

## Acoustic Wave Generation and Amplification in a Plasma\*

UNO INGARD

*Department of Physics and Research Laboratory of Electronics, Massachusetts Institute of Technology,  
Cambridge, Massachusetts*

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Acoustic wave generation resulting from the heating of neutral-gas component by the electrons in a weakly ionized plasma is analyzed. It is shown that under certain conditions wave amplification will result, and criteria for amplification and for spontaneous excitation of normal modes are derived. The results are used to explain sound emission in traveling striations in a glow discharge and acoustic modulation of a plasma afterglow. Recently reported experimental observations of acoustical effects in discharges are discussed in the light of the present analysis.

### INTRODUCTION

IN experiments with electrically modulated discharges several investigators have observed effects that appear to result from the excitation of ordinary acoustic waves in the neutral-gas component in the plasma. For example, Strickler and Stewart,<sup>1</sup> while modulating dc glow discharges in argon and krypton at pressures between 13 and 38 mm Hg, noticed a pronounced displacement or "kinking" of the constricted discharge path at a series of discrete modulation frequencies. They were able to identify these frequencies as those of radial and azimuthal acoustic modes of oscillation of the neutral-gas components. Similarly, in afterglow experiments with pulsed helium and neon plasmas, Berlande, Goldan, and Goldstein<sup>2</sup> found that during the decay period of the discharge the electron density and the light emission were periodically modulated. They proposed that this modulation is caused by pressure waves produced by the discharge itself. The frequency of modulation was found to be of the order of the fundamental resonance frequency of a lateral acoustic mode of the discharge tube. Similar observations of modulation of light emission<sup>3</sup> and ion density<sup>4</sup> by sound waves transmitted into a plasma from sound sources outside the plasma have been reported. Intimately related to these effects, no doubt, are the emission of sound by an electrically modulated corona discharge and the response of such a discharge to an external sound field.<sup>5</sup>

Even in the absence of external electrical modulation of a discharge, macroscopic fluctuations of electron density and temperature in a glow discharge are frequently present in the form of traveling striations or other instabilities, and it is to be expected that these will couple to the neutral gas also and give rise to acoustic waves. Actually, the occurrence of such

pressure fluctuations has recently been reported by Carretta and Moore.<sup>6</sup>

In view of these observations, it is of interest to carry through a systematic study of the mechanism involved in the sound emission by fluctuations in charge density and electron temperature in a weakly ionized gas; we propose to show how the acoustic field in the neutral-gas component can be calculated from the energy transfer of the elastic collisions between the charged particles and the neutrals. In particular, the sound field produced in a discharge with traveling striations and in a plasma afterglow will be calculated. Furthermore, the possibility of an acoustic-wave instability will be demonstrated which may serve to explain spontaneous acoustic oscillations that have been found in discharges under certain conditions by Alexeff and Neidigh.<sup>7</sup> This instability is of interest as a possible means of amplifying acoustic waves.

### ACOUSTIC SOURCES

Since we are interested mainly in the motion of the neutral-gas component in the plasma, we start by deriving the wave equation for the pressure in the neutral gas and express the interaction with the electrons and the ions in terms of sources in the wave equation. The equation follows from the relations expressing balance of mass, momentum, and energy, together with the equation of state for the gas  $P = P(\rho, S)$ , the pressure  $P$  being regarded as a function of density  $\rho$  and entropy  $S$ . The linearized versions of these equations are the following:

$$\partial\delta/\partial t + \rho_0 \operatorname{div} \mathbf{u} = Q, \quad (1)$$

$$\rho_0 \partial \mathbf{u} / \partial t + \operatorname{grad} p = \mathbf{F}, \quad (2)$$

$$\rho_0 T_0 \partial s / \partial t = H, \quad (3)$$

$$\delta = \left( \frac{\partial \rho}{\partial P} \right)_S p + \left( \frac{\partial \rho}{\partial S} \right)_P s = (1/c^2) p - (\rho_0/c_P) s. \quad (4)$$

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<sup>1</sup> S. D. Strickler and A. B. Stewart, *Phys. Rev. Letters* **11**, 527 (1963).

<sup>2</sup> J. Berlande, P. D. Goldan, and L. Goldstein, *Appl. Phys. Letters* **5**, 51 (1964).

<sup>3</sup> K. Wojacek, *Beitr. Plasma Physik* **1**, 127 (1960/61).

<sup>4</sup> K. W. Gentle and U. Ingard, *Appl. Phys. Letters* **5**, 105 (1964).

<sup>5</sup> B. W. Blum, I. Dyer, and U. Ingard, *J. Acoust. Soc. Am.* **26**, 139 (1954).

<sup>6</sup> A. A. Carretta, Jr., and W. N. Moore, Master's thesis, U. S. Naval Postgraduate School, Monterey, California, 1965 (unpublished).

<sup>7</sup> I. Alexeff and R. V. Neidigh, *Phys. Rev.* **129**, 516 (1963).

Here the perturbations in the field variables, density, pressure, velocity, and entropy per unit mass are  $\delta = \rho - \rho_0$ ,  $p = P - P_0$ ,  $\mathbf{u}$ , and  $s = S - S_0$ . The speed of sound is  $c$  and the specific heat per unit mass at constant pressure  $c_P$ . The source terms  $Q$ ,  $\mathbf{F}$ , and  $H$  are the rates of transfer of mass, momentum, and energy to the neutral gas per unit volume. If we neglect the fluctuations in the neutral-particle density resulting from the unbalance of ionization and recombination rates, we have  $Q = 0$ , and the wave equation for pressure resulting from Eqs. (1)–(4) is

$$\frac{1}{c^2} \frac{\partial^2 p}{\partial t^2} - \nabla^2 p = \frac{(\gamma - 1)}{c^2} \frac{\partial H}{\partial t} - \text{div} \mathbf{F}, \quad (5)$$

where  $\gamma$  is the specific heat ratio  $\gamma = c_P/c_V$ . The energy transfer to the neutrals is due largely to the elastic collisions with the electrons. In an ordinary laboratory discharge the electron temperature is considerably higher than the neutral-gas temperature, and it is a good approximation to consider the neutrals to be stationary in the elastic collisions between the electrons and the neutrals. Then, if we introduce an average elastic scattering cross section  $\langle \sigma \rangle$  for the electron-neutral collision and an average electron (thermal) speed  $v_e$ , the rate of energy transfer from the electrons to the neutrals per unit volume may be expressed as

$$H = (4m_e/m_n)(m_e v_e^3/2) N_e N_n \langle \sigma \rangle. \quad (6)$$

Here  $m$  and  $N$  stand for particle mass and particle density and the subscripts  $e$  and  $n$  refer to electrons and neutral particles, respectively. Introducing the electron temperature  $T_e$  defined by  $3kT_e/2 = m_e v_e^2/2$ , we can determine the average cross section  $\langle \sigma \rangle$  in terms of  $T_e$  in the usual way by evaluating the energy transfer integral involving the electron velocity distribution function and the differential elastic cross section.

Since the main energy loss of the electrons ordinarily is due to the elastic collisions with the neutrals, we may as an alternative express  $H$  approximately as the rate of energy gained by the electrons from the external electric field  $E$ . In a dc discharge this energy rate per unit volume of the plasma is  $N_e e E V_e$ , where  $V_e = b_e E$  is the drift velocity of the electrons,  $b_e$  being the mobility. The energy transfer from the ions can be expressed in a similar manner, but since the mobility of the ions usually is considerably smaller than the mobility of the electrons, the energy transfer from the ions will be comparatively small and it will be omitted in the present analysis.

In regard to the effect of the momentum transfer, represented by  $\text{div} \mathbf{F}$  in the wave equation, we note that the contributions from the electrons and the ions have opposite signs and tend to cancel each other at least at the comparatively low acoustic frequencies of interest here. Furthermore, only a spatial variation of  $\mathbf{F}$  leads to sound generation, and in the problems discussed here this contribution is negligible.

In the subsequent discussion of sound waves in a plasma afterglow, the relative pressure fluctuations caused by the sound are of particular importance, and therefore it is convenient to introduce as a reference the static gas pressure  $P_0$  in the source term in the wave equation. Thus with  $c^2 = \gamma P_0/\rho_0 = \gamma k T_n/m_n$  and  $v_e^2 = 3kT_e/m_e$ , where  $T_n$  and  $T_e$  are the temperatures of the neutrals and the electrons, the acoustic source term can be expressed as

$$\frac{(\gamma - 1)}{c^2} \frac{\partial H}{\partial t} = P_0 \left( \frac{m_e}{m_n} \right)^{1/2} \frac{1}{c} \frac{\partial}{\partial t} \left[ \left( \frac{T_e}{T_n} \right)^{3/2} N_e \langle \sigma' \rangle \right]. \quad (7)$$

The numerical constant that appears when we express  $H$  in terms of  $T_e$  and  $T_n$  has been absorbed in  $\langle \sigma' \rangle$ , in fact,  $\langle \sigma' \rangle = 6(\gamma - 1)(3/\gamma)^{1/2} \langle \sigma \rangle$ .

Having obtained the expression for the source term in the wave equation for the acoustic pressure in the neutral gas, we can calculate the sound-pressure field in terms of the space-time dependence of the electron temperature and the electron density if these quantities are assumed to be known *a priori*; this is considered to be the case in the following two specific problems to be analyzed. Later, in the discussion of wave amplification and spontaneous oscillations the effect of the neutral density fluctuation on the electron density will be accounted for.

## TRAVELING STRIATIONS

It is well known that an ordinary glow discharge frequently contains traveling striations, that is, self-sustained wavelike periodic perturbations in the electron density, electron temperature, and electric field. They usually travel in the direction from the anode to the cathode with a velocity  $V$  of approximately  $10^4$  cm/sec. The frequency of oscillation typically is in the range  $10^3$  to  $5 \times 10^4$  sec $^{-1}$ . Under such conditions, the electron density  $N_e$  and the electron temperature  $T_e$  in the discharge tube can be expressed as  $N_e = N_{e0}[1 + \epsilon_1 \sin \omega(t - x/V)]$  and  $T_e = T_{e0}[1 + \epsilon_2 \sin \omega(t - x/V)]$ , where  $x$  is the coordinate along the axis of the tube. For simplicity, we shall consider a somewhat idealized geometry of the plasma such that the unperturbed values  $N_{e0}$  and  $T_{e0}$  of the electron density and electron temperature are both constant along the tube in the interval  $-L < x < L$  and zero outside this region, with only the neutral-gas component present. If we consider coupling only to one-dimensional acoustic waves traveling along the axis of the discharge tube, only the spatial averages of  $T_{e0}$  and  $N_{e0}$  across the tube are relevant.

Assuming  $\epsilon \ll 1$ , we shall retain terms only of first order in  $\epsilon$  in the source term (7). Furthermore, we neglect the perturbation in the scattering cross section produced by the perturbation in the electron temperature. The source term in (5) then is a constant times  $\cos \omega[t - (x/V)]$  for  $-L < x < L$  and zero for  $|x| > L$ . The corresponding solution to the wave equation is

standard, and we find that the amplitude of the sound-pressure wave emitted from the plasma into the region  $|x| > L$  outside the plasma is

$$p = p_0(\epsilon_1 + \frac{3}{2}\epsilon_2) \sin(\omega L/c)(c/V \mp 1) / (\omega L/c)(c/V \mp 1), \quad (8)$$

where  $c$  is the speed of sound, and the characteristic sound pressure  $p_0$  is given by

$$p_0/P_0 = (L/l_e)(m_e/m_n)^{1/2}(T_{e0}/T_n)^{3/2}(N_{e0}/N_n). \quad (9)$$

As before,  $P_0$  is the unperturbed gas pressure and  $l_e = 1/N_n \langle \sigma' \rangle$  is of the order of the electron mean-free path. The minus and plus signs in (8) refer to the acoustic waves emitted in the positive and negative  $x$  directions, respectively. The amplitude of the wave that travels in the same direction as the striations (positive  $x$  direction) has a maximum value  $p_{\max} = p_0(\epsilon_1 + 3\epsilon_2/2)$  when the speed  $V$  of the striations is coincident with the speed of sound  $c$  in the neutral gas. It is interesting to note that the ratio between the characteristic sound pressure  $p_0$  and the static gas pressure  $P_0$  increases with decreasing gas temperature as  $T_n^{-3/2}$ . In other words, if  $T_{e0}$  and  $N_{e0}$  are held constant, the fractional value of the perturbation in the pressure of the neutral gas can be approximately two orders of magnitude greater if the plasma is brought from room temperature to cryogenic surroundings.<sup>8</sup>

To get an idea of the order of magnitude of the sound-pressure amplitudes obtained, we consider a He plasma at a pressure  $\approx 1$  mm Hg and  $N_{e0}/N_n \approx 10^{-6}$ . With an electron temperature  $\approx 1$  eV, we have  $T_{e0}/T_n \approx 40$ , and with  $\langle \sigma \rangle \approx 6 \times 10^{-16}$  cm<sup>2</sup> the corresponding electron mean-free path is  $l_e \approx 0.05$  cm. Then, assuming  $\epsilon_1 = \epsilon_2 \approx 0.1$  and  $L \approx 10$  cm, we find the largest possible value of the sound-pressure amplitude for  $V=c$  to be approximately  $10^{-3}$  times the static pressure, that is,  $\approx 1$  dyn/cm<sup>2</sup>. If under the same electrical operating conditions, the neutral-gas temperature is lowered, say, to  $10^\circ\text{K}$ , the possible maximum sound-pressure amplitude would be approximately 0.1 times the static pressure.

### PLASMA AFTERGLOW

As a second example, we choose the problem of the modulation of plasma afterglow as observed by Berlande, Goldan, and Goldstein.<sup>2</sup> A rectangular discharge tube was used in their experiments, and the observed periodic modulation of the decay of the plasma was believed to be a result of acoustic oscillations in the direction transverse to the tube axis. In the analysis of the excitation of such waves, we let the tube axis be along the  $z$  axis and the tube walls be parallel with the  $x$ - $z$  and  $y$ - $z$  planes. We will consider the normal modes

<sup>8</sup> It should be borne in mind that for a given neutral particle density the static pressure is proportional to the temperature. Therefore, if the relative pressure change depends on temperature as  $T_n^{-3/2}$ , the sound pressure itself will change with temperature as  $T_n^{-1/2}$ .

corresponding to motion in the  $y$  direction. The  $y$  dependence of  $T_{e0}$  and  $N_{e0}$  then plays an important role, and we express this  $y$  dependence in terms of a Fourier series in which the first (and dominant) harmonic term is of the form  $\sin(\pi y/D)$ , the reflecting tube walls being located at  $y=0$  and  $y=D$ . Since we are interested only in the oscillations in the  $y$  direction, we may consider  $T_{e0}$  and  $N_{e0}$  to be independent of the two remaining coordinates.

As a plasma is turned off, the electron temperature quickly decreases to the value of the surrounding gas temperature. The decay in the electron density is less rapid and, as far as sound generation is concerned, of minor importance. Thus let the time dependence of the electron temperature be expressed as  $T_{e0} \exp(-bt)$  for  $t > 0$  and  $T_{e0}$  for  $t < 0$ . The source term (7) in the wave equation will then contain a time dependence expressed by  $\exp(-1.5bt)$ . If, for simplicity, only the first Fourier component  $\sin(\pi y/D)$  of the Fourier expansion of  $(T_{e0}^{3/2} N_{e0})$  is considered, the wave equation (5) will be of the form

$$\frac{1}{c^2} \frac{\partial^2 p}{\partial t^2} - \nabla^2 p = \left[ -\frac{3}{2} b P_0 \left( \frac{m_e}{m_n} \right)^{1/2} \left( \frac{T_{e0}}{T_n} \right)^{3/2} N_{e0} \langle \sigma' \rangle \right] e^{-3bt/2} \sin(\pi y/D),$$

in which the brackets indicate the amplitude of the first Fourier component of the quantity inside the brackets; it differs but little from the average value of this quantity over the cross section of the tube, and we shall not distinguish between them in the sequel. With this source term used, the solution to the wave equation will be a sum of normal modes symmetrical with respect to the axis of the tube. The pressure amplitude of the  $m$ th symmetrical mode is found to be, assuming  $b \gg c/D$ ,<sup>9</sup>

$$p_m = 2p_0/\pi^2 m(4m^2 - 1), \quad (m=1, 2, \dots). \quad (10)$$

The characteristic pressure  $p_0$  is the same characteristic pressure that was defined in (9), with  $L$  replaced by  $D$ .

As will be discussed further in the next section, the modulation of the neutral-gas density caused by the sound wave produces a modulation of the electron density such that the fractional changes in both are the same. The relative modulation of the intensity of a microwave signal transmitted through the decaying plasma in turn depends on the relative change of the density and hence on the relative change in the pressure produced by the sound wave. Under the experimental conditions of Berlande, Goldan, and Goldstein, the relative change in the transmitted microwave intensity should be of the same order as the relative change in sound pressure. As in the case of traveling

<sup>9</sup> This assumption is equivalent to a steplike change of  $H$  and hence a delta-function source in time in Eq. (5).

striations this relative pressure change is proportional to  $(T_{e0}/T_n)^{3/2}(N_{e0}/N_n)$  as given in Eq. (9). At room temperature and under ordinary glow-discharge conditions, the modulation is small and would be difficult to observe, at least for tubes of the order of 1-cm cross dimension or smaller. However, if for a given value of  $T_e$  the neutral gas temperature is decreased, for example, by placing the plasma in a cryogenic environment, the relative pressure change can be large enough, say, 10%, to account for the observed modulation. Furthermore, as discussed in the Appendix, low temperatures are required also to make the decay rate of the acoustic modes sufficiently small. These observations are consistent with the experiments of Berlande, Goldan, and Goldstein, who encountered the modulation effect in the afterglow in their work with cryogenic plasmas. Another feature of the experimental observations that is consistent with (10) is that only the fundamental ( $m=1$ ) mode was observed. From (10) it follows that the amplitudes of oscillation decrease approximately as  $m^{-3}$  so that higher mode contributions would be small compared with the fundamental.

#### ACOUSTIC WAVE AMPLIFICATION

Thus far, we have considered the perturbations in electron temperature and electron density to be given functions and independent of the acoustic-pressure fluctuations in the neutral gas. But, to discuss the possibility of spontaneous excitation of acoustic waves, it will be necessary to investigate how the source term might depend on the acoustic field itself. Without going into the formidable analysis of the general dynamics of, say, a three-fluid model of the weakly ionized gas, it is sufficient in this context to point out that in the ordinary acoustic mode of motion the neutrals, electrons, and ions all move in phase with each other, and the relative density fluctuations in all three components are the same.<sup>10</sup> This statement is valid at frequencies of interest here, smaller than the plasma frequency and the neutral-neutral collision frequency. Then a perturbation  $n_n$  of the neutral particle density will produce a perturbation  $n_e$  in the electron density such that

$$n_e = (N_e/N_n)n_n.$$

We have already seen that the source term in the acoustic wave equation is proportional to the total electron density. In the presence of a sound wave that produces a neutral density perturbation  $n_n$ , the total electron density is of the form  $N_e + n_e = N_e + (N_e/N_n)n_n$ . Consequently, a sound wave in the plasma produces a spatial source distribution in the medium with a strength proportional to the local amplitude of the sound wave. The phase relationship between the fluctuation in density in the sound wave and the acoustic source strength turns out to be such as to produce wave

growth. To study this quantitatively we replace  $N_e$  in the source term (7) by  $N_e + (N_e/N_n)n_n$  and express  $n_n$  in terms of the sound pressure  $p = c^2 m_n n_n = (\gamma n_n/N_n)P_0$ . We find that (5) will be of the form

$$\frac{1}{c^2} \frac{\partial^2 p}{\partial t^2} - \nabla^2 p = \frac{\partial A(t)}{\partial t} + B(t) \frac{\partial p}{\partial t}, \quad (11)$$

where

$$\begin{aligned} B(t) &= (1/\gamma c)(T_e/T_n)^{3/2}(m_e/m_n)^{1/2}N_e\langle\sigma'\rangle, \\ A(t) &= \gamma P_0 B(t). \end{aligned} \quad (12)$$

$T_e$  and  $N_e$  now represent the values of electron temperature and particle density in the absence of a sound field, and they may well be functions of time. In the foregoing discussion of generation of waves this time dependence, corresponding to the term  $\partial A/\partial t$  in (11), was essential. Since we are now interested in the transmission of an acoustic wave through a quiescent plasma,  $A$  and  $B$  are time-independent and therefore  $\partial A/\partial t = 0$ . We shall consider only the behavior of a plane traveling wave. The analysis of other motions applicable to the normal modes in various discharge tubes adds nothing essential. Thus, with  $p \sim \exp(ikx - i\omega t)$  we obtain, from (11) (with  $\partial A/\partial t = 0$ ), the dispersion relation

$$\kappa = (\omega/c)(1 - i/\omega\tau)^{1/2} \equiv \alpha - i\beta, \quad (13)$$

where  $(1/\tau) = c^2 B$ . The expressions for  $\alpha$  and  $\beta$  are:

$$\begin{aligned} \alpha &= \frac{(\omega/c)}{\sqrt{2}} \{ [1 + (\omega\tau)^{-2}]^{1/2} + 1 \}^{1/2} \\ &\simeq (\omega B/2)^{1/2} \quad (\omega\tau \ll 1) \\ &\simeq (\omega/c) [1 + \frac{1}{2}(\omega\tau)^{-2}], \quad (\omega\tau \gg 1) \\ \beta &= \frac{(\omega/c)}{\sqrt{2}} \{ [1 + (\omega\tau)^{-2}]^{1/2} - 1 \}^{1/2} \\ &\simeq (\omega B/2)^{1/2} = \beta_1 \quad (\omega\tau \ll 1) \\ &\simeq 1/2c\tau = \beta_2 \quad (\omega\tau \gg 1). \end{aligned} \quad (14)$$

We see that there is a spatial growth of the wave at a rate  $\beta$ , which, for  $\omega \ll (1/\tau)$ , increases with frequency as  $\sqrt{\omega}$ , and in the limit of high frequencies approaches a constant value  $\beta_2$ .

In obtaining these results, we have neglected the attenuation of the acoustic wave resulting from various loss mechanisms. When sufficiently small, the growth and the attenuation rates are additive so that we can treat them separately. For a monatomic gas the acoustic losses are caused solely by viscosity and heat conduction. Normally, the plasma is contained in a tube, and we must consider the attenuation caused by the dissipation at the tube walls, as well as in the bulk of the gas. The corresponding well-known Kirchhoff attenuation in

<sup>10</sup> U. Ingard and K. W. Gentle, Phys. Fluids 8, 1396 (1965).

a circular tube of diameter  $d$  can be expressed as<sup>11</sup>

$$\alpha_1 = \frac{(\omega/c)}{2d} \left[ \left( \frac{2\eta}{\rho\omega} \right)^{1/2} + \left( \frac{2K}{\omega\rho c_p} \right)^{1/2} \right] \\ \simeq \frac{1}{d} [2(\omega/c)(\eta/\rho c)]^{1/2}, \quad (15)$$

$$\alpha_2 = \frac{1}{2} (\omega/c)^2 \left[ \frac{4}{3} \frac{\eta}{\rho c} + (\gamma-1) \frac{K}{\rho c_p c} \right] \\ \simeq 1.16 (\omega/c)^2 (\eta/\rho c),$$

where  $K$  is the heat-conduction coefficient, and  $\eta$  is the coefficient of viscosity. In obtaining the approximate expressions in (15), we have set  $\eta \simeq 0.67 (K/c_p)$ , which is approximately valid for a monatomic gas.

At low frequencies the attenuation and growth rates both have the same frequency dependence,  $\sqrt{\omega}$ . At high frequencies, on the other hand, the attenuation  $\alpha_2$  increases with frequency as  $\omega^2$ , whereas the growth rate levels off to a constant value  $\beta_2$ . Consequently, it is clear that above a certain frequency the attenuation will predominate,  $\alpha_2 > \beta_2$ , and a net amplification is not possible. Whether or not an amplification is possible at lower frequencies depends on the relative magnitude of  $\beta_1$  and  $\alpha_1$ . Since they both are proportional to  $\sqrt{\omega}$ , it follows from (12), (14), and (15), using the approximate expression for  $\alpha_1$  in (15), that the necessary criterion for wave amplification,  $\beta_1 > \alpha_1$ , can be expressed as

$$(1/4\gamma)(d^2/l_n l_e)(T_e/T_n)^{3/2}(m_e/m_n)^{1/2}(N_e/N_n) > 1, \quad (16)$$

where  $l_n = (\eta/\rho c)$  and  $l_e = 1/N_n(\sigma')$  are of the order of the mean-free path of the neutral particles and of the electrons, respectively. If this criterion is fulfilled, the wave growth is ensured in a frequency range from zero to an upper limit determined by the relationship between the high-frequency value  $\beta_2$  for the growth rate [see Eq. (14)] and the corresponding attenuation. For example, if bulk rather than boundary attenuation is predominant, the upper frequency is obtained by equating  $\beta_2$  and  $\alpha_2$ .

To study the possibility of *spontaneous excitations* of acoustic oscillations, we again consider the modes between the two rigid parallel walls in a rectangular tube as in the previous discussions of the plasma afterglow. The pressure field of a particular mode is then  $\cos(m\pi y/D) \exp(-i\omega_n t)$ , and we obtain from (11), with  $\partial A/\partial t = 0$ ,

$$\omega_m = (cm\pi/D)[1 - (D/2\pi m c \tau)^2]^{1/2} + i/2\tau. \quad (17)$$

As before,  $\tau = (1/Bc^2)$ , where  $B$  is given in (12).

Under the conditions considered here, we have  $(c\tau/D) \gg 1$  and (17) implies a growth in time at a rate

given by  $(1/2\tau)$ . However, to obtain a criterion for the onset of spontaneous oscillations, we must compare the growth rate with the decay caused by acoustic losses in the gas. The decay constant can be expressed as  $\delta/T_1$ , where  $T_1$  is the fundamental period of oscillation, and  $\delta$  is a quantity that depends on the viscosity and heat conduction and other plasma parameters, as discussed in the Appendix. The criterion for spontaneous onset of oscillations, that the decay rate be smaller than the growth rate,  $(\delta/T_1) < (1/2\tau)$ , leads to a criterion of the same form as (16) except for a numerical constant. The value of this constant depends on the particular mode considered and, of course, on the geometry of the discharge chamber. The discussions here are all related to one-dimensional wave motions, but the criterion (16) should be qualitatively applicable also to other waves. For example, we note that for a given tube geometry and for given values of  $(T_e/T_n)$  and  $(N_e/N_n)$  the pressure dependence of the threshold of spontaneous oscillations is expressed by the lengths  $l_n$  and  $l_e$ , which are approximately the neutral and electron mean-free paths, respectively. Under such conditions a critical pressure exists above which spontaneous oscillations should occur. This may explain the fact that Alexeff and Neidigh observed an acoustic wave instability in their spherical discharge chamber at a comparatively high pressure ( $\simeq 1$  mm Hg) but not at lower pressures.

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#### APPENDIX

In the discussion of both the plasma afterglow and the spontaneous excitation of acoustic eigenmodes in an enclosure, results concerning the decay of these modes were used which will be commented upon here. If the gas is monatomic, the losses are due to viscosity and heat conduction, and the calculation of the decay rate can be carried out in much the same way as for the well-known spatial decay rate given in Eq. (15). If the mean-free path of the neutrals is small compared with the typical size of the discharge chamber, the viscous and thermal boundary layers in the acoustic field are also small compared with the chamber dimensions. The energy loss in the field is then conveniently separated into losses in the bulk of the gas outside the acoustic boundary layers and the losses in the layers.

In the following we will discuss the result of such a calculation for the lateral modes perpendicular to the axes of a square tube. We consider a tube with square cross section, the length of the side being  $D$ , and a pressure mode of the form  $\cos(m\pi y/D) \exp(-i\omega_n t)$ , the tube walls being at  $y=0$  and  $y=D$ . Then, if we use the approximate relation  $K/\eta c_p \simeq 0.67$  for a monatomic gas, the numerical value for the decay time  $\tau_d$ , corresponding

<sup>11</sup> P. M. Morse and U. Ingard, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1961), Vol. XI, Chap. 1, pp. 14 and 22.

to the rate  $\exp(-t/\tau_d)$ , is found to be

$$\tau_d \simeq T_1 [26(n^2 l_n / D) + 8(n l_n / D)^{1/2}]^{-1} \equiv T_1 / \delta.$$

Here  $T_1 = 2D/C$  is the period of the fundamental mode,  $l_n = \eta/\rho c$  is of the order of the mean-free path,  $n$  is the mode number ( $n=1$  being the fundamental), and  $D$  is the length of the side of the square tube considered. The first term in the brackets comes from the losses outside the boundary layer and the second term from the losses inside the layer.

The quantity  $l_n = \eta/\rho c$  depends on both the density and the temperature of the gas. At room temperature and a pressure of approximately 1 mm Hg, we have for He,  $l_n \simeq 10^{-2}$  cm. Then, if  $D \simeq 1$  cm, we obtain for the first mode  $n=1$ ,  $\tau_d \simeq T_1$ . Under these conditions the decay of the acoustic waves will be quite rapid, and it would be very difficult to observe acoustic resonances in a plasma at room temperature and pressure  $\approx 1$  mm Hg or less. This may explain why the acoustic resonances

described by Strickler and Stewart were observed only at comparatively high pressures ( $\simeq 13$  mm Hg), in which case we get  $\tau_d \simeq 5 T_1$ .

In the afterglow experiments of Berlande, Goldan, and Goldstein, the number density of the neutrals was  $N_n \simeq 7 \times 10^{17}$  cm $^{-3}$ . The plasma was submerged in a bath with a temperature of 4.2°K, and at the corresponding low gas temperature in the discharge the coefficient of viscosity is estimated to be lower than the value at room temperature ( $\eta \simeq 1.9 \times 10^{-4}$  cgs units) by more than a factor of ten (using, say, Sutherland's formula<sup>12</sup>). Therefore  $l_n = \eta/\rho c$  will be less than  $10^{-4}$  cm. The corresponding value of the decay time is then found to be  $\tau_d \geq 10 T_1$ , which is consistent with the experimental observations.

<sup>12</sup> J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird, *Molecular Theory of Gases and Liquids* (John Wiley & Sons, Inc., New York, 1954), p. 565.

## Nonperturbative Proof of Some Results in Classical Nonequilibrium Statistical Mechanics

RAPHAEL ARONSON

*Department of Nuclear Engineering, New York University, New York, New York*

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Some results of interest in classical nonequilibrium statistical mechanics, previously proved to all orders in the interaction, are proved without recourse to perturbation expansions, making it possible to avoid convergence questions. One theorem so proved is that when the interaction is switched on slowly, a Maxwellian distribution goes into the canonical distribution at the same temperature. The two steps in the Prigogine theory of the approach to equilibrium that originally depended on a perturbation proof are also demonstrated nonperturbatively. Lastly, the statement that in equilibrium cluster expansions in the density there are no articulation points in the graphs contributing to the reduced distributions is proved from a time-dependent point of view.

### I. INTRODUCTION

IN classical-statistical mechanics, as in quantum-statistical mechanics, perturbation theory has been a convenient tool for investigations into nonequilibrium and transport problems. It has been possible in some cases to sum perturbation series in the interaction strength to all orders to obtain sensible results. However, because it is extremely difficult even for finite interactions to prove convergence, it is useful to avoid perturbation expansions when possible. A procedure which treats the time development of the classical distribution function nonperturbatively is developed here. It is used to justify certain results originally obtained by perturbation methods. The approach is formally quite similar to time-dependent scattering theory.

Two problems, the calculation of thermal averages and the approach to equilibrium, are reconsidered. It

has been shown that it is convenient to compute thermal averages of many-time quantities by introducing an interaction picture.<sup>1</sup> The distribution function can be regarded as resulting under the mechanical motion of the system out of an initial unperturbed distribution function when the interaction is turned on slowly.<sup>2</sup> If the distribution function is canonical, the initial distribution function before the interaction is switched on is Maxwellian at the same temperature, and conversely. A proof not involving perturbation theory is given here as an alternative to the perturbation proof given previously.<sup>1</sup>

A second application is to the Prigogine treatment of the approach to equilibrium. The theory, as developed

<sup>1</sup> R. Aronson, *J. Math. Phys.* **7**, 221 (1966).

<sup>2</sup> The term "slowly" means the same as the more commonly used "adiabatically," which we avoid to eliminate possible confusion.