Predictions for sound propagation in disparate-mass gas mixtures

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DISPARATE-MASS gases can behave differently from more familiar gases in the continuum regime, because in some circumstances the light and heavy components of the mixture can have different temperatures. Sound propagation in such gases is considered here. At moderately high frequencies, the existence of two different forced-sound modes is predicted.

Mieszaniny gazów o wysoce zróżnicowanych masach mogą zachowywać się inaczej niż lepiej znane gazy w opisie kontynualnym, ponieważ w pewnych warunkach lekkie i ciężkie składniki mieszaniny mogą mieć różne temperatury. Rozpatrzono przenoszenie dźwięku w takich mieszaninach. Przy umierkowanych częstościach przewiduje się możliwość istnienia dwóch różnych postaci drgań wymuszonych.

Смеси газов этого типа могут иметь другое поведение чем более известные газы в континуальном описании, т.к. в некоторых условиях легкие и тяжелые компоненты смеси могут иметь разные температуры. Рассмотрен перенос звука в таких смесях. При умеренных частотах предсказывается возможность существования двух разных видов вынужденных колебаний.

DISPARATE-MASS gases are those in which the molecules of one component of the mixture are very much lighter than those of the other component ($m_1 \ll m_2$). As discussed by GRAD [1], the relative inefficiency with which kinetic energy is interchanged between the heavy and light species in such a mixture can lead to a temperature separation $\Delta \equiv$ $\equiv (T_2 - T_1)$ between the species. The relaxation frequency ω_{Δ} for this temperature-difference is low compared to other characteristic relaxation frequencies in the gas. (Thus, if ω_i is characteristic self-relaxation frequency for species i, then $\omega_{\Delta} \sim (m_1/m_2)^{1/2} \omega_2 \sim$ $\sim (m_1/m_2)\omega_1$.).

The presence of such a low characteristic relaxation frequency in disparate-mass mixtures means that there are circumstances in which the equations of ordinary hydrodynamics do not give a correct description of such gases even though they may correctly be used for other, more ordinary gases. Continuum equations which describe such mixtures in these circumstances have been developed by a number of authors, from several points of view [2–7], and there is now agreement about the form which these equations should take.

We consider here the way sound propagates in a disparate-mass gas mixture when it is excited at a given frequency ω. We suppose ω low enough so that a continuum treatment is justified: $\omega \ll \omega_2$. As discussed in Ref. [2], the continuum regime consists of two parts: the *normal* regime, for which $\omega \ll \omega_{\Delta}$, and in which the usual hydrodynamic equations apply, and the *near-normal* regime, $\omega \gtrsim \omega_{\Delta}$, in which a correct description of the gas necessarily involves separate species temperatures. The latter near-normal equations are correct throughout the continuum regime, reducing to the usual equations of hydrodynamics when $\omega \ll \omega_{\Delta}$. When $\omega \gtrsim \omega_{\Delta}$, however, their nature is somewhat different. To zero

order (Euler level) the flow equations necessarily include several dissipative terms: light-species heat flux, mutual diffusion and species temperature-separation. To first order (Navier-Stokes level) these equations also include the heat flux and viscosity contributions of the heavy species (light species viscosity, a higher-order-term, is negligible). These equations, taken to first order, are expected to have the same validity for a disparate-mass gas as the Navier-Stokes equations have for more ordinary gases.

To obtain disparate-mass sound propagation predictions, we use the near-normal equations in the form given by GOEBEL, HARRIS, and JOHNSON, Eqs. (24)–(31) of Ref. [4]. (This implies a restriction to mixtures of Maxwell molecules, repelling each other with forces proportional to r^{-5}, where r is the intermolecular separation. This restriction must be improved upon, but is useful for obtaining initial results). In the presence of a small disturbance of the form $\exp i(kx - \omega t)$, these equations may be linearized and nondimensionalized to give

(1)
$$n_1 = z(U - W), \quad n_2 = zU,$$

(2)
$$U = z\left[\sum_i x_i n_i + T + P\right],$$

(3)
$$T = (2/3)\sum_i [x_i n_i + z q_i],$$

for the deviations from equilibrium of the hydrodynamic variables (species i number density, flow velocity, and temperature, respectively), and

(4)
$$W = (i\omega z)d[n_1 + T - x_2 \Delta],$$

(5)
$$P = -(i\omega z)mU,$$

(6)
$$q_1 = -(i\omega z)l_1[T - x_2 \Delta], \quad q_2 = -(i\omega z)l_2[T + x_1 \Delta],$$

(7)(*)
$$\Delta = -(i\omega z)\tau[W - (q_1/x_1)], \quad \tau = 2/3(1 - i\omega),$$

for the dissipative variables (diffusion velocity $W \equiv U_2 - U_1$, heavy species pressure deviator, species i heat flux, and temperature separation, respectively). Here x_i is the mole fraction of species i. Variables have been nondimensionalized with respect to equilibrium number density, equilibrium temperature, and reference speed $c \equiv (p_0/\varrho_0)^{1/2}$ (p_0 and ϱ_0 are equilibrium pressure and mass-density, respectively, and $c = (3/5)^{1/2}V_0$, where V_0 is the equilibrium speed of sound). The reference frequency is taken to be ω ; it has been shown [2–4] that

(8)
$$\omega_\Delta = 2c^2 x_2/D,$$

where D is the usual coefficient of diffusion [8]. The reduced wave number z is by definition

(9)
$$z \equiv kc/\omega.$$

The nondimensional coefficients of diffusion, viscosity, and thermal conductivity are, respectively,

(10)
$$d = D\omega_\Delta/c^2 x_2 = 2,$$

(*) See note on p. 710.

(11)
$$m = 8\mu_2 x_2/3\varrho_0 D,$$

(12)
$$l_i = \lambda_i T_0 \omega_{,1}/p_0 c^2,$$

where μ_2 and λ_i are coefficients of viscosity and thermal conductivity given by Eqs. (27.a) and (27.b) of Ref. [4].

The predictions of these equations for forced sound propagation is found from the dispersion relation, which gives the (complex) wave number k as a function of the frequency ω; the latter is constrained to be real and positive. The dispersion relation itself is obtained by setting the determinant of the coefficients of Eqs. (1)–(7) equal to zero (a numerical solution is necessary). Usually, one root of this equation corresponds to sound propagation in the $+x$ direction. The sound-propagation root is identified by its low-frequency behaviour: the absorption coefficient α has the behaviour

(13)
$$\alpha \equiv (5/3)^{\frac{1}{2}} \operatorname{Im}(z) \to 0^+,$$

$$\omega \to 0$$

while the dispersion V_0/V goes as

(14)
$$(V_0/V) = (5/3)^{\frac{1}{2}} \operatorname{Re}(z) \to 1,$$

$$\omega \to 0$$
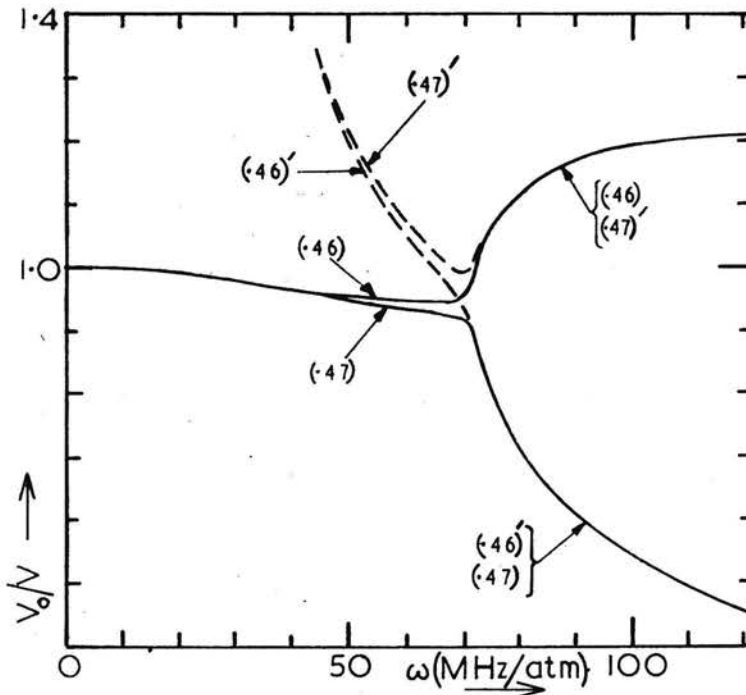

FIG. 1. Dispersion in Xe–He for helium mole-fractions $0.46 < x_c$ and $0.47 > x_c$. Solid curve: sound root; dashed curve: interfering root. For $\omega \gtrsim 75$ MHz/atm, the dispersion of the sound mode for $x_1 = 0.46$ (0.47) overlaps that of the interfering mode with $x_1 = 0.47$ (0.46), to within graphical accuracy

where z is the reduced wave number of Eq. (9), and V is the speed of propagation at frequency ω.

To facilitate comparison with experiment, we have considered mixtures of xenon with helium, for which $m_1/m_2 = 4/131.3$. As specific experimental input we have used the pure-gas viscosities, and the Xe—He coefficient of diffusion, as given in Ref. [8], Chs. 12 and 14. The present predictions refer to mixtures at standard temperature and pressure.

Perhaps the most striking thing to emerge from the present study is the prediction of double sound propagation in disparate-mass gases. Specifically, there is predicted to be a range of compositions in the region of a critical He concentration x_c for which two propagating modes are predicted if the input frequency ω is higher than a critical frequency ω_c. The two modes are predicted to have comparable absorption, but very different sound speeds. The situation is illustrated in Fig. 1 which shows the dispersion predicted in two
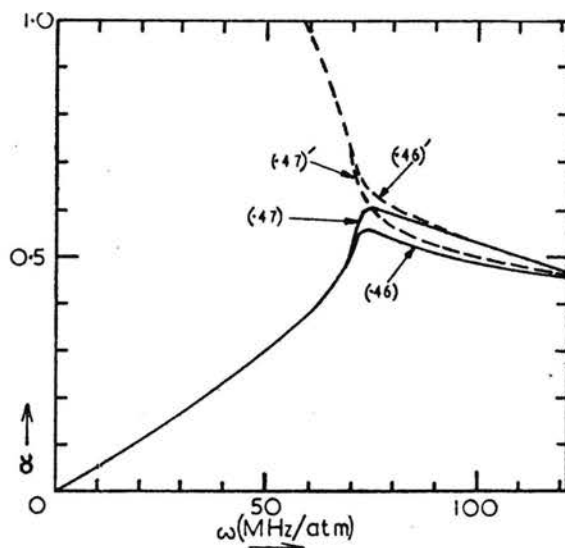

FIG. 2. Absorption curves associated with Fig. 1.

Xe—He mixtures of helium mole fraction $x_1 = 0.460 < x_c$ and $x_1 = 0.470 > x_c$. Figure 2 shows the associated absorption.

The existence of two propagating modes is made possible through the interference of two roots of the dispersion relation. For a mixture of composition $x_1 = x_c$, the sound root of the dispersion relation coincides with a second root of the dispersion relation (in both real and imaginary parts) at the critical frequency ω_c. For lower frequencies, this second root describes a highly-damped dissipative mode in the mixture; for frequencies higher than ω_c, however, there are two different but comparably-damped modes, both of which may properly be called sound modes. For compositions *near* the critical composition, the sound root changes its behaviour *qualitatively* as x_1 goes through the critical value, showing clear signs of the interference of the second root. Indeed, the sound propagation predictions for a range of compositions in the neighborhood of the critical com-

position cannot be understood without reference to the behavior of this second root. These facts make it physically reasonable to call the general effect, for x_1 in the neighborhood of x_c and $\omega \gtrsim \omega_c$, *double sound propagation.*

Figure 3 illustrates the way in which the two relevant roots of the dispersion relation interact with each other in the vicinity of the critical values $0.46 < x_c < 0.465$ and $0.64 <$
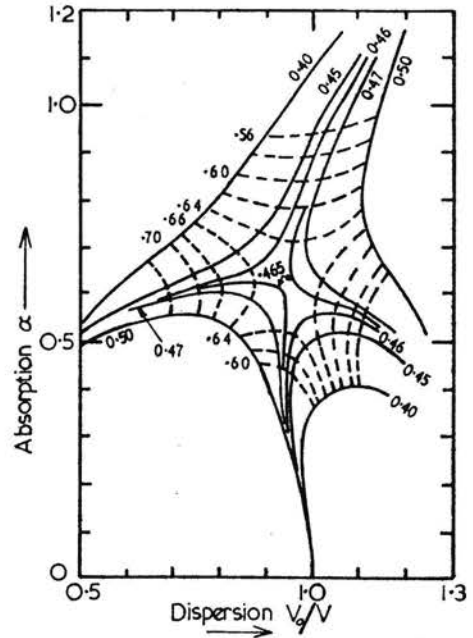

FIG. 3. Argand diagram for $(5/3)^{\frac{1}{2}} z$ as a function of x_1 (solid curves) and (ω/ω_A) (dashed curves) in the vicinity of x_c, ω_c.
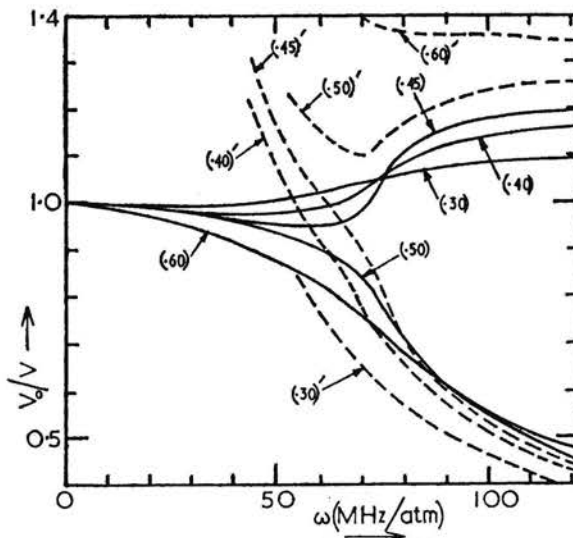

FIG. 4. Dispersion in Xe–He for various compositions x_1. Solid curve: sound root; dashed curve: interfering root.

$< (\omega_c/\omega_\Delta) < 0.66$ (for Xe—He, $\omega_\Delta = 110$ MHz/atm). The figure shows contour lines of the function $(5/3)^{\frac{1}{2}} z(x_1, \omega)$. Solid lines are lines of constant x_1, and dashed lines are lines of constant ω.

Figures 4 and 5 show the predicted dispersion and absorption curves, respectively, in Xe—He mixtures, for a range of compositions. For He fractions less than critical,
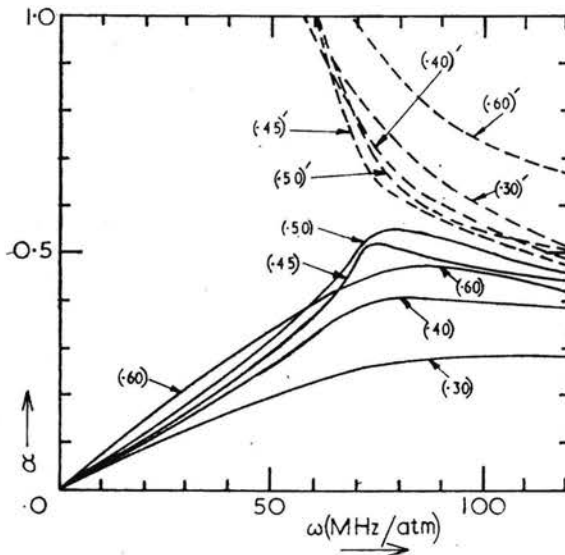

Fig. 5. Absorption curves associated with Fig. 4.

the sound mode is seen to tend to the slow wave ($V < V_0$) as ω increases above ω_c, while for He fractions greater than critical, the sound mode tends toward the fast wave ($V > V_0$) as ω increases above ω_c.

The effects predicted here are expected to be even more striking for mixtures in which the mass-ratio (m_1/m_2) is smaller than its value for Xe—He. In mixtures of He or H_2 with UF_6, for instance, to the extent that internal excitations could be neglected, double sound propagation is predicted to occur at lower frequencies, and with smaller damping. To see this in a qualitative way, we may assume we are dealing with mixtures of Maxwell molecules for which the intermolecular force constants K_{ij} scale with mass in the manner discussed by Johnson [5]:

(15) $$K_{ij} \sim (m_i m_j)^{\frac{1}{4}}.$$

This mass-scaling of forces is roughly true in practice, as pointed out, e.g. by Fox and Eaton [9]. We may thus generalize the present predictions to mixtures of arbitrary (but small) mass ratio m_1/m_2, at least qualitatively, by using xenon-helium input data, as before, together with the mass-scaling of Eq. (15). The frequency ω_Δ (and therefore also the critical frequency ω_c) is found to be lower than ω_{Δ} (Xe—He), since

$$\omega_\Delta = \left[\frac{m_1}{m_2} \frac{m_{Xe}}{m_{He}} \right]^{\frac{3}{4}} \omega_\Delta(\text{Xe—He}),$$

if $(m_1/m_2) < (m_{He}/m_{Xe})$. A second effect concerns the importance of first-order corrections, due to the thermal conductivity l_2 and viscosity m of the heavy species. These transport coefficients are smaller than their values in Xe—He by the factor

$$\left[\frac{m_1}{m_2}\frac{m_{Xe}}{m_{He}}\right]^{\frac{1}{4}},$$

so that first-order corrections to the zero-order predictions become progressively less important as (m_1/m_2) decreases. Calculations show that the presence of first-order corrections contributes greatly to the absorption of the slow wave (in the double sound regime), while scarcely affecting that of the fast wave. Thus, for $(m_1/m_2) < (m_{He}/m_{Xe})$, the damping of the slow wave is smaller than its damping in Xe—He, at a given value of x_1 and of (ω/ω). Since the damping of the slow wave increases with increasing x_1, we see that the slow wave should be more readily observable, up to higher values of x_1, in more highly disparate-mass mixtures.

In conclusion, we emphasize that the present predictions could be tested by experiment at the present time. The frequencies of interest are well within present experimental capability, as well as being within the range of validity of the basic equations. A two-temperature calculation using more realistic intermolecular potentials might be expected to give slightly different predictions for x_c and ω_c from those reported here. (In this connection, we mention that calculations using the usual equations of hydrodynamics, with the simplified transport coefficients appropriate to disparate-mass mixtures [4], also predict double sound propagation in Xe—He, with a similar critical frequency $\omega_c \approx 84$ MHz/atm, but at a much lower critical composition $0.225 < x_c < 0.250$).

It is now of interest to explain the existence of two propagating sound modes in disparate-mass gases in simple physical terms. In this contex, it is of interest to note that double sound propagation at moderate frequencies was predicted by LIBOFF [10], on the basis of simple model equation considerations, for gas mixtures characterized by a set of intrinsic frequencies of disparate magnitudes.

Acknowledgements

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Note added in proof:

In order to include first-order corrections, Eq. (7) should read

$$\Delta = -(i\omega z)\tau[W + (q_2/x_2) - (q_1/x_1)].$$

This change has negligible effects except for an increase of $\sim 11\%$ in the slow-wave absorption shown in Figs. 2 and 5, for frequencies $\omega \gtrsim 80$ MHz/atm.

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