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1979-08-15

# Heating-effect minimization in dye lasers

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## Original Publication Citation

Matheson, Kenneth L. and James M. Thorne. "Heating-effect minimization in dye lasers." *Applied Physics Letters* 35 (1979): 314-317.

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- <sup>24</sup>One device was started ~ 150 h before the other. The number of devices under test is presently limited by available equipment.
- <sup>25</sup>At  $t \sim 623$  h, the  $N_2$  cooling flow increased to zero and the device operated without cooling for ~ 2–3 h. As a consequence, the junction temperature increased and the laser output decreased markedly. After the  $N_2$  flow was reestablished, the output power of the laser was reduced by a factor of ~ 0.97 relative to the power emitted at  $t = 0$  h. This is the cause of the decrease in the power at  $t \sim 620$  h.
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## Heating-effect minimization in dye lasers

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(Received 22 March 1979; accepted for publication 6 June 1979)

Optical pumping of dye lasers deposits heat nonuniformly in the active medium. Because the refractive index of a liquid or gas is a function of temperature, gradients are created which refract the laser beam. This effect is sometimes serious enough to quench laser action. In this paper, we evaluate the effects caused by optical pumping with a coaxial flashlamp and suggest ultraviolet absorbers and wavelength shifters as materials for minimizing the problem.

The formation of refractive-index gradients due to non-uniform heating is a serious problem with dye lasers. These gradients distort the wave front of the laser beam, limiting the pulse repetition rate and the attainable linewidth.<sup>1</sup> The objectives of this paper are to examine the causes of internal heating for a typical dye solution and to suggest methods for alleviating the problem.

Balucani and Tognetti<sup>2</sup> derived expressions for the heat effects in a slab dye laser. We have extended this work by using cylindrical coordinates to describe heating effects in a coaxial flash-pumped dye laser.

The absorbed flashlamp radiation is converted to heat in the solution by two processes, labeled here  $Q_1$  and  $Q_2$ .  $Q_1$  is the heat arising from radiationless deactivation of molecules in excited states. The number of molecules which undergo this process is  $1-\phi$  where  $\phi$  is the quantum efficiency for fluorescence.  $Q_2$  arises from the Stokes shift. Emitted photons are less energetic than absorbed photons, and the energy difference becomes heat in the solution.  $Q_1$  and  $Q_2$  can be expressed mathematically as:

$$Q_1(r,z,t,\lambda) = C\epsilon(\lambda)(1-\phi)I(r,z,t,\lambda), \quad (1)$$

$$Q_2(r,z,t,\lambda) = C\epsilon(\lambda)\phi(1-\lambda/\lambda'_{\max})I(r,z,t,\lambda). \quad (2)$$

In these expressions,  $C$  is the molar concentration of the dye,  $\epsilon(\lambda)$  is the molar extinction coefficient,  $\phi$  is the quantum yield,  $\lambda'_{\max}$  is the wavelength of maximum absorption, and  $I(r,z,t,\lambda)$  is the intensity of the pump light at a distance  $r$  from the cylinder axis at time  $t$ . The total heating power absorbed per unit volume,  $Q(r,z,t)$ , is the sum of these two contributions integrated over the absorption band of the dye:

$$Q(r,z,t) = \int_{\text{abs band}} C\epsilon(\lambda)(1-\phi\lambda/\lambda'_{\max})I(r,z,t,\lambda)(d\lambda). \quad (3)$$

The temperature increase in the solution at time  $t$  is obtained by integrating  $Q(r,z,t)$  up to time  $t$ :

$$\Delta T(r,z,t) = (\rho_0 c_p)^{-1} \int_0^t Q(r,z,t') dt', \quad (4)$$

where  $\rho_0$  is the solvent density and  $c_p$  is the specific heat of the solvent. The change in refractive index is directly related to  $\Delta T(r,z,t)$  by the expression:

$$n(r,z,t) - n^0 = \frac{\partial n}{\partial T} \Delta T(r,z,t), \quad (5)$$

where  $\partial n/\partial T$  is the change in refractive index with temperature at constant pressure.

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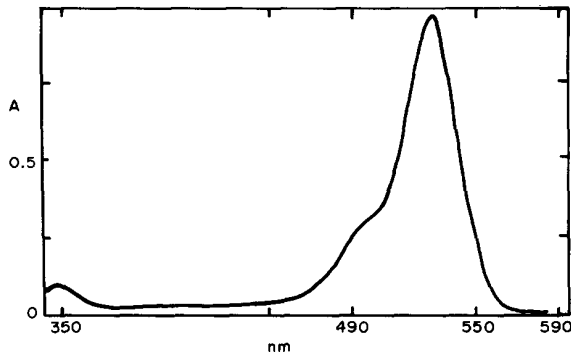


FIG. 1. Absorption spectrum of rhodamine 6G laser dye in ethanol, showing the narrow band and the wide band of wavelengths used in this study to calculate heating effects.

In order to calculate values of  $\Delta T(r, z, t)$  an expression for  $I(r, z, t, \lambda)$  is needed. McKenna<sup>3</sup> gives an exact expression for  $I(r, z, t, \lambda)$ , but we use a simple approximation which can be rationalized as follows: The pump light in a coaxial flashlamp-pumped dye cell converges radially inward to the cylinder axis, hence an exponential factor  $\exp[-C\epsilon(\lambda) \times (r^0 - r)]$ , where  $r^0 - r$  is the distance of penetration, is needed to describe, in part, the attenuation of the pump light due to absorption. Furthermore, the lens effect of the cylindrical dye cell focuses the pump light as it converges to the center of the cylinder. This can be described by a cylindrical focusing function  $g(r)$  which is just a special case of McKenna's exact expression for a nonabsorbing dielectric cylinder. These two factors are combined in the following approximate expression for  $I(r, z, t, \lambda)$ :

$$I(r, z, t, \lambda) \cong g(r)H(z) \exp[-C\epsilon(\lambda)(r^0 - r)] \times e_p 8\pi h - C^2 f(t) / \{\lambda_s \exp[(hc/kT^b\lambda) - 1]\}. \quad (6)$$

In this expression  $c$  is the speed of light,  $r^0$  is the radius of the dye cell in cm,  $e_p$  is the blackbody emittance,  $T_b$  is the blackbody temperature,  $k$  is the Boltzmann constant, and  $g(r)$  is the cylindrical focusing function. In addition,  $f(t) = 0$  when  $t < 0$ ,  $\exp(-t/\tau') - \exp(-t/\tau)$ , where  $\tau'$  and  $\tau$  are pulse-shaping parameters with  $\tau' > \tau$ , and  $H(z) = 0$  for  $z < 0$ ,  $H(z) = 1$  for  $0 < z < h$ , where  $h$  is the dye cell length, and  $H(z) = 0$  for  $z > h$ .

The function  $H(z)$  is included because the dye cell is of finite length, but end effects should be a problem only within a distance of  $r^0$  from either end. Also, in this expression, the flashlamp is assumed to be an idealized blackbody radiator<sup>4</sup> with average emittance  $e_p$ .

TABLE I. Cylindrical focusing function values.

$r$	$g(r)$	$r$	$g(r)$
0	2.28	0.6	2.01
0.1	2.27	0.7	1.79
0.2	2.26	0.8	1.41
0.3	2.22	0.9	1.15
0.4	2.19	1.0	1.00
0.5	2.11		

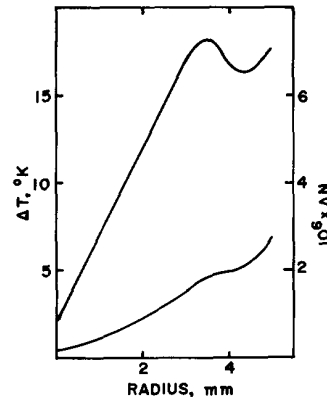


FIG. 2. Temperature change versus radial distance from cell axis. Upper curve is for wide-band irradiation, lower curve for narrow.

To determine how the absorbed heat depends on the various wavelengths of the flashlamp radiation, an idealized system with  $10^{-4}M$  rhodamine 6G solution in ethanol was modeled. Values of  $\Delta T(r, z, t)$  were calculated using Eq. (4) for two wavelength ranges of pump radiation: 350–590 nm and 490–550 nm. Figure 1 shows how these correlate with the absorption spectrum of rhodamine 6G.

The parameters used in the calculation of  $\Delta T$  for the rhodamine 6G solution are as follows:  $C = 0.0001M$ ,  $\phi = 0.83$ ,  $\lambda'_{\max} = 530$  nm,  $T_b = 12\,000$  °K,  $\tau = 10^{-8}$  sec,  $e_p = 1$ ,  $\tau' = 2.0 \times 10^{-7}$  sec,  $t = 1.0 \times 10^{-6}$  sec,  $p_0 = 0.78504$  g/cm<sup>3</sup>, and  $c_p = 0.594$  cal/°K g.

The values of  $g(r)$  were calculated for a dielectric cylinder with  $n = 1.3594$ , the refractive index of ethanol, and they are listed in Table I.

The results of the calculations for  $\Delta T$  and  $\Delta n$ , according to Eqs. (4) and (5), are plotted in Figs. 2 and 3 for the two wavelength ranges. In each case the three curves represent the time evolution of the heat effect. There are two important points worth noting. First, the cylindrical focusing function causes the  $Q(r, z, t)$  and the  $\Delta T(r, z, t)$  functions to have maxima and minima at surfaces with the cylinder. Without  $g(r)$  the functions would simply increase smoothly from a minimum at  $r = 0$  to a maximum at  $r = r_0$ . These maxima and minima cause wide variations in  $\Delta n/\Delta r$ , which indicates defocusing effects in the solution. The other important point is that only one-third as much heat accumulates if irradiation occurs only in the main long-wavelength absorption band

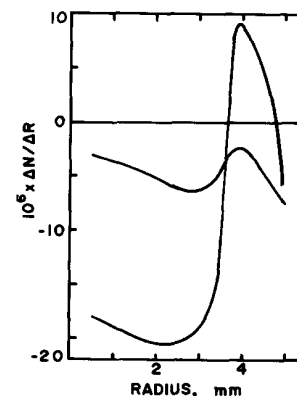


FIG. 3. Refractive index change per cm (radially) versus radial distance from cell axis for a 10-cm dye cell. Upper curve is for wide-band irradiation, lower curve for narrow.

TABLE II. Spectral and decomposition data on uv absorbers in ethanol. Samples of 20 cm<sup>3</sup> were used for decomposition studies.

Compound	Concentration (10 <sup>4</sup> × molarity)	Absorption peaks (nm)	Decomposition rate (picomoles/flash)	Source of chemical
Phenyl salicylate	7.4	310,242	802	Mallinkrodt
2,4-dihydroxybenzophenone	5.8	326,291	21	Aldrich
2-hydroxy-4-methoxybenzophenone	5.8	326,288	66	Aldrich
2,2'-dihydroxy-4,4'-dimethoxybenzophenone	5.0	343,284	49	Aldrich
2,2', 4,4'-tetrahydroxybenzophenone	5.5	350,286	47	Aldrich
Uvinul MS-40	5.2	327,288	42	GAF
Tinuvin P	5.2	338,298	37	Ciba-Geigy
Tinuvin 320	5.4	342,303	28	Ciba-Geigy
Tinuvin 326	5.1	350,310	45	Ciba-Geigy
Tinuvin 327	5.1	348,311	39	Ciba-Geigy
Uvinul N-35	7.7	302	1186	Ciba-Geigy
Cyasorb UV-1988	3.4	313,229	212	American Cyanimid

(490–550 nm). Shorter wavelengths penetrate deeply and deposit excessive heat because of their large Stokes shifts. Eliminating these short wavelengths would be very beneficial.

The triaxial flashlamp dye cell system lends itself well to elimination of short wavelengths through the dissolving of uv absorbers or wavelength shifters in the coolant which circulates in the annular region between the flashlamp and the dye. By absorbing uv radiation, these compounds would decrease the heating, protect the dye from uv-induced photodecomposition, and enable the pump radiation to be used more efficiently. Fluorescent wavelength shifters in the annular region would absorb flashlamp radiation on the short-wavelength side of the dye absorption band and then fluoresce in the absorption band of the laser dye.

Twelve uv absorbers and twelve wavelength shifters have been evaluated on the basis of absorption spectra, resistance to photodegradation under simulated flashlamp-pumped laser conditions, and, where applicable, fluorescence spectra (Tables I and II). The photodegradation tests were carried out using an ILC linear xenon flashlamp L 484

LCT 953 pulsed at 2 Hz at 5 kV. Solutions of each compound at about 10<sup>-3</sup>M concentration were placed in a glass tube surrounding the flashlamp to provide maximum exposure to the radiation. Solution volume was 20 cm<sup>3</sup>.

If a uv absorber is to be selected, then the compound should be chosen so that its long-wavelength absorption peak is just to the short-wavelength side of the absorption peaks of the laser dye. If a wavelength shifter is to be used, then the compound should be chosen so there is maximum overlap between the fluorescence spectrum of the shifter and the absorption spectrum of the dye. For example, the laser dye perylene absorbs strongly from 390 to 440 nm, but inefficiently uses pump light below 390 nm. Tinopal 5BM (Table III) would be an excellent wavelength shifter for perylene because it absorbs the short wavelengths (below 390 nm) and reradiates the light at 430 nm, in the strong absorption region for perylene. Tinopal 5BM has an additional advantage in its very high resistance to photodecomposition.

This work was supported by the U.S. Energy Research and Development Administration.

TABLE III. Fluorescent wavelength shifters: spectral and decomposition data. Samples of 20 cm<sup>3</sup> were used for decomposition studies. Ethanol was used as the solvent for all but Leucophor B, Tinopal CBS, and Tinopal 5BM, where water was used.

Compound	Concentration (10 <sup>4</sup> × molarity)	Absorption peaks (nm)	Fluorescence peaks (nm)	Decomposition rate (picomoles/flash)	Source of shifters
Leucophor B	2.4	348,278	450	28	Sandoz
Tinopal RBS	2.5	362	430	122	Ciba-Geigy
Rinopal CBS	1.4	350	435	140	Ciba-Geigy
Tinopal 5BM	1.5	333,270	430	16	Ciba-Geigy
Tinopal AMS	1.7	331,272	431	22	Ciba-Geigy
PPF	2.2	325	367	375	Eastman
PPO	2.8	303	362	467	Eastman
α-NPO	3.5	330	405	457	Eastman
PBD	2.0	302	360	58	Eastman
BBOT	1.7	375	435	205	Eastman
Bis MSB	1.6	345	419	209	Eastman
Coumarin 4	5.3	323	390	545	Eastman

## Hollow-cathode excitation of ion laser transitions in noble-gas mixtures

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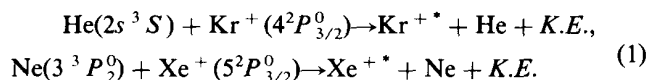
(Received 3 May 1979; accepted for publication 6 June 1979)

Excitation of laser transitions in He-Kr, He-Ar, Ne-Xe, and He-Ne-Xe mixtures was investigated in hollow-cathode discharges. The dominant excitation mechanism of the noble-gas ions was found to be collisions of the second kind between the ground-state noble-gas ions and helium or neon metastables. Based on gain and absorption spectroscopy measurements, the cross section for this mechanism was estimated to be  $\geq 10^{-14}$  cm<sup>2</sup>. New laser transitions in noble-gas and copper ions are also reported.

PACS numbers: 42.55.Fn

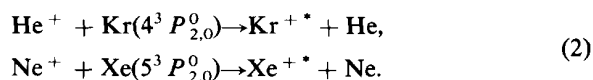
Hollow-cathode discharges have been used primarily for obtaining laser action in metal-ion transitions, where they have proved to be especially efficient. The required metal vapor may be generated by external heating<sup>1</sup> or cathode sputtering,<sup>2,3</sup> and excitation of laser transitions occurs via charge-transfer reactions between ground-state metal atoms and buffer gas ions. The sputtering systems have the advantage that no external heating devices are required, but independent control of the metal density and current is not possible.

Recently, it was demonstrated that hollow-cathode discharges could also be used to excite noble-gas (Ar<sup>+</sup>, Kr<sup>+</sup>, Xe<sup>+</sup>) ion lasers<sup>4-9</sup> with helium or neon as the buffer gas. Compared to metal-vapor lasers, these systems are simpler because optimum ratios of the interacting gases can be controlled independent of the discharge current. Most of these rare-gas-ion transitions were first observed by Laures, Dana, and Frapard in the afterglow of pulsed longitudinal discharge.<sup>10,11</sup> They suggested that the excitation mechanism for these lasers was excitation transfer between ground-state noble-gas ions and helium or neon metastables, for example:



Studies of pulsed microwave He-Kr discharges have been carried out by Kato *et al.* in which determinations of the plasma parameters were made.<sup>12</sup> In their studies they assumed that reaction (1) was the predominant excitation mechanism.

A second possible excitation mechanism which has not been discussed in previous work is charge-transfer reaction, e.g.,



Charge-transfer reactions are responsible for the efficient excitation in hollow-cathode metal-ion lasers<sup>1-3</sup> and therefore might be expected to occur in hollow-cathode rare-gas-ion lasers. Both reactions (1) and (2) have sufficient energy for populating the same upper energy levels of Kr<sup>+</sup>, Xe<sup>+</sup>, and Ar<sup>+</sup>. The question is then of the relative importance of these reactions under laser conditions.

We have investigated laser lines in He-Ar, He-Kr, Ne-Xe, and He-Ne-Xe mixtures. The hollow-cathode laser tube used was composed of a 75-cm-long cylindrical copper cathode and a stainless-steel anode. This system is described in more detail elsewhere.<sup>13</sup> The cavity optics consisted of two high-reflectivity (99.5%) mirrors, which were mounted internally. The wavelength measurements were made in air with a Jarrell Ash 0.5-m Ebert spectrometer, which had an accuracy of  $\pm 0.2$  Å. The laser lines were observed under quasi-cw conditions (pulse width 120  $\mu$ sec, pulse rate 40 Hz). Lasing occurred during the current pulse and up to 50  $\mu$ sec into the afterglow. Some of these laser lines were also observed to oscillate in the cw mode. Typical gas ratios were 1 : 600 for Kr-He, 1 : 40 for Ar-He, 1 : 300 for Xe-Ne, and 1 : 80 : 760 for Xe-Ne-He mixtures. The optimum total pressure was around 30 Torr for all systems.

Our results are summarized in Table I. We have listed all the observed laser lines, including new laser transitions and those which, as far as we know, have lased for the first time in a hollow-cathode discharge.

In order to determine the relative importance of reactions (1) and (2) in exciting upper laser levels, a third noble gas which had a lower ionization potential than the helium or neon metastables was introduced into the discharge. For example, in the case of He-Kr, Ar was added to the mixture. The He(2<sup>3</sup>S) density should be significantly reduced by the addition of Ar since Penning reactions between He(2<sup>3</sup>S) and ground-state Ar atoms have large cross sections.<sup>14</sup> The He<sup>+</sup> density should be less affected because of the absence of near-resonant Ar<sup>+</sup> excited states which could be populated by charge-transfer reactions involving ground-state Ar atoms. We simultaneously monitored the spontaneous emis-

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