\sigma_{em}(\nu)\iint n_2(t,\theta,\varphi)\sin\theta\cos^2\xi\,d\theta\,d\varphi - 3\sigma_{ab}(\nu)\iint n_1(t,\theta,\varphi)\sin\theta\cos^2\xi\,d\theta\,d\varphi, \qquad (1)$$

where t is the time; ν is the frequency; ψ is the angle specifying the sense of polarization vector of the radiation flux in the laboratory system of coordinates; θ and φ are the spherical coordinates specifying the direction of the oscillator axis; $n_1(t,\theta,\varphi)$ and $n_2(t,\theta,\varphi)$ are the distribution functions of molecules over their orientations in the ground and excited states, respectively; $\sigma_{em}(\nu)$ and $\sigma_{ab}(\nu)$ are the emission and absorption cross sections of the molecule; ξ is the angle between the molecular oscillator and the electric vector of the radiation component

$$\cos\xi = \sin\theta\sin\varphi\sin\psi + \cos\theta\cos\psi.$$

To write Eq. (1), we used a simplified scheme of two-electron molecular energy levels: the ground level with population density $n_1(t,\theta,\varphi)$ and the first singlet excited level with population density $n_2(t,\theta,\varphi)$. The process of DL generation in this approximation is described by the system of generalized kinetic equations

$$\frac{dU(t,\psi,\nu)}{dt} = (cl/L)\,U(t,\psi,\nu)\times[K_g(t,\psi,\nu) - K_l(\psi,\nu)] + G(t,\psi,\nu), \qquad (2=)$$

$$\frac{dn_2(t,\theta,\varphi)}{dt} = W(t)\,n_1(t,\theta,\varphi) - n_2(t,\theta,\varphi)\times\left[\tau_s^{-1}\int 3\sigma_{em}(\nu)\int U(t,\psi,\nu)\cos^2\xi(\psi)\,d\psi\,d\nu\right] + n_1(t,\theta,\varphi)\int 3\sigma_{ab}(\nu)\int U(t,\psi,\nu)\cos^2\xi(\psi)\,d\psi\,d\nu, \qquad (2b)$$

$$n_2(t,\theta,\varphi) + n_1(t,\theta,\varphi) = N/4\pi, \qquad (2c)$$

where $U(t,\psi,\nu)$ is the number density of emitted photons, c is the light velocity, l is the length of the active medium, L is the cavity length, K_l is the loss factor for the radiation component ψ, $W(t)$ is the pumping rate, τ_s is the lifetime of the excited state, N is the molecule concentration in the solution, $G(t,\psi,\nu)$ is the luminescence power of a unit volume of solution along the cavity axis.

0235-6880/98/ 02-03 172-04 $02.00

Further the problem was solved in the stationary approximation. In so doing, it was generally assumed that $dn_2(t, \varphi, \theta) = 0$ and that stationary generation is excited for the component with $\psi = 0$. In this case, within the time over which the pulse acts, the following condition is satisfied for this component:

$$K_g(0, \nu_g) = K_l(0, \nu_g). \quad (3)$$

Hence

$$\begin{cases} U_g = U(0, \nu_g) \neq 0, \\ U(\psi \neq 0, \nu = \nu_g) = 0 \end{cases}$$

and then Eq. (2*b*) with consideration of Eq. (2*c*) takes the form

$$(N/4\pi)\,[W + 3c\,\sigma_{ab}(\nu_g)\cos^2\theta] - n_2(\theta, \varphi)\,\{W + \tau_s^{-1} + 3c\,[\sigma_{ab}(\nu_g) + \sigma_{em}(\nu_g)]\,U_g\cos^2\theta\} = 0. \quad (4)$$

Let us assume that $3c\,\sigma_{ab}(\nu_g)\cos^2\theta \ll W \ll \tau_s^{-1}$. Then

$$n_2(\theta, \varphi) = W N \tau_s/4\pi\,[1 + 3\,\tau_s\,c\,\sigma_{em}(\nu_g)\,U_g\cos^2\theta]. \quad (5)$$

By substituting Eq. (5) in Eq. (1) and integrating, we derive

$$K_g(0, \nu) = 3\,W N\,\sigma_{em}(\nu)\,\tau_s \times [1 - (\arctan\sqrt{a})/\sqrt{a}]/a - N\,\sigma_{ab}(\nu), \quad (6)$$

$$K_g(\pi/2, \nu) = 3\,W N\,\sigma_{em}(\nu)\,\tau_s \times (\arctan\sqrt{a})/2\sqrt{a} - [K_g(0, \nu) + 3\,N\,\sigma_{ab}(\nu)]/2, \quad (7)$$

where $a = 3c\,\sigma_{em}(\nu_g)\,U_g\,\tau_s^{-1}$ is the ratio of the probability of induced transitions to the probability of spontaneous transitions. The parameter a can be estimated if we consider that from condition (3) the equation follows:

$$[1 - (\arctan\sqrt{a})/\sqrt{a}]/a = [N\,\sigma_{ab}(\nu_g) + K_l(0, \nu_g)]/3\,W N\,\sigma_{em}(\nu_g)\,\tau_s \equiv A(\nu_g).$$

A comprehensive idea of the character of the gain gives the relation

$$K_g(\psi, \nu) = K_g(0, \nu)\cos^2\psi + K_g(\pi/2, \nu)\sin^2\psi. \quad (8)$$

In order that with the help of Eqs. (6), (7), and (8) to calculate the dependences of the gain from ψ and ν and to analyze its change attendant to variations of the DL parameters, the generation frequency ν_g should be known. In connection with this, we consider separately two regimes of DL operation: with the broadband cavity and with the dispersion cavity.

To determine ν_g of the DL with the broadband cavity, we take advantage of the fact that generation in this case occurs at the frequency for which the condition

$$dK_g(0, \nu_g)/d\nu = 0 \quad (9)$$

is satisfied.

Together with Eqs. (3) and (1), this condition permits us to write down the system of equations from which ν_g can be determined

$$3\,\sigma_{em}(\nu_g)\iint n_2(\theta, \varphi)\sin\theta\cos^2\theta\,d\theta\,d\varphi - N\,\sigma_{ab}(\nu_g) = K_l(0, \nu_g),$$

$$\iint n_2(\theta, \varphi)\sin\theta\cos^2\theta\,d\theta\,d\varphi\,(d\sigma_{em}/d\nu) - N\,(d\sigma_{ab}/d\nu) = 0. \quad (10)$$

In so doing, the profiles $\sigma_{em}(\nu)$ and $\sigma_{ab}(\nu)$ can be approximated by the formulas[6]

$$\sigma_{em}(\nu) \approx \sigma_{em}^{max}\exp\{-\ln 2\,[2\,(\nu - \nu_0^f)/\Delta\nu]^2\},$$
$$\sigma_{ab}(\nu) \approx \sigma_{em}(\nu)\exp[h\,(\nu - \nu_{00})/kT, \quad (11)$$

where σ_{em}^{max} is the maximum value of $\sigma_{ab}(\nu)$, ν_0^f is the frequency of maximum $\sigma_{em}(\nu)$, $\Delta\nu$ is the halfwidth of the profile $\sigma_{em}(\nu)$, h is the Planck constant, ν_{00} is the frequency of pure electron transition, k is the Bolzmann constant, and T is the temperature. Solution of the system of equations (10) with the use of approximation (11) gives

$$\nu_g = \{[h/kT + (8\,\nu_0^f\ln 2)/(\Delta\nu) + 1/\Delta\nu] - \{[h/kT - (8\,\nu_0^f\ln 2)/(\Delta\nu)^2 + 1/\Delta\nu^2]^2 - (16\ln 2)\,[h\nu_{00}/kT + (4\,\nu_0^{f\,2}\ln 2)/(\Delta\nu)^2 + \nu_0^f/\Delta\nu + \ln(K_l(0, \nu_g)\,kT\,8\ln 2)/N\sigma_{em}\,h\Delta\nu + 1]/(\Delta\nu)^2\}^{1/2}\}\,(\Delta\nu)^2/8\ln 2\,. \quad (12)$$

In the derivation of Eq. (12) it was assumed that $(\nu_0^f - \nu)/\Delta\nu < 1$. Formulas (6)–(8) and (12) can be used to calculate $K_g(\psi, \nu)$ directly, thereby analyzing the character of the gain of the medium. Corresponding analysis was made and gain anisotropy was established, as expected; however, in contrast with the single-frequency approximation, it became clear that the maximum of $K_g(\psi, \nu)$ is achieved at the frequency $\nu_{max} \neq \nu_g$. Moreover, for each component ψ, ν_{max} was different. The frequency ν_{max} can be determined from the condition $dK_g(\psi, \nu)/d\nu = 0$. Using Eqs. (6)–(8) and approximation (11), we find

$$\nu_{max}(\psi) \simeq \left[\ln \frac{F(\psi)\ kT\ 8\ln 2}{h\Delta\nu} + \frac{h\nu_{00}}{kT} + \frac{\nu_0^f}{\Delta\nu} - 1 \right] \times$$

$$\times \left(\frac{h}{kT} + \frac{1}{\Delta\nu} \right)^{-1}, \quad (13)$$

where

$$F(\psi) = 3W\ \tau_s \times \{A(\nu_g)\cos^2\psi + [B(\nu_g) - A(\nu_g)]/2\};$$

$$B(\nu_g) = (\arctan\sqrt{a})/\sqrt{a}.$$

The frequency dependence of the gain anisotropy, or in other words, gain dichroism is clearly demonstrated by the dependences $\nu_{max}(\psi)$ and $K_g(\psi, \lambda_{max})/K_g(0, \lambda_g)$ shown in Fig. 1. Here and further we bear in mind that $\lambda = c/\nu$. These dependences were calculated for the laser on rhodamine 6G with the following values of the molecular parameters: $\sigma_{em}^{max} = 1.85{\cdot}10^{-16}$ cm^2, $\tau_s = 7.4$ ns, $\nu_{00} = 5.45{\cdot}10^{14}$ Hz, $\nu_0^f = 5.26{\cdot}10^{14}$ Hz, and $\Delta\nu = 8{\cdot}10^{13}$ Hz. Calculations were done for two rates of pumping and two values of the parameter $K_g(0, \nu_g)/N$ to demonstrate the dependence of the dichroism on the Q-factor of the cavity and the pumping rate. From Fig. 1 it can be seen that the frequencies of maximum gain for each ψ are different and significantly detuned from ν_g. The maximum shift ν_{max} occurs at $\psi = \pi/2$. The decrease of the cavity losses or the increase of the pumping rate increases the shift ν_{max} and anisotropy of the gain.

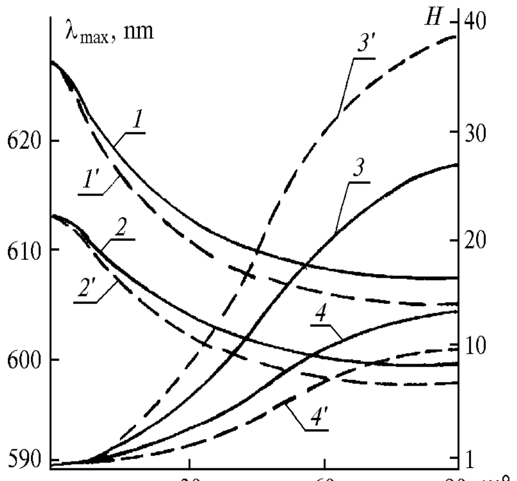

FIG. 1. Broadband cavity. Dependence of λ_{max} *(curves 1, 1′ and 2, 2′) and H (curves 3, 3′ and 4, 4′) on* ψ. $K_1/N = 10^{-20}$ *(1, 1′, 3, 3′)* and 10^{-19} cm^2 *(2, 2′, 4, 4′); W* = $3{\cdot}10^6$ *(1–4) and* $7{\cdot}10^6$ s^{-1} *(1′–4′).*

All problems discussed above refer to the general case in which the absorption and emission spectra of the dye overlap, that it, $\sigma_{ab} \neq 0$. If they do not overlap, then, as our calculations have shown, the frequency dependence of anisotropy does not arise and hence gain dichroism is absent. In this case, only gain anisotropy is observed which can be described, in analogy with the polarization degree, by the degree of anisotropy D defined as

$$D = \frac{K_g(\pi/2, \nu) - K_g(0, \nu)}{K_g(\pi/2, \nu) + K_g(0, \nu)}.$$

Then

$$D = \frac{3\beta \arctan\sqrt{a}/\sqrt{a} - 3}{3\beta \arctan\sqrt{a}/\sqrt{a} + 1}, \quad (14)$$

where $\beta = W/W_t$ is the excess of pumping rate W over the threshold W_t. It can be seen that the degree of gain anisotropy depends on the amount of excess of pumping over the threshold. The curve $D(\beta)$ is shown in Fig. 2, from which it can be seen that with the increase of the amount of threshold excess the degree of gain anisotropy is increased.

Now let us examine the DL with dispersion cavity. In this case, the generation frequency ν_g is determined by a dispersion element inserted in the cavity, and in general is not equal to the frequency of the gain maximum for generated polarization mode $\nu_{max}(\psi = 0)$. Therefore, to analyze the character of $K_g(\psi)$ in this regime, formulas (6)–(8) and (13) are also applicable considering that ν_g is no longer dependent on the laser parameters and is specified arbitrary.

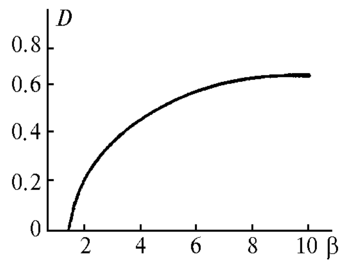

FIG. 2. Dependence of the degree of anisotropy D on the excess of pumping rate β *at* $\sigma_{ab} = 0$.

The results of calculations have shown that the behavior of $K_g(\psi, \nu)$ for DL with dispersion and broadband cavities is similar. In particular, dichroism of the gain exists. The main distinction is that along with the dependence on the Q-factor of the cavity and on the pumping rate the dependence arises on the frequency of detuning of the dispersion cavity. This is vividly illustrated by Figs. 3 and 4.

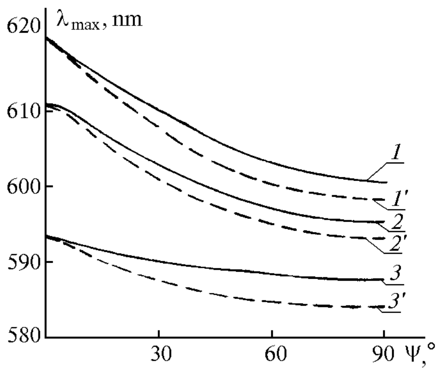

FIG. 3. Dispersion cavity. Dependence of λ_{max} *on* ψ *for* λ_g *a* 615 *(1, 1′),* 600 *(2, 2′), and* 585 nm *(3, 3′);* $W = 3\cdot10^6$ *(1, 3)* and $7\cdot10^6$ s^{-1} *(1′–3′).*

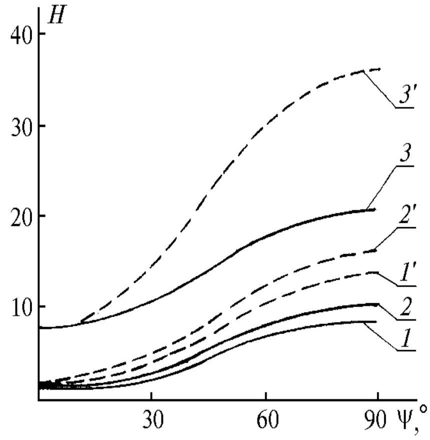

FIG. 4. Dispersion cavity. Dependence of the gain anisotropy H on ψ *for* λ_g = 615 *(1, 1′),* 600 *(2, 2′), and* 585 nm *(3, 3′).*

Figure 3 shows the dependences $\lambda_{max}(\psi)$ calculated for three different λ_g for laser on rhodamine 6G and for two different pumping rates. In our calculations we assumed that $K_l(0, \nu_g)/N = 10^{-20}$ cm^{-2}. As for DL with broadband cavity, λ_{max} is decreased with the increase of ψ so that the condition $\lambda_{max}(\pi/2) < \lambda_{max}(0)$ is always valid. However, this decrease of λ_{max} is different for different wavelengths of cavity detuning λ_g. Its magnitude decreases for shorter generation wavelengths, which is connected, like the resultant effect of gain dichroism, with the magnitude of $\sigma_{ab}(\lambda)$. The same is true for the gain anisotropy, as show the dependences $H = K_g(\psi, \lambda_{max})/K_g(0, \lambda_g)$ for different λ_g in Fig. 4.

Thus, on the basis of our analysis we can draw the following conclusions. In the active medium of DL generating polarized radiation in the presence of re-absorption of emitted radiation by unexcited molecules, gain dichroism arises. The degree of gain anisotropy and the frequency shift of the gain maximum that characterize this effect for each component ψ depend on the *Q*-factor of the cavity and the pumping rate. This is true for DLs with broadband and dispersion cavities. For the DL with dispersion cavity, the dependence of dichroism on the wavelength of cavity tuning is also added.

In the absence of re-absorption in the medium, the frequency dependence of gain anisotropy vanishes, which means the absence of dichroism in this case.

REFERENCES

1. A.M. Ratner, Kvantovaya Elektronika (Naukova Dumka, Kiev), No. 2, 91–105 (1967).
2. S.V. Nikolaev and V.V. Pozhar, "*Some peculiarities in the formation of polarization characteristics of dye lasers with flash-lamp pumping*,B Preprint No. 5, Radio Electronics Institute of the Ukrainian Academy of Sciences, Khar'kov (1995), 21 pp.
3. S.V. Nikolaev and V.V. Pozhar, Atmos. Oceanic Opt. **8**, No. 11, 925–926 (1995).
4. F.I. Morgan and H. Dugan, Appl. Opt. **18**, No. 24, 4112–4115 (1979).
5. L.G. Pikulik and O.I. Yaroshenko, Zh. Prikl. Spektrosk. **27**, No. 1, 53–59 (1977).
6. E.A. Tikhonov and M.T. Shpak, *Nonlinear Optic Phenomena in Organic Compounds* (Naukova Dumka, Kiev, 1979), 384 pp.