Brownian motion of finite-inertia particles in a simple shear flow

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Simultaneous diffusive and inertial motion of Brownian particles in laminar Couette flow is investigated via Lagrangian and Eulerian descriptions to determine the effect of particle inertia on diffusive transport in the long-time limit. The classical fluctuation dissipation theorem is used to calculate the amplitude of random-force correlations, thereby neglecting corrections of the order of the molecular relaxation time to the inverse shear rate. In the diffusive limit (time much greater than the particle relaxation time) the fluctuating particle-velocity autocorrelations functions are found to be stationary in time, the correlation in the streamwise direction being an exponential multiplied by an algebraic function and the cross correlation nonsymmetric in the time difference. The analytic, nonperturbative, evaluation of the particle-phase total pressure, which is calculated to be second order in the Stokes number (a dimensionless measure of particle inertia), shows that the particle phase behaves as a non-Newtonian fluid. The generalized Smoluchowski convective-diffusion equation, determined analytically from a combination of the particle-phase pressure tensor and the inertial acceleration term, contains a shear-dependent cross derivative term and an additional term along the streamwise direction, quadratic in the particle Stokes number. The long-time diffusion coefficients associated with the particle flux relative to the carrier flow are found to depend on particle inertia such that the streamwise diffusion coefficient becomes negative with increasing Stokes number, whereas one of the cross coefficients is always negative. The total diffusion coefficients measuring the rate of change of particle mean-square displacement are always positive as expected from general stability arguments.

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I. INTRODUCTION

Combined inertial and diffusive motion of Brownian particles in a flowing fluid is important in a number of aerosol processes, including filtration, aerosol sampling, deposition in bends, and particulate deposition in the human respiratory tract. The limit of negligible inertial effects, where the particles follow closely the motion of the fluid, has been extensively studied in sheared colloidal suspensions [1,2]. These two-phase systems, which consist of a dispersed particulate phase and a continuous fluid phase (gas for aerosols), have long been of interest for their important industrial and engineering applications. Pure diffusive particle motion is usually described by a convective gradient-diffusion equation from which inertial effects are absent. The limit of inertial transport, where diffusion is neglected and particle trajectories deviate significantly from the fluid stream lines, is most conveniently described in terms of the particle equations of motion in a Lagrangian formulation. In the transition regime between the diffusion limit and the inertia-dominated limit the two particle-transport mechanisms have to be considered simultaneously.

The effect of particle inertia on the diffusive motion of noninteracting Brownian particles in nonuniform fluids has been examined via numerous approaches. Continuum descriptions in terms of mass and momentum conservation equations (macroscopic "hydrodynamic" equations) require constitutive relations for the particle-phase total pressure tensor. A frequently made approximation is to consider the Brownian particles as an "ideal gas," thereby using a phenomenological expression for the particle-phase pressure tensor [3,4].

Mesoscopic descriptions of Brownian motion involve stochastic particle equations of motion and the associated Fokker-Planck equation, as, for example, in Subramanian and Brady [5] where a multiple-scale analysis of the Fokker-Planck equation in a simple shear flow is presented. In mesoscopic descriptions in terms of Langevin equations the fluctuation dissipation theorem (FDT) is an essential ingredient of the calculation. Santamaría-Holek *et al.* [6] used an alternative approach by considering the motion of a Brownian-particle in an external flow field as an example of a driven, far-from-equilibrium system. They used mesoscopic nonequilibrium thermodynamics, an approach that does not require the specification of the stochastic properties of the random force, to obtain the Fokker-Planck equation for the nonequilibrium Brownian particle distribution function in a simple shear flow. They found that the diffusion tensor in the Fokker-Planck equation depends on the shear rate, concluding that fluctuations about the nonequilibrium steady state lead to a violation of the classical (equilibrium) FDT.

Kinetic theory has also been used to obtain the Fokker-Planck equation for the motion of Brownian particles in rarefied nonuniform gases. Fernández de la Mora and Mercer [7] expanded the Boltzmann collision operator in the ratio of the light-gas molecular mass to the Brownian-particle mass; they approximated the light-gas distribution function by the first two terms in the Chapman-Enskog expansion to derive a

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Fokker-Planck equation whose diffusion tensor was found independent of light-gas velocity gradients [7]. However, Rodrígez *et al.* [8], using a similar low-mass-ratio expansion of the collision operator for Maxwellian fluid molecules, obtained a Fokker-Planck equation in a uniform shear flow with a diffusion tensor that depended on the irreversible fluid stress tensor.

Herein, we consider a dilute suspension of noninteracting Brownian particles in a two-dimensional, simple shear (laminar, plane Couette flow). Inertial Brownian particles are considered: namely, particles whose Stokes number (the ratio of the particle relaxation time to the inverse shear rate, or more properly strain rate, a dimensionless number that describes particle response to spatial changes in the carrier flow velocity) is finite (at least of order unity). The limit of small Stokes number corresponds to pure diffusive motion, whereas for large Stokes numbers inertial transport dominates. A mesoscopic approach is adopted, whereby both Lagrangian and Eulerian descriptions are presented to investigate the effect of particle inertia on the convective-diffusion equation (equivalently, the Smoluchowski equation) in the long-time, diffusive limit. The classical FDT is used since we argue that the modification of the classical FDT derived in Refs. [6,8] is of the order of the ratio of the molecular (fluid) relaxation time (a molecular time scale) to the inverse shear rate (a macroscopic time scale) and thus negligible for typical shear rates.

A linear flow field was chosen as the underlying carrier-gas velocity field because the stochastic particle equations of motion and the associated Fokker-Planck equation can be solved exactly for linear flow fields. Thus, the closure problem associated with the continuum equations for the dispersed phase is avoided, and analytic, nonperturbative expressions may be derived in the long-time limit (for example, for the mean particle velocity). The specific case of a simple shear is investigated because Brownian motion in a simple shear has been studied extensively; see, for example, Refs. [5,6,8,9]. These analyses have been limited in either perturbative calculations [5,6] or in calculations of the long-time behavior of equal-time correlation functions without considerations of the associated convective-diffusion equation [9]. The long-time, analytic expressions obtained herein extend these previous analyses (under well-specified approximations): for example, the nonperturbative, generalized Smoluchowski equation for diffusing, inertial Brownian particles in a simple shear is derived (see Ref. [5]), and analytic expressions for the particle-phase total pressure tensor and the diffusion tensor are obtained (see Ref. [6]). Moreover, the methodology presented for a simple shear may be used to investigate Brownian motion of finite-inertia particle in any other linear two-dimensional flow, be it a symmetric or antisymmetric (rotational) shear.

Our approach has much in common with the so-called PDF approach used to describe particle dispersion in inhomogeneous turbulent flows [10]. Moreover, a Langevin equation formally equivalent to the one used in this work has also been used to model turbulence [11] by considering an analogy between the action of the dissipating scales of turbulence and that of the molecular white noise driving force in Brownian motion. Hence, the results obtained herein apply *mutatis mutandis* for turbulent dispersion in a simple shear with the proviso that FDT is not a property of the turbulent motion. Similarly, a Langevin equation equivalent to the one presented in this work for the fluctuating Brownian particle velocities has also been used to describe the effect of shear on fluid velocity fluctuations [12]. In the identification of similarities and differences between Brownian-particle motion and fluid-point motion the Stokes number used in this work corresponds to the dimensionless ratio of the fluid time scale to the inverse shear rate.

The Lagrangian description is presented in Sec. II where the Brownian-particle velocity autocorrelation functions are derived. Section III contains the Eulerian description and the analytic solution of the Fokker-Planck equation. The analytic expression for the total particle-phase pressure tensor as a function of the Stokes number is derived. The generalized Smoluchowski equation is calculated in Sec. IV, as well as the Green-Kubo expressions for the diffusion coefficients. The conclusions are summarized in Sec. V, whereas technical details are presented in the Appendix.

II. LAGRANGIAN DESCRIPTION

Consider, thus, a Brownian particle of mass m in a two-dimensional unbounded laminar, plane Couette shear flow—namely, a simple shear flow with the fluid velocity $\boldsymbol{u}$ along the y direction, $\boldsymbol{u}=\overleftrightarrow{\boldsymbol{\alpha}}\boldsymbol{x}$; the shear rate[1] tensor $\overleftrightarrow{\boldsymbol{\alpha}}$ has only one nonzero element $\alpha_{xy}=\alpha$. The general equation of motion for a small rigid sphere in a nonuniform flow [13] simplifies considerably for most incompressible gas-particle systems because particle density is much greater than fluid density. Accordingly, the pressure gradient force, virtual mass, Basset history integral, and Faxen's modification to Stokes' drag may be neglected. Moreover, the effect of gravity will be neglected since gravitational settling becomes significant only for very large particles ($r_p \gg 50\ \mu$m). The Saffman lift force will also be neglected, it being negligible with respect to Stokes drag for small-diameter, low-inertia particles. Therefore, the particle equations of motion in a Lagrangian description become

$$\frac{d\boldsymbol{v}}{dt} = \beta(\overleftrightarrow{\boldsymbol{\alpha}}\boldsymbol{x} - \boldsymbol{v}) + \boldsymbol{f}(t), \tag{1}$$

where the time derivative is a total derivative following the moving Brownian particle with $\boldsymbol{v}(t)$ the particle velocity, $\boldsymbol{x}(t)$ the particle position, and $\boldsymbol{f}(t)$ the random force per unit particle mass. As argued, the friction force is assumed to be the Stokes drag on the particle. Hence, the friction coefficient β is the inverse particle relaxation time, $\beta=1/\tau_p =9\mu_f/(2\rho_p r_p^2)$, the particle relaxation time being τ_p, the particle material density ρ_p, the particle radius r_p, and the fluid dynamic viscosity μ_f. As in the Langevin description of Brownian motion in a quiescent fluid the random force will be taken to be white in time,

[1]Strictly speaking α is the strain rate, but we will follow the standard practice of referring to it as the shear rate.

$$\langle f_i(t) f_j(t')\rangle = q\delta_{ij}\delta(t-t') \quad (i,j=x,y), \tag{2}$$

with zero mean $\langle f_i(t)\rangle = 0$ and of an unspecified, at the moment, strength q. Angular brackets $\langle\cdot\rangle$ denote an ensemble average over all particle trajectories.

The Langevin equations are linear: hence, the particle velocity and position may be solved formally as functionals of the random force. The formal solutions for Brownian particles injected at the origin with zero initial velocity (the choice of the initial conditions is not important since we are interested in the long-time behavior) are

$$v_x(t) = e^{-\beta t}\int_0^t dt_1 e^{\beta t_1}[\beta\alpha y(t_1) + f_x(t_1)], \quad x(t) = \int_0^t dt_1 v_x(t_1), \tag{3a}$$

$$v_y(t) = e^{-\beta t}\int_0^t dt_1 e^{\beta t_1} f_y(t_1), \quad y(t) = \int_0^t dt_1 v_y(t_1). \tag{3b}$$

The formal solution allows the analytic evaluation of ensemble averages of products of particle position and velocity, two-point correlation functions, in terms of the random-force strength q. Their evaluation requires the calculation of time integrals whose integrands include the causal correlation

$$\langle y(t) f_y(t')\rangle = \theta(t-t')\frac{q}{\beta}[1-e^{-\beta(t-t')}]. \tag{4}$$

The Heaviside θ function in the previous equation arises naturally via the explicit evaluation of the time-dependent correlation function using Eqs. (3b); it ensures causality.

In the diffusive limit $(t \gg \beta^{-1})$ exponential terms in the equal-time, two-point correlation functions may be neglected. If only polynomial terms in time are kept, the correlations evaluate to

$$\langle \boldsymbol{x}(t)\boldsymbol{x}(t)\rangle = \frac{q}{2\beta^2}\begin{pmatrix} 2t - \frac{3}{\beta} + \alpha^2\left[\frac{2}{3}t^3 - \frac{4t^2}{\beta} + \frac{8t}{\beta^2} - \frac{3}{2\beta^3}\right] & \alpha\left[t^2 - \frac{4t}{\beta} + \frac{11}{2\beta^2}\right] \\ \alpha\left[t^2 - \frac{4t}{\beta} + \frac{11}{2\beta^2}\right] & 2t - \frac{3}{\beta}\end{pmatrix}, \tag{5a}$$

$$\langle \boldsymbol{v}(t)\boldsymbol{x}(t)\rangle = \frac{q}{2\beta^2}\begin{pmatrix} \alpha^2\left[t^2 - \frac{4t}{\beta} + \frac{4}{\beta^2}\right] + 1 & \alpha\left[2t - \frac{9}{2\beta}\right] \\ \frac{\alpha}{2\beta} & 1\end{pmatrix}, \tag{5b}$$

$$\langle \boldsymbol{v}(t)\boldsymbol{v}(t)\rangle = \frac{q}{2\beta}\begin{pmatrix} \frac{2\alpha^2}{\beta}\left[t - \frac{11}{4\beta}\right] + 1 & \frac{\alpha}{2\beta} \\ \frac{\alpha}{2\beta} & 1\end{pmatrix}. \tag{5c}$$

In a quiescent fluid the fluctuation strength is specified by invoking the fluctuation-dissipation theorem. Its classical form may be expressed as (see, for example, Reeks [14])

$$\beta\delta_{ij} = \frac{m}{k_B T}\int_0^\infty ds\langle f_i(s) f_j(0)\rangle. \tag{6}$$

The use of the classical FDT in sheared systems has been questioned. Whereas its classical form as shown in Eq. (6) has been used in the past [5,9,15], it has also been argued [16] that the fluctuation strength may be determined by the *ad hoc* requirement that energy equipartition hold in the local, comoving reference frame. Energy equipartition in the local reference frame implies local equilibrium and hence the Brownian velocity distribution function becomes locally Maxwellian. As we show in Sec. III, if the Brownian velocity distribution function is a local Maxwellian distribution, then the Brownian particles behave as an ideal "particle" gas with no shear stresses or shear viscosity.

More recently, Santamaría-Holek *et al.* [6] derived a Fokker-Planck equation for the nonequilibrium distribution function of Brownian particles in stationary flow. They avoided the use of a Langevin equation (and the associated problem of the specification of the statistics of the random force) by using arguments based on mesoscopic nonequilibrium thermodynamics. They found that the flow modifies the diffusion tensor in the Fokker-Planck equation by a term proportional to the imposed velocity gradient. They concluded that due to this additional term the fluctuation-dissipation theorem in its classical form does not hold for fluctuations about the nonequilibrium steady state. However, they did not provide an estimate of the relative magnitude of the nonequilibrium correction to the classical FDT.[2] Such an estimate may be deduced from earlier kinetic theory calculations of particle motion in a nonuniform light gas. Rodríguez *et al.* [8] used Fokker-Planck and Langevin descriptions of fluctuations in uniform shear flow to conclude that the diffusion tensor in the Fokker-Planck equation and the corresponding properties of the random force in the Langevin

[2]The estimate provided in Ref. [27] refers to the complete modification of the diffusion coefficient due to the shear under conditions relevant to nucleation experiments (laminar flow diffusion cloud chamber) and not to the relative magnitude of the equilibrium-to-nonequilibrium terms.

description are modified by a term proportional to the traceless part of the fluid pressure tensor. Thus, the nonequilibrium modification of the FDT implies that in a Langevin description the stochastic properties of the random force are modified by the shear flow. However, in a Langevin description the time scale of the Brownian white noise driving force is considered much shorter than the time scale of the imposed flow, suggesting that the nonequilibrium correction would be of the order of the ratio of the molecular relaxation time to the time scale of the imposed shear.[3] In fact, the nonequilibrium correction derived by Rodríguez *et al.* [8] can be shown to be proportional to the ratio of the fluid molecular relaxation time to the time scale of the imposed shear (a ratio of a microscopic to a macroscopic time scale) by expressing the (dimensionless) correction in terms of the velocity gradient (shear rate) and the ratio of the fluid viscosity to the fluid pressure. Similarly, Fernández de la Mora and Mercer [7] used a Chapman-Enskog expansion of the light-gas velocity distribution function to show that the nonequilibrium modification of the Fokker-Planck equation is of the order of the ratio of the light-gas relaxation time ($\tau_f \sim \mu_f / p_f$) to the macroscopic fluid deceleration time (the inverse shear rate). Thus, it becomes of the order of the light-gas Knudsen number and therefore it may be neglected, only becoming important for rarefied gases [7]. Since in this work we are interested in cases where time scales are clearly separated, in what follows we shall use the classical form of the FDT that does not introduce additional random-force correlations.

The classical form of the FDT and Eq. (2) specify the fluctuation amplitude to be

$$\frac{q}{2\beta} = \frac{k_B T}{m}. \tag{7}$$

With this identification some of Eq. (5) have been previously reported [9], whereas the full time-dependent correlations for δ-function initial conditions are given in Ref. [5]. Particle velocity correlations along the y direction (in the long-time limit), being independent of the shear flow, satisfy the normal equipartition theorem. This is expected since the shear flow is only along the x direction, and the (x,y) components of the random force have been assumed uncorrelated. Moreover, a kinetic temperature, as opposed to the thermodynamic temperature T in Eqs. (5c), may be defined by relating the average particle kinetic energy to $k_B T_{\rm kin}$. Then, the kinetic temperature along the direction of the shear becomes time dependent and a function of particle mass (through the dependence on β) and of properties of the fluid flow (the local fluid gradient α).

The long-time limit of the equal time correlation $\langle y^2(t)\rangle$ may be used to obtain the Stokes-Einstein expression for the diffusion coefficient,

$$D_0 = \frac{1}{2}\frac{d}{dt}\langle y^2(t)\rangle = \frac{k_B T}{\beta m}. \tag{8}$$

Note that the diffusion coefficient is first order in β^{-1}, a result that will be used later in Sec. IV.

The shear flow also modifies the time-dependent particle-velocity autocorrelation functions. Ensemble averages of the formal solutions of the equations of motions (3) give in the diffusive limit ($t \gg \beta^{-1}$), neglecting terms involving powers of $\exp(-\beta t)$,

$$\langle v_x(t+\tau)v_x(t)\rangle = \begin{cases} \dfrac{k_B T}{m}\left\{ e^{-\beta\tau} + \dfrac{1}{2}\mathrm{St}^2[4\beta t - 8 - e^{-\beta\tau}(\beta\tau+3)]\right\} \\ \quad \text{for } \tau \geq 0, \\ \dfrac{k_B T}{m}\left\{ e^{\beta\tau} + \dfrac{1}{2}\mathrm{St}^2[4\beta(t+\tau) - 8 + e^{\beta\tau}(\beta\tau-3)]\right\} \\ \quad \text{for } \tau < 0, \end{cases} \tag{9a}$$

$$\langle v_x(t+\tau)v_y(t)\rangle = \begin{cases} 2\dfrac{k_B T}{m}\mathrm{St}\left[1 - \dfrac{1}{4}e^{-\beta\tau}(2\beta\tau+3)\right] & \text{for } \tau \geq 0, \\ \dfrac{k_B T}{m}\dfrac{1}{2}\mathrm{St}e^{\beta\tau} & \text{for } \tau < 0, \end{cases} \tag{9b}$$

$$\langle v_y(t+\tau)v_y(t)\rangle = \frac{k_B T}{m}e^{-\beta|\tau|}\ \forall\ \tau. \tag{9c}$$

Hence, the combined effects of particle inertia and shear flow modify both the amplitude of the autocorrelation functions and their time dependence. The dependence of the autocorrelation functions on particle inertia and shear rate has been expressed in terms of the Stokes number, $\mathrm{St} = \alpha/\beta$. The shear flow not only breaks spatial symmetry but also (macroscopic) time reversibility and stationarity: the particle-velocity autocorrelation function in the streamwise direction is nonstationary. Of course, the velocity correlations perpendicular to the shear decay exponentially in time as expected for a Gaussian, stationary Markov process, an Ornstein-Uhlenbeck process. The shear-induced modifications of the autocorrelation functions become more transparent if the contribution of the underlying shear flow is subtracted. Specifically, the autocorrelation functions of the fluctuating particle velocities with respect to the shear flow, $v_x''(t) = v_x(t) - \alpha y(t)$, become (in the long-time limit)

$$\langle v_x''(\tau)v_x''(0)\rangle = \frac{k_B T}{m}e^{-\beta|\tau|}\left[1 + \frac{1}{2}\mathrm{St}^2(\beta|\tau|+1)\right]\ \forall\ \tau, \tag{10a}$$

[3] Time-scale separation is also implicit in the separation of the fluid velocity into a mean part and a fluctuating part, which gives rise to the random force in the particle equations of motion (1).

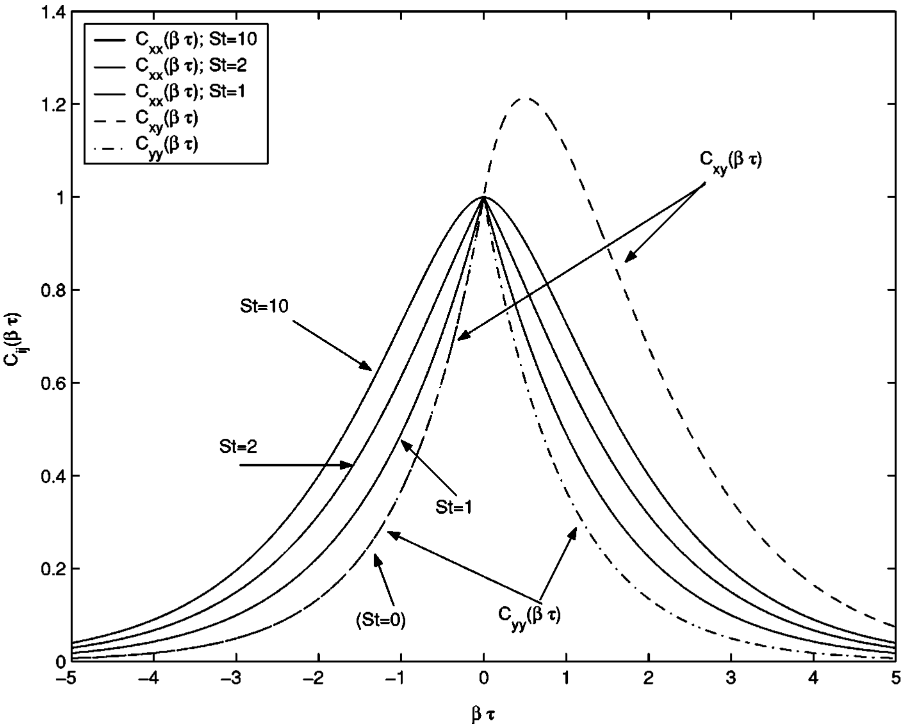

FIG. 1. Time dependence of the fluctuating particle-velocity autocorrelation functions in the diffusive limit, parametrized by the Stokes number: solid lines are $C_{xx}(\tau)$, the dashed line $C_{xy}(\tau)$, and the dash-dotted line is $C_{yy}(\tau)$.

$$\langle v_x''(\tau)v_y(0)\rangle = \begin{cases} -\dfrac{k_BT}{m}\dfrac{1}{2}\mathrm{St}e^{-\beta\tau}(2\beta\tau+1) & \text{for } \tau \geqslant 0, \\ -\dfrac{k_BT}{m}\dfrac{1}{2}\mathrm{St}e^{\beta\tau} & \text{for } \tau < 0. \end{cases} \quad (10b)$$

Thus, the autocorrelation functions of the fluctuating particle velocities are stationary and the velocity correlation along the shear is symmetric in the time difference τ, but the cross correlation is nonsymmetric in τ. Since in the local reference frame time stationarity is recovered, the dependence of the autocorrelation functions on t has been dropped: note, however, that these expressions are valid only in the long-time limit ($\beta t \gg 1$). The time decay of the velocity correlation function along the flow direction is not a pure exponential, but it is modified by an algebraic prefactor; hence, the underlying stochastic process $v_x''(t)$ is not an Ornstein-Uhlenbeck process. Furthermore, since the cross correlation is stationary, $\langle v_x''(\tau)v_y(0)\rangle = \langle v_x''(0)v_y(-\tau)\rangle$, but since it is not symmetric, $\langle v_x''(\tau)v_y(0)\rangle \neq \langle v_x''(0)v_y(\tau)\rangle$. The time asymmetry of the cross correlation implies that for a negative time difference $\tau<0$ the correlation decays exponentially, whereas for $\tau \geqslant 0$ the correlation initially increases at short relative times to decrease exponentially at longer time. The maximum occurs at $\beta\tau = 1/2$.

These qualitative observations are summarized in Fig. 1, where the three correlation functions are compared. The normalized (at the origin) autocorrelation functions are plotted, $C_{ij}(\tau) = \langle v_i''(\tau)v_j''(0)\rangle / \langle v_i''(0)v_j''(0)\rangle$, as functions of time rendered dimensionless by a β scaling. The autocorrelation along the streamwise direction $C_{xx}(t)$ has been parametrized by three values of Stokes number; the other two normalized correlations $C_{xy}(\tau)$ and $C_{yy}(\tau)$ are independent of the Stokes number. Note that with increasing Stokes number the cusp at the origin, characteristic of the short-time behavior of $C_{yy}(\tau)$, becomes rounded. The St=10 curve is also the value of $C_{xx}(\tau)$ in the limit the $\mathrm{St}\to\infty$. The velocity autocorrelation functions will be reconsidered in Sec. IV since they are related to particle diffusion coefficients through Green-Kubo relations.

Reeks [17] in an analysis of particle turbulent dispersion in a shear flow reports equations similar to Eqs. (10). It can be shown that the results of Ref. [17] for the case of fluid-point dispersion lead to velocity autocorrelation functions identical to those presented above. Similarly, Eckhardt and Pandit [12] obtained results formally equivalent to ours for the effect of shear on fluid velocity fluctuations. Note, however, that for the system under consideration the effects of the shear flow and particle inertia are described in terms of the particle Stokes number: hence, for a given shear rate changes of the Stokes number imply changes of the particle relaxation time (i.e., of the particle's inertia).

III. EULERIAN DESCRIPTION

The Eulerian description of the motion of N, independent and identical Brownian particles in terms of the average phase space density $P(\boldsymbol{x},\boldsymbol{v};t)$ is an alternative to the Lagrangian description in terms of stochastic differential equations. The Fokker-Planck equation associated with the particle equations of motion (1) may be derived in numerous ways, including the elegant functional derivation of Refs. [18,19]. It becomes

$$\frac{\partial}{\partial t}P + \frac{\partial}{\partial \boldsymbol{x}}\cdot(\boldsymbol{v}P) - \beta\frac{\partial}{\partial \boldsymbol{v}}\cdot[(\boldsymbol{v}-\boldsymbol{u})P] = \beta\frac{k_BT}{m}\frac{\partial^2}{\partial \boldsymbol{v}^2}P, \quad (11)$$

where the fluctuation amplitude, defined in Eq. (7), was used. The probability density function P is normalized on the total number of particles. As mentioned before, Eq. (11) differs from the Fokker-Planck equation derived by Santamaría-Holek *et al.* [6] in the diffusive term since we used the classical FDT.

The probability density function P is used to define average quantities. In particular, the mean, local particle velocity, a quantity defined as in kinetic theory of gases [21], is

$$\rho(\boldsymbol{x};t)\overline{\boldsymbol{v}}(\boldsymbol{x};t) = m \int d\boldsymbol{v} P(\boldsymbol{x},\boldsymbol{v};t)\boldsymbol{v} \triangleq m\langle nv \rangle, \tag{12}$$

with n the "instantaneous" particle number density and $\rho(\boldsymbol{x};t)$ the local, average particle mass density,

$$\rho(\boldsymbol{x};t) = m \int d\boldsymbol{v} P(\boldsymbol{x},\boldsymbol{v};t). \tag{13}$$

The fluctuating component of the particle velocity field, defined with respect to the mean particle velocity,[4] will be denoted by $\boldsymbol{v}' = \boldsymbol{v} - \overline{\boldsymbol{v}}$.

These local averages, denoted by an overbar and known as density-weighted averages in two-fluid descriptions of dispersed flows, are averages with respect to a local, properly normalized velocity probability density distribution

$$\psi(\boldsymbol{x},\boldsymbol{v};t) = \frac{P(\boldsymbol{x},\boldsymbol{v};t)}{\int d\boldsymbol{v} P(\boldsymbol{x},\boldsymbol{v};t)}. \tag{14}$$

The integral of ψ over particle velocities is, thus, unity.

The appropriate equations to describe particle dispersion, also known as "continuum" or "hydrodynamic" equations, are obtained by taking velocity moments of the Fokker-Planck equation. In particular, the particle mass conservation equation is obtained by multiplying Eq. (11) by m, integrating over particle velocities, and using Eq. (13). Multiplying the Fokker-Planck equation by $m\boldsymbol{v}'$ and realizing that $m\langle n\boldsymbol{v}'\boldsymbol{v}'\rangle = \rho\overline{\boldsymbol{v}'\boldsymbol{v}'}$ [see the definition Eq. (14)] gives the momentum conservation equation. Accordingly, they become (see also [18])

$$\frac{d\rho}{dt} = -\rho \boldsymbol{\nabla} \cdot \overline{\boldsymbol{v}}, \tag{15a}$$

$$\frac{d\overline{\boldsymbol{v}}}{dt} = \beta(\boldsymbol{u} - \overline{\boldsymbol{v}}) - \frac{1}{\rho}\frac{\partial}{\partial \boldsymbol{x}} \cdot (\rho \overline{\boldsymbol{v}'\boldsymbol{v}'}), \tag{15b}$$

where the total derivative $d/dt = \partial/\partial t + \overline{\boldsymbol{v}} \cdot \boldsymbol{\nabla}$ describes changes with respect to the mean particle velocity, and the dot denotes matrix multiplication. Equations (15) are a special case for a white noise random force of the general continuum equations for the dispersed phase in inhomogeneous flows derived by Reeks [10].

The momentum equation Eq. (15b) may be compared to the momentum equation that expresses momentum balance in the particulate phase [22,23]:

$$\frac{d\overline{\boldsymbol{v}}}{dt} = \beta(\boldsymbol{u} - \overline{\boldsymbol{v}}) - \frac{1}{\rho}\boldsymbol{\nabla} \cdot \overleftrightarrow{\boldsymbol{P}}_p, \tag{16}$$

where $\overleftrightarrow{\boldsymbol{P}}_p$ is the unspecified, particle-phase total pressure tensor. Comparison yields $\overleftrightarrow{\boldsymbol{P}}_p = \rho\overline{\boldsymbol{v}'\boldsymbol{v}'}$. It is easy to show [via Eq. (14)] that the expression for the particle-phase pressure tensor is identical to the pressure tensor as defined in kinetic theory [20] or in mesoscopic nonequilibrium thermodynamics [6]

$$\overleftrightarrow{\boldsymbol{P}}_p = m \int d\boldsymbol{v}(\boldsymbol{v} - \overline{\boldsymbol{v}})(\boldsymbol{v} - \overline{\boldsymbol{v}})P(\boldsymbol{x},\boldsymbol{v};t). \tag{17}$$

It is apparent from the previous discussion that the continuum equations and the identification of the particle-phase pressure tensor with the particle covariances only require that the random force be white in time. Thus, the previous expressions are valid for a general flow field and not only for a linear flow field (as long as the random force is white). In what follows we restrict the calculation to a linear flow field, eventually evaluating average quantities (for example, the mean particle velocity and the particle-phase pressure tensor) for the specific case of a simple shear flow, as discussed and justified in the Introduction.

For a linear flow field the Fokker-Planck equation defines a Gaussian process for $[\boldsymbol{x},\boldsymbol{v}]$ and thus it has an analytic solution; see, for example, Refs. [18,21]. The Gaussian solution may be used to evaluate explicitly the density-weighted ensemble averages; Swailes and Darbyshire [21] report the analytic expressions. For Brownian particles injected at the origin of the coordinate system with zero initial velocity the spatially dependent particle concentration is (Ref. [21] presents the analytic solutions for arbitrary initial conditions)

$$\rho(\boldsymbol{x};t) = \frac{1}{2\pi[\det(\langle \boldsymbol{xx}\rangle)]^{1/2}} \exp\left[-\frac{1}{2}\boldsymbol{x}^T \cdot \langle \boldsymbol{xx}\rangle^{-1} \cdot \boldsymbol{x}\right], \tag{18}$$

the mean particle velocity

$$\overline{\boldsymbol{v}}(\boldsymbol{x};t) = \langle \boldsymbol{vx}\rangle \cdot \langle \boldsymbol{xx}\rangle^{-1} \cdot \boldsymbol{x}, \tag{19}$$

and the particle-velocity covariances

$$\overline{\boldsymbol{v}'\boldsymbol{v}'}(t) = \langle \boldsymbol{vv}\rangle - \langle \boldsymbol{vx}\rangle \cdot \langle \boldsymbol{xx}\rangle^{-1} \cdot \langle \boldsymbol{xv}\rangle, \tag{20}$$

where the superscript in Eq. (18) denotes transpose. Thus, for the Gaussian process the mean particle velocity is linear in $\boldsymbol{x}$, and the particle covariances are spatially independent. Analytic expressions for the long-time behavior of the mean particle velocity and its approach to the steady-state value (the carrier flow velocity) are presented in the Appendix. For completeness note that explicit evaluation of the necessary correlation functions in the long-time limit shows that $\lim_{t\to\infty} \overline{\boldsymbol{v}'\boldsymbol{v}'} = \langle(\boldsymbol{v}-\boldsymbol{u})^2\rangle$ since $\lim_{t\to\infty}\langle \boldsymbol{v}\rangle = \boldsymbol{u}$.

The long-time particle-phase pressure tensor may then be evaluated using Eqs. (5) and (20): expressed as a function of the Stokes number, it becomes

[4]Primed variables refer to fluctuating velocities with respect to the mean particle velocity, whereas double-primed variables are defined with respect to the carrier fluid velocity; see Eqs. (10).

$$\overleftrightarrow{\boldsymbol{P}}_p = \rho\frac{k_BT}{m}\left[\left(1+\frac{\mathrm{St}^2}{4}\right)\overleftrightarrow{\boldsymbol{I}} - \frac{\mathrm{St}}{2}\begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix} + \frac{\mathrm{St}^2}{4}\begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}\right]. \tag{21}$$

The particle pressure tensor has been decomposed into a form reminiscent of the pressure tensor of a simple liquid undergoing laminar plane Couette flow [2]. The first term is isotropic and proportional to the identity tensor; the other two terms constitute what has been called [24] the "friction pressure tensor."[5] The first part of the isotropic term gives the ideal gas pressure of a collection of Brownian particles ("Brownian-particle gas"), a result that has been previously postulated on phenomenological arguments [3,4]. The ideal gas pressure is modified by a correction dependent on the Stokes number. The other terms, usually neglected, arise from the particle viscous stresses. The second term is proportional to the symmetric rate-of-strain fluid tensor, $2\overleftrightarrow{\epsilon} = \vec{\nabla}\vec{u} + (\vec{\nabla}\vec{u})^{\mathrm{T}}$. Thus, the (negative) proportionality constant defines the shear viscosity of the particle phase to be

$$\eta_p^{\mathrm{shear}} = \frac{1}{2}D_0\rho. \tag{22}$$

Hence, the Brownian shear viscosity, being independent of the shear rate, is a conventional Newtonian viscosity. An alternative expression for Eq. (22) is that the particle Schmidt number, the ratio of momentum diffusivity to mass diffusivity, is $Sc_p = (\eta_p^{\mathrm{shear}}/\rho)/D_0 = 1/2$, as is the case for particle motion in a turbulent flow in which the particle response time is much larger than the turbulent time scale [17]. The third term, which is absent in Newtonian fluids, shows that the particle phase exhibits non-Newtonian rheology, as has been remarked for sheared simple liquids [2,24]. Note that the last term, as well as the correction to the ideal gas pressure, is second order in the Stokes number. Hence to leading order in St the viscous part of the particle pressure tensor is traceless, and the particle phase behaves as a Newtonian fluid. Non-Newtonian behavior becomes evident only to second order. Moreover, according to Eq. (21) [and Eq. (8)] in the absence of diffusion, for sufficiently massive particles, $\overleftrightarrow{\boldsymbol{P}}_p = 0$.

As remarked in Sec. II if local equilibrium is assumed, as, for example, in Ref. [16], $\langle(\boldsymbol{v}-\boldsymbol{u})(\boldsymbol{v}-\boldsymbol{u})\rangle \triangleq k_BT\overleftrightarrow{\boldsymbol{1}}/m$, the Brownian particle distribution becomes locally Maxwellian. Then, from Eqs. (5c) the fluctuation strengths can be calculated to form a symmetric tensor dependent on the shear rate. As extensively argued in Sec. II the dependence of the fluctuation strengths on the shear rate implies that the externally imposed shear modifies the properties of the random force, a condition that is unlikely to hold when the relevant time scales (inverse shear rate and molecular relaxation time scale) are clearly separated. Moreover, for a local equilibrium assumption a similar calculation shows that the particle-phase pressure tensor (in the long-time limit) contracts to the ideal gas result, $\overleftrightarrow{\boldsymbol{P}}_p^{\mathrm{l.eq.}} = \rho(\boldsymbol{x};t)k_BT\overleftrightarrow{\boldsymbol{1}}/m$; i.e., there is no viscous shearing.

Equation (21) along with Eqs. (15) is our main result. These equations are analytic, nonperturbative expressions that describe the long-time behavior of the dispersed phase: they incorporate the effects of the flow field and the combined effects of particle diffusion and particle inertia on the particle-phase total pressure tensor. However, the momentum equation is coupled to the mass conservation equation. The two (steady-state) equations are, usually, decoupled by performing a low-Stokes-number expansion of the momentum equation to obtain a perturbative expression for the mean particle velocity field in terms of the fluid velocity and its gradients [4]. Substitution of the resulting mean particle velocity into the particle mass conservation equation then gives the associated convective-diffusion equation [3]. For the case of a simple shear this procedure leads to a generalized Smoluchowski equation [since the local average density is defined as an integral of the average phase-space density P, Eq. (13)], as we show in the following section.

IV. SMOLUCHOWSKI EQUATION

The momentum equation (15b), an equation valid for a general flow field and a white random force, may be rearranged to obtain an explicit expression for the total time derivative of the mean-particle-velocity field (also referred to as the inertial acceleration term). For the specific case of a simple shear differentiation of the analytic, time-dependent expression for the particle concentration Eq. (18) leads to

$$\rho\boldsymbol{x} = -\langle\boldsymbol{x}\boldsymbol{x}\rangle\cdot\frac{\partial\rho}{\partial\boldsymbol{x}}, \tag{23}$$

which substituted into the equation for the mean-particle-velocity equation (19) expresses the particle mean velocity in terms of the density gradient:

$$\overline{\boldsymbol{v}} = -\langle\boldsymbol{v}\boldsymbol{x}\rangle\cdot\frac{1}{\rho}\frac{\partial\rho}{\partial\boldsymbol{x}}. \tag{24}$$

Moreover, a variant of Eq. (23), obtained by multiplying it by the shear-rate tensor $\overleftrightarrow{\boldsymbol{\alpha}}$, determines the carrier flow velocity

$$\rho\boldsymbol{u} = -\langle\boldsymbol{u}\boldsymbol{x}\rangle\cdot\frac{\partial\rho}{\partial\boldsymbol{x}}. \tag{25}$$

Equations (24) and (25), derived from the time-dependent solution for the local particle density, are generally valid for a simple shear and not only in the long-time limit. Their substitution into the momentum equation (15b), along with the realization that for a linear flow field (and, in particular, a simple shear) the velocity covariances are spatially independent, leads to

$$\rho\frac{d\overline{\boldsymbol{v}}}{dt} = [\beta\langle(\boldsymbol{v}-\boldsymbol{u})\boldsymbol{x}\rangle - \overline{\boldsymbol{v}'\boldsymbol{v}'}]\cdot\frac{\partial\rho}{\partial\boldsymbol{x}} \triangleq \overleftrightarrow{\boldsymbol{M}}\cdot\frac{\partial\rho}{\partial\boldsymbol{x}}. \tag{26}$$

Hence, the inertial acceleration term in a simple shear may be explicitly evaluated: specifically, Eqs. (5) along with Eqs.

[5]If the momentum equation had been expressed in terms of the total particle-phase stress tensor, the negative of $\overleftrightarrow{\boldsymbol{P}}_p$ as defined by Eq. (16), then these terms would constitute the deviatoric particle stress tensor.

(7) and (20) provide analytic expressions for the necessary correlations (in the diffusive limit). Their substitution into Eq. (26) yields

$$\overleftrightarrow{\boldsymbol{M}} = \frac{k_B T}{m}\mathrm{St}\begin{pmatrix} -2\mathrm{St} & -1 \\ 1 & 0 \end{pmatrix}. \tag{27}$$

Thus, the proportionality matrix $\overleftrightarrow{\boldsymbol{M}}$ is of the order of the Stokes number; note, however, that for a simple shear the convective derivatives of the flow field $[(\boldsymbol{u}\cdot\boldsymbol{\nabla})\boldsymbol{u}]$ vanish. In this respect the simple shear differs from the symmetric and antisymmetric shear, the other possible representations of a general two-dimensional linear flow field. Equation (27) for the inertial acceleration term and Eq. (21) for the particle-phase pressure tensor were derived for the specific case of a simple shear. Hence, they may be substituted into the general, particulate-phase, momentum-conservation equation Eq. (16) or, equivalently, into Eq. (15b), rewritten as

$$\overline{\boldsymbol{v}} = \boldsymbol{u} - \frac{1}{\beta\rho}\boldsymbol{\nabla}\cdot\overleftrightarrow{\boldsymbol{P}}_p - \frac{1}{\beta}\frac{d\overline{\boldsymbol{v}}}{dt}, \tag{28}$$

to obtain the analytic, nonperturbative expression for the mean-particle-velocity field in the diffusive limit ($t \gg \beta^{-1}$):

$$\overline{\boldsymbol{v}} = \boldsymbol{u} - \langle(\boldsymbol{v}-\boldsymbol{u})\boldsymbol{x}\rangle\cdot\frac{1}{\rho}\frac{\partial\rho}{\partial\boldsymbol{x}} \tag{29}$$

$$\overset{\beta t \gg 1}{=} \boldsymbol{u} - D_0\left[\overleftrightarrow{\boldsymbol{1}} - \frac{3}{2}\mathrm{St}\begin{pmatrix} \mathrm{St} & 1 \\ -\frac{1}{3} & 0 \end{pmatrix}\right]\cdot\frac{1}{\rho}\frac{\partial\rho}{\partial\boldsymbol{x}}. \tag{30}$$

Note that Eq. (29) may also be derived by a judicious combination of Eqs. (24) and (25) as can be seen by expanding the ensemble average $\langle(\boldsymbol{v}-\boldsymbol{u})\boldsymbol{x}\rangle$. The alternative derivation presented earlier, however, is more instructive because it allows the identification of the contribution of the particle-phase pressure tensor and the inertial acceleration to the diffusion coefficient, as discussed in what follows.

To leading order in the particle relaxation time (β^{-1}) the mean particle velocity is the fluid velocity modified by a diffusion term (Fick diffusion). Fick diffusion stems from the ideal part of the particle-phase pressure tensor. The correction to Fick's law arises from the inertial acceleration and the particle viscous stresses. In fact, the contribution of the inertial acceleration term in Eq. (28) is of the same order as the contribution from the particle-phase viscous stresses. Either contribution to the mean-particle velocity is at least second order in β^{-1} [see, also, Eq. (8)]. Hence, in a simple shear it is inconsistent to retain particle viscous stresses and neglect inertial acceleration and vice versa.

The convective-diffusion equation (Smoluchowski equation) associated with the two coupled equations (15) is obtained by substituting the mean-particle-velocity equation (29) into the mass conservation equation to obtain

$$\frac{\partial\rho}{\partial t} + \boldsymbol{\nabla}\cdot(\rho\boldsymbol{u}) = \langle(\boldsymbol{v}-\boldsymbol{u})\boldsymbol{x}\rangle\cdot\frac{\partial^2\rho}{\partial\boldsymbol{x}^2}. \tag{31}$$

Equation (31) defines the diffusion tensor $\overleftrightarrow{\boldsymbol{D}}=\langle\boldsymbol{v}''\boldsymbol{x}\rangle$ that depends on both particle inertia and the shear rate through the Stokes number. In the long-time limit it is given by the second term on the right-hand side (RHS) of Eq. (30). Note that the diffusion tensor, as well as the matrix expression for the inertial acceleration term, Eq. (27), is not symmetric, reflecting the symmetry-breaking effect of the imposed shear. Therefore, the long-time generalized Smoluchowski equation becomes

$$\frac{\partial\rho}{\partial t} + \alpha y\frac{\partial\rho}{\partial x} = D_0\left(1-\frac{3}{2}\mathrm{St}^2\right)\frac{\partial^2\rho}{\partial x^2} - D_0\mathrm{St}\frac{\partial^2\rho}{\partial x\partial y} + D_0\frac{\partial^2\rho}{\partial y^2}. \tag{32}$$

Since the inertial acceleration term is antisymmetric, it only modifies the diffusion coefficient in the streamwise direction; the modification is second order in the Stokes number. The first-order correction through the appearance of the cross-derivative term arises solely from the viscous part of the particle pressure tensor. The same first-order correction to the Smoluchowski equation was derived by Subramanian and Brady [5], who, however, did not calculate higher-order corrections.

The sign of the long-time diffusion coefficient along the flow direction, D_{xx} in Eq. (32), depends on the value of the shear rate. For large shear rates it becomes negative, the critical Stokes number being $\mathrm{St}=\sqrt{2/3}$. The long-time limit of the off-diagonal diffusion coefficients is such that D_{yx} is always positive, whereas D_{xy} is always negative [See Eq. (30)]. The other diffusion coefficient D_{yy} is always positive. The dependence of the diffusion coefficient on particle inertia (via the Stokes number) is shown in Fig. 2.

The existence of negative diffusion coefficients does not violate any stability conditions since the coefficients that appear in the convective-diffusion equation refer to the particle diffusive flux with respect to the underlying carrier flow. As expected from general stability arguments the total diffusive flux defined with respect to a fixed reference frame, and not with respect to the carrier flow, is always positive. The mean particle velocity expressed in terms of the density gradient, Eq. (24), defines the total diffusion coefficients, which in the diffusive, long-time limit become

$$\overleftrightarrow{\boldsymbol{D}}_{\mathrm{tot}} \triangleq \langle\boldsymbol{v}\boldsymbol{x}\rangle = D_0\begin{pmatrix} 1+\alpha^2t^2 & 2\alpha t \\ \frac{\alpha}{\beta} & 1 \end{pmatrix}. \tag{33}$$

Therefore, the total diffusion tensor is time dependent, nonsymmetric, the corresponding matrix positive definite, and its components are always positive, thereby assuring the stability of the system. The shear-induced modification of the total diffusion coefficients, in particular the quadratic time dependence of the streamwise diffusion coefficient, has been noted before—for example, in Refs. [2,9,15]. Note that the symmetric part of $\overleftrightarrow{\boldsymbol{D}}_{\mathrm{tot}}$ gives the total diffusion coefficients as

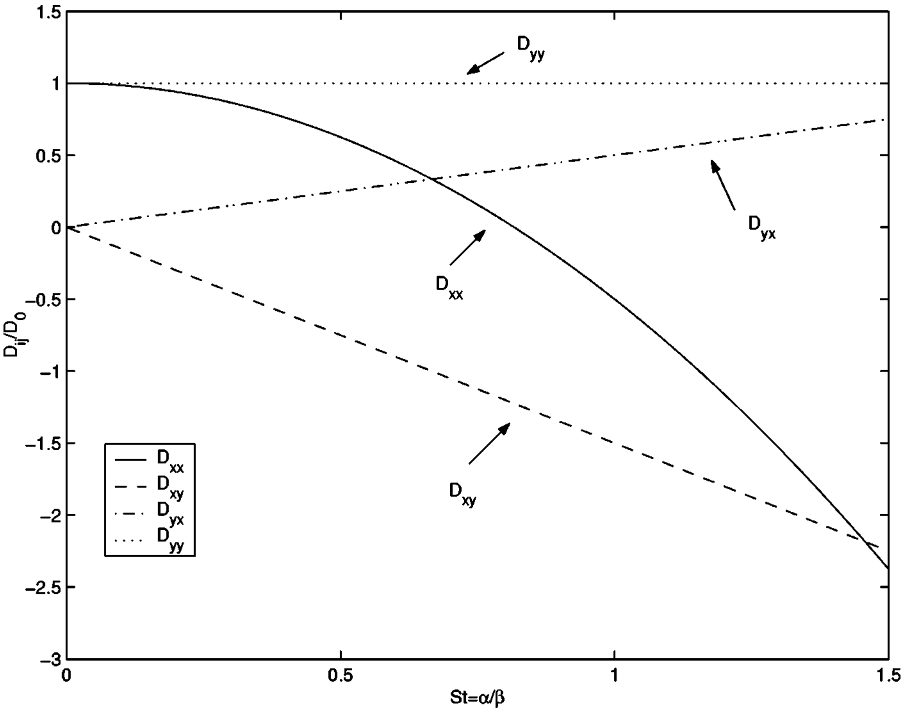

FIG. 2. Dependence of the long-time diffusion coefficients on particle inertia expressed in terms of the Stokes number. The components D_{yx} (dash-dotted line) and D_{yy} (dotted line) are always positive, D_{xy} (dashed line) always negative, whereas the sign of D_{xx} (solid line) changes as a function of the Stokes number.

determined from the rate of change of the particle mean-square displacement.

The long-time diffusion coefficients may also be obtained from the velocity autocorrelation functions presented in Sec. II. The following Green-Kubo relations, also known as Taylor's diffusion formulas for diffusion via continuous movements in theories of turbulent particle dispersion, express the diffusion coefficients in terms of the velocity autocorrelation functions:

$$\overleftrightarrow{\boldsymbol{D}}(t) = \langle \boldsymbol{v}''(t)\boldsymbol{x}(t)\rangle = \int_0^t dt_1 \langle \boldsymbol{v}''(t)\boldsymbol{v}(t_1)\rangle. \tag{34}$$

In the diffusive limit they become

$$\overleftrightarrow{\boldsymbol{D}} = \int_0^\infty dt_1 \lim_{t\to\infty}[\langle \boldsymbol{v}''(t)\boldsymbol{v}(t-t_1)\rangle], \tag{35}$$

expressions that reproduce the diffusion tensor Eq. (30) [see, also, Eq. (A5)].

Equation (35), in addition to providing the long-time diffusion coefficients in terms of velocity autocorrelations, allows a physical interpretation of the negative diffusion coefficients. Specifically, the long-time limit of D_{xy} is always negative because a particle crossing the mean shear flow with positive v_y will have negative streamwise fluctuations (as the mean flow increases with y). Along the streamwise direction D_{xx} becomes negative for large shear rates because streamwise fluctuations $v_x''=v_x-\alpha y$ become negative for large values of the shear rate α (for $v_x>0$).

The results presented in this section may be critically compared to earlier analyses of diffusive motion in nonuniform flows. We showed, via the derivation of analytic, long-time expressions, that in a simple shear the inertial acceleration term (total derivative of the mean particle velocity) and the particle-phase viscous stresses have to be treated consistently. In particular, we argued that in perturbative evaluations of, for example, the Brownian particle diffusive flux in a simple shear, it is inconsistent to retain the particle-phase viscous stresses and neglect inertial acceleration and vice versa. Previous studies differ from ours in the way these two terms ($\overleftrightarrow{\boldsymbol{P}}_p$ and $d\overline{\boldsymbol{v}}/dt$) are treated (and to a lesser degree in the choice of the underlying flow field).

Specifically, Fernández de la Mora and Rosner [4] considered the effect of inertia on diffusional deposition for a general flow field. They solved the steady-state momentum equation, Eq. (28), to leading order in β^{-1}, keeping only the ideal-gas part of $\overleftrightarrow{\boldsymbol{P}}_p$ and the leading-order contribution from the inertial acceleration term $[\beta^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla})\boldsymbol{u}]$. For a general flow field this is a consistent approximation: higher-order corrections would require an expression for the particle viscous stresses. For a simple shear their result reduces to ours to the same order $[\overline{\boldsymbol{v}}=\boldsymbol{u}-D_0\boldsymbol{\nabla}\ln\rho+O(\beta^{-2})]$ since, as noted earlier, the convective derivatives of the flow field vanish in a simple shear.

Ramshaw [3] also analyzed Brownian motion in a general flow field, but his analysis differs from ours in that particles of negligible inertia were considered. He developed a leading order in β^{-1} expansion, neglecting both viscous stresses and inertial acceleration, but keeping other forces—e.g., thermophoresis. Ramshaw [25] extended the original derivation to include phenomenologically viscous stresses in the particle and mixture (particle and fluid) momentum equations, retaining, however, the assumption of negligible particle inertia. He found that inclusion of viscous stresses in the mixture equation and evaluation of the suspension viscosity by Einstein's formula leads to an additional term in the diffusion tensor proportional to the particle-phase volume fraction. Our calculation considers an infinitely dilute suspension, and hence this additional term is absent from Eq. (30). Moreover, his calculation shows that inclusion of viscous stresses only in the particle-phase momentum equation does not modify the diffusion coefficient if the particle shear viscosity is evaluated as if the particles were an ideal gas (and, hence, the shear viscosity is independent of the particle mass density).

The analysis of Santamaría-Holek *et al.* [6], who considered diffusion in a simple shear as in this work, differs from ours primarily in the use of the nonequilibrium FDT (as summarized in Sec. II). Moreover, the diffusion tensor was evaluated perturbatively in the particle relaxation time (β^{-1}), retaining the viscous part of the particle-phase pressure tensor and neglecting the contribution from the inertial acceleration term. As shown earlier, when the classical form of the FDT is used these two terms contribute to the same order (β^{-2}) in the perturbation expansion. However, the resulting, leading-order correction to the Smoluchowski equation is identical to ours (up to the nonequilibrium modification) because the inertial acceleration (being antisymmetric) contributes only to the highest order. A similar remark applies to the leading-order correction to the Smoluchowski equation derived by Subramanian and Brady [5] (who neglected the nonequilibrium modification, as in this work).

V. CONCLUSIONS

The primary question addressed in this work is how particle inertia modifies diffusional transport of particles in a nonuniform flow. The usual convective-diffusion equation describes diffusional transport in the limit where the particles follow the fluid stream lines. It is an Eulerian continuum equation that provides a computationally efficient method to calculate particle transport (and deposition); however, inertial effects are neglected. Inertial transport, where particle trajectories deviate considerably from the fluid stream lines, is best calculated via a Lagrangian description in terms of the particle equations of motion. Diffusive particle motion may be incorporated in the Lagrangian formulation through the addition of a random force, but the numerical solution of the resulting stochastic differential equations is computationally intensive. It is, thus, desirable to derive a continuum equation valid in the transition regime between the diffusion limit and the inertia-dominated limit, incorporating both particle-transport mechanisms.

We considered the coupled diffusive and inertial motion of noninteracting Brownian particles in a simple inhomogeneous fluid flow, a simple shear flow. The long-time, diffusive, behavior of the system was investigated neglecting the short-time regime. Even though the choice of a simple shear as the underlying carrier flow is restrictive [for example, the convective derivatives of the flow vanish, $(\boldsymbol{u}\cdot\boldsymbol{\nabla})\boldsymbol{u}=0$, a condition that does not hold for a symmetric or antisymmetric shear] the choice was motivated by numerous previous investigations of Brownian motion in such a flow. More importantly, the Fokker-Planck equation associated with the particle equations of motion in a linear flow field is of the linear type [8] and, hence, analytically solvable. The solution of the Fokker-Planck equation was used to obtain analytic expressions for average particle properties—for example, the mean particle velocity and particle-velocity correlations. These analytic solutions depend on equal-time ensemble averages that were determined from the formal solutions of the Langevin equations for particle motion. The classical form of the fluctuation-dissipation theorem was used to determine the strength of the random force correlations, neglecting corrections of the order of the ratio of the fluid molecular relaxation time to the time scale of the imposed shear. We showed that the long-time, time-dependent particle-velocity autocorrelation along the streamwise direction is nonstationary. The fluctuating velocity autocorrelations were determined to be stationary in time, but the cross correlation was nonsymmetric in the time difference, reflecting the combined effect of particle inertia and shear on particle-velocity fluctuations.

We used the analytic solution of the Fokker-Planck equation, in conjunction with its first two-velocity-moment equations, to obtain [in the diffusive limit ($t \gg \beta^{-1}$)] the generalized convective-diffusion equation (generalized Smoluchowski equation) that incorporates inertial effects on diffusional transport for dilute suspensions. The coupling of particle inertia to the fluid flow introduces a shear-dependent, linear in (particle) Stokes number, cross-derivative term and an additional term along the streamwise direction, quadratic in the Stokes number. The associated diffusion tensor, thus, depends on the shear rate and particle inertia; the diffusion coefficient along the streamwise direction flow was found to become negative for large particle relaxation times (or, equivalently, for large shear rates), whereas one of the cross-diffusion coefficient was determined to be always negative, the other two being always positive. We argued that stability conditions are not violated since the total diffusion coefficients (not those with respect to the carrier flow) that measure the rate of change of particle mean-square displacement were determined to be always positive.

We showed that in a simple shear and in the long-time limit the contribution of the inertial acceleration term to the diffusion tensor in the generalized Smoluchowski equation is of the same order in the Stokes number as that of the viscous part of the particle-phase pressure tensor. Thus, in perturbative evaluations of, for example, the Brownian particle diffusive flux, it is inconsistent to retain the particle-phase viscous stresses and neglect the inertial acceleration term and vice versa.

As part of the derivation of the Smoluchowski equation we calculated the particle-phase total pressure tensor that was determined to be second order in the Stokes number. Similar to the pressure tensor of simple sheared liquids, the pressure tensor was decomposed into three parts: a part proportional to the identity tensor that gives the ideal pressure of a gas of Brownian particle with an additional term due to particle inertia, and two terms that arise from the particle viscous stresses. We found that the particle phase has a conventional shear viscosity, but it behaves as a non-Newtonian fluid if second-order effects in the Stokes number are considered.

These results will be extended to other two-dimensional linear flows in future work.

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APPENDIX: APPROACH TO THE STEADY STATE

The time-dependent, equal-time correlation functions in conjunction with Eq. (19) [or Eq. (24)] may be used to calculate the approach of the mean particle velocity to its steady-state value. The required calculations were performed with MATHEMATICA [26]. Accordingly, the mean particle velocity approaches the carrier fluid velocity as follows:

$$\lim_{t\to\infty} \overline{v_x} = \alpha y\left(1 - \frac{3}{\beta t}\right) + \left[\frac{9x}{2\beta} - \frac{3y}{\alpha}\left(1 - \frac{3}{2}\mathrm{St}^2\right)\right]t^{-2} + O(t^{-3}), \tag{A1a}$$

$$\lim_{t\to\infty} \overline{v_y} = \frac{2y}{t} - \frac{3}{\alpha}\left(x + \frac{\mathrm{St}}{2}y\right)t^{-2} + O(t^{-3}). \tag{A1b}$$

Note that the long-time limit and the vanishing shear-rate limit do not commute since

$$\lim_{t\to\infty}(\lim_{\alpha\to 0} \overline{v_x}) = \frac{x}{2t}\left(1 + \frac{3}{2\beta t}\right) + \frac{3y}{4}\left[1 - \frac{2}{3\beta t} - \frac{19}{12(\beta t)^2}\right]\alpha + O(\alpha^2), \tag{A2a}$$

$$\lim_{t\to\infty}(\lim_{\alpha\to 0} \overline{v_y}) = \frac{y}{2t}\left(1 + \frac{3}{\beta t}\right) - \frac{x}{4}\left[1 - \frac{2}{\beta t} - \frac{5}{4(\beta t)^2}\right]\alpha + O(\alpha^2). \tag{A2b}$$

The zeroth-order terms of Eqs. (A2) give the mean particle velocity of Brownian particles in a quiescent fluid.

The time-dependent approach of the mean particle velocity to its steady-state value may be used to estimate the relative importance of the two terms in the total derivative of the mean particle velocity: namely, $\partial/\partial t$ and $(\overline{\boldsymbol{v}}\cdot\boldsymbol{\nabla})$. Appropriate differentiation of Eqs. (A1), along with

$$\frac{\partial \log \rho}{\partial x} = \frac{1}{\mathrm{St}}\frac{m}{k_B T}\frac{3y}{t^2} + O(t^{-3}), \tag{A3a}$$

$$\frac{\partial \log \rho}{\partial y} = -\frac{2y}{D_0 t} + O(t^{-2}), \tag{A3b}$$

shows that the explicit time derivative vanishes in the long-time limit (to leading order in t^{-1}) to give $\rho(\overline{\boldsymbol{v}}\cdot\boldsymbol{\nabla})\overline{\boldsymbol{v}} = \overleftrightarrow{\boldsymbol{M}}\cdot\partial\rho/\partial\boldsymbol{x}$. Hence, the leading-order contribution to the total derivative does not arise from the explicit time derivative.

For completeness we also present the approach to steady-state values of the particle-velocity covariances

$$\lim_{t\to\infty} \overline{v_x' v_x'} = \frac{k_B T}{m}\left[1 + \mathrm{St}^2\left(\frac{1}{2} - \frac{9}{2\beta t}\right)\right], \tag{A4a}$$

$$\lim_{t\to\infty} \overline{v_x' v_y'} = -\frac{k_B T}{2m}\mathrm{St}\left(1 - \frac{6}{\beta t}\right), \tag{A4b}$$

$$\lim_{t\to\infty} \overline{v_y' v_y'} = \frac{k_B T}{m}\left(1 - \frac{2}{\beta t}\right). \tag{A4c}$$

The evaluation of the streamwise diffusion coefficient D_{xx} via the Green-Kubo relations (35) requires the nonstationary correlation function

$$\langle v_x(t-\tau)y(t)\rangle = 2\frac{k_B T}{m}\mathrm{St}\left(t - \tau\frac{2}{\beta} - \frac{1}{4\beta}e^{-\beta\tau}\right) \text{ for } \tau \geqslant 0. \tag{A5}$$

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