Light-scattering predictions in the two-temperature regime for disparate-mass gas mixtures

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Predictions are obtained for light-scattering spectra for disparate-mass gas mixtures at wave numbers such that species temperature separation produces strong deviations from hydrodynamic predictions. Recent light-scattering experiments in xenon-helium mixtures by Letamendia *et al.* show good qualitative agreement with the present approximate predictions, thus providing the first experimental confirmation of the existence of a two-temperature regime in disparate-mass neutral gases. Physical reasons are suggested for the strong disagreement between two-temperature and hydrodynamic spectral predictions in the two-temperature regime.

I. INTRODUCTION

For a pure monatomic gas in equilibrium, the spectral intensity $S(k,\omega)$ of light scattered from density fluctuations in the gas behaves in a well-known fashion,[1,2] as the wave number k of the density fluctuations being probed increases from $k \ll l^{-1}$ to $k \gg l^{-1}$, where l is a characteristic microscopic length of the order of a mean-free path in the gas. The small-k spectrum may be correctly derived from hydrodynamics,[3,4] but deviations from hydrodynamic behavior[2,4] are expected to become important when k becomes as large as $\sim l^{-1}$.

For more complicated gases, it can be more illuminating to characterize possible internal processes by relaxation times, rather than by "mean-free paths." In these terms, a convenient definition of l for a pure monatomic gas is

$$l \equiv \frac{c}{\omega_0} , \tag{1}$$

where

$$c \equiv \left[\frac{p_0}{\rho_0} \right]^{1/2} \tag{2}$$

is proportional to the equilibrium speed of sound in the gas, and ω_0 is a frequency characteristic of the relaxation to equilibrium of a disturbance (say, in momentum) in the gas; p_0 and ρ_0 are the equilibrium static pressure and the mass density, respectively.

For pure monatomic gases, and usual mixtures of such gases, the criterion $k \ll l^{-1}$ for hydrodynamic behavior is well defined because all intrinsic relaxa-

tion frequencies in the gas have the same order of magnitude. It was pointed out some time ago by Grad,[5] however, that in a binary mixture of gases of very different molecular masses, so that

$$\left[\frac{m_1}{m_2} \right]^{1/2} = \delta \ll 1 , \tag{3}$$

there is a hierarchy of intrinsic relaxation frequencies, of different orders of magnitude. If species number densities and intermolecular forces are comparable, a disparate-mass mixture of monatomic gases can be characterized by the three intrinsic relaxation frequencies ω_1, ω_2, and ω_Δ, where[5]

$$\delta^2 \omega_1 \simeq \delta \omega_2 \simeq \omega_\Delta . \tag{4}$$

In Eq. (4) ω_1 is a frequency characterizing the relaxation of the light species to local equilibrium at species temperature T_1, ω_2 characterizes relaxation of the heavy species to local equilibrium at its own temperature T_2; and the relaxation of the species temperature-separation Δ,

$$\Delta \equiv T_2 - T_1 , \tag{5}$$

is characterized by the still lower frequency ω_Δ. The epochal relaxation implied by Eq. (4) arises from the inefficient exchange of kinetic energy in collisions between light and heavy molecules, a fact which can lead to separate species temperatures in some circumstances which would otherwise be called "hydrodynamic."

Light-scattering behavior in a gas will only be governed by ordinary hydrodynamics for wave numbers

$$k \ll \left| \frac{\omega'}{c} \right| , \tag{6}$$

where ω' is the lowest intrinsic relaxation frequency in the gas. Although ω' is commonly supposed to be of the same order as the relaxation frequency of a species to local equilibrium, Eq. (4) shows that this is not true for a disparate-mass gas, and that the predictions of hydrodynamics should begin to break down at the much lower wave numbers $k \simeq (\omega_\Delta / c)$.

Equations governing disparate-mass gases have been derived by a number of authors,[6-11] with essential agreement. It has been shown that there should exist a two-temperature regime in which the gas is governed by "near-normal" equations similar to those of hydrodynamics except that they allow for temperature separation between species. For light scattering, this regime is determined roughly by

$$\left| \frac{\omega_\Delta}{c} \right| \lesssim k \ll \left| \frac{\omega_2}{c} \right| . \tag{7}$$

Here, approximate spectral predictions for xenon-helium mixtures are derived from two-temperature equations for disparate-mass gases. The predictions are compared with those of conventional hydrodynamics (using the same approximations), and with recent experiments of Letamendia et al.[12] It will be seen that present predictions are in good qualitative agreement with experiment, and in strong disagreement with hydrodynamic predictions, in the two-temperature regime. The experimental results of Letamendia et al.[12] thus give what appears to be the first experimental confirmation of significant epochal relaxation effects in disparate-mass mixtures of ordinary neutral gases.

II. DETAILS OF THE CALCULATION

Theoretical calculations are based on the two-temperature equations for disparate-mass gas mixtures derived by Goebel, Harris, and Johnson,[6-8] linearized for small deviations from absolute equilibrium, and taken to Navier-Stokes level [i.e., to first order in the small parameter δ of Eq. (3)]. The equations apply to mixtures of Maxwell molecules (molecules interacting with repulsive forces $\propto r^{-5}$, where r is the intermolecular separation). It is also assumed that species number densities n_i are of comparable magnitudes, as are all intermolecular forces.

The linear two-temperature equations obtained thus from Refs. 6−8 may be written as follows.
Flow equations:

$$\frac{\partial n_1}{\partial t} + n_{10} \vec{\nabla} \cdot (\vec{U} - \vec{W}) = 0 , \tag{8}$$

$$\frac{\partial n_2}{\partial t} + n_{20} \vec{\nabla} \cdot \vec{U} = 0 , \tag{9}$$

$$\rho_2 \frac{\partial \vec{U}}{\partial t} + \vec{\nabla} p + \vec{\nabla} \cdot \{ \overleftrightarrow{P}_2 \} = 0 , \tag{10}$$

$$\frac{\partial T}{\partial t} + \tfrac{2}{3} T_0 \vec{\nabla} \cdot (\vec{U} - x_1 \vec{W}) + \frac{2}{3 n_0 k} \vec{\nabla} \cdot (\vec{q}_1 + \vec{q}_2) = 0 . \tag{11}$$

Constitutive relations:

$$\vec{W} = (D / x_2) \vec{\nabla} \ln p_1 , \tag{12}$$

$$\{ \overleftrightarrow{P}_2 \} = -2 \mu_2 \{ \vec{\nabla} \vec{U} \} , \tag{13}$$

$$\vec{q}_1 = -\lambda_1 \vec{\nabla} T_1 , \tag{14}$$

$$\vec{q}_2 = -\lambda_2 \vec{\nabla} T_2 . \tag{15}$$

Relaxation equation for Δ:

$$\frac{\partial \Delta}{\partial t} - \frac{2}{3 k n_0} \vec{\nabla} \cdot \left[\frac{\vec{q}_1}{x_1} - \frac{\vec{q}_2}{x_2} \right] + \tfrac{2}{3} T_0 \vec{\nabla} \cdot \vec{W} = -\omega_\Delta \Delta . \tag{16}$$

Here ρ_i is mass density, p_i partial pressure, $\vec{q}_i$ heat flux, and x_i the equilibrium mole fraction, for species i. The overall number density is $n = n_1 + n_1$, and $\vec{W}$ is the diffusion velocity

$$\vec{W} \equiv \vec{U}_2 - \vec{U}_1 , \tag{17}$$

where $\vec{U}_i$ is the flow velocity of species i. $\vec{U}$ and T are the overall flow velocity and temperature, respectively, according to the conventional kinetic-theory definitions, and k is Boltzmann's constant. Suffix "0" indicates a value taken at absolute equilibrium. $\{ \overleftrightarrow{P}_2 \}$ indicates the symmetric traceless part of the pressure tensor of species 2 only. D is the usual coefficient of diffusion[13]; μ_2 and λ_2 are the conventional coefficients of viscosity and thermal conductivity, respectively, for the pure heavy species, while λ_1 is given by Eqs. (37), (38) of Ref. 6. Finally, the relaxation frequency ω_Δ for the temperature separation is given[6] by

$$\omega_\Delta = \frac{2 c^2 x_2}{D} . \tag{18}$$

Values for the transport coefficients in the governing equations (8)−(16) have been obtained by using as input the experimental values[14] for the dif-

fusion coefficient D for a xenon-helium mixture, and the helium and xenon pure-gas viscosities, at 0 °C, together with all the Maxwell-molecule relations given in Ref. 6. These fix the values of all coefficients in Eqs. (8)—(16) for any temperature, pressure, and composition.

The way in which the spectral function $S(k,\omega)$ may be obtained from the governing equations for a binary gas mixture is reviewed by Boley and Yip,[15] and Clark,[16] (among others). One further piece of experimental information necessary for deriving spectral predictions from Eqs. (8)—(16) is the ratio of species polarizabilities α_1/α_2. The value used in the present calculations is[17]

$$a \equiv \frac{\alpha_1}{\alpha_2} = 0.051\,41 \ , \tag{19}$$

the ratio of He-Xe polarizabilities for light of wavelength 6328 Å.

It is well known[18] that the monatomic pure-gas spectral intensity $S(k,\omega)$ for density fluctuations is a function of only two dimensionless variables, a reduced wave number and reduced frequency. Here it is convenient to define the reduced wave number $\tilde{k}$ to be

$$\tilde{k} \equiv \frac{kc}{\omega_\Delta} \tag{20}$$

and the reduced frequency to be $\tilde{\omega}/\tilde{k}$, where

$$\tilde{\omega} \equiv \frac{\omega}{\omega_\Delta} \ , \tag{21}$$

and

$$\frac{\tilde{\omega}}{\tilde{k}} = \frac{\omega}{kc} = \left[\frac{kT_0}{x_2 m_2} \right]^{1/2} \frac{\omega}{k} \tag{22}$$

correct to order δ. The spectral shape $S(k,\omega)$ for a given disparate-mass mixture is then a function only of $\tilde{k}$, $(\tilde{\omega}/\tilde{k})$, and composition x_i. The equilibrium sound peak is fixed at $(\tilde{\omega}/\tilde{k}) = (5/3)^{1/2}$, with this choice of variables. The two-temperature predictions of Eqs. (8)—(16) should begin to disagree with those of hydrodynamics as $\tilde{k}$ approaches unity. The two-temperature predictions themselves should become invalid as $\tilde{k}$ approaches ω_2/ω_Δ, according to Eq. (7). One may choose ω_2 as the relaxation frequency for the heavy-species pressure tensor,[6]

$$\omega_2 = \frac{p_{20}}{\mu_2} \ . \tag{23}$$

Then Eq. (23), together with the experimental values for the coefficients of mixture diffusion and pure species viscosity, implies that for xenon-

helium mixtures the two-temperature spectral predictions derived from Eqs. (8)—(16) should be valid so long as $\tilde{k} \ll 6.95 x_2$.

The hydrodynamic predictions also obtained here are those derived from single-temperature equations for binary mixtures of Maxwell molecules, using the same experimental input, and with the same approximation that terms higher than first order in the small parameter δ have been neglected.

III. THEORETICAL PREDICTIONS

Figures 1—4 show two-temperature predictions for the spectral function $S(k,\omega)$ for light scattering from density fluctuations in xenon-helium mixtures, together with the comparable hydrodynamic predictions, calculated as described in Sec. II. For each graph, the spectra are normalized to equal areas, but normalization between graphs is arbitrary. For small $\tilde{k}$ (not shown), two-temperature predictions agree with those of hydrodynamics.

When $\tilde{k}$ is as big as 0.3, mild discrepancies begin to appear between two-temperature and hydrodynamic predictions, especially for smaller helium proportions x_1. Also observable at $\tilde{k} = 0.3$ is the strong broadening of the Brillouin peak that occurs as x_1 increases [see Figs. 1(a)—1(c)]. Such a broadening was observed experimentally by Gornall and Wang[19] for $x_1 = 0.6$ at reduced wave number $\tilde{k} = 0.297$ (for experiment at 0 °C).

As $\tilde{k}$ increases, the two-temperature spectra differ increasingly from those predicted by hydrodynamics, so much so that for $\tilde{k} \approx 1$ there is a major qualitative difference between the predictions of the two theories. Roughly, hydrodynamics predicts a strong suppression of the central peak at wave numbers $\tilde{k} \approx 1$, with a resultant line shape similar to that of a pure isothermal gas[20] of the heavy species at its own partial pressure. In contrast, a strong central peak remains a feature of the two-temperature predictions throughout the wave numbers considered here.

IV. COMPARISON WITH EXPERIMENT

A. Qualitative Comparison

Extensive experimental results for light-scattering spectra for xenon-helium mixtures have recently been published by Letamendia *et al.*[12] Large-scale systematic differences between experiment and the predictions of hydrodynamics have been observed in
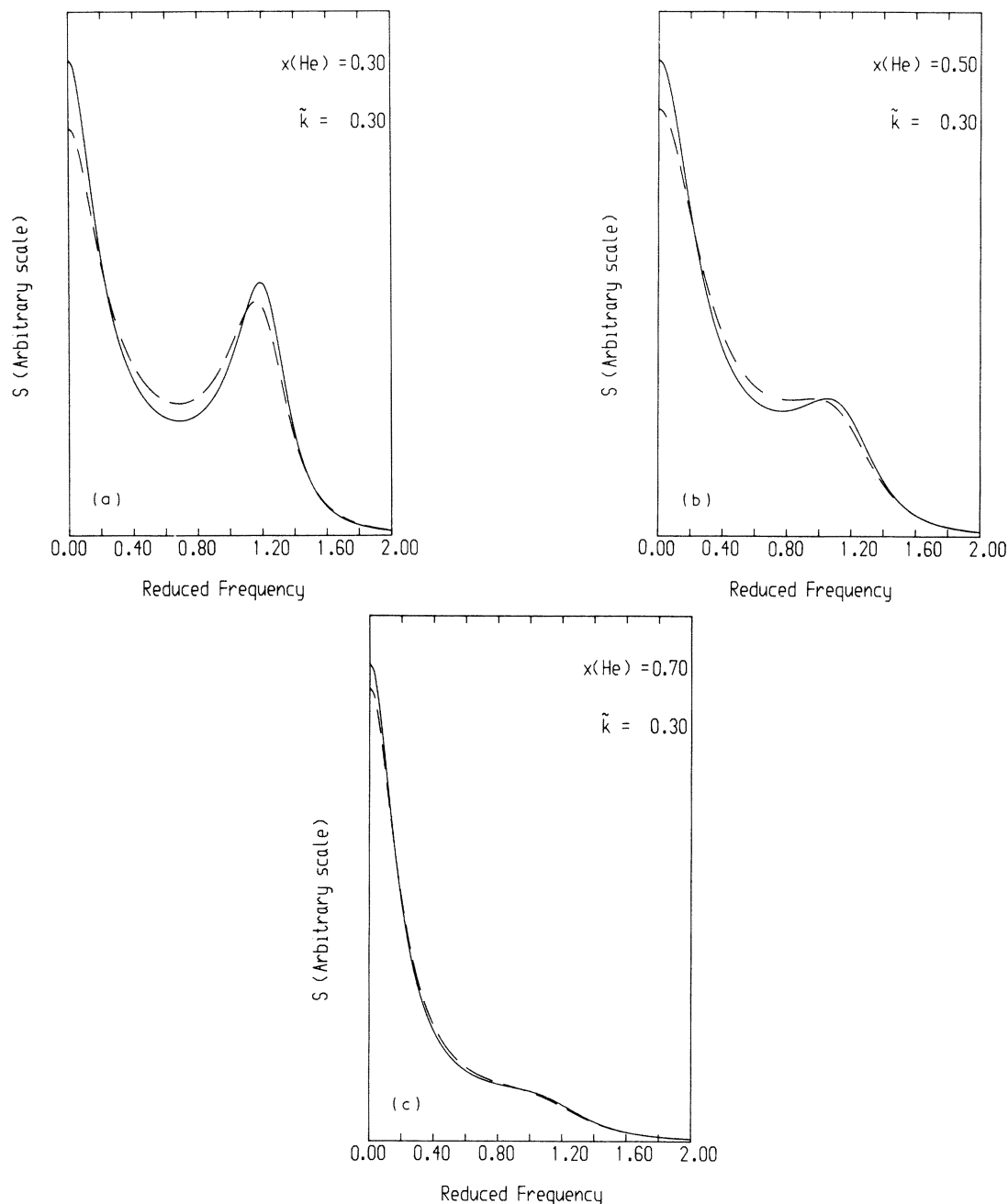
FIG. 1. Theoretical light-scattering spectra $S(k,\omega)$ for xenon-helium mixtures at reduced wave number $\tilde{k}=0.3$, for helium mole fractions (a) $x(\text{He})=0.3$, (b) $x(\text{He})=0.5$, (c) $x(\text{He})=0.7$. Solid lines, two-temperature predictions; dashed lines, hydrodynamic predictions calculated as described in Sec. II herein. Vertical scale is arbitrary. Curves on each graph preserve equal areas.

these experiments, for some ranges of the experimental conditions. A list of the results of Ref. 12 for which $n_1 \sim n_2$ is given in Table I, together with pertinent experimental conditions and the corresponding reduced wave number defined by Eq. (20). In the final column is reproduced the assessment of

Letamendia et al.[12] as to whether the given experimental spectrum did (H) or did not (NH) agree with the predictions of hydrodynamics (as calculated by those authors).

The most striking comparisons between theory and experiment arise at large wave numbers, where
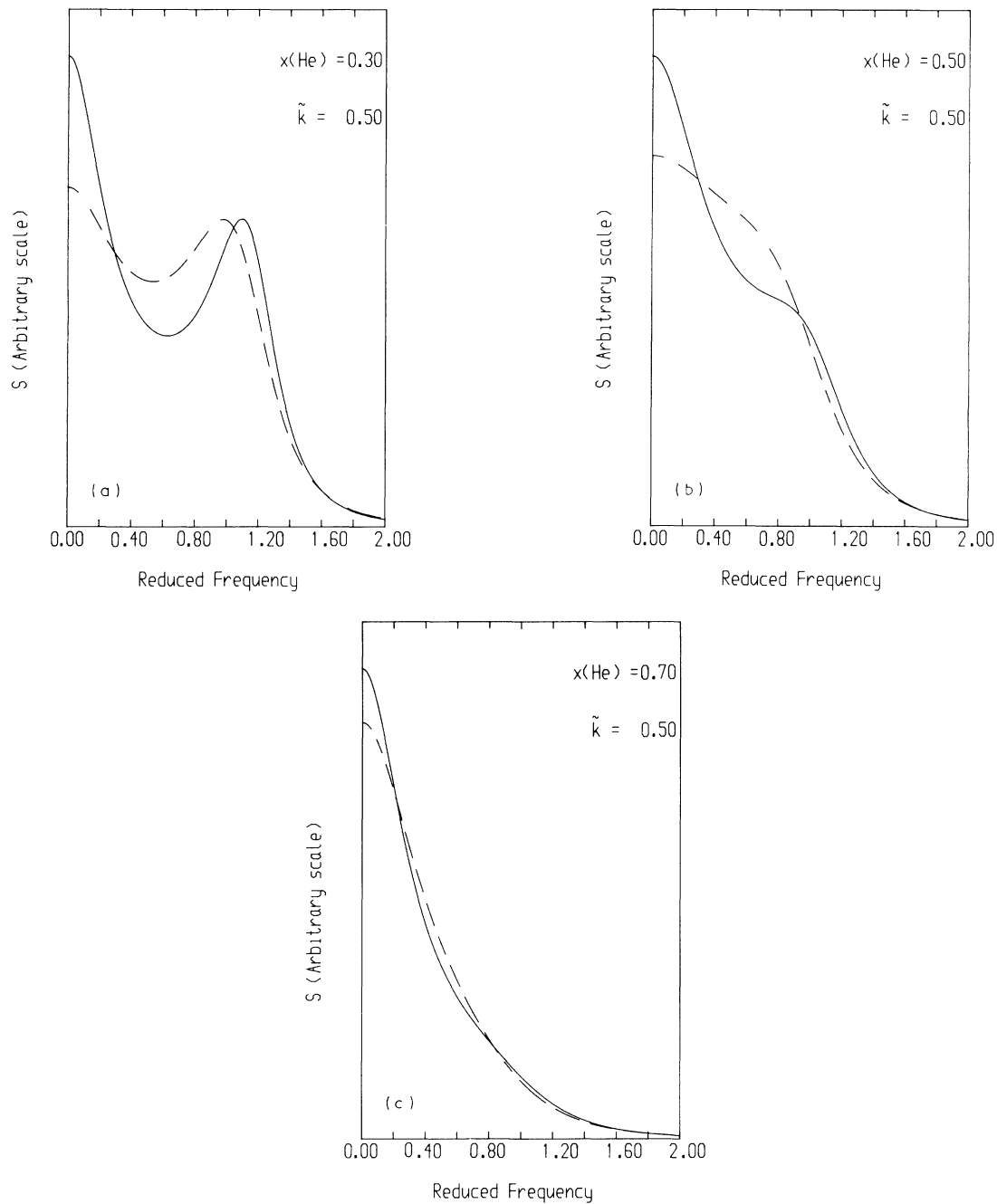
FIG. 2. As in Fig. 1, for $\tilde{k} = 0.5$.

hydrodynamics predicts strong suppression, or absence, of a central peak, in contradiction both to experiment and to the predictions of the present two-temperature predictions.

Other qualitative points may also be made. (i) Wave-number dependence: Both experimentally[12] and theoretically, disagreement of spectra with hydrodynamic predictions becomes noticeable roughly for $\tilde{k} \gtrsim 0.3$. (ii) Composition dependence: Both experiment[12] and theory show that nonhydrodynamic behavior becomes significant at lower $\tilde{k}$ for those mixtures containing less helium. (iii) Sharpening of Brillouin peaks, compared to hydrodynamic predictions, for intermediate wave numbers $0.4 \lesssim \tilde{k} \lesssim 0.6$: This is evident in both experiment[12] and theory, especially for $x_1 \lesssim 0.5$.
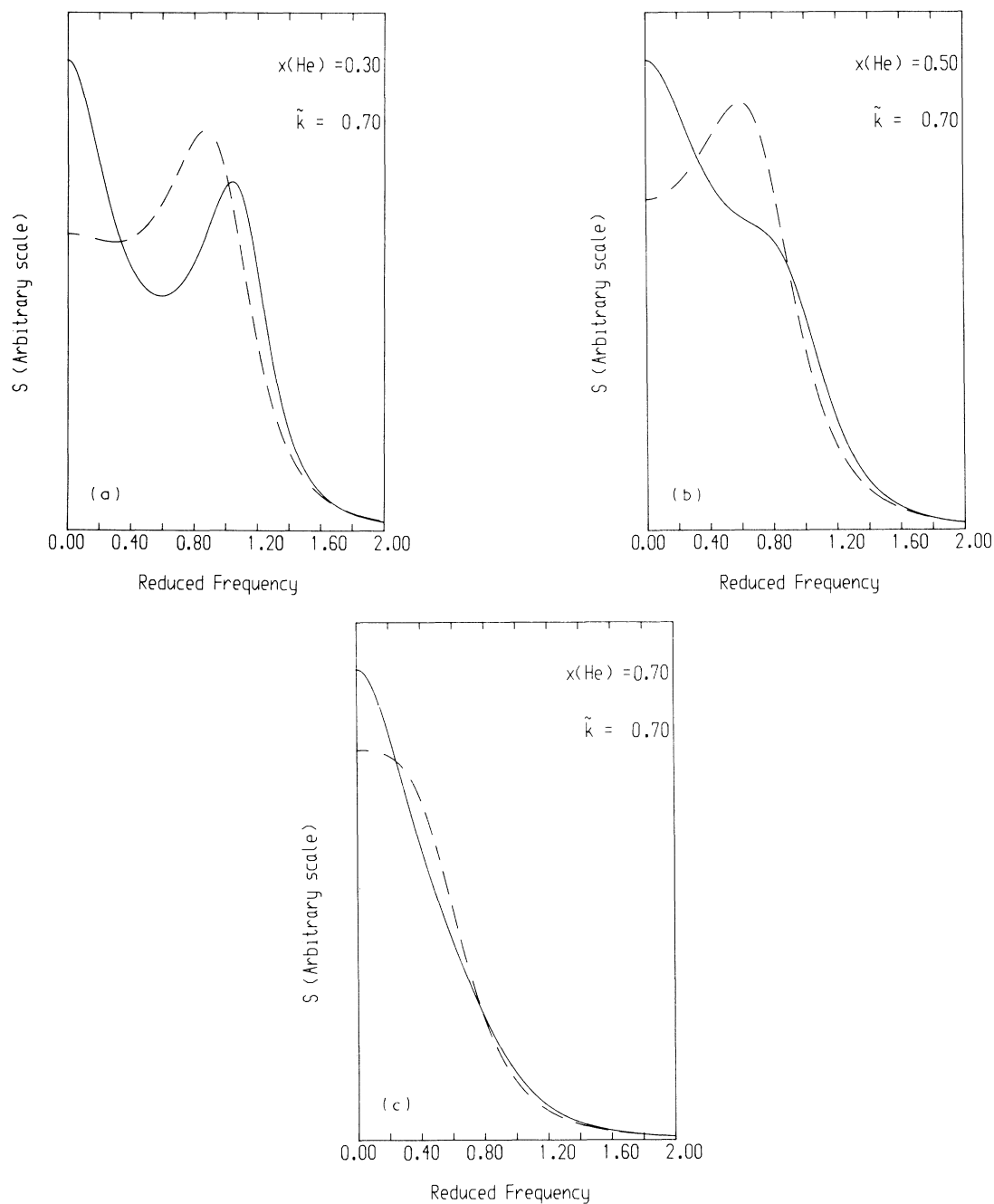
FIG. 3. As in Fig. 1, for $\tilde{k}=0.7$.

B. Quantitative Comparisons

In Figs. 5 — 8 are shown quantitative comparisons of present predictions with representative experimental spectra from Ref. 12, together with the theoretical spectra calculated on the basis of single-temperature hydrodynamics by the same authors.[12] The experimental spectra and hydrodynamic predic-

tions have been taken from the relevant published graphs of Ref. 12. Two-temperature spectral predictions have been normalized to fit experiment at zero frequency shift.

Although good quantitative agreement cannot be expected between experiment and the present two-temperature predictions, because of the approximations made in deducing the latter, it is evident from
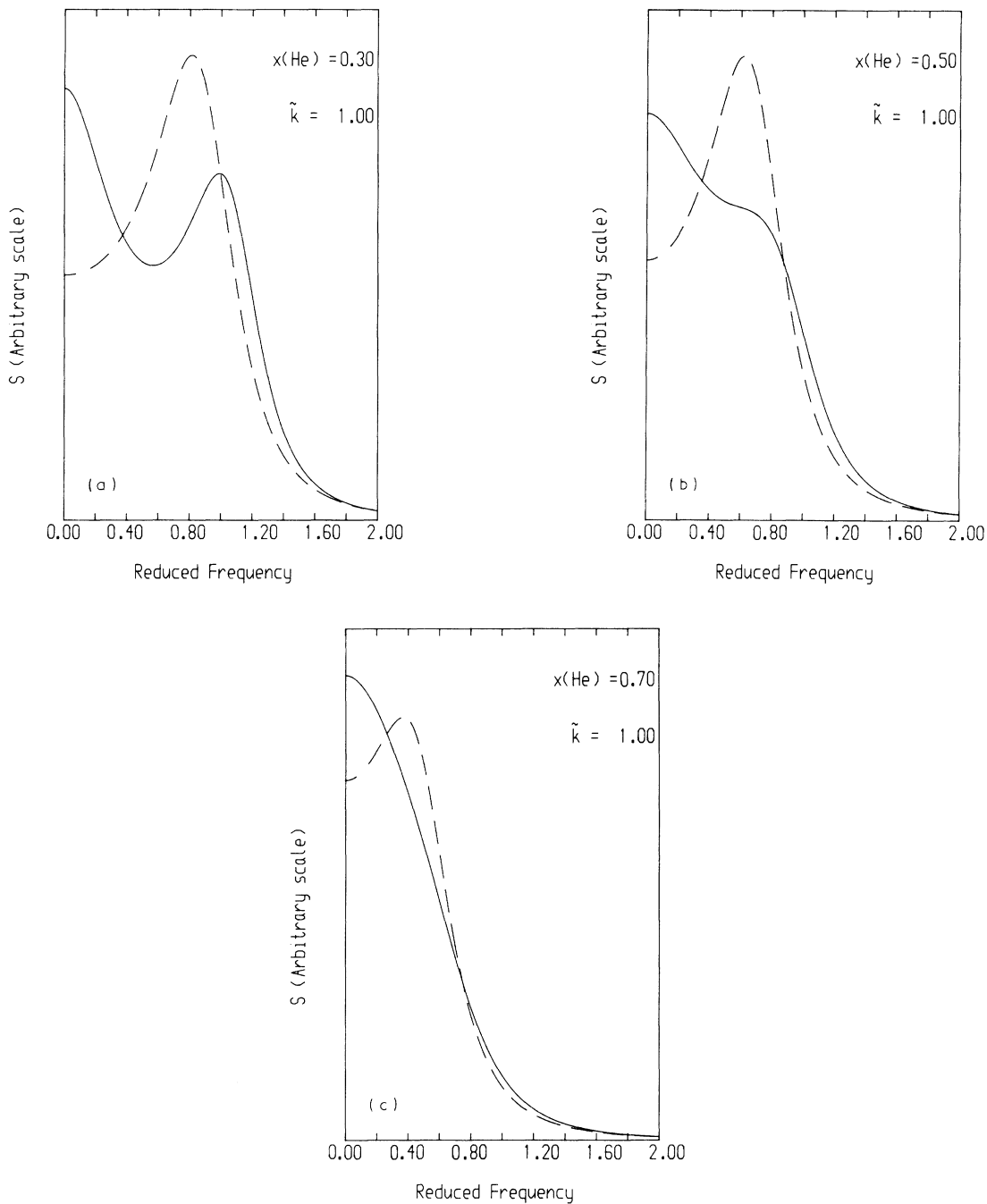
FIG. 4. As in Fig. 1, for $\tilde{k} = 1.0$.

Figs. 5—8 that a large measure of agreement is, in fact, present. Possible sources of the remaining disagreements include the neglect of thermal diffusion (inherent in the assumption of Maxwell intermolecular forces), and "convolution"-induced discrepancies.

1. Possible convolution-induced discrepancies

At large frequency shifts, Figs. 5—8 show that calculations fall off much more rapidly than experimental spectra or the theoretical predictions of Ref.

TABLE I. List of relevant experiments from Ref. 12, showing reduced wave numbers $\tilde{k}$ together with agreement (H) or nonagreement (NH) of experiment with hydrodynamic predictions, according to Ref. 12, Table II and Figs. 3—7.

Fig. no. (Ref. 12)	p_2 (atm)	p_1 (atm)	k (cm^{-1})	x_1	$\tilde{k}$	H or NH (Ref. 12)
3(b)	1.82	0.39	1.725×10^5	0.18	1.81	NH
(c)	1.82	2.65	1.725×10^5	0.59	1.27	NH
4(c)[a]	3.75	1.81	1.727×10^5	0.33	0.80	NH
(d)[a]	3.75	3.51	1.727×10^5	0.48	0.70	NH
(e)[a]	3.75	6.45	1.727×10^5	0.63	0.59	NH
5(d)	5.97	3.80	1.727×10^5	0.39	0.48	NH
(e)	5.97	4.16	1.727×10^5	0.41	0.47	NH
(f)	5.97	9.17	1.727×10^5	0.61	0.38	H
6(b)	7.32	1.59	1.727×10^5	0.18	0.45	NH
(c)	7.32	3.28	1.727×10^5	0.31	0.41	NH
(d)	7.32	7.14	1.727×10^5	0.49	0.35	NH
(e)	7.32	10.42	1.727×10^5	0.59	0.32	H
(f)	7.32	13.14	1.727×10^5	0.64	0.30	H
7(b)	2.66	1.11	0.6316×10^5	0.29	0.42	NH
(c)	2.66	6.9	0.6316×10^5	0.72	0.26	H
(d)	8.5	4.0	0.6316×10^5	0.32	0.13	H
(e)	8.5	4.99	0.6316×10^5	0.37	0.12	H
(f)	8.5	6.8	0.6316×10^5	0.44	0.12	H

[a]These figures have been assumed to correspond to the experimental conditions given in Table II of Ref. 12 rather than to the conditions stated in the caption to Fig. 4 therein.

12. This slow falloff of the latter spectra at large frequency shifts might result from a contribution from overlapping spectral orders which has been incorporated into the theoretical spectra shown in
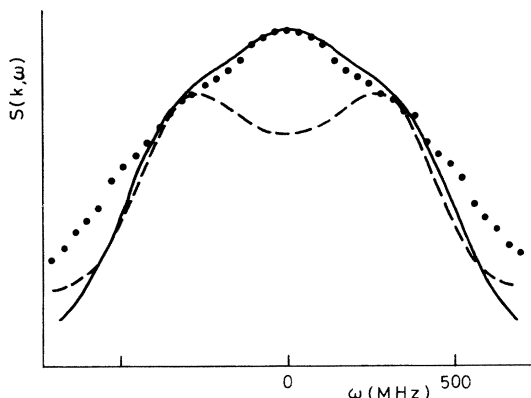

FIG. 5. Light-scattering spectra for xenon-helium mixture under experimental conditions given in Fig. 3(c) of Ref. 12: $k = 1.725 \times 10^5 \text{cm}^{-1}$, $T = 293$ K, $p(\text{Xe}) = 1.82$ atm, $p(\text{He}) = 2.65$ atm. Solid line, two-temperature prediction; dashed line, hydrodynamic prediction of Ref. 12; dots, experimental points (Ref. 12).

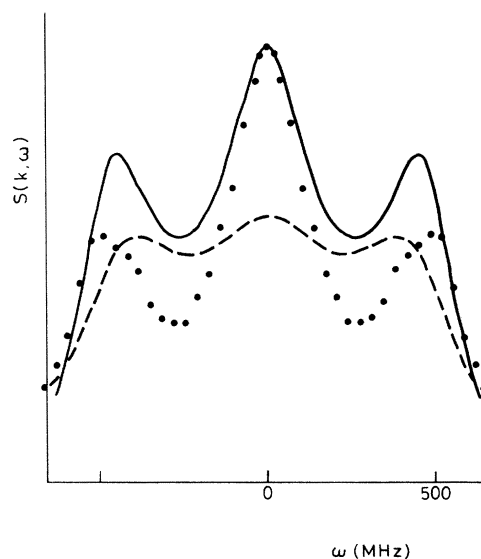FIG. 6. As in Fig. 6, for Fig. 4(c) of Ref. 12: $k = 1.727 \times 10^5$ cm^{-1}, $p(\text{Xe}) = 3.75$ atm, $p(\text{He}) = 1.81$ atm. [It is assumed that Fig. 4(c) of Ref. 12 refers to experimental conditions given in Table II of that reference rather than to the conditions stated in the caption to Fig. 4 therein.]
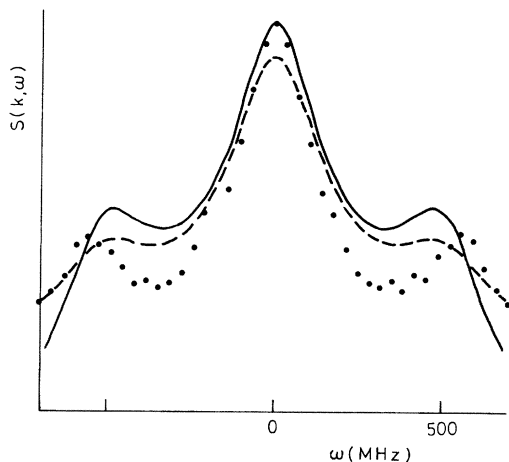
FIG. 7. As in Fig. 6, for Fig. 5(d) of Ref. 12: $k = 1.727 \times 10^5$ cm^{-1}, $p(Xe) = 5.97$ atm, $p(He) = 3.80$ atm.

Ref. 12 and reproduced here. Reference 12 does not contain sufficiently detailed information to allow this suggestion to be checked.

2. Effect of thermal diffusion

Letamendia et al.[12] have shown that spectral shape in xenon-helium mixtures is very sensitive to the presence and magnitude of thermal-diffusion effects (see, in particular, Fig. 10 of Ref. 12).[21] The results presented here apply to mixtures of Maxwell molecules, for which thermal diffusion is automatically absent. One may, nevertheless, estimate the
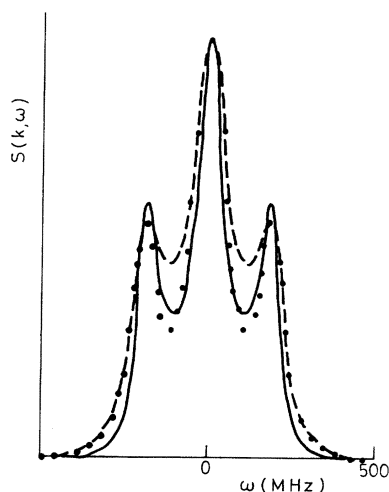

FIG. 8. As in Fig. 6, for Fig. 7(b) of Ref. 12: $k = 0.6316 \times 10^5$ cm^{-1}, $p(Xe) = 2.66$ atm, $p(He) = 1.11$ atm.

degree to which these effects would modify present results.

The generalizations of the two-temperature equations (8)—(16), for general intermolecular forces, may be obtained from Burgers.[22] Apart from minor redefinitions of the coefficients appearing in Eqs. (12)—(16), the only modifications are those to Eqs. (12) and (14), giving instead

$$\vec{W} + z(\vec{q}_1/p_{10}) = (D/x_2)\vec{\nabla}\ln p_1 \ , \qquad (12')$$

$$\vec{q}_1 + bz(p_{10}\vec{W}) = -\lambda_1\vec{\nabla}T_1 \ . \qquad (14')$$

Here b is of order unity, and z depends on the intermolecular forces via a mass-independent ratio of collision integrals.[23] The presence of new terms in z in Eqs. (12') and (14') indicates that the associated thermal-diffusion effect is present to zero order in δ, and thus if $z \sim 1$ it should be comparable with the other zero-order dissipative effects (ordinary diffusion, light-species heat flux, and relaxation of Δ). z is small, however. For inverse ν-power forces[24]

$$z = \frac{(5 - \nu)}{5(\nu - 1)} \ , \qquad (24)$$

so that it is reasonable to expect

$$|z| \lesssim (1/5) \ , \qquad (25)$$

its value for hard spheres. This should be compared with the value taken by δ for xenon-helium mixtures,

$$\delta = (1/5.7) \ . \qquad (26)$$

Equations (25) and (26) indicate that thermal diffusion contributes effects comparable in magnitude to other first-order dissipative contributions (heavy-species heat flux and viscosity), in xenon-helium mixtures. Calculations with and without these other first-order dissipative corrections have shown that their effect is quantitatively significant (in peak broadening and dispersion), but that they do not change the major qualitative features of the predicted spectra.

V. PHYSICAL DISCUSSION

There are large-scale qualitative differences between two-temperature spectral predictions and the corresponding hydrodynamic predictions, for reduced wave numbers $\tilde{k} \sim 1$, with experimental results confirming the former predictions rather than the latter. An understanding of these differences is needed in physical terms.

For xenon-helium mixtures with $x_1 \sim x_2$, light-

scattering probes the behavior of the heavy species alone. This follows from decomposition of $S(k,\omega)$ into the sum[15]

$$S = N(x_1 a^2 S_{11} + 2x_1 a S_{12} + S_{12} + S_{22}) \ , \qquad (27)$$

where N is an overall normalization, S_{ii} characterizes the i-species density fluctuation self-correlation, and S_{ij} the cross-correlation; since $a \ll 1$ [Eq. (19)], the spectrum is dominated by S_{22}.

In the two-temperature regime, S_{22} describes a heavy species which is significantly decoupled from the light species, a decoupling also predicted for sound propagation, as predicted by either hydrodynamics or the two-temperature equations, as shown by Huck and Johnson.[25,26] This decoupling may also be observed in Fig. 9, which compares different spectral contributions S_{ij} for $x_1 = x_2 = 0.5$ and $\tilde{k} = 0.7$.

Finally it has been shown[6-8] that for $\tilde{k} \sim 1$, dissipative mechanisms available to the heavy species are strongly suppressed,[27] with the sole exception of relaxation of the temperature difference Δ. Since temperature separation is not allowed by conventional hydrodynamics, only weak dissipation is predicted for the heavy species in a regime in which it is decoupled to a high degree from the light species, with the resulting strong suppression of a central peak in the hydrodynamic spectral predictions.

The observation of a strong central peak in the experiments of Letamendia et al.[12] may thus be taken as evidence of the important role of temperature separation (and its relaxation) in disparate-mass mixtures in this regime.
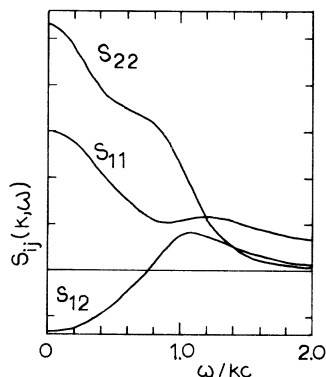
VI. RELATION TO KINETIC CALCULATIONS

Alternative spectral calculations for xenon-helium mixtures have recently been made by Letamendia et al.,[28] using a high-order model of the full coupled Boltzmann equations. These theoretical spectra show good qualitative agreement with experiment, and in particular retain the strong central peak predicted by the two-temperature approach. This seems to imply that a model equation of high enough order includes sufficient information about the full mixture Boltzmann equation to reproduce two-temperature behavior in the regime in which it should be expected. If this is correct, such a model calculation (if it can be modified to include thermal-diffusion effects) should be capable of yielding detailed agreement with experiment, even for disparate-mass gas mixtures.

The present two-temperature approach, on the other hand, is analogous to a hydrodynamic approximation to the full mixture Boltzmann equations, except that its region of validity extends into the two-temperature regime $\tilde{k} \sim 1$. Even if a complete kinetic model calculational procedure is perfected, the importance of the present approach would continue to be the fact that it highlights the physical processes characteristic of the two-temperature regime, that it makes clear the necessity for such a regime to exist (and gives the conditions for its existence), and that it provides a simple and accessible way of calculating two-temperature effects.

VII. CONCLUSIONS

The recent experiments of Letamendia et al.[12] confirm the assertion that there is a regime in which light scattering in disparate-mass gas mixtures must be understood in terms of two-temperature, near-normal behavior. This appears to be the first experimental confirmation of the importance of the epochal relaxation effects originally predicted by Grad,[5,29] in neutral disparate-mass gases.

The approximate predictions obtained here for xenon-helium spectra in the two-temperature regime show encouraging agreement with experiment. It is evident, however, that proper agreement can only be obtained with a more complete theoretical calculation. In particular, it seems likely that the inclusion of thermal-diffusion effects should greatly

FIG. 9. Spectral contributions $S_{ij}(k,\omega)$, for $x_1 = x_2 = 0.5$ and $\tilde{k} = 0.7$ (two-temperature predictions).

improve the quantitative agreement between two-temperature predictions and experiment.

ACKNOWLEDGMENTS

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[21]The results shown in Fig. 10 of Ref. 12 cannot refer to the experimental conditions $p_{Xe} = 5.97$ atm, $p_{He} = 3.78$ atm claimed on p. 1582 of that reference, since that physical situation is shown in Fig. 5(d) of Ref. 12 to behave quite differently.

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[27]To order δ, the diffusion velocity is a light-species velocity which is driven by a gradient in the light-species partial pressure (see Refs. 6—8).

[28]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), Fig. 6. Comparison with Ref. 12 suggests that the figure captions to Figs. 6(a) and 6(b) should be exchanged.

[29]See also J. M. Fitzpatrick and B. Shizgal, Phys. Fluids 22, 436 (1979).