

Measurements of the gain factor in laser active media of dyes

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The technique is proposed to measure the gain in the laser active media of organic compounds under conditions of high-power pumping. The gain factors in substituted pyridyloxazole 4PyPO, 4PyPO-N(CH₃)₂, 4PyPO-OCH₃ and substituted *para*-terphenyl LOS-1 were measured. The gain and efficiency are studied as functions of the pump power density.

Introduction

One of the problems in the development of high-power dye lasers is the decrease in efficiency at high pump intensity.¹⁻⁴ Since the gain $K_g^0 = n_1 \sigma_{10}(\lambda_r)$, the direct measurements of it as a function of pump power density allows one to judge on the processes running in a molecule under these conditions.

The Sank method, which is usually applied to laser dyes, allows measurements of K_g^0 to be performed at the power density $< 1 \text{ MW/cm}^2$, when the signal gain equal to the ratio I/I_0 (where I_0 is the input signal intensity, and I is the intensity at the output from the lasing medium) increases linearly with the pump.⁵⁻⁶ At high pump power density in the media characterized by high gain, the exponential dependence of (I/I_0) on the length of the active zone breaks down, what makes the Sank method inapplicable. In the general case one should take into account the dependence of the gain on the intensity of radiation to be amplified. Thus, in a two-level system the dependence of the gain on the radiation intensity takes the form⁷:

$$K_g = K_g^0 / [1 + \alpha_{ij} u_{ij}(I)] \quad (1)$$

where K_g is the gain of a high-power flux; α_{ij} is the parameter of nonlinearity; u_{ij} is the density of stimulating radiation for the laser levels ij . At a low flux of the stimulating radiation, the gain K_g is equal to K_g^0 .

Technique of determination of K_g^0

The method of measuring K_g^0 is based on the method of calibrated losses,⁸ in which K_g^0 is considered as a negative loss. The measurement technique is the following: calibrated losses are introduced into a plane-parallel cavity until the lasing stops. At that moment the total loss (introduced loss, natural loss, the loss due to the radiation escape from the cavity) is equal to the gain. For active media characterized by low gain, such that the law of gain evolution can be considered as the exponential one all over the length of an active

medium (that is, $K_g = K_g^0 = \text{const}$), the threshold condition of lasing is

$$K_g^0 - \rho = \frac{1}{2L} \ln (R_1 R_2 T^2)^{-1}, \quad (2)$$

where ρ is the natural loss; L is the length of an active medium; T is the coefficient of transmission of the introduced loss; R_1 and R_2 are the reflectances of the cavity mirrors.

Active media based on organic compounds are characterized by high gain, what leads to a decrease in the gain depending on the length of the active medium as the light flux intensity grows. To describe the process of amplification in such a medium, the following model was proposed.

Let the flux I be amplified at the point $L = 0$ of the inverted medium. Let L_0 be the maximum length of the active medium, at which the exponential law holds, that is, the integral Bouguer law

$$I(L_0) = I(0) e^{(K_g^0 - \rho)L_0} \quad (3)$$

is fulfilled.

In the active medium of the length L_0 all molecules are considered emitting, that is, the population inversion is completely "removed. B Then the radiation intensity after passage of the next layer of the length L_0 is equal to $I(L_0)$, that is, for radiation having passed the layer $2L_0$ we can write

$$I(2L_0) = 2I(0) e^{K_g^0 L_0} e^{-2\rho L_0}. \quad (4)$$

And for the whole length of the active zone L

$$I(L) = I(0) e^{K_g^0 L_0} (L/L_0) e^{-2\rho L}. \quad (5)$$

Thus, the threshold condition of lasing for the radiation having traveled round the entire cavity with the introduced loss has the form

$$e^{K_g^0 L_0 - \rho L} (R_2 + 1) (L/L_0) T^2 R_1 = 1; \quad (6)$$

$$K_g^0 = (-\ln [\frac{L}{L_0} T^2 (R_2 + 1) R_1] + 2\rho L) / L_0. \quad (7)$$

For most optical compounds $\rho \ll K_g^0$ under conditions of lasing. With the active zone length such that $2\rho L/L_0 \ll K_g^0$, let us denote $K_0 = K_g^0 - 2\rho L/L_0$

for further transformations to be simpler. Then the working equation takes the form

$$K_0 = (1/L_0) \ln (L/L_0) T^2(R_2 + 1) R_1. \quad (8)$$

To find the unknown parameter L_0 , let us consider the case $L = L_0$. Two models of radiation amplification must be valid in this case: the model based on the integral Bouguer law (that is, $I(L)$ follows the exponential law) and the above-described model. By equating the threshold conditions of lasing of both of the models for a laser with the active medium of the length L_0 , we derive

$$(1/2L_0) \ln R_1 R_2 T^2 = (1/L_0) \ln (R_2 + 1) R_1 T^2. \quad (9)$$

Upon solution of Eq. (9), we obtain the relationship between T and R

$$T_0 = \sqrt{R_1 R_2 / [(R_2 + 1) R_1]}, \quad (10)$$

at which both of the models are valid at the point $L = L_0$. Thus, there is an opportunity to experimentally determine L_0 at the given R_1 and R_2 .

The technique for determining L_0 : the loss T_0 given by Eq. (10) is introduced into a plane-parallel cavity formed by mirrors with the reflectances R_1 and R_2 . Then lasing is stopped by changing (decreasing) the length of the active zone. At that moment the length of the active zone is equal to L_0 .

The technique for determining K_0 : the threshold loss T_{thr} (neutral density filters) are introduced into a plane-parallel cavity formed by the mirrors R_1 and R_2 with the length of the active zone $L > L_0$ until lasing stops. The experimentally obtained T_{thr} and L_0 at a given pump power density are substituted into Eq. (8).

Experimental setup

Figure 1 shows the block-diagram of the experimental setup. The quartz cell 4 cm long, 0.5 cm thick, and 2 cm high was filled with a solution under study. The quartz cylindrical lenses with the focal lengths $F = 250$ and 450 mm were used. The cavity was formed by the mirrors R_1 and R_2 .

To change the length of the excited zone, a set of rectangular diaphragms of different length ($L = 0.57 - 2.04$ cm) and a slit with the changeable length ($L = 0 - 0.4$ cm with the step of 0.001 cm) were used. The pumping radiation was focused so that the pump power density was constant, regardless of the active zone length. Neutral density filters were used to attenuate the pump radiation.

The exiplex XeCl* laser with the following characteristics: $\lambda_{\text{pump}} = 308$ nm; $e_{\text{pump}} = 20-25$ mJ; $\tau_{\text{pump}} = 10$ ns was used for pumping.

The pump and lasing energies were measured with a highly sensitive KTP-2 calorimeter head with the F-138 nanovoltmeter calibrated against IMO-2N accurate to $\pm 1\%$. Focusing allowed us to obtain the excitation intensity of 70 MW/cm² as high.

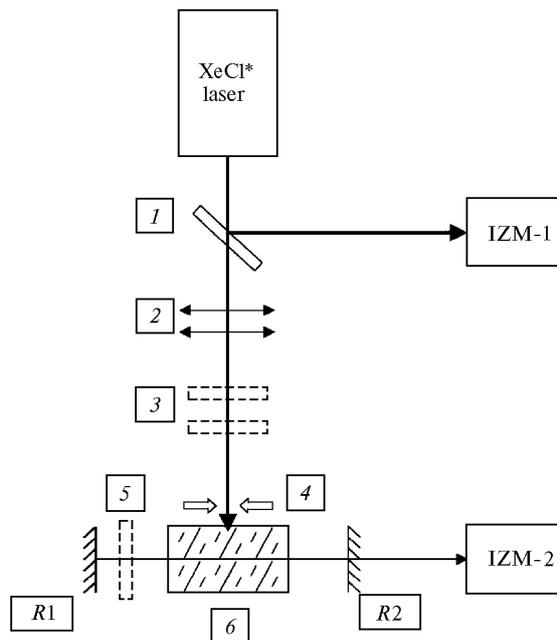


Fig. 1. Block-diagram of the experimental setup: beam splitter 1; cylindrical lenses 2; attenuators 3; diaphragm 4; introduced loss 5; cell filled with a dye 6; cavity mirrors R_1 and R_2 ; energy meters IZM-1 and IZM-2.

Objects of the study and experimental results

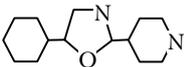
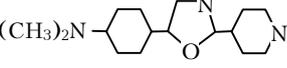
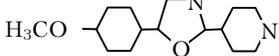
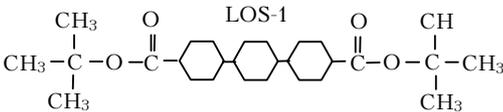
The measurements were conducted in the laser active media based on molecules with different structure and processes proceeding in them and thus having different spectral and lasing properties. The structure formulas and characteristics of the compounds under study are given in the Table 1.

LOS-1 is the derivative of *para*-terphenyl. This compound ideally suits the conversion of XeCl*-laser radiation into the tunable UV radiation, because the pumping comes into the maximum of the long-wave absorption band, and the quantum yield of fluorescence is equal to 0.96. The efficiency of lasing in our excitation scheme is 35% with $\lambda_{\text{max}} = 375$ nm.

The substituted pyridyloxazole 4PyPO is the well-known laser dye for the region of 400 nm, as well as two its derivatives 4PyPO-OCH₃ and 4PyPO-N(CH₃)₂. They are less efficient in fluorescence and lasing than LOS-1. The quantum yield of fluorescence is equal to 0.9, 0.8, and 0.3, respectively. The lasing efficiencies are lower too; their values are given in the Table. For 4PyPO-N(CH₃)₂ molecule it was found that in ethanol solutions TICT-state is formed at excitation, that is, structure reconstruction takes place, which leads to the anomalously large Stokes shift and worsening of the lasing properties.⁹

The difference in spectral-luminescence and lasing properties of these compounds must lead to different gain. Experimental values of K_0 , which correlate with the lasing efficiency, would allow us to judge on the sensitivity and reliability of the technique proposed.

Table 1. Spectral-luminescent characteristics of the compounds under study

Objects of study	Characteristics of longwave absorption			Characteristics of fluorescence		$A_{21} \cdot 10^8, \text{ s}^{-1}$	$\sigma_{10} \times 10^{-16}, \text{ s}^{-2}$	Concentration, $\frac{\text{mmol}}{\text{l}}$	$K_0, \text{ cm}^{-1}$ (exp.)	Efficiency, %	$K_g, \text{ cm}^{-1}$ (limiting)
	$\nu_{\text{abs}}, \text{ cm}^{-1}$	$\Delta\nu_{\text{abs}}, \text{ cm}^{-1}$	$\frac{\epsilon, \text{ l}}{\text{cm} \cdot \text{mol}}$	$\lambda_{\text{fl}}, \text{ nm}$	$\Delta\lambda_{\text{fl}}, \text{ nm}$						
4PyPO 	31000	6000	22600	390	57	4.7	3.5	0.5	55±10%	16	105
4PyPON(CH ₃) 	26200	5200	21200	560	105	2.5	4.2	0.5	40±10%	9	126
4PyPO(OCH ₃) 	29400	5800	22400	440	72	3.25	3.0	2.0	45±10%	2	361
LOS-1 	32600	5400	44000	373	50	8.2	5.7	0.5	125±10%	35	172

The results of investigations into the dependence of K_0 on the length of the excited zone L are shown in Fig. 2.

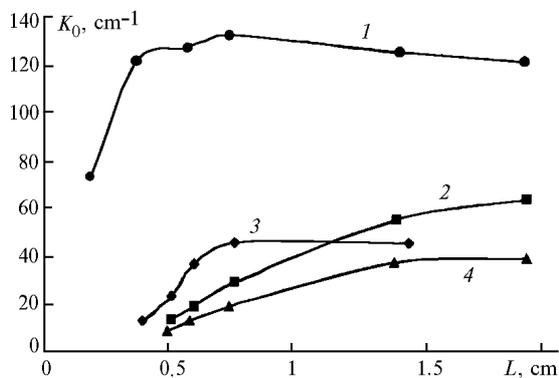


Fig. 2. Dependence of the gain on the length of the active medium in ethanol solutions: LOS-1, concentration of 0.5 mmol/l (curve 1); 4PyPO, concentration of 0.5 mmol/l (curve 2); 4PyPO(OCH₃), concentration of 2 mmol/l (curve 3); and 4PyPO-(NCH₃)₂, concentration of 0.5 mmol/l (curve 4).

As the length increases up to some value of L , the gain for all the compounds increases, then the increase stops and the gain remains constant. Such a behavior of K_0 can be related to the increase of losses at decreasing L , since the value actually determined in experiments is $K_0 = K_g^0 - 2(L/L_0) \rho$. The decrease in the length of the active zone at unchanged cavity base leads to the increase of one component of the natural losses,

namely, the loss due to the radiation escape through the lateral surfaces of the cavity. Since this loss is equal to $\sim 1/L$, at large enough L the loss is small and changes only slightly as L increases. Thus, for such lengths of the active zone that K_0 is unchanged, we can consider the natural losses small in comparison with K_g^0 and use K_0 as an intramolecular characteristic of the active medium.

One can see from the Table 1 that the changes in the experimentally measured gain correspond to the change in the efficiency and spectral-luminescent characteristics of the studied compounds. Thus, for the solution of LOS-1, which has the maximum efficiency, the measured value of K_0 equal to 125 cm^{-1} is also maximum. It should be noted that for LOS-1 the limiting gain equal to $n\sigma_{21}$ (n is the solution concentration), that is, determined from the condition that all molecules are in the excited state, is closest to the measured K_0 as compared with the other compounds. This is indicative of small intramolecular losses, which decrease the number of molecules being at the lasing level. For the 4PyPO solution these values differ by almost two times. For 4PyPO-(OCH₃) and 4PyPO-N(CH₃)₂ the measured value of K_0 differs from the limiting gain even more strongly. Such a difference between the limiting gain and the experimentally measured K_0 agrees with the decrease in the lasing efficiency and is connected with the presence of significant intramolecular losses. The losses can be caused either by the transition of molecules into the triplet state or structure (spatial) reconstruction of

molecules in the excited state, as it was shown for 4PyPO-N(CH₃)₂ (see Ref. 9).

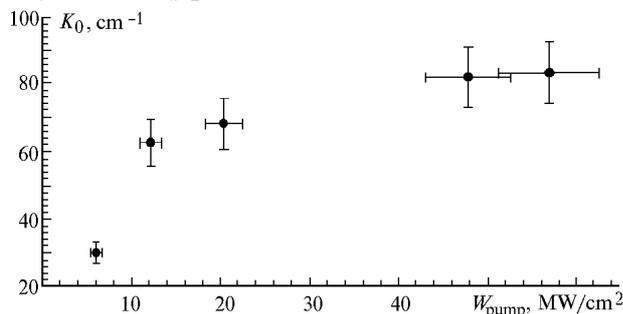


Fig. 3. Dependence of the gain on the pump power density for the ethanol solution of LOS-1 (concentration of 0.37 mmol/l) at the length of the active zone $L = 1.44$ cm.

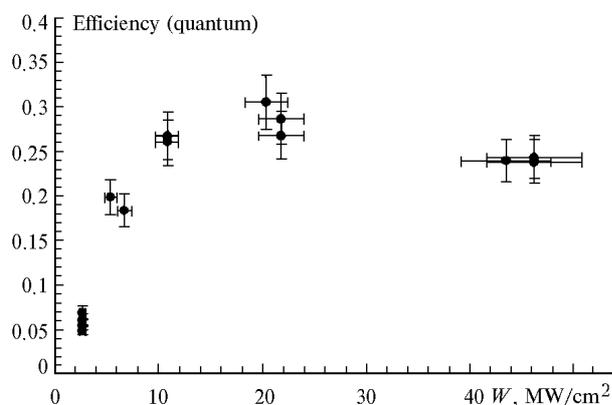


Fig. 4. Dependence of the quantum efficiency on the pump power density for the ethanol solution of LOS-1 (concentration of 0.37 mmol/l) at the length of the active zone $L = 1.44$ cm.

Figures 3 and 4 show the dependence of K_0 and efficiency for the LOS-1 on the pump power density W , respectively. One can see from the figures that as W increases up to ≈ 20 MW/cm², K_0 and the efficiency increase too due to the growth of inversion. At the further growth of W the value of K_0 practically

does not change. The dependence of the efficiency on W is of somewhat different character: at high values of the pump power density $W > 30$ MW/cm², some decrease in the efficiency is observed. The cause of this difference requires further investigation.

The results presented show that the changes in K_0 for the studied compounds represent the actual physical pattern of the set of photophysical processes proceeding in these molecules and determining their lasing capability. These results allow the conclusion to be drawn that the proposed technique is applicable to measurements of the gain with due regard for the above-mentioned restrictions on the length of the active zone.

Acknowledgments

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