STUDY OF THE GAIN OF 2-(4-PYRIDYL)-5-PHENYL OXAZOLE DILUTED IN ETHANOL WITH TRANSVERSE LASER EXCITATION

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Received June 10, 1997

The measurements are presented of the gain of 4PyPO *diluted in ethanol by the calibrated inserted cavity loss method. The gain is measured as a function of the power density of pumping radiation.*

Improvements in the output parameters of tunable lasers on organic compounds call for their pumping by high-power solid-state and gas lasers. High-power light fluxes of lasers used for pumping produce high concentration of excited molecules within the time over which the pumping pulse acts. As a result, the optical characteristics of excited medium are changed not only due to photophysical reasons (clearing of solutions on the lasing wavelength and light absorption by the excited molecules in various states and forms), but also due to the change of thermal parameters of the solutions[1] (formation of thermal lenses and spatial lattices). This causes the increase of the output energy of the tunable lasers on diluted organic compound with the increase of the excitation power.[2–4] Elucidation of reasons for the decrease in the lasing efficiency calls for the study of the optical characteristics of active media with high-power excitation.

An ethanol solution of 2-(4-pyridyl)-5-phenyl oxazole (4PyPO) at a concentration of 10^{-3} mol/l was chosen as an examined active medium. Its characteristic feature is the decrease of the efficiency of conversion of the exciplex XeCl*-laser radiation with the increase of the pumping power density.[5]

The Sank method[6] commonly used to measure the gain of active media based on dye solutions with transverse pumping yields accurate results only for a narrow range of variations of the pumping power (up to 1 MW/cm^2), that is, when the gain is proportional to the pumping power. In case of absorption saturation, this method based on comparison of intense luminescence for different lengths of the active medium yields gross errors, because some excited molecules have no time to make transitions to the ground state during a single radiation passage (for high pumping power density). As a result, the gain is underestimated for high excitation power densities.[7]

To measure the gain of the active medium (the coefficient of negative absorption $K_g + K_l$), in our experiment we used the method of calibrated losses based on insertion of known losses into an optical cavity and their increase until the lasing stopped.[8] Then the net losses would be equal to the gain of the active medium. The condition of self-excitation of the active medium in the presence of the inserted losses assumes the following form:

$$K_g + K_l = \frac{1}{2L} \ln \frac{1}{R_1 R_2 T^2}, \qquad (1)$$

where K_g is the gain due to photophysical characteristics of the medium and K_l is the loss factor in the cavity and medium due to scattering on optical inhomogeneities of the laser material, absorption by inactive centers, and so on; R_1 and R_2 are the reflection coefficients of cavity mirrors; T is the transmission coefficient of the inserted losses. Hence, to obtain K_g, we should know K_l.

To measure the intrinsic losses, we used the method proposed by Aristov et al.[9] It is based on measuring the quantum yield of lasing (η) attendant to changes in the transmission of the cavity mirrors. The mathematical dependence of the parameter $1/\eta$ on the useful mirror loss factor (K_r) is given by the following equation[9]:

$$(1 - 1/X)/\eta = G/(K_r \eta^{\max}) \{K_{\text{abs}} + \alpha_a (1 - \eta^{\max})\}, \qquad (2)$$

where X is the excess of the pumping energy over the threshold, $1/G$ is the usage factor of pumping absorbed in the active medium (when $G = 1$, every absorbed photon excites molecules), $\eta^{\max}$ is the maximum quantum yield,[9] α_a is the coefficient of absorption of generated radiation by the excited dye molecules, $K_r = \frac{1}{2L} \ln \left(\frac{1}{R_1 R_2} \right)$ is the useful loss factor, and K_{abs} is the absorption coefficient specifying additional losses (due to scattering, inactive absorption, and so on) in the solution and cell (except for losses due to absorption by excited molecules and useful mirror losses). Equation (2) was derived by Aristov et al.[9] on the basis of the data published in Ref. 10 under assumptions of stationary generation and uniform excitation. The validity of this formula for pulsing regime was established by Aristov et al.[9] It follows from Eq. (2) that for $X \gg 1$, when the parameters G, $\eta^{\max}$, K_{abs}, and $\alpha_a(1 - \eta^{\max})$ are independent of K_r,

0235-6880/97/11 864-03 $02.00

the dependence of $1/\eta$ on $1/K_r$ is described by the straight line whose slope to the abscissa is

$$\tan\alpha = \frac{G}{\eta^{max}} \{K_{abs} + \alpha_a (1 - \eta^{max})\}. \quad (3)$$

This straight line cuts the ordinate to length G/η^{max}. Thus, the parameter K_l can be easily estimated experimentally using the formula

$$K_l = \frac{\eta^{max} \tan\alpha}{G} = \{K_{abs} + \alpha_a (1 - \eta^{max})\}. \quad (4)$$

The parameters $K_g + K_l$ and K_l were measured for the same geometry of the experiment. The dye was pumped with the XeCl* laser having the parameters λ = 308 nm, t_p = 10 ns, and E = 40 mJ. The pumping radiation was focused with cylindrical lenses into a beam with transverse sizes 2×0.05 cm. The beam was directed onto the end of a cell filled with diluted dye. Neutral light filters were used as attenuators of pumping radiation and as inserted losses. The measurements were carried out for different mirror reflection coefficients (R_1 = 100%, R_2 = 7, 20, 38, and 83%). The relative error in measuring the quantum yield of lasing η was ~10%. To measure $K_g + K_l$ (at the fixed pumping energy), losses were inserted into the cell until lasing stopped, and the sum of the molecular gain and of the loss factor was then calculated for each pumping energy.

The neutral light filters used as inserted losses were oriented to prevent the build-up of spurious lasing from their faces.

As a result, the dependences of $K_g + K_l$ on W were obtained analogous to the dependence shown in Fig. 1. It can be seen that at small pumping power densities, $K_g + K_l$ first increases practically linearly and then saturates at ~5 MW/cm^2. The curves measured for different mirror reflection coefficients coincided within the limits of the experimental error.

The dependences $\eta(W)$ are shown in Fig. 2. It can be seen that η sharply increases near the threshold, reaches its maximum, and then decreases when the pumping power density increases up to 40–50 MW/cm^2.

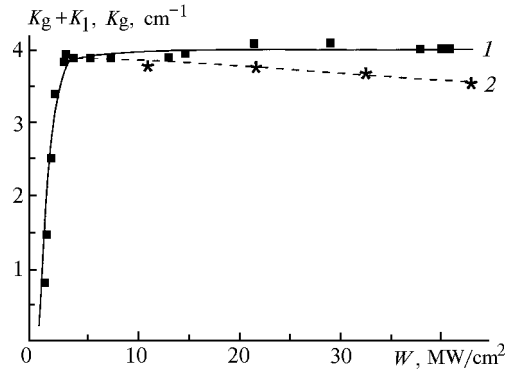

FIG. 1. Dependence of the gain on the pumping power density: 1) $K_g + K_l$; 2) K_g.

To draw the dependence of $1/\eta$ on $(1/K_r)$, the values of η for identical pumping power densities were used (see the curves in Fig. 2). The dependence $1/\eta$□$(1/K_r)$ was linear starting from 10 MW/cm^2. Then we draw the dependences for W = 10, 20, 30, and 40 MW/cm^2.

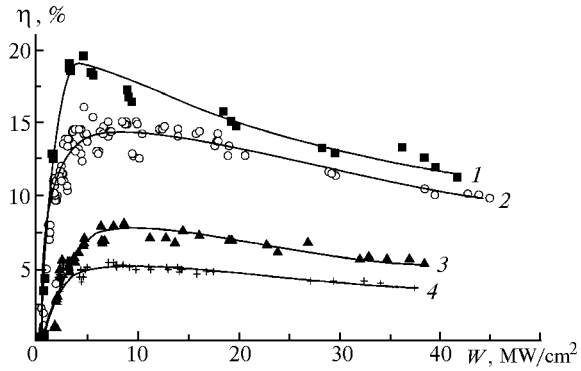

FIG. 2. Dependence of the quantum yield of lasing on the pumping power density for different R_2 = 7 (1), 20 (2), 38 (3), and 83% (4).

Figure 3 shows the dependence $1/\eta$ $(1/K_r)$ for 10 MW/cm^2. In this case, K_l was determined from Eq. (4) using the dependences analogous to those shown in Fig. 3. The values of K_l varied from 0.18 cm^{-1} for 10 MW/cm^2 to 0.42 cm^{-1} for 40 MW/cm^2. For the obtained values of K_l, the molecular gain K_g was drawn (the dashed curve in Fig. 1).

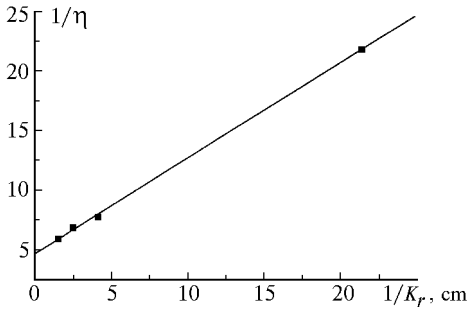

FIG. 3. Dependence of $1/\eta$ on $1/K_r$ for W = 10 MW/cm^2.

An analysis of the obtained results shows that K_g decreases as the pumping power density increases, whereas K_l increases, which agrees with the results obtained by Batishche[2] and Aristov et al.[9] for rhodamine 6 J.

There are several reasons for this behavior of K_g and K_l in high-power light fields, namely, the increase of scattering of generated radiation with the increase of the pumping density,[9] incoherent excess luminescence,[11,12] and so on. Elucidation of the degrees of their effect on $K_g(W)$ and $K_l(W)$ as well as the effect of K_g and K_l on $\eta(W)$ calls for further investigations.

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