

Enskog Theory for Polydisperse Granular Mixtures II. Sonine Polynomial Approximation

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(Dated: January 3, 2014)

The linear integral equations defining the Navier-Stokes (NS) transport coefficients for polydisperse granular mixtures of smooth inelastic hard disks or spheres are solved by using the leading terms in a Sonine polynomial expansion. Explicit expressions for all the NS transport coefficients are given in terms of the sizes, masses, compositions, density and restitution coefficients. In addition, the cooling rate is also evaluated to first order in the gradients. The results hold for arbitrary degree of inelasticity and are not limited to specific values of the parameters of the mixture. Finally, a detailed comparison between the derivation of the current theory and previous theories for mixtures is made, with attention paid to the implication of the various treatments employed to date.

I. INTRODUCTION

In the first portion [1] of this two-paper effort on the development of a kinetic-theory-based description of mixtures, a rigorous Chapman-Enskog (CE) expansion of the revised Enskog theory (RET) for an s -component mixture of inelastic hard spheres was carried out to first (Navier-Stokes) order in spatial gradients. The expansion was performed about a homogeneous cooling state (HCS), which is the zeroth-order solution of the kinetic equation for the single-particle velocity distribution function f_i of species i . Unlike previous theories for mixtures which derive from expansions about an elastic base state [2, 3, 4, 5, 6, 7, 8, 9, 10], this CE expansion does not impose any constraints on the level of dissipation. The resulting theory is thus expected to be applicable to a wide range of restitution coefficients. Furthermore, because the derivation used the RET as its starting point (as opposed to the Boltzmann equation as used in Ref. [11]), the results are expected to be applicable to dilute and moderately dense systems. The theory is not expected to be applicable to systems dense enough for ring collisions to play a significant role, since such velocity correlations are not accounted for in the RET.

This formally exact analysis for Navier-Stokes order hydrodynamics, reported in the companion paper, resulted in integral-differential equations for the zeroth-order $f_i^{(0)}$ and first-order $f_i^{(1)}$ distribution functions as well as integral expressions (in terms of $f_i^{(0)}$ and $f_i^{(1)}$) for each of the equations of state (cooling rate and pressure) and the transport coefficients $\{D_{ij}, D_i^T, D_{ij}^F, \eta, \kappa, \lambda, D_{q,ij}, L_{ij}\}$. Of these, only the pressure could be directly evaluated and cast in algebraic expressions of the macroscopic (hydrodynamic) variables. Hence, in this second part of the work, approximate methods are used to obtain algebraic equations for the kinetic and collisional contributions to the cooling rate and transport coefficients. In particular, the equations for $f_i^{(0)}$ have previously been solved [12] via a combination of scaling arguments and an approximation to the distribution function based on leading-order Sonine polynomials. Recent results derived for binary granular mixtures at low-density [11, 13] have shown that the influence of the non-Gaussian (higher-order) corrections of $f_i^{(0)}$ to the transport coefficients is in general negligible, except for quite large values of dissipation and velocities. For this reason, we use leading order in the Sonine polynomial expansion (Maxwellians) at different temperatures to evaluate integrals over the distributions $f_i^{(0)}$. The solutions for the $f_i^{(1)}$ are also found using a truncated polynomial expansion. These forms for $f_i^{(0)}$ and $f_i^{(1)}$ are then used to obtain practical expressions for the cooling rate and transport coefficients. Namely, the resulting, algebraic constitutive equations depend explicitly on the hydrodynamic variables only and not on the distribution functions of the mixture. Collision integrals are reduced to Gaussian forms and evaluated using standard integration techniques; a computer package of symbolic

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calculations (Mathematica) was used to check their analytical evaluation. Wherever possible, limiting values of the equations of state and transport coefficients were verified by comparison with previously published works in the cases of mechanically equivalent particles [14, 15], binary mixtures at low-density [11, 16], and simple shear flow states for binary mixtures at moderate density [17]. All analytic calculations of both papers were performed independently and cross-checked.

Finally, as mentioned in the Introduction of the companion paper, a myriad of treatments has appeared in the literature for the derivation of kinetic-theory-based descriptions of granular mixtures. To help put the current effort in the context of previous contributions, Sec. VI contains a breakdown of the theoretical contributions to date, along with a critical discussion of the ramifications of each treatment.

II. EULER ORDER PARAMETERS

The hydrodynamic equations to first order in the spatial gradients (Euler order) have as unknown (phenomenological) parameters the pressure and the cooling rate,

$$p = p(T, \{n_i\}), \quad \zeta(T, \{n_i\}) = \zeta^{(0)}(T, \{n_i\}) + \zeta_U(T, \{n_i\})\nabla \cdot \mathbf{U}. \quad (2.1)$$

The pressure $p(T, \{n_i\})$ has been evaluated exactly in the previous paper [1] [Eqs. (7.18) and (7.19)] with the result

$$p(T, \{n_i\}) = nT + \frac{\pi^{d/2}}{d\Gamma\left(\frac{d}{2}\right)} \sum_{i=1}^s \sum_{j=1}^s \mu_{ji} \sigma_{ij}^d \chi_{ij}^{(0)} n_i n_j (1 + \alpha_{ij}) T_i, \quad (2.2)$$

where henceforth, for the sake of convenience, we will use the notation $T_i \equiv T_i^{(0)}$ with $T_i^{(0)}$ defined by Eq. (7.5) of Ref. [1]. Detailed forms for the functions $\chi_{ij}^{(0)}$ are discussed in Appendix C. The leading order cooling rate, $\zeta^{(0)}(T, \{n_i\})$, also is given there by Eq. (7.2) as an integral over $f_i^{(0)}$. For reasons just mentioned above, these integrals are performed using the leading order Sonine approximation for $f_i^{(0)}$

$$f_i^{(0)}(\mathbf{V}) \rightarrow f_{i,M}(\mathbf{V}) = n_i \left(\frac{m_i}{2\pi T_i} \right)^{d/2} \exp\left(-\frac{m_i V^2}{2T_i}\right), \quad (2.3)$$

and $\mathbf{V} = \mathbf{v} - \mathbf{U}$ is the peculiar velocity. The result is [14, 16]

$$\begin{aligned} \zeta^{(0)}(T, \{n_i\}) &= \zeta_i^{(0)}(T, \{n_i\}) \\ &= \frac{4\pi^{(d-1)/2}}{d\Gamma\left(\frac{d}{2}\right)} v_0 \sum_{j=1}^s \chi_{ij}^{(0)} n_j \mu_{ji} \sigma_{ij}^{d-1} \left(\frac{\theta_i + \theta_j}{\theta_i \theta_j} \right)^{1/2} (1 + \alpha_{ij}) \left[1 - \frac{\mu_{ji}}{2} (1 + \alpha_{ij}) \frac{\theta_i + \theta_j}{\theta_j} \right], \end{aligned} \quad (2.4)$$

where $n = \sum_i n_i$ is the total density, $v_0(t) = \sqrt{2T/m}$ is a thermal velocity, $m = (\sum_j m_j)/s$, $\mu_{ij} = m_i/(m_i + m_j)$, and $\theta_i = m_i T/m T_i$. The $\zeta_i^{(0)}(T, \{n_i\})$ are species cooling rates, measuring the decrease of kinetic energy of each species. This is an implicit definition of $\zeta^{(0)}(T, \{n_i\})$. The equality of cooling rates for all i gives $s-1$ equations for the species temperatures T_i in terms of T . A final equation is given by the condition that the total kinetic energy is the sum of species energies $nT = \sum_{i=1}^s n_i T_i$. With the species temperatures determined as functions of T and $\{n_i\}$, Eq. (2.4) gives the cooling rate $\zeta^{(0)}(T, \{n_i\})$. In all of the following expressions, it is understood that the $T_i(T, \{n_i\})$ have been determined in this way.

At first order in gradients, there is a contribution to the cooling rate from $\nabla \cdot \mathbf{U}$. The proportionality coefficient ζ_U is a new transport coefficient for granular fluids. Two different contributions can be identified

$$\zeta_U = \zeta^{(1,0)} + \zeta^{(1,1)}. \quad (2.5)$$

The coefficient $\zeta^{(1,0)}$ is given by an integral over $f_i^{(0)}$ and its velocity derivative. It has been evaluated explicitly in Appendix F of Ref. [1] with the result

$$\zeta^{(1,0)} = -\frac{3}{nT} \frac{\pi^{d/2}}{d^2 \Gamma\left(\frac{d}{2}\right)} \sum_{i=1}^s \sum_{j=1}^s n_i n_j \mu_{ji} \sigma_{ij}^d \chi_{ij}^{(0)} (1 - \alpha_{ij}^2) T_i. \quad (2.6)$$

The second term of Eq. (2.5), $\zeta^{(1,1)}$, is given by

$$\zeta^{(1,1)} = \frac{1}{nT} \frac{\pi^{(d-1)/2}}{d\Gamma\left(\frac{d+3}{2}\right)} \sum_{i=1}^s \sum_{j=1}^s \sigma_{ij}^{d-1} \chi_{ij}^{(0)} m_j \mu_{ij} (1 - \alpha_{ij}^2) \int d\mathbf{v}_1 \int d\mathbf{v}_2 g^3 f_i^{(0)}(\mathbf{V}_1) \mathcal{D}_j(\mathbf{V}_2), \quad (2.7)$$

where the unknown functions $\mathcal{D}_i(\mathbf{V})$ are the solutions to the linear integral equations

$$\left(\left(\mathcal{L} + \frac{1}{2} \zeta^{(0)} \right) \mathcal{D} \right)_i + \frac{1}{2} \zeta^{(1,1)}(\mathcal{D}_i) \nabla_{\mathbf{V}} \cdot (\mathbf{V} f_i^{(0)}) = \overline{D}_i. \quad (2.8)$$

Here, $\nabla_{\mathbf{V}} \equiv \partial/\partial\mathbf{V}$ and the linear operator \mathcal{L} is

$$(\mathcal{L}X)_i = \frac{1}{2} \zeta^{(0)} \nabla_{\mathbf{V}} \cdot (\mathbf{V} X_i) + (LX)_i, \quad (2.9)$$

where L is the linearized Enskog collision operator

$$(LX)_i = - \sum_{j=1}^s \left(J_{ij}^{(0)} \left[\mathbf{v}_1 \mid X_i, f_j^{(0)} \right] + J_{ij}^{(0)} \left[\mathbf{v}_1 \mid f_i^{(0)}, X_j \right] \right), \quad (2.10)$$

$$\begin{aligned} J_{ij}^{(0)} [\mathbf{v}_1 \mid X_i, f_j] &\equiv \chi_{ij}^{(0)} (\sigma_{ij}; \{n_i(\mathbf{r}_1, t)\}) \sigma_{ij}^{d-1} \int d\mathbf{v}_2 \int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) \\ &\times [\alpha_{ij}^{-2} X_i(\mathbf{V}_1'') f_j(\mathbf{V}_2'') - X_i(\mathbf{V}_1) f_j(\mathbf{V}_2)], \end{aligned} \quad (2.11)$$

and $\mathbf{v}_1'', \mathbf{v}_2''$ are the precollision velocities associated with $\mathbf{v}_1, \mathbf{v}_2$ for the colliding pair of species i and j . The same linear operator \mathcal{L} appears below in the definitions of all other transport coefficients as well. The inhomogeneity of Eq. (2.8), $\overline{D}_i(T, \{n_i\})$, is given by

$$\begin{aligned} \overline{D}_i &= \left(\frac{1}{d} \left(1 - \frac{p}{nT} \right) - \frac{1}{2} \zeta^{(1,0)} \right) \nabla_{\mathbf{V}} \cdot (\mathbf{V} f_i^{(0)}) - f_i^{(0)} + \sum_{j=1}^s n_j \frac{\partial f_i^{(0)}}{\partial n_j} \\ &+ \frac{1}{d} \sum_{j=1}^s \mathcal{K}_{ij,\beta} [\partial_{V_\beta} f_j^{(0)}], \end{aligned} \quad (2.12)$$

and the operator $\mathcal{K}_{ij,\beta}$ is given by (B1) of Appendix B. An approximate solution to this integral equation is obtained by using $f_i^{(0)}$, p , $\zeta^{(0)}$, and $\zeta^{(1,0)}$ as determined above, and the leading order term in an expansion of $\mathcal{D}_i(\mathbf{V})$ in a complete set of Sonine polynomials

$$\mathcal{D}_i(\mathbf{V}) \rightarrow e_{i,D} f_{i,M}(\mathbf{V}) F_i(\mathbf{V}). \quad (2.13)$$

The lowest order Sonine polynomial contributing in this case is

$$F_i(\mathbf{V}) = \left(\frac{m_i}{2T_i} \right)^2 V^4 - \frac{d+2}{2} \frac{m_i}{T_i} V^2 + \frac{d(d+2)}{4}, \quad (2.14)$$

as determined by the fact that $\mathcal{D}_i(\mathbf{V})$ is a scalar and orthogonal to 1 and V^2 . Finally, $e_{i,D}$ is the projection of \mathcal{D}_i along F_i

$$e_{i,D} = \frac{2}{d(d+2)} \frac{1}{n_i} \int d\mathbf{v} \mathcal{D}_i(\mathbf{V}) F_i(\mathbf{V}). \quad (2.15)$$

Note that the polynomials are defined for each species in terms of the weight factor $f_{i,M}(\mathbf{V})$ characterized by the mass and temperature of that species [11]. The coefficients $e_{i,D}$ are determined by substituting (2.13) into the integral equation (2.8), multiplying by $F_i(\mathbf{V})$, and integrating over \mathbf{V} (this assures that the integral equation is exactly satisfied in the subspace spanned F_i) to get the algebraic equations

$$\sum_{j=1}^s \left(\psi_{ij} - \frac{3}{2} \zeta^{(0)} \delta_{ij} \right) e_{j,D} = \overline{e}_{i,D}. \quad (2.16)$$

The collision frequencies ψ_{ij} are defined by

$$\psi_{ii} = -\frac{2}{d(d+2)} \frac{1}{n_i} \left(\sum_{j=1}^s \int d\mathbf{v} F_i J_{ij}^{(0)} [f_{i,M} F_i, f_j^{(0)}] + \int d\mathbf{v} F_i J_{ii}^{(0)} [f_i^{(0)}, f_{i,M} F_i] \right), \quad (2.17)$$

$$\psi_{ij} = -\frac{2}{d(d+2)} \frac{1}{n_i} \int d\mathbf{v} F_i J_{ij}^{(0)} [f_i^{(0)}, f_{j,M} F_j], \quad (i \neq j). \quad (2.18)$$

The inhomogeneity on the right-hand side of (2.16) is computed in Appendix B with the result

$$\begin{aligned} \bar{e}_{i,D} &= \frac{1}{d} \sum_{j=1}^s \int d\mathbf{v} F_i(\mathbf{V}) \mathcal{K}_{ij,\gamma} [\partial_{V_\gamma} f_j^{(0)}] \\ &= \frac{\pi^{d/2}}{4d\Gamma(\frac{d}{2})} \sum_{j=1}^s n_i n_j \chi_{ij}^{(0)} \sigma_{ij}^d \mu_{ji} (1 + \alpha_{ij}) [8(d+2)(\mu_{ij} - 1) \\ &\quad + 4(13 + 2d + 9\alpha_{ij})\mu_{ji} - 48\mu_{ji}^2 \theta_j^{-1} (\theta_i + \theta_j)(1 + \alpha_{ij})^2 \\ &\quad + 15\mu_{ji}^3 \theta_j^{-2} (\theta_i + \theta_j)^2 (1 + \alpha_{ij})^3]. \end{aligned} \quad (2.19)$$

The explicit form of the collision frequencies ψ_{ij} are displayed in Appendix A, so the coefficients $e_{i,D}$ can be determined by solving the algebraic Eqs. (2.16) with all coefficients determined.

Finally, $\zeta^{(1,1)}$ is given in terms of $e_{i,D}$ by substitution of Eq. (2.13) into (2.7), with the result

$$\zeta^{(1,1)} = e_{i,D} \frac{3\pi^{(d-1)/2}}{4d\Gamma(\frac{d}{2})} \frac{v_0^3}{nT} \sum_{i=1}^s \sum_{j=1}^s \sigma_{ij}^{d-1} \chi_{ij}^{(0)} m_j \mu_{ij} (1 - \alpha_{ij}^2) \theta_i^{-3/2} \theta_j^{1/2} (\theta_i + \theta_j)^{-1/2}. \quad (2.20)$$

III. NAVIER-STOKES ORDER TRANSPORT COEFFICIENTS

The hydrodynamic equations to second order in the spatial gradients have contributions to the mass flux, pressure tensor, and heat flux from the Chapman-Enskog expansion proportional to gradients of the species densities, temperature, and flow velocity and to an external applied force (of the same order of magnitude as the gradients). For fluid symmetry, this leads to $(s-1)(2s+1)$ transport coefficients for the mass flux, the shear and bulk viscosity for the pressure tensor, and $2s^2 + 1$ coefficients for the heat flux. The mass flux coefficients are determined from integral equations in the same way as $\zeta^{(1,1)}$, just described. The pressure tensor and heat fluxes have “kinetic” and “collisional” transfer parts, i.e. of the forms

$$\omega = \omega^k + \omega^c.$$

The contributions ω^k are determined from integral equations like that for $\zeta^{(1,1)}$; the collisional transfer contributions have been reduced in the previous paper to specific integrals over $f_i^{(0)}$ and can be computed explicitly by using the multi-temperature Maxwellians (2.3). The analysis is similar to that for the cooling rate and many of the details are transferred to the Appendices.

A. Mass Flux

To first order in the spatial gradients, the mass flux $\mathbf{j}_{0i}^{(1)}$ is given by

$$\mathbf{j}_{0i}^{(1)} = - \sum_{j=1}^s \frac{m_i m_j n_j}{\rho} D_{ij} \nabla \ln n_j - \rho D_i^T \nabla \ln T - \sum_{j=1}^s D_{ij}^F \mathbf{F}_j, \quad (3.1)$$

where $\rho = \sum_i \rho_i = \sum_i m_i n_i$ is the total mass density of the mixture, D_{ij} are the mutual diffusion coefficients, D_i^T are the thermal diffusion coefficients and D_{ij}^F are the mobility coefficients. These transport coefficients are defined as

$$D_{ij} = -\frac{\rho}{dm_j n_j} \int d\mathbf{v} \mathbf{V} \cdot \mathcal{B}_i^j(\mathbf{V}), \quad (3.2)$$

$$D_i^T = -\frac{m_i}{d\rho} \int d\mathbf{v} \mathbf{V} \cdot \mathcal{A}_i(\mathbf{V}), \quad (3.3)$$

$$D_{ij}^F = -\frac{m_i}{d} \int d\mathbf{v} \mathbf{V} \cdot \mathcal{E}_i^j(\mathbf{V}). \quad (3.4)$$

The functions $\mathcal{A}_i(\mathbf{V})$, $\mathcal{B}_i^j(\mathbf{V})$, and $\mathcal{E}_i^j(\mathbf{V})$ verify the linear integral equations

$$\left(\left(\mathcal{L} - \frac{1}{2} \zeta^{(0)} \right) \mathcal{A} \right)_i = \mathbf{A}_i, \quad (3.5)$$

$$(\mathcal{L} \mathcal{B}^j)_i - n_j \frac{\partial \zeta^{(0)}}{\partial n_j} \mathcal{A}_i = \mathbf{B}_i^j, \quad (3.6)$$

$$\left(\left(\mathcal{L} + \zeta^{(0)} \right) \mathcal{E}^j \right)_i = \mathbf{E}_i^j, \quad (3.7)$$

where the linear operator \mathcal{L} and the inhomogeneous terms \mathbf{A}_i , \mathbf{B}_i^j , and \mathbf{E}_i^j are given by Eqs. (6.16), (6.17), and (6.20) of the preceding paper [1]. The approximate solutions to these equations are obtained in the same manner as that for (2.8). The leading order Sonine polynomial in all of these cases is \mathbf{V} , since all are vectors

$$\mathcal{A}_i(\mathbf{V}) \rightarrow -\frac{\rho}{n_i T_i} D_i^T f_{i,M}(\mathbf{V}) \mathbf{V}, \quad (3.8)$$

$$\mathcal{B}_i^j(\mathbf{V}) \rightarrow -\frac{m_i m_j n_j}{\rho n_i T_i} D_{ij}^F f_{i,M}(\mathbf{V}) \mathbf{V}, \quad (3.9)$$

$$\mathcal{E}_i^j(\mathbf{V}) \rightarrow -\frac{1}{n_i T_i} D_{ij}^F f_{i,M}(\mathbf{V}) \mathbf{V}, \quad (3.10)$$

The coefficients are now projections of \mathcal{A}_i , \mathcal{B}_i^j , and \mathcal{E}_i^j along \mathbf{V} , which are identified in terms of the transport coefficients through Eqs. (3.2)–(3.4). Multiplication of Eqs. (3.5)–(3.7) by $m_i \mathbf{V}$ and integrating over the velocity yields the algebraic equations determining all mass flux transport coefficients

$$\sum_{j=1}^s (\nu_{ij} - \zeta^{(0)} \delta_{ij}) D_j^T = -\frac{1}{d\rho} \int d\mathbf{v} m_i \mathbf{V} \cdot \mathbf{A}_i(\mathbf{V}), \quad (3.11)$$

$$\sum_{\ell=1}^s \left(\nu_{i\ell} - \frac{1}{2} \zeta^{(0)} \delta_{i\ell} \right) \frac{m_\ell}{m_i} D_{\ell j}^T - \frac{\rho^2}{m_i m_j} \frac{\partial \zeta^{(0)}}{\partial n_j} D_i^T = -\frac{1}{d} \frac{\rho}{m_i m_j n_j} \int d\mathbf{v} m_i \mathbf{V} \cdot \mathbf{B}_i^j(\mathbf{V}), \quad (3.12)$$

$$\sum_{\ell=1}^s \left(\nu_{i\ell} + \frac{1}{2} \zeta^{(0)} \delta_{i\ell} \right) \frac{m_\ell}{m_i} D_{\ell j}^F = -\frac{1}{d} \int d\mathbf{v} m_i \mathbf{V} \cdot \mathbf{E}_i^j(\mathbf{V}). \quad (3.13)$$

The new collision frequencies ν_{ij} are

$$\nu_{ii} = -\frac{1}{dn_i T_i} \sum_{j \neq i}^s \int d\mathbf{v} m_i \mathbf{V} \cdot J_{ij}^{(0)} [f_{i,M} \mathbf{V}, f_j^{(0)}], \quad (3.14)$$

$$\nu_{ij} = -\frac{1}{dn_j T_j} \int d\mathbf{v} m_i \mathbf{V} \cdot J_{ij}^{(0)} [f_i^{(0)}, f_{j,M} \mathbf{V}], \quad (i \neq j). \quad (3.15)$$

Note that the self-collision terms of ν_{ii} arising from $J_{ii}^{(0)} [f_{i,M} \mathbf{V}, f_i^{(0)}]$ do not occur in (3.14) since these conserve momentum for species i . The above collision frequencies were already evaluated in the Boltzmann limit (except for

the factors $\chi_{ij}^{(0)}$ [11, 16]. The details will not repeated here and only the results are quoted in Appendix A. The integrals on the right hand side of Eqs. (3.11), (3.12), and (3.13) can be performed using the definitions of \mathbf{A}_i , \mathbf{B}_i^j , and \mathbf{E}_i^j . The result is

$$\int d\mathbf{v} m_i \mathbf{V} \cdot \mathbf{A}_i = dp \frac{\rho_i}{\rho} \left(1 - \frac{\rho n_i T_i}{\rho_i p} \right) + \frac{1}{2} \sum_{j=1}^s \int d\mathbf{v} m_i V_\gamma \mathcal{K}_{ij,\gamma} [\nabla \mathbf{v} \cdot (\mathbf{V} f_j^{(0)})], \quad (3.16)$$

$$\begin{aligned} \int d\mathbf{v} m_i \mathbf{V} \cdot \mathbf{B}_i^j &= -dn_j \frac{\partial}{\partial n_j} (n_i T_i) + d \frac{\rho_i}{\rho} n_j \frac{\partial p}{\partial n_j} \\ &\quad - \sum_{\ell=1}^s \int d\mathbf{v} m_i V_\gamma \mathcal{K}_{i\ell,\gamma} \left[\left(n_j \partial_{n_j} + \frac{1}{2} \left(n_j \partial_{n_j} \ln \chi_{i\ell}^{(0)} + I_{i\ell j} \right) \right) f_\ell^{(0)} \right], \end{aligned} \quad (3.17)$$

$$\int d\mathbf{v} m_i \mathbf{V} \cdot \mathbf{E}_i^j = d \frac{n_i m_i}{m_j} \left(\delta_{ij} - \frac{n_j m_j}{\rho} \right). \quad (3.18)$$

The integrals appearing in Eqs. (3.16) and (3.17) that involve the operator $\mathcal{K}_{ij,\gamma}[X]$ have been evaluated in Appendix B. They are given by Eqs. (A18), (B6), and (B7). With these results, Eqs. (3.11), (3.12), and (3.13) become

$$\sum_{j=1}^s (\nu_{ij} - \zeta^{(0)} \delta_{ij}) D_j^T = -\frac{p \rho_i}{\rho^2} \left(1 - \frac{\rho n_i T_i}{\rho_i p} \right) + \frac{\pi^{d/2}}{d\Gamma(\frac{d}{2})} \frac{n_i}{\rho} \sum_{j=1}^s n_j \mu_{ij} \chi_{ij}^{(0)} \sigma_{ij}^d T_j (1 + \alpha_{ij}), \quad (3.19)$$

$$\begin{aligned} \sum_{\ell=1}^s \left(\nu_{i\ell} - \frac{1}{2} \zeta^{(0)} \delta_{i\ell} \right) \frac{m_\ell}{m_i} D_{\ell j} &= \frac{\rho^2}{m_i m_j} \frac{\partial \zeta^{(0)}}{\partial n_j} D_i^T + \frac{\rho}{m_i m_j} \frac{\partial}{\partial n_j} (n_i T_i) - \frac{n_i}{m_j} \frac{\partial p}{\partial n_j} \\ &\quad + \frac{\pi^{d/2}}{d\Gamma(\frac{d}{2})} \frac{\rho n_i}{m_j} \sum_{\ell=1}^s \chi_{i\ell}^{(0)} \sigma_{i\ell}^d \mu_{\ell i} (1 + \alpha_{i\ell}) \left\{ \left(\frac{T_i}{m_i} + \frac{T_\ell}{m_\ell} \right) \right. \\ &\quad \times \left[\delta_{j\ell} + \frac{1}{2} \frac{n_\ell}{n_j} \left(n_j \frac{\partial}{\partial n_j} \ln \chi_{i\ell}^{(0)} + I_{i\ell j} \right) \right] + \frac{n_\ell T_\ell}{m_\ell} \frac{\partial}{\partial n_j} \ln \gamma_\ell \left. \right\}, \end{aligned} \quad (3.20)$$

$$\sum_{\ell=1}^s \left(\nu_{i\ell} + \frac{1}{2} \zeta^{(0)} \delta_{i\ell} \right) \frac{m_\ell}{m_i} D_{\ell j}^F = -\frac{n_i m_i}{m_j} \left(\delta_{ij} - \frac{n_j m_j}{\rho} \right), \quad (3.21)$$

where $\gamma_i \equiv T_i/T$ is the temperature ratio. An explicit form for $I_{i\ell j}$ is chosen in Appendix C. The solution of the set of algebraic equations (3.19)–(3.21) gives the dependence of the coefficients D_{ij} , D_i^T , and D_{ij}^F on the restitution coefficients α_{ij} and the composition, the density, and the sizes and masses of the constituents of the mixture.

B. Pressure Tensor

The constitutive equation for the pressure tensor $P_{\alpha\beta}^{(1)}$, proportional to the velocity gradients, is

$$P_{\alpha\beta}^{(1)} = -\eta \left(\partial r_\alpha U_\beta + \partial r_\beta U_\alpha - \frac{2}{d} \delta_{\alpha\beta} \nabla \cdot \mathbf{u} \right) - \kappa \delta_{\alpha\beta} \nabla \cdot \mathbf{u}. \quad (3.22)$$

Here, η is the shear viscosity coefficient and κ is the bulk viscosity. The coefficient η has kinetic and collisional contributions while κ only has a collisional contribution κ^c (and so, vanishes for dilute gases)

$$\eta = \eta^k + \eta^c, \quad \kappa = \kappa^c. \quad (3.23)$$

The collisional transfer contributions have been analyzed in the preceding paper [1] [Eqs. (7.21) and (7.22)]. These expressions reduce to those previously derived in the monodisperse case [14, 15], and in the case of binary mixtures of

hard spheres [17]. The integrals over $f_i^{(0)}(\mathbf{V})$ are easily performed in the multi-temperature Maxwellian approximation (2.3) with the results

$$\kappa^c = \frac{\pi^{(d-1)/2}}{d^2 \Gamma(\frac{d}{2})} \sum_{i=1}^s \sum_{j=1}^s \frac{m_i m_j}{m_i + m_j} n_i n_j v_0 \sigma_{ij}^{d+1} \chi_{ij}^{(0)} (1 + \alpha_{ij}) \left(\frac{\theta_i + \theta_j}{\theta_i \theta_j} \right)^{1/2}, \quad (3.24)$$

$$\eta^c = \frac{2\pi^{d/2}}{\Gamma(\frac{d}{2})} \frac{1}{d(d+2)} \sum_{i=1}^s \sum_{j=1}^s n_j \sigma_{ij}^d \chi_{ij}^{(0)} \mu_{ji} (1 + \alpha_{ij}) \eta_i^k + \frac{d}{d+2} \kappa^c. \quad (3.25)$$

1. Kinetic contribution η^k

As noted above, there is no kinetic part of the bulk viscosity, $\kappa^k = 0$, so κ is given entirely by (3.24). The kinetic contribution to the shear viscosity, η^k , is defined by

$$\eta^k = \sum_{i=1}^s \eta_i^k = -\frac{1}{(d-1)(d+2)} \sum_{i=1}^s \int d\mathbf{v} m_i V_\alpha V_\beta \mathcal{C}_{i,\alpha\beta}(\mathbf{V}). \quad (3.26)$$

The second equality identifies the partial contribution η_i^k of the species i to the shear viscosity η^k in terms of $\mathcal{C}_{i,\alpha\beta}(\mathbf{V})$, which is the solution to the integral equation

$$\left(\left(\mathcal{L} + \frac{1}{2} \zeta^{(0)} \right) \mathcal{C}_{\alpha\beta} \right)_i = C_{i,\alpha\beta}, \quad (3.27)$$

where $C_{i,\alpha\beta}$ is given by Eq. (6.18) in the preceding paper [1]. It is symmetric and traceless so the leading Sonine approximation for the function $\mathcal{C}_{i,\alpha\beta}(\mathbf{V})$ is

$$\mathcal{C}_{i,\alpha\beta}(\mathbf{V}) \rightarrow -f_{i,M}(\mathbf{V}) \frac{\eta_i^k}{n_i T_i^2} R_{i,\alpha\beta}(\mathbf{V}), \quad (3.28)$$

where

$$R_{i,\alpha\beta}(\mathbf{V}) = m_i \left(V_\alpha V_\beta - \frac{1}{d} V^2 \delta_{\alpha\beta} \right). \quad (3.29)$$

The partial contributions η_i^k are obtained by multiplying the integral equation (3.27) with $R_{i,\alpha\beta}$ and integrating over the velocity to get the set of equations:

$$\sum_{j=1}^s (\tau_{ij} - \frac{1}{2} \zeta^{(0)} \delta_{ij}) \eta_j^k = n_i T_i - \frac{1}{(d-1)(d+2)} \sum_{j=1}^s \int d\mathbf{v} R_{i,\alpha\beta} \mathcal{K}_{ij,\alpha} [\partial_{V_\beta} f_j^{(0)}], \quad (3.30)$$

The collision frequencies τ_{ii} are

$$\tau_{ii} = -\frac{1}{(d-1)(d+2)} \frac{1}{n_i T_i^2} \left(\sum_{j=1}^s \int d\mathbf{v} R_{i,\alpha\beta} J_{ij}^{(0)} [f_{i,M} R_{i,\alpha\beta}, f_j^{(0)}] + \int d\mathbf{v} R_{i,\alpha\beta} J_{ii}^{(0)} [f_i^{(0)}, f_{i,M} R_{i,\alpha\beta}] \right), \quad (3.31)$$

$$\tau_{ij} = -\frac{1}{(d-1)(d+2)} \frac{1}{n_j T_j^2} \int d\mathbf{v} R_{i,\alpha\beta} J_{ij}^{(0)} [f_i^{(0)}, f_{j,M} R_{j,\alpha\beta}], \quad (i \neq j). \quad (3.32)$$

The explicit forms of τ_{ii} and τ_{ij} are displayed in Appendix A, and the integral appearing on the right hand side of Eq. (3.30) has been evaluated in Appendix B with the result

$$\begin{aligned} \int d\mathbf{v} R_{i,\alpha\beta} \mathcal{K}_{ij,\alpha} [\partial_{V_\beta} f_j^{(0)}] &= -\frac{\pi^{d/2}}{\Gamma(\frac{d}{2})} \frac{d-1}{d} m_i n_i n_j \mu_{ji} \sigma_{ij}^d \chi_{ij}^{(0)} (1 + \alpha_{ij}) \\ &\times \left[\mu_{ji} (3\alpha_{ij} - 1) \left(\frac{T_i}{m_i} + \frac{T_j}{m_j} \right) - 4 \frac{T_i - T_j}{m_i + m_j} \right]. \end{aligned} \quad (3.33)$$

In the case of a three-dimensional system ($d = 3$), Eq. (3.33) reduces to the one previously derived for hard spheres [17]. In addition, for identical particles previous results [12, 15] obtained for monodisperse gases are also recovered.

With the right side of Eqs. (3.30) now determined the algebraic equations

$$\begin{aligned} \sum_{j=1}^s (\tau_{ij} - \frac{1}{2} \zeta^{(0)} \delta_{ij}) \eta_j^k &= n_i T_i + \frac{m_i n_i n_j \mu_{ji}}{d(d+2) \Gamma(\frac{d}{2})} \pi^{d/2} \sigma_{ij}^d \chi_{ij}^{(0)} (1 + \alpha_{ij}) \\ &\times \left[\mu_{ji} (3\alpha_{ij} - 1) \left(\frac{T_i}{m_i} + \frac{T_j}{m_j} \right) - 4 \frac{T_i - T_j}{m_i + m_j} \right], \end{aligned} \quad (3.34)$$

can be solved to determine the partial contributions η_i^k . Their sum then gives the kinetic contribution to the shear viscosity, η^k . Finally, adding this to the collisional transfer contribution of (3.25) gives the total shear viscosity.

C. Heat Flux

The constitutive equation for the heat flux $\mathbf{q}^{(1)}$ has contributions proportional to gradients of the densities and the temperature, and terms proportional to an applied force (taken to have the same order of magnitude as the gradients)

$$\mathbf{q}^{(1)} = - \sum_{i=1}^s \sum_{j=1}^s (T^2 D_{q,ij} \nabla \ln n_j + L_{ij} \mathbf{F}_j) - T \lambda \nabla \ln T, \quad (3.35)$$

where λ is the thermal conductivity coefficient and $D_{q,ij}$ are the Dufour coefficients. As in the case of the shear viscosity, the transport coefficients $D_{q,ij}$, L_{ij} and λ have kinetic and collisional contributions

$$D_{q,ij} = D_{q,ij}^k + D_{q,ij}^c, \quad L_{ij} = L_{ij}^k + L_{ij}^c, \quad \lambda = \lambda^k + \lambda^c. \quad (3.36)$$

The collisional transfer contributions λ^c , $D_{q,ij}^c$, and L_{ij}^c are given in the preceding paper by Eqs. (7.14)–(7.16)

$$\begin{aligned} \lambda^c &= \sum_{i=1}^s \sum_{j=1}^s \frac{1}{8} (1 + \alpha_{ij}) m_j \mu_{ij} \sigma_{ij}^d \chi_{ij}^{(0)} \left\{ 2B_4 (1 - \alpha_{ij}) (\mu_{ij} - \mu_{ji}) n_i \left[\frac{2}{m_j} \lambda_j^k + (d+2) \frac{T_i}{m_i m_j T} \rho D_j^T \right] \right. \\ &\quad \left. + \frac{8B_2}{2+d} n_i \left[\frac{2\mu_{ij}}{m_j} \lambda_j^k - (d+2) \frac{T_i}{m_i m_j T} (2\mu_{ij} - \mu_{ji}) \rho D_j^T \right] - T^{-1} C_{ij}^T \right\}, \end{aligned} \quad (3.37)$$

$$\begin{aligned} D_{q,ij}^c &= \sum_{p=1}^s \frac{1}{8} (1 + \alpha_{ip}) m_p \mu_{ip} \sigma_{ip}^d \chi_{ip}^{(0)} \left\{ 2B_4 (1 - \alpha_{ip}) (\mu_{ip} - \mu_{pi}) \right. \\ &\quad \times n_i \left[\frac{2}{m_p} D_{q,pj}^k + (d+2) \frac{T_i}{T^2} \frac{m_j n_j}{\rho m_i} D_{pj} \right] \\ &\quad \left. + \frac{8B_2}{d+2} n_i \left[\frac{2\mu_{pi}}{m_p} D_{q,pj}^k - (d+2) (2\mu_{ip} - \mu_{pi}) \frac{T_i}{T^2} \frac{n_j m_j}{\rho m_i} D_{pj} \right] - T^{-2} C_{ipj}^T \right\}, \end{aligned} \quad (3.38)$$

$$\begin{aligned} L_{ij}^c &= \sum_{p=1}^s \frac{1}{8} (1 + \alpha_{ip}) m_p \mu_{ip} \sigma_{ip}^d \chi_{ip}^{(0)} \left\{ 2B_4 (1 - \alpha_{ip}) (\mu_{ip} - \mu_{pi}) \right. \\ &\quad \times n_i \left[\frac{2}{m_p} L_{pj}^k + (d+2) \frac{T_i}{m_i m_p} D_{pj}^F \right] \\ &\quad \left. + \frac{8B_2}{d+2} n_i \left[\frac{2\mu_{pi}}{m_p} L_{pj}^k - (d+2) (2\mu_{ip} - \mu_{pi}) \frac{T_i}{m_i m_p} D_{pj}^F \right] \right\}. \end{aligned} \quad (3.39)$$

The constants, B_k , are defined by Eq. (A19) of Appendix A, and

$$\begin{aligned} C_{ij}^T &= -\frac{2B_3}{d} \int d\mathbf{v}_1 \int d\mathbf{v}_2 f_i^{(0)}(\mathbf{V}_1) f_j^{(0)}(\mathbf{V}_2) \left\{ g G_{ij}^2 + g^{-1} (\mathbf{g} \cdot \mathbf{G}_{ij})^2 + (1 + \mu_{ji}) g (\mathbf{g} \cdot \mathbf{G}_{ij}) \right. \\ &\quad \left. + \mu_{ji} \mu_{ij} g^3 + \frac{3}{4} (1 - \alpha_{ij}) (\mu_{ji} - \mu_{ij}) [g (\mathbf{g} \cdot \mathbf{G}_{ij}) + g^3] \right\}, \end{aligned} \quad (3.40)$$

$$C_{ipj}^T = \frac{B_3}{d} \int d\mathbf{v}_1 \int d\mathbf{v}_2 \left[-(1 - \alpha_{ip})(\mu_{ip} - \mu_{pi})g^3 + 4g(\mathbf{g} \cdot \mathbf{G}_{ip}) \right] f_i^{(0)}(\mathbf{V}_1) n_j \partial_{n_j} f_p^{(0)}(\mathbf{V}_2), \quad (3.41)$$

where $\mathbf{G}_{ip} = \mu_{ip}\mathbf{V}_1 + \mu_{pi}\mathbf{V}_2$. In the first Sonine approximation these integrals have the explicit forms

$$\begin{aligned} C_{ij}^T = & -\frac{2\pi^{(d-1)/2}}{d\Gamma\left(\frac{d}{2}\right)} n_i n_j v_0^3 (\theta_i + \theta_j)^{-1/2} (\theta_i \theta_j)^{-3/2} \\ & \times \left\{ 2\beta_{ij}^2 + \theta_i \theta_j + (\theta_i + \theta_j) [(\theta_i + \theta_j)\mu_{ij}\mu_{ji} + \beta_{ij}(1 + \mu_{ji})] \right\} \\ & - \frac{3\pi^{(d-1)/2}}{2d\Gamma\left(\frac{d}{2}\right)} n_i n_j v_0^3 (1 - \alpha_{ij})(\mu_{ji} - \mu_{ij}) \left(\frac{\theta_i + \theta_j}{\theta_i \theta_j} \right)^{3/2} [\mu_{ji} + \beta_{ij}(\theta_i + \theta_j)^{-1}], \end{aligned} \quad (3.42)$$

$$\begin{aligned} C_{ipj}^T = & \frac{4\pi^{(d-1)/2}}{d\Gamma\left(\frac{d}{2}\right)} n_i n_p v_0^3 (\theta_i + \theta_p)^{-1/2} (\theta_i \theta_p)^{-3/2} \{ \delta_{jp} \beta_{ip} (\theta_i + \theta_p) \\ & - \frac{1}{2} \theta_i \theta_p \left[1 + \frac{\mu_{pi}(\theta_i + \theta_p) - 2\beta_{ip}}{\theta_p} \right] \frac{\partial \ln \gamma_p}{\partial \ln n_j} \} \\ & + \frac{\pi^{(d-1)/2}}{d\Gamma\left(\frac{d}{2}\right)} n_i n_p v_0^3 (1 - \alpha_{ip})(\mu_{pi} - \mu_{ip}) \left(\frac{\theta_i + \theta_p}{\theta_i \theta_p} \right)^{3/2} \left(\delta_{jp} + \frac{3}{2} \frac{\theta_i}{\theta_i + \theta_p} \frac{\partial \ln \gamma_p}{\partial \ln n_j} \right), \end{aligned} \quad (3.43)$$

where $\beta_{ip} \equiv \mu_{ip}\theta_p - \mu_{pi}\theta_i$. With these expressions, the collisional contributions to the heat flux are explicitly known. For mechanically equivalent particles, all the above expressions for the collision transfer contributions reduce again to the previous results obtained for monocomponent gases [14, 15].

1. Kinetic contributions

The kinetic parts of the transport coefficients λ^k , $D_{q,ij}^k$, L_{ij}^k are defined respectively as

$$\lambda^k = -\frac{1}{dT} \sum_{i=1}^s \int d\mathbf{v} \frac{m_i}{2} V^2 \mathbf{V} \cdot \mathcal{A}_i(\mathbf{V}), \quad (3.44)$$

$$D_{q,ij}^k = -\frac{1}{dT^2} \int d\mathbf{v} \frac{m_i}{2} V^2 \mathbf{V} \cdot \mathcal{B}_i^j(\mathbf{V}), \quad (3.45)$$

$$L_{ij}^k = -\frac{1}{d} \int d\mathbf{v} \frac{m_i}{2} V^2 \mathbf{V} \cdot \mathcal{E}_i^j(\mathbf{V}). \quad (3.46)$$

where $\mathcal{A}_i(\mathbf{V})$, $\mathcal{B}_i^j(\mathbf{V})$, and $\mathcal{E}_i^j(\mathbf{V})$ are again the solutions to (3.5)–(3.7). The leading Sonine approximations of (3.8)–(3.10) are not adequate to determine the general leading order for these transport coefficients, since the coefficients D_i^T , D_{ij} , and D_{ij}^F vanish for a simple one component fluid. This would imply a vanishing thermal conductivity λ as well. Consequently, it is necessary to include here the next (second order) Sonine polynomial

$$\mathcal{A}_i(\mathbf{V}) \rightarrow f_{i,M}(\mathbf{V}) \left[-\frac{\rho}{n_i T_i} \mathbf{V} D_i^T - \frac{2}{d+2} \frac{T m_i}{n_i T_i^3} \lambda_i \mathbf{S}_i(\mathbf{V}) \right], \quad (3.47)$$

$$\mathcal{B}_i^j(\mathbf{V}) \rightarrow f_{i,M}(\mathbf{V}) \left[-\frac{m_i m_j n_j}{\rho n_i T_i} \mathbf{V} D_{ij} - \frac{2}{d+2} \frac{T^2 m_i}{n_i T_i^3} d_{q,ij} \mathbf{S}_i(\mathbf{V}) \right], \quad (3.48)$$

$$\mathcal{E}_i^j(\mathbf{V}) \rightarrow f_{i,M}(\mathbf{V}) \left[-\frac{1}{n_i T_i} \mathbf{V} D_{ij}^F - \frac{2}{d+2} \frac{m_i}{n_i T_i^3} \ell_{ij} \mathbf{S}_i(\mathbf{V}) \right], \quad (3.49)$$

where the next order polynomial $\mathbf{S}_i(\mathbf{V})$ is

$$\mathbf{S}_i(\mathbf{V}) = \left(\frac{1}{2} m_i V^2 - \frac{d+2}{2} T_i \right) \mathbf{V}. \quad (3.50)$$

In the above equations, it is understood that the transport coefficients D_i^T , D_{ij} , and D_{ij}^F are given by Eqs. (3.19)–(3.21), respectively. The coefficients λ_i , $d_{q,ij}$, and ℓ_{ij} are the projections along \mathbf{S}_i :

$$\lambda_i = -\frac{1}{dT} \int d\mathbf{v} \, \mathbf{S}_i(\mathbf{V}) \cdot \mathcal{A}_i(\mathbf{V}), \quad (3.51)$$

$$d_{q,ij} = -\frac{1}{dT^2} \int d\mathbf{v} \, \mathbf{S}_i(\mathbf{V}) \cdot \mathcal{B}_i^j(\mathbf{V}), \quad (3.52)$$

$$\ell_{ij} = -\frac{1}{d} \int d\mathbf{v} \, \mathbf{S}_i(\mathbf{V}) \cdot \mathcal{E}_i^j(\mathbf{V}). \quad (3.53)$$

In terms of λ_i , $d_{q,ij}$ and ℓ_{ij} , the transport coefficients λ^k , $D_{q,ij}^k$, and L_{ij}^k become

$$\lambda^k = \sum_{i=1}^s \lambda_i + \frac{d+2}{2T} \frac{\rho T_i}{m_i} D_i^T, \quad (3.54)$$

$$D_{q,ij}^k = d_{q,ij} + \frac{d+2}{2T^2} \frac{m_j n_j T_i}{\rho} D_{ij}. \quad (3.55)$$

$$L_{ij}^k = \ell_{ij} + \frac{d+2}{2} \frac{T_i}{m_i} D_{ij}^F. \quad (3.56)$$

Since D_i^T , D_{ij} , and D_{ij}^F are known from the analysis above it remains to determine λ_i , $d_{q,ij}$ and ℓ_{ij} . The algebraic equations determining them are obtained by substituting (3.47)–(3.49) into the integral equations (3.5)–(3.7), multiplying Eq. (3.5) by $\mathbf{S}_i(\mathbf{V})$ and integrating over the velocity. The results are

$$\sum_{j=1}^s (\gamma_{ij} - 2\zeta^{(0)} \delta_{ij}) \lambda_j = \bar{\lambda}_i, \quad (3.57)$$

$$\sum_{\ell=1}^s \left(\gamma_{i\ell} - \frac{3}{2} \zeta^{(0)} \delta_{i\ell} \right) d_{q,\ell j} = \bar{d}_{q,ij} \quad (3.58)$$

$$\sum_{k=1}^s \left(\gamma_{ik} - \frac{1}{2} \zeta^{(0)} \delta_{ik} \right) \ell_{kj} = \bar{\ell}_{ij}, \quad (3.59)$$

with the inhomogeneities given by

$$\begin{aligned} \bar{\lambda}_i = & -\frac{d+2}{2} \frac{\rho}{T_i} \frac{n_i T_i^3}{T m_i} \sum_{j=1}^s \frac{\omega_{ij} - \zeta^{(0)} \delta_{ij}}{n_j T_j} D_j^T + \frac{d(d+2)}{2d} \frac{n_i T_i^2}{m_i T} \\ & - \frac{1}{2Td} \sum_{j=1}^s \int d\mathbf{v} \, S_{i,\beta}(\mathbf{V}) \mathcal{K}_{ij,\beta} [\nabla \mathbf{v} \cdot (\mathbf{V} f_j^{(0)})], \end{aligned} \quad (3.60)$$

$$\begin{aligned} \bar{d}_{q,ij} = & -\frac{d+2}{2} \frac{n_i n_j T_i^3}{m_i T^2} \left(\frac{m_j}{\rho T_i} \sum_{\ell=1}^s m_\ell \frac{\omega_{i\ell} - \zeta^{(0)} \delta_{i\ell}}{n_\ell T_\ell} D_{\ell j} + \frac{\partial \zeta^{(0)}}{\partial n_j} \lambda_i - \frac{1}{T_i} \frac{\partial \ln T_i}{\partial n_j} \right) \\ & + \frac{1}{dT^2} \sum_{\ell=1}^s \int d\mathbf{v} \, S_{i,\gamma} \left(\mathcal{K}_{i\ell,\gamma} [n_j \partial_{n_j} f_\ell^{(0)}] + \frac{1}{2} \left(n_\ell \partial_{n_j} \ln \chi_{i\ell}^{(0)} + I_{i\ell j} \right) \mathcal{K}_{i\ell,\gamma} [f_\ell^{(0)}] \right), \end{aligned} \quad (3.61)$$

$$\bar{\ell}_{ij} = -\frac{d+2}{2} \frac{n_i T_i^2}{m_i} \sum_{k=1}^s \frac{\omega_{ik} - \zeta^{(0)} \delta_{ik}}{n_k T_k} D_{kj}^F. \quad (3.62)$$

Use has been made of the results

$$\int d\mathbf{v} \mathbf{S}_i \cdot \mathbf{A}_i(\mathbf{V}) = -\frac{d(d+2)}{2} \frac{n_i T_i^2}{m_i} + \frac{1}{2} \sum_{j=1}^s \int d\mathbf{v} S_{i,\beta}(\mathbf{V}) \mathcal{K}_{ij,\beta} [\nabla_{\mathbf{V}} \cdot (\mathbf{V} f_j^{(0)})], \quad (3.63)$$

$$\begin{aligned} \int d\mathbf{v} \mathbf{S}_i \cdot \mathbf{B}_i^j(\mathbf{V}) &= -\frac{d(d+2)}{2} \frac{n_i n_j T_i}{m_i} \frac{\partial T_i}{\partial n_j} \\ &\quad - \sum_{\ell=1}^s \int d\mathbf{v} S_{i,\beta}(\mathbf{V}) \mathcal{K}_{i\ell,\beta} \left[\left(n_j \partial_{n_j} + \frac{1}{2} \left(n_\ell \frac{\partial \ln \chi_{i\ell}^{(0)}}{\partial n_j} + I_{i\ell j} \right) \right) f_\ell^{(0)} \right], \end{aligned} \quad (3.64)$$

$$\int d\mathbf{v} \mathbf{S}_i(\mathbf{V}) \cdot \mathbf{E}_i^j(\mathbf{V}) = 0. \quad (3.65)$$

The collision frequencies introduced here are

$$\gamma_{ii} = -\frac{2}{d(d+2)} \frac{m_i}{n_i T_i^3} \left(\sum_{j=1}^s \int d\mathbf{v} \mathbf{S}_i \cdot J_{ij}^{(0)} [f_{i,M} \mathbf{S}_i, f_j^{(0)}] + \int d\mathbf{v} \mathbf{S}_i \cdot J_{ii}^{(0)} [f_i^{(0)}, f_{i,M} \mathbf{S}_i] \right), \quad (3.66)$$

$$\gamma_{ij} = -\frac{2}{d(d+2)} \frac{m_j}{n_j T_j^3} \int d\mathbf{v} \mathbf{S}_i \cdot J_{ij}^{(0)} [f_i^{(0)}, f_{j,M} \mathbf{S}_j], \quad (i \neq j). \quad (3.67)$$

$$\omega_{ii} = -\frac{2}{d(d+2)} \frac{m_i}{n_i T_i^2} \left(\sum_{j=1}^s \int d\mathbf{v} \mathbf{S}_i \cdot J_{ij}^{(0)} [f_{i,M} \mathbf{V}, f_j^{(0)}] + \int d\mathbf{v} \mathbf{S}_i \cdot J_{ii}^{(0)} [f_i^{(0)}, f_{i,M} \mathbf{V}] \right), \quad (3.68)$$

$$\omega_{ij} = -\frac{2}{d(d+2)} \frac{m_i}{n_i T_i^2} \sum_{j=1}^s \int d\mathbf{v} \mathbf{S}_i \cdot J_{ij}^{(0)} [f_i^{(0)}, f_{j,M} \mathbf{V}]. \quad (i \neq j). \quad (3.69)$$

The explicit expressions for the above collision frequencies are also displayed in Appendix A.

The inhomogeneous terms (3.60) and (3.61) involve integrals over the operator $\mathcal{K}_{ij,\gamma}$. These are evaluated in Appendix B

$$\begin{aligned} \int d\mathbf{v} S_{i,\gamma}(\mathbf{V}) \mathcal{K}_{ij,\gamma} [\nabla_{\mathbf{V}} \cdot (\mathbf{V} f_j^{(0)})] &= -\frac{\pi^{d/2}}{\Gamma(\frac{d}{2})} n_i n_j \mu_{ij} \chi_{ij}^{(0)} \sigma_{ij}^d T_j (1 + \alpha_{ij}) \left\{ \frac{T_i}{m_i} [(d+2)(\mu_{ij}^2 - 1) \right. \\ &\quad \left. + (2d-5-9\alpha_{ij})\mu_{ij}\mu_{ji} + (d-1+3\alpha_{ij}+6\alpha_{ij}^2)\mu_{ji}^2] + 6\frac{T_j}{m_j} \mu_{ji}^2 (1 + \alpha_{ij})^2 \right\}, \end{aligned} \quad (3.70)$$

$$\begin{aligned} \int d\mathbf{v} S_{i,\gamma}(\mathbf{V}) \mathcal{K}_{ij,\gamma} [f_j^{(0)}] &= \frac{\pi^{d/2}}{2\Gamma(\frac{d}{2})} m_i n_i n_j \chi_{ij}^{(0)} \sigma_{ij}^d \mu_{ji} (1 + \alpha_{ij}) \\ &\quad \times \left\{ [(d+8)\mu_{ij}^2 + (7+2d-9\alpha_{ij})\mu_{ij}\mu_{ji} + (2+d+3\alpha_{ij}^2-3\alpha_{ij})\mu_{ji}^2] \frac{T_i^2}{m_i^2} \right. \\ &\quad \left. + 3\mu_{ji}^2 (1 + \alpha_{ij})^2 \frac{T_j^2}{m_j^2} + [(d+2)\mu_{ij}^2 + (2d-5-9\alpha_{ij})\mu_{ij}\mu_{ji} + (d-1+3\alpha_{ij}+6\alpha_{ij}^2)\mu_{ji}^2] \right. \\ &\quad \left. \times \frac{T_i T_j}{m_i m_j} - (d+2) \left(\frac{T_i}{m_i} + \frac{T_j}{m_j} \right) \frac{T_i}{m_i} \right\}, \end{aligned} \quad (3.71)$$

$$\int d\mathbf{v} S_{i,\gamma}(\mathbf{V}) \mathcal{K}_{i\ell,\gamma} [n_j \partial_{n_j} f_\ell^{(0)}] = \int d\mathbf{v} S_{i,\gamma}(\mathbf{V}) \left(\mathcal{K}_{i\ell,\gamma} [\delta_{\ell j} f_\ell^{(0)}] - \frac{1}{2} \mathcal{K}_{i\ell,\gamma} [\nabla_{\mathbf{V}} \cdot (\mathbf{V} f_\ell^{(0)})] \frac{\partial \ln \gamma_\ell}{\partial \ln n_j} \right). \quad (3.72)$$

This completely determines the parameters of the integral equations (3.57)–(3.59). Their solution determines λ_i , $d_{q,ij}$ and ℓ_{ij} , and hence the transport coefficients λ^k , $D_{q,ij}^k$, and L_{ij}^k through Eqs. (3.54)–(3.56). In the special case of mechanically equivalent particles, this description reduces to those previously obtained for a monodisperse gas [14, 15].

IV. SUMMARY AND DISCUSSION

First, a brief summary of the results obtained in the two papers presented here is given. The revised Enskog kinetic equation was used as the basis for deriving exact balance equations for the hydrodynamic fields. The pressure, cooling rate, mass flux, momentum flux, and energy flux were determined from the kinetic theory as linear combinations of the gradients of these fields and an applied force. These are the constitutive equations that convert the exact balance equations into closed equations for the hydrodynamic fields (Navier-Stokes equations, at this order of gradients). The coefficients in these equations are the pressure, leading order cooling rate, and the transport coefficients. Formally exact expressions for these coefficients were obtained in the first paper, expressed as integrals over solutions to integral equations via the Chapman-Enskog method for constructing a solution to the kinetic equation. In the present paper, these exact expressions were evaluated using approximate solutions to the integral equations. The approximation consists of expanding the unknown functions in a complete set of polynomials, and truncating that expansion to convert the integral equations to algebraic equations that can be solved by standard matrix methods. Here, the detailed forms for those equations or their solutions have been given in terms of the hydrodynamic fields and other parameters of the problem. The result is a complete description of the hydrodynamic equations to Navier-Stokes order with all parameters determined explicitly from the theory. Due to the complexity of the analysis it may be useful to give the specific location of the final results. The pressure is given by Eq. (2.2) and the lowest order cooling rate by Eq. (2.4). The equivalence of all species cooling rates determine the species temperatures as well (to this order in the gradients). The only transport coefficient for the cooling rate to first order is given by (2.20) and the solution to the algebraic equations (2.16). The transport coefficients characterizing the mass flux are the solutions to the algebraic equations (3.19)–(3.21). The momentum flux has two transport coefficients, the shear and bulk viscosities. These are given by Eqs. (3.23)–(3.26) and the solutions to the algebraic equations (3.34). Finally, the transport coefficients for the heat flux are given by Eqs. (3.36)–(3.39), (3.54)–(3.56), and the solutions to the algebraic equations (3.57)–(3.59). Detailed forms for the collision frequencies and other input functions are presented in the Appendices. These explicit “constitutive relations” together with the exact macroscopic balance equations for species mass densities, flow velocity, and temperature (Eqs. (4.12), (4.14), and (4.24) of the previous paper) complete the practical description of Navier-Stokes hydrodynamics for a moderately dense, multi-component granular mixture based on the revised Enskog kinetic equation.

In the remainder of this Discussion the relationship to several previous works on kinetic-theory-based descriptions of granular mixtures in the literature is considered. These contributions, which are abbreviated according to the initials of authors and the final two digits of the publication year, are listed in Tables I and II. To help put the current effort in the context of previous works, Tables I and II also list the applicability of each contribution (dimensionality and number of species), the starting kinetic equation (Boltzmann or Enskog), and the specific mechanics and assumptions used in the derivation process: the standard Enskog theory (SET) vs. the revised Enskog theory (RET), pair correlation function $\chi_{ij}^{(0)}$, solution method, order and base state of expansion, single particle velocity distribution function [Maxwellian (M) and non-Maxwellian (nM)], energy distribution [energy equipartition (EE) and non-energy equipartition (nEE)], and hydrodynamic variables. In the subsections below, each of these various treatments are detailed and their implications are discussed.

Dimensionality

The number contained in Table I refers to the dimensionality of the particles. Namely, 2D refers to circular particles (disks) that are constrained to motion in a plane, while 3D refers to spherical particles that can move in all three dimensions. Although systems of practical importance are three-dimensional, two-dimensional theories have appeared in the literature for purposes of comparing with molecular-dynamics (MD) simulations. Specifically, early MD simulations were performed in two dimensions due to computational constraints, whereas three-dimensional simulations are now common. The WA99 theory is applicable to 2D only, the JM87 and GHD theories are derived for both 2D and 3D, while all remaining theories JM89, Z95, H06, GD02, R03, IA05, and S06 are for 3D systems.

Number of Species

Most mixture theories to date have been targeted at binary mixtures, in which the two species can differ in size, mass, restitution coefficient, and density (JM87, JM89, WA99, H01, GD02, R03, and S06). Some recent theories have been derived for a more general system of s distinct species (Z95, IA05, and GHD).

Kinetic Theory

Two kinetic theories have been used in the development of hydrodynamic equations for mixtures, namely the Boltzmann equation and the Enskog equation for hard spheres. The difference between these two equations stems

from the treatment of the two-particle distribution function f_{ij} . For the Boltzmann equation, f_{ij} is assumed equal to the product of the two single-particle distribution functions ($f_{ij} = f_i f_j$). This lack of spatial and pre-collisional velocity correlations between the two particles restricts the Boltzmann equation to dilute systems. The Enskog equation, on the other hand, accounts for positional correlations (but not pre-collisional velocity correlations) via the equilibrium pair correlation function at contact χ_{ij} , namely $f_{ij} = \chi_{ij} f_i f_j$ (the Enskog approximation). More specifically, χ_{ij} accounts for excluded volume effects encountered in denser flows, and thus the corresponding Enskog kinetic theory is applicable to moderately dense flows. The Enskog approximation is expected to deteriorate at higher densities as ring collisions and their associated velocity correlations become important. The prevalence of such correlations has also been found to depend on the restitution coefficient; namely, velocity correlations have been observed to increase as the restitution coefficient decreases. Thus, theories based on the Enskog equation are applicable to both dilute and moderately dense flows (JM87, JM89, Z95, WA99, H01, R03, IA05, and GHD), while theories using the Boltzmann equation as a starting point are restricted to dilute flows (GD02 and S06).

From a practical perspective, the upper limit of concentration that a given kinetic theory should be applied to depends on the desired level of accuracy. As a quick gauge to the range of validity of the Boltzmann equation, the limiting case of a monodisperse system is considered using the theories of Garzó and Dufty [14] and Lutsko [15], which are based on the Enskog equation and thus are applicable to both dilute and dense flows. According to these theories, the dense collisional contributions to the pressure (absent in the Boltzmann equation) are 4% and 18% for solids volume fractions of 0.01 and 0.05, respectively, at $\alpha = 0.9$. A similar estimate of the range of validity of the Enskog equation is not available, since the impact of velocity correlations on granular hydrodynamics has not been extensively studied.

SET vs. RET

For those contributions listed in Table I that employ the Enskog equation, two different approaches are possible – the SET and the RET. As mentioned previously, the difference between SET and RET traces to the choice of the pair correlation function, $\chi_{ij}^{(0)}$. In SET, $\chi_{ij}^{(0)}$ is a *function* of concentration (i.e., depends on local value only) at a single position of interest, whereas for RET $\chi_{ij}^{(0)}$ is treated as a *functional* of concentration (i.e., depends on the local value and its gradient) at the two particle centers. In SET, the location (e.g., midpoint) at which to evaluate $\chi_{ij}^{(0)}$ for mixtures is unclear [18]. Regardless of the choice, however, the resulting diffusion force is found to be inconsistent with irreversible thermodynamics, unlike in RET [19].

The implications of the SET vs. RET treatment on the resulting theory depends on the type system being examined. For the case of monodisperse systems, SET and RET lead to the same Navier-Stokes transport coefficients [19, 20, 21, 22] but different inhomogeneous equilibrium states [19, 20, 21]. For mixtures, however, different Navier-Stokes transport coefficients are obtained from SET and RET. More specifically, although the fluxes appearing in the momentum and energy balances are the same for SET and RET [\mathbf{q} and $P_{\gamma\beta}$ in Eqs. (4.13) and (4.14) in the companion paper], the diffusion flux [\mathbf{j}_{0i} in Eq. (4.12) in companion paper] takes on different forms [23]. Although the quantitative impact of such differences on segregation predictions has not been investigated in detail, it is clear that RET is the appropriate approach since it is consistent with irreversible thermodynamics. The Enskog-based theories of JM89, Z95, WA99, and GHD use RET, whereas JM87, H01, R03, and IA05 utilize SET.

Pair Correlation Function

As noted in Table I, various forms of the pair correlation function $\chi_{ij}^{(0)}$ have been used in conjunction with mixtures. Its explicit forms can be found in the references displayed in the table.

Solution Method

With the exception of Zamankhan [4], all of the efforts to date on mixtures have implemented a Chapman-Enskog (CE) expansion to solve the kinetic equation. By definition, the CE expansion involves a perturbative expansion about low Knudsen numbers (where Kn is defined as the ratio of the mean free path to the characteristic length of the mean-flow gradients) or “small gradients,” and thus is not applicable to systems in which free-molecular (non-continuum) effects play a non-negligible role. The Grad moment method, as was employed by Zamankhan [4] does not contain similar restrictions, though the derivation is necessarily more complex and thus has not been performed without resorting to other simplifying assumptions (e.g., equipartition of energy).

Order of Chapman-Enskog Expansion

For each of the entries in Table I that employ the CE expansion to solve the kinetic theory equation, all expansions are carried out to first order in spatial gradients – i.e., to the Navier Stokes (NS) order. Nonetheless, evidence of higher-order effects has been noted in a range of granular flows, and Burnett-order effects (second order in spatial gradients) in particular have been shown to be linked to the anisotropy in the stress tensor [24]. Examples of systems in which significant higher-order effects have been identified include the upper region of an open-ended, vibro-fluidized bed [25, 26], the vanishing heap of a vibrated, granular material [27], dilute flow around an immersed cylinder [28], simple shear flow [29], the Knudsen layer adjacent to a thermal (energy-providing) boundary [30], and in the continuum interior of a thermally-driven granular gas [31]. For the case of monodisperse systems, several theories [24, 32, 33, 34] and boundary conditions [25, 26, 35] have been developed that account for such higher order effects in a limited class of systems (dilute, sheared flow, open boundary, etc.). Analogous work has not been reported for mixtures [36, 37], however, and thus the incorporation of higher-order effects does not serve as a differentiator between those theories listed in Table II that employ the CE expansion. Nonetheless, the possible presence of such higher-order effects should be kept in mind when comparing these theories with experiments and simulations for purposes of validation.

Base State of Chapman-Enskog Expansion

As described in the companion paper, hydrodynamics results from a “normal solution” to the kinetic equation, whose space and time dependence occurs only through the hydrodynamic fields. The CE expansion is a systematic method for constructing this normal solution as an expansion in powers of the Knudsen number, or spatial gradients of the fields. At zeroth order in these fields, the kinetic theory determines the form of the distribution function to be the “local equilibrium” Maxwellian for molecular fluids. However, in the presence of dissipation the kinetic theory requires a different solution at zeroth order, the “local homogeneous cooling” (HCS) distribution. The HCS distribution agrees with the local Maxwellian only for $\alpha = 1$. It is possible to make an expansion in both Knudsen number and $(1 - \alpha)$, in which case the lowest order term is indeed the Maxwellian. Such a double expansion is necessarily limited to asymptotically weak dissipation. The theories of JM87, JM89, WA99, H01, R03, IA05, and S06 are of this type with the corresponding implicit limitation. The theories of GD02 and GHD, on the other hand, expand only in the Knudsen number, with HCS as the leading order solution and hence no a priori limitation on the degree of dissipation. Quantitatively, the difference between the two types of expansions has been examined in monodisperse systems via a comparison of the dissipation rate obtained in MD simulations [31]. As an example, at a volume fraction of 0.3 and $\alpha = 0.75$, an error of 23% is obtained when using a theory [38] based on an expansion about $\alpha = 1$, while an error of 7% is obtained when using a theory [14] based on an expansion about the HCS. For both theories, the level of mismatch is found to increase with concentration and dissipation levels.

Order of Sonine polynomial expansion

To make the analytical evaluation of the collision integrals possible, a truncated Sonine polynomial expansion is employed. All theories listed in Table I employ the lowest, non-zero order of the polynomial expansion (leading term), except for the weak dissipation theory of S06 which carries out the expansion to third order. Nonetheless, the transport coefficients of S06 agree with those of GD02 in the common domain of validity, namely in the nearly elastic limit [10].

The accuracy of a given Sonine polynomial approximation can be tested via comparison with discrete simulation Monte Carlo (DSMC) results. DSMC provides a numerical solution of the starting kinetic (Boltzmann or Enskog) equation for a specific system, and thus provides a check for the existence of a normal solution, the order of the gradient expansion (e.g., Navier Stokes), and the truncated Sonine polynomial. For dilute granular mixtures, good agreement between DSMC and the theory of Garzó and Dufty [11] was found for the shear viscosity, even for strong dissipation [39]. This agreement for the shear viscosity is also present for moderately dense systems [17]. A similar comparison was carried out for the diffusion coefficient for a system with an impurity [40]. The agreement is again excellent, except when the size or mass disparity is large, in which case the second Sonine approximation leads to a significant improvement over the first Sonine approximation.

Single Particle Velocity Distribution

In an effort to simplify the evaluation of collision integrals, some previous works have assumed that the single particle velocity distribution function is Maxwellian (M). Strictly speaking, such an assumption is valid only for perfectly elastic spheres in equilibrium [41]. Numerous experimental, theoretical, and simulation studies of inelastic, monodisperse systems have indicated that the distribution function departs from Maxwellian [42, 43, 44, 45, 46, 47, 48, 49]. For the case of perfectly elastic mixtures, an estimate of the impact of this effect is given by Willits and Arnarson [6], who compare the shear viscosity predictions of two theories with that of MD simulations. For this particular system, the

only effective difference between the two theories is that one contains a Maxwellian assumption and the other does not. For disks with a diameter ratio of 1.25 and over a range of solids fractions from 0.05 to 0.4, the non-Maxwellian (nM) theory exhibits very good agreement with the simulations. The Maxwellian-based predictions, on the other hand, are significantly lower in value since the Maxwellian assumption precludes the kinetic contribution to the shear stress and simplifies its collisional counterpart. Although similar studies on the impact of non-Maxwellian effects on other constitutive quantities are not available, it is clear that a non-Maxwellian treatment is critical for the accurate prediction of shear stress. Of the mixture theories presented in Table I, those of JM87, H01, and R03 are based on a Maxwellian assumption, while those of JM89, WA99, Z95, GD02, S06, and GHD incorporate non-Maxwellian effects. The theory of IA05 takes a hybrid approach, in which collision integrals involving unlike particles are evaluated based on Maxwellian distributions, whereas collisions integrals involving like particles account for non-Maxwellian effects.

Energy Distribution

Similar to the assumption of a Maxwellian velocity distribution, the assumption of an equipartition of energy (EE) has also been made periodically in an effort to simplify the evaluation of collision integrals. Again, an equipartition of energy between unlike particles is only expected for a perfectly elastic system in equilibrium. Numerous theoretical [12, 39, 50], simulation [51, 52, 53, 54, 55, 56], and experimental [57, 58, 59] studies of granular materials provide evidence that an equipartition of energy does not exist, and that the level of non-equipartition of energy (nEE) increases as the restitution coefficient decreases (dissipation increases) and as the mass ratio gets further from unity. The impact of such non-equipartition has recently been evaluated in the context of species segregation. In particular, Galvin, Dahl and Hrenya [60] have found that the driving forces for species segregation that arise from non-equipartition are significant over a moderate range of parameters for the case of a thermally-driven system. Furthermore, for the case of an intruder particle in the presence of gravity, both Brey et al.[61], Garzó [62], and Yoon and Jenkins[63] have found that the direction of species segregation may reverse due to non-equipartition effects. Together, these studies indicate the importance of non-equipartition in a variety of systems. The mixture theories of JM87, H01, GD02, R03, IA05, and GHD include the effects of non-equipartition, while those of JM89, WA99, Z95, and S06 do not account for non-equipartition.

Hydrodynamic Variables

The appropriate choice of hydrodynamic variables to include in a hydrodynamic theory depends on the timescale associated with a given variable [64]. First consider the case of a molecular gas, in which two timescales are relevant – the “kinetic” time scale and the “hydrodynamic” time scale. The kinetic time scale is “fast” and representative of the time between collisions (i.e., mean free time). The hydrodynamic time scale is “slow” and is set by the gradients in the system; this is the timescale over which macroscopic variables change.

It is important to note that the velocity distribution function f_i (and thus the Boltzmann and Enskog equations) carries information with it on the kinetic time scale. However, the macroscopic variables (n_i , U , and T for the case of a molecular gas) correspond to velocity moments of f_i that are *collisional invariants* – i.e., n_i , U , and T are conserved upon collision. As a result, they will stay constant over a time on the order of a mean free path, and vary in time over the much longer hydrodynamic time scale. It is this property which defines the appropriate (minimum set of) hydrodynamic variables needed to describe a system.

To illustrate the above point, consider the time evolution of mixture temperature (T) and species temperature (T_i), again for the case of a molecular gas with more than one species present:

$$\begin{aligned}\frac{DT}{Dt} &= (\text{energy flux}) \\ \frac{DT_i}{Dt} &= (\text{energy flux}) + \left(\begin{array}{c} \text{collisional exchange (source/sink)} \\ \text{between unlike species} \end{array} \right)\end{aligned}$$

Note that T is a collisional invariant (the total energy is conserved upon collision and thus no “source” term is present), but that T_i is not a collisional invariant since the energy associated with a given species can change upon collision with another species. Also note that the time scale of the flux term is set by gradients (e.g., Fourier’s law), whereas the time scale of the source term is collisional in nature and thus relatively “fast.” Hence, the mixture temperature T is characterized by one (slow) timescale and the species temperature is characterized by two (slow and fast) timescales. The corresponding physical picture is the following: each species is expected to have a rapid relaxation after a few collisions to a distribution near local equilibrium (local mixture temperature), and then all temperatures evolve according to the slow hydrodynamic mode. Thus, the relevant hydrodynamic (macroscopic) variable is T since it is associated with a conserved quantity; all other variables (T_i) are enslaved to it. Thus, solving a separate balance for T_i is superfluous – indeed this is not done for molecular gases.

Next, consider a granular system in which the particles engage in dissipative collisions. Three timescales need to be considered, as illustrated by the balances below:

$$\begin{aligned}\frac{DT}{Dt} &= (\text{energy flux}) + \left(\begin{array}{c} \text{inelastic} \\ \text{dissipation} \\ (\text{sink}) \end{array} \right) \\ \frac{DT_i}{dt} &= (\text{energy flux}) + \left(\begin{array}{c} \text{inelastic} \\ \text{dissipation} \\ (\text{sink}) \end{array} \right) + \left(\begin{array}{c} \text{collisional exchange (source/sink)} \\ \text{between unlike species} \end{array} \right)\end{aligned}$$

As with the molecular systems, the energy flux is characterized by a slow (hydrodynamic) time scale and the collisional exchange between species is characterized by a fast (kinetic) time scale. In the granular system, inelastic dissipation also occurs, and its corresponding time scale is not as obvious. More specifically, the dissipation rate depends not only on the collisional frequency (kinetic time scale), but also on the value of restitution coefficient. As α approaches unity, the dissipation rate goes to zero, and the time scale becomes large (hydrodynamic time scale). In other words, it is unclear *a priori* whether this term is characterized by a fast or slow timescale.

The previous observation leads to the question: for moderate (smaller) values of α , does the time scale become fast enough to approach the kinetic time scale or is there still a separation of time scales? If the latter is assumed (that the time scale associated with inelastic dissipation is much longer than that of the kinetic time scale), Garzó and Dufty [12] have shown that in a HCS two-component system, (i) the cooling rates associated with mixture and species temperatures are equal and (ii) the ratio of species temperature (T_1/T_2) remains constant – i.e., all temperatures decay at the same rate but their ratios remain constant. Further note that T_1/T_2 is not equal to one for unlike particles – i.e., a non-equipartition of energy is predicted. To test this assumption of the separation of timescales, MD simulations of a binary mixture in HCS were performed by Dahl et al. [54]. The simulation results indicate that a constant value of T_1/T_2 is achieved after only a few collisions, and this behavior was confirmed over a wide range of parameters.

As a result of the aforementioned finding that the timescale associated with inelastic dissipation is hydrodynamic (slow) in nature, the physical picture is analogous to that of a molecular system. Namely, each species has a rapid relaxation to a local “equilibrium” state (now characterized by a constant value of non-equipartition) – HCS – and is then enslaved to slow (hydrodynamic) evolution. Thus, the relevant hydrodynamic variables are the same – n_i , \mathbf{U} , and T – and the balance of additional variables (such as T_i) would be superfluous.

The practical implications of using n_i , \mathbf{U} , and T as the hydrodynamic variables in granular flows are twofold. First, only a single balance for T is needed, instead of separate balance for each T_i . The reduction in the number of governing equations is expected to lead to a considerable decrease in computational overhead. Second, the level of non-equipartition is in a state of local “equilibrium” due to the fast time time scale of T_i , and thus T_1/T_2 depends on local values of flow field variables and particle properties (mixture composition, α , etc.). Hence, even though a non-equipartition of energy is indeed present, it does not appear explicitly in the transport coefficients. Instead, its dependency on the flow field variables is incorporated into the transport coefficients – i.e., the effect of non-equipartition is *implicitly* contained in the transport coefficients. It can be solved for explicitly once the flow field variables have been solved for using, for example, the relation derived by Garzó and Dufty [12].

Of the non-equipartition theories listed in Table II, GD02 and GHD use T as the hydrodynamic variable for the granular energy field, whereas the other non-equipartition theories use T_i (JM87, H01, R03, IA05). The choice of hydrodynamic variable(s) is a non-issue for theories which invoke an equipartition assumption (JM89, Z95, WA99, S06) since they inherently assume $T = T_i$ and thus use T as the energy variable. (It is worthwhile to note that S06 defines temperature differently than others, namely $T_S = 3T$. Furthermore, IA06 uses a species temperature $T_{i,IA}$, which is defined in terms of velocity fluctuations relative the mean species velocity, rather than the mass-averaged velocity used for T .)

Recapitulation

Unlike previous Enskog-based (dense) theories, the current effort is based on an expansion about the homogeneous cooling state and employs n_i , \mathbf{U} , and T as the hydrodynamic variables. The former extends the range of validity to strong dissipation levels while the latter results in fewer balance equations, thereby reducing the computational cost. This approach was first put forth by Garzó and Dufty [11] who instead used the Boltzmann equation as their starting kinetic equation; the current work thus extends the domain of applicability from dilute to moderately dense flows.

TABLE I: Hydrodynamic Descriptions of Granular Mixtures

References	Abbrev.	Dimension	Number of Species	Kinetic Theory	SET vs. RET	Pair Correlation Function $\chi_{ij}^{(0)}$
[2]	JM87	2D and 3D	2	Enskog	SET	2D [2], 3D [65]
[3] and [5]	JM89	3D	2	Enskog	RET	3D [65]
[4]	Z95	3D	s	Enskog	RET	[66]
[6] and [67]	WA99	2D	2	Enskog	RET	2D [2]
[7]	H01	3D	2	Enskog	RET	[65]
[11]	GD02	3D	2	Boltzmann	–	–
[8]	R03	3D	2	Enskog	SET	[68]
[9]	IA05	3D	s	Enskog	SET	[69]
[10]	S06	3D	2	Boltzmann	–	–
current work	GHD	2D and 3D	s	Enskog	RET	2D [70], 3D [71]

TABLE II: Hydrodynamic Descriptions of Granular Mixtures

Ref.	Solution Method	Chapman-Enskog Expansion Order	Chapman-Enskog Expansion Base State	Sonine Expansion Order	Single Particle Velocity Distribution	Energy Distributon	Hydro-dynamic Variables
JM87	CE	NS	$\alpha = 1$	1st	M	nEE	n_i, \mathbf{U}_i, T_i
JM89	CE	NS	$\alpha = 1$	1st	nM	EE	n_i, \mathbf{U}_i, T
Z95	Grad's method	–	–	–	nM	EE	13 moments
WA99	CE	NS	$\alpha = 1$	1st	nM	EE	n_i, \mathbf{U}_i, T
H01	CE	NS	$\alpha = 1$	1st	M	nEE	n_i, \mathbf{U}_i, T_i
GD02	CE	NS	HCS	1st	nM	nEE	n_i, \mathbf{U}_i, T
R03	CE	NS	$\alpha = 1$	1st	M	nEE	n_i, \mathbf{U}_i, T_i
IA05	CE	NS	$\alpha = 1$	1st	M/nM	nEE	$n_i, \mathbf{U}_i, T_{i,IA}$
S06	CE	NS	$\alpha = 1$	3rd	nM	EE	n_i, \mathbf{U}_i, T_s
GHD	CE	NS	HCS	1st	nM	nEE	n_i, \mathbf{U}_i, T

Acknowledgments

V. G. acknowledges partial support from the Ministerio de Ciencia y Tecnología (Spain) through Grant No. FIS2007–60977. C.M.H. is grateful to the National Science Foundation for providing financial support of this project through grant CTS-0318999 with additional support provided by the American Chemical Society Petroleum Research Fund (Grant 43393-AC9) and the Engineering and Physical Sciences Research Council (Grant EP/DO30676/1). C. M. H. and J. W. D. are also grateful to the organizers and participants of the Granular Physics Workshop at the Kavli Institute of Theoretical Physics (with partial support from the National Science Foundation under grant PHY99-07949), which provided a starting forum for much of this work.

APPENDIX A: COLLISION FREQUENCIES

In this Appendix we display the expressions for the collision frequencies appearing in the evaluation of the kinetic contributions to the transport coefficients and in the first order contributions to the cooling rate. As noted in the main text, most of these frequencies (those corresponding to the transport coefficients) have been obtained in the low-density limit (except for the factors $\chi_{ij}^{(0)}$) [16] for arbitrary dimensions by considering the Maxwellian approximation for the zeroth-order distribution functions $f_i^{(0)}$. Their expressions are

$$\nu_{ii} = \frac{2\pi^{(d-1)/2}}{d\Gamma\left(\frac{d}{2}\right)} \sum_{j \neq i}^s n_j \sigma_{ij}^{d-1} \chi_{ij}^{(0)} \mu_{ji} v_0 (1 + \alpha_{ij}) \left(\frac{\theta_i + \theta_j}{\theta_i \theta_j} \right)^{1/2}, \quad (\text{A1})$$

$$\nu_{ij} = -\frac{2\pi^{(d-1)/2}}{d\Gamma\left(\frac{d}{2}\right)} n_i \sigma_{ij}^{d-1} \chi_{ij}^{(0)} \mu_{ij} v_0 (1 + \alpha_{ij}) \left(\frac{\theta_i + \theta_j}{\theta_i \theta_j} \right)^{1/2}, \quad (\text{A2})$$

$$\begin{aligned} \tau_{ii} = & \frac{2\pi^{(d-1)/2}}{d(d+2)\Gamma\left(\frac{d}{2}\right)} v_0 \left\{ n_i \sigma_i^{d-1} \chi_{ii}^{(0)} (2\theta_i)^{-1/2} (3 + 2d - 3\alpha_{ii}) (1 + \alpha_{ii}) \right. \\ & + 2 \sum_{j \neq i}^s n_j \chi_{ij}^{(0)} \sigma_{ij}^{d-1} \mu_{ji} (1 + \alpha_{ij}) \theta_i^{3/2} \theta_j^{-1/2} \left[(d+3) \beta_{ij} \theta_i^{-2} (\theta_i + \theta_j)^{-1/2} \right. \\ & \left. \left. + \frac{3 + 2d - 3\alpha_{ij}}{2} \mu_{ji} \theta_i^{-2} (\theta_i + \theta_j)^{1/2} + \frac{2d(d+1) - 4}{2(d-1)} \theta_i^{-1} (\theta_i + \theta_j)^{-1/2} \right] \right\}, \end{aligned} \quad (\text{A3})$$

$$\begin{aligned} \tau_{ij} = & \frac{4\pi^{(d-1)/2}}{d(d+2)\Gamma\left(\frac{d}{2}\right)} v_0 n_i \chi_{ij}^{(0)} \sigma_{ij}^{d-1} \mu_{ij} \theta_j^{3/2} \theta_i^{-1/2} (1 + \alpha_{ij}) \\ & \times \left[(d+3) \beta_{ij} \theta_j^{-2} (\theta_i + \theta_j)^{-1/2} + \frac{3 + 2d - 3\alpha_{ij}}{2} \mu_{ji} \theta_j^{-2} (\theta_i + \theta_j)^{1/2} \right. \\ & \left. - \frac{2d(d+1) - 4}{2(d-1)} \theta_j^{-1} (\theta_i + \theta_j)^{-1/2} \right], \end{aligned} \quad (\text{A4})$$

$$\begin{aligned} \omega_{ii} = & \frac{\pi^{(d-1)/2}}{\Gamma\left(\frac{d}{2}\right)} \frac{2}{d\sqrt{2}} \sigma_i^{d-1} n_i \chi_{ii}^{(0)} v_0 \theta_i^{-1/2} (1 - \alpha_{ii}^2) \\ & + \frac{\pi^{(d-1)/2}}{\Gamma\left(\frac{d}{2}\right)} \frac{2}{d(d+2)} \sum_{j \neq i}^s n_j \chi_{ij}^{(0)} \sigma_{ij}^{d-1} v_0 \mu_{ji} (1 + \alpha_{ij}) (\theta_i + \theta_j)^{-1/2} \theta_i^{1/2} \theta_j^{-3/2} A_{ij}, \end{aligned} \quad (\text{A5})$$

$$\omega_{ij} = \frac{\pi^{(d-1)/2}}{\Gamma\left(\frac{d}{2}\right)} \frac{2}{d(d+2)} n_j \chi_{ij}^{(0)} \sigma_{ij}^{d-1} v_0 \mu_{ji} (1 + \alpha_{ij}) (\theta_i + \theta_j)^{-1/2} \theta_i^{1/2} \theta_j^{-3/2} C_{ij}, \quad (\text{A6})$$

$$\begin{aligned} \gamma_{ii} = & \frac{\pi^{(d-1)/2}}{\Gamma\left(\frac{d}{2}\right)} \frac{8}{d(d+2)} \sigma_i^{d-1} n_i \chi_{ii}^{(0)} v_0 (2\theta_i)^{-1/2} (1 + \alpha_{ii}) \left[\frac{d-1}{2} + \frac{3}{16} (d+8) (1 - \alpha_{ii}) \right] \\ & + \frac{\pi^{(d-1)/2}}{\Gamma\left(\frac{d}{2}\right)} \frac{1}{d(d+2)} \sum_{j \neq i}^s n_j \chi_{ij}^{(0)} \sigma_{ij}^{d-1} v_0 \mu_{ji} (1 + \alpha_{ij}) \left(\frac{\theta_i}{\theta_j (\theta_i + \theta_j)} \right)^{3/2} \left[E_{ij} - (d+2) \frac{\theta_i + \theta_j}{\theta_i} A_{ij} \right], \end{aligned} \quad (\text{A7})$$

$$\gamma_{ij} = -\frac{\pi^{(d-1)/2}}{\Gamma\left(\frac{d}{2}\right)} \frac{1}{d(d+2)} n_i \chi_{ij}^{(0)} \sigma_{ij}^{d-1} v_0 \mu_{ij} (1 + \alpha_{ij}) \left(\frac{\theta_j}{\theta_i (\theta_i + \theta_j)} \right)^{3/2} \left[F_{ij} + (d+2) \frac{\theta_i + \theta_j}{\theta_j} C_{ij} \right]. \quad (\text{A8})$$

In the above equations, $v_0 = \sqrt{2T/m}$ is the thermal velocity, $\theta_i = m_i T / m T_i$, and we have introduced the dimensionless quantities

$$A_{ij} = (d+2)(2\beta_{ij} + \theta_j) + \mu_{ji}(\theta_i + \theta_j) \left\{ (d+2)(1 - \alpha_{ij}) - [(11+d)\alpha_{ij} - 5d - 7]\beta_{ij}\theta_i^{-1} \right\} \\ + 3(d+3)\beta_{ij}^2\theta_i^{-1} + 2\mu_{ji}^2 \left(2\alpha_{ij}^2 - \frac{d+3}{2}\alpha_{ij} + d+1 \right) \theta_i^{-1}(\theta_i + \theta_j)^2 - (d+2)\theta_j\theta_i^{-1}(\theta_i + \theta_j), \quad (\text{A9})$$

$$C_{ij} = (d+2)(2\beta_{ij} - \theta_i) + \mu_{ji}(\theta_i + \theta_j) \left\{ (d+2)(1 - \alpha_{ij}) + [(11+d)\alpha_{ij} - 5d - 7]\beta_{ij}\theta_j^{-1} \right\} \\ - 3(d+3)\beta_{ij}^2\theta_j^{-1} - 2\mu_{ji}^2 \left(2\alpha_{ij}^2 - \frac{d+3}{2}\alpha_{ij} + d+1 \right) \theta_j^{-1}(\theta_i + \theta_j)^2 + (d+2)(\theta_i + \theta_j), \quad (\text{A10})$$

$$E_{ij} = 2\mu_{ji}^2\theta_i^{-2}(\theta_i + \theta_j)^2 \left(2\alpha_{ij}^2 - \frac{d+3}{2}\alpha_{ij} + d+1 \right) [(d+2)\theta_i + (d+5)\theta_j] \\ - \mu_{ji}(\theta_i + \theta_j) \left\{ \beta_{ij}\theta_i^{-2}[(d+2)\theta_i + (d+5)\theta_j][(11+d)\alpha_{ij} - 5d - 7] \right. \\ \left. - \theta_j\theta_i^{-1}[20 + d(15 - 7\alpha_{ij}) + d^2(1 - \alpha_{ij}) - 28\alpha_{ij}] - (d+2)^2(1 - \alpha_{ij}) \right\} \\ + 3(d+3)\beta_{ij}^2\theta_i^{-2}[(d+2)\theta_i + (d+5)\theta_j] + 2\beta_{ij}\theta_i^{-1}[(d+2)^2\theta_i + (24 + 11d + d^2)\theta_j] \\ + (d+2)\theta_j\theta_i^{-1}[(d+8)\theta_i + (d+3)\theta_j] - (d+2)(\theta_i + \theta_j)\theta_i^{-2}\theta_j[(d+2)\theta_i + (d+3)\theta_j], \quad (\text{A11})$$

$$F_{ij} = 2\mu_{ji}^2\theta_j^{-2}(\theta_i + \theta_j)^2 \left(2\alpha_{ij}^2 - \frac{d+3}{2}\alpha_{ij} + d+1 \right) [(d+5)\theta_i + (d+2)\theta_j] \\ - \mu_{ji}(\theta_i + \theta_j) \left\{ \beta_{ij}\theta_j^{-2}[(d+5)\theta_i + (d+2)\theta_j][(11+d)\alpha_{ij} - 5d - 7] \right. \\ \left. + \theta_i\theta_j^{-1}[20 + d(15 - 7\alpha_{ij}) + d^2(1 - \alpha_{ij}) - 28\alpha_{ij}] + (d+2)^2(1 - \alpha_{ij}) \right\} \\ + 3(d+3)\beta_{ij}^2\theta_j^{-2}[(d+5)\theta_i + (d+2)\theta_j] - 2\beta_{ij}\theta_j^{-1}[(24 + 11d + d^2)\theta_i + (d+2)^2\theta_j] \\ + (d+2)\theta_i\theta_j^{-1}[(d+3)\theta_i + (d+8)\theta_j] - (d+2)(\theta_i + \theta_j)\theta_j^{-1}[(d+3)\theta_i + (d+2)\theta_j], \quad (\text{A12})$$

where $\beta_{ij} \equiv \mu_{ij}\theta_j - \mu_{ji}\theta_i$.

It only remains to evaluate the collision frequencies ψ_{ij} needed to get the first order contribution $\zeta^{(1,1)}$ to the cooling rate. These frequencies have not been previously determined in the Boltzmann limit. To compute them, we use the property

$$\int d\mathbf{v}_1 h(\mathbf{v}_1) J_{ij}^{(0)}[\mathbf{V}_1 | \Phi_i, \Phi_j] = \chi_{ij}^{(0)} \sigma_{ij}^{d-1} \int d\mathbf{v}_1 \int d\mathbf{v}_2 \Phi_i(\mathbf{v}_1) \Phi_j(\mathbf{v}_2) \\ \times \int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g})(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) \left[h(\mathbf{V}_1') - h(\mathbf{V}_1) \right], \quad (\text{A13})$$

with

$$\mathbf{V}_1' = \mathbf{V}_1 - \mu_{ji}(1 + \alpha_{ij})(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g})\hat{\boldsymbol{\sigma}}. \quad (\text{A14})$$

To determine ψ_{ij} , let us consider first the integral

$$I_\psi = \int d\mathbf{v} \left(\frac{m_i V^2}{2T_i} \right)^2 J_{ij}^{(0)}[f_i^{(0)}, f_{j,M} F_j]. \quad (\text{A15})$$

Use of Eqs. (A13) and (A14) gives

$$I_\psi = \chi_{ij}^{(0)} \sigma_{ij}^{d-1} \frac{m_i^2}{4T_i^2} \int d\mathbf{v}_1 \int d\mathbf{v}_2 f_i^{(0)}(\mathbf{V}_1) f_{j,M}(\mathbf{V}_2) F_j(\mathbf{V}_2) \int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g})(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) (V_1'^4 - V_1^4). \quad (\text{A16})$$

The collision rule (A14) yields

$$V_1'^4 - V_1^4 = 2\mu_{ji}^2(1 + \alpha_{ij})^2(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g})^2 \left[2(\hat{\boldsymbol{\sigma}} \cdot \mathbf{V}_1)^2 + V_1^2 + \frac{\mu_{ji}^2}{2}(1 + \alpha_{ij})^2(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g})^2 \right] - 4\mu_{ji}(1 + \alpha_{ij})(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g})(\hat{\boldsymbol{\sigma}} \cdot \mathbf{V}_1) [V_1^2 + \mu_{ji}^2(1 + \alpha_{ij})^2(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g})^2]. \quad (\text{A17})$$

The integration over the solid angle in (A16) leads to

$$\int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g})(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) (V_1'^4 - V_1^4) = \frac{4B_3}{d+3} \mu_{ji}^2(1 + \alpha_{ij})^2 g \left[3(\mathbf{V}_1 \cdot \mathbf{g})^2 + \frac{d+5}{2} g^2 V_1^2 + \mu_{ji}^2(1 + \alpha_{ij})^2 g^4 \right] - \frac{4B_3}{d+3} \mu_{ji}(1 + \alpha_{ij}) g(\mathbf{V}_1 \cdot \mathbf{g}) [(d+3)V_1^2 + 4\mu_{ji}^2(1 + \alpha_{ij})^2 g^2], \quad (\text{A18})$$

where

$$B_k \equiv \int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) (\hat{\boldsymbol{\sigma}} \cdot \hat{\mathbf{g}})^k = \pi^{(d-1)/2} \frac{\Gamma\left(\frac{k+1}{2}\right)}{\Gamma\left(\frac{k+d}{2}\right)}. \quad (\text{A19})$$

Therefore, the integral (A16) can be written as

$$I_\psi = \frac{B_3}{d+3} \chi_{ij}^{(0)} \sigma_{ij}^{d-1} n_i n_j (\theta_i \theta_j)^{d/2} v_0 \mu_{ji} (1 + \alpha_{ij}) \theta_i^2 \Delta_j I_\psi^*(\theta_i, \theta_j), \quad (\text{A20})$$

where we have taken the Maxwellian form for $f_i^{(0)}$ and have introduced the dimensionless integral

$$I_\psi^*(\theta_i, \theta_j) = \pi^{-d} \int d\mathbf{c}_1 \int d\mathbf{c}_2 e^{-(\theta_i c_1^2 + \theta_j c_2^2)x} \times \left\{ 4\mu_{ji}(1 + \alpha_{ij}) \left[3(\mathbf{c}_1 \cdot \mathbf{x})^2 + \frac{d+5}{2} g^2 c_1^2 + \mu_{ji}^2(1 + \alpha_{ij})^2 x^4 \right] - 4(\mathbf{c}_1 \cdot \mathbf{x}) [(d+3)c_1^2 + 4\mu_{ji}^2(1 + \alpha_{ij})^2 x^2] \right\}, \quad (\text{A21})$$

and the operator

$$\Delta_j \equiv \theta_j^2 \frac{\partial^2}{\partial \theta_j^2} + (d+2)\theta_j \frac{\partial}{\partial \theta_j} + \frac{d(d+2)}{4}. \quad (\text{A22})$$

In addition, $\mathbf{c}_i \equiv \mathbf{V}_i/v_0$ and $\mathbf{x} \equiv \mathbf{g}/v_0$. The integral (A21) can be performed by the change of variables $\{\mathbf{c}_1, \mathbf{c}_2\} \rightarrow \{\mathbf{x}, \mathbf{y}\}$ where $\mathbf{y} \equiv \theta_i \mathbf{c}_1 + \theta_j \mathbf{c}_2$ and the Jacobian is $(\theta_i + \theta_j)^{-d}$. With this change the integrations can be done quite efficiently by using a computer package of symbolic calculations. A lengthy and careful algebra gives

$$I_\psi^*(\theta_i, \theta_j) = (d+3) \frac{\Gamma\left(\frac{d+3}{2}\right)}{\Gamma\left(\frac{d}{2}\right)} (\theta_i \theta_j)^{-d/2} (\theta_i \theta_j)^{-5/2} (\theta_i + \theta_j)^{-1/2} \times \left\{ -2\theta_j^2 [(d+3)\theta_j + (d+2)\theta_i] + \mu_{ji}(1 + \alpha_{ij}) \theta_j (\theta_i + \theta_j) \right. \\ \times \left[(11+d)\theta_j + \frac{d^2+5d+6}{d+3} \theta_i \right] \\ \left. - 8\mu_{ji}^2(1 + \alpha_{ij})^2 \theta_j (\theta_i + \theta_j)^2 + 2\mu_{ji}^3(1 + \alpha_{ij})^3 (\theta_i + \theta_j)^3 \right\}. \quad (\text{A23})$$

Use of (A20) in (A23) leads to the final expression for I_ψ :

$$I_\psi = \frac{\pi^{(d-1)/2}}{4\Gamma\left(\frac{d}{2}\right)} n_i n_j \chi_{ij}^{(0)} \sigma_{ij}^{d-1} v_0 \mu_{ji} (1 + \alpha_{ij}) \theta_i^{3/2} \theta_j^{-3/2} (\theta_i + \theta_j)^{-5/2} \times \left\{ [(d-1)\theta_j + (d+2)\theta_i] [2\theta_j + 3\mu_{ji}(1 + \alpha_{ij})(\theta_i + \theta_j)] \right. \\ \left. - 24\mu_{ji}^2(1 + \alpha_{ij})^2 (\theta_i + \theta_j)^2 + 30\mu_{ji}^3(1 + \alpha_{ij})^3 \theta_j^{-1} (\theta_i + \theta_j)^3 \right\}. \quad (\text{A24})$$

Following similar mathematical steps as made before for I_ψ , one obtains

$$\int d\mathbf{v} \frac{m_i V^2}{2T_i} J_{ij}^{(0)}[f_i^{(0)}, f_{j,M} F_j] = \frac{\pi^{(d-1)/2}}{4\Gamma(\frac{d}{2})} n_i n_j \chi_{ij}^{(0)} \sigma_{ij}^{d-1} v_0 \mu_{ji} (1 + \alpha_{ij}) \theta_i^{3/2} \theta_j^{-3/2} (\theta_i + \theta_j)^{-3/2} \times [2\theta_j + 3\mu_{ji} (1 + \alpha_{ij}) (\theta_i + \theta_j)]. \quad (\text{A25})$$

Combining Eqs. (A24) and (A25), one gets the result

$$\int d\mathbf{v} F_i(V) J_{ij}^{(0)}[f_i^{(0)}, f_{j,M} F_j] = \frac{\pi^{(d-1)/2}}{4\Gamma(\frac{d}{2})} n_i n_j \chi_{ij}^{(0)} \sigma_{ij}^{d-1} v_0 \mu_{ji} (1 + \alpha_{ij}) \theta_i^{3/2} \theta_j^{-3/2} (\theta_i + \theta_j)^{-5/2} \times \{[(d-1)\theta_j + (d+2)\theta_i] [2\theta_j + 3\mu_{ji} (1 + \alpha_{ij}) (\theta_i + \theta_j)] - 24\mu_{ji}^2 (1 + \alpha_{ij})^2 (\theta_i + \theta_j)^2 + 30\mu_{ji}^3 (1 + \alpha_{ij})^3 \theta_j^{-1} (\theta_i + \theta_j)^3 - (d+2) (\theta_i + \theta_j) [2\theta_j + 3\mu_{ji} (1 + \alpha_{ij}) (\theta_i + \theta_j)]\}. \quad (\text{A26})$$

The remaining integrals needed to determine the collision frequencies ψ_{ii} and ψ_{ij} can be also obtained by performing identical mathematical steps. Their expressions are

$$\int d\mathbf{v} F_i(\mathbf{V}) J_{ij}^{(0)}[f_{i,M} F_i, f_j^{(0)}] = \frac{\pi^{(d-1)/2}}{4\Gamma(\frac{d}{2})} n_i n_j \chi_{ij}^{(0)} \sigma_{ij}^{d-1} v_0 \mu_{ji} (1 + \alpha_{ij}) (\theta_i \theta_j)^{-1/2} (\theta_i + \theta_j)^{-5/2} \times \{-2 [(45 + 15d)\theta_j^3 + 3(38 + 13d)\theta_i \theta_j^2 + 8(11 + 4d)\theta_i^2 \theta_j + 8(2 + d)\theta_i^3] + 3\mu_{ji} (1 + \alpha_{ij}) (\theta_i + \theta_j) [(55 + 5d)\theta_j^2 + 9(10 + d)\theta_i \theta_j + 4(8 + d)\theta_i^2] - 24\mu_{ji}^2 (1 + \alpha_{ij})^2 (\theta_i + \theta_j)^2 (5\theta_j + 4\theta_i) + 30\mu_{ji}^3 (1 + \alpha_{ij})^3 (\theta_i + \theta_j)^3 + (d+2)\theta_j (\theta_i + \theta_j) [2(4\theta_i + 3\theta_j) - 3\mu_{ji} (1 + \alpha_{ij}) (\theta_i + \theta_j)]\}, \quad (\text{A27})$$

$$\int d\mathbf{v} F_i(\mathbf{V}) J_{ii}^{(0)}[f_i^{(0)}, f_{i,M} F_i] = \frac{3\sqrt{2}}{64} \frac{\pi^{(d-1)/2}}{\Gamma(\frac{d}{2})} n_i^2 \chi_{ii}^{(0)} \sigma_i^{d-1} (1 + \alpha_{ij}) v_0 \theta_i^{-1/2} \times [10\alpha_{ii}^3 + 22\alpha_{ii}^2 + 11\alpha_{ii} - 3], \quad (\text{A28})$$

$$\int d\mathbf{v} F_i(\mathbf{V}) J_{ii}^{(0)}[f_{i,M} F_i, f_i^{(0)}] = \frac{\sqrt{2}}{64} \frac{\pi^{(d-1)/2}}{\Gamma(\frac{d}{2})} n_i^2 \chi_{ii}^{(0)} \sigma_i^{d-1} (1 + \alpha_{ij}) v_0 \theta_i^{-1/2} \times [30\alpha_{ii}^3 - 126\alpha_{ii}^2 + 177\alpha_{ii} + 16d(3\alpha_{ii} - 7) - 137]. \quad (\text{A29})$$

The expressions for the frequencies ψ_{ii} and ψ_{ij} can be easily obtained from Eqs. (A27)–(A30) when one takes into account their definitions (2.17) and (2.18).

In the case of mechanically equivalent particles ($\chi_{ij}^{(0)} = \chi^{(0)}$, $\sigma_i = \sigma$, and $\alpha_{ij} = \alpha$), Eqs. (A28) and (A29) yield

$$\int d\mathbf{v} F(\mathbf{V}) \left(J^{(0)}[f^{(0)}, f_M F(\mathbf{V})] + J^{(0)}[f_M F(\mathbf{V}), f^{(0)}] \right) = \frac{\pi^{(d-1)/2}}{8\Gamma(\frac{d}{2})} \chi^{(0)} \sigma^{d-1} n_i n_j \frac{1 + \alpha}{2} \times (30\alpha^3 - 30\alpha^2 + 105\alpha + 24d\alpha - 56d - 73). \quad (\text{A30})$$

This expression coincides with the one previously derived for a monodisperse granular gas [15].

APPENDIX B: COLLISION INTEGRALS

In this Appendix, we provide some of the mathematical steps to compute the different collision integrals involving the operator $\mathcal{K}_{ij,\gamma}[X]$. This operator is defined as

$$\mathcal{K}_{ij,\gamma}[X_j] = \sigma_{ij}^d \chi_{ij}^{(0)} \int d\mathbf{v}_2 \int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) \hat{\sigma}_\gamma \times [\alpha_{ij}^{-2} f_i^{(0)}(\mathbf{V}_1'') X_j(\mathbf{V}_2'') + f_i^{(0)}(\mathbf{V}_1) X_j(\mathbf{V}_2)]. \quad (\text{B1})$$

To simplify all these type of integrals, we use the property

$$\begin{aligned} \int d\mathbf{v}_1 h(\mathbf{V}_1) \mathcal{K}_{ij,\gamma} [X_j(\mathbf{V}_2)] &= -\chi_{ij}^{(0)} \sigma_{ij}^d \int d\mathbf{v}_1 \int d\mathbf{v}_2 f_i^{(0)}(\mathbf{V}_1) X_j(\mathbf{V}_2) \\ &\times \int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g})(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) \hat{\sigma}_\gamma [h(\mathbf{V}'_1) - h(\mathbf{V}_1)] , \end{aligned} \quad (\text{B2})$$

where \mathbf{V}'_1 is defined by Eq. (A14).

Let us start with the collision integrals appearing in the evaluation of the mass flux. One of them is

$$I_D \equiv \int d\mathbf{v}_1 m_i V_{1,\gamma} \mathcal{K}_{ij,\gamma} [\nabla_{\mathbf{v}_2} \cdot (\mathbf{V}_2 f_j^{(0)})]. \quad (\text{B3})$$

Use of the identity (B2) in (B3) gives

$$I_D = B_2 \sigma_{ij}^d \chi_{ij}^{(0)} m_i \mu_{ji} (1 + \alpha_{ij}) \int d\mathbf{V}_1 \int d\mathbf{V}_2 f_i^{(0)}(\mathbf{V}_1) \nabla_{\mathbf{V}_2} \cdot (\mathbf{V}_2 f_j^{(0)}(\mathbf{V}_2)) g^2. \quad (\text{B4})$$

The integral (B4) can be exactly evaluated and the result is

$$I_D = -2dB_2 n_i n_j \sigma_{ij}^d \chi_{ij}^{(0)} \mu_{ji} (1 + \alpha_{ij}) T_j. \quad (\text{B5})$$

The remaining integrals corresponding to the mass flux can be computed by using similar mathematical steps as those made before for I_D . The results are

$$\int d\mathbf{v}_1 m_i V_{1,\beta} \mathcal{K}_{ij,\beta} [f_j^{(0)}] = dB_2 n_i n_j \sigma_{ij}^d \chi_{ij}^{(0)} m_i \mu_{ji} (1 + \alpha_{ij}) \left(\frac{T_i}{m_i} + \frac{T_j}{m_j} \right), \quad (\text{B6})$$

$$\begin{aligned} \int d\mathbf{v}_1 m_i V_{1,\beta} \mathcal{K}_{i\ell,\beta} [n_j \partial_{n_j} f_\ell^{(0)}] &= \int d\mathbf{v}_1 m_i V_{1,\beta} \left(\mathcal{K}_{i\ell,\beta} [\delta_{j\ell} f_\ell^{(0)}] - \frac{1}{2} \mathcal{K}_{i\ell,\beta} [\nabla_{\mathbf{v}_2} \cdot (\mathbf{V}_2 f_\ell^{(0)})] n_j \partial_{n_j} \ln \gamma_\ell \right) \\ &= dB_2 n_i n_j \sigma_{i\ell}^d \chi_{i\ell}^{(0)} m_i \mu_{\ell i} (1 + \alpha_{i\ell}) \left[\delta_{j\ell} \left(\frac{T_i}{m_i} + \frac{T_\ell}{m_\ell} \right) \right. \\ &\quad \left. + \frac{n_\ell T_\ell}{n_j m_\ell} \frac{\partial \ln \gamma_\ell}{\partial \ln n_j} \right]. \end{aligned} \quad (\text{B7})$$

The collision integral involved in the evaluation of the pressure tensor is of the form

$$\begin{aligned} I_\eta &\equiv \int d\mathbf{v}_1 m_i V_{1,\gamma} V_{1,\beta} \mathcal{K}_{ij,\gamma} [\partial_{V_\beta} f_j^{(0)}] \\ &= \chi_{ij}^{(0)} \sigma_{ij}^d m_i \int d\mathbf{v}_1 \int d\mathbf{v}_2 f_i(\mathbf{V}_1) \left(\partial_{V_{2,\beta}} f_j^{(0)}(\mathbf{V}_2) \right) \\ &\times \int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g})(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) \hat{\sigma}_\alpha (V'_{1,\gamma} V'_{1,\beta} - V_{1,\gamma} V_{1,\beta}), \end{aligned} \quad (\text{B8})$$

where the identity (B2) has been used. The scattering rule (A14) gives

$$\begin{aligned} V'_{1,\gamma} V'_{1,\beta} - V_{1,\gamma} V_{1,\beta} &= -\mu_{ji} (1 + \alpha_{ij}) (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) [G_{ij,\gamma} \hat{\sigma}_\beta + G_{ij,\beta} \hat{\sigma}_\gamma + \mu_{ji} (g_\gamma \hat{\sigma}_\beta + g_\beta \hat{\sigma}_\gamma \\ &\quad - \mu_{ji} (1 + \alpha_{ij}) (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) \hat{\sigma}_\gamma \hat{\sigma}_\beta], \end{aligned} \quad (\text{B9})$$

where $\mathbf{G}_{ij} = \mu_{ij} \mathbf{V}_1 + \mu_{ji} \mathbf{V}_2$. Substitution of Eq. (B9) into (B8) allows the angular integral to be performed with the result

$$\begin{aligned} \int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g})(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) \hat{\sigma}_\gamma (V'_{1,\gamma} V'_{1,\beta} - V_{1,\gamma} V_{1,\beta}) &= -\frac{B_2}{d+2} \mu_{ji} (1 + \alpha_{ij}) [(d+3) g^2 G_{ij,\beta} \\ &\quad + \mu_{ji} (1 + d - 3\alpha_{ij}) g^2 g_\beta + 2(\mathbf{g} \cdot \mathbf{G}_{ij}) g_\beta]. \end{aligned} \quad (\text{B10})$$

With this result the integral I_η becomes

$$\begin{aligned}
I_\eta &= -\frac{dB_2}{d+2} n_i n_j \chi_{ij}^{(0)} \sigma_{ij}^d m_i \mu_{ji} (1 + \alpha_{ij}) \int d\mathbf{v}_1 \int d\mathbf{v}_2 f_i^{(0)}(\mathbf{V}_1) f_j^{(0)}(\mathbf{V}_2) \\
&\quad \times [(d+2)\mu_{ji}(3\alpha_{ij}-1)g^2 - 4(d+2)(\mathbf{g} \cdot \mathbf{G}_{ij})] \\
&= -dB_2 n_i n_j \chi_{ij}^{(0)} \sigma_{ij}^d m_i \mu_{ji} (1 + \alpha_{ij}) m_i \left[\mu_{ji} \left(\frac{T_i}{m_i} + \frac{T_j}{m_j} \right) (3\alpha_{ij}-1) - 4 \frac{T_i - T_j}{m_i + m_j} \right].
\end{aligned} \tag{B11}$$

From Eq. (B11) it is easy to get the expression (3.34).

To evaluate the collision integrals appearing in the determination of the heat flux one needs the partial results

$$\begin{aligned}
\mathbf{S}_i(\mathbf{V}'_1) - \mathbf{S}_i(\mathbf{V}_1) &= \frac{m_i}{2} (1 + \alpha_{ij}) \mu_{ji} (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) \left\{ [(1 - \alpha_{ij}^2) \mu_{ji}^2 (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g})^2 - G_{ij}^2 - \mu_{ji}^2 g^2 \right. \\
&\quad \left. - 2\mu_{ji} (\mathbf{g} \cdot \mathbf{G}_{ij}) + 2(1 + \alpha_{ij}) \mu_{ji} (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) (\hat{\boldsymbol{\sigma}} \cdot \mathbf{G}_{ij}) + (d+2) \frac{T_i}{m_i} \right] \hat{\boldsymbol{\sigma}} \\
&\quad \left. - [(1 - \alpha_{ij}) \mu_{ji} (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) + 2(\hat{\boldsymbol{\sigma}} \cdot \mathbf{G}_{ij})] (\mathbf{G}_{ij} + \mu_{ji} \mathbf{g}) \right\},
\end{aligned} \tag{B12}$$

$$\begin{aligned}
\int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) \quad \hat{\boldsymbol{\sigma}} \cdot [\mathbf{S}_i(\mathbf{V}'_1) - \mathbf{S}_i(\mathbf{V}_1)] &= -\frac{3}{2} \frac{B_2}{d+2} (1 + \alpha_{ij}) \mu_{ji} \\
&\times \left\{ \frac{1}{3} \mu_{ji}^2 [(d+2) - 3\alpha_{ij}(1 - \alpha_{ij})] g^4 + \frac{d+4}{3} g^2 G_{ij}^2 \right. \\
&\quad \left. + \frac{1}{3} \mu_{ji} (7 + 2d - 9\alpha_{ij}) g^2 (\mathbf{g} \cdot \mathbf{G}_{ij}) + \frac{4}{3} (\mathbf{g} \cdot \mathbf{G}_{ij})^2 - \frac{(d+2)^2}{3} \frac{T_i}{m_i} g^2 \right\}.
\end{aligned} \tag{B13}$$

The corresponding integrals associated with the heat flux can be explicitly evaluated by using Eqs. (B12) and (B13) and the same mathematical steps as before. After a lengthy algebra, one gets the expressions (3.70), (3.71), and (3.72).

Let us consider now the integral appearing in the evaluation of the cooling rate. To do that, we compute first the collision integral

$$I_\zeta = \int d\mathbf{v} V^4 \mathcal{K}_{ij,\gamma} [\partial_{V_\gamma} f_j^{(0)}]. \tag{B14}$$

In order to evaluate it we use Eq. (A17) and the angular integrations

$$\begin{aligned}
\int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}) \hat{\sigma}_\beta (V_1'^4 - V_1^4) &= \frac{3B_2}{(d+2)(d+4)} \mu_{ji} (1 + \alpha_{ij}) \\
&\times \left\{ 4\mu_{ji} (1 + \alpha_{ij}) [2(\mathbf{V}_1 \cdot \mathbf{g})^2 g_\beta + 2(\mathbf{V}_1 \cdot \mathbf{g}) g^2 V_{1,\beta} \right. \\
&\quad \left. + \frac{1}{2} (d+6) g^2 V_1^2 g_\beta \right] + 5\mu_{ji}^3 (1 + \alpha_{ij})^3 g^4 g_\beta \\
&\quad - \frac{4}{3} (d+4) V_1^2 [2(\mathbf{V}_1 \cdot \mathbf{g}) g_\beta + g^2 V_{1,\beta}] \\
&\quad \left. - 4\mu_{ji}^2 (1 + \alpha_{ij})^2 g^2 [4(\mathbf{V}_1 \cdot \mathbf{g}) g_\beta + g^2 V_{1,\beta}] \right\}.
\end{aligned} \tag{B15}$$

With these results, the integral I_ζ becomes

$$\begin{aligned}
I_\zeta &= -3dB_2n_in_j\chi_{ij}^{(0)}\sigma_{ij}^d\mu_{ji}(1+\alpha_{ij})\int d\mathbf{v}_1\int d\mathbf{v}_2f_i^{(0)}(\mathbf{V}_1)f_j^{(0)}(\mathbf{V}_2) \\
&\times \left\{ \mu_{ji}(1+\alpha_{ij}) \left[2(d+4)g^2V_1^2 + 8(\mathbf{V}_1 \cdot \mathbf{g})^2 + 5\mu_{ji}^2(1+\alpha_{ij})^2g^4 - 16\mu_{ji}(1+\alpha_{ij})g^2(\mathbf{V}_1 \cdot \mathbf{g}) \right] \right. \\
&\quad \left. - \frac{8}{3}(d+2)V_1^2(\mathbf{V}_1 \cdot \mathbf{g}) \right\} \\
&= -3dB_2n_in_j\chi_{ij}^{(0)}\sigma_{ij}^d\mu_{ji}(1+\alpha_{ij}) \left\{ \frac{T_i^2}{m_i^2} [\mu_{ji}(1+\alpha_{ij})(16+2d-16\mu_{ji}(1+\alpha_{ij})) \right. \\
&\quad \left. + 5\mu_{ji}^2(1+\alpha_{ij})^2] - \frac{8}{3}(d+2) \right\} + 5\mu_{ji}^3(1+\alpha_{ij})^3 \frac{T_j^2}{m_j^2} \\
&\quad \left. + \frac{2}{d+2} \frac{T_iT_j}{m_im_j} \mu_{ji}(1+\alpha_{ij}) [d(d+4)+4-8(d+2)\mu_{ji}(1+\alpha_{ij})+5(d+2)\mu_{ji}^2(1+\alpha_{ij})^2] \right\}.
\end{aligned} \tag{B16}$$

The final expression (2.19) can be easily obtained from Eqs. (B11) and (B16).

Finally, note that the integrals C_{ij}^T and C_{ipj}^T defined by Eqs. (3.40) and (3.41), respectively, can be easily computed by using the change of variables written below Eq. (A22). After some algebra and using the Maxwellian approach for the distributions $f_i^{(0)}$ one gets the results (3.42) and (3.43).

APPENDIX C: CHOICE OF $\chi_{ij}^{(0)}$ AND I_{ilj}

This Appendix deals with the choice of the pair correlation function $\chi_{ij}^{(0)}$ and the functional derivative I_{ilj} . A good approximation for $\chi_{ij}^{(0)}$ in two dimensions ($d=2$) is given by [72]

$$\chi_{ij}^{(0)} = \frac{1}{1-\phi} + \frac{10-\phi}{16} \frac{\beta}{(1-\phi)^2} \frac{\sigma_i\sigma_j}{\sigma_{ij}} - \frac{1}{16} \frac{\beta^2}{\phi(1-\phi)} \left(\frac{\sigma_i\sigma_j}{\sigma_{ij}} \right)^2, \tag{C1}$$

where $\phi = \sum_{i=1}^s \phi_i$ is the total solid volume fraction, $\phi_i = n_i\pi\sigma_i^2/4$ is the species volume fraction of component i and $\beta = \pi(\sum_{i=1}^s n_i\sigma_i)/4$. In the case of hard-spheres ($d=3$) we take the following approximation for $\chi_{ij}^{(0)}$ [71]

$$\chi_{ij}^{(0)} = \frac{1}{1-\phi} + \frac{3}{2} \frac{\beta}{(1-\phi)^2} \frac{\sigma_i\sigma_j}{\sigma_{ij}} + \frac{1}{2} \frac{\beta^2}{(1-\phi)^3} \left(\frac{\sigma_i\sigma_j}{\sigma_{ij}} \right)^2, \tag{C2}$$

where now $\phi_i = n_i\pi\sigma_i^3/6$ and $\beta = \pi(\sum_{i=1}^s n_i\sigma_i^2)/6$.

The parameter I_{ilj} is chosen to recover the results derived by López de Haro *et al.* [73] for ordinary polydisperse mixtures in the context of the RET. To do that, for the sake of simplicity, we assume that the temperature, the pressure and the flow velocity are homogeneous so that only the spatial gradients associated with the partial densities will be considered. In this simple case, for elastic collisions ($\alpha_{ij}=1$), the first-order distribution function is given by $f_i^{(1)} = \sum_{j=1}^s \mathcal{B}_i^j \cdot \nabla \ln n_j$ where $\mathcal{B}_i^j(\mathbf{V})$ verifies the integral equation

$$(\mathcal{LB}^j)_{i,\gamma} = -V_\gamma n_j \partial_{n_j} f_i^{(0)} - \sum_{\ell=1}^s \left(\mathcal{K}_{i\ell,\gamma} [n_j \partial_{n_j} f_\ell^{(0)}] + \frac{1}{2} \left(n_\ell \partial_{n_j} \ln \chi_{i\ell}^{(0)} + I_{i\ell j} \right) \mathcal{K}_{i\ell,\gamma} [f_\ell^{(0)}] \right). \tag{C3}$$

In the elastic case, $n_j \partial_{n_j} f_\ell^{(0)} = \delta_{j\ell} f_\ell^{(0)}$ and the linear operator $\mathcal{K}_{i\ell} [f_\ell^{(0)}]$ can be explicitly written as

$$\mathcal{K}_{i\ell,\gamma} [f_\ell^{(0)}] = 2B_2 n_\ell \chi_{i\ell}^{(0)} \sigma_{i\ell}^d V_\gamma f_i^{(0)}(\mathbf{V}). \tag{C4}$$

With this result, Eq. (C3) becomes

$$-(\mathcal{LB}^j)_{i,\gamma} = V_\gamma f_i^{(0)} \delta_{ij} + 2B_2 \sum_{\ell=1}^s n_\ell \chi_{i\ell}^{(0)} \sigma_{i\ell}^d \left[\delta_{j\ell} + \frac{1}{2} \left(n_\ell \partial_{n_j} \ln \chi_{i\ell}^{(0)} + I_{i\ell j} \right) \right] V_\gamma f_i^{(0)}. \tag{C5}$$

Comparison with the results derived by López de Haro *et al.* [73] allow us to identify $I_{ij\ell}$ to be defined through the relation

$$\sum_{\ell=1}^s n_{\ell} \chi_{i\ell}^{(0)} \sigma_{i\ell}^d \left(n_{\ell} \partial_{n_j} \ln \chi_{i\ell}^{(0)} + I_{ij\ell} \right) = \frac{n_j}{B_2} \left[\frac{1}{T} \left(\frac{\partial \mu_i}{\partial n_j} \right)_{T, n_k \neq j} - \frac{1}{n_i} \delta_{ij} - 2B_2 \chi_{ij}^{(0)} \sigma_{ij}^d \right], \quad (\text{C6})$$

where μ_i is the chemical potential of species i .

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