

Radiative and photochemical properties of organic compounds excited by high-power XeCl laser radiation

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Abstract. Radiative and photochemical properties of a number of laser dyes excited by focused radiation of a XeCl laser with intensity up to 200 MW cm^{-2} were studied. A method for measuring the gain of organic molecules under high-power excitation is proposed. The dependence of the dye transmittance for the pump radiation on its intensity was studied. It is shown that changes in energy, spectral, and time characteristics of radiation and the photostability of compounds under high-power excitation are associated with the formation of superluminescence.

1. Introduction

To obtain a high output power in tunable dye lasers, high-power pump radiation is commonly used. This poses problems of retaining a high radiation conversion efficiency and photostability of active laser media under high-power excitation. In recent years, specific features of radiative properties of organic compounds under high-power laser excitation have been discussed in the literature [1–5]. In particular, the possibility of changes in the Einstein coefficient for spontaneous emission under these conditions was discussed in Refs [1, 2].

In this paper we will discuss specific features of radiative properties of organic molecules, estimate the gain of laser media on the basis of organic compounds pumped by high-power XeCl laser radiation and their photostability under conditions of spontaneous emission, superluminescence, and lasing.

2. Objects of study and the experimental technique

The choice of molecules for our study (see Table 1) is determined by the fact that they have different spectral, luminescence, and lasing properties because of the various photo-processes occurring in them. It is interesting in this connection to find specific features of the action of high-power exciting radiation on these molecules. Radiation in the near-UV region can be produced with high efficiency by lasing in LOS-1 (substituted paraterphenyl). This compound has a quantum efficiency of 51% for lasing in this

region, and its effective cross section for stimulated emission at the emission band centre is $\sigma_{10} = 5.7 \times 10^{16} \text{ cm}^2$. A high quantum efficiency is the result of optimum excitation conditions. The XeCl^{*} laser radiation at 308 nm corresponds to the absorption band maximum of LOS-1. The next group of compounds represents pyridyl oxazoles lasing in a spectral region that is important for practical applications. In particular, 4PyPO has an emission maximum at 400 nm, which is of considerable importance for the production of substances of high purity. Substituents in the phenyl cycle considerably expand the emission region of pyridyl oxazoles. For the dimethylamino-substituted 4PyPO in polar solvents, an anomalous Stokes shift and a decrease in the quantum yield of fluorescence in comparison with the compound in nonpolar solvents are observed, which is caused by the formation of states with intramolecular charge transfer and 'twisted' geometry (TICT conformations) [7]. Lasing characteristics of such conformations under high-power excitation were not studied.

The radiative properties of these molecules under high-power excitation were studied on the setup described in Ref. [8]. An XeCl laser beam of size $21 \text{ mm} \times 8 \text{ mm}$ ($\lambda_g = 308 \text{ nm}$, $E_g \leq 50 \text{ mJ}$, $\tau_{1/2} = 10 \text{ ns}$) was focused by two crossed cylindrical lenses with focal lengths 25 cm and 50 cm either to a rectangle of size $8 \text{ mm} \times 0.5 \text{ mm}$ (version 1) or to a spot 1.5–2 mm in diameter through a 1.2 mm aperture (version 2). A cell of thickness 1 mm with a dye solution was used. The aperture cut out the central region of the incident beam, which made it possible to obtain sufficiently uniform excitation representing a parallelepiped of size $8 \text{ mm} \times 0.5 \text{ mm} \times 1 \text{ mm}$ in version 1, or a cylinder 1 mm high with a base 1.2 mm in diameter in version 2. Version 1 has a well-pronounced preferred direction of the excited region, which is virtually absent in version 2.

The focusing allowed us to obtain a pump radiation intensity W up to 60 MW cm^{-2} in version 1 and up to 250 MW cm^{-2} ($3.9 \times 10^{26} \text{ photon cm}^{-2} \text{ s}^{-1}$) in version 2. In the experiments we simultaneously measured the pump radiation intensity and the solution transmittance at the pump wavelength with respect to the solvent transmittance. The photostability of molecules in high-intensity light fields was studied by the technique described in Ref. [9]. From measurements of the absorption spectra of solutions before and after irradiation, we determined the quantum yield of phototransformation ϕ and the relative yield of photoproducts produced as a result of the phototransformation. The lasing photostability, which characterises the resource for operation of a medium, was determined by the total pump energy P_{80} absorbed in a unit volume of a lasing solution until the moment when the efficiency decreased to 80% of the initial value.

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Table 1. Spectral, luminescence, and lasing properties of the compounds under study

Compound	$\lambda_{\text{fl}}^{\text{max}}/\text{nm}$	$\sigma_{01}^{308}/10^{16} \text{ cm}^2$	$\sigma_{10}/10^{16} \text{ cm}^2$ *	η_{fl}	Efficiency** (%)	K_0/cm^{-1} (experiment)	Efficiency****	$K_{\text{am}}^{\text{max}}/\text{cm}^{-1}$	$C \text{ mmol l}^{-1}$
Rhodamine 6G	560	0.53	13.8	0.75	36	–	–	–	–
p-Terphenyl	340	0.28	4.6	0.92	27.5	–	–	–	–
LOS-1	373	1.67	5.7	0.96	51	125	35	172	0.5
4ø	395	0.76	3.5	0.9	20.7	55	16	105	0.5
4PyPOOCH ₃	440	–	3.0	0.8	–	45	2	361	2
4PyPON(CH ₃) ₂	560	0.32	4.2	0.27	27.8	40	9	126	0.5
	417***	0.95***	2.5***	0.8***	3.3***	–	–	–	–
Coumarin 2	440	0.1	2.2	0.84	39	–	–	–	–
trans-Stilbene	348	1	2.8	0.05	5	–	–	–	–
Bifluorophore (trans-Stilbene– CH ₂ –coumarin 120)	440	1.1	2.2	–	39	–	–	–	–

*Estimated by the formula proposed in [6]; **for optimum concentrations; ***hexane solutions, all the rest remaining data correspond to ethanol solutions; ****obtained under the same experimental conditions as K_0 .

One of the main molecular characteristics of lasing media is their gain. The known methods of its estimation [10] have certain restrictions and cannot be used for high-power excitation. Because of this, we developed a modification based on the calibrated-loss method [11]. For active media with low gain, where a signal is amplified exponentially along the length, the threshold lasing condition for a cavity with inserted loss is of form

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

where $K_{\text{am}}^{(0)}$ is the small-signal gain; ρ is the intrinsic loss; L is the length of an active medium; T is the transmittance of inserted loss; and R_1 and R_2 are the cavity mirror reflectivities.

Active media on the basis of organic compounds are characterised by high gain, which leads to nonlinear effects, i.e., a decrease in the gain for a large effective length of the active region at high radiation intensities [12]. To describe gain in such media, we proposed the following model. Consider optical radiation of intensity I entering an amplifying medium at the point $L = 0$. Let L_0 be the maximum length of the active region where the integrated Bouguer law

$$I(L_0) = I(0) \exp[(K_{\text{am}}^{(0)} - \rho)L_0]. \quad (2)$$

is satisfied. The length L_0 is chosen in a way giving the maximum possible radiation intensity at its output (for a given volume of the active medium), which is limited by the number of molecules in the excited state. After passage through the second layer, we have

$$I(2L_0) = 2I(0)e^{K_{\text{am}}^{(0)}L_0}e^{-2\rho L_0}. \quad (3)$$

As a result, one can write for the total length L of the active medium the expression

$$I(L) = I(0)e^{K_{\text{am}}^{(0)}L_0}e^{-\rho L} \left(\frac{L}{L_0} \right). \quad (4)$$

The threshold lasing condition, for radiation making a round trip through a cavity with inserted threshold loss T and mirror reflectivities R_1 and R_2 , takes the form [11]

$$(R_2 + 1) \frac{L}{L_0} T^2 R_1 \exp[(K_{\text{am}}^{(0)}L_0 - 2\rho L)] = 1. \quad (5)$$

Taking the logarithm, we obtain

$$K_{\text{am}}^{(0)} = - \frac{[\ln\{(L/L_0)T^2(R_2 + 1)R_1\}]}{L_0} + 2\rho \frac{L}{L_0}. \quad (6)$$

To simplify the calculation, let us introduce the notation $K_0 = K_{\text{am}}^{(0)} - 2\rho L/L_0$. As a result, we obtain the formula

$$K_0 = \frac{\ln[(L/L_0)T^2(R_2 + 1)R_1]}{L_0}. \quad (7)$$

To find the unknown parameter L_0 , consider the case $L = L_0$. Equating the threshold lasing conditions for two laser models having an active medium of length L_0 , we obtain

$$\frac{\ln(R_1 R_2 T^2)}{2L_0} = \frac{\ln[(R_2 + 1)R_1 T^2]}{L_0}. \quad (8)$$

The solution of Eqn (8) gives the relation

$$T_0 = \frac{(R_1 R_2)^{1/2}}{(R_2 + 1)R_1}, \quad (9)$$

which determines the validity of both models at the point $L = L_0$. The length L_0 can be determined experimentally. For this purpose, one introduces loss T_0 given by Eqn (9) into the plane-parallel cavity formed by the mirrors with reflectivities R_1 and R_2 . The length of the active region is then decreased until lasing fails. At this point, the length of the active region is L_0 .

To determine K_0 , we introduce the threshold loss T_{th} (neutral optical filters) into the plane-parallel cavity, formed by the mirrors with reflectivities R_1 and R_2 and which has an active region of length $L > L_0$, until lasing fails. The experimental values of T_{th} and L_0 obtained for the given pump intensity are substituted in formula (7).

This method of estimating K_0 has a limitation. In fact, one estimates from Eqn (7) that $K_0 = K_{\text{am}}^{(0)} - 2\rho L/L_0$, and $L > L_0$. As a result, we should have $\rho \ll K_{\text{am}}^{(0)}$. In this case, the quantity K_0 measured by the given method may characterise the amplification properties of a medium. If $\rho \sim K_{\text{am}}^{(0)}$, we can thus determine ρ .

3. Discussion of the results

3.1 Radiation intensity

For all the compounds with different quantum yields of fluorescence that were studied here, the dependence $\lg I(\lg W)$ deviates at a certain value of W from a linear dependence. However, the pump radiation intensity at which the deviation is observed is different for different molecules. As the concentration of molecules changes from 0.001 to 0.1 mmol litre⁻¹, the dependence remains almost unchanged. On going from version 2 to version 1, in which a preferred direction for the development of lasing appears, we find that the dependence still does not change. However, an increase in the LOS-1 concentration from 0.01 to 0.1 mmol litre⁻¹ causes an increase in the rate of growth of intensity I . The dependence $I(W)$ in version 1 is nearly quadratic. In version 2, where a preferred direction for the development of lasing is absent, the function $I(W)$ is linear; i.e., the excitation of an extended spatial region produces amplification of radiation with increasing concentration.

This shows that stimulated processes strongly affect the emission probability of organic molecules in the range of excitation intensities under consideration even at concentrations below the threshold value. An increase in the rate of growth of intensity I in version 1 may be associated with the amplification of spontaneous emission, i.e., superluminescence [13]. This assumption is supported by the results of the study of transmission of pump radiation by solutions and of the time and spectral characteristics of the radiation.

3.2 Transmission of pump radiation by solutions

Our study of the dependences on the pump radiation intensity of the transmission of XeCl laser radiation by solutions (version 2) showed that absorption at 308 nm for the pump intensities W under study differed from the linear absorption measured at $W < 1$ MW cm⁻² with a spectrophotometer.

For 4PyPON(CH₃)₂, the transmittance decreased, as compared with the linear absorption law, with increasing W (Fig. 1), which is caused by induced absorption from excited states at 308 nm during a 10 ns excitation pulse. For LOS-1 and 4PyPO, we observed an increase in transmittance in the region where $W \leq 50$ MW cm⁻², further saturation, and a subsequent decrease in transmittance with increasing W up to 150–200 MW cm⁻² (Fig. 1).

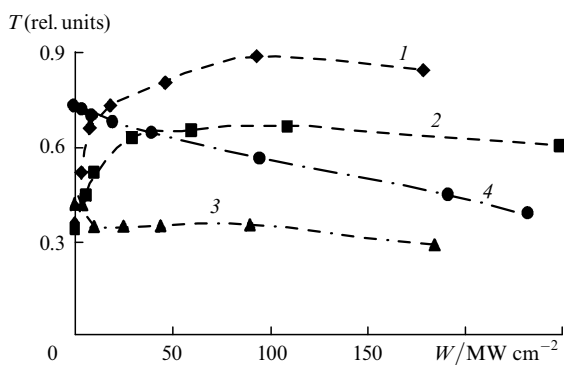


Figure 1. Dependences of the transmission of XeCl laser radiation T on the pump intensity W for LOS-1 in ethanol (1), 4PyPO in ethanol (2), and 4PyPON(CH₃)₂ in ethanol (3) and hexane (4) at the concentration $C = 0.01$ mmol litre⁻¹.

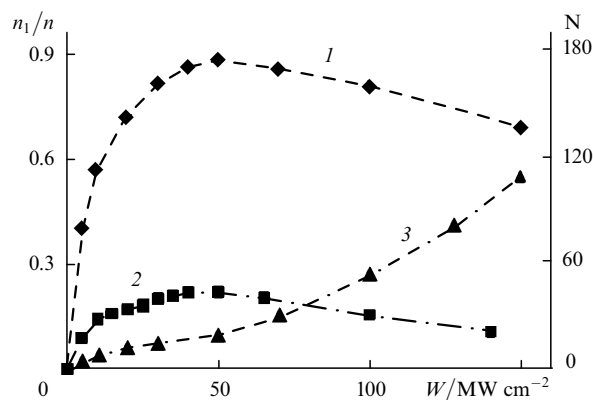


Figure 2. Dependences of the population n_1/n of state S_1 for $\sigma_{10}/\sigma_{01} = 0$ (1) and 3.4 (2) and of the number of photons N absorbed by each molecule during an excitation pulse (3) on the pump radiation intensity.

A decrease in absorption at 308 nm in comparison with linear absorption for LOS-1 and 4PyPO (Fig. 1) suggests that $\sigma_{1n}^{308}/\sigma_{01}^{308} \leq 0.01$ (σ_{1n}^{308} and σ_{01}^{308} are the effective cross sections for absorption at 308 nm from the states S_1 and S_0 , respectively). For LOS-1 in version 2, we made qualitative estimates of the pulse-averaged population n_1/n (n and n_1 are the concentrations of molecules in the ground and excited states) of the state S_1 for $\sigma_{1n}/\sigma_{01} = 0.01$ by the technique proposed in Ref. [1] and of the number of photons absorbed by each molecule during a pulse as functions of W (Fig. 2).

Analysis of the curves in Fig. 2 shows that the magnitude of n_1/n depends on whether or not stimulated processes involved in the population of state S_1 are taken into account (curves 1 and 2), but the form of the curves remains qualitatively unchanged. As photons with $\lambda = 308$ nm are absorbed, an increase in the population of state S_1 takes place mainly at $W < 50$ MW cm⁻². In the region where $W > 50$ MW cm⁻², an increase in the number of absorbed photons of pump radiation leads to the depopulation of state S_1 . The dependence $\lg I(\lg W)$ for LOS-1 correlates with the form of the dependence of n_1/n . A decrease in I and n_1/n is observed for $W > 70$ MW cm⁻². The depopulation of state S_1 at $W > 70$ MW cm⁻² may be caused by transitions to the lower state, i.e., the formation of superluminescence, which is supported by the spectral and temporal characteristics of emission.

3.3 Spectral characteristics of emission

Variations in pump intensity W and the spatial shape of the excited volume (versions 1 and 2) cause changes in the emission spectra of dye solutions (Fig. 3), which manifest themselves in a shift of the band maximum and a change in the band half-width. The concentration at which these changes occur is different for different compounds and depends on the gain of a medium and the excitation conditions. Under our experimental conditions, it was equal to 0.1 mmol litre⁻¹ for LOS-1 and 1 mmol litre⁻¹ for 4PyPON(CH₃)₂.

For the latter compound, which has a large Stokes shift (~ 9000 cm⁻¹), an increase in W leads to band narrowing and a shift of the intensity maximum to the long-wavelength region, which corresponds to the lasing region (Fig. 3b). For LOS-1, the Stokes shift is 5300 cm⁻¹, the overlap of absorption and fluorescence spectra at the given concentration is

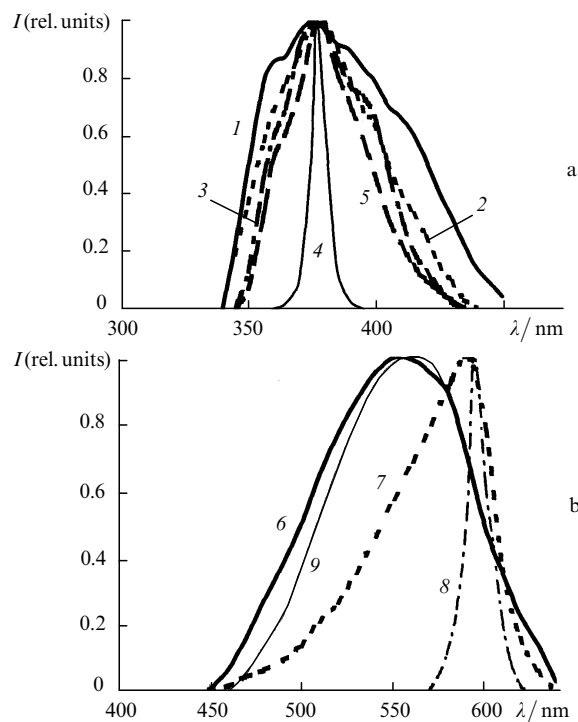


Figure 3. Emission spectra of LOS-1 (a) and 4PyPON(CH₃)₂ in ethanol (b) excited by a XeCl laser for the concentrations $C = 0.01$ (1, 2), 0.1 (3–5), and 1 mmol l^{-1} (6–9) and the pump intensities $W = 1$ (1, 6), 50 (2–4, 7–9), and 150 MW cm^{-2} (5) for versions 1 [1, 2, 4, 6–8 (8 corresponds to the cavity)] and 2 (3, 5, 9).

insignificant, and therefore an increase in W leads only to the narrowing of spectra, without their shift, i.e., lasing is observed at the maximum of the fluorescence spectrum (Fig. 3a).

Thus the spectral changes observed in our experiments with increasing W are caused by the formation of superluminescence.

3.4 Temporal characteristics of emission

The changes observed in the spectral characteristics of emission with increasing W are consistent with the changes in the emission pulse shape under these conditions. In version 2 at $W < 10 \text{ MW cm}^{-2}$, the shape of the emitted pulse is close to that of the pump pulse. The maximum of the emission pulse is slightly delayed, and its duration is increased by the lifetime of the fluorescent state (1–1.5 ns); i.e., this emission represents fluorescence, which is supported by the spectral characteristics (Fig. 3a). In version 1, a similar behaviour is observed at $W < 1 \text{ MW cm}^{-2}$. As the excitation intensity is increased up to $10–50 \text{ MW cm}^{-2}$, the emission pulse length decreases, which is attributed to superluminescence in the case of excitation in an extended spatial region.

The pulse lengthening and the appearance of a shoulder in version 2 at $W > 60–100 \text{ MW cm}^{-2}$ is observed for all the compounds under study, which is likely to support the conclusion that this change in the emission pulse is caused by the onset of the formation of superluminescence. Provided that the conditions for development of superluminescence are favourable [an extended shape of the excited region (version 1) and an increase in W] this leads to the fact that superluminescence becomes the dominant factor, resulting in classical changes in the pulse shape and duration.

3.5 Photostability of LOS-1

The spectral manifestation of LOS-1 phototransformations consists of a decrease in the absorption efficiency in the long-wavelength band and the formation of diffuse absorption in the long-wavelength region (360–450 nm). In addition we observed the additional absorption in the 250–260-nm region after irradiation. The emission spectra of irradiated solutions of LOS-1, when excited in the region of the diffuse band, show that at least two photoproducts absorb in this region. The first photoproduct absorbs at 370–390 nm and emits in the region 430–440 nm, whereas the second absorbs at $\lambda > 400 \text{ nm}$ and its maximum lies in the region 470–480 nm. Our studies showed that the nature of photoproducts formed in LOS-1 solutions was independent of excitation conditions and the kind of emission (lasing or spontaneous emission). However, the yields of phototransformations and their proportion are substantially changed.

In version 1 the quantum yield ϕ of phototransformations of the lasing LOS-1 solution ($C = 1 \text{ mmol litre}^{-1}$) changed only slightly in the range of W from 5 to 50 MW cm^{-2} : $\phi = (1.5–3) \times 10^{-4}$ (curve 1 in Fig. 4), the minimum quantum yield corresponding to the maximum lasing efficiency (curve 1 in Fig. 5). The same solution lases in version 2 as well, but the lasing efficiency at $W = 170 \text{ MW cm}^{-2}$

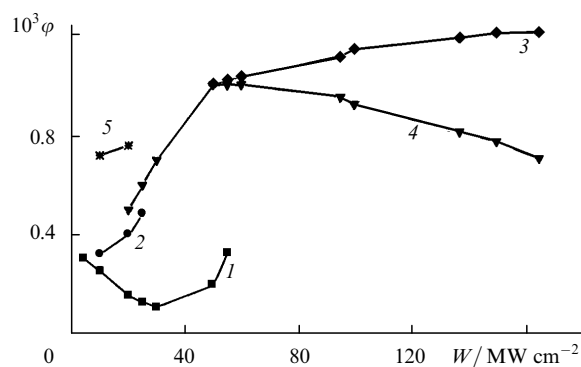


Figure 4. Dependences of the quantum yield of phototransformations ϕ on the pump radiation intensity for versions 1 (1, 2) and 2 (3–5) in the presence (1–3) and absence of lasing (4, 5) for $C = 1$ (1, 3, 5) and $0.1 \text{ mmol litre}^{-1}$ (2, 4).

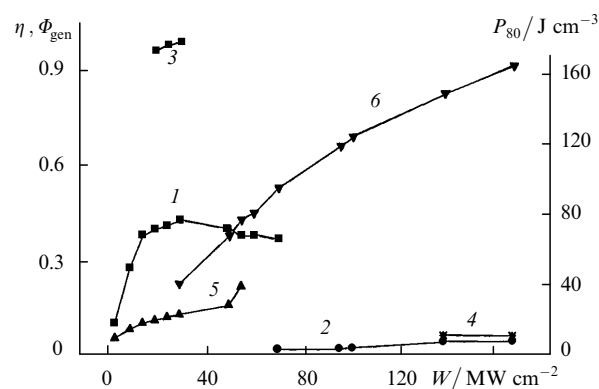


Figure 5. Dependences of the efficiency η (1, 2), the resource for operation P_{80} (3, 4) and the quantum yield of the first photoproduct ϕ_{gen} (5, 6) on the pump radiation intensity at $C = 1 \text{ mmol litre}^{-1}$ for versions 1 (1, 3, 5) and 2 (2, 4, 6).

is 4.5%, and it decreases with decreasing pump intensity (curve 2 in Fig. 5). The threshold intensity is 35–40 MW cm⁻². The quantum yield of phototransformations in the region $W = 40 - 170 \text{ MW cm}^{-2}$ is $\varphi = (1 - 1.2) \times 10^{-3}$.

As for the first photoproduct, its relative yield Φ_{gen} also depends on W . As the pump intensity was increased, we obtained $\Phi_{\text{gen}} \approx 0.05 - 0.2$ in version 1 (curve 5 in Fig. 5) and an increase in Φ_{gen} from 0.1 to 0.9 in version 2 (curve 6 in Fig. 5). The change in the operational life of the LOS-1 laser medium corresponds to the change in Φ_{gen} . We obtained $P_{80} = 175 - 180 \text{ J cm}^{-3}$ at $W = 20 - 30 \text{ MW cm}^{-2}$ for version 1 and $P_{80} = 11 \text{ J cm}^{-3}$ at $W = 160 \text{ MW cm}^{-2}$ for version 2.

Analysis of the curves in Fig. 4 shows that the phototransformation efficiency increased with increasing W (with the exception of curve 4 in Fig. 4). However, the quantum yields of phototransformations of the same solution at a fixed value of W depend substantially on the emission conditions. An increase in the phototransformation efficiency and its subsequent decrease in the absence of lasing ($C = 0.1 \text{ mmol litre}^{-1}$) with increasing W in version 2 (curve 4 in Fig. 4) are caused by changes in the population of the excited state S_1 : the increase in the population at $W \leq 50 \text{ MW cm}^{-2}$ in the case of spontaneous emission and its decrease at $W = 50 - 180 \text{ MW cm}^{-2}$ at the initial stage of superluminescence formation under these excitation conditions.

3.6 Gain for high-power excitation

The dependences of the gain of molecules on the length of the active region are presented in Fig. 6. For all the compounds studied here, we initially observed an increase in the measured gain K_0 with increasing L up to a certain length L_1 ; the increase was then stopped and K_0 remained constant. This may be explained by the increase in loss with decreasing L . A decrease in length of the active region, with the resonator length being fixed, leads to an increase in loss through out-coupling of radiation through the lateral cavity surfaces, which represents one of the components of intrinsic loss. Because this loss is proportional to $1/L$ its value at sufficiently large L is small and changes only weakly with increasing L . Therefore, in the range of lengths of the active region where K_0 is unchanged, the intrinsic loss may be considered to be small in comparison with $K_{\text{am}}^{(0)}$, and one may use K_0 as an intramolecular characteristic of amplification properties of an

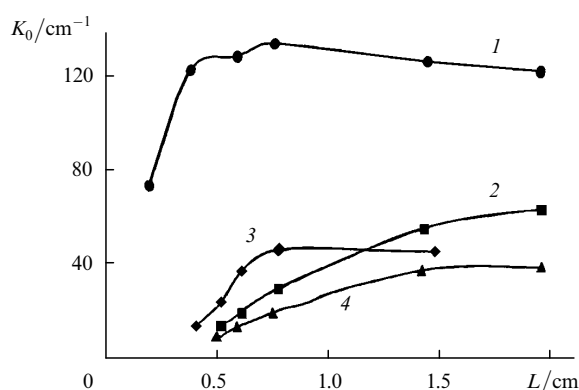


Figure 6. Dependences of the gain K_0 on the active-region length L for ethanol solutions of LOS-1 ($C = 0.5 \text{ mmol l}^{-1}$, 1), 4PyPO ($C = 0.5 \text{ mmol l}^{-1}$, 2), 4PyPOOCH₃ ($C = 2 \text{ mmol l}^{-1}$, 3), and 4PyPON(CH₃)₂ ($C = 0.5 \text{ mmol l}^{-1}$, 4).

active medium. These values of K_0 at $W = 20 \text{ MW cm}^{-2}$ are presented in the table, which shows that the changes in the experimentally measured gain correspond to changes in the efficiency. In the particular case of the LOS-1 solution, which has the maximum efficiency, the measured value of K_0 also has a maximum, which is equal to 125 cm^{-1} .

Note that the limiting gain of LOS-1, which is equal to $n\sigma_{10}$, is very close to the measured value of K_0 , which suggests that the laser level is highly populated ($n_1/n \approx 0.71$) and the intramolecular loss is negligible. For 4PyPO, the limiting gain and the experimentally measured K_0 value differ by a factor of about two. For 4PyPOOCH₃ and 4PyPON(CH₃)₂, the difference is even greater. This difference agrees with a decrease in the lasing efficiency and is caused by considerable intramolecular loss. The loss may be caused either by transitions of molecules to the triplet state or by a change in the geometry of molecules in the excited state, as was shown in Ref. [7] for 4PyPON(CH₃)₂.

Fig. 7 presents the dependences of K_0 and the quantum efficiency on the pump intensity W for LOS-1. One can see that as W is increased up to $\sim 20 \text{ MW cm}^{-2}$, K_0 and the efficiency increase due to an increase in inversion. As W was further increased, K_0 remained almost unchanged, and the efficiency at $W > 30 \text{ MW cm}^{-2}$ decreased slightly. The latter effect may be caused by several reasons, such as the thermal lens formation, pump radiation defocusing, spatial grating formation, etc. The explanation of the effect of these and other mechanisms calls for further studies.

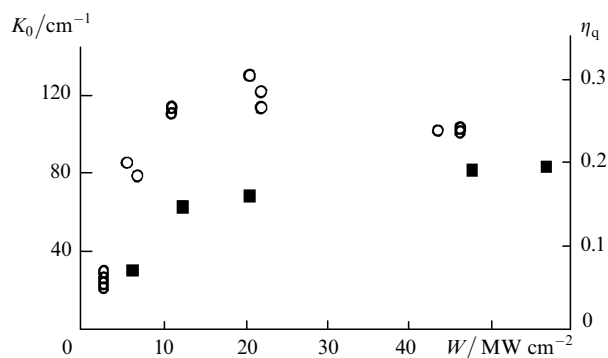


Figure 7. Dependences of K_0 (■) and the quantum efficiency η_q (○) for LOS-1 with $C = 0.37 \text{ mmol l}^{-1}$ on the pump intensity.

Thus the specific features observed in the spectral and temporal characteristics of the emission of organic compounds (the shift of the emission spectrum, a change in its half-width, and a change in the pulse shape) upon excitation by high-power XeCl laser radiation are related to the formation of superluminescence. The best photostability of LOS-1 corresponds to optimum lasing, and it deteriorates under conditions of spontaneous emission and low-efficiency lasing. This suggests that the method proposed here can be used to estimate the gain, taking into account the restrictions imposed on the length of the active region which were mentioned above. The role of intrinsic loss in the determination of gain calls for further studies.

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