Shear-rate dependent transport coefficients in a binary mixture of Maxwell molecules

C. Marín
Departamento de Matemáticas, Universidad de Extremadura, E-06071 Badajoz, Spain

V. Garzó[a)]
Department of Physics, University of Florida, Gainesville, Florida 32611

M. López de Haro[b)]
Centro de Investigación en Energía, Universidad Nacional Autónoma de México, Apdo. Postal 34, Temixco 62580, México

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Mass and heat transport in a dilute binary mixture of Maxwell molecules under steady shear flow are studied in the limit of small concentration gradients. The analysis is made from the Gross–Krook kinetic model of the Boltzmann equation. This model is solved by means of a perturbation solution around the steady shear flow solution [Phys. Fluids **8**, 2756 (1996)], which applies for arbitrary values of the shear rate. In the first order of the expansion the results show that the mass and heat fluxes are proportional to the concentration gradient but, due to the anisotropy of the problem, mutual diffusion and Dufour tensors can be identified, respectively. Both tensors are explicitly determined in terms of the shear rate and the parameters of the mixture (particle masses, concentrations, and force constants). A comparison with the results derived from the exact Boltzmann equation at the level of the diffusion tensor shows a good agreement for a wide range of values of the shear rate. © *2000 American Institute of Physics.* [S1070-6631(00)02102-4]

I. INTRODUCTION

When a fluid mixture is simultaneously subjected to both weak velocity and concentration gradients, the Curie principle[1] states that the shear field (tensorial quantity) cannot modify vectorial quantities such as mass and heat fluxes, which are generated by a concentration gradient ∇n_s. As a consequence, the mutual diffusion coefficient (which couples the mass current and the concentration gradient) and the Dufour coefficient (which couples the heat flux and the concentration gradient) do not depend on the shear field. Nevertheless, when the shear rate is large, nonlinear effects become important so that the Curie principle does not apply and the above-mentioned transport coefficients may be modified by the shear flow. If the concentration gradient is weak, one expects that the mass and heat fluxes are still proportional to ∇n_s although the corresponding transport coefficients must be replaced by second-rank tensors with elements that are nonlinear functions of the shear rate.

In order to gain some insight into this complex nonlinear problem, it is useful to consider a somewhat simpler situation for which greater progress can be made. Here, we consider a binary mixture in the low-density regime for which nonequilibrium phenomena are well described by the nonlinear Boltzmann equation.[2] For states near equilibrium, the Boltzmann equation may be solved for general potentials by means of the Chapman–Enskog expansion[3] of the distribution function around the local equilibrium distribution. In the first order of the concentration gradient (Navier–Stokes domain), one gets explicit expression for the mutual diffusion and Dufour coefficients. Although this method can be in principle applied to states far from equilibrium as well, the evaluation of the Chapman–Enskog expansion to higher orders is extremely difficult. In addition, questions about its convergence remain still unknown. This gives rise to look for alternative approaches. One possibility is to expand in small concentration gradients around a more relevant reference state than local equilibrium. Since we are interested in computing the influence of shear flow on the diffusion and Dufour coefficients, we choose the uniform shear flow (USF) state as the reference state. The USF state is probably the simplest fluid flow problem since the only nonzero hydrodynamic gradient is $\partial u_x/\partial y = a = \text{const}$, where $\mathbf{u}$ is the flow velocity and a is the constant shear rate. The relevant transport coefficients of USF are the nonlinear shear viscosity coefficient and viscometric functions which are related to the pressure tensor (second-degree moment). The USF state is one of the rare exceptions for which the Boltzmann equation admits an exact solution.[4] In the special case of Maxwell molecules (repulsive potential of the form r^{-4}), Ikenberry and Truesdell[5] obtained explicit expressions of the pressure tensor for *arbitrary* values of the shear rate. Recently, this solution has been extended to the case of a multicomponent system with arbitrary values of masses, concentrations, and force constants.[6]

[a)]Author to whom all correspondence should be addressed at Departmento de Física, Universidad de Extremadura, E-06071 Badajoz, Spain; electronic mail: vicenteg@unex.es
[b)]Also consultant at Programa de Simulación Molecular of the Instituto Mexicano del Petróleo.

As previously mentioned, in this paper we are interested in analyzing heat flow and mass diffusion in a strongly shearing binary mixture. The physical situation is such that an arbitrary shear rate coexists with a weak concentration gradient. Nevertheless, in practice this program cannot be carried out analytically by using the Boltzmann equation. The main reason is that we need the fourth-degree moments of USF (whose explicit expressions are not known in the Boltzmann equation) to evaluate the shear-rate dependent Dufour tensor. In order to overcome such difficulty, here we use a convenient kinetic model that preserves the essential features of the Boltzmann equation but admits more practical analysis. Specifically, we consider the well-known Gross–Krook (GK) model[7] for a binary mixture, for which an exact solution of the USF state has also been recently found.[8] The comparison of these results with those from the Boltzmann equation[6] at the level of the rheological properties shows good agreement, in general. This fact indicates again the reliability of this kinetic model in computing transport properties in a binary mixture. Very recently, in the context of the BGK equation for a single gas, a similar study of spatially inhomogeneous states near USF has been made.[9]

The plan of the paper is as follows. In Sec. II, we introduce the model and define the transport coefficients to be evaluated. In Sec. III, we give a brief summary of relevant results concerning the USF problem in the framework of the GK model. Since we are interested in making connection with computer simulations, we also introduce thermostat forces to compensate for the viscous heating and achieve a steady state. Section IV contains the main results of the paper. We describe the perturbation scheme around the USF distribution and explicitly compute the mass and heat fluxes to linear order in the concentration gradient. The associated transport coefficients are identified and given as nonlinear functions of the shear rate and the parameters of the mixture (mass ratio, concentration ratio, and force constants ratios). The shear-rate dependence of these coefficients is illustrated and compared with some previous results obtained from the Boltzmann equation for the diffusion tensor.[10] Such a comparison shows that the GK predictions are in a reasonably good agreement with the Boltzmann ones. We close the paper in Sec. V with a discussion of the results presented.

II. KINETIC MODEL AND TRANSPORT COEFFICIENTS

Let us consider a binary mixture in the low-density regime. In this limit, the time evolution of the system is described by the set of nonlinear Boltzmann equations.[2] Nevertheless, exact or even approximate solutions to the Boltzmann equation far from equilibrium are scarce, due basically to the intricacy of its collision operator.[4] Therefore, for practical purposes, it is convenient to introduce kinetic models that replace the Boltzmann collision operator with a more tractable operator that retains the essential features of the exact one, such as the conservation laws. In the case of a binary mixture, a popular model is the one proposed by Gross and Krook (GK).[7] The GK model is given by the set of two equations:

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\cdot\frac{\partial}{\partial \mathbf{r}}+\frac{1}{m_1}\frac{\partial}{\partial \mathbf{v}}\cdot\mathbf{F}_1\right)f_1=-\nu_{11}(f_1-f_{11})-\nu_{12}(f_1-f_{12}), \tag{1}$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\cdot\frac{\partial}{\partial \mathbf{r}}+\frac{1}{m_2}\frac{\partial}{\partial \mathbf{v}}\cdot\mathbf{F}_2\right)f_2=-\nu_{22}(f_2-f_{22})-\nu_{21}(f_2-f_{21}). \tag{2}$$

Here, $f_s(\mathbf{r},\mathbf{v};t)$ is the one-particle velocity distribution function of species s, m_s is the mass of a particle of species s, and $\mathbf{F}_s$ is an external force acting on particles of species s. Also, the model introduces the distributions f_{rs} as

$$f_{rs}=n_r\left(\frac{m_r}{2\pi k_{\mathrm{B}}T_{rs}}\right)^{3/2}\exp\left[-\frac{m_r}{2k_{\mathrm{B}}T_{rs}}(\mathbf{v}-\mathbf{u}_{rs})^2\right], \tag{3}$$

where

$$\mathbf{u}_{rs}=\frac{m_r\mathbf{u}_r+m_s\mathbf{u}_s}{m_r+m_s}, \tag{4}$$

$$T_{rs}=T_r+2\frac{m_rm_s}{(m_r+m_s)^2}\left[(T_s-T_r)+\frac{m_s}{6k_{\mathrm{B}}}(\mathbf{u}_r-\mathbf{u}_s)^2\right]. \tag{5}$$

In Eqs. (4) and (5) the number density n_s, the mean velocity $\mathbf{u}_s$, and temperature T_s of species s, are defined, respectively, as

$$\{n_s,n_s\mathbf{u}_s,n_sk_{\mathrm{B}}T_s\}=\int d\mathbf{v}\left(1,\mathbf{v},\frac{1}{3}m_s(\mathbf{v}-\mathbf{u}_s)^2\right)f_s. \tag{6}$$

From the partial quantities appearing in (6), one can define the total number density $n=n_1+n_2$, the flow velocity $\mathbf{u}=(\rho_1\mathbf{u}_1+\rho_2\mathbf{u}_2)/(\rho_1+\rho_2)$, $\rho_s=m_sn_s$ being the mass density of species s, and the temperature of the mixture T (which is the relevant one at a hydrodynamic level) as

$$nk_{\mathrm{B}}T=\sum_{s=1}^{2}\left(n_sk_{\mathrm{B}}T_s+\frac{1}{3}\rho_s(\mathbf{u}_s-\mathbf{u})^2\right). \tag{7}$$

The quantities $\mathbf{u}_{rs}$, and T_{rs} are determined by imposing that the total momentum and energy are conserved and that the first five collisional moments as computed with the GK collision term be the same as those computed with the exact Boltzmann collision operator for Maxwell molecules, namely, a repulsive potential of the form $\kappa_{rs}r^{-4}$. In this case, the effective collision frequency ν_{rs} can be identified as

$$\nu_{rs}=An_s\left(\kappa_{rs}\frac{m_r+m_s}{m_rm_s}\right)^{1/2}, \tag{8}$$

where A is a constant to be fixed by requiring that the model reproduces some transport coefficient of the Boltzmann equation. Although the GK model can be extended to more general repulsive interactions,[11] for the sake of concreteness, we will restrict ourselves to Maxwell molecules.

The corresponding balance equations associated with n_s, $\mathbf{u}$, and T define the dissipative fluxes of mass

$$\mathbf{J}_s=\int d\mathbf{v}\,m_s(\mathbf{v}-\mathbf{u})f_s=\rho_s(\mathbf{u}_s-\mathbf{u}), \tag{9}$$

momentum (pressure tensor)

$$\mathsf{P}=\sum_{s=1}^{2}\int d\mathbf{v}\, m_s(\mathbf{v}-\mathbf{u})(\mathbf{v}-\mathbf{u})f_s=\sum_{s=1}^{2}\mathsf{P}_s, \tag{10}$$

and energy (heat flux)

$$\mathbf{q}=\sum_{s=1}^{2}\int d\mathbf{v}\frac{m_s}{2}(\mathbf{v}-\mathbf{u})^2(\mathbf{v}-\mathbf{u})f_s. \tag{11}$$

These fluxes define the relevant transport coefficients of the mixture. In particular, in a mixture the presence of a concentration gradient not only induces a mass current but also a heat flux. According to linear irreversible thermodynamics, the macroscopic linear relations governing such fluxes in a binary mixture can be written as[1]

$$\mathbf{J}_s=-\frac{m_1m_2n}{\rho}D_0\nabla n_s, \tag{12}$$

$$\mathbf{J}_q\equiv\mathbf{q}-\frac{5}{2}k_BT\sum_{s=1}^{2}\frac{\mathbf{J}_s}{m_s}=-\frac{k_B\rho}{\rho_1\rho_2}L_0\nabla n_1. \tag{13}$$

In writting these equations, for simplicity, we have assumed that n and T are constants and have chosen $-\nabla[(\mu_1-\mu_2)]/T\equiv-(k_B\rho/\rho_1\rho_2)\nabla n_1$ as the force conjugate to $\mathbf{J}_s$ and $\mathbf{J}_q$, μ_s being the chemical potential of species s. In Eqs. (12) and (13), we have introduced the mutual diffusion coefficient D_0 and the Dufour coefficient L_0. Microscopic expressions for these coefficients can be obtained from the GK model by using, for instance, the Chapman–Enskog expansion.[3] In this case, it is easy to get the following results:

$$D_0=\frac{k_BT}{n\tau}, \tag{14}$$

$$L_0=0, \tag{15}$$

where

$$\tau=\frac{m_1m_2}{m_1+m_2}\frac{\nu_{12}}{n_2}. \tag{16}$$

The fact that the Dufour coefficient vanishes is due to the interaction potential considered (Maxwell molecules), since this result is identical to the one obtained from the Boltzmann equation for Maxwell molecules. For more general interaction potentials, the Boltzmann equation leads to additional crossed contributions to the heat flux.[3] With respect to the diffusion coefficient, it has the same structure as the one obtained from the Boltzmann equation. As a matter of fact, Eq. (14) coincides with the Boltzmann result if one conveniently adjusts the constant A appearing in the definition of ν_{12}, cf. Eq. (8). In this case, if $A=1.69\pi$, the mutual diffusion coefficient given by the GK model is the same as the one given from the Boltzmann equation.[3] Henceforth, we will take this choice for the constant A.

III. UNIFORM SHEAR FLOW IN A BINARY MIXTURE

The fact that the distributions f_{rs} depend on space and time through their dependence on the densities, velocities, and temperatures (which are functionals of the actual distributions f_s) makes the GK equation more nonlinear than the actual Boltzmann equation. Nevertheless, in some cases the GK model can be explicitly solved. One of them is the so-called uniform shear flow (USF).[4,12] The USF state is a planar flow where the only nonzero hydrodynamic gradient corresponds to the x component of the flow velocities along the y direction:

$$u_{s,i}=u_i=a_{ij}r_j, \quad a_{ij}=a\,\delta_{ix}\delta_{jy}, \tag{17}$$

a being the constant shear rate. This parameter (which may be arbitrarily large) measures the distance of the mixture from equilibrium. This state is generated by a periodic boundary condition in the Lagrangian frame.[13] These boundary conditions induce viscous heating so that the temperature increases in time. In order to prevent this effect, it is usual in computer experiments to introduce external forces which remove the heat at the same rate as it is produced. The simplest choice (which is based on Gauss' principle of least constraint) is a nonconservative force proportional to the peculiar velocity $V_i=v_i-a_{ij}r_j$, i.e.,[14]

$$\mathbf{F}=-m_s\alpha\mathbf{V}, \tag{18}$$

where the thermostat parameter α is adjusted to maintain a constant temperature. In the case of Maxwell molecules, it is important to remark that in the USF problem there is an exact equivalence between the results obtained with and without a thermostat force.[15,6,8] In addition, the uniform shear flow becomes spatially homogeneous in the frame moving with the flow velocity $\mathbf{u}$. Thus, in this frame the velocity distribution function adopts the form $f_s(\mathbf{r},\mathbf{v})=f_s(\mathbf{V})$.

Under the above-mentioned conditions, the set of GK equations (1) and (2) becomes

$$-\frac{\partial}{\partial V_i}(a_{ij}V_j+\alpha V_i)f_1=-\nu_{11}(f_1-f_{11})-\nu_{12}(f_1-f_{12}) \tag{19}$$

and a similar equation holds for f_2. To get the rheological properties of the mixture it is convenient to define the reduced velocity moments corresponding to each species. They are defined as

$$M_{k,l,m}=\frac{1}{n_1}\left(\frac{2k_BT}{m_1}\right)^{-(1/2)(k+l+m)}\int d\mathbf{V}\,V_x^kV_y^lV_z^mf_1(\mathbf{V}), \tag{20}$$

$$N_{k,l,m}=\frac{1}{n_2}\left(\frac{2k_BT}{m_2}\right)^{-(1/2)(k+l+m)}\int d\mathbf{V}\,V_x^kV_y^lV_z^mf_2(\mathbf{V}). \tag{21}$$

Recently, explicit expressions of these moments have been obtained.[8] They are given by

$$M_{k,l,m}=\sum_{q=0}^{k}\frac{k!}{(k-q)!}(-a)^q[\nu_1+(k+l+m)\alpha]^{-(1+q)}\times A_{k-q,l+q,m}[\nu_{11}\chi_1^{(k+l+m)/2}+\nu_{12}\chi_{12}^{(k+l+m)/2}], \tag{22}$$

where

$$A_{k,l,m}=\pi^{-3/2}\Gamma\left(\frac{k+1}{2}\right)\Gamma\left(\frac{l+1}{2}\right)\Gamma\left(\frac{m+1}{2}\right) \tag{23}$$

if (k,l,m) are even, being zero otherwise. Further, we have introduced the quantities

$$\chi_1\equiv\frac{T_1}{T}=\left[1+\frac{3\alpha(\nu_1+2\alpha)^2-a^2\nu_1}{M\nu_{12}(1+\delta)[3(\nu_1+2\alpha)^2+2a^2]}\right]^{-1}, \tag{24}$$

$\nu_1=\nu_{11}+\nu_{12}$, $M\equiv\mu/(1+\mu)^2$, and $\chi_{12}\equiv T_{12}/T=\chi_1+2M(1+\delta)(1-\chi_1)$. In these expressions, $\mu\equiv m_1/m_2$ is the mass ratio, and $\delta\equiv n_1/n_2$ is the concentration ratio. The moments $N_{k,l,m}$ can be easily obtained from Eq. (22) by the adequate changes: $\mu\leftrightarrow\mu^{-1}$, $\delta\leftrightarrow\delta^{-1}$, $\kappa_{11}\leftrightarrow\kappa_{22}$. Finally, the thermostat parameter α can be obtained from the consistency condition: $-(1-\chi_1)\delta+\chi_2=1$. This leads to a sixth-degree algebraic equation, whose largest (real) root gives α as a function of the shear rate and the parameters of the mixture. However, from a practical point of view, it is convenient to take α as independent variable (instead of a) and express the shear rate in terms of α. The result is

$$a^2=-\tfrac{1}{2}(P+\sqrt{P^2-4Q}), \tag{25}$$

where

$$\begin{aligned}P=&\frac{3}{\nu_1\nu_2(1+\delta)-2M\zeta(\delta\nu_1+\nu_2)}\{4\alpha^3(1+\delta)(2M\zeta\\&-\nu_1-\nu_2)+4\alpha^2[M\zeta(\delta\nu_1+\nu_2)-2\nu_1\nu_2(1+\delta)]\\&+\alpha[2M\zeta(\delta\nu_1^2+\nu_2^2)-4M\zeta\nu_1\nu_2(1+\delta)\\&-\zeta\nu_1\nu_2(\nu_1+\nu_2)(1+\delta)]-M\zeta\nu_1\nu_2(\nu_1+\delta\nu_2)\},\end{aligned} \tag{26}$$

$$Q=\frac{9\alpha(1+\delta)(\alpha+\zeta M)(2\alpha+\nu_1)^2(2\alpha+\nu_2)^2}{\nu_1\nu_2(1+\delta)-2M\zeta(\delta\nu_1+\nu_2)}, \tag{27}$$

with $\zeta=(n/n_2)\nu_{12}$. According to Eqs. (22)–(27), it is apparent that the moments exhibit a highly nonlinear dependence on all the parameters of the problem, i.e., a, μ, δ, and κ_{rs}. The shear-rate dependence of the second-degree moments presents an excellent agreement with the one obtained from the Boltzmann equation.[6]

The use of a kinetic model allows us to derive the explicit expressions of the velocity distribution functions f_s. This is one of the main advantages of using kinetic models. In the steady shear flow state, the distribution f_1 can be written as $f_1(\mathbf{V})=n_1(m_1/2k_BT)^{3/2}g_1(\boldsymbol{\xi})$, where $\boldsymbol{\xi}\equiv(m_1/2k_BT)^{1/2}\mathbf{V}$ and the reduced distribution g_1 is

$$\begin{aligned}g_1(\boldsymbol{\xi})=\pi^{-3/2}\int_0^\infty d\tau\, e^{-(\nu_1-3\alpha)\tau}[&\nu_{11}\chi_1^{-3/2}\\&\times\exp(-\chi_1^{-1}e^{2\alpha\tau}\boldsymbol{\xi}\cdot\boldsymbol{\Gamma}_\tau\cdot\boldsymbol{\xi})+\nu_{12}\chi_{12}^{-3/2}\\&\times\exp(-\chi_{12}^{-1}e^{2\alpha\tau}\boldsymbol{\xi}\cdot\boldsymbol{\Gamma}_\tau\cdot\boldsymbol{\xi})].\end{aligned} \tag{28}$$

Here, $\boldsymbol{\Gamma}_\tau$ is the matrix defined as $\Gamma_{ij}(\tau)=\delta_{ij}+a^2\tau^2\delta_{iy}\delta_{jy}+a\tau(\delta_{ix}\delta_{jy}+\delta_{iy}\delta_{jx})$. According to (28), g_1 has a highly nonlinear dependence on the parameters of the problem.

Since all the velocity moments and the distribution functions of the GK equation in the steady USF problem are explicitly known, we are in a position to evaluate the diffusion and Dufour coefficients when the mixture is strongly sheared. This will be done in Sec. IV.

IV. SHEAR-RATE DEPENDENT TRANSPORT COEFFICIENTS

In this section we are interested in studying what is the effect of the shear flow on the mass and heat fluxes in the limit of small concentration gradients. In this case, one expects that the above-mentioned fluxes are still proportional to the concentration gradient, Eqs. (12) and (13), although the coefficients D_0 and L_0 must be replaced by their corresponding shear-rate dependent mutual diffusion and Dufour tensors, respectively. The presence of new transport coefficients (which do no exist for hydrodynamics near equilibrium) is a consequence of the anisotropy introduced in the fluid by the presence of the shear flow. The evaluation of the above-mentioned tensors is the main goal of this paper.

Let us assume that we perturb the steady USF by introducing a weak concentration gradient $\boldsymbol{\nabla}n_s$. On physical grounds, we also assume that the total density n and temperature T are constant. As a consequence, the concentration gradients are not independent but satisfy the relation $\boldsymbol{\nabla}n_1=-\boldsymbol{\nabla}n_2$. These are the typical experimental conditions for measuring the mutual diffusion coefficient in a binary mixture close to equilibrium. At a kinetic level, we will look for solutions in which all the space dependence occurs through a functional dependence on the densities n_s, since the space dependence on the flow velocity is completely absorbed by the peculiar velocity $\mathbf{V}$. In other words, we look for *normal* solutions of the form $f_s(\mathbf{r},\mathbf{v})=f_s(n_1(\mathbf{r}),n_2(\mathbf{r});\mathbf{V})$. Therefore, in the steady state, the set of GK Boltzmann equations are obtained from Eqs. (1) and (2) by

$$\begin{aligned}-\frac{\partial}{\partial V_i}(a_{ij}V_j+\alpha V_i)f_1+(V_i+a_{ij}r_j)\frac{\partial}{\partial r_i}f_1+\nu_1 f_1\\=\nu_{11}f_{11}+\nu_{12}f_{12},\end{aligned} \tag{29}$$

and similarly for f_2. Here, we have introduced again the thermostat force (18) to keep the temperature constant. In the following we will focus on the properties of species 1. In the same spirit as the usual Chapman–Enskog expansion,[3] we solve Eq. (29) by means of an expansion in powers of $\boldsymbol{\nabla}n_1$ but taking the pure shear flow distribution (28) as the zeroth-order approximation. This is the main feature of our method. Thus, we write

$$f_1=f_1^{(0)}+f_1^{(1)}+\cdots, \tag{30}$$

where $f_1^{(k)}$ is of order k in $\boldsymbol{\nabla}n_1$ but retains all the hydrodynamic orders in the shear rate. The distribution function $f_1^{(0)}$ is the same as in Eq. (28) except that now the densities n_s are nonuniform. In this paper, we will only consider the first order of the expansion.

In this first-order approximation, the kinetic equation of $f_1^{(1)}$ becomes

$$-\frac{\partial}{\partial V_i}(a_{ij}V_j+\alpha V_i)f_1^{(1)}+(V_i+a_{ij}r_j)\frac{\partial}{\partial r_i}f_1^{(0)}+\nu_1 f_1^{(1)}$$
$$=\nu_{11}f_{11}^{(1)}+\nu_{12}f_{12}^{(1)}, \tag{31}$$

where

$$f_{11}^{(1)}=\frac{1}{n_1 k_B T_1}\mathbf{V}\cdot\tilde{\mathbf{J}}_1^{(1)}f_{11}^{(0)}, \tag{32}$$

$$f_{12}^{(1)}=\frac{1}{n_1 k_B T_{12}}\left(\mu\frac{1-\delta}{1+\mu}\mathbf{V}\cdot\tilde{\mathbf{J}}_1^{(1)}+\rho_1\frac{1+\mu\delta}{1+\mu}\mathbf{V}\cdot\mathbf{u}^{(1)}\right)f_{12}^{(0)}, \tag{33}$$

$$f_{ij}^{(0)}=n_i\left(\frac{m_i}{2\pi k_B T_{ij}}\right)^{3/2}\exp\left(-\frac{m_i V^2}{2k_B T_{ij}}\right), \tag{34}$$

and

$$\tilde{\mathbf{J}}_1^{(1)}=m_1\int d\mathbf{v}\,\mathbf{V}f_1^{(1)}. \tag{35}$$

The quantity $\tilde{\mathbf{J}}_1^{(1)}$ represents the mass flux of species 1 with respect to a reference frame moving with the linear velocity profile $u_i^{(0)}=a_{ij}r_j$. As we will see later, the linear shear flow can be disturbed by the presence of the concentration gradient. For this reason, we have introduced the first-order perturbation to the velocity of the mixture $\mathbf{u}^{(1)}$, which is defined as

$$\mathbf{u}^{(1)}=\frac{1}{\rho}\sum_{s=1}^{2}\int d\mathbf{v}\,m_s\mathbf{V}f_s^{(1)}. \tag{36}$$

To first order in the expansion the conservation laws of mass and momentum imply that

$$a_{ij}r_j\frac{\partial}{\partial r_i}n_1=0, \tag{37}$$

$$a_{ik}u_k^{(1)}+\alpha u_i^{(1)}=-\frac{1}{\rho}\frac{\partial}{\partial r_k}P_{ik}^{(0)}, \tag{38}$$

where $\mathsf{P}^{(0)}$ refers to the total pressure tensor of the mixture in the USF, whose expression can be readily obtained from Eq. (22) and its counterpart for $f_2^{(0)}$, but will be omitted. In addition, in writting such balance equations we have taken into account that $\mathsf{P}_s^{(1)}=\mathbf{0}$ since $f_s^{(1)}$ is an odd function on $\mathbf{V}$.

Equation (37) implies that, in order to keep the mixture in a steady state, the concentration gradient must be orthogonal to the direction of the shear flow, i.e., $\partial n_1/\partial x=0$. On the other hand, according to Eq. (38), only in the case that the total pressure tensor is uniform the velocity field is not perturbed by the presence of the concentration gradient. If $a=0$, $P_{ij}^{(0)}=nk_BT\delta_{ij}=\text{const}$, so $\mathbf{u}^{(1)}=0$. For nonzero shear rates, there are only two limit cases for which $\mathsf{P}^{(0)}$ is constant: the case of mechanically equivalent particles[16] ($\mu=1$, $\kappa_{11}=\kappa_{22}=\kappa_{12}$) and the tracer limit ($n_1\ll n_2$).[17] In both cases, the pressure tensor can be written as $P_{ij}^{(0)}=nk_BTF(a)$, $F(a)$ being a nonlinear function of the *constant* shear rate a. Beyond these limit cases the pressure tensor depends on space through its dependence on the partial densities, and consequently the velocity field is modified by the concentration gradient. The solution to Eq. (38) is

$$u_i^{(1)}=-\frac{1}{\rho\alpha}\left(\delta_{ik}-\frac{a_{ik}}{\alpha}\right)\frac{\partial}{\partial r_j}P_{kj}^{(0)}. \tag{39}$$

We are interested in computing the mass and heat fluxes. At this order, taking into account relations (9) and (11), they are given, respectively, by

$$J_{1,i}^{(1)}=m_1\int d\mathbf{v}\,V_i f_1^{(1)}-\rho_1 u_i^{(1)}\equiv\tilde{J}_{1,i}^{(1)}-\rho_1 u_i^{(1)}, \tag{40}$$

$$q_i^{(1)}=\sum_{s=1}^{2}\frac{m_s}{2}\int d\mathbf{v}\,\mathbf{V}^2V_i f_s^{(1)}-\frac{3}{2}pu_i^{(1)}-P_{ij}^{(0)}u_j^{(1)}$$
$$\equiv\sum_{s=1}^{2}\tilde{q}_{s,i}^{(1)}-\frac{3}{2}pu_i^{(1)}-P_{ij}^{(0)}u_j^{(1)}, \tag{41}$$

where the partial contribution $\tilde{\mathbf{q}}_s^{(1)}$ to the heat flux is defined as

$$\tilde{\mathbf{q}}_s^{(1)}=\frac{m_s}{2}\int d\mathbf{v}\,\mathbf{V}^2\mathbf{V}f_s^{(1)}. \tag{42}$$

In order to get the fluxes $\tilde{\mathbf{J}}_1^{(1)}$ and $\tilde{\mathbf{q}}_s^{(1)}$, it is convenient to introduce the dimensionless moments $M_{k,l,m}^{(1)}$. They are similarly defined as in Eq. (20), but setting $f_1^{(1)}$ instead of f_1. The corresponding hierarchy that such moments obey can be obtained by multiplying both sides of Eq. (31) by $V_x^kV_y^lV_z^m$ and integrating over the velocity space. Thus, one finds that

$$akM_{k-1,l+1,m}^{(1)}+[\nu_1+\alpha(k+l+m)]M_{k,l,m}^{(1)}=R_{k,l,m}, \tag{43}$$

where

$$R_{k,l,m}=-\zeta\left[1+(1+\delta)\delta\frac{\partial}{\partial\delta}\right](M_{k,l+1,m}^{(0)}\epsilon_{1,y}+M_{k,l,m+1}^{(0)}\epsilon_{1,z})+2\nu_{11}\chi_1^{(1/2)(k+l+m-1)}[A_{k+1,l,m}M_{1,0,0}^{(1)}+A_{k,l+1,m}M_{0,1,0}^{(1)}$$
$$+A_{k,l,m+1}M_{0,0,1}^{(1)}]+2\frac{\mu}{1+\mu}(1-\delta)\nu_{12}\chi_{12}^{(1/2)(k+l+m-1)}[A_{k+1,l,m}M_{1,0,0}^{(1)}+A_{k,l+1,m}M_{0,1,0}^{(1)}+A_{k,l,m+1}M_{0,0,1}^{(1)}]$$
$$-\frac{\mu\delta}{1+\mu}(1+\delta)^2\nu_{12}\chi_{12}^{(1/2)(k+l+m-1)}[A_{k+1,l,m}\Lambda_{xy}\epsilon_{1,y}+A_{k,l+1,m}\Lambda_{yy}\epsilon_{1,y}+A_{k,l,m+1}\Lambda_{zz}\epsilon_{1,z}]. \tag{44}$$

Here, $M_{k,l,m}^{(0)}$ refers to the moments of the pure USF state given by Eq. (22) and we have introduced the reduced concentration gradient

$$\boldsymbol{\epsilon}_1=\left(\frac{2k_BT}{m_1}\right)^{1/2}\frac{1}{\zeta}\nabla\ln n_1, \tag{45}$$

and the dimensionless tensor

$$\Lambda_{ij}=\frac{1}{nk_BT}\frac{\zeta}{\alpha}\left(\delta_{ik}-\frac{a_{ik}}{\alpha}\right)\frac{\partial}{\partial\delta}P_{kj}^{(0)}. \tag{46}$$

The solution to Eq. (43) can be written as

$$M_{k,l,m}^{(1)}=\sum_{q=0}^{k}\frac{k!}{(k-q)!}(-a)^q[\nu_1+(k+l+m)\alpha]^{-(1+q)}\times R_{k-q,l+q,m}. \tag{47}$$

Equation (47) is still formal since we need to know the first degree moments, which are related to the flux $\tilde{\mathbf{J}}_1^{(1)}$. After some manipulations, it is easy to see that these moments are given by

$$M_{0,1,0}^{(1)}=-\tilde{D}_{yy}\boldsymbol{\epsilon}_{1,y}, \tag{48}$$

$$M_{0,0,1}^{(1)}=-\tilde{D}_{zz}\boldsymbol{\epsilon}_{1,z}, \tag{49}$$

$$M_{1,0,0}^{(1)}=-\tilde{D}_{xy}\boldsymbol{\epsilon}_{1,y}, \tag{50}$$

where

$$\tilde{D}_{yy}=\tilde{D}_{zz}=\frac{(1+\mu)\zeta\left[1+(1+\delta)\delta\frac{\partial}{\partial\delta}\right]M_{0,2,0}^{(0)}+\frac{1}{2}\delta\mu(1+\delta)^2\nu_{12}\Lambda_{yy}}{(1+\mu)\alpha+\nu_{12}(1+\delta\mu)}, \tag{51}$$

$$\tilde{D}_{xy}=\frac{1}{2[(1+\mu)\alpha+\nu_{12}(1+\delta\mu)]}\times\left\{\delta(1+\delta)^2\mu\nu_{12}\Lambda_{xy}-2a(1+\mu)\tilde{D}_{yy}+2(1+\mu)\zeta\left[1+(1+\delta)\delta\frac{\partial}{\partial\delta}\right]M_{1,1,0}^{(0)}\right\}. \tag{52}$$

From Eqs. (40), and (48)–(52), the mass flux $\mathbf{J}_1^{(1)}$ can be finally obtained. It can be written in the form of a generalized Fick's law, Eq. (12),

$$J_{1,i}^{(1)}=-\frac{m_1m_2n}{\rho}D_{ij}\frac{\partial}{\partial r_j}n_1, \tag{53}$$

with a mutual diffusion tensor given by

$$D_{ij}=D_0\frac{1}{1+\mu}\left[2\frac{1+\mu\delta}{1+\delta}\tilde{D}_{ij}-\mu\delta(1+\delta)\Lambda_{ij}\right], \tag{54}$$

D_0 being the mutual diffusion coefficient at equilibrium, Eq. (14). According to Eq. (54), $D_{xz}=D_{yz}=D_{zy}=0$, which is consistent with the symmetry of the problem. In addition, since $\partial n_1/\partial x=0$, the only relevant components of this tensor are $D_{yy}=D_{zz}$ and D_{xy}. They are nonlinear functions of the shear rate a and the parameters of the mixture μ, δ, κ_{11}/κ_{12}, and κ_{22}/κ_{12}. When $a=0$, $D_{ij}=D_0\delta_{ij}$ and one gets the expected result. For small shear rates, $D_{yy}\approx D_0-D_{yy}^{(2)}a^2$ and $D_{xy}\approx -D_{xy}^{(1)}a$, where $D_{yy}^{(2)}$ and $D_{xy}^{(1)}$ depend on the parameters of the mixture. Notice that the equality $P_{s,yy}=P_{s,zz}$, which is also present in the case of the exact Boltzmann equation,[10] implies that $D_{yy}=D_{zz}$. This is probably a peculiarity of the Maxwell interaction since Monte Carlo simulations[18] for a dilute single gas of hard spheres show that $P_{s,yy}\neq P_{s,zz}$.

Figures 1 and 2 show the dependence of the reduced diffusion tensor $D_{ij}^*=D_{ij}/D_0$ on the reduced shear rate $a^*=a/\zeta$ for $\delta\equiv n_1/n_2=5$, $\kappa_{11}=\kappa_{22}=\kappa_{12}$, and $\mu\equiv m_1/m_2=0.5$, and 2. The behavior for $\delta<1$ can be easily inferred by taking into account that D_{ij} is invariant under the changes $\mu\leftrightarrow\mu^{-1}$, $\delta\leftrightarrow\delta^{-1}$, and $\kappa_{11}\leftrightarrow\kappa_{22}$. We have also included the results obtained from the Boltzmann equation.[10] We observe that the GK predictions present a qualitative good agreement with the Boltzmann ones, especially when the mass of the excess component is larger than that of the defect component. The influence of the shear flow on the mass transport is quite significant in the region of shear rates analyzed. In the case of D_{yy}^*, we see that this element decreases as the shear rate increases so that the presence of the shear

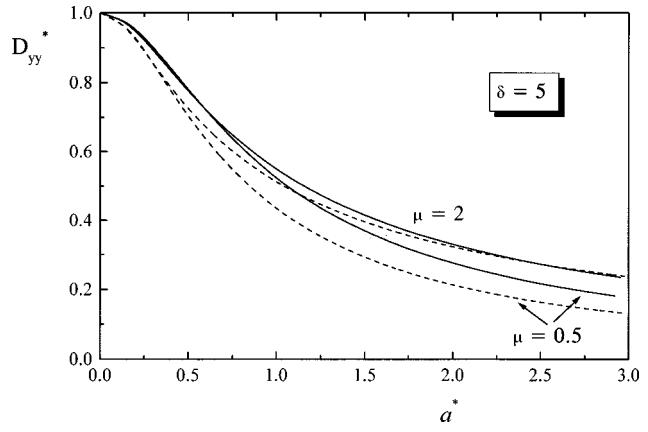

FIG. 1. Plot of the reduced diagonal element of the diffusion tensor $D_{yy}^*=D_{yy}/D_0$ as a function of the reduced shear rate $a^*=a/\zeta$ for $\kappa_{11}=\kappa_{22}=\kappa_{12}$, $\delta\equiv n_1/n_2=5$, and $\mu\equiv m_1/m_2=0.5$, and $\mu=2$. The solid lines correspond to the GK results while the dashed lines refer to the Boltzmann results.

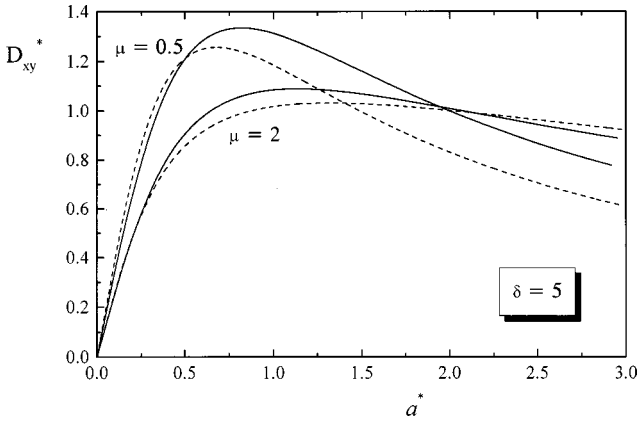

FIG. 2. Plot of the reduced diagonal element of the diffusion tensor $D_{xy}^{*}=-D_{xy}/D_0$ as a function of the reduced shear rate $a^{*}=a/\zeta$ for $\kappa_{11}=\kappa_{22}=\kappa_{12}$, $\delta\equiv n_1/n_2=5$, and $\mu\equiv m_1/m_2=0.5$, and $\mu=2$. The solid lines correspond to the GK results while the dashed lines refer to the Boltzmann results.

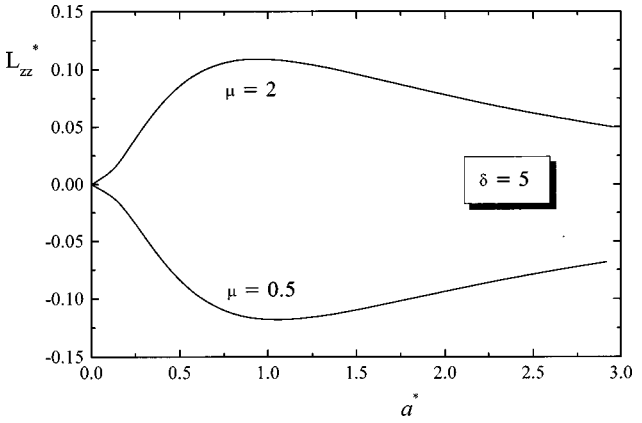

FIG. 4. Plot of the reduced diagonal element of the Dufour tensor L_{zz}^{*} as a function of the reduced shear rate $a^{*}=a/\zeta$ for $\kappa_{11}=\kappa_{22}=\kappa_{12}$, $\delta\equiv n_1/n_2=5$, and $\mu\equiv m_1/m_2=0.5$, and $\mu=2$.

flow inhibits the mass transport along the direction of the gradient of the flow velocity (y axis). This inhibition becomes more significant when the defect species is heavier than the excess species. The component D_{xy}^{*} (which is zero in the absence of shear flow) gives the mass flux along the x direction due to a concentration gradient along the y direction. It can be seen as a measure of the anisotropy generated in the system by the action of the shear field. It is negative and its dependence on the shear rate is quite similar, regardless of which mass ratio considered: for small shear rates $-D_{xy}^{*}$ increases with a^{*} while the opposite happens for large shear rates. As noted in the previous Boltzmann work,[10] the dependence of D_{ij} on a found here agrees qualitatively well with the one observed in molecular dynamics simulations[19] in a strongly shearing Lennard-Jones binary mixture.

Once the mass fluxes $\mathbf{J}_s^{(1)}$ have been obtained, all the velocity moments of $f_s^{(1)}$ can be explicitly determined from Eq. (47) and its corresponding counterpart for the component 2. An interesting moment corresponds to the heat flux created by the gradient of concentration. The total heat flux is $\mathbf{q}^{(1)}=\mathbf{q}_1^{(1)}+\mathbf{q}_2^{(1)}$ where the partial contributions $\mathbf{q}_s^{(1)}$ are given by Eq. (41) and its counterpart. The fluxes $\tilde{\mathbf{q}}_s^{(1)}$ can be obtained from (47) after some simple algebra and their explicit expressions are given in the Appendix. By collecting all the contributions coming from both species, the flux $\mathbf{J}_q$ can be obtained and consequently, the generalized Dufour tensor L identified. The calculation of such tensor is one of the main achievements of this paper. Its explicit expression is

$$L_{ij}=\frac{k_B T^2}{m_1\zeta}\frac{\rho_1\rho_2}{\rho}\left[2(\Sigma_{1,ij}-\mu\Sigma_{2,ij})-\frac{\mu(1+\delta)^3}{1+\mu\delta}\times\left(\frac{3}{2}\Lambda_{ik}+P_{ik}^{(0)*}\right)\Lambda_{kj}-\frac{5}{2}\frac{(1-\mu^2)(1+\delta)}{1+\mu\delta}D_{ij}^{*}\right], \tag{55}$$

where $P_{ij}^{(0)*}\equiv P_{ij}^{(0)}/nk_BT$ and the nonzero elements of the tensor $\boldsymbol{\Sigma}_s$ are given in the Appendix.

In the same way as the diffusion tensor, the tensor L has three relevant elements: two diagonals L_{yy}, and L_{zz}, and one off-diagonal L_{xy}. However, in contrast to D, its diagonal elements are different, which is a consequence of the high anisotropy induced by the shear field. For zero shear rate,

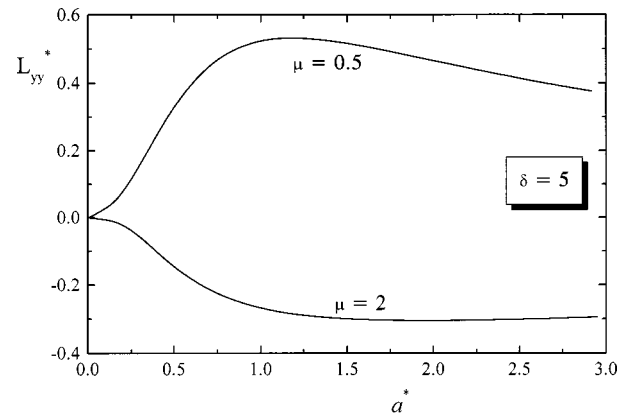

FIG. 3. Plot of the reduced diagonal element of the Dufour tensor L_{yy}^{*} as a function of the reduced shear rate $a^{*}=a/\zeta$ for $\kappa_{11}=\kappa_{22}=\kappa_{12}$, $\delta\equiv n_1/n_2=5$, and $\mu\equiv m_1/m_2=0.5$, and $\mu=2$.

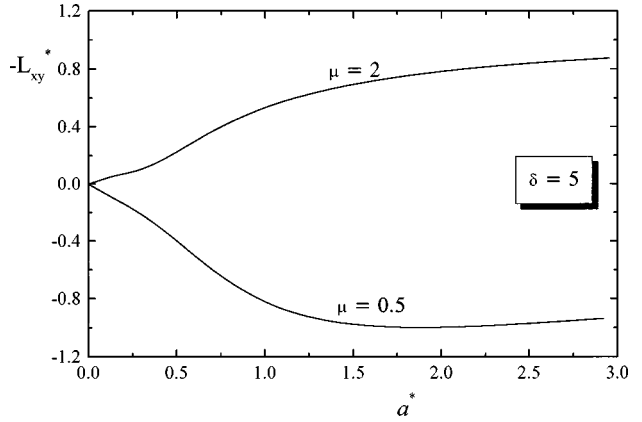

FIG. 5. Plot of the reduced off-diagonal element of the Dufour tensor $-L_{xy}^{*}$ as a function of the reduced shear rate $a^{*}=a/\zeta$ for $\kappa_{11}=\kappa_{22}=\kappa_{12}$, $\delta\equiv n_1/n_2=5$, and $\mu\equiv m_1/m_2=0.5$, and $\mu=2$.

one gets the well-known result for Maxwell binary mixtures near equilibrium, i.e., $L_{ij}=0$.[3] Also, if $\mu=1$, the result is again $L_{ij}=0$ even for $a\neq 0$. In the limit of small shear rates, $L_{yy}\approx L_{yy}^{(2)}a^2$, $L_{zz}\approx L_{zz}^{(2)}a^2$, and $L_{xy}\approx -L_{xy}^{(1)}a$, where $L_{yy}^{(2)}$, $L_{zz}^{(2)}$, and $L_{xy}^{(1)}$ are nonlinear functions of the mass ratio, the concentration ratio, and the potential parameters. In order to illustrate the dependence of the Dufour tensor on a and the parameters of the mixture, it is convenient to reduce it in a proper way. Here, we define the dimensionless Dufour tensor L^* as

$$L_{ij}^*=\frac{m_1m_2}{m_1+m_2}\frac{\zeta}{k_BT^2}\frac{\rho}{\rho_1\rho_2}L_{ij}. \tag{56}$$

The reduced tensor L_{ij}^* possesses the same invariant properties as the Dufour tensor L_{ij}. In Figs. 3, 4, and 5, we plot L_{yy}^*, L_{zz}^*, and L_{xy}^* as a function of a^*, respectively. We consider again $\delta=5$, $\kappa_{11}=\kappa_{22}=\kappa_{12}$, and the same values of the mass ratio as before, namely $\mu=0.5$ and $\mu=2$. To the best of our knowledge, we are not aware of any previous simulation or calculation of this tensor so that no comparison is possible at this stage. As Figs. 3–5 show, the various components L_{ij}^* are nonmonotonic functions of a^*, reaching either a maximum or a minimum for a given value of the shear rate. Note that depending on the value of the mass ratio, the shear-rate dependent components of the generalized Dufour tensor may either be negative or positive.

V. DISCUSSION

In a binary mixture, the presence of a concentration gradient induces mass and heat fluxes. Both fluxes define two relevant transport coefficients: the mutual diffusion coefficient and the Dufour coefficient. While the description of such processes is well developed when the mixture is close to equilibrium, much less is known when the system is far from equilibrium. In particular, an interesting question is how the diffusion and Dufour transport coefficients are affected by the action of a shear field with arbitrary strength. In this paper we have addressed this question in the context of a dilute binary mixture where the Boltzmann equation provides a controlled formulation of the problem. However, due to the complex mathematical structure of this equation, there are still difficulties for practical applications, so a kinetic model has been used to allow a detailed analysis. Specifically, we have considered the GK model of the Boltzmann equation whose reliability has been shown in the past few years in different problems.[17,20]

The physical situation is such that a linear profile of the x component of the flow velocity along the y direction coexists with a weak concentration gradient. The (constant) shear rate is arbitrary so that the mass flux $\mathbf{J}_s$ and the heat flux $\mathbf{J}_q$ can be modified by the shear flow. In addition, thermostat forces are introduced to prevent the viscous heating effect and get an stationary state. Under these conditions, $\mathbf{J}_s$ and $\mathbf{J}_q$ are still proportional to the concentration gradient but the diffusion and Dufour coefficients become shear-rate dependent tensors. The determination of these tensors in the case of Maxwell molecules has been the objective of this paper. Apart from the limitation of the interaction considered, our results apply to arbitrary values of masses, concentrations, and force constants. The solution has been obtained from a generalization of the usual Chapman–Enskog method[3] where the reference state corresponds to the stationary shear flow distribution[8] instead of the local equilibrium distribution. In the first-order approximation, we explicitly get the diffusion D_{ij} and Dufour L_{ij} tensors. These results extend previous works done in the limit cases of mechanically equivalent particles[16] and tracer particles.[17,11]

In the special case of Maxwell molecules, it is well known that for a dilute binary mixture the heat flux $\mathbf{J}_q=\mathbf{0}$ in the absence of shear flow.[3] The results obtained here show that the shearing induces a nonzero heat flux proportional to the concentration gradient even for the Maxwell interaction. Furthermore, the mass flux is highly disturbed with respect to its equilibrium value. In general, the nonzero elements of the mutual diffusion and Dufour tensors present a complex nonlinear dependence on the shear rate and the parameters of the mixture and, in particular, cross effects in the transport of mass and energy are generated by the presence of the shear flow. For instance, there exist nonzero contributions to the x component of the mass and heat fluxes due to a concentration gradient along the y direction. Concerning the diffusion tensor, $D_{yy}=D_{zz}$ and $D_{xy}<0$. The net effect of the shear field on the transport of mass is to inhibit the transport of particles along the direction of the flow velocity (y axis). In the case of x direction, $-D_{xy}$ is not a monotonic function of the shear rate and has a maximum for a given value of a^*. These results agree with those obtained from the Boltzmann equation[10] and from molecular dynamics simulations of dense mixtures.[19]

With respect to the Dufour tensor, the results show that in the presence of the shear flow $L_{yy}\neq L_{zz}$, that $L_{ij}=0$ both for $a=0$ or $\mu=1$, and that depending on the mass ratio the L_{ij}, which are nonmonotonic functions of the shear rate, may be either positive or negative, reaching a maximum or minimum for given a^*. It is interesting to point out that the question of the sign of the components L_{ij} is not, in principle, at odds with the second law of thermodynamics. Nonetheless, the actual proof of the compatibility of this feature with irreversible thermodynamics would involve the verification of an inequality that generalizes Eq. (221) in Ref. 1 to the case of tensorial thermal conductivity, diffusion, and thermal diffusion. This verification is not feasible at this stage due to the fact that neither the precise inequality nor the shear dependent thermal conductivity for the mixture are presently available. But the point is certainly interesting and we may address it in the future. To provide a proper perspective in which the importance of the calculations involving L_{ij} may be further assessed, the following comments are in order. As said before, to our knowledge this is the first derivation of an explicit expression of the shear-rate dependent Dufour tensor. It could be argued that consideration of Maxwell molecules somewhat restricts the usefulness of such an expression. However, it is important to remark that this choice was motivated by the fact that, in the absence of shear flow, the GK model yields *always* a zero Dufour coefficient irrespective of the intermolecular potential, whereas the Boltzmann equation only does it for the case of Maxwell

molecules. Therefore it seemed only natural to consider a potential in which the correct Dufour coefficient is obtained at zero shear rate. On the other hand, at least for repulsive potentials, our previous findings[8] indicate that other transport coefficients are rather insensitive to the choice of the power law and on these grounds we would expect this also to be the case for the Dufour tensor.

The results reported here can also be of relevance in connection with computer simulations. We have already mentioned the simulation performed by Sarman, Evans, and Baranyai[19] to analyze the shear rate dependence of the mutual diffusion tensor of a dense mixture. Nevertheless, when one extrapolates our definition of the collision frequency ζ to dense fluids, one estimates that the shear rates applied in this simulation are not large enough to clearly observe nonlinear effects. In the case of the Dufour tensor no simulation data are known to date, even for dense systems. As we have indicated in previous works, one possibility to overcome the difficulties inherent to molecular dynamics to achieve large shear rates in the low density regime is to use the direct simulation Monte Carlo method.[21] We hope that the results derived here for the mutual diffusion and Dufour tensors stimulates the performance of computer simulations to check the accuracy of our predictions.

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APPENDIX: PARTIAL CONTRIBUTIONS TO THE HEAT FLUX

In this appendix we explicitly write the nonzero elements L_{yy}, L_{zz}, and L_{xy} of the Dufour tensor L. First, the partial contributions to the heat flux $\tilde{\mathbf{q}}_s^{(1)}$ can be written as

$$\tilde{\mathbf{q}}_s^{(1)} = -\frac{m_1 n_1}{2}\left(\frac{2k_B T}{m_1}\right)^{3/2} \boldsymbol{\Sigma}_s \cdot \boldsymbol{\epsilon}_s, \tag{A1}$$

where the relevant elements of $\boldsymbol{\Sigma}_s$ are given by

$$\begin{aligned}\Sigma_{1,xy} = &-\frac{1}{z_1^4}\Delta[6a^3 M_{0,4,0}^{(0)} + a z_1^2 (M_{0,2,2}^{(0)} + M_{0,4,0}^{(0)} \\ &+ 3M_{2,2,0}^{(0)}) - z_1^3 (M_{3,1,0}^{(0)} + M_{1,1,2}^{(0)}) \\ &- z_1 (z_1^2 + 6a^2) M_{1,3,0}^{(0)}] \\ &- A_1 \frac{(7z_1^2 + 18a^2) a \tilde{D}_{yy} - (5z_1^2 + 6a^2) z_1 \tilde{D}_{xy}}{2(1+\mu) z_1^4} \\ &- B_1 \frac{(7z_1^2 + 18a^2) a \Lambda_{yy} - (5z_1^2 + 6a^2) z_1 \Lambda_{xy}}{4(1+\mu) z_1^4},\end{aligned} \tag{A2}$$

$$\begin{aligned}\Sigma_{1,yy} = &\frac{1}{z_1}\Delta\left[M_{0,2,2}^{(0)} + M_{2,2,0}^{(0)} + \frac{2a^2 + z_1^2}{z_1^2} M_{0,4,0}^{(0)} \right. \\ &\left. - \frac{2a}{z_1} M_{1,3,0}^{(0)}\right] + A_1 \frac{(5z_1^2 + 6a^2)\tilde{D}_{yy} - 2a z_1 \tilde{D}_{xy}}{2(1+\mu) z_1^3} \\ &+ B_1 \frac{(5z_1^2 + 6a^2)\Lambda_{yy} - 2a z_1 \Lambda_{xy}}{4(1+\mu) z_1^3},\end{aligned} \tag{A3}$$

$$\begin{aligned}\Sigma_{1,zz} = &\frac{1}{z_1}\Delta\left[M_{2,0,2}^{(0)} + M_{0,0,4}^{(0)} + \frac{2a^2 + z_1^2}{z_1^2} M_{0,2,2}^{(0)} \right. \\ &\left. - \frac{2a}{z_1} M_{1,1,2}^{(0)}\right] + A_1 \frac{5z_1^2 + 2a^2}{2(1+\mu) z_1^3} \tilde{D}_{zz} \\ &+ B_1 \frac{5z_1^2 + 2a^2}{4(1+\mu) z_1^3} \Lambda_{zz}.\end{aligned} \tag{A4}$$

Here, we have introduced the operator

$$\Delta[g(\delta)] = \left(1 + \delta(1+\delta)\frac{\partial}{\partial \delta}\right) g(\delta), \tag{A5}$$

and the quantities

$$z_1 = \nu_1 + 3\alpha, \tag{A6}$$

$$A_1 = (1+\mu)\nu_{11}\chi_1 + (1-\delta)\mu\nu_{12}\chi_{12}, \tag{A7}$$

$$B_1 = \mu\nu_{12}\delta(1+\delta)^2\chi_{12}. \tag{A8}$$

The relevant elements of $\boldsymbol{\Sigma}_2$ can be obtained from Eqs. (A2) to (A8) by the changes: $\mu \leftrightarrow \mu^{-1}$, $\delta \leftrightarrow \delta^{-1}$, and $\kappa_{11} \leftrightarrow \kappa_{22}$. The Dufour tensor can be identified from the flux $\mathbf{J}_q$ defined in Eq. (13). By collecting all the contributions, it is straightforward to get the expression (55) appearing in the main text.

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