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Plasma Stability of Electric Discharges in Molecular Gases

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The results of an analytical treatment of the local low-frequency stability of weakly ionized molecular-gas mixtures consisting of a diatomic molecular species and an atomic diluent are presented. Plasma conditions typical of high-power electric-discharge-laser technology are emphasized. The calculations indicate that small-amplitude fluctuations present within these discharges excite several different wave modes. These have been identified as a space-charge relaxation mode, an electron thermal mode, an ionization mode, a negative-ion-production mode, an electronically-excited-species-production mode, a sound mode, a vibrational-energy relaxation mode, a heavy-particle thermal mode, and a vorticity mode. The stability of these modes is treated in detail with particular emphasis placed on illustrating the influence on stability of charged-particle kinetics, energy transfer, and transport processes. The influence of auxiliary ionization and aerodynamic techniques is also considered.

I. INTRODUCTION

Current interest in the development of powerful electrically excited molecular-gas lasers has focused attention on the problem of producing high-energy-density large-volume convectively cooled glow discharges in molecular-gas mixtures. Of particular interest are the electrically-excited-CO_2 and -CO molecular lasers because of their high efficiency. Theoretical considerations[1–3] indicate that the optical power density in these lasers should increase with increasing electron density and gas pressure provided the gas temperature remains sufficiently low. However, the occurrence of bulk plasma instabilities has inhibited the scaling of laboratory devices to larger sizes and higher pressures.[4–7] These instabilities produce local current constriction within the medium resulting in highly nonuniform excitation and excessive local heating of the gas.

In an effort to circumvent these laser-discharge stability problems, recent attention has been directed toward development of dynamic stabilization techniques such as the introduction of gas dynamic turbulence,[4,5] the superposition of rf and dc electric fields,[5] and the use of high-energy proton[6] and electron[7] beams to provide ionization. The proton- and electron-beam sustained discharges have the additional advantage of permitting independent adjustment of the discharge E/n ratio (electric field intensity/neutral-gas density) and therefore of the electron temperature. The varying degrees of success achieved using these methods of discharge stabilization suggest that the stability of high-power-density discharges is intimately associated with the energy transfer and particle production and loss processes.

In order to develop an understanding of the fundamental physical mechanisms influencing electric discharge stability, an analytical treatment of the local low-frequency stability of a weakly ionized molecular-gas mixture has been developed. In Sec. II the equations, which describe the dynamics of a weakly ionized molecular-gas mixture, are presented. Conditions representative of high-power electric-laser applications are emphasized. Coupling of the translational and molecular vibrational internal degrees of freedom[8,9] and the non-Maxwellian nature of the electron distribution function[10–14] have been included in treating the collisional energy-transfer and particle-production processes. Because of the complexity of the particle-production and -loss mechanisms within molecular-gas discharges and the absence of detailed data regarding these processes, certain aspects of the particle kinetics are treated in a more general fashion.

The analysis of Sec. III shows that small-amplitude fluctuations present within molecular discharge plasmas can excite several different wave modes. The physical conditions for onset of instability and the instability growth characteristics of these different wave modes are treated in detail in Sec. IV, with particular emphasis placed on illustrating the influence on stability of collisional charged-particle-production, energy-transfer, and transport processes. The influence of auxiliary ionization and of aerodynamic techniques on discharge-plasma stability are also considered in this section.

Although the analysis presented here does not attempt to "solve" the complete plasma stability problem for high-power electric discharge lasers, this investigation provides physical insight into the mechanisms that can lead to the discharge instability observed in laboratory devices and suggests areas where further research will prove most useful in overcoming these problems.

II. PLASMA MODEL

Glow discharges[4–7] in mixtures of molecular gases are usually operated at total pressures in the 10–100-torr range. These plasmas are characterized by values of fractional ionization below 10^{-6}, a mean electron energy of approximately 1 eV, and effective vibrational temperatures that occasionally exceed 5000 °K.[2,3,15] In addition, a host of positive and negative ions and neutral minority species are produced in the discharge as a result of numerous plasma-chemical reactions.[16] The general behavior of such a plasma is governed by an exceedingly complex network of energy-transfer and particle-conservation interactions, the most important of which are illustrated by the processes indicated in Fig. 1. Because of the highly nonequilibrium nature of the plasma, several sources of instability exist, and a rather extensive description of the plasma is required in order to determine the dominant physical processes corresponding to a particular combination of experimentally interesting circumstances. Nevertheless, a detailed computer calculation[17] of the stability of N_2-CO_2-He discharge plasmas has shown that assumptions are possible which permit retention of the essential physical features pertinent to the present discussion, while greatly reducing the complexity of the analysis. On the basis of such considerations, in the present analysis the gas mixture is taken to be composed of a single diatomic molecular species and an atomic diluent. In addition, it is assumed that there exists only one species each of positive and negative ions and a single electronically excited species.[18] In the paragraphs to follow, the particle continuity, momentum, and energy equations appropriate to the description of such a plasma are derived and certain other features of the model discussed.

A. Dynamics of the Heavy Particles

Particle Conservation

When chemical decomposition and diffusive separation of the initial mixture are small then the equation of continuity for the gas may be written

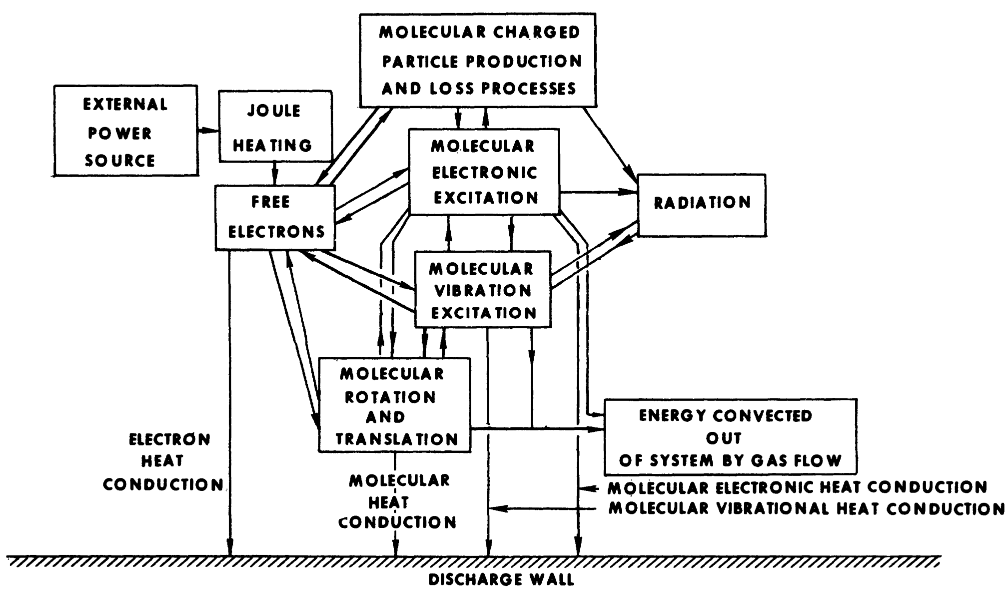

FIG. 1. Energy-transfer processes in molecular-gas discharges.

$$\frac{Dn}{Dt} + n\nabla\cdot\vec{u} = 0, \qquad (1)$$

where $D/Dt = \partial/\partial t + \vec{u}\cdot\nabla$, $\vec{u}$ is the mass average velocity, and n is the sum of the atom and molecule densities, $n = n_a + n_m$.

For conditions typical of low-pressure, weakly ionized discharges in molecular gases, ion-production and -loss processes are generally of importance.[16] These along with their respective rate coefficients are: electron-impact ionization of molecules (k_i) and electronically excited species (k_i^*),[18] electron-molecule attachment (k_a), electron excitation of electronic species (k_*), quenching of electronic species (k_q), two-body electron-ion recombination ($k_r^{(e)}$), positive-ion–negative-ion recombination ($k_r^{(i)}$), and detachment by neutral impact (k_d). Accordingly, the conservation equations for the positive-ion, negative-ion, and electronically-excited-species densities are, respectively,

$$\frac{Dn_p}{Dt} + n_p\nabla\cdot\vec{u} + \nabla\cdot(n_p\vec{U}_p) = n_e n k_i(T_e) + n_e n_* k_i^*(T_e) - n_e n_p k_r^{(e)}(T_e) - n_p n_n k_r^{(i)}(T) + n\frac{S_{e\,\mathrm{ext}}}{n}, \qquad (2)$$

$$\frac{Dn_n}{Dt} + n_n\nabla\cdot\vec{u} + \nabla\cdot(n_n\vec{U}_n) = n_e n k_a(T_e) - n_p n_n k_r^{(i)}(T) - n_n n k_d(T), \qquad (3)$$

and

$$\frac{Dn_*}{Dt} + n_*\nabla\cdot\vec{u} = n_e n k_*(T_e) - n_e n_* k_i^*(T_e) - n_* n k_q(T) + n\frac{S_{*\,\mathrm{ext}}}{n}, \qquad (4)$$

where the dependence of the rate coefficients k_j on electron kinetic temperature[19] T_e and gas temperature T is indicated. The density of the electronically excited species is determined by collisional instead of radiative processes. The effect of vibrational excitation on the ionization, attachment, detachment, and quenching processes has been omitted but may be important. In addition, the existence of an auxiliary, independently controllable source of ionization and excitation has been assumed and is designated $S_{j\,\mathrm{ext}}$. Such a source may be in the form of an electron beam,[7] a photo source, etc. Production of negative ions by these means is assumed to be relatively unimportant. Consistent with the neglect of diffusion of neutral species, only the contribution from the electric field is included in describing the diffusion velocities of the ionic species, i.e., $\vec{U}_p = \mu_p\vec{E}$ and $\vec{U}_n = -\mu_n\vec{E}$, where μ_p and μ_n are the positive- and negative-ion mobilities.

Momentum Equation

The momentum equation for the mixture may be expressed

$$\rho\frac{D\vec{u}}{Dt} = -\nabla p + \mu\nabla^2\vec{u} + (\mu_B + \tfrac{1}{3}\mu)\nabla(\nabla\cdot\vec{u}) + \rho_c\vec{E}, \qquad (5)$$

where $\rho = m_a n_a + m_m n_m$, $\rho_c = e(n_p - n_n - n_e)$, and $p = n\kappa T$ are the mass density, charge density, and pressure of the gas. In the absence of magnetic effects, the last term in Eq. (5) represents the transfer of momentum from the electric field to the ions and electrons. The kinematic and bulk viscosity coefficients are μ and μ_B, respectively. The bulk viscosity arises as a consequence of the rapid molecular rotational relaxation[20] during dilatation of fluid elements.

Translation-Rotation and Vibrational Energy Equations

Since the translational and rotational degrees of freedom of the heavy particles are closely coupled and are therefore near equilibrium, it is appropriate to introduce an energy equation governing energy transfer to and from these coupled degrees of freedom. When the contributions from diffusive separation, electronic quenching, and ion Joule heating are small, the translation-rotation energy equation[21] may be written

$$\frac{D}{Dt}(n\mathcal{E}_{\mathrm{TR}}) + (n\mathcal{E}_{\mathrm{TR}} + p)\nabla\cdot\vec{u} = -\nabla\cdot\vec{q}_{\mathrm{TR}} + \mu\Phi + \frac{n_m}{\tau_{\mathrm{VT}}}[\mathcal{E}_V(T_V) - \mathcal{E}_V(T)] + n_e n\frac{\nu_{\mathrm{TR}}(T_e)}{n}\kappa T_e, \qquad (6)$$

where $\mathcal{E}_{\mathrm{TR}} = (3n_a + 5n_m)\kappa T/(2n)$ is the average translation-rotation energy per particle, $\mathcal{E}_V(T_V) = \epsilon[e^{\epsilon/\kappa T_V} - 1]^{-1}$ is the average vibrational energy per molecule,[22] and ϵ is the quantum of vibrational energy. The characteristic time for vibrational relaxation, due predominantly to the atomic diluent, is $\tau_{\mathrm{VT}} = [n_a k_{10}(T)(1 - e^{-\epsilon/\kappa T})]^{-1}$, where k_{10} is the rate coefficient[8,9] for deactivation of the first vibrational level. The quantity ν_{TR} is the translation-rotation contribution to the total electron-energy-exchange-collision frequency[12–14] ν_u, and Φ is the viscous dissipation function.[20] If contributions from diffusion and vibrational temperature gradients are small, the translation-rotation energy flux[21] is given by

$$\vec{q}_{\mathrm{TR}} = -\lambda_T^T\nabla T, \qquad (7)$$

where λ_T^T is the thermal conductivity including translation and rotation contributions.

To the same degree of approximation the vibrational energy equation may be written

$$\frac{D}{Dt}[n_m \mathcal{E}_V(T_V)] + n_m \mathcal{E}_V(T_V)\nabla\cdot\vec{u}$$
$$= -\nabla\cdot\vec{q}_V + n_e n \frac{\nu_V(T_e)}{n}\kappa T_e - \frac{n_m}{\tau_{VT}}[\mathcal{E}_V(T_V) - \mathcal{E}_V(T)], \quad (8)$$

where ν_V is the vibrational contribution to the electron-energy-exchange-collision frequency and the contribution from electronic quenching is assumed to be small. The vibrational energy-flux vector is[21]

$$\vec{q}_V = -\lambda_V^V \nabla T_V \quad (9)$$

and λ_V^V is the vibrational conductivity. The relative contributions to the energy-flux vectors, Eqs. (7) and (9), due to diffusion will be small when fluctuations in gas and vibrational temperatures are large relative to fluctuations in mixture partial pressures.

B. Dynamics of the Electrons and Electromagnetic Field

Electron Continuity and Energy Equations

The electron continuity equation for the plasma kinetic model under consideration may be written

$$\frac{Dn_e}{Dt} + n_e\nabla\cdot\vec{u} + \nabla\cdot(n_e\vec{U}_e) = n_e n k_i(T_e) + n_e n_* k_i^*(T_e) + n_n n k_d(T) - n_e n_p k_r^{(e)}(T_e) - n_e n k_a(T_e) + n\frac{S_{e\,\mathrm{ext}}}{n}, \quad (10)$$

where the electron drift velocity $\vec{U}_e$ is given by

$$n_e\vec{U}_e = -\nabla(n_e D_e) - n_e D_e \nabla(\ln n) - n_e\mu_e\vec{E}. \quad (11)$$

The diffusion coefficient D_e and mobility μ_e are related to the effective electron-momentum-transfer-collision frequency[12-14] ν_m by the relations $D_e = (\kappa T_e/m_e)\nu_m^{-1}$ and $\mu_e = (e/m_e)\nu_m^{-1}$.

The corresponding electron energy equation is expressed

$$\frac{D}{Dt}(\tfrac{3}{2}n_e\kappa T_e) + (\tfrac{3}{2}n_e\kappa T_e + p_e)\nabla\cdot\vec{u} = \vec{j}_e\cdot\vec{E} - \nabla\cdot\vec{q}_e - n_e n[\nu_u(T_e)/n]\kappa T_e, \quad (12)$$

where $p_e = n_e\kappa T_e$ is the electron pressure, $\vec{j}_e = -en_e\vec{U}_e$ is the electron current density, and ν_u is the electron-energy-exchange-collision frequency.[23] For the present purposes the electron-energy-flux vector may be written approximately as

$$\vec{q}_e = -\lambda_e\nabla T_e - \tfrac{5}{2}(\kappa T_e/e)\vec{j}_e, \quad (13)$$

where $\lambda_e = \frac{5}{2}\kappa n_e D_e$ is the effective-electron thermal conductivity.

Maxwell's Equations

For the low-frequency disturbances of interest in the present investigation, the electromagnetic field is quasistatic and hence magnetic interactions are unimportant. Thus the electric field is determined by

$$\nabla\cdot\vec{E} = \rho_c/\epsilon_0 \quad\text{and}\quad \nabla\times\vec{E} = 0. \quad (14)$$

III. STABILITY ANALYSIS

A. Small-Amplitude Fluctuations

The equations of motion developed in Sec. II can be used to provide a self-consistent locally valid description of nonlinear-collision-dominated disturbances within the plasma. In large-volume fast-flow electric discharge devices the transport processes are sufficiently slow that large gradients in plasma properties are confined to relatively narrow regions adjacent to the walls of the device. In regions away from the boundaries the plasma properties vary relatively smoothly. Owing to electron translation-rotation excitation and vibrational relaxation there exists a gradient in gas temperature and consequently plasma properties along the direction of flow through the discharge; however, the local plasma state for the most part is determined by collisional rather than transport processes.

In practice, the interior of a high-energy discharge is never locally steady or uniform, because of the presence of plasma fluctuations, noise generated by flow turbulence,[4,5] power-supply ripple, etc. When the amplitude of the fluctuations is sufficiently small, it is useful to decompose plasma properties into the sum of a nearly spatially invariant steady-state value, $\Psi(\vec{x})$, and a spatially and temporally varying fluctuation, $\psi'(\vec{x},t)$ normalized by Ψ, i.e., $\Psi(\vec{x},t) = \Psi(\vec{x})[1+\psi'(\vec{x},t)]$. The symbol $\Psi(\vec{x},t)$ denotes the set of fundamental plasma properties required to describe the behavior of the plasma, $\Psi \equiv \{n, n_e, n_p, \ldots, T, T_V\}$. Using this form for the disturbance, the governing conservation equations developed in Sec. II may be decomposed into their zero and first-order linear components which describe, respectively, the steady-state and the fluctuating quantities. The stability of these small-amplitude fluctuations is the subject of the present analysis.

B. Steady State

As a consequence of the energy deposition within the gas during its passage through the discharge, the steady state is governed by a system of differential equations derivable directly from Eqs. (1)–(14) by deletion of the partial time derivatives. In the plasmas of interest for laser application the residence time of a fluid element within the discharge region is generally long compared with the characteristic times (Table I) associated with plasma kinetic processes, and the spatial derivatives in the steady continuity equations for electrons, positive ions, negative ions, electronically excited species, the electron energy, and the vibrational energy are small and may be neglected. Consequently, the local plasma state is determined by the local gas temperature, E/n, gas mixture, and pressure together with

$$\left(\frac{1}{\tau_i}+\frac{1}{\tau_i{}^*}-\frac{1}{\tau_r^{(e)}}-\frac{1}{\tau_a}\right)+\frac{n_n}{n_e}\frac{1}{\tau_d}+\frac{S_{e\,\mathrm{ext}}}{n_e}=0\,, \quad (15)$$

$$\frac{n_e}{n_n}\frac{1}{\tau_a}-\frac{1}{\tau_r^{(i)}}-\frac{1}{\tau_d}=0\,, \quad (16)$$

$$\frac{n_e}{n_*}\left(\frac{1}{\tau_*}-\frac{1}{\tau_i^*}\right)-\frac{1}{\tau_q}+\frac{S_{*\,\mathrm{ext}}}{n_*}=0\,, \quad (17)$$

$$\frac{n_m}{\tau_{\mathrm{VT}}}[\mathcal{E}_V(T_V)-\mathcal{E}_V(T)]=n_e n\frac{\nu_V}{n}\kappa T_e\,, \quad (18)$$

and

$$J_e^2/\sigma_e=n_e n(\nu_u/n)\kappa T_e\,. \quad (19)$$

The positive-ion density is related to the electron and negative-ion densities by the charge-neutrality condition. The characteristic plasma kinetic times for direct ionization τ_i, ionization of electronically excited species τ_i^*, detachment by neutral impact τ_d, electron attachment τ_a, electron-ion recombination $\tau_r^{(e)}$, ion-ion recombination $\tau_r^{(i)}$, electronic excitation τ_*, and electronic quenching τ_q are defined by the relations given in Table I.

C. Description of the Perturbed State

For the purposes of the present treatment, the equations governing the temporal and spatial evolution of a disturbance are determined by collecting the first-order components of the equations obtained by substituting $\Psi(\vec{x},t)$ into the equations of Sec. II. If the characteristic dimension l of a spontaneous disturbance within the plasma is small compared to the dimensions of the discharge, the effects of steady-state gradients are small and may be neglected in the equations describing the initial development of the fluctuation. However, the gradients of the fluctuating quantities must be retained, since they represent the local spatial development (i.e., propagation, diffusion) of a fluctuation.

Mixture Continuity and Momentum Equations

Under these conditions the equation of continuity for the neutral-gas fluctuations may be written

$$\frac{Dn'}{Dt}+\frac{1}{\tau_s}\nabla\cdot\vec{u}'=0\,, \quad (20)$$

where the spatial variable has been normalized by the characteristic length l and the velocity fluctuation has been normalized to the local frozen sound speed $a=(\gamma\kappa T/m)^{1/2}$ with the specific-heat ratio $\gamma=C_p/C_v$. The specific heats at constant volume C_v and pressure C_p are given by $C_v=(3n_a+5n_m)\kappa/2n$ and $C_p=\kappa+C_v$. The characteristic time $\tau_s\equiv l/a$ is a measure of the time for sound to cross the disturbance.

Introducing the vorticity fluctuation $\vec{\Omega}'=\nabla\times\vec{u}'$, the momentum equation governing fluctuations may be replaced by equations governing vorticity and pressure fluctuations,

$$\frac{D\vec{\Omega}'}{Dt}-\left(\frac{\mu}{\rho l^2}\right)\nabla^2\vec{\Omega}'=\left(\frac{en_pE}{\rho a^2/l}\right)\frac{1}{\tau_s}(\nabla\rho_c')\times\left(\frac{\vec{E}}{E}\right) \quad (21)$$

and

$$\frac{D^2p'}{Dt^2}-\frac{1}{\tau_s{}^2}\nabla^2p'=\gamma\left(\frac{\mu_B+\frac{4}{3}\mu}{\rho l^2}\right)\nabla^2\left(\frac{Dn'}{Dt}\right)+\left(\frac{\lambda_T^T}{nC_v l^2}\right)\nabla^2\left(\frac{DT'}{Dt}\right)-\left(\frac{en_pE}{\rho a^2/l}\right)\frac{\gamma}{\tau_s^2}(\nabla\rho_c')\cdot\left(\frac{\vec{E}}{E}\right)+\frac{D\dot{Q}'_{\mathrm{TR}}}{Dt}\,, \quad (22)$$

where $\dot{Q}'_{\mathrm{TR}}$ is the fluctuation in the volumetric heating of the translation-rotation degree of freedom of the neutral gas and

$$\frac{1}{\gamma}\frac{D\dot{Q}'_{\mathrm{TR}}}{Dt}=\frac{1}{\tau_T}(2P_V+P_{\mathrm{TR}})\frac{Dn'}{Dt}+\frac{1}{\tau_T}\left[-P_V\hat{\tau}_{\mathrm{VT}}-\frac{C_v^V(T)T}{C_v^V(T_V)T_V}\left(\frac{\tau_V}{\tau_{\mathrm{VT}}}\right)\right]\frac{DT'}{Dt}+\frac{1}{\tau_{\mathrm{VT}}}\left(\frac{n_mC_v^V(T_V)T_V}{nC_pT}\right)\frac{DT_V'}{Dt}+\frac{P_{\mathrm{TR}}}{\tau_T}\frac{Dn_e'}{Dt}+\frac{P_{\mathrm{TR}}}{\tau_T}(1+\hat{\nu}_{\mathrm{TR}})\frac{DT_e'}{Dt}. \quad (23)$$

The charge-density fluctuation ρ_c' is normalized by en_p. The quantities P_V and P_{TR} are defined by

$$P_V=\frac{n_m}{\tau_{\mathrm{VT}}}\frac{\mathcal{E}_V(T_V)-\mathcal{E}_V(T)}{J_e^2/\sigma_e}\sim\frac{n_e n(\nu_V/n)\kappa T_e}{J_e^2/\sigma_e} \quad (24)$$

and

$$P_{\mathrm{TR}}=\frac{n_e n(\nu_{\mathrm{TR}}/n)\kappa T_e}{J_e^2/\sigma_e}\,, \quad (25)$$

and consequently are a measure of the fractional power transfer from electrons into vibration and into translation-rotation excitation. The time scales $\tau_T\equiv nC_pT/J_e^2/\sigma_e$ and $\tau_V\equiv n_mC_v^V(T_V)T_V/J_e^2/\sigma_e$ (Table I) represent the characteristic times for heating the gas and the vibrational degree of freedom within the discharge. The vibrational specific heat is defined by $C_v^V(T_V)\equiv\partial\mathcal{E}_V(T_V)/\partial T_V$. The

TABLE I. Characteristic times. Typical discharge conditions (diatomic molecule-atom mixture, $p \sim 10$–100 torr, $T \sim 300$–600 °K, $T_e \sim 0.5$–2.0 eV, $T_V \sim 1000$–5000 °K, $n_e/n \sim 10^{-8}$–10^{-6}, $l \sim 1$ cm; for a plane wave with wave number k the characteristic length $l = k^{-1}$).

	Process	Characteristic time	Range of values (sec)
I.	Space-charge relaxation	$\tau_C = \epsilon_0/\sigma \sim \nu_m/\omega_{pe}^2$	10^{-10}–10^{-9}
II.	Collisional energy transfer		
i.	electron heating	$\tau_e = (\frac{3}{2})n_e \kappa T_e/(J_e^2/\sigma_e) = \frac{3}{2}\nu_u^{-1}$	10^{-9}–10^{-8}
ii.	translation-rotation heating	$\tau_T = n C_p T/(J_e^2/\sigma_e) \sim (n/n_e)(C_p/\kappa)(T/T_e)\nu_u^{-1}$	10^{-3}–10^{-2}
iii.	vibration heating	$\tau_V = n_m C_v^V(T_V)T_V/(J_e^2/\sigma_e) \sim (n_m/n_e)[C_v^V(T_V)/\kappa](T_V/T_e)\nu_u^{-1}$	10^{-3}–10^{-2}
iv.	vibrational relaxation	$\tau_{VT} = [n_a k_{10}(1 - e^{-\epsilon/\kappa T})]^{-1}$	10^{-4}–10^{-2}
III.	Plasma kinetic processes		
i.	direct ionization	$\tau_i = [n k_i]^{-1}$	10^{-6}–10^{-5}
ii.	electronic state ionization	$\tau_i^* = [n_* k_i^*]^{-1}$	10^{-6}–10^{-4}
iii.	attachment (dissociative)	$\tau_a = [n k_a]^{-1}$	10^{-6}–10^{-5}
iv.	detachment (direct neutral and associative)	$\tau_d = [n k_d]^{-1}$	10^{-6}–10^{-5}
v.	electronic excitation	$\tau_* = [n k_*]^{-1}$	10^{-6}–10^{-4}
vi.	electronic quenching	$\tau_q = [n_* k_q]^{-1}$	10^{-6}–10^{-4}
vii.	electron-ion dissociative recombination	$\tau_r^{(e)} = [n_p k_r^{(e)}]^{-1}$	10^{-6}–10^{-5}
viii.	ion-ion recombination	$\tau_r^{(i)} = [n_p k_r^{(i)}]^{-1}$	10^{-6}–10^{-5}
IV.	Transport and other processes		
i.	sound propagation	$\tau_{S_T} = l/a$	10^{-5}–10^{-4}
ii.	translation-rotation energy conduction	$\tau_T = (n C_v l^2/\lambda_T^T)$	10^{-3}–10^{-2}
iii.	viscous dissipation	$\tau_\mu = \rho l^2/\mu$	10^{-3}–10^{-2}
iv.	vibrational energy conduction	$\tau_V = n_m C_v^V(T_V) l^2/\lambda_V^V$	10^{-2}–10^{-1}
v.	electron thermal conduction	$\tau_{Te} = (\frac{3}{2})n_e \kappa l^2/\lambda_e$	10^{-7}–10^{-6}
vi.	ambipolar diffusion	$\tau_{amb} = l^2/[(1 - \sigma_e/\sigma)D_e]$	10^{-5}–10^{-4}
vii.	negative-ion diffusion	$\tau_{neg} = l^2/[(\sigma_n/\sigma)(n_e/n_n)D_e]$	10^{-5}–10^{-4}

quantities $\hat{\tau}_{VT} \equiv \partial \ln\tau_{VT}/\partial \ln T$ and $\hat{\nu}_{TR} \equiv \partial \ln\nu_{TR}/\partial \ln T_e$ reflect the sensitivity of the volumetric heating rate of the gas translation-rotation degree of freedom to fluctuations in gas temperature and electron kinetic temperature, respectively. For the sake of notational simplicity in the discussion to follow the logarithmic derivatives will be written using the caret notation, e.g., $\hat{a} = \partial \ln a/\partial \ln T_j$. From Eq. (21) gradients in charge-density fluctuation normal to the applied electric field can generate vorticity in the fluid. However, the vorticity is also damped by viscous shearing stresses. Note that the bulk viscosity associated with expansion and contraction of fluid elements due to rotational relaxation plays no direct role in vorticity dissipation. From Eq. (22) pressure fluctuations are dissipated by viscosity and thermal conduction but may be amplified by the gradient of the charge-density fluctuation in the direction of the applied electric field and by the rate of volumetric heating of the translation-rotation degree of freedom.

Translation-Rotation and Vibrational Energy Equations

The translation-rotation and vibrational energy equations governing small amplitude fluctuations obtained from Eqs. (6) and (8) may be expressed

$$\frac{DT'}{Dt} - (\gamma - 1)\frac{Dn'}{Dt} = \left(\frac{\lambda_T^T}{nC_v l^2}\right)\nabla^2 T' + \frac{\gamma}{\tau_T}\left(2P_V + P_{TR}\right)n' + \frac{\gamma}{\tau_T}\left[-P_V\hat{\tau}_{VT} - \frac{C_v^V(T)T}{C_v^V(T_V)T_V}\left(\frac{\tau_V}{\tau_{VT}}\right)\right]T'$$
$$+ \frac{1}{\tau_{VT}}\left(\frac{n_m C_v^V(T_V)T_V}{nC_v T}\right)T_V' + \frac{\gamma P_{TR}}{\tau_T}n_e' + \frac{\gamma P_{TR}}{\tau_T}(1 + \hat{\nu}_{TR})T_e' \tag{26}$$

and

$$\frac{DT_V'}{Dt} = \left(\frac{\lambda_V^V}{n_m C_v^V(T_V)l^2}\right)\nabla^2 T_V' - \frac{P_V}{\tau_V}n' + \frac{1}{\tau_V}\left[P_V\hat{\tau}_{VT} + \frac{C_v^V(T)T}{C_v^V(T_V)T_V}\left(\frac{\tau_V}{\tau_{VT}}\right)\right]T' - \frac{1}{\tau_{VT}}T_V' + \frac{P_V}{\tau_V}n_e' + \frac{P_V}{\tau_V}(1 + \hat{\nu}_V)T_e' , \tag{27}$$

where $\hat{\nu}_V$ reflects the sensitivity of vibrational-excitation-energy transfer from electrons due to fluctuations in electron kinetic temperature. From Eqs. (26) and (27) it can be seen that τ_T, τ_V, and τ_{VT} are the characteristic times for variation of energy residing in the translation-rotation and vibrational degrees of freedom during a disturbance.

Conservation of Charge

In slowly changing disturbances the charge-density fluctuation is small, $n_p \simeq n_e + n_n$, and the number of charged-particle equations can be reduced by one. It is therefore more convenient to replace the positive-ion continuity equation with the equation governing charge-density fluctuations,

$$\frac{D\rho_c'}{Dt} + \frac{\sigma}{\epsilon_0}\rho_c' + \frac{\sigma}{\epsilon_0}\left(\frac{\epsilon_0\vec{E}}{en_p l}\right)\cdot\nabla\left[\frac{\sigma_p}{\sigma}\rho_c' + \left(\frac{\sigma_e}{\sigma} + \frac{n_e}{n_p}\frac{\sigma_p}{\sigma}\right)n_e' + \left(\frac{\sigma_n}{\sigma} + \frac{n_n}{n_p}\frac{\sigma_p}{\sigma}\right)n_n' - n' - \frac{\sigma_e}{\sigma}\hat{\nu}_m T_e' + \frac{\sigma_i}{\sigma}\hat{\sigma}_i T'\right] + \frac{n_e}{n_p}\frac{D_e}{l^2}\nabla^2\eta_e' = 0 , \tag{28}$$

where $\eta_e' = n_e' + (1 - \hat{\nu}_m)T_e'$ and the electrical conductivity of the plasma is $\sigma = \sigma_i + \sigma_e$ with the ion contribution given by $\sigma_i = \sigma_p + \sigma_n$. The individual conductivities are related to the appropriate particle mobilities by $\sigma_e = en_e\mu_e$, $\sigma_n = en_n\mu_n$, and $\sigma_p = en_p\mu_p$. The second term in Eq. (28) represents the relaxation of space charge due to the induced electric field. For typical discharges where $\sigma \sim \sigma_e$, the characteristic time for space-charge relaxation, $\tau_c \equiv \epsilon_0/\sigma$, is given approximately by $\nu_m\omega_{pe}^{-2}$, where ω_{pe} is the electron-plasma frequency. The third term in Eq. (28) represents the transport of charge-density fluctuations due to particle drift. Typically $\epsilon_0 E/en_p l \ll 1$, and the charge-density fluctuations do not drift very far during relaxation. The last term represents the dissipation of charge-density fluctuations by diffusion and is negligible compared to the other terms except for very small disturbances.

Electron, Negative-Ion, and Electronically-Excited-Species Conservation

The continuity equations governing fluctuations in electron, negative-ion, and electronically-excited-species densities are obtained from the corresponding continuity equations for these species combined with Poisson's equation and the equation of charge continuity and may be expressed for electrons,

$$\frac{Dn'_e}{Dt} + \frac{1}{\tau_s}\nabla\cdot\vec{u}' - \left(1 - \frac{\sigma_e}{\sigma}\right)\frac{D_e}{l^2}\nabla^2\eta'_e - \frac{\sigma_e}{\epsilon_0}\left(\frac{\epsilon_0\vec{E}}{en_e l}\right)\cdot\nabla\left[-\frac{\sigma_p}{\sigma}\rho'_c + \left(1 - \frac{\sigma_e}{\sigma} - \frac{n_e}{n_p}\frac{\sigma_p}{\sigma}\right)n'_e - \left(\frac{\sigma_n}{\sigma} + \frac{n_n}{n_p}\frac{\sigma_p}{\sigma}\right)n'_n - \left(1 - \frac{\sigma_e}{\sigma}\right)\hat{\nu}_m T'_e - \frac{\sigma_i}{\sigma}\hat{\sigma}_i T'\right]$$
$$= -\frac{\sigma_e}{\sigma}\frac{n_p}{n_e}\frac{D\rho'_c}{Dt} - \frac{1}{\tau_r^{(e)}}\rho'_c - \left(\frac{n_e}{n_p}\frac{1}{\tau_r^{(e)}} + \frac{n_n}{n_e}\frac{1}{\tau_d} + \frac{S_{e\,\mathrm{ext}}}{n_e}\right)n'_e + \left(\frac{n_n}{n_e}\frac{1}{\tau_d} - \frac{n_n}{n_p}\frac{1}{\tau_r^{(e)}}\right)n'_n + \frac{1}{\tau_i^*}n'_* + \left(\frac{1}{\tau_r^{(e)}} - \frac{1}{\tau_i^*}\right)n'$$
$$+ \frac{1}{\tau_i}\left(\hat{k}_i + \frac{\tau_i}{\tau_i^*}\hat{k}_i^* - \frac{\tau_i}{\tau_r^{(e)}}\hat{k}_r^{(e)} - \frac{\tau_i}{\tau_a}\hat{k}_a\right)T'_e + \frac{n_n}{n_e}\frac{1}{\tau_d}\hat{k}_d T', \tag{29}$$

for negative ions,

$$\frac{Dn'_n}{Dt} + \frac{1}{\tau_s}\nabla\cdot\vec{u}' + \frac{\sigma_n}{\sigma}\frac{n_e}{n_n}\frac{D_e}{l^2}\nabla^2\eta'_e - \frac{\sigma_n}{\epsilon_0}\left(\frac{\epsilon_0\vec{E}}{en_n l}\right)\cdot\nabla\left[-\frac{\sigma_p}{\sigma}\rho'_c - \left(\frac{\sigma_e}{\sigma} + \frac{n_e}{n_p}\frac{\sigma_p}{\sigma}\right)n'_e + \left(1 - \frac{\sigma_n}{\sigma} - \frac{n_n}{n_p}\frac{\sigma_p}{\sigma}\right)n'_n + \frac{\sigma_e}{\sigma}\hat{\nu}_m T'_e + \left(\hat{\sigma}_n - \frac{\sigma_i}{\sigma}\hat{\sigma}_i\right)T'\right]$$
$$= -\frac{\sigma_n}{\sigma}\frac{n_p}{n_n}\frac{D\rho'_c}{Dt} - \frac{1}{\tau_r^{(i)}}\rho'_c + \left(\frac{n_e}{n_n}\frac{1}{\tau_a} - \frac{n_e}{n_p}\frac{1}{\tau_r^{(i)}}\right)n'_e - \left(\frac{n_n}{n_p}\frac{1}{\tau_r^{(i)}} + \frac{n_e}{n_n}\frac{1}{\tau_a}\right)n'_n + \frac{1}{\tau_r^{(i)}}n' + \frac{n_e}{n_n}\frac{1}{\tau_a}\hat{k}_a T'_e - \frac{1}{\tau_r^{(i)}}\left(\hat{k}_r^{(i)} + \frac{\tau_r^{(i)}}{\tau_d}\hat{k}_d\right)T', \tag{30}$$

and for electronically excited species,

$$\frac{Dn'_*}{Dt} + \frac{1}{\tau_s}\nabla\cdot\vec{u}' = \left(\frac{1}{\tau_q} - \frac{S_{*\,\mathrm{ext}}}{n_*}\right)n'_e - \left(\frac{n_e}{n_*}\frac{1}{\tau_i^*} + \frac{1}{\tau_q}\right)n'_* + \frac{n_e}{n_*}\frac{1}{\tau_i^*}n' + \frac{n_e}{n_*}\frac{1}{\tau_*}\left(\hat{k}_* - \frac{\tau_*}{\tau_i^*}\hat{k}_i^*\right)T'_e - \frac{1}{\tau_q}\hat{k}_q T', \tag{31}$$

where the logarithmic derivatives $\hat{k}_j$ of the rate coefficients k_j represent the sensitivity of electron, negative-ion, and electronically-excited-species production and loss to fluctuations in electron and gas temperature. The term $\tau_s^{-1}\nabla\cdot\vec{u}'$ in these equations represents the coupling between the velocity field of the gas and the production and loss of charged particles. Since volumetric dilatation of fluid elements occurs on a time scale τ_s, the velocity field associated with sound waves can couple effectively to charged-particle-production processes only if $\tau_s \leq \tau_i, \tau_a, \ldots,$ etc. In general this will occur only at low pressure and for disturbances with sufficiently small size. The third and fourth terms on the left-hand side of the electron and negative-ion continuity equations (29) and (30), represent the contributions from ambipolar diffusion and particle drift due to plasma gradients, respectively. For electrons the ambipolar diffusion results in a local loss; however, the negative ions tend to accumulate owing to the direction of the ambipolar field established between the fast electrons and slow positive ions. The first term on the right-hand sides of the electron and negative-ion continuity equations represents the contribution from space-charge fluctuations and is only large during the initial stage of evolution of the disturbance. Since $\sigma_n/\sigma \ll 1$, the contribution of negative ions to space-charge fluctuation is small.

The remaining terms on the right-hand sides of these equations are the contributions from fluctuations in particle kinetic processes. In conventional laser discharges the cross sections[10–14] for electronic excitation and ionization have thresholds in the high-energy portion of the electron-energy distribution, and consequently the rate coefficients for electronic excitation (k_*) and ionization (k_i, k_i^*) exhibit exceptionally strong dependences on electron temperature. For example, values of $\hat{k}_*$, $\hat{k}_i^*$, and $\hat{k}_i$ in the range of 10 to 20 are typical, indicating that small changes in electron temperature can produce relatively large fluctuations in the rates of electron, positive-ion, and electronically-excited-species production and loss. The electron-molecule cross sections[24–26] for attachment, on the other hand, may be large for low- as well as high-energy electrons, and consequently the dependence of the attachment rate on electron temperature depends critically on molecular species. Values of $\hat{k}_a$ from slightly negative to large positive values in the range of 10 to 20 are possible.[24] The temperature dependences of the electron-ion[27–29] and ion-ion recombination rates[29] are usually relatively small, with values of $\hat{k}_r^{(e)}$ and $\hat{k}_r^{(i)}$ in the range $-\frac{1}{2}$ to -1. The gas-temperature dependences of the electronic quenching and detachment rate coefficients[30] vary significantly with electronic state and type of negative-ion species present within the discharge.

Equations (29)–(31) indicate that fluctuations in the densities of charged particles and electronically excited species are significantly affected by electron- and gas-temperature fluctuations due to the magnitude and the sign of quantities $\hat{k}_j$. However, under certain conditions the significance of some of these quantities can be greatly reduced by using an independently controllable source of ionization, such as an e-beam system,[7] to maintain the discharge. In this case the volumetric rate of external ionization $S_{e\,\mathrm{ext}}$ may be made large enough to sustain the plasma against recombination losses and the discharge E/n ratio can be lowered thus lowering the electron temperature to the point where $\{\tau_i^{-1}, \tau_*^{-1}, \tau_i^{*\,-1}\} \ll \{S_{e\,\mathrm{ext}}/n_e, S_{*\,\mathrm{ext}}/n_*\}$. Since in the continuity equations (29)–(31) the temporal density fluctuations are proportional to the quantities $\hat{k}_j/\tau_j$, the ability to sustain the plasma with arbitrarily large values of τ_j negates the otherwise dominant influence of the terms $\hat{k}_j$. Thus in the presence of an auxiliary source of particle production the fluctuations in charged-particle and electronically-excited-species densities can be made virtually insensitive to fluctuations in electron temperature. It should be noted that the factors influencing the energy-transfer processes and the production and maintenance of a population inversion in the case of molecular lasers remain essentially the same in the presence of e-beam ionization. The consequences of these conditions as regards discharge stability will be discussed in Sec. IV.

Electron-Energy and Electromagnetic Field Equation

The electron-energy equation for fluctuating quantities is obtained from Eq. (12) and may be written

$$\frac{DT_e'}{Dt} + \frac{Dn_e'}{Dt} + \frac{5}{3}\frac{1}{\tau_s}\nabla\cdot\vec{u}' = \frac{1}{\tau_e}\Omega_e' + \left(\frac{\lambda_e}{\frac{3}{2}n_e\kappa l^2}\right)\nabla^2 T_e' + \frac{5}{3}\left(1-\frac{\sigma_e}{\sigma}\right)\frac{D_e}{l^2}\nabla^2\eta_e' - \frac{5}{3}\frac{n_p}{n_e}\frac{\sigma_e}{\sigma}\frac{D\rho_c'}{Dt}$$
$$+\frac{5}{3}\frac{\sigma_e}{\epsilon_0}\left(\frac{\epsilon_0\vec{E}}{en_e l}\right)\cdot\nabla\left\{-\frac{\sigma_p}{\sigma}\rho_c' + \left(1-\frac{\sigma_e}{\sigma}-\frac{n_e}{n_p}\frac{\sigma_p}{\sigma}\right)n_e' - \left(\frac{\sigma_n}{\sigma}+\frac{n_n}{n_p}\frac{\sigma_p}{\sigma}\right)n_n' + \left[1-\left(1-\frac{\sigma_e}{\sigma}\right)\hat{\nu}_m\right]T_e'\right.$$
$$\left. - \frac{\sigma_i}{\sigma}\hat{\sigma}_i T'\right\} - \frac{1}{\tau_e}n_e' - \frac{1}{\tau_e}n' - \frac{1}{\tau_e}(1+\hat{\nu}_u)T_e', \tag{32}$$

where $\hat{\nu}_u$ denotes the sensitivity of electron energy loss to changes in electron temperature and is typically in the range of 1 to 5 for most common molecular gases. The contribution from fluctuations in Joule heating is

$$\Omega_e' = 2\left(\frac{\vec{E}}{E}\right)\cdot\vec{E}' + n_e' - n' - \hat{\nu}_m T_e' + \tfrac{2}{3}\tau_e\left(\frac{\sigma_e}{\epsilon_0}\right)\frac{\epsilon_0\vec{E}}{en_e l}\cdot\nabla\eta_e'. \tag{33}$$

The time scale $\tau_e \equiv \frac{3}{2}n_e\kappa T_e/(J_e^2/\sigma_e) = \frac{3}{2}\nu_u^{-1}$ represents the characteristic time for heating the electron gas. Typically, $\tau_e \ll \tau_s$, and the third term in Eq. (32) representing the coupling to the velocity field is negligible. The second, third, fourth, and fifth terms on the right-hand side of the electron-energy equation represent the combined effect of electron-energy transfer due, respectively, to thermal transport, ambipolar diffusion, space-charge fluctuations, and electron drift. The last three terms of Eq. (32) constitute the contributions due to collisional energy losses such as rotation, vibration, and electronic excitation. Fluctuations in the electric field are determined by

$$(\epsilon_0 E/en_p l)\nabla\cdot\vec{E}' = \rho_c' \tag{34}$$

and

$$\nabla\times\vec{E}' = 0. \tag{35}$$

D. Plane-Wave Approximation

Up to this point only arbitrary small amplitude fluctuations have been considered. However, in order to proceed further it is necessary to specify the spatial and temporal nature of the disturbance. If Ψ_0 is the source of the disturbance such that $\Psi_0(\vec{x}, t) = 0$ for $t < 0$, then, introducing Fourier–Laplace transforms, the general response of the plasma to this initial fluctuation may be written

$$\Psi(\vec{x}, t) = \int_{-\infty - i\beta}^{\infty - i\beta}\frac{d\omega}{2\pi}\int_{-\infty}^{\infty}\frac{d^3k}{(2\pi)^3}\left(\frac{\Psi_0(\vec{k}, \omega)}{D(\vec{k}, \omega)}e^{i(\omega t - \vec{k}\cdot\vec{x})}\right), \tag{36}$$

where β is chosen so that the contour in the ω plane lies below all singularities of the integrand to guarantee causality. In general, for a localized initial disturbance, $\Psi_0(\vec{k}, \omega)$ is analytic. The dispersion relation for the present plasma model is of the polynomial form, $D(\vec{k}, \omega) = \prod_n[\omega - \omega_n(\vec{k})]$, where ω_n are all distinct. Therefore, the integration in Eq. (36) over ω is readily completed,

$$\Psi(\vec{x}, t) = \sum_n \frac{i}{(2\pi)^3} \int_{-\infty}^{\infty} d^3k \left\{ \left[\Psi_0(\vec{k}, \omega_n(\vec{k})) / \frac{\partial D}{\partial \omega}(\vec{k}, \omega_n(\vec{k})) \right] \times e^{i[\omega_n(\vec{k})t - \vec{k}\cdot\vec{x}]} \right\} = \sum_n \Psi_n(\vec{x}, t). \tag{37}$$

This result indicates that the low-frequency response of the plasma to an arbitrary fluctuation within the plasma may be decomposed[31] into a superposition of normal modes determined by $D(\vec{k}, \omega_n(\vec{k})) = 0$. Furthermore, studies[32,33] of the asymptotic character of relations similar to Eq. (37) indicate that a necessary and sufficient condition for a normal wave mode to be unstable is $\omega_{n_i} = \mathrm{Im}[\omega_n(\vec{k})] < 0$ for some real value of $\vec{k}$. In addition, at long times, the system is dominated by the mode giving the root ω_n ($\vec{k}$ real) with the minimum imaginary part $(\omega_{n_i})_{\min}$. This corresponds to the slowest-decaying mode for a stable plasma $[(\omega_{n_i})_{\min} \geq 0]$, or the fastest-growing mode for an unstable plasma $[(\omega_{n_i})_{\min} < 0]$. Equation (37) also indicates that, although the individual Fourier components may be unstable, the form of the instability during development need not be wavelike.

As a consequence of these results the stability of a large-volume high-energy electric discharge plasma is ultimately tied to the stability of the normal wave modes which may be excited by fluctuations inherent in the discharge. Taking account of the physical processes associated with their evolution, these normal modes are identified in Sec. IV and ordered according to their characteristic time scales as follows: a space-charge relaxation mode (10^{-10}–10^{-8} sec), an electron thermal mode (10^{-8}–10^{-7} sec), an ionization mode (10^{-6}–10^{-5} sec), an electronically-excited-species-production mode (10^{-6}–10^{-4} sec), a sound mode (10^{-5}–10^{-4} sec), a vibrational energy relaxation mode (10^{-4}–10^{-3} sec), a heavy-particle thermal mode (10^{-4}–10^{-3} sec), and a vorticity mode (10^{-3}–10^{-2} sec). In equilibrium (i.e., $T = T_V = T_e$), the heavy-particle thermal, vibrational energy, and electron thermal modes become the thermal or entropy mode considered by Chu and Kovasnay.[34] The splitting is simply a consequence of the nonequilibrium between the electron translational, vibrational, and heavy-gas translation-rotation degrees of freedom within the discharge. The electron thermal, ionization, and electronically-excited-species modes have been considered previously to some extent in studies of striations[35–38] observed in noble-gas discharges. The propagation of sound and development of the thermal mode in weakly ionized noble gases has been considered by Schulz and Ingard[39] and Ecker *et al.*,[40,41] respectively.

IV. NORMAL MODES

A. Characteristic Times

Fourier–Laplace analysis of the self-consistent set of equations (20)–(35) governing the behavior of the fluctuating quantities leads to a tenth-order dispersion relation for the complex frequency ω, analytical solution of which is a practical impossibility. However, as displayed in Table I, the characteristic times associated with the several processes of importance are spread over a very large range (10^{-10}–10^{-2} sec), permitting significant simplifications of the problem. As a result, the general dispersion relation can be partitioned into a set of relatively independent dispersion relations of order three or less, indicative of the fact that for a wide range of physical conditions the various normal modes are effectively decoupled.

As a consequence of the large differences in characteristic times indicated in Table I, the dissipation or growth of a disturbance involves different modes of plasma behavior depending on the type of disturbance and time scale involved. From the space-charge continuity equation, Eq. (28), the relaxation of space-charge fluctuations occurs on a time scale τ_c for typical conditions and involves primarily the electrons. Typically, τ_c is the shortest characteristic time in the plasma. The next-shortest time is τ_e, which measures the electron-energy relaxation in the electric field. Usually, τ_c is 10^{-10}–10^{-9} sec, whereas τ_e is 10^{-9}–10^{-8} sec, and hence space-charge relaxation occurs faster than or on a time scale comparable with adjustments in electron energy. Somewhat longer than the electron-energy relaxation time are a host of characteristic times ($\tau_i, \tau_i^*, \tau_a, \tau_d, \ldots$) which characterize the charged and electronically-excited-species collisional kinetic and transport processes. These times are typically in the range 10^{-8}–10^{-5} sec and characterize the temporal evolution of the particle production modes. With the possible exception of short-wavelength sound waves, the longest characteristic times are those that characterize the response of the neutral-gas properties, i.e., n, T, T_V, and $\vec{u}$. The times τ_T, τ_V, τ_{VT}, and τ_s, which measure translation-rotation heating, vibrational heating, vibrational relaxation, and sound propagation, are usually in the range 10^{-5}–10^{-2} sec.

These considerations suggest that during evolution of the normal wave modes associated with the neutral-gas energy and momentum transfer processes the charged-particle properties respond instantaneously. On the other hand, during evolution of the modes associated with the charged-particle and electronically-excited-species production processes, neutral-gas properties are effectively frozen. In addition, the electron energy and space-charge fluctuations adjust on a quasisteady basis producing nearly quasineutral plasma motions. Finally, the space-charge relaxation and electron thermal modes occur on time scales that are so rapid that charged- and neutral-particle collisional kinetic processes remain essentially frozen. Only electron-energy loss and elastic scattering collisions are of consequence in this case. In the following, the physics of the individual Fourier components of the normal modes is investigated. The instability criterion is taken to be $\mathrm{Im}[\omega_n(\vec{k})] < 0$ for real $\vec{k}$.

B. Space-Charge Relaxation and Electron Thermal Modes

In considering the characteristic wave modes associated with the space-charge and electron thermal relaxation processes, since the time scales for these processes are so short compared to the other time scales governing plasma processes, only fluctuations in electron density and temperature and the electric field vary significantly. The evolution of these quantities is described by the charge-conservation equation, Eq. (28), with $\rho_c' \simeq -(n_e/n_p)n_e'$, the electron-energy equation, Eq. (32), and Maxwell's equations, Eqs. (34) and (35). Upon Fourier analysis of these equations, it can be shown that as a result of the electrostatic nature of the relaxation the kth Fourier component of the electric field $\vec{E}_{\vec{k}} \simeq -i(\vec{k}/k)(en_e/\epsilon_0 kE)n_{e\vec{k}}$ is in the direction of the wave vector. Except for quite large values of k, the quantity $(en_e/\epsilon_0 kE)$ is much greater than unity and the electric field fluctuations are typically large and approximately $3\pi/2$ radians out of phase with electron-density fluctuations. As a consequence of Joule heating by this field, the corresponding electron-temperature fluctuations are also relatively large. For conditions of interest in high-power electrically excited lasers, and again except for quite large values of k, $\sigma_e/\epsilon_0 \gg \tau_e^{-1} \gtrsim (\sigma_e/\epsilon_0)(\epsilon_0\vec{k}\cdot\vec{E}/en_e)$ and the dispersion relation for the coupled space-charge and electron thermal modes may be further simplified.

Space-Charge Mode

Keeping only first-order terms, under these conditions the dispersion relation for the space-charge relaxation mode may be written[42]

$$i\omega_{\vec{k}} \simeq -\sigma_e/\epsilon_0 + i\tfrac{7}{3}(1-\tfrac{8}{7}\hat{\nu}_m)\vec{k}\cdot\vec{U}_e. \tag{38}$$

Consequently, the Fourier components of this mode are damped in a time ϵ_0/σ_e and drift relative to the gas in the general direction of electron flow with phase velocity

$$V_{\vec{k}} \simeq \tfrac{7}{3}(1-\tfrac{8}{7}\hat{\nu}_m)\left(\frac{\vec{k}}{k}\cdot\vec{U}_e\right) . \tag{39}$$

Since $\sigma_e/\epsilon_0 \gg \vec{k}\cdot\vec{U}_e$, in general the damping occurs in less than one oscillation period.

Electron Thermal Mode

To a similar degree of approximation the dispersion relation for the electron thermal mode is given by

$$i\omega_{\vec{k}} \simeq -\left(\frac{1}{\tau_e}\hat{\nu}_u' + \frac{\lambda_e k^2}{\frac{3}{2}n_e\kappa}\right) + i(1+\tfrac{2}{3}\hat{\nu}_m)\vec{k}\cdot\vec{U}_e , \tag{40}$$

where $\hat{\nu}_u' = (1+\hat{\nu}_u) - \hat{\nu}_m\cos 2\phi$. From this result, the electron thermal mode is unstable if

$$\tau_e\left(\frac{\lambda_e k^2}{\frac{3}{2}n_e\kappa}\right) < -\hat{\nu}_u' = -(1+\hat{\nu}_u - \hat{\nu}_m\cos 2\phi) . \tag{41}$$

Thus the electron thermal mode is unstable if the quantity $\hat{\nu}_u'$ is negative, i.e., if $\hat{\nu}_u'$ decreases with increasing electron temperature. For most situations of interest[10–14] $|\hat{\nu}_m| \lesssim 1$, and the criterion $\hat{\nu}_u' < 0$ reduces in practice to the condition $\hat{\nu}_u < 0$ regardless of the angle ϕ. If $\hat{\nu}_m > 0$, the condition for instability is most easily satisfied for those Fourier components associated with motion along the direction of electron flow. For most common[10–14] molecular species and their mixtures the electron-energy-exchange collision frequency ν_u is an increasing function of electron temperature, indicating that for most practical circumstances the electron thermal mode is stable.

From Eq. (40) the phase velocity of the electron thermal mode is

$$V_{\vec{k}} \simeq (1+\tfrac{2}{3}\hat{\nu}_m)\left(\frac{\vec{k}}{k}\cdot\vec{U}_e\right) , \tag{42}$$

and consequently this mode is transported relative to the gas by drifting electrons. For discharges in molecular gases τ_e^{-1} is generally comparable with or larger than $\vec{k}\cdot\vec{U}_e$, and the waves move only a short distance during damping or growth. Also τ_e^{-1} is small compared with σ_e/ϵ_0, and the associated space-charge fluctuations are relatively small. In addition, since τ_e is typically much less than the characteristic times of other wave modes with the exception of stable space-charge relaxation, when the electron thermal wave mode is unstable it dominates plasma stability.

It should be noted that this treatment of space-charge and electron-energy relaxation using velocity moments of the electron distribution function and neglecting electron inertia effects is only approximate. In many cases of interest the electron-momentum-transfer collision frequency is comparable to the electron plasma frequency, and hence ϵ_0/σ_e is approximately ω_{pe}, for which it follows that electron-inertia effects are important during space-charge relaxation. In addition, the characteristic time scales of both of these modes are comparable to the characteristic time for evolution of the electron distribution and a more accurate treatment of them requires consideration of the spatially and temporally dependent electron Boltzmann equation. A formulation of this problem for discharges in noble gases, including electron-distribution-function effects, has been given by Gentle.[38] For discharges in noble gases the characteristic time for electron-energy transfer, τ_e, is comparable to the characteristic times for charged and electronically excited species production, and the simplifying assumptions employed in the present treatment of molecular-gas discharges are no longer valid.

C. Quasineutral Modes

On the basis of the previous results, the treatment of the remaining normal wave modes of the plasma may be significantly simplified. In particular, space-charge effects dissipate on a time scale that is very much faster than other time scales of the plasma. Consequently, for wave processes other than space-charge relaxation and electron thermal modes, the time derivative of the charge-density fluctuation is small compared to other terms in the charge-density [Eq. (28)], electron [Eq. (29)], and negative-ion [Eq. (30)] continuity equations, and the electron-energy equation [Eq. (32)]. Thus when

$$\epsilon_0 E/en_p\, l \ll 1\,, \tag{43}$$

the space-charge-density fluctuations rapidly relax to a relatively small value,

$$\rho_c' \simeq -\left(\frac{\epsilon_0 \vec{\mathrm{E}}}{en_p\, l}\right)\cdot \nabla\left[\left(\frac{\sigma_e}{\sigma}+\frac{n_e}{n_p}\frac{\sigma_p}{\sigma}\right)n_e' + \left(\frac{\sigma_n}{\sigma}+\frac{n_n}{n_p}\frac{\sigma_p}{\sigma}\right)n_n' - n' - \frac{\sigma_e}{\sigma}\hat{\nu}_m T_e' + \frac{\sigma_i}{\sigma}\hat{\sigma}_i T'\right] - \frac{\epsilon_0}{\sigma}\frac{n_e}{n_p}\frac{D_e}{l^2}\nabla^2\eta_e'\,, \tag{44}$$

and the plasma becomes quasineutral. The corresponding irrotational electric field fluctuation is determined by substituting Eq. (44) into Poisson's equation, Eq. (34). The effect of this quasineutral-plasma condition is to reduce the effectiveness of particle transport processes; i.e., particle transport is inhibited by the space-charge electric field that maintains quasineutrality. Although the transport terms in Eqs. (29)–(33) are of importance in many cases of practical importance, their complexity prevents a simple treatment. However, transport phenomena generally produce a stabilizing effect, and their omission for the sake of clarity is not of qualitative significance. In addition, for characteristic disturbance dimensions sufficiently large ($l \gtrsim 1$–10 cm), transport processes are of relatively little significance.

For most discharges of interest, the mass of the ions is very much greater than the mass of the electrons, and the electron temperature is significantly elevated above the gas temperature. In this limit, σ_p/σ and σ_n/σ are vanishingly small, and ion transport effects disappear. In fact, the presence of massive ions is destabilizing in the sense that the transport terms become ineffective for much smaller values of l. In the following calculations, this heavy-ion or no-ion-slip approximation is adopted and ion transport effects are not treated in any further detail, i.e., $\sigma_n/\sigma = \sigma_p/\sigma = 0$.

In addition to the condition of quasineutrality, for normal modes other than space-charge relaxation and electron thermal, the rate of change of electron energy with time in the electron-energy equation, Eq. (32), is small compared to the Joule heating, transport, and collisional energy-loss terms and may therefore be neglected in the analysis of all modes which develop over a longer time scale. This quasisteady electron-energy equation and the electric field equations can be used to determine the relations among the Fourier components of the electron-temperature, electron-density, and gas-density fluctuations. These relationships are defined by the equation

$$T_{e\vec{k}} = \frac{d\ln T_e}{d\ln n_e} n_{e\vec{k}} + \frac{d\ln T_e}{d\ln n} n_{\vec{k}}\,, \tag{45}$$

where if $\hat{\nu}_u'' = \hat{\nu}_u' + \tau_e[\lambda_e k^2/(\frac{3}{2}n_e\kappa)]$ then

$$\frac{d\ln T_e}{d\ln n_e} = -\frac{2\cos^2\phi + i\frac{2}{3}(\tau_e \vec{k}\cdot\vec{\mathrm{U}}_e)}{\hat{\nu}_u'' - i(\tau_e\vec{k}\cdot\vec{\mathrm{U}}_e)(1+\frac{2}{3}\hat{\nu}_m)} \tag{46}$$

and

$$\frac{d\ln T_e}{d\ln n} = \frac{-2\sin^2\phi}{\hat{\nu}_u'' - i(\tau_e\vec{k}\cdot\vec{\mathrm{U}}_e)(1+\frac{2}{3}\hat{\nu}_m)}\,, \tag{47}$$

in which ϕ is the angle between the wave vector $\vec{k}$ and the direction of the applied electric field. For wavelengths greater than a millimeter, τ_e/τ_{Te} and $(\tau_e\vec{k}\cdot\vec{\mathrm{U}}_e)^2 \sim O(\tau_e/\tau_{Te})$ are small compared to unity for the plasma conditions under consideration and therefore $d\ln T_e/d\ln n_e$ and $d\ln T_e/d\ln n$ may be written

$$\frac{d\ln T_e}{d\ln n_e} \simeq \frac{-2\cos^2\phi}{\hat{\nu}_u'} - i\,\frac{(\tau_e\vec{k}\cdot\vec{\mathrm{U}}_e)}{\hat{\nu}_u'}\left[\frac{2}{3} + \frac{2\cos^2\phi}{\hat{\nu}_u'}(1+\tfrac{2}{3}\hat{\nu}_m)\right] \tag{48}$$

and

$$\frac{d\ln T_e}{d\ln n} \simeq \frac{-2\sin^2\phi}{\hat{\nu}_u'} - i\,\frac{(\tau_e\vec{k}\cdot\vec{\mathrm{U}}_e)}{\hat{\nu}_u'} 2\sin^2\phi(1+\tfrac{2}{3}\hat{\nu}_m)\,. \tag{49}$$

The quantities $d\ln T_e/d\ln n_e$ and $d\ln T/d\ln n$ are a measure of the magnitude and phase of the electron-temperature fluctuations due to electron-density and gas-density fluctuations, respectively. Since the electron thermal mode is unstable and dominates plasma behavior only if $\hat{\nu}_u' < 0$, a situation not often realized in practice, in the following analysis of the charged-particle-production and neutral-gas modes it is assumed that $\hat{\nu}_u' > 0$. Consequently from Eqs. (48) and (49), the electron-temperature fluctuations increase when the electron- and gas-density fluctuations are decreasing. In the following, conditions such that τ_e/τ_{Te} is small compared to unity are considered in detail.

D. Ionization, Negative-Ion, and Electronically-Excited-Species Modes

During the characteristic motions of the charged particles and electronically excited species the neutral degrees of freedom remain nearly frozen and the equations describing these motions in the heavy-ion approximation are obtained from the previous results by neglecting terms involving fluctuations in gas density, temperature, and velocity in Eqs. (29)–(33). Under these conditions the dispersion relation for the charged-particle-production modes is a complicated cubic polynomial. To illustrate some of the important physics associated with the ionization, negative-ion-production, and electronically-excited-species-production modes characterized by the roots of this polynomial, three relatively simple cases are considered.

Ionization Mode

In the absence of negative ions and electronically excited species, only positive ions and electrons constitute the plasma, and the dispersion relation for the resulting ionization mode associated with them is given by

$$i\omega_{\vec{k}} \simeq -\frac{2}{\tau_i}\left(\frac{\hat{k}_i}{\hat{\nu}_u'}\right)\cos^2\phi - \left(\frac{1}{\tau_r^{(e)}} + \frac{S_{e\,\mathrm{ext}}}{n_e}\right) - i\left[\tfrac{2}{3} + \frac{2\cos^2\phi}{\hat{\nu}_u'}(1+\tfrac{2}{3}\hat{\nu}_m)\right]\frac{\tau_e}{\tau_i}\left(\frac{\hat{k}_i}{\hat{\nu}_u'}\right)(\vec{k}\cdot\vec{U}_e) . \qquad (50)$$

For most molecular gases $|\hat{k}_r^{(e)}|$ is less than $\hat{k}_i$ and $|\hat{\nu}_u'|$ and therefore the contribution due to the electron-temperature dependence of the two-body recombination coefficient is neglected here and in the remainder of this paper. Examination of the right-hand side of Eq. (50) shows that the ionization mode is always stable and decays on a time scale comparable to the ionization and electron–positive-ion recombination times. The contribution to the electron-production rate due to electron-temperature fluctuations is out of phase with the electron-density fluctuations and represents an effective loss in addition to the recombination loss associaated with electron-density fluctuations. From Eq. (50), the phase velocity of the kth component is given by

$$V_{\vec{k}} = -\left[\tfrac{2}{3} + \frac{2\cos^2\phi}{\hat{\nu}_u'}(1+\tfrac{2}{3}\hat{\nu}_m)\right]\left(\frac{\tau_e}{\tau_i}\right)\left(\frac{\hat{k}_i}{\hat{\nu}_u'}\right)\left(\frac{\vec{k}}{k}\cdot\vec{U}_e\right) , \qquad (51)$$

and the individual components move relative to the gas in the direction of current flow at a velocity which is small compared to the electron drift velocity.

When an external source is used to maintain the discharge and E/n is lowered, thereby substantially increasing the ionization time, the effect of electron-temperature fluctuations on the particle-production process and the phase velocity of the waves may be significantly reduced. In this limit the dispersion relation becomes $i\omega_{\vec{k}} \sim -2/\tau_r^{(e)}$, damping by recombination dominates, and the phase velocity is very small.

Summarizing previous results, the ionization mode is stable in the absence of negative ions and electronically excited species for conditions of interest. Transport processes, within the heavy-ion limit, have a negligible effect on wave damping except for very small wavelengths.

Coupled Ionization and Negative-Ion Modes

If collisional attachment is important and substantial numbers of negative ions are present relative to the electron density, essential features of the ionization mode are modified and an additional wave mode associated with the negative ions appears. The first-order dispersion relation governing the evolution of these modes is obtained from the combined negative-ion [Eq. (30)] and electron [Eq. (29)] continuity equations, the quasisteady-electron-energy equation, and the electric field equations [Eqs. (34) and (35)] using the quasineutral approximation [Eq. (44)]. This dispersion relation is quadratic with solutions given by

$$i\omega_{\vec{k}} = \tfrac{1}{2}[-b \pm (b^2-4c)^{1/2}] , \qquad (52)$$

where

$$b = \left(\frac{n_e}{n_p}\frac{1}{\tau_r^{(e)}} + \frac{n_n}{n_e}\frac{1}{\tau_d} + \frac{n_n}{n_p}\frac{1}{\tau_r^{(i)}} + \frac{n_e}{n_n}\frac{1}{\tau_a} + \frac{S_{e\,\mathrm{ext}}}{n_e}\right) - \frac{\hat{k}_i}{\tau_i}\frac{d\ln T_e}{d\ln n_e}\left(1 - \frac{\tau_i}{\tau_a}\hat{k}_a(\hat{k}_i)^{-1}\right) \qquad (53)$$

and

$$c = \left[\frac{n_e}{n_n}\frac{1}{\tau_a}\frac{1}{\tau_r^{(e)}} + \frac{n_n}{n_e}\frac{1}{\tau_r^{(i)}}\frac{1}{\tau_d} + \left(\frac{n_n}{n_p}\frac{1}{\tau_r^{(i)}} + \frac{n_e}{n_n}\frac{1}{\tau_a}\right)\frac{S_{e\,\mathrm{ext}}}{n_e}\right] - \frac{\hat{k}_i}{\tau_i}\frac{d\ln T_e}{d\ln n_e}\left\{\left(\frac{n_n}{n_p}\frac{1}{\tau_r^{(i)}} + \frac{n_e}{n_n}\frac{1}{\tau_a}\right) - \left[\left(1+\frac{n_n}{n_p}\right)\frac{1}{\tau_r^{(i)}} + \frac{n_e}{n_p}\frac{1}{\tau_r^{(e)}}\right]\frac{\tau_i}{\tau_a}\hat{k}_a(\hat{k}_i)^{-1}\right\} . \qquad (54)$$

The complexity of these results prohibits a simple general treatment such as was given in the previous subsection for the ionization mode alone. However, for very long wavelengths propagation and transport effects are small; b and c are real. For these situations consideration of the physical processes contributing to the coefficients b and c indicates that when $b>0$ and $c<0$ the negative-ion mode is unstable. When $b<0$ the ionization mode is unstable and, if $c>0$, the negative-ion mode is unstable as well. To examine the dispersion relations for the ionization and negative-ion modes in more detail, suppose that detachment dominates negative-ion collisional loss. Then the effect of negative-ion formation on the steady-state electron kinetics vanishes, i.e., $n_e/n_n=\tau_a/\tau_d$ and $\tau_i^{-1}+\tau_r^{(e)\,-1}-S_{e\,\mathrm{ext}}/n_e=0$. In addition, under many cases of practical importance, $\hat{k}_i \gg \hat{\nu}'_u$, and the ionization and negative-ion dispersion relations are decoupled.

Ionization Mode

When detachment dominates negative-ion loss and $\hat{k}_i \gg \hat{\nu}'_u$, the dispersion relation for the ionization mode, from Eqs. (52)–(54), may be expressed

$$i\omega_{\vec{k}}^{*}=-\frac{2}{\tau_i}\left(1-\frac{\tau_i}{\tau_a}\hat{k}_a(\hat{k}_i)^{-1}\right)\left(\frac{\hat{k}_i}{\hat{\nu}'_u}\right)\cos^2\phi-\left(\frac{n_p}{n_n}\frac{1}{\tau_a}+\frac{n_e}{n_p}\frac{1}{\tau_r^{(e)}}+\frac{S_{e\,\mathrm{ext}}}{n_e}\right)-i\left[\tfrac{2}{3}+\frac{2\cos^2\phi}{\hat{\nu}'_u}(1+\tfrac{2}{3}\hat{\nu}_m)\right]$$
$$\times\left(1-\frac{\tau_i}{\tau_a}\hat{k}_a(\hat{k}_i)^{-1}\right)\left(\frac{\tau_e}{\tau_i}\right)\left(\frac{\hat{k}_i}{\hat{\nu}'_u}\right)(\vec{k}\cdot\vec{U}_e)\,. \tag{55}$$

From Eq. (55) the ionization mode will be unstable if

$$\frac{2}{\tau_i}\left(1-\frac{\tau_i}{\tau_a}\hat{k}_a(\hat{k}_i)^{-1}\right)\hat{k}_i+\left(\frac{n_p}{n_n}\frac{1}{\tau_a}+\frac{n_e}{n_p}\frac{1}{\tau_r^{(e)}}+\frac{S_{e\,\mathrm{ext}}}{n_e}\right)\hat{\nu}'_u<0\,, \tag{56}$$

the characteristic growth time is faster than the ionization time, and the phase velocity of the kth component is given by

$$V_{\vec{k}}^{*}=-\left[\tfrac{2}{3}+\frac{2\cos^2\phi}{\hat{\nu}'_u}(1+\tfrac{2}{3}\hat{\nu}_m)\right]\times\left(1-\frac{\tau_i}{\tau_a}\hat{k}_a(\hat{k}_i)^{-1}\right)\left(\frac{\tau_e}{\tau_i}\right)\left(\frac{\hat{k}_i}{\hat{\nu}'_u}\right)\left(\frac{\vec{k}}{k}\cdot\vec{U}_e\right)\,. \tag{57}$$

The instability criterion, Eq. (56), indicates that in the absence of an external source the ionization instability does not depend directly on the discharge power density and gas pressure because of the relative importance of kinetic processes involving the electron-temperature dependence of the ionization and attachment rate coefficients of the electrons. A necessary, but not sufficient, condition for instability is $[1-(\tau_i/\tau_a)\hat{k}_a(\hat{k}_i)^{-1}]<0$, which requires that the electron attachment rate increase with electron temperature. In addition since $\hat{k}_i$ is typically large, 10 to 20, the condition for instability will be satisfied only if $\tau_i \gg \tau_a$ or if $\hat{k}_a \gtrsim \hat{k}_i$ when the ionization and attachment times are comparable in magnitude. As the negative-ion density increases, the second term in Eq. (56) tends to decrease and the recombination processes are reduced in importance compared with detachment. The instability occurs because the electron-density and temperature fluctuations are out of phase, as displayed by Eq. (48). Under these conditions, instability appears when the loss of electrons due to recombination, attachment, etc., is less than the effective electron-production rate due to electron-temperature fluctuations. Negative-ion-density fluctuations are relatively small. From Eq. (57) when the ionization mode is stable the phase velocity is in the direction of current flow; however, as the instability boundary is approached the phase velocity reverses direction and unstable Fourier components drift in the direction of electron flow. When an external source is employed to maintain the discharge the contribution due to electron-temperature fluctuations, the first term in Eq. (56), is unimportant and the ionization mode is damped by recombination processes.

Negative-Ion Mode

When detachment is the dominant negative-ion collisional loss and $\hat{k}_i \gg \hat{\nu}'_u$ the dispersion relation for the negative-ion mode, from Eqs. (52)–(54), is

$$i\omega_{\vec{k}}^{*}=-\frac{n_e}{n_n}\frac{1}{\tau_a}\frac{2[1-(n_n/n_p)(\tau_i/\tau_r^{(e)})\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i\cos^2\phi+[(\tau_i/\tau_r^{(e)})+\tau_i S_{e\,\mathrm{ext}}/n_e]\hat{\nu}'_u}{2[1-(\tau_i/\tau_a)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i\cos^2\phi+[(n_p/n_n)(\tau_i/\tau_a)+(n_e/n_p)(\tau_i/\tau_r^{(e)})+\tau_i S_{e\,\mathrm{ext}}/n_e]\hat{\nu}'_u}$$
$$-i\frac{n_e}{n_n}\frac{1}{\tau_a}\frac{[\tfrac{2}{3}+2\cos^2\phi(1+\tfrac{2}{3}\hat{\nu}_m)/\hat{\nu}'_u][1-(n_n/n_p)(\tau_i/\tau_r^{(e)})\hat{k}_a(\hat{k}_i)^{-1}](\tau_e/\tau_i)(\hat{k}_i/\hat{\nu}'_u)}{2[1-(\tau_i/\tau_a)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i\cos^2\phi+[(n_p/n_n)(\tau_i/\tau_a)+(n_e/n_p)(\tau_i/\tau_r^{(e)})+\tau_i S_{e\,\mathrm{ext}}/n_e]\hat{\nu}'_u}(\vec{k}\cdot\vec{U}_e)\,. \tag{58}$$

From Eq. (58) the negative mode will be unstable if

$$\frac{2}{\tau_i}\left(1-\frac{\tau_i}{\tau_a}\hat{k}_a(\hat{k}_i)^{-1}\right)\hat{k}_i\cos^2\phi + \left(\frac{n_p}{n_n}\frac{1}{\tau_a}+\frac{n_e}{n_p}\frac{1}{\tau_r^{(e)}}+\frac{S_{e\,\mathrm{ext}}}{n_e}\right)\hat{\nu}_u' > 0 \tag{59}$$

and

$$\frac{2}{\tau_i}\left(1-\frac{n_n}{n_p}\frac{\tau_i}{\tau_r^{(e)}}\hat{k}_a(\hat{k}_i)^{-1}\right)\hat{k}_i\cos^2\phi + \left(\frac{1}{\tau_r^{(e)}}+\frac{S_{e\,\mathrm{ext}}}{n_e}\right)\hat{\nu}_u' < 0\,, \tag{60}$$

or vice versa. When the latter instability condition holds, i.e., the inequality signs are reversed in Eqs. (59) and (60), the ionization mode is also unstable, has the largest growth rate, and will evolve sufficiently rapidly to dominate plasma temporal behavior. Therefore, this case is not discussed further. The phase velocity of the kth component of the negative-ion mode is much less than the phase velocity of the ionization mode and is given by

$$V_{\vec{k}} = -\frac{n_e}{n_n}\frac{1}{\tau_a}\frac{[\frac{2}{3}+2\cos^2\phi(1+\frac{2}{3}\hat{\nu}_m)/\hat{\nu}_u'][1-(n_n/n_p)(\tau_i/\tau_r^{(e)})\hat{k}_a(\hat{k}_i)^{-1}](\tau_e/\tau_i)(\hat{k}_i/\hat{\nu}_u')}{2[1-(\tau_i/\tau_a)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i\cos^2\phi+[(n_p/n_n)(\tau_i/\tau_a)+(n_e/n_p)(\tau_i/\tau_r^{(e)})+\tau_i S_{e\,\mathrm{ext}}/n_e]\hat{\nu}_u'}\left(\frac{\vec{\mathrm{k}}}{k}\cdot\vec{\mathrm{U}}_e\right)\,. \tag{61}$$

From Eqs. (59) and (60) the negative-ion mode may be unstable because of the relative importance of the electron-temperature dependences of the ionization and attachment rate coefficients. Under many cases of practical importance $\hat{k}_i$ is sufficiently greater than $\hat{\nu}_u'$ that the negative-ion mode is convected with the gas flow, and the stability criteria, in the absence of an external source, become simply

$$[1-(\tau_i/\tau_a)\hat{k}_a(\hat{k}_i)^{-1}]>0$$

and

$$[1-(n_n/n_p)(\tau_i/\tau_r^{(e)})\hat{k}_a(\hat{k}_i)^{-1}]<0\,,$$

which may be combined to yield the condition

$$\frac{n_p}{n_n}\frac{\tau_r^{(e)}}{\tau_a}<\frac{\tau_i}{\tau_a}\hat{k}_a(\hat{k}_i)^{-1}<1\,. \tag{62}$$

Therefore when $(\tau_i/\tau_a)\hat{k}_a(\hat{k}_i)^{-1}<1$ and the negative-ion density exceeds $n_p(\tau_r^{(e)}/\tau_i)\hat{k}_i(\hat{k}_a)^{-1}$, the negative-ion mode will be unstable. The physical reason for this result may best be understood by noting that, under these conditions, the negative-ion mode evolves sufficiently slowly that the electron continuity equation is quasisteady and thus the Fourier components of the electron-temperature, electron-density, and negative-ion-density fluctuations are related by

$$T_{e_{\vec{k}}} \sim -\frac{2\cos^2\phi}{\hat{\nu}_u'}n_{e_{\vec{k}}} \sim \left(\frac{\tau_i}{\tau_a}\right)\frac{(n_n/n_p)(\tau_a/\tau_r^{(e)})-1}{[1-(\tau_i/\tau_a)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i}n_{n_{\vec{k}}}\,. \tag{63}$$

Consequently the electron-temperature fluctuations and negative-ion-density fluctuations are in phase and the electron-temperature fluctuations increase relative to the negative-ion-density fluctuations with increasing negative-ion density when $(n_p/n_n)\times(\tau_r^{(e)}/\tau_a)<1$ provided $[1-(\tau_i/\tau_a)\hat{k}_a(\hat{k}_i)^{-1}]>0$. According to Eq. (30), these electron-temperature fluctuations may produce large fluctuations in negative-ion density if $\hat{k}_a$ is large. Further, when Eq. (62) holds, these fluctuations are sufficient to overcome detachment losses and the negative-ion-density fluctuations may increase thereby further increasing the electron temperature, etc. Thus, the stability of the negative-ion mode is intimately related to the negative-ion density and the relative importance of ionization and attachment kinetics with no direct dependence on discharge power density or gas pressure.

As in the case with the ionization mode, an external source may be employed to significantly reduce the effect of electron-temperature fluctuations on electron-density and negative-ion-density fluctuations and, from Eqs. (59) and (60), thereby stabilize the negative-ion mode. When $\hat{k}_i$ is not sufficiently greater than $\hat{\nu}_u'$ to justify the previous approximations, the ionization and negative-ion modes may couple together; however, the essential physical features just described remain important. In either case, application of an external source will eliminate these instabilities.

In summary, the presence of negative ions within the discharge significantly degrades plasma stability for long-wavelength disturbances under conditions of interest to high-power lasers. Under these circumstances, the growth times for the ionization and negative-ion modes are comparable to the characteristic times for charged-particle production and loss by collision processes and therefore short compared to characteristic times of the neutral modes. Consequently, when they occur, the negative-ion and ionization instabilities dominate plasma behavior. However, maintenance of the plasma ionization processes by application of an external source and subsequent lowering of E/n will stabilize these modes.

Coupled Ionization and Electronically-Excited-Species Modes

If collisional attachment is unimportant, within the context of the present plasma model the density of negative ions and consequently their effect on discharge stability will be negligible. Under these conditions, in addition to the ionization mode a wave mode associated with the metastable electronic species appears. Using the quasineutral-charge-density fluctuation [Eq. (44)], the first-order quadratic dispersion relation governing evolution of these modes is obtained from the combined electronically-excited-species [Eq. (31)] and electron [Eq. (29)] continuity equations, the quasisteady-electron-energy equation, and the electric field equations [Eqs. (34) and (35)]. Examination of the roots of this dispersion relation shows that the conditions for instability are generally not satisfied for conditions of interest in molecular-gas-laser discharges. Therefore, in the absence of negative ions, independent metastable electronic species present in molecular-gas discharges exert no destabilizing influence on particle-production wave modes. It should be noted, however, that this result does not imply that cumulative or multistep ionization processes involving several electronically excited species cannot lead to particle-production instabilities. Indeed, the importance of the electron-temperature dependence of the ionization-rate coefficient in the stability criteria for the ionization and negative-ion modes suggests that, in the presence of negative ions, electronically excited species may be important as regards discharge stability.[24]

E. Vorticity, Sound, Thermal, and Vibrational Relaxation Modes

With the possible exception of the sound mode at low pressure, the evolution of the neutral-gas translation, rotation, and vibrational degrees of freedom occurs on time scales that are long compared to the characteristic times associated with charged-particle and electronically-excited-species dynamics. Consequently, charged-particle properties adjust in a quasisteady fashion to changes in gas properties, i.e., the time derivatives in the charged-particle [Eqs. (29) and (30)] and electronically-excited-species [Eq. (31)] continuity equations, and the electron-energy equation [Eq. (32)] are vanishingly small compared to the other terms in these equations. In addition, the contribution from volumetric dilatation of the fluid due to fluctuations in the velocity field is also small. Under these conditions, fluctuations in the properties of the neutral gas couple to the charged-particle kinetics and energy transfer primarily through changes in gas temperature and density.

Using the quasisteady electron, negative-ion, and electronically-excited-species continuity equations, the quasisteady-electron-energy equation, and the quasineutrality condition [Eq. (44)], the Fourier components of the electron density, gas density, and gas temperature can be expressed as

$$n_{e\vec{k}} = \frac{d\ln n_e}{d\ln n}\, n_{\vec{k}} + \frac{d\ln n_e}{d\ln T}\, T_{\vec{k}}\ . \tag{64}$$

The quantities $d\ln n_e/d\ln n$ and $d\ln n_e/d\ln T$ are a measure of the magnitude and phase of electron-density fluctuations due to fluctuations in gas density and temperature, respectively. Examining the effects of negative ions and electronically excited species separately, it can be shown that

$$\frac{d\ln n_e}{d\ln n} = \frac{-2\zeta_1 \sin^2\phi \hat{k}_i + \zeta_2 \hat{\nu}_u'}{2\zeta_1 \cos^2\phi \hat{k}_i + \zeta_3 \hat{\nu}_u'} \tag{65}$$

and

$$\frac{d\ln n_e}{d\ln T} = \frac{\zeta_4 \hat{\nu}_u'}{2\zeta_1 \cos^2\phi \hat{k}_i + \zeta_3 \hat{\nu}_u'}\ , \tag{66}$$

where if negative ions are present

$$\zeta_1 = \frac{1}{\tau_i}\left(\frac{n_n}{n_p}\frac{1}{\tau_r^{(i)}} + \frac{n_e}{n_n}\frac{1}{\tau_a}\right) - \left[\left(1+\frac{n_n}{n_p}\right)\frac{1}{\tau_r^{(i)}} + \frac{n_e}{n_p}\frac{1}{\tau_r^{(e)}}\right]\frac{1}{\tau_a}\hat{k}_a(\hat{k}_i)^{-1}\ , \tag{67}$$

$$\zeta_2 = \frac{n_e}{n_n}\frac{1}{\tau_a}\frac{1}{\tau_r^{(e)}} + \frac{n_n}{n_e}\frac{1}{\tau_r^{(i)}}\frac{1}{\tau_d}\ , \tag{68}$$

$$\zeta_3 = \left(1+\frac{n_e}{n_n}\right)\frac{n_e}{n_p}\frac{1}{\tau_a}\frac{1}{\tau_r^{(e)}} + \left(1+\frac{n_n}{n_e}\right)\frac{n_n}{n_p}\frac{1}{\tau_r^{(i)}}\frac{1}{\tau_d} + \left(\frac{n_n}{n_p}\frac{1}{\tau_r^{(i)}} + \frac{n_e}{n_n}\frac{1}{\tau_a}\right)\frac{S_{e\,\mathrm{ext}}}{n_e}, \tag{69}$$

$$\zeta_4 = \frac{n_n}{n_e}\frac{1}{\tau_d}\left\{\left[\left(1+\frac{n_n}{n_p}\right)\frac{1}{\tau_r^{(i)}} + \frac{n_e}{n_p}\frac{1}{\tau_r^{(e)}}\right]\hat{k}_d - \left(\frac{1}{\tau_d} - \frac{n_e}{n_p}\frac{1}{\tau_r^{(e)}}\right)\frac{\tau_d}{\tau_r^{(i)}}\hat{k}_r^{(i)}\right\}\ , \tag{70}$$

and if independent electronically excited species[18] dominate charged-particle kinetics,

$$\zeta_1 = \left(\frac{n_e}{n_*}\frac{1}{\tau_i^*} + \frac{1}{\tau_q}\right)\frac{1}{\tau_i} + \frac{1}{\tau_i^*}\left(\frac{n_e}{n_*}\frac{1}{\tau_*}\hat{k}_* + \frac{1}{\tau_q}\hat{k}_i^*\right)(\hat{k}_i)^{-1}, \tag{71}$$

$$\zeta_2 = \frac{n_e}{n_*}\frac{1}{\tau_i^*}\frac{1}{\tau_r^{(e)}} + \frac{1}{\tau_q}\left(\frac{1}{\tau_i} + \frac{S_{e\,\mathrm{ext}}}{n_e}\right), \tag{72}$$

$$\zeta_3 = \frac{1}{\tau_i^*}\frac{S_{*\,\mathrm{ext}}}{n_*} + \left(\frac{n_e}{n_*}\frac{1}{\tau_i^*} + \frac{2}{\tau_q}\right)\frac{S_{e\,\mathrm{ext}}}{n_e} + \frac{n_e}{n_*}\frac{1}{\tau_i^*}\frac{1}{\tau_r^{(e)}} + \frac{1}{\tau_q}\frac{1}{\tau_i}\ , \tag{73}$$

$$\zeta_4 = -\frac{1}{\tau_q}\frac{1}{\tau_i^*}\hat{k}_q\ , \tag{74}$$

provided the wave number is such that electron thermal conduction and transport effects are unimportant, i.e., the imaginary parts of Eqs. (48)

and (49) are neglected.

Before proceeding with further analysis of the neutral-gas modes using these results, their consequences are addressed. If negative ions and electronically excited species are not present within the discharge, then $d\ln n_e/d\ln T = 0$ and

$$\frac{d\ln n_e}{d\ln n} = \frac{-2\hat{k}_i \sin^2\phi + (\tau_i/\tau_r^{(e)})\hat{\nu}_u'}{2\hat{k}_i \cos^2\phi + (\tau_i/\tau_r^{(e)} + \tau_i S_{e\,\mathrm{ext}}/n_e)\hat{\nu}_u'}, \tag{75}$$

i.e., the electron density responds only to gas-density changes. For a conventional discharge $\hat{k}_i$ is generally much greater than $\hat{\nu}_u'$. Then, from Eq. (75), except for propagation in the direction of current flow where the response is weak, the electron density in a fluctuation increases with decreasing gas density. The maximum response occurs for propagation nearly normal to the direction of current flow where $d\ln n_e/d\ln n \sim -2\hat{k}_i/\hat{\nu}_u'$. The magnitude of this response is due to the rapid increase of the ionization rate with electron temperature and the sensitivity of electron temperature to gas-density fluctuations expressed by Eqs. (47) and (49).

When an external source is employed, the significance of the rapid increase of the ionization rate with electron temperature can be neutralized and, from Eq. (75), $d\ln n_e/d\ln n$ approaches one-half. Consequently, the application of an external source significantly reduces the sensitivity and changes the sign of the dependence of electron-density fluctuations on gas-density fluctuations.

To examine the effect of negative ions, suppose that detachment dominates the negative-ion collisional loss. Under these conditions, Eqs. (65) and (66) become

$$\frac{d\ln n_e}{d\ln n} = \frac{-2[1-(n_n/n_p)(\tau_i/\tau_r^{(e)})\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i \sin^2\phi + (\tau_i/\tau_r^{(e)})\hat{\nu}_u'}{2[1-(n_n/n_p)(\tau_i/\tau_r^{(e)})\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i \cos^2\phi + [(\tau_i/\tau_r^{(e)}) + \tau_i S_{e\,\mathrm{ext}}/n_e]\nu_u'} \tag{76}$$

and

$$\frac{d\ln n_e}{d\ln T} = \frac{(n_n/n_p)(\tau_i/\tau_r^{(e)})\hat{k}_d \hat{\nu}_u'}{2[1-(n_n/n_p)(\tau_i/\tau_r^{(e)})\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i \cos^2\phi + [(\tau_i/\tau_r^{(e)}) + \tau_i S_{e\,\mathrm{ext}}/n_e]\nu_u'}\,. \tag{77}$$

For a conventional discharge $\hat{k}_i$ is generally much greater than $\hat{\nu}_u'$ and therefore, except for values of ϕ near zero or integral multiples of $\frac{1}{2}\pi$, $d\ln n_e/d\ln n \simeq -\tan^2\phi$ and $d\ln n_e/d\ln T \simeq (n_n/n_p)\hat{k}_d \hat{\nu}_u'/\{2[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i\cos^2\phi\}$. In this limit, the dependence of electron-density fluctuations on gas-density fluctuations is unaffected by the presence of the negative ions. However, the presence of negative ions can have a pronounced effect on the response of the electron density to changes in gas temperature due to the increase of the detachment rate with gas temperature. When the gas temperature increases, the production rate of electrons due to detachment increases. If in addition $[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}] > 0$, the quasisteady electron kinetics condition requires the electron temperature to decrease, thereby reducing the production rate of electrons due to electron temperature variations. Accordingly from Eq. (48), the electron density must necessarily increase. On the other hand, when the negative-ion density becomes sufficiently large that $[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}] < 0$, the electron temperature must increase as the gas temperature increases in order to increase the loss rate of electrons due to attachment processes. Under these circumstances, from Eq. (48), the electron density must necessarily decrease. For Fourier components oriented along the direction of the applied electric field, the expression for $d\ln n_e/d\ln T$ remains unchanged and

$$\frac{d\ln n_e}{d\ln n} \simeq \hat{\nu}_u'/\{2[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i\cos^2\phi\}.$$

Since $(\hat{\nu}_u'/\hat{k}_i)$ is small, the corresponding electron-density fluctuation is relatively small. This is a consequence of the fact that for these values of ϕ, again from Eq. (49), electron-temperature fluctuations due to gas-density fluctuations are negligible. When $2[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]$ is less than $-\hat{\nu}_u'$, the denominator in Eqs. (76) and (77) vanishes at certain values of ϕ, determined by

$$2[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i\cos^2\phi_c + (1+\hat{\nu}_u - \hat{\nu}_m\cos^2\phi_c) = 0,$$

owing to the quasisteady and long-wavelength approximations made in the derivation of these equations. Although a more complete treatment removes this singular behavior the electron density remains very sensitive to fluctuations in gas density and temperature for ϕ near ϕ_c, and therefore the present results are sufficient for the purposes of this paper. As ϕ approaches ϕ_c such that $\cos^2\phi > \cos^2\phi_c$, the response of the electron density to changes in gas density and temperature increases rapidly. Small decreases in gas density and temperature during a fluctuation produce large increases in electron density. Correspondingly, for values of ϕ approaching odd integral multiples of $\pi/2$ such that $\cos^2\phi < \cos^2\phi_c$ the response of the electron density to changes in gas density and temperature reverses sign and consequently small decreases in gas density and temperature produce large decreases in electron density due to enhanced attachment loss resulting from the corresponding electron-temperature rise. From Eqs. (76) and (77), for propagation nearly normal to the direction of the applied electric field

$$\frac{d\ln n_e}{d\ln T} \simeq (n_n/n_p)\hat{k}_d$$

and

$$\frac{d\ln n_e}{d\ln n} \simeq -2[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]\sin^2\phi(\hat{k}_i/\hat{\nu}_u') \ .$$

The electron density continues to respond strongly to changes in gas density and temperature. In particular, the electron density increases with increasing gas temperature due to detachment. When the gas density decreases, from Eq. (49), the electron temperature increases. If, in addition, $[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]>0$, the production rate of electrons due to the electron-temperature variation also increases, and the quasisteady condition requires both the electron density and the recombination loss to increase as well. Correspondingly, if $[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]<0$, the electron density decreases with decreasing gas density.

From these results, the presence of negative ions can have a pronounced effect on electron-density fluctuations produced by changes in gas density and temperature which accompany the development of the sound, thermal, and vibrational relaxation modes. This response of the electron density to gas-temperature and -density changes may be quite large for discharge operating conditions in which $\hat{k}_a$, $\hat{k}_i$, and $\hat{k}_d$ are large, the response being greatest for propagation nearly normal to the direction of current flow.

If an external souce is employed to sustain the discharge ionization process, then Eqs. (76) and (77) become

$$\frac{d\ln n_e}{d\ln n} \simeq \frac{2(n_n/n_p)\hat{k}_a\sin^2\phi+\hat{\nu}_u'}{-2(n_n/n_p)\hat{k}_a\cos^2\phi+2\hat{\nu}_u'} \tag{78}$$

and

$$\frac{d\ln n_e}{d\ln T} \simeq \frac{(n_n/n_p)\hat{k}_d\,\hat{\nu}_u'}{-2(n_n/n_p)\hat{k}_a\cos^2\phi+2\hat{\nu}_u'}. \tag{79}$$

If $\hat{k}_a$ is large and positive as the electron temperature is reduced by lowering the discharge E/n, (n_n/n_p) becomes small, and the sensitivity of the electron density to changes in gas density and temperature may be significantly reduced. If $\hat{k}_a$ is small compared to $\hat{\nu}_u'$, the effect of negative ions on $d\ln n_e/d\ln n$ is eliminated. However, the response of the electron density to gas-temperature fluctuations is enhanced if the detachment rate is a strong function of gas temperature. If $\hat{k}_a$ is large and negative, the effectiveness of the external source is greatly reduced, since $(-\hat{k}_a)$ behaves similarly to $\hat{k}_i$ and n_n/n_p tends toward unity.

In summary, under certain conditions the use of an external ionization source,[6,7] such as an electron-beam sustainer,[7] significantly reduces the dependence of electron-density fluctuations on gas-density and -temperature fluctuations.

When electronic species[18] play a significant role in electron kinetics and negative ions are absent, from Eqs. (65), (66), and (71)–(74), ζ_1, ζ_2, and ζ_3 are generally positive and $d\ln n_e/d\ln n$ behaves similar to the way it behaves in the absence of the electronically excited species. On the other hand, since the quenching rate tends to increase with increasing gas temperature, the electron density decreases with increasing gas temperature within a fluctuation. This is a consequence of the reduction in ionizable electronically excited species due to quenching as the gas temperature rises. If an external source is employed and the electron temperature is lowered significantly (a few tenths of an electron volt, or more) the ionization and electronic excitation times increase sufficiently that the sensitivity of electron density to gas-density and -temperature fluctuations is greatly reduced.

Sound Mode

From the quasineutrality condition [Eq. 44], the normalized charge-density fluctuation ρ_c' is on the order of $\epsilon_0 E/en_p l$, and consequently for molecular-laser discharge conditions

$$\left(\frac{en_pE}{\rho a^2/l}\right)\frac{\gamma}{\tau_s^2}\nabla\rho_c'\cdot\frac{\vec{\mathrm{E}}}{E}\sim\left(\frac{\epsilon_0E^2}{\rho al}\right)\frac{1}{\tau_s}\ll\frac{1}{\tau_s^2}.$$

Space-charge fluctuations have no direct effect on the evolution of gas-pressure fluctuations, i.e., sound waves, within these discharges. In addition, when the characteristic time associated with the sound disturbance τ_s is much less than the characteristic times associated with gas heating τ_T vibrational relaxation, τ_{VT}, and dissipation of vorticity, τ_μ, the motion of the gas is nearly irrotational, isentropic, and the vibrational degree of freedom is nearly frozen. This condition is satisfied if the characteristic dimension of the disturbance is bounded from above and below,

$$\left(\frac{\mu}{\rho a}\right),\left(\frac{\lambda_V^V}{n_mC_v^Va}\right),\left(\frac{\lambda_T^T}{nC_va}\right)\ll l\ll\left(\frac{nC_vTa}{J_e^2/\sigma_e}\right)\left(\frac{n_mC_v^VT_Va}{J_e^2/\sigma_e}\right),$$

where the bounds depend on the appropriate viscosity, conductivities, gas density, gas temperature, degree of vibrational excitation, and discharge electrical power density. Consistent with this condition, electron transport effects in the quasisteady-electron-energy equation are small, i.e., $\tau_e/\tau_{T_e} \ll 1$. When these conditions are not satisfied, the sound, vorticity, thermal, and vibrational relaxation modes couple strongly together, severely complicating the analysis.

When coupling effects are unimportant the dispersion relation for sound waves may be written $i\omega_{\mathbf{k}}^{\pm} \simeq \alpha_{\mathbf{k}}^{\pm} \pm iak$, where

$$2\alpha_{\mathbf{k}}^{\pm} \simeq -\left[\left(\frac{\mu_B+\frac{4}{3}\mu}{\rho}\right)+(\gamma-1)\left(\frac{\lambda_T^T}{nC_p}\right)\right]k^2+\frac{1}{\tau_T}\left\{P_V[2-(\gamma-1)\hat{\tau}_{\text{VT}}]-(\gamma-1)\left(\frac{C_v^V(T)}{C_v^V(T_V)}\right)\frac{T}{T_V}\left(\frac{\tau_V}{\tau_{\text{VT}}}\right)\right.$$
$$+P_{\text{TR}}\left[1+\left(\frac{d\ln n_e}{d\ln n}+(\gamma-1)\frac{d\ln n_e}{d\ln T}\right)\left(1+\frac{d\ln T_e}{d\ln n_e}(1+\hat{\nu}_{\text{TR}})\right)\right.$$
$$\left.\left.+\frac{d\ln T_e}{d\ln n}(1+\hat{\nu}_{\text{TR}})\right]\right\}. \tag{80}$$

The sound propagation velocity is essentially unaffected by the weakly ionized plasma and the damping or growth of sound waves is the same for propagation in directions separated by π radians.

In the absence of a discharge plasma the gas and vibrational temperatures are equal,

$$\alpha_{\mathbf{k}}^{\pm} \simeq -\frac{1}{2}\left[\left(\frac{\mu_B+\frac{4}{3}\mu}{\rho}\right)+(\gamma-1)\left(\frac{\lambda_T^T}{nC_p}\right)\right]k^2 - \frac{(\gamma-1)}{2}\frac{n_m C_v^V(T)}{nC_p\tau_{\text{VT}}}, \tag{81}$$

and the sound waves are damped by viscous and thermal transport processes and energy absorption by molecular vibration.

In the presence of a discharge plasma the electron temperature exceeds the vibrational temperature which, in turn, is elevated above the gas temperature, and the sound waves will be unstable if $\alpha_{\mathbf{k}}^{\pm} > 0$. This instability appears when the rate of heating of the coupled translational-rotational degree of freedom due to vibrational relaxation and electron excitation of translation-rotation is in phase with pressure fluctuations in the sound wave. For conditions of interest in electrically excited molecular lasers, the fractional electron power transfer into vibration and electronic excitation significantly exceeds that into rotation and translation and the condition for instability may be written

$$\frac{P_V}{\tau_T}[2-(\gamma-1)\hat{\tau}_{\text{VT}}] > \left[\left(\frac{\mu_B+\frac{4}{3}\mu}{\rho}\right)+(\gamma-1)\left(\frac{\lambda_T^T}{nC_p}\right)\right]k^2 + (\gamma-1)\frac{n_m C_v^V(T)}{nC_p}\frac{1}{\tau_{\text{VT}}}. \tag{82}$$

A necessary but not sufficient condition for instability is $[2-(\gamma-1)\hat{\tau}_{\text{VT}}] > 0$. When Eq. (82) holds, the waves are destabilized if energy transfer due to vibrational relaxation exceeds the ability of thermal conduction and the thermal capacity of the vibrational degree of freedom to dissipate this energy. The growth time of the instability is on the order of τ_T, the characteristic time for gas heating. If $\tau_T > L_d/a$, where L_d is the discharge length, the sound waves may propagate out of the system before they are amplified sufficiently to disturb the plasma. In supersonic flows, the possibility of standing waves and convection must be considered.

In terms of more fundamental physical parameters, Eq. (82) may be written

$$\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\left(\frac{T_e}{T}\right)[2-(\gamma-1)\hat{\tau}_{\text{VT}}] > \left[\left(\frac{\mu_B+\frac{4}{3}\mu}{\rho}\right)+(\gamma-1)\left(\frac{\lambda_T^T}{nC_p}\right)\right]k^2 + (\gamma-1)\frac{n_m C_v^V(T)}{nC_p\tau_{\text{VT}}}. \tag{83}$$

For discharge conditions of interest in molecular lasers, $\epsilon \gg \kappa T$. Then from the vibrational relaxation data of Taylor and Bitterman[9] and the correlations of Millikan and White[8] for many diatomic-molecule–noble-gas mixtures, $\hat{\tau}_{\text{VT}}$ is approximately −5 to −10 for gas temperatures in the 300–600 °K range. In addition $\hat{\tau}_{\text{VT}} \sim -T^{-1/3}$, and becomes more negative as the discharge-gas temperature is lowered. Therefore, as the degree of nonequilibrium, such as the fractional ionization and/or the ratio of electron to gas temperature, is increased, the condition, Eq. (83), for sound wave instability is eventually satisfied. At higher pressures and lower gas temperatures, the right-hand side, associated primarily with transport processes, decreases rapidly while the left-hand side increases, and the sound mode becomes less stable. Provided $(n_e/n)\nu_V(\kappa/C_p)(T_e/T)[2-(\gamma-1)\hat{\tau}_{\text{VT}}] > (\gamma-1)(n_m/n) \times (C_v^V/C_p)\tau_{\text{VT}}^{-1}$, from Eq. (83) a critical wavelength

$$\lambda_{\rm crit}^2=(2\pi)^2\left[\left(\frac{\mu_B+\frac{4}{3}\mu}{\rho}\right)+(\gamma-1)\left(\frac{\lambda_T^T}{nC_p}\right)\right]\left[\frac{n_e}{n}\nu_V\left(\frac{\kappa}{C_p}\right)\times\left(\frac{T_e}{T}\right)[2-(\gamma-1)\hat{\tau}_{\rm VT}]-\frac{(\gamma-1)n_mC_v^V(T)}{nC_p\tau_{\rm VT}}\right]^{-1} \quad (84)$$

exists such that if $\lambda>\lambda_{\rm crit}$ the sound mode is unstable.

When an external source is used to maintain the discharge ionization process the left-hand side of Eq. (83) may be reduced. However, for discharge conditions of interest, $\hat{\nu}_V$ is on the order of unity, and this reduction is not very large. Consequently, the use of an external source is expected to have only a small effect on sound-mode stability.

When electron excitation of rotation and translation dominates the electron-energy loss during a disturbance, for instance when plasma properties are very sensitive to changes in gas density and temperature or at very low molecular fractions, it can be seen from Eq. (80), and from Eqs. (85)–(87) in the next subsection, that charged-particle kinetics play a fundamental role in the evolution of the sound and thermal modes. However, these conditions are not considered in any detail in this paper.

Coupled Thermal and Vibrational Relaxation Modes

As indicated in Sec. I local heating of the gas is due predominantly to vibrational relaxation and rotation-translation excitation by electron impact, whereas the vibrational mode of the molecules is excited by electron collisions and deexcited by collisions with the atoms of the diluent. The characteristic wave modes associated with the evolution of these degrees of freedom of the plasma are referred to as the thermal and vibrational relaxation modes, respectively. In general, these modes couple together with sound waves to represent the relaxation of the neutral gas following a disturbance. In the following, for the purpose of illustrating the fundamental physical processes which influence the evolution of these modes, conditions are considered such that these modes are decoupled from the sound mode. These conditions are often satisfied in practice.

When the characteristic time τ_s associated with the sound disturbance is much less than the characteristic times for the thermal, τ_T, and vibrational, τ_V and $\tau_{\rm VT}$, relaxation modes, from Eq. (22) $\tau_s^{-2}\nabla^2p'\simeq 0$. Consequently, these phenomena do not generate pressure fluctuations and, to first order, $n'\simeq -T'$. Thus, while the density varies, only a weak irrotational velocity field necessary for the conservation of mass is generated.

Since for many conditions of practical interest τ_T, τ_V, and $\tau_{\rm VT}$ are of comparable magnitudes, the relaxation of the gas and vibration temperatures is coupled. Under these circumstances, the dispersion relation for the coupled thermal and vibrational relaxation modes is obtained from the translation-rotation, Eq. (26), and vibrational internal, Eq. (27), energy equations using the constant-pressure condition. As in the case with the treatment of other neutral-gas modes, the charged-particle dynamics is quasisteady and quasineutral. Then the dispersion relation for the coupled thermal and vibrational relaxation modes is a quadratic with solutions given by

$$i\omega_{\rm K}^{\pm}=\tfrac{1}{2}[-b\pm(b^2-4c)^{1/2}], \quad (85)$$

where

$$b\equiv\left[\left(\frac{\lambda_T^T}{nC_p}\right)+\left(\frac{\lambda_V^V}{n_mC_v^V(T_V)}\right)\right]k^2+\left(1+\frac{n_mC_v^V(T)}{nC_p}\right)\frac{1}{\tau_{\rm VT}}+\frac{1}{\tau_T}\left\{P_V(2+\hat{\tau}_{\rm VT})+P_{\rm TR}\left[1+\frac{d\ln T_e}{d\ln n}(1+\hat{\nu}_{\rm TR})+\left(\frac{d\ln n_e}{d\ln n}-\frac{d\ln n_e}{d\ln T}\right)\left(1+\frac{d\ln T_e}{d\ln n_e}(1+\hat{\nu}_{\rm TR})\right)\right]\right\} \quad (86)$$

and

$$c\equiv\left(\frac{\lambda_V^V}{n_mC_v^V(T_V)}\right)k^2\left(\left(\frac{\lambda_T^T}{nC_p}\right)k^2+\left(\frac{n_mC_v^V(T)}{nC_p}\right)\frac{1}{\tau_{\rm VT}}+\frac{1}{\tau_T}\left\{P_V(2+\hat{\tau}_{\rm VT})+P_{\rm TR}\left[1+\frac{d\ln T_e}{d\ln n}(1+\hat{\nu}_{\rm TR})+\left(\frac{d\ln n_e}{d\ln n}-\frac{d\ln n_e}{d\ln T}\right)\left(1+\frac{d\ln T_e}{d\ln n_e}(1+\hat{\nu}_{\rm TR})\right)\right]\right\}\right)$$
$$+\frac{1}{\tau_{\rm VT}}\left\{\left(\frac{\lambda_T^T}{nC_p}\right)k^2+\frac{1}{\tau_T}\left[P_{\rm TR}+P_V+\frac{d\ln T_e}{d\ln n}[P_{\rm TR}(1+\hat{\nu}_{\rm TR})+P_V(1+\hat{\nu}_V)]+\left(\frac{d\ln n_e}{d\ln n}-\frac{d\ln n_e}{d\ln T}\right)\times\left(P_{\rm TR}+P_V+\frac{d\ln T_e}{d\ln n_e}[P_{\rm TR}(1+\hat{\nu}_{\rm TR})+P_V(1+\hat{\nu}_V)]\right)\right]\right\}. \quad (87)$$

In the absence of a discharge, from Eqs. (85)-(87), the thermal-mode dispersion relation is simply

$$i\omega_{\bar{k}}^{T} \simeq -\left(\frac{\lambda_T^T}{nC_p}k^2 + \frac{n_m C_v^V(T)}{nC_p \tau_{VT}}\right), \tag{88}$$

and the thermal mode is associated with the diffusion of hot spots in a heat-conducting-fluid medium. The dispersion relation for the vibrational relaxation mode is

$$i\omega_{\bar{k}}^{T} \simeq -\left(\frac{\lambda_V^V k^2}{n_m C_v^V(T)} + \frac{1}{\tau_{VT}}\right), \tag{89}$$

and it characterizes the dissipation of internal energy concentrations within the fluid by diffusion and V-T relaxation. For many cases of interest in molecular-discharge-laser technology, electron rotation-translation excitation is insignificant. Thus, owing to the presence of the atomic diluent, $(\lambda_V^V/n_m C_v^V)k^2$ is much less than $[(\lambda_T^T/nC_p)k^2, \tau_{VT}^{-1}]$, and consequently

$$b \simeq \left(\frac{\lambda_T^T}{nC_p}\right)k^2 + \left(1 + \frac{n_m C_v^V(T)}{nC_p}\right)\frac{1}{\tau_{VT}} + \frac{P_V}{\tau_T}(2 + \hat{\tau}_{VT}) \tag{90}$$

and

$$c \simeq \frac{1}{\tau_{VT}}\left\{\left(\frac{\lambda_T^T}{nC_p}\right)k^2 + \frac{P_V}{\tau_T}\left[1 + \frac{d\ln T_e}{d\ln n}(1+\hat{\nu}_V) + \left(\frac{d\ln n_e}{d\ln n} - \frac{d\ln n_e}{d\ln T}\right)\left(1 + \frac{d\ln T_e}{d\ln n_e}(1+\hat{\nu}_V)\right)\right]\right\}. \tag{91}$$

From Eq. (85), taking account of the physical processes contributing to the coefficients b and c indicates that, if $b > 0$ and $c < 0$, the plasma is unstable because of the vibrational relaxation mode. If $b < 0$, the thermal mode is unstable, and, if $c > 0$, the vibrational relaxation mode is unstable also.

Thermal Mode

From the previous results, the thermal mode is unstable if

$$-\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}(2 + \hat{\tau}_{VT}) > \left[\left(\frac{\lambda_T^T}{nC_p}\right)k^2 + \left(1 + \frac{n_m C_v^V(T)}{nC_p}\right)\frac{1}{\tau_{VT}}\right]. \tag{92}$$

A necessary condition for instability is $(2 + \hat{\tau}_{VT}) < 0$, i.e., the V-T relaxation time must decrease rapidly with increasing gas temperature. The instability in this case is driven by local thermal heating due to vibrational relaxation. As the gas temperature rises the V-T relaxation time τ_{VT} decreases, increasing the energy release within the gas due to vibrational relaxation. If thermal conduction is unable to transport this energy away and back pumping of vibration is inefficient in reabsorbing this energy, the gas temperature will continue to rise, producing a runaway condition. The characteristic time for evolution of the instability is τ_T, generally 10^{-3}–10^{-2} sec. Since to first order the pressure remains nearly constant during the linear stage of the instability, the gas density decreases and the local electrical conductivity rises sharply producing local concentrations in the current flow. From Eq. (92) it can be seen that discharges with high degree of ionization and high electron-temperature–to–gas-temperature ratio, associated with high electrical power density, are most likely to be unstable to the thermal mode. Furthermore, at low gas temperature and high pressure the left-hand side of Eq. (92) is large while the right-hand side is decreased, and these conditions also favor instability. Provided

$$-(n_e/n)\nu_V(\kappa/C_p)(T_e/T)(2 + \hat{\tau}_{VT}) > \{1 + [n_m C_v^V(T)/(nC_p)]\}\tau_{VT}^{-1},$$

a critical wavelength

$$\lambda_{\text{crit}}^2 = (2\pi)^2\left(\frac{\lambda_T^T}{nC_p}\right)\left[-\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\left(\frac{T_e}{T}\right)(2 + \hat{\tau}_{VT}) - \left(1 + \frac{n_m C_v^V(T)}{nC_p}\right)\frac{1}{\tau_{VT}}\right]^{-1} \tag{93}$$

exists such that if $\lambda > \lambda_{\text{crit}}$ the thermal mode is unstable. As the degree of nonequilibrium increases the ability of thermal conduction to dissipate local hot spots decreases and the critical wavelength decreases.

An external source is expected to be relatively ineffective in stabilizing this mode. It should be noted, however, that if the gas temperature is increased at constant pressure and E/n, $\hat{\tau}_{VT}$, and T_e/T are decreased, the gas thermal conductivity is increased, τ_{VT} and n are decreased and, from Eq. (92), this mode and the sound mode can be stabilized. The temperature rise required, however, may exceed acceptable conditions for efficient laser operation.

Vibrational Relaxation Mode

If electron vibrational excitation exceeds electron translation-rotation excitation, $P_V \gg P_{TR}$, from Eqs. (85)-(87) the vibrational relaxation mode is unstable when

$$-\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\left(\frac{T_e}{T}\right)(2 + \hat{\tau}_{VT}) - \left(1 + \frac{n_m C_v^V(T)}{nC_p}\right)\frac{1}{\tau_{VT}} \lessgtr \left(\frac{\lambda_T^T}{nC_p}\right)k^2 \tag{94}$$

and

$$\left(\frac{\lambda_T^T}{nC_p}\right)k^2 \lessgtr -\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}\left\{1+\frac{d\ln T_e}{d\ln n}(1+\hat{\nu}_V) + \left(\frac{d\ln n_e}{d\ln n}-\frac{d\ln n_e}{d\ln T}\right)\left[1+\frac{d\ln T_e}{d\ln n_e}(1+\hat{\nu}_V)\right]\right\}. \tag{95}$$

If the plasma conditions are such that the upper inequality signs in Eqs. (94) and (95) are satisfied the unstable vibrational relaxation mode is convected with the gas flow. However, when the lower inequality conditions in Eqs. (94) and (95) hold, and if the electron-density and -temperature fluctuations are sufficiently large, coupling between the unstable thermal and vibrational relaxation modes results in equal and oppositely directed phase velocities, relative to the gas, for the Fourier components of the two modes. In this case the vibrational relaxation mode drifts relative to the gas in the direction of electron flow.

The vibrational-relaxation-mode instability is driven by electron vibrational excitation. As the vibrational temperature increases during the instability, the gas temperature increases when the upper inequality signs hold in Eqs. (94) and (95), and decreases when the lower inequality signs hold. The changing gas density and temperature, at nearly constant pressure, produce fluctuations in electron temperature and electron density which, in turn, lead to enhanced vibrational excitation. Vibrational energy relaxation, due to collisions with the atomic diluent, is insufficient to compensate for the enhanced pumping of vibration by electrons during the disturbance. The vibrational degree of freedom becomes a bottleneck, and the energy stored in molecular vibration increases. As a consequence of the variation in gas density, electron density, and temperature, the local conductivity increases usually producing local concentrations in the current flow.

To consider the physical conditions leading to the appearance of vibrational-relaxation-mode instability, Eq. (95) is rewritten using Eqs. (65) and (66) to give

$$\left(\frac{\lambda_T^T}{nC_p}\right)k^2 \lessgtr -\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}\left(\frac{2\zeta_1\hat{k}_i\cos 2\phi - 2[(\zeta_2-\zeta_4)\cos^2\phi+\zeta_3\sin^2\phi](1+\hat{\nu}_V)+(\zeta_2+\zeta_3-\zeta_4)\hat{\nu}_u'}{2\zeta_1\hat{k}_i\cos^2\phi+\zeta_3\hat{\nu}_u'}\right), \tag{96}$$

where the ζ_j's have been defined previously, Eqs. (67)–(74). For a conventional discharge, in the absence of negative ions and electronically excited species, Eq. (96) becomes

$$\left(\frac{\lambda_T^T}{nC_p}\right)k^2 \lessgtr -2\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}\left(\frac{\hat{k}_i\cos 2\phi+(\hat{\nu}_u'-1-\hat{\nu}_V)}{2\hat{k}_i\cos^2\phi+\hat{\nu}_u'}\right). \tag{97}$$

This expression may be further simplified by noting that the upper and lower inequality signs in Eq. (97) are first satisfied for ϕ equal to odd multiples of $\pi/2$ and even multiples of π, respectively, since $\hat{k}_i \gg \hat{\nu}_u'$ and $\hat{\nu}_u > \hat{\nu}_V$ in most molecular-gas discharges. Combining these results with Eq. (94), the vibrational mode is unstable for a bounded range of wave numbers in the absence of negative ions and electronically excited species when

$$-\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}(2+\hat{\tau}_{VT})-\left(1+\frac{n_m C_v^V(T)}{nC_p}\right)\frac{1}{\tau_{VT}} < \left(\frac{\lambda_T^T}{nC_p}\right)k^2 < 2\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}\left(\frac{\hat{k}_i}{(1+\hat{\nu}_u+\hat{\nu}_m)}\right) \tag{98}$$

or

$$-\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T} < \left(\frac{\lambda_T^T}{nC_p}\right)k^2 < -\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}\left(2+\hat{\tau}_{VT}\right)-\left(1+\frac{n_m C_v^V(T)}{nC_p}\right)\frac{1}{\tau_{VT}}. \tag{99}$$

The possibility of instability exists because of enhanced electron pumping of vibration. When Eq. (98) is satisfied, this enhanced electron pumping is due to the large variations in electron density produced by the electron-temperature fluctuations generated by the decreasing gas density during the evolution of the vibrational relaxation mode. Correspondingly, when Eq. (99) is satisfied the enhanced electron pumping is due to the increase in electron density generated by the increasing gas density.

When the discharge is maintained using an external source, the dependence of electron-density fluctuations and, hence, fluctuations in vibrational excitation on gas-density fluctuations can be significantly reduced, and the instability conditions, Eqs. (94) and (95), become

$$\frac{1}{2}\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}(1+4\hat{\nu}_V-3\hat{\nu}_u-3\hat{\nu}_m)>\left(\frac{\lambda_T^T}{nC_p}\right)k^2 > -\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}(2+\hat{\tau}_{\mathrm{VT}}) - \left(1+\frac{n_m C_v^V(T)}{nC_p}\right)\frac{1}{\tau_{\mathrm{VT}}} \tag{100}$$

or

$$-\frac{1}{2}\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}(1+3\hat{\nu}_u-3\hat{\nu}_m-2\hat{\nu}_V)<\left(\frac{\lambda_T^T}{nC_p}\right)k^2 < -\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}(2+\hat{\tau}_{\mathrm{VT}}) - \left(1+\frac{n_m C_v^V(T)}{nC_p}\right)\frac{1}{\tau_{\mathrm{VT}}}\,. \tag{101}$$

Under these conditions the possibility of instability is significantly reduced but not eliminated. A necessary, but not sufficient, condition for Eq. (100) to be satisfied is $[1+4\hat{\nu}_V-3(\hat{\nu}_u+\hat{\nu}_m)]>0$, which is usually satisfied when vibrational excitation dominates electron collisional energy loss and $\hat{\nu}_V$ is large compared to $\hat{\nu}_m$. When Eq. (101) is satisfied the thermal mode is also unstable.

When negative ions are present within the discharge in sufficient quantity, from Eqs. (67)–(70) it is clear that the instability condition Eq. (96) becomes very complicated. To simplify the discussion, suppose detachment dominates negative-ion collisional loss. Then Eq. (96) for a conventional discharge becomes

$$\left(\frac{\lambda_T^T}{nC_p}\right)k^2 \lessgtr -\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}\left(\frac{[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i\cos 2\phi-[1-\quad /n_p)\hat{k}_d\cos^2\phi](1+\hat{\nu}_V)+[1-(n_n/2n_p)\hat{k}_d]\hat{\nu}_u'}{[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i\cos^2\phi+\frac{1}{2}\hat{\nu}_u'}\right). \tag{102}$$

From Eqs. (94) and (102) it is clear that when negative ions are present the possibility of vibrational-relaxation-mode instability depends on the relative magnitudes of $\hat{k}_a$,

$\hat{k}_i$, $\hat{k}_d$, $(2+\hat{\tau}_{\mathrm{VT}})$, and (n_n/n_p). When $[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]>0$ the instability first appears for ϕ near odd integral multiples of $\pi/2$ when

$$2\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}\left(\frac{[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i+(1+\hat{\nu}_V)+[(n_n/2n_p)\hat{k}_d-1](1+\hat{\nu}_u+\hat{\nu}_m)}{(1+\hat{\nu}_u+\hat{\nu}_m)}\right) > \left(\frac{\lambda_T^T}{nC_p}\right)k^2 > -\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}(2+\hat{\tau}_{\mathrm{VT}}) - \left(1+\frac{n_m C_v^V(T)}{nC_p}\right)\frac{1}{\tau_{\mathrm{VT}}}, \tag{103}$$

and for ϕ near even multiples of π when

$$-\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}\left(\frac{[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i+(\hat{\nu}_u-\hat{\nu}_V-\hat{\nu}_m)+(n_n/n_p)\hat{k}_d[\hat{\nu}_V+(1-\hat{\nu}_u+\hat{\nu}_m)/2]}{[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i+(1+\hat{\nu}_u-\hat{\nu}_m)/2}\right) < \left(\frac{\lambda_T^T}{nC_p}\right)k^2 < -\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}(2+\hat{\tau}_{\mathrm{VT}}) - \left(1+\frac{n_m C_v^V(T)}{nC_p}\right)\frac{1}{\tau_{\mathrm{VT}}}\,. \tag{104}$$

Since $\hat{k}_a$ is usually positive, the attachment process has a stabilizing influence as the negative-ion density increases. Correspondingly detachment destabilizes the vibrational relaxation mode. If $\hat{k}_d$ is much greater than 1, this may be a very significant effect according to Eqs. (103) and (104).

If $-(1+\hat{\nu}_u+\hat{\nu}_m)<[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i<0$ the treatment of the conditions for vibrational-relaxation-mode instability is complicated and requires detailed consideration of Eqs. (94) and (102). However, if $[1-(n_n/n_p)\hat{k}_a(\hat{k}_i)^{-1}]\hat{k}_i<-(1+\hat{\nu}_u+\hat{\nu}_m)$ the denominator in Eq. (102) vanishes at ϕ_c owing to the singular behavior, discussed previously, of $d\ln n_e/d\ln T$ and $d\ln n_e/d\ln n$ at ϕ_c. Under these conditions Eqs. (94) and (102) are readily satisfied for all wavelengths. Since typically $\hat{k}_i \gg \hat{\nu}_u'$ the just mentioned condition reduces to $[1-(n_n/n_p)\times\hat{k}_a(\hat{k}_i)^{-1}]<0$ and the instability first appears when ϕ is equal to ϕ_c, which is near an integral multiple of $\pi/2$.

When an external source is employed to sustain the discharge if $\hat{k}_a$ is large and positive, as the electron temperature is reduced (n_n/n_p) becomes small and the conditions for instability are given by Eqs. (100) and (101). The effect of the negative ions is eliminated; however, the vibrational relaxation mode is not necessarily stabilized. If $\hat{k}_a$ is small compared to $\hat{\nu}_u'$ and $\hat{\nu}_V$ when an external ionization source is used the instability first appears for ϕ near integral multiples of $\pi/2$ when

$$\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}\left(\frac{2(1+\hat{\nu}_V)+[(n_n/n_p)\hat{k}_d-3](1+\hat{\nu}_u+\hat{\nu}_m)/2}{(1+\hat{\nu}_u+\hat{\nu}_m)}\right)>\left(\frac{\lambda_T^T}{nC_p}\right)k^2$$
$$>-\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}(2+\hat{\tau}_{\mathrm{VT}})-\left(1+\frac{n_m C_v^V(T)}{nC_p}\right)\frac{1}{\tau_{\mathrm{VT}}}, \tag{105}$$

and for ϕ near even multiples of π when

$$\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}\left(\frac{[\hat{\nu}_V+(1-\hat{\nu}_u+\hat{\nu}_m)/2][1-(n_n/n_p)\hat{k}_d]-(1+\hat{\nu}_u-\hat{\nu}_m)}{(1+\hat{\nu}_u-\hat{\nu}_m)}\right)<\left(\frac{\lambda_T^T}{nC_p}\right)k^2$$
$$<-\left(\frac{n_e}{n}\right)\nu_V\left(\frac{\kappa}{C_p}\right)\frac{T_e}{T}(2+\hat{\tau}_{\mathrm{VT}})-\left(1+\frac{n_m C_v^V(T)}{nC_p}\right)\frac{1}{\tau_{\mathrm{VT}}}. \tag{106}$$

The importance of the gas-temperature dependence of the detachment-rate constant is evident. If $\hat{k}_a$ is large and negative no improvement in the stability of the vibrational relaxation mode is possible through the use of auxiliary ionization.

When electronic species play a significant role in electron kinetics and negative ions are absent, Eq. (96) remains valid if the ζ_j's are calculated using Eqs. (71)–(74). Since ζ_1, ζ_2, and ζ_3 are positive, the vibrational relaxation mode behaves in the absence of quenching in a manner similar to its behavior in the absence of electronically excited species and negative ions. The presence of quenching tends to stabilize the plasma vibrational relaxation process as a consequence of the reduction in ionizable electronically excited species. The electron density decreases with increasing gas temperature during evolution of this mode because $\hat{k}_q > 0$. If an external source is employed and the electron temperature is lowered significantly, the ionization and electronic excitation times increase sufficiently that the sensitivity of electron density to gas-density and -temperature fluctuations is greatly reduced, and Eqs. (100) and (101) hold.

Vorticity Mode

From Eq. (21), the vorticity fluctuations associated with a disturbance in the plasma are damped by a diffusion process associated with viscous dissipation and interact with charged-particle processes through the gradient in charge-density fluctuation. From the quasineutrality condition, Eq. (44), ρ_c' is on the order of $(\epsilon_0 E/en_p l)$. Consequently the last term in Eq. (21) is

$$\left(\frac{en_p E}{\rho a^2/l}\right)\frac{1}{\tau_s}(\nabla\rho_c')\times\frac{\vec{E}}{E}\sim\left(\frac{\epsilon_0 E^2}{\rho a l}\right)\ll\frac{1}{\tau_\mu},$$

and viscous dissipation dominates the evolution of vorticity fluctuations. Thus Eq. (21) becomes essentially identical to the equation describing the production, convection, and dissipation of weak vorticity fluctuations in a viscous incompressible medium. Such a flow field does not generate pressure fluctuations of the same order, since the pressure fluctuation is proportional to the square of the velocity fluctuation and hence is negligible when the vorticity fluctuation is weak. For a similar reason there is no heating of the gas or vibrational mode; viscous dissipation is again second order, and the velocity field is essentially solenoidal as it should be for an incompressible motion. Consequently, coupling to charged-particle kinetics, which occurs through volumetric dilatation and gas-density and -temperature fluctuations, is necessarily negligible, and the dispersion relation for vorticity fluctuations is simply $i\omega_{\mathrm{k}}^{\dagger}\simeq-(\mu/\rho)k^2$.

Influence of Aerodynamic Techniques

Several investigators[4,5] have observed that aerodynamic conditioning, such as introduction of turbulence generators upstream of the discharge, has a significant stabilizing effect on molecular-laser discharges at low pressure. Experiments by Garosi, Bekefi, and Schulz[43] on a constricted argon arc discharge have shown that the plasma of a weakly ionized gas cannot appreciably change the turbulent field of the neutral gas, although ion-neutral collisions readily transfer turbulent motions from the uncharged to the charged particles. Therefore, in addition to enhancing neutral-particle transport processes, the small-scale (high-frequency) eddies of the turbulence were observed[43] to significantly increase the transport properties of the plasma medium. The resulting increase in particle loss was also observed to increase the electron temperature. On the basis of the results of the stability analysis of this paper, these effects under certain conditions should lead to improved stability. The enhanced transport properties increase the critical wavelength for instabilities of all types. In addition, under many conditions of practical interest an increase in electron temperature can significantly reduce the ratio $\tau_i\hat{k}_a/(\tau_a\hat{k}_i)$. Therefore the increased electron temperature produced by the turbulent flow should favorably alter the conditions for instability onset of the ionization, negative-ion, and vibrational relaxation modes when negative ions are present within the discharge.

The explanation of high-power-laser-discharge results[4,5] in this fashion, however, is complicated by several other possible effects. Firstly, Garosi *et al.*[43] also observed that the large-scale (low-frequency) eddies present within the turbulence produced large-scale fluctuations of the constricted column, causing an expansion of the effective radius of the plasma column with increased gas flow. This effect would complicate the interpretations of high-power laser experiments[4,5] since the observed improvement in stability could simply be due to the thrashing of the discharge column in the turbulent gas flow. Finally, the observed improvements in discharge performance[4,5] may simply be that induced turbulence compensates for nonuniformities produced by the discrete electrode sets used in these discharges, thereby improving discharge uniformity and stability. Therefore, although on the basis of the stability theory developed in this paper gas dynamic turbulence should lead to improved stability, in the absence of detailed measurements the interpretation of experimental results[4,5] in this fashion is complicated by other possible effects.

In addition to gas turbulence, aerodynamic expansion should also produce a stabilizing effect on the discharge plasma. Aerodynamic expansion is more effective at high pressures. In the equations of motion, the effect of aerodynamic expansion enters through the volumetric dilatation $\nabla \cdot \mathbf{u}$. The characteristic time for local volume change of a fluid element during expansion is $\tau = (\nabla \cdot \mathbf{u})^{-1}$. For practical discharge conditions and flow expansion, τ is much larger than the characteristic times associated with charged-particle kinetics, and under these conditions fluid expansion has little effect on the ionization and negative-ion modes of instability. On the other hand, values of $\tau \ll \tau_T, \tau_V$ are possible, and under these conditions it can be shown that aerodynamic expansion could be employed to stabilize the thermal, vibrational relaxation, and possibly the sound modes. Finally, as a consequence of the slow growth rates of the thermal and vibrational relaxation modes, the possibility exists that they may be convected out of the discharge region by simply increasing the flow speed.

V. SUMMARY AND COMMENTS

The development of high-power-density, large-volume electric-discharge-excited molecular lasers has been impeded by the appearance of bulk-plasma instability. The results of the stability analysis of Sec. IV suggest that as a consequence of the highly nonequilibrium nature of the discharge plasma, several different instabilities are possible. The conditions for onset and the driving mechanisms of these instabilities are directly related to fundamental charged- and neutral-particle collisional-particle-production, energy-transfer, and transport processes.

It has been shown that for a locally uniform plasma, fluctuations within the plasma excite several fundamental wave modes. The physical characteristics of these different wave modes have been analyzed using the self-consistent, tenth-order, dispersion relation for the Fourier–Laplace components of an arbitrary disturbance. However, the characteristic time scales (Table I) associated with plasma processes of importance cover a very large range (10^{-10}–10^{-2} sec), permitting significant simplifications in the analysis of this dispersion relation. Taking account of the physical processes associated with their evolution, these normal wave modes have been identified and ordered according to their characteristic time scales as follows: a space-charge relaxation mode (10^{-10}–10^{-8} sec), an electron thermal mode (10^{-8}–10^{-7} sec), an ionization mode (10^{-6}–10^{-5} sec), a negative-ion-production mode (10^{-6}–10^{-5} sec), an electronic-species-production mode (10^{-6}–10^{-4} sec), a sound mode (10^{-5}–10^{-4} sec), a vibrational energy relaxation mode (10^{-4}–10^{-3} sec), a heavy-particle thermal mode (10^{-4}–10^{-3} sec), and a vorticity mode (10^{-3}–10^{-2} sec).

The stability criteria derived from this study indicate that the space-charge relaxation, electron thermal, and vorticity modes are stable for plasma conditions of interest in high-power electrically excited molecular lasers. However, the stability of the ionization, negative-ion production, and vibrational relaxation modes is critically dependent on: (i) the electron- and gas-temperature dependences of the rate coefficients for all the charged-particle production and loss processes, particularly those involving ionization, attachment, and detachment; (ii) the electron-temperature dependence of the electron-energy-loss collision frequency due to vibrational and electronic excitation; (iii) the gas-temperature dependence of the V-T relaxation rate coefficient and the magnitude of the gas thermal conductivity (vibrational relaxation mode only); and (iv) the steady-state-plasma properties such as the gas mixture, the degree of ionization (which is related to the discharge power density), the discharge E/n or equivalently the electron temperature, the relative negative-ion density which is sensitive to minority-species levels, and the ratios of electron and vibrational to gas temperature. In the absence of negative ions, the presence of a metastable electronically excited species[18] has been found to have no significant effect on discharge stability; i.e., the ionization and electronic species production modes are stable. However, the sensitivity of the stability of these modes to ionization kinetics in the presence of negative ions suggests[24] that the existence of electronically excited species may be important when negative ions are produced within the discharge. In general, for sufficiently small wavelengths, the ionization, negative-ion, and vibrational relaxation modes are stabilized by transport processes. In addition, it has been shown that, for a wide range of physical conditions, the application of an external source of alter steady-state-plasma conditions significantly reduces the sensitivity of electron-density fluctuations to electron-temperature, gas-density, and gas-temperature fluctuations, and thereby has a stabilizing influence on the ionization, negative-ion, and vibrational relaxation modes. The stability of the thermal and sound modes is shown in the analysis to be critically dependent on: (i) the gas-temperature dependence of the V-T relaxation rate coefficient, (ii) the magnitude of the gas viscosity and thermal conductivity, and (iii) the steady-state-plasma properties, such as the gas mixture, the degree of ionization, the gas temperature, and the ratios of electron and vibrational to gas temperature. When the thermal and sound modes are unstable, a critical wavelength exists below which these modes are stabilized by gas transport processes. These modes are often relatively insensitive to the details of the charged- and electronic-species kinetics. Consequently, their stability has been found to be insensitive to the application of external-source techniques, and they therefore constitute a general limitation to e-beam and other auxiliary ionization devices.[6,7] The approximations used in the present analysis of the dispersion relation have been evaluated by comparison with detailed computer solutions of the complete dispersion relation for the N_2-CO_2-He system obtained by Haas and Guckel.[17] Good agreement has been obtained.

The purpose of the present investigation has been to provide a physical understanding of the mechanisms that can lead to the instabilities observed in laboratory electrically excited molecular lasers. The accuracy and usefulness of this analysis depends on several fundamental experimental and theoretical considerations. The present lack of experimental data necessary for the calculation of charged-particle and electronically-excited-species rate coefficients and their temperature dependences limits the quantitative evaluation of some of the criteria and the assessment of the significance of certain kinetic processes such as detachment, quenching, etc. Evaluation of the importance of electronically excited species in the ionization, vibrational excitation, and gas-heating processes requires additional experimental data. Direct and detailed measurements of instability characteristics, such as onset conditions and initial growth rates under controlled discharge conditions, and comparison with analytical results are clearly needed since several types of instabilities may be present simultaneously. The instabilities occur on significantly different time scales, and hence experimental measurements of discharge temporal behavior should provide the determination of the class of instability.

Although the present analysis describes certain essential physical characteristics of the local behavior of molecular-gas discharge plasmas, the possibility exists that the presence of boundaries and plasma-flow gradients introduce effects of equal or greater significance as regards stability.[33] In addition, it is known that the instabilities observed manifest themselves in the form of a constriction of the discharge column. Precisely how the instabilities discussed here lead to constriction, particularly the charged-particle modes which are unstable only along or opposite to the direction of current flow, is not fully understood and requires asymptotic analysis of the complete integral representation, Eq. (37). Such an analysis would determine how and under what conditions constriction develops from an arbitrary disturbance. In addition to these considerations, nonlinear effects associated with the initial development of instability may quench certain modes of instability or destabilize the plasma in the presence of finite-amplitude fluctuations. The resolution of these theoretical and experimental questions by comparison of the previous analytical results, or an extension of them to include additional kinetic processes where appropriate, with experimental measurements for specific gas-discharge conditions represents a most important area for further research.

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[23]For a uniform plasma Phelps *et al.* (see Refs. 12–14) have defined the electron-energy-exchange collision frequency $\nu_u \equiv (j_e^2/\sigma_e)/\,[n_e(\epsilon_\kappa - \kappa T)]$ as the ratio of the power input per electron to the excess energy of the electron over its thermal value. The characteristic electron energy is $\epsilon_\kappa \equiv e D_e/\mu_e$. For many cases of interest $\epsilon_\kappa \sim \kappa T_e \gg \kappa T$ and superelastic collisions of electrons with vibrationally and rotationally excited molecules are unimportant (see Ref. 10). Then the volumetric rate of electron energy loss is $n_e n(\nu_u(T_e)/n)\kappa T_e$ and contains contributions from translation rotation ν_{TR}, vibration ν_V, and electronic excitation ν_e; $\nu_u = \nu_{TR} + \nu_V + \nu_e$. When vibrational excitation dominates electron energy loss ν_u is related to Nighan's (see Ref. 10) effective energy-loss collision frequency ν_{eff} by $\nu_u \simeq (\epsilon/\kappa T_e)\nu_{eff}$ provided $\epsilon_\kappa \sim \kappa T_e \gg \kappa T$.

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