

COMPARISON BETWEEN ELECTRON BEAM AND OPTICALLY PRODUCED MERCURY EXCIMER FLUORESCENCE*

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Significant differences have been observed between the mercury excimer fluorescence produced by electron beam and optical excitation. We have used time resolved fluorescent spectroscopy to study the evolution of the excimer system following a 4 ns optical excitation pulse.

When mercury vapor is optically excited at or near the resonance line (253.7 nm), prominent excimer fluorescence bands are observed at 335 nm and 500 nm. These bands have been the subject of extensive study by numerous groups [1] and the excimer to ground state dissociation transitions which produce them have been suggested as potential high power laser candidates [2]. This has led several groups to use high power, short pulse, beams of relativistic electrons to excite mercury vapor in an attempt to reach a high excimer concentration and possible laser action. This type of excitation leads to fluorescence emissions which differ in time and wavelength from those produced optically [3,4].

We have been working on a system of measurement techniques based on optical excitation to characterize metal vapor excimer systems such as mercury. With this excitation scheme we use photons with energy slightly less than the atomic resonance transitions to produce excimers directly in the lowest bound state which is dipole connected to the ground state (see fig. 1). This is a very clean excitation scheme and avoids the complicating presence of electrons and population in highly excited states. Part of this work has involved

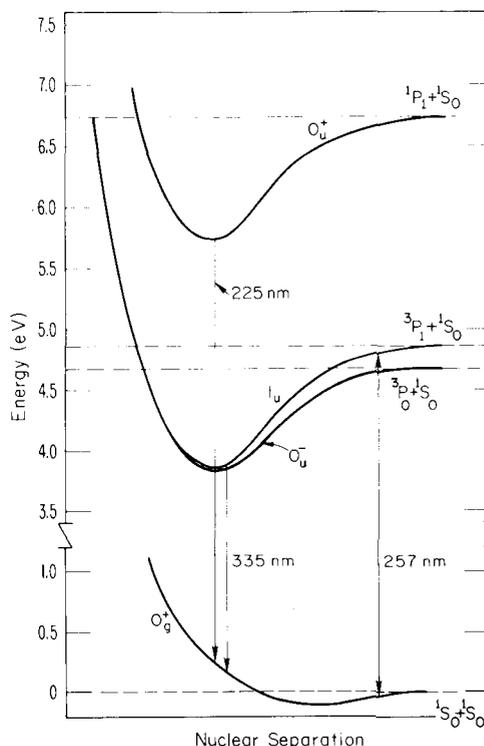


Fig. 1. Pertinent Hg_2 potential curves. The source of the 485 nm band has not been shown because we believe it is an Hg_3 surface which lies some 6500 cm^{-1} below the bottom of the 31_u curve [5].

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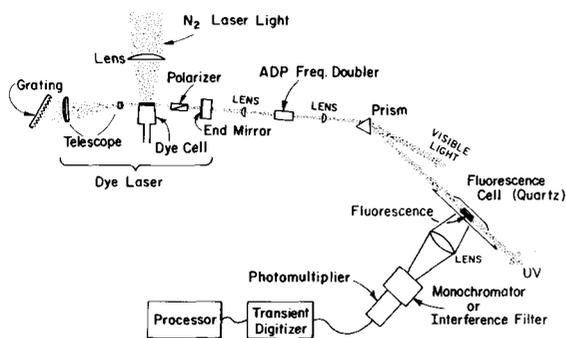


Fig. 2. Experimental setup for time resolved spectroscopy of optically pumped Hg_2 .

short pulse excitation of the excimers by a nitrogen pumped, frequency doubled, dye laser (pulse length ≈ 4 ns) and subsequent time resolved spectroscopy of the evolving excimer population. The experimental apparatus is shown in fig. 2.

The spectra are acquired in the following manner: A portion of the fluorescence spectrum is observed through a monochromator (resolution ≈ 2 nm) and detected on a fast photomultiplier. A transient digitizer records the fluorescence intensity with a time resolution of 10 ns and subsequent shots are averaged and normalized to the laser energy in a minicomputer. After each average is acquired, it is stored on magnetic tape and the spectrometer is stepped to a new wavelength. Once the spectral region of interest has been covered, the magnetic tape is read into a large computer where spectra are reconstructed by taking constant time slices through the data. These spectra are then calibrated for the spectral response of the monochromator-photomultiplier system to return intensity (in photons/s) versus wavelength plots. Spectra generated in this manner are shown in fig. 3 (solid curve) along with the time resolved spectra following e-beam excitation of ref. [4] (dashed curve).

The time integrated spectra following e-beam excitation [4] shows a pronounced hole at 500 nm. This feature shows up as a dip at 510 nm in the time resolved spectra which quickly fills in as the wings collapse. This behavior has been interpreted by Schlie et al. [4] as being due to radiation from a non-thermal population distribution at early times following the e-beam pulse. However, under identical conditions of pressure

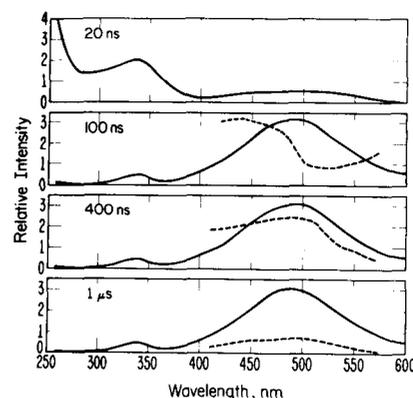


Fig. 3. Time resolved spectra: optically excited (solid curve), electron beam excited (dashed curve). Experimental conditions of temperature and vapor density are: $T = 680$ K, $N = 2.5 \times 10^{19} \text{ cm}^{-3}$.

and temperature, our spectra (fig. 3) show vibrational equilibration within the 485 nm band takes less than 20 ns. The fact that we see vibrational equilibrium within the 485 nm band even at very early times, combined with the decay rates of the "440 nm" and "530 nm" bands which are much too fast to be related to the 485 nm band and different even from each other, leads us to the conclusion that they arise from radiative relaxation of higher levels, populated as a result of the e-beam pulse.

The time integrated fluorescence emission in ref. [4] is wholly dominated by the "440 nm" and "530 nm" bands. This would imply each cascading excimer does not produce an excimer in the lowest bound state and subsequent radiation to the ground state, as has been assumed [3] since that would result in a "485 nm" band of intensity equal to that of the "440 nm" and "530 nm" bands. This apparent low quantum yield on the lowest excimer to ground state transition could be accounted for if the cascading excimers largely get trapped in the $1^1O_u^+$ state and radiate to the ground state at 225 nm (see fig. 1). This region has not been observed when e-beam excitation has been used.

A second conclusion which follows from the cascading argument concerns the bi-excimer destruction rate derived in ref. [3]. If the dominant radiation at 457 nm is not from the 485 nm band as assumed but rather from the cascading "440 nm" band, then the observed non-exponential decay could well have some explanation other than bi-excimer destruction.

References

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