X. PHYSICAL ACOUSTICS*

Academic and Research Staff

Prof. K. U. Ingard
Prof. L. W. Dean III

Graduate Students

R. H. Katyl
C. Krischer
W. M. Manheimer

A. POSSIBILITY OF RADIATION-INDUCED SOUND AMPLIFICATION IN A GAS

The possibility of observing acoustic-wave amplification made possible through the heating of neutral particles by electrons in a weakly ionized gas has received some recent attention.[1-4] The idea is that an existing sound wave, by inducing a space-time variation in the electron density, would create an acoustic source, thereby generating a wave that interferes constructively with the original sound wave. At moderately high sound frequencies ($\omega \gtrsim 10^4$ sec^{-1}) the electrons behave isothermally under typical discharge conditions. Then, if acoustic losses resulting from viscosity and heat conduction in the neutral gas are ignored, the resulting exponential growth in the wave intensity in the linear theory is characterized by a time constant $\tau' = \tau(1+x^2)$. The parameter τ (~1 sec under typical discharge conditions) represents a time of the order of that required for the gas to absorb from the electrons an amount of energy equal to the thermal energy of the gas, while x is the tangent of the phase angle between relative density perturbations in the electrons and neutrals.[4] An analogous calculation for ion waves in a fully ionized gas shows only a very slight wave amplification.[5] Here, also, there is no guarantee that the gain would offset the expected losses. In fact, one of the principal difficulties in observing energy-transfer wave amplification is the considerable loss rate associated with the relatively low-pressure gas required to obtain an electrical discharge.

This difficulty could be overcome if the energy to be transferred into the translational modes of the gas could be carried internally in the gas molecules themselves. Discharge conditions would not then be required, and one could increase the gas pressure so as to reduce ordinary acoustic losses in the completely un-ionized gas. By carrying the energy internally, moreover, the gas molecules would eliminate the phase difference[4] between relative density perturbations in the gas component that is responsible for heating and the component that is being heated through collisions with the former. The purpose of

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the present report is to point out the existence of this kind of acoustic-instability mechanism in a high-pressure gas, completely un-ionized, having internal modes, for example, molecular rotational or vibrational states or atomic-electron states, excited selectively by an external radiation field so as to be out of thermal equilibrium with the translational modes of the gas. The possibility of nonradiative decay through atomic or molecular collisions in such a gas constitutes a positive-feedback condition for an acoustic wave in the gas, since the decay energy would go into the translational modes and the rate of energy transfer would be proportional to the collision frequency. The result could be either the spatial amplification of a wave propagating in a relatively lossless high-pressure gas or the spontaneous oscillation of a high-Q acoustic cavity.

The level separation in such a gas is large enough that at room temperature, or at least at temperatures easily attained by cooling the gas, nearly all of the molecules are in the ground state under conditions of thermal equilibrium, that is, they are not in excited rotational, vibrational or electronic states. Intense illumination of the gas, by electromagnetic radiation having a narrow bandwidth about a transition frequency, for example, a line of the rotational, vibrational or electronic spectrum, would lead to a nonequilibrium distribution of the gas molecules among the energy levels of the internal mode, or to an internal temperature, which we hope may be much larger than the temperature associated with the translational modes.

The effect can be imagined as occurring in a one-dimensional electromagnetic resonant cavity driven externally, for instance, by a large number of parallel laser beams, so as to possess an energy-density spectrum sufficiently peaked at the transition frequency that a uniform nonthermal fraction f of the molecules (or atoms) are in the excited state. For this to be true it is necessary that a random molecule (or atom) be much more likely to interact with a photon from the radiation field than with another molecule (or atom). Thus, the radiation field must have, on the average, an energy-flux density $J \gg \omega_{nn} h\nu/\sigma$, where ω_{nn} is the collision frequency, h is Planck's constant, $h\nu$ is the transition energy for the internal mode, and σ is the cross section for the absorption of a photon at that energy. Under these conditions, the time required for the translational modes of the gas to absorb their own thermal energy from the radiatively excited internal modes is of the order of $\tau \approx m_n c_n^2/h\nu f\omega_{nn}$, where m_n is the mass of a gas molecule, and c_n is the speed of sound. Within a factor of order unity, this time constant would also characterize the growth of the intensity of a sound wave in the absence of acoustic losses.

H. M. Schulz III

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B. LIGHT SCATTERING OFF SURFACE WAVE PHONONS

The dynamics of surface waves on liquids can now be measured in the wavelength region of $\sim 10^{-5}$ cm by light-scattering techniques.[1] In this wavelength region the damping of the waves is quite high and first-order theories of surface waves cannot be used. We shall outline the exact solution to the surface wave problem,[2] using a linearized theory, and use this solution to interpret some experimental results.

We work here with an incompressible fluid of mass density ρ, surface tension σ, and shear viscosity μ. The liquid surface is taken to be the x-y plane, with the z-axis extending out of the fluid. The velocity field is $\vec{u}(r,t)$ and the surface displacement $\zeta(x,y,t)$. Following Lamb,[3] the boundary conditions required of the stress tensor T_{ij} at the surface $z = 0$ are

$$T_{zz} = -\sigma\left(\frac{\partial^2\zeta}{\partial x^2} + \frac{\partial^2\zeta}{\partial y^2}\right) \tag{1}$$

$$T_{xz} = T_{yz} = 0, \tag{2}$$

where T_{ij} is defined as $T_{ij} = p\delta_{ij} - \mu\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right) + \frac{2}{3}\mu\delta_{ij}(\vec{\nabla}\cdot\vec{u})$.

We now resolve the velocity field u into its independent longitudinal and transverse parts by $\vec{u} = -\vec{\nabla}\phi + \vec{\nabla}\times\vec{b}$, where $\vec{\nabla}\cdot\vec{b} = 0$ specifies the gauge. With this replacement for u, the linearized equation of motion, $\rho\partial u_i/\partial t = -\partial T_{ij}/\partial x_j$, is separated into two equations. With the use of the incompressibility condition, $\vec{\nabla}\cdot\vec{u} = 0$, these are

$$\rho\frac{\partial\phi}{\partial t} = p \tag{3}$$

$$\rho\frac{\partial\vec{b}}{\partial t} = \mu\nabla^2\vec{b}. \tag{4}$$

To simplify the problem, we look for the motion of a one-dimensional disturbance of the form

$$\phi = \phi_o e^{at} e^{iKx} e^{Kz} \tag{5}$$

$$\vec{b} = \vec{b}_o e^{at} e^{iKx} e^{\beta z}. \tag{6}$$

Here, ϕ satisfies $\nabla^2\phi = 0$, and (4) requires $\beta^2 = K^2 + \rho a/\mu$. For this one-dimensional wave $u_y = 0$, and the velocity field becomes

$$\vec{u} = \begin{bmatrix} u_x \\ 0 \\ u_z \end{bmatrix} = \left\{ -\phi_o e^{Kz} \begin{bmatrix} iK \\ 0 \\ K \end{bmatrix} + b_o e^{\beta z} \begin{bmatrix} -\beta \\ 0 \\ iK \end{bmatrix} \right\} e^{iKx} e^{at}. \tag{7}$$

With this form of $\vec{u}$, condition (2) results in

$$\frac{ib_o}{\phi_o} = \frac{1}{1 + \dfrac{\rho a}{2\mu K^2}}, \tag{8}$$

and upon taking the time derivative of (1) and using (3), we obtain

$$\sigma \frac{\partial^2 u_z}{\partial x^2} = -\rho \frac{\partial^2 \phi}{\partial t^2} + 2 \frac{\partial^2 u_z}{\partial z \partial t}. \tag{9}$$

With the use of (7) and (8), this reduces to the following equation for a:

$$a^2 + \frac{2\mu K^2}{\rho} a + \left(\frac{2\mu K^2}{\rho}\right)^2 \left(1 + \frac{\rho a}{2\mu K^2} - \sqrt{1 + \frac{\rho a}{\mu K^2}}\right) + \frac{\sigma K^3}{\rho} = 0. \tag{10}$$

With a change of variables, this becomes

$$S^2 + 2S - \sqrt{1 + 2S} + 1 + y = 0, \tag{11}$$

where $S = a\tau_o$, $\tau_o = \rho/2\mu K^2$, $\omega_o = \sqrt{\sigma K^3/\rho}$, and $y = (\omega_o \tau_o)^2$. This is the determining equation for the time dependence of surface waves of a particular wavelength.

Equation 11 can be squared, and the resultant quartic equation solved either exactly or numerically. Of the four roots obtained in the solution for S(y) only two are consistent with the requirement $\beta > 0$. These are shown in Fig. X-1. For $y \leq 0.145$, both roots are real and negative, and for $y > 0.145$, the roots are complex conjugates with a negative real part. Thus, for $y \leq 0.145$, the decay of the disturbance is nonoscillatory; there are two modes of decay with differing time constants. But for $y > 0.145$, the decay is damped oscillatory.

This solution for the time dependence means that the spectrum of the scattered light will be split into a doublet, the so-called Brillouin doublet, for $y > 0.145$. The doublet frequency separation is $(S_i/\pi\tau_o)$, and the half-intensity width of each line is $(S_r/(\pi\tau_o))$.

But for $y \leq 0.145$, no splitting occurs; the spectrum consists of two unshifted components whose peak intensities are in inverse proportion to their widths.

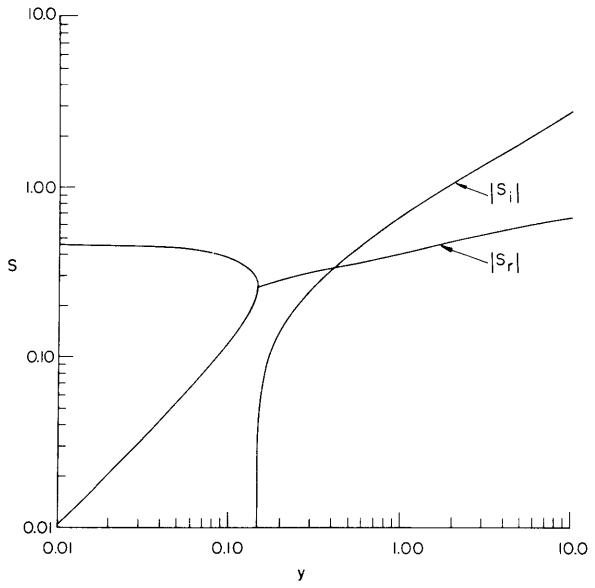

Fig. X-1. Solution of Eq. 11 for time dependence of surface waves. The wave propagates as $e^{iKx}e^{S(t/\tau_o)}$, $\tau_o = \rho/2\mu K^2$, $\omega = \sqrt{\sigma K^3/\rho}$, $y = (\omega_o \tau_o)^2$.

This absence of a Brillouin splitting under certain conditions can explain the results of an experiment that we have performed recently. Light-scattering measurements were made with two different liquids with conditions chosen so that a doublet was expected for the less viscous liquid (methanol), and nonoscillatory decay was expected for the other (isopropanol). Although the doublet could not be resolved, the spectrum for methanol was noticeably wider than for isopropanol (see Fig. X-2).

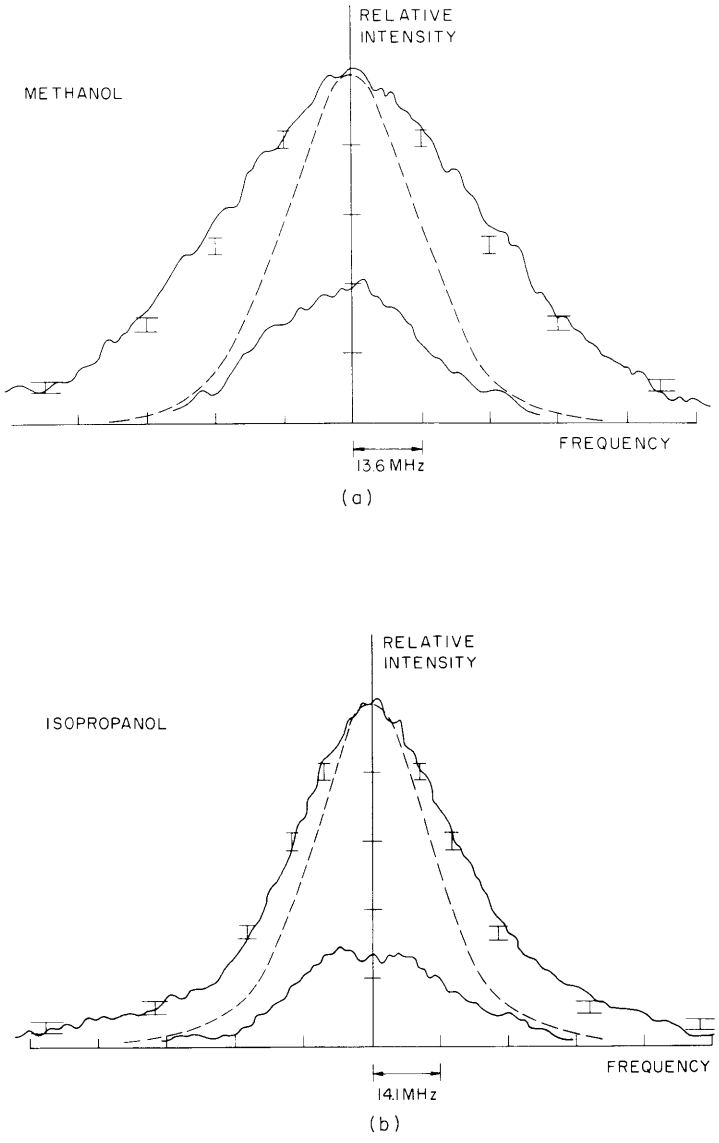

Fig. X-2. (a) Results for methanol. (b) Results for isopropanol. Upper solid curve: surface scattering; lower solid curve, scattering in bulk of fluid; dashed curve, instrument response; points (I), theoretical curve resulting from the solution of Eq. 11 when the effects of instrumental broadening and bulk scattering are included.

Note that the theory that we have outlined here does not consider the effects of any changes in structure of the liquid near the surface. Such effects can occur in polar or associated liquids over length scales approaching a light wavelength, and would be expected to greatly modify the scattered light spectrum. Higher resolution experiments, which are needed to examine this possibility, are now in progress.

R. H. Katyl, U. Ingard

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C. ION-DENSITY RESPONSE TO A RECTANGULAR PULSE OF SOUND IN A PLASMA

Recent investigations of the acoustic-wave mode in a weakly ionized gas[1-5] have shown this to be a slightly perturbed version of ordinary sound in the neutral-gas component. The charged particles are dragged along by the neutrals, but ambipolar diffusion in the direction of wave propagation causes a phase and amplitude difference between the relative density perturbations in the neutral gas and in the electrons or ions.[5] In the present report the frequency dependence of this phase and amplitude difference is applied to the calculation of the ion-density response to a rectangular pulse of sound in a weakly ionized gas. It is shown, in particular, that the wavefront in the ionic component is not sharp, but has a width in time inversely proportional to the neutral-to-electron temperature ratio and the ion-neutral collision frequency, a result qualitatively consistent with previously reported probe studies of the time dependence of the ion density during the passage of a weak shock wave.[6]

As has recently been shown,[5] the relative density perturbations (n_j/N_j) (the subscripts $j = n, i, e$ refer to the neutrals, ions, and electrons, respectively) in the various fluid components are given by

$$(n_1/N_i) \approx (n_e/N_e) \approx [1+i\omega(T_e/\gamma_n T_n \omega_{in})]^{-1} (n_n/N_n) \qquad (1)$$

for a sound wave of frequency $\omega/2\pi$, where T is the temperature, ω_{in} is (essentially) the ion-neutral collision frequency, and γ_n is the specific-heat ratio (1.67 for a monatomic gas). The initial condition for a small-amplitude rectangular pulse in the three-component fluid undoubtedly depends on the method of generation, and is satisfied by a

superposition of Fourier spectra of the various density and entropy waves that can exist in a weakly ionized gas. Of these, however, the only mode that really propagates for $\omega \lesssim 10^{-6}$ sec^{-1} is the acoustic mode; all the others are strongly damped.[1,2] Since the Fourier spectrum of a rectangular pulse is dominated by the low-frequency components, an accurate description of the pulse for times $t \gtrsim 10^{-6}$ sec after generation is achieved by considering the acoustic Fourier components only.

Under the simplifying assumptions that the plane pulse generated at $z = 0$ and traveling in the $+z$ direction is of small amplitude and substantially free from ordinary acoustic losses such as viscosity and heat conduction, the rectangular pulse shape thereby being maintained for all times of experimental interest, the relative density perturbation in the neutral gas can be represented by $(n_n/N_n) = \epsilon\theta(t-z/c)$, where $\theta(x) = 1$ for $x \geq 0$, and $\theta(x) = 0$ for $x < 0$. The corresponding relative ion-density perturbation for times of experimental interest is given by

$$\frac{n_i(z,t)}{N_i} = \frac{\epsilon}{2\pi}\int_{-\infty}^{+\infty}\int_{0}^{+\infty}\frac{\exp(-i\omega t'+i\omega z/c)\,dt'd\omega}{1+i\omega(T_e/\gamma_n\omega_{in})}$$

$$= \epsilon\begin{cases}\exp[(\gamma_n T_n\omega_{in}/T_e)(t-z/c)], & t < z/c\\ 1 & t > z/c\end{cases} \qquad (2)$$

Thus, the leading edge of the pulse traveling in the neutral gas is preceded by a disturbance in the charged particles. This disturbance, caused by ambipolar diffusion of charged particles ahead of the wavefront in the neutral gas, is characterized by a time $\tau = T_e/\gamma_n T_n\omega_{in} \sim 10^{-4} - 10^{-3}$ sec, and so the wavefront associated with the ion density is not at all sharply defined, even for a perfectly sharp wavefront in the neutral gas.[7]

This lack of sharpness in the ion-density wavefront is consistent with a waveform obtained in recent experiments designed to measure the speed of a weak, spark-generated shock wave in an argon discharge.[6] The structure of the wavefront was not then of interest, and no comment was made on it, although the experimental conditions should have produced a sharp wavefront in the neutral gas. In particular, the previous experiments did not firmly establish whether the disturbance in the charged particles trails the wavefront in the neutral gas or leads it, as predicted here. This matter can be decided only by future experiments specifically designed to resolve the question.

U. Ingard, H. M. Schulz III

Footnotes and References

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7. It might be mentioned parenthetically that the total number of ions and electrons is conserved by an appropriate disturbance preceding the trailing edge of a real pulse. This is not of essence in the present discussion, since one may imagine a pulse of duration much longer than τ.