Self-diffusion in a dilute gas under heat and momentum transport

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Diffusion of tagged particles in a dilute gas in a stationary state with temperature and velocity gradients (planar Couette flow) is analyzed. The results are obtained from a kinetic model for a multicomponent gas constructed in the same spirit as in the Bhatnagar-Gross-Krook equation for a single gas. The kinetic equation for the velocity distribution function of the tagged particles is solved by using a Chapman-Enskog-like expansion around the steady-Couette-flow state. Up to first order in the concentration gradient of tagged particles, explicit expressions for the self-diffusion coefficient and the partial pressure have been derived. Both quantities are nonlinear functions of the shear rate. In addition, the distribution function of tagged particles is explicitly obtained. It exhibits a highly nonlinear dependence on both the shear rate and the thermal gradient. In the absence of shear rate, self-diffusion in pure heat flow is also considered with more detail.

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I. INTRODUCTION

The analysis of diffusion in dilute binary gas mixtures has been a subject of interest for many years. For states near equilibrium, the Chapman-Enskog [1] method provides an adequate way to solve the Boltzmann equations [1,2] for a general interaction law. The constitutive equation for the mass flux is known (Fick's law) and the linear hydrodynamic equation can be solved for several boundary conditions. Nevertheless, the systematic expansion given by the Chapman-Enskog theory is very complicated and not very useful beyond the linear order. Consequently, much less is known about the description of the mass transport in systems far from equilibrium.

In order to gain some insight into this general problem, specific physical situations must be considered. In particular, the planar shear flow at uniform temperature and density [3] (usually referred to as "uniform shear flow") has provided an adequate framework to describe such nonlinear diffusion problems. One reason is that the Boltzmann equation admits a solution in the uniform-shear-flow state for Maxwell molecules. Thus, in the case of mechanically identical Maxwell molecules, Marchetti and Dufty [4] and Garzó, Santos, and Brey [5] have explicitly obtained an expression for the nonlinear shear-rate-dependent self-diffusion tensor. More recently, Garzó and López de Haro [6] have extended these results to the case of binary mixtures with arbitrary mass ratio in the tracer limit. Their expressions reduce to previous results obtained from a generalized Green-Kubo formula [7] in the Fokker-Planck limit [8]. More detailed information has been given for both situations (self-diffusion and tracer limit) using kinetic models [9] for the nonlinear Boltzmann equations. In the context of dense fluids, generalized Green-Kubo relations for diffusion [10] and thermal conductivity [11] under shear flow have recently been analyzed. In addition, Cummings *et al.* [12] have carried out molecular-dynamics simulations on a Lennard-Jones fluid in the non-Newtonian regime in order to analyze self-diffusion in the presence of shear flow. All these results refer to a system (dilute or dense) subject to mass and momentum transport.

The aim of this paper is to describe self-diffusion of a dilute gas in a steady state in the presence of heat and momentum transport (steady Couette flow). The physical situation of the system as a whole is that of a steady state of a gas enclosed between two parallel plates in relative motion and kept at different temperatures. In this problem there are two parameters measuring the departure from equilibrium: the shear rate and the thermal gradient. Due to the mathematical difficulties embodied in the steady-Couette-flow problem, no solution has been found to the Boltzmann equation. However, an exact description has been given when one uses the Bhatnagar-Gross-Krook (BGK) [13] kinetic model. From this equation, an exact normal solution has been obtained for arbitrary values of the velocity and temperature gradients [14,15]. Therefore, assuming that the system is in steady Couette flow, our goal is to describe a self-diffusion problem under these conditions. Self-diffusion involves only like-particle motion and is somewhat simpler to treat from a theoretical view as well as from nonequilibrium computer simulations [12]. In this paper the analysis will be achieved by using a kinetic model suitable to study transport properties for mixtures of similar masses [16]. This model is consistent with the BGK equation for the total distribution function when the particular case of mechanically identical components (self-diffusion case) is considered. This property is not verified by several kinetic models previously proposed [17].

The kinetic equation for the distribution function of the tagged particles is solved by a Chapman-Enkog-type of expansion around the Couette flow state. In this way, the different approximations will be highly nonlinear functions of the shear rate and the thermal gradient. In this paper we will restrict the calculations to first order in the gradient of the concentration of the tagged particles. We get a shear-rate-dependent self-diffusion coefficient

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and a generalized partial pressure. In addition, the use of a kinetic model allows us to explicitly obtain the velocity distribution function of the tagged particles. In absence of shear rate (pure heat flow), the self-diffusion coefficient reduces to the one given by the linear theory and all velocity moments are polynomials in the thermal gradient.

The plan of the paper is as follows. In Sec. II we present the kinetic model to be used in the paper. It is shown that it is consistent with the BGK equation for a single gas in the self-diffusion case and the parameters that define the collision term are explicitly written for a binary mixture of identical particles. Section III concerns with the description of the steady Couette flow at the level of the BGK equation. Section IV deals with the general problem of self-diffusion in Couette flow, while the particular case of pure heat flow is addressed in Sec. V. Finally, some concluding remarks are made in Sec. VI.

II. KINETIC MODEL FOR MULTICOMPONENT GASES

Let us consider an N-component dilute mixture. Let $f_1(\mathbf{r},\mathbf{v};t)$ be the one-particle velocity distribution function of species i. The corresponding local number density and mean velocity are defined, respectively, by

$$n_i=\int d\mathbf{v}\, f_i \ , \tag{2.1}$$

$$\mathbf{u}_i=\frac{1}{n_i}\int d\mathbf{v}\, \mathbf{v} f_i \ . \tag{2.2}$$

A local temperature T_i for species i can also be introduced through the relation

$$\tfrac{3}{2}n_i k_B T_i=\int d\mathbf{v}\tfrac{1}{2}m_i(\mathbf{v}-\mathbf{u}_i)^2 f_i \ , \tag{2.3}$$

where k_B is the Boltzmann constant and m_i is the mass of a particle of species i. The local velocity $\mathbf{u}$ and the local temperature T of the mixture as a whole are given by

$$\rho\mathbf{u}=\sum_i \rho_i \mathbf{u}_i \ , \tag{2.4}$$

$$\tfrac{3}{2}nk_B T=\sum_i \int \mathbf{d}\tfrac{1}{2}m_i(\mathbf{v}-\mathbf{u})^2 f_i = \sum_i [\tfrac{3}{2}n_i k_B T_i+\tfrac{1}{2}m_i n_i(\mathbf{u}_i-\mathbf{u})^2] \ , \tag{2.5}$$

where $\rho_i=m_i n_i$, $n=\sum_i n_i$, and $\rho=\sum_i \rho_i$.

In the low-density limit, the functions f_i, $i=1,2,\ldots,N$, satisfy a coupled set of N nonlinear Boltzmann equations [1]. A kinetic model retaining the main physical properties of the Boltzmann description has been recently proposed [16],

$$\frac{\partial}{\partial t}f_i+\mathbf{v}\cdot\nabla f_i=-\sum_{j=1}^{N}\zeta_{ij}(f_i-f_{ij}^R) \ , \tag{2.6}$$

where ζ_{ij} is an effective collision frequency of a particle of species i with particles of species j. Consequently, $n_i\zeta_{ij}=n_j\zeta_{ji}$. The reference distribution function f_{ij}^R reads

$$f_{ij}^R=n_i\left[\frac{m_i}{2\pi k_B T}\right]^{3/2}(1+A_{ij}+\mathbf{B}_{ij}\cdot\mathbf{V}+C_{ij}V^2) \times\exp\left[-\frac{m_i}{2k_B T}V^2\right] \ , \tag{2.7}$$

where $\mathbf{V}\equiv\mathbf{v}-\mathbf{u}$ is the peculiar velocity, and the parameters A_{ij}, $\mathbf{B}_{ij}$, and C_{ij} are given by

$$A_{ij}=-\frac{3k_B T}{m_i}C_{ij} \ , \tag{2.8}$$

$$\mathbf{B}_{ij}=\frac{m_i}{k_B T}(\mathbf{u}_{ij}-\mathbf{u}) \ , \tag{2.9}$$

$$C_{ij}=\frac{m_i}{2k_B T}\left[\frac{T_{ij}-T}{T}+\frac{m_i}{3k_B T}(\mathbf{u}_{ij}-\mathbf{u})^2\right] \ . \tag{2.10}$$

In Eqs. (2.9) and (2.10) we have introduced the quantities

$$\mathbf{u}_{ij}=(m_i\mathbf{u}_i+m_j\mathbf{u}_j)/(m_i+m_j) \ , \tag{2.11}$$

$$T_{ij}=T_i+2[m_i m_j/(m_i+m_j)^2] \times[(T_j-T_i)+(m_j/6k_B)(\mathbf{u}_i-\mathbf{u}_j)^2] \ . \tag{2.12}$$

The kinetic model described by Eqs. (2.6)–(2.12) has the following features: (i) preserves conservation of number of particles for each species, as well as of total momentum and total energy for the mixture, (ii) reproduces the collisional transfer of momentum and energy given by the Boltzmann equation for Maxwell molecules, and (iii) is compatible with the BGK kinetic model for a single gas when the case of mechanically indistinguishable particles is considered. Let us show the latter point with detail. If all the particles are mechanically equivalent, $m_i=m$ and $\zeta_{ij}=\zeta n_j/n$, ζ being the collision frequency of the system. Consequently,

$$\mathbf{u}_{ij}=(\mathbf{u}_i+\mathbf{u}_j)/2 \ , \tag{2.13}$$

$$T_{ij}=\tfrac{1}{2}(T_i+T_j)+\frac{m}{12k_B}(\mathbf{u}_i-\mathbf{u}_j)^2 \ , \tag{2.14}$$

and it is easy to prove that

$$\sum_i\sum_j n_i\zeta_{ij}A_{ij}=\sum_i\sum_j n_i\zeta_{ij}\mathbf{B}_{ij}=\sum_i\sum_j n_i\zeta_{ij}C_{ij}=0 \ . \tag{2.15}$$

Therefore, by adding the kinetic equations (2.6) for all species i one gets

$$\frac{\partial}{\partial t}f+\mathbf{v}\cdot\nabla f=-\zeta(f-f^{\mathrm{LE}}) \ , \tag{2.16}$$

where $f=\sum_i f_i$ is the distribution function of the whole mixture and f^{LE} is the local equilibrium distribution function

$$f^{\mathrm{LE}}=n(m/2\pi k_B T)^{3/2}\exp(-mV^2/2k_B T) \ . \tag{2.17}$$

Equation (2.16) is the usual BGK model kinetic equation [13] of the Boltzmann equation for a single gas. Thus, our model (2.6) can be seen as a consistent extension of

the BGK equation to a multicomponent system. In fact, the physical idea behind Eq. (2.6) is that the main global effect of the collisions on particles of species i is to produce a tendency towards the local equilibrium state characterized by the hydrodynamic fields of the whole mixture. The details of the collisions with particles of species j give rise to a correction through the parameters A_{ij}, $\mathbf{B}_{ij}$, and C_{ij}. According to this spirit, although the model is not *a priori* restricted to any range of mass ratio, one expects it to be more suitable in the case of species with similar masses.

In this work, we deal with a binary mixture of identical particles. The only distinction between species 1 and 2 is a tag or "label" with no influence on the mechanical behavior of the particles. This is the appropriate framework to study self-diffusion. In this case, the total distribution function $f=f_1+f_2$ obeys the BGK equation (2.16), while f_1 verifies Eq. (2.6)

$$\frac{\partial}{\partial t}f_1+\mathbf{v}\cdot\nabla f_1=-\zeta_{11}(f_1-f_{11}^R)-\zeta_{12}(f_1-f_{12}^R)$$
$$=-\zeta(f_1-\Phi_1)\ , \tag{2.18}$$

where

$$\Phi_1=x_1f_{11}^R+x_2f_{12}^R$$
$$=x_1f^{\mathrm{LE}}\left[1+\frac{m}{2x_1p}\mathbf{V}\cdot\mathbf{J}_i+\frac{1}{2}\left[\frac{mV^2}{2k_BT}-\frac{3}{2}\right]\left[\frac{p_1}{x_1p}-1\right]\right]\ . \tag{2.19}$$

In Eq. (2.19), $x_i=n_i/n$ is the molar fraction,

$$\mathbf{J}_i=n_i(\mathbf{u}_i-\mathbf{u})$$
$$=\int d\mathbf{v}\mathbf{V}f_i \tag{2.20}$$

is the flux of particles, and

$$p_i=n_ik_BT_i+\tfrac{1}{3}mn_i(\mathbf{u}_i-\mathbf{u})^2$$
$$=\tfrac{1}{3}m\int d\mathbf{v}\,V^2f_i \tag{2.21}$$

is the partial pressure of species i, while $p=p_1+p_2=nk_BT$ is the pressure of the mixture. It is worth remarking that Eq. (2.18) holds for any relative number of tagged particles. On the other hand, Eq. (2.18) is not a closed equation since it depends on f through its moments n, $\mathbf{u}$, and T. Once Eq. (2.16) is solved, Eq. (2.18) becomes a linear equation for f_1.

III. STEADY COUETTE FLOW

We consider a dilute gas enclosed between two infinite parallel walls in relative motion and, in general, at different temperatures. Let the x axis be parallel to the direction of motion and the y axis be orthogonal to the walls. A nonequilibrium steady normal state exists if one increases the velocity difference and/or the temperature difference at the walls as the system size increases. In this way, the Knudsen number vanishes near the walls and the boundary layers are made asymptotically small.

If the velocity distribution function f is assumed to obey the BGK model, Eq. (2.16), its normal solution is consistent with the following profiles for the hydrodynamic fields [14]:

$$\frac{\partial}{\partial y}p=0\ , \tag{3.1}$$

$$\frac{1}{\zeta(y)}\frac{\partial}{\partial y}u_x=a=\text{const}\ , \tag{3.2}$$

$$\left[\frac{1}{\zeta(y)}\frac{\partial}{\partial y}\right]^2T=-\frac{2m}{k_B}\gamma(a)=\text{const}\ . \tag{3.3}$$

The reduced shear rate a appearing in Eq. (3.2) is one of the uniformity parameters of the system. It measures the departure from equilibrium as the mean free path relative to the spatial variation of the flow velocity u_x. There exists another independent uniformity parameter related to the spatial variation of the temperature:

$$\epsilon(y)=\frac{1}{\zeta(y)}\left[\frac{2k_BT}{m}\right]^{1/2}\frac{1}{T}\frac{\partial T}{\partial y}\ . \tag{3.4}$$

In contrast to a, ϵ depends on the space variable y. The dimensionless parameter $\gamma(a)$ appearing in Eq. (3.3) is a nonlinear function of the shear rate a given implicitly by the equation

$$a^2=\gamma\frac{2F_2(\gamma)+3F_1(\gamma)}{F_1(\gamma)}\ , \tag{3.5}$$

where

$$F_r(\gamma)=\left[\frac{d}{d\gamma}\gamma\right]^rF_0(\gamma)$$

and

$$F_0(\gamma)=\frac{2}{\gamma}\int_0^\infty dt\,t\exp(-t^2/2)K_0(2\gamma^{-1/4}t^{1/2})\ , \tag{3.6}$$

K_0 being the zeroth-order modified Bessel function [18].

The explicit expression for the distribution function is [15]

$$f(\mathbf{r},\mathbf{v})=n(m/2\pi k_BT)^{3/2}\frac{2\alpha(1+\alpha)}{\epsilon|\xi_y|}^{3/2}$$
$$\times\int_{t_0}^{t_1}dt[2t-(1-\alpha)t^2]^{-5/2}\exp\left\{-\frac{2\alpha}{1+\alpha}\frac{1-t}{\epsilon\xi_y}-\frac{1+\alpha}{2t-(1-\alpha)t^2}\left[\left[\xi_x+\frac{2a\alpha}{1+\alpha}\frac{1-t}{\epsilon}\right]^2+\xi_y^2+\xi_z^2\right]\right\}\ , \tag{3.7}$$

where $(t_0,t_1)=(0,1)$ if $\xi_y>0$ and $(t_0,t_1)=(1,2/(1-\alpha))$ if $\xi_y<0$. In Eq. (3.7),

$$\alpha(a,\epsilon)\equiv\frac{\epsilon}{(\epsilon^2+8\gamma)^{1/2}}\ , \tag{3.8}$$

$$\xi\equiv(m/2k_BT)^{1/2}\mathbf{V}\ . \tag{3.9}$$

From Eq. (3.7), fluxes such as the pressure tensor

$$P_{ij}=\int d\mathbf{v}\, mV_iV_jf \tag{3.10}$$

and the heat flux

$$\mathbf{q}=\int d\mathbf{v}\frac{m}{2}V^2\mathbf{V}f \tag{3.11}$$

can be obtained. The results are (see Appendix A)

$$P_{xx}=p[1+4\gamma(F_1+F_2)]\ , \tag{3.12}$$

$$P_{yy}=p[1-2\gamma(F_1+2F_2)]\ , \tag{3.13}$$

$$P_{zz}=[1-2\gamma F_1]\ , \tag{3.14}$$

$$P_{xz}=P_{yz}=0\ , \tag{3.15}$$

$$P_{xy}=-\frac{p}{\zeta}F_0(\gamma)\frac{\partial u_x}{\partial y}\ , \tag{3.16}$$

$$\mathbf{q}=-\frac{pk_B}{2m\zeta}\frac{a^2}{\gamma}F_0(\gamma)\nabla T\ . \tag{3.17}$$

It is worth mentioning that Eq. (3.7) includes as a particular case the pure heat flow (Fourier flow), where both walls are at rest and no flow velocity exists [19]. This case is obtained by taking the limit $a\to 0$ (which implies $\gamma\to 0$ and $\alpha\to 1$) with $\epsilon>0$ fixed:

$$f(\mathbf{r},\mathbf{v})=n(m/2\pi k_BT)^{3/2}\frac{1}{\epsilon|\xi_y|}\times\int_{t_0}^{t_1}dt\, t^{-5/2}\exp\left[-\frac{1-t}{\epsilon\xi_y}-\frac{\xi^2}{t}\right]\ , \tag{3.18}$$

where $(t_0,t_1)=(0,1)$ if $\xi_y>0$ and $(t_0,t_1)=(1,\infty)$ if $\xi_y<0$. Also, Eq. (3.3) reduces to

$$\frac{1}{\zeta(y)}\frac{\partial}{\partial y}T=\text{const} \tag{3.19}$$

and Eq. (3.17) coincides with the Fourier law [19].

IV. SELF-DIFFUSION IN STEADY COUETTE FLOW

Let us consider a dilute gas that is arbitrarily far from equilibrium in the state described in the previous section. Although all the particles are mechanically equivalent, we assume that some of them are distinguished by a certain tag or label and belong to the same species 1. The physical problem we are interested in is the diffusion of the tagged particles when the system as a whole is in steady Couette flow.

We shall suppose that the temporal evolution of the velocity distribution function f_1 of the tagged particles is governed by Eq. (2.18), according to the model described in Sec. II. In order to solve Eq. (2.18), we shall follow a perturbation scheme in the same spirit as in the Chapman-Enskog method. Assuming that the molar fraction $x_1(\mathbf{r},t)$ of tagged particles is only slightly nonuniform, we perform an expansion taking the gradient ∇x_1 as the perturbation parameter. The main difference from the usual Chapman-Enskog expansion [1] is that the reference zeroth-order state is not that of local equilibrium, but the one corresponding to the steady Couette flow. In this way, the successive approximations will be highly nonlinear functions of the uniformity parameters a and ϵ. Here, we restrict ourselves to first order in ∇x_1. This kind of expansion has been previously used in other problems [5,9].

Following the usual techniques described by the Chapman-Enskog method, the operator $\partial/\partial t$ must be consistently computed at each order according to the hydrodynamic balance equations. In our case, the balance equation for the concentration of the tagged particles reads

$$\frac{\partial}{\partial t}x_1=-\frac{1}{n}\nabla\cdot\mathbf{J}_1-\mathbf{u}\cdot\nabla x_1\ , \tag{4.1}$$

where we have taken into account that in the Couette flow $\nabla\cdot\mathbf{u}=0$ and $\mathbf{u}\cdot\nabla n=0$. Since we are interested in a steady state, now we particularize to a concentration gradient parallel to the temperature and flow velocity gradients, i.e., $\nabla x_1=(\partial x_1/\partial y)\hat{\mathbf{y}}$. Thus, the last term in the right-hand side of Eq. (4.1) vanishes. Further, up to the first order in the concentration gradient, $\partial x_1/\partial t=0$ since $\mathbf{J}_1=0$ at the zeroth order (generalized Euler order). Consequently, Eq. (2.18) becomes

$$v_y\frac{\partial}{\partial y}f_1^{(1)}=-\zeta(f_1^{(1)}-\Phi_1^{(1)})\ , \tag{4.2}$$

where $f_1^{(1)}$ denotes the function f_1 up to the first-order approximation, i.e., $f_1=f_1^{(1)}+O(\nabla^2x_1)$. In Eq. (4.2),

$$\Phi_1^{(1)}=x_1f^{\mathrm{LE}}+\left[\frac{m}{2p}v_yJ_1^{(1)}+\frac{1}{2}\left[\frac{mV^2}{2k_BT}-\frac{3}{2}\right]\left[\frac{p_1^{(1)}}{p}-x\right]\right]\times f^{\mathrm{LE}}\ , \tag{4.3}$$

where

$$J_1^{(1)}=\int d\mathbf{v}\, v_yf_1^{(1)}\ , \tag{4.4}$$

$$p_1^{(1)}=\tfrac{1}{3}\int d\mathbf{v}\, mV^2f_1^{(1)}\ . \tag{4.5}$$

The formal solution of Eq. (4.2) is

$$f_1^{(1)}=\left[1+v_y\frac{\partial}{\partial s}\right]^{-1}\Phi_1^{(1)}\ , \tag{4.6}$$

where $ds\equiv\zeta(y)dy$. It is worth remarking that when the operator $(1+v_y\partial/\partial s)^{-1}$ acts on the function $\Phi_1^{(1)}$, one must take into account that only terms up to first order in the concentration gradient need to be retained. The solution (4.6) is still formal, not only because of the presence of the operator $(1+v_y\partial/\partial s)^{-1}$, but also because $\Phi_1^{(1)}$ de-

pends on $f_1^{(1)}$ through its moments $J_1^{(1)}$ and $p_1^{(1)}$. The calculation of $J_1^{(1)}$ and $p_1^{(1)}$ is rather involved and is made in Appendix B. The result is

$$J_1^{(1)} = -\frac{2p}{m} D(a) \frac{\partial x_1}{\partial s} , \tag{4.7}$$

$$p_1^{(1)} = p \left[x_1 + 10\Omega(a)\epsilon(k_B T/2m)^{1/2} \frac{\partial x_1}{\partial s} \right] , \tag{4.8}$$

where the functions $D(a)$ and $\Omega(a)$ are given by Eqs. (B32) and (B35), respectively. Equations (4.7) and (4.8) have been derived keeping all the hydrodynamic orders in a and ϵ. Expression (4.7) is a generalization of the usual Fick's law (valid in the absence of shear rate and thermal gradient). It gives the mass transport of tagged particles when the concentration gradient is weak but the system is otherwise subject to arbitrarily large velocity and temperature gradients. The generalized self-diffusion coefficient $D(a)$ is a highly nonlinear function of the shear rate, although it does not depend on the thermal gradient ϵ. This feature may be due to the particular Maxwell interaction, since in the linear theory a crossed contribution to the mass flux due to the presence of the thermal gradient (Soret effect) does not appear either [1,16]. For $a=0$, $D=1$, i.e., one recovers the usual self-diffusion coefficient given by the linear theory [16]. The behavior of $D(a)$ is shown in Fig. 1. As happens in the case of uniform shear flow [5,9], the coefficient $D(a)$ decreases as the shear rate increases. On the other hand, Eq. (4.8) shows that the deviation of the partial pressure from its equilibrium value is at least of Burnett order, since it is proportional to $\epsilon \partial x_1/\partial s$. The coefficient $\Omega(a)$ is again a highly nonlinear function of the shear rate. For $a=0$, $\Omega=1$, and Eq. (4.8) defines the corresponding Burnett transport coefficient. The function $\Omega(a)$ is also plotted in Fig. 1. It is also a monotonically decreasing function of the shear rate. This means that for a given value of the concentration gradient, the partial pressure approaches its equilibrium value as the shear rate increases.

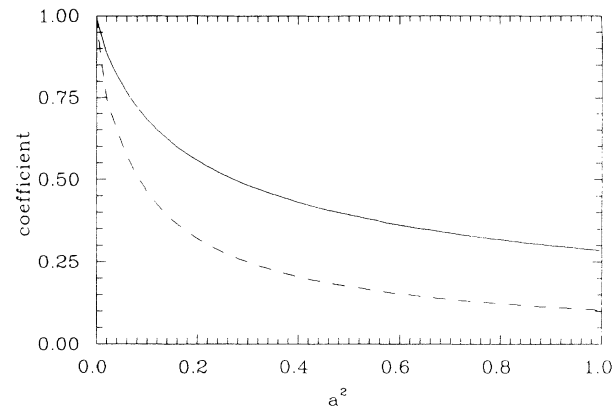

FIG. 1. Shear rate dependence of the self-diffusion coefficient (solid line) $D(a)$. The same for $\Omega(a)$ (dashed line).

It still remains to obtain the distribution function of the tagged particles. From the knowledge of the first moments, we are in conditions to explicitly write $f_1^{(1)}$. It is proved in Appendix C that

$$\begin{aligned} f_1^{(1)} = x_1 f - \frac{\partial x_1}{\partial s}(2k_B T/m)^{1/2}(\xi_y \mathcal{D}\xi_y + D(a)\xi_y \\ + 5\Omega(a)\{\tfrac{1}{4}\epsilon(\xi^2\mathcal{D}_z + 3) + [\tfrac{1}{2}a\epsilon\xi_x\mathcal{D}_z + \gamma(\xi^2\mathcal{D}_z + 3)]\xi_y\mathcal{D}\xi_y \\ + a(2\gamma\xi_x + \tfrac{1}{4}a\epsilon)\mathcal{D}_z\xi_y^2\mathcal{D}^2\xi_y^2 + \tfrac{5}{3}a^2\gamma\mathcal{D}_z\xi_y^3\mathcal{D}^3\xi_y^3\})f , \end{aligned} \tag{4.9}$$

where f is given by Eq. (3.7) and $\mathcal{D}_z$ and $\mathcal{D}$ are operators defined as

$$\mathcal{D}_z \equiv \xi_z^{-1}\frac{\partial}{\partial \xi_z}, \quad \mathcal{D} \equiv \frac{\partial}{\partial \xi_y} - \xi_y \mathcal{D}_z . \tag{4.10}$$

Equation (4.9) represents the major result of this paper. It may be considered as a generalization of the Navier-Stokes distribution function in the self-diffusion problem. It gives the velocity distribution function of tagged particles up to first order in the concentration gradient when the system is globally drawn out from equilibrium by the presence of arbitrary shear rate a and thermal gradient ϵ. The function $f_1^{(1)}$ is expressed in terms of a complicated operator acting on the distribution function f of the system seen as a whole. The nonlinear dependence of $f_1^{(1)}$ on a and ϵ appears both in the operator (which turns out to be linear in ϵ) and in f itself.

V. SELF-DIFFUSION IN PURE HEAT FLOW

As said in Sec. III, the case of pure heat flow is obtained in the limit $a \to 0$ ($\gamma \to 0$) with $\epsilon > 0$ fixed. In that particular case, the coefficients D and Ω are given by their near-equilibrium values. Nevertheless, the distribution function $f_1^{(1)}$ is still a highly nonlinear function of ϵ,

$$f_1^{(1)}=x_1 f-(2k_BT/m)^{1/2}\frac{\partial x_1}{\partial s}[\xi_y\mathcal{D}\xi_y+\xi_y+\tfrac{5}{4}\epsilon(\xi^2\mathcal{D}_z+3)]f$$
$$=x_i f-(2k_BT/m)^{1/2}\frac{\partial x_1}{\partial s}n\left[\frac{m}{2\pi k_BT}\right]^{3/2}\varphi(\xi,\epsilon)\ , \tag{5.1}$$

where f is given by Eq. (3.18) and

$$\varphi(\xi,\epsilon)=\frac{1}{\epsilon|\xi_y|}\int_{t_0}^{t_1}dt\,t^{-5/2}\left[\frac{1-t}{\epsilon}+\xi_y+\tfrac{5}{4}\epsilon\left[3-2\frac{\xi^2}{t}\right]\right]\exp\left[\frac{1-t}{\epsilon\xi_y}-\frac{\xi^2}{t}\right]\ . \tag{5.2}$$

Here, the limits of integration are the same as those of Eq. (3.18). The first two terms in the expansion of φ in powers of ϵ are

$$\varphi(\xi,\epsilon)=2\xi_y e^{-\xi^2}+\epsilon[\tfrac{5}{2}(\tfrac{3}{2}-\xi^2)+3\xi_y^2(\tfrac{5}{2}-\xi^2)]e^{-\xi^2}+O(\epsilon^2)\ , \tag{5.3}$$

as can be easily obtained from the corresponding expansion of f. Since the latter expansion is only asymptotic, so is the expansion (5.3). Thus, for moderate and large values of ϵ one has to use Eq. (5.2). In Fig. 2, we plot the function

$$\bar{\varphi}(\xi_y,\epsilon)=\frac{\int_{-\infty}^{\infty}d\xi_x\int_{-\infty}^{\infty}d\xi_z\varphi(\xi,\epsilon)}{\int_{-\infty}^{\infty}d\xi_x\int_{-\infty}^{\infty}d\xi_z e^{-\xi^2}} \tag{5.4}$$

for $\epsilon=1$. The shape of $\bar{\varphi}$ clearly differs from the one given by the Navier-Stokes limit ($\bar{\varphi}=2\xi_y$). However, it is noticeable that both functions lead to the same self-diffusion coefficient.

Although $\varphi(\xi,\epsilon)$ is a complicated function of ϵ, its velocity moments are just polynomials in the thermal gradient. We define the moments $N_{k,l}$ as

$$N_{k,l}(\epsilon)=\pi^{-3/2}\int d\xi\,\xi^{2k}\xi_y^l\varphi(\xi,\epsilon)\ . \tag{5.5}$$

Equation (5.1) implies that

$$N_{k,l}=\tfrac{15}{4}\epsilon M_{k,l}-2kM_{k-1,l+3}-lM_{k,l+1}+\tfrac{5}{4}\epsilon Q_{k+1,l}-Q_{k,l+3}\ , \tag{5.6}$$

where

$$M_{k,l}=\int d\xi\,\xi^{2k}\xi_y^l\frac{1}{n}\left[\frac{m}{2k_BT}\right]^{-3/2}f\ , \tag{5.7}$$

$$Q_{k,l}=\int d\xi\,\xi^{2k}\xi_y^l\mathcal{D}_z\frac{1}{n}\left[\frac{m}{2k_BT}\right]^{-3/2}f\ . \tag{5.8}$$

The moments $M_{k,l}$ have been calculated elsewhere [19] and here we only quote the result

$$M_{k,l}=(-1)^l 2^{-(2k+l)}\sum_{\substack{r=0\\(l+r)\text{even}}}^{2(k-1)+l}\frac{(2k+l+r+1)!}{\left[k-1+\frac{l-r}{2}\right]!\left[k+\frac{l+r}{2}\right](l+r+1)}\left[\frac{\epsilon}{2}\right]^r\ . \tag{5.9}$$

The moments $Q_{k,l}$ are worked out in Appendix D and they are given by

$$Q_{k,l}=(-l)^{l+1}2\sum_{\substack{r=0\\(l+r)\text{even}}}^{2k+l+4}\frac{(2k+l+r+1)!!\left[k+\frac{l+r}{2}-2\right]!}{2^{k+(l+r)/2}(l+r+1)\left[k+\frac{l-r}{2}-2\right]!}\epsilon^r\ . \tag{5.10}$$

VI. CONCLUSIONS

In this paper we have addressed the problem of diffusion of tagged particles in a dilute gas subject to arbitrarily large velocity and temperature gradients (steady Couette flow). The analysis is made from a kinetic model proposed for a multicomponent gas which reduces to the BGK equation for a single gas. The knowledge of the velocity distribution function of the system as a whole enables us to solve the kinetic equation for the distribution of the tagged particles. A perturbation expansion in the same spirit as in the Chapman-Enskog method is carried out to first order in the gradient of the tagged particles, the steady Couette flow being the reference state. Therefore, the successive approximations are nonlinear functions of both the shear rate and the thermal gradient. The main transport coefficients have been computed. In particular, we have derived an explicit expression for the

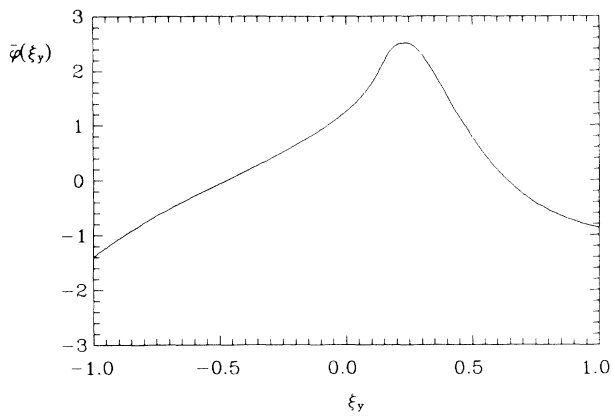

FIG. 2. Plot of the distribution function $\bar{\varphi}(\xi_y)$ for $\epsilon=1$.

self-diffusion coefficient, which is independent of the thermal gradient and nonlinearly depends on the shear rate. This coefficient decreases with increasing shear rate. A similar behavior has been found for the uniform-shear-flow problem [5,9]. Further, a generalized partial pressure of the tagged particles has been obtained which is linear in the thermal gradient and highly nonlinear in the shear rate. In addition, we have been able to explicitly write the velocity distribution function of the tagged particles. It is given in terms of a complicated operator acting on the distribution of the whole system. In absence of shear rate (pure heat flow), the self-diffusion coefficient reduces to the one given in the linear regime and the partial pressure is of Burnett order. All the velocity moments are polynomials in the thermal gradient ϵ and the distribution function is a nonlinear function of ϵ.

Although the results presented in this paper have been obtained from a kinetic model, it seems reasonable to conjecture that the qualitative features given here are also present in the Boltzmann description. On the other hand, the problem can be also extended to the case of arbitrary mass ratio in the tracer limit in the same way as in the uniform shear flow problem [9]. Work is now in progress along these lines.

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APPENDIX A

This appendix contains the calculation of the pressure tensor and heat flux in the steady Couette flow. Equations (3.16) and (3.17) have already been obtained elsewhere [14]. Equation (3.15) is an obvious consequence of the fact that the distribution function given by Eq. (3.7) is an even function of ξ_z. Equation (3.12) is obtained from Eqs. (3.13) and (3.14) by taking into account the identity $P_{xx}+P_{yy}+P_{zz}=3p$. Thus, only Eqs. (3.13) and (3.14) need to be proved here.

Rather than from direct integration of Eq. (3.7), expressions for the moments of f are much more easily found by considering the formal solution of the BGK equation (2.16) for steady Couette flow,

$$f=\left[1+v_y\frac{\partial}{\partial s}\right]^{-1}f^{\mathrm{LE}}=\sum_{k=0}^{\infty}(-v_y)^k\left[\frac{\partial}{\partial s}\right]^k f^{\mathrm{LE}}\ , \tag{A1}$$

where $ds\equiv\zeta(y)dy$. Let us start with the calculation of P_{yy},

$$\begin{aligned}P_{yy}&=m\sum_{k=0}^{\infty}\left[-\frac{\partial}{\partial s}\right]^k\int d\mathbf{v}\,v_y^{k+2}f^{\mathrm{LE}}\\&=m\sum_{k=0}^{\infty}\left[\frac{\partial}{\partial s}\right]^{2k}(2k+1)!!n(k_BT/m)^{k+1}\\&=p\sum_{k=0}^{\infty}(2k)!(2k+1)!!(-\gamma)^k\ ,\end{aligned} \tag{A2}$$

where in the last step use has been made of Eqs. (3.1) and (3.3). Similarly,

$$\begin{aligned}P_{zz}&=m\sum_{k=0}^{\infty}\left[-\frac{\partial}{\partial s}\right]^k\int d\mathbf{v}\,v_z^2v_y^kf^{\mathrm{LE}}\\&=p+m\sum_{k=1}^{\infty}\left[\frac{\partial}{\partial s}\right]^{2k}(2k-1)!!n(k_BT/m)^{k+1}\\&=p\left[1+\sum_{k=1}^{\infty}(2k)!(2k-1)!!(-\gamma)^k\right]\ .\end{aligned} \tag{A3}$$

The asymptotic expansion in powers of γ of the function $F_0(\gamma)$ defined in Eq. (3.6) is [14]

$$F_0(\gamma)=\sum_{k=0}^{\infty}(2k+1)!(2k+1)!!(-\gamma)^k\ . \tag{A4}$$

Therefore, the expansion of

$$F_r(\gamma)\equiv\left[\frac{d}{d\gamma}\gamma\right]^rF_0(\gamma)$$

is

$$F_r(\gamma)=\sum_{k=0}^{\infty}(k+1)^r(2k+1)!(2k+1)!!(-\gamma)^k\ . \tag{A5}$$

Notice that $F_r(\gamma)$, $r\geq 3$ is a linear combination of F_0, F_1, and F_2. For instance,

$$F_3=\frac{1}{8\gamma}(1-F_0)-F_2-\tfrac{1}{4}F_1\ , \tag{A6}$$

$$F_4=\frac{d}{d\gamma}(\gamma F_3)=\frac{1}{8\gamma}(2F_0-F_1-1)+\tfrac{1}{4}F_1+\tfrac{3}{4}F_2\ . \tag{A7}$$

Comparison between Eqs. (A3) and (A5) gives Eq. (3.14). Similarly, Eq. (A2) becomes Eq. (3.13).

APPENDIX B

The expressions for $J_1^{(1)}$ and $p_1^{(1)}$ will be derived in this appendix. First, notice that integration of Eq. (4.2) over velocity shows that $\partial J_1^{(1)}/\partial s=0$. Furthermore, we assume, to be verified later, that $\partial^2(p_1^{(1)}/p-x_1)/\partial s^2=0$. Following similar steps as in Appendix A, one has

$$J_1^{(1)}=\sum_{k=0}^{\infty}\left[-\frac{\partial}{\partial s}\right]^k\int d\mathbf{v}\, v_y^{k+1}\Phi_1^{(1)}$$

$$=-\sum_{k=0}^{\infty}(2k+1)!!\left[\frac{\partial}{\partial s}\right]^{2k+1}x_1 n(k_BT/m)^{k+1}+\frac{m}{2p}J_1^{(1)}\sum_{k=0}^{\infty}(2k+1)!!\left[\frac{\partial}{\partial s}\right]^{2k}n(k_BT/m)^{k+1}$$

$$-\frac{1}{2}\sum_{k=0}^{\infty}(k+1)(2k+1)!!\left[\frac{\partial}{\partial s}\right]^{2k+1}\left[\frac{p_1^{(1)}}{p}-x_1\right]n(k_BT/m)^{k+1}\,. \tag{B1}$$

Now, taking into account Eqs. (3.1) and (3.3), one gets

$$J_1^{(1)}=-\frac{\partial x_1}{\partial s}\frac{p}{m}\sum_{k=0}^{\infty}(2k+1)!(2k+1)!!(-\gamma)^k+\tfrac{1}{2}J_1^{(1)}\sum_{k=0}^{\infty}(2k)!(2k+1)!!(-\gamma)^k$$

$$-\frac{1}{4}\left[\frac{\partial}{\partial s}\left[\frac{p_1^{(1)}}{p}-x_1\right]\right]\frac{p}{m}\sum_{k=0}^{\infty}(2k+2)!(2k+1)!!(-\gamma)^k\,. \tag{B2}$$

All the three summations have already appeared in Appendix A. Thus, Eq. (B2) can be conveniently rewritten as

$$J_1^{(1)}=-\frac{\partial x_1}{\partial s}\frac{p}{m}F_0+\tfrac{1}{2}J_1^{(1)}[1-2\gamma(F_1+2F_2)]-\frac{p}{2m}F_1\frac{\partial}{\partial s}\left[\frac{p_1^{(1)}}{p}-x_1\right]\,. \tag{B3}$$

Let us consider now $p_1^{(1)}$

$$p_1^{(1)}=\tfrac{1}{3}m\sum_{k=0}^{\infty}\int d\mathbf{v}\, V^2\left[-\frac{\partial}{\partial s}\right]^k v_y^k\Phi_1^{(1)}$$

$$=\tfrac{1}{3}m\sum_{k=0}^{\infty}\left[-\frac{\partial}{\partial s}\right]^k\int d\mathbf{v}\, V^2 v_y^k\Phi_1^{(1)}-\tfrac{2}{3}ma\sum_{k=1}^{\infty}k\left[-\frac{\partial}{\partial s}\right]^{k-1}\int d\mathbf{v}\, V_x v_y^k\Phi_1^{(1)}$$

$$+\frac{1}{3}ma^2\sum_{k=2}^{\infty}k(k-1)\left[-\frac{\partial}{\partial s}\right]^{k-2}\int d\mathbf{v}\, v_y^k\Phi_1^{(1)}\,, \tag{B4}$$

where in the last step we have taken into account that

$$\left[\frac{\partial}{\partial s}\right]^k V^2=V^2\left[\frac{\partial}{\partial s}\right]^k+k\frac{\partial V^2}{\partial s}\left[\frac{\partial}{\partial s}\right]^{k-1}+\frac{k(k-1)}{2}\frac{\partial V^2}{\partial s^2}\left[\frac{\partial}{\partial s}\right]^{k-2}$$

$$=V^2\left[\frac{\partial}{\partial s}\right]^k-2ak\left[\frac{\partial}{\partial s}\right]^{k-1}V_x-a^2k(k-1)\left[\frac{\partial}{\partial s}\right]^{k-2}\,, \tag{B5}$$

since V^2 is a quadratic function of the variable s. We must evaluate each separate term in the right-hand side of Eq. (B4). The first one is

$$\sum_{k=0}^{\infty}\left[-\frac{\partial}{\partial s}\right]^{k}\int d\mathbf{v}\, V^2 v_y^k \Phi_1^{(1)}$$

$$=\sum_{k=0}^{\infty}\frac{(2k+3)!!}{2k+1}\left[\frac{\partial}{\partial s}\right]^{2k} x_1 n(k_BT/m)^{k+1}-\frac{m}{2p}J_1^{(1)}\sum_{k=0}^{\infty}\frac{(2k+5)!!}{2k+3}\left[\frac{\partial}{\partial s}\right]^{2k+1} n(k_BT/m)^k$$

$$+\frac{1}{2}\sum_{k=0}^{\infty}(k+1)\frac{(2k+3)!!}{2k+1}\left[\frac{\partial}{\partial s}\right]^{2k}\left[\frac{p_1^{(1)}}{p}-x_1\right]n(k_BT/m)^{k+1}$$

$$=x_1\frac{p}{m}\sum_{k=0}^{\infty}(2k)!\frac{(2k+3)!!}{2k+1}(-\gamma)^k+\frac{pk_B}{2m^2}\frac{\partial x_1}{\partial s}\frac{\partial T}{\partial s}\sum_{k=0}^{\infty}(2k+5)(2k+2)(2k+2)!(2k+1)!!(-\gamma)^k$$

$$-J_1^{(1)}\frac{k_B}{4m}\frac{\partial T}{\partial s}\sum_{k=0}^{\infty}(2k+5)(2k+2)!(2k+1)!!(-\gamma)^k$$

$$+\left[\frac{p^{(1)}}{p}-x_1\right]\frac{p}{2m}\sum_{k=0}^{\infty}\frac{k+1}{2k+1}(2k)!(2k+3)!!(-\gamma)^k$$

$$+\left[\frac{\partial}{\partial s}\left[\frac{p_1^{(1)}}{p}-x_1\right]\right]\frac{pk_B}{4m^2}\frac{\partial T}{\partial s}\sum_{k=0}^{\infty}(2k+5)(2k+4)(k+1)(2k+2)!(2k+1)!!(-\gamma)^k\,, \tag{B6}$$

where in the last equality we have used the result

$$\left[\frac{\partial}{\partial s}\right]^{2k+1}(k_BT/m)^{k+1}=\frac{k_B}{m}\frac{\partial T}{\partial s}\frac{(2k+2)!}{2}(-\gamma)^k\,. \tag{B7}$$

Since $\Phi_1^{(1)}$ is an even function of V_x, the second term in the right-hand side of Eq. (B4) vanishes. The third term is

$$\sum_{k=0}^{\infty}(k+2)(k+1)\left[-\frac{\partial}{\partial s}\right]^{k}\int d\mathbf{v}\, v_y^{k+2}\Phi_1^{(1)}$$

$$=\sum_{k=0}^{\infty}(2k+2)(2k+1)(2k+1)!!\left[\frac{\partial}{\partial s}\right]^{2k} x_1 n(k_BT/m)^{k+1}$$

$$-\frac{m}{2p}J_1^{(1)}\sum_{k=0}^{\infty}(2k+3)(2k+2)(2k+3)!!\left[\frac{\partial}{\partial s}\right]^{2k+1} n(k_BT/m)^{k+2}$$

$$+\sum_{k=0}^{\infty}(k+1)^2(2k+1)(2k+1)!!\left[\frac{\partial}{\partial s}\right]^{2k}\left[\frac{p_1^{(1)}}{p}-x_1\right]n(k_BT/m)^{k+1}$$

$$=x_1\frac{p}{m}\sum_{k=0}^{\infty}(2k+2)!(2k+1)!!(-\gamma)^k+\frac{pk_B}{m^2}\frac{\partial x_1}{\partial s}\frac{\partial T}{\partial s}\sum_{k=0}^{\infty}(k+1)(2k+4)!(2k+3)!!(-\gamma)^k$$

$$-\tfrac{1}{2}J_1^{(1)}\frac{k_B}{m}\frac{\partial T}{\partial s}\sum_{k=0}^{\infty}(k+1)(2k+3)!(2k+3)!!(-\gamma)^k+\frac{p}{2m}\left[\frac{p_1^{(1)}}{p}-x_1\right]\sum_{k=0}^{\infty}(k+1)(2k+2)!(2k+1)!!(-\gamma)^k$$

$$+\frac{pk_B}{2m^2}\left[\frac{\partial}{\partial s}\left[\frac{p_1^{(1)}}{p}-x_1\right]\right]\frac{\partial T}{\partial s}\sum_{k=0}^{\infty}(k+1)(k+2)(2k+4)!(2k+3)!!(-\gamma)^k\,. \tag{B8}$$

Substitution of Eqs. (B6) and (B8) into Eq. (B4) yields

$$\frac{p_1^{(1)}}{p}-x_1=\frac{k_B}{m}\frac{\partial T}{\partial s}\frac{\partial x_1}{\partial s}[A_1(\gamma)+a^2B_1(\gamma)]-\frac{k_B}{p}J_1^{(1)}\frac{\partial T}{\partial s}[A_2(\gamma)+a^2B_2(\gamma)]$$

$$+\left[\frac{p_1^{(1)}}{p}-x_1\right][A_3(\gamma)+a^2B_3(\gamma)]+\frac{k_B}{m}\frac{\partial T}{\partial s}\left[\frac{\partial}{\partial s}\left[\frac{p_1^{(1)}}{p}-x_1\right]\right][A_4(\gamma)+a^2B_4(\gamma)]\,, \tag{B9}$$

where

$$A_1(\gamma)=\tfrac{1}{6}\sum_{k=0}^{\infty}(2k+5)(2k+2)(2k+2)!(2k+1)!!(-\gamma)^k\,, \tag{B10}$$

$$A_2(\gamma)=\tfrac{1}{12}\sum_{k=0}^{\infty}(2k+5)(2k+2)!(2k+1)!!(-\gamma)^k\ , \tag{B11}$$

$$A_3(\gamma)=\tfrac{1}{6}\sum_{k=0}^{\infty}\frac{k+1}{2k+1}(2k)!(2k+3)!!(-\gamma)^k\ , \tag{B12}$$

$$A_4(\gamma)=\tfrac{1}{12}\sum_{k=0}^{\infty}(2k+5)(2k+4)(k+1)(2k+2)!(2k+1)!!(-\gamma)^k\ , \tag{B13}$$

$$B_1(\gamma)=\tfrac{1}{3}\sum_{k=0}^{\infty}(k+1)(2k+4)!(2k+3)!!(-\gamma)^k\ , \tag{B14}$$

$$B_2(\gamma)=\tfrac{1}{6}\sum_{k=0}^{\infty}(k+1)(2k+3)!(2k+3)!!(-\gamma)^k\ , \tag{B15}$$

$$B_3(\gamma)=\tfrac{1}{6}\sum_{k=0}^{\infty}(k+1)(2k+2)!(2k+1)!!(-\gamma)^k\ , \tag{B16}$$

$$B_4(\gamma)=\tfrac{1}{6}\sum_{k=0}^{\infty}(k+1)(k+2)(2k+4)!(2k+3)!!(-\gamma)^k\ . \tag{B17}$$

All these functions can be expressed in terms of F_0, F_1, and F_2,

$$A_1=\tfrac{4}{3}F_3+2F_2=\frac{1}{6\gamma}(1-F_0)-\tfrac{1}{3}F_1+\tfrac{2}{3}F_2\ , \tag{B18}$$

$$A_2=\tfrac{1}{2}F_1+\tfrac{1}{3}F_2\ , \tag{B19}$$

$$\begin{aligned}A_3&=\tfrac{1}{2}-\gamma(\tfrac{2}{3}F_3+\tfrac{5}{3}F_2+F_0)\\&=\tfrac{5}{12}+\tfrac{1}{12}F_0-\gamma(F_0-\tfrac{1}{6}F_1+F_2)\ ,\end{aligned} \tag{B20}$$

$$\begin{aligned}A_4&=F_2+\tfrac{5}{3}F_3+\tfrac{2}{3}F_4\\&=\frac{1}{24\gamma}(3-F_0-2F_1)-\tfrac{1}{4}F_1-\tfrac{1}{6}F_2\ ,\end{aligned} \tag{B21}$$

$$B_1=\frac{2}{3\gamma}(F_1-F_2)\ , \tag{B22}$$

$$B_2=\frac{1}{6\gamma}(F_0-F_1)\ , \tag{B23}$$

$$B_3=\frac{1}{3}F_2\ , \tag{B24}$$

$$\begin{aligned}B_4&=\frac{1}{3\gamma}(F_2-F_3)\\&=\frac{1}{\gamma}(\tfrac{1}{12}F_1+\tfrac{2}{3}F_2)+\frac{1}{24\gamma^2}(F_0-1)\ .\end{aligned} \tag{B25}$$

In Eq. (B9) it is the combination $C_r=A_r+a^2B_r$ that really matters. By making use of Eq. (3.5), one has

$$C_1=\frac{1}{6\gamma F_1}[2\gamma(5F_1^2-4F_2^2)-F_1(F_0-1)]\ , \tag{B26}$$

$$C_2=\frac{F_0}{6F_1}(3F_1+2F_2)\ , \tag{B27}$$

$$C_3=\tfrac{1}{12}(F_0+5)-\frac{\gamma}{6F_1}[6F_0F_1-(F_1^2+4F_2^2)]\ , \tag{B28}$$

$$C_4=\frac{1}{12\gamma F_1}[8\gamma F_2(3F_1+2F_2)+F_0(F_1+F_2)-F_1^2-F_2]\ . \tag{B29}$$

Derivation of Eq. (B9) with respect to s gives

$$\begin{aligned}\frac{\partial}{\partial s}\left[\frac{p_1^{(1)}}{p}-x_1\right]&=-2\gamma C_1\frac{\partial x_1}{\partial s}+2\gamma C_2\frac{m}{p}J_1^{(1)}\\&\quad+C_3\frac{\partial}{\partial s}\left[\frac{p_1^{(1)}}{p}-x_1\right]\\&\quad-2\gamma C_4\frac{\partial}{\partial s}\left[\frac{p_1^{(1)}}{p}-x_1\right]\ ,\end{aligned} \tag{B30}$$

where use has been made of Eq. (3.3) and terms of order $\partial^2x_1/\partial s^2$ have been neglected. Equation (B30) confirms that $\partial(p_1^{(1)}/p-x_1)/\partial s=\text{const}$. From Eqs. (B3) and (B30), the flux of tagged particles $J_1^{(1)}$ is given by

$$J_1^{(1)}=-\frac{2p}{m}D(a)\frac{\partial x_1}{\partial s}\ , \tag{B31}$$

where

$$D(a)=\frac{F_0(2\gamma C_4+1-C_3)-\gamma F_1C_1}{2\gamma F_1C_2-(2\gamma F_1+4\gamma F_2+1)(C_3-2\gamma C_4-1)}\ . \tag{B32}$$

The first terms in the power expansion of $D(a)$ are

$$D(a)=1-34\gamma+\cdots=1-\tfrac{34}{5}a^2+\cdots\ . \tag{B33}$$

Once $J_1^{(1)}$ is known, Eqs. (B9) and (B30) enable one to get $p_1^{(1)}$. The result is

$$p_1^{(1)}=p[x_1+10\Omega(a)\epsilon(k_BT/2m)^{1/2}\frac{\partial x_1}{\partial s}\ , \tag{B34}$$

where

$$\Omega(a)=\tfrac{1}{10}D(a)\frac{(2\gamma F_1+4\gamma F_2+1)C_1+2F_0C_2}{F_0(2\gamma C_4+1-C_3)-\gamma F_1C_2}\ , \tag{B35}$$

and its first power expansion terms are

$$\Omega(a)=1-\tfrac{82}{5}a^2+\cdots\ . \tag{B36}$$

APPENDIX C

Here we derive the explicit solution of Eq. (4.2). First, it is convenient to prove by induction the identity

$$\hat{\mathcal{D}}^{r+1}v_y g(\mathbf{v})=r\hat{\mathcal{D}}^r g(\mathbf{v})+\hat{\mathcal{D}}v_y\hat{\mathcal{D}}^r g(\mathbf{v})\ , \tag{C1}$$

where $g(\mathbf{v})$ is an arbitrary function and $\hat{\mathcal{D}}$ is the operator

$$\hat{\mathcal{D}}\equiv\frac{\partial}{\partial v_y}-\frac{v_y}{v_z}\frac{\partial}{\partial v_z}\ . \tag{C2}$$

Equation (C1) is obviously true for $r=0$. Provided that Eq. (C1) holds for a given integer value of r, let us see that it also holds for the next integer value,

$$\begin{aligned}\hat{\mathcal{D}}^{r+2}v_y g&=\hat{\mathcal{D}}^{r+1}(g+v_y\hat{\mathcal{D}}g)\\&=(r+1)\hat{\mathcal{D}}^{r+1}g+\hat{\mathcal{D}}v_y\hat{\mathcal{D}}^{r+1}g\ ,\end{aligned} \tag{C3}$$

where in the first step we have taken into account that $\hat{\mathcal{D}}v_y g=g+v_y\hat{\mathcal{D}}g$, and in the last step we have used Eq. (C1) for $g\to\hat{\mathcal{D}}g$.

Now we can prove the relation

$$\sum_{k=0}^{\infty}\frac{(k+r)!}{k!}v_y^k\left[-\frac{\partial}{\partial s}\right]^k f^{\mathrm{LE}}=\hat{\mathcal{D}}^r v_y^r f\ . \tag{C4}$$

For $r=0$, Eq. (C4) becomes Eq. (A1). For $r\to r+1$, the left-hand side of Eq. (C4) becomes

$$\begin{aligned}&\sum_{k=0}^{\infty}\frac{(k+r+1)!}{k!}v_y^k\left[-\frac{\partial}{\partial s}\right]^k f^{\mathrm{LE}}\\&=r\hat{\mathcal{D}}^r v_y^r f+\sum_{k=0}^{\infty}\frac{(k+r)!}{k!}\left[\frac{\partial}{\partial v_y}v_y^{k+1}\right]\left[-\frac{\partial}{\partial s}\right]^k f^{\mathrm{LE}}\\&=r\hat{\mathcal{D}}^r v_y^r f+\hat{\mathcal{D}}v_y\hat{\mathcal{D}}^r v_y^r f\ ,\end{aligned} \tag{C5}$$

where we have used Eq. (C4) and the identity $\partial f^{\mathrm{LE}}/\partial v_y=(v_y/v_z)\partial f^{\mathrm{LE}}/\partial v_z$. Insertion of Eq. (C1) into Eq. (C5) proves Eq. (C4) for $r\to r+1$.

Let us consider in Eq. (4.6) the first piece of $\Phi_1^{(1)}$,

$$\begin{aligned}&\left[1+v_y\frac{\partial}{\partial s}\right]^{-1}x_1 f^{\mathrm{LE}}\\&=x_1\sum_{k=0}^{\infty}v_y^k\left[-\frac{\partial}{\partial s}\right]^k f^{\mathrm{LE}}\\&\quad-\frac{\partial x_1}{\partial s}\sum_{k=0}^{\infty}(k+1)v_y^{k+1}\left[-\frac{\partial}{\partial s}\right]^k f^{\mathrm{LE}}\\&=x_1 f-\frac{\partial x_1}{\partial s}v_y\hat{\mathcal{D}}v_y f\ ,\end{aligned} \tag{C6}$$

where in the first step terms of order $\partial^2 x_1/\partial s^2$ have been neglected.

The next contribution in Eq. (4.6) is

$$\begin{aligned}\left[1+v_y\frac{\partial}{\partial s}\right]^{-1}\frac{m}{2p}v_y J_1^{(1)}f^{\mathrm{LE}}&=\frac{m}{2p}v_y J_1^{(1)}f\\&=-D(a)v_y f\frac{\partial x_1}{\partial s}\ .\end{aligned} \tag{C7}$$

Finally, the last contribution is

$$\begin{aligned}&\frac{1}{2}\left[1+v_y\frac{\partial}{\partial s}\right]^{-1}\left[\frac{mV^2}{2k_BT}-\frac{3}{2}\right]\left[\frac{p_1^{(1)}}{p}-x\right]f^{\mathrm{LE}}\\&=-\frac{1}{4}\left[1+v\frac{\partial}{y\partial s}\right]^{-1}\left[V^2\hat{\mathcal{D}}_z+3\right]\left[\frac{p_1^{(1)}}{p}-x_1\right]f^{\mathrm{LE}}\\&=-\tfrac{1}{4}(V^2\hat{\mathcal{D}}_z+3)\left[\frac{p_1^{(1)}}{p}-x_1\right]\sum_{k=0}^{\infty}v_y^k\left[-\frac{\partial}{\partial s}\right]^k f^{\mathrm{LE}}\\&\quad+\frac{1}{4}\left[\frac{\partial}{\partial s}(V^2\hat{\mathcal{D}}_z+3)\left[\frac{p_1^{(1)}}{p}-x_1\right]\right]\sum_{k=0}^{\infty}(k+1)v_y^{k+1}\left[-\frac{\partial}{\partial s}\right]^k f^{\mathrm{LE}}\\&\quad-\frac{1}{8}\left[\frac{\partial^2}{\partial s^2}(V^2\hat{\mathcal{D}}_z+3)\left[\frac{p_1^{(1)}}{p}-x_1\right]\right]\sum_{k=0}^{\infty}(k+1)(k+2)v_y^{k+2}\left[-\frac{\partial}{\partial s}\right]^k f^{\mathrm{LE}}\\&\quad+\frac{1}{24}\left[\frac{\partial^3}{\partial s^3}(v^2\hat{\mathcal{D}}_z+3)\left[\frac{p_1^{(1)}}{p}-x_1\right]\right]\sum_{k=0}^{\infty}(k+1(k+2)(k+3)v_y^{k+3}\left[-\frac{\partial}{\partial s}\right]^k f^{\mathrm{LE}}\ ,\end{aligned} \tag{C8}$$

with $\hat{\mathcal{D}}_z\equiv v_z^{-1}\partial/\partial v_z$. Taking into account Eqs. (B5), (B34), and (C4), the last expression can be written in the form

$$-5\Omega(a)\frac{\partial x_1}{\partial s}\{\epsilon(k_BT/2m)^{1/2}\tfrac{1}{2}(V^2\hat{\mathcal{D}}_z+3)+[a\epsilon(k_BT/2m)^{1/2}V_x\hat{\mathcal{D}}_z+\gamma(V^2\hat{\mathcal{D}}_z+3)]v_y\hat{\mathcal{D}}v_y$$

$$+[2a\gamma V_x\hat{\mathcal{D}}_z+\tfrac{1}{2}a^2\epsilon(k_BT/2m)^{1/2}\hat{\mathcal{D}}_z]v_y^2\hat{\mathcal{D}}^2v_y^2+\tfrac{5}{3}a^2\gamma\hat{\mathcal{D}}_zv_y^3\hat{\mathcal{D}}^3v_y^3\}f\ . \quad \text{(C9)}$$

Putting together Eqs. (C6), (C7), and (C9), one gets Eq. (4.9).

APPENDIX D

In this appendix the moments defined in Eq. (4.17) are evaluated. By making use of Eq. (A1), we have

$$Q_{k,l}=-\frac{1}{n}\left[\frac{m}{2k_BT}\right]^{k-1+l/2}\sum_{r=0}^{\infty}\left[-\frac{\partial}{\partial s}\right]^r\frac{m}{k_BT}n\left[\frac{2k_BT}{m}\right]^{k+[(l+r)/2]}\pi^{-3/2}\int d\xi\,\xi^{2k}\xi_y^{l+r}e^{-\xi^2}$$

$$=(-1)^{l+1}2T^{-(k+l/2)+2}\sum_{\substack{r=0\\(r+l)\text{even}}}^{\infty}\left[\frac{k_B}{m}\right]^{r/2}\frac{(2k+l+r+1)!!}{2^{k+l/2}(l+r+1)}\left[\frac{\partial}{\partial s}\right]^rT^{k+[(l+r)/2]-2}\ . \quad \text{(D1)}$$

Since T is a linear function of s, one finally gets

$$Q_{k,l}=(-1)^{l+1}2\sum_{\substack{r=0\\(l+r)\text{even}}}^{2k+l-4}\frac{(2k+l+r+1)!!\left[k+\dfrac{l+r}{2}-2\right]!}{2^{k+(l+r)/2}(l+r+1)\left[k+\dfrac{l-r}{2}-2\right]!}\epsilon^r\ . \quad \text{(D2)}$$

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