

Coupling Between Rayleigh and Brillouin Scattering in a Disparate-Mass Gas Mixture*

W. S. Gornall,† C. S. Wang, C. C. Yang, and N. Bloembergen

Gordon McKay Laboratory, Harvard University, Cambridge, Massachusetts 02138

(Received 11 March 1971)

Brillouin and Rayleigh scattering have been studied in a gas mixture of SF₆ and He at various concentrations. A strong coupling exists between the concentration and pressure fluctuations in the mixture that produces a heavy damping of the thermal sound-wave excitations and prevents them from propagating adiabatically. Consequently, a fully coupled solution of the hydrodynamic equations is necessary to describe the spectrum accurately.

The spectrum of light scattered from thermal fluctuations provides a means of probing the low-lying excitations of a fluid system. Theoretical descriptions of light scattering from fluid mixtures have been presented by several authors,^{1,2} which indicate that the transport coefficients, particularly the binary diffusion coefficient D , can be readily obtained from the spectra. In each case it has been assumed that the coupling between the sound mode and modes associated with thermal and mass diffusion can be neglected. For liquid mixtures where this coupling is generally weak, the approximation is valid. The spectral contributions from individual modes are easy to identify and can be represented by Lorentzian line shapes. On this basis, the binary diffusion coefficients in some liquid mixtures have been accurately determined.³ However, for gas mixtures at low pressure, but still within the hydrodynamic regime, D is typically three orders of magnitude larger than in liquids, and sound velocities are lower. Consequently, the coupling between the sound wave and the other hydrodynamic modes cannot be neglected.

In this Letter we report the first systematic study of thermal Brillouin scattering in a disparate-mass gas mixture in the hydrodynamic regime. It is found that strong coupling between hydrodynamic modes of the mixture causes a dramatic broadening of the Brillouin components and reduces their frequency shift by as much as 20% below that calculated from the adiabatic sound velocity.

It is well known^{1,2} that the intensity distribution of light scattered from a thermally fluctuating medium can be obtained from the dynamical structure factor

$$S(\vec{k}, \omega) = 2 \operatorname{Re}[\langle \epsilon(\vec{k}, z) \epsilon(-\vec{k}) \rangle_{z=i\omega}], \quad (1)$$

where \vec{k} is the momentum transfer in the scattering process and ω is the angular-frequency shift. The Fourier-Laplace transform of the correla-

tion function of the dielectric fluctuation can be expressed in terms of similar correlation functions of any complete set of dynamical variables N_i which describe the system, viz.,

$$\begin{aligned} \langle \epsilon(\vec{k}, z) \epsilon(-\vec{k}) \rangle \\ = \sum_{ij} \frac{\partial \epsilon}{\partial N_i} \frac{\partial \epsilon}{\partial N_j} \langle N_i(\vec{k}, z) N_j(-\vec{k}) \rangle. \end{aligned} \quad (2)$$

For a binary system one possible choice for the N_i is the pressure, temperature, and concentration (p, T, c).⁴

Following the methods of Kadanoff and Martin⁵ and Mountain,⁶ the Fourier-Laplace-transformed hydrodynamic equations can be written in matrix form¹:

$$M(\vec{k}, z) N(\vec{k}, z) = T(\vec{k}, z) N(\vec{k}), \quad (3)$$

from which the Fourier-Laplace transforms of the time-correlation functions for the dynamical variables can be expressed in terms of the equal-time correlation functions:

$$\begin{aligned} \langle N_i(\vec{k}, z) N_j(-\vec{k}) \rangle \\ = [\det M(\vec{k}, z)]^{-1} \sum_j P_{ij}(\vec{k}, z) \langle N_j(\vec{k}) N_i(-\vec{k}) \rangle. \end{aligned} \quad (4)$$

Here, $P_{ij}(\vec{k}, z)$ is the determinant of the matrix obtained from M by replacing the i th column with the j th column of T . The equal-time correlation functions $\langle N_j(\vec{k}) N_i(-\vec{k}) \rangle$ are obtained from the equipartition theorem.⁷

For dilute gas mixtures the transport coefficients can be accurately evaluated.⁸ The coupling constants $\partial \epsilon / \partial N_i$ between the electromagnetic field and the dynamical variables are also readily obtained.⁹ Thus, the above equations can be computer programmed to calculate the *exact hydrodynamic spectrum* for gas mixtures.

Mountain and Deutch¹ have derived an expression for $S(\vec{k}, \omega)$ from the approximate roots of $\det M(\vec{k}, z)$ in Eq. (4). Their analysis assumes that the sound-wave mode can be decoupled from

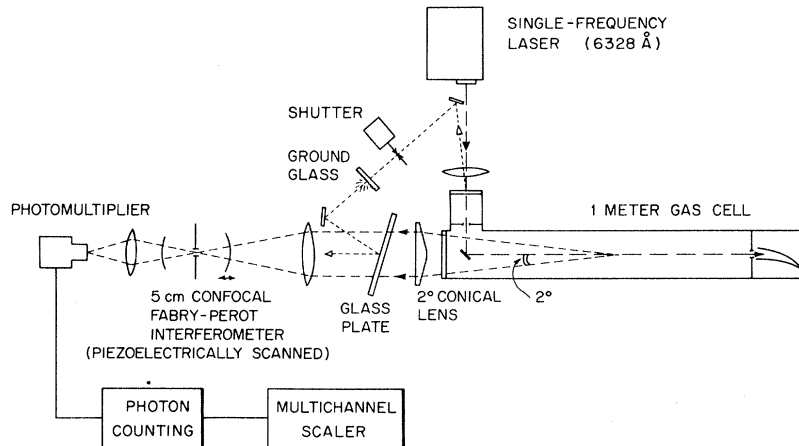


FIG. 1. Experimental arrangement for studying backward Rayleigh and Brillouin scattering from gases.

the other hydrodynamic modes and so gives rise to two Lorentzian-shaped Brillouin components at frequency shifts of $\omega = \pm v_s k$, where v_s is the adiabatic sound velocity. The full width at half-maximum of the Brillouin components is given by Kohler's expression.¹⁰

The experimental apparatus used to study the Brillouin scattering from gas mixtures is shown in Fig. 1. A stabilized single-frequency He-Ne laser (0.2 mW) was focused down the axis of a long pressure cell. Light scattered at an angle of 178° was collected by a 2° conical lens and analyzed using a piezoelectrically scanned, spherical Fabry-Perot interferometer. The spectrum was accumulated on a 1024-channel scaler. To compensate for thermal drift in the interferometer, each sweep of the scaler was triggered by the transmission of laser light (reflected from the cell window) through the interferometer. The trigger pulse also activated a shutter which blocked the triggering beam so that the spectrum was recorded for the remainder of the sweep. Fabry-Perot mirrors coated to 99% reflectivity provided an instrumental finesse of approximately 100 which did not change throughout the accumulation of many thousand scans. Typical spectra obtained for mixtures of SF_6 and He are shown in Fig. 2. Starting with 5 atm of SF_6 , He was added to obtain the He-number concentrations indicated. A full spectral free range is shown in each case, with the unshifted components of adjacent orders at either end. A small amount of stray-light intensity superimposed on the unshifted components is easily distinguished by its narrow (instrumental) linewidth.

With increasing He concentration the three components of the pure- SF_6 spectrum broaden

rapidly because of the increased damping caused by the lighter He-gas atoms. In addition, even at low He concentrations a broad background as-

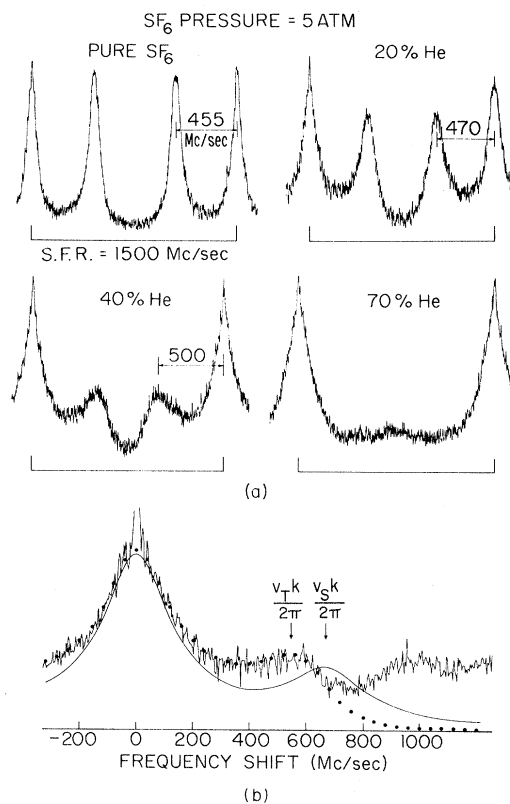


FIG. 2. Spectra scattered from mixtures of SF_6 and He for various He number concentrations. (a) One spectral free range is shown in each case with overlapping spectral orders. (b) The spectrum for 50% He concentration is compared with the uncoupled approximation of Ref. 1 (solid line) and the exact hydrodynamic spectrum (closed circles). The arrows indicate the shift corresponding to the adiabatic sound velocity v_s and the isothermal sound velocity v_T .

sociated with concentration fluctuations is evident centered about the exciting frequency. Its intensity increases dramatically with He concentration until at 70% it dominates the entire spectrum. The influence of this "concentration mode" on the rest of the spectrum is particularly striking at 50% concentration as shown in Fig. 2(b). The Brillouin component occurs at a much lower frequency shift than that corresponding to the adiabatic sound velocity (indicated by the arrow) or that derived from the spectrum of Mountain and Deutch [Eq. (3.18) of Ref. 1] based on the approximation that the sound wave is uncoupled.

The lowering of the Brillouin frequency occurs due to the strong coupling between the diffusive "concentration mode" and the sound wave. The observed Brillouin shift and that computed from the adiabatic and isothermal velocities of sound in the mixture are plotted as a function of He concentration in Fig. 3. It is apparent that with the addition of He the sound propagation ceases to be adiabatic and tends to become isothermal as a result of the high diffusion rate ($D \sim 0.1 \text{ cm}^2/\text{sec}$). Unlike the case for liquid mixtures considered in Ref. 1, in gas mixtures $Dk/v_s \sim 1$. Consequently, energy associated with pressure fluctuations can relax through mass diffusion in times on the order of the period of the sound wave. When this occurs the sound wave cannot propagate adiabatically but only isothermally. By the same mechanism the sound wave is heavily damped as predicted by Kohler.¹⁰ The experimental Brillouin linewidths showed good agreement with Kohler's result at low He concentrations where they could be measured.

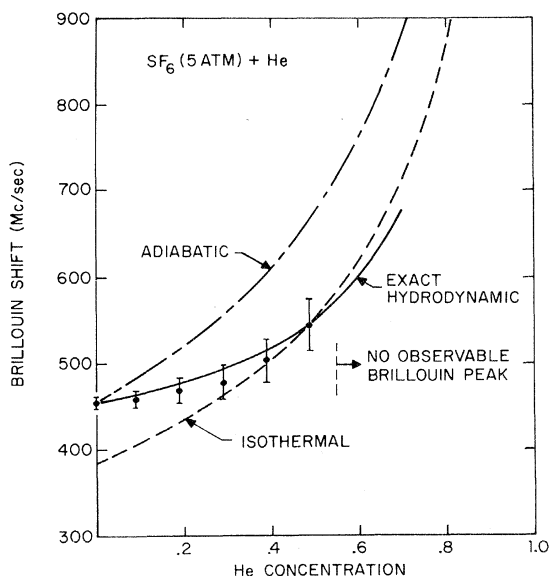


FIG. 3. Brillouin peak shift as a function of He concentration. The experimental points follow the solid curve determined from the calculated exact hydrodynamic spectrum. The dot-dashed and dashed curves correspond to adiabatic and isothermal sound velocities, respectively.

ically but only isothermally. By the same mechanism the sound wave is heavily damped as predicted by Kohler.¹⁰ The experimental Brillouin linewidths showed good agreement with Kohler's result at low He concentrations where they could be measured.

The *exact hydrodynamic spectrum* was also calculated for the SF_6 -He mixtures, taking into account the internal degrees of freedom of SF_6 and appropriate imperfect-gas corrections. One such spectrum (convoluted with the instrumental profile) is shown in Fig. 2(b). Similar agreement was found for all concentrations. Thus it is concluded that the mixture is in the hydrodynamic regime and is accurately described by a fully coupled solution of the hydrodynamic equations.

The original impetus for these experiments came from recent observations of stimulated scattering¹¹ from SF_6 and He mixtures. In turn, the more detailed information from these spontaneous scattering experiments can be used for a refined analysis of the stimulated scattering results, as will be discussed in a separate publication.

*Work supported jointly by the Joint Services Electronics Program under Contract No. 44-923-7309-2 and by Advanced Research Projects Agency Interdisciplinary Laboratories Program under Contract No. DAHC 15-67-C-0219.

†Holder of a National Research Council of Canada Postdoctoral Fellowship 1970-1971.

¹R. D. Mountain and J. M. Deutch, *J. Chem. Phys.* **50**, 1103 (1969).

²L. Blum, *J. Chem. Phys.* **50**, 17 (1969); see also P. C. Martin, in *Statistical Mechanics of Equilibrium and Nonequilibrium*, edited by J. Meixner (North Holland, Amsterdam, 1965), p. 122; P. C. Jordan and J. R. Jordan, *J. Chem. Phys.* **45**, 2492 (1966).

³P. Bergé, P. Calmettes, M. DuBois, and C. Laj, *Phys. Rev. Lett.* **24**, 89 (1970); M. DuBois and P. Bergé, *Phys. Rev. Lett.* **26**, 121 (1971); S. H. Chen and N. Polonsky-Ostrowsky, *Opt. Commun.* **1**, 64 (1969).

⁴Mountain and Deutch (Ref. 1) introduced a set of statistically independent variables (p, φ, c). If the spectrum is derived including all cross-correlation functions in Eq. (2), the choice of variables is not important. However, a set of statistically independent variables facilitates the calculation of an approximate analytical solution.

⁵L. P. Kadanoff and P. C. Martin, *Ann. Phys. (New York)* **24**, 419 (1963).

⁶R. D. Mountain, *Rev. Mod. Phys.* **38**, 205 (1966).

⁷L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Addison-Wesley, Reading, Mass., 1958).

⁸S. Chapman and T. G. Cowling, *The Mathematical*

Theory of Nonuniform Gases (Cambridge U. Press, Cambridge, England, 1952); or J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird, *Molecular Theory of Gases and Liquids* (Wiley, New York, 1954).

⁹See for example N. Bloembergen, W. H. Lowdermilk, M. Matsuoka, and C. S. Wang, *Phys. Rev. A* **3**, 404 (1971). The explicit form of the coupling constants has also been obtained independently by N. A. Clark,

thesis, Massachusetts Institute of Technology, 1970 (unpublished); Bergé, Calmettes, DuBois, and Laj (Ref. 3) obtained equivalent expressions for liquid mixtures.

¹⁰M. Kohler, *Ann. Phys. (Leipzig)* **39**, 209 (1941); also see Refs. 1 and 2.

¹¹N. Bloembergen, W. H. Lowdermilk, and C. S. Wang, *Phys. Rev. Lett.* **25**, 1476 (1970).

Anomalous Resistivity Due to Electrostatic Turbulence

S. Peter Gary

Department of Physics, College of William and Mary, Williamsburg, Virginia 23185

and

J. W. M. Paul

Culham Laboratory, Abingdon, Berkshire, England

(Received 22 March 1971)

We calculate a general expression for anomalous resistivity due to weak electrostatic turbulence in a plasma. In the case of ion acoustic turbulence, it is shown to reduce to the heuristic result of Sagdeev. Applications to perpendicular collisionless shock experiments are discussed.

The heating of electrons by superthermal fluctuations in a plasma has been observed in several experiments.^{1,2} This turbulent heating may be described in terms of an anomalous resistivity η^* which can be orders of magnitude larger than the resistivity due to electron-ion collisions.

To calculate η^* , we consider a stochastic model in which energy is transferred from the waves to the particles by small-angle, random scatterings of the electrons by the fluctuations. We assume a homogeneous plasma and a given spectrum of turbulence. (The inclusion of self-consistent fields is a much more difficult problem and will not be considered here.) Then the time development of the ensemble-averaged electron distribution function

$$F(\vec{v}, t) = \langle f(\vec{r}, \vec{v}, t) \rangle$$

is given by a Fokker-Planck equation³⁻⁵

$$\frac{DF(\vec{v}, t)}{Dt} = \frac{\partial}{\partial v_\mu} \left[D_{\mu\nu} \frac{\partial F(\vec{r}, t)}{\partial v_\nu} \right], \quad (1)$$

where D/Dt represents the time derivative along the zeroth-order trajectory of an electron. If there are no fluctuating magnetic fields,⁴

$$D_{\mu\nu} = (e^2/m^2) \langle \delta E_\mu(\vec{r}, t) \int_{-\infty}^t \delta E_\nu(\vec{r}', t') dt' \rangle, \quad (2)$$

where δE_α is the α th component of the fluctuating electric field and the t' integration is over a zeroth-order trajectory [$\vec{r}' = \vec{r}(t')$, $\vec{v}' = \vec{v}(t')$].

We consider only electrostatic waves so that

$$\delta \vec{E}(\vec{r}, t) = -\nabla \varphi(\vec{r}, t), \quad (3)$$

and we work in terms of the Fourier transform of the potential-potential correlation function,

$$S(\vec{k}, \omega) = \int d^3\xi \int d\tau \exp[-i(\vec{k} \cdot \vec{\xi} - \omega\tau)] R(\vec{\xi}, \tau), \quad (4)$$

where

$$R(\vec{\xi}, \tau) \equiv \langle \varphi(\vec{r}, t) \varphi(\vec{r} + \vec{\xi}, t + \tau) \rangle. \quad (5)$$

Then

$$D_{\mu\nu} = [e^2/(2\pi)^4 m^2] \int d^3k \int d\omega k_\mu k_\nu S(\vec{k}, \omega) \times \int_{-\infty}^t dt' \exp\{i[\vec{k} \cdot (\vec{r}' - \vec{r}) - \omega(t' - t)]\}. \quad (6)$$

For a plasma in which there are uniform electric magnetic fields \vec{E}_0 and \vec{B}_0 ,

$$\frac{D}{Dt} = \frac{\partial}{\partial t} + \vec{v} \cdot \frac{\partial}{\partial \vec{r}} - \frac{e}{m} \left(\vec{E}_0 + \frac{\vec{v} \times \vec{B}_0}{c} \right) \cdot \frac{\partial}{\partial \vec{v}}. \quad (7)$$

We now assume a steady-state situation in which the force exerted on the electrons by \vec{E}_0 is balanced by the "friction" force due to particle scattering by waves. Then from Eq. (1),

$$\frac{en_0 \vec{E}_0}{m} + \frac{en_0}{m c} (\vec{v}_0 \times \vec{B}_0) = - \int d^3v \overline{D} \cdot \frac{\partial F(\vec{v})}{\partial \vec{v}}, \quad (8)$$

where $\vec{v}_0 = n_0^{-1} \int d^3v \vec{v} F(\vec{v}) = v_0 \hat{e}_3$ is the electron drift. Then, by analogy with magnetohydrody-