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## INVESTIGATION OF OPTIMUM LASER PUMPING

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| An important task of the Optimum Pumping Technology program is the investigation of the feasibility of discharge pumping electronic transition lasers. Such an investigation requires a detailed knowledge of the secondary electron transport properties that may be obtained from a numerical solution of the Boltzmann code. An example of an electronic transition discharge pumped laser is HgCl*. |                       |                                                             |

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## 20. Abstract (continued)

A theoretical investigation of the e-beam controlled discharge pumping of the mercury halide lasers requires a coupled solution of the electron transport Boltzmann Equation and dominant kinetics rate equations. The general only the lowest order diffusion approximation of the Boltzmann Equation is used for this purpose. This is because the more general Boltzmann Equation is complex in nature. To insure the validity of the  $P_1$  approximation an analysis to determine the error of the lowest order diffusion equation is made. The analysis shows that if the spatial derivative of the distribution function can be neglected, i.e., for a distributed source of electrons, the  $P_1$  approximation is valid for all the cases considered. When the spatial derivative cannot be neglected, which is the case for a spatially localized source of electrons, then the region of validity of the  $P_1$  approximation is much more restrictive. High pressure discharges are examples of a distributed source. Electron beams generated for swarm experiments are examples of a spatially localized source.

In order to address the key technical issue of discharge stability in mercury halide lasers; a theoretical study is made to predict the stability criteria in recombination and attachment-dominated metal vapor discharges. In mercury halide laser discharges, a relatively large excited state population exists which dominates the stability of these discharges. This occurs because the excited states have a smaller ionization potential and larger ionization cross-section than the ground state. As a result, multi-step ionization is the dominant ionization. When three step-ionization is dominant and electron mixing of the excited states is unimportant, attachment-dominated discharges will be stable if the attachment rate  $\beta \geq m \nu_{i0}$  where  $2 < m \leq 3$  and  $\nu_{i0}$  is the equilibrium ionization rate. For a recombination dominated discharge the criterion in this limit is  $\alpha n_{e0} \geq m \nu_{i0}$  where  $1.0 < m \leq 1.5$  and  $\alpha$  is the recombination coefficient and  $n_{e0}$  is the equilibrium electron density.



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## I. INTRODUCTION

In the last three years ARPA/ONR sponsored Visible/UV laser programs of AERL have led to the discovery and successful development of important rare gas halide lasers. Specifically, we have obtained 12 J/liter with a 10% intrinsic efficiency from KrF\* by pure e-beam pumping and 10 J/liter with a 9.5% intrinsic efficiency from e-beam controlled discharge pumping. In the last year another class of visible lasers, the mercury halides, have been discovered.<sup>(1,2)</sup> These molecules, like the rare gas halides, have ionic upper levels. Hence, the formation kinetics of the upper laser level should be rapid and efficient due to the long range attractive forces involved in ion-ion interactions. Since the ionization energy of mercury is lower than that of the rare gases, the mercury halides radiate at a longer wavelength than the rare gas halides. To date, lasing action has been obtained in HgCl at 5576 Å and HgBr at 5018 Å.

E-beam controlled discharge pumping of the mercury halide lasers could lead to very high overall laser system efficiencies—efficiencies as high as 20%. In order to achieve these high efficiencies, efficient production of mercury 3<sub>p</sub> manifold, Hg\* (<sup>3</sup>P), in a stable discharge run at high enhancement ratios (discharge power/e-beam power) must be demonstrated. The key technical issues are:

- 1) Hg ( $^3P$ ) production efficiency
- 2) Discharge stability
- 3) Discharge enhancement ratio

In order to theoretically predict the characteristics of the mercury halide lasers a coupled solution of the electron transport equation and kinetic rate equations that model the key formation and quenching kinetics is required. Such a solution is necessary because the electron impact excitation and ionization rates are dependent on the discharge power deposited in the gas. The solution of the Boltzmann Equation for this purpose is generally obtained by considering only the lowest order diffusion or  $P_1$  approximation. This is because the more general form of the Boltzmann Equation is not amenable to analytic solutions. The  $P_1$  approximation of the Boltzmann Equation has proved most useful in the understanding of high pressure discharges. However such an approximation is not necessarily valid for all cases under consideration. Hence, the first part of this report investigates the regions of validity of the  $P_1$  approximation.

One of the major technical issues of the e-beam controlled discharge pumping of the mercury halide lasers is the discharge stability. The mercury halide laser discharges which are typical of attachment-dominated discharges are characterized by a relatively large excited state population. These excited states dominate the stability of these discharges. Typically they have a smaller ionization potential and a larger ionization cross



section than the ground state. As a consequence these effects lead to a condition where multi-step ionization dominates. A theoretical study to establish the stability criteria (for both attachment and recombination dominated discharges) is reported in the second part of this report.

## II. ON THE DIFFUSION APPROXIMATION OF THE BOLTZMANN EQUATION FOR SLOW ELECTRONS

The lowest order diffusion or  $P_1$  approximation of the Boltzmann equation has proved most useful in the understanding of high pressure discharges. There have been several efforts addressing the domain of applicability of this approximation.<sup>(1)</sup> In one of the earliest discussion Holstein<sup>(2)</sup> has shown that the diffusion approximation is valid when the elastic cross section for electron heavy particle collisions is large compared to the inelastic cross section. This criterion has also been used in the Bethe Age theory for electron and neutron diffusion.<sup>(3,4)</sup> While, for most gases, this criterion is well satisfied, there are instances when the magnitude of inelastic and elastic cross sections are comparable. For example, recent measurements<sup>(5)</sup> of the vibrational cross section of  $N_2$  evaluates the peak cross section in the energy range of 2-3 eV, to about  $12 \text{ \AA}^2$ , which is comparable to the peak elastic cross section of  $30 \text{ \AA}^2$  for  $N_2$  evaluated by Englehardt, Phelps and Risk.<sup>(6)</sup>

In this article we investigate the applicability of the  $P_1$  approximation by considering a narrow band of electrons in energy space. It is shown that the angular distribution function depends on two dimensionless parameters,  $\gamma$  and  $\xi$ .  $\gamma$  is the ratio of the energy gained by an electron in the electric field per transport mean free path to the electron energy and  $\xi$  is the ratio of the

energy lost by inelastic collisions per transport mean free path along its trajectory to the electron energy. The former is a familiar parameter in plasma physics, while the later is proportional to the ratio of the inelastic to the elastic cross sections. The analysis presented suggests that when the spatial derivative of the distribution function cannot be neglected, the  $P_1$  approximation is not valid for values of  $\gamma$  and  $\xi$  that approach unity. However, when the spatial derivative can be neglected, the error in  $P_1$  approximation is limited to 5% for all electron energies and for all values of  $\gamma$  and  $\xi$  considered. These results imply that the applicability of the  $P_1$  approximation depends strongly on whether the sources of electrons are spatially localized or uniformly distributed throughout the volume under consideration. This is because for the localized source, the spatial variation of the electron density is important, while for a uniformly distributed source the spatial variation may be neglected. Spatially localized sources of electrons that diffuse between two electrodes are used in swarm experiments.<sup>(7)</sup> Hence, for gases like  $N_2$ , for which values of  $\gamma$  and  $\xi$  are large, the evaluation of impact parameters from swarm data using the  $P_1$  approximation will be suspect. In a high pressure discharge, however, secondary electrons are created uniformly (at least on the scale length of the transport mean free path) through the discharge volume. For such cases, therefore, the spatial derivative of the distribution function may be neglected.

The transport of a broad area electron source in the presence of collisions can be described by the one dimensional Boltzmann equation, (8)

$$2\mu \lambda \frac{\partial f}{\partial x} + 2 eE \lambda \mu \frac{\partial f}{\partial w} + \gamma (1-\mu^2) \frac{\partial f}{\partial \mu} = 2\lambda \left\{ \frac{\partial f}{\partial s} \Big|_i + \frac{\partial f}{\partial s} \Big|_e \right\} \quad (1)$$

where the distribution function  $f$  is a function of the kinetic energy  $w$ , angular coordinate  $\mu$  and spatial coordinate  $x$ .  $\mu$  is the cosine of the angle between electron trajectory and  $\bar{E}$ , which is assumed to be along the  $x$  direction. The transport mean free path  $\lambda$  is defined in the usual manner. The parameter  $\gamma$  is the ratio of the energy gained per transport mean free path to the electron energy, i.e.,

$$\gamma = \frac{eE\lambda}{w} \quad (2)$$

For isotropic elastic scattering,  $\frac{\partial f}{\partial s} \Big|_e$  is given by (8)

$$2\lambda \frac{\partial f}{\partial s} \Big|_e = -2f + \int_{-1}^{+1} f(\mu') d\mu \quad (3)$$

We will assume that an inelastic scattering process changes only the energy of the electron. Then for the  $J^{\text{th}}$  excitation process

$$\frac{\partial f}{\partial s} \Big|_i = \sum_J \left\{ P_J(w + E_J) f(w + E_J) - P_J(w) f(w) \right\} \quad (4)$$

where  $P_J(w)$  is the average number of collisions per unit distance along the electron trajectory.  $E_J$  is the energy lost after the collision.  $P_J(w)$  is zero for  $w < E_J$ .



The usual method of solving the Boltzmann equation for high pressure discharges is to neglect the variations of  $f$  with  $x$  and then expand  $f$  as a sum of Legendre polynomials. Because of the complexity only the first two polynomials are generally retained. This procedure is valid provided, of course, that  $f$  is nearly isotropic. A method for checking the validity of such an approach is to simplify Eq. (1) by considering a narrow beam of electrons (of width  $\Delta w$ ) in energy space having a mean energy  $w_0$ . Integrating Eq. (1) over all energy, we obtain

$$2\mu\lambda_0 \frac{\partial F}{\partial x} + 2\xi_0 F - 2\gamma_0 \mu F + \gamma_0 (1 - \mu^2) \frac{\partial F}{\partial \mu} = -2F + \int_{-1}^{+1} F(\mu) d\mu \quad (5)$$

where

$$F = \int_0^{\infty} dw w f(w) \quad (6)$$

The dimensionless parameter  $\xi$  is the ratio of energy lost by the electrons per transport mean free path to the energy of the electron (8)

$$\xi_0 = \frac{\chi_0 \lambda_0}{w_0} \text{ when } E_J \sim \Delta w \ll w_0 \quad (7)$$

$$\xi_0 = \frac{\chi_0 \lambda_0}{E_J} \text{ when } E_J \lesssim w_0 \quad (8)$$

The two definitions of  $\xi_0$  are obtained from Eq. (6) for the two limiting conditions. Equation (7) is obtained by a Taylor expansion of the first term on RHS of Eq. (4), while for a narrow beam

this term will have a negligible contribution when  $E_J \approx w_0$ .  $\chi_0$  is the energy lost by the electron per unit path length along its trajectory and is given as

$$\chi_0 = \sum_J E_J P_J = N \sum_J Q_J E_J \quad (9)$$

where  $Q_J$  is the inelastic cross section for the  $J^{\text{th}}$  process and  $N$  is the number density of scatterers. From Eqs. (7), (8) and (9), it is clear that  $\xi_0$  is proportional to the ratio of the inelastic cross section to the elastic cross section. In fact, when  $E_J \approx w_0$ , which is near or just above the threshold of the excitation process,  $\xi_0$  is precisely the ratio of the inelastic cross section to the elastic cross section. Note that  $\xi_0$  evaluated by Eq. (7) is always less than when evaluated by Eq. (8), except when  $w_0 = E_J$ . As the two values are similar and we are only making estimates of the error, we will use Eq. (7) in the subsequent discussion.  $\xi_0$ ,  $\gamma_0$ , and  $\chi_0$  are all evaluated at the particular energy  $w_0$ .

Dropping the subscript 0 and integrating Eq. (5) over all  $\mu$  we get

$$\lambda \frac{dJ}{dx} + \frac{\xi J}{\langle \mu \rangle} = 0 \quad (10)$$

where  $J$  is the electron current density and

$$\langle \mu \rangle = \int_{-1}^{+1} F \mu d\mu \left\{ \int_{-1}^{+1} F d\mu \right\}^{-1}$$

or

$$J = J_0 \exp - \left( \int dx' \frac{\xi}{\langle \mu \rangle} \frac{1}{\lambda} \right) \quad (11)$$

Assuming

$$F \sim g(\mu) \exp \left( - \int dx' \frac{\xi}{\langle \mu \rangle} \frac{1}{\lambda} \right) \quad (12)$$

Eq. (5) reduces to

$$\frac{-2\mu g}{\langle \mu \rangle} + 2\xi g - 2\gamma \mu g + \gamma (1 - \mu^2) \frac{dg}{d\mu} = -2g + \int_{-1}^{+1} g(\mu') d\mu' \quad (13)$$

Making a Legendre polynomial expansion of  $g(\mu)$

$$g(\mu) = \sum_{\ell} A_{\ell} P_{\ell}(\mu) \quad (14)$$

Equation (13) can be represented by an infinite set of simultaneous equations

$$A_{n+1} \left( \frac{n+1}{2n+3} \right) \left( \gamma n - \frac{2\xi}{\langle \mu \rangle} \right) + A_n [2(1 - \delta_{0,n}) + 2\xi] - A_{n-1} \left( \frac{n}{2n-1} \right) \left[ \frac{2\xi}{\langle \mu \rangle} + (n+1) \gamma \right] = 0 \quad (15)$$

Using Eq. (15) we may solve for the coefficients  $A_{\ell}$  by truncating the series at some arbitrary  $\ell = L$ . An iterative procedure is necessary as  $\langle \mu \rangle$  is not known a priori. In this manner one may obtain a solution of  $g(\mu)$  to any desired accuracy. Note that when the variation of  $f$  with  $x$  is neglected then this equation takes a simpler form ( $n > 0$ )

$$A_{n+1} \left( \frac{n+1}{2n+3} \right) \gamma n + A_n (2+2\xi) - A_{n-1} \left( \frac{n}{2n-1} \right) (n+1) \gamma = 0 \quad (16)$$

for which an iterative procedure is not necessary. The error in the  $P_1$  approximation may be evaluated by the following ratio

$$\sum_{n=2} A_n^2 / \sum_{n=0} A_n^2 \quad (17)$$

It is interesting to note that the series represented by Eq. (16) converges much more rapidly than Eq. (15) for a given  $\gamma$  and  $\xi$ , signifying that when the spatial derivative is neglected the distribution function is much more isotropic. The error for the case when electron distribution is not a function of  $x$ , is  $\leq 5\%$  for all  $\gamma$  and  $\xi$  values considered in Figure 1. However, the same argument does not apply to the situation when the variation of  $f$  with  $x$  cannot be neglected, as in the case of electron swarm experiments. This result is supported through Figure 1 that shows the contours of the error as a function of  $\gamma$  and  $\xi$ . As one expects, the error increases as both  $\gamma$  and  $\xi$  increase. Figure 2 shows the distribution function obtained from our Boltzmann code along with our estimate of error and the parameters  $\gamma$  and  $\xi$  for an electric field of 1 kV/cm amagat in nitrogen. The error  $< 1\%$  for most of the electrons having an energy  $> 0.2$  eV. The error is large for a few electrons near zero energy as Holstein<sup>2</sup> has pointed out. This is because  $\gamma$  is large in this region. Since relatively large numbers of electrons have energies beyond 0.2 eV for this electric field (as is observed from the distribution



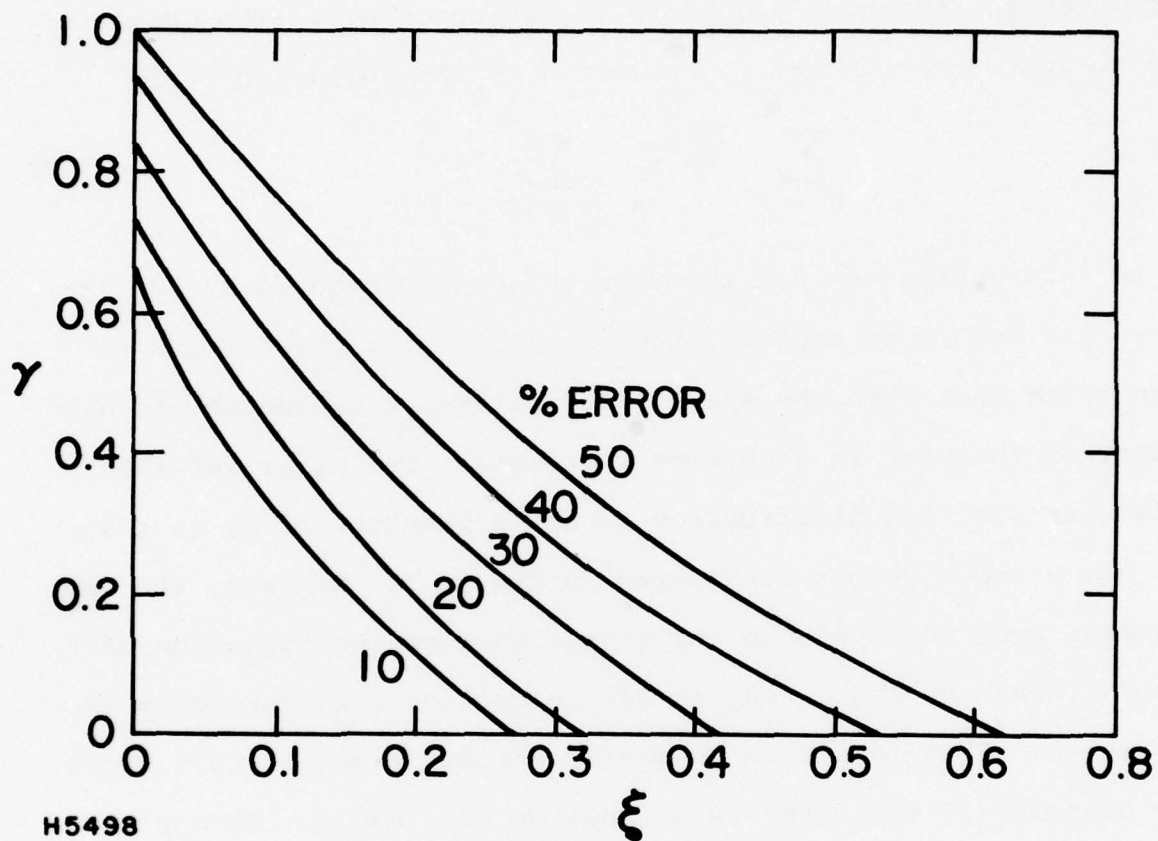


Figure 1 % Error in Using  $P_1$  Approximation for Various Values of Parameters  $\gamma$  and  $\xi$

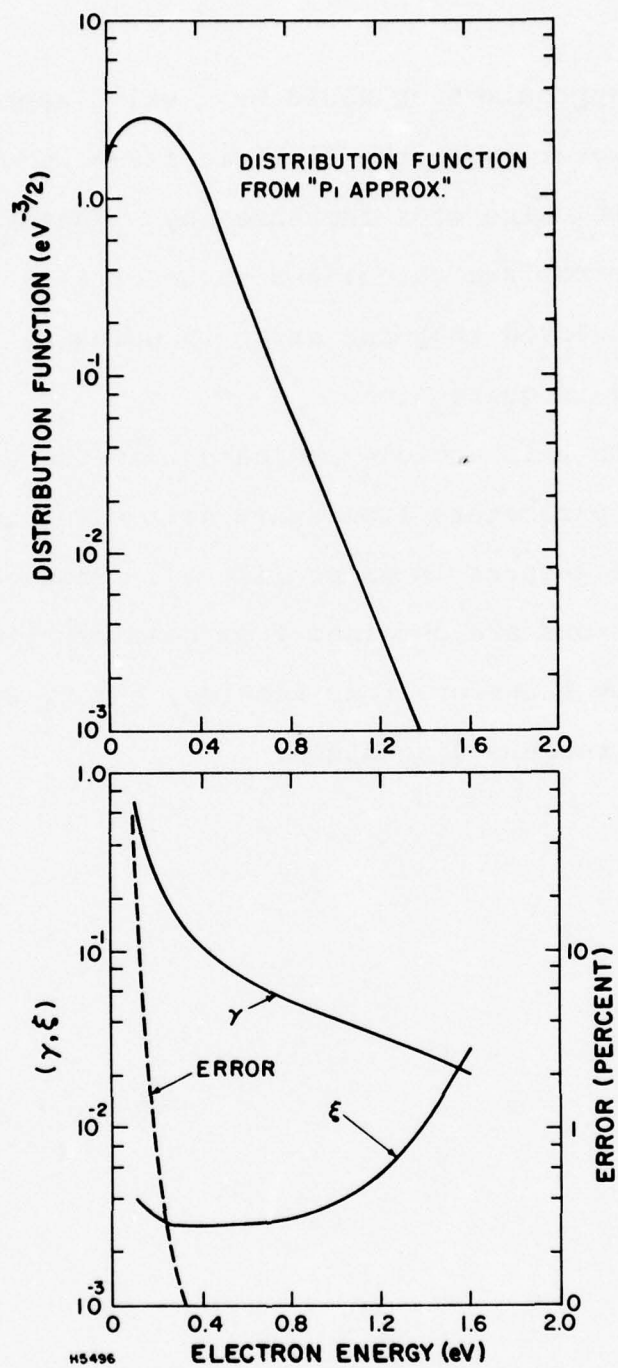


Figure 2 % Error in Using  $P_1$  Approximation for Nitrogen at 1 kV/cm Amagat at Various Electron Energies

function), the  $P_1$  approximation would be a valid approximation for this case. However, when the electric field is increased to 20 kV/cm amagat, the value of  $\gamma$  increases by a factor of 20 and the corresponding error and calculated value of  $\xi$  is shown in Figure 3. It is observed that the error in using  $P_1$  approximations for this case is quite high.

The results in this article indicate that the evaluation of electron impact parameters from swarm data using the  $P_1$  approximation will be suspect in gases like  $N_2$ . However, if the detailed cross sections are obtained from beam experiments, these include the momentum transfer cross section, the  $P_1$  approximation is valid for high pressure discharges.

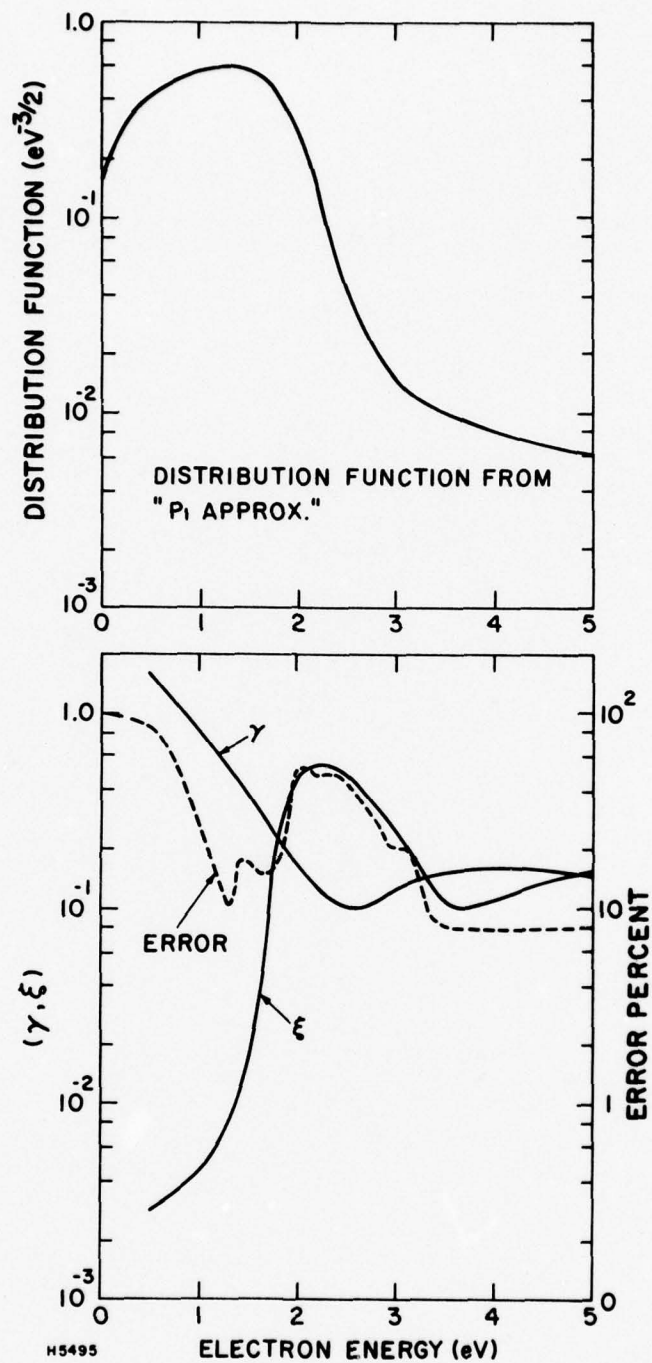


Figure 3 % Error in Using P<sub>1</sub> Approximation for nitrogen at 20 kV/cm Amagat at Various Electron Energies



### III. STABILITY CRITERIA IN METAL VAPOR DISCHARGES

The stability criterion for discharges where excited state (two-step) ionization plays an important role has been previously investigated.<sup>(9)</sup> Such a stability criterion has been shown<sup>(10)</sup> to describe high power rare gas halide laser discharges when the discharge electric field is held constant during the discharge pulse. In this letter, the stability of metal vapor discharges is investigated. Interest in metal vapor lasers has been heightened since the observation of lasing action in  $\text{HgCl}^*$  at  $5576 \text{ \AA}$  by both pure e-beam pumping<sup>(11)</sup> and e-beam controlled discharge pumping.<sup>(12)</sup> In the high power metal vapor discharges, there exists a relatively large excited state population<sup>(13)</sup> which dominates the stability of these discharges. This occurs because typically the excited states have a smaller ionization potential and a larger ionization cross section than the ground state. For high power discharges such as those used in pumping electronic transition lasers, these effects lead to a condition where multi-step ionization dominates. Both attachment and recombination metal vapor discharges have been considered in this paper. The mercury halide lasers are examples of attachment dominated metal vapor discharges. The  $\text{CdHg}^*$  laser<sup>(14)</sup> is typical of recombination dominated metal vapor discharges. In the subsequent discussion on discharge stability, conditions typical of those found in these lasers will be considered as examples of the stability criteria derived.

Figure 4 shows the energy levels of cadmium, mercury and argon (used as a buffer gas). In cadmium for example, it requires 3.7 eV to create  $\text{Cd}^*$  and another 5.3 eV to ionize  $\text{Cd}^*$  whereas it requires only 2.6 eV to produce  $\text{Cd}^{**}$  from  $\text{Cd}^*$  or to ionize  $\text{Cd}^{**}$ . As indicated in Figure 4,  $\text{Cd}^*$  and  $\text{Cd}^{**}$  are a number of relatively closely spaced levels. As the electron temperature will typically be larger than this spacing, we assume that the manifold of states represented by  $\text{Cd}^*$  and  $\text{Cd}^{**}$  will be completely mixed and hence can be represented by a single level. Since the average electron energy in these discharges is 1-1.5 eV three-step ionization tends to dominate at the electron densities of interest. However, if the electron density is large such that the mixing of the two excited states ( $\text{Cd}^*$  and  $\text{Cd}^{**}$ ) by collisions of the first and second kind is far more rapid than excited state reactions with other heavy particles or ionization then the  $\text{Cd}^*$  and  $\text{Cd}^{**}$  may be treated as a single level and the two-step ionization criterion<sup>(9)</sup> derived previously is valid. In the cadmium mercury system, stabilization of the discharge current will be achieved through loss of secondary electrons by recombination. In the case of the  $\text{HgCl}^*$  laser, the excited states  $\text{Hg}^*$  and  $\text{Hg}^{**}$  will play a dominant role in discharge stability and discharge stabilization will be achieved through loss of electrons by attachment.

Three equations are important in determining the stability of the discharge in the presence of rapid excited state ionization. The first describes the production and loss of discharge electrons.

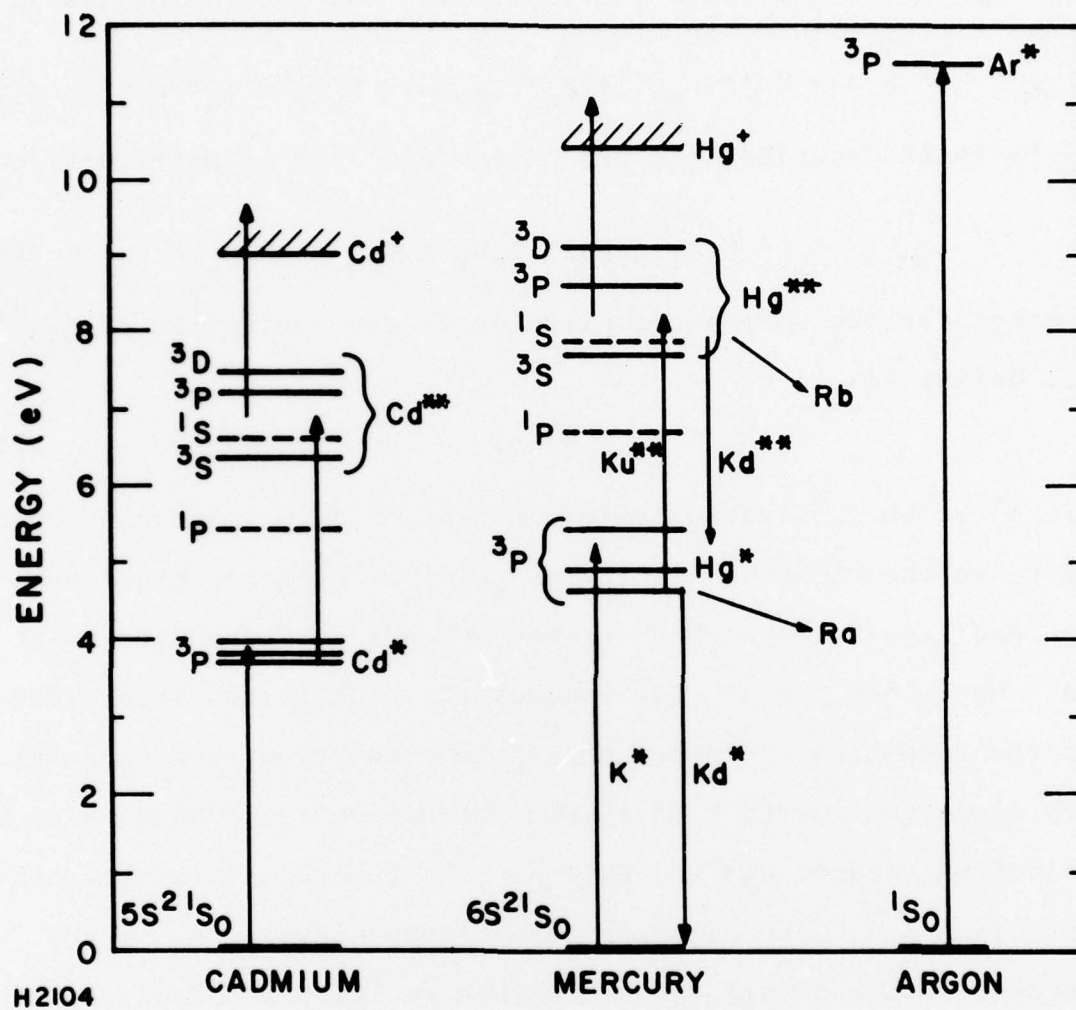


Figure 4 Energy Levels for Cadmium, Mercury and Argon

$$\frac{dn_e}{dt} = S_{eb} + (v_i - \beta)n_e \quad (18)$$

while the second describes the production and loss of  $Hg^*(Cd^*)$ ,

$$\frac{dn_1}{dt} = K^* n_e n - K_u^{**} n_e n_1 + K_d^{**} n_e n_2 - R_a n_1 - K_d^* n_1 n_e \quad (19)$$

and the third describes the production and loss of  $Hg^{**}(Cd^{**})$ ,

$$\frac{dn_2}{dt} = K_u^{**} n_e n_1 - K_d^{**} n_e n_2 - R_b n_2 - v_i n_e \quad (20)$$

Assuming that the dominant ionization is the ionization of  $n_2$ ,<sup>(15)</sup>  $v_i$  is defined as

$$v_i = K^+ n_2 \quad (21)$$

where  $K^+$  is the ionization rate constant of  $n_2$  by electron impact. Here  $n_e$  is the electron density;  $S_{eb}$  is the electron production rate resulting from the high energy e-beam;  $\beta$  is the attachment rate. Note that  $\beta = \alpha n_e$  for recombination dominated laser ( $CdHg^*$ );  $\alpha$  is the recombination rate;  $K^*$  is the production rate constant of  $n_1$  by electron impact;  $K_u^{**}$  is the production rate constant of  $n_2$  by electron impact;  $K_d^*$  and  $K_d^{**}$  are the superelastics rate constant of  $n_1$  and  $n_2$  by electron impact respectively;  $R_a$  is the inverse lifetime of  $n_1$ ;  $R_b$  is the inverse lifetime of  $n_2$ ;  $n$ ,  $n_1$  and  $n_2$  represent the number densities  $Hg(Cd)$ ,  $Hg^*(Cd^*)$  and  $Hg^{**}(Cd^{**})$  respectively. Figure 4 depicts schematically the terminology of the rates described above. In the present analysis we will assume that the ground state density  $n$  is constant. Further, only transitions between adjacent levels have been considered as these rate constants will be by far the most rapid.



Equations (18) to (20) are three nonlinear simultaneous differential equations. If the discharge electric field is a constant in time, we can linearize by letting,

$$n_e = n_{e0} + n_e(t), n_1 = n_{10} + n_1(t), n_2 = n_{20} + n_2(t) \quad (22)$$

The equilibrium conditions,  $n_{e0}$ ,  $n_{10}$  and  $n_{20}$  is readily obtained from Eqs. (18) to (20). The normalized perturbed Eqs. (18) to (20) are given by,

$$\frac{d\bar{n}_e}{dt} = -C_1\bar{n}_e + C_2\bar{n}_2 \quad (23)$$

$$\frac{d\bar{n}_1}{dt} = R_a\bar{n}_e + C_3\bar{n}_2 - C_4\bar{n}_1 \quad (24)$$

$$\frac{d\bar{n}_2}{dt} = R_b\bar{n}_e + C_5\bar{n}_1 - C_5\bar{n}_2 \quad (25)$$

where  $\bar{n}_e = n_e/n_{e0}$ ,  $\bar{n}_1 = n_1/n_{10}$ ,  $\bar{n}_2 = n_2/n_{20}$   
and

$$C_1 = \frac{\alpha n_{e0}^2 + S_{eb}}{n_{e0}} \quad \text{Recombination dominated} \quad (26a)$$

$$= \frac{S_{eb}}{n_{e0}} \quad \text{Attachment dominated} \quad (26b)$$

$$C_2 + K^+ n_{20}, C_3 = \frac{K_d^{**} n_{e0} n_{20}}{n_{10}}, C_4 = R_a + K_u^{**} n_{e0}, C_5 \quad (27a-d)$$

$$= K_d^{**} n_{e0} + R_b$$

Equations (23), (24) and (25) can be combined by elimination to yield

$$\frac{d^3 \bar{n}_e}{dt^3} + A \frac{d^2 \bar{n}_e}{dt^2} + B \frac{d\bar{n}_e}{dt} + C\bar{n}_e = 0 \quad (28)$$

where A, B and C are functions of  $C_1, C_2, C_3, C_4$  and  $C_5$ . For Eq. (28) to be of stable type (i.e., the characteristic equation has real negative roots or the real part of the complex roots be negative) the following conditions<sup>(16)</sup> must be satisfied,

$$A > 0, B > 0, C > 0, AB - C > 0 \quad (29a-d)$$

We have analyzed Eq. (29a-d) in order to establish stability criteria for recombination and attachment dominated discharges. The most restrictive condition is obtained from Eq. (29c) (i.e.,  $C > 0$ ) and is given as,

$$\alpha n_{e0} \geq (1 + \epsilon/2) v_{i0} \quad \text{Recombination dominated} \quad (30a)$$

$$\beta \geq (2 + \epsilon) v_{i0} \quad \text{Attachment dominated} \quad (30b)$$

$$\text{where } v_{i0} = K^+ n_{20}, \quad \epsilon = \frac{A\delta_1\delta_2 - 1}{1 + A(\delta_1 + \delta_2) + A\delta_1\delta_2}, \quad \delta_1 = \frac{R_a}{(K_u^{**} + K_d^*)n_{e0}}$$

$$\delta_2 = \frac{R_b}{(K_d^{**} + K^+)n_{e0}}$$

$$\text{and } A = (K_d^{**} + k_d^+)(K_u^{**} + K_d^*) / (K^+K_u^{**} + K_d^*K^+ + K_d^{**}K_d^*)$$

It can be shown that Eq. (30) is sufficient to ensure the validity of Eq. (29a-d). In such cases Eq. (28) would predict temporally decaying solutions indicating stable discharges. The parameter  $\delta_1$  and  $\delta_2$  are the ratios of characteristic time for loss of electrons by electron impact to that due to heavy particle collisions for the two excited states. It is observed that when  $\delta_2$  or  $\delta_1$

approaches zero (with  $A\delta_1$  or  $A\delta_2 \gg 1$ ), two-step ionization limit similar<sup>(17)</sup> to that derived by Daugherty, Mangano and Jacob<sup>(9)</sup> is recovered. This corresponds to a situation when  $n_1$  and  $n_2$  are completely mixed by electrons. When  $\delta_1$  and  $\delta_2$  both approach zero, the usual single-step stability criteria ( $\beta \geq v_{i_0}$ , and  $\alpha n_{e_0} \geq v_{i_0}$ ) is recovered. For this case the excited state population will come into thermal equilibrium with the secondary electrons.<sup>(18)</sup> It is noticed from the expression for  $\epsilon$  that when electron mixing of the excited states is unimportant (i.e.,  $\delta_1, \delta_2 > 1$ ), its value is bounded (note that  $A \geq 1$ ) by  $0 < \epsilon \leq 1$ . The stability criteria for three-step dominant ionization, therefore, is given by  $\alpha n_{e_0}$  (or  $\beta$ )  $\geq m v_{i_0}$  where  $1 < m \leq 3/2$  for recombination and  $2 < m \leq 3$  for attachment dominated discharge. Our numerical calculations for laser discharges typically indicate  $\delta_1 \approx 2$ ,  $\delta_2 \approx 1.0$  and  $A \approx 1.1$  for CdHg\* and  $\delta_1 \approx 13$ ,  $\delta_2 \approx 1$  and  $A \approx 2$  for HgCl\* discharge. These correspond to values of  $m = 1.1$  and  $2.5$ , respectively. In general, knowledge of the parameters  $\delta_1$ ,  $\delta_2$ ,  $A$  and the equilibrium values,  $n_{e_0}$ ,  $v_{i_0}$ , etc. (obtained by solving Eqs. (18) to (20), would determine the value of  $m$  below which the discharge will go unstable. Note also that when the electron production rate  $S_{eb} = 0$ , then from Eq. (18) the equilibrium condition for an attachment dominated discharge is  $\beta = v_{i_0}$ , which is in violation of Eq. (30) when  $\epsilon > -1$ . Hence, an external source for electron production is essential for discharge stability of an attachment dominated discharge when  $\epsilon > -1$ . By a similar reasoning, a

nonzero  $S_{eb}$  is essential for the stability of a recombination dominated discharge when  $\epsilon > 0$ .

In order to demonstrate the validity of the linearized analysis presented earlier, we have also numerically solved the system of Eqs. (18) to (20). Figure 5 shows the results of such an analysis. Discharge conditions were selected such that  $m \simeq 3$  for this case. On the left-hand side we have the stable discharge condition, i.e., one-third of the attachment rate is slightly greater than the equilibrium ionization rate. Note that the discharge current reaches a constant value asymptotically. If we keep everything constant but decrease the attachment rate by 5%, we observe that the ionization rate increases and after about 0.8  $\mu\text{sec}$  becomes greater than one-third of the attachment rate. The discharge current grows and shows stronger than exponential growth after about 0.8  $\mu\text{sec}$ . In Figure 6 we show a similar behavior for recombination dominated discharge. In this case, discharge conditions were selected such that  $m \simeq 1.5$ . On the left-hand side we have again the stable discharge condition as long as  $\alpha n_e \geq 3/2 v_i$  and the discharge current reaches a constant value asymptotically. If the recombination rate is reduced by 10%, we observe that the ionization rate increases and becomes  $> 3/2 \alpha n_e$  at about 0.7  $\mu\text{sec}$ . The discharge current exhibits a stronger than exponential growth after about 0.7  $\mu\text{sec}$ .

The volumetric instability described in this letter predicts a temporal increase of the secondary electron density that is faster than exponential in a spatially uniform manner. For spatial



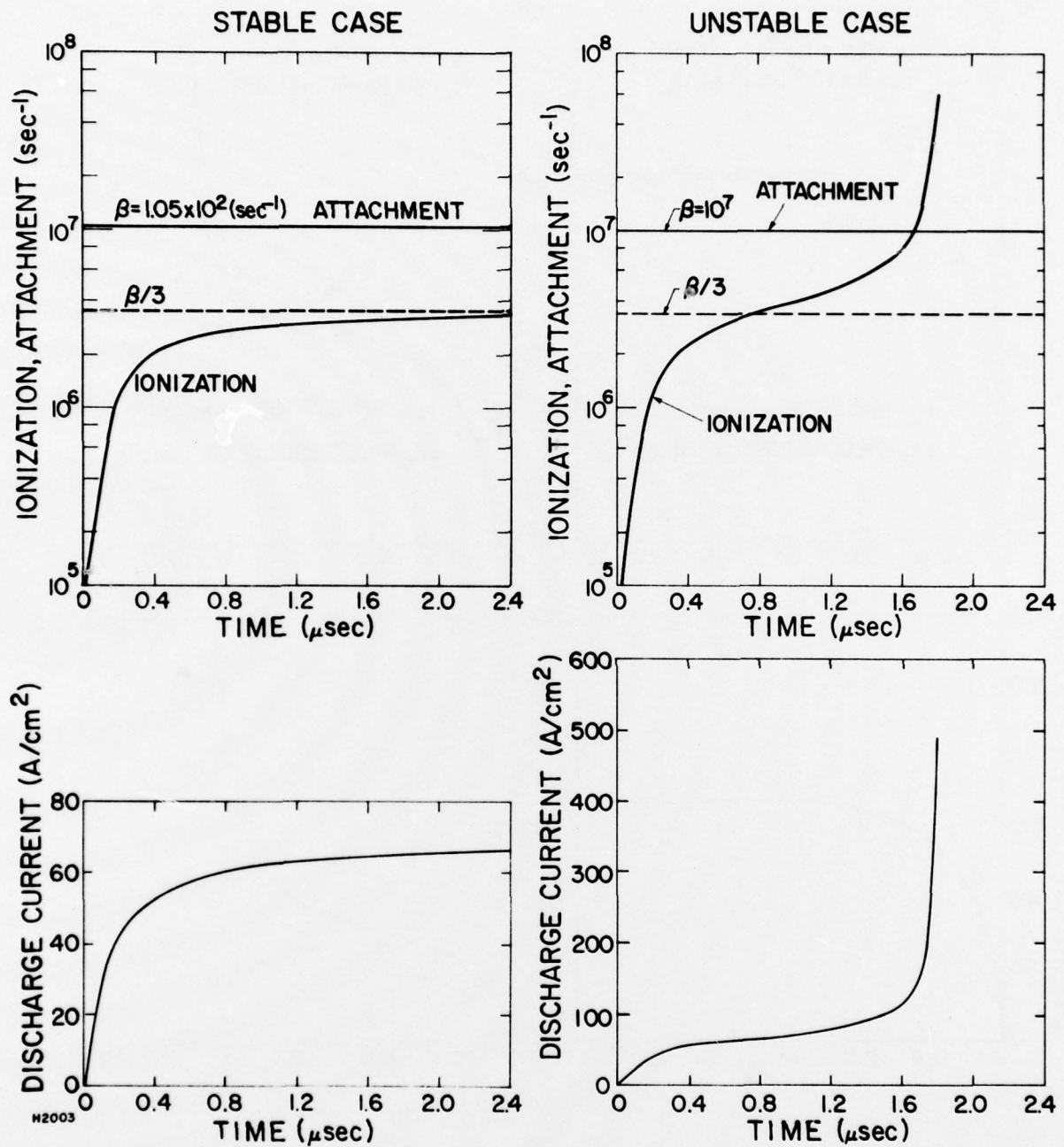


Figure 5 Results of a Numerical Analysis of a System of Eqs. (1)-(3) for Attachment Dominated Discharge

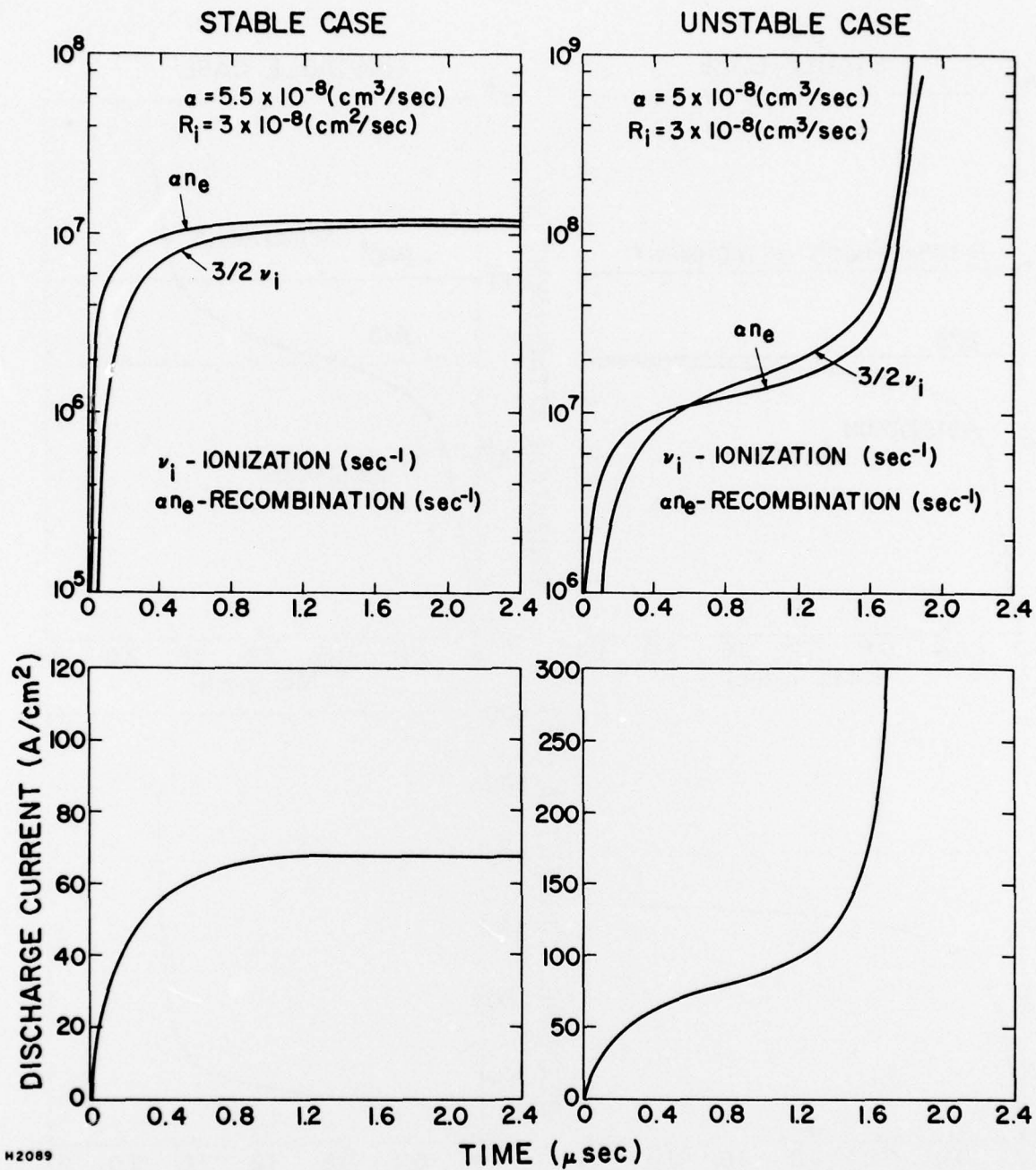


Figure 6 Results of a Numerical Analysis of a System of Eqs. (1)-(3) for Recombination Dominated Discharge

regions in the discharge where the stability criteria derived above are not satisfied, the discharge will go unstable. These locally unstable regions will lead to arc formation if the variation in the power deposition is in the dimension transverse to the electric field. Consequently, for a constant electric field, stability is guaranteed only if the criteria derived above are satisfied at every point in space. In principle, resistive or inductive ballasting can be used to stabilize the discharge. However, the stabilizing influence of the ballast depends on the ratio of the transverse area which is unstable to the total area of the discharge. As this ratio decreases, the effectiveness of resistive or inductive ballasting decreases.

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15. The technique presented here can be further extended by including ionization from other levels.



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17. It can be shown by including the superelastic and ionization loss terms, which were neglected in Eq. (2) of Ref. 1 that the two-step stability criterion for attachment dominated discharges is given as  $\beta \geq \left(\frac{1+2\delta}{1+\delta}\right) v_{i0}$  where  $\delta = R_a / (K_d^* + K^+) n_{e0}$ .
18. It appears that when the excited states are well mixed by the secondary electron the stability criteria given by Eq. (13) is least restrictive. Before such a conclusion can be reached, other factors, such as the variation of  $v_{i0}$  and the efficiency of producing the first excited state, have to be considered. These effects will be discussed more fully in a forthcoming report.

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