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TLHG LASER DEVELOPMENT PROGRAM.(U)
APR 79 C S LIU, R K WILLIAMS, L A WEAVER N00014-78-C-0131

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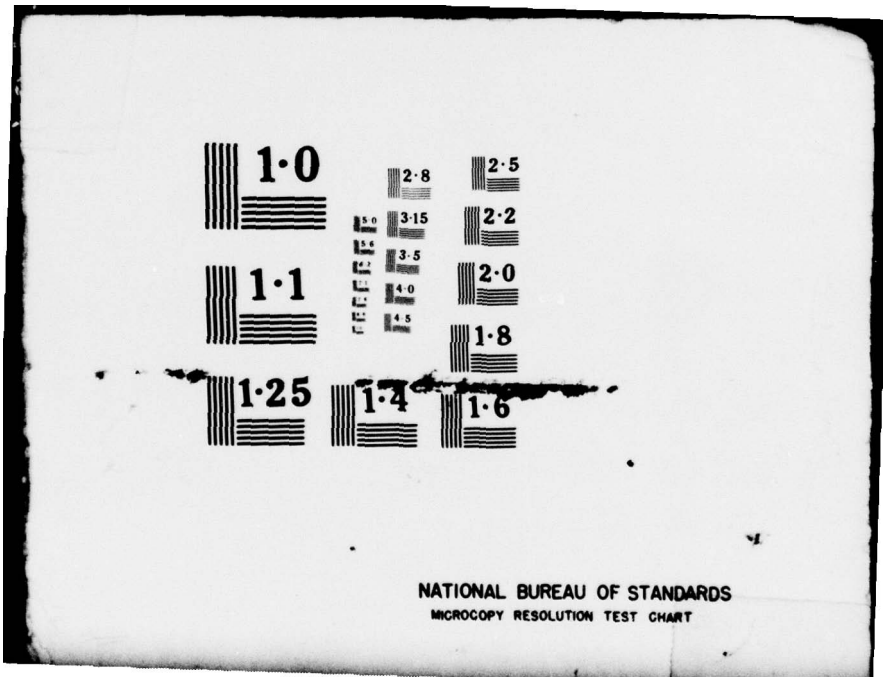
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(10) C. S. Liu, R. K. Williams and L. A. Weaver

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TlHg LASER DEVELOPMENT PROGRAM

C. S. Liu, R. K. Williams, and L. A. Weaver

FINAL REPORT (PHASE I)

(period between February 1, 1978 and January 31, 1979)

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April 10, 1979

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TlHg LASER DEVELOPMENT PROGRAM

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SUMMARY

This report describes the results of an experimental study of the TlHg excimer system. A comparative evaluation of TlXe and TlHg candidates has indicated that the TlHg excimer is the most promising system for detailed study. An all-hot, sealed-off transverse discharge tube and a pressurized oven assembly have been fabricated and tested successfully. This experimental apparatus will permit the study of most metal excimer discharges at temperatures up to 1000°C and pressures up to 10 atm. Preliminary tests have shown that pulsed TlHg discharges suitable for excimer laser feasibility studies can be produced in the experimental apparatus. It is recommended that fluorescence and gain measurements of TlHg discharges proceed immediately, with laser tests to follow if warranted.

1. INTRODUCTION

This report summarizes research work performed at the Westinghouse R&D Center under ONR Contract No. N00014-79-C-0131, "Thallium Mercury Laser Development", for the period between February 1, 1978 and January 31, 1979. The effort has as its goal the performance of definitive gain measurements on discharge-excited TlHg laser mixtures using specialized test apparatus. This apparatus will also be employed to demonstrate TlHg laser output at 459 nm if the gain measurements indicate that laser tests are warranted. Lasers emitting in the blue-green spectral region offer many potential advantages for Navy undersea applications, and the metal excimer family represented by the TlHg molecule continues to hold high promise as an efficient, scalable blue-green laser source.

During the past several years encouraging data have been accumulated which support the promising laser potential of the TlHg excimer. In 1976 York and Carbone[1] demonstrated high pressure, high temperature mercury glows in UV-initiated, transverse discharge geometries. High densities of excited Hg_2^* molecules were generated at attractive excitation efficiencies, although gain measurements revealed no population inversions in the Hg_2^* species. More recently Hamil et al.[2] measured a 30% discharge efficiency in the production of $Tl(7^2S_{1/2})$ and $TlHg(B^2\Sigma^+_{1/2})$ states in pulsed TlHg discharges. Thus it appears that Hg_2^* excimers do not quench $TlHg^*$ laser states substantially in the discharge, but serve rather to enhance the production of $TlHg^*$ molecules and the atomic precursor, Tl^*. The three-body collisional processes that form $TlHg^*$ and Tl^* states apparently occur quite efficiently with the Hg_2^* molecule acting as a near-resonant intermediate energy storage species.

The encouraging results of these studies have motivated the development of a versatile experimental apparatus which is capable of

providing definitive feasibility studies of hot metal excimer laser candidates. This in turn has led to several important recommendations regarding the directions in which further work should proceed. Specifically, the major accomplishments to date are:

- The relative merits of TlXe, TlHg, TlIHg and TlIXe excimer systems have been evaluated. The TlHg system is recommended as the most promising blue-green excimer candidate for further detailed studied.
- UV-preionized transverse discharge electrode assemblies having full sealed-off TlHg compatibility have been fabricated and tested successfully. These electrode structures have produced diffuse, 0.5 x 1.0 x 8 cm (4 cm^3) discharges at pressures up to 5 atm within a room temperature Plexiglas enclosure.
- A stainless steel, high pressure oven assembly has been fabricated and tested successfully. The oven passed qualification tests at hydrostatic pressures of 250 psia, and has attained operating tube temperatures up to 1000°C.
- The entire discharge tube and pressurized oven apparatus has been tested as a unit. Preliminary results indicate that high pressure, high temperature pulsed discharges can be achieved in the intended TlHg mixtures.

The experimental apparatus appears to be operating properly, and is ready for definitive spectroscopic measurements. It is recommended that the program effort proceed immediately to assembling the appropriate diagnostic apparatus and measuring the fluorescence/gain features of the TlHg excimer system.

2. TECHNICAL RESULTS

Figure 2.1 illustrates the flow of work for the TlHg Laser Development Program. A considerable amount of preliminary effort has been expended to identify the discharge and laser kinetic features of candidate metal excimer blue-green laser candidates. The TlXe, TlHg, TlI:Xe and TlI:Hg systems have been studied in part by research groups at JILA and AFWL as well as at Westinghouse. The consensus recommendation by these scientific groups is that the TlHg excimer system offers the best hope of exhibiting laser gain in discharge-excited tests. The iodide constituent could improve laser excitation conditions, but the exact effects of this additive remain uncertain. As a consequence, the TlHg excimer has been selected as the system of choice for these studies.

Design of the laser test apparatus has sought to satisfy the stringent requirements for TlHg excimer formation, while preserving the device flexibility to test the other metal excimer candidates. This constraint has been satisfied in the approach selected. Basically, two parallel efforts of device design, fabrication and test have been pursued, as shown in Figure 2.1. The first approach involves the development of a pre-ionized, transverse discharge electrode assembly enclosed in a sealed-off quartz envelope. The second effort involves the development of a high pressure stainless steel oven to enclose the heating elements and the discharge test cell. Both development activities have been completed, with integration and full TlHg laser discharge tests scheduled for the next work phase.

2.1 Comparison of TlXe and TlHg Systems

The ground state of alkali metal-noble gas molecules generally shows a weak van der Waals' minimum at large internuclear separations, and is repulsive at shorter radii. The excited states are bound by several thousand wave numbers, and the potential minimum occurs at an

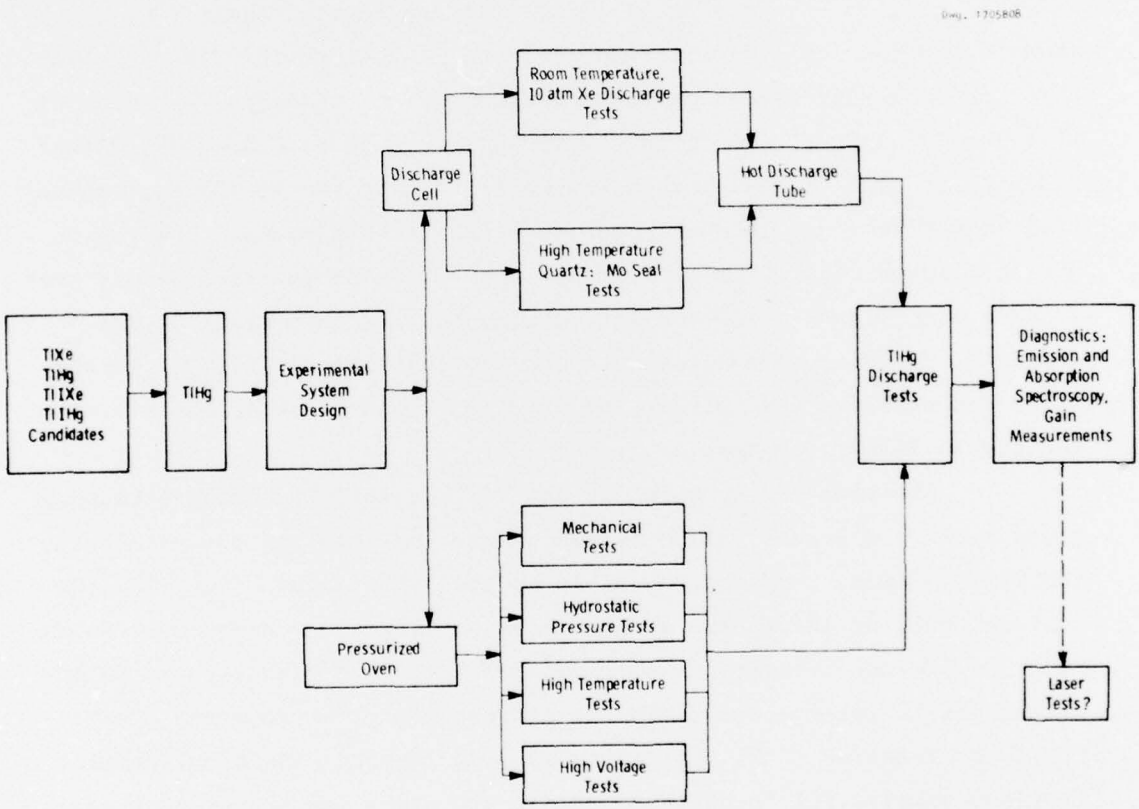

Figure 2.1. Flow diagram of the work elements required to produce definitive laser gain and output tests in TlHg excimer lasers.

internuclear separation where the ground state is repulsive. This molecular structure gives rise to the possibility of achieving significant population inversions within the excimer species.

Several reports in the literature on TlXe[3] excimers have indicated that the TlXe system might be a good candidate for an efficient visible laser if one could achieve a 20% excitation fraction for $Tl^*(7^2S_{1/2})/Tl(6^2P_{1/2} + 6^2P_{3/2})$. The thallium excited state $7^2S_{1/2}$ atoms would then collide with ground state Xe to form the $TlXe^*$ excimer through three-body recombination processes. The stimulated emission of $TlXe^*$ would begin at 410 nm and be tunable to ~450 nm. However, recent findings at JILA[4] indicate that the generation of the $Tl^*(7^2S_{1/2})$ state from TlXe discharges is much lower than earlier estimates. The reason for this lower excitation efficiency is that discharges in TlXe mixtures produce significantly lower electron temperatures than required for efficient $Tl^*(7^2S_{1/2})$ excitation. Thus to increase $Tl^*(7^2S_{1/2})$ populations one must increase either the electron temperature or the electron density in TlXe discharges.

The electron temperature can be increased by adding attaching gases such as halogens, which is equivalent to replacing the metal with thallium halides. However, possible adverse effects of using thallium halides, such as absorption at the laser wavelength, are not well known and thus the net benefit is uncertain. The electron density can be increased by increasing the discharge current density. However, due to the large fraction of Tl ionization at high current densities, atomic thallium vapors will be depleted unless the operating temperature can be raised to provide additional Tl vapor density. Moreover, the likelihood of discharge arcing increases substantially at higher current densities in the presence of lower ionization potential excited species such as Tl^*. Therefore, further work is needed to determine whether the TlXe excimer can be a practical laser system, given the somewhat disappointing nature of preliminary spectroscopic observations.

The thallium-mercury system basically belongs to the same excimer family as TlXe, but is characterized by significantly different excitation mechanisms. Mercury has one less electron than thallium and

behaves as a noble gas in many respects. The TlHg ground state should be only weakly bound relative to the excited TlHg state; the ionization potential for Hg is 10.4 eV (cf., 12.1 eV for Xe), further enhancing the characterization of Hg as an inert gas.

The emission spectrum of the TlHg excimer is concentrated in two narrow bands, one centered at 459 nm and the other at 656 nm. The binding energy[5,6] of the excited $TlHg^*$ ($B^2\Sigma_{1/2}$) state is about 4100 cm^{-1}, three times that of $TlXe^*$. Therefore, the stimulated emission cross section of $TlHg^*$ is expected to be enhanced by the greater binding energy as well as by the narrow ($\sim$4 nm) bandwidth.

The TlHg discharge has been studied recently by Hamil, Drummond, Schlie, and Benedict[2] at AFWL. Their experimental results indicate that a uv-preionized TlHg discharge can efficiently produce $Tl^*(7^2S_{1/2})$ and $TlHg^*(B^2\Sigma_{1/2})$ states. Hamil et al.[2] and Gallagher[4] have hypothesized that these highly efficient discharges are due to rapid energy transfer of Hg_2^* into Tl^* and $TlHg^*$. The transfer cross sections from Hg_2^* to $Tl^*(7^2S_{1/2})$ and $TlHg^*(B^2\Sigma_{1/2})$ are found to be about 1.2×10^{-14} cm^2. Studies of self-sustained discharges in mercury by York and Carbone[1] at LASL have indicated that diffuse stable mercury discharge glows can produce Hg_2^* densities as high as 2×10^{15} cm^{-3}. Thus formation rates of 6×10^5 sec^{-1} for Tl^* (7S) and $TlHg^*$ (B) can be obtained easily due to efficient energy transfer processes of Hg_2^* into Tl^* and $TlHg^*$. Because of this efficient transfer process the TlHg excimer becomes a more promising blue-green metal excimer laser system than the corresponding TlXe system.

Laser excitation in the TlHg system can be accomplished through two channels. One is a result of pumping ground state Tl to the $Tl^*(7^2S_{1/2})$ level followed by three-body recombination with ground state Hg to form $TlHg^*$ (the same as in the TlXe system). The other method is to pump ground state Hg to the $Hg^*(^3P)$ levels to form Hg_2^*, which can then react with ground state Tl to form $Tl^*(7^2S_{1/2})$, or to form $TlHg^*$ directly. The $Hg(6^2P_1)$ and (6^3P_2) electron impact excitation cross sections[7] at $\sim$6 eV are 1.8 and 3.2×10^{-16} cm^2, respectively, and the $Tl^*(7^2S_{1/2})$ excitation

cross section is 2.3 x 10^{-16} cm^2 at the same energy.[8] Thus the 10^2 to 10^3 density ratio of Hg:Tl should channel most of the initial energy into $Hg^*(6^3P)$ states, and then collisional processes will lead to the formation of Hg_2^* and subsequently $TlHg^*$. This is probably the most efficient way to generate $Tl^*(7^2S_{1/2})$ and the $TlHg^*$ excimer because the collisional processes are near-resonant. Measured discharge efficiencies of 30% obtained by Hamil et al.[2] in producing $Tl^*(7^2S_{1/2})$ and $TlHg^*(B^2\Sigma_{1/2})$ states indicate that TlHg excimer laser emission should be possible in the blue-green region with attractively high efficiencies.

The TlHg and TlXe comparative studies have concentrated upon understanding the fundamental kinetic processes involved in producing excited $TlXe^*$ and $TlHg^*$ excimers. This information was obtained primarily from fluorescence measurements of TlXe and TlHg discharges. Since the $TlHg^*$ excimer system appears to be the more promising for blue-green laser applications, diagnostic studies will favor this system. Gain and absorption measurements using a tunable dye laser source will further quantify $TlHg^*$ production, as well as intermediate species such as Hg_2^* and Tl^*.

2.2 Tl-Hg Gain Cell

The discharge tube design for the TlHg gain cell has been guided by two fundamental considerations: first, the formation of $TlHg^*$ excimers depends on three-body recombination processes of various forms, which suggests the necessity of high Hg vapor pressure; secondly, reasonable Tl vapor density requires fairly high operating temperatures (~900°C). As is well known from previous experience, mercury and thallium compatibility is an important factor in material selection. Both quartz and molybdenum are known to be inert to Hg and Tl even at elevated temperatures; in previous laser research efforts we developed reliable quartz-to-molybdenum high temperature seals, and these techniques have been used successfully in fabricating the required all-hot, sealed-off quartz gain cell module.

Electrodes are made of molybdenum, and are profiled according to the Rogowski criteria. Preionization sparks can be placed either behind

a screen anode or along side the discharge, as shown in Figures 2.2 and 2.3. The active volume of the proposed gain cell will be of the order of 0.5 cm wide by 1 cm gap by 50 cm long in five modules of 10 cm long each. For initial studies the uv-preionized transverse discharge electrode modules have undergone qualification tests in a high pressure chamber shown in Figure 2.4. This chamber, fabricated from Plexiglas, provides valuable design information at room temperature in various inert gases up to pressures of 5 atm.

A "Mark I" gain cell electrode module has been fabricated to assess the operating characteristics of the tube design, and is shown in Figure 2.5. The envelope is made entirely from quartz, including flat optical quality windows glassblown to each end of the cylindrical body. These windows are tilted slightly from the perpendicular to suppress spurious optical feedback into the discharge gain region. With little difficulty, Brewster angle windows can be fabricated. The transverse electrode assembly is fabricated entirely from molybdenum, and includes two near-Rogowski profiled electrodes about 10 cm in length spaced about 1 cm apart. These electrodes are bolted to thin Mo plates which reduce fringing electric fields; the electrode assemblies are fastened to the two quartz:Mo cup seal feedthroughs with a threaded Mo rod. Four Mo ribbon feedthrough seals serve as uv preionizers by producing sparks from the bare ends of the Mo ribbon conductors to the adjacent Mo electrode plate. The entire tube can operate sealed-off for long periods at temperatures up to $\sim$1000°C and pressures up to $\sim$1 atm in the presence of most corrosive materials, including Tl, Hg and their halides. This cell not only demonstrated the fabrication feasibility of a rather complex Mo:quartz transverse discharge assembly, but produced satisfactory diffuse glow discharges in He at 1 atm pressure.

Since the quartz envelope with flat windows cannot withstand a pressure differential exceeding $\sim$1 atm, this gain cell module must be tested with TlHg mixtures at elevated temperatures and pressures in the pressurized oven. Definitive gain measurements and possible laser output tests, if warranted, will be performed after successful qualification of the electrode assemblies under high temperature and high pressure conditions.

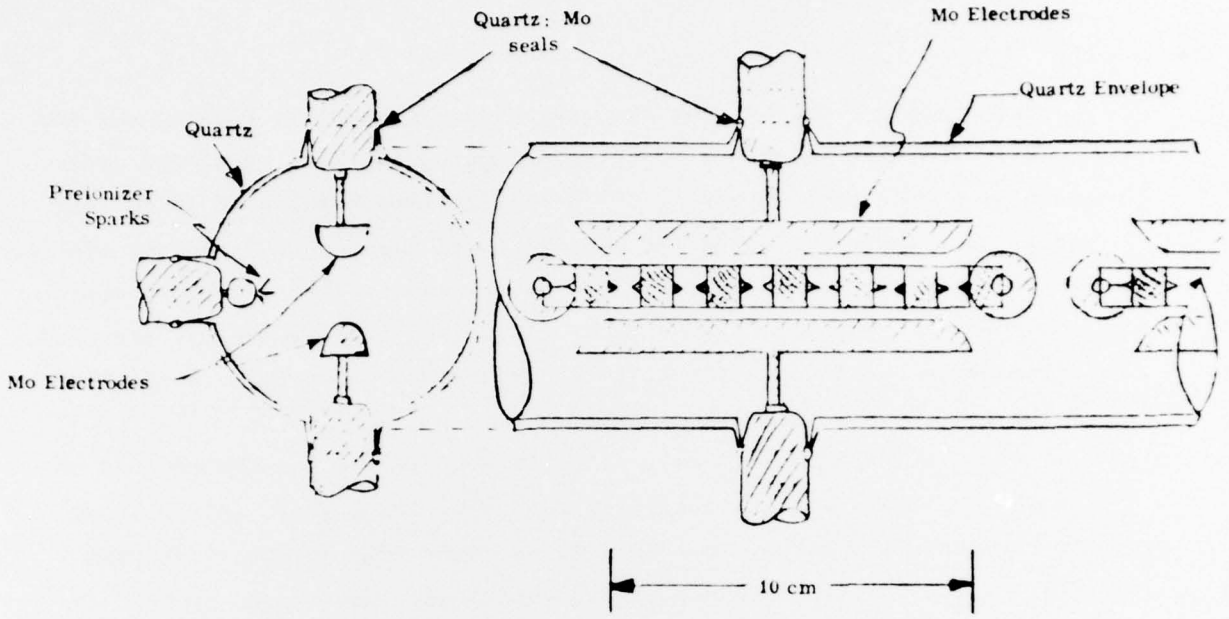

Figure 2.2. TlHg discharge tube with side preionization.

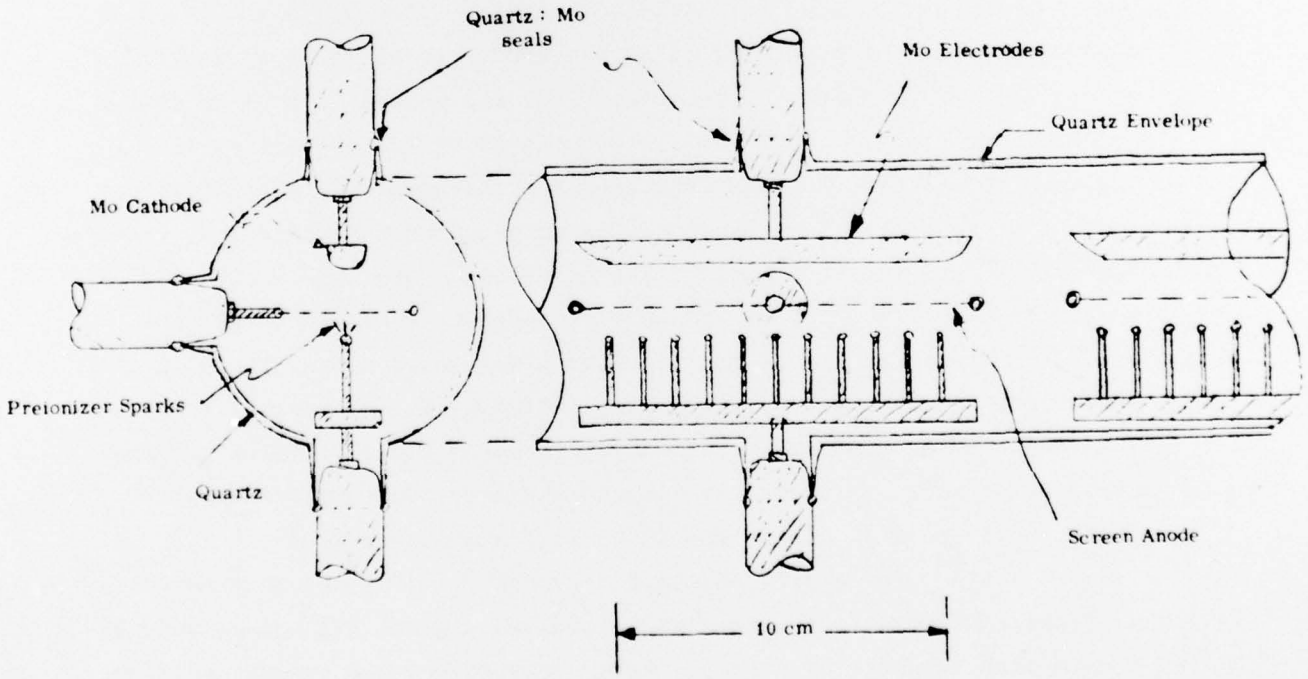

Figure 2.3. TlHg discharge tube with preionizers behind the screen anode.

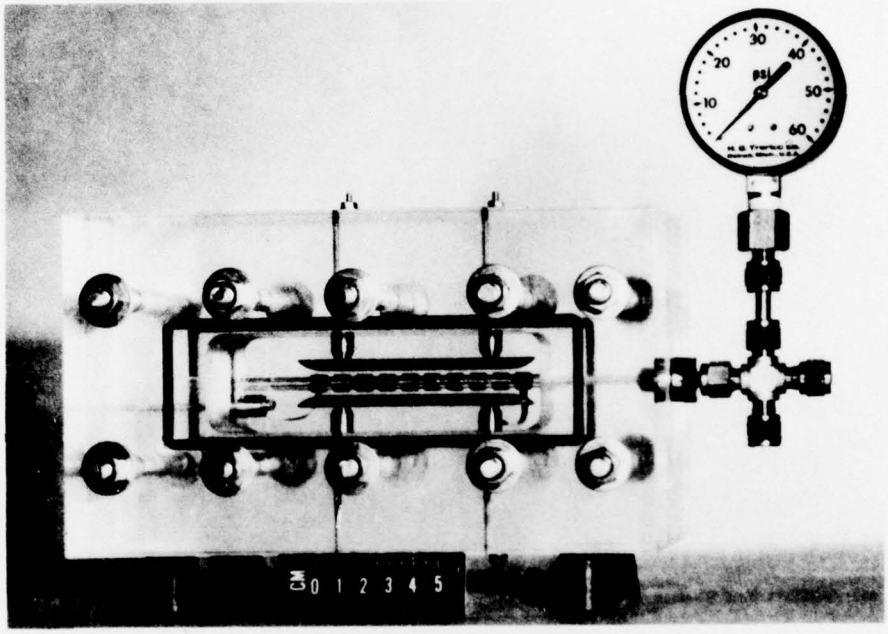

Figure 2.4. High pressure (5 atm) Plexiglas chamber for testing UV-preionized, transverse discharge electrode configurations at room temperature prior to final tests with TlHg laser mixtures.

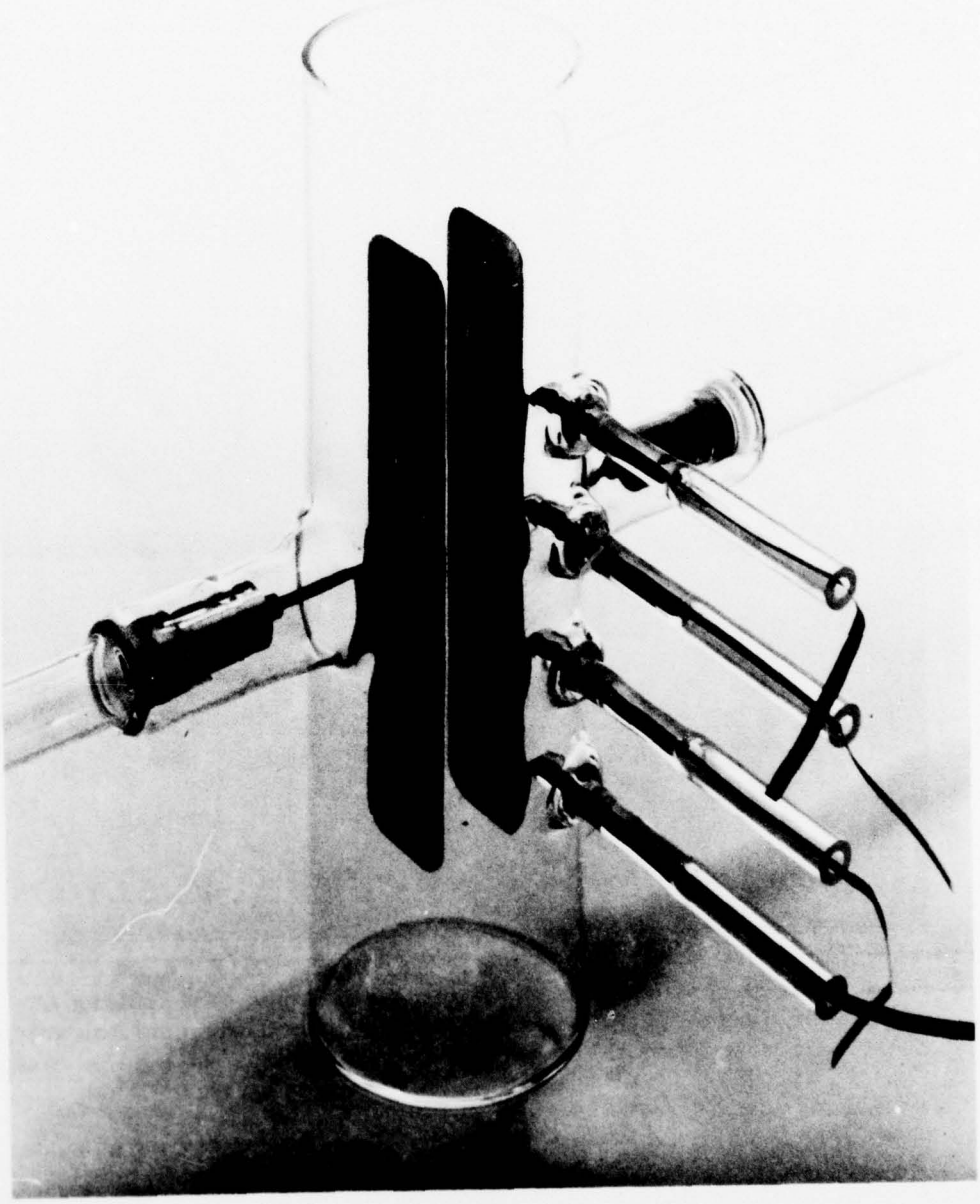

Figure 2.5. Trial quartz:Mo transverse discharge cell used to establish fabrication and electrical design techniques.

2.3 Pressurized Oven

The high operating pressures required for TlHg excimer formation, in the range of 3 to 5 atm, place a severe demand upon a test apparatus which must also function at high temperatures (up to ∿900°C). To satisfy this requirement, a pressurized oven has been designed and fabricated to contain the quartz laser test cell. The stainless steel cylinder and end flanges are designed to withstand internal pressures up to ∿200 psi, and in actual operation the pressure of the excimer laser mixture within the quartz cell will be counterbalanced by an equal gas pressure within the oven container. Thus no net pressure differential will exist across the quartz cell wall. Appropriate electrical feedthroughs and optical windows in the high pressure container provide experimental access to the discharge cell.

The pressurized oven is designed for maximum flexibility in testing new high temperature, high pressure metal excimer systems. Figure 2.6 is a photograph of the high pressure, stainless steel enclosure for the TlHg laser, shown with the flange and oven assembly withdrawn and the top half of the oven removed. The oven enclosure is made of 1/2 inch thick, 14 inch diameter stainless steel pipe with two flanged end plates. This vessel, qualified under OSHA specifications for operating pressures up to 250 psia, can contain discharge chambers up to 1 m in length operating at temperatures up to ∿1000°C required to test the TlHg excimer laser potential. Four high voltage feedthroughs and eight thermocouple feedthroughs are shown on the end flange, as well as a special optical window . The electrical pulser components can be seen adjacent to the withdrawn end flange. On the other flange, not visible in the photograph, are a nitrogen inlet, an evacuation outlet, a safety pressure relief valve and a second optical window. These two special windows on the end flanges are placed along the optical axis of the discharge cell, and can be used for absorption and gain measurements as well as laser tests. Three additional viewing windows can be seen on the enclosure cylinder. Heating elements, temperature sensors, and the discharge tube cradle are mounted on one of the end plates, which can be slid easily in and out to allow necessary modifications.

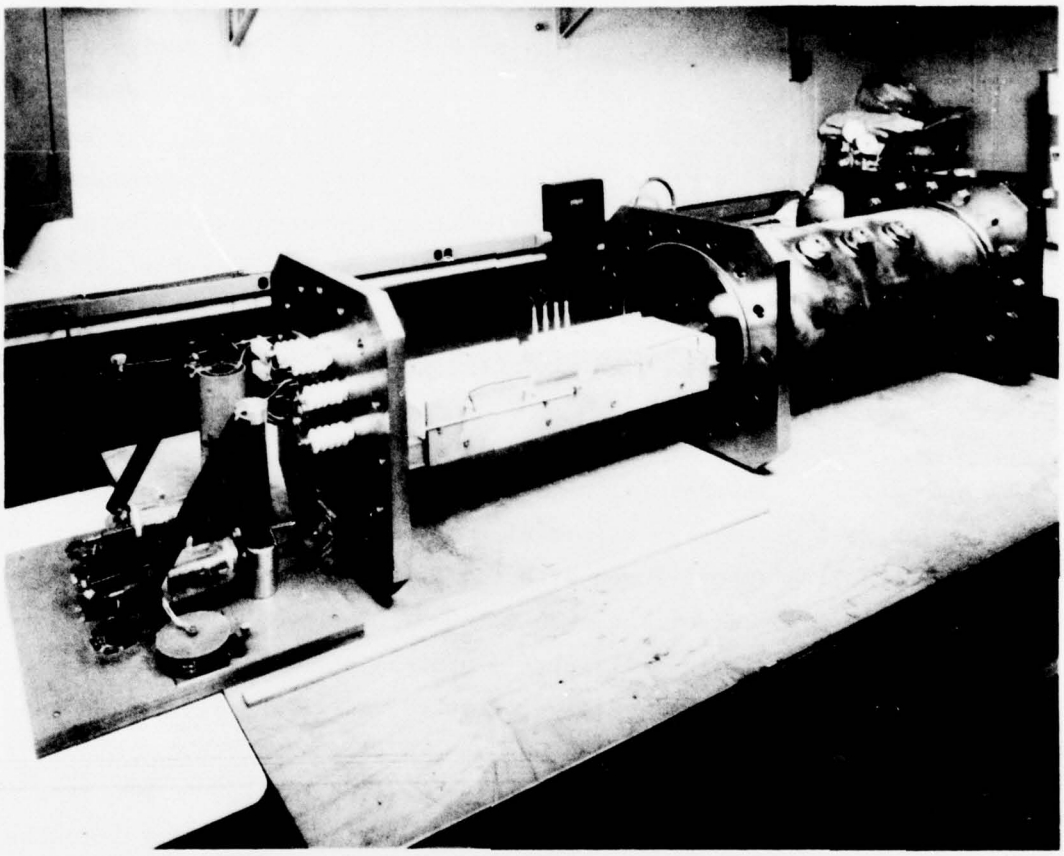

Figure 2.6. High pressure vessel employed to contain UV-preionized, transverse TlHg laser discharges at pressures up to 250 psia and temperatures up to 1000°C. The top half of the oven has been removed for clarity.

The oven has passed all mechanical, hydrostatic pressure, high temperature and high voltage tests. The total system is now completely assembled and ready for TlHg discharge tests under high vapor pressures and high temperature conditions. Full discharge, fluorescence and gain tests will commence during the next phase of this contract effort.

3. FUTURE WORK AND RECOMMENDATIONS

To assess the feasibility of the TlHg excimer laser system, one must measure the number density of $TlHg^*$ excimers created within the discharge. This quantity determines the gain coefficient of the $TlHg^*$ laser system. Additionally, one must measure discharge characteristics such as the electric field-to-particle density ratio E/N and the current density which determine the excimer excitation efficiency. The various excitation cross sections and excited state lifetimes of which determine $TlHg^*$ excimer populations are of critical importance for realistic laser kinetic calculations, and for assessing ultimate $TlHg^*$ laser performance. Techniques for measuring temporally-resolved densities and lifetimes of $Tl^*(7s)$, $TlHg^*$ and Hg_2^* species are described in the following paragraphs.

Figure 3.1 is a schematic diagram of the proposed spectroscopic apparatus. The TlHg discharge comprises a uv-preionized self-sustained transverse discharge contained in a pressurized high temperature oven. The pressure in the oven enclosure varies according to the mercury vapor pressure, which in turn is monitored by measuring the discharge tube temperature. The transmitted and fluorescence signals are detected through either a monochromator by a gated detector or an optical multichannel analyzer (OMA) system. The processed signals are displayed on either an oscilloscope or a chart recorder.

The fluorescence studies will be carried out to find the optimum conditions for the TlHg laser gain measurements. The dependence of the thallium line intensities at 377.6 and 535 nm and the Hg_2^* band intensities peaked at 380 and 458 nm will be monitored to describe Tl^* and Hg_2^* densities. In addition, $TlHg^*$ emission peaked at 459 and 656 nm will be measured as a function of mercury pressure, and the thallium vapor density will be studied independently. From these studies we will obtain not only the relative temporally-resolved densities and

lifetimes of $Tl^*(7S)$, $TlHg^*$ and Hg_2^*, species, but also will learn the energy transfer rates from Hg_2^* states to the $Tl^*(7S)$ and $TlHg^*(B)$ states. The $TlHg^*$ excitation mechanism through the energy transfer of Hg_2^* excimers is significant, and the magnitude of this rate may well determine the success or failure of the proposed TlHg laser system.

Gain or/and absorption measurements of the TlHg system will be performed with the same experimental setup shown in Figure 3.1. A tunable dye laser that can be triggered by the discharge with a variable delay will be employed as a probe source. The wavelength of the probe will be tuned to 459 nm, which is the peak of the $TlHg^*(B)$ emission band. The length of the active gain region will be of the order of 50 cm. The effects of beam steering and defocussing caused by the non-uniformity of gas densities and shock waves can be eliminated by using a small probing beam diameter with a large detecting surface, and by properly designing the optical system. From the laser gain measurements we will be able to conclude both the feasibility and the potential scalability of the TlHg excimer laser system. If gain is measured at 459 nm in the TlHg discharge system, then an actual laser test using discharges up to 100 cm in length and external reflecting mirrors will be attempted with this apparatus.

The measured discharge current and electric field coupled with theoretical expressions for the drift velocity of the electrons in the gas mixture should lead to a deduction of electron density and electron temperature. Discharge parameters such as electron density and E/N are important in determining the laser kinetics and excitation rates. Thus measurements of electrical current and voltage will be performed to obtain a better understanding of the discharge characteristics.

The apparatus fabricated is sufficiently versatile that it is capable of handling almost any hot metal excimer system up to temperatures of 1000°C and pressures of 10 atm. Thus it is recommended that this experimental facility be employed to re-examine TlXe and TlI-Hg systems, and to explore new excimer molecules as potential laser candidates.

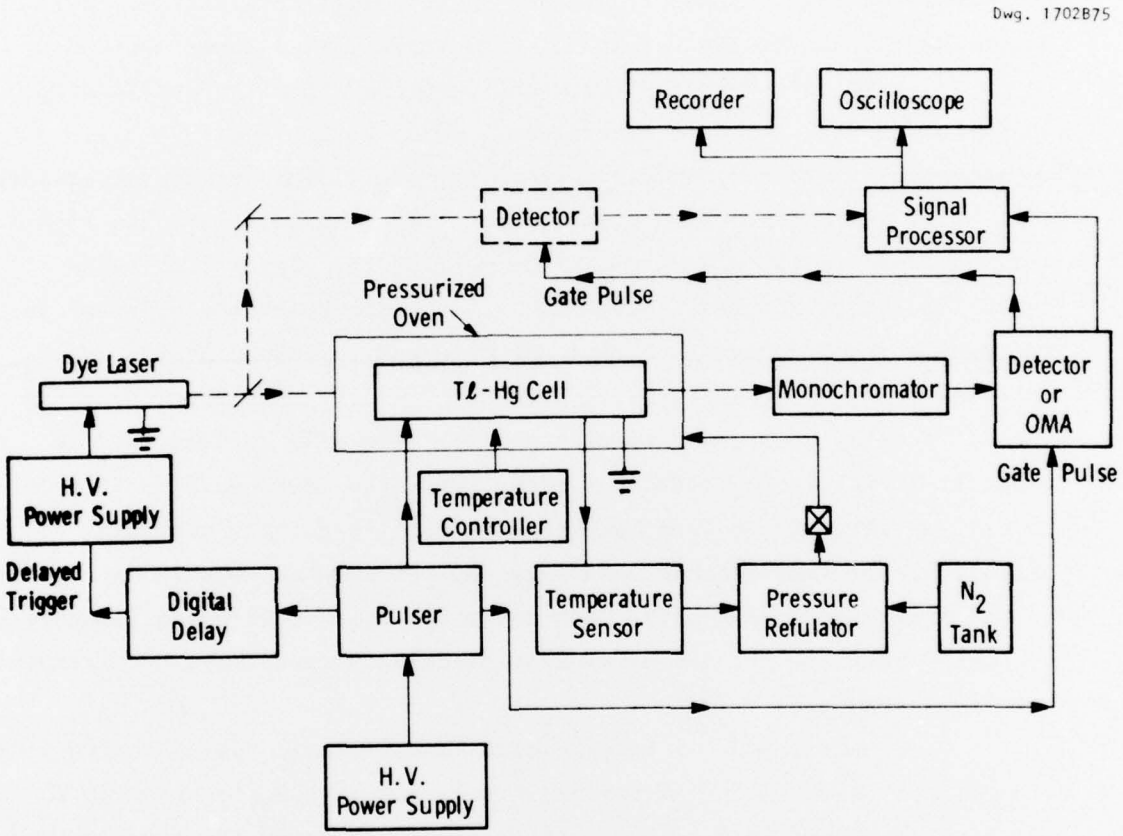

Figure 3.1. Spectroscopic apparatus designed to measure the fluorescence, absorption and gain features of pulsed TlHg discharges.

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This report has been typed by Martha A. Fischer.