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Superradiant emission from electron-beam-excited POPOP vapor[a)]

G. Marowsky

Max-Planck-Institut für Biophysikalische Chemie, D-3400 Göttingen, Federal Republic of Germany

R. Cordray, F. K. Tittel, and W. L. Wilson

Department of Electrical Engineering, Rice University, Houston, Texas 77001
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For the first time superradiant emission from an electron-beam-excited dye vapor has been observed. Temporal and spectral characteristics are described that confirm the presence of stimulated emission. The wavelength integrated net gain of an optimized POPOP dye vapor–buffer gas mixture is found to be 0.17 cm^{-1} at 1 Torr and 4 atm argon pressures for an absorbed power input of 40 MW into an active volume of 10 cm^3. Under these conditions an estimate of the conversion efficiency from electronic energy to coherent light yields 5%. These results suggest that an efficient tunable electronically pumped dye vapor laser system is feasible.

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In this paper we wish to report the first observation of electron-beam-excited superradiant emission from the organic dye POPOP [*p*-phenylene-bis-(5.7 phenyl-2.-oxazole)] in the vapor phase buffered by high-pressure pure argon. In a series of earlier papers,[1-3] we have described procedures for establishing optimum electron-beam-excitation conditions of various organic dye vapor—buffer gas mixtures. Electron-beam pumping may be considered an important intermediate step in the development of a dye laser excited by direct electric discharge. Such a laser would be a more efficient system than an optically pumped liquid or vapor dye laser and would also result in a new tunable ultraviolet light source. The observed superradiant emission appears to be efficient when considering the energy processes involved in channeling stored energy from a suitable dense buffer gas to the dye vapor. Hence, electron-beam-excited dye vapor lasers appear feasible. A few technical problems, such as the need for a dye flow system to minimize quenching effects by fragmented dye molecules and a high-temperature solid compact optical resonator, still remain to be solved.

The key experimental parameter for studying potential laser performance of a rare-gas buffer—dye vapor mixture was found to be the effective gain of such a system and not its fluorescence yield. The experimental arrangement used to probe the gain along the optical axis of the reaction cell is shown in Fig. 1. The optical axis was located as close as mechanically feasible to the 3-mil-thick titanium anode foil of the field emission diode of the electron-beam accelerator (PI Pulserad 110) to ensure a maximum current density in the active region. The light emission was monitored by a photodiode placed 65 cm from the center of the reaction cell. The procedure for probing the gain essentially follows a method described in Refs. 4 and 5. In this method the fluorescence created in one-half the active volume irradiated by the electron beam serves as the source for probing the other half of the electron-beam-excited volume. If I_1 is the fluorescence intensity of an irradiated region 3 cm long and I_2 is the intensity of the entire 6-cm irradiated region, then the net unsaturated gain is $\alpha_{net} = (2/L)\ln(I_2/I_1 - 1)$. Opening and closing the left half of the reaction cell was achieved by means of a magnetically driven internal shutter system. A pair of samarium-cobalt alloy magnets with a high Curie temperature was used to withstand cell temperatures of up

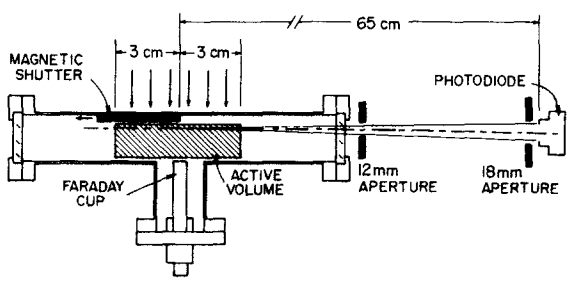

FIG. 1. Experimental arrangement for measurement of net gain using a magnetically driven shutter technique to probe gain in the active volume.

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to 450°C. The measured gain is corrected for the geometric factor associated with the finite distance of the photodiode from the center of the active region. The gain, α_{net}, is the measured gain figure when the system has overcome the various internal loss processes, such as singlet state reabsorption and triplet losses[6] by POPOP dye molecules and absorption of the buffer gas system. The suitability of various buffer gases was investigated in terms of minimum absorption and optimum energy transfer to the POPOP vapor. Several cw laser sources at 354, 441, and 476 nm were used to characterize the absorption of argon, xenon, and nitrogen. Xenon should be a good buffer gas because of its superior electron stopping power,[7] but from Fig. 2 it is evident that xenon introduces considerably higher losses than either argon or nitrogen. In the pressure range 3—4 atm, argon shows a typical decrease in transmitted laser intensity of 25% in a 6-cm-long optical cavity when pumped with a 10-kA 100-J electron beam. Based on this result and on earlier fluorescence studies,[3,8] argon was selected as the most suitable buffer gas.

Gain measurements were performed for different dye vapor and argon pressures. Homogeneity of the electron-pumped active region was verified by examining fluorescence from several 1-cm lengths of the active region by means of the externally operated shutter system. Two small apertures restricted the observed volume to minimize geometric effects due to changing the pumped length and to superradiant spatial narrowing. It was established that optimum gain conditions exist (at the axis of a potential intracell laser resonator) at POPOP vapor pressures between 0.5 and 5 Torr and at argon pressures of 3—4 atm. Figure 3(a) illustrates typical oscilloscope traces taken with an ITT FW114 photodiode—R7912 transient digitizer data acquisition system. The photodiode outputs obtained for both the partially irradiated 3-cm region and the entire 6-cm-long region are superimposed in Fig. 3(a). A considerable degree of temporal narrowing from 12 to 7 nsec and a faster rise and decay time of the pulse originating from the entire 6-cm-long active region is clearly evident. This pulse narrowing behavior occurred only for argon pressures greater than 1 atm. This can be considered a threshold condition of energy deposition required for the onset of superradiant emission. For

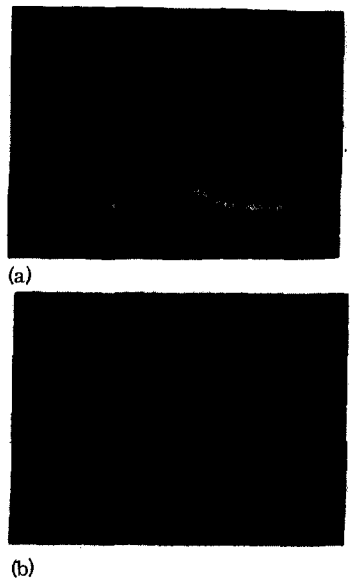

FIG. 3. Photodiode response to (a) POPOP dye fluorescence with 3-cm- and 6-cm-long active region and (b) Faraday cup current. Time scale for both (a) and (b) is 5 nsec/div.

comparison, the electron-beam-pump pulse behavior as monitored by an intracell Faraday cup probe is shown in Fig. 3(b). Additional evidence for superradiant emission from such a system is obtained from spectrographic measurements. Figure 4 shows the extremely high degree of spectral narrowing of the electron-beam-excited fluorescence spectrum centered at 385 nm from a bandwidth of 50 nm at 1 atm argon to 2 nm at 4 atm. The 385-nm peak shifts slightly to shorter wavelengths when decreasing the dye vapor pressure to 0.5 Torr and to longer wavelengths with pressures up to 5 Torr. Such a wavelength shift is also observed in optically pumped organic dyes and is due to the interaction between singlet-singlet absorption and fluorescence.[9] The spectral characteristics were determined with a grating spectrograph of 0.1 nm resolution. Both spectra in Fig. 4 are plotted from densitometric evaluation of spectral data without taking into account the non-

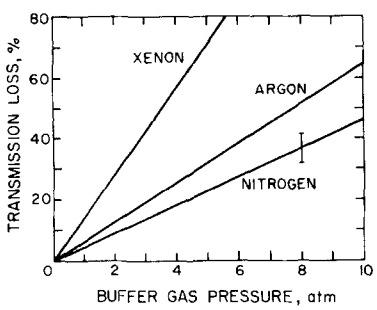

FIG. 2. Dependence of transmission loss on buffer gas pressure for xenon, argon, and nitrogen obtained by averaging absorption data taken at 354, 441, and 476 nm. Typical scatter of data is shown by error bar for nitrogen.

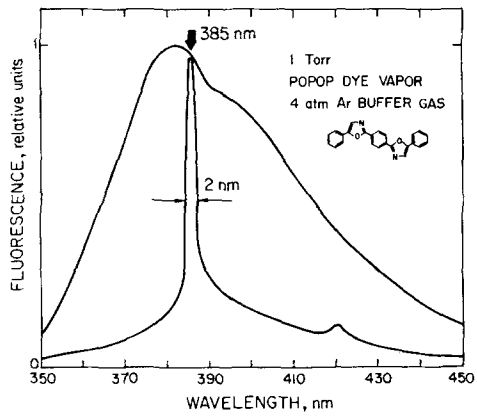

FIG. 4. Comparison of electron-beam-excited dye vapor fluorescence spectra, taken below superradiant laser threshold (1 atm argon buffer gas pressure) and above (4 atm argon buffer gas pressure) in relative units. Wavelength integrated fluorescence power is four times higher above threshold than below.

linear characteristics of the recording material. The contribution from the original fluorescence distribution is apparent as a background in the superradiant emission spectra. The wavelength integrated output increased typically by a factor of 4 from 250 W below threshold to 1 kW above. However, the spectral density increased by a factor of at least two orders of magnitude at the line center. It was found that superradiant emission was quenched by impurities in the buffer gas. Hence, the dye cell was pumped out to less than 10^{-3} Torr before all experiments, and ultrahigh-purity argon (99.999%) was used. The addition of 0.1 Torr of nitrogen reduced the fluorescence intensity for 1 Torr of POPOP dye vapor by 50% and completely quenched its net gain. The presence of a small peak at 420 nm in the fluorescence spectra can be attributed to a nitrogen line from nitrogen produced by dye fragmentation due to intense electron-beam pumping. In fact, the fluorescence output and net gain decreased with each successive electron beam pulse. Hence, both dye and buffer gas were removed from the cell and replaced after each pulse.

So far we have only observed net gain between 0.5 and 5 Torr dye vapor pressures. A typical gain for POPOP dye at 1 Torr and 4 atm argon buffer gas pressure is 0.17 cm^{-1}. This wavelength integrated net gain value is considerably higher than the gain measured for optically excited dye vapors[10] if one takes into account the dye vapor pressure dependence and integrates the gain over the whole fluorescence band. The efficiency, in terms of the ratio of optical output energy to the electron energy deposited in an active volume of 10 cm^3, can be estimated by considering the known amount of energy deposited in the active volume and the threshold requirements for population inversion.[9] The energy deposition in the argon buffer gas is approximately 0.4 J and the power input 40 MW. From these two quantities and the lifetime of an excited dye molecule, an inversion of 7% of the dye molecules present can be estimated. This inversion requires deexcitation of three argon metastables to create one excited POPOP dye molecule which subsequently can release a photon energy of 3.8 eV. Since the production of one argon metastable requires an energy of above 25 eV,[11] the estimated efficiency appears to be 3.8/75 or 5% at a dye vapor pressure of 1 Torr. At higher dye pressures this efficiency figure should increase. It should also be higher under laser conditions in the presence of optical feedback from a resonator, when the competing quenching processes (absorption by impurities and breakdown products) will be less effective.[12]

In conclusion, electron-beam-excited POPOP dye vapor has exhibited superradiant emission in a carefully optimized dye vapor–argon buffer gas mixture that minimizes various loss processes associated with intrinsic dye and rare-gas buffer absorption. The observed high degree of spectral and temporal narrowing together with considerable net gain and efficiency make electron-beam-excited organic dye vapors attractive for the development of efficient scalable tunable lasers.

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