

Anomalous dependence of the lasing parameters of dye solutions on the spectrum of microsecond pump laser pulses

V.V. Tarkovsky, V.Yu. Kurstak, S.S. Anufrik

Abstract. The anomalous dependence of the lasing parameters of ethanol solutions of coumarin, rhodamine, oxazine, and laser dyes of other classes on the spectrum of microsecond pump laser pulses is found. The dependence is determined by the shape of the induced singlet–singlet absorption spectra and absorption spectra of short-lived photoproducts. The elucidation of the influence of these factors makes it possible to choose optimal pump spectra and to enhance the efficiency and stability of microsecond dye lasers.

Keywords: converter dye laser, induced absorption, singlet–singlet absorption, triplet–triplet absorption, photoproducts.

1. Introduction

At present different pulsed lasers are used to pump dye solutions. The most popular are the N_2 , XeCl, KrF, XeF, copper vapour lasers, as well as Nd^{3+} :YAG, Cr^{3+} : Al_2O_3 , and Ti^{3+} : Al_2O_3 lasers. Among gas lasers, excimer lasers (using rare gas halides) are most promising for this purpose. These lasers are very convenient to pump dyes because they emit UV radiation in the region of UV absorption bands of most dyes [1]. The disadvantage of excimer laser pump is that not all the dyes are sufficiently photostable upon UV irradiation.

Harmonics of a Nd^{3+} :YAG laser provide higher peak and average powers than a nitrogen laser. Due to a good quality of a beam produced by a Nd^{3+} :YAG laser, radiation from this laser can be efficiently mixed with radiation of dye lasers to obtain tunable lasing in a broad spectral range from 217 to 5000 nm [2]. However, to produce lasing in a broad spectral range, a number of costly frequency converters should be used, which are sensitive to external perturbations; this poses great difficulties in the manufacturing and use of such systems.

An advantage of a copper vapour laser as a pump source for dye lasers is a very high pulse repetition rate (~ 10 kHz) and a high average power (~ 100 W) [3]. Its disadvantage is

lasing at long wavelengths, which restricts the short-wavelength boundary of dye laser tuning by the green region.

Note that the above-mentioned lasers, which are used to pump dye solutions, emit at discrete, strongly separated lines [4]. That is why the authors of paper [4] did not study the dependence of the lasing efficiency of dye solutions on the pump wavelength. Pump sources available at present make it possible to study this dependence in more detail.

Recent studies have revealed the anomalous dependence of the lasing efficiency of some dyes on the wavelength of nanosecond pump pulses. It was assumed earlier that the maximum lasing efficiency is achieved by pumping into the maximum of the most intense absorption band of a dye [5, 6]. It was shown in Refs [7–10] that the lasing efficiency increased when the pump wavelength was continuously tuned over the long-wavelength wing of the absorption spectrum, reached a maximum and then slowly decreased. It was found that this effect was observed in high- Q resonators at high pump intensities and high dye concentrations in solutions. However, the mechanism of this effect remains unclear, and it is unknown whether a similar dependence is inherent in dyes of other classes excited by pulses of different durations.

Such studies were performed only using excitation by nanosecond pulses of the second harmonic of a Ti^{3+} : Al_2O_3 laser [8, 9] in the spectral range from 350 to 450 nm. The authors of paper [7] studied the POPOP solution pumped into the first absorption band by continuously tuning the pump wavelength. Later, similar studies were also performed for some other organic dyes (coumarin 120, perylene, and naphthalimides) [10]. Some authors believe that losses induced in the channel of excited triplet [11] and singlet [12] levels (Fig. 1) play a significant role in the effects observed in experiments. On the contrary, the author of Ref. [6] concluded that the influence of absorption of the pump energy in the system of excited singlet–singlet and triplet–triplet states on the lasing efficiency as a function of the pump wavelength was weak [6].

In earlier papers [13, 14], pairs of dyes were selected and optimal excitation conditions were determined for a two-stage laser, in which a converter dye laser was pumped by another flashlamp-pumped microsecond dye laser. This laser system produced ~ 1 -J, 1- μ s light pulses tunable in the spectral range between 495 and 835 nm. The dye pairs were selected so that the emission wavelength of the pump dye laser coincided with the maximum of the absorption band of the dye in the second stage.

This laser system has a number of advantages over a usual flashlamp-pumped microsecond laser. Because of

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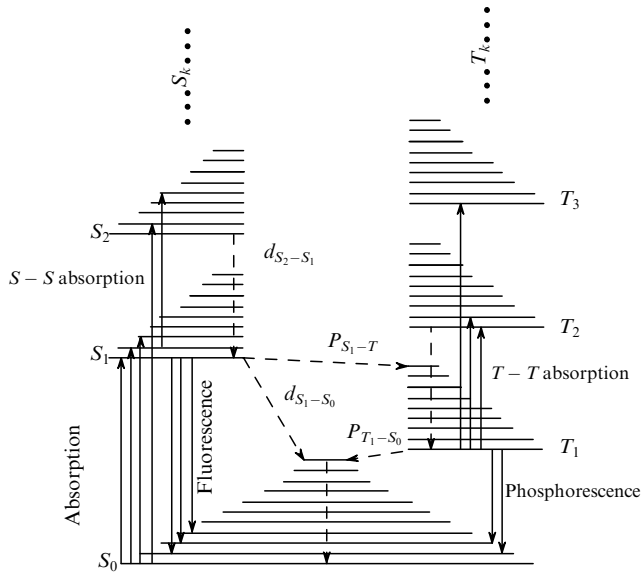


Figure 1. Energy level diagram of singlet (S_k) and triplet (T_k) states of a laser dye (nonradiative transitions are shown by dashed arrows).

weak Stokes losses and the absence of the UV and IR components in the pump spectrum, the angular divergence of the laser emission as small as ~ 1 mrad is provided, resulting in the increase in the emission brightness by 140–700 times. In addition, the operating life of the second-stage dye in this system substantially increases. It was shown in Ref. [15] that a master oscillator with two-stage excitation and amplifiers based on this excitation scheme provide ~ 0.5 -J, ~ 1 - μ s tunable pulses of spectral width $\sim 10^{-3}$ nm with a beam divergence of $\sim 10^{-3}$ rad. In addition, the converter laser has a high spatial coherence and, hence, can be used for art holography (for example, to make portraits and shoot pictures). The outlook for applications of such lasers in this field is determined by many requirements imposed on the laser pulse duration, which should correspond both to the properties of photographic materials and to characteristic rates of variation in the surface structure of objects being photographed. This results in a decrease in the contrast of holograms being detected. In particular, the fastest movements in the portrait holography is the involuntary tremor of eyelids and face muscles, which, as follows from estimates, requires the use of ~ 1 - μ s pulses [16].

In addition, microsecond lasers are used in spectroscopy, nonlinear optics, photochemistry, biology, and medicine. For example, flashlamp-pumped microsecond lasers have found a wide application in intracorporeal lithotripsy. Medical units based on such lasers are developed and produced by Candela (USA), Tecnomed (France), and Telemet Electronic (Germany) [17, 18].

The advantage of these lasers is, first, that they are tunable and thus can provide the selective action on a kidney stone of a particular type [19]. Second, unlike nanosecond pulses, microsecond radiation pulses do not damage an optical fibre, whereas after a standard lithotripsy procedure the fine particles of a silica fibre, whose length is ~ 50 cm, and of its polymer cladding can remain in the ureter [13]. In addition, because the efficiency of intracorporeal lithotripsy depends to a great extent on the efficiency of laser radiation transport through an optical fibre, while the efficiency of coupling of laser radiation to the fibre is

determined by the radiation divergence, the microsecond converter laser considered in this paper is promising in this field as well because its radiation has good spatial-angular characteristics.

In this paper, we consider the reasons for the anomalous dependence of the efficiency of the converter laser based on the ethanol solutions of coumarin, rhodamine, and oxazine dyes, and dyes of some other classes on the pump radiation spectrum. We also determined the conditions providing high lasing efficiency of the dyes pumped by microsecond pulses.

2. Experimental

The experimental setup, whose scheme is shown in Fig. 2, consisted of a two-stage-pumped dye laser and a system for measuring the energy and spectral parameters of laser radiation. Pumping was performed by a microsecond dye laser based on the modification of a coaxial lamp-cell designed by Dzyubenko [20] (Fig. 3). The discharge channel of the lamp-cell was formed by two coaxial quartz tubes (11) and (12) spaced by ~ 1 mm. Quartz cell (6) with an internal diameter of 8 mm was located inside tube (12). The pulsed current of the electric discharge of a storage capacitor bank passed through lamp-cell (2), which plays the role of a reverse conductor, lamp electrode (14) and the discharge gap of lamp (4). To concentrate the pump radiation, the cell was filled with magnesium oxide between the lamp body and quartz tube (11). The lamp was evacuated and filled with xenon through branch pipe (1). The dye solution was pumped in the cell through branch pipes (10), and the cooling liquid or radiation converter

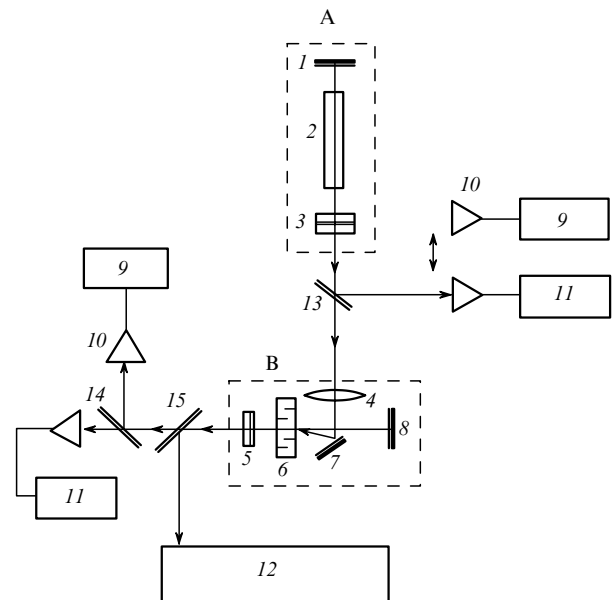


Figure 2. Scheme of the experimental setup: (A) dye laser based on a coaxial lamp-cell; (B) dye laser pumped by coherent microsecond pulses (converter laser); (1) highly reflecting mirror of the resonator of the flashlamp-pumped dye laser; (2) coaxial lamp-cell; (3) output mirror (stack) of the flashlamp-pumped dye laser; (4) focusing lens; (5) output mirror of the converter laser; (6) cell with the ethanol solution of a dye under study; (7) deflecting mirror; (8) highly reflecting mirror of the converter laser; (9) C8-14 two-beam storage oscilloscope; (10) FK-22 photodetector; (11) IMO-2H power meter; (12) DFS-8 diffraction spectrograph; (13, 14, 15) glass beamsplitters.

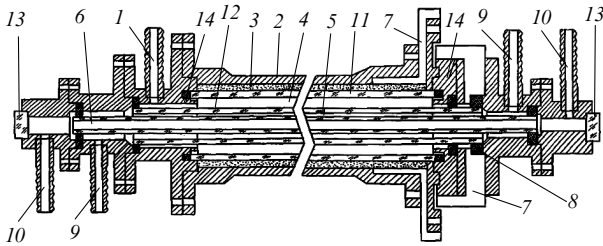


Figure 3. Scheme of a coaxial lamp-cell: (1) air evacuation and xenon admission channel; (2) housing of the lamp-cell; (3) MgO light reflecting coating; (4) discharge gap; (5) cell jacket; (6) quartz cell; (7) fluoroplastic insulators; (8) silicon rubber seals; (9) cooling liquid circulation channel; (10) dye circulation channel; (11) reflector quartz tube; (12) cell jacket quartz tube; (13) cell windows; (14) lamp electrodes.

solution was pumped through branch pipes (9). All seals (8) of the lamp-cell were made of silicon rubber. The cell had glued windows with antireflection coatings.

The lamp-cell was evacuated and filled with xenon with the help of a gas-vacuum system whose scheme is shown in Fig. 4. At first all the vacuum valves were opened and the vacuum system was evacuated, up to a valve of balloon (6) with xenon, with rough-vacuum pump (7) down to a pressure of 5×10^{-3} Torr. Then, storage tank (4) was filled with xenon with the help of reducer (5) up to a pressure of ~ 1 atm (the vacuum post was closed with the help of a vacuum valve). Lamp-cell (1) was filled then with xenon [the filling was controlled with vacuum gauge (8)]. To provide the stability of the parameters of the lamp-cell, the lamp was refilled with xenon when needed.

The resonator of the dye laser was formed by a plane dielectric mirror with $R = 100\%$ and a stack of three plane-parallel plates made of a K8 glass, which was an optimal output mirror in this case. The discharge circuit contained ten parallel low-inductance K75-48 capacitors of

the total capacitance $2.2 \mu\text{F}$ and an IRT-6 ignitron. The power supply voltage was 17 kV. The discharge gap of the lamp was filled with xenon at a pressure of 20–30 Torr. The FWHM duration of the pump pulse was $2 \mu\text{s}$, the duration of the leading edge of the pulse was $0.8 \mu\text{s}$ and the electric energy of the pump pulse was 300 J.

The resonator of the converter laser in the second stage (laser B in Fig. 2) was formed by a highly reflecting plane mirror and a stack as the output mirror. Lasing was excited almost longitudinally at an angle of $5-7^\circ$ to the resonator axis. In the first-stage laser (laser A in Fig. 2), we used ethanol solutions of most efficient laser dyes at the concentration of 1.2×10^{-4} M (Table 1), which covered a broad spectral range (within the most intense absorption band of dyes under study). The concentration of the dyes under study was chosen to provide the absorption coefficient $K \sim 15-25 \text{ cm}^{-1}$ at the pump wavelength. Such an absorption coefficient for typical dyes corresponds to the concentration $\sim 10^{-4}$ M ($5 \times 10^{16} \text{ cm}^{-3}$) [21]. The dye solutions were poured into a plane-parallel cell of thickness 5 mm. Pump radiation was focused on the cell into a ~ 4 -mm spot, the required excitation level was adjusted with the help of neutral filters. The output energy was detected with an IMO-2N power meter, and the temporal parameters of the pump and laser pulses were measured with an FK-22 detector, whose output signal was fed into a C8-14 two-beam storage oscilloscope. The power meter was protected from high-power laser radiation with neutral filters. The radiation was delivered to the power meter through a silica fibre. The emission spectra of the laser were recorded with a DFS-8 diffraction spectrograph. The laser radiation was also coupled into the spectrograph through an optical fibre. The absorption spectra of dye solutions were recorded with a Specord M-40 or a SF-26 spectrophotometers.

3. Experimental results and discussion

It was shown earlier [13, 14] that the lasing efficiency of rhodamine 6G excited by microsecond pulses into the

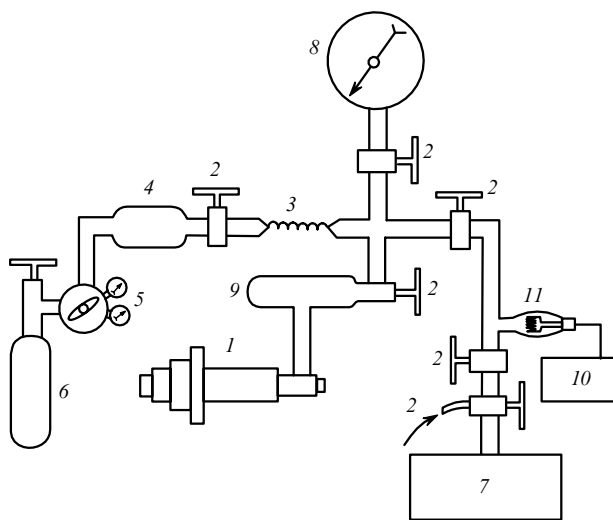


Figure 4. System for evacuating the lamp-cell and filling it with a working gas: (1) lamp-cell; (2) vacuum valves; (3) capillary leak; (4) storage tank; (5) reducer; (6) vessel with xenon; (7) vacuum gauge; (8) vacuum gauge; (9) ballast tank; (10) thermocouple vacuum gauge; (11) measuring lamp.

Table 1. Laser dyes used in the pump laser.

| Laser dye | $\lambda_p^{\text{max}}/\text{nm}$ |
|---|------------------------------------|
| Coumarin 120 | 440 |
| Coumarin 47 | 460 |
| Coumarin 102 | 485 |
| Coumarin 30 | 505 |
| Coumarin 334 | 520 |
| 3-carbamidojulolidine-2-iminocoumarin | 525 |
| Coumarin 6 | 535 |
| 2-(7-diethylaminocoumarin-3-il) benzimidazolyl chloride | 550 |
| Rhodamine 110 | 560 |
| 2-(julolidinecoumarin-3-il) benzimidazole chloride | 570 |
| Rhodamine 6G | 590 |
| Rhodamine B | 610 |
| Rhodamine 3B | 630 |
| Oxazine 17 | 665 |

maximum of the absorption band at 525 nm increased first with increasing pump energy density and then decreased. Figure 5 shows the lasing efficiency of the rhodamine 6G converter laser pumped by a flashlamp-pumped coumarin 334 laser at ~ 520 nm through the output mirror as functions of the pump energy density. The FWHM pump pulse durations were 1.0, 2.0, and 5.0 μs (the pulse duration was varied by changing the parameters of the discharge circuit of the flashlamp-pumped dye laser; the pulse energy was 1.2–2.2 J depending on its duration).

The studies were performed using the optimal concentrations of the dye and the optimal reflectivity of the output mirror of the plane resonator [13]. One can see from the dependences in Fig. 5 that the lasing efficiency first increases with increasing the pump energy density and then decreases. In all the cases, the optimal pump energy density is ~ 2 –4 J cm^{-2} . The lasing efficiency achieves 36% upon pumping by 1- μs pulses and decreases down to $\sim 20\%$ upon pumping by 5- μs pulses. It is known from the literature that the maximum efficiency (65%–68%) of a laser on the ethanol solution of rhodamine 6G excited by nanosecond 530-nm radiation pulses is achieved at the pump power density equal to 20–40 MW cm^{-2} . In this case, the lasing efficiency is determined under optimal conditions by losses of the pump and lasing radiation in the channel of excited singlet levels [22].

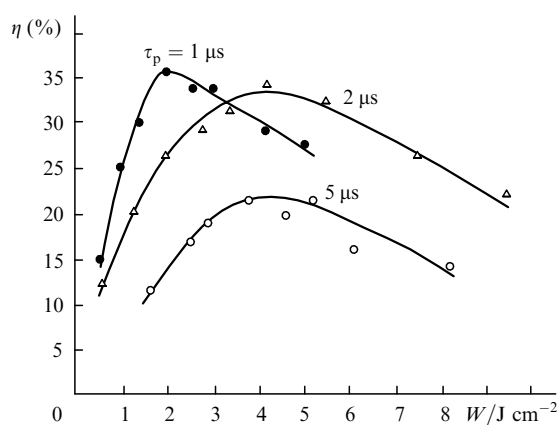


Figure 5. Dependences of the lasing efficiency η of the rhodamine 6G converter laser on the pump energy density W for different pump pulse durations τ_p .

Comparison of the experimental data showed that the lasing efficiency achieved upon pumping by 1- μs pulses at the optimal pump energy is virtually the same as that obtained upon pumping by nanosecond pulses. This fact and the equality of the lasing onset and termination thresholds for the rhodamine 6G converter laser under optimal pumping conditions show (Fig. 6) that no time-dependent losses appear during lasing. This suggests that upon optimal pumping by 1- μs pulses, the losses related to triplet–triplet absorption play a minor role compared to the losses associated with singlet–singlet absorption. When the pump energy density exceeds the optimal value, the triplet–triplet absorption losses begin to play a significant role, as well as losses related to the thermal optic and acoustic distortions of the active medium, which reduce the lasing efficiency.

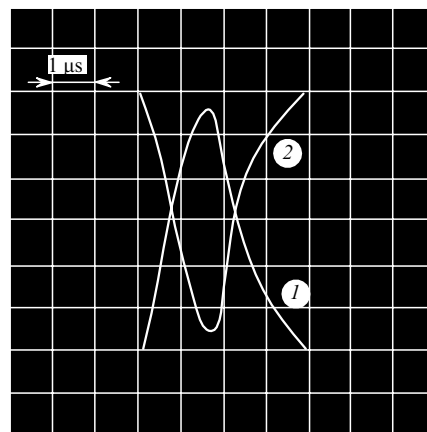


Figure 6. Oscillograms of the pump (1) and output (2) pulses of the converter laser on the ethanol solution of rhodamine 6G pumped at 525 nm by the flashlamp-pumped laser on the ethanol solution of 3-carbomidojulolidine-2-iminocoumarin.

The dominating influence of singlet–singlet absorption losses on the efficiency of the converter laser is confirmed by the studies of the dependence of the lasing efficiency of ethanol solutions of most efficient laser dyes on the spectrum of microsecond pump pulses. As mentioned above, the dyes were pumped into the most intense absorption band. The results of these experiments are presented in Fig. 7. We found the anomalous dependences of the lasing efficiency and spectral characteristics of ethanol solutions of coumarin, rhodamine and oxazine dyes, and DCM and phenolemine 160 on the pump radiation wavelength. The lasing efficiency first increases with increasing the pump wavelength, then decreases near the maximum of the absorption band and again increases at the long-wavelength side of the absorption maximum. The lasing spectra exhibit the blue shift, their long-wavelength boundary being shifted only weakly (as shown for rhodamine 6G in Fig. 8).

As the pump wavelength is increased, the laser emission band broadens to the blue by 10–12 nm, and two-band lasing is observed in the ‘hole’ region. The blue shift of laser emission spectra suggests that losses increase during lasing [23]. To elucidate the nature of these losses, we studied the time dependences of the pump and laser pulses as functions of the pump pulse spectrum. We found that the onset and termination lasing thresholds for the converter laser were the same in all the cases studied (Fig. 6). Therefore, no additional time-dependent losses appeared during lasing, and as mentioned above, the triplet–triplet absorption losses play a minor role compared to singlet–singlet absorption losses.

The experimental data show that, to explain the observed effects in detail, in particular, the nature of photoproducts, comprehensive studied should be performed. However, the available data on excited singlet–singlet absorption for ethanol solutions of some coumarin, rhodamine, and oxazine dyes allow one to explain satisfactorily these effects [24, 25].

Curves (3) in Figs 7e, f, j show the spectra of the Einstein coefficient $B_{s_1 \rightarrow s_2}(\nu)$ for excited singlet–singlet absorption observed in solutions of rhodamine 6G, rhodamine B, and cresyl violet, respectively [24]. Comparison of the spectral dependence of losses in the channel of excited

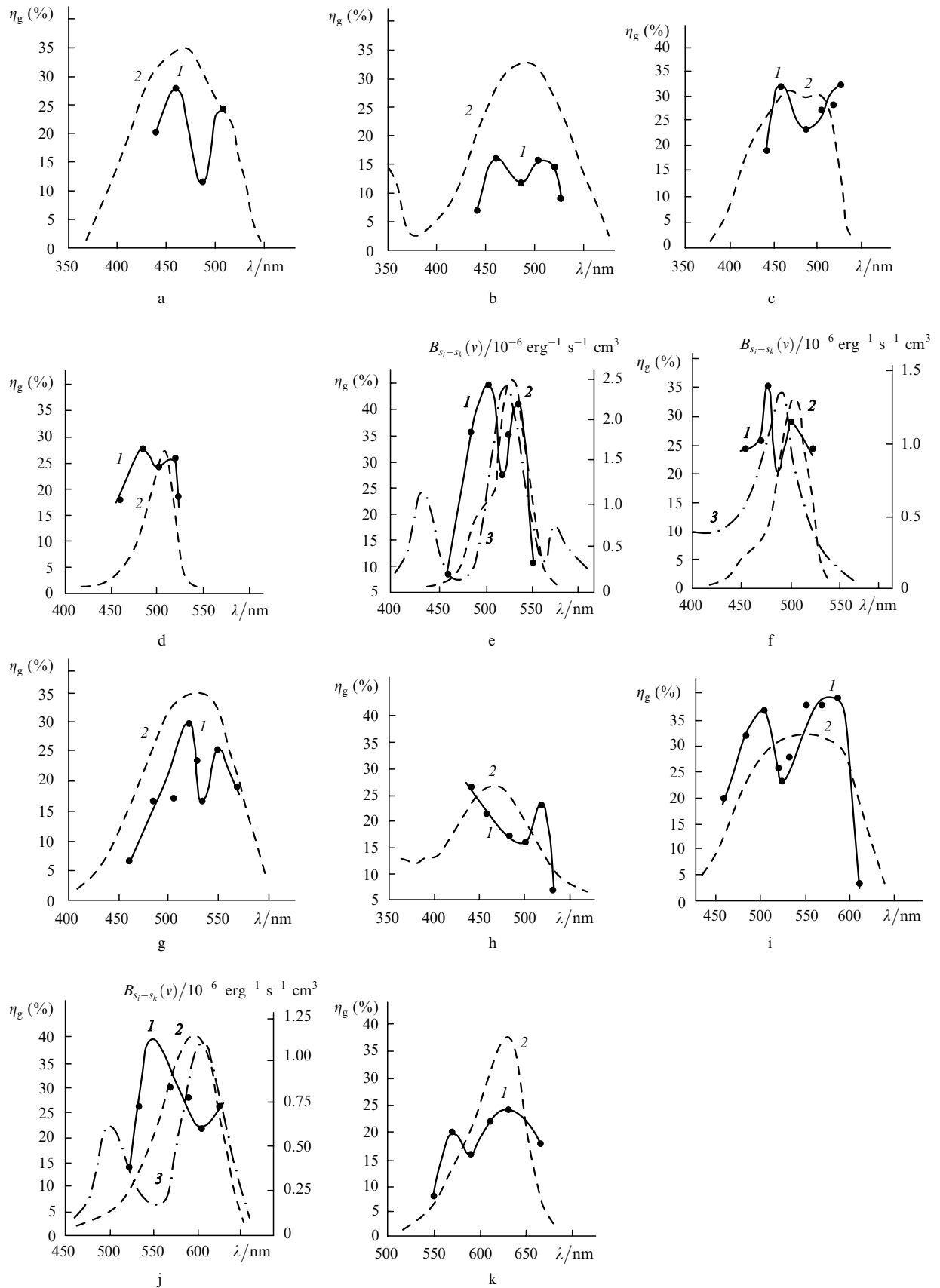


Figure 7. Dependences of the lasing efficiency of ethanol solutions of laser dyes on the excitation wavelength: curves (1): (a) 3-benzimidazolil-7-julolidine iminocoumarin perchlorate; (b) 3-benzimidazo [4,5 v]-pyridil-7-julolidine coumarin hydrochloride; (c) 2-(julolidinecoumarin-3-il) benzimidazolyl perchlorate; (d) rhodamine 110; (e) rhodamine 6G; (f) rhodamine B; (g) phenolemine 160; (h) DCM, (i) oxazine 17; (j) cresyl violet; and (k) oxazine 1. Absorption bands of the dyes in relative units [curves (2)] and the Einstein coefficients for excited singlet-singlet absorption for rhodamine 6G (e), rhodamine B (f), and cresyl violet (j) [curves (3)].

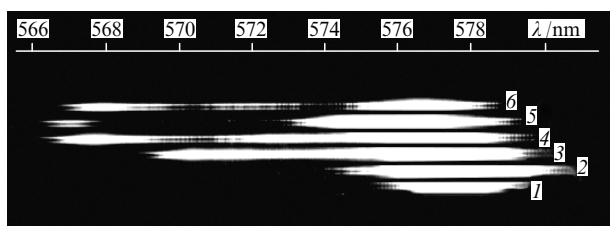


Figure 8. Lasing spectra of the ethanol solution of rhodamine 6G obtained upon pumping at 460 (1), 485 (2), 505 (3), 520 (4), 525 (5) and 535 nm (6).

singlet–singlet absorption with the dependence of the lasing efficiency of these dyes on the pump wavelength suggests that upon pumping of the dyes by microsecond pulses, reversible photoproducts are formed due to excited singlet–singlet absorption of pump radiation, as upon pumping by nanosecond pulses. The photoproducts have strong absorption in a certain spectral gain region, which reduces the lasing efficiency [25, 26].

The results of laser flash photolysis of the ethanol solution of rhodamine 6G performed at different excitation intensities are reported in Ref. [27]. The integrated absorption spectra of photoproducts were obtained in the range from 550 to 670 nm upon probing synchronously with pumping or with a time delay of ~ 25 ns. The distinct structure of the spectrum observed for $W \geq 1.5 \text{ J cm}^{-2}$ and the difference between the spectra recorded without or with the time delay indicate to the appearance of short- and long-lived photoproducts, which absorb in a broad spectral range, including the lasing region. The estimates made in Ref. [27] showed that the probability of phototransformations of rhodamine 6G upon three-stage excitation at 532 nm was rather high ($\sim 0.005 - 0.007$).

4. Conclusions

We have shown that two-stage pumping of a converter dye laser by $\sim 1\text{-}\mu\text{s}$ pulses from a flashlamp-pumped dye laser makes it possible to obtain the same lasing efficiency as upon pumping by nanosecond pulses of the corresponding power density. This suggests that the energy losses related to excited triplet–triplet absorption and the products of irreversible photochemical transformations play a minor role compared to losses related to excited singlet–singlet absorption.

The study of the main classes of laser dyes in the converter laser has shown that the lasing efficiency decreases when the pump radiation has a certain spectrum, which depends on the dye under study. This can be explained by reabsorption of the pump radiation in the system of excited single levels. The reabsorption of pump radiation leads to the formation of reversible photoproducts strongly absorbing in a certain gain region. The optimal choice of the pump spectrum results in a significant improvement of the energy parameters of the converter dye laser pumped by microsecond pulses.

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