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Diffusion in a gaseous dilute solution under heat and momentum transport

V. Garzó and A. Santos
Departamento de Física, Universidad de Extremadura, E-06071 Badajoz, Spain

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A diffusion problem in a gaseous dilute solution in a steady state with both temperature and velocity gradients is studied. The results are obtained from the Gross-Krook model [Phys. Rev. **102**, 593 (1956)] of the Boltzmann equation for a binary mixture. A perturbation expansion around a nonequilibrium state with both arbitrary velocity and temperature gradients is applied to get the diffusion tensor of the solute particles. This tensor is given in terms of the shear rate, the mass ratio, and the force constant ratio. In addition, the velocity distribution function corresponding to the tracer species is explicitly written.

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I. INTRODUCTION

The understanding of diffusion processes taking place in fluid mixtures far from equilibrium is still an open problem. The general description of these systems is much more complicated than that of a single gas since the transport properties are now functions of the mass ratios, the molar fractions, and the size ratios. This complexity leads to consider tractable examples to gain insight into the general behavior of the mass transport under arbitrary nonequilibrium conditions.

In the past few years a great effort has been devoted to the study of diffusion problems in fluids subjected to strong shear flows. In the context of dense gases, generalized Green-Kubo relations for self-diffusion [1] and mutual diffusion tensors [2] under shear flow have been derived. Similar studies have been carried out for dilute binary mixtures in the case of mechanically equivalent particles [3] as well as in the case of unequal masses when one of the components is present in tracer concentration [4]. All these results refer to situations in which a weak concentration gradient coexists with an arbitrarily large *linear* velocity profile, the temperature being uniform.

The aim of this paper is to analyze a diffusion problem in a low-density binary mixture under steady planar Couette flow. The physical situation corresponds to a mixture with a nonuniform molar fraction enclosed between two parallel plates in relative motion and at different temperatures. Our goal is to describe the influence of both velocity and temperature gradients on the diffusion in the system. Since the analysis of the general problem seems to be unapproachable, we will consider again a mixture with a solute molar fraction much smaller than 1 (tracer limit). Consequently, the state of the solvent is not significantly modified by collisions with the solute particles and, in addition, one can neglect the effect on the state of the solute of collisions among the solute particles themselves.

Unfortunately, the program outlined above cannot be carried out analytically using the Boltzmann equation [5], since no explicit solution for the steady Couette flow is known, even for the single gas. Therefore, one must resort to numerical solutions or to the use of simplified kinetic models. Here we will follow the second approach by considering the Gross-Krook (GK) kinetic model [6] for binary mixtures. This model is constructed in the same spirit as the Bhatnagar-Gross-Krook (BGK) model of a single gas [7], for which an exact solution has been found for the steady Couette flow [8,9].

Since the state of the solvent component is well characterized, we shall solve the kinetic equation corresponding to the solute component by performing a perturbation expansion around a nonequilibrium state with arbitrary values for the shear rate and the thermal gradient. To first order in the gradient of molar fraction, we get explicit expressions for the relevant transport properties associated with the solute particles, namely, the mutual diffusion tensor and the partial pressure. Both quantities are nonlinear functions of the shear rate and also of the ratios of mass and force constant. These expressions extend previous results obtained in the case of mechanically identical particles (i.e., equal masses and force constants) [10].

The plan of the paper is as follows. The physical problem is introduced in Sec. II. A brief summary of relevant results concerning the steady Couette problem at the level of the BGK equation is also included. In Sec. III we obtain the main transport properties of the solute. Further, its corresponding velocity distribution function is explicitly written up to first order in the expansion. Finally, the results are summarized and discussed in Sec. IV.

II. DESCRIPTION OF THE PROBLEM

Let us consider a dilute binary mixture. Let $f_i(\mathbf{r},\mathbf{v};t)$ be the velocity distribution function of species i ($i=1,2$). The local number density and mean velocity of species i are defined, respectively, as

$$n_i = \int d\mathbf{v}\, f_i \,, \tag{1}$$

$$\mathbf{u}_i = \frac{1}{n_i}\int d\mathbf{v}\,\mathbf{v} f_i \ . \tag{2}$$

We also define a local temperature T_i for each species through

$$\frac{3}{2}n_i k_B T_i = \frac{m_i}{2}\int d\mathbf{v}\,(\mathbf{v}-\mathbf{u}_i)^2 f_i \ , \tag{3}$$

where k_B is the Boltzmann constant and m_i is the mass of a particle of species i.

In the case of a dilute mixture, the master equation for f_i is the Boltzmann equation [5]. However, due to its mathematical complexity, it is a very difficult task to solve such an equation, especially in far from equilibrium situations. For this reason several kinetic models have been proposed. Here we consider the well-known GK model [6], in which the Boltzmann collision operators $J_{ij}[f_i, f_j]$ are replaced by relaxation terms of the form

$$J_{ij}^{\mathrm{GK}} = -\nu_{ij}(f_i - f_{ij}) \ , \tag{4}$$

where ν_{ij} is an effective collision frequency and

$$f_{ij} = n_i\left(\frac{m_i}{2k_B T_{ij}}\right)^{3/2}\exp\left[-\frac{m_i}{2k_B T_{ij}}(\mathbf{v}-\mathbf{u}_{ij})^2\right] \ . \tag{5}$$

The parameters $\mathbf{u}_{ij}$ and T_{ij} are determined by imposing the conditions

$$\int d\mathbf{v} \times \left\{\begin{matrix}\mathbf{v}\\ v^2\end{matrix}\right\} \times (J_{ij}^{\mathrm{GK}} - J_{ij}) = \left\{\begin{matrix}\mathbf{0}\\ 0\end{matrix}\right\} \ , \tag{6}$$

in the special case of Maxwell molecules [i.e., particles interacting via a potential $V_{ij}(r) = \kappa_{ij} r^{-4}$]. Under these conditions one obtains that

$$\mathbf{u}_{ij} = \frac{m_i\mathbf{u}_i + m_j\mathbf{u}_j}{m_i + m_j} \ , \tag{7}$$

$$T_{ij} = T_i + 2\frac{m_i m_j}{(m_i+m_j)^2}\left[(T_j - T_i) + \frac{m_j}{6k_B}(\mathbf{u}_i - \mathbf{u}_j)^2\right] \ , \tag{8}$$

and one can identify the effective collision frequency ν_{ij} by

$$\nu_{ij} = A n_j\left[\kappa_{ij}\frac{m_i + m_j}{m_i m_j}\right]^{1/2} \ , \tag{9}$$

where $A = 4\pi \times 0.422$.

We describe now the problem we are interested in. We consider a dilute binary mixture in which the masses of both species are arbitrary. Since the general description of diffusion in arbitrary nonequilibrium conditions is a complex problem, we will choose a case that shares the simplicity of the single gas problem but yet introduces the mass ratio as a new ingredient. This case corresponds to a gaseous dilute solution, in which the concentration of one of the components (the solute) is much smaller than that of the other component (the solvent). In this limit (tracer limit), one expects that the state of the solvent, say, 2, is not disturbed by collisions with the tracer species, so that the velocity distribution function f_2 verifies a closed equation (the BGK equation). Further, the concentration of the solute particles is so small that their mutual interactions can be neglected versus the solute-solvent collisions.

Under the above conditions, we assume that the solvent component is in steady planar Couette flow, namely, it is enclosed between two parallel plates (normal to the y axis) in relative motion (along the x axis) and maintained at different temperatures. In this state, an exact solution to the BGK model has been found [8]. This solution is characterized by a uniform pressure p_2, and linear velocity and parabolic temperature profiles with respect to a scaled space variable, i.e.,

$$p_2 \equiv n_2 k_B T_2 = \mathrm{const} \ , \tag{10}$$

$$\frac{1}{\nu_{22}(y)}\frac{\partial}{\partial y}u_{2,x} = a = \mathrm{const} \ , \tag{11}$$

$$\left[\frac{1}{\nu_{22}(y)}\frac{\partial}{\partial y}\right]^2 T_2 = -\frac{2m_2}{k_B}\gamma(a) = \mathrm{const} \ . \tag{12}$$

The dimensionless parameter γ is a nonlinear function of the reduced shear rate a through the implicit equation

$$a^2 = \gamma\frac{2F_2(\gamma) + 3F_1(\gamma)}{F_1(\gamma)} \ , \tag{13}$$

where $F_s(\gamma) = [(d/d\gamma)\gamma]^s F_0(\gamma)$ and

$$F_0(\gamma) = \frac{2}{\gamma}\int_0^\infty dt\, t\exp(-t^2/2)K_0(2\gamma^{-1/4}t^{1/2}) \ , \tag{14}$$

K_0 being the zeroth-order modified Bessel function.

Taking into account the profiles (10)–(12), the relevant fluxes associated with the solvent particles can be obtained. For instance, the partial pressure tensor

$$\mathsf{P}_2 = m_2\int d\mathbf{v}\,(\mathbf{v}-\mathbf{u}_2)(\mathbf{v}-\mathbf{u}_2)f_2 \tag{15}$$

and the partial heat flux

$$\mathbf{q}_2 = \frac{m_2}{2}\int d\mathbf{v}\,(\mathbf{v}-\mathbf{u}_2)^2(\mathbf{v}-\mathbf{u}_2)f_2 \tag{16}$$

are given by [8,10]

$$P_{2,xx} = p_2\{1 + 4\gamma[F_1(\gamma) + F_2(\gamma)]\} \ , \tag{17}$$

$$P_{2,yy} = p_2\{1 - 2\gamma[F_1(\gamma) + 2F_2(\gamma)]\} \ , \tag{18}$$

$$P_{2,zz} = p_2[1 - 2\gamma F_1(\gamma)] \ , \tag{19}$$

$$P_{2,xy} = -\frac{p_2}{\nu_{22}}F_0(\gamma)\frac{\partial}{\partial y}u_{2,x} \ , \tag{20}$$

$$q_{2,y} = -\frac{p_2 k_B}{2m_2\nu_{22}}\frac{a^2}{\gamma}F_0(\gamma)\frac{\partial}{\partial y}T_2 \ . \tag{21}$$

In addition, the velocity distribution function f_2 can be obtained. Its explicit form can be found in Ref. [9].

Since the state of the solvent component is well known, our goal is now to compute the main transport properties of the solute component (tracer particles) when diffusion takes place. This will be done in the next section.

III. TRACER DIFFUSION UNDER HEAT AND MOMENTUM TRANSPORT

Under the assumptions established in the tracer limit, the steady kinetic equation for the velocity distribution function f_1 becomes

$$\mathbf{v}\cdot\nabla f_1 = -\nu_{12}(f_1 - f_{12}) \;, \tag{22}$$

where, according to Eq. (9),

$$\nu_{12} = \omega^2 \left(\frac{1+\mu}{2}\right)^{1/2} \nu_{22} \;, \tag{23}$$

$\omega \equiv (\kappa_{12}/\kappa_{22})^{1/4}$ and $\mu \equiv m_2/m_1$ being the force constant ratio and the mass ratio, respectively [11]. We are interested in analyzing the effect of the steady Couette flow on the diffusion of solute particles in terms of both velocity and temperature gradients and the parameters of the mixture. To this end we shall solve Eq. (22) by following an approach already used in our previous works [4,10]. Assuming that the molar fraction $x_1 = n_1/n_2$ is only slightly nonuniform, we perform an expansion around a nonequilibrium state (with arbitrarily large velocity and temperature gradients) by taking ∇x_1 as the perturbation parameter. As a consequence, the transport coefficients obtained from this method will be nonlinear functions of the shear rate and the thermal gradient. Since we are interested in deriving an explicit expression for the tracer diffusion tensor, our calculations will go up to the first order in ∇x_1. We assume that f_1 can be written in the form

$$f_1 = f_1^{(k)} + O(\nabla^{k+1} x_1) \;, \tag{24}$$

where the approximation $f_1^{(k)}$ contains the contributions up to order k in the gradient of molar fraction. In a similar way the fields $\mathbf{u}_{12}$ and T_{12} must be expanded as

$$\mathbf{u}_{12} = \mathbf{u}_{12}^{(k)} + O(\nabla^{k+1} x_1) \;, \tag{25}$$

$$T_{12} = T_{12}^{(k)} + O(\nabla^{k+1} x_1) \;, \tag{26}$$

where the approximations $\mathbf{u}_{12}^{(k)}$ and $T_{12}^{(k)}$ define the corresponding reference function $f_{12}^{(k)}$. By substituting the above expansions into Eq. (22), one gets a hierarchy of equations for the successive functions $f_1^{(k)}$. Now we are ready to examine the first two approximations.

A. Zeroth-order approximation

This approximation is concerned with a situation where no mutual diffusion appears in the system. This reference state has been recently analyzed by the present authors [12] and now we offer a brief summary of the most important results. In the absence of diffusion, i.e., $x_1 = \text{const}$ and $\mathbf{u}_1^{(0)} = \mathbf{u}_2$, the state of the solute is characterized by the profiles

$$p_{12}^{(0)} \equiv n_2 k_B T_{12}^{(0)} = \text{const} \;, \tag{27}$$

$$\frac{1}{\nu_{12}}\frac{\partial}{\partial y} u_{12,x}^{(0)} = \widetilde{a} = \text{const} \;, \tag{28}$$

$$\left[\frac{1}{\nu_{12}}\frac{\partial}{\partial y}\right]^2 T_{12}^{(0)} = -\frac{2m_1}{k_B}\widetilde{\gamma} = \text{const} \;. \tag{29}$$

Here, $\mathbf{u}_{12}^{(0)} = \mathbf{u}_1^{(0)} = \mathbf{u}_2$ and $T_{12}^{(0)}/T_2 = \chi + 2M(1-\chi)$ with $M \equiv \mu/(1+\mu)^2$ and $\chi \equiv T_1^{(0)}/T_2$. The ratio χ between the temperatures of the solute and the solvent is the crucial quantity at this stage of description. Equations (28) and (29) imply that

$$\widetilde{a} \equiv \frac{a}{\omega^2[(1+\mu)/2]^{1/2}} \;, \tag{30}$$

$$\widetilde{\gamma} \equiv \frac{2\mu}{1+\mu}\frac{\chi + 2M(1-\chi)}{\omega^4}\gamma \;. \tag{31}$$

The self-consistency of the solution leads to the following implicit equation for $\widetilde{\gamma}$:

$$2F_2(\widetilde{\gamma}) + \left(3 - \frac{\widetilde{a}^2}{\widetilde{\gamma}}\right) F_1(\widetilde{\gamma}) = \frac{3}{\widetilde{\gamma}}\frac{M(1-\chi)}{\chi + 2M(1-\chi)} \;. \tag{32}$$

The solution of this equation gives χ as a function of a, μ, and ω. In general, χ varies monotonically from 1 (limit of zero shear rate) to $1/\mu$ (large shear rates). From the knowledge of this quantity, the partial shear viscosity and thermal conductivity can be computed [12].

In addition, an explicit expression for the solute distribution function has been derived. As a matter of fact, this function represents the reference state around which we perform our expansion. It can be written as $f_1^{(0)} = n_1(m_1/2\pi k_B T_2)^{3/2}\Phi(\boldsymbol{\xi})$, where

$$\begin{aligned}\Phi(\boldsymbol{\xi}) = {} & \omega^2\left(\frac{1+\mu}{2\mu}\right)^{1/2}[\chi + 2M(1-\chi)]^{-3/2}\,\frac{2\alpha(1+\alpha)^{3/2}}{\epsilon|\xi_y|}\int_{t_0}^{t_1} dt[2t-(1-\alpha)t^2]^{-5/2}\\ & \times\exp\left\{-\omega^2\left(\frac{1+\mu}{2\mu}\right)^{1/2}\frac{2\alpha}{1+\alpha}\frac{1-t}{\epsilon\xi_y} - [\chi + 2M(1-\chi)]^{-1}\,\frac{1+\alpha}{2t-(1-\alpha)t^2}\right.\\ & \left.\times\left[\left(\xi_x + \frac{2a\alpha}{1+\alpha}\frac{1-t}{\mu^{1/2}\epsilon}\right)^2 + \xi_y^2 + \xi_z^2\right]\right\} .\end{aligned} \tag{33}$$

Here $(t_0, t_1) = (0, 1)$ if $\xi_y > 0$ and $(t_0, t_1) = [1, 2/(1-\alpha)]$ if $\xi_y < 0$. Besides, $\boldsymbol{\xi} \equiv (m_1/2k_BT_2)^{1/2}(\mathbf{v} - \mathbf{u}_2)$,

$$\epsilon \equiv \frac{1}{\nu_{22}}\left(\frac{2k_BT_2}{m_2}\right)^{1/2}\frac{1}{T_2}\frac{\partial}{\partial y}T_2 \tag{34}$$

is the reduced thermal gradient, and

$$\alpha \equiv \frac{\epsilon}{(\epsilon^2 + 8\gamma)^{1/2}} \,. \tag{35}$$

B. First-order approximation

Let us assume now that we disturb the above state by introducing a weak molar fraction gradient ∇x_1. We are interested in obtaining the diffusion tensor when only terms up to first order in ∇x_1 are retained. In this approximation, and according to Eq. (22), the balance equation for the concentration of the solute component implies that $\mathbf{u}_2 \cdot \nabla x_1 = 0$. Consequently, we will take the concentration gradient parallel to both temperature and velocity gradients, i.e., $\nabla x_1 \equiv (\partial x_1/\partial y)\hat{\mathbf{y}}$. Under this geometry, the kinetic equation for $f_1^{(1)}$ reads

$$v_y \frac{\partial}{\partial y} f_1^{(1)} = -\nu_{12}(f_1^{(1)} - f_{12}^{(1)}) \,, \tag{36}$$

where $f_{12}^{(1)}$ is given by

$$f_{12}^{(1)} = \left[1 + \frac{m_1}{n_1k_BT_{12}^{(0)}}\frac{1}{1+\mu}\mathbf{V}\cdot\mathbf{j}_1^{(1)} + \left(\frac{m_1V^2}{2k_BT_{12}^{(0)}} - \frac{3}{2}\right)\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}} - 1\right)\right] f_{12}^{(0)} \,. \tag{37}$$

Here $\mathbf{V} \equiv \mathbf{v} - \mathbf{u}_2$ and we have introduced the flux of tracer particles $\mathbf{j}_1^{(1)}$

$$\mathbf{j}_1^{(1)} = \int d\mathbf{v}\,\mathbf{V} f_1^{(1)} \tag{38}$$

and the reference function

$$f_{12}^{(0)} = n_1\left(\frac{m_1}{2\pi k_BT_{12}^{(0)}}\right)^{3/2}\exp\left(-\frac{m_1V^2}{2k_BT_{12}^{(0)}}\right) \,. \tag{39}$$

The ratio $T_{12}^{(1)}/T_{12}^{(0)}$ is related to the partial pressure of the tracer species through the relation

$$x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}} - 1\right) = (1 - 2M)\frac{p_1^{(1)} - p_1^{(0)}}{p_{12}^{(0)}} \,, \tag{40}$$

where

$$p_1^{(k)} = \frac{m_1}{3}\int d\mathbf{v}\,V^2 f_1^{(k)} \,. \tag{41}$$

The formal solution of Eq. (36) is

$$f_1^{(1)} = \left(1 + \frac{v_y}{\nu_{12}}\frac{\partial}{\partial y}\right)^{-1} f_{12}^{(1)} \,. \tag{42}$$

Notice that when the operator $[1 + (v_y/\nu_{12})(\partial/\partial y)]^{-1}$ acts on the function $f_{12}^{(1)}$ only terms of first order in ∇x_1 need to be retained. The solution (42) is still formal as $f_{12}^{(1)}$ depends on $f_1^{(1)}$ through its first nontrivial moments $\mathbf{j}_1^{(1)}$ and $p_1^{(1)}$. The calculation of these moments is quite tedious and is made in the Appendix. The result is

$$j_{1,i}^{(1)} = -\frac{1+\mu}{\mu}\frac{p_2}{m_1\nu_{12}}D_{ik}(a)\frac{\partial}{\partial r_k}x_1 \,, \tag{43}$$

$$p_1^{(1)} - p_1^{(0)} = \frac{5}{6}\frac{(1+\mu)^2(1+2\mu)}{\mu^2}\frac{p_2k_B}{m_1\nu_{12}^2}\Omega(a)\frac{\partial T_2}{\partial y}\frac{\partial x_1}{\partial y} \,. \tag{44}$$

The explicit expressions of the reduced tracer diffusion tensor D and the function Ω are given in the Appendix. These expressions extend previous results derived in the particular case of mechanically equivalent particles (i.e., $\omega = 1$, $\mu = 1$) [13]. Equation (43) can be seen as a generalization of the usual Fick's law (valid in the absence of both velocity and temperature gradients). It describes the mass transport of solute particles when the solvent is subject to arbitrarily large velocity and temperature gradients. Due to the anisotropy induced by the shear flow, a diffusion tensor rather than a scalar is identified. Since no concentration gradient exists along the x and z directions, the relevant elements in this problem are D_{yy} and D_{xy}. These elements are given in terms of the shear rate, the mass ratio, and the force constants ratio, but they do not explicitly depend on the thermal gradient. The fact that the mass flux does not depend on the thermal gradient (which implies the absence of thermal diffusion) is probably due to the particular Maxwell interaction since a similar property also appears in the linear regime [5]. For $a = 0$, $D_{ik} = \delta_{ik}$, so that one recovers the familiar mutual diffusion coefficient for the tracer species.

In Figs. 1 and 2 we show the shear-rate dependence of D_{yy} and $-D_{xy}$ for $\omega = 1$ and three values of the mass ratio. The diagonal element D_{yy} decreases as the

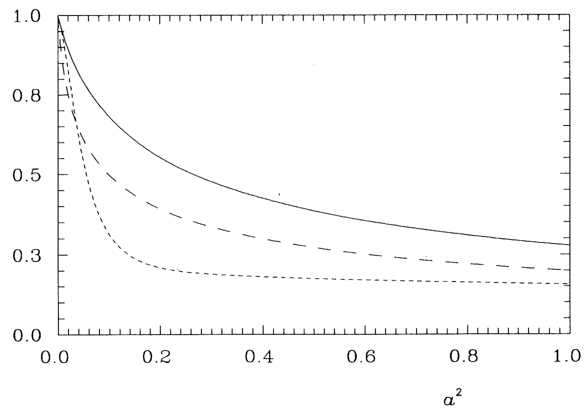

FIG. 1. Shear-rate dependence of the reduced element D_{yy} for $\omega = 1$ and three values of the mass ratio: $\mu = 1$ (—), $\mu = 10$ (– – –), and $\mu = 0.1$ (- - -).

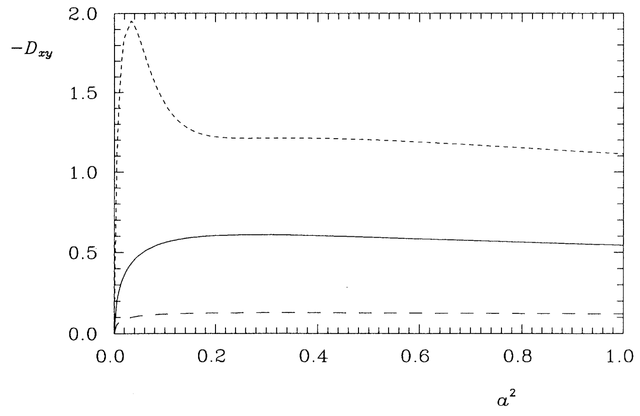

FIG. 2. Same as in Fig. 1, but for $-D_{xy}$.

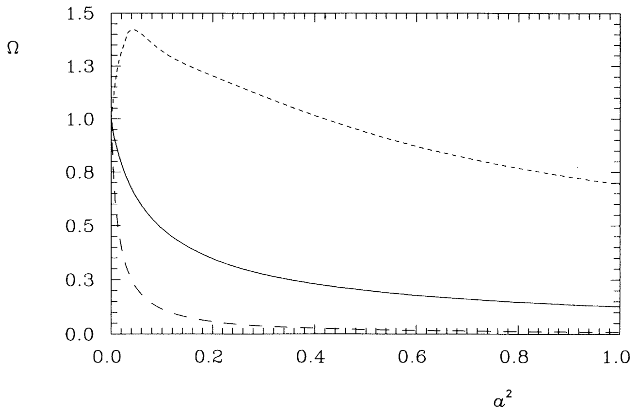

FIG. 3. Same as in Fig. 1, but for Ω.

shear rate increases whatever the mass ratio considered is. Consequently, the mass transport along the y direction is inhibited by the shear flow. For finite values of the shear rate, this inhibition becomes more noticeable when the mass ratio is different from 1 than in the particular case of identical particles. The off-diagonal element D_{xy} measures cross effects induced by the shear flow in the mass transport of tracer particles. We see that this coefficient is always negative and its absolute value increases as the mass of the tracer particles increases. In particular, for light tracer particles, this value is practically zero. In addition, we find that there exists a small region of shear rates for which $-D_{xy}$ increases with a, while the opposite occurs for larger shear rates. This decrease is not completely monotonic in the case of small mass ratios since there is a region of shear rates for which $-D_{xy}$ increases slightly again with a.

Equation (44) indicates that the deviation of the partial pressure of the solute component from its equilibrium value is at least of second order in the gradients (Burnett order) since it is proportional to $(\partial T_2/\partial y)(\partial x_1/\partial y)$. The function Ω measures the combined effect of the shear rate and the mass ratio on the partial pressure. It is again a nonlinear function of both parameters. In Fig. 3 we have plotted Ω for the same cases as in Figs. 1 and 2. According to the behavior of Ω for small shear rates (see the Appendix), we find that this function monotonically decreases as the shear rate increases if $\mu \gtrsim 0.468$. Thus, for $\mu = 0.1$, there is a region in which Ω increases with a. We observe again that Ω tends quickly to zero in the case of light tracer particles.

The knowledge of the first moments of the velocity distribution function of the solute allows one to explicitly write it. Following mathematical steps identical to those done in Appendix C of Ref. [10], it is straightforward to prove that

$$\begin{aligned} f_1^{(1)} - f_1^{(0)} = & -\left(\frac{2k_BT_2}{m_1}\right)^{1/2} (\partial_s \ln x_1) \Bigg\{ \xi_y \mathcal{D}\xi_y + \frac{T_2}{\mu T_{12}^{(0)}} (D_{xy}\xi_x + D_{yy}\xi_y + \widetilde{a} D_{xy}\xi_y \mathcal{D}\xi_y) \\ & + \frac{5}{6}(1-2M)\frac{(1+\mu)^2(1+2\mu)}{\mu^2}\frac{T_2}{T_{12}^{(0)}}\Omega \Bigg[\frac{\mu^{1/2}}{4}\frac{\nu_{22}}{\nu_{12}}(\xi^2\mathcal{D}_z + 3)\epsilon \\ & + \left(\frac{\mu^{1/2}}{2}\frac{\nu_{22}}{\nu_{12}}\widetilde{a}\epsilon\xi_x\mathcal{D}_z + \frac{T_2}{T_{12}^{(0)}}\widetilde{\gamma}(\xi^2\mathcal{D}_z + 3)\right)\xi_y\mathcal{D}\xi_y \\ & + \left(\frac{\mu^{1/2}}{4}\frac{\nu_{22}}{\nu_{12}}\widetilde{a}^2\epsilon\mathcal{D}_z + 2\frac{T_2}{T_{12}^{(0)}}\widetilde{\gamma}\widetilde{a}\xi_x\mathcal{D}_z\right)\xi_y^2\mathcal{D}^2\xi_y^2 + \frac{T_2}{T_{12}^{(0)}}\widetilde{\gamma}\widetilde{a}^2\mathcal{D}_z\xi_y^3\mathcal{D}^3\xi_y^3 \Bigg] \Bigg\} f_1^{(0)} , \end{aligned} \tag{45}$$

where $\partial_s \equiv (1/\nu_{12})\partial/\partial y$ and we have introduced the operators

$$\mathcal{D}_z \equiv \xi_z^{-1}\frac{\partial}{\partial \xi_z} , \quad \mathcal{D} \equiv \frac{\partial}{\partial \xi_y} - \xi_y \mathcal{D}_z . \tag{46}$$

Equation (45) extends the one derived in the self-diffusion problem [10] since it applies to both arbitrary mass ratio and force constants ratio. The function $f_1^{(1)}$ is expressed in terms of a complicated operator acting on the reference distribution function $f_1^{(0)}$. It represents a generalization of the familiar Navier-Stokes distribution function corresponding to the mutual diffusion problem [5].

IV. SUMMARY AND DISCUSSION

A mutual diffusion problem in a gaseous dilute solution far from equilibrium has been analyzed. The solution is assumed to be so dilute that the state of the solvent is not disturbed by the presence of the solute and the state of the solute is only affected by cross collisions among particles of the solvent and the solute. This situation is usually referred to as the tracer limit. Under these conditions, and for a low-density binary mixture, the solvent distribution f_2 verifies a closed Boltzmann equation and the solute distribution f_1 obeys a Boltzmann-Lorentz equation. The physical situation we have addressed corresponds to a system enclosed between two parallel plates in relative motion and kept at different temperatures (steady planar Couette flow). In addition, we assume that the molar fraction x_1 is nonuniform, so that tracer diffusion appears in the system.

Due to the mathematical complexity embodied in the Boltzmann collision operators, we use the nonlinear GK model for binary mixtures of Maxwell molecules. In this case, the kinetic equation for f_2 becomes the BGK model of a single gas, for which an exact solution in the steady Couette flow is known [8,9]. Consequently, our goal now is to solve the corresponding kinetic equation for the velocity distribution function f_1 of the solute. By assuming that the molar fraction gradient is weak, we perform an expansion around a nonequilibrium state with arbitrary velocity and temperature gradients. Therefore, each approximation $f_1^{(k)}$ will neglect terms of order higher than k in ∇x_1, but will retain the full nonlinear dependence on both the shear rate a and the thermal gradient ϵ, as well as on the mass ratio μ and the size ratio ω. Here we have restricted ourselves to the first order in ∇x_1 and an explicit expression for the mass flux of tracer particles $\mathbf{j}_1^{(1)}$ has been obtained. It obeys a generalized Fick law from which the mutual diffusion tensor D_{ij} is identified. This tensor happens to be a nonlinear function of the shear rate, but it is independent of the temperature gradient. In addition, the partial pressure of the tracer particles has also been obtained, being linear in ϵ and nonlinear in a.

According to the symmetries of the problem, the transport coefficients D_{xy} and D_{yy} provide all the information on the physical mechanisms involved in the mutual diffusion of tracer particles under steady Couette flow. While the diagonal element can be interpreted as a generalization of the usual mutual diffusion coefficient (as it *couples* the yth component of the mass flux with the yth component of the concentration gradient), the off-diagonal element takes into account the cross couplings generated by the presence of the Couette flow. For small shear rates, $D_{yy} \approx 1$ and $D_{xy} \approx -\omega^{-2}\sqrt{2/(1+\mu)}[(1+2\mu)/\mu]a$. From a physical point of view, one expects that the action of the shear does not alter the qualitative behavior of the mass transport in the sense that $D_{yy} > 0$ and $D_{xy} < 0$. Our results confirm the above predictions. At a quantitative level, we observe that the net consequence of the presence of the Couette flow on the mass transport is to inhibit the diffusion of the tracer species along the y direction. In general, this inhibition is more significant as the mass ratio differs from 1. With respect to the x direction, $-D_{xy}$ is not a monotonic function of the shear rate and its absolute value increases as the mass of the tracer particle increases at a given value of a. Furthermore, we have been able to explicitly get the velocity distribution function $f_1^{(1)}$. It is given in terms of a complicated operator acting on the reference distribution $f_1^{(0)}$. The velocity distribution $f_1^{(1)}$ is a linear function of the concentration gradient but it presents a highly nonlinear dependence on both the shear rate and the thermal gradient as well as on the mass and size ratios.

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APPENDIX: EXPLICIT EXPRESSIONS OF THE TRACER DIFFUSION TENSOR AND THE PARTIAL PRESSURE

In this appendix we derive the expressions for $\mathbf{j}_1^{(1)}$ and $p_1^{(1)}$. Introducing the operator $\partial_s \equiv (1/\nu_{12})\partial/\partial y$, Eq. (36) can be rewritten as

$$(1+v_y\partial_s)f_1^{(1)} = f_{12}^{(1)} \ . \tag{A1}$$

Direct integration of this equation implies that $\partial_s j_{1,y}^{(1)} = 0$. Further, we assume, to be verified later, that $\partial_s^2 x_1(T_{12}^{(1)}/T_{12}^{(0)}) = 0$. In order to get the moments of $f_1^{(1)}$, it is more convenient to consider the formal solution to Eq. (A1) given by

$$f_1^{(1)} = (1+v_y\partial_s)^{-1} f_{12}^{(1)} = \sum_{k=0}^{\infty}(-v_y)^k\partial_s^k f_{12}^{(1)} \ . \tag{A2}$$

Let us start with the calculation of the components of the mass flux vector. The x component is given by

$$\begin{aligned} j_{1,x}^{(1)} &= \int d\mathbf{v}\, V_x f_1^{(1)} \\ &= \sum_{k=0}^{\infty}(-\partial_s)^k \int d\mathbf{v}\, v_y^k[V_x - \widetilde{a}(k+1)v_y]f_{12}^{(1)} \\ &= \frac{j_{1,x}^{(1)}}{1+\mu}\left[1+\sum_{k=1}^{\infty}(2k-1)!!\partial_s^{2k}(k_BT_{12}^{(0)}/m_1)^k\right] \\ &\quad +2\widetilde{a}\frac{p_{12}^{(0)}}{m_1}\sum_{k=0}^{\infty}(k+1)(2k+1)!!\partial_s^{2k+1}x_1(k_BT_{12}^{(0)}/m_1)^k - \frac{\widetilde{a}}{1+\mu}j_{1,y}^{(1)}\sum_{k=0}^{\infty}(2k+1)(2k+1)!!\partial_s^{2k}(k_BT_{12}^{(0)}/m_1)^k \\ &\quad +2\widetilde{a}\frac{p_{12}^{(0)}}{m_1}\sum_{k=0}^{\infty}(k+1)^2(2k+1)!!\partial_s^{2k+1}x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)(k_BT_{12}^{(0)}/m_1)^k \ , \end{aligned} \tag{A3}$$

where use has been made of Eqs. (27) and (28). Now, taking into account Eq. (29) and retaining terms up to first order in $\partial_s x_1$, one gets

$$j_{1,x}^{(1)} = \frac{j_{1,x}^{(1)}}{1+\mu}\left[1+\sum_{k=1}^{\infty}(2k)!(2k-1)!!(-\widetilde{\gamma})^k\right] + 2\widetilde{a}\frac{p_{12}^{(0)}}{m_1}\partial_s x_1 \sum_{k=0}^{\infty}(k+1)(2k+1)!(2k+1)!!(-\widetilde{\gamma})^k$$
$$-\frac{\widetilde{a}}{1+\mu}j_{1,y}^{(1)}\sum_{k=0}^{\infty}(2k+1)!(2k+1)!!(-\widetilde{\gamma})^k + \frac{p_{12}^{(0)}}{m_1}\widetilde{a}\partial_s\left[x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)\right]\sum_{k=0}^{\infty}(k+1)(2k+2)!(2k+1)!!(-\widetilde{\gamma})^k\,. \quad \text{(A4)}$$

All these summations can be expressed in terms of the functions $F_s(\widetilde{\gamma})$ defined in Sec. II and whose asymptotic series expansion is [8]

$$F_s(\widetilde{\gamma}) = \sum_{k=0}^{\infty}(k+1)^s(2k+1)!(2k+1)!!(-\widetilde{\gamma})^k\,. \quad \text{(A5)}$$

Therefore, Eq. (A4) can be rewritten as

$$j_{1,x}^{(1)} = \frac{j_{1,x}^{(1)}}{1+\mu}(1-2\widetilde{\gamma}\widetilde{F}_1) + 2\frac{p_{12}^{(0)}}{m_1}\widetilde{a}\widetilde{F}_1\partial_s x_1 - \frac{\widetilde{a}}{1+\mu}\widetilde{F}_0 j_{1,y}^{(1)} + 2\frac{p_{12}^{(0)}}{m_1}\widetilde{a}\widetilde{F}_2\partial_s\left[x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)\right]\,, \quad \text{(A6)}$$

where, for the sake of brevity, we have called $\widetilde{F}_s \equiv F_s(\widetilde{\gamma})$.

The y component of the mass flux can be obtained in a similar way. Following identical steps, one has that

$$j_{1,y}^{(1)} = \sum_{k=0}^{\infty}(-\partial_s)^k\int d\mathbf{v}v_y^{k+1}f_{12}^{(1)}$$
$$= -\frac{p_{12}^{(0)}}{m_1}\sum_{k=0}^{\infty}(2k+1)!!\partial_s^{2k+1}x_1(k_BT_{12}^{(0)}/m_1)^k + \frac{j_{1,y}^{(1)}}{1+\mu}\sum_{k=0}^{\infty}(2k+1)!!\partial_s^{2k}(k_BT_{12}^{(0)}/m_1)^k$$
$$-\frac{p_{12}^{(0)}}{m_1}\sum_{k=0}^{\infty}(k+1)(2k+1)!!\partial_s^{2k+1}x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)(k_BT_{12}^{(0)}/m_1)^k\,. \quad \text{(A7)}$$

Now, collecting all the terms up to first order in $\partial_s x_1$ one obtains

$$j_{1,y}^{(1)} = -\frac{p_{12}^{(0)}}{m_1}\partial_s x_1\sum_{k=0}^{\infty}(2k+1)!(2k+1)!!(-\widetilde{\gamma})^k + \frac{j_{1,y}^{(1)}}{1+\mu}\sum_{k=0}^{\infty}(2k)!(2k+1)!!(-\widetilde{\gamma})^k$$
$$-\frac{p_{12}^{(0)}}{2m_1}\partial_s\left[x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)\right]\sum_{k=0}^{\infty}(2k+2)!(2k+1)!!(-\widetilde{\gamma})^k$$
$$= -\frac{p_{12}^{(0)}}{m_1}\widetilde{F}_0\partial_s x_1 + \frac{j_{1,y}^{(1)}}{1+\mu}[1-2\widetilde{\gamma}(\widetilde{F}_1+2\widetilde{F}_2)] - \frac{p_{12}^{(0)}}{m_1}\widetilde{F}_1\partial_s\left[x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)\right]\,. \quad \text{(A8)}$$

Since $\partial_s j_{1,y}^{(1)} = 0$, Eqs. (A6) and (A8) imply that $\partial_s^2 x_1(T_{12}^{(1)}/T_{12}^{(0)}) = 0$ and $\partial_s j_{1,x}^{(1)} = 0$.

Let us consider now the partial pressure. It is defined as

$$p_1^{(1)} = \frac{1}{3}m_1\sum_{k=0}^{\infty}\int d\mathbf{v}\,V^2(-\partial_s)^k v_y^k f_{12}^{(1)}$$
$$= \frac{1}{3}m_1\sum_{k=0}^{\infty}(-\partial_s)^k\int d\mathbf{v}v_y^k[V^2 - 2\widetilde{a}(k+1)V_xv_y + \widetilde{a}^2(k+2)(k+1)v_y^2]f_{12}^{(1)}\,, \quad \text{(A9)}$$

where in the last step we have taken into account that $\partial_s V^2 = -2\widetilde{a}V_x$ and $\partial_s^2 V^2 = 2\widetilde{a}^2$. Now we are going to evaluate each separate term on the right-hand side of Eq. (A9). For that, we will follow again steps similar to those made in the problem of mechanically equivalent particles [10]. The first term is

$$\begin{aligned}\sum_{k=0}^{\infty}(-\partial_s)^k\int d\mathbf{v}\,V^2v_y^kf_{12}^{(1)} &= \frac{p_{12}^{(0)}}{m_1}\sum_{k=0}^{\infty}\frac{(2k+3)!!}{(2k+1)}\partial_s^{2k}x_1(k_BT_{12}^{(0)}/m_1)^k-\frac{j_{1,y}^{(1)}}{1+\mu}\sum_{k=0}^{\infty}\frac{(2k+5)!!}{2k+3}\partial_s^{2k+1}(k_BT_{12}^{(0)}/m_1)^{k+1}\\ &\quad+\frac{p_{12}^{(0)}}{m_1}\sum_{k=0}^{\infty}(k+1)\frac{(2k+3)!!}{2k+1}\partial_s^{2k}x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)(k_BT_{12}^{(0)}/m_1)^k\\ &= x_1\frac{p_{12}^{(0)}}{m_1}\sum_{k=0}^{\infty}(2k)!\frac{(2k+3)!!}{2k+1}(-\widetilde{\gamma})^k\\ &\quad+\frac{k_Bp_{12}^{(0)}}{2m_1^2}(\partial_sx_1)(\partial_sT_{12}^{(0)})\sum_{k=0}^{\infty}(2k+5)(2k+2)(2k+2)!(2k+1)!!(-\widetilde{\gamma})^k\\ &\quad-\frac{k_B}{2m_1}\frac{j_{1,y}^{(1)}}{1+\mu}\partial_sT_{12}^{(0)}\sum_{k=0}^{\infty}(2k+5)(2k+2)!(2k+1)!!(-\widetilde{\gamma})^k\\ &\quad+x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)\frac{p_{12}^{(0)}}{m_1}\sum_{k=0}^{\infty}\frac{k+1}{2k+1}(2k)!(2k+3)!!(-\widetilde{\gamma})^k\\ &\quad+\partial_s\left[x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)\right]\frac{k_Bp_{12}^{(0)}}{2m_1^2}\partial_sT_{12}^{(0)}\sum_{k=0}^{\infty}(2k+5)(2k+4)(k+1)\\ &\quad\times(2k+2)!(2k+1)!!(-\widetilde{\gamma})^k\,, \end{aligned}\tag{A10}$$

where use has been made of the identity

$$\partial_s^{2k+1}{T_{12}^{(0)}}^{k+1}=(k+1)(2k+1)(\partial_sT_{12}^{(0)})\partial_s^{2k}{T_{12}^{(0)}}^{k}\;.\tag{A11}$$

The second term is

$$\begin{aligned}\sum_{k=0}^{\infty}(k+1)(-\partial_s)^k\int d\mathbf{v}\,V_xv_y^{k+1}f_{12}^{(1)} &= -2\frac{j_{1,x}^{(1)}}{1+\mu}\sum_{k=0}^{\infty}(k+1)(2k+1)!!(\partial_s)^{2k+1}(k_BT_{12}^{(0)}/m_1)^{k+1}\\ &= -\frac{j_{1,x}^{(1)}}{1+\mu}\frac{k_B}{m_1}\partial_sT_{12}^{(0)}\sum_{k=0}^{\infty}(k+1)(2k+2)!(2k+1)!!(-\widetilde{\gamma})^k\;.\end{aligned}\tag{A12}$$

The third term is

$$\begin{aligned}\sum_{k=0}^{\infty}(k+2)(k+1)(-\partial_s)^k\int d\mathbf{v}\,v_y^{k+2}f_{12}^{(1)} &= \frac{p_{12}^{(0)}}{m_1}\sum_{k=0}^{\infty}(2k+2)(2k+1)(2k+1)!!\partial_s^{2k}x_1(k_BT_{12}^{(0)}/m_1)^k\\ &\quad-\frac{j_{1,y}^{(1)}}{1+\mu}\sum_{k=0}^{\infty}(2k+3)(2k+2)(2k+3)!!\partial_s^{2k+1}(k_BT_{12}^{(0)}/m_1)^{k+1}\\ &\quad+2\frac{p_{12}^{(0)}}{m_1}\sum_{k=0}^{\infty}(k+1)^2(2k+1)(2k+1)!!\partial_s^{2k}x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)(k_BT_{12}^{(0)}/m_1)^k\\ &= x_1\frac{p_{12}^{(0)}}{m_1}\sum_{k=0}^{\infty}(2k+2)!(2k+1)!!(-\widetilde{\gamma})^k\\ &\quad+\frac{k_Bp_{12}^{(0)}}{m_1^2}(\partial_sx_1)(\partial_sT_{12}^{(0)})\sum_{k=0}^{\infty}(k+1)(2k+4)!(2k+3)!!(-\widetilde{\gamma})^k\\ &\quad-\frac{k_B}{m_1}\frac{j_{1,y}^{(1)}}{1+\mu}\partial_sT_{12}^{(0)}\sum_{k=0}^{\infty}(k+1)(2k+3)!(2k+3)!!(-\widetilde{\gamma})^k\\ &\quad+\frac{p_{12}^{(0)}}{m_1}x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)\sum_{k=0}^{\infty}(k+1)(2k+2)!(2k+1)!!(-\widetilde{\gamma})^k\\ &\quad+\frac{k_Bp_{12}^{(0)}}{m_1^2}\partial_s\left[x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)\right]\partial_sT_{12}^{(0)}\sum_{k=0}^{\infty}(k+1)(k+2)\\ &\quad\times(2k+4)!(2k+3)!!(-\widetilde{\gamma})^k\;.\end{aligned}\tag{A13}$$

By substituting Eqs. (A10), (A12), and (A13) into Eq. (A9) and taking into account the relation (40) between $p_1^{(1)}$ and $T_{12}^{(1)}$, one arrives at

$$\begin{aligned}\frac{1}{1-2M}x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right) &= \frac{k_B}{m_1}(\partial_s T_{12}^{(0)})(\partial_s x_1)C_1 - 2\frac{k_B}{p_{12}^{(0)}}\frac{1}{1+\mu}\partial_s T_{12}^{(0)} j_{1,y}^{(1)} C_2 + 2x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)C_3 \\ &\quad +2\frac{k_B}{m_1}(\partial_s T_{12}^{(0)})\partial_s\left[x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)\right]C_4 + 2\frac{k_B}{p_{12}^{(0)}}\frac{1}{1+\mu}\partial_s T_{12}^{(0)}\frac{j_{1,x}^{(1)}}{\widetilde{a}}C_5 \ , \end{aligned} \tag{A14}$$

where the coefficients C_i are given by

$$C_1 = \frac{1}{6\widetilde{F}_1\widetilde{\gamma}}[2\widetilde{\gamma}(5\widetilde{F}_1^2 - 4\widetilde{F}_2^2) - \widetilde{F}_1(\widetilde{F}_0 - 1)] - \frac{2}{\widetilde{F}_1\widetilde{\gamma}}(\widetilde{F}_1 - \widetilde{F}_2)\beta \ , \tag{A15}$$

$$C_2 = \frac{\widetilde{F}_0}{6\widetilde{F}_1}(3\widetilde{F}_1 + 2\widetilde{F}_2) - \frac{2}{\widetilde{F}_1\widetilde{\gamma}}(\widetilde{F}_0 - \widetilde{F}_1)\beta \ , \tag{A16}$$

$$C_3 = \frac{1}{12}(\widetilde{F}_0 + 5) - \frac{\widetilde{\gamma}}{6\widetilde{F}_1}[6\widetilde{F}_0\widetilde{F}_1 - (\widetilde{F}_1^2 + 4\widetilde{F}_2^2)] - \frac{\widetilde{F}_2}{\widetilde{F}_1}\beta \ , \tag{A17}$$

$$C_4 = \frac{1}{12\widetilde{\gamma}\widetilde{F}_1}[8\widetilde{\gamma}\widetilde{F}_2(3\widetilde{F}_1 + 2\widetilde{F}_2) + \widetilde{F}_0(\widetilde{F}_1 + \widetilde{F}_2) - \widetilde{F}_1^2 - \widetilde{F}_2] - \frac{1}{8\widetilde{\gamma}^2\widetilde{F}_1}(\widetilde{F}_0 + 2\widetilde{F}_1\widetilde{\gamma} + 16\widetilde{F}_2\widetilde{\gamma} - 1)\beta \ , \tag{A18}$$

$$C_5 = \frac{2}{3}\frac{\widetilde{F}_2}{\widetilde{F}_1}\widetilde{\gamma}(2\widetilde{F}_2 + 3\widetilde{F}_1) - 2\frac{\widetilde{F}_2}{\widetilde{F}_1}\beta \ , \tag{A19}$$

with

$$\beta = \frac{M(1-\chi)}{\chi + 2M(1-\chi)} \ . \tag{A20}$$

Notice that in deriving Eq. (A14) use has been made of Eq. (32). Equations (A6), (A8), and (A14) constitute a set of three coupled equations. By applying the operator ∂_s on both sides of Eq. (A14), one gets

$$\begin{aligned}\frac{1}{1-2M}\partial_s x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right) &= -2\widetilde{\gamma}C_1\partial_s x_1 + \frac{m_1}{p_{12}^{(0)}}\frac{4}{1+\mu}\widetilde{\gamma}C_2 j_{1,y}^{(1)} + 2C_3\partial_s\left[x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)\right] \\ &\quad -4\widetilde{\gamma}C_4\partial_s\left[x_1\left(\frac{T_{12}^{(1)}}{T_{12}^{(0)}}-1\right)\right] - \frac{m_1}{p_{12}^{(0)}}\frac{4}{1+\mu}C_5\widetilde{\gamma}\frac{j_{1,x}^{(1)}}{\widetilde{a}} \ . \end{aligned} \tag{A21}$$

From Eqs. (A6), (A8) and (A21), one can obtain the explicit expression of the mass flux. It may be written in the form of a generalized Fick law Eq. (43), where the relevant elements of the tracer diffusion tensor are given by

$$D_{yy} = [\chi + 2M(1-\chi)]\mu\frac{N_1}{N_2} \ , \tag{A22}$$

$$D_{xy} = [\chi + 2M(1-\chi)]\mu\frac{N_3}{N_2}\widetilde{a} \ , \tag{A23}$$

where

$$\begin{aligned}N_1 &= 8C_5\widetilde{\gamma}\lambda(\widetilde{F}_0\widetilde{F}_2 - \widetilde{F}_1^2) \\ &\quad -(2\widetilde{F}_1\widetilde{\gamma} + \mu)[2C_1\widetilde{F}_1\widetilde{\gamma}\lambda + \widetilde{F}_0(2C_3\lambda - 4C_4\widetilde{\gamma}\lambda + 1)] \ , \end{aligned} \tag{A24}$$

$$\begin{aligned}N_2 &= (2\widetilde{F}_1\widetilde{\gamma} + \mu)[4C_2\widetilde{F}_1\widetilde{\gamma}\lambda - (2\widetilde{F}_1\widetilde{\gamma} + 4\widetilde{F}_2\widetilde{\gamma} + \mu) \\ &\quad \times(2C_3\lambda - 4C_4\widetilde{\gamma}\lambda + 1)] \\ &\quad +4C_5\widetilde{\gamma}\lambda[\widetilde{F}_0\widetilde{F}_1 + 2\widetilde{F}_2(2\widetilde{F}_1\widetilde{\gamma} + 4\widetilde{F}_2\widetilde{\gamma} + \mu)] \ , \end{aligned} \tag{A25}$$

$$\begin{aligned}N_3 &= 8C_2\widetilde{\gamma}\lambda(\widetilde{F}_0\widetilde{F}_2 - \widetilde{F}_1^2) + 2(2\widetilde{F}_1\widetilde{\gamma} + 4\widetilde{F}_2\widetilde{\gamma} + \mu) \\ &\quad \times[2C_1\widetilde{F}_2\widetilde{\gamma}\lambda + \widetilde{F}_1(2C_3\lambda - 4C_4\widetilde{\gamma}\lambda + 1)] \\ &\quad +2C_1\widetilde{\gamma}\lambda\widetilde{F}_0\widetilde{F}_1 + \widetilde{F}_0^2(2C_3\lambda - 4C_4\widetilde{\gamma}\lambda + 1) \ . \end{aligned} \tag{A26}$$

Here $\lambda \equiv 2M - 1 = -(1+\mu^2)/(1+\mu)^2$. In the limit of small shear rates, one has that

$$D_{yy} \approx 1 - \frac{2}{5}\frac{5\mu^2 + 18\mu + 11}{\omega^4(1+\mu)}a^2 , \tag{A27}$$

$$D_{xy} \approx -\frac{1}{\omega^2}\left(\frac{2}{1+\mu}\right)^{1/2}\frac{1+2\mu}{\mu}a . \tag{A28}$$

From the knowledge of the mass flux, the partial pressure $p_1^{(1)}$ can be obtained. It can be written in the form of Eq. (44) where Ω is given by

$$\Omega = -\frac{6}{5}[\chi + 2M(1-\chi)]^2 \frac{\mu^2}{(1+\mu)^2(1+2\mu)}\frac{N_4}{N_2} , \tag{A29}$$

with

$$N_4 = (2\widetilde{F}_1\widetilde{\gamma} + \mu)[2C_2\widetilde{F}_0 + C_1(2\widetilde{F}_1\widetilde{\gamma} + 4\widetilde{F}_2\widetilde{\gamma} + \mu)] \\ +2C_5[\widetilde{F}_0^2 + 2\widetilde{F}_1(2\widetilde{F}_1\widetilde{\gamma} + 4\widetilde{F}_2\widetilde{\gamma} + \mu)] . \tag{A30}$$

The first terms in the power expansion of Ω are

$$\Omega \approx 1 - \frac{4}{75}\frac{200\mu^4 + 1562\mu^3 + 553\mu^2 - 505\mu - 55}{\omega^4\mu(1+\mu)(1+2\mu)}a^2 . \tag{A31}$$

Consequently, Ω monotonically decreases as the shear rate increases if $\mu \gtrsim 0.468$.

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