Photonics of complex molecules under conditions of nonlinear absorption at high-power laser excitation.
Part 1. Peculiarities of spontaneous and stimulated molecular emission in strong light fields

T.N. Kopylova, R.T. Kuznetsova, G.V. Maier, V.A. Svetlichnyi, E.N. Tel'minov, and D.N. Filinov

V.D. Kuznetsov Siberian Physical-Technical Institute at the Tomsk State University

Emission from concentrated (10^{18}–10^{19} cm^{-3}) solutions of complex organic compounds (rhodamine 6G, substituted paraterphenyl) in strong light fields of XeCl laser radiation (power density up to $5 \cdot 10^{26}$ phot/s·cm^2) is studied. Under our experimental conditions, four types of emission were observed; three of them are interpreted unambiguously (fluorescence, lasing, lasing on a thermal lens), while the nature of the fourth type is still unclear. This type of emission ("strip") is presumably interpreted as amplified spontaneous emission, but it also may be collective emission of an ensemble of excited molecules. Further investigations aimed at elucidating the nature of this emission are planned.

It is known that optical properties of molecules may change in strong light fields, thus giving rise to some phenomena, whose nature is complicated and still not fully understood.[1–3] With the advent of lasers and their further intense development, fine physical experiments became possible, and their results had a significant effect on the interpretation of interaction of resonance optical radiation with atoms and molecules. In recent years, series of works by Russian scientists well known in this field (Shilov, Klochkov with colleagues) have been done. Since publication of the first papers by Klochkov with co-authors[5] (1988), the nature of phenomena observed at interaction of high-power laser radiation with organic molecules has been widely discussed in the literature, and the number of experimental data on the peculiarities of this interaction has increased continuously. However, there are a number of problems that are still unclear and whose solution require new experiments with the use of exciting laser beams having different parameters, as well as investigations into the effect of peculiarities of spontaneous emission of organic molecules (laser dyes) in strong light fields on the development of their stimulated emission.

Let us say some words on the essence of the observed phenomena. In 1988 Klochkov and Verkhovskii[5] discovered the anomalous growth of molecular fluorescence under conditions of absorption saturation when studying spontaneous emission (fluorescence) of organic molecules (rhodamines, cyanins, and other laser active media) under the exposure to second-harmonic Nd:YAG laser radiation. It was found that the dependence has a threshold character, and the threshold power density depends on the properties of a medium, concentration of the solution, and conditions of formation of the exciting

laser beam (focused–unfocused). The threshold level of the power density increases considerably in the case of excitation by unfocused radiation. Based on the characteristics of the observed phenomenon, called the incoherent superfluorescence, Klochkov assumed that it is connected with the increase of the probability of spontaneous molecular emission with the increasing power density of the exciting radiation as a result of four-wave mixing of the two pumping waves, the wave of the fluctuating field of vacuum, and the wave conjugate with the latter. The possibility of the increase in spontaneous emission at interaction of the vacuum field with the pump radiation was predicted by Milonni with co-authors[20] and by Ho Send-Tiong with co-authors[21] in the 80s.

The observed phenomena have been widely discussed by scientists. In some papers (Aristov, Stoilov[22–24]), the results obtained are called in question; other interpretation is given to them, etc. This only underlines the complexity of the experiments and the urgency of the research.

In 1996, Shilov with co-authors[13] has found the significant discrepancy between the experimental curves of the optical clearing up of organic molecules at high pump intensity and the theoretical calculations following from the classical concept of the interaction of the laser field with the extended multilevel systems, i.e., they also observed anomalously high fluorescence of organic molecules in strong light fields. However, they believed that the cause of this phenomenon was collective interactions of molecules in the strong resonance field, rather than the increased probability of spontaneous molecular emission (in their opinion, this interpretation needs for fundamental justification). Collective interactions may change the dynamics of population and fluorescence. If they include zero-point oscillations of vacuum and only in the case, if these oscillations

have a finite length of spatial coherence, the glow is considered within the framework of four-wave collective interaction with the participation of two oscillations of vacuum (the first theoretical paper devoted to this phenomenon was published at the same time by Fain[3]).

In recent papers by Klochkov and Verkhovskii,[17,19] the glow of solutions of rhodamine 6G and rhodamine B at high concentration ($C > 10^{18}$ cm^{-3}) exposed to high-power laser radiation (power density of $10^{25} - 10^{26}$ cm^{-2} s^{-1}) was studied.

Depending on the concentration of solutions and the radiation power density, several types of glow were observed: collective spontaneous emission, amplified spontaneous emission, and lasing.

The intensity of directed collective glow changes as a square of the power density of the exciting radiation and is interpreted by the authors as a collective spontaneous emission of the Dicke type. Its basic feature is the need for a very short time for the correlation establishment and very short pulse delay, because of the high cross relaxation rate of the dye molecules ($\sim 10^{12} - 10^{13}$ s^{-1}). This requirement can be fulfilled at high concentrations (C) of the excited molecules. At $C = 10^{17} - 10^{19}$ cm^{-3} the correlation can be established in $\sim 5 \cdot 10^{-14} - 5 \cdot 10^{-15}$ s; this time is much shorter than the time of dephasing due to the cross relaxation in molecules.

The observed types of emission are spectrally separated, and the intensity ratio between their bands changes depending on the above parameters. In our opinion, these types of emission occur not simultaneously. The collective glow has the shortest emission time, separate pulses of this glow in rhodamine 6G are emitted for the time $< 10^{-13}$ s, while the process of collective emission continues during the entire exciting pulse − a group of femtosecond pulses is emitted. The collective emission decreasing the concentration of excited molecules prevents the development of amplified spontaneous emission and lasing. Appearance of lasing with the decreasing concentration of the solution is explained by the longer time of development of the collective emission, the lower concentration of in-phase emitters because of vibrational relaxation, and the higher concentration of excited molecules, which are out-phase emitters. The excited molecules (out-phase emitters) are responsible for amplified spontaneous emission and lasing.

Shilov and his colleagues continue the study of anomalous fluorescence of molecules in the strong laser pump fields. Thus, in Refs. 13 and 15, they studied anomalous light-induced fluorescence of solutions of multiatomic molecules under the exposure to high-intensity laser radiation of subnano- and picosecond duration. The effect of fluorescence condensation in the pump field was observed for the first time. This effect manifests itself in that the kinetics of the fluorescence approach the kinetics of the pump pulse and in the increase of the quantum yield with the growing excitation power density. In developing their previous ideas, they put forward the hypothesis on the mechanism of the observed phenomena − four-wave collective interaction in the system "ensemble of optical centers − pump field" with the participation of zero-point oscillations of vacuum with their partial local coherence.

The understanding of the causes of these phenomena is needed not only from the standpoint of obtaining basic knowledge, but also from the standpoint of optimizing power parameters of lasers operating at organic compounds under high-power excitation. Thus, for example, it was found that the efficiency of radiation conversion by laser active medium decreases, as the pump power increases.[23] The cause of this process is too complicated being determined by the nonlinear optical processes characterizing mostly the experimental conditions (superluminescence in the near-wall layer, luminescence excited by pumping radiation scattered in the cell volume, irreversible worsening of the transmission of the cell windows, nonlinear scattering of different nature) and by changes in the intramolecular properties of the molecule itself (incoherent superluminescence, induced absorption and reversible photochemical processes [Batishche, Aristov],[25,22,23] two-stage transitions caused by absorption of the pump radiation in the channel of excited singlet states [Tikhonov][26]). The probability of photochemical transformations in the laser active medium under the action of strong light fields changes as well. Thus, it was found[29] that the quantum yield of photodecomposition of a molecule at lasing is lower than that under conditions of spontaneous emission, the parameters of the pumping radiation being the same.

Klochkov and Verkhovskii[11] showed that the threshold power of dye lasers decreases if the power exceeds the excitation threshold of incoherent superfluorescence. This is connected with the fact that the increase in the rate of spontaneous emission not only decreases the population of the excited state (thus leading to the increase of the lasing power threshold), but also decreases the time for which the threshold power is achieved.

Recent paper by Shilov in co-authorship with Klochkov[18] shows that the collective interactions in optical ensembles affect the lasing threshold characteristics of organic molecules at coherent optical pumping. They have found that, as the quantum yield of fluorescence decreases, the pumping threshold increases slower as compared with the predictions by the classic theory of lasers. This agrees with the concept of collective processes developing in concentrated media at intense coherent excitation.

Thus, we can see the direct relation between the luminescence characteristics of organic molecules at high-power excitation and the lasing properties of these molecules determining the optimal parameters of lasers operating at them.

We began to study the peculiarities of emission of organic molecules in strong light fields of excimer laser pumping radiation in 1995 (Refs. 27−30).

Table 1. Spectral-luminescence and lasing properties of the studied compounds

Compound	Compound	λ_{ab}^{max}, nm	λ_{fl}^{max}, nm	$\sigma_{01}^{308} \cdot 10^{-16}$, cm^2	$\sigma_{10} \cdot 10^{-16}$, cm^2	$A_{10} \cdot 10^8$, s^{-1}	η_{fl}	Quantum efficiency of lasing, %
Rhodamine 6G	Ethanol	532	560	0.53	13.8	2.7	0.75	36
Para-terphenyl	Ethanol	280	340	0.28	4.6	9.9	0.92	27.5
LOS-1	Ethanol	309	373	1.67	5.7	8.2	0.96	51
4PyPO	Ethanol	323	395	0.76	3.5	4.7	0.9	20.7
4PyPON(CH$_3$)$_2$	Ethanol	382	560	0.32	4.2	2.5	0.27	27.8
	Hexane	365	417	0.95	2.5	2.8	0.8	3.3
Coumarin-2	Ethanol	370	440	0.1	2.2	1.9	0.84	39
Trans-stilbene	Ethanol	309	348	1	2.8	5.27	0.05	5
Biphosphor (T-stilbene-CH$_2$-K120)	Ethanol	370	440	1.1	2.2	1.9		39

Object of the study: organic molecules having different structure and spectral-luminescence properties (Table 1).
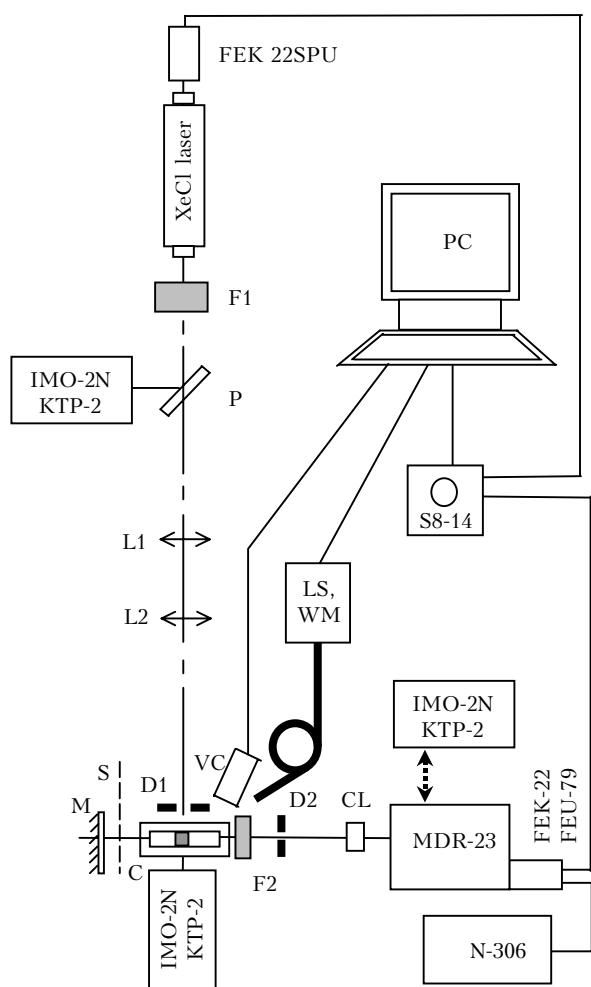

Fig. 1. Experimental setup: S8-14 oscilloscope, beam-splitting plate P, filters F1 and F2, lenses L1 and L2, screen S, mirror M, cell C with the solution under study, diaphragms D1 and D2, collimator CL, FEK−22SPU and FEU−79 photodetectors, IMO−2N and KTP−2 calorimeters, MDR−23 monochromator, N−306 plotter, video camera VC for measuring the size of the excited volume, laser spectrometer LS, wavelength meter WM, personal computer PC.

As is seen, the quantum yield of molecular fluorescence ranges from 0.05 (trans-stilbene) to 0.96 (LOS-1). The difference in the spectral-luminescence and lasing characteristics of the molecules under study is caused by different photochemical processes proceeding in these molecules. Therefore, it is interesting to study the effect of high-power laser radiation on these processes.

Excitation source: XeCl* excimer laser (λ_{las} = = 308 nm, E_{las} up to 50 mJ, $\tau_{1/2}$ = 10 ns, beam size of 21×8 mm). The spatial, power, and temporal characteristics of this laser differ widely from those of the Nd:YAG laser used by Klochkov and Shilov[4−19] (thus, its spatial coherence, according to our estimates, is 12 μm at visible interference pattern).

Characteristics to be studied: transmission, temporal, power, and spectral characteristics of radiation in light fields of the exciting radiation from 1 to 350 MW/cm^2 (10^{23}–10^{27} phot/(cm$^2 \cdot$s)).

The experimental setup shown in Fig. 1 is described in detail in Refs. 27 and 28. Here let us only note that we used this setup to study different types of emission: spontaneous emission, superfluorescence, and lasing.

Among the obtained scientific results, we would like to note the following:

1. The dependence of the luminescence intensity of ethanol solutions of 4PyPO that emit rather intensely in the blue spectral region on the power density of the pumping XeCl* laser radiation (wavelength of 308 nm, pulse duration at halfmaximum of 10 ns, P = = 10^{23}–10^{27} phot/(cm$^2 \cdot$s)). We observed the anomalous growth of luminescence at high-intensity pumping, as was reported by Klochkov. However, it may be caused, in our opinion, by the increasing role of stimulated emission.[27]

2. The intensity, pulse duration, and emission spectra of trans-stilbene and 4PyPO were studied under the exposure to XeCl* laser radiation with the power density from 10^{23} to 10^{26} phot/(cm$^2 \cdot$s). We have found anomalous growth of the intensity in the ethanol solutions of 4PyPO, trans-stilbene, photocation form of 4PyPO in aqueous solution, as well as the shortwave (<10 nm) spectral shift, as the intensity of the pumping radiation increased. The obtained experimental data

suggested that the anomalous character of the observed emission might be connected with both the stimulated emission and collective interactions of molecules.[28]

3. The emitting and photochemical properties of laser dyes [4PyPO, its dimethyl derivative, and para-terphenyl (LOS-1)] excited by focused XeCl* laser radiation with the power density up to 200 MW/cm^2 have been studied. The technique for measuring the gain factor of organic molecules under high-power excitation has been proposed. The transmission of the studied molecules for the pumping radiation has been investigated as a function of power density of the pumping radiation.

It has been shown that peculiarities of spectral and temporal emission characteristics of the studied organic compounds (spectral shift and change of the halfwidth of the emission spectrum, change of the pulse shape) excited by high-power XeCl laser radiation are connected with the formation of amplified spontaneous emission – superluminescence.[29]

4. The spectral-luminescence properties of eight organic dyes, including rhodamine 6G, exposed to focused XeCl laser radiation with the power density up to 200 MW/cm^2 have been studied. The dye concentration varied from 5 to 1000 μmol/l depending on the efficiency of absorption of pumping radiation and fluorescence intensity at laser excitation.

It was shown that the peculiarities of emission of the molecules studied in a strong light field of the XeCl* laser under conditions of our experiments were connected with the formation of superluminescence.[30]

Thus, the observed peculiarities of molecular emission in strong light fields can hardly be interpreted unambiguously; different types of emission may compete with each other.

The paper by Klochkov and Verkhovskii[17] and their observations of the collective spontaneous molecular emission stimulated our study of the concentrated solutions of some organic compounds (systems with high dephasing rate,[19] which include laser dyes) at high-power excitation by XeCl laser radiation.

References 2 and 32 consider the cooperative emission in the case of extended volumes $V \gg \lambda^3$, when the time of action of the pumping radiation exceeds the time of cross relaxation. In this case, the characteristics of cooperative emission[3] may differ markedly from those of the Dicke emission.[1] Along with the cooperative emission, other types of emission can develop simultaneously in the medium (competing with each other). They are spontaneous emission, multi-pass lasing, amplified spontaneous emission (superfluorescence).

As the objects of our study, we took rhodamine 6G (studied by Klochkov with colleagues), whose concentration varied from $5 \cdot 10^{-3}$ mol/l ($3 \cdot 10^{18}$ cm^{-3}) to 10^{-1} mol/l ($6 \cdot 10^{19}$ cm^{-3}), and phenalemine 512 with the concentration from 10^{-2} mol/l ($6 \cdot 10^{18}$ cm^{-3}) to $5 \cdot 10^{-3}$ mol/l ($3 \cdot 10^{18}$ cm^{-3}). They are characterized by strong reabsorption of radiation. For LOS-1

reabsorption is absent, but its solubility in ethanol is somewhat lower, and the maximum concentration of LOS-1 was $5 \cdot 10^{-3}$ mol/l ($3 \cdot 10^{18}$ cm^{-3}).

The corresponding experiments were conducted with the same setup (Fig. 1). The solution was excited through a round diaphragm 0.5 mm in diameter, and the excited volume in this case was a cylinder with the height equal to the penetration depth of the pumping radiation into the solution and the diameter of 0.5 mm. The radiation was recorded in all directions except for the frontal one. The spatial distribution of radiation was observed on a screen, and its characteristics were recorded with a laser spectrometer through an optical waveguide of 0.4 mm in diameter.

For the compounds with strong reabsorption (rhodamine 6G and phenalemine 512), four types of emission were observed:

1. Spontaneous emission. At frontal excitation, it is ordinary fluorescence; the radiation is undirected with a wide spectrum ($\lambda_{max} = 570$ nm) coincident with the spectrum of fluorescence recorded with a Hitachi–850 at low-intensity excitation. If recording is normal to pimping, spontaneous emission is strongly reabsorbed, and the shift is 30 nm. At the rhodamine 6G concentration $C = 10^{-1}$ mol/l, fluorescence decreases, and not emitting dimers are observed.

2. Lasing. It occurs in a low-Q cavity formed by cell faces in the direction normal to pumping. Lasing is characterized by a high threshold ($20-100$ MW/cm^2) due to high loss in the low-Q cavity, diffraction loss because of absorption in a thin layer, and loss due to reabsorption (at the concentration of $5 \cdot 10^{-2}$ mol/l and 10^{-1} mol/l, lasing occurs only in the cavity with the base length $L = 0.2$ cm and is absent at $L = 1$ cm), and low efficiency ($\approx 1\%$).

3. "Strip." It is the radiation propagating from a circle in all directions in the plane perpendicular to the direction of excitation. On a screen, it is observed as a narrow vertical strip. The emission spectrum is wider than the lasing spectrum (for rhodamine 6G $\Delta\lambda = 10-20$ nm). The maximum of the strip emission spectrum does not coincide with the maximum of the lasing spectrum (it is shifted to the shortwave region; for example, at the rhodamine 6G concentration of 10^{-2} mol/l the strip maximum is at 573 nm, whereas the maximum of the lasing spectrum falls on 596 nm). If the radiation in the strip is recorded at a small angle to the laser beam ($\alpha = 10-15°$), then one can observe the pattern shown in Fig. 2. The emission spectrum consists of two bands. If a bushing is placed in the cell, the longwave band corresponding to scattered lasing disappears. Thus, spectrally, the strip radiation for rhodamine 6G coincides with the band of the collective emission observed by Klochkov.[19] However, we are not sure that the strip radiation we observed is of the same nature. We plan to study the dependence of the radiation intensity (I) in the strip on the power density of the exciting radiation (W) and on the concentration of the excited particles (N). Only after analysis of the

dependence $I = f(N)$, we can draw some conclusions on the nature of this emission.
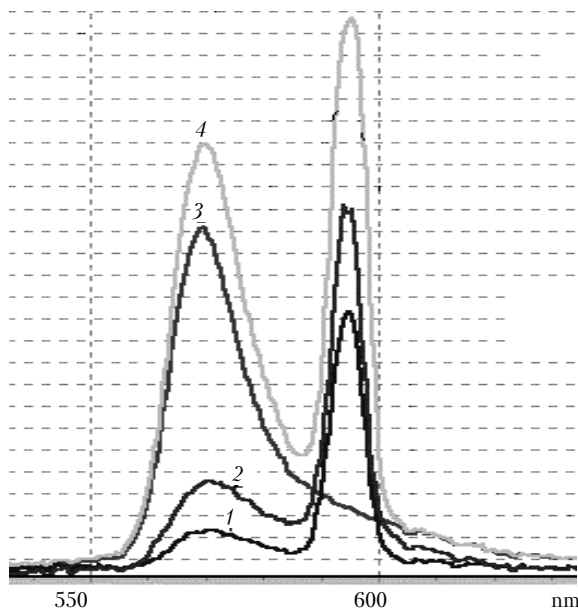

Fig. 2. Strip emission spectra: $W = 50$ (*1*), 120 (*2*), and 250 MW/cm^2 (*3, 4*); without bushing (*1, 2, 4*) and with bushing (*3*).

The observed deviations from the linear dependence of the strip radiation intensity on the power density of the exciting radiation for LOS-1 at $C = 5 \cdot 10^{-3}$ mol/l (maximum solubility) are most likely connected with the formation of the amplified spontaneous emission competing with the collective emission,[17,19] because at $C < 3 \cdot 10^{18}$ ($5 \cdot 10^{-3}$ mol/l) the processes of formation of amplified spontaneous emission and lasing prevail.

However, it should be emphasized that the parameters of the exciting XeCl laser radiation strongly differ from those of the Nd:YAG laser radiation, for example, by its coherence. This undoubtedly affects the characteristics of collective emission, for example, the concentration of organic molecules in solutions. Now we believe that the most probable cause for appearance of this emission is spontaneous emission (superfluorescence) amplified during a single pass. This emission is formed within the excited zone, in which reabsorption is much lower than in the unexcited areas of the cell; therefore, its band shifts to the shortwave region as compared to lasing, which is formed at multiple passes.

From the viewpoint of interpreting the nature of the strip emission, the results obtained for LOS-1 are of interest (Fig. 3*b*). The strip emission spectrum consists of a 20–25-nm wide base and a narrow band (3–4 nm) at the center, and the band is observed only at large angles with respect to the laser beam. The limiting gain factor for LOS-1 in the given concentration is higher than 10^3 cm^{-1}, the lifetime of the fluorescence state is ~1 ns (for rhodamine 6G it is 3–5 ns), and the concentration quenching of fluorescence is not observed; therefore, narrow-band emission (superfluorescence) is formed during a single pass. As is seen from Fig. 3*b*, with the increase in the intensity, superfluorescence is formed smoothly, while the formation of lasing is of a threshold character (Fig. 3*a*).
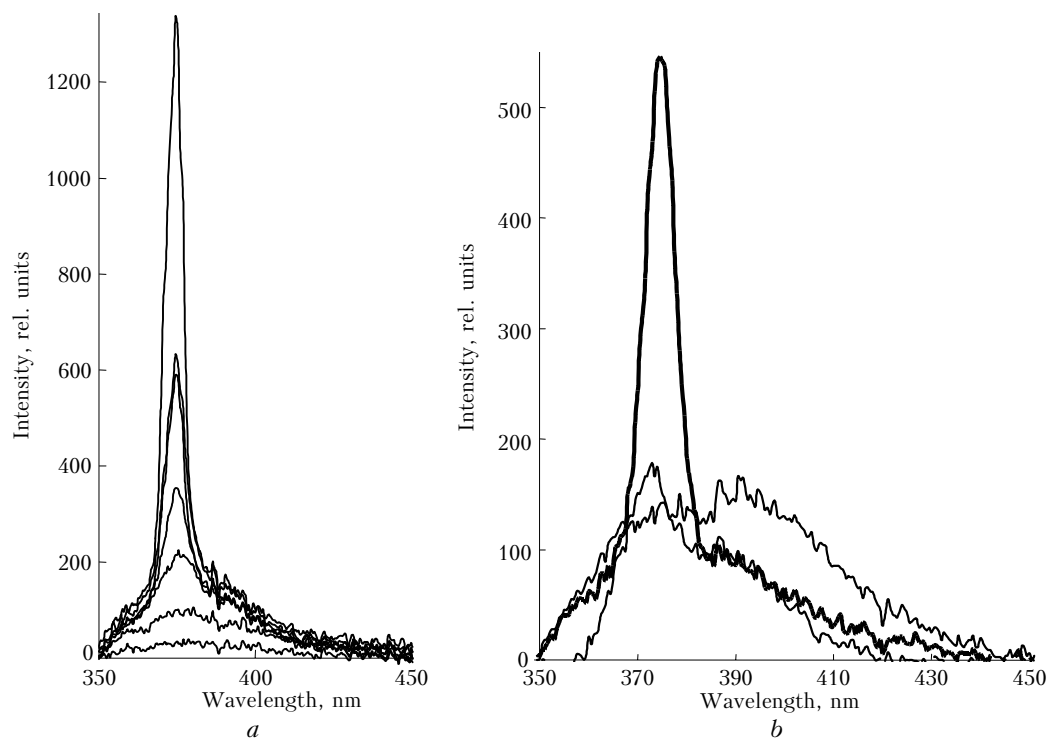

Fig. 3. Formation of lasing (*a*) and strip (*b*) in LOS-1. The parameter W changes by no more than 20% (*a*) and two times (*b*).

It seems to be interesting to give the following estimate: the penetration depth for the pumping radiation in the solution was calculated from the width of the superfluorescence band by the technique proposed by Klochkov in Ref. 19, and the dimensions of the excited zone were estimated. According to these estimates, at the pump power of 200 MW/cm^2 every molecule in the excited volume absorbs no less than 10^4 photons during the pump pulse. Thus, the absorption-relaxation cycle lasts ~1 ps. It is much shorter than the lifetime of the fluorescent state, but this time is more typical of the amplified emission than of the cooperative processes. At the pump power density of 1–30 MW/cm^2, the intensity of LOS-1 emission in the strip nonlinearly depends on the pump power density. However, we believe that at such pump power density the nonlinear dependence can be connected with the formation of superfluorescence.

4. "Conical" emission. It is the directed radiation propagating along the directrix of a cone with the vertex at the excitation point. The spectrum consists of two bands. Thus, for rhodamine 6G the conic emission is observed at the concentration of $5 \cdot 10^{-2}$ mol/l (as this takes place, no lasing is observed in the cavity with the base length $L = 1$ cm), and the peaks of the emission band are at 588 and 600 nm.

The possible cause for this emission is, in our opinion, induction of the gradient of the refractive index at the interface between the excited and unexcited volumes, as well as formation of a "fast" thermal lens at nanosecond pumping.[26,31] These two processes may lead to appearance of an optical system forming the amplified stimulated emission in this direction. This point of view is confirmed by the fact that the conical emission was not observed for LOS-1, because for this to take place, both the threshold and the time needed for induction of the efficiently scattering thermal lens must be much larger.

Thus, for concentrated solutions of rhodamine 6G in ethanol at high-power excitation by the XeCl laser radiation, the emission of unknown nature is observed. Based on the observed characteristics (band narrowing, pulse shortening), we presumably classify this emission as the amplified spontaneous emission. To confirm the nature of strip emission, the dependence of its intensity on the concentration of molecules is to be studied.

Acknowledgments

References

1. R.H. Dicke, Phys. Rev. **93**, No.1, 99–110 (1954).
2. A.V. Andreev, V.I. Emel'yanov, and Yu.A. Il'inskii, *Cooperative Phenomena in Optics* (Nauka, Moscow, 1988), 180 pp.
3. B. Fain, SPIE **2798**, 9–20 (1996).
4. A.G. Makogonenko and V.P. Klochkov, Opt. Spektrosk. **64**, No. 2, 244–246 (1988).
5. V.P. Klochkov and E.B. Verkhovskii, Opt. Spektrosk. **65**, No. 1, 237–240 (1988).
6. V.P. Klochkov, E.B. Verkhovskii, and V.L. Bogdanov, Opt. Spektrosk. **70**, No. 3, 547–551 (1991).
7. A.G. Makogonenko, I.N. Myakisheva, V.A. Smirnov, and V.B. Shilov, Opt. Spektrosk. **70**, No. 4, 795–800 (1991).
8. A.G. Makogonenko and V.P. Klochkov, Opt. Spektrosk. **70**, No. 2, 439–455 (1991).
9. V.P. Klochkov, Opt. Spektrosk. **74**, No. 4, 676–694 (1993).
10. V.P. Klochkov and E.B. Verkhovskii, Opt. Spektrosk. **75**, No. 6, 1183–1187 (1993).
11. V.P. Klochkov and E.B. Verkhovskii, Opt. Spektrosk. **77**, No. 3, 394–397 (1994).
12. V.P. Klochkov and E.B. Verkhovskii, Opt. Spektrosk. **81**, No. 4, 613–615 (1996).
13. V.B. Shilov, V.A. Smirnova, E.G. Korsakova, et al., Opt. Spektrosk. **81**, No. 5, 767–773 (1996).
14. V.P. Klochkov and E.B. Verkhovskii, Opt. Spektrosk. **85**, No. 3, 427–433 (1998).
15. G.M. Ermolaeva, E.G. Gregg, V.A. Smirnov, and V.B. Shilov, Opt. Spektrosk. **84**, No. 3, 393–397 (1998).
16. N.V. Vysotina, E.G. Gregg, G.M. Ermolaeva, V.V. Kuznetsov, V.N. Kulyasov, V.A. Smirnov, and V.B. Shilov, Opt. Spektrosk. **86**, No. 4, 598–603 (1999).
17. V.P. Klochkov and E.B. Verkhovskii, Opt. Spektrosk. **88**, No. 5, 768–771 (2000).
18. G.M. Ermolaeva, V.P. Klochkov, G.E. Nikolaev, V.A. Svetlichnyi, V.A. Smirnov, and V.B. Shilov, Opt. Spektrosk. **89**, No. 1, 157–159 (2000).
19. V.P. Klochkov and E.B. Verkhovskii, Opt. Spektrosk. **89**, No. 1, 114–118 (2000).
20. P.W. Milonni, E.J. Bochove, and R.J. Cook, J. Opt. Soc. Am. B **6,** No. 10, 1932–1941 (1989).
21. Ho Send-Tiong, P. Kumar, J.H. Shapiro, J. Opt. Soc. Am. B. **1**, No. 1, 37–57 (1991).
22. A.V. Aristov, D.A. Kozlovskii, and A.B. Nikolaev, Opt. Spektrosk. **67**, No. 2, 327–331 (1989).
23. A.V. Aristov, D.A. Kozlovskii, and A.B. Nikolaev, Opt. Spektrosk. **68**, No. 2, 335–338 (1990).
24. A.Yu. Belonogov, A.V. Startsev, Yu.Yu. Stoilov, and Cho San-Dzyu, Kvant. Elektron. **23**, No. 6, 571–573 (1996).
25. S.A. Batishche, V.A. Ganzha, N.A. Malevich, et al., Zh. Prikl. Spektrosk. **39**, No. 6, 934–938 (1983).
26. V.I. Bezrodnyi, O.V. Przhonskaya, E.A. Tikhonov, and M.T. Shpak, Zh. Eksp. Teor. Fiz. **80**, No. 2, 512–523 (1981).
27. T.N. Kopylova, R.T. Kuznetsova, E.N. Tel'minov, K.M. Degtyarenko, V.A. Svetlichnyi, and G.V. Maier, Izv. Vyssh. Uchebn. Zaved., Ser. Fizika, No. 4, 69–74 (1997).
28. T.N. Kopylova, R.T. Kuznetsova, V.A. Svetlichnyi, E.N. Tel'minov, and L.G. Samsonova, Opt. Spektrosk. **85**, No. 5, 778–782 (1998).
29. T.N. Kopylova, R.T. Kuznetsova, V.A. Svetlichnyi, A.K. Sergeev, E.N. Tel'minov, and D.N. Filinov, Kvant. Elektron. **30**, No. 6, 489–494 (2000).
30. R.T. Kuznetsova, V.A. Svetlichnyi, T.N. Kopylova, and E.N. Tel'minov, Opt. Spektrosk. **89**, No. 2, 261–268 (2000).
31. E. Berik and V. Davydenko, Izvestiya Akad. Nauk Estonii, Ser. Fiz. Matem. **39**, No. 1, 46–51 (1990).
32. L.I. Men'shikov, Usp. Fiz. Nauk **169**, No. 2, 113–154 (1999).