Solution of the Multicomponent Boltzmann Equation Based on an Extended Set of Observables

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Abstract

We present the perturbative solution of the multicomponent Boltzmann kinetic equation based on the set of observables including the hydrodynamic velocity and temperature for each component. The solution is obtained by modifying the formal density scaling scheme by Enskog, such that the density of each component is scaled independently. As a result we obtain the species momentum and energy balance equations with the source terms describing the transfer of corresponding quantities between different components. In the zero order approximation those are the Euler equations with the momentum and heat diffusion included in the form of the classical Maxwell-Stefan diffusion terms. The first order approximation results in equations of a Navier-Stokes type with the partial viscosity and heat conductivity including only the correlations of the particles of the same component. The first order corrections to the Maxwell-Stefan terms as well as the contributions bilinear in gradients and differences of observables are calculated. The first order momentum source term is shown to include thermal diffusion. The nondiagonal (in component indexes) components of viscosity and heat conductivity appear as second order contributions.

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I. INTRODUCTION

The theory of transport phenomena in gases based on a kinetic theory dates back to the works of Chapman and Enskog [1, 2, 3] and is generally accepted as mature. It provides the perturbative solution of Boltzmann kinetic equation for the single- as well as multicomponent gases, resulting apart from other things in the microscopic basis for the Navier-Stokes equations. In spite of that the applicability of Chapman-Enskog solution to some particular problems is limited and a number of works has been published on modified and alternative derivation of transport equations from the microscopic kinetics [4, 5, 6, 7]. Dynamics of multicomponent flows has also attracted considerable interest of the statistical mechanical community in the past. The works on the subject include the derivations of species momentum and heat balance equations [8, 9], as well as the works devoted to discussion and modification of diffusion equations [10, 11, 12, 13]. A recent overview of multicomponent transport theories can be found in [14, 15].

The common recipe in modeling \( n \)-component transport problems is to solve the Navier-Stokes equation for the mixture, combined with \((n-1)\) Maxwell-Stefan equations, all with appropriate boundary conditions. Application of this principle to binary counterdiffusion in capillaries [16] leads however to large discrepancies with experimental results (see for example [7]). On the other hand, instead of the set of observables including the number density of each component \( (n_i) \), the hydrodynamic velocity \( (\mathbf{v}_0) \) and the temperature \( (T) \) of the mixture one may wish to use the set including the velocities and temperatures for every component \( (\mathbf{v}_{i0}, T_i) \). This choice of observables was made already in the works of Maxwell and Stefan [17, 18], but this has become obscured by the success of the Chapman-Enskog theory. The species momentum and heat balance equations resulting from such an approach can be also considered as a basis for multifluid models used in plasma theory [19]. It should also be noted that that the tools to solve multifluid transport equations numerically are well established [20].

Multifluid models with application to the disperate-mass binary mixtures were studied before in the framework of kinetic theory. The two-fluid theory was developed by Goldman and Sirovich [21] for the special case of Maxwell molecules, and later attempts were made to extend it to arbitrary interaction potentials [22]. However, quite elegant but rather ad-hoc derivation of de la Mora and Fernandez-Feria [22] is restricted to binary mixtures, and, in
spite of the similarity of the resulting transport equations with the present work, the use of "unmatched Maxwellians" that are not a solution of any equation creates a lot of confusion throughout the treatment, especially when dealing with momentum and energy transfer between the components. The problem was recently approached also by Kerkhof and Geboers \[7\]. Relating each component distribution function to its own hydrodynamic velocity, they obtained the momentum balance in the form similar to the Maxwell-Stefan equations. However, in their derivation inaccuracies are present \[23\] and thus serious reconsideration of the result is required. We like to stress that the present derivation is not related to ref. \[7\], although Kerkhof has also contributed to this work. We should also mention the little-known paper of Struminskii \[24\] that we came across at the latest stages of development. To derive the equations for multicomponent gas transport Struminskii employed the same basic idea of modified scaling that is used in the present work. However our treatment is very different from given there, and we claim our results to be more general and accurate compared to those of Struminskii. More detailed comparison with this and other papers is given in the text where appropriate.

In the present paper we aim to obtain the perturbative solution of the Chapman-Enskog type for the Boltzmann equation based on extended set of observables \(n_i, v_{i0}, T_i\), and to establish equations of motion for these variables, relating the transport coefficients to the microscopic properties of the mixture components. The paper is organized as follows: In section II we suggest a modification to the standard density scaling scheme of solving the Boltzmann equation. Equations for the distribution functions as well as the momentum and energy balance equations are given there for the arbitrary order in the formal scaling parameter. In section III we derive explicit expressions in the 0-th order in formal scaling parameter, while section IV contains the results of the higher orders of perturbation theory. Discussion of the results is given in section V.

II. MODIFIED DENSITY SCALING

We consider the multicomponent Boltzmann equation

\[
\mathcal{D} f_i = \sum_j J\{f_i f_j\},
\]  

(1)
which describes the evolution $D f_i = (\partial_t + v_i \cdot \nabla_x + F_i \cdot \nabla v_i) f_i$ of the component $i$ distribution function $f_i$ due to collisions with particles of the same and other components. The pairwise collisional operator is given by

$$J\{f_i f_j\} = \int d\mathbf{v}_j d\mathbf{k}_{ij} |\mathbf{v}_j - \mathbf{v}_i| \sigma_{ij}(|\mathbf{v}_j - \mathbf{v}_i|, \mathbf{k}_{ij}) (f'_i f'_j - f_i f_j),$$

where $\mathbf{k}_{ij}$ is the unit vector in the direction of relative velocity, and primed quantities denote those after collision. Here we only consider the case of spherically symmetric interactions, such that the cross-section $\sigma_{ij}(|\mathbf{v}_i - \mathbf{v}_j|, \mathbf{k}_{ij})$ to change the direction of relative velocity from $\mathbf{k}_{ij}$ to $\mathbf{k}'_{ij}$ depends only on $\mathbf{k}'_{ij} \cdot \mathbf{k}_{ij}$. We look for the solution of equation (1) based on the set of observables $\beta_i = (n_i, v_{i0}, T_i)$ including the hydrodynamic velocities ($v_{i0}$) and temperatures ($T_i$) for each of the components.

Following the standard procedure of Enskog we introduce the formally small scaling parameter $\varepsilon$, put equal to 1 at the end of derivation. The solution is then given in terms of a series in this parameter $f_i = \sum_n \varepsilon^n f_i^{(n)}$. In the classical treatment this parameter is used to scale the left-hand part of equation (1) which is equivalent to scaling the density of the mixture. Such a scaling is very natural if one is going to use the set of observables including the hydrodynamic velocity and the temperature of the mixture as a whole ($n_i, v_0, T$). Here, instead, we suggest a bit different scaling of equation (1)

$$\varepsilon[D f_i - \sum_{j \neq i} J\{f_i f_j\}] = J\{f_i f_i\}. \tag{2}$$

This corresponds to scaling only the chosen $i$-th component density, and allows the zero order solutions to be centered around different velocities, which is convenient for deriving the species momentum and energy balance equations. It should be noted that the similar reordering was proposed yet by Lorentz [25] for the heavy species in disparate mass binary mixture, and in modified form used later by many researchers [26, 27, 28]. However they use such a reordering of the terms to collect all the relevant contributions in equations of the same order, thus requiring different scaling patterns for different components, and as a consequence, the different transport equations. Here instead, we use the same form of scaling for every component, employing it as a formal tool, which allows to construct the contributions from observable derivatives up to desired order. Namely, in the present paper we limit ourselves to the thermal conductivity, thermal diffusion and viscous contributions. Using the same form of scaling independently of component masses allows to treat the mixtures with arbitrary number of components, but on the other hand it leads to slower convergence of the series in $m_i/\mu_{ij}$ for some transport coefficients in the case of very light
Expanding unknown functions $f_i$, operators $\mathcal{D}$ and $J\{f_if_j\}$ as well as the time derivatives of observables $\partial \beta_i/\partial t$ in series in scaling parameter $\epsilon$, and assuming after Enskog that the time enters only implicitly through $\beta$ and the spatial gradients of $\beta$

$$f_i(r, v_i, t) = f_i(r, v_i; \beta, \nabla_r \beta, \ldots) = \sum_n \epsilon^n f_i^{(n)},$$

$$\frac{\partial}{\partial t} \beta_i(r, t) \equiv \Phi_i(r; \beta, \nabla_r \beta, \ldots) = \sum_n \epsilon^n \Phi_i^{(n)}.$$  \hspace{1cm} \hspace{1cm} \hspace{1cm} (3)

we arrive at the following equations for the contribution of the order $n+1$ in a scaling parameter

$$J\{f_i^{(n+1)} f_i^{(0)}\} + J\{f_i^{(0)} f_i^{(n+1)}\} = \mathcal{D}^{(n)} f_i - \sum_{j \neq i} J^{(n)} \{f_i f_j\} - \sum_{k=1}^{n} J\{f_i^{(k)} f_i^{(n-k+1)}\},$$

$$J\{f_i^{(0)} f_i^{(0)}\} = 0,$$  \hspace{1cm} \hspace{1cm} \hspace{1cm} (4)

where the following notation is introduced

$$\mathcal{D}^{(n)} f_i = \sum_{m=0}^{n} (\Phi^{(m)} \cdot \nabla_{\beta} + \nabla_{v_i} \Phi^{(m)} : \nabla_{v_i} \beta + \ldots) f^{(n-m)} + (v_i \cdot \nabla_r + F \cdot \nabla_v) f^{(n)},$$

$$J^{(n)} \{f_i f_j\} = \sum_{m=0}^{n} J\{f_i^{(m)} f_j^{(n-m)}\}.$$  \hspace{1cm} \hspace{1cm} \hspace{1cm} (5)

The right-hand side of equations (4) contains only the functions which are known from the lower order calculations. The left-hand side, containing the functions to be calculated, includes only collisional operators for the species of the same type.

The solubility condition for equations (4), demanding that the right part of equation must be orthogonal to all solutions of the homogeneous equation, read as (the scaling parameter $\epsilon$ is already put equal to 1 here)

$$\frac{\partial \beta_i}{\partial t} + \sum_n \int d^3 v_i \psi_i(v_i \cdot \nabla_r + F \cdot \nabla_v) f^{(n)} + \sum_{j \neq i} \sum_n \int d^3 v_i \psi_i J^{(n)} \{f_i f_j\} = 0.$$  \hspace{1cm} \hspace{1cm} \hspace{1cm} (6)

As long as the solution of the homogeneous equation (4) consists of collisional invariants $\psi_i = (1, m_i v_i, \frac{1}{2} m_i c_i^2)$, with $c_i = v_i - v_{i0}$ the velocity related to coordinate set moving with the average $i$-th component velocity, the solubility condition provides us with the
corresponding conservation laws. After some manipulation this can be rewritten as

\[
\frac{1}{\rho_i} \frac{d\rho_i}{dt} = -\nabla \cdot \mathbf{v}_{i0},
\]

\[
\rho_i \frac{d\mathbf{v}_{i0}}{dt} = \rho_i \mathbf{F}_i - \sum_n \nabla \cdot \mathbf{P}^{(n)} + m_i \sum_{n,j} \int d^3 c_i c_j J^{(n)} \{f_i f_j\},
\]

\[
\rho_i \frac{du_i}{dt} = -\sum_n \nabla \cdot \mathbf{q}^{(n)} + \sum_n \mathbf{P}^{(n)} \cdot \nabla \mathbf{v}_{i0} + \frac{m_i}{2} \sum_{n,j} \int d^3 c_i c_j^2 J^{(n)} \{f_i f_j\}.
\] (7)

with partial pressure and the heat flow defined as

\[
\mathbf{P}^{(n)}_i = m_i \int d^3 c_i c_i f_i^{(n)},
\]

\[
\mathbf{q}^{(n)}_i = \frac{m_i}{2} \int d^3 c_i c_i^2 f_i^{(n)}.
\] (8)

Here it should be noted that \( \frac{1}{2} m_i c_i^2 \) can be considered as a collisional invariant only in a limited sense as it conserves only under collisions of the particles of the same type. This leads to the sum of the sources \( \sum_{i,j} \int d^3 c_i \frac{m_i c_i^2}{2} J^{(n)} \{f_i f_j\} \) in the heat balance equation of the set (7) being not equal to zero. However there is no contradiction here with the energy conservation as the temperatures for each component are defined in its own reference frame, and thus the energy balance would have also included the terms of the \( \rho_i v_{i0}^2 / 2 \) type. Additionally, these source terms can only be calculated exactly in some special cases, and in what follows we expand them assuming the velocity \( \mathbf{v}_{i0} - \mathbf{v}_{j0} \) and temperature \( T_i - T_j \) differences are small and keep only the first nonvanishing term of expansion. Thus, resulting equations should be understood as a linear in above mentioned differences approximation, while the sum of the source terms in (7) is obviously a contribution of the next order. Equations (7) can be seen as the single-component balance equations with the momentum and energy transport due to interaction between the components included as a source terms. This approach is equivalent to considering the single component system in a field of hydrodynamic forces exerted by the flow through the other components. We like to note however that quantities \( \mathbf{P}_i \) and \( \mathbf{q}_i \) can depend on the properties of the other components through the functions \( f_i^{(n)} \).

Similarly to original Enskog approach we derive here the transport equations in a general form with all contributions determined from equations (5,8). Calculation of these contributions is then straightforward (although elaborate), with all assumptions and approximations being very clear. This should be contrasted to the previous derivations, which often require cumbersome argumentation to justify some steps in there.
III. ZERO-ORDER APPROXIMATION

The solution of the equation (4) in the 0 order approximation and resulting in the proper observables values is the Maxwell distribution related to the frame moving with the average velocity of the \(i\)-th component \(\mathbf{v}_{i0}\)

\[
f_i^{(0)} = n_i(\gamma_i/2\pi)^{3/2} \exp \left\{ -\gamma_i(\mathbf{v}_i - \mathbf{v}_{i0})^2/2 \right\},
\]

where \(n_i\) is the number density of the \(i\)-th component, \(\gamma_i = m_i/T_i\) with \(m_i\) being the mass of the component’s molecules and \(T_i\) its kinetic temperature (we prefer using the energetic temperature scale, thus saving on \(k_B\)). The resulting partial pressure and the heat flow are

\[
P_i^{(0)} = p_iI = n_iT_iI,
\]

\[
q_i^{(0)} = 0,
\]

where \(I\) is the unit tensor.

The collision integral \(J^{(0)}\{f_if_j\}\) can be expanded in series in velocity and temperature difference between the components. Then, assuming these differences are small, compared to the thermal velocity and the temperature of the component correspondingly, we keep only the first nonvanishing terms of expansion. The contributions of the zero order in velocity and temperature differences are identically zero, while the most general form of the first order contribution would be

\[
J^{(0)}\{f_if_j\} = -f_i^{(0)} Q^{(v)}(c_i)(\mathbf{v}_{i0} - \mathbf{v}_{j0}) \cdot c_i - f_i^{(0)} Q^{(T)}(c_i)(T_i - T_j).
\]

Explicit form of the scalar functions \(Q^{(v)}(c_i)\) and \(Q^{(T)}(c_i)\) is rather complicated, and for the time being we keep them undefined. In the end it turns out that we only need to know the even moments of these functions with respect to \(c_i\) and not the functions themselves. Calculation of these moments, using equation (11) gives

\[
\int d^3c_i c_i^{2n} f_i^{(0)} Q^{(T)}(c_i) = -n_i \frac{\mu_{ij}}{T_iT_j} \frac{\Gamma(2n+2)}{2^{2n}} \left( \frac{\gamma_{ij}}{2} \right)^{-1} \left( \frac{\gamma_i + \gamma_j}{2} \right)^{-n} \sum_{m=1}^{n} \frac{\nu_{ij}^{(1;m)}}{\Gamma(n-m+1)},
\]

\[
\int d^3c_i c_i^{2n+2} f_i^{(0)} Q^{(v)}(c_i) = -n_i \frac{\Gamma(2n+4)}{2^{2n+1}(n+1)} \left( \frac{\gamma_{ij}}{2} \right)^{-n} \sum_{m=1}^{n+1} \frac{\nu_{ij}^{(1;m)}}{\Gamma(n-m+2)},
\]

where the hierarchy of frequencies is introduced

\[
\nu_{ij}^{(1;m)} = \frac{n_j m_{2m+3}}{(2\pi\gamma_{ij})^{1/2} \Gamma(2m+2)} \left( \frac{\mu_{ij}}{m_i} \right)^{2m-1} \left( \frac{\gamma_i + \gamma_j}{\gamma_{ij}} \right)^{m-1} \int_0^\infty d\xi \xi^{2m+3} e^{-\xi^2} Q_{ij}^{(l)}(\xi),
\]

\[
\nu_{ij}^{(1;m)} = \frac{n_j m_{2m+3}}{(2\pi\gamma_{ij})^{1/2} \Gamma(2m+2)} \left( \frac{\mu_{ij}}{m_i} \right)^{2m-1} \left( \frac{\gamma_i + \gamma_j}{\gamma_{ij}} \right)^{m-1} \int_0^\infty d\xi \xi^{2m+3} e^{-\xi^2} Q_{ij}^{(l)}(\xi),
\]
with \( \xi = \gamma_{ij}g/2 \), \( \mu_{ij} \) is the reduced mass of the components \( i \) and \( j \), \( \gamma_{ij} \) is similarly introduced reduced quantity, and \( Q_{ij}^{(l)}(g) = \int d\Omega \sigma_{ij}(g,\theta)(1 - \cos^l \theta) \) is the \( ij \) transport cross section of the \( l \)-th order. \( Q_{ij}^{(1)}(g) \) entering the zero order collision integral is sometimes referred to as a diffusion or momentum transfer cross section. When all components of the mixture have the same kinetic temperature these frequencies can be easily related to familiar Chapman-Cowling \( \Omega \)-integrals: \( \nu_{ij}^{(l,m)}|_{T_i = T_j} = n_j(m_j/m_i)^m(\mu_{ij}/m_j)Q_{ij}^{(l,m)} \). The finite series for the moments of collision integrals were calculated by Kolodner [29] for the special case of Maxwell molecules, and were used by Goldman and Sirovich [21] to develop the two-fluid theory for the mixtures of those molecules. Here we use the similar formalism to derive the multifluid theory for the species with arbitrary interaction potential. This should be contrasted to the work of Struminskii [24], where similar quantities were also calculated for the case of Maxwell molecules, and implicit assumption made that the same expression can be applied to the species with arbitrary interaction potential.

Extracting the required moments of \( J(0)\{f_i,f_j\} \) from (12) we can write down the transport equations in the following form

\[
\begin{align*}
\frac{1}{\rho_i} \frac{d\rho_i}{dt} &= -\nabla \cdot \mathbf{v}_{i0}, \\
\rho_i \frac{d\mathbf{v}_{i0}}{dt} &= \rho_i F_i - \nabla p_i - n_i \sum_j \mu_{ij} \nu_{ij}^{(1;1)}(\mathbf{v}_{i0} - \mathbf{v}_{j0}), \\
\rho_i \frac{du_i}{dt} &= -p_i \nabla \cdot \mathbf{v}_{i0} - 3n_i \sum_j \frac{\mu_{ij}}{m_i + m_j} \nu_{ij}^{(1;1)}(T_i - T_j). 
\end{align*}
\]  

(14)

The \( \nu_{ij}^{(1;1)} \) frequency here coincides with the effective collisional frequency of the electron commonly used in the plasma theory [19]. Thus in the zero order approximation we obtain the Euler equations for the ideal liquid including the Maxwell-Stefan terms for the heat and momentum diffusion due to interaction between different components. The zero order results are in total agreement with Struminskii [24] and binary Mora [22] equations, allowing to recover the relationship between diffusion rates, obtained by the Enskog method in the first approximation.

IV. FIRST ORDER APPROXIMATION

The first order contribution to the distribution function is conveniently put in the form \( f_i^{(1)} = f_i^{(0)} \phi_i^{(1)} \), introducing the new unknown function \( \phi_i^{(1)} \). Using the results of the previous
section to calculate $\mathcal{D}^{(0)} f_i$ and substituting expression for $J^{(0)}\{f_i f_j\}$ we can rewrite equation (4) for the unknown function $\phi_i$ as follows

$$n_i^2 I\{\phi_i^{(1)}\} = -f_i^{(0)} \left[ \gamma_i (c_i c_i - \frac{1}{3} c_i^2 I) : \nabla v_{i0} + \left( \frac{\gamma_i c_i^2}{2} - \frac{5}{2} \right) c_i \cdot \nabla \ln T_i + \sum_j \left( Q^{(e)}(c_i) - \frac{\mu_{ij}}{T_i} \nu_{ij}^{(1;1)} \right) c_i \cdot (v_{i0} - v_{j0}) + \sum_j \left( Q^{(T)}(c_i) + \frac{3}{T_i} \left( \frac{\gamma_i c_i^2}{2} \right) \frac{\mu_{ij}}{m_i + m_j} \nu_{ij}^{(1;1)} \right) (T_i - T_j) \right],$$

where $I\{\phi\} = n^{-2} \int d^3 c d\Omega |c - c_1| f^{(0)} \phi f_i^{(0)} (\phi + \phi_1 - \phi'_1)$ is the linearized collision operator \( \square \). This expression differs from the similar single component equation only by the ‘friction’ terms proportional to $(v_{i0} - v_{j0})$ and $(T_i - T_j)$, therefore the corresponding contributions should be included into the function $\phi_i^{(1)}$. The structure of this expression is similar to that used by Goldman and Sirovich \[21\], however in the later work of de la Mora \[22\] the $Q$-terms arising from collision integrals were omitted. Taking into account rotational invariance of collision operator we can write the function $\phi_i$ in the form

$$\phi_i = -\frac{1}{n_i} A(c_i) c_i \cdot \nabla \ln T_i - \frac{1}{n_i} B(c_i) \left( c_i c_i - \frac{1}{3} c_i^2 I \right) : \nabla v_{i0} - \frac{1}{n_i} \sum_{j \neq i} D_{ij}(c_i) c_i \cdot (v_{i0} - v_{j0}) - \frac{1}{n_i} \sum_{j \neq i} E_{ij}(c_i) (T_i - T_j). \quad (16)$$

The form of the $\phi_i$ function is similar to that used in the work of de la Mora \[22\], but differs from the one in the paper of Struminskii \[24\], where the unknown functions were related not to the driving forces, or independent parameters of solution, but rather chosen in accordance with tensorial order of $c_i$ weights. In our opinion this can not be done if one is looking for solution based on extended set of observables.

In order for $\phi_i^{(1)}$ to satisfy equation (15) the following identities should hold

$$n_i I\{A(c_i) c_i\} = f^{(0)} \left( \frac{\gamma_i c_i^2}{2} - \frac{5}{2} \right) c_i,$$

$$n_i I\{B(c_i)(c_i c_i - \frac{1}{3} c_i^2 I)\} = \gamma_i f^{(0)} (c_i c_i - \frac{1}{3} c_i^2 I),$$

$$n_i I\{D_{ij}(c_i) c_i\} = f^{(0)} \left( Q^{(e)}(c_i) - \frac{\mu_{ij}}{T_i} \nu_{ij}^{(1;1)} \right) c_i,$$

$$n_i I\{E_{ij}(c_i)\} = f^{(0)} \left( Q^{(T)}(c_i) + \frac{3}{T_i} \left( \frac{\gamma_i c_i^2}{2} \right) \frac{\mu_{ij}}{m_i + m_j} \nu_{ij}^{(1;1)} \right). \quad (17)$$

Since the proper values of all observables are obtained from the 0-order distribution function, several conditions should be applied to the function $\phi_i$, such that $f_i^{(1)}$ provides no
contribution to observables. In terms of functions $A(c_i), B(c_i)$ and $E(c_i)$ these conditions read as

\[
\int d^3c \ c^2 f^{(0)} A(c) = 0,
\]
\[
\int d^3c \ c^2 f^{(0)} D_{ij}(c) = 0,
\]
\[
\int d^3c \ c^2 f^{(0)} E_{ij}(c) = \int d^3c f^{(0)} E_{ij}(c) = 0.
\]

which determines the function $\phi_i$ uniquely.

To determine the functions $A(c), B(c), D(c), E(c)$ we use the variational approach \cite{3,30}, i.e. we look for the functions that maximize the corresponding bracket integrals and satisfy \eqref{17,18}. Here we should mention that unlike in the classical treatment it is not immediately obvious that the entropy production would also be maximized by this procedure, and the links between the current development and nonequilibrium thermodynamics require a separate study. Expanding unknown functions into series we can obtain the simple equations to determine the expansion coefficients. The functions $A(c)$ and $B(c)$ here are exactly the same as in classical single-component approach, thus resulting in the ‘partial’ viscosity and thermal conductivity, equal to those the $i$ component would have by its own at the same number density $n_i$ and temperature $T_i$ it has in a mixture. Expressions for these functions are well known, but we provide them here again for completeness. Similarly to $A(c)$ and $B(c)$ the functions $D(c)$ and $E(c)$ are most easily determined if we expand them in series of associated Laguerre polynomials \cite{3,31}

\[
A_i(c_i) = -(\gamma_i/2)^{1/2} \sum_{n=1}^{\infty} a_{ij}^{(n)} L_{n}^{(3/2)} (\gamma_i c_i^2/2),
\]
\[
B_i(c_i) = (\gamma_i/2) \sum_{n=0}^{\infty} b_{ij}^{(n)} L_{n}^{(5/2)} (\gamma_i c_i^2/2),
\]
\[
D_{ij}(c_i) = -(\gamma_i/2) \sum_{n=1}^{\infty} d_{ij}^{(n)} L_{n}^{(3/2)} (\gamma_i c_i^2/2),
\]
\[
E_{ij}(c_i) = \sum_{n=2}^{\infty} e_{ij}^{(n)} L_{n}^{(1/2)} (\gamma_i c_i^2/2).
\]

Some terms in these series are omitted here to satisfy conditions \eqref{18}.

Using the moments of the the functions $Q^{(v)}(c_i)$ and $Q^{(T)}(c_i)$ \eqref{12} we can write down the simple matrix equations to determine the expansion coefficients $e_{ij}^{(m)}$ and $d_{ij}^{(m)}$ (the equations

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for \( a_i \) and \( b_i \) coefficients are again provided here for completeness)

\[
\sum_{m=1}^{\infty} a_i^{(m)} \left[ \left( \frac{\gamma_i}{2} \right)^{1/2} c_i L_m^{(3/2)} \left( \frac{\gamma_i c_i^2}{2} \right), \left( \frac{\gamma_i}{2} \right)^{1/2} c_i L_n^{(3/2)} \left( \frac{\gamma_i c_i^2}{2} \right) \right] = \frac{4}{5} \delta_{n1},
\]

\[
\sum_{m=0}^{\infty} b_i^{(m)} [(\gamma_i/2)(c_i, c_i - \frac{1}{3} c_i^2 I) L_m^{(5/2)}(\gamma_i c_i^2/2), (\gamma_i/2)(c_i, c_i - \frac{1}{3} c_i^2 I) L_n^{(5/2)}(\gamma_i c_i^2/2)] = \frac{2}{T_i} \delta_{n0},
\]

\[
\sum_{m=1}^{\infty} d_{ij}^{(m)} [(\gamma_i/2)^{1/2} c_i L_m^{(3/2)}(\gamma_i c_i^2/2), (\gamma_i/2)^{1/2} c_i L_n^{(3/2)}(\gamma_i c_i^2/2)] = K_{ij}^{(n)},
\]

\[
\sum_{m=2}^{\infty} e_{ij}^{(m)} [L_m^{(1/2)}(\gamma_i c_i^2/2), L_n^{(1/2)}(\gamma_i c_i^2/2)] = \frac{1}{T_i m_j} K_{ij}^{(n-1)}.
\]

(20)

where the bracket integral is defined as \([F, G] = Tr \int d^3 c_i G \otimes I \{F\}\) and the shorthand notation \(K_{ij}^{(n)} = \frac{4\Gamma(n+5/2)}{\sqrt{\pi}} \sum_{l=0}^{n} \frac{(-)^{n-l}}{l!(n-l+1)!} \left(1 - \frac{\mu_i}{\mu_j}\right)^{n-l} \left(\frac{\mu_i}{\mu_j}\right)^l\) is introduced.

Provided equations (20) are solved and thus the functions \(A(c), B(c), D(c)\) and \(E(c)\) are known, we can calculate the first order contributions to equations (17). For the pressure and the heat flow those read as

\[
P_i^{(1)} = -b_i^{(0)} T_i S_i,
\]

\[
q_i^{(1)} = -\frac{5}{4} a_i^{(1)} \nabla T_i - \frac{5T_i}{4} \sum_j d_{ij}^{(1)} (v_{i0} - v_{j0}).
\]

(21)

Here, the partial viscosity and heat conductivity are given by the first expansion coefficients \(b_i^{(0)} T_i/2\) and \(5a_i^{(1)}/4\) correspondingly. There is also a term describing the heat flow due to interaction of the flows moving with different velocities, however temperature differences do not contribute to the pressure. Instead there appears a term in the first order momentum source integral having the same effect. The partial viscosity and heat conductivity here are determined by only the first nonvanishing coefficients from the expansion (19). On contrary the contributions from the friction terms, are given in terms of series including the whole list of expansion coefficients. Up to the terms bilinear in gradients of observables and their differences the source terms in the first order calculate

\[
m_i \int d^3 c_i J_i^{(1)} \{f_i, f_j\} = -\mu_{ij}^{(VT)} \nabla T_i - \mu_{ij}^{(VT)} \nabla T_j - (\mu_{ij}^{(TVT)} \nabla T_i + \mu_{ij}^{(TVT)} \nabla T_j)(T_i - T_j)
\]

\[
- (\mu_{ij}^{(SV)} S_i + \mu_{ij}^{(SV)} S_j) \cdot (v_{i0} - v_{j0}) - \sum_k (\mu_{ijk}^{(v)} (v_{i0} - v_{k0}) + \mu_{ijk}^{(v)} (v_{j0} - v_{k0})),
\]

(22)
and
\[
\frac{m_i}{2} \int d^3 c_i c_i^2 J^{(1)} \{ f_i f_j \} = -(\lambda^{(v\nabla T)}_{ij} \nabla T_i + \lambda^{(v\nabla T)}_{ji} \nabla T_j) \cdot (\mathbf{v}_i - \mathbf{v}_j) \\
- \sum_k (\lambda^{(T)}_{ijk} (T_i - T_k) + \lambda^{(T)}_{jik} (T_j - T_k)).
\] (23)

The transport coefficients \( \mu \) and \( \lambda \) include the whole expansions \{a^{(n)}\}, \{b^{(n)}\}, \{d^{(n)}\} and \{e^{(n)}\}, and are given in appendix A. The bilinear terms here are not really important and most probably can be omitted for most of the practical applications. The most interesting contribution is probably the one describing the thermal diffusion which explicitly appears as a result of interactions between different components in the current treatment. If the components share the same value of \( m_i \) and \( T_i \) (that is for example if the single-component flow is virtually divided in two flows of similar species), this contribution disappears in total agreement with the classical solution. Summing the equations to obtain the momentum balance for the mixture as a whole also eliminates this term.

The other interesting contributions are those with transport coefficients with 3 indexes. Those not only include the first order corrections to the Maxwell-Stefan diffusion terms, but also describe indirect interactions of two components through the impact on a third one. Appearance of such contributions derived from the Boltzmann equation, allowing only the pairwise interactions is fascinating but not totally unexpected. Use of the reduced description with some degrees of freedom integrated out often leads to many-body effective interactions. The most famous example of that is probably the interaction of the ions of the same sort in electrolytes.

Thus, in the first order approximation we obtain the Navier-Stokes equations which include the corrected Maxwell-Stefan terms, thermal diffusion and also several new terms bilinear in gradients and differences of observables. The partial viscosity and heat conductivity depend only on the properties of the component \( i \), thus totally neglecting \( ij \) correlations. Also the time evolution of \( v_{i0} \) depends on the strain tensor of the component \( i \) and does not include the strain of the other components, which is not physical. This situation can be understood if we notice that the formal density scaling we are using, leads to mixing the terms of different orders of the scaling parameter \( \epsilon \) as compared to the standard treatment, and tends to give more weight to \( ii \) interactions. Thus, although, the second order distribution function does not contribute to viscous forces in the classical approach, here we need
to proceed to higher orders to include the $ij$ viscous interactions.

As many contributions from the first order treatment can already be considered excessive, we are not going to do the complete analysis of the second order solution. Instead we are only interested in correcting the viscosity and heat conductivity including the terms resulting from $ij$ correlations. As previously, introducing the auxiliary function $\phi_i^{(2)}$ such that $f_i^{(2)} = f_i^{(2)} \phi_i^{(2)}$ we rewrite equation (4) in the form

$$- n_i^2 I \{ \phi_i^{(2)} \} = \frac{\partial_t f_i^{(0)}}{\partial t} + \frac{\partial_b f_i^{(1)}}{\partial t} + (c \cdot \nabla r + F \cdot \nabla v_i) f_i^{(1)} - \sum_{j \neq i} J^{(1)} \{ f_i f_j \} - J \{ f_i^{(1)} f_j^{(1)} \}. \quad (24)$$

Here $\sum_{j \neq i} J^{(1)} \{ f_i f_j \}$ is the only term containing the required contributions, the most general form of which is given by

$$J^{(1)} \{ f_i f_j \} \propto -f_i^{(0)} \left[ Q_{ij}^{(S_i)} (c_i) (c_i c_i - \frac{1}{3} c_i^2 I) : S_i + Q_{ij}^{(\nabla T_i)} (c_i) c_i \cdot \nabla T_i \right] \right]. \quad (25)$$

We use the sign $\propto$ to indicate that all irrelevant contributions are omitted here. The functions $Q_{ij}^{(l)} (c_i)$ can be left undefined for the time being, as for the later analysis we will only need their moments with respect to even powers of $c_i$ which are easier to calculate then the functions themselves.

Similarly to the previous section, the viscous contribution to the second order pressure tensor is found

$$P_i^{(2)} = -T_i \sum_{j \neq i} b_{ij}^{(2;0)} S_i - T_i \sum_{j \neq i} b_{ij}^{(2;0)} S_j, \quad (26)$$

and nondiagonal part of the heat conductivity reads as

$$q_i^{(2)} = -\frac{5}{4} \sum_{j \neq i} a_{ij}^{(2;1)} \nabla T_j - \frac{5}{4} \sum_{j \neq i} a_{ij}^{(2;1)} \nabla T_j. \quad (27)$$

The expansion coefficients again can be found from the simple matrix equations

$$\sum_{m=0}^{\infty} b_{ij}^{(2;m)} [(c_i c_i - \frac{1}{3} c_i^2 I) L_m^{(5/2)} (\gamma_i c_i^2 / 2), (c_i c_i - \frac{1}{3} c_i^2 I) L_m^{(5/2)} (\gamma_i c_i^2 / 2)] = K_{ij}^{(2;0;m)},$$

$$\sum_{m=1}^{\infty} a_{ij}^{(2;m)} [(\gamma_i / 2)^{1/2} c_i L_m^{(3/2)} (\gamma_i c_i^2 / 2), (\gamma_i / 2)^{1/2} c_i L_n^{(3/2)} (\gamma_i c_i^2 / 2)] = K_{ij}^{(2;a;m)} \quad (28)$$

Expressions for the coefficients $K_{ij}^{(2;m)}$ are given in Appendix B. When the mixture is treated as a whole and thus all components have approximately the same rate of shear and temperature gradient, the viscosity and heat conductivity for the mixture can be found simply as a sum of all the components of corresponding matrices.
V. DISCUSSION

We investigate the perturbative solution to the Boltzmann multicomponent kinetic equation based on the set of observables including each component velocity and the temperature. The corresponding set of the species balance equations is derived. The new solution is obtained through modification of the formal density scaling scheme of Enskog, such that the density of each component is scaled independently. This approach allows for successive development, with clear assumptions and approximations, which should be contrasted to rather ad-hoc previous attacks on the problem. Omitting or addition of the terms in some intermediate equations as compared to the similar works, as discussed in the text, follows directly from the scaling equation \(2\).

We show that the distribution functions related to each component velocity appear naturally using this approach, and the solubility conditions immediately provide us with the balance equations for species. The zero order equations are the Euler equations for each component including the heat and momentum transport between components through the familiar Maxwell-Stefan diffusion term. As a next approximation we obtain the Navier-Stokes equations with partial viscosities equal to those the component would have in absence of the other. Apart from that there are viscous contributions from the shear of other components. The interaction between components leads to appearance of the heat diffusion and also some corrections to the Maxwell-Stefan diffusion term. These corrections include apart from direct interactions between the two components indirect ones, through perturbing the other components flows. The heat balance equations possess the similar properties.
To summarize we provide here the final equations describing the multicomponent flow

$$\frac{1}{\rho_i} \frac{d\rho_i}{dt} = - \nabla \cdot \mathbf{v}_{i0},$$
$$\rho_i \frac{d\mathbf{v}_{i0}}{dt} = \rho_i \mathbf{F}_i - \nabla p_i + 2\eta_i \nabla \mathbf{S}_i + 2 \sum_{j \neq i} (\eta_{ij} \nabla \mathbf{S}_i + \eta_{ji} \nabla \mathbf{S}_j) - \sum_{j \neq i} (\mu_{ij}^{(T)} \nabla T_i + \mu_{ji}^{(T)} \nabla T_j)$$

$$- n_i \sum_j \mu_{ij}^{(1;1)} (\mathbf{v}_{i0} - \mathbf{v}_{j0}) - \sum_k (\mu_{ij}^{(v)} (\mathbf{v}_{i0} - \mathbf{v}_{k0}) + \mu_{ji}^{(v)} (\mathbf{v}_{j0} - \mathbf{v}_{k0}));$$

$$\rho_i \frac{d\mathbf{u}_i}{dt} = - p_i \nabla \cdot \mathbf{v}_{i0} + \nabla \lambda_i \nabla T_i + \sum_{j \neq i} (\nabla \lambda_{ij} \nabla T_i + \nabla \lambda_{ji} \nabla T_i)$$
$$+ 2\eta_i \mathbf{S}_i : \nabla \mathbf{v}_{i0} + 2 \sum_{j \neq i} (\eta_{ij} \mathbf{S}_i + \eta_{ji} \mathbf{S}_j) : \nabla \mathbf{v}_{i0}$$

$$- 3n_i \sum_j \frac{\mu_{ij}}{m_i + m_j} \nu_{ij}^{(1;1)} (T_i - T_j) - \sum_k (\lambda_{ijk}^{(T)} (T_i - T_k) + \lambda_{jik}^{(T)} (T_j - T_k)),$$ (29)

where the transport coefficients are defined in the text through the properties of the components.

The convergence of the scheme is not questioned in the paper, however one may argue that apart from using an extended set of observables the whole procedure can be seen as mixing the terms of different orders (compared to the classical treatment), and thus convergence of both solutions should be about the same, suggesting the range of Knudsen numbers where it is applicable. Another issue is the convergence of the transport coefficients which depend on the whole first order distribution function, and not just include the first expansion coefficient. As we see, convergence of these series depend strongly on the value of $0 < \gamma_{ij}/\gamma_j < 1$. In cases when the temperatures of the different components are about the same, and the impact of the temperature gradient is more significant, which is true for many chemical engineering applications, the above ratio can be replaced by $\mu_{ij}/m_j$, thus removing the complicated temperature dependency from the transport coefficients. In the case when the gradients are small, but the temperatures themselves are very different, as it may happen for example in plasma, the $\gamma_{ij}/\gamma_j$ ratio should be kept, but it varies slowly again. The only potential problems may appear when both gradients and temperature differences are big enough, leading to complicated temperature dependency of the transport coefficients, however this case is clearly out of scope of the present theory. The present study can also be seen as a basis for the multifluid model used to describe kinetic processes in plasma. The other potential applications of the method presented here include reactive flows and the boundary
layer problem for mixtures of rarified gases, that still generates a lot of discussion in the literature (see for example [32, 33]). Some authors also tend to employ the multicomponent transport equations to describe the shock-waves or even turbulence phenomena [34].

Apart from modified scaling, the presented solution follows closely the original Enskog scheme, with which it has more in common than any other work on multicomponent transport cited here. However some features appear to be very different. The question of entropy production which is not obviously maximized by the present solution requires separate study to establish the links with the nonequilibrium thermodynamics. The fact that there are no \(ij\) collisions in the left part of the equation (4) eliminates the need of \(ij\) bracket integrals. In some sense they are replaced by the set of collisional frequencies \(\nu^{(m:n)}_{ij}\), closely related to the Chapman-Enskog \(\Omega\)-integrals. The \(\nu^{(1;1)}_{ij}\) is recognized as an effective collisional frequency of electrons from the plasma theory [19]. Unlike in the classical solution the first order solution does not provide us with all the relevant contributions, and the contributions of the same nature, such as viscous, are divided between the first and the second order contributions. This is the consequence of the suggested scaling, which gives more weight to collisions of the species of the same type. On the other hand the \(ii\) and \(ij\) contributions to the viscosity and heat conductivity are explicitly separated, which makes it convenient to compare the present results with the Green-Kubo theory for the transport coefficients. It should also be noted that for the special case of binary mixtures the form of the transport equations derived here coincide with those obtained by de la Mora [22]. The structure of the equations for the transport coefficients however is somewhat different, although we do not expect large discrepancies in the numerical values. The detailed numerical study revealing importance of different contributions, and containing comparison with counterflow and other experiments (simulations) is the topic of the future research.

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APPENDIX A

Below we provide the expressions for the transport coefficients entering the first order equations of motion. Lower signs correspond to switching i and j indexes, and $F(a; b; c; x)$ is the hypergeometric function.

$$\mu_{ij}^{(VT)} = \mp \frac{4}{3\pi^{1/2}} \left(\frac{\gamma_i}{2}\right)^{1/2} \sum_n a_{ij}^{(n)} \Gamma(n + 5/2) \sum_{s=0}^{n} (-)^s \left(\frac{\gamma_{ij}}{\gamma_j}\right)^s \cdot \left(\frac{F(s+3/2, s-n; s+5/2; \gamma_{ij}/\gamma_j)}{\Gamma(n-s+1)} \Gamma(n-s) (s+5/2) \right) \nu_{ij}^{(1; s+1)}$$

$$\mu_{ij}^{(TVT)} = \frac{2}{3\pi^{1/2}} \left(\frac{\gamma_i}{2}\right)^{1/2} \frac{\gamma_i}{T_j} \sum_n a_{ij}^{(n)} \Gamma(n + 5/2) \sum_{s=0}^{n} (-)^s \left(\frac{\gamma_{ij}}{\gamma_j}\right)^s \cdot \left(- \frac{F(s+3/2, s-n; s+5/2; \gamma_{ij}/\gamma_j)}{\Gamma(n-s+1)} \Gamma(n-s) (s+5/2) \right) \nu_{ij}^{(1; s+1)}$$

$$\mu_{ij}^{(SV)} = \frac{8\mu_{ij}}{15\pi^{1/2}} \sum_n b_{ij}^{(n)} \Gamma(n + 7/2) \sum_{s=0}^{n} (-)^s \left(\frac{\gamma_{ij}}{\gamma_j}\right)^s \cdot \left(- \frac{m_{ij}}{m_j} \sum_{s=0}^{n} \frac{F(s+5/2, s-n+1; s+9/2; \gamma_{ij}/\gamma_j)}{\Gamma(n-s+1)} \Gamma(n-s) (s+5/2) \right) \nu_{ij}^{(1; s+1)}$$

$$\mu_{ijk}^{(V)} = \mp \frac{4}{3\pi^{1/2}} \gamma_i \left(\frac{\gamma_i}{2}\right)^{1/2} \sum_n a_{ijk}^{(n)} \Gamma(n + 5/2) \sum_{s=0}^{n} (-)^s \left(\frac{\gamma_{ij}}{\gamma_j}\right)^s \cdot \left(\frac{F(s+3/2, s-n; s+5/2; \gamma_{ij}/\gamma_j)}{\Gamma(n-s+1)} \Gamma(n-s) (s+5/2) \right) \nu_{ij}^{(1; s+1)}$$

$$\lambda_{ij}^{(VT)} = \frac{4}{3\pi^{1/2}} \left(\frac{\gamma_i}{2}\right)^{1/2} \sum_n a_{ij}^{(n)} \Gamma(n + 5/2) \sum_{s=0}^{n} (-)^s \left(\frac{\gamma_{ij}}{\gamma_j}\right)^s \cdot \left(- \frac{\mu_{ij} - \gamma_{ij}}{\gamma_j} \frac{F(s+3/2, s-n; s+5/2; \gamma_{ij}/\gamma_j)}{\Gamma(n-s+1)} \Gamma(n-s) (s+5/2) \right) \nu_{ij}^{(1; s+1)}$$

$$\lambda_{ij}^{(T)} = \mp \frac{4}{\pi^{1/2}} \sum_{n} c_{ij}^{(n)} \sum_{s=0}^{n-1} (-)^s \gamma_j \sum_{s=0}^{n} \sum_{s=0}^{n} \left(1 - \frac{\gamma_{ij}}{\gamma_j}\right)^{n-s-1} \left(\frac{\gamma_{ij}}{\gamma_j}\right)^{s+1} \nu_{ij}^{(1; s+1)}.$$
APPENDIX B

Here we provide the expressions for coefficients appearing in the right hand side of equations (28) which determine the nondiagonal (in component indexes) viscosity and heat conductivity. Here again the \(ij\) coefficients are obtained by simultaneous replacing all \(i\) by \(j\) and contrary, no sign changes required here. These expressions are rather awkward, and in principle several sums should be possible to calculate analytically, however we failed to further simplify these expressions, which anyway would not make significant difference for the numerical calculations of the coefficients.

\[
K_{ij}^{(2b;n)} = - \frac{1}{n_i} \sum_{l=0}^{m} b_l^{(m)} \sum_{p=0}^{n} \frac{(-)^{l+p}}{l!p!} \left( \frac{m + 5/2}{m - l} \right) \left( \frac{n + 5/2}{n - p} \right) \left( \frac{\gamma_{ij}}{\gamma_j} \right) \sum_{k=0}^{l} \sum_{q=0}^{p} \left( \frac{l}{k} \right) \left( \frac{p}{q} \right) \times \sum_{r=0}^{l+p-k-q} \left( \frac{l + p - k - q}{r} \right)
\]

\[
K_{ij}^{(2a;n)} = \frac{1}{n_i} \sum_{l=0}^{m} a_l^{(m)} \sum_{p=0}^{n} \frac{(-)^{l+p}}{l!p!} \left( \frac{m + 3/2}{m - l} \right) \left( \frac{n + 3/2}{n - p} \right) \left( \frac{\gamma_{ij}}{\gamma_j} \right) \sum_{k=0}^{l} \sum_{q=0}^{p} \left( \frac{l}{k} \right) \left( \frac{p}{q} \right) \times \sum_{r=0}^{p+l-k-q} \left( \frac{p + l - k - q}{r} \right)
\]

Here we have introduced an auxiliary function \(W_{l,m,n}^{q} = \frac{2^{-2n} \Gamma(l-m+2n+q) \Gamma(m+2n+4)}{(m+r+1)}\), and it is also implied that only contributions with integer upper indexes of \(\nu_{ij}^{(lm)}\) enter the summation.

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