Two-fluid Euler theory of sound dispersion in gas mixtures of disparate masses

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The suitability of an Euler-level two-fluid theory to describe the behaviour of gas mixtures with disparate masses is explored for the problem of sound propagation at frequencies high enough that dispersion effects are important. The determination of the speed of propagation is reduced to solving a quadratic equation in the complex plane. The model leads to small errors of the order of the molecular mass ratio M when the molar fraction x_p of the heavy gas is small $(x_p = O(M))$, becoming increasingly inaccurate at larger values of x_p . Yet agreement with He-Xe experiments is excellent for the whole range of frequencies tested, up to values of $x_p = 0.4$. For values of x_p above 0.5 our quantitative results become poorer but they still agree qualitatively with experiments, predicting small and negative dispersion coefficients and the presence of a bifurcation at critical values of the frequency and the composition. It is concluded that this generalized Euler theory provides an excellent framework within which to develop a two-fluid boundary-layer description of the peculiar dynamics of disparate-mass mixtures in the region of parameters of greatest industrial interest.

1. Introduction

The non-equilibrium behaviour of mixtures of two gases with large disparities in their molecular weights is considerably richer than that of mixtures made of similar gases, because very different internal relaxation timescales are present simultaneously. The two fast timescales occurring are those of Maxwellization of each species, given roughly by the time between self-collisions, of the order of μ_i/p_i , where μ_i and p_i are the coefficient of viscosity and the partial pressure of one of the species (i) alone. Because the viscosity coefficient of a pure neutral gas is roughly mass-independent, these two self-equilibration times are comparable with each other, \dagger unless one of the species is in a small molar proportion in the mixture (i.e. $p_i \ge p_j$). The slow scale is that characterizing the transfer of energy between the two species, which, for purely mechanical reasons, is slower than the Maxwellization scale by a factor of the order of m_p/m , the ratio of masses between the heavy (subscript p for particle) and the light molecule. Such a picture suggests that the system may be characterized by the standard laws of hydrodynamics only when the external excitation time is large

† In an interesting paper advancing many of the ideas underlying the treatment of disparate-mass mixtures, Grad (1960) stated that the rates of self-relaxation would be faster for the light than for the heavy gas by a factor of the order of the square root of the mass ratio. Although the validity of such a scaling has often been accepted in the literature, it does not hold at all for the important case of the noble gases, whose viscosity coefficients at 20 °C are 194.1, 311.1, 221.7, 250.4 and 226.0 micropoises for He, Ne, A, Kr and Xe, respectively. The self-collision relaxation times are thus practically independent of the molecular weight. relative to the slow scale. In the much broader region in which the characteristic excitation time is large compared to the fast relaxation time and comparable to the slow one, the problem may still be described within a generalized hydrodynamic framework, because the two species have near-Maxwellian distribution function with two different temperatures, and perhaps also two different mean velocities. To proceed with rigour, one has to go through a sort of two-fluid Chapman-Enskog procedure in order to generate hydrodynamic equations with correct transport coefficients. Short of that laborious exercise, several two-fluid equations have been derived for the special case of Maxwell molecules by Goldman & Sirovich (1967), Hamel (1966), Goebel, Harris & Johnson (1976), and others (see Goebel *et al.* 1976).

Obviously, the acoustic problem is, by its simplicity, best suited to check the validity of these theories against experiments. A first test of Goldman's (1968) predictions for sound absorption was made by Prangsma, Jackson & Beenakker (1970) on the basis of their own measurements in He–Ar mixtures, with good results for Argon molar fractions $x_{\rm Ar} = 0.1$ and 0.25; but less good for $x_{\rm Ar} = 0.05$, 0.5 and 0.75. Pointing out that, unlike absorption, dispersion cannot be predicted by the Navier–Stokes equations (not even at small frequencies), Fuentes Losa & Foch (1972) compared their measurements on initial (low frequency) sound dispersion for He–Ar mixtures with a direct solution of the Boltzmann equation (Foch, Uhlenbeck & Fuentes Losa 1972). Such a test gave excellent results for all compositions, but was confined to frequencies small compared to the inverse of the small relaxation time, because the Boltzmann equation had been solved in an expansion in powers of the frequency.

Perhaps the most interesting findings of Fuentes Losa & Foch are those on dispersion in He-Xe mixtures (unpublished), available only in Fuentes Losa's (1972) thesis, and showing negative dispersion coefficients in Xe-rich mixtures. More recently, Huck & Johnson (1980) have explored the predictions of the two-temperature model of Goebel et al. (1967) for He-Xe mixtures. Their striking predictions on two different propagating modes at frequencies still within the range of validity of the model are a remarkable exponent of the richness of the problem. A number of these unusual features have been confirmed experimentally by Bowler (1984) and Bowler & Johnson (1985), which explores a range of concentrations and frequencies broader than that studied by Fuentes Losa. At this point, the finest available experiments to test two-fluid theories are those involving He-Xe mixtures. With a ratio m_p/m more than three times larger than for He-Ar mixtures, such data ought to give excellent results when compared to a theory exploiting systematically the existing mass disparity. The purpose of this paper is, therefore, to compare such data with the lowest order (Euler level) two-fluid theory, and to suggest ways to systematically improve this model.

The fluid-dynamic importance of the Euler equations, particularly when coupled to boundary-layer theory, is too monumental for us to try to make its defence here. Obviously, the additional complexities involved in two-fluid hydrodynamic equations provide a greater stimulus towards exploring the possible usefulness of a generalized two-fluid Euler theory, as a first step prior to the development of a two-fluid boundary-layer theory. Some novel technologies have started to exploit the very remarkable behaviour of heavy gases (carried in He or H₂), whose large inertia can be used to induce energetic surface collisions (tens of eV) at extremely high fluxes (Fernandez de la Mora 1985). These potential new applications call for improvements in existing techniques for computing the evolution of such exotic gas mixtures, and a first step in this direction is to explore whether a two-fluid Euler theory can play a similarly fruitful role in this new field to that which Euler's equations play in ordinary fluid mechanics. We shall conclude that, for the acoustic problem, a two-fluid Euler theory yields excellent results over the whole frequency range of interest and up to a molar fraction of Xe in He of around 0.4, fully covering the region of parameter values where disparate-mass mixtures are of practical relevance.

2. Governing equations

The governing equations are derived by assuming that the two species have different Maxwellian velocity distribution functions, with mean velocities $U_{
m p}$ and Uand temperatures $T_{\rm p}$ and T for the heavy and the light gas, respectively. One may write the convective terms in the conservation equations for the mass, momentum and energy for each of the gases, in total analogy with the case of a pure gas, by realizing that, in the particular reference frame moving with the mean speed of one of the species, its viscous stress tensor and heat flux are zero for a Maxwellian distribution function. However, there is now a net interchange of momentum and energy between the two species which may be computed from the appropriate Boltzmann collision integral (Burgers 1969) since the velocity distribution functions of the two species are specified. When the velocity difference $U_p - U$ is small compared to the sound speed of the light gas, these momentum and energy coupling terms are linear in the velocity and temperature differences. Such a condition is obviously satisfied in the acoustic problem, where we may also linearize around the unperturbed densities ρ_0 , ρ_{p0} , temperature T_0 (the same for the two species) and null mean velocities:

$$\rho = \rho_0 + \rho',\tag{1}$$

$$\rho_{\mathbf{p}} = \rho_{\mathbf{p}\mathbf{0}} + \rho_{\mathbf{p}}',\tag{2}$$

$$T = T_0 + T', \tag{3}$$

$$T_{\rm p} = T_0 + T_{\rm p}^{\prime},\tag{4}$$

where the quantities with and without a subscript p refer to the heavy and the light species respectively. The linearized conservation equations for the two monatomic gases are

$$\partial_{\mathbf{t}} \boldsymbol{\rho}' + \boldsymbol{\rho}_{\mathbf{0}} \boldsymbol{\nabla} \cdot \boldsymbol{U} = 0, \tag{5}$$

$$\rho_{\mathbf{0}}\partial_{\mathbf{t}} \boldsymbol{U} + \boldsymbol{\nabla} \boldsymbol{p} = -\frac{\rho_{\mathbf{p}\mathbf{0}}(\boldsymbol{U} - \boldsymbol{U}_{\mathbf{p}})}{\tau}, \qquad (6)$$

$$\partial_{\mathrm{t}} T + {}^{2}_{3}T_{0} \nabla \cdot U = \frac{2\rho_{\mathrm{p}0} m}{\rho_{0} m_{\mathrm{p}}} \frac{T_{\mathrm{p}} - T}{\tau}, \qquad (7)$$

$$\partial_{\mathbf{t}} \rho_{\mathbf{p}}' + \rho_{\mathbf{p}\mathbf{0}} \nabla \cdot \boldsymbol{U}_{\mathbf{p}} = 0, \tag{8}$$

$$\rho_{\mathbf{p}\mathbf{0}} \partial_{\mathbf{t}} U_{\mathbf{p}} + \nabla p_{\mathbf{p}} = \frac{\rho_{\mathbf{p}\mathbf{0}} (U - U_{\mathbf{p}})}{\tau}, \qquad (9)$$

$$\partial_{\mathrm{t}} T_{\mathrm{p}} + \frac{2}{3} T_{\mathrm{0}} \nabla \cdot \boldsymbol{U}_{\mathrm{p}} = -\frac{2(T_{\mathrm{p}} - T)}{\tau}, \qquad (10)$$

and the partial pressures are, for Maxwellian distributions,

$$p = \frac{\rho kT}{m},\tag{11}$$

$$p_{\rm p} = \frac{\rho_{\rm p} \, kT_{\rm p}}{m_{\rm p}}.\tag{12}$$

The constant τ is the slow relaxation time governing the rates of interspecies momentum and energy transfer. It is related to the mixture diffusion coefficient Dthrough Einstein's relation

$$\tau = \frac{Dm_{\rm p}}{kT} \frac{n + n_{\rm p}}{n},\tag{13}$$

where $n_i = \rho_i / m_i$ is the number density of species *i*.

The principal limitation of these equations arises from our neglect of heat fluxes and viscous stresses. The relative importance of viscous effects measured by the dimensionless frequency $\mu_i \omega/p_i$, which may be written

$$\frac{\mu\omega}{p} = \frac{mSc}{m_{\rm p}}\omega\tau,$$
$$\frac{\mu_{\rm p}\omega}{p_{\rm p}} = \frac{mSc_{\rm p}}{m_{\rm p}}\frac{p}{p_{\rm p}}\omega\tau$$

for the light and heavy gas, respectively. Sc and Sc_p are the diffusivity ratios

$$Sc_i = \frac{\mu_i}{m(n+n_{\rm p})\,D},$$

which take the values 2.33 and 2.71 for He and Xe respectively, in a He–Xe mixture. Accordingly, the viscous corrections are some $M \equiv m/m_{\rm p}$ times smaller than $\omega\tau$, and (6) and (9) contain errors of order M when $\omega\tau$ takes values up to order one. A similar conclusion may be reached for the heavy-gas energy equation (10), where the neglected heat flux is also of order $M\omega\tau$. But the heat flux term neglected in the light-gas energy equation (7) is of order $\mu\omega/(\rho c_{\rm R}^2)$ with respect to the $\partial T/\partial t$ term, where $c_{\rm R}$ is the mixture sound speed. This term is thus negligible only if $\rho c_{\rm R}^2$ is comparable to p, a condition which becomes less and less true as the heavy gas is more concentrated. For that reason, the present two-fluid Euler model is correct to lowest order in M only for moderate values of the molar fraction of the heavy gas.

Let us now assume plane waves in which the linearized hydrodynamic quantities depend on time and position through the factor

$$f' = f_0 \exp(i\omega t - i\lambda x), \tag{14}$$

and introduce the notation

$$-i\alpha = \omega \tau,$$
 (15)

$$c \equiv \frac{\omega}{\lambda} \equiv \left(\frac{\gamma k T_0}{m}\right)^{\frac{1}{2}},\tag{16}$$

$$M \equiv \frac{m}{m_{\rm p}},\tag{17}$$

$$\epsilon \equiv \frac{\rho_{\rm p0}}{\rho_0},\tag{18}$$

$$y \equiv \frac{U}{c},\tag{19}$$

$$y_{\mathbf{p}} \equiv \frac{U_{\mathbf{p}}}{c},\tag{20}$$

$$z \equiv \frac{T'}{T_0},\tag{21}$$

$$z_{\rm p} \equiv \frac{T_{\rm p}'}{T_0},\tag{22}$$

where M and ϵ are the mass and density ratios, α is a (purely imaginary) frequency parameter, and γ is a dimensionless complex speed of sound entering into the problem as an eigenvalue. Eliminating the densities by means of the mass conservation equations we obtain

$$y - \frac{(y+z)}{\gamma} + \frac{\epsilon \left(\theta - \theta_{\rm p}\right)}{\alpha} = 0, \tag{23}$$

$$y_{\mathbf{p}} - \frac{M(y_{\mathbf{p}} + z_{\mathbf{p}})}{\gamma} - \frac{(y - y_{\mathbf{p}})}{\alpha} = 0, \qquad (24)$$

$$z - \frac{2}{3}y - \frac{2\epsilon M(z_{\rm p} - z)}{\alpha} = 0, \qquad (25)$$

$$z_{\rm p} - \frac{2}{3}y_{\rm p} + \frac{2(z_{\rm p} - z)}{\alpha} = 0.$$
 (26)

Before evaluating the speed of sound, it is worth taking the difference between the two equations for the temperatures ((25), (26)) to obtain

$$z - z_{\mathbf{p}} = \Sigma^{-1} (y - y_{\mathbf{p}}), \qquad (27)$$

$$\Sigma = \frac{3}{2} + \frac{3}{\alpha} (1 + M\epsilon). \tag{28}$$

Accordingly, the velocity and the temperature differences for the acoustic problem are in a constant ratio, with the proportionality coefficient Σ . The group ϵM is the ratio of unperturbed number densities, $n_{\rm p}/n$, so that Σ is a quantity of order unity except in the low-frequency limit, and in the Lorentz limit $n \ll n_n$. Accordingly, except in these two extreme cases, it is not consistent to allow for two different temperatures without having also two different velocities. This conclusion seems to contradict the common view that the energy, and not the momentum, is inefficiently transferred between different species, so that more temperature than velocity separation should be expected. Indeed, if one considers a homogeneous medium $(\partial/\partial x = 0)$ where two species have different initial velocities and temperatures, combining (6) and (9), and (7) and (10), respectively, one obtains the relaxation times $\tau_v = \tau/(1+\epsilon)$ and $2\tau_T = \tau/(1+M\epsilon)$ for the velocity and temperature slip respectively. Therefore, except in the limit where the heavy gas is dilute ($\epsilon \simeq 1$), the velocity differences do relax much faster than the differences in temperatures. But this conclusion holds only in the absence of spatial gradients. In our acoustic problem there are velocity gradients which lead to compressive heating of each gas proportionally to its own velocity gradient. Indeed, the only driving force leading to temperature differences in our model is precisely the velocity difference, as expressed in (27). The dimensionless temperature slip is thus, at best, two-thirds of the dimensionless velocity slip, and the conclusions reached for a homogeneous situation cannot be extrapolated into the acoustic problem. An interesting corollary of this fact is that, for ϵ large, both velocity and temperature slip are small, and dispersive effects will necessarily be small.

where

3. The speed of propagation

The condition for non-trivial solutions of the system (23)–(26) fixes γ through the quadratic equation

$$q\sigma^{2} - (l+q+1)\sigma + l = 0, \qquad (29)$$

where we have introduced the new velocity variable

$$\sigma \equiv \frac{5}{3\gamma} \frac{1 + M\epsilon}{1 + \epsilon},\tag{30}$$

which is equal to one in the equilibrium limit $(\alpha = 0)$, and

$$q \equiv \frac{3M}{5\epsilon} \frac{(1+\epsilon)^2}{(1-M)^2} [1+\Sigma^{-1}],$$
$$l \equiv \left(1+\frac{\epsilon+1}{\alpha}\right) \frac{(1+M\epsilon)^2}{(1-M)^2} \frac{1}{\epsilon}.$$

For the present situation where the frequency ω is real, the speed of propagation $c_{\rm R}$ is given by

$$\frac{c_{\rm e}}{c_{\rm R}} = Re(\sigma^{\frac{1}{2}}). \tag{31}$$

where c_{e} is the zero-frequency speed of sound for the mixture,

$$c_{\rm e}^2 = \frac{5}{3} \frac{kT_0}{m} \frac{1+M\epsilon}{1+\epsilon}.$$
(32)

Therefore, the problem of finding the sound speed of the mixture reduces to solving a quadratic equation in the complex plane, and evaluating the inverse of the real part of the square root of σ . The two resulting roots are characterized by very different absorption coefficients at low frequencies, so that only the least damped of the modes may be observed. But at higher frequencies, both modes have comparable absorption, and may be present simultaneously (see Huck & Johnson 1980). Some results are shown in figure 1 for the mode dominating at low frequency and the particular case M = 4/131.3 (He-Xe).

4. Comparison between theory and experiments

Because of the considerable differences in precision and range of parameters covered by the two available sources of experimental data on sound speed in He–Xe mixtures, we will discuss them separately, starting with Fuentes Losa.

In order to avoid corrupting the accuracy of the sound speed results with the less precise information available on species composition, the data will be made dimensionless in a concentration-independent fashion. We will thus use $c_{\rm R}/c_0$ for our dependent variable, where c_0 is the equilibrium sound speed of the pure light gas, defined by

$$c_0^2 = \frac{5}{3} \frac{kT_0}{m}.$$
 (33)

Similarly, we introduce the frequency variable

$$s' = \omega \tau (1 - x_{\rm p}), \tag{34}$$



FIGURE 1. Non-dimensional inverse sound speed $c_0/c_R vs$. the frequency parameter $s' = \omega \tau (1-x_p)$ for various values of the Xe molar fraction x_p . Dotted lines correspond to the predictions of the two-fluid Euler theory, while open data symbols are from Fuentes Losa (1972) for nominal values of $x_p = 0.01, 0.03, 0.05, 0.1, 0.15, 0.2, 0.3, 0.7, 0.8$ and 0.9, increasing upwards. Only the mode dominating at low frequency is displayed. Filled data symbols from Bowler (1984) for reported concentrations x_p of 0.2, 0.3, 0.4, 0.55, 0.6, 0.7 and 0.8. The abscissa gives values of $(s')^2$.

where $x_{\rm p}$ is the molar fraction of the heavy species

$$x_{\rm p} = \frac{n_{\rm p}}{(n+n_{\rm p})}.\tag{35}$$

From (13), s' may be expressed as

$$s' = \frac{\omega m_{\rm p} D(n+n_{\rm p})}{p_{\rm t}},\tag{36}$$

where $p_t = p + p_p$ is the total pressure of the mixture, reported in the experiments, while the group $D(n+n_p)$ is pressure and composition independent to the first approximation in the Chapman-Enskog theory. Therefore s' may be obtained from the data without knowledge of the value of x_p . The temperature dependence of the diffusion coefficient D can be fitted to the data reported by Vargaftik (1975) for He–Xe mixtures at one atmosphere as

$$D = 0.473 \left(\frac{T}{273.3 \text{ K}}\right)^{1.7} \text{ cm}^2/\text{s},$$
(37)

leading to
$$s' = 2.077 \left(\frac{T'}{273.3 \text{ K}}\right)^{0.7} \frac{\omega(\text{M Hertz})}{p_t(\text{torr})}.$$
 (38)

The converted experimental data have been plotted also in figure 1.

Notice first the general trends predicted by the model. For small values of $x_{\rm p}$, $c_0/c_{\rm R}$ falls monotonically from the zero-frequency (equilibrium) value, to the high-frequency asymptote (unity), corresponding to the limit where the light gas oscillates on its own, uncoupled from the heavy molecules. For values of $x_{\rm p}$ close to unity, $c_0/c_{\rm R}$ increases monotonically (negative dispersion, first observed and predicted by Fuentes Losa) towards the pure heavy-gas equilibrium speed of sound.[†] Obviously, there must be a critical value of $x_{\rm p}$ separating the curves which asymptote at high frequencies towards the pure light-gas sound speed from those asymptoting to the pure heavy-gas speed of propagation. Indeed, at a value of $s' \simeq 1.36$ and $x_{\rm p} \simeq 0.52$, the discriminant of the quadratic equation (29) vanishes, and the roots cross through a singular point where the curve $c_{\rm R} vs. s'$ bends itself abruptly through 90 degrees. This singular behaviour is similar to that described in greater detail by Huck & Johnson (1980).[‡] Obviously, there is no dispersion for the two extreme cases of pure gases, since the governing equations reduce to Euler's.

Notice that the theoretical curves for $x_p = 0.0086$, 0.0274, 0.0411, 0.0884, 0.1291, 0.1796 and 0.275 go nearly through the experimental points for which x_p was reported to be 0.01, 0.03, 0.05, 0.1, 0.15, 0.20 and 0.30 respectively. Such nominal concentrations cannot be correct because the equilibrium sound speed corresponding to these reported concentrations does not coincide with the value of c_R resulting from extrapolating the data to zero frequency (see Fernandez de la Mora 1984, table 1). We therefore attribute the disagreement in the x_p label to inaccuracies in the measurement of x_p . But once the experimental value of x_p is inferred from the zero-frequency value of c_R extrapolated from the data, the agreement between theory and experiments is excellent for concentrations up to 0.3. For concentrations above 0.6, experiments and theory show that dispersion is relatively small and negative. But the predicted values are clearly larger than those observed. This disagreement

[†] This simple asymptotic behaviour at large frequencies arises from considering (23)-(26) in the limit $\alpha \ge i$. To lowest order $(\alpha = \infty)$ the coupling terms disappear, and the gases behave as if they were alone. The propagation speeds of the two modes thus degenerate into the sound speeds for the two pure gases, with $(c_0/c_R)^2$ becoming 1 and M^{-1} , while their corresponding eigenvectors (y, z, y_p, z_p) are $(1, \frac{2}{3}, 0, 0)$ and $(0, 0, 1, \frac{2}{3})$ respectively. To lowest order the amplitude of oscillation is null for the gas which is not directly responsible for the propagation. The coupling can be obtained to first order in the inverse frequency, together with the corresponding coefficients of absorption, which scale with x and x_p respectively. This result is consistent with the fact that the mode observed in Xe-rich mixtures is the slow one, while the fast mode occurs in He-rich mixtures. But the value of x_p at which the two asymptotic absorptions coincide is different from the critical one.

¹To lowest order in M, the condition that the discriminant of (29) vanishes takes the form $x_p = 5(\alpha'+2) [7\alpha'+8-(5\alpha'+6)^{\frac{1}{2}}(9\alpha'+10)^{\frac{1}{2}}]/(8\alpha')$. The critical values x_p^* and α'_* may be found as the intersection of this expressions's right-hand side with the real axis when α' is purely imaginary, yielding $s'_* = 1.3159740$ and $x_p^* = 0.518790$. Bowler & Johnson (1985) report predictions for the critical values s'_* and x_p^* of 1.387 and 0.505 for Maxwellian molecules, and 1.276 and 0.495 for hard spheres. The agreement between their model and ours is thus quite good, even though our results cannot be very precise for such high concentrations of Xe ($\rho_p/\rho = 35$).

will be rationalized below, and used to motivate future refinements for the present theory.

Figure 1 shows also the experimental points of Bowler (1984), also reported by Bowler & Johnson (1985). These data have the extremely interesting feature of reaching the critical region $s' \simeq 2$, covering also the range of concentrations $0.4 < x_{\rm p} < 0.6$ left unexplored by Fuentes Losa. Clearly, our previous conclusions are confirmed further by these new data. Provided that $x_p \lesssim 0.4$, the two-fluid Euler theory yields excellent predictions in all the frequency range tested. For $x_n \approx 0.5$, the dispersions obtained by Bowler & Johnson (1985) are far smaller than ours, and if the present model seemed poor against the data of Fuentes Losa, it leaves even more to be desired at these high frequencies. Bowler & Johnson have compared their experimental results with a two-temperature theory based on Grad's 13 polynomial expansion. Their predictions fit the experimental data reasonably well for the supercritical values of x_{p} , if one takes into account how abruptly the sound speed depends on all parameters near the (branch point) singularity. Our predictions are superior, however, for $x_p \gtrsim 0.4$, although the comparison is clouded again by what appears to be inaccuracy in the measurement of x_p : the extrapolated experimental zero-frequency sound speeds do not agree with the equilibrium values of (32). It is striking that the data for which both Bowler and Fuentes Losa have reported concentrations x_p of 0.2, 0.3, 0.7, and 0.8 agree with each other in missing the equilibrium sound speed by similar amounts (figure 1). Such a coincidence has made us wonder whether there might be a fundamental reason behind this low-frequency mismatch between experiments and equilibrium sound speed. The only source of error involved in (32) of which we can think is associated with non-ideality of the gas. But this effect appears to be negligible, because, for Xe (the most non-ideal component of the mixture, with a critical temperature of 289.7 K), the product $B_p n_p$ between the virial coefficient and the number density takes the value 2.4×10^{-4} for the highest pressures of Bowler & Johnson. Furthermore, the observed mismatch shows trends opposite to those one would expect from non-idealities, increasing for decreasing partial pressures of Xe. The alternative hypothesis that the Xe might have been contaminated is ruled out (except for H₂ contamination) because the observed sound speed is greater than the equilibrium value. Therefore, unless the Xe was contaminated with a lighter gas, we must conclude that both Fuentes Losa and Bowler & Johnson overmeasure the value of $x_{\rm p}$. Such an interpretation finds strong support in the good agreement obtained between experiments and our own theory in the concentration range where our errors are of the order of M (see also Fernandez de la Mora 1984). But because neither of the two experimental sources have addressed the question of accuracy in the measurement of $x_{\rm p}$, the matter must remain unresolved for the moment.

5. Dispersion at low frequencies

Additional insight on why the two-fluid Euler theory works so well below values of $x_p = 0.3$, and why it is deficient for $x_p \gtrsim 0.6$, may be obtained through a study of the behaviour at the low frequencies at which the most accurate experiments are available. Let us define the dispersion coefficient as the initial slope of the curve $c_e/c_R vs. s'^2$,

$$d_{2} = \frac{\mathrm{d}(c_{\mathrm{e}}/c_{\mathrm{R}})}{\mathrm{d}s'^{2}} = \frac{\mathrm{d}\,Re(\sigma^{\frac{1}{2}})}{\mathrm{d}s'^{2}}.$$
(39)

To determine d_2 , let us obtain σ in a power expansion in terms of the frequency, using the imaginary variable

$$\boldsymbol{\alpha}' = \mathrm{i}\boldsymbol{s}',\tag{40}$$

$$\sigma = 1 + \sigma_1 \alpha' + \sigma_2 \alpha'^2 + \dots$$
(41)

The corresponding expansions for the terms l and q entering into (29) are

$$l = l_0 + \frac{l'}{\alpha'},\tag{42}$$

with

$$l_0 \equiv \frac{(1+M\epsilon)^2}{(1-M)^2} \frac{1}{\epsilon},\tag{43}$$

$$l' \equiv l_0 \frac{1+\epsilon}{1+M\epsilon},\tag{44}$$

and

$$q = q_0 (1 + \frac{1}{3}\alpha' - \frac{1}{12}\alpha'^2 + \dots), \tag{45}$$

with
$$q_0 \equiv \frac{3M}{5\epsilon} \frac{(1+\epsilon)^2}{(1-M)^2}.$$
 (46)

Substituting these expressions into (29) and grouping together the terms of equal power in α' , there results

$$\sigma_1 = \frac{-N}{l_0},\tag{47}$$

$$\sigma_2 = \frac{N^2 (1 + l_0 - q_0)}{l_0^2},\tag{48}$$

where we have used the definition

$$N \equiv \frac{(1+M\epsilon)}{(1+\epsilon)}.$$
(49)

We may also expand the term $\sigma^{\frac{1}{2}}$ entering into (39),

$$\sigma^{\frac{1}{2}} = 1 + \frac{1}{2}\sigma_1 \alpha' + \frac{1}{8}(4\sigma_2 - \sigma_1^2) \alpha'^2 + \dots$$
 (50)

leading finally to

$$d_2 = N^2 \frac{3 + 4l_0 - 4q_0}{8l_0^2}.$$
 (51)

Although it is not easy to extract an accurate slope from the experimental data in order to determine an experimental value for d_2 , we will still plot approximate values determined with errors around 10–15%. These data are shown in figure 2 along with the theoretical predictions of (51), and confirm our previous observation on the deficiency of the theory for values of x_p beyond 0.6.

There are two main sources of error in our Euler model. Firstly, viscosity and heat-conduction effects have been neglected. Secondly, even if one were to keep these terms in a rigorous two-fluid Chapman-Enskog theory, the corresponding predictions would still be in error, at least for the two limits of pure gases, $x_p = 0$ and $x_p = 1$. This is a well-known consequence of the fact that dispersion is a second-ord reffect in the frequency, while the Navier-Stokes equations are correct only to first order. Interestingly enough, however, dispersion effects for pure gases are of order M^2 smaller than those due to the coupling of the two gases. For a pure gas, the only characteristic speed available is the sound speed, c_e , and the frequency can be made

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FIGURE 2. Dispersion coefficient $d_2 = [d(c_e/c_R)/ds'^2]_{s'=0}$ as a function of the Xenon molar fraction x_p . Data symbols are extracted from the measurements of Fuentes Losa (1972), except for those at $x_p = 0$ and $x_p = 1$, which are taken from the calculation of Wang Chang & Uhlenbeck (1970) for Maxwell molecules.

dimensionless only through the ratio of the pressure and the viscosity coefficient. The group d(x, y)

$$a_2 \equiv \frac{\mathrm{d}(c_{\mathrm{e}}/c_{\mathrm{R}})}{\mathrm{d}(\mu\omega/p)^2},\tag{52}$$

is therefore a quantity of order one, which Wang Chang & Uhlenbeck (1970) and Foch & Fuentes Losa (1972) report to be equal to 1.075 for the special case of Maxwell molecules, and which appears to be nearly independent of the intermolecular potential. Accordingly, our dispersion parameter d_2 can be obtained straightforwardly for the case of pure gases as

$$d_{2} = a_{2} \left[\frac{m}{m_{\rm p}} \frac{\mu}{mD(n+n_{\rm p})} \right]^{2}, \tag{53}$$

which is clearly a quantity of order M^2 , taking at room temperature the values $d_2 = 0.0054$ and $d_2 = 0.0073$ for pure He and pure Xe, respectively. Considering that the values of d_2 due to the interaction between the two species is as large as $0.22 (x_p = 0.08)$, these pure-gas dispersion effects are negligible for all concentrations in the interval $0 < x_p < 0.4$. But in the region beyond $x_p = 0.4$ the dispersive mechanisms due to energy and momentum transfer between the two species are relatively weak (at most four times greater than d_2 for pure Xe), so that viscosity and heat conductivity effects can no longer be neglected.

In conclusion, the model correctly predicts dispersion effects whenever these are important. But it contains errors of the order of the prediction itself in the region $x_{\rm p} \gtrsim 0.5$ where dispersion phenomena are small and comparable to those due to the pure heavy gas. Perfecting our treatment requires second-order corrections to the present Euler model. A two-fluid Chapman-Enskog theory would not be sufficient, although it would contain errors of order M^2 only.

Also, it is worth commenting on the validity of our closure in the region $x_p \leq 1$. There, the heavy gas is so dilute and has such a long self-equilibration time that it relaxes towards a Maxwellian velocity distribution function due mainly to collisions with the light gas, within times of order τ . Then, the viscous contributions to the partial pressure of the heavy species are comparable to $n_p kT_p$, and the Euler closure $p_p = n_p kT_p$ is very inaccurate. Nonetheless, the model still works well because, at small x_p , γ is of order unity, and the pressure term in (24) is small, of order M. Fortunately, the situation is such that the closure for the pressure tensor of the heavy species fails only in the region where the term is altogether negligible. In the opposite (Lorentz) limit, $l-x_p \leq 1$, the Maxwellization of the light gas is still very fast, and our closure remains valid.

Finally, we do remark again that a theory predicting dispersion in mixtures for the whole range of concentrations is available for the case of Maxwell molecules (Foch et al. 1972), and agrees well with experiments (Fuentes Losa & Foch 1972; Fuentes Losa 1972). But such a theory is based on a frequency expansion, and holds only for small values of s'. It did, nonetheless, succeed in predicting negative dispersion coefficients well before any two-temperature or two-fluid theory. Bowler & Johnson (1985) have also pointed out that, in the frequency expansion of Foch *et al.* (1977), the slow mode degenerates into the diffusion mode at low frequencies, while it degenerates into the thermal mode in their own theory. These authors attribute this disagreement to the 'approximate calculations' involved in the work of Foch et al. (1972). Yet, their frequency expansion solution is exact (for Maxwell molecules) at the low frequencies where the diffusion mode emerges. One cannot therefore so easily discard the conclusions of Foch et al. (1972). It is interesting to note that the thermal mode is absent from the two-fluid Euler model; but the diffusion mode is included, because the laws of diffusion result from a low-frequency momentum balance fully contained in our two momentum conservation equations. Indeed, the $s' \rightarrow 0$ limit of our second mode leads to the dispersion relation $\lambda^2 = -i\omega/D$, confirming its identification as a diffusion mode. But in that case the neglected heat flux of (7) is comparable to the terms retained, even in the limit where x_n is small. We cannot then take our own predictions on the small frequency behaviour of the slow mode very seriously.

6. Strengths and limitations of the theory, and higher-order extensions

A first look at the present problem has revealed the existence of two dimensionless frequencies: $\omega \mu_i / p_i$ and $\omega \tau$, of which the first is smaller than the second by a factor M, and measures the importance of viscosity and heat conductivity, while the group $\omega \tau$ measures the importance of temperature relaxation (also of slip velocity when x_p is small). Such a picture gives the impression that a two-fluid theory ignoring viscosity and heat conductivity (Euler level) would contain errors of the order of M only, while temperature separation would dominate over velocity differences except for small values of x_p . Yet, for the acoustic problem, both of these expectations are deceptive. Because the main driving force for temperature differences is the velocity slip, temperature and velocity differences are of similar magnitude, being dominant over viscosity and heat conductivity only in the region of small x_p , where the velocity slip relaxation time $\tau_v = \tau/(1+\epsilon)$ is small with respect to μ_i/p_i (that is, $M^{-1} \ge \epsilon$). A second surprise comes at the finding that a two-fluid Euler theory is not uniformly valid to order M in all the range of concentrations, but only for small values of x_n . Upon closer examination one realizes that, again, spatial gradients are responsible for this behaviour. A Knudsen number analogous to $\mu_i \omega/p_i$, but now based on the wavenumber λ , promotes the importance of heat conductivity when the wavelength becomes smaller as a result of the decrease in sound speed upon increase of x_n . Of our four governing equations three are correct to first order in M; but the heat conductivity ignored in the energy equation for the light gas is comparable to the other terms retained when x_n is of order one. This observation has some interesting implications; aside from making our theory highly suspect when $x_n = O(1)$, it also tells us that first-order theories (including heat conductivity and viscosity) will not yield accurate predictions either unless their transport coefficients are correct, since such 'first order' terms have been seen to be comparable to the convective terms in the energy equation. Unfortunately, the correct transport coefficients needed are not those appearing in the 13-moment approach of Goebel et al., because Grad's method is not a systematic perturbation theory with errors of the order of a small parameter (a Knudsen number). Furthermore, the alternative approach of Goldman & Sirovich's (1967) two-fluid theory for Maxwell molecules fails also to yield correct transport coefficients. Therefore, a complete two-fluid Chapman-Enskog theory with realistic intermolecular potentials is called for before an accurate prediction of sound propagation speeds in He-Xe mixtures may be given over the whole range of concentrations. This fact is in itself a good motivation to undertake such a laborious kinetic exercise. On the other hand, if one is interested in the behaviour of disparate-mass mixtures diluted in the heavy component, the present analysis shows that, for He-Xe mixtures, the two-fluid Euler level is excellent up to an unexpectedly high value of $x_{\rm n} = 0.4$. Such a conclusion has the very considerable importance of making it possible, in principle, to extend the methods of boundary layer theory to problems involving relaxing disparate-mass mixtures. Also, the mathematical problem posed by the two-fluid Euler equations becomes physically relevant. It might even be possible to extend classical potential theory to a two-fluid system, as Robinson (1956) has already done for the case of an incompressible dust-laden gas (see also Fernandez de la Mora & Rosner 1981).

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