A Dye Laser Pumped by an Air Laser

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A modified dye laser system operating at UNARC is discussed. The dye laser (peak wavelength ≈ 440 nm) is pumped by an air/N_2-laser. The dye is a new one, Tinopal, provided by CIBA Co. in Egypt. The dye laser/air laser characteristics are presented.

INTRODUCTION

The possibility of achieving optical gain (negative absorption) in complex molecules was noticed by Ivanov (1960) even before the development of lasers. Dye lasers have subsequently been pumped by flashlamps (Snavely, 1969), nitrogen lasers, and many other types of lasers (Schafer, 1973). N_2 lasers are very attractive lasers for excitation of a variety of dyes. The high efficiency of the N_2 laser pumping and its pulse characteristics allow efficient excitation of dyes with significant output. The continuously tunable wavelength output of a dye laser has made it possible to carry out experiments in high resolution spectroscopy. In these experiments radiation could be tuned to excite, selectively, any given transition within the wavelength range of the dye.

   In this article, we report preliminary detection and gain analysis of the radiation from a new dye (Tinopal) pumped by an air/N_2 laser. A solution of the radiative transfer equation in the dye allows experimental measurements of both absorption and emission coefficients. The pumping power of the system is large enough to exceed the threshold power required to lase dyes placed inside a Littrow cavity. Details of the pumping system and the dye laser arrangement are described.

APPARATUS

A schematic diagram of the dye laser and measuring system is shown in Figure 1. The pumping laser is an AVCO EVERETT N_2-laser. The gain of nitrogen lasers is so high that only one mirror is required to produce a pulse of power over 20 kWatt and duration less than 15 nanoseconds. This pulse duration is shorter than that necessary to cause significant excitation of the triplet states of dyes and hence triplet states losses are considered insignificant. The pulse shapes for two adjustments of the back reflection mirror are shown in Figure 2. A detailed account of calibration and characterisation apparatus is given by El-Raey and Amer (1982). Due to a small air leakage into the system and use of commercial nitrogen, the system produces low power. A power of near 10 kwatt/pulse is estimated. The relative average pumping power of the N_2 enriched air laser is monitored by a radiometer having a pyroelectric detector of flat spectral response. The N_2/air laser spectrum contains, in addition to the prominant 3371 A line of the second positive system of $N_2(C\,^3\Pi_u \rightarrow B\,^3\Pi_g)$, several ultraviolet lines of longer wavelengths associated with other components of air. Figure 3 shows the spectrum of the pumping air/N_2-laser at three different excitation voltages. The observed line
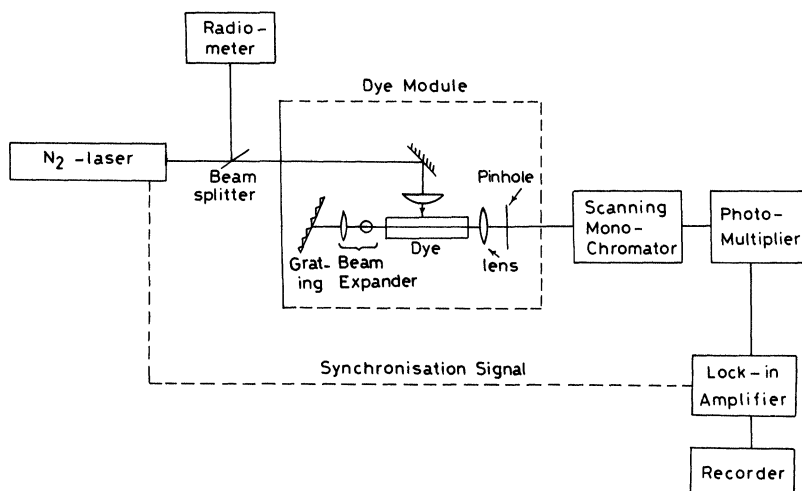

FIGURE 1   A schematic diagram of the dye laser and the pumping system.
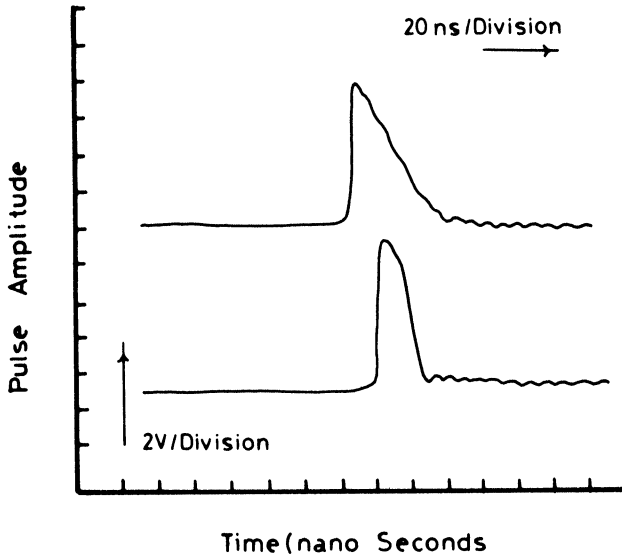
FIGURE 2   Pulse shape of the pumping laser for two adjustments of the back mirror of the air/N$_2$-laser.

profile represents the convolution of the real profile $I(\nu)$ with monochromator response function $A(\nu)$ of a bandwidth $\sim$3 A, according to

$$g(\nu) = \int I(\nu - \tau)A(\tau)\, d\tau \qquad (1)$$

This is the main factor responsible for the observed low amplitude and large bandwidth of the 3371 A line as compared to other lines.

The output of the air/N$_2$-laser is incident on a dye cell placed inside a cavity in a Littrow arrangement. A shutter is introduced inside the cavity so as to block the beam from reaching the grating. The amplified spontaneous emission (ASE) from the dye is synchronously detected by using a lock-in amplifier tuned to the pulse repetition frequency of the pumping laser. The gain spectra of several dyes of industrial interest are measured by the length dependence of ASE (Dienes and Madden, 1973; Shank *et al.*, 1970; Dienes *et al.*, 1973; Urisu and Kajiyama, 1976) based on a modified solution of the radiative transfer equation. The shutter is then removed and the dye laser cavity
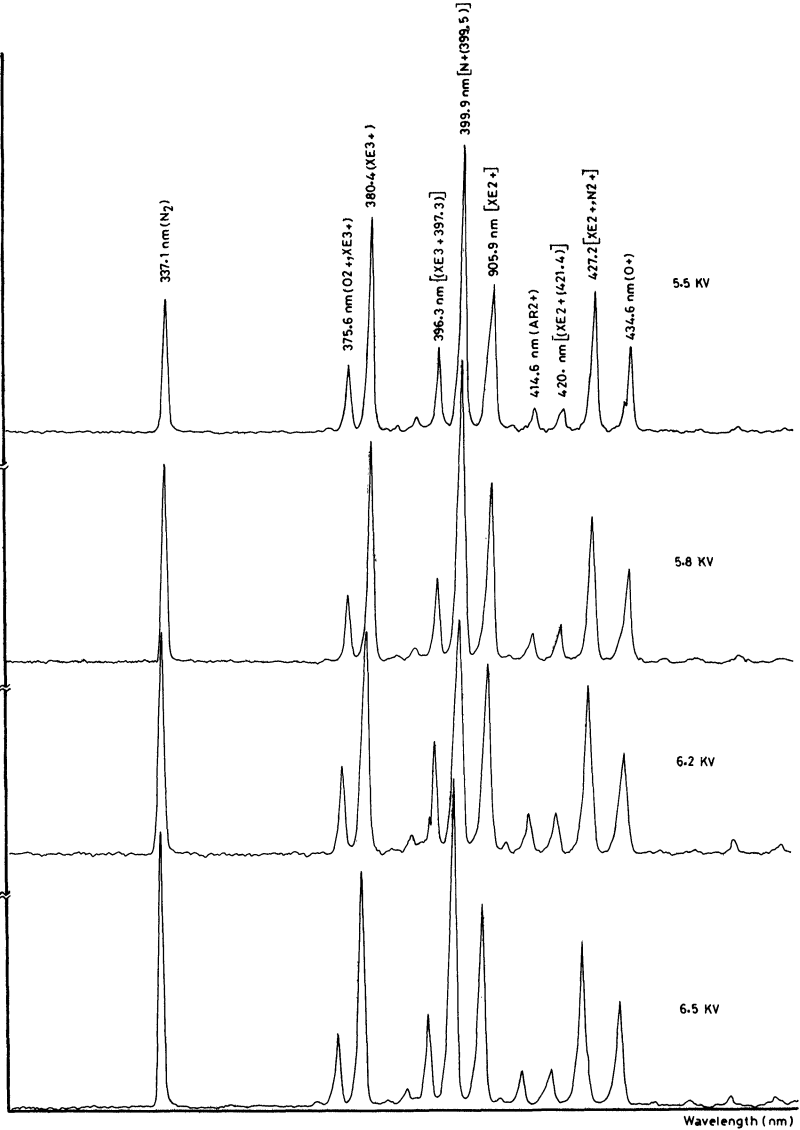
FIGURE 3   Spectrum of the pumping air/Nitrogen-laser broadened by convolution with instrumental response function. Lines are identified.

adjusted with the grating blazed near an angle of 54°. Spectral narrowing due to stimulated emission is observed.

GAIN SPECTRAL ANALYSIS

The gain spectrum of a dye solution is related to the fundamental parameters of the dye, and hence is a direct measure of the microscopic properties of the dye. The radiation incident on the cell excited molecules of the dye over the focusing line of the cylindrical lens. The spontaneous emission over all parts of the line is amplified by passing through the line to the detector. The intensity of the amplified radiation output (amplified spontaneous emission, ASE), is controlled by the radiative transfer equation inside the dye (Efthimiopoulos and Garside, 1981).

$$\frac{dI^{\pm}(x,\lambda)}{dx} = N_1(x)\left\{\sigma_e(\lambda) - \sigma_{01}\frac{N_0(x)}{N_1(x)} - \sigma_t(\lambda)\frac{N_t(x)}{N_1(x)} - \sigma_{12}(\lambda)\right\}I^{\pm}(x,\lambda)$$

$$+\frac{N_1(x)E(\lambda)F(x)}{\tau} \tag{2}$$

with usual notation and the geometry of Figure 4. The emission cross section $\sigma_e(\lambda)$ is related to the fluorescence spectrum $E(\lambda)$ via the equation

$$\sigma_e(\lambda) = \lambda^4 E(\lambda)/8\Pi\tau n^2 c \tag{3}$$
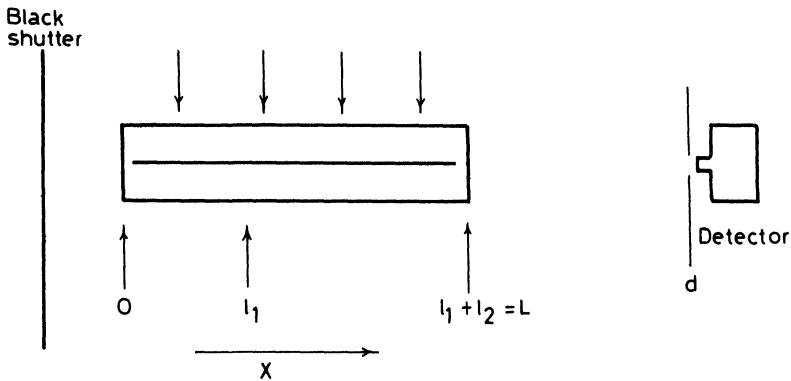

FIGURE 4   Pumping geometry of the dye.

where τ is the lifetime of the excited state and n is the refractive index of the solution. The function $F(x)$ is given by

$$F(x) = FA/2(L-x)^2$$

where A and L are the cross sectional area and length of the amplifier, and F is a parameter whose exact value is specified by direct comparison of experimental and theoretical results. If $I(x, \lambda)$ is the total photon flux per unit wavelength (photons s^{-1} cm^{-2} cm^{-1}) at a position x and wavelength λ, then

$$I(x, \lambda) = I^+(x, \lambda) + I^-(x, \lambda) \tag{4}$$

From (2), and (4), using (3), we get

$$\frac{dI(x, \lambda)}{dx} = [\alpha_\lambda(x) - \beta_\lambda(x)]I(x, \lambda) + S(x, \lambda) \tag{5}$$

where;

$$\alpha_\lambda(x) = N_1(x)\sigma_e(\lambda)$$

$$\beta(x) = N_1(x)\left\{\sigma_{01}(\lambda)\frac{N_0(x)}{N_1(x)} + \sigma_t(\lambda)\frac{N_t(x)}{N_1(x)} + \sigma_{12}(\lambda)\right\} \tag{6}$$

are the emission and absorption coefficients. The term $S(x, \lambda)$ in the right-hand side of the radiative transfer equation (5) is the spontaneous emission term and is given by

$$S(x, \lambda) = \frac{M_\lambda N_1(x)}{(d-x)^2} \tag{7}$$

where M_λ is a constant. The solution of the radiative transfer Eq. (5) when the back part of the cell of length l_1 is pumped is

$$I(\lambda) = M_\lambda e^{-\beta_\lambda l_2} \int_0^{l_1} \frac{N_1(x) \exp\left(\int^x [\alpha_\lambda(x') - \beta_\lambda(x')] dx'\right) dx}{(d-x)^2} \tag{8}$$

If we assume that the distribution of molecules is constant through the dye, then Eq. (8) could be written:

$$I_1(\lambda) = M_\lambda e^{-Bl_2} \int_0^{l_1} \frac{e^{(\alpha_\lambda - B_\lambda)x}}{(d-x)^2} dx \tag{9}$$

In our experiment $l_1 = l_2 = L/2$, and $L \ll d$. Under these conditions Eq. (9) becomes

$$I_1(\lambda) = K_\lambda \frac{e^{-B_\lambda L/2}}{(\alpha_\lambda - B_\lambda)} [e^{(\alpha_\lambda - B_\lambda)L/2} - 1] \qquad (10)$$

where K_λ is another constant. We have assumed that the gain is small enough so that the amplified spontaneous emissions do not saturate the transition. Similarly, the fluorescence intensity $I_2(\lambda)$ detected when the front $\frac{1}{2}$ of the cell is pumped, is

$$I_2(\lambda) = \frac{K_\lambda}{\alpha_\lambda - B_\lambda} [e^{(\alpha_\lambda - B_\lambda)L/2} - 1] \qquad (11)$$

when the entire cell is pumped

$$I_{1+2}(\lambda) = \frac{K_\lambda}{\alpha_\lambda - B_\lambda} [e^{(\alpha_\lambda - B_\lambda)L} - 1] \qquad (12)$$

From Eqs. (10) and (11), we get

$$B_\lambda = \frac{2}{L} \ln [I_2(\lambda)/I_1(\lambda)] \qquad (13)$$

and from (10), (11) and (12), the net gain coefficient;

$$\alpha_\lambda - B_\lambda = \frac{2}{L} \ln \left[\frac{I_{1+2}(\lambda)}{I_2(\lambda)} - 1 \right] \qquad (14)$$

Therefore, the emission coefficient is given by

$$\alpha_\lambda = \frac{2}{L} \left[\ln \left(\frac{I_2(\lambda)}{I_1(\lambda)} \right) + \ln \left(\frac{I_{1+2}(\lambda)}{I_2(\lambda)} - 1 \right) \right] \qquad (15)$$

from which both absorption and emission coefficients are determined.

Preliminary results of gain analysis gave unsaturated gain coefficients in excess of 1.0 cm^{-1} in Tinopal solutions near $\lambda = 450$ nm. To find the effective pump power density and hence obtain a measure of the excitation transfer distance, it is necessary to plot the variations of gain with pump power. Such a plot is shown in Figure 5, and is a straight line. The extrapolated y-axis intercept should equal the ground state loss (Dienes and Madden, 1973).
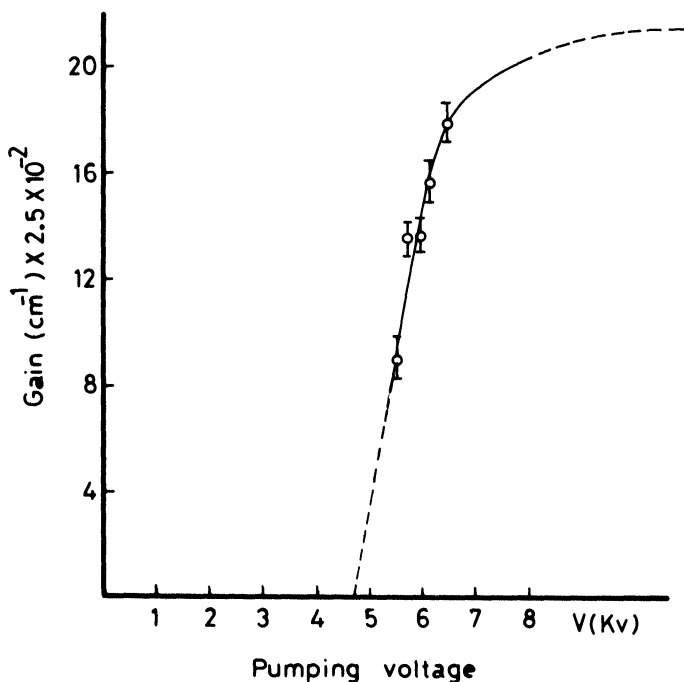
FIGURE 5   Gain variation with pumping power.

ANALYSIS OF THE DYE LASER

If the shutter inside the cavity is removed and the grating angle is adjusted, feedback and hence stimulated emission occurs with consequent spectral narrowing of the output. Figure 6 shows the dye laser output spectrum for three different pumping powers, superimposed on the amplified spontaneous emission of the dye (Tinopal of concentration 0.8 gm/one litre of water of pH = 7). The peak power of the dye laser output decreases nearly linearly with the pumping power. It should also be noted that the observed line widths are broadened by several factors: (a) convolution with the monochromator response function, (b) dye cell heating, (c) the cavity parameters. Adjustment of the expanding telescope and increasing the order of diffraction results in narrower bandwidths. It is planned to carry out measurements to increase pumping power of the dye by
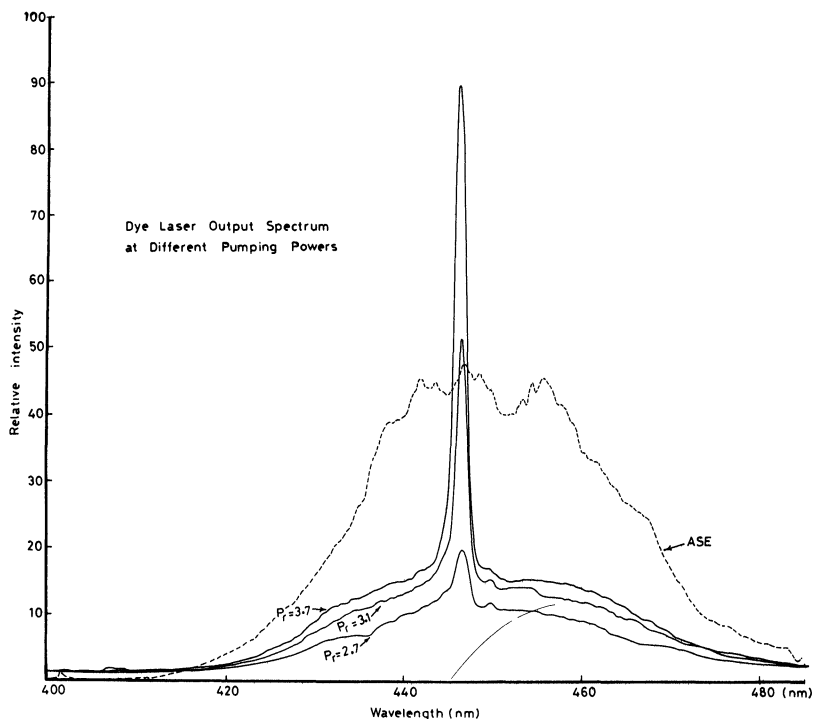
FIGURE 6   Dye laser output spectrum (convoluted with instrumental response). Amplified spontaneous emission of the same dye is shown for comparison.

increasing the rate of flow of the N$_2$ gas. Reduction of bandwidths is then possible for higher diffraction orders by using internal and/or external Fabry–Perot etalons.

CONCLUSION

A dye laser beam of tunable wavelength is detected from organic Tinopal solution, pumped by an air/N$_2$-laser. Preliminary measurements of gain spectra and pumping power variations are carried out. Study of polarisation effects, bandwidth narrowing and gain enhancements are in progress. Applications in detection of trace pollutants, high resolution spectroscopy and lifetime measurements are planned.

Acknowledgement

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