VISIBLE LASER DISCHARGE STUDIES

FINAL TECHNICAL REPORT

for period June 1, 1976 - November 29, 1977

B. N. Srivastava, J. H. Jacob, J. A. Mangano, and J. C. Hsia

AVCO EVERETT RESEARCH LABORATORY, INC.
a Subsidiary of Avco Corporation
Everett, Massachusetts 02149

Contract No. EY-76-C-02-2922

prepared for

THE U. S. DEPARTMENT OF ENERGY
9800 South Cass Ave.
Argonne, Illinois 60439

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

VISIBLE LASER DISCHARGE STUDIES

FINAL TECHNICAL REPORT

for period June 1, 1976 - November 29, 1977

B. N. Srivastava, J. H. Jacob, J. A. Mangano, and J. C. Hsia

AVCO EVERETT RESEARCH LABORATORY, INC.
a Subsidiary of Avco Corporation
Everett, Massachusetts 02149

Contract No. EY-76-C-02-2922

prepared for

THE U. S. DEPARTMENT OF ENERGY
9800 South Cass Ave.
Argonne, Illinois 60439

ABSTRACT

Modeling of the discharge physics of a laser system requires the coupled solution of electron transport Boltzmann equation and kinetic rate equations. Such a coupled solution is necessary because the electron impact excitation and ionization rates are dependent on the discharge power deposited in the gas. The technique is applied to predict characteristics in a discharge created by plasma return current in a mixture of helium, krypton and fluorine. The comparisons of the predicted electric field generated by plasma return current with experimental measurements indicate that the technique provides an adequate means of theoretically predicting the discharge characteristics. Plasma return current discharge characteristics in pure argon that is representative of discharges in rare gases and argon with small percentage of SF_6 that is representative of attachment dominated discharge are presented for various pressures and e-beam energies. An analysis is also presented to identify the dominant discharge physics and kinetics for metal vapor excimers like cadmium-mercury (CdHg^*). These studies indicate that the excited states Cd^* and Cd^{**} will play a dominant role in discharge physics of CdHg^* laser.

TABLE OF CONTENTS

<u>Section</u>	<u>Page</u>
Abstract	iii
List of Illustrations	vii
I. NUMERICAL SOLUTION OF THE BOLTZMANN EQUATION FOR ELECTRONIC TRANSITION LASER MODELING	1
1. Introduction	1
2. Theory	2
3. Finite Difference Formulation	4
4. Results	10
5. Code Operation on a CDC 3000 Machine	22
6. User Oriented Code	22
II. PLASMA RETURN CURRENT DISCHARGE	25
1. Introduction	25
2. Theory	25
3. Results	29
4. Experiment	45
III. THEORETICAL ANALYSIS FOR METAL VAPOR EXCIMERS	51
1. Introduction	51
2. Electron Impact Cross Sections for Cadmium and Mercury	52
3. Buffer Gas for CdHg ⁺ Laser	54
4. Discharge Kinetics	56
5. Results and Discussion	64
REFERENCES	69

LIST OF ILLUSTRATIONS

<u>Figure</u>		<u>Page</u>
1	Drift Velocity of Electrons in Argon	11
2	Drift Velocity of Electrons in Argon	13
3	Drift Velocity of Electrons in Helium	14
4	Electron Temperature in Helium	15
5	Electron Energy Distribution Function in Krypton	17
6	Electron Energy Distribution Function in Krypton Including Superelastic Collisions	18
7	Details of Electron Impact Processes Shown in Fig. 6	19
8	Influence of Electron-Electron Collision on Electron Distribution Function in Krypton	21
9	Plasma Return Current Discharge - Representative Geometry	26
10	Calculated and Measured E-field vs F_2 Number Density at Various Total Pressures	31
11	Temporal Variation of Electron Density for E-beam Current Density (J_{eb}) of 100 A/cm^2	32
12	Temporal Variation of Electric Field for $J_{eb} = 100 \text{ A/cm}^2$	33
13	Temporal Variation of Electron Density for $J_{eb} =$ 10 A/cm^2	35
14	Temporal Variation of Electric Field for $J_{eb} =$ 10 A/cm^2	36
15	Temporal Variation of Electron Density for $J_{eb} = 1 \text{ A/cm}^2$	37
16	Temporal Variation of Electric Field for $J_{eb} = 1 \text{ A/cm}^2$	38
17	Temporal Variation of Electron Density for $J_{eb} = 10 \text{ A/cm}^2$	39

<u>Figure</u>		<u>Page</u>
18	Temporal Variation of Electric Field for Jeb = 10 A/cm ²	40
19	Temporal Variation of Electron Density for Jeb = 1 A/cm ²	41
20	Temporal Variation of Electric Field for Jeb = 1 A/cm ²	42
21	Temporal Evaluation of Electron Density in Argon and SF ₆	43
22	Temporal Evolution of Electric Field in Argon and SF ₆ Mixture	44
23	Discharge Chamber and Diagnostic Probes	46
24	Magnetic and Electric Probe Oscillograms	47
25	KrF Laser Pumping with Plasma Return Current	49
26	Energy Levels of Cadmium Mercury and Neon	53
27	Percentage of Discharge Energy into Cd*, Hg*, He	55
28	Percentage of Discharge Energy into Cd*, Hg*, Ar	57
29	Percentage of Discharge Energy into Cd*, Hg*, Ne	58
30	Dominant Formation Kinetics for CdHg*	60
31	Dominant Quenching Kinetics for CdHg*	63
32	Temporal Variation of Electron Density for a Mixture of Cd/Hg/Ne	67
33	Production Efficiency of Cd* as a Function of Time	68

I. NUMERICAL SOLUTION OF THE BOLTZMANN EQUATION FOR ELECTRONIC TRANSITION LASER MODELING

1. INTRODUCTION

The use of high-power electrical discharges for pumping electronic transition lasers typically results in large values for the electron density ($\geq 10^{14}/\text{cm}^3$) and the fractional population of the excited electronic states ($>10^{-5}$). As a consequence, the effect of electron collisions on excited atomic and molecular states can be significant in highpower electrical discharges. These processes include excitation and ionization of excited states and collisions of the second kind or superelastic collisions. Because of the large cross sections for these processes, the electron excited state interaction may modify the electron energy distribution function in the discharge. In particular, this interaction has two-fold effect:

- (1) it gives a greatly enhanced ionization rate
- (2) it cools the electrons thereby decreasing the efficiency of pumping the electronic state of interest.

At high electron densities, the electron-electron and electron-ion collisions become important. The former tend to drive the distribution function toward a Maxwellian. The totality of Coulomb collisions will result in a larger momentum transfer cross section resulting in a higher plasma resistivity. In order to evaluate these effects, it is necessary to numerically solve the Boltzmann transport equation for electrons including the following effects:

- (1) elastic collisions
- (2) excitation of ground and excited states
- (3) superelastic collisions
- (4) attachment
- (5) ionization from ground and excited states
- (6) electron-electron collisions

Difficulties associated with any closed form solution of the Boltzmann transport equation for a mixture of gases in the presence of an applied electric field necessitates the use of a finite difference technique. The purpose of the present effort is to develop a user-oriented code, capable of handling arbitrary gas mixtures for visible and uv laser discharge studies. There have been several efforts in the past^(1, 2, 3) towards generating such solutions. The present formulation is similar to that of Ref. 3.

2. THEORY

The time evolution of the number density $n(\epsilon, t) d\epsilon$ of electrons with energy between ϵ and $\epsilon + d\epsilon$ in a mixture of gases of total number density N is determined by⁽³⁾

$$\begin{aligned} \frac{\partial n}{\partial t} = & - \frac{\partial J_f}{\partial \epsilon} - \frac{\partial J_{el}}{\partial \epsilon} \\ & + \sum_{s,j} N_s^0 \left(Q_{sj}(\epsilon + \epsilon_{sj}^*) n(\epsilon + \epsilon_{sj}^*) + Q_{sj}^-(\epsilon - \epsilon_{sj}^*) n(\epsilon - \epsilon_{sj}^*) \frac{N_s^J}{N_s^0} \right. \\ & + Q_s^i(\epsilon + \epsilon_{si}^*) n(\epsilon + \epsilon_{si}^*) + \delta(\epsilon) \int_{\epsilon_{si}^*}^{\infty} Q_s^i(\epsilon') n(\epsilon') d\epsilon' - \left[Q_{sj}(\epsilon) + Q_{sj}^-(\epsilon) \right. \\ & \left. \left. + \bar{Q}_s(\epsilon) + Q_s^i(\epsilon) \right] n(\epsilon) \right) \end{aligned} \quad (1)$$

where N_s^0 is the neutral density of species s , N_s^j is the density of excited state j . $Q_{sj}(\epsilon)$ is the rate at which the electrons with velocity $v(\epsilon)$ produce excitation from the ground state of species s to excited state j losing ϵ_{sj}^* and $Q'_{sj}(\epsilon)$ represents the rate at which electrons at ϵ suffer superelastic collisions with molecules in state N_s^j and gains energy, ϵ_{sj}^* . These rates are given as

$$Q_{sj}(\epsilon) = \sigma_{sj}(\epsilon) v(\epsilon) \quad (2a)$$

$$Q'_{sj}(\epsilon) = (\epsilon + \epsilon_{sj}^*)/\epsilon \sigma_{sj}(\epsilon + \epsilon_{sj}^*) v(\epsilon) \quad (2b)$$

where $\sigma_{sj}(\epsilon)$ represents the cross section for excitation from ground state of species s to excited state j . Q_s^i is the ionization rate for species s , $\bar{Q}_s$ is the loss rate of electron by attachment to species s and $\delta(\epsilon)$ is the delta function indicating that all electrons resulting from electron impact ionization are produced at zero energy. The first term on the right-hand side of Eq. (1) is the flux of electrons in energy space driven by the applied field E , given by

$$J_f = a_f \epsilon^{1/2} \left(\frac{n}{2\epsilon} - \frac{\partial n}{\partial \epsilon} \right) / \sum_s q_s \sigma_s(\epsilon) \quad (3)$$

where

$$a_f = \frac{\sqrt{2} N e^2 (E/N)^2}{3 \sqrt{m}}$$

and q_s is the mole fraction of species, $\sigma_s(\epsilon)$ the cross section for momentum transfer from electrons at energy ϵ to molecules N_s . The second term on the right hand side is the flux of electrons in energy space driven by elastic collisions. This is given as,

$$J_{el} = a_{el} \epsilon^{1/2} \left[n \left(\frac{kT}{2} - \epsilon \right) - kT \epsilon \frac{\partial n}{\partial \epsilon} \right] \sum_s q_s \sigma_s(\epsilon) / M_s \quad (4)$$

where

$$a_{el} = 2 m N \left(\frac{2}{m} \right)^{1/2}$$

M_s - mass of species, s

T - gas temperature

3. FINITE DIFFERENCE FORMULATION

A finite difference representation of the energy derivatives is given at an energy mesh point "k", as

$$\left(\frac{\partial J_f}{\partial \epsilon} \right)_k = \left[\frac{J_f(\epsilon_{k+1/2}) - J_f(\epsilon_{k-1/2})}{\Delta \epsilon} \right] + O(\Delta \epsilon^2) \quad (5)$$

where

$$J_f(\epsilon_{k+1/2}) = a_f \epsilon_{k+1/2}^{1/2} \left(\frac{n_k + n_{k+1}}{4 \epsilon_{k+1/2}} - \frac{n_{k+1} - n_k}{\Delta \epsilon} \right) / \sum q_s \sigma_s(\epsilon_{k+1/2}) + O(\Delta \epsilon^2) \quad (6)$$

However, notice that a combination of Eqs. (5) and (6) would decrease the accuracy of the scheme to $O(\Delta \epsilon)$. Thus, written in this form, the differencing scheme is first order accurate. The differential equation can now be written, at an energy station k as,

$$\begin{aligned} \dot{n}_k = & - \left(\frac{\partial J_f}{\partial \epsilon} \right)_k - \left(\frac{\partial J_{el}}{\partial \epsilon} \right)_k + \sum_{sj} N_s^o (Q_{sjk+m_{sj}} n_{k+m_{sj}} + Q'_{sjk-m_{sj}} N_s^j / N_s^o \\ & + Q_{sk+m_{si}}^i n_{k+m_{si}} \\ & + \delta_{ik} \sum_m Q_{sm}^i n_m - (Q_{sjk} + Q'_{sjk} + \bar{Q}_{sk} + Q_{sk}^i) n_k \end{aligned} \quad (7)$$

where $\left(\frac{\partial J_f}{\partial \epsilon} \right)_k$ is given by combining Eqs. (5) and (6) and a similar expression for $\left(\frac{\partial J_{el}}{\partial \epsilon} \right)_k$ can be obtained. Other notations appearing in Eq. (7) can be

defined as

$$m_{sj} = \epsilon_{sj}^* / \Delta \epsilon$$

$$\dot{n}_k = \left(\frac{\partial n}{\partial t} \right)_k$$

Notice that Eq. (5) contains n_k , n_{k+1} and n_{k-1} energy mesh points. As a consequence, the first two terms of Eq. (7) can be reduced to a tri-diagonal form as

$$a_{k-1} n_{k-1} + b_{k+1} n_{k+1} - (a_k + b_k) n_k$$

where

$$a_k = a_f \left(\frac{1}{\Delta \epsilon} \right)^2 \left(\epsilon_{k+1/2} + \frac{\Delta \epsilon}{4} \right) / \left(\sum_s q_s \sigma_s (\epsilon_{k+1/2}) \times \epsilon_{k+1/2}^{1/2} \right) + a_{el} \epsilon_{k+1/2}^{1/2} \left(\frac{kT}{2} - \epsilon_{k+1/2} + \frac{2kT}{\Delta \epsilon} \epsilon_{k+1/2} \right) / (2 \Delta \epsilon) \quad (8)$$

$$b_{k+1} = a_f \left(\frac{1}{\Delta \epsilon} \right)^2 \left(\epsilon_{k+1/2} - \frac{\Delta \epsilon}{4} \right) / \left(\sum_s q_s \sigma_s (\epsilon_{k+1/2}) \times \epsilon_{k+1/2}^{1/2} \right) + a_{el} \epsilon_{k+1/2}^{1/2} \left(-\frac{kT}{2} + \epsilon_{k+1/2} + \frac{2kT}{\Delta \epsilon} \epsilon_{k+1/2} \right) \quad (9)$$

It is interesting to note the symmetry obtained in representing these two terms by a first order accurate difference scheme. In this form a_k can be interpreted as the rate at which electrons at energy ϵ_k are promoted to energy ϵ_{k+1} while b_k is the rate for loss from ϵ_k to ϵ_{k-1} .

Equation (7), thus represents the finite difference form of the differential Eq. (2). It is readily seen that Eq. (1) represents a system of linear simultaneous equations for K unknowns, where K is the number of energy points selected along the energy axis. This system can be written mathematically as,

$$(I - B \Delta t) \cdot \vec{n}(t + \Delta t) = \vec{n}(t) \quad (10)$$

where B represents the matrix obtained from

$$\dot{n}_k = \sum_l B_{kl} n_l \quad (11)$$

Notice, however, that the matrix B is of upper Hessenberg form when superelastic collisions are not included whereas it is not a sparse matrix of upper Hessenberg form when superelastic collisions are included. In either case, a solution of Eq. (10) can be obtained by using Gauss-Jordan reduction technique, explicitly accounting for the fact that when superelastics are not included, the matrix is of Hessenberg type. Equation (10) will be of linear form in both cases since it would be assumed that the excited state number density N_s^j does not vary during the time the electron distribution n_k relaxes to steady state. In practice this is a good approximation because the time scale for establishing a steady-state electron distribution is more rapid than the time scale for molecular or atom excitation (Ref. 3).

a. Electron-Electron Collisions

Following the work of Ref. 3, one can write the electron-electron collision terms in a flux divergent form.

$$\frac{\partial n}{\partial t} = - \frac{\partial J_{ee}}{\partial \epsilon} \quad (12)$$

where

$$J_{ee} = a \left[P \left(\frac{n}{2\epsilon} - \frac{\partial n}{\partial \epsilon} \right), - Qn \right]$$

$$P(\epsilon, t) = 2\epsilon^{-1/2} \int_0^\epsilon x n(x, t) dx$$

$$+ 2\epsilon \int_\epsilon^\infty x^{-1/2} n(x, t) dx$$

$$Q(\epsilon, t) = 3\epsilon^{-1/2} \int_0^\epsilon n(x, t) dx$$

$$a = \frac{2}{3} \pi e^4 \left(\frac{2}{m}\right)^{1/2} \ell \ln \Lambda, \quad \Lambda = \frac{(kT/4\pi n_0 e^2)^{1/2}}{2e^2/mv^2}$$

A finite difference representation of Eq. (12) is given as

$$J_{ee}(k + 1/2) = a \left[\frac{P_{k+1} + P_k}{2} \left(\frac{n_{k+1} + n_k}{4\epsilon_{k+1/2}} - \frac{n_{k+1} - n_k}{\Delta\epsilon} \right) - \frac{Q_{k+1} + Q_k}{2} \frac{n_{k+1} + n_k}{2} \right] \quad (13)$$

Substituting Eq. (13) into Eq. (12) one would reduce the finite difference equations as

$$\dot{n}_k = a'_{k-1} n_{k-1} + b'_{k+1} n_{k+1} - (a'_k + b'_k) n_k$$

where

$$a'_k = \sum A_{k\ell} n_\ell, \quad b'_k = \sum B_{k\ell} n_\ell$$

and

$$\begin{aligned} A_{k\ell} &= a \left\{ \left(\epsilon_{k+1}^{-1/2} H_{k+1,\ell} + \epsilon_k^{-1/2} H_{k,\ell} \right) \left(\epsilon_\ell U_k^+ - 0.75 \right) \right. \\ &\quad \left. + \left[\left(1 - H_{k,\ell} \right) \epsilon_{k+1} + \left(1 - H_{k-1,\ell} \right) \epsilon_k \right] \left(\epsilon_\ell^{-1/2} U_k^+ \right) \right\} \\ B_{k+1,\ell} &= a \left\{ \left(\epsilon_{k+1}^{-1/2} H_{k+1,\ell} + \epsilon_k^{-1/2} H_{k,\ell} \right) \left(\epsilon_\ell U_k^- + 0.75 \right) \right. \\ &\quad \left. + \left[\left(1 - H_{k,\ell} \right) \epsilon_{k+1} + \left(1 - H_{k-1,\ell} \right) \epsilon_k \right] \left(\epsilon_\ell^{-1/2} U_k^- \right) \right\} \\ U_k^\pm &= \left(\frac{1}{\Delta\epsilon} \pm \frac{0.25}{\epsilon_{k+1/2}} \right), \quad H_{k\ell} = \begin{cases} 0 & k < \ell \\ 1 & k \geq \ell \end{cases} \end{aligned} \quad (14)$$

The matrix element $A_{k\ell}$ may be interpreted as the rate for excitation of electrons from ϵ_k to ϵ_{k+1} while electrons at ϵ_ℓ go to $\epsilon_{\ell-1}$. Similarly, $B_{k\ell}$ is the rate for de-excitation from ϵ_k to ϵ_{k-1} by electrons at ϵ_ℓ going to $\epsilon_{\ell+1}$. It is observed that Eq. (12) can be combined with Eq. (7) when electron-electron collision terms are to be included in the analysis. However, notice that Eq. (12) is nonlinear in nature and thus an iterative scheme must be adopted whenever electron-electron interactions are included in the analysis.

Equation (12) when expressed in finite difference form does not ensure the stationarity of a Maxwellian distribution nor the conservation of energy preserved to all orders. It is, therefore, necessary to modify Eq. (14) to incorporate these properties. It can be shown⁽³⁾ that the following conditions must be satisfied.

$$A_{k\ell} = B_{\ell k} \quad (15)$$

$$A_{jk} = A_{k-1, j+1} \left(\frac{\epsilon_{k-1}}{\epsilon_k} \right)^{1/2} \left(\frac{\epsilon_{j+1}}{\epsilon_j} \right)^{1/2} \quad (16)$$

From the rate interpretation of $A_{k\ell}$ and $B_{k\ell}$, the following conditions must be satisfied,

$$A_{j1} = B_{jk} = A_{kj} = B_{lj}, \quad (J = 1, k) \quad (17)$$

The solution procedure when such interactions are included is as follows. The elements $[A_{k\ell}]$ are computed from Eq. (14) and then condition Eq. (16) is used for evaluating modified form of $A_{k\ell}$. $[B_{k\ell}]$ is then determined from transpose of $[A_{k\ell}]$ in order to satisfy Eq. (15). These elements are used to determine a'_k and b'_k , which are added to Eqs. (8) and (9) in order to get the electron-electron interaction effect.

b. Higher Order Accurate Finite Difference Schemes

The overall finite difference technique presented above is first order accurate in $\Delta\epsilon$. A more accurate way to represent the derivatives is to first rewrite $\frac{\partial J_f}{\partial \epsilon}$ as,

$$\frac{\partial J_f}{\partial \epsilon} = A \left[B(\epsilon) \sum_s q_s \sigma_s(\epsilon) + C(\epsilon) \frac{\partial}{\partial \epsilon} \sum_s q_s \sigma_s(\epsilon) \right] \quad (18)$$

where A is a constant dependent on the applied electric field and

$$B(\epsilon) = - \left[\frac{1}{4\epsilon^{3/2}} n(\epsilon) + \epsilon^{1/2} \frac{\partial^2 n}{\partial \epsilon^2} \right] \quad (19)$$

$$C(\epsilon) = \left[\frac{1}{2\epsilon^{1/2}} n(\epsilon) - \epsilon^{1/2} \frac{\partial n}{\partial \epsilon} \right] \quad (20)$$

A central difference representation of Eqs. (19) and (20) at any energy point k would yield a second order scheme. It is, however, noted that the symmetric representation obtained by substituting Eq. (5) in Boltzmann Eq. (1) will be lost. Nonetheless, in principle, such a formulation is possible. The present technique was restricted to a first order accurate formulation. It was later determined through a step size ($\Delta\epsilon$) study that the marginal gain in computer time would not justify the additional effort of formulating a technique accurate to second order. Similarly, a variable grid finite difference scheme along the energy axis in regions of sharp changes could improve the accuracy of the present scheme. However, again a step size study indicated a posterior that such gain would not be enough to require the use of variable step size. From the finite difference standpoint, the first formulation was found to be sufficiently accurate.

4. RESULTS

The results of the computer code using the above discussed formulation are divided into the following categories:

- (a) Test cases in order to determine the accuracy of the code when electron-electron and superelastic collisions are not included. The code predictions are compared with published results.
- (b) Investigation of the effects of superelastic collisions. An example using krypton and krypton metastables is presented.
- (c) Investigation of the effects of electron-electron collisions. For this case also a typical example of krypton and krypton metastables is presented.

In order to investigate the accuracy of the present code when electron-electron and superelastic collisions are neglected; two cases were investigated. The first case was that of argon and the second case was that of helium. The cross-sections and some experimental data for these gases are readily available from literature. We have used the available cross-sections for argon^(4,5) and helium^(4,6) in our code; in order to predict electron drift velocity as a function of the applied electric field. Figure 1 shows the electron drift velocity in argon for $3 \leq E/P \leq 24$ (V/cm-Torr). Three cases are presented in this figure. The first case indicated by solid lines are those calculated in Ref. 7. This calculation corresponds to a numerical solution of the Boltzmann transport equation. The calculations indicated by broken lines are the predictions of the present code whereas the circles represent a similar calculation using an existing AERL code. The comparisons between the three cases are within $\pm 4\%$ for the whole

Fig. 1 Drift Velocity of Electrons in Argon

range considered. Figure 2 shows a similar calculation for a lower range of $0 \leq E/P \leq 4.0$ (V/cm-Torr) for argon. The results of the present code are compared here with electron drift velocity data⁽⁸⁾ indicated by solid lines and by the existing AERL code. Figure 3 shows a similar comparison for the case of helium. The range of electric field considered was $2 \leq E/P \leq 60$ (V/cm-Torr). The range of electric fields chosen is wide enough to cover most electronic transition laser discharges. The predictions of the present code indicated by broken lines are seen to compare well with calculations of Ref. 9 and existing AERL code. We have also compared the electron temperature in helium predicted by the present code with those of Ref. 9 and existing AERL code. This is presented in Fig. 4. These results indicate that the predictions of the present code is accurate enough for electronic transition discharge investigations. In all the numerical calculations mentioned above the energy step size, $\Delta\epsilon$, were varied till a change of less than one percent in the quantity being compared was observed. This would correspond to $\Delta\epsilon = 0.2$ for the cases examined above. For these cases the energy balance was confined within 1-2%.

In the following text the effect of superelastic collisions will be investigated using the present code. The test gas chosen will be Kr because of recent interest in KrF laser discharge.⁽¹⁰⁾ It has been determined⁽¹⁰⁾ that the excited state Kr^* plays an important role in KrF laser discharge. For this reason, the effect of excited state fractional population, Kr^*/Kr , on the electron energy distribution function for a given electric field will be investigated. The cross-sectional data for ground state Kr and excited state Kr^* were taken to be that given in Refs. 10, 11 and 5. In order to

Fig. 2 Drift Velocity of Electrons in Argon

Fig. 3 Drift Velocity of Electrons in Helium

Fig. 4 Electron Temperature in Helium

isolate the effect of superelastic collisions of the excited states with electrons; the electron-electron collisions were neglected in the present investigation. The effect of electron-electron collisions are described later in the report. Two different electric fields of $E/N = 4.23 \times 10^{-16}$ and 1.41×10^{-16} (V-cm²) were chosen for these calculations. This was done to demonstrate the effect of electric field on the electron energy distribution function.

Figure 5 shows the electron energy distribution function when excited state fractional populations are negligibly small ($< 10^{-5}$) for the two electric fields. It is observed that the distribution function for higher electric field is larger than that for the lower electric field at a given energy. This shape, therefore, indicates an overall higher energy associated with the electrons for larger electric field as one would expect. For excited state fractional population less than 10^{-5} the shape of the distribution function was found to be virtually identical. This indicates that energy loss due to ionization and excitation of excited state, Kr*, is negligible at this fractional population. Figure 6 shows the effect of excited state fractional population and superelastic collisions on the distribution function for an electric field of $E/N = 1.41 \times 10^{-16}$ (V-cm²). The explanation of the various excitation processes considered for the calculations shown in this figure is depicted schematically in Fig. 7. The following conclusions seem relevant from Fig. 6:

- (1) The tail of the distribution function is depleted more rapidly when ionization and excitation of the excited state, Kr*, is an important energy loss. This becomes clear by comparing curves (1) and (3) in Fig. 6. At fractional excited state population of 10^{-3} the electrons at high energy are depleted due to excitation and ionization of excited states.

Fig. 5 Electron Energy Distribution Function in Krypton

Fig. 6 Electron Energy Distribution Function in Krypton Including Superelastic Collisions

H1299

Fig. 7 Details of Electron Impact Processes Shown in Fig. 6

- (2) When superelastic collisions are also included at fractional excited state population of $Kr^*/Kr = 10^{-3}$ and $Kr^{**}/Kr = 10^{-5}$ the electrons gain energy due to de-excitation process. This results in high energy tail of the distribution function as depicted by curve (4) in Fig. 6.
- (3) The de-excitation process indicated by high energy tail of the distribution function is also seen to be important when excitation and ionization of the excited state is ignored. This is depicted through curve (2) for the distribution function.

These results indicate clearly the influence of various excitation processes on the calculated distribution function using the present code. It would appear that excited state collisions have a considerable impact on the electron energy distribution function at fractional excited state populations larger than 10^{-5} .

In order to assess the impact of electron-electron collisions on the distribution function; numerical predictions were obtained using the total number of electrons (cm^{-3}) of 10^{14} and 10^{16} for krypton. All other effects such as excitation and ionization from excited state and superelastic collisions were neglected. Figure 8 shows the distribution function for these cases for an electric field of $E/N = 1.41 \times 10^{-16}$ ($\text{V}\cdot\text{cm}^2$). It is observed from this figure that the electron distribution function changes markedly when the electron density is increased by a factor of 100. This indicates that the electron-electron collisions are apparently of significant importance, at least under the conditions modeled here. In any discharge calculations, therefore, an assessment of the impact of electron-electron collisions must be made.

Fig. 8 Influence of Electron-Electron Collision on Electron Distribution Function in Krypton

5. CODE OPERATION ON A CDC-3000 MACHINE

This objective first required a step size study in order to assess the finite difference accuracy with respect to the number of energy points that must be included with a given step size $\Delta\epsilon$ (present test case was taken to be a single specie Xenon). It was found that a step size of $\Delta\epsilon = 0.2$ with 90 points on the energy axis provides an accurate solution (within 1%) for the case considered. This case was, therefore, chosen to be run on a smaller machine, the CDC 3000 available at Avco Everett Research Laboratory, Inc. This effort required an efficient overlaying of the existing code without introducing substantial changes in the coding format. This has been achieved and earlier solutions obtained on CDC 6600 have been reproduced on this machine. It must be understood that the most general case cannot be run on this smaller machine. However, the objective was to generalize as much as is allowed by the size of the machine and learn the constraints imposed by machine size. It was determined that the machine size restricts the use of more than one species when electron-electron and superelastic collisions are included in the present code.

6. USER ORIENTED CODE

A user-oriented code was developed with the following characteristics of the input and output format:

- (a) Ability to store the cross-sections for various gases on different tapes identifiable by their names. Users could be able to use those by specifying the name.
- (b) Ability to generate computer plots for drift velocity, overage energy, rates, etc.

The plotting routine has been coupled to the Boltzmann Code for user's convenience. A specific instruction in the form of an input parameter (zero or one) is needed in order to get one or all of the plots of the quantities mentioned below:

- (1) Plots of normalized distribution function $f(\epsilon)$ ($\text{eV}^{-3/2}$) vs ϵ ,
energy (ev) for each input E/N (volt - cm^2).
- (2) Drift velocity (cm/sec) vs E/N
- (3) Average Energy (eV) vs E/N
- (4) Inelastic Rate (cm^3/sec) vs E/N
- (5) Superelastic Rate (cm^3/sec) vs E/N
- (6) All input cross sections

II. PLASMA RETURN CURRENT DISCHARGE

1. INTRODUCTION

Plasma return current discharge technique is based on the utilization of the return current to produce a volumetrically large plasma. This discharge technique has two important uses;

- (a) It is a very convenient way of studying processes such as ionization, attachment and discharge stability over a wide range of pressures and discharge conditions.
- (b) It is also useful for pumping visible and UV lasers at low pressures. The discharge operation for this case is not limited to high pressures (≥ 1 atm) since energy is coupled into the discharge through the return current. Furthermore, because the e-beam current is exactly cancelled by the return current in steady state, current and aperture scaling are not limited by; (a) beam pinching due to the magnetic field of the discharge; and (b) skin effect or inductance.

2. THEORY

The plasma return current discharge is a method in which an e-beam is fired through a grounded wire mesh and collected on a floating plate some distance, L , from the wire mesh as shown in Fig. 9. On the way to the plate, the beam ionizes the neutral gas (laser mixture) in its path so that a tenuous plasma is formed. Since the beam collector plate is floating, it will quickly charge negatively until it reaches a potential that is sufficient to drive a

Fig. 9 Plasma Return Current Discharge - Representative Geometry

plasma return current equal in magnitude but in the opposite direction of the e-beam current. The plasma return current is driven by an electric field E given by

$$E = - \rho J_b = \rho J_p \quad (21)$$

where ρ is the plasma resistivity J_b , the beam current density and J_p , the plasma return current density. Initially, the secondary electron density is low and consequently the resistivity and, the electric field are large. Very rapidly, however, the electric field heats the plasma electrons and avalanches the plasma density. The electric field then self-adjusts to a value, which in the case of constant beam current, is just sufficient to produce a total ionization rate equal to the loss of electrons due to recombination or attachment. Because of this mechanism, the discharge is inherently stable.

The purpose of the present effort is to develop a numerical model for the plasma return current discharge for a wide range of discharge conditions likely to be encountered in making visible lasers. The resulting model will be used to calculate the temporal variation of the electric field and secondary electron density as a function of the following parameters:

- (a) e-beam current density
- (b) gas composition
- (c) gas pressure

A representative numerical formulation would proceed as follows. Since the plasma return current density, J_p , is equal and opposite to the e-beam current density, J_b , hence

$$J_b = e n_e V_D (E/N, n^*/n) \quad (22)$$

where n_e is the plasma electron density, e the electronic charge and V_D , the drift velocity. In general the drift velocity is a function of the electric field, neutral density (i. e., gas pressure) and the fractional population of the excited states. In addition for cases where electron-electron collisions play an important role; the effect of these collisions on the drift velocity must be accounted for. Equation (22) when used in conjunction with the Boltzmann code and relevant kinetics code would provide the time evolution of the electric field and secondary electron density. In general, a complete theoretical description requires the solution of the coupled Boltzmann transport equation for the electrons and the kinetic equations for the excited and ionic states of the atoms. Although such numerical techniques have been developed; considerable computation time and expenses can be saved by using a quasi-steady technique. This technique employs a procedure in which various rates of excitation and ionization of species constituting the gas mixture is determined by seeking steady state solution of the Boltzmann equation. These rates are determined for different a priori selected values of the parameters E/N and n^*/n . Thus determined values are utilized in the solution of the kinetic rate equations for excited and ionic states. The coupling of the results from the Boltzmann code and kinetic rate equations is based on utilizing Eq. (22). If an initial value of electron density is assumed, Eq. (22) can be used to determine V_D for a given J_b . Using this value of V_D and an initial value of n^*/n ; the relevant rates to be used in the kinetic rate equations are determined by a two-dimensional interpolation technique. The results of this interpolation are then used in the kinetics code which yields a new value of n_e and n^*/n for a small increment in time. Repetition of the above procedure leads to the time evolution of the electron density and discharge electric field.

3. RESULTS

Using the procedure described in Section 2; the following studies for plasma return current discharge were made:

- (1) Helium, Krypton with small percentage of fluorine added
(representative of attachment-dominated discharges).
- (2) Pure Argon at 1, 5 and 10 torr (representative of discharges
in the rare gases). Two different high e-beam energies of .15
MeV and 1.5 MeV were considered for this case.
- (3) Argon at 10 and 100 torr with small percentages of SF_6 added
(representative of attachment-dominated discharges).

In order to demonstrate the concept of pumping through plasma re-
turn current discharge, we have made an analytical and experimental investi-
gation in mixtures of $\text{F}_2/\text{Kr}/\text{He}$. The details of the experimental setup and
discussion is provided in Section 4. We will discuss, here, the analytical
modeling and compare the results of this modeling to measured data. Un-
fortunately there is no published data on the electron impact cross-sections
of F_2 . For this reason we have neglected the effects of fluorine on the
secondary electron distribution. The electron impact cross-sections of
 F_2 could be large enough to change the Boltzmann code predictions of exci-
tation and ionization rates by as much as 20%. The cross-sections for
Helium and Krypton were taken to be those given in Refs. 4, 5, 6 and 11.
The excited state Krypton, Kr^* , plays an important role in such discharges⁽¹⁰⁾
and therefore, we have included the Krypton metastable in our Boltzmann
code calculations. The cross-sections for Krypton metastable were taken
to be those from Ref. 10. We have used the various cross-sections for a

mixture of Kr/Kr*/He in our Boltzmann code in order to predict the relevant rates (that are required in our kinetics code) as a function of:

- (1) electric field (E/N)
- (2) fractional Krypton metastable population (Kr^*/Kr)

The output of the Boltzmann code is then coupled to the kinetics code that solves the kinetic rate equations for the time evolution of ionic and excited states through a two-dimensional interpolation routine. Figure 10 shows the results of our calculation. We have plotted the measured electric field at 400 ns after initiation of the e-beam versus F_2 number density at various total gas pressures. Also shown are the detailed computer kinetic model calculations of the self-consistent electric field. In general, experimental results agree well with theoretical predictions for high F_2 concentrations and low pressures. The disagreement at low F_2 concentration and high total pressure is probably due to two effects:

- (1) The effects of F_2 on the secondary electron distribution.
- (2) Our kinetic model neglects the electron impact ionization from high lying excited states of Kr.

In order to predict the time evolution of the self-consistent electron density of plasma return current discharge in pure argon, we have made kinetic calculations at various total pressures and current densities for a monoenergetic e-beam of 0.15 MeV. Discharges of this type are representative of the discharges in rare gases. Figures 11 and 12 show the electron density and electric field at total pressures of 1-10 torrs and e-beam current density of 100 A/cm^2 . It is observed from Fig. 12 that initially the electric field generated in the plasma is large which heats and avalanches the electron

Fig. 10 Calculated and Measured E-field vs F₂ Number Density at Various Total Pressures

Fig. 11 Temporal Variation of Electron Density for E-beam Current Density (J_{eb}) of 100 A/cm^2

Fig. 12 Temporal Variation of Electric Field for $J_{eb} = 100 \text{ A/cm}^2$

density. The electric field subsequently rapidly drops. It is noted from this figure that the equilibrium condition has not been achieved for this case until about $.5 \mu\text{sec}$. Ultimately, however, we expect the electric field to asymptote at $.7 \mu\text{sec}$. These figures also show the results at an increased pressure of 5.0 torr. The qualitative behavior of the electron density and the electric field is similar to that of 1.0 torr, however, it is observed that the electron density is larger (Fig. 11) and the electric field is lower at higher pressures at all times. This trend continues when the mixture pressure was further increased to 10.0 torr. We have also calculated the time evolution of electron density and the electric field for e-beam current densities of 10 A/cm^2 and 1 A/cm^2 for different total pressures of 1.0, 5.0 and 10.00 torr. The results for these calculations are fundamentally similar in character. These predictions can be fruitfully used in future plasma return current discharge studies. Figures 13-16 show the results for these calculations. We have also made discharge calculations for a monoenergetic e-beam of 1.5 MeV for different total gas pressures and current densities. The results of these calculations are shown in Figs. 17-20. From these analytical predictions the relevant parameters for discharge pumping by return current can be selected. Figures 21-22 show a similar calculation for Argon when a small percentage of SF_6 (0.5%) was added. We have not included SF_6 in our Boltzmann analysis due to lack of sufficient information on electron impact cross-sections. The attachment rate for SF_6 were obtained from Ref. 12. From these calculations it is observed that the attaching species, SF_6 , tends to equilibrate the electron density on a shorter time scale (see Figs. 21-22 as compared to Figs. 13-14).

Fig. 13 Temporal Variation of Electron Density for $J_{eb} = 10 \text{ A/cm}^2$.

Fig. 14 Temporal Variation of Electric Field for $Jeb = 10 \text{ A/cm}^2$

Fig. 15 Temporal Variation of Electron Density for $J_{EB} = 1 \text{ A/cm}^2$

Fig. 16 Temporal Variation of Electric Field for $J_{EB} = 1 \text{ A/cm}^2$

Fig. 17 Temporal Variation of Electron Density for $J_{eb} = 10 \text{ A/cm}^2$

Fig. 18 Temporal Variation of Electric Field for $J_{eb} = 10 \text{ A/cm}^2$

Fig. 19 Temporal Variation of Electron Density for $J_{EB} = 1 \text{ A/cm}^2$

Fig. 20 Temporal Variation of Electric Field for $J_{EB} = 1 \text{ A/cm}^2$

Fig. 21 Temporal Evaluation of Electron Density in Argon and SF₆

Fig. 22 Temporal Evolution of Electric Field in Argon and SF₆ Mixture

4. EXPERIMENT

The experimental apparatus is schematically shown in Fig. 23 for performing return current discharge investigations in a mixture of $F_2/Kr/He$. The cold cathode e-gun produced a 250 kV/1.2 μ sec/10 cm x 100 cm e-beam with average transmitted current densities of 5 A/cm². The spacing between the floating beam collector (discharge cathode) and the grounded wire mesh (discharge cathode) was 9 cm. A magnetic field in the direction of the e-beam was provided by two field coils. The field was 800 gauss at the center of the discharge chamber and uniform to within 7% throughout the discharge region. The magnetic field prevented beam spreading due to foil and gas scattering and assured a uniform e-beam. Plasma floating potential and electric field were measured with tungsten-tipped electric probes situated at various distances from the grounded mesh anode as shown. Pulse magnetic fields generated by the e-beam or discharge were monitored by a magnetic probe just outside the discharge region.

Typical oscillograms of electric probe voltages, transmitted e-beam current, and magnetic probe signal obtained with 1/2 atm of 1.0% F_2 /5% Kr/94.5% He are shown in Fig. 24. Also shown for comparisons are corresponding oscillograms obtained when the beam collector was externally grounded. Note that no pulse magnetic field was detected when the current returned through the discharge. Small residual electric probe voltages measured with a shorted beam collector were due to e-beam current intercepted by the probe. Experimental results and comparison to our kinetic model was shown earlier in Fig. 10. To show that the return current is indeed an effective method of coupling energy into the discharge, an optical resonator was put on the device and an

Fig. 23 Discharge Chamber and Diagnostic Probes

FLOATING POTENTIAL AND MAGNETIC FIELD PROBES

FLOATING POTENTIAL PROBE 2.5 cm FROM GROUND PLANE
 (1%F₂/5%K/94%He @ 0.5 ATM)

FLOATING BEAM COLLECTOR PLATE

200 ns/div

EXPECTED 3-FIELD

$$B_c = \frac{\mu_0 J_{eb} h}{2} = 31 \text{ GAUSS}$$

200 ns/div

SHORTED BEAM COLLECTOR PLATE

200 ns/div

MAGNETIC FIELD

17 G/div

FLOATING POTENTIAL

1kV/div

(GUN CURRENT

COLLECTED BY PROBE)

200 ns/div

GUN CATHODE

VOLTAGE

(150 kV/div)

GUN CATHODE

CURRENT

5kA/div

69446

Fig. 24 Magnetic and Electric Probe Oscillograms

attempt was made to obtain laser action at $2485 \text{ \AA}$. Since the current density required to reach laser threshold is higher than that which can be supplied by the e-beam, a concentrator beam collector (discharge cathode) shown in Fig. 25 was used to yield a fourfold increase in return current density in the lasing region. Laser action was immediately obtained with a resonator consisting of a 95% reflector and an 89% reflective output coupler. Laser energy output was 350 mJ at an average e-beam current density of 6 A/cm^2 and a return current density of 24 A/cm^2 . When the beam collector was grounded so that no return current flowed through the discharge, no laser output was detected.

69445

Fig. 25 KrF Laser Pumping with Plasma Return Current

III. THEORETICAL ANALYSIS FOR METAL VAPOR EXCIMERS

1. 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^* .⁽¹³⁾ 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.

2. 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 26 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 $3P$ of cadmium that have a threshold of 3.7 eV while $3P$ 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 of cadmium and mercury. The momentum transfer and inelastic cross-sections for the ground state Hg have been taken from Rockwood's calculation.⁽³⁾ 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⁽¹⁴⁾ 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.⁽¹⁵⁾ For cadmium inelastic cross-sections of ground state, we have scaled the mercury cross-sections used by Rockwood.⁽³⁾ 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. 26 Energy Levels of Cadmium Mercury and Neon

play an important role instead of two-step as for rare gas monohalide lasers.¹⁶ 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.

3. 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 Cd*/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 27 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 Cd/Hg/He at number densities of 2 x 10¹⁷, 2 x 10¹⁸ and 10¹⁹ (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. 27 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 28 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 29 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.

4. 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. 28 Percentage of Discharge Energy into Cd^* , Hg^* , Ar

Fig. 29 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. 30. 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. 26 shows that the lowest excited manifold of states are the $3P$ 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 $3 \times 10^{-31} \text{ cm}^6/\text{sec}$.⁽¹⁷⁾ There are other possible channels for formation of Cd^* . One such possible channel is through

H3827

Fig. 30 Dominant Formation Kinetics for CdHg^+ . M represents the third body.

TABLE I
FORMATION KINETICS FOR CdHg^*

<u>No.</u>	<u>Reaction</u>	<u>Rate Constant</u>
1	$\bar{e} + \text{Cd} \rightarrow \text{Cd}^+ + \bar{e} + e_s$	(1-P) Seb
2	$\bar{e} + \text{Hg} \rightarrow \text{Hg}^+ + \bar{e} + e_s$	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).⁽¹⁸⁾ 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.⁽¹⁰⁾

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. 31 and

H3828

Fig. 31 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} cm⁶/sec. The rate constant for reaction (2) has been measured by McGeech, Fournier and Ewart (Ref. 17) to be 4×10^{-11} cm³/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 5×10^{-10} cm³/sec.

5. 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 32-33 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. 32, 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. 32 Temporal Variation of Electron Density for a Mixture of Cd/Hg/Ne

Fig. 33 Production Efficiency of Cd* as a Function of Time

REFERENCES

1. A.G. Engelhardt and A.V. Phelps, *Phys. Rev.* 131, 2115 (1963).
2. W.A. Proctor and G.H. Canavan, *Bull. Am. Phys. Soc.* 17, 979 (1972).
3. S.D. Rockwood, *Phys. Rev. A*, Vol. 8, No. 5, Nov. 1973.
4. A. Gilardini, *Low Energy Electron Collisions in Gases*, John Wiley and Sons, N.Y., 1972.
5. D. Rapp and P. Englanders-Golden, *J. Chem. Phys.* 43, 1464 (1965).
6. S.T.B. Corrigan and Engel, *Pro. Soc. Lond.*, 72, 786 (1958).
7. O. Judd, *J. Appl. Phys.*, Vol. 47, No. 12, 5297 (1976).
8. S.C. Brown, *Basic Data of Plasma Physics*, Wiley and Sons, NY.
9. A. Nishitsuji, K. Kobayaski and M. Matanoka, U.S.-Japan Joint Seminar on The Glow Discharge and its Fundamental Processes, July 12-15, 1977.
10. J.H. Jacob and J.A. Mangano, *Appl. Phys. Lett.*, Vol. 28, No. 12, 724 (1976).
11. L.S. Frost and A.V. Phelps, *Phys. Rev.*, 136, A 1538-1545 (1964).
12. L.G. Christophorou, D.L. McCorkle and J.G. Carter, *J. Chem. Phys.*, Vol. 54, 1, 253 (1971).
13. J.H. Parks, *Appl. Phys. Lett.*, 31, 192 (1977). Also J.H. Jacob, J.A. Mangano, M. Rokni and B.N. Srivastava, presented at 20th Gaseous Electronic Conference, *Bull. Am. Phys. Soc.*, Vol. 23, 2 (1978).
14. L. Vriens, *Phys. Lett.* 8, 260 (1964).
15. H. Hyman, private communication.
16. B.N. Srivastava, J.H. Jacob, J.A. Mangano and M. Rokni, to be published in *Appl. Phys. Lett.*
17. M.W. McGeoch, G.R. Fournier and P. Ewart, *J. Phys. B., Atom. Molec. Phys.*, 9, L121 (1976). Also *J. Appl. Phys.* - To be published.
18. M.A. Biondi, *Phys. Rev.*, 90, 730 (1953) known for Hg_2^+ . Also "Mercury Excimer Studies," MSNW 78-1077-2 Mathematical Sciences Northwest, Inc., January 1978.