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Variational determination of the coefficient of sound dispersion in binary gas mixtures

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The propagation of plane linear acoustic wave in a mixture of inert gases is considered by means of a variational formulation of the Boltzmann equations, through which the sound speed c is expressed with errors of order ϵ^2 in terms of trial functions determined with errors of order ϵ. This feature allows the exact determination of the coefficient of sound dispersion $d_2 \equiv [\mathrm{d}c/\mathrm{d}\omega^2]$ at zero frequency ($\omega = 0$), in terms of trial functions known from the Chapman–Enskog theory. Explicit results for d_2 are given for all combinations of noble gases from He to Xe, assumed to interact through the Lennard–Jones potential. Comparison with previous approximate descriptions and with experiments is made.

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1. Introduction

We consider the problem of determining the initial-frequency dependence of the speed $c(\omega)$ of propagation of a plane sound wave of infinitesimal amplitude and given frequency ω progressing in a uniform mixture of monoatomic gases. When the problem is treated by means of the Navier–Stokes equations for a pure gas or an inert mixture of gases, a known result is that $\mathrm{d}c/\mathrm{d}\omega = 0$ at $\omega = 0$, so that dispersion phenomena at small frequencies are at least quadratic in ω. Finding the coefficient of initial dispersion $d_2 \equiv [\mathrm{d}c/\mathrm{d}\omega^2]_{\omega=0}$ thus requires the use of a theory correct at least to second order in the frequency ω, a quality not enjoyed by the Navier–Stokes equations. One could, in principle, use higher-order hydrodynamic formalisms (Burnett, super-Burnett) to determine d_2; however, because of the complexity of the theory, a complete description of all relevant Burnett transport coefficients is only available for pure gases (Greenspan 1965). Alternatively, the acoustic problem can be studied directly at all frequencies for plane waves of infinitesimal amplitude by direct attack on the Boltzmann equations, linearized around Maxwellian velocity distribution functions with uniform velocity temperature and densities, corresponding to the conditions of the unperturbed gas.

For gas mixtures Foch, Uhlenbeck & Fuentes Losa (1972), following previous work for pure gases (Foch & Ford 1970), expanded the linearized Boltzmann equations for the acoustic problem in powers of the sound frequency, in an attempt to test the validity of the Boltzmann equations under conditions increasingly distant from equilibrium. For the particular case of molecules interacting with Maxwellian potentials, these authors obtained an explicit expression for the dispersion coefficient in terms of the standard first-order transport coefficients (viscosity, heat conductivity, etc.) for Maxwell molecules, as well as other parameters arising in the theory only to second order.

Although the Maxwellian interaction model does not provide a realistic description of transport in real gases, Foch & Ford (1970) have shown that the dispersion coefficient d_2 of a pure gas depends weakly on the potential of interatomic interaction provided that the exact value of the viscosity coefficient is used in place of its Maxwellian counterpart. A similar result may be expected for mixtures whose constituents have similar masses, but not for gas mixtures with widely different molecular weights, whose transport coefficients differ markedly from their Maxwellian counterparts (even when the parameters of intermolecular interaction are selected at every temperature such as to yield correct values for the viscosities of the two pure gases and for the mixture diffusion coefficient).

For the case of gas mixtures with disparate masses, a number of two-fluid theories have been developed and applied to the acoustic problem (Goldman 1968; Huck & Johnson 1980; Fernandez-Feria & Fernandez de la Mora 1986). Even though they are only valid to first order in the frequency, these treatments have been quite successful because, for disparate-mass mixtures, second-order self-collision (Burnett) effects become small compared with irreversibilities originating in the slow momentum and energy transfer in cross-collisions between the components. An interesting feature of two-fluid theories is that they have a frequency range of validity far broader than that of the standard Navier–Stokes equations, because they account properly for such slow relaxation phenomena. However, two-fluid theories do not improve over the Navier–Stokes level in situations where contributions of second order in the frequency arising from viscosity and heat conductivity become dominant; for instance in the case of high dilution where the mixture degenerates into a pure gas, or when the mass ratio is of order unity.

The simplest available theory able to provide an exact description of the initial dispersion in binary mixtures for all mass ratios and concentrations is the perturbed eigenvalue solution of the linearized Boltzmann equation used by Foch *et al.* (1972). Though simpler in higher order than the Chapman–Enskog approach, this method is still extremely complex algebraically, which so far has precluded its development for any model of intermolecular interaction with the exception of the Maxwellian potential. For the case of pure gases with the same interaction potential, Foch & Ford (1970) have carried the expansion to higher orders (up to order 16 in Foch & Fuentes Losa 1972). By appropriate manipulations and exploiting the symmetry of the linearized Boltzmann operator, order by order, Foch & Fuentes-Losa (1972) have succeeded in expressing the sound-speed eigenvalue of order $2n$ in terms of the velocity distribution function eigenfunctions of order n. However, this efficient scheme has not been extended to the case of binary gas mixtures.

In what follows, the eigenvalue problem is reconsidered using a well-known variational formulation, through which eigenvalues correct to second order in a small parameter ϵ are written in terms of trial functions correct only to order ϵ. Variational methods have been widely used in kinetic problems: a very general variational principle applicable to the steady linearized Boltzmann equation in a pure gas can be found in previous works by Cercignani & Pagani (1966) and Cercignani (1969). Bernstein (1969) mentions several other gaskinetic applications of variational principles in his own variational calculation of transport coefficients in a binary gas mixture. In this paper, the results of Foch & Ford (1970) are condensed into a single expression valid to any order in the frequency expansion, and are automatically extended to the case of mixtures with an arbitrary number of components. As an example, the known first-order solutions of the Chapman–Enskog theory are used to obtain an exact explicit expression for the dispersion coefficient in binary mixtures of gases interacting with arbitrary intermolecular potentials. This result provides an exact standard against which higher-order hydrodynamic theories for gas mixtures can be tested.

2. Variational principle for the acoustic linearized Boltzmann problem

Consider a perturbation of very small amplitude propagating in a binary gas mixture originally at rest with equilibrium temperature T_0 and component densities ρ_{10} and ρ_{20} for species 1 and 2 respectively ($\rho_0 = \rho_{10}+\rho_{20}$). The velocity distribution function f_i for component i can be written conveniently in the form:

$$f_i = f_{0i}(1+\phi_i), \tag{1}$$

where $f_{0i} = n_{0i}(m_i/2\pi kT_0)^{\frac{3}{2}} \exp(-m_i u_i^2/2kT_0)$ is the space- and time-independent equilibrium distribution function associated with the system at rest. Let the perturbation be a plane acoustic wave propagating in the positive x-direction with a given frequency ω, so that $\phi_i \propto \exp[\mathrm{i}\omega(t-x/c)]$ and therefore, $\boldsymbol{U}, \rho_i - \rho_{i0}, T-T_0 \propto \exp[\mathrm{i}\omega(t-x/c)]$, where the ρ_i are the instantaneous values of the species densities and $\boldsymbol{U}$ and T are the mixture mean velocity and temperature. The sound speed c of the mixture can be written in terms of the non-dimensional variable γ:

$$c = \left(\frac{\gamma\, kT_0}{m_0}\right)^{\frac{1}{2}}, \tag{2}$$

which takes the value $\gamma = \gamma_0 = \frac{5}{3}$ at equilibrium, wile $m_0 = (m_1\, n_{01} + m_2\, n_{02})/n_0$, with $n_0 = n_{01}+n_{02}$. The dimensionless molecular velocity is introduced as

$$\xi_i = \frac{\boldsymbol{u}_i}{(2kT_0/m_0)^{\frac{1}{2}}}.$$

The Boltzmann equation can now be linearized to obtain

$$\mathrm{i}\omega\left(1-\frac{u_{ix}}{c}\right) f_{0i}\,\phi_i = K_{ii}\,\phi_i + K_{ij}\,(\phi_i+\phi_j), \quad i = 1, 2, \tag{3}$$

where K_{ii} and K_{ij} are related to the linearized collision integrals I_{ij} defined in Chapman & Cowling (1970) and provide a most convenient notation for the mixture problem (Bernstein 1969):

$$K_{ii}\,\phi_i = -n_i{}^2 I_i(\phi_i), \tag{4}$$

$$K_{ij}(\phi_i+\phi_j) = -n_i n_j I_{ij}(\phi_i+\phi_j), \tag{5}$$

valid for mixtures with an arbitrary number of components.

Equation (3) can be rewritten in the more compact vector form

$$\mathrm{i}\omega(\mathbf{L}-\zeta\mathbf{M})\,\boldsymbol{\Phi} = \mathbf{K}\boldsymbol{\Phi}, \tag{6}$$

where $\boldsymbol{\Phi}$ is the vector $(\phi_1, \phi_2)^{\mathrm{T}}$, the eigenvalue ζ is defined as $(2/\gamma)^{\frac{1}{2}}$, while for binary mixtures the matrices $\mathbf{K}$, $\mathbf{L}$ and $\mathbf{M}$ are, respectively,

$$\begin{pmatrix} K_{11}+K_{12} & K_{12} \\ K_{21} & K_{22}+K_{21} \end{pmatrix}, \quad \begin{pmatrix} f_{01} & 0 \\ 0 & f_{02} \end{pmatrix}, \quad \begin{pmatrix} M_1^{-\frac{1}{2}}\xi_1 f_{01} & 0 \\ 0 & M_2^{-\frac{1}{2}}\xi_2 f_{02} \end{pmatrix},$$

where $M_i = m_i/m_0$. By introducing the inner product

$$\{\boldsymbol{\Phi}, \boldsymbol{\Psi}\} \equiv [\phi_1, \Psi_1]_1 + [\Phi_2, \Psi_2]_2 \tag{7}$$

where $[\phi_i, \psi_i]_i = \int \phi_i \psi_i \, d^3u_i$, the problem can be reformulated variationally. A functional whose Euler–Lagrange equation is (6) can be obtained by taking a variation $\delta\boldsymbol{\Phi}$ around the solution $\boldsymbol{\Phi}$ and forming the inner product with (6):

$$\mathrm{i}\omega\{\delta\boldsymbol{\Phi}, (\mathbf{L}-\zeta\mathbf{M})\boldsymbol{\Phi}\} = \{\delta\boldsymbol{\Phi}, \mathbf{K}\boldsymbol{\Phi}\}. \tag{8}$$

Now $\mathbf{K}$ is a symmetric operator (Chapman & Cowling 1970), so that

$$\delta\{\boldsymbol{\Phi}, \mathbf{K}\boldsymbol{\Phi}\} = \{\delta\boldsymbol{\Phi}, \mathbf{K}\boldsymbol{\Phi}\} + \{\boldsymbol{\Phi}, \mathbf{K}\,\delta\boldsymbol{\Phi}\} = 2\{\delta\boldsymbol{\Phi}, \mathbf{K}\boldsymbol{\Phi}\},$$

and writing a similar equation for $(\mathbf{L}-\zeta\mathbf{M})$, since this is a symmetric matrix of given functions which commutes with δ, (8) can be expressed as

$$\delta\{\boldsymbol{\Phi}, \mathrm{i}\omega(\mathbf{L}-\zeta\mathbf{M})\boldsymbol{\Phi} - \mathbf{K}\boldsymbol{\Phi}\} = 0, \tag{9}$$

so that the functional $\Lambda(\boldsymbol{\Phi}) = \{\boldsymbol{\Phi}, \mathrm{i}\omega(\mathbf{L}-\zeta\mathbf{M})\boldsymbol{\Phi} - \mathbf{K}\boldsymbol{\Phi}\}$ is stationary when $\boldsymbol{\Phi}$ is a solution to (6). Furthermore its value there is $\Lambda^* = 0$, since in that case it follows from (6) that $\mathrm{i}\omega(\mathbf{L}-\zeta\mathbf{M})\boldsymbol{\Phi} - \mathbf{K}\boldsymbol{\Phi} = 0$. The equality $\Lambda^* = 0$ can be expressed as

$$\mathrm{i}\omega(\{\boldsymbol{\Phi}, \mathbf{L}\boldsymbol{\Phi}\} - \zeta\{\boldsymbol{\Phi}, \mathbf{M}\boldsymbol{\Phi}\}) = \{\boldsymbol{\Phi}, \mathbf{K}\boldsymbol{\Phi}\}, \tag{10}$$

from which ζ can be written in terms of the eigenfunction $\boldsymbol{\Phi}$ as

$$\zeta = \frac{\{\boldsymbol{\Phi}, \mathbf{L}\boldsymbol{\Phi}\} - \{\boldsymbol{\Phi}, \mathbf{K}\boldsymbol{\Phi}\}/\mathrm{i}\omega}{\{\boldsymbol{\Phi}, \mathbf{M}\boldsymbol{\Phi}\}}. \tag{11}$$

The functional above is also stationary when $\boldsymbol{\Phi}$ satisfies (6), a feature that may be exploited for the optimal determination of ζ. Assume that an approximation $\boldsymbol{\Phi}^* = \boldsymbol{\Phi} - \boldsymbol{\Phi}'$ is inserted into (11) instead of the exact solution $\boldsymbol{\Phi}$, where $\boldsymbol{\Phi}'$ is small. Let ζ^* be the resulting value obtained for ζ and define $\zeta' = \zeta - \zeta^*$. If (5), which is satisfied by the exact solution $\boldsymbol{\Phi}$, is combined with the quotient formula particularized for $\boldsymbol{\Phi}$ and $\boldsymbol{\Phi}^*$, it is found that ζ' is quadratic in $\boldsymbol{\Phi}'$:

$$\zeta' = \frac{-\{\boldsymbol{\Phi}', \mathbf{L}\boldsymbol{\Phi}'\} + \zeta\{\boldsymbol{\Phi}', \mathbf{M}\boldsymbol{\Phi}'\} + \{\boldsymbol{\Phi}', \mathbf{K}\boldsymbol{\Phi}'\}/\mathrm{i}\omega}{\{\boldsymbol{\Phi}, \mathbf{M}\boldsymbol{\Phi}\}}. \tag{12}$$

Consequently, if the trial function is in error with respect to $\boldsymbol{\Phi}$ by a term of the order of the nth power of the frequency, the numerator would be dominated by the collision term $O(\omega^{2n-1})$ with respect to the denominator. Therefore (12) provides an approximation for the eigenvalue which is of higher order ($\zeta' = O(\omega^{2n-1})$) than the trial function $\boldsymbol{\Phi}^*$ (except in the case $n = 1$). This is a general property of perturbation theory for symmetric operators (Bethe & Salpeter 1957).

3. Navier–Stokes trial function applied to the determination of the dispersion coefficient

From the previous section it follows that it is possible to exploit the symmetry properties of the operator $\mathbf{K}$ in order to express the sound speed of the mixture with errors of order ϵ^2 in terms of a trial distribution function known with errors of order ϵ. Such approximate functions may be generated in a variety of ways. Foch & Ford

(1970) used velocity polynomials to create a family of trial functions in the pure-gas case. Alternatively, the approximate solutions found by Sirovich & Thurber (1965) based on model equations could in principle be extended to binary mixtures and used as trial functions to improve their results over the whole frequency range. However, by their *ad hoc* approximate nature, these and other similar methods do not yield exact results. For that reason we will base our computation of the speed of sound on the results from the Chapman–Enskog theory, which are exact asymptotically in the low-frequency range, with errors $O(\omega^2)$.

In the Chapman–Enskog theory an expression is derived in terms of a small parameter, the Knudsen number Kn, proportional to the frequency ω in the acoustic problem and characterizing the degree of non-equilibrium. For a pure gas there is only one collision frequency, and $Kn = \mu\omega/p$, where μ and p are the viscosity coefficient and pressure of the mixture. For gas mixtures, other relaxation scales appear, depending on the molecular masses and molar concentrations, leading to a variety of Knudsen numbers. Typically, the region of validity of the Chapman–Enskog theory for mixtures is such that the product of ω and the longest microscopic relaxation scale should be small compared with one. The subject has been discussed at great length in the literature on two-fluid theories, particularly by Fernandez de la Mora & Fernandez-Feria (1987) and by Fernandez-Feria & Fernandez de la Mora (1986) for the acoustic problem. In what follows the term 'small frequency' will be used loosely to refer to frequencies small compared with the smallest microscopic relaxation time; that is, to frequencies contained within the region of convergence of the Chapman–Enskog theory.

If $\boldsymbol{\Phi}^*$ is obtained from the Euler-level solution, where $\boldsymbol{\Phi}' = O(\omega)$, the variational formulation yields $\gamma = \frac{5}{3}$, adding nothing to the hydrodynamical analysis. But choosing $\boldsymbol{\Phi}^*$ at the Navier–Stokes level, where $\boldsymbol{\Phi}' = O(\omega^2)$, the resulting error in ζ is of order ω^3, from which an asymptotically exact value for the dispersion coefficient will be derived. Let us write $\boldsymbol{\Phi}^* = \boldsymbol{\Phi}_0 + \boldsymbol{\Phi}_1$, where $\boldsymbol{\Phi}_0$ is an exact solution of the homogeneous problem $\mathbf{K}\boldsymbol{\Phi} = 0$ which contains all the hydrodynamic information, that is

$$\Sigma_i\, m_i[f_{0i}(1+\phi_{0i}),\, \psi_i]_i = Y, \tag{13}$$

where $\psi_i = (\delta_{i1}, \delta_{i2}, \boldsymbol{u}_i, \frac{1}{2}(\boldsymbol{u}_i - \boldsymbol{U})^2)$ and $Y = (\rho_1, \rho_2, \rho \boldsymbol{U}, \frac{3}{2}nkT)$. In this notation, δ_{ij} takes the value 1 for $i = j$ and 0 otherwise. $\boldsymbol{\Phi}_0$ corresponds to the zeroth-order solution in the Chapman–Enskog method while $\boldsymbol{\Phi}_1$ is determined from $\boldsymbol{\Phi}_0$ by solving

$$\mathrm{i}\omega(\mathbf{L} - \zeta\mathbf{M})^{(0)}\, \boldsymbol{\Phi}_0 = \mathbf{K}\boldsymbol{\Phi}_1, \tag{14}$$

where $\mathrm{i}\omega(\mathbf{L} - \zeta\mathbf{M})^{(0)}$ is the corresponding zeroth-order term in the subdivision of the $\mathrm{i}\omega(\mathbf{L} - \zeta\mathbf{M})$ operator (Chapman & Cowling 1970) chosen so as to ensure solvability of the non-homogeneous problem (14). Uniqueness in the solution results from the orthogonality condition

$$\Sigma\, m_i[f_{0i}\,\phi_{1i},\, \psi_i]_i = 0, \tag{15}$$

which is equivalent to the statement in (13). Now the solvability condition for the next order can be shown to be equivalent to the fulfilment of the hydrodynamic Navier-Stokes equations, that is

$$\mathrm{i}\omega\{\boldsymbol{\Psi}, (\mathbf{L} - \zeta_{\mathrm{N}}\mathbf{M})\,\boldsymbol{\Phi}^*\} = 0, \tag{16}$$

where ζ_{N} is the Navier-Stokes solution for ζ and $\boldsymbol{\Psi}$ is the vector $(\psi_1, \psi_2)^{\mathrm{T}}$. If $\boldsymbol{\Phi}^*$ is inserted in the quotient (11) to obtain the refined value ζ^* and (15) and (16) are taken into account, the following results:

$$\zeta^* - \zeta_N = \frac{\{\boldsymbol{\Phi}_1, (\mathbf{L} - \zeta_N \mathbf{M})\boldsymbol{\Phi}_1)\}}{\{\boldsymbol{\Phi}^*, \mathbf{M}\boldsymbol{\Phi}^*\}}. \tag{17}$$

Introducing the following dimensionless variables

$$\eta_i = \frac{n_i - n_{0i}}{n_{0i}}, \quad \theta_i = \frac{T - T_0}{T_0},$$

$$x_i = \frac{n_{0i}}{n_0}, \quad \epsilon_i = \frac{\rho_{0i}}{\rho_0} = M_i x_i, \quad X = \frac{U}{c},$$

ϕ_{0i} and ϕ_{1i} can be expressed as

$$\phi_{0i} = (\xi_i^2 - \tfrac{3}{2})\,\theta + \left(\frac{2}{\gamma M_i}\right)^{\frac{1}{2}} \xi_{ix} X + \eta_i \tag{18}$$

and

$$\phi_{1i} = \phi_{Ai} + \phi_{Bi} + \phi_{Di}, \tag{19}$$

with

$$\phi_{Ai} = -\boldsymbol{A}_i \cdot \boldsymbol{\nabla} \ln T, \tag{20}$$

$$\phi_{Bi} = -2\boldsymbol{B}_i \boldsymbol{:} \boldsymbol{\nabla} U, \tag{21}$$

$$\phi_{Di} = -\boldsymbol{D}_i \cdot \boldsymbol{d}_{12}, \tag{22}$$

where $\boldsymbol{\nabla} \ln T = -\mathrm{i}\omega\theta\ \boldsymbol{e}_x/c$, $\boldsymbol{\nabla} U = -\mathrm{i}\omega U\ \boldsymbol{e}_x \boldsymbol{e}_x/c$, $\boldsymbol{d}_{12} = -\mathrm{i}\omega d'\ \boldsymbol{e}_x/c$, $d' = x_1(\eta_1 + \theta) - \epsilon_i(x_1\ \eta_1 + x_2\ \eta_2 + \theta)$ and $\boldsymbol{A}_i$, $\boldsymbol{B}_i$ and $\boldsymbol{D}_i$ can be expanded in Sonine polynomials (Chapman & Cowling 1970) as follows:

$$\boldsymbol{A}_i = \sum_p (a_p + k_T d_p)\, \boldsymbol{a}_i^{(p)},$$

$$\boldsymbol{B}_i = \textstyle\sum_p' b_p \boldsymbol{b}_i^{(p)},$$

$$\boldsymbol{D}_i = \textstyle\sum_p' d_p \boldsymbol{a}_i^{(p)}.$$

In the above formulae, sums over p extend from $-\infty$ to ∞, while the prime indicates that the $p = 0$ term is not included. k_T is the thermal diffusion ratio. Formulae giving $\boldsymbol{a}_i^{(p)} \propto \boldsymbol{\xi}$, $\boldsymbol{d}_i^{(p)} \propto \boldsymbol{\xi}$ and $\boldsymbol{b}_i^{(p)} \propto \boldsymbol{\xi}^0\boldsymbol{\xi}$ (where $\boldsymbol{\xi}^0\boldsymbol{\xi} = \boldsymbol{\xi}\boldsymbol{\xi} - \tfrac{1}{3}\xi^2 \boldsymbol{I}$) as a function of Sonine polynomials can be found in Chapman & Cowling (1970), who also include the system of equations leading to the determination of the coefficients a_p, b_p and d_p.

Returning to (17), the Navier-Stokes eigenvalue ζ_N involved in it can be readily obtained to any order from the acoustic hydrodynamic Navier-Stokes equations. In the formulation leading to (17), ζ_N is specified to at least second order. A complete specification of ζ_N is not usually found in the literature (see Foch *et al.* 1972) because it is not correct beyond first order. The so-called Kohler formula is obtained by truncating at first order in the frequency: $\zeta_N^{(1)} = (2/\gamma_N^{(1)})^{\frac{1}{2}}$, where

$$\frac{\gamma_N^{(1)}}{\gamma_0} = 1 + \tfrac{4}{25}\Lambda \mathrm{i} + \tfrac{4}{5}\Omega \mathrm{i} + (x_1 - \epsilon_1 + \tfrac{2}{5}k_T)^2 \Delta \mathrm{i} \tag{23}$$

and $\Lambda = \lambda\omega m_0/n_0 k^2 T_0$, $\Delta = \omega D m_0/k T_0 x_1 x_2$, $\Omega = \mu\omega/n_0 k T_0$, where λ, μ and D are respectively the mixture coefficients of heat conduction, viscosity and diffusion. In the initial dispersion problem, Λ, Δ and Ω are small parameters characterizing the degree of non-equilibrium. Ω is the Knudsen number of the mixture while $\Lambda = 2\Omega/$

$5Pr$, where $Pr = \mu c_p/\lambda$ is the Prandtl number, of order unity. Δ is related to the interspecies non-equilibrium and can be written as $\Delta = \Omega/Sc x_1 x_2$, where $Sc = \mu/\rho_0 D$ is the Schmidt number, also of order unity. All three parameters are therefore of the same order, except in the limit where one of the species is very dilute in molar fraction; this limit is relevant in the case of disparate-mass mixtures, where even a small volume fraction of the heavy component may lead to a mass fraction of order unity.

According to the arguments in (12), ζ^* is correct to second order, that is, $\zeta^* - \zeta = O(\omega^3)$. Therefore if $O(\omega^3)$-terms are neglected in (17), a simplified version is obtained:

$$\frac{\zeta^* - \zeta_N}{\zeta_0} = \frac{\{\Phi_1^0, (\mathbf{L} - \zeta_0 \mathbf{M})\Phi_1^0\}}{\zeta_0\{\Phi_0^0, \mathbf{M}\Phi_0^0\}}, \tag{24}$$

where $\zeta_0 = (\frac{6}{5})^{\frac{1}{2}}$, while Φ_0^0 and Φ_1^0 are given by (18) and (20)–(22) when the temperature, velocity and density ratios are taken at the zeroth (Euler) level:

$$\eta_1 = \eta_2 = X, \quad \theta = \tfrac{2}{3}X, \quad d' = \tfrac{5}{3}X(x_1 - \epsilon_1). \tag{25}$$

Using (25), the denominator of (24) is shown to be

$$2(x_1\epsilon_1 + x_2\epsilon_2 + \theta)X = \tfrac{10}{3}X^2,$$

and making use of the vector equalities in Appendix A one gets

$$\begin{aligned}\frac{\zeta^* - \zeta_N}{\zeta_0} = \{&\tfrac{1}{3}\beta_1 \nabla \ln T \cdot \nabla \ln T + \tfrac{4}{5}\beta_2 \nabla^0 \boldsymbol{U} : \nabla^0 \boldsymbol{U} + \tfrac{1}{3}\beta_3 \boldsymbol{d}_{12} \cdot \boldsymbol{d}_{12} + \tfrac{2}{3}\beta_4 \boldsymbol{d}_{12} \cdot \nabla \ln T \\ &- \tfrac{4}{5}\zeta_0(\beta_5 \nabla^0 \boldsymbol{U} : \boldsymbol{e}_x \nabla \ln T + \beta_6 \nabla^0 \boldsymbol{U} : \boldsymbol{e}_x \boldsymbol{d}_{12})\}/\tfrac{10}{3}X^2.\end{aligned} \tag{26}$$

The β-coefficients involved here are defined as follows:

$$\beta_1 = \Sigma \frac{[\boldsymbol{A}_i \cdot, \boldsymbol{A}_i f_{0i}]_i}{n}, \quad \beta_2 = \Sigma \frac{[\boldsymbol{B}_i :, \boldsymbol{B}_i f_{0i}]_i}{n}, \quad \beta_3 = \Sigma \frac{[\boldsymbol{D}_i \cdot, \boldsymbol{D}_i f_{0i}]_i}{n}, \quad \beta_4 = \Sigma \frac{[\boldsymbol{A}_i \cdot, \boldsymbol{D}_i f_{0i}]_i}{n},$$

$$\beta_5 = \Sigma \frac{[\boldsymbol{A}_i \boldsymbol{u}_i :, \boldsymbol{B}_i f_{0i}]_i}{na}, \quad \beta_6 = \Sigma \frac{[\boldsymbol{D}_i \boldsymbol{u}_i :, \boldsymbol{B}_i f_{0i}]_i}{na},$$

where $a = (2kT/m_0)^{\frac{1}{2}}$. They play a similar role to the transport coefficients arising in a Burnett theory, although the unavailability of Burnett results for the dispersion of sound in mixtures has prevented any direct identification. However, the fact that the Burnett dispersion must be identical with our results and depend on a combination of first- and second-order (Burnett) transport coefficients, permits an identification from which at least one of these second-order coefficients could be derived just from the first-order Chapman–Enskog eigenfunctions. Dimensionless transport groups (expressed in terms of the Sonine coefficients a_p, b_p and d_p in Appendix B) can be introduced:

$$\beta_1^* = \frac{\beta_1(nk/\lambda)^2 kT}{m_0}, \quad \beta_2^* = \beta_2\left(\frac{nkT}{\mu}\right)^2, \quad \beta_3^* = \frac{\beta_3(x_1 x_2/D)^2 kT}{m_0},$$

$$\beta_4^* = \frac{\beta_4(nkx_1x_2/D\lambda)kT}{m_0}, \quad \beta_5^* = \beta_5 \frac{n^2k^2T}{\mu\lambda}\left(\frac{kT}{m_0}\right)^{\frac{1}{2}}, \quad \beta_6^* = \beta_6 \frac{nkx_1x_2}{D\lambda}\left(\frac{kT}{m_0}\right)^{\frac{1}{2}},$$

whereupon (26) becomes

$$\frac{\zeta^* - \zeta_{\mathrm{N}}}{\zeta_0} = -\tfrac{2}{75}\beta_1^* \Lambda^2 - \tfrac{4}{25}\beta_2^* \Omega^2 - \tfrac{1}{6}\beta_3^* \Delta^2(x_1-\epsilon_1)^2 - \tfrac{2}{15}\beta_4^* \Lambda\Delta(x_1-\epsilon_1) + \left(\frac{8\sqrt{2}}{125}\right)\beta_5^* \Lambda\Omega + \left(\frac{16\sqrt{2}}{27}\right)\beta_6^* \Delta\Omega. \tag{27}$$

This result can be compared with the one obtained from the two-fluid theory given by Fernandez-Feria & Fernandez de la Mora (1986):

$$\frac{\zeta_{2\mathrm{F}} - \zeta_{\mathrm{N}}}{\zeta_0} = \tfrac{1}{2}\epsilon_1\epsilon_2(\epsilon_1 - x_1 - \tfrac{2}{5}k_T)^2 \Delta^2 + \tfrac{2}{5}(\epsilon_1 - x_1 - \tfrac{2}{5}k_T)\,[(\Omega_{22} - \Omega\epsilon_2) - (\Omega_{11} - \Omega\epsilon_1)]\,\Delta, \tag{28}$$

where $\Omega_{ij} = \omega\ \mu_{ij}/n_0 kT_0$ and the μ_{ij} are two-fluid partial viscosities. In the limit where the two-fluid theory gives a good approximation ($\zeta_{2F} \approx \zeta^*$), that is, for $M = m_1/m_2 \ll 1$ and $\epsilon_2 < 1$ (and therefore $x_2 \sim M \ll 1$), (27) becomes

$$\frac{\zeta^* - \zeta_{\mathrm{N}}}{\zeta_0} \approx -\tfrac{1}{6}\beta_3^* \Delta^2(x_1-\epsilon_1)^2,$$

while (28) is

$$\frac{\zeta_{2\mathrm{F}} - \zeta_{\mathrm{N}}}{\zeta_0} \approx -\tfrac{1}{2}\epsilon_1\epsilon_2(\epsilon_1 - x_1 - \tfrac{2}{5}k_T)^2 \Delta^2. \tag{29}$$

if the sum giving β_3^* in terms of the d_p coefficients is truncated at the first term and the definition of d_0 in Chapman & Cowling (1970) is used, the following approximation for β_3^* results:

$$\beta_3^* \approx 3\,\epsilon_1\epsilon_2, \tag{30}$$

which, taking into account the smallness of k_T, shows the identity between both limits, confirming the accuracy of first-order two-fluid theories for disparate-mass mixtures where the heavy component is not dominant with respect to density.

In order to compare our results with experiments, the sound-speed ratio is expanded in terms of the dimensionless quantity f/p, where f is the frequency in MHz and p the mixture pressure in atm:

$$\frac{\zeta}{\zeta_0} = \frac{c_0}{c} = 1 - \mathrm{i}d_1\frac{f}{p} - d_2\left(\frac{f}{p}\right)^2 + \dots. \tag{31}$$

The Navier–Stokes solution ζ_{N} is correct to first order and therefore predicts the absorption coefficient d_1, while the variationally improved ratio ζ^* is exact to second order and yields the dispersion coefficient d_2. In the case of a pure gas, similar calculations can be performed leading to the following results:

$$\frac{\zeta}{\zeta_0} = 1 - \frac{\left(2+\dfrac{1}{Pr}\right)\mathrm{i}\Omega}{5} - \left\{\tfrac{6}{25} + \frac{\tfrac{1}{5} - \tfrac{3}{50}\alpha}{Pr^2} + \tfrac{2}{5}\beta + \frac{(\tfrac{2}{5} - \tfrac{8}{25}\delta)}{Pr}\right\}\Omega^2 + \dots. \tag{32}$$

Pr is again the Prandtl number, while α, β and δ are defined in Appendix C and take the value 1 when only the first Sonine polynomial is kept, or in the case where the interaction potential is Maxwellian. In this case $Pr = \tfrac{2}{3}$ and (32) becomes

$$\frac{\zeta}{\zeta_0} = 1 - 0.7\,\mathrm{i}\Omega - 1.075\,\Omega^2 + \dots, \tag{33}$$

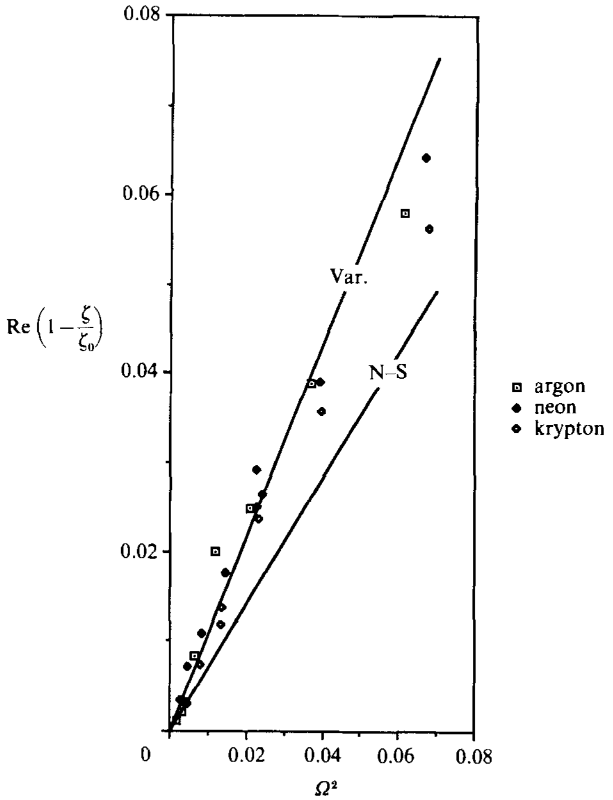

FIGURE 1. Sound-speed velocity ($\mathrm{Re}\,(1-\zeta/\zeta_0)$) in pure noble gases as a function of the square of the frequency Ω^2. Symbols are from experiments of Greenspan (1956).

in agreement with the results given in the literature from second-order hydrodynamic formulations such as Burnett equations (Greenspan 1965). Foch & Ford (1970) gave an approximate result for ζ/ζ_0 using their perturbed eigenvalue solution:

$$\frac{\zeta}{\zeta_0} = 1-0.7\,\mathrm{i}\Omega - 1.075\left(1+\frac{19\Delta_1}{215}\right)\Omega^2+\ldots, \tag{34}$$

where Δ_1 depends on the interaction potential and on the temperature, taking maximum values below unity (therefore, the influence of the interaction potential in the dispersion coefficient is slight, at most around 5%) and becoming zero in the case of Maxwell molecules, in agreement with (33). Therefore, given the considerable scatter observed in the experimental data available for the pure-gas sound speed (Greenspan 1956; replotted in Foch & Ford 1970), going to higher order in Sonine polynomials in (32) would be futile as far as comparison between theory and available pure-gas experiments is concerned. Figure 1 represents experimental measurements of $\mathrm{Re}(1-\zeta/\zeta_0)$ as a function of Ω^2 for Ne, Ar and Kr (Greenspan 1956), as well as the lowest order in Sonine polynomials, Navier–Stokes and variational expressions ($0.705\Omega^2$ and $1.075\Omega^2$ respectively). The limitation of the theory to small frequencies ($\Omega \ll 1$) is patent from the figure.

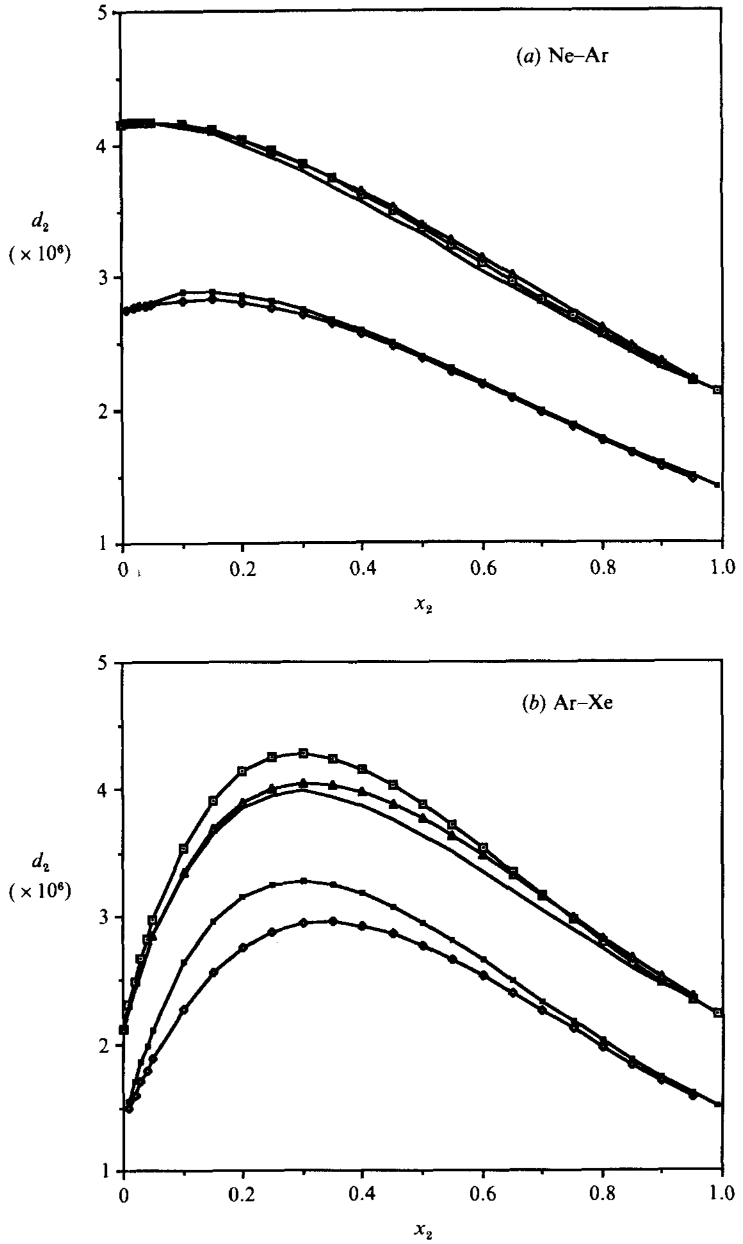

FIGURE 2(*a*, *b*). For caption see p. 216.

4. Discussion of results and comparison with experiments

The dispersion coefficient d_2, as defined in (31) is evaluated using (27). The Sonine expansions giving the new transport coefficients β_i are truncated, keeping only terms with $p = 1$, 0, 1. The interaction model adopted is the (12–6) Lennard-Jones potential with the ϵ/k and σ parameters given by Hogervorst (1971), who found excellent agreement between calculated and experimental values of the viscosity and thermal conductivity of noble-gas mixtures. The dispersion coefficient d_2 is plotted in

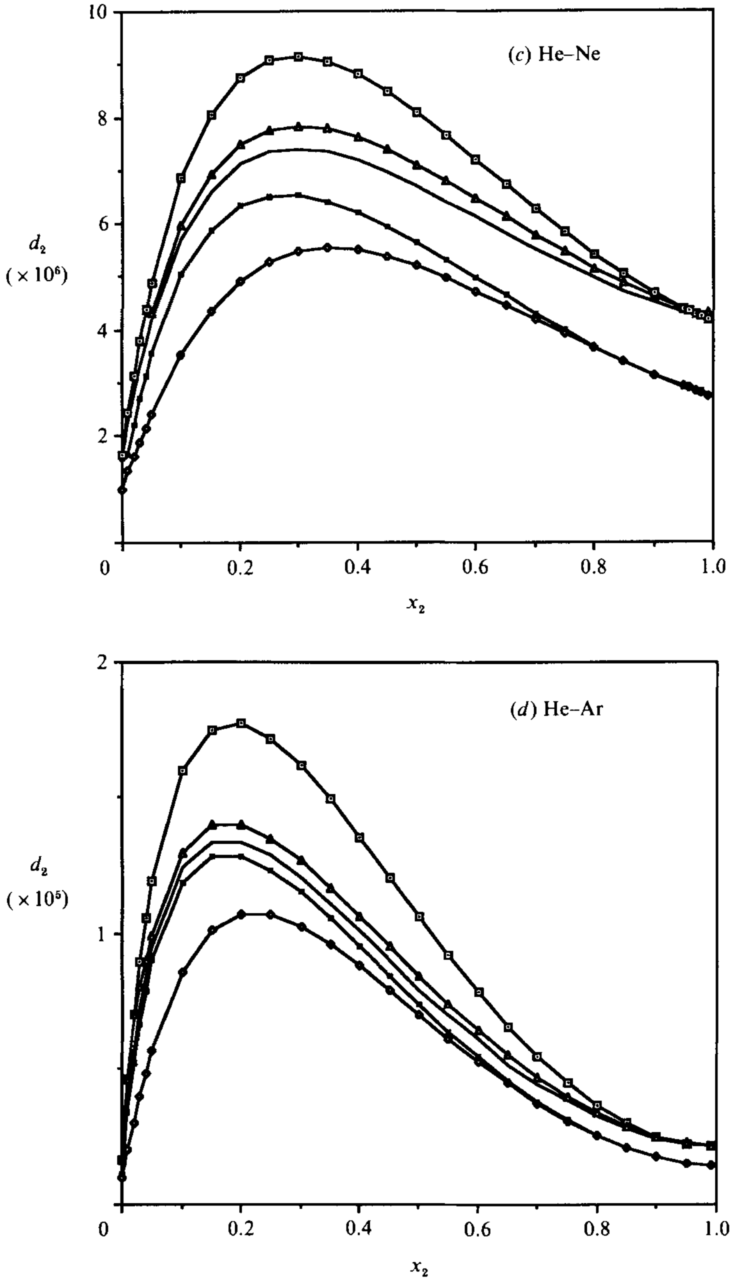

FIGURE 2(*c*, *d*). For caption see p. 216.

the whole range of concentrations for some of the binary combinations of He, Ne, Ar, Kr and Xe in figure 2 which is presented in ascending order of mass ratios m_2/m_1. The convention is used of denoting the heavier component in the mixture by the subscript 2, so that $M = m_1/m_2 < 1$. The coefficient of dispersion is computed at $T_0 = 299$ K. The results from different theories are compared with those from the variational method. The Navier-Stokes equations, though only valid to first order, yield a formula for the dispersion coefficient d_{NS} which is not correct (35% below the

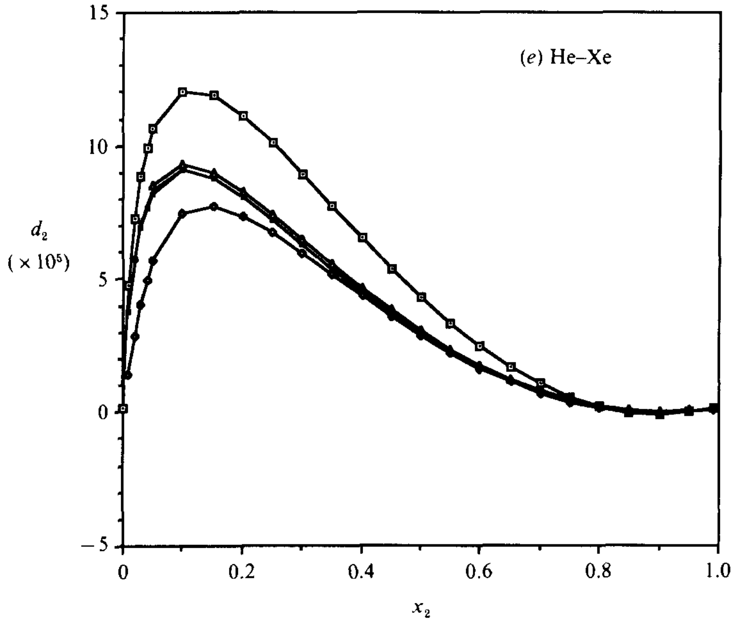

FIGURE 2. Initial dispersion d_2 (as defined in (31), in $(\mathrm{atm/MHz})^2$) for binary mixtures of noble gases, ordered in ascending mass ratio, as a function of the molar fraction x_2 of the heavier component: ——△——, d_F (direct solution of the Boltzmann equation (from the expression given by Foch *et al.* 1972), with first-order coefficients evaluated using the Lennard–Jones potential); ----□----, d_M (same as d_F with Maxwell mixture first-order coefficients (Fuentes Losa 1972); ——, d_2 (variational formula); ----■----, d_{2F} (two-fluid solution (Fernandez-Feria & Fernandez de la Mora 1986)); ——◇——, d_{NS} (Kohler formula). (*a*) Ne–Ar; (*b*) Ar–Xe; (*c*) He–Ne; (*d*) He–Ar; (*e*) He–Xe.

actual value for pure gases); however it is plotted here for reference. The results obtained by Foch *et al.* (1972) (d_F) using a direct solution of the linearized Boltzmann equation are also included. The set of explicit formulae giving d_2 in terms of the mixture transport coefficients in this reference has been used. Rather than using the Maxwell model to calculate the first-order coefficients λ, μ, k_T and D, those have been computed using the $(12-6)$ Lennard–Jones model (complete experimental data for all the spectrum of concentrations in noble-gas mixtures are not available). The remaining (second-order) coefficients are evaluated, as in Foch *et al.* (1972) under the assumption of the mixture being Maxwellian. The fact that the agreement between d_F and the variational results d_2 (where first- and second-order transport coefficients are evaluated using the Lennard–Jones potential) is quite good indicates that dispersion is mainly dependent on first-order transport coefficients and their combinations. However, results are very sensitive to the interaction model used for the evaluation of the first-order coefficients, as can be observed by noting the large difference between d_2 and d_M where d_M is the dispersion coefficient give in Fuentes Losa (1972), obtained using the direct solution of the linearized Boltzmann equation with all the transport coefficients evaluated with the Maxwell mixture assumption. Also included in the figure is the coefficient of dispersion d_{2F} from the first-order two-fluid theory (Fernandez-Feria & Fernandez de la Mora 1986) whose expression was compared with the variational one in the previous section.

Some observations follow from the figure. The magnitude of d_2 increases with the mass-disparity m_2/m_1. This shows the prevalent role of cross-collisions as a source of irreversibility causing dispersion; the energy transfer process is increasingly slow with m_2/m_1. As shown by Fernandez de la Mora & Puri (1986) the ratio between the pure-gas dispersion, caused by viscous and thermal dissipation – self-collisions – and the maximum dispersion for the mixture is of the order of M^2 and therefore becomes very small for disparate-mass gas mixtures. Besides, given that the mixture behaviour becomes dominated by self-collisions as soon as the heavy–light density ratio ρ_2/ρ_1 is of order unity, that is for $x_2 \sim M$, the pronounced change in d_2 from pure-gas to mixture dispersion takes place in a thin boundary layer of thickness M close to the pure-light-gas limit. However, as ϵ_2 increases, the pure-gas behaviour gradually sets in again and the value of d_2 slowly decays to the pure-heavy-gas limit. Therefore the curves representing d_2 as a function of x_2 are increasingly non-symmetric as m_2/m_1 grows. This skewness would disappear if the data were plotted as a function of ϵ_2 instead.

The two-fluid method is accurate for large mass disparity where the momentum and energy transfer become the main source of irreversibility. However, for mixtures with similar molecular weights, or, in general, close to the pure-gas limit, the method reduces to the Navier–Stokes solution and the two curves d_{NS} and $d_{2\mathrm{F}}$ come together.

Some of the most precise data on dispersion at low frequency are reported in Fuentes Losa (1972), where the value of the sound speed is measured for He–Xe and He–Ar mixtures at different dimensionless frequencies Ω and molar concentrations x_2. As discussed by Fernandez de la Mora & Puri (1986), the accuracy of the measured sound speed is excellent but the determination of the concentration x_2 is somewhat less precise. Therefore, the frequency is made dimensionless here through the same concentration-independent factor used by Fernandez de la Mora & Puri (1986) and Fernandez-Feria & Fernandez de la Mora (1987), leading to the group

$$s = \frac{\omega D m_2}{kT} = \frac{\Omega}{Sc(x_1 m_1/m_2 + x_2)}, \tag{35}$$

while a good approximation for x_2 is obtained from the zero-frequency speed of sound extrapolated by Fuentes Losa from his experimental measurements. Because the extraction of a dispersion coefficient from the data is subject to considerable arbitrariness (for instance, Fuentes Losa's use of a parabolic fitting systematically overestimates the slope and underestimates the zero-frequency speed of sound), we shall provide a check of the accuracy of the variational method by directly comparing the theoretical and experimental sound speeds at low frequencies. Notice, however, that the figure is not meant to compare the full experimental and theoretical curves, but only their respective slopes at the origin of frequencies (the only 'exact' quantity that the nature of the trial function used allows).

Figure 3 corresponds to initial-dispersion experiments for He–Xe mixtures. Nominal Xe concentrations x_2 are indicated at each curve, though the concentration used for the calculation of the variational sound-speed curve is obtained from the extrapolated zero-frequency speed give by Fuentes Losa. In the case He–Xe, $m_1 \ll m_2$, so that $s \approx \Omega/(Scx_2)$ and the range of validity of the variational predictions ($\Omega \ll 1$) is increasingly wide in terms of s^2 as x_2 decreases (as is apparent from the figure). The agreement is in general good, even though slight differences are noticeable for intermediate values of x_2. The cause of this mismatch is not clear, but a possible source might be the absence of experimental data close enough to zero

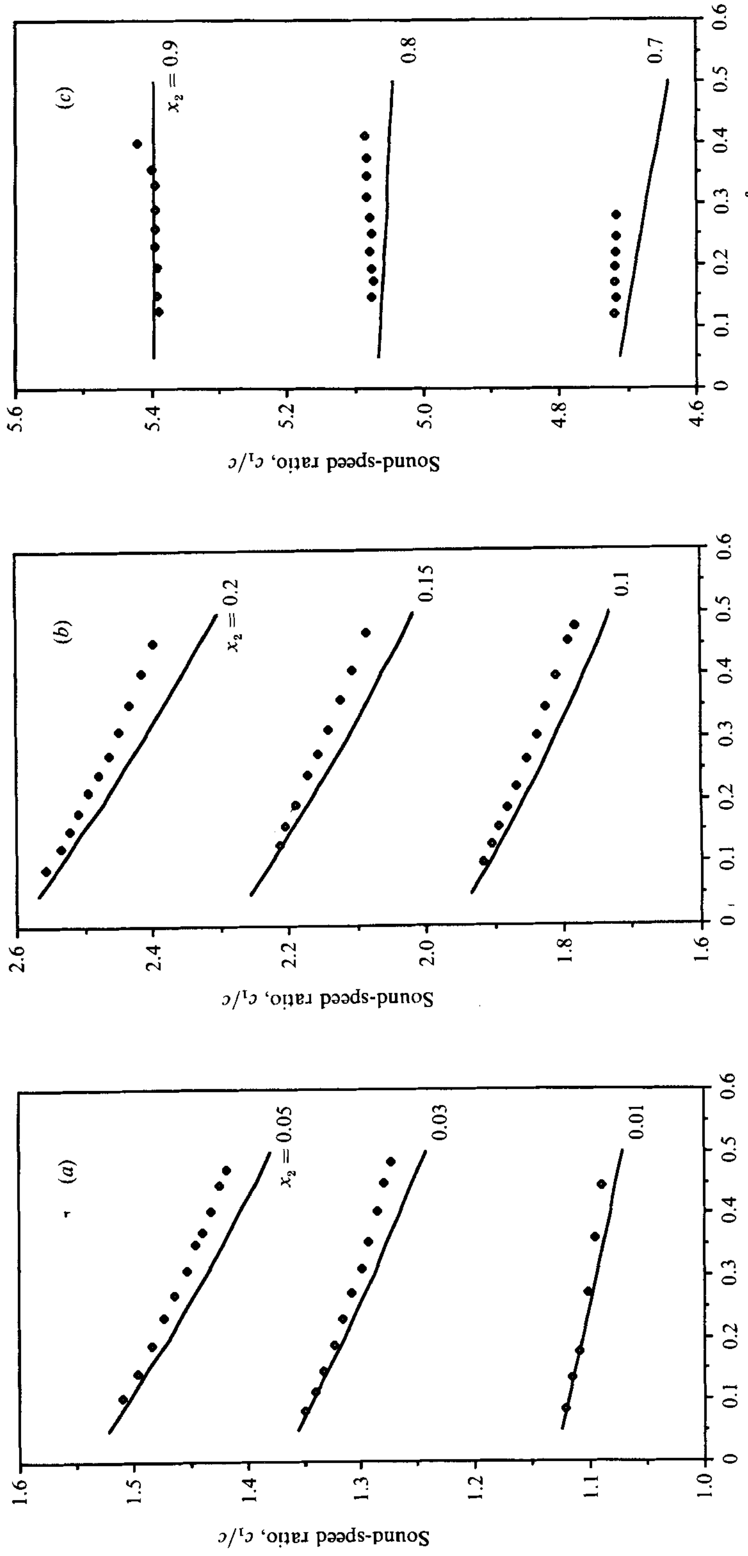

FIGURE 3. (a–c). Sound-speed ratio c_1/c in a He–Xe mixture (c_1: equilibrium sound speed of the pure light gas; c: actual speed of sound in the mixture at the given frequency) as a function of dimensionless frequency s^2 for different nominal concentrations (x_2 = molar fraction of Xe). ——, Variational calculation; ◆ ◆ ◆ ◆ ◆ ◆, experiments by Fuentes Losa (1972).

frequency, or our truncation of Sonine expansions for the coefficients β_j at the first term. A similar set of figures for the He–Ar case, showing an equally satisfactory agreement, is not presented here for brevity's sake.

Notice finally that the predictions are valid only at small values of s, as a result of the narrow frequency range of validity of the Chapman–Enskog theory for mixtures with disparate masses. An asymptotically exact result valid in the far broader region $s \leqslant 1$ could be derived by using the predictions from the two-fluid Chapman–Enskog theory (Fernandez Feria & Fernandez de la Mora 1986) as trial functions.

This work owes much to the variational advice of Professor I. B. Bernstein. P.R.Ch. was sponsored by the US–Spanish Joint Committee for Cultural and Educational Cooperation. Much stimulus was derived during the course of this research from unfounded hopes to eventually obtain some financial support.

Appendix A

Some integral relations used in the derivation of (26) are given here. Let $f(\xi)$ be an arbitrary function of ξ, $\boldsymbol{M}$ a constant matrix, and consider integrals taken over the whole space. Because of symmetry considerations, the following equalities hold:

$$\int \xi\xi f(\xi)\,\mathrm{d}^3\xi = \tfrac{1}{3}\int \xi^2 f(\xi)\,\mathrm{d}^3\xi\,\boldsymbol{I} \tag{A 1}$$

$$\begin{aligned}\int\!\left(\boldsymbol{M}:\xi^0\xi\right)\left(\boldsymbol{M}:\xi^0\xi\right)f(\xi)\,\mathrm{d}^3\xi &= \tfrac{1}{5}\boldsymbol{M}:\boldsymbol{M}^0\int\!\left(\xi^0\xi:\xi^0\xi f(\xi)\,\mathrm{d}^3\xi\right.\\ &= \int\!\left(\boldsymbol{M}:\xi\xi\right)\left(\boldsymbol{M}:\xi^0\xi\right)f(\xi)\,\mathrm{d}^3\xi = \tfrac{1}{5}\boldsymbol{M}:\boldsymbol{M}^0\int \xi^0\xi:\xi\xi f(\xi)\,\mathrm{d}^3\xi,\end{aligned} \tag{A 2}$$

where $\xi^0\xi$ and $\boldsymbol{M}^0$ stand for the symmetrized traceless parts of $\xi\xi$ and $\boldsymbol{M}$, respectively.

Appendix B

The dimensionless transport groups $\beta_i{}^*$ can be expressed in terms of the Sonine coefficients. To that end, (19)–(22) are used and a dimensionless form of the bracket product is introduced:

$$[f_{0i}\phi_i, \psi_i]_i = n_i\left(\exp\left(-\xi_1^2\right)\phi_i, \psi_i\right), \tag{B 1}$$

where
$$(f_i, g_i) = \pi^{-\frac{3}{2}}\int f_i(\xi_i)\,g_i(\xi_i)\,\mathrm{d}^3\xi_i.$$

The coefficients a_p, b_p, d_p of the Sonine expansions of $\boldsymbol{A}_i$, $\boldsymbol{B}_i$ and $\boldsymbol{D}_i$ are also non-dimensionalized

$$a_p^* = a_p\frac{nk}{\lambda}\left(\frac{kT}{m_0}\right)^{\frac{1}{2}}, \quad b_p^* = \frac{b_p\,nkT}{\mu}, \quad d_p^* = d_p\frac{x_1x_2}{D}\left(\frac{kT}{m_0}\right)^{\frac{1}{2}},$$

and the equality

$$\left[\exp\left(-\xi^2\right)S_m^{(p)}\left(\xi^2\right)\xi^{2m-1}, S_m^{(p)}\left(\xi^2\right)\right] = 2\delta_{pq}\frac{\Gamma'(m+p+1)}{p!} \tag{B 2}$$

is used, where $S_m^{(p)}$ are the Sonine polynomials as defined by Chapman & Cowling (1970), and we define $\Gamma'(x) = \Gamma(x)\,\pi^{-\frac{1}{2}}$.

The following is obtained:

$$\beta_1^* = \tfrac{3}{2}\epsilon_1\epsilon_2 k_T^2\, d_0^{*2}\left(\frac{\Delta}{\Lambda}\right)^2 + 2\Sigma\, x_i\, \Sigma^{(i)}\left(a_p^* + k_T\frac{\Delta}{\Lambda}d_p^*\right)^2\frac{\Gamma'(\frac{5}{2}+p)}{p!}, \tag{B 3}$$

$$\beta_2^* = \tfrac{4}{3}\Sigma\, x_i\, \Sigma^{(i)}\, b_P^{*2}\frac{\Gamma'(\frac{5}{2}+p)}{(p-1)!}, \tag{B 4}$$

$$\beta_3^* = \tfrac{3}{2}\epsilon_1\epsilon_2 k_T^2\, d_0^{*2} + 2\Sigma x_i \Sigma^{(i)}\, d_p^{*2}\frac{\Gamma'(\frac{5}{2}+p)}{p!}, \tag{B 5}$$

$$\beta_4^* = \tfrac{3}{2}\epsilon_1\epsilon_2 k_T d_0^{*2}\frac{\Delta}{\Lambda} + 2\Sigma\, x_i\, \Sigma^{(i)}\, (a_p^* + k_T\frac{\Delta}{\Lambda}d_p^*)\, d_p^*\frac{\Gamma'(\frac{5}{2}+p)}{p!}, \tag{B 6}$$

$$B_5^* = \tfrac{2}{3}\Sigma^{(i)} x_i M_i^{-\frac{1}{2}}\, \boldsymbol{v}_i \boldsymbol{b}_i : \boldsymbol{M}, \tag{B 7}$$

$$\beta_6^* = \tfrac{2}{3}\Sigma^{(i)}\, x_i M_i^{-\frac{1}{2}}\, \boldsymbol{w}_i \boldsymbol{b}_i : \boldsymbol{M}, \tag{B 8}$$

where the $\Sigma^{(i)}$ represent sums over index p (from $p = -\infty$ to $p = 0$ when $i = 2$, and from $p = 0$ to $p = \infty$ when $i = 1$), while the unlabelled symbols Σ are sums over the two species, from $i = 1$ to $i = 2$. In (B7)–(B 8) we denote

$$\boldsymbol{v}_1 = k_T\frac{\Delta}{\Lambda}\boldsymbol{w}_1 + (a_0^*, a_1^*, a_2^*, \ldots),$$

$$\boldsymbol{v}_2 = k_T\frac{\Delta}{\Lambda}(-R_2^{\frac{1}{2}}\epsilon_1 d_0^*, d_{-1}^*, d_{-2}^*, \ldots) + (a_0^*, a_{-1}^*, a_{-2}^*, \ldots),$$

$$\boldsymbol{w}_1 = (R_1^{\frac{1}{2}}\epsilon_2 d_0^*, d_1^*, d_2^*, \ldots),$$

$$\boldsymbol{w}_2 = (-R_2^{\frac{1}{2}}\epsilon_1 d_0^*, d_{-1}^*, d_{-2}^*, \ldots),$$

where $R_i = m_i/(m_1+m_2)$, and $\boldsymbol{M}$ is an infinite matrix whose first terms are $M_{11} = -M_{21} = 2\Gamma'\frac{7}{2}$, $M_{22} = 2\Gamma'\frac{9}{2}$ and $M_{12} = 0$.

Appendix C

In the pure-gas limit, using the Chapman–Enskog first-order solution, ϕ_1 becomes

$$\phi_1 = -\frac{1}{n}\left(\frac{2kT}{m}\right)^{\frac{1}{2}}\boldsymbol{A}\cdot\nabla\ln T - (2/n)\,\boldsymbol{B}:\nabla\boldsymbol{U}. \tag{C 1}$$

(Observe the different choice of $\boldsymbol{A}$ and $\boldsymbol{B}$ as compared with the expressions for a gas mixture.) $\boldsymbol{A}$ and $\boldsymbol{B}$ are expanded in terms of Sonine polynomials

$$\boldsymbol{A} = \Sigma\, a_p \boldsymbol{a}^{(p)}, \tag{C 2}$$

$$\boldsymbol{B} = \Sigma\, b_p \boldsymbol{b}^{(p)}, \tag{C 3}$$

where $\boldsymbol{a}^{(p)}$ and $\boldsymbol{b}^{(p)}$ are defined in Chapman & Cowling, and the sums extend from $p = 1$ to $p = \infty$. The following dimensionless coefficients are introduced:

$$a_p^* = a_p\frac{5k^2T}{2m\lambda}, \quad b_p^* = \frac{b_p kT}{\mu}. \tag{C 4}$$

When the $\boldsymbol{A}$ and $\boldsymbol{B}$ expansions are truncated at $p = 1$, $a_1^* = -1$ and $b_1^* = 1$. The coefficients α, β and δ of (32) are defined as

$$\alpha = \Sigma\, a_p^{*2} \frac{\Gamma(\frac{5}{2}+p)}{p\,!\,\Gamma\frac{7}{2}}, \tag{C 5}$$

$$\beta = \Sigma\, b_p^{*2} \frac{\Gamma(\frac{5}{2}+p)}{(p-1)\,!\,\Gamma\frac{7}{2}}, \tag{C 5}$$

$$\delta = \boldsymbol{vb}:\boldsymbol{M}/\Gamma(7/2), \tag{C 6}$$

where $\boldsymbol{M}$ is as in Appendix B, and $\boldsymbol{v} = (a_1^*, a_2^*, a_3^*, \ldots)$, $\boldsymbol{b} = (b_1^*, b_2^*, b_3^*, \ldots)$.

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