Two-fluid Chapman-Enskog theory for binary gas mixtures

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As a result of the increasing inefficiency in the transfer of energy in collisions between species with a decreasing ratio of molecular masses, the Knudsen number range of validity of the Chapman-Enskog (CE) theory for binary gas mixtures decreases linearly with the molecular mass ratio. To remedy the situation, a two-fluid CE theory uniformly valid in the molecular mass ratio is constructed here. The analysis extends previous two-fluid theories to allow for arbitrary potentials of intermolecular interaction and arbitrary mass ratios. The treatment differs from the CE formulation in that the mean velocities and temperatures of the two gases are not required to be identical to lowest order. To first order, the streaming terms of the Boltzmann equation are thus computed in terms of the derivatives of the two-fluid hydrodynamic quantities, rather than the mean mixture properties as in the CE theory. As a result, associated with the nonconservation of momentum and energy for each species alone, two new "driving forces" appear in the first-order integral equations. The amount of momentum and energy transferred per unit time between the species appear in the theory as free constants, which allow satisfying the constraint that all hydrodynamic information be contained within the lowest-order two-fluid Maxwellians, Simultaneously, this constraint fixes the rate of momentum and energy interchange in terms of the two-fluid hydrodynamic quantities and their gradients. The driving force d_{12} of the CE theory is directly related to the rate of interspecies momentum transfer, and the corresponding CE functions D_1 and D_2 appear here unmodified. But the physical interpretation of d_{12} is very different in the two pictures. On the CE side there is only one momentum equation, while d_{12} provides constitutive information fixing the diffusion flux (velocity differences) in the mass conservation equation. Here, the similar constitutive information associated to d_{12} is used to couple two different momentum equations. Although the CE theory captures some of the two-velocity aspects of the problem, no CE analog exists with the functions E_1 and E_2 associated here with temperature differences, which now require solving new integral equations. Finally, the presence of two velocities and two temperatures leads to four coefficients of viscosity and of thermal conductivity for the two stress tensors and heat flux vectors. Also, two thermal diffusion factors enter now into the expression for d_{12} . Although all these new coefficients arise as portions of the overall CE transport coefficients, their independent optimal determination requires new developments. The corresponding variational formulation is presented here and used to first order to obtain explicit expressions for all two-fluid transport coefficients by means of Sonine polynomials as trial functions.

I. INTRODUCTION

Transport phenomena in pure gases or mixtures are described in the Chapman-Enskog (CE) theory by an asymptotic solution of the Boltzmann equations in terms of a Knudsen number parameter Kn formed as the ratio between microscopic and macroscopic characteristic times or lengths, and measuring the departure from thermodynamic equilibrium. It has been pointed out by Grad, Hamel, 2 Goldman and Sirovich, Goebel et al., Tiem, and others that, in the case of binary mixtures of gases with very different masses, the range of validity of the CE theory is reduced considerably as a result of the appearance of a slow internal relaxation scale associated with the small interspecies energy transfer occurring in heavy-light collisions. However, the fact that each species still equilibrates efficiently by selfcollisions permits the development of a broader perturbation theory in which, to lowest order, each gas has a Maxwellian velocity distribution function with its own temperature T_i

and mean velocity U_i. Based on this scheme, a two-fluid theory has been developed in all rigor by Goldman and Sirovich³ for the special case of Maxwell molecules. In a recent paper, 6 we have extended this work to arbitrary interaction potentials, in the limit where the ratio of molecular masses $M = m_1/m_2$ between the two species is a small parameter. Surprisingly, although our treatment has a broader range of validity than the CE theory for the limit $M \leq 1$, one discovers that many (if not all) of the new transport coefficients arising in the two-fluid theory coincide with the $M \rightarrow 0$ limit of seemingly physically irrelevant integrals appearing in the computational apparatus of the CE approach. Given the complexity of the theory, such an isomorphism can hardly be taken as fortuitous: it points strongly to the possibility of extending the CE theory, converting it into a two-fluid theory uniformly valid for binary mixtures of arbitrary mass ratios. The generalization will be achieved by allowing the lowest-order solutions to the CE problem to be "unmatched

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Maxwellians" centered at different velocities and temperatures for the two gases.

In what follows, our objective is to present a two-fluid CE theory valid for arbitrary interaction potentials and uniformly valid for arbitrary mass ratios (in the sense that the Knudsen number range of validity of the theory is independent of M). It will be seen that a slight change of scope within the same structure of the CE building permits the desired generalization with practically no fundamental modifications in the resulting mathematical problem. Simultaneously, many aspects of the CE theory now acquire a newer and richer physical significance.

II. TWO-FLUID CHAPMAN-ENSKOG THEORY

Let us define the velocity distribution functions $f_1(\mathbf{u})$ and $f_2(\mathbf{u})$ for the two components in a binary mixture of gases and assume that they are slightly perturbed from two Maxwellian distributions f'_{0i} , each with its own number density n_i , temperature T_i , and mean velocity \mathbf{U}_i , which coincide exactly with the first three moments of the functions f_i :

$$f_i(\mathbf{u}) \equiv f'_{0i}(1 + \Phi'_i) \quad (i = 1, 2),$$
 (1)

$$f'_{0i} = n_i (2\pi kT_i/m_i)^{-3/2}$$

$$\times \exp[-m_i|\mathbf{u} - \mathbf{U}_i|^2/(2kT_i)]$$
 (*i* = 1,2), (2)

where k is Boltzmann's constant and m_i is the molecular mass of species i. Because the functions f'_{0i} contain all the two-fluid hydrodynamic information relevant to species i, the Φ'_i do not contribute at all to either n_i , U_i , or T_i [see Eqs. (41) below for a precise statement of these conditions].

The need to follow a perturbation scheme based to lowest order on two unmatched Maxwellian distributions becomes evident from the following considerations.

- (1) The CE theory for gas mixtures is an asymptotic expansion in the Knudsen number that is not uniformly convergent with respect to the ratio M of molecular weights, in the sense that the relative ordering of the terms in the expansion depends on the mass ratio M, with O(Kn/M) contributions appearing to first order. Accordingly, the region of validity of the CE theory is severely limited for small values of M by the condition $Kn \ll M$.
- (2) Following Grad,¹ it has been pointed out that the mechanism of self-equilibration of each species leads to their relaxation toward Maxwellian distributions within times of the order of the interval between self-collisions. Accordingly, $\Phi'_i = O(Kn)$ uniformly, independently of the value of M, 6 and the two-fluid theory resulting from determining Φ'_i in Eq. (1) will be constrained by the condition that $Kn \ll 1$ rather than the stronger condition $Kn \ll M$ to which the results from the CE theory are subject.

We shall determine the corrections Φ'_i only to first order in a Knudsen number expansion, so that the collision operators may be substituted by their linearized forms. A difficulty of having introduced distributions f'_{0i} with unmatched velocities and temperatures is that one would be obliged to rederive the standard properties of the linearized Boltzmann operators for the case of these peculiar Maxwellians. Such an approach, however, would not use optimally the available computational machinery of the CE theory. On the other

hand, if the differences in species mean velocities and temperatures are small compared with the thermal speed of the light gas and the mean mixture temperature T, respectively, one may still follow the road of linearizing the Boltzmann operators around "matched Maxwellian distributions" f_{0i} with a common temperature T and mean velocity \mathbf{U} , which we leave unspecified for the moment:

$$f_{0i} = n_i (2\pi kT/m_i)^{-3/2} \exp[-m_i (\mathbf{u} - \mathbf{U})^2/(2kT)]$$
(i = 1,2), (3)

in terms of which.

$$f_i = f_{0i}(1 + \Phi_i). \tag{4}$$

Then, the linearized Boltzmann equations may be written schematically as

$$Df_1 = K_{11}(\Phi_1) + K_{12}(\Phi_1 + \Phi_2), \tag{5}$$

$$Df_2 = K_{21}(\Phi_1 + \Phi_2) + K_{22}(\Phi_2), \tag{6}$$

where D denotes the streaming operator, while the K_{ij} are the usual linearized Boltzmann collision operators characterizing the rate of change of f_i by collisions with molecules of species j. In a linear theory consistent with the linearization of the Boltzmann collision integrals, our primed and unprimed functions differ by small quantities proportional to the velocity and temperature slips

$$f'_{0i} = f_{0i}(1 + \Delta_i), \tag{7}$$

$$\Phi_i' = \Phi_i - \Delta_i, \tag{8}$$

with

$$\Delta_i \equiv (-1 + T_i/T) \left[-\frac{3}{2} + m_i (\mathbf{u} - \mathbf{U})^2 / (2kT) \right] + m_i (\mathbf{U}_i - \mathbf{U}) \cdot (\mathbf{u} - \mathbf{U}) / kT.$$
(9)

Clearly, if Δ_i is small, Φ_i will be small whenever Φ_i' is small, and the choice of linearizing the collision operators around the matched functions f_{0i} is perfectly justified. As things have been introduced, although the functions Φ_i' are tied by the constraint that $f'_{0i}\Phi_i'$ does not contribute to any of the hydrodynamic quantities n_i , \mathbf{U}_i , T_i , the functions $f_{0i}\Phi_i$ do actually contribute to both \mathbf{U}_i and T_i [Eqs. (42) and (43) below].

The perturbation procedure we shall use to solve Eqs. (5) and (6) is based on the smallness of the Φ'_i functions with respect to unity. Whether there are one or two temperatures and velocities, the distribution function of each species approaches a Maxwellian, with errors of the order of its own Knudsen number. Accordingly, to first order, the terms Df_i in Eqs. (5) and (6) may be approximated by Df'_{0i} :

$$Df'_{01} = K_{11}(\Phi_1) + K_{12}(\Phi_1 + \Phi_2), \tag{10a}$$

$$Df'_{02} = K_{21}(\Phi_1 + \Phi_2) + K_{22}(\Phi_2),$$
 (10b)

whose left-hand side can be written in terms of the derivatives of the two-fluid hydrodynamic quantities n_i , U_i , and T_i . Because we are looking for normal solutions of the Boltzmann equation (that is solutions whose only time dependence come through their moments), time derivatives may be eliminated, as in the CE theory, by means of the hydrodynamic equations (in fact, in the notation of Ref. 7, we will eliminate $\partial_0/\partial t$, see below). But now, such an operation brings a key physical difference with the CE theory, because

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the present conservation equations are written independently for each one of the two gases, thus containing extra terms caused by the interspecies transfer of energy and momentum. Defining the standard inner product

$$[g(\mathbf{u}), f(\mathbf{u})] \equiv \int f(\mathbf{u}) g(\mathbf{u}) d^3\mathbf{u},$$

and taking moments in the Boltzmann equations we obtain

$$\partial_i \rho_i + \nabla \cdot (\rho_i \mathbf{U}_i) = 0, \tag{11}$$

$$\partial_t(\rho_i \mathbf{U}_i) + \nabla \cdot (\rho_i \mathbf{U}_i \mathbf{U}_i + \mathbf{P}_i) = \lambda_i, \tag{12}$$

$$\partial_t \left[\rho_i (e_i + U_i^2/2) \right]$$

$$+ \nabla \cdot \left[\rho_i \mathbf{U}_i (\mathbf{e}_i + U_i^2/2) + \mathbf{P}_i \cdot \mathbf{U}_i + \mathbf{Q}_i \right] = \sigma_i, \quad (13)$$

where the quantities ρ_i , \mathbf{U}_i , \mathbf{P}_i , and \mathbf{Q}_i are the moments of f_i defined as

$$\rho_i \equiv n_i m_i \equiv [m_i, f_i], \qquad (14)$$

$$\rho_i \mathbf{U}_i \equiv [m_i \mathbf{u}, f_i] , \qquad (15)$$

$$\mathbf{P}_i \equiv [m_i(\mathbf{u} - \mathbf{U}_i)(\mathbf{u} - \mathbf{U}_i), f_i], \qquad (16)$$

$$\rho_i e_i \equiv \frac{1}{2} \mathbf{P}_i : \mathbf{I} \equiv 3p_i / 2 \equiv 3n_i k T_i / 2, \tag{17}$$

$$\mathbf{Q}_i \equiv \left[\frac{1}{2}m_i|\mathbf{u}_i - \mathbf{U}_i|^2(\mathbf{u} - \mathbf{U}_i), f_i\right]. \tag{18}$$

The new terms λ_i and σ_i are, respectively, the net rate of momentum and of energy transferred per unit time to species i by collisions with molecules of the other gas. They are given by the expressions

$$\lambda_i \equiv [m_i \mathbf{u}, K_{ii} (\Phi_1 + \Phi_2)], \quad i \neq j, \tag{19}$$

$$\sigma_i \equiv [m_i u^2 / 2, K_{ii} (\Phi_1 + \Phi_2)], \quad i \neq j,$$
 (20)

and satisfy

$$\lambda_1 + \lambda_2 = 0, \tag{21}$$

$$\sigma_1 + \sigma_2 = 0, \tag{22}$$

because of total momentum and energy conservation for the mixture. Evaluating Df'_{0i} and eliminating temporal derivatives by means of the hydrodynamic equations (11)–(13) but with $\mathbf{P}_i = p\mathbf{I}$ and $\mathbf{Q}_i = 0$ (that is, eliminating $\partial_0/\partial t$ in the notation of Ref. 7), we obtain

$$Df'_{0i} = \operatorname{Grad}_{i}' + p_{i}^{-1} f'_{0i} \mathbf{c}_{i} \cdot \lambda_{i} + p_{i}^{-1} f'_{0i} \left[-1 + m_{i} c_{i}^{2} / (3kT) \right] \sigma'_{i},$$
 (23)

with

$$\mathbf{c}_i \equiv \mathbf{u} - \mathbf{U}_i, \tag{24}$$

$$\sigma_i' \equiv \sigma_i - \lambda_i \cdot \mathbf{U}_i = \left[\frac{1}{2} m_i c_i^2, K_{ij} (\Phi_1 + \Phi_2) \right], \quad i \neq j, \quad (25)$$

and where the term $Grad_i$ is the same one appearing in the CE theory for a pure gas,

Grad'_i
$$\equiv f'_{0i} \left[2m_i \mathbf{c}_i \mathbf{c}_i : \nabla \circ \mathbf{U}_i + \mathbf{c}_i \cdot \nabla \ln T_i \right] \times (m_i c_i^2 - 5kT_i) \left[/(2kT_i), \right]$$
 (26)

in which $\nabla \circ \mathbf{U}_i$ denotes the traceless, symmetrized gradient tensor of \mathbf{U}_i . The mathematical problem is thus completely specified: one has to solve Eqs. (10) when their left-hand sides are given by Eq. (23), with the constraint that the Φ_i' functions do not contribute to n_i, \mathbf{U}_i , or T_i . In principle, the λ and σ' terms complicate the problem because λ_i and σ'_i are linear functionals of the solutions Φ_1 and Φ_2 that ought to be grouped on the right-hand side of Eqs. (10), thus breaking

their original symmetry. Alternatively, one may consider λ_i and σ_i' as given constants [subject to Eqs. (21) and (22)] and then treat Eqs. (19) and (25) as constraints on the solution, as one does with n_i , U_i , and T_i , which are also functionals of Φ_i . Remarkably enough, proceeding in this fashion, the two constraints are satisfied automatically for any value of λ_1 and σ_1' . Let us assume that λ_i and σ_i' have been chosen arbitrarily [subject to Eqs. (21) and (22) but free from the constraints (19) and (25)]. In that case we shall subsequently show that solutions Φ_1 and Φ_2 exist such that

$$K_{ii}\Phi_{i} + K_{ij}(\Phi_{1} + \Phi_{2})$$

$$= \operatorname{Grad}_{i}' + p_{i}^{-1}f_{0i}'\mathbf{c}_{i}\cdot\lambda_{i} + \sigma_{i}'p_{i}^{-1}f_{0i}'$$

$$\times \left[-1 + m_{i}c_{i}^{2}/(3kT) \right] \quad (i = 1, 2, \quad j \neq i). \quad (27)$$

Forming the inner product of this expression with $m_i \mathbf{c}_i$, the contribution of the term Grad' and that proportional to σ'_i are null, while $[K_{ii}\Phi_i,\mathbf{c}_i]$ is also null because the operator K_{ii} is symmetric (i.e., $[\Phi,K_{ii}\Psi]=[K_{ii}\Phi,\Psi]$) and $K_{ii}\mathbf{c}_i=0$. It thus results that

$$[m_i \mathbf{c}_i, K_{ij}(\Phi_1 + \Phi_2)] = \lambda_i [f'_{0i} \mathbf{c}_i, \mathbf{c}_i]/p_i = \lambda_i,$$

so that the constraint (19) is satisfied for any value of λ_i subject to Eq. (21). That the constraint (25) for σ'_i is satisfied automatically for any σ'_i [subject to Eqs. (21) and (22)] results from forming the inner product of Eq. (27) with $m_i c_i^2/2$, realizing that the inner product of c_i^2 with both Grad' and the λ_i terms vanish; also $K_{ij}c_i^2 = 0$, and furthermore, $[m_i c_i^2, p_i^{-1} f'_{0i} (-1 + m_i c_i^2/3kT)] = 1$. Equation (27) may thus be treated as if λ_i and σ'_i were arbitrary constants [except for Eqs. (21) and (22)]. The presence of these two free parameters is an essential feature of the present theory, as they will make it possible to satisfy the condition that $f'_{0i}\Phi'_i$ does not contribute to m_i , U_i , and T_i . Simultaneously, that condition fixes λ_1 and σ'_1 univocally, providing constitutive expressions for the momentum and energy transfer terms that close the two-fluid hydrodynamic equations (11)–(13).

Notice that the independent variable enters in the right-hand sides of Eq. (27) as \mathbf{c}_i or $\mathbf{c}_i/(2kT_i/m_i)$, rather than \mathbf{c} or $\mathbf{c}/(2kT/m_i)$, where

$$c \equiv u - U$$
,

as in the CE theory. However, complete analogy with the equations arising in the CE theory may be achieved by realizing that substitution in the right-hand side of Eq. (27) of our f'_{0i} , \mathbf{c}_i , and $\mathbf{c}_i/(2kT_i/m_i)$ by f_{0i} , \mathbf{c} , and $\mathbf{c}/(2kT/m_i)$ introduces second-order errors proportional to Δ_i^2 [Eq. (9)]. Accordingly, consistent with the linearization of the Boltzmann collision integrals, we obtain

$$K_{11}\Phi_{1} + K_{12}(\Phi_{1} + \Phi_{2})$$

$$= f_{01}[2m_{1}\mathbf{cc}:\nabla \circ \mathbf{U}_{1} + \mathbf{c}\cdot\nabla \ln T_{1}(m_{1}c^{2} - 5kT)]/(2kT)$$

$$+ f_{01}[\mathbf{c}\cdot\lambda + \sigma'(-1 + m_{1}c^{2}/3kT)]/(n_{1}kT), \quad (28)$$

$$K_{22}\Phi_{2} + K_{21}(\Phi_{1} + \Phi_{2})$$

$$= f_{02}[2m_{2}\mathbf{cc}:\nabla \circ \mathbf{U}_{2} + \mathbf{c}\cdot\nabla \ln T_{2}(m_{2}c^{2} - 5kT)]/(2kT)$$

$$- f_{02}[\mathbf{c}\cdot\lambda + \sigma'(-1 + m_{2}c^{2}/3kT)]/(n_{2}kT), \quad (29)$$

where we have substituted

$$-\lambda_2 = \lambda_1 \equiv \lambda, \quad -\sigma_2' = \sigma_1' \equiv \sigma'. \tag{30}$$

Because $\sigma_2' + \sigma_1' = \lambda \cdot (\mathbf{U}_2 - \mathbf{U}_1)$, interchanging σ_2' with σ_1' is not exact; but the differences induced by the term proportional to λ are second-order quantities, comparable to those resulting from putting \mathbf{c} instead of \mathbf{c}_i . The substitution of σ' for $-\sigma_2'$ is essential in order for Eqs. (28) and (29) to satisfy the required compatibility conditions associated with a singular nonhomogeneous linear equation.

Notice finally that Δ_i is small provided that $(T_1 - T_2)$ and $(\mathbf{U}_1 - \mathbf{U}_2)$ are small with respect to T and the thermal speed of the light gas, respectively. But the time derivatives or the gradients of T_1 and T_2 (or \mathbf{U}_1 and \mathbf{U}_2) may be completely different from each other when the mass ratio differs significantly from unity. For instance, $\nabla \ln T_1$ and $\nabla \ln T_2$ are both first-order quantities; but their difference may be of the same order as either of them.

In conclusion, Eqs. (28) and (29) are mathematically nearly identical to those arising in the CE theory for a binary mixture. Yet there are important physical differences between the two theories. Here, the terms λ and σ' are free parameters providing the liberty to select the two velocities U_i and temperatures T_i independently, each satisfying its own dynamical equation (with time derivatives). The σ' term is absent from the CE theory, where only one temperature is allowed. The term d_{12} associated to diffusion in the CE theory is directly related to the present λ as we shall discuss below (end of Sec. III B). The remarkable thing is that the generalization to a two-fluid theory can be made within the same mathematical framework of the CE theory.

In order to solve Eqs. (28) and (29), we introduce the notation $\underline{K} \Phi$ for the left-hand sides of Eqs. (28) and (29), where \underline{K} is the matrix operator

$$\underline{K} = \begin{vmatrix} K_{11} + K_{12} & K_{12} \\ K_{21} & K_{21} + K_{22} \end{vmatrix}$$
 (31)

and Φ is the column vector

$$\underline{\Phi} = \begin{vmatrix} \Phi_1 \\ \Phi_2 \end{vmatrix} . \tag{32}$$

For these functions we introduce the inner product

$$\{\Phi,\Psi\} = [\Phi_1,\Psi_1] + [\Phi_2,\Psi_2],$$

in terms which the operator \underline{K} is symmetric and nonpositive (see, for instance, Chapman and Cowling, 7 Sec. 4.4, noticing that their operators $-n_1^2I_1, -n_1n_2I_{12}, -n_1n_2I_{21}$, and $-n_2^2I_2$ are, in our notation, K_{11}, K_{12}, K_{21} , and K_{22} , respectively; notice also that the Chapman-Cowling $^7 - n^2\{\Phi, \Psi\}$ is our $\{\Phi, K\Phi\}$). That is, \underline{K} satisfies

$$\{\underline{\Phi}, \underline{K}\underline{\Psi}\} = \{\underline{\Psi}, \underline{K}\underline{\Phi}\} \tag{33}$$

and

$$\{\Phi, K\Phi\} \leqslant 0 \tag{34}$$

for any pair of functions Φ and Ψ .

The solutions to Eqs. (28) and (29) may be obtained by superposition as

$$\underline{\Phi} = \underline{B}_1 \mathbf{c} \circ \mathbf{c} : \nabla \mathbf{U}_1 + \underline{B}_2 \mathbf{c} \circ \mathbf{c} : \nabla \mathbf{U}_2 + \underline{A}_1 \mathbf{c} \cdot \nabla \ln T_1 + \underline{A}_2 \mathbf{c} \cdot \nabla \ln T_2 + (\underline{D} \mathbf{c} \cdot \lambda + \underline{E} \sigma') / kT,$$
 (35)

where the vector functions \underline{B}_i , \underline{A}_i , \underline{D} , and \underline{E} are defined,

except for a linear combination of the collisional invariants whose transposed vector functions are (1,0), (0,1), $(m_1\mathbf{c},m_2\mathbf{c})$, (m_1c^2,m_2c^2) , by the solutions of the integral equations:

$$KB_i \mathbf{c} \circ \mathbf{c} = b_i \mathbf{c} \circ \mathbf{c}, \quad i = 1, 2,$$
 (36a)

$$\underline{K}\underline{A}_{i}\mathbf{c} = \underline{a}_{i}\mathbf{c}, \quad i = 1, 2, \tag{36b}$$

$$KD\mathbf{c} = d\mathbf{c},\tag{36c}$$

$$KE = e, (36d)$$

with

$$\underline{b}_{1} \equiv \begin{vmatrix} b_{11} \\ b_{12} \end{vmatrix} \equiv \frac{m_{1}}{kT} \begin{vmatrix} f_{01} \\ 0 \end{vmatrix}, \quad \underline{b}_{2} \equiv \begin{vmatrix} b_{21} \\ b_{22} \end{vmatrix} \equiv \frac{m_{2}}{kT} \begin{vmatrix} 0 \\ f_{02} \end{vmatrix}, \quad (37)$$

$$\underline{a}_{1} \equiv \begin{vmatrix} a_{11} \\ a_{12} \end{vmatrix} \equiv \left(\frac{m_{1}c^{2}}{2kT} - \frac{5}{2} \right) \begin{vmatrix} f_{01} \\ 0 \end{vmatrix},
\underline{a}_{2} \equiv \begin{vmatrix} a_{21} \\ a_{22} \end{vmatrix} \equiv \left(\frac{m_{2}c^{2}}{2kT} - \frac{5}{2} \right) \begin{vmatrix} 0 \\ f_{02} \end{vmatrix},$$
(38)

$$\underline{d} \equiv \begin{vmatrix} d_1 \\ d_2 \end{vmatrix} \equiv \begin{vmatrix} f_{01}/n_1 \\ -f_{02}/n_2 \end{vmatrix}, \tag{39}$$

$$\underline{e} \equiv \begin{vmatrix} e_1 \\ e_2 \end{vmatrix} \equiv \begin{vmatrix} (m_1 c^2 / 3kT - 1) f_{01} / n_1 \\ (-m_2 c^2 / 3kT + 1) f_{02} / n_2 \end{vmatrix}. \tag{40}$$

There is a similarity between these equations and the corresponding ones in the CE theory [Ref. 7, Eqs. (8.31, 4, 5, 6)]. In particular, the functions A, B, and D of Ref. 7 are our functions $-(A_1+A_2)$, $-(B_1+B_2)/2$, and -nD, respectively, while our function E does not appear in the CE theory. However, except for the D function, this equivalence is meaningful only in the limit when both temperatures and mean velocities coincide ($T_1 = T_2 = T$, $U_1 = U_2 = U$) because the CE functions $\underline{A}_1 + \underline{A}_2$, $\underline{B}_1 + \underline{B}_2$ are split in our function Φ accounting for the two different temperatures and the two different mean velocities. Moreover, there are important differences in the constraints imposed to the functions Φ_i versus those in the CE theory. The present Φ'_1 and Φ'_2 are such that they do not contribute to the mean temperature and mean velocity. Such a restriction could not be satisfied in the CE theory; but it may be here, because our freedom to vary λ controls the difference of mean speed between species, while changes in σ' lead to variations in the temperature difference (see below). The ordinary collision invariants of the CE theory allow the other necessary control on the mean mixture temperature and mean velocity, so that our solution provides a complete specification of λ and σ in terms of $T_2 - T_1$, $U_2 - U_1$, $\nabla \ln T_i$, and ∇U_i , thus closing the two-fluid equations.

The condition that $f'_{0i}\Phi'_i$ does not contribute to n_i , \mathbf{U}_i and T_i ,

$$[\Psi, f'_{0i}\Phi'_{i}] = 0, \quad \Psi = 1, \mathbf{c}, c^{2} \quad (i = 1, 2)$$
 (41)

may be rewritten in terms of the unprimed function $f_{0i}\Phi_i$. Using the fact that $f'_{0i}\Phi'_i = f_{0i}\Phi_i + f_{0i} - f'_{0i}$ we find

$$[1, f_{0i}\Phi_i] = n_i - n_i = 0 \quad (i = 1,2),$$
(42)

$$[\mathbf{c}, f_{0i}\Phi_i] = n_i(\mathbf{U}_i - \mathbf{U}) \quad (i = 1,2);$$

$$[c^2, f_{0i}\Phi_i] = 3n_i k(T_i - T)/m_i \quad (i = 1, 2),$$
 (43)

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where the second-order term $n_i(\mathbf{U}_i - \mathbf{U})^2/m_i$ has been dropped from the last equations. The first of these constraints can be satisfied, like in the CE theory, by appropriate combination of Φ with the collisional invariants whose transposes are $(\bar{1},0)$ and (0,1). The second two vectorial conditions may be reexpressed in terms of the d_i functions [Eq. (39)] as

$$\mathbf{U}_1 - \mathbf{U} = [d_1 \mathbf{c}, \Phi_1], \tag{44a}$$

$$\mathbf{U}_2 - \mathbf{U} = -[d_2, \mathbf{c}, \Phi_2].$$
 (44b)

Their difference yields

$$\mathbf{U}_{1} - \mathbf{U}_{2} = \{\mathbf{c}d, \Phi\},$$
 (45)

which fixes λ after substituting the expression (35) for Φ :

$$\lambda = \lambda_1 \nabla \ln T_1 + \lambda_2 \nabla \ln T_2 + \lambda_u (\mathbf{U}_1 - \mathbf{U}_2), \tag{46}$$

while

$$\lambda_u \equiv 3kT / \{Dc^2, d\},\tag{47}$$

$$\lambda_i = -\lambda_{ii} \{ A_i c^2, d \} / 3. \tag{48}$$

Similarly, the two conditions (43) may be rewritten in terms of the e_i functions [Eq. (40)] as

$$(T_1 - T)/T = [e_1, \Phi_1],$$
 (49a)

$$(T_2 - T)/T = -[e_2, \Phi_2],$$
 (49b)

whose difference yields

$$(T_1 - T_2)/T = \{\underline{e}, \underline{\Phi}\},\tag{50a}$$

determining σ' as

$$\sigma' = k(T_1 - T_2) / \{E, e\}. \tag{50b}$$

The second-independent constraints in Eqs. (42) and (43) can be satisfied, like in the CE theory, by suitable combination of Φ with the collisional invariants whose transposes are $(m_1\mathbf{c}, m_2\mathbf{c})$ and (m_1c^2, m_2c^2) . It is convenient to choose this combination such that U is the center-of-mass velocity of the mixture and T is the mixture temperature, that is,

$$\rho \mathbf{U} = \rho_1 \mathbf{U}_1 + \rho_2 \mathbf{U}_2$$

and

$$nT = n_1T_1 + n_2T_2,$$

which means that

$$[m_1\mathbf{c}, f_{01}\Phi_1] + [m_2\mathbf{c}, f_{02}\Phi_2] = 0$$

and

$$[m_1c^2, f_{01}\Phi_1] + [m_2c^2, f_{02}\Phi_2] = 0,$$

or

$$\rho_1[d_1\mathbf{c},\Phi_1] = \rho_2[d_2\mathbf{c},\Phi_2],\tag{51a}$$

$$n_1[e_1, \Phi_1] = n_2[e_2, \Phi_2].$$
 (51b)

Notice that the above addition of collisional invariants leaves the transport coefficients unaffected because they can be expressed (see next section) in terms of quantities of the form $\{L,I\}$, where KL=I are Eqs. (36a)–(36d).

III. TRANSPORT COEFFICIENTS AND THEIR VARIATIONAL DETERMINATION

The closure of the two-fluid hydrodynamic equations requires expressing the interspecies momentum and energy terms λ and σ' as well as the heat fluxes \mathbf{Q}_i and stress tensors \mathbf{P}_i in terms of the hydrodynamic variables and their gradi-

ents. All these objects, like Φ , are linear in the nonequilibrium driving forces $\nabla \circ \mathbf{U}_i, \nabla \ln T_i, (T_i - \mathbf{T})$, and $(\mathbf{U}_i - \mathbf{U})$, with proportionality coefficients (transport coefficients) given by integral functionals of the solutions to the integral Eqs. (36a)–(36d). In the present section we follow Bernstein⁸ in expressing these transport coefficients in terms of quantities that can be determined optimally through a variational approach. Equations (36a)–(36d) may all be written schematically as

$$KL = I. (52)$$

In view of the symmetry properties (33) of \underline{K} , the solution \underline{L} to the above equation extremalizes the functional

$$\Lambda[L] \equiv \{L, KL - 2I\},\tag{53}$$

which takes the (positive) extremal value

$$\Lambda_{\text{ext}} = -\{\underline{L},\underline{I}\} = -\{\underline{L},\underline{K}\underline{L}\}. \tag{54}$$

Consequently, if Λ is evaluated by means of a trial function \underline{L}_t , where $\underline{L}_t = \underline{L} + \underline{\epsilon}$, which differs from the exact solution \underline{L} by the small quantity $\underline{\epsilon}$, the resulting value of the functional differs from the exact value by a quantity of order ϵ^2 only:

$$\Lambda[L+\epsilon] = \Lambda_{\rm ext} + \{\epsilon, K\epsilon\}. \tag{55}$$

Furthermore, because the operator \underline{K} is nonpositive, $\{\underline{\epsilon}, \underline{K}\underline{\epsilon}\} \leqslant 0$, the extremal value of Λ is actually a maximum. Accordingly, objects of the form $\{\underline{L}, \underline{K}\underline{L}\}$ may be evaluated optimally by maximizing the functional $\Lambda\{\underline{L}\}$ above. In order to exploit this feature for the computation of the transport coefficients, we shall proceed to write them in terms of such curly bracket integrals. This program has already been initiated for the momentum and energy transfer terms λ and σ' , because the inner products $\{\underline{D}\mathbf{c},\underline{d}\mathbf{c}\}$ and $\{\underline{E},\underline{e}\}$ entering in Eqs. (47) and (50b) have the desired form $\{\underline{D}\mathbf{c},\underline{K}\underline{D}\mathbf{c}\}$ and $\{\underline{E},\underline{K}\underline{E}\}$. The other mixed quantities $\{\underline{A}_1\mathbf{c},\underline{c}\underline{d}\}$ and $\{\underline{A}_2\mathbf{c},\underline{c}\underline{d}\}$ may also be obtained optimally by noticing the identity

$$2\{\underline{L}_{1},\underline{K}\underline{L}_{2}\} = \{\underline{L}_{1} + \underline{L}_{2},\underline{K}(\underline{L}_{1} + \underline{L}_{2})\} - \{L_{1},KL_{1}\} - \{L_{2},KL_{2}\},$$
 (56)

where each one of the three right-hand side terms may be found variationally. Accordingly, we proceed to express all remaining transport coefficients in terms of integrals of the forms $\{L, KL\}$ or $\{L_1, KL_2\}$.

A. Stress tensors

The stress \mathbf{P}_i of species i is given by $\mathbf{P}_i = [m_i \mathbf{c}_i \mathbf{c}_i, f_i]$. Here \mathbf{P}_i may be decomposed into an isotropic pressure $p_i \mathbf{I}$ (I is the unit tensor) and a traceless viscous stress tensor Π_i , where

$$3p_i \equiv [m_i c_i^2, f_i] = [m_i c_i^2, f'_{0i}] = 3n_i k T_i,$$
 (57)

while

$$\Pi_{i} \equiv \left[\mathbf{c}_{i} \circ \mathbf{c}_{i} m_{i} f_{0i}, 1 + \Phi_{i} \right] = \left[\mathbf{c}_{i} \circ \mathbf{c}_{i} m_{i} f_{0i}, \Phi_{i} \right]
\simeq \left[\mathbf{c} \circ \mathbf{c} m_{i} f_{0i}, \Phi_{i} \right],$$
(58)

where the second-order corrections have been neglected in the last equality above. Substituting Eq. (35) for Φ in the expression for Π_i , we obtain after writing the $\mathbf{c} \circ \mathbf{c} \, m_i \, \bar{f}_{0i}$ functions in terms of the b function of Eq. (37)

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$$\Pi_i = \{b_i \mathbf{c} \circ \mathbf{c} : \mathbf{c} \circ \mathbf{c}, B_1 \nabla \circ \mathbf{U}_1 + B_2 \nabla \circ \mathbf{U}_2\} kT / 5.$$
 (59)

Introducing the definitions for the four viscosity coefficients

$$\Pi_i = -2\mu_{i1} \nabla \cdot \mathbf{U}_1 - 2\mu_{i2} \nabla \cdot \mathbf{U}_2, \tag{60}$$

one obtains after identifying coefficients in Eqs. (59) and (60)

$$\mu_{ii} \equiv -\{b_i \mathbf{c} \circ \mathbf{c} : \mathbf{c} \circ \mathbf{c}, B_i\} kT/10, \tag{61}$$

which from the symmetry of \underline{K} (notice that $KB_i \mathbf{c} \circ \mathbf{c} = b_i \mathbf{c} \circ \mathbf{c}$) satisfy

$$\mu_{ij} = \mu_{ji}. \tag{62}$$

The sum of the four viscosity coefficients (61) is equal to the viscosity μ of the CE theory for binary mixtures [Ref. 7, Eq. (8.42,1)]. Note that this sum may be written as

$$-\left[m_{1}\int d^{3}c f_{01}\mathbf{c}\circ\mathbf{c}:\mathbf{c}\circ\mathbf{c}(B_{11}+B_{21})\right]$$

$$+m_{2}\int d^{3}c f_{02}\mathbf{c}\circ\mathbf{c}:\mathbf{c}\circ\mathbf{c}(B_{12}+B_{22})$$
/10,

while Eq. (8.42,1) of Ref. 7 reads

$$5\mu \equiv 2n^2kT\{B,B\}$$

$$= m_1 \int d^3c \, \mathbf{c} \circ \mathbf{c} : \mathbf{c} \circ \mathbf{c} B_1 + m_2 \int d^3c \, \mathbf{c} \circ \mathbf{c} : \mathbf{c} \circ \mathbf{c} B_2,$$

and their B_i is equal to our $-(B_{1i} + B_{2i})/2$. Thus in the limit $U_1 = U_2 = U$, the mixture pressure tensor **P** may be expressed as

$$\mathbf{P} = \mathbf{P}_1 + \mathbf{P}_2 = p\mathbf{I} - 2\mu \nabla \cdot \mathbf{U},$$

with $p = p_1 + p_2$, in accordance with the CE theory.

B. Heat fluxes and momentum transfer

In terms of the variable $\mathbf{c} = \mathbf{u} - \mathbf{U}$, rather than $\mathbf{c}_i = \mathbf{u} - \mathbf{U}_i$, it may be shown that the heat flux $\mathbf{Q}_i \equiv \left[\frac{1}{2}m_i|\mathbf{u} - \mathbf{U}_i|^2(\mathbf{u} - \mathbf{U}_i), f_i\right]$ can be written as

$$\mathbf{Q}_{i} = kT \left[\mathbf{c} (m_{i}c^{2} - 5kT) f_{0i}, \Phi_{i} \right] / 2kT, \tag{63}$$

with errors of order $|\mathbf{U}_i - \mathbf{U}|^2$. By means of the vector functions a_i defined in Eq. (38), \mathbf{Q}_i may be reexpressed as

$$\mathbf{Q}_i = kT\{a_i\mathbf{c},\Phi\},\tag{64}$$

or, substituting Φ from Eq. (35),

$$\mathbf{Q}_{i} = kT\{\underline{a}_{i}\mathbf{c},\underline{A}_{1}\mathbf{c}\cdot\nabla\ln T_{1} + \underline{A}_{2}\mathbf{c}\cdot\nabla\ln T_{2} + \underline{D}\mathbf{c}\cdot\lambda/kT\}.$$
(65)

The linearity of \mathbf{Q}_i in the quantities $\nabla \ln T_i$ and $\mathbf{U}_1 - \mathbf{U}_2$ suggests defining the transport coefficients λ_{ij} and λ_{iu} through

$$\mathbf{Q}_{i} \equiv -\lambda_{i1} \nabla T_{1} - \lambda_{i2} \nabla T_{2} + \lambda_{iu} (\mathbf{U}_{1} - \mathbf{U}_{2}), \tag{66}$$

with

$$-3\lambda_{ij} = k\{\underline{a}_i c^2, \underline{A}_j\} + \{\underline{D}c^2, \underline{a}_i\}\lambda_j,$$

$$3\lambda_{iu} = \{\underline{a}_i c^2, \underline{D}\}\lambda_u,$$
(67)

where the factor T/T_j in λ_{ij} has been taken to be unity according to our first-order theory and λ_u , λ_j are given by Eqs. (47) and (48). In virtue of the symmetry of \underline{K} , λ_{iu} reduces to $-\lambda_i$:

$$\lambda_{iu} = -\lambda_i. \tag{68}$$

By substituting λ_i from Eq. (48) and using the symmetry properties of \underline{K} , the thermal conductivity matrix λ_{ij} may be reexpressed as

$$-3\lambda_{ij} = k\{\underline{A}_{j}\mathbf{c},\underline{K}\underline{A}_{i}\mathbf{c}\}:\mathbf{I} - \lambda_{u}\{\underline{A}_{i},\underline{d}c^{2}\}\{\underline{A}_{j},\underline{d}c^{2}\}/3, \quad (69)$$

where its symmetry becomes manifest:

$$\lambda_{ij} = \lambda_{ji}. \tag{70}$$

Let us compare these results with the CE theory for binary mixtures. The first approximation of the total heat flux may be written as

$$\mathbf{Q} = [m_1 \mathbf{c}c^2 / 2, f_1] + [m_2 \mathbf{c}c^2 / 2, f_2]$$

$$\simeq \mathbf{Q}_1 + \mathbf{Q}_2 + \frac{5}{2}kT \{n_1(\mathbf{U}_1 - \mathbf{U}) + n_2(\mathbf{U}_2 - \mathbf{U})\}$$

$$= -(\lambda_{11} + \lambda_{21})\nabla T_1 - (\lambda_{12} + \lambda_{22})\nabla T_2$$

$$+ (\lambda_{1u} + \lambda_{2u})(\mathbf{U}_1 - \mathbf{U}_2)$$

$$+ 5kT \{n_1(\mathbf{U}_1 - \mathbf{U}) + n_2(\mathbf{U}_2 - \mathbf{U})\} / 2. \tag{71}$$

Using the symmetry of \underline{K} and the relations of Eqs. (47) and (48) for λ_u and λ_i , the sum λ of the four thermal conductivities (69) can be expressed as

$$3\lambda = -k \{ \{ (\underline{A}_1 + \underline{A}_2)c^2, \mathbf{a}_1 + \mathbf{a}_2 \}$$
$$- \{ Dc^2, (a_1 + a_2) \}^2 / \{ Dc^2, d \} \},$$
(72)

which is equal to the mixture thermal conductivity of the CE theory for binary mixtures [Ref. 7, Eq. (8.41,4), noticing the equivalence of functions and inner products described in Sec. II]. Since, in addition, the mixture thermal diffusion ratio k_T of the CE theory is equal to our

$$(\lambda_{1u} + \lambda_{2u})/nkT = n^{-1}\{(\underline{a}_1 + \underline{a}_2)c^2,\underline{D}\}/\{\underline{D}c^2,\underline{d}\}$$

[Ref. 7, Eq. (8.4, 6)], the total heat flux given by Eq. (71) coincides with that in the CE theory in the limit $T_1 = T_2 = T$ [compare with Eq. (8.41, 3) of Ref. 7]. This suggests the introduction of the coefficients k_{Ti} ,

$$k_{Ti} \equiv \lambda_{iu} / nkT = n^{-1} \{\underline{a}_i c^2, \underline{D}\} / \{\underline{D}c^2, \underline{d}\}.$$
 (73)

Moreover, instead of the coefficient λ_u , it is convenient to use a new one λ_h defined by

$$\rho_1 \rho_2 \lambda_b = -m_2 \lambda_u = -3k T m_2 / \{Dc^2, d\}, \tag{74}$$

which, as we shall see in the next section, depends on n_1 and n_2 through the molar fractions $x_i \equiv n_i/n$ (the minus sign is because $\lambda_b > 0$). In terms of these coefficients, the expressions for \mathbf{Q}_i and λ become

$$\mathbf{Q}_i = -\lambda_{i1} \nabla T_1 - \lambda_{i2} \nabla T_2 + nkTk_{Ti} (\mathbf{U}_1 - \mathbf{U}_2) \quad (75)$$

and

$$\lambda = -nkT(k_{T1}\nabla \ln T_1 + k_{T2}\nabla \ln T_2)$$
$$-\rho_1\rho_2\lambda_h(\mathbf{U}_1 - \mathbf{U}_2)/m_2. \tag{76}$$

Finally, notice that the last expression coincides with the CE equation for $U_1 - U_2$ [Ref. 7, Eq. (8.4, 7)] if we identify the vector \mathbf{d}_{12} of the CE theory with λ/p in the limit $T_1 = T_2$, since by Eq. (74) and Eq. (8.4, 4) of Ref. 7, $\lambda_b = kT/(m_1nD_{12})$, where D_{12} is the binary diffusion coefficient of the CE theory.

IV. VARIATIONAL CALCULATION OF TRANSPORT COEFFICIENTS

As explained at the beginning of Sec. III, the transport coefficients may be obtained via variational principles as the maxima of functionals of the form $\{\underline{L}, \underline{K}\underline{L}\}$. In particular, we define the functionals⁹

$$\Lambda_{Bi} \left[\Xi_{B} \mathbf{c} \circ \mathbf{c} \right] \equiv \{ \Xi_{B} \mathbf{c} \circ \mathbf{c} : \underline{K} \Xi_{B} \mathbf{c} \circ \mathbf{c} - 2\underline{b}_{i} \mathbf{c} \circ \mathbf{c} \}, \quad i = 1, 2,$$
(77a)

$$\Lambda_{Ai} \left[\Xi_A \mathbf{c} \right] \equiv \left\{ \Xi_A \mathbf{c} \cdot K \Xi_A \mathbf{c} - 2a_i \mathbf{c} \right\}, \quad i = 1, 2, \tag{77b}$$

$$\Lambda_D \left[\Xi_D \mathbf{c} \right] \equiv \left\{ \Xi_D \mathbf{c} \cdot K \Xi_D \mathbf{c} - 2d \mathbf{c} \right\}, \tag{77c}$$

and

$$\Lambda_E[\Xi_E] = \{\Xi_E, K\Xi_E - 2e\},\tag{77d}$$

which from Eqs. (33), (34), and (36) have the following absolute maxima:

$$\Lambda_{Bi}^* \equiv \Lambda_{Bi} \left[\underline{B}_i \mathbf{c} \circ \mathbf{c} \right] = -\left\{ \underline{B}_i \mathbf{c} \circ \mathbf{c} : \mathbf{c} \circ \mathbf{c}, \underline{b}_i \right\}, \quad i = 1, 2,
\Lambda_{Ai}^* \equiv \Lambda_{Ai} \left[\underline{A}_i \mathbf{c} \right] = -\left\{ \underline{A}_i c^2, \underline{a}_i \right\}, \quad i = 1, 2,
\Lambda_D^* \equiv \Lambda_D \left[D\mathbf{c} \right] = -\left\{ Dc^2, d \right\},$$

and

$$\Lambda_E^* \equiv \Lambda_E[E] = -\{E, e\}. \tag{78}$$

These quantities are non-negative in virtue of the nonpositivity of K and are related to the transport coefficients.

For the viscosities we have

$$\mu_{11} = kT\Lambda_{B1}^*/10, \quad \mu_{22} = kT\Lambda_{B2}^*/10,$$
 (79)

whence μ_{11} and μ_{22} are optimally calculated using the functions Ξ_{B1}^* and Ξ_{B2}^* which maximize the functionals Λ_{B1} and Λ_{B2} within the class of functions Ξ_{B} . These two functions also yield μ_{12} optimally since the absolute maximum of the functional

$$\Lambda_{B}\left[\Xi_{B}\mathbf{c}\circ\mathbf{c}\right] \equiv \left\{\Xi_{B}\mathbf{c}\circ\mathbf{c}:\underline{K}\Xi_{B}\mathbf{c}\circ\mathbf{c} - 2(\underline{b}_{1} + \underline{b}_{2})\mathbf{c}\circ\mathbf{c}\right\}$$
(80)

$$\Lambda_B^* \equiv \Lambda_B \left[(\underline{B}_1 + \underline{B}_2) \mathbf{c} \circ \mathbf{c} \right]
= -\left\{ (\underline{B}_1 + \underline{B}_2) \mathbf{c} \circ \mathbf{c} : \mathbf{c} \circ \mathbf{c}, \underline{b}_1 + \underline{b}_2 \right\}
= 10\mu/kT.$$
(81)

Thus using the same class of functions, the maximum of Λ_B is reached with $\Xi_{B1}^* + \Xi_{B2}^*$ and if μ_{11} , μ_{22} , and μ are optimally obtained, so is μ_{12} because $\mu = \mu_{11} + \mu_{22} + 2\mu_{12}$.

Let us rewrite the transport coefficients appearing in the heat flux and in the momentum transfer [Eqs. (67), (47), (48), (73), and (74)] as

$$\lambda_{ii} = -(k/3)(\{\underline{A}_{i}c^{2},\underline{a}_{i}\} - \{\underline{D}c^{2},\underline{a}_{i}\}^{2}/\{\underline{D}c^{2},\underline{d}\}),$$

$$i = 1,2,$$
(82)

$$\lambda_{12} = \lambda_{21} = -(k/3)(\{\underline{A}_{2}c^{2},\underline{a}_{1}\} - \{Dc^{2},a_{1}\}\{Dc^{2},a_{2}\}/\{Dc^{2},d\}), \tag{83}$$

$$\lambda_b = -3kT/\rho_1 n_2 \{\underline{D}c^2, \underline{d}\}, \tag{84}$$

$$k_{T_i} = \{Dc^2, a_i\} / n\{Dc^2, d\}, \quad i = 1, 2.$$
 (85)

Notice that λ_b , λ_{11} , and λ_{22} are non-negative (λ_{ii} can be written as $-k\{\underline{T}_ic^2,t_j\}/3$ with $\underline{T}_i=\underline{A}_i-\underline{D}\{\underline{D}c^2,\underline{a}_i\}/\{\underline{D}c^2,\underline{d}\}$ and $\underline{K}\underline{T}_i\mathbf{c}=\underline{t}_i\mathbf{c}$). Clearly, λ_b is optimally calculated by the function $\underline{\Xi}_D^*$, which makes the functional Λ_D a maximal Λ_D

mum. Once we have fixed $\underline{D} = \underline{\Xi}_D^*$, k_{T1} and k_{T2} are fixed and λ_{11} and λ_{22} are optimally calculated by the functions $\underline{\Xi}_{A1}^*$ and $\underline{\Xi}_{A2}^*$ that maximize the functionals Λ_{A1} and Λ_{A2} , respectively. By a similar argument to that used for μ_{12} , these functions also determine optimally λ_{12} since the mixture thermal conductivity λ [which is also non-negative since $\lambda = -\{(\underline{T}_1 + \underline{T}_2)c^2, (t_1 + t_2)\}$; see Eq. (72)] is optimally obtained with $\underline{\Xi}_{A1}^* + \underline{\Xi}_{A2}^*$ (for D fixed and within the class of functions $\underline{\Xi}_A$).

Finally, for the transfer of energy it is convenient to introduce the coefficient σ_T defined as

$$\sigma' = -n^2 \sigma_T (T_1 - T_2) \tag{86}$$

or, from Eqs. (50b) and (78),

$$n^2 \sigma_T = -k/\{E,e\} = k/\Lambda_E^*. \tag{87}$$

This coefficient has the advantage of depending on the densities n_i only through the molar fractions x_i . Using the variational principle, maximizing Λ_E yields σ_T optimally.

For calculation purposes it is usual to take Sonine polynomial expansions as trial functions (for a brief description of the Sonine polynomials see, for instance, Ref. 7, p. 127). Thus we choose the functions

$$\Xi = \sum_{n=0}^{N} \begin{vmatrix} \alpha_n^1 & S_v^{(n)} \\ \alpha_n^2 & S_v^{(n)} \end{vmatrix}, \tag{88}$$

with $v = \frac{5}{2}$, $\frac{3}{2}$, and $\frac{1}{2}$ for Ξ_B , Ξ_A and Ξ_D , and Ξ_E , respectively, because of the special orthogonality properties for each one of the particular cases. Introducing these functions in Eqs. (77), we are led to the optimal functions

$$\Xi_{B_i}^* = \sum_{n=0}^{N} \begin{vmatrix} b_{ni}^1 & S_{5/2}^{(n)} \\ b_{ni}^2 & S_{5/2}^{(n)} \end{vmatrix}, \quad i = 1, 2,$$
 (89)

$$\underline{\Xi}_{A_i}^* = \sum_{n=0}^{N} \begin{vmatrix} a_{ni}^1 & S_{3/2}^{(n)} \\ a_{ni}^2 & S_{3/2}^{(n)} \end{vmatrix}, \quad i = 1, 2,$$
 (90)

$$\Xi_{D}^{*} = \sum_{n=0}^{N} \begin{vmatrix} d_{n}^{1} & S_{3/2}^{(n)} \\ d_{n}^{2} & S_{3/2}^{(n)} \end{vmatrix}, \tag{91}$$

$$\Xi_E^* = \sum_{n=0}^{N} \begin{vmatrix} e_n^1 & S_{1/2}^{(n)} \\ e_n^2 & S_{1/2}^{(n)} \end{vmatrix}, \tag{92}$$

with the coefficients given by the solution of the linear algebraic equations

$$\sum_{m=0}^{N} \left(b_{mi}^{1} L_{nm}^{11} + b_{mi}^{2} L_{nm}^{12} \right) = l_{ni} \delta_{i1},$$

$$\sum_{m=0}^{N} \left(b_{mi}^{1} L_{nm}^{21} + b_{mi}^{2} L_{nm}^{22} \right) = l_{ni} \delta_{i2};$$
(93a)

$$\sum_{m=0}^{N} \left(a_{mi}^{1} Q_{nm}^{11} + a_{mi}^{2} Q_{nm}^{12} \right) = q_{ni} \delta_{i1},$$

$$\sum_{m=0}^{N} \left(a_{mi}^{1} Q_{nm}^{21} + a_{mi}^{2} Q_{nm}^{22} \right) = q_{ni} \delta_{i2};$$
(93b)

$$\sum_{m=0}^{N} \left(d_{m}^{1} Q_{nm}^{11} + d_{m}^{2} Q_{nm}^{12} \right) = p_{n}^{1},$$

$$\sum_{m=0}^{N} \left(d_{m}^{1} Q_{nm}^{21} + d_{m}^{2} Q_{nm}^{22} \right) = p_{n}^{2};$$

$$\sum_{m=0}^{N} \left(e_{m}^{1} E_{nm}^{11} + e_{m}^{2} E_{nm}^{12} \right) = \epsilon_{n}^{1},$$
(93c)

$$\sum_{m=0}^{N} \left(e_m^1 E_{nm}^{21} + e_m^2 E_{nm}^{22} \right) = \epsilon_n^2, \quad n = 0, 1, 2, ..., N.$$
(93d)

In these equations δ_{ij} is the Kronecker delta,

$$L_{nm}^{11} = \left[S_{5/2}^{(n)} \mathbf{c}^{\circ} \mathbf{c}; (K_{11} + K_{12}) S_{5/2}^{(m)} \mathbf{c}^{\circ} \mathbf{c} \right],$$

$$L_{nm}^{12} = L_{nm}^{21} = \left[S_{5/2}^{(n)} \mathbf{c}^{\circ} \mathbf{c}; K_{12} S_{5/2}^{(m)} \mathbf{c}^{\circ} \mathbf{c} \right],$$

$$L_{nm}^{22} = \left[S_{5/2}^{(n)} \mathbf{c}^{\circ} \mathbf{c}; (K_{22} + K_{21}) S_{5/2}^{(m)} \mathbf{c}^{\circ} \mathbf{c} \right],$$
(94a)

and similarly Q_{nm}^{ij} , E_{nm}^{ij} with $S_{3/2}$ **c** and $S_{1/2}$, respectively;

$$l_{ni} = 10n_i kT/m_i, \quad n = 0, \quad l_{ni} = 0, \quad n \neq 0,$$

$$q_{ni} = -15n_i kT/2m_i, \quad n = 1, \quad q_{ni} = 0, \quad n \neq 1,$$

$$p_n^i = (-1)^{i+1} 3kT/m_i, \quad n = 0, \quad p_n^i = 0, \quad n \neq 0,$$

$$\epsilon_n^i = (-1)^i, \quad n = 1, \quad \epsilon_n^i = 0, \quad n \neq 0.$$
(94b)

Notice that Eqs. (93b)-(93d) should be completed with the constraints given by Eqs. (51), since the matrices Q and E are singular. The transport coefficients calculated with these functions are

$$\mu_{ij} = -kTl_{0i}b_{0j}^{i}/10,$$

$$\lambda_{ij} = 5k^{2}n_{i}T(a_{1j}^{i} - d_{1}^{i}a_{0j}^{i}/d_{0}^{i})/2m_{i},$$

$$k_{T1} = -5M_{2}x_{1}x_{2}d_{1}^{1}/2x_{0}d_{0}^{1},$$

$$k_{T2} = 5M_{1}x_{1}x_{2}d_{1}^{2}/2x_{0}d_{0}^{2},$$

$$\lambda_{b} = -(n_{1}d_{0}^{1})^{-1}, \quad \sigma_{T} = kx_{2}/n^{2}e_{1}^{1},$$
(95a)

where $M_i = m_i/(m_1 + m_2)$ and $x_0 = M_1x_1 + M_2x_2$. The coefficients L_{nm}^{ij} , Q_{nm}^{ij} , and E_{nm}^{ij} may be expressed in terms of the "bracket integrals." For example, in the notation of Ferziger and Kaper's $[\ ,\]_1, [\ ,\]_2, [\ ,\]_{12}^{\prime\prime}$, and $[\ ,\]_{12}^{\prime\prime}$ (Ref. 10, Secs. 7.1 and 7.2),

$$Z_{nm}^{11} = -n_1^2 \left[S_v^{(n)} z_1, S_v^{(m)} z_1 \right]_1 - n_1 n_2 \left[S_v^{(n)} z_1, S_v^{(m)} z_2 \right]_{12}^{\prime},$$
(95b)
$$Z_{nm}^{12} = -n_1 n_2 \left[S_v^{(n)} z_1, S_v^{(m)} z_2 \right]_{12}^{\prime\prime},$$
(95c)

$$Z_{nm}^{22} = -n_2^2 \left[S_v^{(n)} z_2, S_v^{(m)} z_2 \right]_2 - n_1 n_2 \left[S_v^{(n)} z_1, S_v^{(m)} z_2 \right]_{12}^{\prime},$$
(95d)

where Z stands for L, Q, and E [with $v = \frac{5}{2}, \frac{3}{2}, \frac{1}{2}$ and $z_i = \xi_i \circ \xi_i$, ξ_i , 1, respectively; $\xi_i \equiv \mathbf{c}_i (m_i/2kT)^{1/2}$]. Some of these bracket integrals for L and Q are tabulated in terms of the Ω integrals (see, i.e., Ref. 10, Sec. 7.2). However, the bracket integrals corresponding to E_{nm}^{ij} do not appear in the CE theory for binary mixtures, and have to be calculated. Up to n = m = 2, the following values are obtained:

$$\begin{split} E_{00}^{ij} &= E_{01}^{ij} = E_{10}^{ij} = 0, \\ E_{11}^{11} &= E_{11}^{22} = -E_{11}^{12} = -E_{11}^{21} = -16n_1n_2M_1M_2\Omega_{12}^{(1,1)}, \\ E_{12}^{11} &= E_{21}^{12} = -E_{11}^{12} = -E_{11}^{21} = -16n_1n_2M_1M_2\Omega_{12}^{(1,1)}, \\ E_{12}^{11} &= E_{21}^{12} = 16n_1n_2M_1M_2\left[5(M_1 - 1)\Omega_{12}^{(1,1)}/2 + M_2\Omega_{12}^{(1,2)}\right], \\ E_{12}^{12} &= E_{21}^{12} = 16n_1n_2M_1M_2\left[5\Omega_{12}^{(1,1)}/2 - \Omega_{12}^{(1,2)}\right], \\ E_{12}^{12} &= E_{21}^{21} = 16n_1n_2M_1M_2\left[5\Omega_{12}^{(1,1)}/2 - \Omega_{12}^{(1,2)}\right], \\ E_{21}^{22} &= E_{12}^{22} \\ &= 16n_1n_2M_1M_2\left[5(M_2 - 1)\Omega_{12}^{(1,1)}/2 + M_1\Omega_{12}^{(1,2)}\right], \\ E_{22}^{11} &= n_1^2\left(37\Omega_1^{(1,1)}/4 - 2\Omega_1^{(2,2)}\right) \end{split}$$

$$\begin{split} &-16n_{1}n_{2}M_{1}M_{2}\big[\big(1+53(M_{1}-1)^{2}/2\big)\Omega_{12}^{(1,1)}\\ &+5M_{2}(M_{1}-1)\Omega_{12}^{(1,2)}+M_{2}^{2}\Omega_{12}^{(1,3)}\\ &+M_{1}M_{2}\Omega_{12}^{(2,2)}\big],\\ E_{22}^{12}&=E_{22}^{21}=-16n_{1}n_{2}M_{1}^{2}M_{2}^{2}\big[-53\Omega_{12}^{(1,1)}/32\\ &+5\Omega_{12}^{(1,2)}-\Omega_{12}^{(1,3)}+\Omega_{12}^{(2,2)}\big],\\ E_{22}^{22}&=n_{2}^{2}\big(37\Omega_{2}^{(1,1)}/4-2\Omega_{2}^{(2,2)}\big)\\ &-16n_{1}n_{2}M_{1}M_{2}\big[\big(1+53(M_{2}^{2}-1)^{2}/2\Omega_{12}^{(1,1)}\big)\\ &+5M_{1}(M_{2}-1)\Omega_{12}^{(1,2)}+M_{1}^{2}\Omega_{12}^{(1,3)}+M_{1}M_{2}\Omega_{12}^{(2,2)}\big]. \end{split}$$

These integrals have been calculated using the method of Ref. 10, Appendix B.

From Eqs. (95) and (94a), to obtain the first approximation of the transport coefficients we have to retain one term in Eq. (93a) (that is, set N = 1), and two terms in the remaining equations (93b)-(93d) (N = 2). We obtain

$$[\mu_{11}]_1 = \left[x_1^2(\frac{2}{3} + M_1A_1/M_2) + x_1x_2B_1/2\mu_2\right]/Q_1,$$

$$[\mu_{22}]_1 = \left[x_2^2(\frac{2}{3} + M_2A_1/M_1) + x_1x_2B_1/2\mu_1\right]/Q_1,$$

$$[\mu_{12}]_1 = [\mu_{21}]_1 = x_1x_2(\frac{2}{3} - A_1)/Q_1,$$

$$[\lambda_{11}]_1 = (75x_1k^2T/64m_1Q_2)(P_2 - P_1P_6/P_5),$$

$$[\lambda_{22}]_1 = (75x_2k^2T/64m_2Q_2)(P_4 - P_1P_7/P_5),$$

$$[\lambda_{12}]_1 = [\lambda_{21}]_1 = -(75x_2k^2T/64m_2Q_2)(P_3 + P_1P_7/P_5),$$

$$[k_{T1}]_1 = -5M_2x_1x_2P_6/2x_0P_5,$$

$$[k_{T2}]_1 = -5M_1x_2^2P_7/2x_0P_5,$$

$$[\lambda_b]_2 = -16M_2Q_2/3x_0P_5,$$

$$[\sigma_T]_1 = 16kM_1M_2x_1x_2\Omega_{12}^{(1,1)},$$
with
$$Q_1 = x_1^2(\frac{2}{3} + M_1A_1/M_2)/\mu_1 + x_2^2(\frac{2}{3} + M_2A_1/M_1)/\mu_2$$

$$+ x_1x_2(B_1/2\mu_1\mu_2 + 4A_1/3B_1M_1M_2),$$

$$\mu_1 = 5kT/8\Omega_1^{(2,2)}, \quad \mu_2 = 5kT/8\Omega_2^{(2,2)},$$

$$A_1 = \Omega_{12}^{(2,2)}/5\Omega_{12}^{(1,1)}, \quad B_1 = kT/8M_1M_2\Omega_{12}^{(1,1)},$$

$$Q_2 = BM_2x_0P_1 + (x_1E + M_2x_2C)P_2 + M_1^2M_2x_2GP_3,$$

$$P_1 = M_1^{3/2}M_2^{3/2}B(x_2F/x_1M_1 + D - M_1^2G),$$

$$P_2 = M^{1/2}x_0[M_1(M_1^2B^2 - AD)/x_2 - AF/x_1],$$

$$P_3 = (M_1M_2)^{3/2}(AG - B^2)x_0/x_2,$$

$$P_4 = M^{1/2}x_0[M_2(M_2^2B^2 - AC)/x_1 - AE/x_2],$$

$$P_5 = M^{1/2}[(x_1E/x_2 + M_2C)(x_2F/x_1 + M_1D) - M_1^3M_2^3G^2],$$

$$P_6 = -M^{1/2}x_0B[M_2F/x_1 + M_1M_2(D - M_1^2G)/x_2],$$

$$P_7 = M^{1/2}x_0B[Ex_1M_1/x_2 + M_1M_2(C - M_2^2G)]/x_2,$$

$$A = \Omega_{11}^{(1,1)}, \quad B = 5\Omega_{11}^{(1,1)}/2 - \Omega_{12}^{(1,2)},$$

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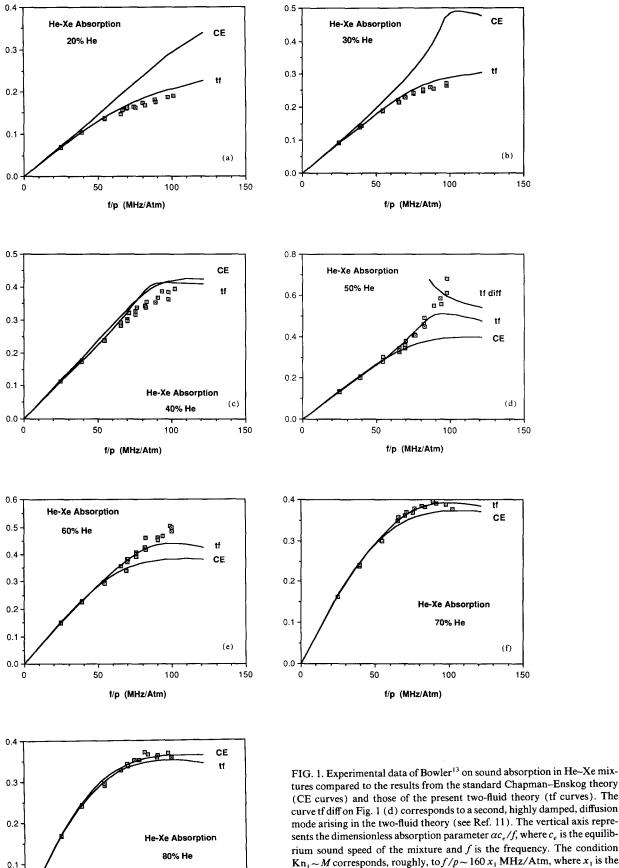
 $C = 5(6M_1^2 + 5M_2^2)\Omega_{12}^{(1,1)}/4 - 5M_2^2\Omega_{12}^{(1,2)}$

 $D = 5(6M_2^2 + 5M_1^2)\Omega_{12}^{(1,1)}/4 - 5M_1^2\Omega_{12}^{(1,2)}$

 $+M_{2}^{2}\Omega_{12}^{(1,3)}+2M_{1}M_{2}\Omega_{12}^{(2,2)},$

 $+M_1^2\Omega_{12}^{(1,3)}+2M_1M_2\Omega_{12}^{(2,2)}$

 $E = \Omega_1^{(2,2)}/2, \quad F = \Omega_2^{(2,2)}/2.$



(CE curves) and those of the present two-fluid theory (tf curves). The curve tf diff on Fig. 1 (d) corresponds to a second, highly damped, diffusion mode arising in the two-fluid theory (see Ref. 11). The vertical axis represents the dimensionless absorption parameter $\alpha c_e/f$, where c_e is the equilibrium sound speed of the mixture and f is the frequency. The condition $Kn_1 \sim M$ corresponds, roughly, to $f/p \sim 160 x_1$ MHz/Atm, where x_1 is the He molar fraction (this explains why the frequency range of validity of the standard CE theory increases with x_1).

f/p (MHz/Atm)

100

50

(g)

150

$$G = 55\Omega_{12}^{(1,1)}/4 - 5\Omega_{12}^{(1,2)} + \Omega_{12}^{(1,3)} - 2\Omega_{12}^{(2,2)},$$

$$x_0 = x_1 M_1 + x_2 M_2, \quad M_i = m_i/(m_1 + m_2),$$

$$M = M_1/M_2.$$

Notice that the second rather than the first approximation of λ_b has been given. This is because two terms in the matrix Q_{nm}^{ij} are needed in order to obtain the first approximations of λ_{ij} and k_{Ti} , while with these two terms the second approximation of λ_b is obtained [see Eq. (95a)]. The first approximation of λ_b is

$$[\lambda_b]_1 = 16M_2A/3.$$

In the limit $M \le 1$ $(M_1 \le 1, M_2 \ge 1)$ and $n_1/n_2 = O(1)$, the above transport coefficients coincide with the those calcalculated in Ref. 6 using an expansion of the Boltzmann equation in the small parameter M. (Notice that the coefficient λ_b of this work is $-n_p\lambda_b$ of Ref. 6.) Also, it is easy to verify that $[\mu_{11}]_1 + [\mu_{22}]_1 + 2[\mu_{12}]_1$ coincides with the expression of $[\mu]_1$ given by Chapman and Cowling [Ref. 7, Eq. (9.84,2)] and that $[\lambda_b]_1$ is equal to $kT/nm_1[D_{12}]_1$ of Ref. 7 [Eq. (9.81,1)]. For the thermal conductivity and the thermal diffusion coefficients we have not made the direct comparison of $[\lambda_{11}]_1 + [\lambda_{22}]_1 + 2[\lambda_{12}]_1$ and $[k_{T1}]_1$ + $[k_{T2}]_1$ with $[\lambda]_1$ and $[k_T]_1$ of Chapman and Cowling because of the complexity of the corresponding expressions. However, this check has been made numerically for a number of noble gases binary mixtures with perfect agreement. The first approximation of σ_T is related to the first approximation of D_{12} through $[\sigma_T]_1 = 3k^2 T x_1 x_2 /$ $n(m_1 + m_2)[D_{12}]_1$ [Ref. 7, Eq. (9.81,1)], which coincides with the exact expression for the case of molecules interacting with Maxwellian potentials (i.e., Ref. 3).

With the coefficients E_{nm}^{ij} given above and the bracket integrals tabulated in Ref. 10, Sec. 7.2 [after using the relations (95)], the second approximations of the transport coefficients can be obtained without much additional work by solving Eqs. (93).

V. COMPARISON WITH EXPERIMENTS ON SOUND ABSORPTION IN He-Xe MIXTURES

The two-fluid CE theory previously developed has been recently applied to the acoustic problem, and its results are compared in Ref. 11 with available experimental data 12,13 on absorption and dispersion of sound in He–Xe mixtures $(M \simeq 0.03)$. In this section we present an additional comparison of the two-fluid results and those of the standard Navier–Stokes equations with experimental data of Bowler¹³ on absorption of sound in He–Xe mixtures.

Figures 1(a)-1(g) illustrate the frequency range (Knudsen number range) of validity of the standard Chapman-Enskog theory and that of the present two-fluid theory for disparate-mass mixtures. At low frequencies $(Kn \ll M)$ both theories predict the same values for the absorption (Kohler expressions for low frequencies, e.g., Ref. 11), and the agreement with the experiments is excellent. However, as the frequency increases, the predictions of the standard CE theory become poor for most concentrations, while the two-fluid theory agrees reasonably well with the experiments. The agreement of the two-fluid theory with experiments.

mental data^{12,13} on dispersion is also excellent, as shown in Ref. 11.

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APPENDIX: HYDRODYNAMIC EQUATIONS

From Eqs. (11)–(13),

$$\begin{split} & \frac{\partial \rho_i}{\partial t} + \nabla \cdot (\rho_i \mathbf{U}_i) = 0, \\ & \frac{\partial \rho_i \mathbf{U}_i}{\partial t} + \nabla \cdot \left[\rho_i \mathbf{U}_i \mathbf{U}_i + \mathbf{P}_i \right] = (-1)^{i-1} \lambda, \\ & \frac{\partial \left[\rho_i (e_i + U_i^2/2) \right]}{\partial t} + \nabla \cdot \left[\rho_i \mathbf{U}_i (e_i + U_i^2/2) + \mathbf{P}_i \cdot \mathbf{U}_i + \mathbf{Q}_i \right] \\ & = (-1)^{i-1} \sigma' \quad (i = 1, 2), \end{split}$$

where

$$\rho_i e_i \equiv \frac{1}{2} \mathbf{P}_i : \mathbf{I} \equiv 3p_i/2 \equiv 3n_i k T_i/2.$$

To first order, these equations may be closed in terms of the two fluid hydrodynamic quantities by means of Eqs. (57), (60), (75), (76), and (86):

$$\begin{split} \mathbf{P}_i &= n_i k T_i \mathbf{I} - 2\mu_{i1} \nabla \cdot \mathbf{U}_1 - 2\mu_{i2} \nabla \cdot \mathbf{U}_2, \\ \mathbf{Q}_i &= -\lambda_{i1} \nabla T_1 - \lambda_{i2} \nabla T_2 + nk T k_{Ti} (\mathbf{U}_1 - \mathbf{U}_2), \\ \boldsymbol{\lambda} &= -nk T (k_{T1} \nabla \ln T_1 + k_{T2} \nabla \ln T_2) \\ &- \rho_1 \rho_2 \lambda_b (\mathbf{U}_1 - \mathbf{U}_2) / m_2, \\ \boldsymbol{\sigma}' &= -n^2 \sigma_T (T_1 - T_2). \end{split}$$

The expressions for the transport coefficients μ_{ij} , λ_{ij} , k_{Ti} , λ_b , and σ_T , in the first approximation of the Sonine polynomial expansion, are given at the end of Sec. IV.

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