

# Forced Modes of Dilute Binary Gas Mixtures<sup>1</sup>

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Anomalous forced sound dispersion in dilute binary gas mixtures is studied as a function of the mass ratio of the two components, using one- and two-temperature theories as well as different interparticle potentials. For a disparate-mass mixture, such as He-Xe, the results are compared with previous work due to Johnson et al. It is suggested that even for nondisparate-mass mixtures, a one-temperature treatment is not appropriate.

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**KEY WORDS:** anomalous sound dispersion; dilute binary gas mixtures; disparate-mass mixtures; forced modes; He-Xe mixtures; two-temperature effects.

## 1. INTRODUCTION

The forced modes of a fluid determine its response to external disturbances. When the frequency,  $\omega$ , of the disturbance is low, the modes obtained from the usual hydrodynamic equations are sufficient. At higher frequencies, however, the modes must be obtained from kinetic equations. For dilute binary gas mixtures, these equations are two coupled Boltzmann equations (see Burgers [1], for instance). Of particular interest are disparate-mass binary mixtures, that is, mixtures with very different molecular masses. In such mixtures, because of the slow exchange of kinetic energy between the two components, there exists a frequency regime (just beyond the hydrodynamic region) which is characterized by each component having its own temperature [2]. An interesting example, viz., that of a He-Xe mixture, where two-temperature effects appear, has been discussed in great detail by Johnson and co-workers [3, 4].

In fact, Huck and Johnson [3] have predicted a peculiar behavior in the forced sound propagation in dilute disparate-mass binary gas mixtures. Thus, at frequencies higher than a critical frequency  $\omega_c$  and concentrations

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of the light component  $x_1$ , exceeding a critical concentration  $x_c$ , the speed of sound increases steeply with external frequency  $\omega$ , while for  $x_1 < x_c$  the speed slightly decreases with  $\omega$  (cf. Fig. 1).

Although this phenomenon also occurs in an ordinary hydrodynamic treatment of such mixtures, the critical frequency  $\omega_c$  is too high for hydrodynamics to be valid and a two-temperature extension of hydrodynamics is necessary. On the basis of such a theory, Huck and Johnson predicted a behavior of He-Xe mixtures that was subsequently confirmed by sound velocity measurements of Bowler and Johnson [4]. They related this behavior to the fact that the sound mode and another hydrodynamic mode become identical at  $\omega = \omega_c$  for  $x_1 = x_c$ .

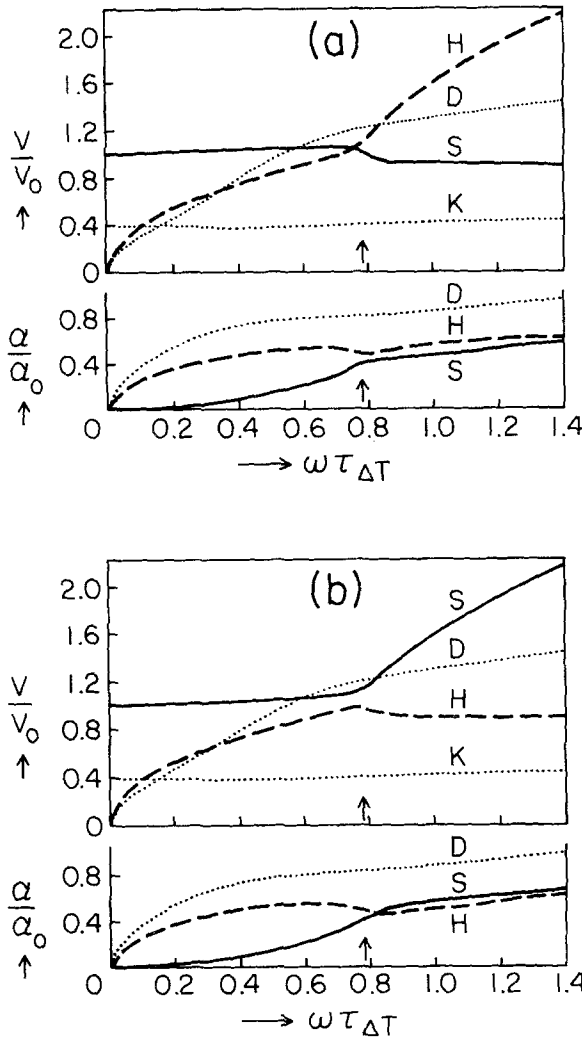
In this work, we investigate two-temperature effects on the predictions for  $x_c$  and  $\omega_c$ , not only for disparate-mass mixtures but also for mixtures with arbitrary mass ratios  $\mu = m_1/m_2$ , where  $m_1$  and  $m_2$  are the masses of light and heavy molecules, respectively. In Section 2, we briefly discuss the forced modes in the hydrodynamic regime and their extensions to higher frequencies. In Section 3, theoretical predictions for  $x_c$  and  $\omega_c$  for mixtures of He-Xe, based on one- and two-temperature theories are compared with experimental estimates. And in Section 4, we present our results for mixtures with arbitrary mass ratios.

## 2. FORCED MODES OF DILUTE BINARY MIXTURES

We consider the behavior of a dilute binary gas mixture in equilibrium due to a small external disturbance of frequency  $\omega$ . The disturbance propagates through the gas by exciting its forced modes. To obtain these modes, we assume that the disturbance is sufficiently weak that only linear deviations from equilibrium,  $\delta\mathbf{a} = \mathbf{a} - \mathbf{a}_0$ , need be taken into account and that the dependence on position  $\vec{r}$  and time  $t$  of the perturbed variables  $\delta\mathbf{a}(\vec{r}, t)$  that describe the fluid are of the form  $\delta\mathbf{a}(\vec{r}, t) = \delta\mathbf{a}(k, \omega) e^{i(\vec{k} \cdot \vec{r} - \omega t)}$ . Then by requiring that the linearized fluid equations have nontrivial solutions for the  $\delta\mathbf{a}(k, \omega)$ , we obtain a dispersion relation of the form  $F(k, \omega) = 0$ , where  $k = |\vec{k}|$  (cf. Foch and Ford [5]). For forced modes we solve the dispersion relation for  $k = k(\omega)$ , where  $\omega$  is real, from which we may calculate the phase velocity  $v = \omega/\text{Re } k$  and the spatial damping  $\alpha = \text{Im } k$ . In this work, we consider only the longitudinal (i.e., parallel to  $\vec{k}$ ) part of the dispersion relation, which contains the sound modes.

### 2.1. Forced Modes at Low Frequencies

When the frequency of the disturbance is low such that  $\omega\tau_k \ll 1$ , where  $\tau_k$  is the relaxation time of the slowest of the variables,  $\mathbf{a}(\vec{r}, t)$ , the



**Fig. 1.** Reduced propagation velocity  $v/v_0$  and reduced damping  $\alpha/\alpha_0$  of the forced modes of dilute mixtures of He-Xe at  $p = 1$  atm and  $T = 25^\circ\text{C}$ , as a function of the reduced frequency  $\omega\tau_{dT}$ . The modes are calculated from the two-temperature 13-moment equations for a hard-sphere interparticle potential; (a)  $x_{\text{He}} = 0.445 < x_c$ ; (b)  $x_{\text{He}} = 0.465 > x_c$ . The modes are indicated by (S) sound, (H) heat, (D) diffusion, and (K) kinetic.  $v_0$  is the velocity of sound at zero frequency and  $\alpha_0 = (v_0\tau_{dT})^{-1}$ . Not plotted is the kinetic mode damping, which runs from 1.6 at  $\omega\tau_{dT} = 0$  to 2 at  $\omega\tau_{dT} = 1.4$ . The arrows indicate the value of  $\omega_c\tau_{dT}$ .

hydrodynamic equations describe the behavior of the system. In that case, the variables,  $\mathbf{a}(\vec{r}, t)$ , are the local number densities, the local temperature, and the local longitudinal velocity, which are related to the first three velocity moments of the nonequilibrium distribution functions that describe the mixture.

The dispersion relation obtained from the linearized hydrodynamic equations [6] has the following form:

$$A_3 k^6 + A_2 k^4 + A_1 k^2 + A_0 = 0 \quad (1)$$

where the  $A_i$  are functions of the frequency as well as of the transport and thermodynamic properties. Equation (1) yields three types of modes: sound, heat, and diffusion. There are two of each type, propagating in opposite directions, so that a total of six forced modes is present. Here, we consider only one set of modes, say, those that propagate in the  $+\vec{k}$  direction. According to hydrodynamics, these forced modes have the following frequency dependence as  $\omega \rightarrow 0$ : sound mode  $v_S \sim \text{const.}$ ,  $\alpha_S \sim \omega^2$ ; heat mode  $v_H \sim \omega^{\frac{1}{2}}$ ,  $\alpha_H \sim \omega^{\frac{1}{2}}$ ; and diffusion mode  $v_D \sim \omega^{\frac{1}{2}}$ ,  $\alpha_D \sim \omega^{\frac{1}{2}}$  [4].

## 2.2. Forced Modes at Higher Frequencies

In order to describe the response of the fluid to higher-frequency disturbances, one must, in addition to the hydrodynamic variables, take into account explicitly more velocity moments of the distribution function. For the case of a dilute simple gas, as a first step beyond hydrodynamics, Grad [7] derived from the Boltzmann equation a set of 13 equations, whereby in addition to the usual hydrodynamic variables, also the stress tensor and the heat flux vector were included in the description of the fluid. Burgers [1] generalized the 13-moment method to the case of binary mixtures, where the usual assumption of a common local equilibrium of the two components was not made. In fact, Burgers allowed each component to have its own density, temperature, flow velocity, stress tensor, and heat flux vector. The linearized two-temperature 13-moment equations derived from Burgers' nonlinear equations are given in the Appendix.

The case of disparate-mass binary mixtures is especially interesting. Grad [2] already noted that in such mixtures the exchange of kinetic energy between light and heavy molecules is very slow. Thus, he conjectured that the approach to equilibrium will take place in three stages. First the light molecules reach local equilibrium with temperature  $T_1$ , in a time  $\tau_1$ ,<sup>3</sup> then the heavy component reaches local equilibrium with temperature

<sup>3</sup>In principle, in a disparate-mass mixture, a different local equilibrium for the two components means different values for their number densities, velocities and temperatures. While

$T_2$  in a time  $\tau_2$ , finally followed by the equilibration of the two different species temperatures in a time  $\tau_{dT}$ , after which the system behaves according to the usual one-temperature equations of hydrodynamics. The times  $\tau_1$  and  $\tau_2$  are the mean-free times associated with the light and heavy components, respectively, and  $\tau_{dT} \sim \tau_2/\sqrt{\mu} \sim \tau_1/\mu$  [8], with  $\mu = m_1/m_2$  the mass ratio. Therefore, Grad suggested that the first step beyond ordinary hydrodynamics for disparate-mass mixtures should be a two-temperature hydrodynamics.

Goebel et al. [8] have derived two-temperature hydrodynamic equations. Starting from the two-temperature 13-moment equations of Burgers, and expanding in powers of the mass ratio  $\mu$  (in fact  $\mu^{1/2}$ ), they found that in a disparate-mass mixture, of all the nonhydrodynamic moments, the temperature difference, indeed, had the longest relaxation time. Thus by retaining only the temperature-difference equation and approximating other moment equations by linear laws, they obtained two-temperature hydrodynamic equations.

The dispersion relation derived from both the two-temperature hydrodynamic equations and the two-temperature 13-moment equations have the following form:

$$B_4 \tilde{k}^8 + B_3 \tilde{k}^6 + B_2 \tilde{k}^4 + B_1 \tilde{k}^2 + B_0 = 0 \quad (2)$$

where the coefficients  $B_i$  differ in the two cases and  $\tilde{k}$  is a dimensionless wavenumber defined in Eq. (A11). Thus besides the six hydrodynamic modes discussed before, an additional pair of kinetic modes, propagating in opposite directions, appear. The coefficients  $B_i$  are essentially determined by the dimensionless frequency  $\tilde{\omega}$ , defined in Eq. (A11), the molar fraction  $x_1$  of the light component, the mass ratio  $\mu$ , and the diameter ratio  $\sigma = \sigma_1/\sigma_2$  (or force constant ratio), where  $\sigma_1$  and  $\sigma_2$  are the diameters of the light and heavy molecules, respectively.<sup>4</sup>

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their number densities are fixed, the local velocities can be shown to relax much faster (in fact by a factor  $\mu$ ) to a common value than their local temperatures [8], so that only two-temperature, and not two (local)-velocity, extension of hydrodynamics has to be considered.

<sup>4</sup> For hard spheres there are three relevant diameters:  $\sigma_1$ ,  $\sigma_2$ , and  $\sigma_{12}$ .  $\sigma_1$  and  $\sigma_2$  are usually derived from pure gas viscosities, while  $\sigma_{12}$  can be determined either from the mutual diffusion coefficient [9] or from the mixing rule  $\sigma_{12} = (\sigma_1 + \sigma_2)/2$ . The  $B_i$  or the  $x_c$  and  $\omega_c$  are not very sensitive to either choice of  $\sigma_{12}$ ; similarly for the force constants in the case of Maxwell molecules.

### 3. RESULTS FOR He-Xe MIXTURES

In their experiments on He-Xe mixtures, Bowler and Johnson [4] observed the sudden change in the sound propagation velocity near a particular concentration by varying the molar fraction of helium. They estimated the critical molar fraction  $x_c \approx 0.5$  and the critical frequency  $\omega_c \approx 4.4 \times 10^8 \text{ s}^{-1}$ , at a pressure  $p = 1 \text{ atm}$  and a temperature  $T = 25^\circ\text{C}$ .

In Table I, we report, for comparison, theoretical predictions for  $x_c$  and  $\omega_c$  calculated with various methods, for hard-sphere and Maxwell molecule interactions. The experimental information used, in addition to the molecular mass ratio  $\mu = 4/131.3 \approx 0.03$ , is the pure gas viscosities [10] and the mutual diffusion coefficient [11] to obtain the diameter ratios or force constant ratios [9].

The most striking feature is that the difference between one-temperature and two-temperature theories is much larger than that between hydrodynamics and the 13-moment method or between different interparticle potentials.

The predictions of one-temperature theories, i.e., the usual hydrodynamic equations ( $1T-H$ ) and the one-temperature 13-moment equations ( $1T-13$ ), which are the Burgers equations with a common temperature for both species,<sup>5</sup> are very close to each other and in clear disagreement with the experimental results. On the other hand, the predictions of two-temperature theories, i.e., the two-temperature hydrodynamic equations ( $2T-H$ ) and two-temperature 13-moment equations ( $2T-13$ ),

<sup>5</sup> The difference between the predictions of the 13-moment Burgers equations for a disparate-mass mixture with a common temperature and with a common temperature as well as a common velocity for both species is small: 6% for  $x_c$  and 2% for  $\omega_c$  for He-Xe mixtures.

Table I. Predictions for Mixtures of He-Xe at  $p = 1 \text{ atm}$ ,  $T = 25^\circ\text{C}$

	Maxwell		Hard sphere	
	$x_c$	$\omega_c (10^8 \text{ s}^{-1})$	$x_c$	$\omega_c (10^8 \text{ s}^{-1})$
$1T-H$	$0.253 \pm 0.001^a$	$5.5^a$	$0.310 \pm 0.001$	6.7
$1T-13$	$0.256 \pm 0.003$	5.4	$0.322 \pm 0.003$	5.8
$2T-H$	$0.457 \pm 0.002^a$	$4.8^a$	$0.435 \pm 0.002$	5.3
$2T-13$				
Johnson	$0.495 \pm 0.005$	4.7	$0.505 \pm 0.005$	4.3
This work	$0.482 \pm 0.002$	4.7	$0.455 \pm 0.002$	5.2

<sup>a</sup> In agreement with Johnson and co-workers.

are not only close to each other but close to the experimental values as well. This clearly indicates the existence of different species temperatures in a disparate-mass mixture.

We obtain somewhat different numerical results than those reported by Bowler and Johnson [4] for the two-temperature 13-moment method. We do not know the source of this difference. We should remark, however, that we have used the SMP (symbolic manipulation program) to obtain an analytic expression for the dispersion relation for an arbitrary mass ratio from the 13-moment equations, then later substituted specific values appropriate for He-Xe mixtures. The predictions of Bowler and Johnson are very close to their experimental estimates (and even overlapping for hard-sphere molecules), suggesting that the 13-moment approximation is sufficient to explain the experiment. However, our results seem to indicate that the inclusion of other higher moments, in particular, the flux of the heat flux—which has a relaxation time equal to that of the heat flux itself [12]—will still make significant contributions. This could be at the root of the difference for  $x_c$  and  $\omega_c$  of our results and experiment. We note that the difference in  $x_c$  and  $\omega_c$  due to the interparticle potential is comparable to the difference between two-temperature hydrodynamic and two-temperature 13-moment theories (similarly for the one-temperature theories). In view of this and the considerable experimental uncertainty in  $x_c$  and  $\omega_c$ , we believe that a complete quantitative agreement between theory and experiment has not yet been obtained.

In Fig. 1, we plot the propagation velocities and dampings of all the forced modes calculated from the two-temperature 13-moment equations for hard-sphere molecules as a function of the external frequency, for mixtures of He-Xe under the physical conditions mentioned above. The sudden change in the velocity of the sound mode at higher frequencies can be seen as the molar fraction of helium is increased from below  $x_c$  (Fig. 1a) to above  $x_c$  (Fig. 1b). The interfering mode which becomes identical to the sound mode for  $x = x_c$  at  $\omega = \omega_c$  is also indicated. In this case, it is the mode which has a heat mode-like behavior at low frequencies. As pointed out by Bowler and Johnson [4], the nature of the interfering mode is sensitive to the transport properties of the mixture and may be different in different theories. For example, in two-temperature hydrodynamics it would be the diffusion mode that interferes.

#### 4. BINARY MIXTURES WITH ARBITRARY MASS RATIOS

The same effect that is described in Fig. 1 is also present in non-disparate-mass binary mixtures, although less pronounced. As a means of

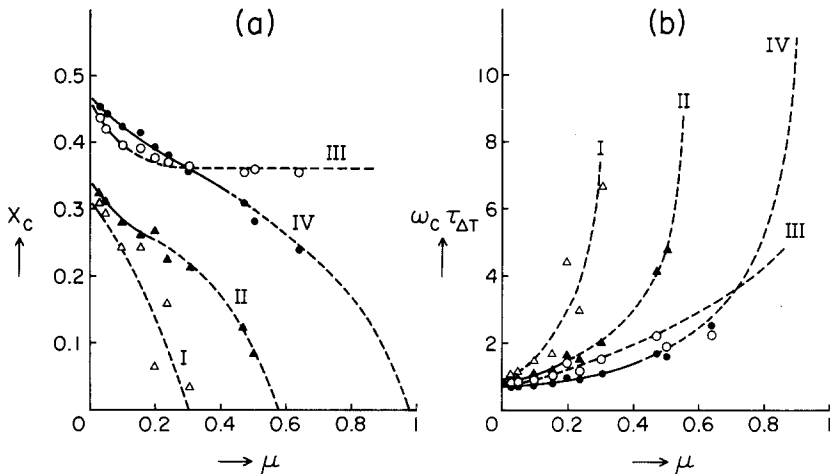


Fig. 2. (a)  $x_c$  and (b)  $\omega_c \tau_{DT}$  as a function of the mass ratio  $\mu$  from various theories for hard-sphere molecules. The curves are calculated assuming that  $\mu = \sigma^3$ ; the points refer to noble gas mixtures using experimental values for  $\sigma$ . The results from various theories are shown as one-temperature hydrodynamics (I,  $\Delta$ ), one-temperature 13-moment (II,  $\blacktriangle$ ), two-temperature hydrodynamics (III,  $\circ$ ), and two-temperature 13-moment (IV,  $\bullet$ ).

investigating the importance of two-temperature effects in more normal (nondisparate-mass) mixtures, we calculated  $x_c$  and  $\omega_c$  in mixtures with arbitrary mass ratios from one- and two-temperature theories. The results are shown in Fig. 2, where  $x_c$  and  $\omega_c \tau_{DT}$ <sup>6</sup> are plotted as a function of the mass ratio  $\mu$ , assuming hard-sphere interactions. The points correspond to mixtures of noble gases with diameter ratios deduced from experiment [13], and the curves are calculated with the assumption that the mass ratio  $\mu$  and the diameter ratio  $\sigma$  are related by  $\mu = \sigma^3$ . The various curves have been drawn up until values of  $\mu$  for which an  $x_c$  and an  $\omega_c$  exist. The rather good agreement between noble gas mixtures and the curves may indicate that  $x_c$  and  $\omega_c$  are not too sensitive to the diameter ratio. We should remark that the two-temperature hydrodynamic equations are valid only when  $\mu \ll 1$ , while the two-temperature 13-moment method is valid for frequencies for which  $\omega \tau_{DT}$  is not much in excess of unity. The one-temperature curves are valid as long as  $\omega \tau_i < 1$ , where  $\tau_i$  is the longest of the two mean free times  $\tau_1$  and  $\tau_2$ . Hence, we have dotted the portions of the curves that cannot be trusted.<sup>7</sup>

<sup>6</sup> Although  $\tau_{DT}$  has no special physical significance in one-temperature theories, it can, nevertheless, be calculated from the mutual diffusion coefficient as given in the Appendix.

<sup>7</sup> To extend these curves further, i.e., to obtain reliable results for higher values of  $\omega$ , one needs to include more moments than we have considered here.



One can deduce the following, however.

(i) By comparing the one-temperature hydrodynamic with the one-temperature 13-moment curves, we see the increasing importance of higher velocity moments as the mass ratio approaches unity, while comparison of the one-temperature and the two-temperature 13-moment curves shows the importance of the two-temperature contribution.

(ii) The somewhat surprising result is that two-temperature effects continue to remain important even in nondisparate-mass mixtures. The two-temperature effect, however, is then not the dominant effect for, say, a mass ratio  $\mu = 0.2$ , since its contribution is comparable to that of other moments. Hence these other moments have also to be taken into account to give a satisfactory theory.

### APPENDIX

The nonlinear two-temperature 13-moment equations can be found in Section 2 of Burgers' book [1]. Here we give the linearized version of these equations, and only those that contribute to the longitudinal part of the dispersion relation. The original 26 equations reduce, then, to 10.

Burgers' equations are given in terms of the species number density  $n_i$ , temperature  $T_i$ , flow velocity  $\vec{u}_i$ , stress tensor  $\vec{P}_i$ , and heat flux vector  $\vec{q}_i$  of species  $i = 1, 2$ . Instead of  $n_i$ ,  $T_i$ , and  $\vec{u}_i$  we use the following variables: total number density  $n = n_1 + n_2$ , density difference  $\Delta n = n_1/x_1 - n_2/x_2$ , average temperature  $T = x_1 T_1 + x_2 T_2$ , temperature difference  $\Delta T = T_1 - T_2$ , average flow velocity  $\vec{u} = (x_1 m_1 \vec{u}_1 + x_2 m_2 \vec{u}_2)/m$ , and velocity difference  $\vec{w} = \vec{u}_1 - \vec{u}_2$ , where  $x_i$  is the molar fraction of species  $i$  and  $m = x_1 m_1 + x_2 m_2$ . We also define  $m_0 = m_1 + m_2$ .

We give the equations in a nondimensional form, where the densities are reduced by  $n_0$ , temperatures by  $T_0$ , velocities by  $(k_B T_0/m)^{1/2}$ , stress tensors by  $p_0$ , and heat fluxes by  $(k_B T_0/m)^{1/2} p_0$ , where  $n_0$ ,  $T_0$ , and  $p_0$  are the equilibrium density, temperature, and pressure, respectively, and  $k_B$  is the Boltzmann constant. The nondimensional form of a quantity  $y$  is denoted by  $\tilde{y}$ , and its deviation from equilibrium by  $\delta\tilde{y}$ . Thus the linearized two-temperature 13-moment equations (the longitudinal part) are

$$-i\tilde{\omega} \delta\tilde{n} + i\tilde{k} \delta\tilde{u} = 0 \tag{A1}$$

$$-i\tilde{\omega} \delta\Delta\tilde{n} + i\tilde{k} \delta\tilde{w} = 0 \tag{A2}$$

$$-i\tilde{\omega} \delta\tilde{T} + \frac{2}{3} i\tilde{k} \delta\tilde{u} + \frac{2}{3} x_1 x_2 \frac{(m_2 - m_1)}{m} i\tilde{k} \delta\tilde{w} + \frac{2}{3} i\tilde{k} \delta\tilde{q}_1 + \frac{2}{3} i\tilde{k} \delta\tilde{q}_2 = 0 \tag{A3}$$

$$-i\tilde{\omega} \delta\tilde{u} + i\tilde{k} \delta\tilde{n} + x_1 x_2 \frac{(m_2 - m_1)}{m} i\tilde{k} \delta\Delta\tilde{n} + i\tilde{k} \delta\tilde{T} + i\tilde{k} \delta\tilde{P}_1 + i\tilde{k} \delta\tilde{P}_2 = 0 \quad (\text{A4})$$

$$(-i\tilde{\omega} + 1) \delta\Delta\tilde{T} + \frac{2}{3} i\tilde{k} \delta\tilde{w} + \frac{2}{3x_1} i\tilde{k} \delta\tilde{q}_1 - \frac{2}{3x_2} i\tilde{k} \delta\tilde{q}_2 = 0 \quad (\text{A5})$$

$$\begin{aligned} & \left(-i\tilde{\omega} + \frac{mm_0}{2m_1 m_2}\right) \delta\tilde{w} + \left(\frac{m}{m_1} - \frac{m}{m_2}\right) i\tilde{k} \delta\tilde{n} + \left(\frac{x_2 m_2}{m_1} + \frac{x_1 m_1}{m_2}\right) i\tilde{k} \delta\Delta\tilde{n} \\ & + \left(\frac{m}{m_1} - \frac{m}{m_2}\right) i\tilde{k} \delta\tilde{T} + \frac{m^2}{m_1 m_2} i\tilde{k} \delta\Delta\tilde{T} + \frac{m}{x_1 m_1} i\tilde{k} \delta\tilde{P}_1 \\ & - \frac{m}{x_2 m_2} i\tilde{k} \delta\tilde{P}_2 + \frac{C}{2} \frac{m}{x_1 m_1} \delta\tilde{q}_1 - \frac{C}{2} \frac{m}{x_2 m_2} \delta\tilde{q}_2 = 0 \end{aligned} \quad (\text{A6})$$

$$\begin{aligned} & (-i\tilde{\omega} + \tilde{\omega}_{P_1}) \delta\tilde{P}_1 + \frac{4}{3} x_1 i\tilde{k} \delta\tilde{u} + \frac{4}{3} x_1 x_2 \frac{m_2}{m} i\tilde{k} \delta\tilde{w} + \frac{8}{15} i\tilde{k} \delta\tilde{q}_1 \\ & - \frac{x_1}{2} (4 - 3A) \delta\tilde{P}_2 = 0 \end{aligned} \quad (\text{A7})$$

$$\begin{aligned} & (-i\tilde{\omega} + \tilde{\omega}_{P_2}) \delta\tilde{P}_2 + \frac{4}{3} x_2 i\tilde{k} \delta\tilde{u} - \frac{4}{3} x_1 x_2 \frac{m_1}{m} i\tilde{k} \delta\tilde{w} + \frac{8}{15} i\tilde{k} \delta\tilde{q}_2 \\ & - \frac{x_2}{2} (4 - 3A) \delta\tilde{P}_1 = 0 \end{aligned} \quad (\text{A8})$$

$$\begin{aligned} & (-i\tilde{\omega} + \tilde{\omega}_{q_1}) \delta\tilde{q}_1 + \frac{5}{2} x_1 \frac{m}{m_1} i\tilde{k} \delta\tilde{T} + \frac{5}{2} x_1 x_2 \frac{m}{m_1} i\tilde{k} \delta\Delta\tilde{T} + \frac{m}{m_1} i\tilde{k} \delta\tilde{P}_1 \\ & + \frac{5}{4} C x_1 x_2 \frac{m_2}{m_1} \delta\tilde{w} - \frac{x_1 m_2}{4 m_0} (11 - 4B - 8A) \delta\tilde{q}_2 = 0 \end{aligned} \quad (\text{A9})$$

$$\begin{aligned} & (-i\tilde{\omega} + \tilde{\omega}_{q_2}) \delta\tilde{q}_2 + \frac{5}{2} x_2 \frac{m}{m_2} i\tilde{k} \delta\tilde{T} - \frac{5}{2} x_1 x_2 \frac{m}{m_2} i\tilde{k} \delta\Delta\tilde{T} + \frac{m}{m_2} i\tilde{k} \delta\tilde{P}_2 \\ & - \frac{5}{4} C x_1 x_2 \frac{m_1}{m_2} \delta\tilde{w} - \frac{x_2 m_1}{4 m_0} (11 - 4B - 8A) \delta\tilde{q}_1 = 0 \end{aligned} \quad (\text{A10})$$

where  $\delta\tilde{u}$ ,  $\delta\tilde{w}$ ,  $\delta\tilde{q}_1$ , and  $\delta\tilde{q}_2$  stand for the  $\vec{k}$  component of the velocities and heat fluxes, and  $\delta\tilde{P}_1$  and  $\delta\tilde{P}_2$  for the  $\vec{k}\vec{k}$  component of the stress tensors.

Here the reduced frequency  $\tilde{\omega}$  and wave number  $\tilde{k}$  are

$$\tilde{\omega} = \omega \tau_{AT}, \quad \tilde{k} = \left(\frac{k_B T_0}{m}\right)^{\frac{1}{2}} \tau_{AT} k \quad (\text{A11})$$

where

$$\tau_{AT} = \frac{m_0 \bar{n}_0 D_{12}}{2p_0} \quad (\text{A12})$$

is the relaxation time of the temperature difference, with  $D_{12}$  the coefficient of mutual diffusion in first Enskog approximation [9]. Furthermore,

$$\tilde{\omega}_{p_1} = x_1 \frac{m_0 n_0 D_{12}}{2\eta_1} + x_2 \left( 1 + \frac{3m_2}{2m_1} A \right) \quad (\text{A13})$$

$$\tilde{\omega}_{q_1} = x_1 \frac{m_0 n_0 D_{12}}{3\eta_1} + x_2 \frac{m_1}{m_0} \left[ \frac{3}{2} + 2A \frac{m_2}{m_1} + \left( \frac{5}{4} - B \right) \frac{m_2^2}{m_1^2} \right] \quad (\text{A14})$$

and similar expressions obtain for  $\tilde{\omega}_{p_2}$  and  $\tilde{\omega}_{q_2}$  with indices 1 and 2 interchanged. Here  $\eta_i$  is the viscosity of species  $i$  in first Enskog approximation [9], and the coefficients  $A$ ,  $B$ , and  $C$  are ratios of collision integrals defined on p. 163 of Ref. 9.

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