INFLUENCE OF DYE CONCENTRATION AND OF THE LASER SYSTEM DESIGN ON ITS LIFETIME

K.M. Degtyarenko, T.N. Kopylova, P.T. Kuznetsova, G.Y. Mayer, and E.N. Tel'minov

V.D. Kuznetsov Siberian Physicotechnical Scientific-Research Institute at the State University, Tomsk

Received December 21,1992

We experimented with laser system consisting of a master oscillator and two light amplifiers and emitting within a narrow spectral band ($\Delta\lambda \le 0.01$ nm). In this system the ethanol solution of coumarine 2 is pumped by radiation of a XeCl excimer laser. The lifetime of such a system is shown to exceed that of a system with nonselective resonator and to depend on cell length and on dye concentration in the amplifier. Our study indicated that laser emission stabilized dye in the system. Laser design should fit its future applications while its lasing efficiency be optimized varying the concentration of dye.

At present the task most important for dye lasers is that of increasing the lifetime of their active media (stabilizing their lasing characteristics). It is well known that dye solutions arc irreversibly photoconverted by high intensity radiation, pumped into them, so that lasing characteristics change in the course of generation, producing operational difficulties. There are extensive reports on the task of increasing lifetime, but, generally these cover lasers with nonselective resonator.[1,2] Theoretical estimates of the effect of conditions of irradiation upon photo-decomposition of laser dyes may be found in Refs. 3 and 4. We studied the lifetime experimentally using a setup, which combined high efficiency of transformation of radiation in an excimer laser with pulse energy up to 0.5 J (efficiency up to 20%) with high quality of the transformed radiation (it produced a line 0.01 nm wide with a divergence below 1.5 mrad).

The pumping XeCl laser radiation is distributed as following: 0.15 E_p goes to the controlling generator (CG), 0.3 E_p — to the first amplifier (A1), and 0.55 E_p — to the second amplifier (A2). Two diffraction gratings are used as the CG selective element, one of them functioning at glancing incidence thus producing a line 0.1 nm wide. The two amplifying cells are total internal reflection prisms, the pumping radiation completely absorbed by the solution of the coumarine 2 dye, circulating in the internal channel of $D = 0.4$ cm diameter and $l = 5$ cm length. We define lifetime as the total energy, pumped into a unit volume A2 while the lasing efficiency drops by 20 or 50%. Since the CG and A1 both operate in throughput mode and the solution volume is $V = 500$ cm^3, while A2 has no throughput and its volume is $V = 1$ cm^3, we assume photoconversions in the CG and A2 to be negligible during the experiment. Lasing characteristics are shown in Table I.

TABLE I. Lasing characteristics of a MZhL–03 laser system

Active medium in the A2 amplifier, irradiation regime	η_0, %	η/η_0	$K^0_{\lambda_p}$, (cm^{-1})	$\frac{\Delta K_{\lambda_p}}{K_{\lambda_p}}$,(%)	$1-\frac{c}{c_0}$	P_{80}		P_{50}		ΔD_{λ_g}
						$\frac{J}{\text{cm}^3}$	$\frac{\text{phot}}{\text{mol}}$	$\frac{J}{\text{cm}^3}$	$\frac{\text{phot}}{\text{mol}}$	
C2 0.6 mmol/1 in ethanol, selective generation	8.1	0.43	1.4	13.6	0.056	3	13	10	43	0.158
C2 1.0 mmol/1 in ethanol, spontaneous	11.8	0.26	2.45	10.4	0.094	5	13	15	39	0.385
C2 1.0 mmol/1 in ethanol, selective generation	11.8	0.35	2.45	10.5	0.036	8	21	32	83	0.316
C2 1.3 mmol/1 in ethanol, selective generation	12.1	0.46	2.9	5.2	0.064	8	16	45	90	0.281
C2 1.3 mmol/1 in ethanol, spontaneous	12.1	0.5	2.9	10.7	0.13	5	10	22	44	0.267
C2 1.3 mmol/1 in ethanol, nonselective generation	12.5	0.53	2.9	2.2	0.055	5	10	35	70	0.253
C2 2.0 mmol/1 in ethanol, selective generation	13.8	0.49	4.9	8.6	0.032	18	23	75	98	0.3
C2 2.0 mmol/1 in ethanol, spontaneous	13.8	0.51	4.9	9	0.12	8	10	46	60	0.295
C2 4.2 mmol/1 in ethanol, selective generation	15.1	0.52	10.2	4	0.019	20	12	95	59	0.348
C2 4.0 mmol/1 in ethanol, spontaneous	15.1	0.51	9.8	2.2	0.08	10	6	40	26	0.416

Coincidentally with lifetime we checked spectral changes in the longwave absorption band, measuring the quantum output of photoconversions, φ_{ph}, i.e., the number of molecules disintergrated per each absorbed photon. In

0235-6880/93/06 421-03 $02.00

addition, we controlled changes in spectral absorption at λ_g and λ_p using the Specord M 40 device, and measured the depletion of the injecting signal I from A1 during generation using an IMO–2N calorimeter. Since $\Delta D_{\lambda_g} = \log (I_0^{A1} / I^{A1})$ is the optical thickness, results obtained using both spectral measurements and extinction coincided. Substituting irradiated solution in A2 after the efficiency halved by the fresh one reclaimed the injecting signal to its previous experimental value, which confirmed photochemical nature of those changes.

Both λ_g and $\Delta\lambda_g$ are prescribed by the injecting signal from the CG, not affected noticeably by photoconversions during the experiment. That is why we discovered no changes of these characteristics during lasing, whereas a small, 1–2 nm longwave shift of generation spectrum was noted for coumarines in the nonselective resonator due to photoconversions. The divergence we measured using the diaphragm in the focal plane of the lens remained constant within the experiment accuracy.

It is seen from Table I that the optimum efficiency of transformation is observed when dye concentration in A2 is about 2–4 mmol/liter. A 50% drop in efficiency from its initial value due to photoconversions results in an order of magnitude smaller than proportion of disintegrated molecules. This fact testifies that the decrease of generation energy results from the formation of photoproducts absorbing radiation in dye lasing and pumping ranges, instead of lower concentration of generating molecules. Figure 1 presents the dependences of efficiency on absorption at λ_g.

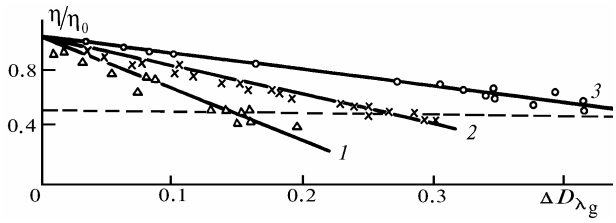

FIG. 1. Generation efficiency vs optical density at ˆ ŭuų generation. Dye concentration in A2 *is: 1)0.6, 2)1 aid 3)* 4.2 mmol/liter.

Since photoproduct absorbing with coefficient ε at λ_g is formed at concentration c by a certain time photoproduction, it follows from relation $D = c\varepsilon l$, that the drop in efficiency is proportional to l, which is the length of active medium in the amplifier. This fall–off depends also on dye concentration: generation energy drops at a slower rate at higher concentrations of dye even if absorption at λ_g remains the same (see Fig. 1).

Thus lifetime increases to one order of magnitude while concentration is raised from 0.6 to 4 mmol/liter, photoconversion output remaining practically the same (see Fig. 2*b*). Curves in Fig. 2 show lifetimes decrease by practically one order of magnitude due to increased output of photoconversions in the case when irradiation is free of generation, i.e., spontaneous or almost spontaneous (without injecting signal). In that case photostabilization by lasing is observed due to shorter lifetime of photodegradating state.[5]

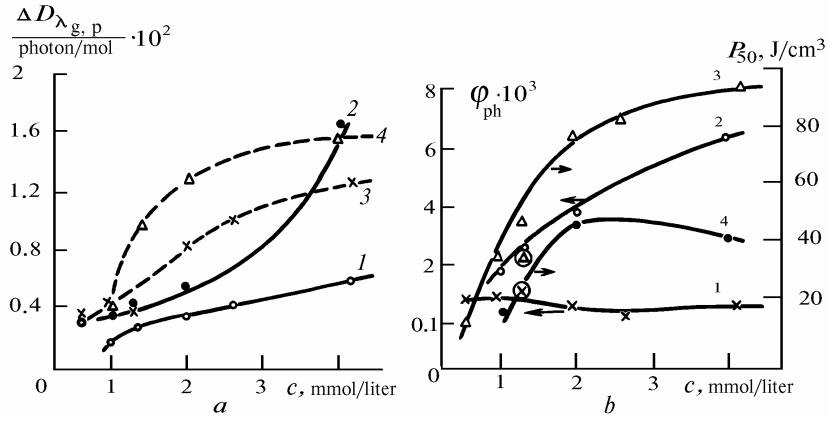

FIG. 2. Dependences of the absorption at λ_g (curves 1 and 2) and at λ_p (curves 3 and 4) per single photon (a) as well as of photoconversion output φ_{ph} (curves 1 and 2) and of lifetime P_{50} (curves 3 and 4) (b) on dye concentration in A2 for different regimes of irradiation: 1 and 3) generation regime; 2 and 4) spontaneous irradiation regime. The symbols inside the circles (b) correspond to generation reginte with a nonselective resonator.

Absorption per each photon absorbed by a single molecule grows quicker depending on concentration in generation range if the regime is spontaneous, in contrast to that of lasing, whereas the respective dependence at λ_p remains practically the same. Thus, in the lasing regime the photoconversion channel more strongly inhibited is that related to photoproducts absorbing at λ_g, instead of at λ_p. One may then conclude that photoproduct absorbing at λ_g is related to photodegradation of dye from the laser level, while the "shortwave" photoproduct is produced in the other channel. The increase in lifetime accompanying the growth in concentration from 0.6 to 4 mmol/liter in generation regime is apparently explained by the system reaching an optimum regime of transformation efficiency, the fraction of spontaneous radiation falling off. The lifetime trend under the conditions of spontaneous irradiation can be explained by lower energy load per molecule within the pumping pulse: 1.3 photon at 0.6 mmol/liter and 0.3 photon at 2 mmol/liter.

Of special interest it is to compare the narrow hand generation regime, $\Delta\lambda \le 0.01$ nm, to that with a nonselective resonator in the CG, when a total reflection miror was used instead of the second grating. Zeroth order diffraction pattern from grating the first was sent to that mirror. In that case the lifetime was shorter than for a

similar solution generating in the narrow line, although photoconversion output remained practically the same (see Fig. 2*b*).

It was supposedly due to an essential growth in superradiation: the wide spectral band ($\Delta\lambda_g$ = 12–13 nm) was amplified, i.e., the fraction of spontaneous processes increased, resulting in stronger absorption at λ_g too. If one compares that result with lifetime for C2 in a common laser system with a nonselective generator, no amplifiers and a cross Xe–Cl laser pump of approximately the same energy, then taking into account the cell length, the lifetime in such a system with nonselective resonator appears to be significantly longer. This is apparently due to a more homogeneous excitation and to shorter generation time, i.e., to smaller contribution from spontaneous processes.

It is noticed that the fraction of photoproducts absorbing at λ_p. is smaller in that system: it reaches 20% in a generator with nonselective resonator and no amplifiers when the efficiency drops by 50%.

Thus, lifetime of a laser system including a cascade of amplifiers exceeds that of a system with nonselective resonator due to lower contribution from spontaneous processes. However, that lifetime decreases for longer cell lengths, i.e., this parameter should be optimized by both efficiency and lifetime. Moreover, the lifetime depends significantly on dye concentration, which also has to be optimized over transformation efficiency.

REFERENCES

1. A.N. Fletcher and M.E. Pietrak, Appl. Phys. **B37,** 151–157 (1985).
2. R.T. Kuznetsova, R.M. Fofonova, T.N. Konylova, et al., Kvant. Elektron. **16**, No. 5,993-996 (1989).
3. E.M. Goryaeva and A.V. Shablya, Opt. Spectrosk. **64**, No. 3, 532–538 (1988).
4. O.I. Ivanenko and O.B. Cherednichenko, Zh. Prikl. Spektrosk. **54**, No. 6, 955–959 (1991).
5. M.V. Bondar, O.V. Przhonskaya, and E.A. Tikhonov, Kvant. Elektron. **16**, No. 11, 2201–2206 (1989).