Anomalous Dispersion in Disparate-Mass Gas Mixtures

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(Received 21 August 1984)

Dispersion of forced sound in Xe-He mixtures is found here to show mode coupling: interference between the sound mode and another excitation mode of the gas. Qualitatively different dispersion is found, depending upon whether the He mole fraction is greater or less than a critical value, if the frequency is higher than a critical frequency. Results also show that "two-temperature hydrodynamics" must be used to describe a disparate-mass gas in this frequency range.

PACS numbers: 51.10.+y, 05.20.Dd, 43.35.+d, 51.40.+p

Forced sound propagation in a gas mixture of very light with very heavy molecules is expected to be particularly interesting. In the first place, Navier-Stokes hydrodynamics fails in disparate-mass gas mixtures at frequencies where it is valid to use a two-temperature generalized hydrodynamics[1] (frequencies high, but lower than any inverse mean free time in the gas). Secondly, according to Huck and Johnson[2,3] mode coupling should occur, within the same frequency range, between the acoustic mode and another mode of excitation of the gas. The present experiments on forced sound propagation in Xe-He mixtures show clear evidence of this predicted mode interference. They also allow one to decide between ordinary hydrodynamics and its two-temperature extension.

The linear acoustic behavior of a gas may be predicted by assumption of small disturbances from equilibrium of the form $\exp[i(kz-\omega t)]$ in the governing equations to obtain a dispersion relation giving the wave number k in terms of the angular frequency ω (for forced sound propagation, ω will be assumed real and positive). Various processes, such as heat conduction or diffusion—not only sound waves—correspond to such disturbances; each is characterized by a different solution k_i of the dispersion relation, but sound is commonly distinguished by being the only propagating mode in the gas. For this acoustic mode, the phase velocity $u(\omega)$ is given by the real part of the appropriate solution k_s,

$$u=\omega/\mathrm{Re}(k_s), \tag{1}$$

while the absorption parameter β is given by

$$\beta=u_0\,\mathrm{Im}(k_s)/\omega, \tag{2}$$

where u_0 is the low-frequency speed of sound.

Linearized hydrodynamics gives dispersion relations correct only for low-frequency deviations from equilibrium[4]:

$$k_s=(1+i\beta)\omega/u_0, \tag{3}$$

with β linear in ω. In this limit, there is no dispersion, $u_0/u=1$. For the diffusion and thermal modes, respectively,[5]

$$k_D^2=i\omega/D, \tag{4}$$

$$k_T^2=i\omega NC_p/\kappa, \tag{5}$$

where D is the coefficient of diffusion, C_p the molar specific heat at constant pressure, N the number of moles, and κ the thermal conductivity.

For disparate-mass gas mixtures, one expects a two-temperature generalization of hydrodynamics to be needed[1] to describe sound propagation in the mixture, when frequencies are lower than any inverse mean free time, but high enough so that ω approximately equals the inverse relaxation time for species kinetic-energy differences.

From such "two-temperature hydrodynamics," Huck and Johnson[2] predict mode coupling in sound in disparate-mass gases. Dispersion is predicted to be a *discontinuous* function of the light-species mole fraction x, the discontinuity resulting from a degeneracy between the sound mode and one other hydrodynamic mode in the gas. The wave numbers k_i of the two modes become equal at a critical frequency ω_c and a critical composition x_c. For frequencies higher than ω_c, the acoustic behavior of the gas should be that of a qualitatively different physical process depending upon whether x is greater than or less than x_c. This also suggests the possibility[2] of observing the simultaneous presence of both competing modes when x is sufficiently near to x_c.

Contradictory predictions of Foch, Uhlenbeck, and Losa[4] based on perturbation theory give k as a (necessarily smooth and single valued) polynomial in ω and in x.

Sound absorption and dispersion measurements[6] are reported here, for mixtures of Xe ($m=131.3$ amu) and He ($m=4$ amu) and for frequency-to-pressure ratios up to 100 MHz/bar and for He mole fractions x from 0.2 to 0.8. Full details will be presented elsewhere. Earlier work of Losa[7] does not go to such high frequencies at the extremes of this composition range, and omits altogether the critical range $x=0.4$ to 0.6.

The experiment uses a continuous sound wave gen-

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erated by a coaxial pair of thin piezoceramic transducers (see Fig. 1) excited at their resonance frequency of 1.005 MHz. Since ω-dependent effects scale as ω/p, the frequency-to-pressure ratio is varied by adjusting the pressure p. The phase and amplitude of the received signal are observed as the separation of the transducer disks is varied.

High absorption in the gas in the critical region makes it important to reduce transmitter-receiver cross-talk. Electrical crosstalk is reduced by gluing each transducer to a copper disk with a thickness of one wavelength at the operating frequency.[8] Mechanical crosstalk is reduced by supporting each disk in a nodal plane and mounting the receiver housing on narrow pins capped by nylon tips to produce an acoustic mismatch. Gas pressure is measured with a diaphragm-type pressure transducer. The temperature is controlled to within 0.4 °C of a nominal 25 °C; a micrometer screw gauge measures linear displacements to 2 μm.

The amplitude and phase of the signal are found by phase rectification. A phase-control loop maintains a null output at the phase-shift detector B, shown in Fig. 2. A change in phase of the received signal produces a nonzero phase-control voltage which is integrated and used to establish reference B in quadrature and reference A in phase with the received signal. The output of phase-shift detector A gives amplitude information, while phase measurements are made across the phase delay unit with use of a digital phase meter accurate to 1° at 1 MHz.

Boundary-layer effects and the influence of standing waves are minimized by putting a lower limit on the transducer separation Δz. Effects due to multiple reflections have a maximum relative magnitude of roughly $\exp(-2\beta\omega\,\Delta z/u_0)$. The standing-wave contribution is made negligible by ensuring that this factor is less than 1%. This also minimizes boundary-layer effects by keeping Δz always greater than an estimated 20 mean free paths.

The data are analyzed under the assumption that only a single mode is excited in the gas, varying as the real part of

$$V(z,t) = V_0 \exp(-\beta k_0 z) \exp[i(2\pi z/\lambda - \omega t)], \quad (6)$$

where $k_0 = \omega/u_0$. For such a wave, both $\ln|V|$ and the phase angle $(2\pi z/\lambda)$ vary linearly with z. This assumption is expected to be inadequate for $\omega > \omega_c$ and $x \simeq x_c$, but was adopted because high absorption and intrinsic experimental errors make meaningful two-mode analysis of the data impossible.

Experimental results are compared in Fig. 3 with predictions[6] of two-temperature hydrodynamics, using exact linearized thirteen-moment two-temperature equations[9] for Maxwell molecules.[10] The pure-gas viscosities and the He-Xe diffusion coefficient are taken from experiment.[11] The critical frequency and critical composition so predicted are $\omega_c/2\pi = 75 \pm 1$ MHz at 1 bar, and $x_c = 0.495 \pm 0.005$, respectively.

Figure 3(a) shows dispersion data for compositions above and below the predicted critical composition $x_c \simeq 0.5$; also shown are theoretical predictions for $x = 0.45$ and $x = 0.60$ (upper and lower dashed lines, respectively). Although there is qualitative disagreement with theory for $x = 0.45$, the main predictions of Huck and Johnson[2] for $\omega \gtrsim \omega_c$ are unambiguously confirmed: (i) a smooth, gentle change in dispersion as x decreases from 0.45, or increases from 0.60; (ii) physically different dispersion for $x < x_c$ (falling sound speed) compared to $x > x_c$ (rising sound speed); (iii) a sharp change in dispersive behavior over a small composition range near $x = 0.5$. The fall in sound speed observed for $\omega \gtrsim \omega_c$ and for compositions $x \leqslant 0.45$, while predicted by the approach of Huck and Johnson,[2,12] is itself noteworthy—dispersion typically *increases* the sound speed.[13]

Figure 3(b) shows that close to the critical composition, at $x = 0.50$, and for $\omega/2\pi p$ above $\simeq 70$ MHz/bar, the data analyzed by a single-mode assumption fall between the dispersion curves for the two competing modes and show large scatter. Here the predicted absorptions for the two modes in question have compar-

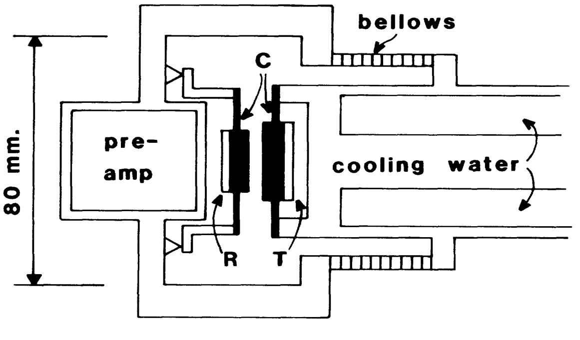

FIG. 1. Test chamber. R (receiver) and T (transmitter), acoustic transducers; C, copper disks.

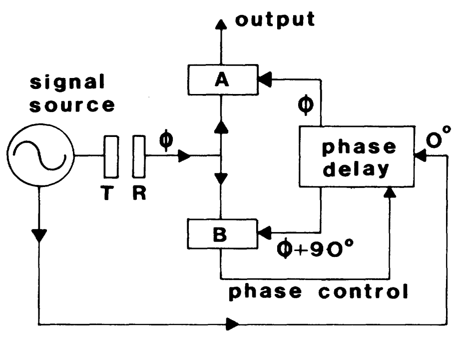

FIG. 2. Signal-detection scheme. A and B, phase-sensitive detectors; R, receiver; T, transmitter.

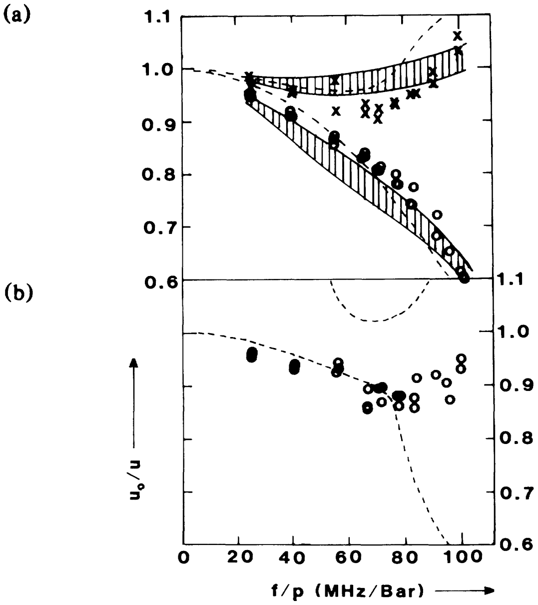

FIG. 3. (a) Upper (lower) shaded band: dispersion data for He mole fraction $x = 0.20$, 0.30, 0.40 (0.70, 0.80). Crosses, $x = 0.45$; circles, $x = 0.60$. Upper (lower) dashed line: theory, $x = 0.45$ (0.60). (b) Circles, $x = 0.50$. Lower (upper) dashed line: theory, acoustic (interfering nonacoustic) mode. ($f \equiv \omega/2\pi$.)

able and large values. Exceptionally large absorption is also observed experimentally.[6] The data are consistent with the presence of appreciable components of both of these modes in the received signal, but more positive conclusions must await substantially better experiments. The smooth behavior of experiment for $0.2 \leqslant x \leqslant 0.4$ and for $0.6 \leqslant x \leqslant 0.8$ seems to be related to the fact that all nonacoustic modes are predicted to be highly attenuated here even for $\omega > \omega_c$.

A rough experimental estimate for the critical parameters may be made from Fig. 3: evidently $x_c \sim 0.5$ and $\omega_c/2\pi p \sim 70$ MHz/bar. This agrees qualitatively with the present calculation, with similar calculations[6] for hard-sphere molecules (including thermal diffusion) giving $x_c = 0.505 \pm 0.005$ and $\omega_c/2\pi p = 69 \pm 1$ MHz/bar, and with the predictions $x_c = 0.46 \pm 0.01$ and $\omega_c/2\pi p \simeq 74$ MHz/bar of Ref. 2. It disagrees markedly, however, with ordinary hydrodynamics, which gives[14] $x_c = 0.24$–0.31 and $\omega_c/2\pi p = 84$–107 MHz/bar.[15]

The perturbation approximation of Foch, Uhlenbeck, and Losa[4] cannot, of its nature, predict the interference effects observed. Neither does it predict the decrease in sound speed observed here for compositions $x \leqslant 0.45$ at high enough frequencies.[13] As will be shown elsewhere, the failure of the perturbation approximation for disparate-mass gases is linked with the appearance of a second small parameter (the square root of the mass ratio); when that parameter and the perturbation parameter have comparable magnitudes the approximation of Ref. 4 is invalidated.

The identity of the interfering nonacoustic mode is also of interest. Present calculations indicate that if one follows this mode to low frequencies it becomes the thermal mode, while it becomes the diffusion mode in the more approximate calculation of Ref. 4. Since these modes have similar dispersion relations, Eqs. (4) and (5), respectively, and since $D \sim \kappa/NC_p$ at the critical composition, low-frequency behavior of the interfering mode is likely to be sensitive to small variations in initial assumptions. In any case, at frequencies $\omega > \omega_c$ the physical nature of the two interfering modes is unambiguous.[3] In a mixture of near-critical composition, the slow wave has the nature of a damped acoustic wave carried by the heavy component of the mixture, which is partly decoupled from the light component, while the fast wave is a disturbance carried by the light species in the presence of semifixed heavy scatterers. Which of these two modes becomes the acoustic mode at low frequencies depends upon whether x is less than or greater than x_c (thus, roughly upon whether the heavy or the light molecules are more numerous).

One of us (J.R.B.) wishes to thank his experimental supervisor R. G. Harlow; this work was supported in part by the British Science and Engineering Research Council.

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[1] C. J. Goebel, S. M. Harris, and E. A. Johnson, Phys. Fluids **19**, 627 (1976); E. A. Johnson, Arch. Mech. (Warsaw) **28**, 803 (1976); C. J. Goebel, S. M. Harris, and E. A. Johnson, in *Rarefied Gas Dynamics,* edited by J. L. Potter (American Institute of Aeronautics and Astronautics, New York, 1977), Vol. 51, Pt. I, p. 109; see also references therein.

[2] R. J. Huck and E. A. Johnson, Phys. Rev. Lett. **44**, 142 (1980); E. A. Johnson, Arch. Mech. (Warsaw) **32**, 703 (1980).

[3] R. J. Huck and E. A. Johnson, in *Rarefied Gas Dynamics,* edited by S. Fisher (American Institute of Aeronautics and Astronautics, New York, 1981), Vol. 74, Pt. I, p. 452.

[4] J. D. Foch, G. E. Uhlenbeck, and M. F. Losa, Phys. Fluids **15**, 1224 (1972); see also references therein.

[5] J. P. Boon and S. Yip, *Molecular Hydrodynamics* (McGraw-Hill, New York, 1980), Eq. (5.5.16) plus neglect of thermal diffusion.

[6] J. R. Bowler, Ph.D. thesis, University of Surrey, 1984 (unpublished).

[7] M. F. Losa, Ph.D. thesis, University of Colorado, 1972 (unpublished).

[8] A technique used successfully by R. Schotter, Phys. Fluids **17**, 1163 (1974).

[9] Bowler, Ref. 6; obtainable from J. M. Burgers, *Flow Equations for Composite Gases* (Academic, New York, 1969),

Chap. 2; this differs from Ref. 2 in retaining terms of the order of the He-Xe mass ratio.

[10] Intermolecular forces proportional to r^{-5}, where r is intermolecular separation.

[11] Viscosity data: J. Kestin and W. Leidenfrost, Physica (Utrecht) **25**, 1033 (1959); diffusion data: P. S. Arora, H. I. Robjohns, and P. J. Dunlop, Physica (Utrecht) **95A**, 561 (1971).

[12] E. A. Johnson, Phys. Fluids **23**, 2558 (1980).

[13] Some indication of this behavior is seen in Losa's experiments, Ref. 7. The approach of Ref. 4 predicts such behavior *only* when x is very close to 0.10 (cf. Ref. 7, Fig. IV.3).

[14] Foch, Uhlenbeck, and Losa, Ref. 4; E. G. D. Cohen and B. Kamgar-Parsi, private communication.

[15] Two-temperature hydrodynamics must also be used in this (k, ω) regime for light scattering in Xe-He mixtures: E. A. Johnson, Phys. Rev. A **27**, 1146 (1983).