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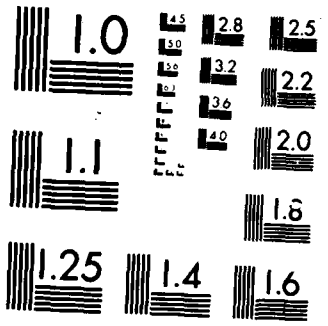

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LASER PROBING AND MODIFICATION OF MOCVD AND VPE REACTOR KINETICS

FINAL REPORT

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OCTOBER 1985

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Experiments are described in which the potential of dye vapor and HgBr as tunable visible lasers is under investigation. A near UV optically pumped HgBr laser has been demonstrated and transient excited-state absorption in HgBr' has been observed. Intense stable discharges in POPOP and coumarin 6 vapor have been obtained and gain measurements are underway.

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J. G. Eden (Book Review), J. Opt. Soc. Am. B 2, p. 390 (1985).
D. P. Greene and J. G. Eden, "Transient absorption in a cadmium monoiodide laser discharge," Opt. Comm. 53, pp. 263-268 (1985).
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I. INTRODUCTION

Over the last two years, the Army Research Office has supported experimental work in laser physics here at the University of Illinois. The goal of this research program is two-fold: 1) to develop new sources of tunable coherent radiation in the visible and ultraviolet and 2) to better understand the fundamental processes occurring in existing sources and particularly those that show promise for improvement in efficiency and output power.

This work has proven to be very fruitful in both areas. Specifically, the following accomplishments have been realized in the last year alone:

1) Stable, streamer-free discharges have been obtained in coumarin 6 and POPOP vapors - glows have been sustained at PRF's of 1-5 Hz for several hours with no noticeable deterioration in fluorescence intensity from the active region.
2) Excited state absorption of the cadmium monoiodide (CdI) and mercury-bromide (HgBr) molecules has been observed and spectroscopically assigned for the first time.
3) The X → B absorption band of HgBr has been observed.
4) An HgBr laser at 502 nm, optically pumped at 351 nm (XeF) or 355 nm (3 X Nd:YAG), has been demonstrated.
5) A flashlamp-pumped HgBr laser has been demonstrated.

II. DESCRIPTION OF EXPERIMENTAL RESULTS

A. HgBr Excited State Absorption

Recent experiments have involved an HgBr laser in which we have observed the analogous excited state (charge transfer) band as that observed previously for CdI. In HgBr, this absorption peaks near 447 nm. Figure 1 shows an expanded view of the blue absorption spectrum superimposed on the HgBr B → X fluorescence profile. An aside that is important insofar as tunable lasers are concerned is that, at the same time these measurements were made, we discovered that the HgBr B → X band exhibits gain as far to the blue as 469 nm! Therefore, this oscillator is potentially tunable over as much as ~ 30 nm.

The identity of the HgBr blue band was also confirmed by exciting the active medium with a dye laser pulse of wavelength $444 \leq \lambda \leq 454$ nm while at the same time monitoring the gain at 502 nm (peak of B → X band). As shown by Figure 2, when the wavelength of the first dye laser pulse coincided with one of the absorption peaks of Figure 1, a strong suppression in the 502 nm gain waveform was observed. At other wavelengths (between the absorption peaks or outside the band), little or no effect on the 502 nm gain was detectable. Of course, the strongest suppression was observed when $\lambda = 446.98$ nm and the gain suppression at other absorption peaks is proportional to the absorption strength.

By varying the laser fluence at $\lambda \sim 447$ nm and measuring the transmitted blue intensity, the "saturation intensity" of the HgBr excited

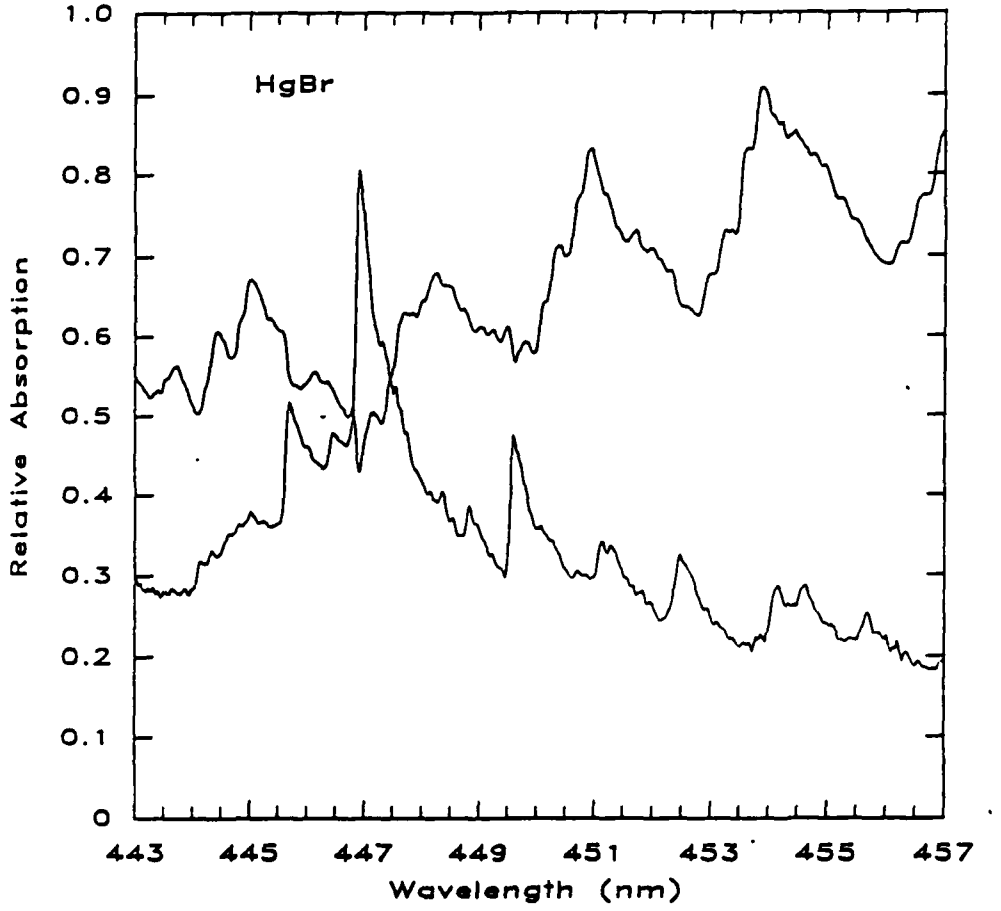

Figure 1. HgBr (B → X) fluorescence spectrum (top trace) overlapped with the $HgBr^*$ (B) absorption spectrum (bottom).

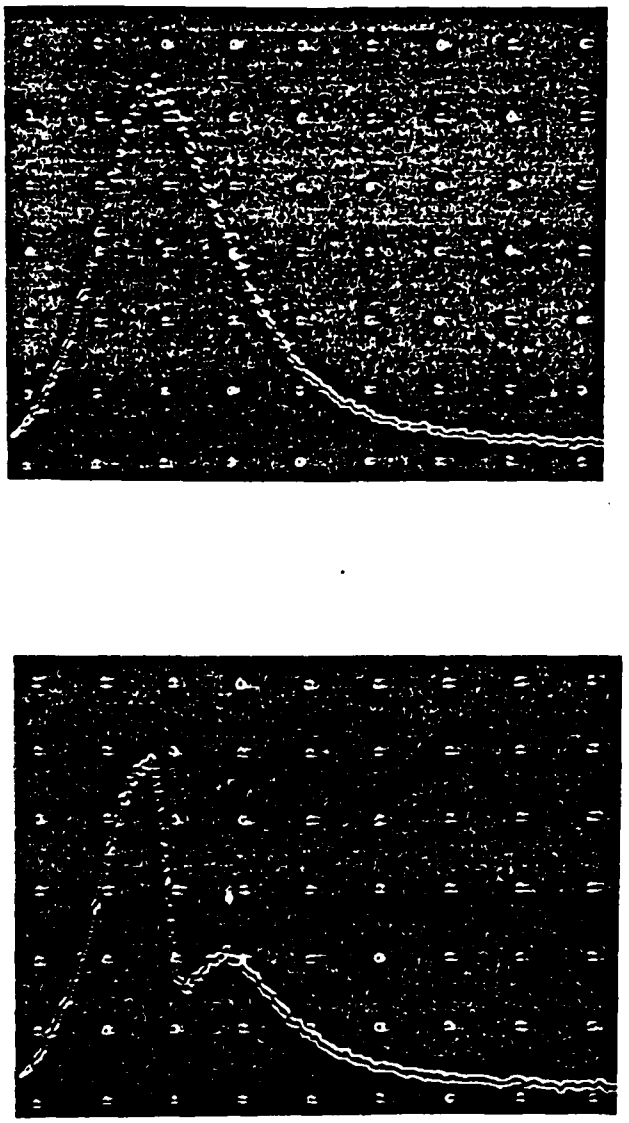

Figure 2. (Top) Gain at 502 nm in HgBr laser discharge; (Bottom) Suppression of gain when laser plasma is irradiated at 446.98 nm. Peak suppression is 70% and the dye laser pulse was timed to arrive at the point of peak gain. Time base: 50 ns/div.

state absorption band was measured to be 710 kW - cm^{-2}. Similarly, the absorption cross-section at 446.98 nm is $3 \cdot 10^{-17}$ cm^2. Finally, it should be mentioned that the structure of the band shown in Figure 1 is consistent with an HgBr absorption band originating from the B state - the energy spacing between alternate peaks in Figure 1 is close to the known ω_e for $B^2\Sigma(\sim 139\ cm^{-1})$.

We conclude that excited state absorption in CdI and HgBr has been observed for the first time though the existence of such bands had been predicted theoretically for at least a decade. These results are currently being written up for publication and will be submitted to the Journal of Chemical Physics.

B. Optically Pumped HgBr Laser

In the last six months, we have uncovered a simple and efficient way to pump the HgBr laser - a method which we believe is amenable to CW operation. In the course of examining the transient absorption spectrum for the HgBr discharge laser, we unexpectedly observed the HgBr X → B absorption band. Figure 3 shows the X → B absorption spectrum of HgBr that was measured with a pulsed lamp and OMA in the afterglow of a discharge pumped HgBr laser based on dissociation of $HgBr_2$.

Because of the relatively low photon energies required to access the $B^2\Sigma$ state from ground (~ 3.5 eV), one can quickly envision an optically-pumped HgBr laser having a high quantum efficiency (2.5 eV/3.5 eV = 71%). Preliminary experiments to explore this possibility have been conducted. An ~ 8 ns pulse from a frequency tripled, Q-switched Nd:YAG (λ = 355 nm) or XeF laser was directed along the axis of a discharge-pumped HgBr laser

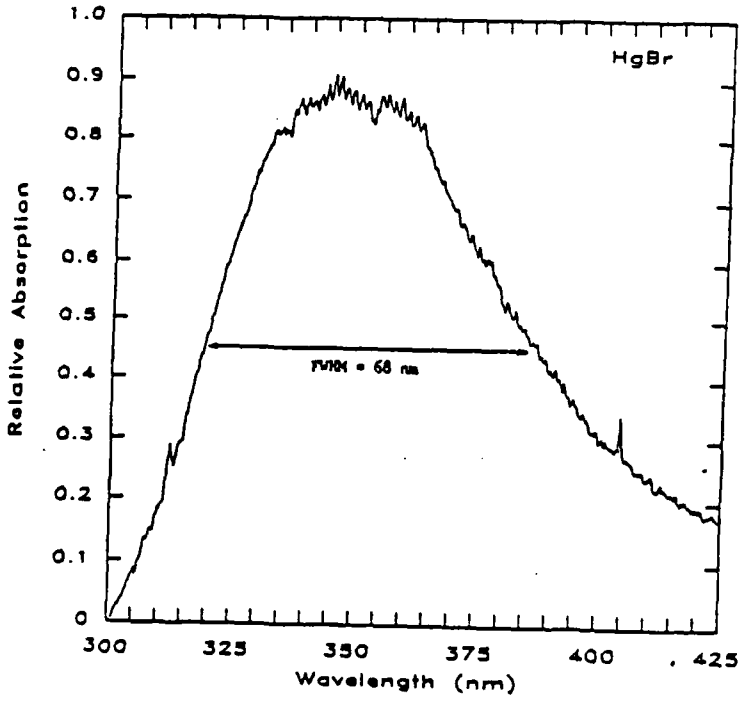

Figure 3. Ground state HgBr absorption spectrum in the near-ultraviolet that was recorded in the afterglow of a pulsed HgBr laser discharge.

(active length of 50 cm $He/N_2/HgBr_2$ gas mixture). Fired by itself, the discharge produces an ~ 40 ns FWHM HgBr laser pulse. Once the 355 nm laser pulse arrives, a second HgBr pulse is generated having a width of ~ 10 ns. The efficiency for converting XeF radiation (λ = 351 nm) into blue-green output is 23% which corresponds to a photon conversion efficiency of approximately 1/3. These data are extremely reproducible.

The obvious match between the peak in the X → B absorption spectrum of Figure 3 and the spectral output of a xenon flashlamp prompted us to explore the possibility of a pulsed, coaxial flashlamp-pumped HgBr laser. Several milligrams of natural abundance $HgBr_2$ and 760 Torr of argon buffer gas were contained in a quartz cell having Suprasil quartz Brewster windows. Lasing was obtained with a high-Q cavity (1% output coupling) - larger couplings have not yet been tried. A pulse of ~ 280 ns FWHM is produced.

C. Dye Vapor Experiments

One of the major objectives of this research program when it was proposed to ARO two years ago was to explore the possibility of a discharge-pumped dye vapor laser. At that time, it was noted that short discharge current risetimes and the proper choice of dye would be essential to minimizing dye fragmentation.

Since then, considerable progress has been made towards this goal. Intense, stable, streamer-free, glow discharges have been obtained in both POPOP and coumarin 6 dye vapor. We consider this alone to be a significant development since the discharge current pulsewidth is 50-80 ns, depending on the buffer gas pressure. At the present time, the discharge cell has

the following parameters: current risetime - 50 ns, approximate specific power loading at 1 atm - 2 to 5 MW - cm^{-3}, equivalent series inductance - 85 nH and an active volume of 10 cm^3.

One of our most significant observations is that even for specific power loadings of ~ 5 MW - cm^{-3}, dye fragmentation is not a serious problem. At a pulse repetition frequency of 1 Hz, the discharge has been operated for several hours without a noticeable deterioration in the fluorescence intensity from the excited dye vapor.

The brightness of the discharges in both POPOP and coumarin 6 vapors prompted us to make a preliminary search for gain. However, the only laser probe source available in our laboratory (until recently) is an Ar^+ ion laser. Therefore, to date, gain measurements have only been carried out at 457.9, 476.5, 488.0 and 514.4 nm in coumarin vapor at ≈ 270°C (coumarin number density ~ $3 \cdot 10^{16}$ cm^{-3}). At these wavelengths, only optical loss is observed which is not surprising since gain would only be expected to exist in the ~ 525-550 nm spectral region.

Therefore, the possible presence of gain in the system is inconclusive and it will be necessary to measure the entire gain absorption spectra of these molecules using an optical multichannel analyzer (OMA) or a CW dye laser.

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