Enskog theory for polydisperse granular mixtures. I. Navier-Stokes order transport

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A hydrodynamic description for an *s*-component mixture of inelastic, smooth hard disks (two dimensions) or spheres (three dimensions) is derived based on the revised Enskog theory for the single-particle velocity distribution functions. In this first part of the two-part series, the macroscopic balance equations for mass, momentum, and energy are derived. Constitutive equations are calculated from exact expressions for the fluxes by a Chapman-Enskog expansion carried out to first order in spatial gradients, thereby resulting in a Navier-Stokes order theory. Within this context of small gradients, the theory is applicable to a wide range of restitution coefficients and densities. The resulting integral-differential equations for the zeroth- and first-order approximations of the distribution functions are given in exact form. An approximate solution to these equations is required for practical purposes in order to cast the constitutive quantities as algebraic functions of the macroscopic variables; this task is described in the companion paper.

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I. INTRODUCTION

Flows of polydisperse particles (mixtures) are ubiquitous in nature and industry alike. Examples of the former include pyroclastic flows, landslides, pollutant transport, and planetary rings. Examples of the latter include pneumatic conveying of grains, ores, and chemicals; fluidized-bed operation for power production and catalytic cracking; and mixing of pharmaceutical powders (medication and binder) and poultry feedstock (grains and vitamins). A nonuniform particle distribution may be a property of the starting material itself, or it may be intentionally utilized in order to improve process performance. For an example of the latter, the addition of fines to a relatively monodisperse material has been shown to (i) decrease attrition in high-speed conveying lines [1], (ii) increase conversion in high-velocity, fluidized-bed reactors [2], and (iii) improve heat transfer efficiency in a circulating fluidized bed (CFB) combustor [3]. Polydisperse materials are also known to exhibit counterintuitive behaviors that have no monodisperse counterpart. For example, agitation of polydisperse materials via vibration, free-fall, or flow down an incline leads to segregation among unlike particles (demixing). Enhancing or suppressing this segregation tendency may be critical to process performance, depending on whether the desired outcome is a separated or well-mixed state, respectively.

In the current effort, attention is restricted to rapid flows, in which particle collisions are assumed to be both binary and instantaneous in nature. For monodisperse systems, kinetic-theory-based treatments have been successful at predicting not only rapid granular flows (in which the role of the interstitial fluid is assumed negligible), but have also been incorporated into models of high-velocity, gas-solid systems. In particular, kinetic-theory-based descriptions are now standard components in both commercial and open-source CFD (computational fluid dynamics) software packages for multiphase flows such as Fluent® and MFIX (http://www.mfix.org/), respectively. Nonetheless, the development and application of kinetic-theory-based descriptions for polydisperse systems are in their infancy relative to their monodisperse counterparts, as has been highlighted in several recent review articles and perspectives [4–7]. The main challenge associated with the derivation of kinetic-theory-based descriptions for mixtures is the increased complexity associated with the additional hydrodynamic fields and associated transport coefficients, and in particular with the accurate evaluation of the collisions integrals. Correspondingly, many of the early contributions resorted to assumptions which are only strictly true in the limit of perfectly elastic spheres in a uniform steady state: a Maxwellian (single-particle) velocity distribution [8–10] or an equipartition of energy [11–14]. However, the presence of a non-Maxwellian velocity distribution in granular flows is well-documented [15–22] and has been shown to have a significant impact on some transport coefficients [23]. Moreover, a nonequipartition of energy between unlike particles is widely established [24–32], and has been shown to significantly contribute to the driving force for segregation [33] and to lead to a reversal of the segregation direction [34–36] in certain systems. A more recent theory [37] involves the lifting of both of these assumptions, except in the evaluation of collision integrals involving two unlike particles, in which case a Maxwellian velocity distribution is assumed for each particle type. Two current theories exist that do not involve either of these as-

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sumptions [38,39], though both are based on the Boltzmann equation and thus are limited to dilute flows. Another key difference between existing polydisperse theories is the base state used in the Chapman-Enskog (CE) expansion. Some theories [8–14,37,39] assume an expansion about a perfectly elastic (molecular equilibrium) base state, and thus are restricted to nearly elastic systems. However, in the CE method the base state must not be chosen *a priori*, but rather it is determined as the solution to the kinetic equation to zeroth order in the gradient expansion. This solution is found to correspond to the *local* homogeneous cooling state (HCS) and was used in Ref. [38] as the reference state to determine the Navier-Stokes transport coefficients of a dilute mixture, without any restriction on the level of inelasticity.

The objective of the current effort is twofold. First, a kinetic-theory-based description for the flow of an s-component mixture in d dimensions is derived which (i) incorporates non-Maxwellian and nonequipartition effects, (ii) is applicable to a wide range of restitution coefficients, and (iii) is applicable to both dilute and (moderately) dense flows. In particular, a CE expansion of the revised Enskog theory for inelastic, hard spheres is carried out for both disks ($d=2$) and spheres ($d=3$) up to the Navier-Stokes order. Second, the derivation of the resulting theory is critically compared and contrasted to that of existing theories, in an effort to clearly reveal the implications of various treatments on both the governing equations and constitutive relations. For this reason, the derivation is presented in a detailed and somewhat pedagogical fashion. This work takes the form of two self-contained, companion papers. In this first paper, the results of the exact analysis are given. The follow-up paper details the leading order approximations needed for the explicit evaluation of all properties derived here: the distribution functions, the "equations of state" (cooling rate and pressure), and the transport coefficients. In addition, the methodology used to obtain these results is critically compared there to that of previous theories.

A confusing issue in the granular community is the context of the Navier-Stokes hydrodynamic equations in freely cooling granular gases derived in this paper. The expressions for the Navier-Stokes transport coefficients are not limited to weak inelasticity and so the calculations provided here apply even for strong dissipation. The Navier-Stokes hydrodynamic equations may or may not be limited with respect to inelasticity, depending on the particular states analyzed. The CE method assumes that the relative changes of the hydrodynamic fields over distances of the order of the mean free path are small. For ordinary (elastic) gases this can be controlled by the initial or boundary conditions. However, in the case of granular fluids the situation is more complicated since in some cases (e.g., steady states such as the simple shear flow problem [40]) the boundary conditions imply a relationship between dissipation and gradients so that both cannot be chosen independently. In these cases, the Navier-Stokes approximation only holds for nearly elastic particles [40]. However, the transport coefficients characterizing the Navier-Stokes hydrodynamic equations are nonlinear functions of the coefficients of restitution, regardless of the applicability of those equations.

In spite of the above cautions, the Navier-Stokes approximation is relevant to describe a wide class of granular flows. One of them corresponds to spatial perturbations of the HCS for an isolated system. Computer simulations have confirmed the accuracy of the Navier-Stokes hydrodynamic equations with their associated transport coefficients to quantitatively describe cluster formation [41]. The same kinetic theory results apply to driven systems as well. This is so since the reference state is a *local* HCS whose parameters change throughout the system to match the physical values in each cell. Other examples of good agreement between theory and simulation [42] and experiments [43,44] include the application of the Navier-Stokes hydrodynamics to describe density-temperature profiles in vertical vibrated gases, supersonic flow past a wedge in real experiments [45], and nonequipartition and size segregation in agitated granular mixtures [27,28,46]. In summary, the Navier-Stokes hydrodynamics with the constitutive equations obtained in this paper constitute an important and useful description for many different physical situations, although more limited than for elastic gases.

II. OVERVIEW OF DERIVATION

The theoretical basis for a hydrodynamic description of molecular gases is most completely established at low density using the Boltzmann kinetic equation. There, the CE solution and its prediction of transport coefficients is well-established from both computer simulation and experiment [47]. For a moderately dense gas there is no accurate and practical generalization of the Boltzmann equation except for the idealized hard sphere fluid. In that case, the Enskog kinetic equation describes the dominant positional corrections to the Boltzmann equation due to excluded volume effects of other particles on a colliding pair [47]. The neglected velocity correlations are important only at much higher densities. The derivation of hydrodynamics and evaluation of transport coefficients based on the Enskog kinetic equation leads to an accurate and unique description of moderately dense gases. The generalization to mixtures requires a revision of the original Enskog theory for thermodynamic consistency [revised Enskog theory (RET)] [48], and its application to hydrodynamics and mixture transport coefficients was accomplished 20 years ago [49]. As noted above, for granular (dissipative) gases, there remains an open problem of predicting transport properties at moderate densities, as occurs in current experiments and simulations. This problem is addressed here in its full generality using the extension of this revised Enskog theory to inelastic collisions without limits on the number of components, densities, temperature, or degree of dissipation. This subsumes all previous analyses for both molecular and granular gases, which are recovered in the appropriate limits.

Due to the extreme length of the derivation, an outline of the steps involved is given here for easy reference.

Section III. The starting point of the derivation process is the revised Enskog kinetic equations for mixtures of inelastic, hard spheres. These equations for the single-particle, position, and velocity distribution functions of each species, $\{f_i\}$, take the form of nonlinear, integrodifferential equations, where the integral portion arises from the collision operator.

Section IV. The macroscopic variables of interest (number density $\{n_i\}$, etc.) are defined exactly in terms of moments of $\{f_i\}$ [e.g., $n_i(\mathbf{r})=\int d\mathbf{v} f_i(\mathbf{r},\mathbf{v},t)$, where $\mathbf{v}$ is the velocity of species i]. Thus the macroscopic balance equations can be obtained by appropriate manipulation of the Enskog kinetic equations (e.g., multiplication by $d\mathbf{v}$ followed by integration over the velocity to obtain the species mass balance). At this stage, all of the constitutive quantities (cooling rate, stress tensor, conduction, and mass flux) appearing in the macroscopic balances are integral functionals of $\{f_i\}$, which depend explicitly on space and time only through their dependence on $\{f_i\}$.

Section V. In order to obtain a hydrodynamic description (one in which the constitutive quantities are determined entirely by the macroscopic or hydrodynamic variables), the concept of a normal solution is introduced. These are special solutions to the Enskog equations for which the $\{f_i\}$ depend on space and time only through an implicit *functional* dependence on the macroscopic fields (or equivalently as explicit *functions* of these local fields and their gradients at the spatial point of interest).

Section VI. An exact analytical solution for $\{f_i\}$ is not a practical objective in the most general case, and thus attention is restricted to states with small spatial gradients. In this case the gradients provide a small parameter, allowing a small spatial gradients, or small Knudsen number, expansion (i.e., the CE expansion). The analysis is carried out here to first (Navier-Stokes) order: $f_i=f_i^{(0)}+f_i^{(1)}$, where $f_i^{(0)}$ is the zeroth order solution and $f_i^{(1)}$ is the first-order correction (zero- and first-order in gradients, respectively). The kinetic equations then become integral-differential equations for the determination of $f_i^{(0)}$ and $f_i^{(1)}$.

Section VII. Correspondingly, the constitutive equations are identified as functions of the hydrodynamic variables and their gradients through their dependence on $\{f_i\}$, with coefficients of the gradients defining the transport coefficients. Hence all equations of state (pressure and reference state cooling rate) and all transport coefficients, which are integrals involving $f_i^{(0)}$, inherit this dependence on the hydrodynamic variables and their gradients. The coefficients are determined from solutions to the integral equations.

This completes the derivation reported in this paper. Up until this point, the results are exact for Navier-Stokes order hydrodynamics (first order in spatial gradients) of the RET. This determines the form of the Navier-Stokes hydrodynamics, but more explicit dependence of the transport coefficients on the macroscopic variables requires a corresponding explicit solution to the integral equations for $f_i^{(0)}$ and $f_i^{(1)}$. One approximate method, known to be accurate for ordinary fluids, is detailed in the follow-up paper [50], resulting in constitutive quantities that are algebraic functions of the macroscopic variables.

III. REVISED ENSKOG KINETIC THEORY

The system considered is a mixture of $\{N_i\}$ smooth hard disks ($d=2$) or spheres ($d=3$) of masses $\{m_i\}$ and diameters $\{\sigma_i\}$, where the subscript i labels one of the s mechanically different species and d is the dimension. In general, collisions among all pairs are inelastic and are characterized by independent constant normal restitution coefficients $\{\alpha_{ij}=\alpha_{ji}\}$, where α_{ij} is the restitution coefficient for collisions between particles of species i and j, $0<\alpha_{ij}\leqslant 1$. The macroscopic (or hydrodynamic) properties of interest (number densities, flow velocity, and energy density) are determined from the single particle position and velocity distribution functions $f_i(\mathbf{r}_1,\mathbf{v}_1;t)$, for $i=1,\ldots,s$, where $f_i(\mathbf{r}_1,\mathbf{v}_1;t)d\mathbf{r}_1 d\mathbf{v}_1$ is proportional to the probability to find a particle of species i in the position and velocity element $d\mathbf{r}_1 d\mathbf{v}_1$ at time t. The fundamental description of any system is based on the probability density for *all* constituent particles and the Liouville equation for its time evolution; this is equivalent to solving the collective equations of motion for all particles in the system and becomes computationally prohibitive for a large number of particles. However, for the macroscopic fields only the reduced distribution functions $\{f_i\}$, obtained from the integration of the probability density over all except one particle's position and velocity for each of the species, are required for calculation of the macroscopic properties. The equations for these reduced distribution functions resulting from the partial integrations of the Liouville equation, give rise to the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy equations. The first level of this hierarchy gives the time dependence of $\{f_i\}$ [51,52]

$$[\partial_t + \mathbf{v}_1\cdot\nabla_{\mathbf{r}_1} + m_i^{-1}\mathbf{F}_i(\mathbf{r}_1)\cdot\nabla_{\mathbf{v}_1}]f_i(\mathbf{r}_1,\mathbf{v}_1;t) = C_i(\mathbf{r}_1,\mathbf{v}_1;t), \tag{3.1}$$

where

$$C_i(\mathbf{r}_1,\mathbf{v}_1;t) = \sum_{j=1}^{s}\sigma_{ij}^{d-1}\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12}) \times[\alpha_{ij}^{-2}f_{ij}(\mathbf{r}_1,\mathbf{v}_1'',\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2'';t) - f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)]. \tag{3.2}$$

The left sides of these equations describe changes in the distribution functions due to motion in the presence of external conservative forces $\mathbf{F}_i(\mathbf{r}_1)$. The right side describes changes due to collisions among the particles. The function $f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_2,\mathbf{v}_2;t)d\mathbf{r}_1 d\mathbf{v}_1 d\mathbf{r}_2 d\mathbf{v}_2$ is proportional to the joint probability of finding a particle of species i in $d\mathbf{r}_1 d\mathbf{v}_1$ and one of species j in $d\mathbf{r}_2 d\mathbf{v}_2$. The position $\mathbf{r}_2$ in these functions appears only for $\mathbf{r}_2=\mathbf{r}_1\pm\boldsymbol{\sigma}_{ij}$, where $\boldsymbol{\sigma}_{ij}=\hat{\boldsymbol{\sigma}}\sigma_{ij}$ and $\sigma_{ij}\equiv(\sigma_i+\sigma_j)/2$; this means that the two particles are at contact. The vector $\hat{\boldsymbol{\sigma}}$ is a unit vector directed along the line of centers from the sphere of species j to that of species i at contact and the integration $d\hat{\boldsymbol{\sigma}}$ is over a solid angle for this contact sphere. The Heaviside step function Θ assures that the relative velocities $\mathbf{g}_{12}=\mathbf{v}_1-\mathbf{v}_2$ are such that a collision takes place, and the "restituting" (precollisional) velocities $\mathbf{v}_1''$ and $\mathbf{v}_2''$ are related to the postcollisional velocities by

$$\mathbf{v}_1'' = \mathbf{v}_1 - \mu_{ji}(1+\alpha_{ij}^{-1})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\hat{\boldsymbol{\sigma}},$$

$$\mathbf{v}_2'' = \mathbf{v}_2 + \mu_{ij}(1+\alpha_{ij}^{-1})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\hat{\boldsymbol{\sigma}}, \tag{3.3}$$

where $\mu_{ij}=m_i/(m_i+m_j)$. It is convenient for the discussion here to write that equation in a more symbolic form by introducing the notation

$$X(\mathbf{v}_1'',\mathbf{v}_2'') = b_{ij}^{-1}X(\mathbf{v}_1,\mathbf{v}_2), \tag{3.4}$$

so that b_{ij}^{-1} is a general substitution operator that changes the argument of a function to its precollision velocities given by Eq. (3.3). Then, changing variables $\hat{\boldsymbol{\sigma}}\to-\hat{\boldsymbol{\sigma}}$ in the second term on the right side of Eq. (3.1) and noting that $b_{ij}^{-1}\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12}=-\alpha_{ij}^{-1}\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12}$ gives the equivalent form [51,52]

$$\begin{aligned}[\partial_t + \mathbf{v}_1\cdot\nabla_{\mathbf{r}_1} + m_i^{-1}\mathbf{F}_i(\mathbf{r}_1)\cdot\nabla_{\mathbf{v}_1}]f_i(\mathbf{r}_1,\mathbf{v}_1;t)&\\ = -\sum_{j=1}^{s}\sigma_{ij}^{d-1}\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}(\alpha_{ij}^{-1}b_{ij}^{-1}+1)(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})&\\ \times\Theta(-\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t).&\end{aligned} \tag{3.5}$$

This demonstrates that the two particle distributions f_{ij} appear only on the contact hemisphere given by $\Theta(-\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})$, correponding to particles that are directed toward each other and hence have a change in their velocities.

Equation (3.1) becomes a kinetic theory (i.e., closed equations for the set of f_i) only after specifying f_{ij} on the right side as a functional of the set of f_i (the alternative of making approximations at higher levels of the BBGKY hierarchy has not been productive in general for molecular gases). As indicated above, this is required for f_{ij} only when the particles are at contact and on that hemisphere for which the relative velocities are directed toward each other. In this restricted context, the Enskog kinetic theory results from a neglect of velocity correlations, i.e., the Enskog approximation

$$f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_2,\mathbf{v}_2;t)\to\chi_{ij}(\mathbf{r}_1,\mathbf{r}_2|\{n_i\})f_i(\mathbf{r}_1,\mathbf{v}_1;t)f_j(\mathbf{r}_2,\mathbf{v}_2;t). \tag{3.6}$$

Spatial correlations arising from volume exclusion effects are retained through the factor $\chi_{ij}(\mathbf{r}_1,\mathbf{r}_2|\{n_i\})$. In the special case of a uniform system, it is simply related to the nonequilibrium pair correlation function $g_{ij}(|\mathbf{r}_1-\mathbf{r}_2|;\{n_i\})$ (probability density to find a particle of species i at $\mathbf{r}_1$ and j at $\mathbf{r}_2$) by [53]

$$g_{ij}(\sigma_{ij};\{n_k\}) = \frac{1+\alpha_{ij}}{2\alpha_{ij}}\chi_{ij}(\sigma_{ij};\{n_k\}). \tag{3.7}$$

This relationship is proved in Appendix A and provides some partial interpretation for χ_{ij}. It is important to note that these correlation functions are *functionals* of the actual species densities $\{n_i\}$ [defined below in Eq. (4.1)]. This functional dependence is what distinguishes the RET from the original "standard" Enskog theory (SET), where the g_{ij} are *functions* of the species densities at the single position of interest, $\mathbf{r}_1$. Some partial justification for the approximation (3.6) for ordinary atomic fluids is given in Appendix A, where it is known to provide accurate results for moderately dense gases, and reasonable estimates even for dense gases. Its use for granular gases is justified largely from expectations based on these results for ordinary fluids.

Substitution of the Enskog approximation (3.6) into the exact first level hierarchy equations (3.1) defines the RET for the distribution functions $\{f_i\}$

$$[\partial_t + \mathbf{v}_1\cdot\nabla + m_i^{-1}\mathbf{F}_i(\mathbf{r}_1)\cdot\nabla_{\mathbf{v}_1}]f_i(\mathbf{r}_1,\mathbf{v}_1;t) = \sum_{j=1}^{s}J_{ij}[\mathbf{r}_1,\mathbf{v}_1|f(t)]. \tag{3.8}$$

The collision operators $\{J_{ij}[\mathbf{r}_1,\mathbf{v}_1|f(t)]\}$ are given by

$$\begin{aligned}J_{ij}[\mathbf{r}_1,\mathbf{v}_1|f(t)] \equiv \sigma_{ij}^{d-1}\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})&\\ \times[\alpha_{ij}^{-2}\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_i\})&\\ \times f_i(\mathbf{r}_1,\mathbf{v}_1'';t)f_j(\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2'';t)&\\ -\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij}|\{n_i\})f_i(\mathbf{r}_1,\mathbf{v}_1;t)&\\ \times f_j(\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)].&\end{aligned} \tag{3.9}$$

The corresponding Boltzmann equations for a dilute mixture follow from this result since $\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_i\})\to 1$ at low density. Furthermore, on length scales of the order of the mean free path or greater, the different centers $(\mathbf{r}_1,\mathbf{r}_2=\mathbf{r}_1\pm\boldsymbol{\sigma}_{ij})$ of the colliding pair in Eq. (3.9) can be neglected $(\mathbf{r}_1\approx\mathbf{r}_2)$ since the diameters of the particles are small compared to the mean free path at low density. As will be shown below, a nonzero distance between the particle centers gives rise to the collisional contributions to the transport coefficients, which are not present in dilute systems. These two modifications to f_{ij} result in the usual Boltzmann description for a granular mixture. The results obtained here therefore encompass earlier work on granular mixtures at low density [38]. In the elastic limit, $\alpha_{ij}\to 1$, these equations become the Enskog theory for mixtures of dense molecular gases studied in Ref. [49].

As happens for elastic collisions, the inelastic Enskog equation provides a semiquantitative description of the hard sphere system that neglects the velocity correlations between the particles that are about to collide (molecular chaos assumption). The Enskog approximation is expected to be valid for short times since as the system evolves corrections to the Enskog equation due to multiparticle collisions, including recollision events ("ring" collisions), should be incorporated. The latter are expected to be stronger for fluids with inelastic collisions where the colliding pairs tend to be more focused. Therefore some deviations from molecular chaos have been observed in molecular dynamics (MD) simulations [54–56] of granular fluids as the density increases. Although the existence of these correlations restricts the range of validity of the Enskog equation, there is substantial evidence in the literature for the validity of the Enskog theory at moderate densities and higher restitution coefficients especially at the level of macroscopic properties (such as transport coefficients). In the case of molecular dynamics (MD) simulations, the Enskog theory compares quite well with simulations for the radial distribution function [53], the self-diffusion coefficient [57,58], the kinetic temperatures of a binary mixture in homogeneous cooling state [59], and the rheological properties of a mixture under

simple shear flow [60,61]. The agreement between the MD and Enskog equation is good for moderate densities (solid volume fraction up to 0.15) and even conditions of strong dissipation (restitution coefficients $\alpha_{ij}>0.7$). For higher densities the α range is more limited but the Enskog theory still captures the relevant qualitative features. The Enskog transport coefficients for a monocomponent gas [62] have also been tested against NMR experiments of a system of mustard seeds vibrated vertically [43,44]. The average value of the coefficient of restitution of the grains used in this experiment is $\alpha=0.87$, which lies outside of the quasielastic limit ($\alpha \approx 0.99$). Comparison between theory and experiments shows that the Enskog kinetic theory successfully models the density and granular temperature profiles away from the vibrating container bottom and quantitatively explains the temperature inversion observed in experiments [63]. All these results clearly show the applicability of the Enskog theory for densities outside the Boltzmann limit and values of dissipation beyond the quasielastic limit. In this context, one can conclude that the Enskog equation provides a unique basis for the description of dynamics across a wide range of densities, length scales, and degrees of dissipation. No other theory with such generality exists.

IV. MACROSCOPIC BALANCE EQUATIONS

In the previous section, the Enskog assumption (3.6) was used to obtain a closed set of kinetic equations (3.8) for a moderately dense mixture of inelastic hard spheres. The result takes the form of nonlinear, integral-differential equations for the distribution function f_i, which contains information on a *microscopic* scale. In this section, this theory will be used to obtain the corresponding description on the *macroscopic* (or hydrodynamic) scale. First, the relevant macroscopic variables will be identified and defined. Next, the corresponding balance equations will be derived. Finally, expressions for the equations of state (pressure and cooling rate) and fluxes will be presented as integral expressions containing f_i.

The variables of interest for a macroscopic description of the mixture are the number densities for all species, $\{n_i(\mathbf{r},t)\}$ [or equivalently, the mass densities $\{\rho_i(\mathbf{r},t)=m_i n_i(\mathbf{r},t)\}$], the total energy density, $e(\mathbf{r},t)$, and the total momentum, $\mathbf{p}(\mathbf{r},t)$. These are expected to be the $s+1+d$ slow variables that dominate the dynamics for long times through a closed autonomous set of equations, the hydrodynamic equations. The reasoning behind this is that these are the densities for global conserved quantities in molecular fluids, and therefore have decay times set by the wavelength of the excitations. Long wavelength (space scales large compared to the mean free path) phenomena therefore persist at long times (compared to a mean free time) after which the complex transient microscopic dynamics has become negligible. For granular fluids, the energy is not conserved but is characterized by a cooling rate at long wavelengths. Still, this cooling rate may be slow compared to the transient dynamics and thus the energy remains a relevant slow variable. This is confirmed by MD simulations showing a rapid approach to this cooling law after only a few collisions [59].

These macroscopic variables will be referred to collectively as the hydrodynamic fields. They are defined without approximation in terms of moments of the distribution functions

$$n_i(\mathbf{r},t) \equiv \int d\mathbf{v} f_i(\mathbf{r},\mathbf{v};t), \quad i=1,\ldots,s, \tag{4.1}$$

$$e(\mathbf{r},t) \equiv \sum_{i=1}^{s} \int d\mathbf{v} \frac{1}{2} m_i v^2 f_i(\mathbf{r},\mathbf{v};t), \tag{4.2}$$

$$\mathbf{p}(\mathbf{r},t) \equiv \sum_{i=1}^{s} \int d\mathbf{v} m_i \mathbf{v} f_i(\mathbf{r},\mathbf{v};t). \tag{4.3}$$

The time dependence occurs entirely through the distribution function and hence is determined from the Enskog kinetic equations (3.8). However, rather than solving the kinetic equation to determine this complete time dependence it is useful for the purposes of deriving the simpler hydrodynamic description to first obtain the balance equations. These equations express the time derivative of the hydrodynamic fields in terms of local fluxes and sources due to collisions or the external force. These equations and the identification of the fluxes follow in detail from the form of the collision operators in Eq. (3.2) as shown in Appendix B [in fact they are obtained there exactly from the first hierarchy equation (3.1) without the Enskog approximation (3.6) and hence are exact]. The results for the balance equations are

$$\partial_t n_i(\mathbf{r},t) + m_i^{-1} \nabla \cdot \mathbf{j}_i(\mathbf{r},t) = 0, \tag{4.4}$$

$$\partial_t e(\mathbf{r},t) + \nabla \cdot \mathbf{s}(\mathbf{r},t) = -w(\mathbf{r},t) + \sum_{i=1}^{s} m_i^{-1} \mathbf{F}_i(\mathbf{r}) \cdot \mathbf{j}_i(\mathbf{r},t), \tag{4.5}$$

$$\partial_t p_\beta(\mathbf{r},t) + \partial_{r_\gamma} t_{\gamma\beta}(\mathbf{r},t) = \sum_{i=1}^{s} n_i(\mathbf{r},t) F_{i\beta}(\mathbf{r}). \tag{4.6}$$

The explicit expressions for $\mathbf{j}_i$, $\mathbf{s}$, w, and $t_{\gamma\beta}$ are contained in Appendix B and not shown here since they are cast in a more convenient form below.

The mass fluxes $\{\mathbf{j}_i(\mathbf{r},t)\}$, energy flux $\mathbf{s}(\mathbf{r},t)$, and momentum flux $t_{\beta\gamma}(\mathbf{r},t)$ describe the rate of transport of the hydrodynamic fields through a given cross-sectional area. They consist of parts due to pure convection and parts due to collision. To identify the convective (kinetic) parts, the local flow field $\mathbf{U}(\mathbf{r},t)$ is defined in terms of the momentum density by

$$\mathbf{p}(\mathbf{r},t) \equiv \rho(\mathbf{r},t)\mathbf{U}(\mathbf{r},t), \quad \rho(\mathbf{r},t) = \sum_{i=1}^{s} m_i n_i(\mathbf{r},t), \tag{4.7}$$

where the second equation defines the mass density. Also, the energy density is written in terms of the internal energy density $e_0(\mathbf{r},t)$ in the local rest frame, plus the energy due to flow

$$e(\mathbf{r},t) = e_0(\mathbf{r},t) + \frac{1}{2}\rho(\mathbf{r},t)U^2(\mathbf{r},t). \tag{4.8}$$

In terms of $\mathbf{U}(\mathbf{r},t)$ the fluxes become

$$\mathbf{j}_i(\mathbf{r},t) = \rho_i(\mathbf{r},t)\mathbf{U}(\mathbf{r},t) + \mathbf{j}_{0i}(\mathbf{r},t), \tag{4.9}$$

$$s_\beta(\mathbf{r},t) = \left(e_0(\mathbf{r},t) + \frac{1}{2}\rho(\mathbf{r},t)U^2(\mathbf{r},t)\right)U_\beta(\mathbf{r},t) + P_{\beta\gamma}(\mathbf{r},t)U_\gamma(\mathbf{r},t) + q_\beta(\mathbf{r},t), \tag{4.10}$$

$$t_{\beta\gamma}(\mathbf{r},t) = \rho(\mathbf{r},t)U_\beta(\mathbf{r},t)U_\gamma(\mathbf{r},t) + P_{\beta\gamma}(\mathbf{r},t). \tag{4.11}$$

The first terms on the right sides describe convective transport, while the diffusion fluxes $\mathbf{j}_{0i}(\mathbf{r},t)$, heat flux $\mathbf{q}(\mathbf{r},t)$, and pressure tensor $P_{\beta\gamma}(\mathbf{r}_1,t)$ describe the residual transport for each fluid element in its local rest frame. Before giving their forms more explicitly, it is instructive to insert Eqs. (4.9)–(4.11) into Eqs. (4.4)–(4.6) to get the equivalent form for the balance equations

$$D_t n_i + n_i\nabla\cdot\mathbf{U} + m_i^{-1}\nabla\cdot\mathbf{j}_{0i} = 0, \tag{4.12}$$

$$D_t e_0 + (e_0\delta_{\gamma\beta} + P_{\gamma\beta})\partial_{r_\gamma}U_\beta + \nabla\cdot\mathbf{q} = -w(\mathbf{r},t) + \sum_{i=1}^{s} m_i^{-1}\mathbf{F}_i(\mathbf{r})\cdot\mathbf{j}_{0i}(\mathbf{r},t), \tag{4.13}$$

$$\rho D_t U_\beta + \partial_{r_\gamma}P_{\gamma\beta} = \sum_{i=1}^{s} n_i(\mathbf{r},t)F_{i\beta}(\mathbf{r}), \tag{4.14}$$

where $D_t = \partial_t + \mathbf{U}\cdot\nabla$ is the material derivative.

The independent hydrodynamic fields are now $\{n_i(\mathbf{r},t)\}$, $e_0(\mathbf{r},t)$, and $\mathbf{U}(\mathbf{r},t)$. The remaining quantities in the balance equations are the energy loss rate $w(\mathbf{r},t)$, the mass fluxes $\{\mathbf{j}_{0i}(\mathbf{r},t)\}$, the heat flux $\mathbf{q}(\mathbf{r},t)$, and the pressure tensor $P_{\beta\gamma}(\mathbf{r}_1,t)$. These quantities, which are defined in terms of the distribution functions, are obtained by the explicit forms for $\mathbf{j}_i$, $\mathbf{s}$, w, and $t_{\gamma\beta}$ given in Appendix B together with Eqs. (4.9)–(4.11).

Specifically, the energy loss rate is due to inelastic collisions

$$w(\mathbf{r},t) \equiv \frac{1}{4}\sum_{i,j=1}^{s}(1-\alpha_{ij}^2)m_i\mu_{ji}\sigma_{ij}^{d-1}\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}} \times \Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})^3 f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t), \tag{4.15}$$

whereas the diffusion flux arises from convective (kinetic) transport

$$\mathbf{j}_{0i}(\mathbf{r}_1,t) \equiv m_i\int d\mathbf{v}_1\mathbf{V}_1 f_i(\mathbf{r}_1,\mathbf{v}_1;t), \tag{4.16}$$

where $\mathbf{V}_1 = \mathbf{v}_1 - \mathbf{U}(\mathbf{r},t)$ is the velocity in the local rest frame. The heat flux has both "kinetic" and "collisional" transfer contributions

$$\mathbf{q}(\mathbf{r}_1,t) \equiv \mathbf{q}^k(\mathbf{r}_1,t) + \mathbf{q}^c(\mathbf{r}_1,t), \tag{4.17}$$

with

$$\mathbf{q}^k(\mathbf{r}_1,t) = \sum_{i=1}^{s}\int d\mathbf{v}_1\frac{1}{2}m_i V_1^2\mathbf{V}_1 f_i(\mathbf{r}_1,\mathbf{v}_1;t), \tag{4.18}$$

$$\mathbf{q}^c(\mathbf{r}_1,t) = \sum_{i,j=1}^{k}\frac{1}{8}(1+\alpha_{ij})m_j\mu_{ij}\sigma_{ij}^d \times\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})^2[(1-\alpha_{ij}) \times(\mu_{ji}-\mu_{ij})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12}) + 4\hat{\boldsymbol{\sigma}}\cdot\mathbf{G}_{ij}]\hat{\boldsymbol{\sigma}}\int_0^1 dx f_{ij} \times(\mathbf{r}_1 - x\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t), \tag{4.19}$$

where $\mathbf{G}_{ij} = \mu_{ij}\mathbf{V}_1 + \mu_{ji}\mathbf{V}_2$ is the center-of-mass velocity.

Similarly, the pressure tensor has both kinetic and collisional contributions

$$P_{\gamma\beta}(\mathbf{r}_1,t) \equiv P_{\gamma\beta}^k(\mathbf{r}_1,t) + P_{\gamma\beta}^c(\mathbf{r}_1,t), \tag{4.20}$$

where

$$P_{\gamma\beta}^k(\mathbf{r}_1,t) = \sum_{i=1}^{s}\int d\mathbf{v}_1 m_i V_{1\beta}V_{1\gamma}f_i(\mathbf{r}_1,\mathbf{v}_1;t), \tag{4.21}$$

$$P_{\gamma\beta}^c(\mathbf{r}_1,t) = \frac{1}{2}\sum_{i,j=1}^{s} m_j\mu_{ij}(1+\alpha_{ij})\sigma_{ij}^d \times\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})^2\hat{\sigma}_\beta\hat{\sigma}_\gamma \times\int_0^1 dx f_{ij}(\mathbf{r}_1 - x\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t). \tag{4.22}$$

Equations (4.12)–(4.14) together with the definitions (4.15)–(4.22) represent the macroscopic balance equations for a granular mixture, without restrictions on the densities or degrees of dissipation. In the case of a three-dimensional system ($d=3$), the above equations reduce to previous results [64] derived for hard spheres. When the approximate form (3.6) is used in the first hierarchy equation and in these expressions for the cooling rate and fluxes, the Enskog theory results.

For historical consistency with the usual constitutive equations for an ordinary fluid, the temperature $T(\mathbf{r},t)$ is used in the following instead of the internal energy density $e_0(\mathbf{r},t)$, with the definition

$$e_0(\mathbf{r},t) \equiv \frac{d}{2}n(\mathbf{r},t)T(\mathbf{r},t). \tag{4.23}$$

As a definition, this amounts only to a change of variables and there are no thermodynamic implications involved in the use of this temperature for a granular fluid. The correspond-

ing hydrodynamic equation for $T(\mathbf{r},t)$ follows directly from Eq. (4.13),

$$\frac{d}{2}n(D_t+\zeta)T+P_{\gamma\beta}\partial_{r_\gamma}U_\beta+\nabla\cdot\mathbf{q}-\frac{d}{2}T\sum_{i=1}^{s}m_i^{-1}\nabla\cdot\mathbf{j}_{0i}=\sum_{i=1}^{s}m_i^{-1}\mathbf{F}_i\cdot\mathbf{j}_{0i}. \tag{4.24}$$

To obtain these results the continuity equation has been used,

$$D_t\rho+\rho\nabla\cdot\mathbf{U}=0. \tag{4.25}$$

This follows from the definitions of ρ and $\mathbf{U}$ and the conservation laws for the $\{n_i(\mathbf{r},t)\}$. A related consequence is

$$\sum_{i=1}^{s}\mathbf{j}_{0i}=0, \tag{4.26}$$

so that only $s-1$ dissipative mass fluxes are independent. Finally, the "cooling rate" ζ has been introduced in Eq. (4.24) by the definition

$$\zeta=\frac{2}{dnT}w=\frac{1}{2dnT}\sum_{i,j=1}^{s}(1-\alpha_{ij}^2)m_i\mu_{ji}\sigma_{ij}^{d-1}\times\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})^3\times f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t). \tag{4.27}$$

V. CONCEPT OF A NORMAL SOLUTION AND HYDRODYNAMICS

The form of the equations of state and fluxes given in the previous section, Eqs. (4.16)–(4.22) and (4.27), are cast as functionals of the distributions $\{f_i\}$, which depend explicitly on space and time. As a result, the macroscopic balance equations are not entirely expressed in terms of the hydrodynamic fields, and thus do not comprise a closed set of equations. If these distributions can instead be expressed as functionals of the hydrodynamic fields (*normal* solution), then $\zeta(\mathbf{r},t)$, $\{\mathbf{j}_{0i}(\mathbf{r},t)\}$, $\mathbf{q}(\mathbf{r},t)$, and $P_{\beta\gamma}(\mathbf{r}_1,t)$ also will become functionals of the hydrodynamic fields through Eqs. (4.16)–(4.22) and (4.27). Such expressions are called "constitutive relations." They provide the missing link between the balance equations and a closed set of equations for the hydrodynamic fields alone. Such a closed set of equations defines "hydrodynamics" in its most general sense.

It is seen, therefore, that any derivation of hydrodynamics proceeds first by construction of normal solutions to the kinetic equations. More precisely, a normal solution is one whose space and time dependence occurs entirely through the hydrodynamic fields, denoted

$$f_i(\mathbf{r}_1,\mathbf{v}_1;t)=f_i(\mathbf{v}_1|\{y_\beta(\mathbf{r}_1,t)\}), \tag{5.1}$$

where $\{y_\beta(\mathbf{r}_1,t)\}$ denotes generically the set of hydrodynamic fields

$$y_\beta\Leftrightarrow\{T,\mathbf{U},\{n_i(\mathbf{r},t)\}\}. \tag{5.2}$$

Therefore the space and time derivatives of the kinetic equation are given by

$$(\partial_t+\mathbf{v}_1\cdot\nabla_{\mathbf{r}})f_i(\mathbf{v}_1|\{y_\beta(\mathbf{r}_1,t)\})=\int d\mathbf{r}\frac{\delta f_i(\mathbf{v}_1|\{y_\beta(t)\})}{\delta y_\eta(\mathbf{r};t)}\times(\partial_t+\mathbf{v}_1\cdot\nabla_{\mathbf{r}})y_\eta(\mathbf{r};t). \tag{5.3}$$

Furthermore, the balance equations for the hydrodynamic fields (4.12)–(4.14) can be used to express $\partial_t y_\eta(\mathbf{r};t)$ in Eq. (5.3) in terms of space derivatives of the hydrodynamic fields. For such a solution for f_i, Eqs. (4.16)–(4.22) and (4.27) give directly by integration the desired constitutive relations.

The determination of $f_i(\mathbf{v}_1|\{y_\beta(\mathbf{r}_1,t)\})$ from the kinetic equations (3.8) is a very difficult task in general, and further restriction on the class of problems considered is required at this point to make progress. Any *functional* of the fields can be represented equivalently as a local *function* of the fields *and* all of their gradients. In many cases, gradients of high degree are small and may be negligible so that the normal distribution becomes

$$f_i(\mathbf{v}_1|\{y_\beta(\mathbf{r}_1,t)\})\to f_i(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t),\nabla_{\mathbf{r}_1}y_\beta(\mathbf{r}_1,t),\ldots\}). \tag{5.4}$$

This representation does not imply that the low degree gradients are small, and f_i may be a nonlinear function of the relevant gradients. This occurs in many important applications for granular fluids [40]. In the limiting case where the low-degree gradients can be controlled by boundary or initial conditions and made small, a further Taylor series expansion can be given,

$$\begin{aligned}f_i(\mathbf{v}_1|\{y_\beta(\mathbf{r}_1,t)\})&\to f_i^{(0)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\})+f_i^{(1)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\})+\cdots\\&\to f_i^{(0)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\})+\mathbf{Y}_{i\alpha}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\})\cdot\nabla_{\mathbf{r}_1}y_\alpha(\mathbf{r}_1,t)+\cdots.\end{aligned} \tag{5.5}$$

It follows that the leading order distributions have the exact properties

$$n_i(\mathbf{r},t)\equiv\int d\mathbf{v}f_i^{(0)}(\mathbf{v};\{y_\beta(\mathbf{r},t)\}),\quad i=1,\ldots,s, \tag{5.6}$$

$$\frac{d}{2}n(\mathbf{r},t)T(\mathbf{r},t)\equiv\sum_{i=1}^{s}\int d\mathbf{v}\frac{1}{2}m_iV^2f_i^{(0)}(\mathbf{v};\{y_\beta(\mathbf{r},t)\}), \tag{5.7}$$

$$\rho(\mathbf{r},t)\mathbf{U}(\mathbf{r},t)\equiv\sum_{i=1}^{s}\int d\mathbf{v}m_i\mathbf{v}f_i^{(0)}(\mathbf{v};\{y_\beta(\mathbf{r},t)\}), \tag{5.8}$$

and the corresponding moments of all higher order terms in Eq. (5.5) must vanish. Generalization of this type of gradient

expansion for the normal solution to include a class of nonlinear gradients in the reference state has been discussed recently [65,66].

As is standard for molecular gases, the gradient expansion will be taken with respect to the reference local HCS, i.e., that resulting from the neglect of all gradients in the functional but evaluated at the value of the fields at the chosen point and time $\{y_\beta(\mathbf{r}_1,t)\}$. This point is crucial in our analysis since most of the previous results have taken elastic Maxwell distributions as the base state. Note that in the CE method the form of $f_i^{(0)}$ comes from the solution to the kinetic equation to zeroth order in gradients and cannot be chosen *a priori*. Accordingly, $f_i^{(0)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\}) \to f_i^{(0)}(V_1;\{y_\beta(\mathbf{r}_1,t)\})$, where $V=|\mathbf{v}-\mathbf{U}(\mathbf{r},t)|$ is homogeneous and isotropic with respect to its velocity dependence. This symmetry implies that the leading (zero) order contributions to Eqs. (4.16) and (4.17) for the vector fluxes $\{\mathbf{j}_{0i}(\mathbf{r},t)\}$ and $\mathbf{q}(\mathbf{r},t)$ must vanish, and this contribution to the pressure tensor $P_{\gamma\beta}$ must be isotropic (proportional to $\delta_{\gamma\beta}$). Similar symmetry considerations to the first order contribution (linear in the gradients) determines the exact structure of the constitutive equation to this order. Based on these symmetry considerations, the constitutive quantities are known to take the forms

$$\zeta(\mathbf{r},t) \to \zeta^{(0)}(\{y_\beta(\mathbf{r},t)\}) + \zeta_U(\{y_\beta(\mathbf{r},t)\})\nabla\cdot\mathbf{U}(\mathbf{r},t), \tag{5.9}$$

$$\mathbf{j}_{0i}(\mathbf{r},t) \to -\sum_{j=1}^{s} m_i m_j \frac{n_j(\mathbf{r},t)}{\rho(\mathbf{r},t)} D_{ij}(\{y_\beta(\mathbf{r},t)\})\nabla \ln n_j(\mathbf{r},t) - \rho(\mathbf{r},t) D_i^T(\{y_\beta(\mathbf{r},t)\})\nabla \ln T(\mathbf{r},t) - \sum_{j=1}^{s} D_{ij}^F(\{y_\beta(\mathbf{r},t)\})\mathbf{F}_j(\mathbf{r}), \tag{5.10}$$

$$\mathbf{q}(\mathbf{r},t) \to -\lambda(\{y_\beta(\mathbf{r},t)\})\nabla T(\mathbf{r},t) - \sum_{i,j=1}^{s} [T^2(\mathbf{r},t) D_{q,ij}(\{y_\beta(\mathbf{r},t)\})\nabla \ln n_j(\mathbf{r},t) + L_{ij}(\{y_\beta(\mathbf{r},t)\})\mathbf{F}_j(\mathbf{r})], \tag{5.11}$$

$$P_{\gamma\lambda}(\mathbf{r},t) = p(\{y_\beta(\mathbf{r},t)\})\delta_{\gamma\lambda} - \eta(\{y_\beta(\mathbf{r},t)\})\left(\partial_{r_\gamma} U_\lambda(\mathbf{r},t) + \partial_{r_\lambda} U_\gamma(\mathbf{r},t) - \frac{2}{d}\nabla\cdot\mathbf{U}(\mathbf{r},t)\right) - \kappa(\{y_\beta(\mathbf{r},t)\})\nabla\cdot\mathbf{U}(\mathbf{r},t). \tag{5.12}$$

The unknown quantities in these constitutive equations (5.9)–(5.12) include the cooling rate $\zeta^{(0)}(\{y_\beta(\mathbf{r},t)\})$, the hydrostatic granular pressure $p(\{y_\beta(\mathbf{r},t)\})$, and the transport coefficients ζ_U, D_{ij}, D_i^T, D_{ij}^F, $D_{q,ij}$, L_{ij}, η, and κ. These quantities can be expressed as explicit functions of the hydrodynamic variables once $f_i^{(0)}$ and $f_i^{(1)}$ are known. The equations governing the solution of $f_i^{(0)}$ and $f_i^{(1)}$ are found using the CE method, as described below.

VI. CHAPMAN-ENSKOG NORMAL SOLUTION

The CE method is a procedure for constructing an approximate normal solution. It is perturbative, using the spatial gradients as the small expansion parameter. More precisely, the small parameter is Knudsen number (Kn), defined as the gradient of the hydrodynamic fields relative to their local value times the mean free path. This means that the conditions for the solution are restricted to small variations of the hydrodynamic fields over distances of the order of the mean free path. In the presence of an external force it is necessary to characterize the magnitude of this force relative to the gradients as well. Here, it is assumed that the magnitude of the force is first order in perturbation expansion. This allows comparison with the results of Ref. [49] for the elastic case.

The perturbation is carried out by considering the Enskog kinetic equations successively at each order in the gradients. As described below, the zeroth order equation is first obtained for $f_i^{(0)}$. Next, the first order equation for $f_i^{(1)}$ is obtained. This expansion leads to integral-differential equations for the determination of $f_i^{(0)}$ and $f_i^{(1)}$, which are solved explicitly in the follow-up paper [50].

As detailed in Appendix C, to zeroth order in the gradients, the kinetic equation (3.8) becomes

$$(\partial_t T)\partial_T f_i^{(0)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\}) = \sum_{j=1}^{s} J_{ij}^{(0)}[\mathbf{v}_1|f^{(0)}(t)], \tag{6.1}$$

where

$$J_{ij}^{(0)}[\mathbf{v}_1|f^{(0)}(t)] \equiv \chi_{ij}^{(0)}(\sigma_{ij};\{n_i(\mathbf{r}_1,t)\})\sigma_{ij}^{d-1} \times \int d\mathbf{v}_2 \int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12}) \times [\alpha_{ij}^{-2} f_i^{(0)}(V_1'';\{y_\beta(\mathbf{r}_1,t)\}) f_j^{(0)}(V_2'';\{y_\beta(\mathbf{r}_1,t)\}) - f_i^{(0)}(V_1;\{y_\beta(\mathbf{r}_1,t)\}) f_j^{(0)}(V_2;\{y_\beta(\mathbf{r}_1,t)\})]. \tag{6.2}$$

All spatial gradients are neglected at this lowest order. Equation (6.1) determines the velocity dependence of $f_i^{(0)}(V_1;\{y_\beta(\mathbf{r}_1,t)\})$; the space and time dependence is local and entirely through the fields $\{y_\beta(\mathbf{r}_1,t)\}$ at the space and time point of interest. This has been exploited by writing the time dependence of $f_i^{(0)}$ in terms of the time dependence of the fields, and recognizing that all time derivatives of the latter are proportional to space gradients, except the temperature, through the balance equations

$$\partial_t n_i = 0, \quad \partial_t T = -\zeta^{(0)} T, \quad \partial_t \mathbf{U} = \mathbf{0}. \tag{6.3}$$

Here, $\zeta^{(0)}$ is the cooling rate (4.27) to zeroth order in the gradients

$$\zeta^{(0)} = \frac{1}{2dnT}\sum_{i,j=1}^{s}(1-\alpha_{ij}^2)m_i\mu_{ji}\chi_{ij}^{(0)}(\sigma_{ij};\{n_i(\mathbf{r}_1,t)\})\sigma_{ij}^{d-1}$$
$$\times\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})^3$$
$$\times f_i^{(0)}(V_1;\{y_\beta(\mathbf{r}_1,t)\})f_j^{(0)}(V_2;\{y_\beta(\mathbf{r}_1,t)\}). \tag{6.4}$$

Similarly, the functional dependence of $\chi_{ij}^{(0)}(\mathbf{r}_1,\mathbf{r}_2|\{n_i\})$ on the compositions to zeroth order in the gradients has the functional dependence on the densities replaced by $\{n_i\}\to\{n_i(\mathbf{r}_1,t)\}$ at the point of interest. The result is translational and rotational invariant $\chi_{ij}^{(0)}(\mathbf{r}_1,\mathbf{r}_2|\{n_i\})\to\chi_{ij}^{(0)}(|\mathbf{r}_1-\mathbf{r}_2|;\{n_i(\mathbf{r}_1,t)\})$, a *function* of the densities. Finally, gradients in the distribution functions of the collision operators must be neglected, e.g., $f_i^{(0)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1+\sigma_{ij},t)\})\to f_i^{(0)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\})$.

A further simplification of these equations for the lowest order distribution functions occurs when they are written in terms of the corresponding dimensionless forms $\{\phi_i\}$

$$f_i^{(0)}(V;\{y_\beta\}) = n_i v_0^{-d}(T)\phi_i(V^*;\{n_i^*\}), \tag{6.5}$$

with the definitions

$$\mathbf{V}^* = \frac{\mathbf{V}}{v_0(T)},\quad v_0(T)=\sqrt{\frac{2T}{m}},\quad n_i^* = n_i\sigma_i^d,\quad m=\frac{1}{s}\sum_{i=1}^{s}m_i. \tag{6.6}$$

The solution depends on the flow field only through the relative velocity $\mathbf{V}$. Furthermore, since there is no external energy scale the temperature can occur only through the scaling of the dimensionless velocity through the thermal velocity $v_0(T)$. Equation (6.1) now takes the dimensionless form

$$-\frac{1}{2}\zeta^*\nabla_{\mathbf{V}^*}\cdot(\mathbf{V}^*\phi_i) = \sum_j J_{ij}^{(0)*}(V^*|\phi_i), \tag{6.7}$$

where $\nabla_{\mathbf{V}^*}\equiv\partial/\partial\mathbf{V}^*$ and

$$\zeta^* = \frac{\ell}{v_0}\zeta^{(0)},\quad J_{ij}^{(0)*} = \frac{\ell}{n}v_0^{d-1}J_{ij}^{(0)},\quad \ell = \frac{1}{n\sigma^{d-1}},$$
$$n=\sum_{i=1}^{s}n_i,\quad \sigma=\frac{1}{s}\sum_{i=1}^{s}\sigma_i. \tag{6.8}$$

The solution to this equation is a universal function of the magnitude of the velocity V^* and is otherwise independent of the temperature and flow field. For a one component fluid it is independent of the density as well. However, for mixtures it is parametrized by the dimensionless species densities through the factors $\chi_{ij}^{(0)}(\sigma_{ij};\{n_i^*\})$. Equation (6.1) has the same form as the corresponding dimensionless Enskog equations for a strictly *homogeneous* state. The latter is called the HCS. Here, however, the state is not homogeneous because of the requirements (5.6)–(5.8). Instead it is a *local* HCS. As said before, an important point to recognize is that the occurrence of this local HCS as the reference state is not an assumption of the CE expansion but rather a consequence of the kinetic equations to lowest order in the gradient expansion.

The analysis to first order in the gradients is similar and the details are given in Appendix C. The result has the form (5.5)

$$f_i^{(1)}\to\boldsymbol{\mathcal{A}}_i(\mathbf{V})\cdot\nabla\ln T+\sum_{j=1}^{s}\boldsymbol{\mathcal{B}}_i^j(\mathbf{V})\cdot\nabla\ln n_j$$
$$+\mathcal{C}_{i,\gamma\eta}(\mathbf{V})\frac{1}{2}\left(\partial_\gamma U_\eta+\partial_\eta U_\gamma-\frac{2}{d}\delta_{\gamma\eta}\nabla\cdot\mathbf{U}\right)$$
$$+\mathcal{D}_i(\mathbf{V})\nabla\cdot\mathbf{U}+\sum_{j=1}^{s}\boldsymbol{\mathcal{E}}_i^j(\mathbf{V})\cdot\mathbf{F}_j. \tag{6.9}$$

The contributions from the flow field gradients have been separated into independent traceless and diagonal components, as follows from fluid symmetry. The velocity dependence of the gradient contributions is contained in the functions $\boldsymbol{\mathcal{A}}_i(\mathbf{V})$, $\boldsymbol{\mathcal{B}}_i^j(\mathbf{V})$, $\mathcal{C}_{i,\gamma\eta}(\mathbf{V})$, $\mathcal{D}_i(\mathbf{V})$, and $\boldsymbol{\mathcal{E}}_i^j(\mathbf{V})$. The kinetic equations determine these functions as the solutions to the integral equations

$$\left(\left(\mathcal{L}-\frac{1}{2}\zeta^{(0)}\right)\boldsymbol{\mathcal{A}}\right)_i = \mathbf{A}_i, \tag{6.10}$$

$$(\mathcal{L}\boldsymbol{\mathcal{B}}^j)_i - n_j\frac{\partial\zeta^{(0)}}{\partial n_j}\boldsymbol{\mathcal{A}}_i = \mathbf{B}_i^j, \tag{6.11}$$

$$\left(\left(\mathcal{L}+\frac{1}{2}\zeta^{(0)}\right)\mathcal{C}_{\gamma\eta}\right)_i = C_{i,\gamma\eta}, \tag{6.12}$$

$$\left(\left(\mathcal{L}+\frac{1}{2}\zeta^{(0)}\right)\mathcal{D}\right)_i = D_i, \tag{6.13}$$

$$((\mathcal{L}+\zeta^{(0)})\boldsymbol{\mathcal{E}}^j)_i = \mathbf{E}_i^j. \tag{6.14}$$

The linear operator $\mathcal{L}$ is given by

$$(\mathcal{L}X)_i = \frac{1}{2}\zeta^{(0)}\nabla_{\mathbf{V}}\cdot(\mathbf{V}X_i)+(LX)_i, \tag{6.15}$$

$$(LX)_i = -\sum_{j=1}^{s}(J_{ij}^{(0)}[\mathbf{v}_1|X_i,f_j^{(0)}]+J_{ij}^{(0)}[\mathbf{v}_1|f_i^{(0)},X_j]), \tag{6.16}$$

and the inhomogeneous terms are defined by

$$A_{i,\gamma}(\mathbf{V}) = \frac{1}{2}V_\gamma\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_i^{(0)})-\frac{p}{\rho}\partial_{V_\gamma}f_i^{(0)}$$
$$+\frac{1}{2}\sum_{j=1}^{s}\mathcal{K}_{ij,\gamma}[\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_j^{(0)})], \tag{6.17}$$

$$B_{i,\gamma}^j(\mathbf{V}) = -V_\gamma n_j\partial_{n_j}f_i^{(0)}-\rho^{-1}(\partial_{V_\gamma}f_i^{(0)})n_j(\partial_{n_j}p)$$
$$-\sum_{\ell=1}^{s}\mathcal{K}_{i\ell,\gamma}\left\{\left[n_j\partial_{n_j}+\frac{1}{2}\left(n_\ell\frac{\partial\ln\chi_{i\ell}^{(0)}}{\partial n_j}+I_{i\ell j}\right)\right]f_\ell^{(0)}\right\}, \tag{6.18}$$

$$C_{i,\gamma\beta}(\mathbf{V}) = \frac{1}{2}\left(V_\gamma \partial_{V_\beta} f_i^{(0)} + V_\beta \partial_{V_\gamma} f_i^{(0)} - \frac{2}{d}\delta_{\beta\gamma}\mathbf{V}\cdot\nabla_{\mathbf{V}} f_i^{(0)} \right) + \frac{1}{2}\sum_{j=1}^{s}\left(\mathcal{K}_{ij,\gamma}[\partial_{V_\beta} f_j^{(0)}] + \mathcal{K}_{ij,\beta}[\partial_{V_\gamma} f_j^{(0)}] - \frac{2}{d}\delta_{\beta\gamma}\mathcal{K}_{ij,\lambda}[\partial_{V_\lambda} f_j^{(0)}] \right), \tag{6.19}$$

$$D_i(\mathbf{V}) = \frac{1}{d}\mathbf{V}\cdot\nabla_{\mathbf{V}} f_i^{(0)} - \frac{1}{2}\left(\zeta_U + \frac{2}{nTd}p\right)\nabla_{\mathbf{V}}\cdot(\mathbf{V} f_i^{(0)}) + \sum_{j=1}^{s}\left(n_j\partial_{n_j} f_i^{(0)} + \frac{1}{d}\mathcal{K}_{ij,\gamma}[\partial_{V_\gamma} f_j^{(0)}] \right), \tag{6.20}$$

$$\mathbf{E}_i^j(\mathbf{V}) = -(\nabla_{\mathbf{V}} f_i^{(0)})\frac{1}{m_j}\left(\delta_{ij} - \frac{n_j m_j}{\rho}\right), \tag{6.21}$$

where the operator $\mathcal{K}_{ij,\gamma}[X]$ is defined by Eq. (C16).

This completes the construction of the normal solution to the revised Enskog equations up through first order in the gradients. Equation (6.7) determines the $f_i^{(0)}$ through the definition (6.5); the solution to the linear integral equations (6.10)–(6.14) determines the $f_i^{(1)}$ through the definition (6.9). The unknown fluxes and cooling rate of the hydrodynamic equations can then be calculated with these solutions. This is made explicit in the next section.

VII. CONSTITUTIVE EQUATIONS AND TRANSPORT COEFFICIENTS

The forms for the constitutive equations to first order in the gradients are given by Eqs. (5.9)–(5.12). The explicit representations for the coefficients in these equations are given in terms of the solutions to the integral equations for $f_i^{(0)}$ and $f_i^{(1)}$ of the previous section. Details of the simplification of these expressions in terms of $f_i^{(0)}$ and $f_i^{(1)}$ are given in Appendix F and only the final results are presented here.

Recall that the results below are based on the assumption that the external force is of the same magnitude as $f_i^{(1)}$; a force of different magnitude would result in different constitutive relations.

A. Cooling rate

The cooling rate is calculated from Eq. (4.27), resulting in the form (5.9)

$$\zeta \to \zeta^{(0)} + \zeta_U \nabla\cdot\mathbf{U}, \tag{7.1}$$

with

$$\zeta^{(0)} = \frac{B_3}{2dnT}\sum_{i,j=1}^{s}(1-\alpha_{ij}^2)\frac{m_i m_j}{m_i+m_j}\chi_{ij}^{(0)}\sigma_{ij}^{d-1} \times \int d\mathbf{v}_1 \int d\mathbf{v}_2 f_i^{(0)}(V_1) f_j^{(0)}(V_2) g_{12}^3, \tag{7.2}$$

and

$$\zeta_U = -\frac{d+2}{dnT}B_4\sum_{i,j=1}^{s}(1-\alpha_{ij}^2)\mu_{ji}\chi_{ij}^{(0)}\sigma_{ij}^{d} n_i n_j T_i^{(0)} + \frac{B_3}{dnT}\sum_{i,j=1}^{s}(1-\alpha_{ij}^2)\frac{m_i m_j}{m_i+m_j}\chi_{ij}^{(0)}\sigma_{ij}^{d-1} \times \int d\mathbf{v}_1 \int d\mathbf{v}_2 g_{12}^3 f_i^{(0)}(\mathbf{V}_1)\mathcal{D}_j(\mathbf{V}_2). \tag{7.3}$$

The constant B_n is defined by

$$B_n \equiv \pi^{(d-1)/2}\frac{\Gamma\left(\frac{n+1}{2}\right)}{\Gamma\left(\frac{n+d}{2}\right)}. \tag{7.4}$$

Also in Eq. (7.3) the species temperatures $\{T_i^{(0)}\}$ have been defined by

$$\frac{d}{2}n_i T_i^{(0)} = \int d\mathbf{v}\frac{1}{2}m_i V^2 f_i^{(0)}(\{n_i\},T,V). \tag{7.5}$$

There is no special significance to these quantities other than naming the integral on the right side, which is a specified function of the hydrodynamic fields $\{n_i\}$ and the global temperature T through $f_i^{(0)}$.

B. Mass fluxes

The mass fluxes are determined from the definition of Eq. (4.16) leading to the form (5.10) to first order in the gradients

$$\mathbf{j}_{0i} \to -\sum_{j=1}^{s} m_i m_j \frac{n_j}{\rho} D_{ij}\nabla \ln n_j - \rho D_i^T \nabla \ln T - \sum_{j=1}^{s} D_{ij}^F \mathbf{F}_j. \tag{7.6}$$

The transport coefficients are identified as

$$D_i^T = -\frac{m_i}{\rho d}\int d\mathbf{v}\mathbf{V}\cdot\boldsymbol{\mathcal{A}}_i(\mathbf{V}), \tag{7.7}$$

$$D_{ij} = -\frac{\rho}{m_j n_j d}\int d\mathbf{v}\mathbf{V}\cdot\boldsymbol{\mathcal{B}}_i^j(\mathbf{V}), \tag{7.8}$$

$$D_{ij}^F = -\frac{m_i}{d}\int d\mathbf{v}\mathbf{V}\cdot\boldsymbol{\mathcal{E}}_i^j(\mathbf{V}). \tag{7.9}$$

C. Energy flux

The energy flux to first order in the gradients is given by Eq. (5.11),

$$\mathbf{q} \to -\lambda\nabla T - \sum_{i,j=1}^{s}(T^2 D_{q,ij}\nabla \ln n_j + L_{ij}\mathbf{F}_j). \tag{7.10}$$

There are both kinetic and collisional transfer contributions according to Eq. (4.17), $\mathbf{q} \equiv \mathbf{q}^k + \mathbf{q}^c$. The kinetic contributions to the transport coefficients are identified as

$$\lambda^k = \sum_{i=1}^{s} \lambda_i^k = -\frac{1}{dT} \sum_{i=1}^{s} \int d\mathbf{v} \frac{1}{2} m_i V^2 \mathbf{V} \cdot \boldsymbol{\mathcal{A}}_i(\mathbf{V}), \quad (7.11)$$

$$D_{q,ij}^k = -\frac{1}{dT^2} \int d\mathbf{v} \frac{1}{2} m_i V^2 \mathbf{V} \cdot \boldsymbol{\mathcal{B}}_i^j(\mathbf{V}), \quad (7.12)$$

$$L_{ij}^k = -\frac{1}{d} \int d\mathbf{v} \frac{1}{2} m_i V^2 \mathbf{V} \cdot \boldsymbol{\mathcal{E}}_i^j(\mathbf{V}). \quad (7.13)$$

For convenience below, the partial thermal conductivities λ_i^k have been introduced in Eq. (7.11). The collision transfer contributions are obtained from Eq. (4.19) to first order in the gradients. These are calculated in Appendix F with the results

$$\lambda^c = \sum_{i,j=1}^{s} \frac{1}{8}(1+\alpha_{ij}) m_j \mu_{ij} \sigma_{ij}^d \chi_{ij}^{(0)} \Bigg\{ 2B_4(1-\alpha_{ij})(\mu_{ij} - \mu_{ji}) n_i \left[\frac{2}{m_j} \lambda_j^k + (d+2) \frac{T_i^{(0)}}{m_i m_j T} \rho D_j^T \right] + \frac{8B_2}{2+d} n_i \left[\frac{2\mu_{ij}}{m_j} \lambda_j^k - (d+2) \frac{T_i^{(0)}}{m_i m_j T} (2\mu_{ij} - \mu_{ji}) \rho D_j^T \right] - T^{-1} C_{ij}^T \Bigg\}, \quad (7.14)$$

$$D_{q,ij}^c = \sum_{p=1}^{s} \frac{1}{8}(1+\alpha_{ip}) m_p \mu_{ip} \sigma_{ip}^d \chi_{ip}^{(0)} \Bigg\{ 2B_4(1-\alpha_{ip})(\mu_{ip} - \mu_{pi}) \times n_i \left[\frac{2}{m_p} D_{q,pj}^k + (d+2) \frac{T_i^{(0)}}{T^2} \frac{m_j n_j}{\rho m_i} D_{pj} \right] + \frac{8B_2}{d+2} n_i \left[\frac{2\mu_{pi}}{m_p} D_{q,pj}^k - (d+2)(2\mu_{ip} - \mu_{pi}) \frac{T_i^{(0)}}{T^2} \frac{n_j m_j}{m_i \rho} D_{pj} \right] - T^{-2} C_{ipj}^T \Bigg\}, \quad (7.15)$$

$$L_{ij}^c = \sum_{p=1}^{s} \frac{1}{8}(1+\alpha_{ip}) m_p \mu_{ip} \sigma_{ip}^d \chi_{ip}^{(0)} \Bigg\{ 2B_4(1-\alpha_{ip})(\mu_{ip} - \mu_{pi}) \times n_i \left[\frac{2}{m_p} L_{pj}^k + (d+2) \frac{T_i^{(0)}}{m_i m_p} D_{pj}^F \right] + \frac{8B_2}{d+2} n_i \left[\frac{2\mu_{pi}}{m_p} L_{pj}^k - (d+2)(2\mu_{ip} - \mu_{pi}) \frac{T_i^{(0)}}{m_i m_p} D_{pj}^F \right] \Bigg\}, \quad (7.16)$$

where the coefficients C_{ij}^T and C_{ipj}^T are given by Eqs. (F27) and (F28). These expressions also depend on the transport coefficients of the mass fluxes D_i^T, D_{ij}, and D_{ij}^F given by Eqs. (7.7)–(7.9), respectively, and on the kinetic contributions λ_i^k, $D_{q,ij}^k$, and L_{ij}^k.

D. Momentum flux

The pressure tensor is evaluated from Eqs. (4.20)–(4.22). To zeroth order in the gradients, one gets the pressure p as

$$p(\{n_i\},T) = \frac{1}{d} P_{\gamma\gamma}^{(0)} \equiv p^k(\{n_i\},T) + p^c(\{n_i\},T) = \frac{1}{d} P_{\gamma\gamma}^{(0)k} + \frac{1}{d} P_{\gamma\gamma}^{(0)c}, \quad (7.17)$$

where

$$p = p^k + p^c, \quad (7.18)$$

$$p^k = nT, \quad p^c = B_2 \sum_{i,j=1}^{s} \mu_{ji}(1+\alpha_{ij}) \sigma_{ij}^d \chi_{ij}^{(0)} n_i n_j T_i^{(0)}. \quad (7.19)$$

Similarly the shear viscosity is $\eta = \eta^k + \eta^c$ where

$$\eta^k = \sum_{i=1}^{s} \eta_i^k, \quad \eta_i^k = -\frac{1}{(d+2)(d-1)} \sum_{i=1}^{s} \int d\mathbf{v} m_i V_\lambda V_\gamma \mathcal{C}_{i,\lambda\gamma}(\mathbf{V}), \quad (7.20)$$

$$\eta^c = \frac{2B_2}{(d+2)} \sum_{i,j=1}^{s} \mu_{ij}(1+\alpha_{ij}) \chi_{ij}^{(0)} n_i \sigma_{ij}^d \eta_j^k + \frac{d}{d+2} \kappa^c. \quad (7.21)$$

Finally, the bulk viscosity is $\kappa = \kappa^k + \kappa^c$ where

$$\kappa^k = 0,$$

$$\kappa^c = \frac{B_3(d+1)}{2d^2} \sum_{i,j=1}^{s} m_j \mu_{ij}(1+\alpha_{ij}) \chi_{ij}^{(0)} \sigma_{ij}^{d+1} \times \int d\mathbf{v}_1 \int d\mathbf{v}_2 f_i^{(0)}(\mathbf{V}_1) f_j^{(0)}(\mathbf{V}_2) g_{12}. \quad (7.22)$$

VIII. DISCUSSION

The most complete and accurate description of mixtures for ordinary fluids is based on the revised Enskog kinetic equations for hard spheres. The explicit construction of solutions to those equations by the CE expansion to first order in the gradients was given more than 20 years ago in Ref. [49]. These solutions, together with the macroscopic balance equations obtained from the kinetic equations, provide a self-consistent derivation of Navier-Stokes hydrodynamics for mixtures and the identification of expressions for all the Navier-Stokes parameters (equations of state, transport coefficients). In the context of the chosen kinetic theory, the analysis and the expressions for these parameters are exact. At this formal level questions of principle could be addressed, prior to the introduction of subsequent approximations for practical evaluations. For example, it was shown that application of the analysis to the original and revised Enskog theories leads to qualitatively different Navier-Stokes hydrodynamics, only one of which is consistent with irreversible thermodynamics. Since no approximations were involved this was sufficient to reject the Enskog kinetic theory in favor of its revised version [48].

The present work is simply an extension of that in Ref.

[49] to *inelastic* hard sphere granular mixtures. Modification of the collisions to account for inelasticity leads to significant differences from ordinary fluids in detail, but the formal structure of the CE expansion remains the same. Similarly, granular Navier-Stokes hydrodynamics results exactly from the CE solution to first order in the gradients and the corresponding modified balance equations. The form of these hydrodynamic equations and expressions for the transport coefficients are exact, as in the ordinary fluid case. The primary motivation for this analysis is to provide the basis for practical applications, as noted in the Introduction, and described in the following paper. However, at the formal level, important fundamental questions can be addressed and clarified as well.

The existence of hydrodynamics for granular fluids has been questioned, due to the many known differences from ordinary fluids: there is no equilibrium or even stationary reference state; the temperature is not a hydrodynamic field (failure of energy conservation), or conversely, multiple temperature fields could be required for mixtures (failure of equilibrium state equipartition for the corresponding granular HCS). In the end, qualitative discussions must be resolved by controlled analysis. Here, the validity of the RET for some range of densities and degree of dissipation has been assumed as a mesoscopic basis for possible macroscopic dynamics in a granular mixture. As shown in the text, sufficient conditions are the macroscopic balance equations (verified) and a normal solution to the kinetic equations. The normal solution is defined in terms of a chosen set of hydrodynamic fields, and the question of hydrodynamics reduces to its existence. The details of the Appendixes give the explicit construction of this solution to first order in the gradients, together with a proof of the existence of solutions to the associated integral equations. It can be concluded from this that a closed set of hydrodynamic equations for the species densities, flow velocity, and a single temperature exist for sufficiently small gradients.

This conclusion is consistent with the observations that the reference state is not equilibrium, depends on the cooling temperature, energy loss can be large at strong dissipation, and the kinetic temperatures of species are different. None of these facts compromises implementation of the CE expansion for solution to the kinetic equation. The parameters of the resulting Navier-Stokes equations incorporate such effects through the integral equations that determine them, and their dependence on the time dependent fields. This in turn affects the solutions to the Navier-Stokes equations under different physical conditions, and is responsible for some of the observed peculiarities of granular fluids. Clearly, it is important to get the details of the Navier-Stokes equations accurately before concluding that any observed experimental phenomenon is hydrodynamic or not. This is another primary motivation for the present work.

These details entail solution to the equation for the reference state and solution to the integral equations for the transport coefficients, to determine them as functions of the hydrodynamic fields (temperature, flow field, and species densities) and the system parameters (restitution coefficients, masses, particle sizes). There has been considerable study of the reference state, as an expansion about a Gaussian for relatively small velocities (asymptotic forms for large velocities are known as well). The integral equations can be solved approximately as truncated expansions in a complete set of polynomials with Gaussian weight factors. For ordinary fluids the leading approximation is generally quite accurate, and the following paper gives its extension to the granular mixture. Still, there are open questions about this approximation for strong dissipation and large mechanical disparity (e.g., mass ratio). Previous results obtained for granular mixtures at low-density [67,68] and for the shear viscosity of a heated granular mixture at moderate density [64] have shown the accuracy of the above approximation, even for strong dissipation.

An accurate solution to the integral equations will predict the transport coefficients as functions of the dissipation. There is only one correct result for this dependence, given by the formulas obtained here. However, its measurement in a given experiment or simulation can entangle and affect this dependence of the transport coefficients due to higher order gradients beyond the Navier-Stokes limit. It may be tempting to compare experimental or simulation data to a corresponding Navier-Stokes solution, adjusting the transport coefficients for a best fit and reporting these as the "measured" values. This can be misleading for granular fluids under conditions where the size of the gradients increases with the degree of dissipation. For such states, strong dissipation can require additional terms in the constitutive equations beyond those of Navier-Stokes order [40,69,70]. This does not mean that the results obtained here are not correct at strong dissipation, only that they must be distinguished carefully from other effects of the same order. A careful tabulation of the Navier-Stokes results given here (e.g., via Monte Carlo simulation) is required for an accurate analysis of experiments of current interest. It is an interesting feature of granular fluids that hydrodynamic states beyond Navier-Stokes order may be the norm rather than the exception.

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APPENDIX A: RET AND SPATIAL CORRELATIONS AT CONTACT

In the case of ordinary fluids, the Enskog approximation can be understood as a short time, or Markovian approxima-

tion. This follows if the initial distribution has the Enskog form

$$f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t=0)=f_i(\mathbf{r}_1,\mathbf{v}_1,t=0)\times f_j(\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t=0)\times\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_k\}). \tag{A1}$$

In fact, this is a quite plausible class of initial conditions since correlations in that case are generally induced by the interparticle structure that is independent of the velocities. Then at finite times, it is assumed that $f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)$ becomes a functional of f_i,

$$f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)=F_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t|f_i(t)). \tag{A2}$$

The Enskog approximation corresponds to evaluating this functional at $t=0$

$$f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)\to F_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t=0|f_i(t)). \tag{A3}$$

Thus for the special class of initial conditions the Enskog approximation is asymptotically exact at short times, and assumes that the generator for dynamics at later times is the same as that initially. This idea provides a simple mean field theory for particles with continuous potentials of interaction, but is more realistic for hard spheres where there is instantaneous momentum transport at the initial time. The presence of inherent velocity correlations for granular fluids suggests that the form (A1) is less justified than in the ordinary fluid case. However, it is noted that velocity correlations are present for any nonequilibrium state even with elastic collisions and it is known that the Enskog equation still provides a good approximation in these latter cases.

An important exact boundary condition for hard spheres is given by [53]

$$\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)=\alpha_{ij}^{-2}b_{ij}^{-1}\Theta(-\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t). \tag{A4}$$

This equation implies that the distribution of particles that have collided is the same as those about to collide, but with their velocities changed according to the collision rules. In general the two particle distribution function can be written as

$$f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)=\Theta(-\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)+\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t). \tag{A5}$$

If the Enskog approximation (A3) is introduced in the first term on the right side of Eq. (A5), then the corresponding approximation on the right side of Eq. (A5) gives the approximate two particle distribution function at contact as

$$\begin{aligned}f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)&\to\Theta(-\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_k\})f_i(\mathbf{r}_1,\mathbf{v}_1;t)f_j(\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)+\alpha_{ij}^{-2}b_{ij}^{-1}\Theta(-\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\\&\quad\times\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_k\})f_i(\mathbf{r}_1,\mathbf{v}_1;t)f_j(\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)\\&=\Theta(-\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_k\})f_i(\mathbf{r}_1,\mathbf{v}_1;t)f_j(\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)+\alpha_{ij}^{-2}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_k\})\\&\quad\times f_i(\mathbf{r}_1,\mathbf{v}_1'';t)f_j(\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2'';t).\end{aligned} \tag{A6}$$

Since $\mathbf{v}_1''$ and $\mathbf{v}_2''$ are functions of both $\mathbf{v}_1$ and $\mathbf{v}_2$ there are velocity correlations on the complementary hemisphere, even when they are neglected on the precollision hemisphere.

An important consequence of Eq. (A6) is the relationship of $\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_i\})$ to the pair correlation function $g_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_k\})$ defined by

$$n_i(\mathbf{r}_1)n_j(\mathbf{r}_1-\boldsymbol{\sigma}_{ij})g_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_k\})=\int d\mathbf{v}_1\int d\mathbf{v}_2 f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t). \tag{A7}$$

Use of the approximation (A6) gives the result

$$n_i(\mathbf{r}_1)n_j(\mathbf{r}_1-\boldsymbol{\sigma}_{ij})g_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_k\})=\frac{1+\alpha_{ij}}{\alpha_{ij}}\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_k\})\int d\mathbf{v}_1\int d\mathbf{v}_2\Theta(-\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\times f_i(\mathbf{r}_1,\mathbf{v}_1;t)f_j(\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t), \tag{A8}$$

where a change of variables has been made in the integration of the second term in Eq. (A6),

$$\int d\mathbf{v}_1\int d\mathbf{v}_2 X(\mathbf{v}_1'',\mathbf{v}_2'')=\alpha_{ij}\int d\mathbf{v}_1''\int d\mathbf{v}_2'' X(\mathbf{v}_1'',\mathbf{v}_2''). \tag{A9}$$

For a uniform system, $g_{ij}(\mathbf{r}_1,\mathbf{r}_2|\{n_k\})\to g_{ij}(|\mathbf{r}_1-\mathbf{r}_2|;\{n_k\})$ and this expression reduces to

$$g_{ij}(\sigma_{ij};\{n_k\}) = \frac{1+\alpha_{ij}}{2\alpha_{ij}}\chi_{ij}(\sigma_{ij};\{n_k\}). \tag{A10}$$

Equation (A10) is the result quoted in the text and provides the interpretation for $\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_k\})$. For elastic collisions $\chi_{ij}(\boldsymbol{\sigma}_{ij};\{n_k\})$ is indeed the pair correlation function at contact. The Enskog theory in that case takes $\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_k\})$ to be the pair correlation function for an equilibrium nonuniform fluid whose densities are equal to those for the actual nonequilibrium state being considered. This assumption is based on the fact that structural correlations for hard spheres are entirely due to excluded volume effects which should be similar for equilibrium and nonequilibrium states. It is reasonable to extend this choice for $\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_k\})$ to granular fluids as well. Its accuracy can be judged by measuring (via MD simulation) the pair correlation given by Eq. (A10) with this choice on the right side. This has been done for the one component fluid, indicating reasonable results over a range of values for the restitution coefficient [53].

APPENDIX B: BALANCE EQUATIONS AND FLUXES

The macroscopic balance equations follow from the definitions (4.1)–(4.3) and the first hierarchy equation (3.1),

$$\partial_t n_i + \nabla_{\mathbf{r}_1}\cdot\int d\mathbf{v}_1\mathbf{v}_1 f_i = \int d\mathbf{v}_1 C_i, \tag{B1}$$

$$\partial_t e + \nabla_{\mathbf{r}_1}\cdot\sum_{i=1}^{s}\int d\mathbf{v}_1\frac{1}{2}m_i v_1^2\mathbf{v}_1 f_i - \sum_{i=1}^{s}\mathbf{F}_i\cdot\int d\mathbf{v}_1\mathbf{v}_1 f_i = \sum_{i=1}^{s}\int d\mathbf{v}_1\frac{1}{2}m_i v_1^2 C_i, \tag{B2}$$

$$\partial_t p_\beta + \partial_{r_{1\gamma}}\sum_{i=1}^{s}\int d\mathbf{v}_1 m_i v_{1\gamma}v_{1\beta}f_i - \sum_{i=1}^{s}n_i F_{i\beta} = \sum_{i=1}^{s}\int d\mathbf{v}_1 m_i v_{1\beta}C_i. \tag{B3}$$

The integrals over the collisional contribution C_i are analyzed below with the results

$$\int d\mathbf{v}_1 C_i = 0, \tag{B4}$$

$$\sum_{i=1}^{s}\int d\mathbf{v}_1\frac{1}{2}m_i v_1^2 C_i = -\nabla\cdot\mathbf{s}^c - w, \tag{B5}$$

$$\sum_{i=1}^{s}\int d\mathbf{v}_1 m_i v_{1\beta}C_i = -\partial_{r_\gamma}t^c_{\gamma\beta}. \tag{B6}$$

Use of these expressions in Eqs. (B1)–(B3) gives the balance equations (4.4)–(4.6) of the text with

$$\mathbf{j}_i = m_i\int d\mathbf{v}_1\mathbf{v}_1 f_i, \tag{B7}$$

$$\mathbf{s} = \sum_{i=1}^{s}\int d\mathbf{v}_1\frac{1}{2}m_i v_1^2\mathbf{v}_1 f_i + \mathbf{s}^c, \tag{B8}$$

$$t_{\gamma\beta} = \sum_{i=1}^{s}\int d\mathbf{v}_1 m_i v_{1\gamma}v_{1\beta}f_i + t^c_{\gamma\beta}. \tag{B9}$$

The terms w, $\mathbf{s}^c$, and $t^c_{\gamma\beta}$ arising from the collisional contribution C_i are identified by further analysis of the left sides of Eqs. (B5) and (B6). To do so consider the general expression for some arbitrary function $\psi_i(\mathbf{v}_1)$

$$\int d\mathbf{v}_1\psi_i C_i = \sum_{j=1}^{s}\sigma_{ij}^{d-1}\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\psi_i(\mathbf{v}_1)[\alpha_{ij}^{-2}f_{ij}(\mathbf{r}_1,\mathbf{v}_1'',\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2'';t) - f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)]. \tag{B10}$$

The restituting velocities are functions of the given velocities, $\mathbf{v}_1''=\mathbf{v}_1''(\mathbf{v}_1,\mathbf{v}_2)$, $\mathbf{v}_2''=\mathbf{v}_2''(\mathbf{v}_1,\mathbf{v}_2)$, defined by Eq. (3.3). These relations can be inverted to get

$$\mathbf{v}_1 = \mathbf{v}_1'' - \mu_{ji}(1+\alpha_{ij})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12}'')\hat{\boldsymbol{\sigma}},$$

$$\mathbf{v}_2 = \mathbf{v}_2'' + \mu_{ij}(1+\alpha_{ij})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12}'')\hat{\boldsymbol{\sigma}}. \tag{B11}$$

Therefore in the first term of Eq. (B10) it is possible to change integration variables from $d\mathbf{v}_1d\mathbf{v}_2$ to $d\mathbf{v}_1''d\mathbf{v}_2''$, with a Jacobian α_{ij} to get

$$\begin{aligned}\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\psi_i(\mathbf{v}_1)\alpha_{ij}^{-2}f_{ij}(\mathbf{r}_1,\mathbf{v}_1'',\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2'';t)\\ = \int d\mathbf{v}_1''\int d\mathbf{v}_2''\int d\hat{\boldsymbol{\sigma}}\Theta(-\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12}'')(-\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12}'')\psi_i(\mathbf{v}_1(\mathbf{v}_1'',\mathbf{v}_2''))f_{ij}(\mathbf{r}_1,\mathbf{v}_1'',\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2'';t)\\ = \int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\psi_i(\mathbf{v}_1'(\mathbf{v}_1,\mathbf{v}_2))f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t),\end{aligned} \tag{B12}$$

where use has been made of $(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})=-\alpha_{ij}(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}''_{12})$. In the last line the dummy variables $(\mathbf{v}''_1,\mathbf{v}''_2)$ have been relabeled $(\mathbf{v}_1,\mathbf{v}_2)$, and a change of integration from $\hat{\boldsymbol{\sigma}}$ to $-\hat{\boldsymbol{\sigma}}$ has been performed. Accordingly $\mathbf{v}_1(\mathbf{v}''_1,\mathbf{v}''_2)$ has been relabeled $\mathbf{v}'_1(\mathbf{v}_1,\mathbf{v}_2)$ with Eq. (B11) becoming in this notation

$$\mathbf{v}'_1=\mathbf{v}_1-\mu_{ji}(1+\alpha_{ij})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\hat{\boldsymbol{\sigma}},\quad \mathbf{v}'_2=\mathbf{v}_2+\mu_{ij}(1+\alpha_{ij})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\hat{\boldsymbol{\sigma}}. \tag{B13}$$

This is the direct scattering law, which differs from the restituting scattering law (3.3) for $\alpha_{ij}\neq 1$. With this transformation the integral (B10) is

$$\int d\mathbf{v}_1\psi_i C_i=\sum_{j=1}^{s}\sigma_{ij}^{d-1}\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})[\psi_i(\mathbf{v}'_1)-\psi_i(\mathbf{v}_1)]f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t). \tag{B14}$$

The special choice $\psi_i(\mathbf{v}_1)=1$ proves Eq. (B4) above.

Next, consider the sum of Eq. (B14) over all species,

$$\begin{aligned}\sum_{i=1}^{s}\int d\mathbf{v}_1\psi_i C_i&=\sum_{i,j=1}^{s}\sigma_{ij}^{d-1}\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})[\psi_i(\mathbf{v}'_1)-\psi_i(\mathbf{v}_1)]f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)\\&=\frac{1}{2}\sum_{i,j=1}^{k}\sigma_{ij}^{d-1}\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\{[\psi_i(\mathbf{v}'_1)-\psi_i(\mathbf{v}_1)]f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)\\&\quad+[\psi_j(\mathbf{v}'_2)-\psi_j(\mathbf{v}_2)]f_{ji}(\mathbf{r}_1,\mathbf{v}_2,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_1;t)\}.\end{aligned} \tag{B15}$$

The second equality is obtained from the first by taking half the sum of the first plus an equivalent form obtained by interchanging $\mathbf{v}_1$ and $\mathbf{v}_2$, interchanging i and j, and changing $\hat{\boldsymbol{\sigma}}$ to $-\hat{\boldsymbol{\sigma}}$. To simplify this further, note the relation $f_{ji}(\mathbf{r}_1,\mathbf{v}_2,\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_1;t)=f_{ij}(\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1,\mathbf{v}_2;t)$ and arrange terms as

$$\begin{aligned}\sum_{i=1}^{s}\int d\mathbf{v}_1\psi_i C_i&=\frac{1}{2}\sum_{i,j=1}^{s}\sigma_{ij}^{d-1}\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\{[\psi_i(\mathbf{v}'_1)+\psi_j(\mathbf{v}'_2)-\psi_i(\mathbf{v}_1)-\psi_j(\mathbf{v}_2)]f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)\\&\quad+[\psi_i(\mathbf{v}'_1)-\psi_i(\mathbf{v}_1)][f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)-f_{ij}(\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1,\mathbf{v}_2;t)]\}.\end{aligned} \tag{B16}$$

The first term of the integrand represents a collisional effect due to scattering with a change in the velocities. The second term is a collisional effect due to the spatial difference of the colliding pair. This second effect is called "collisional transfer." It can be written as a divergence through the identity

$$\begin{aligned}f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)-f_{ij}(\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1,\mathbf{v}_2;t)&=\int_0^1 dx\frac{\partial}{\partial x}f_{ij}(\mathbf{r}_1-x\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)\\&=\nabla_{\mathbf{r}_1}\cdot\boldsymbol{\sigma}_{ij}\int_0^1 dx f_{ij}(\mathbf{r}_1-x\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t).\end{aligned} \tag{B17}$$

Using the identity (B17), Eq. (B16) can be finally written as

$$\begin{aligned}\sum_{i=1}^{s}\int d\mathbf{v}_1\psi_i C_i=\frac{1}{2}\sum_{i,j=1}^{s}\sigma_{ij}^{d-1}\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\Big\{&[\psi_i(\mathbf{v}'_1)+\psi_j(\mathbf{v}'_2)-\psi_i(\mathbf{v}_1)-\psi_j(\mathbf{v}_2)]f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)\\&+\nabla_{\mathbf{r}_1}\cdot\boldsymbol{\sigma}_{ij}[\psi_i(\mathbf{v}'_1)-\psi_i(\mathbf{v}_1)]\int_0^1 dx f_{ij}(\mathbf{r}_1-x\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)\Big\}.\end{aligned} \tag{B18}$$

Now, apply this result to the case $\psi_i=m_i\mathbf{v}_1$. Since the total momentum is conserved in all pair collisions, $\psi_i(\mathbf{v}'_1)+\psi_j(\mathbf{v}'_2)-\psi_i(\mathbf{v}_1)-\psi_j(\mathbf{v}_2)=0$ for this case and Eq. (B18) gives Eq. (B6) with

$$\begin{aligned}t^c_{\gamma\beta}\equiv-\frac{1}{2}\sum_{i,j=1}^{s}\sigma_{ij}^{d}\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\hat{\sigma}_\gamma(m_i v'_{1\beta}-m_i v_{1\beta})\int_0^1 dx f_{ij}(\mathbf{r}_1-x\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)\\=\frac{1}{2}\sum_{i,j=1}^{s}m_i\mu_{ji}(1+\alpha_{ij})\sigma_{ij}^{d}\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})^2\hat{\sigma}_\gamma\hat{\sigma}_\beta\int_0^1 dx f_{ij}(\mathbf{r}_1-x\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t).\end{aligned} \tag{B19}$$

The analysis leading to Eq. (B6) follows from Eq. (B16) in a similar way with $\psi_i=m_i v_1^2/2$. However, since energy is not

conserved in pair collisions the first term on the right side does not vanish. Instead, it represents the collisions energy loss w,

$$w = -\sum_{i,j=1}^{s} \frac{1}{4}\sigma_{ij}^{d-1} \int d\mathbf{v}_1 \int d\mathbf{v}_2 \int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(m_i v_1'^2 + m_j v_2'^2 - m_i v_1^2 - m_j v_2^2) f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)$$

$$= \frac{1}{4}\sum_{i,j=1}^{s} (1-\alpha_{ij}^2) m_i \mu_{ji} \sigma_{ij}^{d-1} \int d\mathbf{v}_1 \int d\mathbf{v}_2 \int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})^3 f_{ij}(\mathbf{r}_1,\mathbf{v}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t). \tag{B20}$$

The second term on the right side of Eq. (B18) gives the collisional transfer contribution to the flux

$$\nabla\cdot\mathbf{s}^c = -\nabla_{\mathbf{r}_1}\cdot\sum_{i,j=1}^{s}\frac{1}{4}m_i\sigma_{ij}^d \int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\hat{\boldsymbol{\sigma}}(v_1'^2 - v_1^2)\int_0^1 dx f_{ij}(\mathbf{r}_1 - x\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)$$

$$= \nabla_{\mathbf{r}_1}\cdot\sum_{i,j=1}^{s}\frac{1}{4}(1+\alpha_{ij})m_i\mu_{ji}\sigma_{ij}^d \int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})^2\hat{\boldsymbol{\sigma}}[\mu_{ji}(1-\alpha_{ij})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})$$

$$+ 2\hat{\boldsymbol{\sigma}}\cdot(\mu_{ij}\mathbf{v}_1 + \mu_{ji}\mathbf{v}_2)]\int_0^1 dx f_{ij}(\mathbf{r}_1 - x\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t). \tag{B21}$$

This confirms Eq. (B5) and identifies $\mathbf{s}^c$, which has the equivalent form (obtained by taking half the sum of forms with i and j interchanged)

$$\mathbf{s}^c = \sum_{i,j=1}^{s}\frac{1}{8}(1+\alpha_{ij})m_i\mu_{ji}\sigma_{ij}^d \int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})$$

$$\times(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})^2\hat{\boldsymbol{\sigma}}[(1-\alpha_{ij})(\mu_{ji}-\mu_{ij})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12}) + 4\hat{\boldsymbol{\sigma}}\cdot(\mu_{ij}\mathbf{v}_1$$

$$+ \mu_{ji}\mathbf{v}_2)]\int_0^1 dx f_{ij}(\mathbf{r}_1 - x\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t). \tag{B22}$$

APPENDIX C: CHAPMAN-ENSKOG SOLUTION

As described in the text a normal solution to the kinetic equation is a nonlocal functional of the hydrodynamic fields $f_i(\mathbf{v}_1|\{y_\beta(t)\})$. This is equivalent to a function of the fields at a point and all their derivatives at that point,

$$f_i(\mathbf{r}_1,\mathbf{v}_1|\{y_\beta(t)\}) = f_i(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\};\{\partial_{\mathbf{r}_1}y_\beta(\mathbf{r}_1,t);\ldots\}). \tag{C1}$$

If the gradients are small, this function can be expanded in the appropriate dimensionless small parameter

$$f_i(\mathbf{v}_1|\{y_\beta(t)\}) = f_i^{(0)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\}) + f_i^{(1)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\};\{\partial_{\mathbf{r}_1}y_\beta(\mathbf{r}_1,t)\}) + \cdots, \tag{C2}$$

where $f_i^{(0)}$ is a function of the fields alone, $f_i^{(1)}$ is a function of the fields and linear in their gradients, and so on. Thus the kinetic equation can be solved perturbatively by requiring that contributions from common order in this gradient expansion vanish.

To perform this ordering it is necessary to expand the collision operators of Eq. (3.9),

$$J_{ij}[\mathbf{r}_1,\mathbf{v}_1|f(t)] \equiv \sigma_{ij}^{d-1}\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})$$

$$\times[\alpha_{ij}^{-2}\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1-\boldsymbol{\sigma}_{ij}|\{n_i\})$$

$$\times f_i(\mathbf{r}_1,\mathbf{v}_1'';t)f_j(\mathbf{r}_1-\boldsymbol{\sigma}_{ij},\mathbf{v}_2'';t)$$

$$-\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1+\boldsymbol{\sigma}_{ij}|\{n_i\})f_i(\mathbf{r}_1,\mathbf{v}_1;t)$$

$$\times f_j(\mathbf{r}_1+\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)]. \tag{C3}$$

For the purposes here it is sufficient to go up through first order. The distribution functions evaluated at $\mathbf{r}_1\pm\boldsymbol{\sigma}_{ij}$ become

$$f_i(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1\pm\boldsymbol{\sigma}_{ij},t)\};\{\partial_{\mathbf{r}_1}y_\beta(\mathbf{r}_1\pm\boldsymbol{\sigma}_{ij},t);\ldots\}) \to (1\pm\boldsymbol{\sigma}_{ij}\cdot\nabla_{\mathbf{r}_1})f_i^{(0)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\}) + f_i^{(1)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\};\{\partial_{\mathbf{r}_1}y_\beta(\mathbf{r}_1,t)\})$$

$$= f_i^{(0)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\}) \pm [\partial_{y_\beta}f_i^{(0)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\})]\boldsymbol{\sigma}_{ij}\cdot\nabla_{\mathbf{r}_1}y_\beta(\mathbf{r}_1,t)$$

$$+ f_i^{(1)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\};\{\partial_{\mathbf{r}_1}y_\beta(\mathbf{r}_1,t)\}). \tag{C4}$$

The functional expansion of $\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1\pm\sigma_{ij}|\{n_i\})$ to this order is obtained by a functional expansion of all species densities about their values at $\mathbf{r}_1$

$$\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1\pm\boldsymbol{\sigma}_{ij}|\{n_i(t)\})=\chi_{ij}^{(0)}(\sigma_{ij}|\{n_k(\mathbf{r}_1,t)\})+\sum_{\ell=1}^{s}\int d\mathbf{r}'\ \left.\frac{\delta\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1\pm\boldsymbol{\sigma}_{ij}|\{n_i\})}{\delta n_\ell(\mathbf{r}',t)}\right|_{\delta n=0}(n_\ell(\mathbf{r}')-n_\ell(\mathbf{r}_1))+\cdots \tag{C5}$$

$$\to\chi_{ij}^{(0)}(\sigma_{ij};\{n_k(\mathbf{r}_1,t)\})+\sum_{\ell=1}^{s}(\nabla_{\mathbf{r}_1}n_\ell(\mathbf{r}_1;t))\int d\mathbf{r}'(\mathbf{r}'-\mathbf{r}_1)\ \left.\frac{\delta\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1\pm\boldsymbol{\sigma}_{ij}|\{n_i\})}{\delta n_\ell(\mathbf{r}',t)}\right|_{\delta n=0}. \tag{C6}$$

The arrow denotes the leading terms of a Taylor series for $(n_\ell(\mathbf{r}')-n_\ell(\mathbf{r}_1))$. The integral can be simplified by noting at $\delta n=0$ the functional integral has translational invariance

$$\left.\frac{\delta\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1\pm\boldsymbol{\sigma}_{ij}|\{n_i\})}{\delta n_\ell(\mathbf{r}',t)}\right|_{\delta n=0}=F_{ij\ell}(\mathbf{r}_1-\mathbf{r}',\mathbf{r}_1\pm\boldsymbol{\sigma}_{ij}-\mathbf{r}') \tag{C7}$$

so

$$\int d\mathbf{r}'(\mathbf{r}'-\mathbf{r}_1)\ \left.\frac{\delta\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1\pm\boldsymbol{\sigma}_{ij}|\{n_i\})}{\delta n_\ell(\mathbf{r}',t)}\right|_{\delta n=0}$$

$$=\int d\mathbf{r}'\left(\mathbf{r}'\pm\frac{1}{2}\boldsymbol{\sigma}_{ij}\right)\ \left.\frac{\delta\chi_{ij}\left(\mp\frac{1}{2}\boldsymbol{\sigma}_{ij},\pm\frac{1}{2}\boldsymbol{\sigma}_{ij}|\{n_i\}\right)}{\delta n_\ell(\mathbf{r}',t)}\right|_{\delta n=0} \tag{C8}$$

$$=\pm\frac{1}{2}\boldsymbol{\sigma}_{ij}\frac{\partial\ln\chi_{ij}^{(0)}(\sigma_{ij};\{n_\ell(\mathbf{r}_1)\})}{\partial n_\ell(\mathbf{r}_1)}+\int d\mathbf{r}'\mathbf{r}'\ \left.\frac{\delta\chi_{ij}\left(\mp\frac{1}{2}\boldsymbol{\sigma}_{ij},\pm\frac{1}{2}\boldsymbol{\sigma}_{ij}|\{n_i\}\right)}{\delta n_\ell(\mathbf{r}',t)}\right|_{\delta n=0}. \tag{C9}$$

The expansion for $\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1\pm\sigma_{ij}|\{n_i(t)\})$ becomes

$$\chi_{ij}(\mathbf{r}_1,\mathbf{r}_1\pm\boldsymbol{\sigma}_{ij}|\{n_i(t)\})=\chi_{ij}^{(0)}(\sigma_{ij};\{n_k(\mathbf{r}_1,t)\})\times\left[1\pm\frac{1}{2}\boldsymbol{\sigma}_{ij}\sum_{\ell=1}^{s}\nabla_{\mathbf{r}_1}\ln n_\ell(\mathbf{r}_1;t)\times\left(n_\ell(\mathbf{r}_1)\frac{\partial\ln\chi_{ij}^{(0)}(\sigma_{ij};\{n_\ell(\mathbf{r}_1)\})}{\partial n_\ell(\mathbf{r}_1)}+I_{ij\ell}(\sigma_{ij};\{n_k(\mathbf{r}_1,t)\})\right)\right]. \tag{C10}$$

The last line defines $I_{ij\ell}(\sigma_{ij};\{n_k(\mathbf{r}_1,t)\})$ as

$$I_{ij\ell}(\sigma_{ij};\{n_k\})\equiv\frac{2n_\ell(\mathbf{r}_1,t)}{\chi_{ij}^{(0)}(\sigma_{ij};\{n_k\})\sigma_{ij}}\int d\mathbf{r}'(\hat{\boldsymbol{\sigma}}_{ij}\cdot\mathbf{r}')\times\left.\frac{\delta\chi_{ij}\left(-\frac{1}{2}\boldsymbol{\sigma}_{ij},+\frac{1}{2}\boldsymbol{\sigma}_{ij}|\{n_i\}\right)}{\delta n_\ell(\mathbf{r}',t)}\right|_{\delta n=0}. \tag{C11}$$

These results give the expansion of $J_{ij}[\mathbf{r}_1,\mathbf{v}_1|f_i]$ to first order in the gradients

$$\sum_{j=1}^{s}J_{ij}[\mathbf{r}_1,\mathbf{v}_1|f]\to\sum_{j=1}^{s}J_{ij}^{(0)}[\mathbf{v}_1|f_i^{(0)},f_j^{(0)}]-(Lf^{(1)})_i \tag{C12}$$

$$-\sum_{j=1}^{s}\mathcal{K}_{ij,\gamma}[\mathbf{v}_1|\partial_{y_\beta}f_j^{(0)}(t)]\partial_\gamma y_\beta(\mathbf{r}_1,t) \tag{C13}$$

$$-\sum_{j,\ell=1}^{s}\left(n_\ell\frac{\partial\ln\chi_{ij}^{(0)}(\sigma_{ij};\{n_k\})}{\partial n_\ell}+I_{ij\ell}(\sigma_{ij};\{n_k\})\right)\times\mathcal{K}_{ij,\gamma}[\mathbf{v}_1|f_j^{(0)}(t)]\partial_\gamma\ln n_\ell(\mathbf{r}_1;t) \tag{C14}$$

with the definitions $\partial_\gamma X\equiv\partial X/\partial r_\gamma$,

$$J_{ij}^{(0)}[\mathbf{v}_1|g_i,f_j]\equiv\chi_{ij}^{(0)}(\sigma_{ij};\{n_i(\mathbf{r}_1,t)\})\sigma_{ij}^{d-1}\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\times(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})[\alpha_{ij}^{-2}g_i(\mathbf{V}_1'')f_j(\mathbf{V}_2'')-g_i(\mathbf{V}_1)f_j(\mathbf{V}_2)], \tag{C15}$$

$$\mathcal{K}_{ij,\gamma}[X_j]=\sigma_{ij}^d\chi_{ij}^{(0)}\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{12})\hat{\sigma}_\gamma\times[\alpha_{ij}^{-2}f_i^{(0)}(\mathbf{r}_1,\mathbf{V}_1'';t)X_j(\mathbf{r}_1,\mathbf{V}_2'')+f_i^{(0)}(\mathbf{r}_1,\mathbf{V}_1;t)X_j(\mathbf{r}_1,\mathbf{V}_2)]. \tag{C16}$$

Finally, L is a linear operator defined over s dimensional vectors $\{X_i\}$ whose components are labeled by the species

$$(LX)_i=-\sum_{j=1}^{s}(J_{ij}^{(0)}[\mathbf{v}_1|X_i,f_j^{(0)}]+J_{ij}^{(0)}[\mathbf{v}_1|f_i^{(0)},X_j]). \tag{C17}$$

It remains to choose the magnitude of the external forces $\mathbf{F}_i$ to consistently order this expansion. To be specific, and for comparison with Ref. [49] it is assumed here to be of first order in the gradients.

1. Zeroth order

At lowest order all gradients of the hydrodynamic fields are neglected, and Eq. (3.8) becomes

$$\partial_t^{(0)} f_i^{(0)}(\mathbf{v}_1;\{y_\beta(\mathbf{r}_1,t)\}) = \sum_{j=1}^{s} J_{ij}^{(0)}[\mathbf{v}_1|f_i^{(0)}(\{y_\beta(\mathbf{r}_1,t)\}), f_j^{(0)}(\{y_\beta(\mathbf{r}_1,t)\})]. \tag{C18}$$

The notation $\partial_t^{(0)}$ for the time derivative means that the balance equations are to be used to zeroth order in the gradients

$$\partial_t^{(0)} f_i^{(0)}(V_1;\{y_\beta(\mathbf{r}_1,t)\}) = [\partial_{y_\beta} f_i^{(0)}(V_1;\{y_\beta(\mathbf{r}_1,t)\})]\partial_t^{(0)} y_\beta(\mathbf{r}_1,t) = -\zeta^{(0)}(\{y_\beta(\mathbf{r}_1,t)\})T\partial_T \times f_i^{(0)}(V_1;\{y_\beta(\mathbf{r}_1,t)\}). \tag{C19}$$

Use of Eq. (C19) in Eq. (C18) gives the zeroth order equation (6.1) of the text.

2. First order

The kinetic equation for contributions of first order in the gradients is

$$\partial_t^{(0)} f_i^{(1)} + (Lf^{(1)})_i = -(\partial_t^{(1)} + \mathbf{v}_1\cdot\nabla_{\mathbf{r}_1} + m_i^{-1}\mathbf{F}_i\cdot\nabla_{\mathbf{v}_1})f_i^{(0)} - \sum_{j=1}^{s}\mathcal{K}_{ij,\gamma}[\mathbf{v}_1|\partial_{y_\beta} f_j^{(0)}]\partial_\gamma y_\beta - \frac{1}{2}\sum_{j,\ell=1}^{s}\mathcal{K}_{ij,\gamma}\left[\mathbf{v}_1\middle|\left(n_\ell\frac{\partial \ln \chi_{ij}^{(0)}}{\partial n_\ell} + I_{ij\ell}\right)f_j^{(0)}\right]\partial_\gamma \ln n_\ell, \tag{C20}$$

where $\nabla_{\mathbf{r}}\equiv\partial/\partial\mathbf{r}$ and $\nabla_{\mathbf{V}}\equiv\partial/\partial\mathbf{V}$. The first term on the right side of Eq. (C20) can be expressed explicitly in terms of the gradients, where now $\partial_t^{(1)}$ means that the balance equations are to be used with only terms of first order in the gradients

$$\begin{aligned}(\partial_t^{(1)} + \mathbf{v}_1\cdot\nabla_{\mathbf{r}_1} + m_i^{-1}\mathbf{F}_i\cdot\nabla_{\mathbf{v}_1})f_i^{(0)} &= m_i^{-1}\mathbf{F}_i\cdot\nabla_{\mathbf{v}_1} f_i^{(0)} + (\partial_{y_\beta} f_i^{(0)})(D_t^{(1)} + \mathbf{V}_1\cdot\nabla_{\mathbf{r}_1})y_\beta \\ &= \left(m_i^{-1}\mathbf{F}_i - \rho^{-1}\sum_{j=1}^{s} n_j\mathbf{F}_j\right)\nabla_{\mathbf{v}_1} f_i^{(0)} - (\nabla_{\mathbf{v}_1} f_i^{(0)})(-\rho^{-1}\nabla_{\mathbf{r}_1} p + \mathbf{V}_1\cdot\nabla_{\mathbf{r}_1}\mathbf{U}) \\ &\quad - (\partial_T f_i^{(0)})\left[\left(T\zeta_U + \frac{2}{nd}p\right)\nabla\cdot\mathbf{U} - \mathbf{V}_1\cdot\nabla_{\mathbf{r}_1} T\right] + \sum_{j=1}^{s}(\partial_{n_j} f_i^{(0)})(-n_j\nabla\cdot\mathbf{U} + \mathbf{V}_1\cdot\nabla_{\mathbf{r}_1} n_j).\end{aligned} \tag{C21}$$

In Eq. (C21), $D_t^{(1)}\equiv\partial_t^{(1)} + \mathbf{U}\cdot\nabla$ and use has been made of the fact that $f_i^{(0)}$ depends on $\mathbf{U}$ only through the combination $\mathbf{V}_1 = \mathbf{v}_1 - \mathbf{U}$, so that

$$\nabla_{\mathbf{U}} f_i^{(0)} = -\nabla_{\mathbf{V}_1} f_i^{(0)}. \tag{C22}$$

The pressure gradient can be expressed in terms of the temperature and density gradients

$$\nabla_{\mathbf{r}_1} p = \frac{p}{T}\nabla_{\mathbf{r}_1} T + \sum_{j=1}^{s}(\partial_{n_j} p)\nabla_{\mathbf{r}_1} n_j, \tag{C23}$$

to give

$$\begin{aligned}(\partial_t^{(1)} + \mathbf{v}_1\cdot\nabla_{\mathbf{r}_1} + m_i^{-1}\mathbf{F}_i\cdot\nabla_{\mathbf{v}_1})f_i^{(0)} &= m_i^{-1}\mathbf{F}_i\cdot\nabla_{\mathbf{v}_1} f_i^{(0)} - \rho^{-1}\sum_{j=1}^{s} n_j\nabla_{\mathbf{v}_1} f_i^{(0)}\cdot\mathbf{F}_j \\ &\quad + \left(\frac{p}{\rho T}\nabla_{\mathbf{v}_1} f_i^{(0)} + \mathbf{V}_1\partial_T f_i^{(0)}\right)\nabla_{\mathbf{r}_1} T + \sum_{j=1}^{s}(\rho^{-1}\partial_{n_j} p\nabla_{\mathbf{v}_1} f_i^{(0)} + \mathbf{V}_1\partial_{n_j} f_i^{(0)})\nabla_{\mathbf{r}_1} n_j \\ &\quad - \left(\left(\zeta_U + \frac{2}{nTd}p\right)T\partial_T f_i^{(0)} + \sum_{j=1}^{s} n_j\partial_{n_j} f_i^{(0)}\right)\nabla\cdot\mathbf{U} - V_{1\gamma}(\partial_{V_{1\eta}} f_i^{(0)})\partial_\gamma U_\eta.\end{aligned} \tag{C24}$$

Equations (C20) for the first order distributions, $f_i^{(1)}$, now become

$$\partial_t^{(0)} f_i^{(1)} + (Lf^{(1)})_i = \mathbf{A}_i(\mathbf{V}_1;\{n_i\})\nabla\ln T + \sum_{j=1}^{s}\mathbf{B}_i^j(\mathbf{V}_1;\{n_i\})\nabla\ln n_j + C_{i,\gamma\eta}(\mathbf{V}_1;\{n_i\})\frac{1}{2}\left(\partial_\gamma U_\eta + \partial_\eta U_\gamma - \frac{2}{d}\delta_{\gamma\eta}\nabla\cdot\mathbf{U}\right) + D_i(\mathbf{V}_1;\{n_i\})\nabla\cdot\mathbf{U} + \sum_{j=1}^{s}\mathbf{E}_i^j(\mathbf{V}_1;\{n_i\})\cdot\mathbf{F}_j. \tag{C25}$$

The functions of velocity on the right side of Eq. (C25) are identified as

$$A_{i,\gamma}(\mathbf{V}) = \frac{1}{2}V_\gamma\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_i^{(0)}) - \frac{p}{\rho}\partial_{V_\gamma} f_i^{(0)} + \frac{1}{2}\sum_{j=1}^{s}K_{ij,\gamma}[\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_j^{(0)})], \tag{C26}$$

$$B_{i,\gamma}^j(\mathbf{V}) = -V_\gamma n_j\partial_{n_j} f_i^{(0)} - \rho^{-1}(\partial_{V_\gamma} f_i^{(0)})n_j(\partial_{n_j} p) - \sum_{\ell=1}^{s}\mathcal{K}_{i\ell,\gamma}\left\{\left[n_j\partial_{n_j} + \frac{1}{2}\left(n_\ell\frac{\partial\ln\chi_{i\ell}^{(0)}}{\partial n_j} + I_{i\ell j}\right)\right]f_\ell^{(0)}\right\}, \tag{C27}$$

$$C_{i,\gamma\beta}(\mathbf{V}) = \frac{1}{2}\left(V_\gamma\partial_{V_\beta} f_i^{(0)} + V_\beta\partial_{V_\gamma} f_i^{(0)} - \frac{2}{d}\delta_{\beta\gamma}\mathbf{V}\cdot\nabla_{\mathbf{V}} f_i^{(0)}\right) + \frac{1}{2}\sum_{j=1}^{s}\left(\mathcal{K}_{ij,\gamma}[\partial_{V_\beta} f_j^{(0)}] + \mathcal{K}_{ij,\beta}[\partial_{V_\gamma} f_j^{(0)}] - \frac{2}{d}\delta_{\beta\gamma}\mathcal{K}_{ij,\lambda}[\partial_{V_\lambda} f_j^{(0)}]\right), \tag{C28}$$

$$D_i(\mathbf{V}) = \frac{1}{d}\mathbf{V}\cdot\nabla_{\mathbf{V}} f_i^{(0)} - \frac{1}{2}\left(\zeta_U + \frac{2}{nTd}p\right)\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_i^{(0)}) + \sum_{j=1}^{s}\left(n_j\partial_{n_j} f_i^{(0)} + \frac{1}{d}\mathcal{K}_{ij,\gamma}[\partial_{V_\gamma} f_j^{(0)}]\right), \tag{C29}$$

$$\mathbf{E}_i^j(\mathbf{V}) = -(\nabla_{\mathbf{V}} f_i^{(0)})\frac{1}{m_j}\left(\delta_{ij} - \frac{n_j m_j}{\rho}\right). \tag{C30}$$

Upon deriving Eqs. (C26)–(C30), use has been made of the relations

$$T\partial_T f_i^{(0)} = -\frac{1}{2}\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_i^{(0)}), \quad \partial_{U_\beta} f_i^{(0)} = -\partial_{V_\beta} f_i^{(0)}. \tag{C31}$$

The tensor derivative of the flow field $\partial_\gamma U_\eta$ has been expressed in terms of its independent trace and traceless parts, using the spherical symmetry of $f_i^{(0)}$, e.g.,

$$\begin{aligned}(V_\gamma\partial_{V_\eta} f_i^{(0)})\partial_\gamma U_\eta &= \hat{V}_\gamma\hat{V}_\eta(V\partial_V)f_i^{(0)}(\partial_\gamma U_\eta)\\ &= \hat{V}_\gamma\hat{V}_\eta(V\partial_V f_i^{(0)})\frac{1}{2}(\partial_\gamma U_\eta + \partial_\eta U_\gamma)\\ &= \hat{V}_\gamma\hat{V}_\eta(V\partial_V f_i^{(0)})\frac{1}{2}\left(\partial_\gamma U_\eta + \partial_\eta U_\gamma - \frac{2}{d}\delta_{\gamma\eta}\nabla\cdot\mathbf{U}\right) + \frac{1}{d}(V\partial_V f_i^{(0)})\nabla\cdot\mathbf{U},\end{aligned} \tag{C32}$$

and a similar analysis of the contribution from $\Sigma_{j=1}^{s}\mathcal{K}_{ij,\gamma}[\mathbf{v}|\partial_{V_\beta} f_j^{(0)}]$. Equation (C25) is an inhomogeneous, linear integral equation, where the inhomogeneity (the right side) is a linear combination of the the external force and the gradients of the hydrodynamic fields. The coefficients of these fields are specified functions of the velocity $\mathbf{V}$. Since by definition $f_i^{(1)}$ is proportional to the external force and the gradients of the hydrodynamic fields, it must have the form

$$\begin{aligned}f_i^{(1)} \to \boldsymbol{\mathcal{A}}_i(\mathbf{V})\cdot\nabla\ln T + \sum_{j=1}^{s}\boldsymbol{\mathcal{B}}_i^j(\mathbf{V})\cdot\nabla\ln n_j\\ + \mathcal{C}_{i,\gamma\eta}(\mathbf{V})\frac{1}{2}\left(\partial_\gamma U_\eta + \partial_\eta U_\gamma - \frac{2}{d}\delta_{\gamma\eta}\nabla\cdot\mathbf{U}\right)\\ + \mathcal{D}_i(\mathbf{V})\nabla\cdot\mathbf{U} + \sum_{j=1}^{s}\boldsymbol{\mathcal{E}}_i^j(\mathbf{V})\cdot\mathbf{F}_j.\end{aligned} \tag{C33}$$

The unknown functions of the peculiar velocity, $\boldsymbol{\mathcal{A}}_i$, $\boldsymbol{\mathcal{B}}_i^j$, $\mathcal{C}_{i,\gamma\eta}$, $\mathcal{D}_i$, and $\boldsymbol{\mathcal{E}}_i^j$ are determined by solving Eq. (C25). By dimensional analysis,

$$\boldsymbol{\mathcal{A}}_i(\mathbf{V}) = v_0^{-d}\ell^{1-d}\boldsymbol{\mathcal{A}}_i^*(\mathbf{V}^*),$$

$$\boldsymbol{\mathcal{B}}_i^j(\mathbf{V}) = v_0^{-d}\ell^{1-d}\boldsymbol{\mathcal{B}}_i^{j*}(\mathbf{V}^*),$$

$$\mathcal{C}_{i,\gamma\eta}(\mathbf{V}) = v_0^{-(d+1)}\ell^{1-d}\mathcal{C}_{i,\gamma\eta}^*(\mathbf{V}^*),$$

$$\mathcal{D}_i(\mathbf{V}) = v_0^{-(d+1)}\ell^{1-d}\mathcal{D}_i^*(\mathbf{V}^*),$$

and

$$\boldsymbol{\mathcal{E}}_i^j(\mathbf{V}) = m^{-1}v_0^{-(d+2)}\ell^{1-d}\boldsymbol{\mathcal{E}}_i^{j*}(\mathbf{V}^*),$$

where ℓ is an effective mean free path and $\boldsymbol{\mathcal{A}}_i^*(\mathbf{V}^*)$, $\boldsymbol{\mathcal{B}}_i^{j*}(\mathbf{V}^*)$, $\mathcal{C}_{i,\gamma\eta}^*(\mathbf{V}^*)$, $\mathcal{D}_i^*(\mathbf{V}^*)$, and $\boldsymbol{\mathcal{E}}_i^{j*}(\mathbf{V}^*)$ are dimensionless functions of the reduced velocity $\mathbf{V}^* = \mathbf{V}/v_0$, $v_0 = \sqrt{2T/m}$ being a thermal speed. Consequently,

$$\partial_t^{(0)}\boldsymbol{\mathcal{A}}_i(\mathbf{V}) = -\zeta^{(0)}T\partial_T\boldsymbol{\mathcal{A}}_i(\mathbf{V}) = \frac{1}{2}\zeta^{(0)}\nabla_{\mathbf{V}}\cdot(\mathbf{V}\boldsymbol{\mathcal{A}}_i(\mathbf{V})), \tag{C34}$$

$$\partial_t^{(0)}\boldsymbol{\mathcal{B}}_i^j(\mathbf{V}) = -\zeta^{(0)}T\partial_T\boldsymbol{\mathcal{B}}_i^j(\mathbf{V}) = \frac{1}{2}\zeta^{(0)}\nabla_{\mathbf{V}}\cdot(\mathbf{V}\boldsymbol{\mathcal{B}}_i^j(\mathbf{V})), \tag{C35}$$

$$\begin{aligned}\partial_t^{(0)}\mathcal{C}_{i,\gamma\eta}(\mathbf{V}) &= -\zeta^{(0)}T\partial_T\mathcal{C}_{i,\gamma\eta}(\mathbf{V})\\ &= \frac{1}{2}\zeta^{(0)}\mathcal{C}_{i,\gamma\eta} + \frac{1}{2}\zeta^{(0)}\nabla_{\mathbf{V}}\cdot(\mathbf{V}\mathcal{C}_{i,\gamma\eta}(\mathbf{V})),\end{aligned} \tag{C36}$$

$$\partial_t^{(0)}\mathcal{D}_i(\mathbf{V}) = -\zeta^{(0)}T\partial_T\mathcal{D}_i(\mathbf{V}) = \frac{1}{2}\zeta^{(0)}\mathcal{D}_i + \frac{1}{2}\zeta^{(0)}\nabla_{\mathbf{V}}\cdot(\mathbf{V}\mathcal{D}_i(\mathbf{V})), \tag{C37}$$

$$\partial_t^{(0)}\boldsymbol{\mathcal{E}}_i^j(\mathbf{V}) = -\zeta^{(0)}T\partial_T\boldsymbol{\mathcal{E}}_i^j(\mathbf{V}) = \zeta^{(0)}\boldsymbol{\mathcal{E}}_i^j(\mathbf{V}) + \frac{1}{2}\zeta^{(0)}\nabla_{\mathbf{V}}\cdot(\mathbf{V}\boldsymbol{\mathcal{E}}_i^j(\mathbf{V})). \tag{C38}$$

In addition,

$$\begin{aligned}\partial_t^{(0)}\nabla\ln T &= \nabla\partial_t^{(0)}\ln T = -\nabla\zeta^{(0)}\\ &= -\frac{1}{2}\zeta^{(0)}\nabla\ln T - \sum_{j=1}^{s}n_j\frac{\partial\zeta^{(0)}}{\partial n_j}\nabla\ln n_j.\end{aligned} \tag{C39}$$

Since the external force and gradients of the fields are all independent, Eq. (C25) can be separated into independent equations for the coefficients of each. This leads to the set of linear, inhomogeneous integral equations

$$\left(\left(\mathcal{L} - \frac{1}{2}\zeta^{(0)}\right)\boldsymbol{\mathcal{A}}\right)_i = \mathbf{A}_i, \tag{C40}$$

$$(\mathcal{L}\boldsymbol{\mathcal{B}}^j)_i - n_j\frac{\partial\zeta^{(0)}}{\partial n_j}\boldsymbol{\mathcal{A}}_i = \mathbf{B}_i^j, \tag{C41}$$

$$\left(\left(\mathcal{L} + \frac{1}{2}\zeta^{(0)}\right)\mathcal{C}_{\gamma\eta}\right)_i = C_{i,\gamma\eta}, \tag{C42}$$

$$\left(\left(\mathcal{L} + \frac{1}{2}\zeta^{(0)}\right)\mathcal{D}\right)_i = D_i, \tag{C43}$$

$$((\mathcal{L}+\zeta^{(0)})\boldsymbol{\mathcal{E}}^j)_i=\mathbf{E}_i^j. \tag{C44}$$

The linear operator $\mathcal{L}$ is

$$(\mathcal{L}X)_i=\frac{1}{2}\zeta^{(0)}\nabla_{\mathbf{V}}\cdot(\mathbf{V}X_i)+(LX)_i. \tag{C45}$$

Notice that Eq. (C40) can be used in Eq. (C41) to give the equivalent representation for the latter

$$[\mathcal{L}(\boldsymbol{\mathcal{B}}^j-(2n_j\partial_{n_j}\ln\zeta^{(0)})\boldsymbol{\mathcal{A}})]_i=\mathbf{B}_i^j-(2n_j\partial_{n_j}\ln\zeta^{(0)})\mathbf{A}_i. \tag{C46}$$

This completes the CE solution up through first order in the gradients and first order in the external force. Once Eq. (6.1) has been solved for $f_i^{(0)}$ the integral equations for $\boldsymbol{\mathcal{A}}_i$, $\boldsymbol{\mathcal{B}}_i^j$, $\mathcal{C}_{i,\gamma\eta}$, $\mathcal{D}_i$, and $\boldsymbol{\mathcal{E}}_i^j$ can be solved for $f_i^{(1)}$. Then, the cooling rate, heat flux, and pressure tensor can be calculated as linear functions of the gradients and the external force, and the explicit forms for the transport coefficients identified.

APPENDIX D: AN EIGENVALUE PROBLEM FOR $\mathcal{L}$

To simplify and interpret the linear integral equations defining the first order solutions $\{f_i^{(1)}\}$ it is useful to identify a special set of eigenvalues and eigenfunctions for the operator $\mathcal{L}$. Consider the equation for $f_i^{(0)}$,

$$-\zeta^{(0)}T\partial_T f_i^{(0)}=\sum_{j=1}^{s}J_{ij}^{(0)}[\mathbf{r}_1,\mathbf{v}_1|f_i^{(0)},f_j^{(0)}]. \tag{D1}$$

Since the temperature occurs through the form (6.10), the temperature derivatives can be expressed as velocity derivatives

$$\frac{1}{2}\zeta^{(0)}\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_i^{(0)})=\sum_{j=1}^{s}J_{ij}^{(0)}[\mathbf{r}_1,\mathbf{v}_1|f_i^{(0)},f_j^{(0)}]. \tag{D2}$$

Noting that $\zeta^{(0)}\propto\sqrt{T}$, the derivative of this equation with respect to T gives directly

$$(\mathcal{L}T\partial_T f^{(0)})_i=\frac{1}{2}\zeta^{(0)}T\partial_T f_i^{(0)}, \tag{D3}$$

where use has been made of Eq. (C34), i.e., $T\partial_T f_i^{(0)}=-\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_i^{(0)})/2$. An equivalent dimensionless form is

$$(\mathcal{L}\nabla_{\mathbf{V}}\cdot(\mathbf{V}f^{(0)}))_i=\frac{1}{2}\zeta^{(0)}\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_i^{(0)}). \tag{D4}$$

In a similar way, differentiation of Eq. (D2) with respect to each component of the flow velocity gives

$$(\mathcal{L}\partial_{U_\gamma}f^{(0)})_i=-\frac{1}{2}\zeta^{(0)}\partial_{U_\gamma}f_i^{(0)}. \tag{D5}$$

Finally, differentiate Eq. (D2) with respect to each of the species densities and noting that the density dependence of all quantities occurs only through the $f^{(0)}$'s and the χ_{ij}'s, one gets

$$\begin{aligned}(\mathcal{L}\partial_{n_\ell}f^{(0)})_i&=\left(\partial_{n_\ell}\sum_{j=1}^{s}J_{ij}^{(0)}[f_i^{(0)},f_j^{(0)}]\right)\Bigg|_{f^{(0)}}\\&-\frac{1}{2}(\partial_{n_\ell}\zeta^{(0)})\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_i^{(0)})\\&=\frac{1}{2}(\partial_{n_\ell}\zeta^{(0)}|_{f^{(0)}}-\partial_{n_\ell}\zeta^{(0)})\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_i^{(0)})\\&=-\partial_{n_\ell}\ln\zeta^{(0)}|_{\chi_{ij}^{(0)}}\frac{1}{2}\zeta^{(0)}\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_i^{(0)}).\end{aligned} \tag{D6}$$

This last form can be simplified by taking into account Eq. (D4) to get

$$[\mathcal{L}(\partial_{n_\ell}f^{(0)}+\partial_{n_\ell}\ln\zeta^{(0)}|_{\chi_{ij}^{(0)}}\nabla_{\mathbf{V}}\cdot(\mathbf{V}f^{(0)}))]_i=0. \tag{D7}$$

In summary, there are $s+d+1$ eigenvalues and eigenfunctions of the operator $\mathcal{L}$. Equations (D4) and (D5) identify these for the eigenvalue $\zeta^{(0)}/2$ and the d-fold degenerate value $-\zeta^{(0)}/2$, respectively. Equation (D7) identifies the eigenfunctions for the s-fold degenerate eigenvalue 0. In dimensionless form this eigenvalue problem is written as

$$(\mathcal{L}\Psi^{(m)})_i=\lambda^{(m)}\Psi_i^{(m)}. \tag{D8}$$

The eigenvectors are

$$\Psi_i^{(\ell)}=n_\ell\partial_{n_\ell}f_i^{(0)}-2n_\ell\,\partial_{n_\ell}\ln\zeta^{(0)}|_{\chi_{ij}^{(0)}}\Psi_i^{(s+1)},\quad \ell=1,\ldots,s,$$

$$\Psi_i^{(s+1)}=-\frac{1}{2}\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_i^{(0)}),\quad \Psi_i^{(\gamma)}=-v_0\partial_{V_\gamma}f_i^{(0)}, \tag{D9}$$

with the corresponding eigenvalues

$$\lambda^{(m)}\Leftrightarrow\left(0,\ldots,0,\frac{1}{2}\zeta^{(0)},-\frac{1}{2}\zeta^{(0)},-\frac{1}{2}\zeta^{(0)},-\frac{1}{2}\zeta^{(0)}\right). \tag{D10}$$

These eigenvalues are the same as those of the linearized hydrodynamic equations in the long wavelength limit. This provides a direct link between hydrodynamics and the spectrum of the linearized Enskog operator. In addition to this physical interpretation, the eigenvalues and eigenfunctions allow a practical formulation of the integral equations, as follows.

1. Biorthogonal set

Define a scalar product by

$$(a,b)=\sum_{i=1}^{s}\int d\mathbf{V}a_i^\dagger(\mathbf{V})b_i(\mathbf{V}), \tag{D11}$$

where the dagger denotes complex conjugation. A biorthogonal basis set is then defined by the eigenfunctions $\Psi_i^{(m)}$ above, and

$$\psi_{\alpha i}=\left[\frac{\delta_{i1}}{n_i},\ldots,\frac{\delta_{is}}{n_i},\left(\frac{2m_i}{dm}V^{*2}-1\right),\frac{m_i}{\rho}\mathbf{V}^*\right], \tag{D12}$$

where $\mathbf{V}^*=\mathbf{V}/v_0$. The orthonormality condition

$$\sum_{i=1}^{s} \int d\mathbf{V}^* \psi_{\alpha i}(\mathbf{V}^*)\Psi_{\beta i}(\mathbf{V}^*) = \delta_{\alpha\beta} \tag{D13}$$

is easily verified. An associated projection operator is given by

$$(\mathcal{P}X)_i = \sum_{\alpha} \Psi_{\alpha i}(\mathbf{V}^*)\sum_{j} \int d\mathbf{V}^* \psi_{\alpha j}(\mathbf{V}^*)X_j(\mathbf{V}^*). \tag{D14}$$

It follows from Eq. (D4) that $\mathcal{P}^2=\mathcal{P}$. The corresponding orthogonal projection is

$$\mathcal{Q} = 1 - \mathcal{P}. \tag{D15}$$

Consider the quantity $(\mathcal{P}\mathcal{L}X)_i$,

$$(\mathcal{P}\mathcal{L}X)_i = \sum_{\alpha} \Psi_{\alpha i}(\mathbf{V}^*)\sum_{j} \int d\mathbf{V}^* \psi_{\alpha j}(\mathbf{V}^*)(\mathcal{L}X)_j$$

$$= \Psi_i^{(s+1)}(\mathbf{V}^*)\sum_{j=1}^{s} \frac{2m_j}{dm} \int d\mathbf{V}^* V^{*2}(\mathcal{L}X)_j. \tag{D16}$$

Only the projection onto $\Psi_i^{(s+1)}$ contributes due to conservation of species number and momentum. It follows then that

$$(\mathcal{P}\mathcal{L}\boldsymbol{\mathcal{A}})_i = 0, \quad (\mathcal{P}\mathcal{L}\boldsymbol{\mathcal{B}}^j)_i = 0, \quad (\mathcal{P}\mathcal{L}\boldsymbol{\mathcal{E}}^j)_i = 0, \quad (\mathcal{P}\mathcal{L}\mathcal{C})_i = 0. \tag{D17}$$

The terms with $\boldsymbol{\mathcal{A}}$, $\boldsymbol{\mathcal{B}}^j$, and $\boldsymbol{\mathcal{E}}^j$ vanish from symmetry since these are all vectors; the last equality follows because $\mathcal{C}$ has zero trace. Next, note that

$$\zeta_U \nabla \cdot \mathbf{U} = \zeta^{(1)} = \frac{2}{dnT}\sum_{i=1}^{s} \int d\mathbf{v}_1 \frac{1}{2} m_i v_1^2 (\mathcal{L}\mathcal{D})_i$$

$$- \frac{2}{dnT}\sum_{i,j=1}^{s} \int d\mathbf{v}_1 \frac{1}{2} m_i v_1^2 \mathcal{K}_{ij,\eta}[\partial_{V_\eta} f_j^{(0)}], \tag{D18}$$

or equivalently

$$\Psi_i^{s+1}\zeta_U = \Psi_{2i}\sum_{j=1}^{s} \int d\mathbf{v}_1 \psi_j^{s+1}\mathcal{L}_j[\mathcal{D}]$$

$$- \Psi_i^{s+1}\frac{2m}{dnT}\sum_{j=1}^{s} \int d\mathbf{v}_1 \frac{1}{2md v_0} m_j v_1^2 \sum_{p} \mathcal{K}_{jp,\eta}[\partial_{V_\eta^*} f_j^{(0)}]$$

$$= \Psi_{2i}\sum_{j=1}^{s} \int d\mathbf{v}_1 \psi_{2j}\mathcal{L}_j[\mathcal{D}] - \mathcal{P}\frac{1}{dv_0}\mathcal{K}_\eta[\partial_{V_\eta^*} f^{(0)}]$$

$$= \mathcal{P}\mathcal{L}_j[\mathcal{D}] + \mathcal{P}\frac{1}{dv_0}\mathcal{K}_\eta[\partial_{V_\eta^*} f^{(0)}]. \tag{D19}$$

This can be used to eliminate the explicit occurrence of the transport coefficient ζ_U in the integral equation (C43). Finally, two additional identities are needed for the proofs of Appendix E

$$-\partial_{V_\gamma^*} f_i^{(0)} \frac{p-nT}{v_0^2\rho} = \mathcal{P}\sum_{j=1}^{s} \frac{1}{2v_0}\mathcal{K}_{ij,\gamma}[\nabla_{\mathbf{V}^*}\cdot(\mathbf{V}^* f_j^{(0)})], \tag{D20}$$

$$(\nabla_{\mathbf{V}^*} f_i^{(0)})(v_0^2\rho)^{-1} n_j \partial_{n_j}(p-nT)$$

$$= -\mathcal{P}\sum_{\ell=1}^{s} \frac{1}{v_0}\mathcal{K}_{i\ell}\left[\left(n_j\partial_{n_j} + \frac{1}{2}I_{i\ell j}\right) f_\ell^{(0)}\right]. \tag{D21}$$

APPENDIX E: SOLUBILITY CONDITIONS AND UNIQUENESS

The results of Appendix D allow proof that the integral equations have solutions and that they are unique. These equations have the generic form

$$(\mathcal{L} - \lambda)\mathcal{X} = X, \tag{E1}$$

where λ is one of the eigenvalues (D10). Let relation $\mathcal{X}$ be a solution to Eq. (E1). Then adding any solution to the corresponding homogeneous integral equation also gives a solution

$$\mathcal{X}' = \mathcal{X} + c\Psi, \tag{E2}$$

where $(\mathcal{L}-\lambda)\ \Psi=0$. However, the property

$$(\mathcal{P}f^{(1)})_i = \sum_{\alpha} \Psi_{\alpha i}(\mathbf{V})\sum_{j} \int d\mathbf{v}\psi_{\alpha j}(\mathbf{V}) f_j^{(1)}(\mathbf{V}) = 0 \tag{E3}$$

follows from the fact that the average densities, temperature, and flow velocity are given exactly by the first order term $f_i^{(0)}$, so that contributions to these averages from all higher order terms must vanish. Equivalently, Eq. (E3) implies

$$\mathcal{P}\begin{pmatrix} \boldsymbol{\mathcal{A}} \\ \boldsymbol{\mathcal{B}}^j \\ \mathcal{C} \\ \mathcal{D} \\ \boldsymbol{\mathcal{E}}^j \end{pmatrix} = 0. \tag{E4}$$

Consequently, the solution to Eq. (E1) with the condition (E4) is unique.

To show that solutions exist the integral equations are written in the equivalent form

$$\left(\mathcal{Q}\left(\mathcal{L} - \frac{1}{2}\zeta^{(0)}\right)\boldsymbol{\mathcal{A}}\right)_i = \mathcal{Q}\mathbf{A}_i, \tag{E5}$$

$$\left(\mathcal{Q}\mathcal{L}\left(\boldsymbol{\mathcal{B}}^j - 2n_j\frac{\partial \ln \zeta^{(0)}}{\partial n_j}\boldsymbol{\mathcal{A}}\right)\right)_i = \mathcal{Q}\left(\mathbf{B}_i^j - 2n_j\frac{\partial \ln \zeta^{(0)}}{\partial n_j}\mathbf{A}_i\right), \tag{E6}$$

$$\left(\mathcal{Q}\left(\mathcal{L} + \frac{1}{2}\zeta^{(0)}\right)\mathcal{C}_{\gamma\eta}\right)_i = \mathcal{Q}C_{i,\gamma\eta}, \tag{E7}$$

$$\left(\mathcal{Q}\left(\mathcal{L} + \frac{1}{2}\zeta^{(0)}\right)\mathcal{D}\right)_i = \mathcal{Q}D_i, \tag{E8}$$

$$(\mathcal{Q}(\mathcal{L}+\zeta^{(0)})\boldsymbol{\mathcal{E}}^j)_i = \mathcal{Q}\mathbf{E}_i^j. \tag{E9}$$

These equations are the same as Eqs. (C40)–(C44). The appearance of the factors of $\mathcal{Q}$ simply represent a convenient rearrangement of those equations, using the identities of Appendix D. They show that the relevant linear operator is $\mathcal{Q}(\mathcal{L}-\lambda)$ where λ is one of the eigenvalues (D10). The orthogonal projection $\mathcal{Q}$ identifies the left eigenfunctions with zero eigenvalue as being those of the biorthogonal set $\psi_{\alpha i}$ in Eq. (D12). According to the Fredholm alternative [71], solutions to these equations exist if and only if the inhomogeneity is orthogonal to the null space of the left eigenfunctions. Here, all the inhomogeneities on the right sides of Eqs. (E5)–(E9) appear explicitly orthogonal to this null space. Hence solutions exist and are unique.

APPENDIX F: DETAILS OF CONSTITUTIVE EQUATIONS

The cooling rate, and fluxes of mass, momentum, and energy are given exactly as explicit integrals of solutions to the kinetic equation. Once the CE solution is obtained, approximately to first order in the gradients, these expressions give the cooling rate and fluxes in the form of the constitutive equations (5.9)–(5.12). The objective of this appendix is to simplify these expressions to the extent possible without making any approximations. This is accomplished in most cases by performing solid angle integrations using the results

$$\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})^n = \pi^{(d-1)/2}\frac{\Gamma\left(\frac{n+1}{2}\right)}{\Gamma\left(\frac{n+d}{2}\right)}g^n \equiv B_n g^n, \tag{F1}$$

$$\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})^n\hat{\boldsymbol{\sigma}} = B_{n+1}g^n\hat{\mathbf{g}}, \tag{F2}$$

$$\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})^n\hat{\sigma}_k\hat{\sigma}_\ell = \frac{B_n}{n+d}g^n(n\hat{g}_k\hat{g}_\ell+\delta_{k\ell}), \tag{F3}$$

$$\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})^n\hat{\sigma}_k\hat{\sigma}_\ell\hat{\sigma}_m = g^n\frac{B_{n+1}}{n+d+1}[(n-1)\hat{g}_k\hat{g}_\ell\hat{g}_m + \hat{g}_m\delta_{k\ell}+\hat{g}_k\delta_{m\ell}+\hat{g}_\ell\delta_{km}], \tag{F4}$$

where d is the dimension ($d\geq 2$), and $\Gamma(x)$ is the usual gamma function

$$\Gamma(x+1)=x\Gamma(x), \quad \Gamma\left(\frac{1}{2}\right)=\sqrt{\pi}, \quad \Gamma(1)=1. \tag{F5}$$

In addition, for the sake of convenience, henceforth we will use the notation $\mathbf{g}_{12}\equiv\mathbf{g}$.

To get the collisional transfer contributions to the fluxes, one has to consider the following expansion:

$$\begin{aligned}\int_0^1 dx f_{ij}(\mathbf{r}_1-x\boldsymbol{\sigma}_{ij},\mathbf{v}_1,\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t) &= \int_0^1 dx\chi_{ij}(\mathbf{r}_1-x\boldsymbol{\sigma}_{ij},\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij}|\{n_i\})f_i(\mathbf{r}_1-x\boldsymbol{\sigma}_{ij},\mathbf{v}_1;t)f_j(\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij},\mathbf{v}_2;t)\\ &\to \chi_{ij}^{(0)}(\sigma_{ij};\{n_i\})f_i^{(0)}(\mathbf{v}_1;t)f_j^{(0)}(\mathbf{v}_2;t)+\frac{1}{2}\chi_{ij}^{(0)}(\sigma_{ij};\{n_i\})[f_i^{(0)}(\mathbf{v}_1;t)\partial_{y_\beta}f_j^{(0)}(\mathbf{v}_2;t)\\ &\quad -f_j^{(0)}(\mathbf{v}_2;t)\partial_{y_\beta}f_i^{(0)}(\mathbf{v}_1;t)]\boldsymbol{\sigma}_{ij}\cdot\nabla y_\beta+\chi_{ij}^{(0)}[f_i^{(0)}(\mathbf{v}_1;t)f_j^{(1)}(\mathbf{r}_1,\mathbf{v}_2;t)\\ &\quad +f_i^{(1)}(\mathbf{v}_1;t)f_j^{(0)}(\mathbf{r}_1,\mathbf{v}_2;t)]+f_i^{(0)}(\mathbf{v}_1;t)f_j^{(1)}(\mathbf{r}_1,\mathbf{v}_2;t)\delta\chi_{ij},\end{aligned} \tag{F6}$$

where $\delta\chi_{ij}$ is defined by

$$\delta\chi_{ij}=\sum_{\ell=1}^{s}(\nabla_{\mathbf{r}_1}n_\ell(\mathbf{r}_1;t))\int_0^1 dx\int d\mathbf{r}'(\mathbf{r}'-\mathbf{r}_1)\left.\frac{\delta\chi_{ij}(\mathbf{r}_1-x\boldsymbol{\sigma}_{ij},\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij}|\{n_i\})}{\delta n_\ell(\mathbf{r}',t)}\right|_{\delta n=0}. \tag{F7}$$

The functional derivative is evaluated at $\delta n=0$ and so it depends only on differences of pairs of coordinates, as in Eq. (C7). A change of variables then makes the dependence on x explicit,

$$\int d\mathbf{r}'(\mathbf{r}'-\mathbf{r}_1)\ \left.\frac{\delta\chi_{ij}(\mathbf{r}_1-x\boldsymbol{\sigma}_{ij},\mathbf{r}_1+(1-x)\boldsymbol{\sigma}_{ij}|\{n_i\})}{\delta n_\ell(\mathbf{r}',t)}\right|_{\delta n=0}=\int d\mathbf{r}''\left(\mathbf{r}''+\frac{1}{2}\boldsymbol{\sigma}_{ij}-x\boldsymbol{\sigma}_{ij}\right)\ \left.\frac{\delta\chi_{ij}\left(-\frac{1}{2}\boldsymbol{\sigma}_{ij},\frac{1}{2}\boldsymbol{\sigma}_{ij}|\{n_i\}\right)}{\delta n_\ell(\mathbf{r}'',t)}\right|_{\delta n=0}$$

$$=-\left(x-\frac{1}{2}\right)\boldsymbol{\sigma}_{ij}\ \left.\frac{\partial\chi_{ij}^{(0)}(\boldsymbol{\sigma}_{ij}|\{n_i\})}{\partial n_\ell(\mathbf{r}'',t)}\right|_{\delta n=0}+\int d\mathbf{r}''\mathbf{r}''\ \left.\frac{\delta\chi_{ij}\left(-\frac{1}{2}\boldsymbol{\sigma}_{ij},\frac{1}{2}\boldsymbol{\sigma}_{ij}|\{n_i\}\right)}{\delta n_\ell(\mathbf{r}'',t)}\right|_{\delta n=0}$$

$$=-\frac{\chi_{ij}^{(0)}}{n_\ell}\boldsymbol{\sigma}_{ij}\left[\left(x-\frac{1}{2}\right)n_\ell\frac{\partial\ln\chi_{ij}^{(0)}(\boldsymbol{\sigma}_{ij};\{n_k\})}{\partial n_\ell}-\frac{1}{2}I_{ij\ell}(\boldsymbol{\sigma}_{ij};\{n_k\})\right],\quad\text{(F8)}$$

where $I_{ij\ell}(\sigma_{ij};\{n_k\})$ is defined in Eq. (C11). Finally, then

$$\delta\chi_{ij}=\frac{1}{2}\chi_{ij}^{(0)}\sum_{\ell=1}^{s}(\nabla_{\mathbf{r}_1}\ln n_\ell(\mathbf{r}_1;t))\cdot\boldsymbol{\sigma}_{ij}(I_{ij\ell}(\sigma_{ij};\{n_k\})).\quad\text{(F9)}$$

1. Cooling rate

Since ζ is a scalar, the only gradient contributions are proportional to $\nabla\cdot\mathbf{U}$, and Eq. (4.27) to first order in the gradients becomes

$$\zeta=\zeta^{(0)}+\zeta_U\nabla\cdot\mathbf{U},\quad\text{(F10)}$$

with

$$\zeta^{(0)}=\frac{1}{2dnT}\sum_{i,j=1}^{s}(1-\alpha_{ij}^2)m_i\mu_{ji}\chi_{ij}^{(0)}\sigma_{ij}^{d-1}\times\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})^3\times f_i^{(0)}(\mathbf{V}_1)f_j^{(0)}(\mathbf{V}_2),\quad\text{(F11)}$$

$$\zeta_U=\frac{1}{2dnT}\sum_{i,j=1}^{s}(1-\alpha_{ij}^2)m_i\mu_{ji}\chi_{ij}^{(0)}\sigma_{ij}^{d-1}\times\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})^3\times\left[\frac{1}{d}f_j^{(0)}(\mathbf{V}_2)\boldsymbol{\sigma}_{ij}\cdot\nabla_{\mathbf{V}_1}f_i^{(0)}(\mathbf{V}_1)+2f_i^{(0)}(\mathbf{V}_1)\mathcal{D}_j(\mathbf{V}_2)\right].\quad\text{(F12)}$$

Performing the solid angle integrals gives

$$\zeta^{(0)}=\frac{B_3}{2dnT}\sum_{i,j=1}^{s}(1-\alpha_{ij}^2)\frac{m_im_j}{m_i+m_j}\chi_{ij}^{(0)}\sigma_{ij}^{d-1}\int d\mathbf{v}_1\int d\mathbf{v}_2g^3f_i^{(0)}\times(\mathbf{V}_1)f_j^{(0)}(\mathbf{V}_2),\quad\text{(F13)}$$

$$\zeta_U=\frac{B_4}{2d^2nT}\sum_{i,j=1}^{s}(1-\alpha_{ij}^2)\frac{m_im_j}{m_i+m_j}\chi_{ij}^{(0)}\sigma_{ij}^{d}\times\int d\mathbf{v}_1\int d\mathbf{v}_2g^2f_j^{(0)}(\mathbf{V}_2)(\mathbf{g}\cdot\nabla_{\mathbf{V}_1}f_i^{(0)}(\mathbf{V}_1))+\frac{B_3}{dnT}\sum_{i,j=1}^{s}(1-\alpha_{ij}^2)\frac{m_im_j}{m_i+m_j}\chi_{ij}^{(0)}\sigma_{ij}^{d-1}\times\int d\mathbf{v}_1\int d\mathbf{v}_2g^3f_i^{(0)}(\mathbf{V}_1)\mathcal{D}_j(\mathbf{V}_2).\quad\text{(F14)}$$

Finally, an integration by parts in the first term of the velocity integrals gives the result quoted in the text,

$$\zeta_U=-\frac{d+2}{2dnT}B_4\sum_{i,j=1}^{s}(1-\alpha_{ij}^2)\frac{m_im_j}{m_i+m_j}\chi_{ij}^{(0)}\sigma_{ij}^{d}n_in_j\left(\frac{T_i^{(0)}}{m_i}+\frac{T_j^{(0)}}{m_j}\right)+\frac{B_3}{dnT}\sum_{i,j=1}^{s}(1-\alpha_{ij}^2)\frac{m_im_j}{m_i+m_j}\chi_{ij}^{(0)}\sigma_{ij}^{d-1}\times\int d\mathbf{v}_1\int d\mathbf{v}_2g^3f_i^{(0)}(\mathbf{V}_1)\mathcal{D}_j(\mathbf{V}_2),\quad\text{(F15)}$$

where the species temperatures are defined by

$$\frac{d}{2}n_iT_i^{(0)}=\int d\mathbf{v}\frac{1}{2}m_iV^2f_i^{(0)}(\mathbf{V}).\quad\text{(F16)}$$

In the case of mechanically equivalent particles, Eq. (F15) reduces to previous results obtained for a monocomponent gas [62,72].

2. Mass flux

The mass fluxes are determined from the definition of Eq. (4.16)

$$\mathbf{j}_{0i}(\mathbf{r}_1,t) \to m_i \int d\mathbf{v}\mathbf{V} f_i^{(1)}(\mathbf{r}_1,\mathbf{v};t)$$
$$= \frac{1}{d}\int d\mathbf{v} m_i \mathbf{V}\cdot\left(\boldsymbol{\mathcal{A}}_i(\mathbf{V})\nabla \ln T + \sum_{j=1}^{s}(\boldsymbol{\mathcal{B}}_i^j(\mathbf{V})\nabla \ln n_j + \boldsymbol{\mathcal{E}}_i^j(\mathbf{V})\mathbf{F}_j)\right), \tag{F17}$$

where the contribution from $f_i^{(0)}$ vanishes. The transport coefficients according to Eq. (5.10) are identified as

$$D_i^T = -\frac{m_i}{\rho d}\int d\mathbf{v}\mathbf{V}\cdot\boldsymbol{\mathcal{A}}_i(\mathbf{V}), \tag{F18}$$

$$D_{ij} = -\frac{\rho}{m_j n_j d}\int d\mathbf{v}\mathbf{V}\cdot\boldsymbol{\mathcal{B}}_i^j(\mathbf{V}), \tag{F19}$$

$$D_{ij}^F = -\frac{m_i}{d}\int d\mathbf{v}\mathbf{V}\cdot\boldsymbol{\mathcal{E}}_i^j(\mathbf{V}). \tag{F20}$$

3. Energy flux

The energy flux to first order in the gradients is obtained from Eqs. (4.18) and (4.19) as

$$\mathbf{q} = \mathbf{q}^k + \mathbf{q}^c. \tag{F21}$$

The kinetic contribution is

$$\mathbf{q}^k = \sum_{i=1}^{s}\int d\mathbf{v}_1 \frac{1}{2} m_i V_1^2 \mathbf{V}_1 f_i^{(1)}(\mathbf{r}_1,\mathbf{v}_1;t)$$
$$= \frac{1}{d}\sum_{i=1}^{s}\int d\mathbf{v}_1 \frac{1}{2} m_i V_1^2 \mathbf{V}_1\cdot\boldsymbol{\mathcal{A}}_i(\mathbf{V}_1)\nabla \ln T$$
$$+ \frac{1}{d}\sum_{i,j=1}^{s}\int d\mathbf{v}_1 \frac{1}{2} m_i V_1^2 \mathbf{V}_1\cdot(\boldsymbol{\mathcal{B}}_i^j(\mathbf{V}_1)\nabla \ln n_j + \boldsymbol{\mathcal{E}}_i^j(\mathbf{V}_1)\mathbf{F}_j). \tag{F22}$$

The contributions proportional to derivatives of the flow velocity vanish from symmetry. The collisional transfer contribution is

$$\mathbf{q}^c = \sum_{i,j=1}^{s}\frac{1}{8}(1+\alpha_{ij})m_j\mu_{ij}\sigma_{ij}^d\chi_{ij}^{(0)}\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})$$
$$\times(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})^2\hat{\boldsymbol{\sigma}}[-(1-\alpha_{ij})(\mu_{ij}-\mu_{ji})(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}) + 4(\mathbf{G}_{ij}\cdot\hat{\boldsymbol{\sigma}})]$$
$$\times\Big[f_i^{(0)}(\mathbf{V}_1)f_j^{(1)}(\mathbf{V}_2) + f_i^{(1)}(\mathbf{V}_1)f_j^{(0)}(\mathbf{V}_2)$$
$$-\frac{1}{2}f_j^{(0)}(\mathbf{V}_2)\partial_{y_\beta}f_i^{(0)}(\mathbf{V}_1)\boldsymbol{\sigma}_{ij}\cdot\nabla y_\beta$$
$$+\frac{1}{2}f_i^{(0)}(\mathbf{V}_1)\partial_{y_\beta}f_j^{(0)}(\mathbf{V}_2)\boldsymbol{\sigma}_{ij}\cdot\nabla y_\beta\Big], \tag{F23}$$

where $\mathbf{G}_{ij}\equiv\mu_{ij}\mathbf{V}_1+\mu_{ji}\mathbf{V}_2$. The contribution from $\delta\chi_{ij}$ in Eq. (F9) vanishes from symmetry. The angular integrals can be performed to get

$$q_\gamma^c = \sum_{i,j=1}^{s}\frac{1}{8}(1+\alpha_{ij})m_j\mu_{ij}\sigma_{ij}^d\chi_{ij}^{(0)}\int d\mathbf{v}_1\int d\mathbf{v}_2\Big\{-B_4(1-\alpha_{ij})$$
$$\times(\mu_{ij}-\mu_{ji})g^2 g_\gamma + \frac{4B_2}{2+d}[2(\mathbf{G}_{ij}\cdot\mathbf{g})g_\gamma + g^2 G_{ij,\gamma}]$$
$$\times(f_i^{(0)}(\mathbf{V}_1)f_j^{(1)}(\mathbf{V}_2) + f_i^{(1)}(\mathbf{V}_1)f_j^{(0)}(\mathbf{V}_2))$$
$$+\frac{B_3}{3+d}[-(1-\alpha_{ij})(\mu_{ij}-\mu_{ji})g^3(3\hat{g}_\gamma\hat{g}_\eta+\delta_{\gamma\eta})$$
$$+4g^2((\mathbf{G}_{ij}\cdot\hat{\mathbf{g}})(\hat{g}_\gamma\hat{g}_\eta+\delta_{\gamma\eta}) + \hat{g}_\gamma G_{ij,\eta} + \hat{g}_\eta G_{ij,\gamma})]$$
$$\times\frac{1}{2}[f_i^{(0)}(\mathbf{V}_1)\partial_{y_\beta}f_j^{(0)}(\mathbf{V}_2) - f_j^{(0)}(\mathbf{V}_2)\partial_{y_\beta}f_i^{(0)}(\mathbf{V}_1)]\partial_\eta y_\beta\Big\}. \tag{F24}$$

Interchanging the labels i,j and $\mathbf{v}_1,\mathbf{v}_2$ it is seen that the contributions from $f_i^{(1)}$ and $f_j^{(1)}$ are the same. For the same reason the contributions from $\partial_{y_\beta}f_i^{(0)}$ and $-\partial_{y_\beta}f_j^{(0)}$ are the same. The first terms of the integrand give velocity moments of $f_j^{(1)}$ of degree one and three, which are proportional to the (partial) mass and kinetic energy fluxes. Finally, the only contributions from $\partial_{y_\beta}f_i^{(0)}$ are those that are scalar functions of the velocities, i.e., those proportional to temperature and species density gradients. The final result is therefore

$$\mathbf{q}^c = \sum_{i,j=1}^{s}\frac{1}{8}(1+\alpha_{ij})m_j\mu_{ij}\sigma_{ij}^d\chi_{ij}^{(0)}\Big[2B_4(1-\alpha_{ij})(\mu_{ij}$$
$$-\mu_{ji})n_i\left(\frac{2}{m_j}\mathbf{q}_j^k + (d+2)\frac{T_i^{(0)}}{m_i m_j}\mathbf{j}_{0j}^{(1)}\right)$$
$$+\frac{8B_2}{2+d}n_i\left(\frac{2\mu_{ji}}{m_j}\mathbf{q}_j^k - (d+2)\frac{T_i^{(0)}}{m_i m_j}(2\mu_{ij}-\mu_{ji})\mathbf{j}_{0j}^{(1)}\right)$$
$$+C_{ij}^T\nabla \ln T + \sum_{p=1}^{s}C_{ijp}^T\nabla \ln n_p\Big], \tag{F25}$$

where $\mathbf{j}_{0i}^{(1)}$ is defined by Eq. (F17) and the partial kinetic energy flux is

$$\mathbf{q}_i^k = \int d\mathbf{v}\frac{m_i}{2}V^2\mathbf{V}f_i^{(1)}(\mathbf{V}). \tag{F26}$$

The constants C_{ij}^T and C_{ijp}^T are

$$C_{ij}^T = \frac{B_3}{d}\int d\mathbf{v}_1\int d\mathbf{v}_2[-(1-\alpha_{ij})(\mu_{ij}-\mu_{ji})g^3$$
$$+4g(\mathbf{g}\cdot\mathbf{G}_{ij})]f_i^{(0)}(\mathbf{V}_1)T\partial_T f_j^{(0)}(\mathbf{V}_2), \tag{F27}$$

$$C_{ijp}^T = \frac{B_3}{d}\int d\mathbf{v}_1\int d\mathbf{v}_2[-(1-\alpha_{ij})(\mu_{ij}-\mu_{ji})g^3$$
$$+4g(\mathbf{g}\cdot\mathbf{G}_{ij})]f_i^{(0)}(\mathbf{V}_1)n_p\partial_{n_p}f_j^{(0)}(\mathbf{V}_2). \tag{F28}$$

The expression of C_{ij}^T can be simplified when one takes into account the relation

$$T\partial_T f_j^{(0)}(\mathbf{V}) = -\frac{1}{2}\nabla_{\mathbf{V}}\cdot(\mathbf{V}f_j^{(0)}(\mathbf{V})), \tag{F29}$$

and integrates by parts in Eq. (F27). The result is

$$C_{ij}^T = -\frac{2B_3}{d}\int d\mathbf{v}_1\int d\mathbf{v}_2 f_i^{(0)}(\mathbf{V}_1)f_j^{(0)}(\mathbf{V}_2)\Bigg\{gG_{ij}^2 + g^{-1}(\mathbf{g}\cdot\mathbf{G}_{ij})^2 + (1+\mu_{ji})g(\mathbf{g}\cdot\mathbf{G}_{ij}) + \mu_{ji}\mu_{ij}g^3 + \frac{3}{4}(1-\alpha_{ij})(\mu_{ji}-\mu_{ij}) \times[g(\mathbf{g}\cdot\mathbf{G}_{ij}) + g^3]\Bigg\}. \tag{F30}$$

On the other hand, no significant further simplification of Eq. (F28) is possible until $f_i^{(0)}$ is specified in detail.

The heat flux is seen to have the form (5.11)

$$\mathbf{q}(\mathbf{r},t) \to -\lambda\nabla T - \sum_{i,j=1}^{s}(T^2 D_{q,ij}\nabla\ln n_j + L_{ij}\mathbf{F}_j), \tag{F31}$$

so the transport coefficients now can be identified,

$$\lambda = \lambda^k + \lambda^c, \quad D_{q,ij} = D_{q,ij}^k + D_{q,ij}^c, \quad L_{ij} = L_{ij}^k + L_{ij}^c. \tag{F32}$$

The kinetic parts are

$$\lambda^k = \sum_{i=1}^{s}\lambda_i^k = -\frac{1}{dT}\sum_{i=1}^{s}\int d\mathbf{v}\frac{m_i}{2}V^2\mathbf{V}\cdot\boldsymbol{\mathcal{A}}_i(\mathbf{V}), \tag{F33}$$

$$D_{q,ij}^k = -\frac{1}{dT^2}\int d\mathbf{v}\frac{m_i}{2}V^2\mathbf{V}\cdot\boldsymbol{\mathcal{B}}_i^j(\mathbf{V}), \tag{F34}$$

$$L_{ij}^k = -\frac{1}{d}\int d\mathbf{v}\frac{m_i}{2}V^2\mathbf{V}\cdot\boldsymbol{\mathcal{E}}_i^j(\mathbf{V}), \tag{F35}$$

while the collisional transport parts are given by Eqs. (7.14)–(7.16).

4. Momentum flux

The momentum flux to first order in the gradients is obtained from Eq. (4.20)–(4.22),

$$P_{\gamma\lambda} \equiv P_{\gamma\lambda}^k + P_{\gamma\lambda}^c, \tag{F36}$$

where

$$P_{\gamma\lambda}^k \to \sum_{i=1}^{s}\int d\mathbf{v}_1 m_i V_{1\gamma}V_{1\lambda}(f_i^{(0)}(\mathbf{V}_1) + f_i^{(1)}(\mathbf{V}_1)) = \delta_{\gamma\lambda}nT + \sum_{i=1}^{s}\int d\mathbf{v}_1 m_i V_{1\lambda}V_{1\gamma}f_i^{(1)}(\mathbf{V}_1), \tag{F37}$$

$$P_{\gamma\lambda}^c = \frac{1}{2}\sum_{i,j=1}^{s} m_j\mu_{ij}(1+\alpha_{ij})\sigma_{ij}^d\chi_{ij}^{(0)}\int d\mathbf{v}_1\int d\mathbf{v}_2\int d\hat{\boldsymbol{\sigma}}\Theta(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}) \times(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g})^2\hat{\sigma}_\lambda\hat{\sigma}_\gamma\Bigg[f_i^{(0)}(\mathbf{V}_1)f_j^{(0)}(\mathbf{V}_2) + 2f_i^{(0)}(\mathbf{V}_1)f_j^{(1)}(\mathbf{V}_2) - \frac{1}{2}(f_j^{(0)}(\mathbf{V}_2)\partial_{y_\beta}f_i^{(0)}(\mathbf{V}_1) - f_i^{(0)}(\mathbf{V}_1)\partial_{y_\beta}f_j^{(0)}(\mathbf{V}_2))\boldsymbol{\sigma}_{ij}\cdot\nabla y_\beta\Bigg]. \tag{F38}$$

A contribution to Eq. (F38) proportional to the density gradients from the expansion of $\chi_{ij}(\mathbf{r}_1 - x\sigma_{ij}, \mathbf{r}_1 + (1-x)\sigma_{ij})$ vanishes from symmetry. For similar reasons, the only gradients contributing to both Eqs. (F37) and (F38) are those from the flow field. The terms proportional to $\mathcal{D}_i$ in Eq. (C33) also do not contribute due to the orthogonality condition (E4). The solid angle integrations can be performed with the results

$$P_{\gamma\lambda}^k \to \delta_{\gamma\lambda}nT + \frac{1}{2}\sum_{i=1}^{s}\int d\mathbf{v}_1 m_i V_{1\lambda}V_{1\gamma}\mathcal{C}_{i,\beta\mu}(\mathbf{V}_1)\left(\partial_\beta U_\mu + \partial_\mu U_\beta - \frac{2}{d}\delta_{\beta\mu}\nabla\cdot\mathbf{U}\right), \tag{F39}$$

$$P_{\gamma\lambda}^c = \delta_{\gamma\lambda}\frac{B_2}{2d}\sum_{i,j=1}^{s} m_j\mu_{ij}(1+\alpha_{ij})\sigma_{ij}^d\chi_{ij}^{(0)}n_i n_j\left(\frac{T_i^{(0)}}{m_i} + \frac{T_j^{(0)}}{m_j}\right) + \frac{B_2}{d+2}\sum_{i,j=1}^{s}\mu_{ij}(1+\alpha_{ij})\chi_{ij}^{(0)}n_i\sigma_{ij}^d\int d\mathbf{v}_2 m_j V_{2\gamma}V_{2\lambda}\mathcal{C}_{j,\beta\mu}(\mathbf{V}_2)\left(\partial_\beta U_\mu + \partial_\mu U_\beta - \frac{2}{d}\delta_{\beta\mu}\nabla\cdot\mathbf{U}\right) + \frac{1}{2}\frac{B_3}{3+d}\sum_{i,j=1}^{s} m_j\mu_{ij}(1+\alpha_{ij})\chi_{ij}^{(0)}\sigma_{ij}^d\int d\mathbf{v}_1\int d\mathbf{v}_2 f_j^{(0)}(V_2)(\partial_{V_{1\ell}}f_i^{(0)}(V_1)) \times g^2(\hat{g}_\lambda\hat{g}_\gamma\hat{g}_\mu + \hat{g}_\mu\delta_{\lambda\gamma} + \hat{g}_\gamma\delta_{\mu\lambda} + \hat{g}_\lambda\delta_{\gamma\mu})\partial_\mu U_\ell. \tag{F40}$$

An integration by parts in the velocity integral, and use of fluid symmetry gives, finally,

$$P_{\gamma\lambda}^{c}=\delta_{\gamma\lambda}\frac{B_2}{2d}\sum_{i,j=1}^{s}m_j\mu_{ij}(1+\alpha_{ij})\sigma_{ij}^{d}\chi_{ij}^{(0)}n_in_j\left(\frac{T_i^{(0)}}{m_i}+\frac{T_j^{(0)}}{m_j}\right)+\frac{B_2}{d+2}\sum_{i,j=1}^{s}\mu_{ij}(1+\alpha_{ij})\chi_{ij}^{(0)}n_i\sigma_{ij}^{d}\int d\mathbf{v}_2m_jV_{2\gamma}V_{2\lambda}\mathcal{C}_{j,\beta\mu}(\mathbf{V}_2)\Bigg(\partial_\beta U_\mu+\partial_\mu U_\beta$$
$$-\frac{2}{d}\delta_{\beta\mu}\nabla\cdot\mathbf{U}\Bigg)-\frac{B_3(d+1)}{2d(d+2)}\sum_{i,j=1}^{s}m_j\mu_{ij}(1+\alpha_{ij})\chi_{ij}^{(0)}\sigma_{ij}^{d}\int d\mathbf{v}_1\int d\mathbf{v}_2f_i^{(0)}(\mathbf{V}_1)f_j^{(0)}(\mathbf{V}_2)g\Bigg[\left(\partial_\lambda U_\gamma+\partial_\gamma U_\lambda-\frac{2}{d}\delta_{\gamma\lambda}\nabla\cdot\mathbf{U}\right)$$
$$+\frac{d+2}{d}\delta_{\gamma\lambda}\nabla\cdot\mathbf{U}\Bigg]. \tag{F41}$$

The pressure tensor therefore has the form (5.12), and the pressure, shear viscosity, and bulk viscosity are identified in terms of their kinetic and collisional transfer contributions. Their expressions are given by Eqs. (7.18)–(7.22), respectively.

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