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APR 80 C S LIU, D W FELDMAN, J L PACK N00014-78-C-0131
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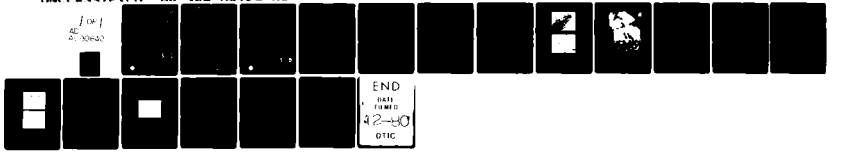

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THALLIUM MERCURY LASER DEVELOPMENT

C. S. Liu, D. W. Feldman and J. L. Pack

FINAL REPORT (PHASE II)

(Period between Feb. 1, 1979 and Jan. 31, 1980)

Contract No. N00014-78-0131

Office of Naval Research

April 17, 1980

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REPORT DOCUMENTATION PAGE

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1. REPORT NUMBER: 80-5-23-007	2. GOVT ACCESSION NO.
3. RECIPIENT'S CATALOG NUMBER	
4. TITLE (and Subtitle): Thallium Mercury Laser Development	5. TYPE OF REPORT & PERIOD COVERED: Final Report (Phase II) Feb. 1, 1979 to Jan. 31, 1980
6. PERFORMING ORG. REPORT NUMBER: 80-9C2-HOTEX-R1	
7. AUTHOR(s): C. S. Liu, D. W. Feldman and J. L. Pack	8. CONTRACT OR GRANT NUMBER(s): N00014-78-C-0131
9. PERFORMING ORGANIZATION NAME AND ADDRESS: Westinghouse R&D Center, 1310 Beulah Road, Pittsburgh, PA 15235	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS
11. CONTROLLING OFFICE NAME AND ADDRESS: ONR, Department of the Navy, 800 N. Quincy Street, Arlington, VA 22217	12. REPORT DATE: April 1980
	13. NUMBER OF PAGES: [redacted] 16
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)	15. SECURITY CLASS. (of this report): Unclassified
	15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Unclassified/Unlimited

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

thallium, laser, pulses, fluorescence, recombination, mercury, excimer, preionization, molecules, ionization, discharges, excitation, emission, visible, vapor, collisions, gain, absorption, mercury

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The major effort was to perform definitive gain measurements on discharge-excited Tl-Hg laser mixtures and to demonstrate TlHg laser output at 459 nm if the gain measurement indicated that laser tests were warranted. Gain and absorption measurements of the discharge containing high densities of mercury (3×10^{19} cm^{-3}) and thallium (6×10^{16} cm^{-3}) were undertaken. No transient gain was observed within the TlHg* excimer band at 459 nm; on the contrary, a strong absorption from Hg_2^* molecules in this region was detected.

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1. INTRODUCTION AND SUMMARY

This report summarizes research work performed at the Westinghouse R&D Center under ONR Contract No. N00014-78-C-0131 "Thallium Mercury Laser Development", for the period between Feb. 1, 1979 and January 31, 1980. The major effort was to perform definitive gain measurements on discharge-excited Tl-Hg laser mixtures and to demonstrate TlHg laser output at 459 nm if the gain measurement indicated that laser tests were warranted. Gain and absorption measurements of the discharge containing high densities of mercury (3×10^{19} cm^{-3}) and thallium (6×10^{16} cm^{-3}) were undertaken. No transient gain was observed within the $TlHg^*$ excimer band at 459 nm; on the contrary, a strong absorption from Hg_2^* molecules[1] in this region was detected.

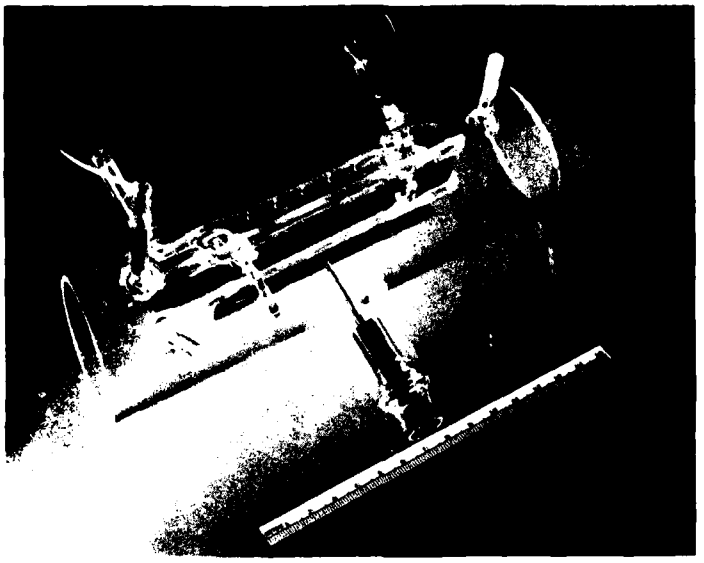
2. EXPERIMENTS AND RESULTS

A self-sustained glow discharge was obtained in Tl-Hg vapors at temperatures up to 900°C and pressures up to 4 atm. These pulsed transverse discharges, preionized by UV spark sources placed behind a screen anode, were diffuse and had dimensions of 8 cm length x 1 cm gap x 0.5 cm width ($V = 4\ cm^3$). At temperature above 700°C, strong TlHg emissions were observed at the 459 nm and 656 nm excimer bands.

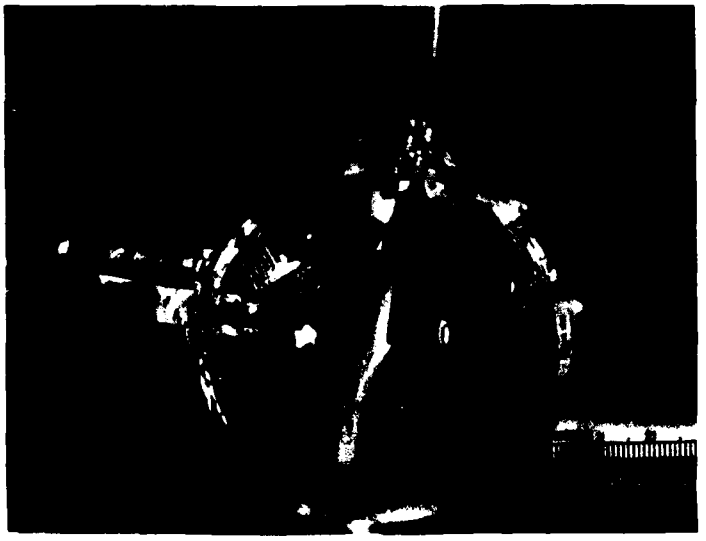
The discharge cell was fabricated entirely from quartz, including two quartz windows which were glass blown to each end for optical measurements along the tube length. Electrical feedthroughs consisted of quartz - molybdenum seals, and internal electrode structures were fabricated from molybdenum as shown in Figure 1. Measured amounts of mercury ($3 \times 10^{19}\ cm^{-3}$) and excess thallium were introduced into the cell under high vacuum conditions and the tube was then sealed. Thus the discharge cell operated under all hot, sealed-off conditions with only quartz and molybdenum in contact with the discharge medium. An external oven was used to raise the tube temperature up to ∿900°C. A stainless steel pressure vessel, as shown in Figure 2, surrounded the tube and oven and was filled with an inert gas to equalize the pressure across the quartz discharge cell walls. Three windows in the pressure vessel side wall permitted visual observation of the diffuse discharge.

A typical glow voltage for Tl-Hg discharges operating at 900°C was 10 kV. Under these conditions, the mercury density was $3 \times 10^{19}\ cm^{-3}$ and the thallium density was $\sim 3 \times 10^{16}\ cm^{-3}$. The electric field-to-vapor density ratio, E/N, is $3.3 \times 10^{-16}\ V\ cm^2$ and the current pulse width is ∿150 nsec.

Figure 3 shows a schematic of the spectroscopic apparatus. The time-integrated fluorescence emitted along the discharge tube axis is shown in Figure 4 for a Tl-Hg discharge operating at 900°C. The observed emission spectra were



(a)



(b)

Figure 1 — TlHg discharge tube with preionizers behind the screen anode.

Figure 2 — High-pressure vessel employed to contain UV-preionized, transverse discharge TlHg laser discharges at pressures up to 250 psia and temperatures up to 1000°C.

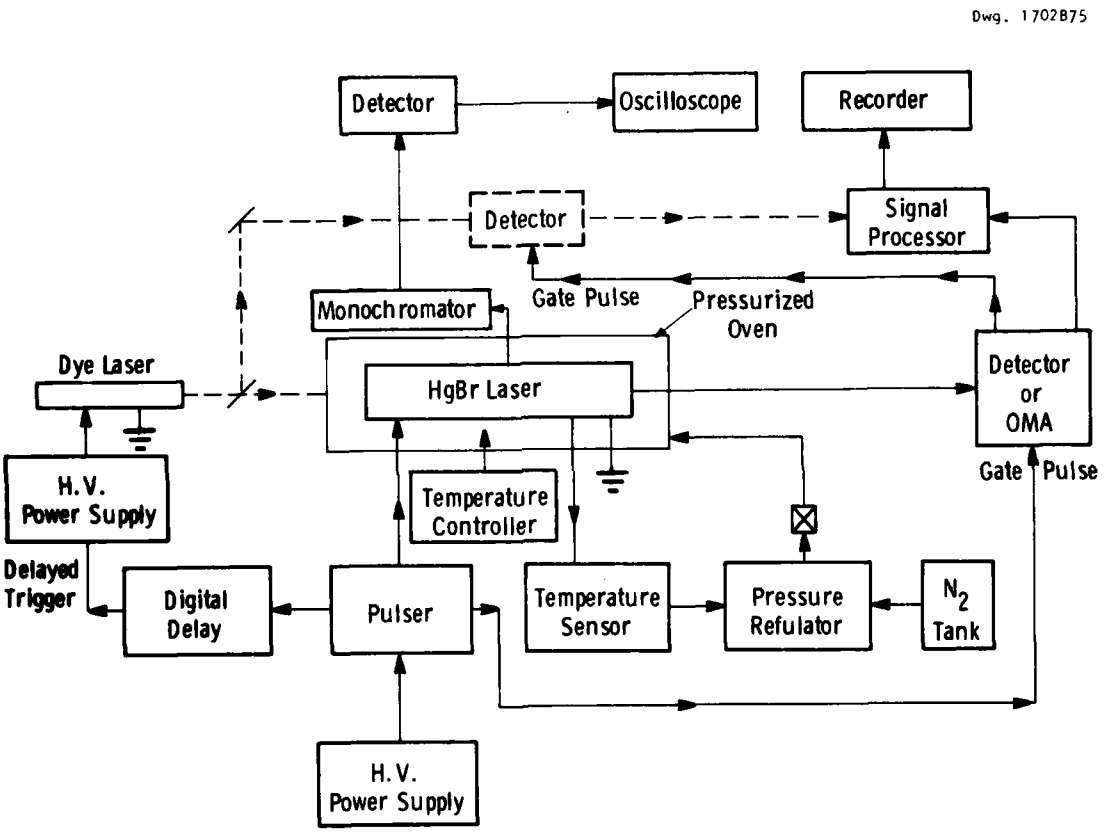


Figure 3. Spectroscopic apparatus designed to measure the fluorescence, absorption and gain features of pulsed TlHg discharges.

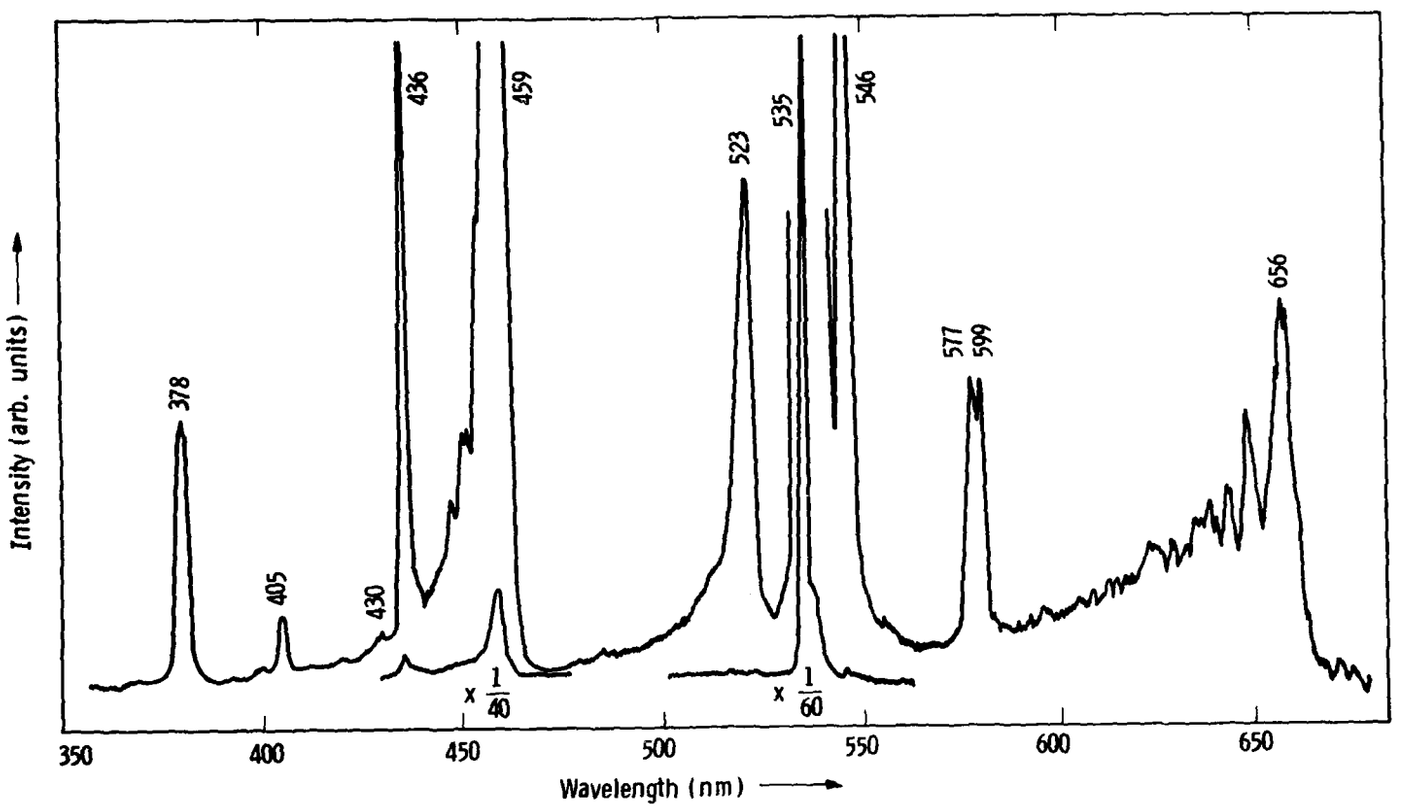


Figure 4. Time-integrated fluorescence from a pulsed glow discharge in TlHg vapors at 900°C. Relative line intensities are uncorrected for S-20 spectral response.

characterized by strong TlHg excimer bands at 459 and 656 nm, originating from the TlHg ($B^2\Sigma_{1/2}$) level. Intense Tl fluorescence at the 535 nm ($7^2s_{1/2} \rightarrow 6^2p_{1/2}$) green transition and the 378 nm ($7^2s_{1/2} \rightarrow 6^2p_{1/2}$) resonance line indicated strong pumping of the desired Tl ($7^2s_{1/2}$) level within the glow discharge. The emission intensities of the 535 nm Tl line and the 459 and 656 nm TlHg excimer bands increased by a factor of ten as the temperature was raised from 700°C to 900°C. However, the 378 nm Tl resonance line intensity merely doubled over this range, and is approximately constant for temperatures beyond ∿825°C. This behavior indicated radiation trapping of the 378 nm Tl line in this temperature regime. Trapping permitted substantial portions of the Tl ($7^2s_{1/2}$) population to be channeled into the 535 nm Tl line and the 459 and 656 nm TlHg excimer bands. Thus glow discharges in Tl-Hg vapors at elevated temperatures and pressures appeared to be very effective in pumping the TlHg excimer ($B^2\Sigma_{1/2}$) level.

The time dependence of emission at several temperatures from the 459 nm TlHg excimer is shown in Figure 5. The decay time constant of the TlHg excimer decreased as the Tl density increased (as temperature increased). The temporal behavior of the emission from Tl^* and $TlHg^*$ states was found to be identical, and fluorescence decay rates increased linearly with increasing Tl density. This indicated that, in high pressure TlHg mixtures, the electron impact can efficiently excite Hg atoms to their ^3p levels. Through a collisional process, most of the Hg excited atoms combines with the Hg ground state to form Hg_m^* (m = 2,3) excimers. Since the Hg_m^* excimers have long lifetimes (>1 μs), they appeared to act as energy reservoirs for the Tl^* and $TlHg^*$; their population relaxed primarily through radiation from Tl^* and $TlHg^*$. From the dependence of the $TlHg^*$ excimer decay rates on Tl density (as shown in Figure 6), the intrinsic lifetime of the Hg_m^* molecule was found to be 1.8 μsec, and the rate coefficient for the reaction

$$Hg_m^* + Tl \rightarrow TlHg^* + Hg$$

was 1.8×10^{-10} cm^{-3} sec^{-1}.

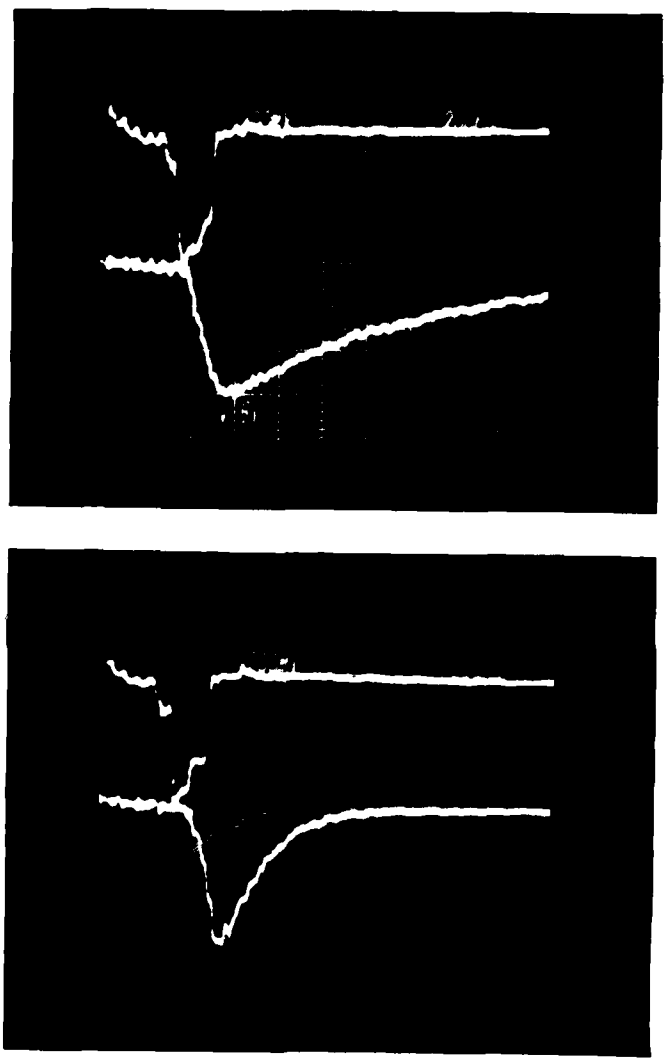


Figure 5 — Time dependence of emission of the TlHg excimer at 458 nm, at various temperatures.

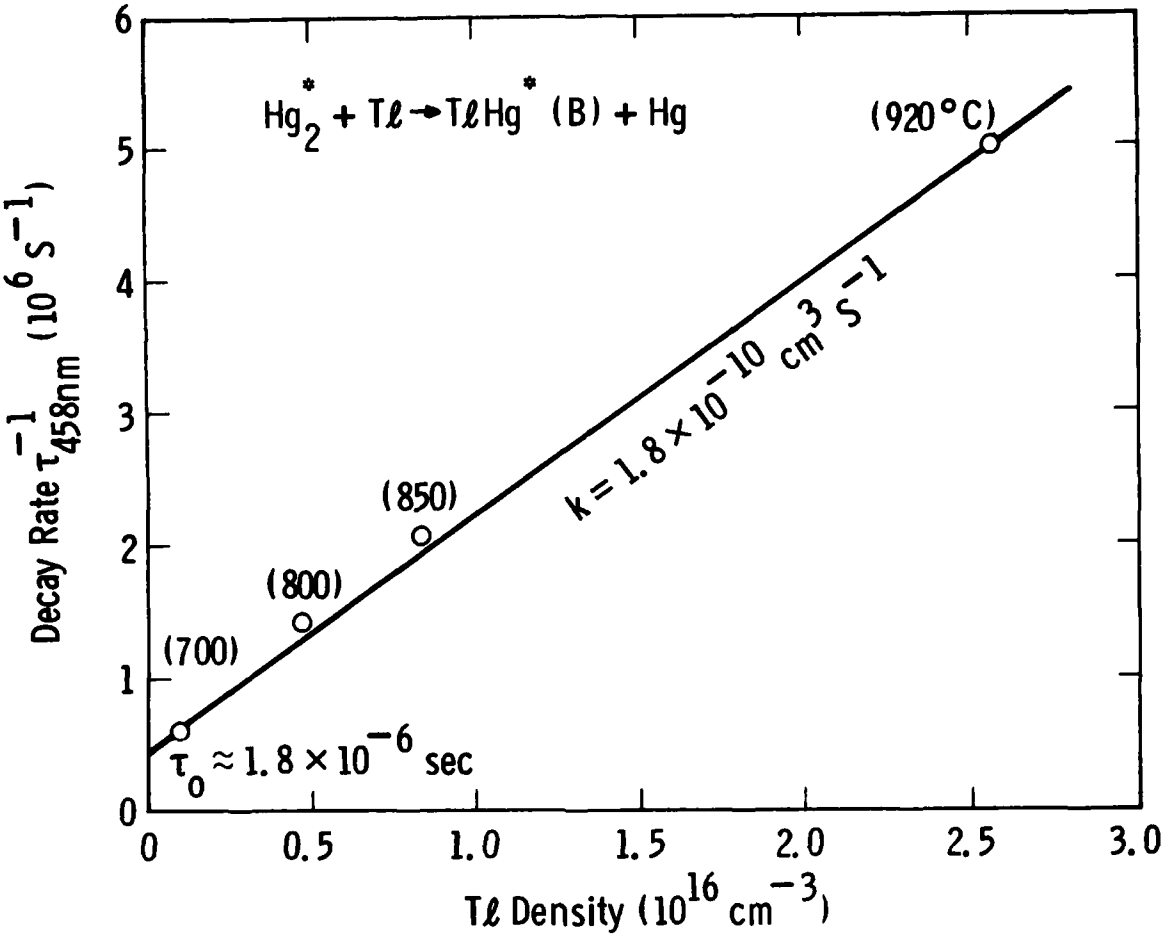
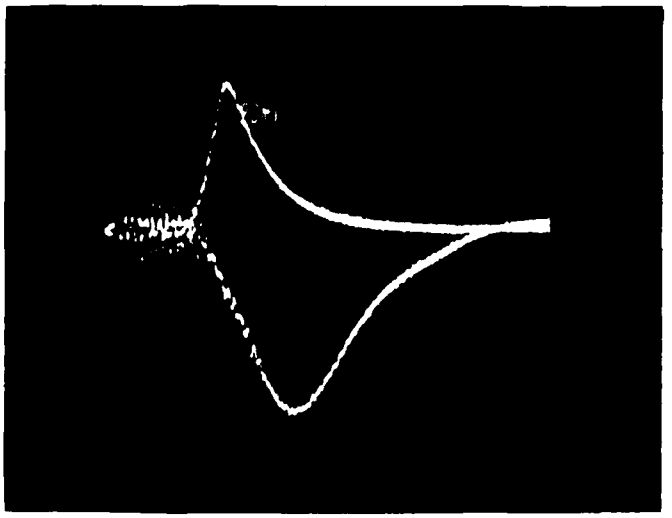


Figure 6. $TlHg^*$ decay rates as a function of Tl density.

Gain and absorption measurements of these discharges were performed using a multiline Ar^+ laser as a probe. No transient gain was observed at the peak of the $TlHg^*$ excimer band at 458 nm; on the contrary, a strong absorption was detected. The time for maximum absorption at 458 nm was delayed by 150 nsec with respect to the time of maximum emission, and the decay of the absorption was considerably slower than that of the emission (as shown in Figure 7). Unlike the constant absorption observed at 488 nm (out of the TlHg excimer band), the 458 nm absorption increased with increased Tl density (see Figure 8). It is believed that the 488 nm absorption was due to Hg_m^* molecules, but that the 458 nm absorption was due to some unknown species (probably ground state TlHg(x)) as well as Hg_m^*.



λ = 458 nm

920°C

Absorption

Figure 7 — Optical absorption at 458 nm as a function of time (the upper trace is emission and the lower trace is absorption).

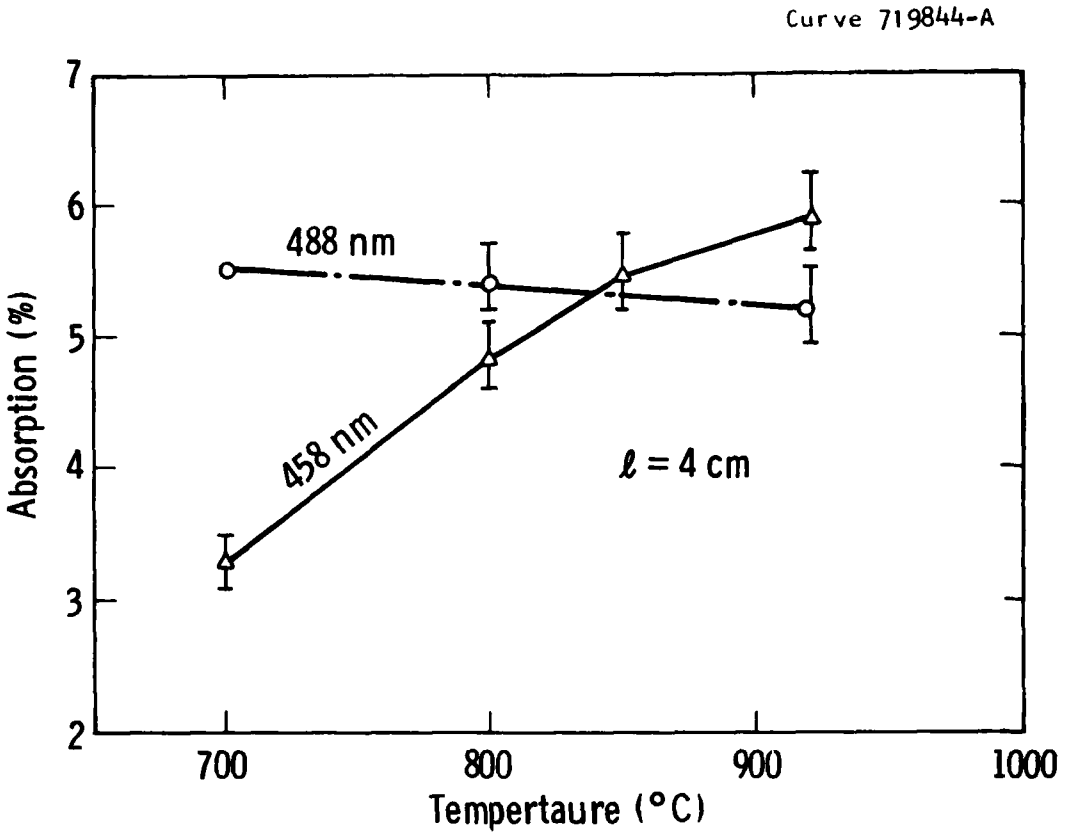


Figure 8. Optical absorption at 458 nm and 488 nm as a function of temperature.

Although the transfer of energy from Hg_m^* (m = 2,3) into TlHg was efficient, the formation rate of the $TlHg^*$ (B) excimer was not fast enough to produce gain in the Tl-doped, high Hg-density, discharge medium because of Hg_m^* (m = 2,3) absorption. Thus, to achieve population inversion in a TlHg excimer system, the excitation must be more rapid and/or the Hg_m^* (m = 2,3) absorption must be minimal. The formation rate of TlHg (B) through a three-body recombination from Tl^* (7s) colliding with Hg is rapid. By substituting a high pressure buffer gas for high density Hg, the Hg density can be reduced; this should promote direct excitation of the Tl as well as reduce the Hg_m^* (m = 2,3) absorption. Through this excitation process, population inversion of TlHg (B) molecules may be achieved.

3. FUTURE WORK AND RECOMMENDATIONS

The results of our experimental study on TlHg indicated that the transfer of energy from Hg_m^* (m = 2,3) into Tl^* and $TlHg^*$ is efficient, but that TlHg laser gain would never be achieved because of Hg_m^* absorptions. Thus, in order to reduce the Hg_m^* absorption in Tl-Hg discharge without decreasing the three-body collision recombination rate of TlHg excimers, one must reduce the Hg density and increase buffer gas pressure. At high buffer gas pressures, TlHg discharges may generate sufficient $TlHg^*$ excimers to produce laser gain. In the future, we recommend dye laser gain measurements of Tl-Hg discharges at moderate Hg density ($\sim 10^{17}$ cm^{-3}) and high buffer gas pressure ($\sim$10 atm) be performed.

4. REFERENCES

1. L. A. Schlie, L. E. Jusinki, P. D. Rathge, and D. L. Drummond, "E-Beam Initiated Discharges in High Pressure Hg Vapors," 37th Gaseous Electronic Conference, Pittsburgh, October 9-12, 1979.

This report was typed by Martha Fischer.