Journal of Statistical Physics, Vol. 57, Nos. 3/4, 1989

Two-Temperature Hydrodynamics and Multiple Sound Modes in Disparate-Mass Gas Mixtures

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Received March 20, 1989

Gas mixtures of heavy plus light molecules can apparently support several different sorts of sound waves (the most recent such prediction is that of Campa and Cohen). The origin of the first such predictions is reviewed, along with the development of the two-temperature hydrodynamic equations which govern these mixtures at moderate wavenumbers and frequencies. Light scattering in the two-temperature regime is also discussed. Experiments in both sound propagation and light scattering are shown to confirm the existence of a two-temperature regime, and two simultaneous sound modes, in these disparate-mass gas mixtures.

KEY WORDS: Gas mixtures; sound propagation; light scattering; hydrodynamics; two-temperature hydrodynamics; mode interference; double sound; disparate-mass gases; He–Xe mixtures.

1. INTRODUCTION

Gas mixtures of heavy plus light molecules can support several different sorts of sound waves (the most recent prediction of an effect of this sort is that of Campa and Cohen(1)). The possibility of such an effect was first pointed out by Huck and Johnson in 1980.(2,3) The present paper reviews the basis of this prediction and its experimental confirmation. The effect occurs at frequencies for which ordinary hydrodynamics is not valid; the development of two-temperature hydrodynamic equations which can correctly describe the regime in question is therefore also reviewed.

At low frequencies, one can expect sound propagation in a gas to behave simply, with a sound velocity independent of frequency, and absorption proportional to frequency. The "low" frequencies in question

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are required to be low in comparison with all the characteristic internal frequencies of the system. For monatomic gases, these are simply the inverse relaxation times for gas parameters such as density or velocity fluctuations.(4) For single-species noble gases, or for mixtures of comparable atomic weights, these frequencies are all of similar magnitude (say, ν_0), of the order of an inverse mean free time. The "low-frequency" regime is thus at least qualitatively well defined: $\omega \ll \nu_0$. In this regime, processes in the gas are correctly described by ordinary hydrodynamics, together with linear constitutive relations, with material parameters (such as thermal conductivity) which are calculable by the well-known methods of Chapman and Enskog.(5)

Although the hydrodynamic equations and their related transport coefficients are derived from the Boltzmann equation,(4) the predictions of that equation have a much broader range of application. In particular, the Boltzmann equation should in principle be capable of giving correct predictions for gas behavior in the kinetic regime, that is, for processes involving frequencies of the order of intrinsic relaxation frequencies (or, alternatively, length scales of the order of a mean free path).(4)

2. TWO-TEMPERATURE HYDRODYNAMICS FOR DISPARATE-MASS GASES

A disparate-mass gas mixture is one composed of very heavy plus very light molecules. Multiple sound modes in gases seem to be one characteristic possibility for systems of this type.

It was first pointed out by Grad(6) that a disparate-mass gas mixture would have an internal relaxation frequency much lower than that of either of its separate constituents. This means that the breakdown of hydrodynamics and the onset of the kinetic regime happen at much lower frequencies in such mixtures than in more ordinary gases and gas mixtures. The multiple sound modes predicted by Huck and Johnson(2,3) occur in precisely such mixtures and at such frequencies. Correct physical predictions will thus depend upon the existence of a theoretical description of these mixtures valid at these frequencies.

In a pure monatomic gas, the inverse mean free time gives the correct order of magnitude for the frequency of any sort of perturbation, for instance, in momentum or energy, to relax to equilibrium. In a disparate-mass mixture, on the other hand, the kinematic effects of two very different atomic masses,

$$m_1/m_2 \ll 1 \tag{1}$$

result in very diffent relaxation times for different molecular properties. As originally pointed out by Grad,$^{(6)}$ if the light-species momentum relaxation time ν_1 is taken as reference, then

$$\nu_1 \approx (\tau_1)^{-1} \tag{2}$$

where τ_1 is the light-species self-collision time. The much lower frequency ν_2 for self-collisions among the heavy molecules

$$\nu_2 \approx (\tau_2)^{-1} \tag{3}$$

is of order

$$\nu_2 \approx \delta \nu_1 \tag{4}$$

for comparable number densities $n_1 \approx n_2$, where

$$\delta \equiv (m_1/m_2)^{1/2} \ll 1 \tag{5}$$

while the lowest frequency of all is that with which the light-species kinetic energy comes to equilibrium with that of the heavy species:

$$\nu_\Delta \approx (m_1/m_2)\, \nu_1, \qquad \nu_\Delta \ll \nu_1, \nu_2 \tag{6}$$

The laws of Newtonian mechanics imply this inefficient exchange of kinetic energy between light and heavy species, a fact which led Grad$^{(6)}$ to predict the possibility that under appropriate circumstances the two components of such a mixture could support different temperatures.

The regime of interest here is the frequency regime $\omega \approx \nu_\Delta$. In this regime, disparate-mass gas behavior indeed departs from that of more normal gases. Nevertheless, an approximate description emerges which is very like that of hydrodynamics, while allowing extra degrees of freedom which include the possibility of different temperatures for the different species in the mixture, as predicted by Grad.$^{(6)}$ In what follows the generalized two-temperature hydrodynamic description developed by Goebel *et al.*$^{(7-9)}$ will be used. These equations have been derived from a Grad 13-moment type expansion$^{(10)}$ of the Boltzmann equations for the two coupled species of the mixture. It is assumed that $(m_1/m_2)^{1/2} \equiv \delta \ll 1$ and that neither mole fraction x_i is so small as to be of order δ.

A key variable in these equations is the temperature separation

$$\Delta \equiv T_2 - T_1 \tag{7}$$

between the species. This is obtained from the defining equations

$$\tfrac{3}{2} n_i k T_i = \int (m_i/2)(\mathbf{v} - \mathbf{U}_i)^2 f_i \, d\mathbf{v} \tag{8}$$

with n_i the number density of species i, $\mathbf{U}_i$ its flow velocity, f_i the distribution function of species i, $\mathbf{v}$ its molecular velocity, and k Boltzmann's constant. Instead of separate temperatures T_i, the alternative variables Δ and T will be used, where T is the common temperature given by

$$\tfrac{3}{2}nkT = \sum_i \int (m_i/2)(\mathbf{v}-\mathbf{U})^2 f_i\, d\mathbf{v} \tag{9}$$

Similarly, the overall mixture flow velocity $\mathbf{U}$ and the diffusion velocity

$$\mathbf{W} \equiv \mathbf{U}_2 - \mathbf{U}_1 \tag{10}$$

will be used in place of the separate species flow velocities $\mathbf{U}_i$ given by

$$n_i \mathbf{U}_i = \int \mathbf{v} f_i\, d\mathbf{v} \tag{11}$$

The flow equations which result from this approach reduce to familiar Euler/Navier–Stokes equations for low frequencies $\omega \ll \nu_\Delta$ [ν_Δ, introduced in (6), is the relaxation time for the temperature difference Δ of (7)]. In the regime of interest ($\omega \approx \nu_\Delta$), however, one obtains instead

$$(D\rho_1/Dt) + \rho_1 \nabla \cdot \mathbf{U} - \nabla \cdot \rho_1 \mathbf{W} = 0 \tag{12a}$$

$$(D\rho_2/Dt) + \rho_2 \nabla \cdot \mathbf{U} = 0 \tag{12b}$$

$$\rho_2(D\mathbf{U}/Dt) + \nabla(n_1 kT_1 + n_2 kT_2) + \nabla \cdot \{\mathbf{P}_2\} = 0 \tag{13}$$

$$(D/Dt)(\tfrac{3}{2}nkT) + \tfrac{5}{2}nkT\, \nabla \cdot \mathbf{U} + \nabla \cdot (\mathbf{q}_{10} + \mathbf{q}_{20}) + \{\mathbf{P}_2\} : \nabla\mathbf{U} = 0 \tag{14}$$

Here $D/Dt = (\partial/\partial t + \mathbf{U} \cdot \nabla)$ is the derivative with respect to flow, $n = (n_1 + n_2)$ the overall number density, and $\rho_i = n_i m_i$ the i-species mass density. The $\mathbf{q}_i$ and $\{\mathbf{P}_i\}$ are, respectively, the heat flux and the symmetrized traceless pressure tensor of species i taken in the rest frame of that species, and $\mathbf{q}_{i0}$ is the heat flux of species i taken with respect to the overall flow velocity of the gas; in the present approximation

$$\mathbf{q}_{10} = \mathbf{q}_1 - \tfrac{5}{2} n_1 kT_1 \mathbf{W}, \qquad \mathbf{q}_{20} = \mathbf{q}_2 \tag{15}$$

In (12)–(14), the only first-order terms are those involving $\{\mathbf{P}_2\}$ and $\mathbf{q}_2$; all other terms are present to *lowest* order ("Euler" level) in the regime $\omega \approx \nu_\Delta$.

Supplementing the flow equations (12)–(14), one has the constitutive equations, which in the present approximation become

$$\mathbf{W} = (D/x_2)\nabla \ln p_1 \qquad (p_i \equiv n_i kT_i) \tag{16}$$

$$\{\mathbf{P}_2\} = -2\mu_2 \{\nabla \cdot \mathbf{U}\} \tag{17}$$

$$\mathbf{q}_i = -\lambda_i \nabla T_i \tag{18}$$

Explicit forms for the transport coefficients D, μ_2, and λ_i are given in the Appendix. The final equation governing temperature separation Δ is peculiar to a disparate-mass gas in the two-temperature regime:

$$\begin{aligned}(D/Dt)\Delta + \tfrac{2}{3}\Delta\nabla\cdot\mathbf{U} - (\tfrac{2}{3}kn_1)\nabla\cdot\mathbf{q}_{10} - (T_1/n_1)\nabla\cdot(n_1\mathbf{W})\\ + (\tfrac{2}{3}kn_2)[\nabla\cdot\mathbf{q}_2 + \{\mathbf{P}_2\}:\nabla\mathbf{U}] = -\nu_\Delta\Delta \qquad (19)\end{aligned}$$

For low-frequency phenomena, (19) is of second order and is therefore not retained in any ordinary hydrodynamic equations. In the two-temperature regime of a disparate-mass gas, however, its contribution (except for the final bracket on the left) is of *lowest* order. The equations (12)–(19) were predicted by Goebel *et al.*$^{(7-9)}$ to govern the behavior of a disparate-mass gas in the two-temperature regime in the same way that the ordinary Euler/Navier–Stokes equations govern ordinary gas mixtures for all phenomena of low enough frequency.

3. TWO-TEMPERATURE HYDRODYNAMICS NEAR EQUILIBRIUM

Given the prediction that there should be a regime in which a disparate-mass gas behaves in accordance with two-temperature hydrodynamics, one may ask what sort of physical effects might bear a characteristic "two-temperature" signature. The simplest should be linear effects (involving only small deviations from equilibrium), but effects characterized either by frequencies $\omega \approx \nu_\Delta$ or by length scales $L \approx (m_2/m_1)l$, where l is a mean free path. The propagation of forced sound of frequency $\omega \approx \nu_\Delta$ is an obvious example of the former type, while light scattering for wavenumbers

$$k \approx \nu_\Delta/V_0 \qquad (20)$$

is an example of the latter (V_0 is the equilibrium speed of sound).

3.1. Sound Propagation

The behavior of small perturbations from equilibrium in the gas in the two-temperature regime can be predicted by linearizing (12)–(14) and (16)–(19) about equilibrium values of the number densities n_{i0}, temperature T_0, and pressure p_0. For sound propagation, one assumes that deviations from equilibrium take the form $\exp[i(kx-\omega t)]$, so that density fluctuations, for instance, are given by

$$n_i = n_{i0}\{1 + \tilde{n}_i \exp[i(kx-\omega t)]\} \qquad (21)$$

The governing equations are then written as equations in the dimensionless amplitudes for these deviations, defined in similar fashion to the amplitudes $\tilde{n}_i$ of (21). The deviation amplitudes of interest here are those for the flow velocity $\tilde{U}$, overall temperature $\tilde{T}$, diffusion velocity $\tilde{W}$, pressure deviator $\tilde{P}$ $(\equiv \{P\}_{xx}/p_0)$, heat fluxes $\tilde{q}_i$, and temperature separation $\tilde{\Delta}$. Temperature amplitudes are made dimensionless by reference to T_0, velocity amplitudes by reference to the characteristic speed

$$c \equiv (p_0/\rho_0)^{1/2} \tag{22}$$

(ρ_0 is the equilibrium mass density $\rho_{10}+\rho_{20}$), and heat fluxes by reference to $(p_0 c)$. Defining the reduced wavenumber z by

$$z \equiv kc/\omega \tag{23}$$

we find that the equations governing sound propagation take the following simple form:

Conservation equations:

$$\tilde{n}_1 = z(\tilde{U} - \tilde{W}), \qquad \tilde{n}_2 = z\tilde{U} \tag{24}$$

$$\tilde{U} = z\left[\sum_i (x_i \tilde{n}_i) + \tilde{T} + \tilde{P}\right] \tag{25}$$

$$\tilde{T} = \frac{2}{3}\sum_i (x_i \tilde{n}_i + z\tilde{q}_i) \tag{26}$$

Constitutive equations:

$$\tilde{W} = (i\tilde{\omega}z)\,\tilde{D}(\tilde{n}_i + \tilde{T} - x_2\tilde{\Delta}) \tag{27}$$

$$\tilde{P} = -(i\tilde{\omega}z)\,\tilde{\mu}\tilde{U} \tag{28}$$

$$\tilde{q}_1 = -(i\tilde{\omega}z)\,\tilde{\lambda}_1(\tilde{T} - x_2\tilde{\Delta}) \tag{29}$$

$$\tilde{q}_2 = -(i\tilde{\omega}z)\,\tilde{\lambda}_2(\tilde{T} + x_1\tilde{\Delta}) \tag{30}$$

$$\tilde{\Delta} = -(i\tilde{\omega}z)\,\tilde{\tau}[\tilde{W} + (\tilde{q}_2/x_2) - (\tilde{q}_1/x_1)] \tag{31}$$

Here x_i is the mole fraction for species i, while $\tilde{D}$, $\tilde{\mu}$, $\tilde{\lambda}_i$, and $\tilde{\tau}$ are dimensionless transport coefficients defined in the Appendix, (A.6)–(A.9). The frequency $\tilde{\omega}$ is made dimensionless with respect to the reference frequency ν_Δ, given by

$$\nu_\Delta = 2c^2 x_2/D \tag{32}$$

[an explicit expression is also given in (A.4a)].

The dispersion relation governing sound propagation is that for $z = z(\tilde{\omega})$ or $\tilde{\omega} = \tilde{\omega}(z)$ obtained from (24)–(31), above; for forced sound propagation at a given frequency, the former is the appropriate relation, with $\tilde{\omega}$ real and positive. At low frequencies, one expects one root of this equation to correspond to plane-wave propagation in the $+x$ direction. That root is then identified by its low-frequency behavior:

$$\text{dispersion:} \qquad \operatorname{Re}[(5/3)^{1/2} z] = V_0/V \underset{\omega \to 0^+}{\to} 1 \tag{33}$$

$$\text{absorption:} \qquad \operatorname{Im}[(5/3)^{1/2} z] = \alpha \underset{\omega \to 0^+}{\to} 0 \tag{34}$$

where V is the speed of propagation at frequency ω.

3.2. Light Scattering

Light scattering probes the behavior of density fluctuations in a gas through the density correlation function $S(k, \omega)$. The information one obtains about a gas from light scattering is complementary to that obtained from sound propagation, in the sense that one is probing a different region of the dispersion relation in the space of complex k and ω. An experiment specifies the wave number of the density fluctuations being probed (fixed, real k), and the spectrum of the scattered light is observed for this wavenumber. The frequency ω of interest is the frequency shift $\omega = \omega_s - \omega_i$ between the scattered (s) and incident (i) radiation; ω is real, and in the range $\pm\infty$.

For $S(k, \omega)$, the hydrodynamic regime is the one in which

$$k \ll \nu_0/c \tag{35}$$

where ν_0 is the lowest intrinsic relaxation frequency in the gas. For these wavenumbers k, one may expect to derive $S(k, \omega)$ from the linearized equations of hydrodynamics.(11) For higher wavenumbers such an approach is no longer adequate. However, for a disparate-mass mixture, there should exist a two-temperature regime

$$\nu_\Delta/c \lesssim k \ll \nu_1/c,\ \nu_2/c \tag{36}$$

for which the linearized two-temperature equations should provide valid predictions.

The analysis is analogous to that followed in Section 3.1 for sound propagation. The method is discussed in detail in, e.g., ref. 11; its generalization to gas mixtures is discussed, for instance, in refs. 12 and 13. The spectral power density $S(k, \omega)$ of light scattered by a dilute binary gas mixture can be written in terms of the i-species density fluctuation self-correlations S_{ii} and the cross-correlations S_{ij} as

$$S(k, \omega) = N(x_1 a^2 S_{11} + 2x_1 a S_{12} + x_2 S_{22}) \tag{37}$$

Here N is a normalization, and

$$a \equiv \alpha_1/\alpha_2 \tag{38}$$

is the ratio of the i-species optical polarizabilities α_i.

Qualitatively, one expects the observed spectrum at low wavenumbers to show a single peak at $\omega = 0$ and two symmetrically-located satellite peaks at $\pm \Delta\omega$, where

$$kV_0 = \pm \Delta\omega \tag{39}$$

The peak at $\omega = 0$, and the widths of all peaks, relate to dissipative processes in the gas.

4. COMPARISON WITH EXPERIMENT

The theoretical approach described here$^{(7-9)}$ [Eqs. (12)–(14) and (16)–(19)] is approximate in several respects: it is based on a limited set of moments of the Boltzmann equation; it discards all terms of order $\delta^2 \equiv m_1/m_2$ or smaller; and it assumes Maxwellian intermolecular forces (repulsion proportional to r^{-5}, where r is intermolecular separation). The first two approximations result in noticeable discrepancies from the results of full multimoment type calculations such as those of Boley and Yip$^{(12)}$ (the final approximation usually has a smaller effect). Detailed agreement with experiment cannot therefore be expected in the present approach. It can, however, be relied upon to offer a relatively simple way of predicting in a large-scale and qualitative way the differences between ordinary hydrodynamics and the two-temperature behavior of disparate-mass gases. In addition, it provides clear pointers to the physical nature of these differences, as well as indicating the correct physical regimes in which these differences should be evident. As will be seen in the following sections, experimental evidence for two-temperature gas behavior and for dual sound modes is, indeed, of such a gross and qualitative kind.

Among the monatomic gases, the best readily-available candidate for a disparate-mass mixture is that of He and Xe, with a mass ratio

$$m_1/m_2 = 4/131.3 \tag{40}$$

The sound frequency range relevant to the existence of a two-temperature regime is of the order of 100 MHz at standard temperature and pressure [this is the frequency given by (32)]. Wave numbers relevant to light scattering in the two-temperature regime are of order $x_2^{1/2}(5 \times 10^4)\,\mathrm{cm}^{-1}$ at standard temperature and pressure, from (36). The optical polarizability of Xe is much greater than that of He:

$$a = \alpha_1/\alpha_2 = 0.0514 \tag{41}$$

for light(14) of wavelength 6328 Å. This fact implies, from (37), that one will only *see* density fluctuations in Xe, while cross-correlation effects, and the behavior of He in the mixture, must be inferred indirectly.

The two-temperature regime is directly accessible to light scattering and to sound propagation experiments using available techniques. The *existence* of such a regime was first verified by the light scattering experiments of Letamendia *et al.*(15,16) and subsequently confirmed via sound propagation by Bowler.(17–19) The phenomenon of dual sound mode interference in this regime was also confirmed by Bowler in these experiments.(17–19) Light scattering will be discussed first.

4.1. Light Scattering

Since light scattering experiments succeed in seeing only Xe in a He–Xe mixture, it is of interest to look at the governing equations insofar as they affect the heavy species alone. Dissipation is associated with transport, and governed by the constitutive equations (16)–(19). As was shown earlier,(7–9) temperature separation Δ, the diffusion velocity $\mathbf{W}$, and the light-species heat flux $\mathbf{q}_l$ comprise the sole zeroth-order dissipative mechanisms. Of these, it is only Δ that directly involves the behavior of the heavy species to lowest order. If ordinary hydrodynamics were instead to describe a disparate-mass mixture in the same (k, ω) regime, there would be *no* zeroth-order dissipation in the heavy species. This difference has a dramatic effect on the predicted spectral intensity. Figure 1 shows the calculated spectral intensity predicted by hydrodynamics(15) for a He–Xe mixture in the two-temperature regime (dashed line). This is essentially the spectrum for Xe alone, and the absence of a visible central peak corresponds clearly to the strong suppression of dissipation predicted by hydrodynamics. The two-temperature calculation(20) (solid line), on the other hand, shows the mark of strong dissipation resulting from the possibility of a temperature separation in the mixture. The dots are the experimental points of Letamendia *et al.*(15): the central peak gives a very clear indication of the dissipation resulting from temperature separation in the mixture.

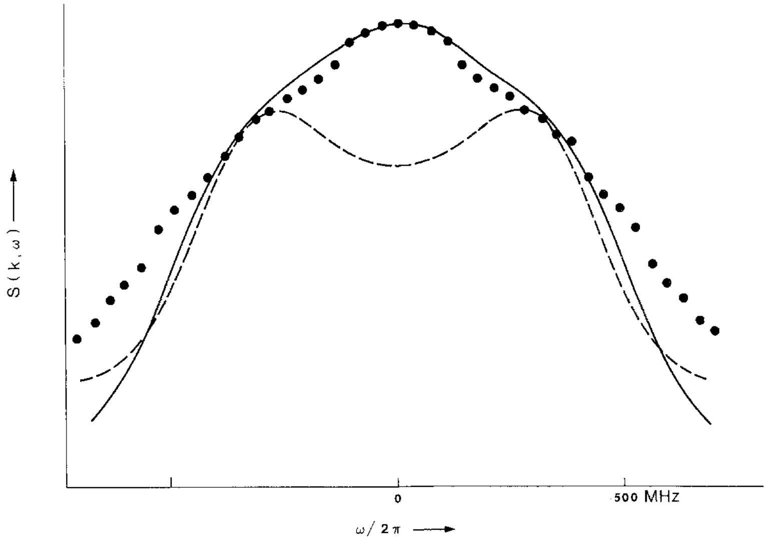

Fig. 1. Light scattering spectrum in He–Xe for experimental conditions of Fig. 3c of ref. 15: $k = 1.725 \times 10^5\,\mathrm{cm}^{-1}$, $T = 293$ K, $p(\mathrm{Xe}) = 1.82$ atm, $p(\mathrm{He}) = 2.65$ atm. Solid line, two-temperature prediction (ref. 20); dashed line, hydrodynamic prediction (ref. 15); dots, experimental points (ref. 15).

The data of ref. 15 also indicate the experimental extent of this regime. The two-temperature regime should occur for reduced wavenumbers $\tilde{k}$ of order unity, where

$$\tilde{k} \equiv kc/v_\Delta \tag{42}$$

The strong discrepancies evident in ref. 15 between experiment and the predictions of hydrodynamics for He–Xe mixtures occur for all values of k in the range

$$0.47 \leqslant \tilde{k} \leqslant 1.8 \tag{43}$$

(1.8 is the highest value of $\tilde{k}$ tested in the mixtures), as expected from the two-temperature picture.

4.2. Sound Propagation

Two-temperature sound propagation predictions were obtained by the approach of Huck and Johnson$^{(2,3,21)}$ outlined in Section 3.1. These proved of more than academic interest, since they suggested the possibility of several simultaneous "sound" modes in a disparate-mass gas mixture, for a

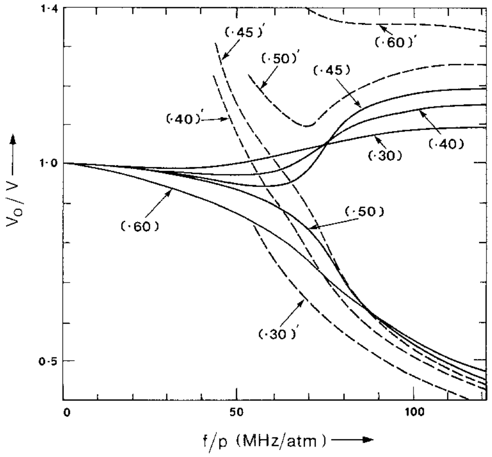

Fig. 2. Dispersion in He–Xe for various He mole fractions x_1. Solid lines: sound mode; dashed lines: interfering mode.

narrow range of compositions at a high enough frequency, or, alternatively, a dispersion which should show a large-scale, discontinuous change of behavior as a function of composition for frequencies in the two-temperature regime.[2]

Figure 2 illustrates these claims more explicitly (the value $f/p = 100$ MHz/atm corresponds approximately to $\tilde{\omega} = 1$ for the reduced frequency of Section 3.1; here $f \equiv \omega/2\pi$). At low frequencies there is no dispersion, while for moderate frequencies ($f/p \lesssim 50$ MHz/atm) the dispersion is seen to be small and to vary smoothly with He mole fraction x_1. Above some critical frequency, however, there is a dramatic change in the dispersion which can only be understood in terms of two competing roots of the dispersion relation, one going over smoothly to that describing ordinary sound propagation at low frequencies, the other a root related near $\tilde{\omega} = 0$ to dissipative processes. Figure 3 shows that within a very narrow composition range these two modes compete so closely at high enough frequencies that it makes no sense to call one or the other the mode describing sound propagation: physically, one must accept both as "sound" modes. The calculations also predict that this effect is characterized by a critical frequency $f/p \approx 70$ MHz/atm and the critical composition $x \approx 0.5$.

[2] Foch *et al.*(22) obtained predictions for sound propagation in gas mixtures in the kinetic regime which showed no sign of this effect. Their approach, however, is based on a perturbation approach in which deviations from hydrodynamic behavior are assumed small. In a disparate-mass gas in the two-temperature regime, deviations from hydrodynamic behavior are so gross that the perturbation approach of ref. 24 has an almost vanishing region (in $\tilde{\omega}$) of applicability.

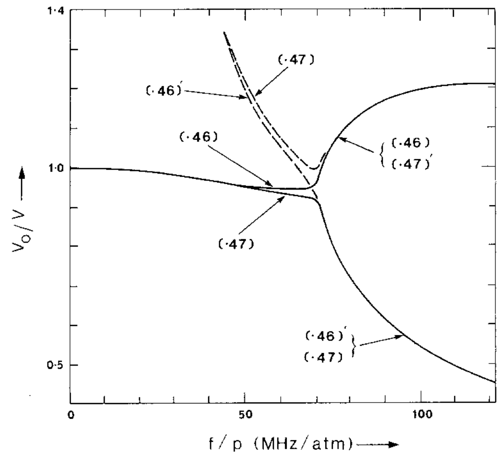

Fig. 3. Dispersion in He–Xe for He mole fractions $x_1 = 0.46 < x_c$ and $x_1 = 0.47 > x_c$. Lines as in Fig. 2. For $f/p > 70$ MHz/atm, dashed and solid lines overlap to graphical accuracy.

Experimental confirmation of this prediction is not, however, as straightforward a process as might first appear, since the calculations leading to Figs. 2 and 3 also predict$^{(21-23)}$ an unusually high absorption in the region of the critical composition and for frequencies $f/p \lesssim 70$ MHz/atm. Thus, sound dispersion in this regime can be expected to be particularly hard to measure, and one needs to take into account a variety of experimental pointers in deciding the degree to which experiment confirms theory. Because of the approximate nature of the basic equations, these pointers should be both qualitative and semiquantitative, while detailed comparison with experiment must await full many-moment kinetic calculations[3] and possibly a more extensive set of experiments.

The following are the salient features one should look for in experiment.

1. There should be a critical frequency ω_c and a critical He mole fraction x_c such that the observed dispersion shows qualitative and large-scale changes (a) for any intermediate composition (say, $0.3 \leqslant x_1 \leqslant 0.7$) as ω goes through ω_c, and (b) for fixed frequencies $\omega > \omega_c$ as x_1 increases through x_c.

2. x_c should be approximately $x_c \approx 0.5$; ω_c should correspond to an f/p value of ~ 70 MHz/atm.

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[3] These would entail a generalization to mixtures of the BGK approximation discussed in detail by Cohen *et al.*,$^{(23)}$ together with the use of realistic intermolecular forces.

3. Absorption should be particularly high near $x_1 = x_c$ and $\omega = \omega_c$.

4. For frequencies above ω_c, the dispersion V/V_0 should rise as a function of x_1 if $x_1 \not\approx x_c$ (note that for lower frequencies it falls uniformly and continuously with x_1). For $x < x_c$, at high enough frequencies, V_0/V should be greater than 1, while for $x > x_c$ the dispersion should change grossly and abruptly to values much less than 1.

Sound propagation experiments were carried out on He–Xe mixtures in the two-temperature regime by Bowler.(17–19) A summary of Bowler's experimental data on dispersion is shown in Fig. 4. The most evident points to note are that there is indeed a large-scale change in dispersion at high frequencies as x_1 goes from 0.2 to 0.8. This change occurs at roughly $x_1 \approx 0.5$ for frequencies above $f/p \approx 70$ MHz/bar (≈ 70 MHz/atm). At the highest frequencies shown, the dispersion is a steeply rising function for $x_1 \leqslant 0.45$ and a steeply falling function for $x_1 \geqslant 0.6$.

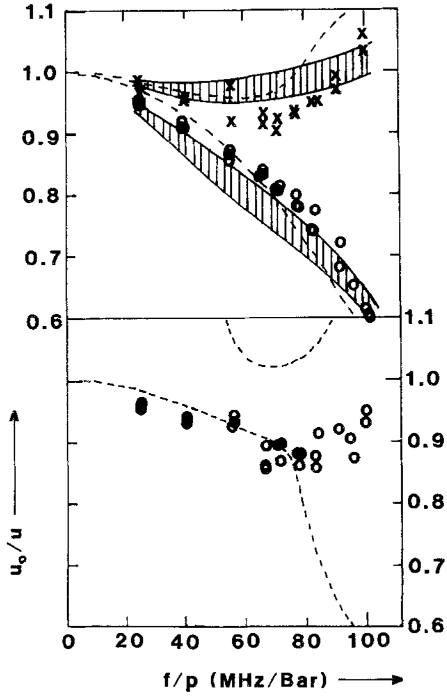

Fig. 4. (a) Upper (lower) shaded band: experimental dispersion for He mole fractions $x_1 = 0.20$, 0.30, 0.40 (0.70, 0.80); crosses, $x_1 = 0.45$; circles, $x_1 = 0.60$. Upper (lower) dashed line: two-temperature prediction for $x_1 = 0.45$ (0.60). (b) Circles, experimental dispersion, $x_1 = 0.50$. Lower (upper) dashed line: two-temperature prediction for acoustic (interfering) mode.

Bowler also measured sound absorption in the two-temperature regime,(17,19) and obtained exceptionally high absorption coefficients for x_1 in the range $0.4 \leqslant x_1 \leqslant 0.6$ and $f/p \gtrsim 70$ MHz/atm. By far the highest absorption was that observed for $x_1 = 0.5$ and frequencies $f/p \approx 100$ MHz/atm.

The dispersion observed by Bowler(17,19) confirms the predictions of Huck and Johnson,(2) in showing clear evidence of a competition between "sound" modes in a disparate-mass gas, and in verifying the predicted values for critical composition and frequency which should characterize this interference.

Interestingly, it can also be shown that a similar mode competition is predicted by the equations of ordinary hydrodynamics.(2,18,19,24) These hydrodynamic calculations predict a critical composition of the order of 0.25–0.3 and a critical frequency corresponding to $f/p \approx 84$–100 MHz/atm. This critical composition, in particular, differs widely from that found in the experiments of Bowler, which instead agrees with that predicted by two-temperature hydrodynamics. Thus, sound propagation experiments, as also light scattering, show clear evidence of the existence and importance of the predicted two-temperature regime in He–Xe gas mixtures.

5. PHYSICAL DISCUSSION

A picture is needed of the physical processes going on in a disparate-mass gas in the presence of the mode-interference effects discussed here. Referring to Figs. 2 and 3, one sees that for frequencies above the critical frequency there can be a slow wave (upper curves) or a fast wave (lower curves) in the gas, with the possibility that both may be present simultaneously if He and Xe are present in nearly equal proportions. As reference to Fig. 2 shows, the slow wave itself is characteristic of a Xe-rich mixture, while the fast wave is characteristic of one in which He predominates.

Closer study of the phase and amplitude for the deviation of the hydrodynamic variables from equilibrium for each component of the mixture separately confirms(21) that the slow wave is a damped soundlike mode primarily carried by the Xe. The predicted sound speed correlates well with that expected in pure Xe at a pressure equal to the partial pressure of the Xe in the mixture. Thus, for high enough frequencies, the heavy component in a disparate-mass gas mixture can decouple from the mixture and support its own sound wave if its partial pressure is high enough.

The fast wave, in He, is not, however, analogous to a sound wave in pure He. Instead, it may be understood as a dusty-gas mode, that is, one which could exist in He in the presence of fixed scattering centers. In such a gas, at high enough frequencies and nearly equal numbers of light-gas molecules and scattering centers, one can show that

$$V_0/V = \alpha = A\omega^{-1/2} \tag{44}$$

where α is the coefficient of absorption and A a known positive constant. One finds[(21)] that the dispersion and absorption predicted by equations (24)–(31) obey this relation well for $\omega > 2\nu_\Delta$, so that at these frequencies the fast mode should be nonpropagating in the usual sense. For lower frequencies above critical, however, the damping per wavelength of the fast wave is lower than that predicted by a dusty-gas model, and it becomes possible to consider the fast mode, too, as a propagating mode of the mixture.

The existence of a fast sound mode in He and a simultaneous slow mode in Xe is also evident in the density self-correlation functions predicted by two-temperature hydrodynamics.[(20)] Figure 5 shows a sound peak (i.e., a maximum at nonzero frequency shift) in the He–He spectral function S_{11} at a much higher frequency shift than that predicted to occur simultaneously in S_{22} for Xe. This result cannot be verified directly in light

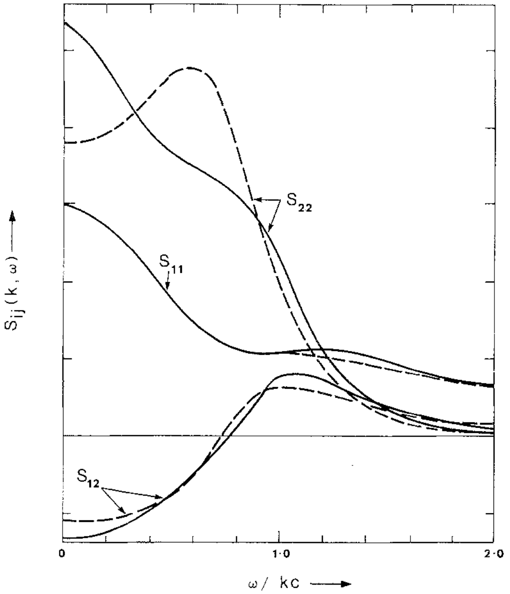

Fig. 5. Density self-correlation functions for He (S_{11}) and Xe (S_{22}), and cross-correlation S_{12} in He–Xe for equal mole fractions at $\tilde{k} = 0.7$. Solid line: two-temperature prediction; dashed line: hydrodynamic prediction.

scattering experiments, however, since these are essentially sensitive to S_{22} alone.

It is also of interest to understand the critical point itself. One finds$^{(21)}$ that in the region of this point, both modes should have identical physical characteristics: a damped soundlike mode is present in both species in the mixture, but the disturbance in the heavy species is $\pi/2$ out of phase with that of the light species. Each wave thus provides maximum damping for the other, and it is therefore not surprising that the absorption should be a maximum in this critical region.

Finally, one may enquire into the origin of the dissipative mode that becomes soundlike near the critical point. The calculations of refs. 2, 3, and 21 suggest that at low frequencies this mode is related to diffusion in the gas, being a mode in which the light and heavy species oscillate in antiphase. This identification, however, seems to depend on the precise approximations used in the calculation, and in particular on the intermolecular force assumed (through the existence of a second critical point at lower frequencies, at which there is interference between two dissipative modes).$^{(17)}$

6. FINAL COMMENTS

Mode interference effects like those discussed here can arise in various contexts. An example with formal similarity to the present one is that of propagation of electromagnetic waves in the waveguide formed by the earth plus the ionosphere.$^{(25)}$ A more direct correspondence can be sought in the propagation of sound in various composite systems. Two compressive modes have in fact been observed experimentally in a system of fluid-saturated glass beads.$^{(26)}$ It has also been suggested recently$^{(27)}$ that a slow and a fast mode can propagate in liquid water at high enough frequency as a result of a certain amount of decoupling in a water molecule between the O atom and its associated H atoms.

Finally, in He–Xe mixtures themselves, a different fast propagating mode has recently been predicted by Campa and Cohen.$^{(1)}$ The possibility of such a mode again arises from mode interference. Its properties are, however, different from that of either of the modes discussed here. The predicted mode appears to be a true He-based sound mode, with a sound velocity appropriate to a gas in which the Xe atoms have been removed; moreover, it is predicted to appear in a very different k, ω regime from that considered here. It is suggested that this mode may correspond in kind to a fast propagating mode observed recently$^{(28)}$ in a computer simulation modeling a liquid $Li_{0.8}Pb_{0.2}$ alloy.

APPENDIX. DISPARATE-MASS GAS TRANSPORT COEFFICIENTS

Transport coefficients for disparate-mass gas mixture are given by Goebel *et al.*[7] based on a 13-moment form for the distribution function of each species. A Maxwell-type (κ_{ij}/r^5) intermolecular repulsive force is assumed to exist between species i and j, where κ_{ij} is a force constant and r the intermolecular separation. In the disparate-mass approximation, one obtains for the diffusion coefficient

$$D = \frac{x_2 p_1}{A \rho_1 \rho_2} \tag{A.1}$$

for the viscosity (heavy species)

$$\mu_2 = \frac{p_2}{3 B_{22} \rho_2} \tag{A.2}$$

and for the heat fluxes

$$\lambda_1 = \frac{5kp_1}{2m_1(2B_{11}\rho_1 + A\rho_2)}, \qquad \lambda_2 = \frac{5kp_2}{4m_2 B_{22}\rho_2} \tag{A.3}$$

The frequency ν_Δ governing the temperature separation is given by

$$\nu_\Delta = 2An\mu, \qquad \mu \equiv m_1 m_2/(m_1 + m_2) \tag{A.4}$$

Here the collision parameters are defined by

$$\begin{aligned} A &= \frac{2\pi \mathcal{A}_1(5)}{m_1 + m_2}\left(\frac{\kappa_{12}}{\mu}\right)^{1/2} \\ B_{ii} &= \frac{\pi \mathcal{A}_2(5)}{m_i}\left(\frac{\kappa_{ii}}{2m_i}\right)^{1/2} \end{aligned} \tag{A.5}$$

The $\mathcal{A}_i(5)$ are tabulated constants$^{(31)}$ of magnitude about 0.4.

The dimensionless transport coefficients of equations (26)–(30) are related to these as follows:

$$\tilde{D} = D\nu_\Delta/c^2 x_2 = 2 \tag{A.6}$$

$$\tilde{\mu} = 8\mu_2 x_2/3\rho_0 D \tag{A.7}$$

$$\tilde{\lambda}_i = \lambda_i T_0/p_0 c^2 \tag{A.8}$$

$$\tilde{\tau} = \tfrac{2}{3}(1 - i\tilde{\omega})^{-1} \tag{A.9}$$

where $\tilde{\omega} \equiv \omega/\nu_\Delta$ is the reduced frequency of the disturbance being studied.

ACKNOWLEDGMENTS

I thank Paul Highton for extensive computing help, and the U.K. Science and Engineering Council for partial support. Much of this work was carried out at the University of Surrey, with partial support from the University of Surrey Research Committee.

REFERENCES

1. A. Campa and E. G. D. Cohen, *Phys. Rev. Lett.* **61**:853 (1988).
2. R. J. Huck and E. A. Johnson, *Phys. Rev. Lett.* **44**:142 (1980).
3. R. J. Huck and E. A. Johnson, *Arch. Mech.* (Warsaw) **32**:703 (1980).
4. J. D. Foch and G. W. Ford, in *Studies in Statistical Mechanics*, Vol. 5, J. de Boer and G. E. Uhlenbeck, eds. (North-Holland, Amsterdam, 1970), p. 101.
5. S. Chapman and T. G. Cowling, *Mathematical Theory of Non-Uniform Gases*, 3rd ed. (Cambridge University Press, 1970).
6. H. Grad, in *Rarefied Gas Dynamics*, Vol. 3, F. M. Devienne, ed. (Pergamon, New York, 1960), p. 100.
7. C. J. Goebel, S. M. Harris, and E. A. Johnson, *Phys. Fluids* **19**:627 (1976).
8. E. A. Johnson, *Arch. Mech.* (Warsaw) **28**:803 (1976).
9. C. J. Goebel, S. M. Harris, and E. A. Johnson, in *Rarefied Gas Dynamics*, Vol. 51, J. L. Potter, ed. (American Institute of Aeronautics and Astronautics, New York, 1977), Part I, p. 109.
10. H. Grad, *Commun. Pure Appl. Math.* **11**:331 (1949); in *Handbuch der Physik*, S. Flügge, ed. (Springer-Verlag, Berlin, 1958), Vol. XII, Section 28, p. 267.
11. J. P. Boon and S. Yip, *Molecular Hydrodynamics* (McGraw-Hill, New York, 1980).
12. C. D. Boley and S. Yip, *Phys. Fluids* **15**:1424 (1972); *Phys. Fluids* **15**:1433 (1972); *J. Phys.* (Paris) **33**:C1-43 (1972).
13. N. A. Clark, *Phys. Rev. A* **12**:2092 (1975).
14. E. W. Washburn, ed., *International Critical Tables* (McGraw-Hill, New York, 1930), Vol. VII, pp. 7–8.
15. L. Letamendia, J. P. Chabrat, G. Nouchi, J. Rouch, C. Vaucamps, and S.-H. Chen, *Phys. Rev. A* **24**:1574 (1981).
16. L. Letamendia, P. Joubert, J. P. Chabrat, J. Rouch, C. Vaucamps, C. D. Boley, S. Yip, and S.-H. Chen, *Phys. Rev. A* **25**:481 (1982).
17. J. R. Bowler, *Harmonic modes in binary monatomic gas mixtures*, Ph.D. thesis, University of Surrey, United Kingdom (1984).
18. J. R. Bowler and E. A. Johnson, *Phys. Rev. Lett.* **54**:329 (1985).
19. J. R. Bowler and E. A. Johnson, *Proc. R. Soc. Lond. A* **408**:79 (1986).
20. E. A. Johnson, *Phys. Rev. A* **27**:1146 (1983).
21. R. J. Huck and E. A. Johnson, in *Rarefied Gas Dynamics*, S. S. Fisher, ed. (American Institute of Astronautics and Aeronautics, New York, 1981), p. 452.
22. J. D. Foch, G. E. Uhlenbeck, and M. Fuentes Losa, *Phys. Fluids* **15**:1224 (1972).
23. E. G. D. Cohen, in *Trends in Applications of Pure Mathematics to Mechanics*, E. Kroner and K. Kirchgasser, eds. (Springer-Verlag, Berlin, 1986), p. 2; B. Kamgar-Parsi, E. G. D. Cohen, and I. M. de Schepper, *Phys. Rev. A* **35**:4781 (1987); B. Kamgar-Parsi and E. G. D. Cohen, *Physica A* **138**:249 (1986).
24. B. Kamgar-Parsi and E. G. D. Cohen, *Int. J. Thermophys.* **7**:395 (1986), and private communication.
25. K. G. Budden and M. Eve, *Proc. R. Soc. Lond. A* **342**:175 (1975) and references therein.
26. J. G. Plona, *Appl. Phys. Lett.* **36**:259 (1981).
27. M. A. Ricci, D. Rocca, G. Ruocco, and R. Vallauri, *Phys. Rev. Lett.* **61**:1958 (1988).
28. J. Bosse, G. Jacucci, M. Ronchetti, and W. Schirmacher, *Phys. Rev. Lett.* **57**:3277 (1986).