VISIBLE LASER DISCHARGE STUDIES

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VISIBLE LASER DISCHARGE STUDIES QUARTERLY PROGRESS REPORT

Task III

Introduction

In order to facilitate the development of metal vapor excimer lasers for inertial fusion, a theoretical study has been undertaken to identify the dominant discharge physics and kinetics for metal vapor excimers like cadmium-mercury (CdHg^*). Interest in such lasers have been heightened since the reported observation of gain in CdHg^* .⁽¹⁾ An efficient method of pumping the first excited state of metal vapors is by secondary electrons created in a discharge. By discharge pumping it is possible to create mainly the lowest excited state, thereby minimizing highly excited electronic states that could photoabsorb. E-beam pumping may have difficulty in pumping metal vapor excimers due to photoabsorption by excited species. To obtain high efficiencies for CdHg^* system by discharge pumping three technical issues must be addressed:

- (a) inefficiencies in discharge pumping due to energy flow into excited states
- (b) discharge stability
- (c) efficient discharge enhancement

The present effort is directed to address the above issues through the development of a kinetics code that models the dominant kinetics of CdHg^* laser system. The bulk of this report is devoted to identification of inefficiencies in discharge pumping due to energy flow into excited states. The present analysis can also be used to predict discharge stability and discharge enhancement. These latter issues are beyond the scope of the present effort; however they will be addressed in the ERDA sponsored metal vapor excimer program at AERL.

Electron Impact Cross Sections for Cadmium and Mercury

The electron impact cross-sections for cadmium plays an important role in discharge physics of CdHg* laser. Figure 1 shows the energy level diagram for cadmium, mercury and neon (used as a buffer gas). As shown, the lowest excited manifold of states are the 3_P of cadmium that have a threshold of 3.7 eV while 3_P of mercury have a threshold of 5.3 eV. It is also observed that the threshold for electron impact excitation of Cd^* and ionization of Cd^{**} is only 2.6 eV. For mercury it is also about 2.6 eV. We have also estimated the electron impact excitation and ionization cross-sections of ground state and excited states cadmium and mercury. The momentum transfer and inelastic cross-sections for the ground state Hg have been taken from Rockwood's calculation.⁽²⁾ The momentum transfer cross-section for the excited Hg species are assumed to be the same as those for the ground state. For ionization from excited state mercury we have used a Gryzinski type calculation⁽³⁾ while the excitation cross section from Hg^* to Hg^{**} has been taken to be same as Cd^* to Cd^{**} ; that has been calculated by H. Hyman.⁽⁴⁾ For cadmium inelastic cross-sections of ground state, we have scaled the mercury cross-sections used by Rockwood.⁽²⁾ Other cross-sections were calculated to be similar to mercury. It was found that the peak value of electron impact ionization cross-section of Cd^{**} is twelve times larger than the peak value of electron impact cross-section for formation of Cd^* from ground state. This coupled with the fact that the ionization threshold for Cd^{**} is less than that for formation of Cd^* from ground state indicates that the excited state Cd^{**} would probably play an important role in discharge physics. From discharge stability considerations; three-step ionization will

Fig. 1 Energy Levels of Cadmium Mercury and Neon

play an important role instead of two-step as for rare gas monohalide lasers. (5) It is, therefore, evident that for CdHg^* laser systems the excited states Cd^* and Cd^{**} will play a dominant role in discharge; much like the role of Krypton metastables in KrF^* laser systems. For KrF^* laser systems the discharge stability (due to dominant two-step ionization) was achieved through loss of electrons by attachment to fluorine while for CdHg^* laser the discharge stability (due to dominant three-step ionization) will be achieved through loss of electrons via recombination.

Buffer Gas for CdHg^* Laser

An important question, however, remains as to the basis of selection for buffer gas in CdHg^* laser system. Buffer gas serves as a third body in excimer formation. Using our Boltzmann code, we have investigated the effects of argon, neon, and helium as a buffer gas in a mixture of cadmium and mercury for different electric fields and a fixed excited state fractional population of $\text{Cd}^*/\text{Cd} = 10^{-3}$. The excited state population Cd^{**}/Cd was assumed to be negligibly small. Our detailed calculations using different values for Cd^*/Cd and Cd^{**}/Cd indicate a similar behavior as presented here with regard to the choice of buffer gas for CdHg^* laser system. Figure 2 shows the percentage of input energy expended as a function of electric field in formation of Hg^* , Cd^* and that into the heating (elastic loss) of helium for a mixture of $\text{Cd}/\text{Hg}/\text{He}$ at number densities of 2×10^{17} , 2×10^{18} and 10^{19} (cm^{-3}) respectively. It is observed that a large percentage of input energy goes into direct heating of helium at electric fields below 1 kV/cm while the peak value of input energy for the formation of Cd^* is also near 1 kV/cm. Hence, for discharge operations near 1 kV/cm and below about 60% of the

Fig. 2 Percentage of Discharge Energy into Cd*, Hg*, He

input energy goes into formation of Cd^* . A large amount of input energy is lost due to the elastic collisions of secondary electrons with helium. Since optimum discharge operation near an electric field of 0.5 - 1 kV/cm is found to be only 60% efficient; it is probably disadvantageous to use helium as a buffer gas.

Figure 3 shows a similar result using argon as buffer gas for identical discharge conditions. This figure shows that unlike helium (a lighter gas); the elastic losses due to secondary electron impact with argon are confined to within 2% of the input energy in the entire range of electric fields considered. The efficiency for formation of Cd^* by electron impact is as high as 85% at 0.5 kV/cm and decreases as the electric field is increased. From these considerations argon may be considered to be a suitable candidate for a buffer gas. However, one disadvantage of using argon as a buffer gas is that the range of the fast electrons in argon is shorter than neon or helium.

Figure 4 shows the results for a mixture of Cd/Hg/Ne at identical discharge conditions. The behavior of the distribution of input energy going into various excited states is similar to the case of a mixture of Cd/Hg/Ar. At 0.5 kV/cm, estimated operating electric field for CdHg* laser system, the percentage of input energy that goes into the formation of Cd^* is as high as 80% and decreases with increase of electric field. The added advantage for using neon is that the fast electrons will travel a further distance in Ne. The issue of the third body for excimer formation will also have to be considered in choosing the buffer gas.

Discharge Kinetics

The major difficulty that was encountered in discharge modeling was the lack of information for various rate constants of many reactions. In our

Fig. 3 Percentage of Discharge Energy into Cd^* , Hg^* , Ar

Fig. 4 Percentage of Discharge Energy into Cd*, Hg*, Ne

attempt to model the discharge kinetics we have used approximate estimations of a few reactions, as we shall discuss later, for which the rate constants have not been determined. There is a clear need for reliable data for many processes. In the following we would identify the dominant formation and quenching kinetics.

a. Formation Kinetics

The formation kinetics of CdHg^* is through two channels:

- (1) CdHg^* formation via excited state Cd^*
- (2) CdHg^* formation via excited mercury Hg^*

In our discharge modeling we will assume that the dominant formation channel is through Cd^* . This will indeed be a dominant formation channel due to fast production of Cd^* as compared to that of Hg^* at electric fields of interest. Our present kinetics code, therefore, has been developed assuming dominant formation kinetics shown schematically in Fig. 5. The corresponding reactions and rate constants are shown in Table I. The laser mixture will be preionized by high energy e-beam. This mixture will contain cadmium and mercury ionic states. The secondary electrons produced by the high energy electrons are heated by the applied electric field. The energy level diagram of Fig. 1 shows that the lowest excited manifold of states are the 3_p of Cd that have a threshold of 3.7 eV.

It is possible by adjusting the mixture ratio and electric field to tailor the secondary electron distribution such that Cd^* is created preferentially by reaction (3). The excited cadmium atom rapidly recombines with mercury to form the excimer CdHg^* via reaction (9). The three-body rate constant for this reaction is found to be $3 \times 10^{-31} \text{ cm}^6/\text{sec}$.⁽⁶⁾ There are other possible channels for formation of Cd^* . One such possible channel is through

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Fig. 5 Dominant Formation Kinetics for CdHg^+ . M represents the third body.

TABLE I
FORMATION KINETICS FOR CdHg^*

No.	Reaction	Rate Constant
1	$\vec{e} + \text{Cd} \rightarrow \text{Cd}^+ + \vec{e} + e_s$	(1-P) Seb
2	$\vec{e} + \text{Hg} \rightarrow \text{Hg}^+ + \vec{e} + e_s$	P Seb
3	$\text{Cd} + e_s \rightarrow \text{Cd}^* + e_s$	R_1
4	$\text{Hg} + e_s \rightarrow \text{Hg}^* + e_s$	R_2
5	$\text{Cd}^* + e_s \rightarrow \text{Cd}^{**} + e_s$	R_3
6	$\text{Cd}^{**} + e_s \rightarrow \text{Cd}^* + e_s$	R_4
7	$\text{Cd}^+ + \text{Cd} + \text{M} \rightarrow \text{Cd}_2^+ + \text{M}$	$2 \times 10^{-31} \text{ cm}^6/\text{sec}$
8	$\text{Cd}_2^+ + e_s \rightarrow \text{Cd}^* + \text{Cd}$	$5.5 \times 10^{-7} \text{ cm}^3/\text{sec}$
9	$\text{Cd}^* + \text{Hg} + \text{M} \rightarrow \text{CdHg}^* + \text{M}$	$3 \times 10^{-31} \text{ cm}^6/\text{sec}$
10	$\text{Hg}^+ + \text{Hg} + \text{M} \rightarrow \text{Hg}_2^+ + \text{M}$	$1.2 \times 10^{-31} \text{ cm}^6/\text{sec}$
11	$\text{Hg}_2^+ + e_s \rightarrow \text{Hg}^* + \text{Hg}$	$5.5 \times 10^{-7} \text{ cm}^3/\text{sec}$
12	$\text{Hg}^* + \text{Hg} + \text{M} \rightarrow \text{Hg}_2^* + \text{M}$	$3 \times 10^{-31} \text{ cm}^6/\text{sec}$
13	$\text{Hg}_2^* + \text{Cd} \rightarrow \text{CdHg}^* + \text{Hg}$	$5 \times 10^{-10} \text{ cm}^3/\text{sec}$
14	$\text{Cd}^{**} + \text{Hg} + \text{M} \rightarrow \text{CdHg}^* + \text{M}$	$3 \times 10^{-32} \text{ cm}^6/\text{sec}$
15	$\text{Hg}_2^* + \text{Hg} + \text{M} \rightarrow \text{Hg}_3^* + \text{M}$	$10^{-30} \text{ cm}^6/\text{sec}$

NOTE

1. The rates R_1 , R_2 , R_3 and R_4 are dependent on fractional population Cd^*/Cd and Cd^{**}/Cd and are determined from Boltzmann code.
2. P Seb represents the total electrons produced per unit time by high energy e-beam due to impact with mercury.
3. Reaction rates for Eqs. (8) and (11) are further scaled by $T_e^{-0.7} T_g^{-1.0}$.

formation of dimer ion Cd_2^+ ; which can be formed due to a reaction of Cd^+ with cadmium atom in the presence of a third body. We have taken the rate constant for this reaction to be $2 \times 10^{-31} \text{ cm}^6/\text{sec}$ by analogy to the rare gases. The dimer ion can form the excited state Cd^* via reaction (8).⁽⁷⁾ A similar parallel but secondary channel for formation of CdHg^* could be through the reaction depicted by Eq. (13) in Table I. This can occur through the reaction of Hg_2^* with cadmium atom. The rate constant for this reaction is estimated to be $5 \times 10^{-10} \text{ cm}^3/\text{sec}$. Hg_2^* formation channel is depicted through Eqs. (2), (10), (11) and (12). The corresponding rates for these reactions are presented in Table I. The electron impact formation of Hg^* is also included in the present modeling. The rate for formation of Hg^* is determined from Boltzmann code predictions. In general, the electron impact rates such as R_1 , R_2 , R_3 and R_4 are functions of Cd^* and Cd^{**} population (which play a dominant role in discharge kinetics) for a given electric field E/N (V-cm^{-2}). Hence for these rates a set of two-dimensional tables are generated using the Boltzmann code for a priori fixed values of Cd^*/Cd and Cd^{**}/Cd (as discussed earlier) and a given electric field. These generated rates are subsequently used in the solution of the kinetic rate equations. Such quasi-steady analysis has been found to be successful in the past.⁽⁸⁾

Apart from these dominant formation reactions, we have also included the formation of Hg^+ and Cd^+ by penning ionization and formation of Cd^* due to deexcitation of Cd^{**} by electron impact (i.e., superelastics).

b. Quenching Kinetics

Quenching processes lead to inefficiencies and so the mixture ratios should be chosen to minimize these losses. The dominant quenching kinetics, the corresponding reactions and rate constants are shown in Fig. 6 and

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Fig. 6 Dominant Quenching Kinetics for CdHg^* . M represents the third body.

Table II. We have assumed that the reaction (1) is similar to the mercury dimer formation which has a rate constant of $10^{-30} \text{ cm}^6/\text{sec}$. The rate constant for reaction (2) has been measured by McGeech, Fournier and Ewart (Ref. 6) to be $4 \times 10^{-11} \text{ cm}^3/\text{sec}$. Reaction (3) and (4) are the effect of quenching of CdHg^* by Cd and Hg respectively in the presence of a third body. Reaction (5) is a displacement reaction. The rate constant for this reaction could be as high as $5 \times 10^{-10} \text{ cm}^3/\text{sec}$.

Results and Discussion

The theoretical model, thus far described was used to predict discharge characteristics of the metal vapor laser system. The procedure adopted in the numerical solution is outlined as follows:

- (1) The sets of cross section described in Section 2 of this report were used to calculate the electron impact excitation and ionization rates for mixtures of gases considered. The Boltzmann equation is used to determine the rates. We have also estimated the effects of electron-electron and superelastic collisions for the laser mixture. For the discharge conditions modeled, we have determined that at electron densities of interest electron-electron collisions are probably not important; however, superelastic collision rates (i.e., deexcitation rates for Cd^* and Cd^{**}) are comparable to other excitation rates. For this reason, our model includes the effect of superelastic collisions.
- (2) A set of rate equations are then solved to calculate the time dependent densities of charged, excited and neutral species in the discharge. These rate equations are solved in conjunction

TABLE II
QUENCHING KINETICS

<u>No.</u>	<u>Reaction</u>	<u>Rate Constant</u>
1	$\text{Cd}^* + \text{Cd} + \text{M} \rightarrow \text{Cd}_2^* + \text{M}$	$10^{-30} \text{ cm}^6/\text{sec}$
2	$\text{CdHg}^* + \text{CdHg}^* \rightarrow \text{Nonradiating}$	$4.0 \times 10^{-11} \text{ cm}^3/\text{sec}$
3	$\text{Cd} + \text{CdHg}^* + \text{M} \rightarrow \text{Cd}_2\text{Hg}^* + \text{M}$	$10^{-30} \text{ cm}^6/\text{sec}$
4	$\text{Hg} + \text{CdHg}^* + \text{M} \rightarrow \text{CdHg}_2^* + \text{M}$	$10^{-31} \text{ cm}^6/\text{sec}$
5	$\text{Cd} + \text{CdHg}^* \rightarrow \text{Cd}_2^* + \text{Hg}$	$5 \times 10^{-10} \text{ cm}^3/\text{sec}$

with a linear interpolation routine which is used to determine the rates from step (1) at every time interval.

Figures 7-8 show the results of time evolution of electron density and efficiency for production of Cd^* for a typical laser mixture of 82% helium, 16.4% mercury and 1.6% cadmium at a mixture temperature of 775°K . These correspond to number densities (cm^{-3}) of 10^{19} , 2×10^{18} and 2×10^{17} for neon, mercury and cadmium respectively. From Fig. 7, it is observed that the electron density reaches a steady-state value of $2 \times 10^{14} \text{ (cm}^{-3}\text{)}$ in $1.0 \mu\text{s}$. However it is observed that the efficiency for production of Cd^* stays nearly 80% for short time ($\approx 0.1 \mu\text{s}$) and then it drops gradually to nearly 50% at larger times ($\approx 0.2 \mu\text{s}$ or greater). The present investigation is based on our estimates of various rate constants which are not yet fully confirmed. Further analytical and experimental work is needed for more accurate information.

Fig. 7 Temporal Variation of Electron Density for a Mixture of Cd/Hg/Ne

Fig. 8 Production Efficiency of Cd* as a Function of Time

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