

# Intense laser emission from electron-beam-pumped ternary mixtures of Ar, N<sub>2</sub>, and POPOP vapor

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per unit strain. In writing Eq. (2), we have assumed that  $D$  does not show much variability among inorganic solids.

<sup>8</sup>See Ref. 6 for a table of structural factors.

<sup>9</sup>R.W.G. Wyckoff, *Crystal Structures* (Wiley, New York, 1965).

<sup>10</sup>References for curves in Fig. 1 as designated: (1) Amersil Pamphlet EM10012. (2) and (4) Naval Research Labs. ARPA Order No. 2031 Report No. 6, 1975 (unpublished). (3), (6), (7), (9), and (10) Raytheon Inc. ARPA Order No. 1180 (unpublished); F.A. Horrigan and T.F. Deutsch (unpublished); T.F. Deutsch and R.I. Rudko (unpublished). (11) S.S. Ballard, K.A. McCarthy, and W.L. Wolfe, *Optical Materials for Infrared Instrumentation* (University of Michigan Press, Ann Arbor, 1959), U.S. Dept. of Commerce PB181087. (5) L.R. Batsanova, G.S. Yurev, and V.P. Doronina, *J. Struct. Chem.* **9**, 63 (1968). (8) C.A. Angell, G.H. Wegdam, and J. van der Elsken, *Spectrochim. Acta* **30A**, 665 (1974).

<sup>11</sup>J.W. Fleming (private communication).

<sup>12</sup>S.H. Wemple (unpublished).

<sup>13</sup>Computed from Eqs. (4) and (6) using  $E_1 \approx 14$  meV ( $\mu = 57.45$ ,  $B = 0.6495$ ,  $d = 3.44$  Å) and  $E_d = 21$  eV;  $E_0 = 5.2$  eV (Ref. 6).

<sup>14</sup>D.L. Wood and J.P. Remeika, *J. Appl. Phys.* **38**, 1038 (1967).

<sup>15</sup>See Ref. 17.

<sup>16</sup>J.N. Plendl, in *Optical Properties of Solids*, edited by S. Nudelman and S.S. Mitra (Plenum, New York, 1969).

<sup>17</sup>In the notation of Ref. 6, the coefficient 42 in Eq. (5) is  $n_e f$ , where  $n_e$  equals the number of valence electrons per anion and  $f \approx 3.0$  eV is a bonding-type parameter describing ionic materials. In  $\text{ZnCl}_2$ , the specific choice for  $n_e$ , which determines  $E_0$ , depends upon the Zn  $d^{10}$  core contribution to interband transitions. The numerical factor in Eq. (5) assumes that  $n_e \approx 14$ , its value in AgCl. The possibility that  $n_e = 8$  (as in  $\text{SiO}_2$ , NaCl, ..., without  $d^{10}$  core contributions) is precluded by the resulting low  $E_0$  value of 5 eV, which is not compatible with a colorless solid. In the unlikely event that  $E_0 = 7$  eV, its value in AgCl (Ref. 6), we find that  $E_d = 12$  eV and  $\lambda_c \approx 3.9$  μm.

<sup>18</sup>*Handbook of Chemistry and Physics*, 53rd ed. (Chemical Rubber Co., Cleveland, 1973).

<sup>19</sup>W. Eppers, in *Handbook of Lasers*, edited by R.J. Pressley (Chemical Rubber Co., Cleveland, 1971), p. 39.

<sup>20</sup>H. Rawson, *Inorganic Glass-Forming Systems* (Academic London, 1967).

<sup>21</sup>M. Poulain, M. Chanthanasinh, and J. Lucas, *Mater. Res. Bull.* **12**, 151 (1977).

<sup>22</sup>I. Schulz, *Naturwissenschaften* **44**, 536 (1957).

## Intense laser emission from electron-beam-pumped ternary mixtures of Ar, N<sub>2</sub>, and POPOP vapor<sup>a)</sup>

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Powerful laser output was observed at 381 nm from electron-beam-pumped ternary mixtures of argon-nitrogen and 2,2'-p-phenylenebis(5-phenyloxazole) (POPOP) dye vapor. The injection of the subthreshold 380.5-nm N<sub>2</sub> line into the electrically excited gain profile of POPOP vapor has been identified as a pumping mechanism.

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Recently we reported superradiant emission from electron-beam-excited POPOP dye vapor buffered by argon.<sup>1</sup> In this paper we report the first observation of strong laser output at 381 nm from ternary mixtures of Ar, N<sub>2</sub>, and 2, 2'-p-phenylenebis (5-phenyloxazole) (POPOP) vapor. Cavity oscillations were found to be supported by a combination of N<sub>2</sub> and POPOP vapor. The energy deposited in the cavity into the argon species was found to be nearly a factor of 3 below the threshold for operation as a conventional Ar-N<sub>2</sub> transfer laser in the absence of the POPOP. Collisional and to a lesser-

degree radiative energy transfer from the electron-beam-excited N<sub>2</sub> are apparently responsible for the excitation of the dye vapor.

The apparatus used in this work consisted of a high-temperature high-pressure vapor cell with both electron-beam and optical access.<sup>2</sup> The cell could be filled with a variety of mixtures of different gases. The POPOP partial pressure was controlled by adjusting the temperature of the cell assembly with an oven enclosure. A prealigned plane-parallel optical resonator with high-reflectivity dielectric coatings ( $R = 96\%$  at  $385 \pm 10$  nm,  $R = 70\%$  at 360 nm) was located inside the cell. The cell was isolated from the cathode of the electron-beam machine by a 0.075-mm-thick titanium foil. This foil acted both as a pressure barrier and as the anode of the field-emission diode. A second 0.025-

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mm-thick foil, located 0.3 cm away from the first one inside the cell, was used as a thermal barrier to prevent condensation of dye on the relatively cool anode foil. All experiments were performed on an S<sup>3</sup> Apex I machine, configured for this experiment to produce a beam of 1-MeV electrons with a peak current of 28 kA in a triangular-shaped 30-nsec pulse which corresponded to an input energy of 500 J. The output from the cell was measured with a spectrograph which had a 1-nm resolution. Temporal characteristics of the output pulse were obtained with a fast-vacuum photodiode and a Tektronix R7912 transient digitizer.

Intense laser emission at 381 nm was observed in a 10-nsec pulse with less than 2-nm bandwidth and a 5-mrad beam divergence from an electron-beam-excited ternary mixture of 5 Torr POPOP, 2 atm Ar, and 4 atm N<sub>2</sub>. No attempt was made in this initial series of measurements to optimize power output from this system. Output powers in excess of 500 kW were observed, indicating a conversion efficiency from deposited electronic energy into the active volume to optical output of at least 0.3%. Laser action in the electron-beam-excited Ar-N<sub>2</sub>-POPOP mixture was found to have three important characteristics.

First, the presence of POPOP vapor at partial pressures of 0.1 to 5 Torr (corresponding to cell temperatures of 215 to 315 °C) significantly reduced the amount of argon buffer gas needed to attain laser action of Ar-N<sub>2</sub> at 357.7 nm, as shown in Fig. 1. Since total pressure of buffer gas, Ar + N<sub>2</sub>, was held constant at 6 atm, the lowering of the argon pressure is also associated with a decrease in stopping power due to the lower atomic number of nitrogen. For example, reduction of the argon buffer gas pressure from 6 to 2 atm and insertion of 4 atm nitrogen correspond to a lowering of the effective stopping power by 20%.<sup>3</sup> However, from Fig. 1 it can be seen that such a change lowered the concentration of Ar required to achieve laser threshold in the POPOP-N<sub>2</sub> system by almost a factor of 3 below that required in the absence of the POPOP. This behavior must be interpreted as indicating that less direct

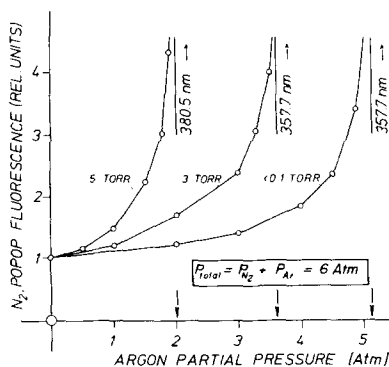


FIG. 1. Dependence of subthreshold fluorescence intensities from an Ar-N<sub>2</sub>-POPOP vapor ternary mixture upon argon partial pressure at three different dye partial pressures. Arrows show wavelength and argon pressures at laser threshold in presence of POPOP.

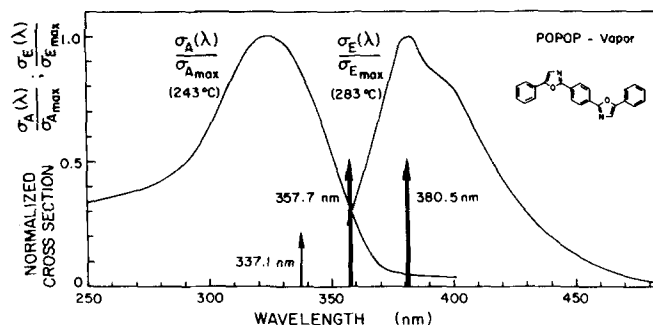


FIG. 2. Normalized absorption and fluorescence of POPOP vapor (Ref. 5). Spectral positions of Ar-N<sub>2</sub> laser lines are indicated.

energy transfer from the argon is necessary in order for N<sub>2</sub> to reach laser threshold. The behavior of an Ar-N<sub>2</sub> laser at elevated temperatures has been reported previously.<sup>4</sup> Based on those results one would have expected *a priori* an increase of laser threshold rather than a decrease with increasing POPOP vapor pressure. The reduction in laser threshold deposition in the argon species actually observed can be attributed to several factors including optical pumping via the directly excited 337.1-nm N<sub>2</sub> line which overlaps the POPOP absorption band as shown in Fig. 2. However, it is evident from Fig. 1 that at the higher-temperature conditions known to produce gain in POPOP,<sup>1</sup> laser emission of the low-gain 380.5-nm N<sub>2</sub> line occurred at an even lower threshold level of deposited energy into the argon kinetics.

The second set of experimental observations is shown in Fig. 3. This figure shows the spectral characteristics of the laser output under identical electron-beam-pumping conditions from a 6 atm Ar-N<sub>2</sub> mixture as the POPOP concentration was increased. With POPOP pressures from zero up to about 0.1 Torr, laser output at both 357.7 and 380.5 nm could be observed. As the POPOP partial pressure was increased above 0.1 Torr, the 380.5-nm line disappeared and only the intense 357.7-nm laser line remained. However, as the temperature was increased further to give a partial pres-

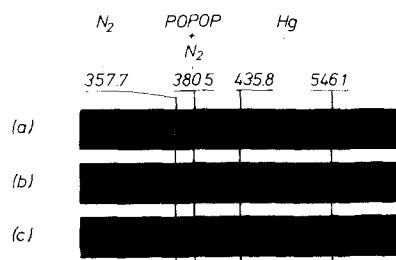


FIG. 3. Optical spectra showing (a) electron-beam-pumped Ar-N<sub>2</sub> laser operating at room temperature with Hg lines indicated for calibration (10% N<sub>2</sub> in Ar at 6 atm); (b) Ar-N<sub>2</sub> laser operating near threshold at 215 °C (≈ 0.1 Torr POPOP partial pressure, 20% N<sub>2</sub> in Ar at 6 atm); and (c) combined laser emission of Ar-N<sub>2</sub>-POPOP vapor mixture near threshold at 315 °C (≈ 5 Torr POPOP partial pressure, 65% N<sub>2</sub> in Ar at 6 atm).

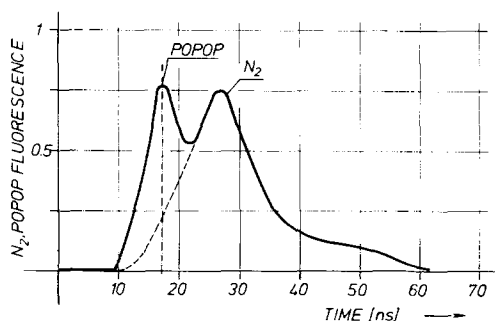


FIG. 4. Temporal characteristics of electron-beam-pumped Ar-N<sub>2</sub>-POPOP fluorescence redrawn from transient digitizer display. Data was taken at 5 Torr POPOP partial pressure, 1 atm Ar, and 5 atm N<sub>2</sub> buffer gas pressure.

sure of 5 Torr POPOP, the 357.7-nm emission vanished. Under these operating conditions, laser emission occurred only at 381 nm. Absorption quenching of the 357.7-nm N<sub>2</sub> line can be ruled out because of the enhanced laser output at reduced amounts of energy input to the argon kinetic pathways as a result of the reduced argon buffer gas requirements for this system. This is also supported by the observation that repeated excitation of the Ar-N<sub>2</sub>-POPOP mixture, without changing the gas in the cell, caused the 381-nm line to disappear as a result of dye degradation.

Finally, the temporal characteristics of the fluorescence output from the Ar-N<sub>2</sub>-POPOP mixture as shown in Fig. 4 provides further physical insight into the pumping mechanism responsible for laser emission. This fluorescence output is characterized by two distinct peaks separated by about 20 nsec. The initial peak with the fast rise and fall times is associated with POPOP fluorescence, while the broader slower peak is characteristic of the argon-buffered nitrogen fluorescence. The kinetics of the Ar-N<sub>2</sub> system are sufficiently slow that fluorescent intensity adequate to start cavity oscillations is not achieved within the first 5 nsec. During this time, however, the POPOP fluorescence reaches maximum intensity. The superposition of the beam-excited Ar-N<sub>2</sub> and POPOP inversion leads to laser action at 381 nm. Furthermore, the POPOP gain increases the chances of laser action at this longer wavelength by amplifying that component of the intensity circulating within the optical resonator. On the other hand, an electron-beam-excited binary mixture of argon-POPOP vapor while showing gain has so far exhibited no laser action, most likely because of the relatively short available interaction region, the selective absorption of the buffering system, and shock-wave perturbation of the plane-parallel optical resonator.

The mechanism by which the POPOP initiates laser action can be compared to injection locking.<sup>6</sup> However, instead of locking two laser lines, the subthreshold

380.5-nm line is enhanced by the broad spectral gain of POPOP vapor. Work is underway at present to perform measurements of the spectral dependence of gain for both the POPOP and nitrogen systems at elevated temperature, so that the relative importance of the two systems may be properly evaluated. Excitation for the intense POPOP fluorescence appears to be due to metastable argon atoms (Ar\*), argon molecules (Ar<sub>2</sub><sup>+</sup>), and metastable N<sub>2</sub> molecules. The most promising candidate for collisional excitation of the POPOP vapor appears to be the N<sub>2</sub>(B) state (for details see Ref. 7). The available excitation energy of 7.3 eV for N<sub>2</sub>(B) closely matches the energy of the second excitation state of POPOP, which could lead to a nearly resonant efficient energy transfer. Such an energy match proved to be an effective excitation scheme for a xenon-xanthene dye N92 vapor mixture.<sup>8</sup> Radiative energy transfer processes within the manifold of the nitrogen states, according to Ref. 7, are less likely at the high nitrogen buffer pressures used. From extrapolation of the rate constants given in Ref. 7, only 1% of the energy stored in the primarily produced N<sub>2</sub>(C) state can be radiatively transferred into the N<sub>2</sub>(B) state and finally into the dye molecule.

The injection-type pumping mechanism described above may also enable laser action to be achieved in other potential laser media with low gain. Organic dye vapors are available which fluoresce in the spectral region from 300 to 650 nm. For example, it should be possible to selectively enhance the 406-nm (0,3) or the 434-nm (0,4) components of the same C → B electronic transition of N<sub>2</sub> by means of N92 dye vapor.<sup>8</sup>

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