Fokker–Planck and Langevin Descriptions of Fluctuations in Uniform Shear Flow[1]

Rosalío F. Rodríguez,[2,3] E. Salinas-Rodríguez,[3] and James W. Dufty[2,4]

The Boltzmann description of the preceding paper for tagged particle fluctuations in a nonequilibrium gas is further analyzed in the limit of small mass ratio between the gas and the tagged particles. For a large class of nonequilibrium states the Boltzmann–Lorentz collision operator for the tagged particle distribution is expanded to leading order in the mass ratio, resulting in a Fokker–Planck operator. The drift vector and diffusion tensor are calculated exactly for Maxwell molecules. The Fokker–Planck operator depends on the nonequilibrium state only through the hydrodynamic variables for the fluid. The diffusion tensor is a measure of the "noise" amplitude and is not simply determined from the nonequilibrium temperature; instead, it depends on the fluid stress tensor components as well. For the special case of uniform shear flow, the Fokker–Planck equation is of the linear type and may be solved exactly. The associated set of Langevin equations is also identified and used to describe spatial diffusion in the Lagrangian coordinates of the fluid. The effect of viscous heating on diffusion is discussed and the dependence of the diffusion coefficient on the shear rate is calculated.

1. INTRODUCTION

The equations for transport and fluctuations of a tagged particle in a nonequilibrium gas were described in the preceding paper.[1] The kinetic equation for the tagged particle is characterized by a Boltzmann–Lorentz

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[2] Thermophysics Division, National Bureau of Standards, Washington, D.C. 20234.
[3] Permanent address: Departamento de Física, Facultad de Ciencias, UNAM, 04510 México D.F., México.
[4] Permanent address: Department of Physics, University of Florida, Gainesville, FL 32611.

collision operator as a functional of the fluid distribution function, and the tagged particle was taken to be mechanically identical to the fluid particles. This description is extended here to the case of unequal masses for the tagged and fluid particles. Specifically, the tagged and fluid particles are assumed to interact via the same force law as holds between fluid particles, but the mass of the tagged particle, M, is considered to be large compared to that of the fluid particle, m. The mass ratio, $\epsilon = m/M$, is therefore a small parameter in terms of which the Boltzmann–Lorentz operator may be expanded. This expansion is carried out to leading order, $\epsilon^{1/2}$, resulting in a second-order differential operator of the Fokker–Planck form. The drift vector and diffusion tensor in this operator are calculated exactly for Maxwell molecules. A special feature of the Maxwell potential is that the Fokker–Planck operator depends on the nonequilibrium state of the gas only through low-order moments of the fluid distribution function, which can be identified in terms of the hydrodynamic variables and irreversible fluxes of the nonequilibrium gas. In particular, the diffusion tensor is proportional to the components of the pressure tensor. This implies that the "noise" in the dynamics of the tagged particle is not simply thermal as in equilibrium. The Langevin description associated with the Fokker–Planck equation is also identified to emphasize this difference between equilibrium and nonequilibrium fluctuations.

In the case of a fluid with uniform shear flow, the Fokker–Planck equation is of the "linear" type. The fluctuations and time correlation function for position and velocity of the tagged particle are easily determined from either the Fokker–Planck equation or the (linear) Langevin equations, in the same way as for equilibrium fluctuations.[2] The results agree with those of Reference 1 obtained directly from the Boltzmann–Lorentz equation. An advantage of the Fokker–Planck description is that the distribution function and joint probability density for the tagged particle may be determined exactly, in contrast to the more general Boltzmann–Lorentz description for which only moments of these functions are tractable. Furthermore, since the "linear" Fokker–Planck equation corresponds to a Gaussian–Markovian process, all multipoint probability densities may be expressed in terms of the distribution function and joint probability density. This provides an example of a system far from equilibrium for which all statistical properties may be calculated exactly. The solutions to both the Fokker–Planck equation and the Langevin equations are given in Section 3. The general results are difficult to interpret owing to the combined effects of anisotropy induced by the shear field and the viscous heating. Consequently attention is focused on the reduced distribution function for spatial coordinates of the tagged particle, referred to the Lagrangian frame of the fluid. The diffusion coefficient is calculated as a function of the shear rate and the nonequilibrium temperature.

A limiting form of the Langevin equations derived here has been considered by San Miguel and Sancho.[3] Also, a similar discussion of the Fokker–Planck equation for a nonequilibrium gas has just been published by Fernandez de la Mora and Mercer.[4] The relationship of the present work to these others is discussed in the last section.

2. FOKKER–PLANCK AND LANGEVIN EQUATIONS

The kinetic equations for transport and fluctuations of a tagged particle in a nonequilibrium gas have the form

$$\left(\frac{\partial}{\partial t} + \mathbf{v} \cdot \nabla \right) F = J[f, F] \tag{2.1}$$

where f is the solution to the nonlinear Boltzmann equation for the gas and F denotes either the tagged particle distribution, h, or the joint distribution for tagged particle fluctuations in phase space, C [see Eq. (2.1) of Ref. 1 for the precise definition]. The operator $J[f, \cdot]$, is the Boltzmann–Lorentz operator as a functional of the fluid distribution, f. This operator is implicitly a function of the masses of the fluid and tagged particles, and for small mass ratio $\epsilon = m/M \ll 1$, describes the small velocity changes of the massive tagged particle due to collisions with the light fluid particles. An expansion of $J[f, \cdot]$, to lowest order in ϵ is carried out in Appendix A with the result

$$J[f, F] \rightarrow \frac{\partial}{\partial v_1} \left[A_i(\mathbf{r}, \mathbf{v}, t) + \frac{1}{2} \frac{\partial}{\partial v_j} D_{ij}(\mathbf{r}, t) \right] F \tag{2.2}$$

The drift vector, A_i, and diffusion tensor, D_{ij}, are given by

$$\mathbf{A}(\mathbf{r}, \mathbf{v}, t) = \nu_1 \frac{n(\mathbf{r}, t)}{n_0} (\mathbf{v} - \mathbf{U}(\mathbf{r}, t)) \tag{2.3}$$

$$D_{ij}(\mathbf{r}, t) = 2\rho_0^{-1} \left[\nu_1 p(\mathbf{r}, t) \delta_{ij} + (\nu_1 - \nu_2) t_{ij}^*(\mathbf{r}, t) \right] \tag{2.4}$$

Here, ν_1 and ν_2 are constants proportional to $\epsilon^{1/2}$, and $n(\mathbf{r}, t)$, $p(\mathbf{r}, t)$, and $U(\mathbf{r}, t)$ are the local density, pressure, and flow velocity of the fluid. The constant n_0 is the average number density, and $\rho_0 = Mn_0$. Also, $t_{ij}^*(\mathbf{r}, t)$ is the traceless part of the fluid pressure tensor and represents the irreversible part of the momentum flux. A complete description of the tagged particle dynamics for small ϵ is therefore provided by the kinetic equation

$$\left(\frac{\partial}{\partial t} + \mathbf{v} \cdot \nabla \right) F = \frac{\partial}{\partial v_i} \left[A_i(\mathbf{r}, \mathbf{v}, t) + \frac{1}{2} \frac{\partial}{\partial v_j} D_{ij}(\mathbf{r}, t) \right] F \tag{2.5}$$

The initial conditions must be specified, together with the state of the fluid. An interesting anomaly of the Maxwell potential is that only low-order moments of the fluid distribution function occur in the Fokker–Planck

limit. As a consequence the drift vector and diffusion tensor depend on only the hydrodynamic state of the fluid, and not on some more detailed features of the distribution, f. This simplification does not occur for any other force law, except as an approximation.

For an equilibrium fluid Eq. (2.5) reduces to the usual description of tagged particle dynamics.[2] Most of the differences associated with a nonequilibrium state are easily understood. The appearance of the flow field in the drift vector is simply due to the fact that the collisional damping of the tagged particle is due to its velocity *relative* to the local fluid velocity. Similarly, the fluid density occurs because the collision rate is proportional to the local number of scatterers surrounding the tagged particle. The first term in Eq. (2.4) for the diffusion tensor is also a straightforward general-ization of the equilibrium diffusion tensor, $D_{ij}^{(0)} = (2\nu_1 k_B T/M)\delta_{ij}$, where T is the temperature. All of these changes are associated with the replacement of the equilibrium distribution for the fluid by a local equilibrium distribu-tion. These local equilibrium modifications are at the basis of several models for the extension of equilibrium theories of fluctuations to nonequi-librium states.[5] In some cases these local equilibrium models can be justified as an approximation for states near equilibrium. In other cases they are simply not valid.[6] The system considered here is an example of the latter, due to the second term in Eq. (2.4), proportional to t_{ij}^*. This not only makes the diffusion tensor anisotropic, but also implies a source of fluctuations other than that characterized by a local equilibrium ensemble. The relative importance of these contributions is illustrated in Fig. 2 of Ref. 1. In the non-Newtonian range the deviations from local equilibrium are typically 10% with a maximum of 44%.

The Fokker–Planck Eq. (2.5) applies for an arbitrary nonequilibrium state of the fluid. The form of the equation is deceptively simple, however. Even if the fluid variables are known functions of $\mathbf{r}$ and t, the Fokker–Planck operator is generally of the "nonlinear" type,[5] and very little can be extracted exactly except in special cases. For example, the average equa-tions of motion of the tagged particle are obtained from the position and velocity moments of Eq. (2.5),

$$\frac{\partial}{\partial t} \langle \mathbf{r} \rangle = \langle \mathbf{v} \rangle$$

$$\frac{\partial}{\partial t} \langle \mathbf{v} \rangle = -\nu_1 n_0^{-1} \langle n(\mathbf{r}, t)(\mathbf{v} - \mathbf{U}(\mathbf{r}, t)) \rangle \tag{2.6}$$

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[5] The Fokker–Planck operator is clearly a linear differential operator over its domain of definition. However, the terminology "linear" and "nonlinear" is often used to distinguish the cases in which the equations for the first moments are linear and nonlinear. More precisely, the Fokker–Planck operator is "linear" if the drift vector is linear in the dependent variables (excluding time) and the diffusion tensor is constant; otherwise it is "nonlinear."[7]

These are not closed equations due to the correlation between the velocity and position dependence of the fluid variables. This leads to the difficult problem of "fluctuation renormalization".[7,8] In the limit of small diffusion tensor a systematic expansion about the deterministic motion,

$$\frac{\partial}{\partial t} \langle \mathbf{v} \rangle = - \nu_1 n_0^{-1} n(\langle \mathbf{r} \rangle, t)(\langle \mathbf{v} \rangle - \mathbf{U}(\langle \mathbf{r} \rangle, t)) \tag{2.7}$$

may be possible,[7,8] but otherwise very little is known.[9] In the remainder of this paper the discussion is limited to a fluid with uniform shear flow, for which there are significant simplifications. The conditions of uniform shear flow are defined by Eqs. (2.8), (3.2), and (3.3) of Ref. 1. Since the density, pressure, and stress tensor are spatially uniform, the Fokker–Planck equation reduces to

$$\left(\frac{\partial}{\partial t} + \mathbf{v} \cdot \nabla \right) F = \frac{\partial}{\partial v_i} \left[\nu_1(v_i - U_i(\mathbf{r})) + \frac{1}{2} \frac{\partial}{\partial v_j} D_{ij}(t) \right] F \tag{2.8}$$

The diffusion tensor is now independent of $\mathbf{r}$ and $\mathbf{v}$:

$$D_{ij}(t) = 2\nu_1 \frac{k_B T(t)}{M} \delta_{ij} + 2(\nu_1 - \nu_2)\rho_0^{-1} t_{ij}^*(t) \tag{2.9}$$

Also, since the flow field is linear in r,

$$U_i(r) = a_{ij} r_j \tag{2.10}$$

where a_{ij} is the shear rate tensor, the Fokker–Planck equation, (2.8), is of the linear type. The equations of motion corresponding to (2.6) are now closed and linear. Furthermore, Eq. (2.8) defines a Gaussian–Markovian process so that all multipoint distribution functions are determined explicitly from the Green's functions for (2.8) (or, equivalently, the two-point conditional probability density). The latter is Gaussian in $\mathbf{r}, \mathbf{v}$ and easily determined by standard methods.[2] Finally, the fluid state in this case is specified by the nonequilibrium temperature, $T(t)$, and irreversible stress tensor, $t_{ij}^*(t)$. As shown in the appendix of Ref. 1 these may be calculated exactly from the nonlinear Boltzmann equation, and therefore all parameters of the Fokker–Planck operator are specified.

Associated with the linear Fokker–Planck equation is an equivalent set of Langevin equations for the same Gaussian–Markovian process,[6]

$$\frac{\partial \mathbf{r}}{\partial t} = \mathbf{v}$$

$$\frac{\partial \mathbf{v}}{\partial t} + \nu_1(\mathbf{v} - \mathbf{U}(\mathbf{r})) = \gamma \tag{2.11}$$

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[6] There is also a set of nonlinear Langevin equations associated with the nonlinear Fokker–Planck equation, (2.5). van Kampen's objections to such a relationship do not apply to the particular form obtained here (see the conditions on p. 246 of Ref. 7).

Here γ is a stochastic vector representing the fluctuations of r and v about their average motion. From the Fokker–Planck equation it follows that γ is Gaussian distributed with the properties

$$\overline{\gamma_i(t)} = 0$$

$$\overline{\gamma_i(t)\gamma_j(t')} = D_{ij}(t)\delta(t - t') \qquad (2.12)$$

$$\overline{v_i(t)\gamma_j(t')} = 0, \qquad \text{for} \quad t < t'$$

The bar over the variables on the left side of Eqs. (2.9) denotes an average over the distribution for γ. Again, the most notable feature of these equations for the nonequilibrium fluid is the modification of the noise amplitude from $2(\nu_1 k_B T/M)\delta_{ij}$ to $D_{ij}(t)$.

3. SOLUTIONS TO FOKKER–PLANCK AND LANGEVIN EQUATIONS

The Langevin equations are a set of coupled linear first-order differential equations and therefore straightforward to solve in terms of the stochastic force, γ. Let z be a six-dimensional vector whose elements are the position and the velocity of the tagged particle,

$$z \leftrightarrow (\mathbf{r}, \mathbf{v}) \qquad (3.1)$$

Then the Langevin equations may be written as

$$\left(\frac{\partial}{\partial t} + \mathscr{L} \right) z = \Gamma \qquad (3.2)$$

where the matrices $\mathscr{L}$ and Γ are

$$\mathscr{L} \equiv \left[\begin{array}{c|c} 0 & -\overset{\leftrightarrow}{I} \\ \hline -\nu_1\overset{\leftrightarrow}{a} & \nu_1\overset{\leftrightarrow}{I} \end{array} \right], \qquad \Gamma \equiv \begin{pmatrix} 0 \\ \overset{\rightarrow}{\gamma} \end{pmatrix} \qquad (3.3)$$

and the noise spectrum is given by

$$\overline{\Gamma(t)} = 0$$

$$\overline{\Gamma(t)\Gamma(t')} = \mathscr{D}(t)\delta(t - t') \qquad (3.4)$$

$$\overline{z(t)\Gamma(t')} = 0, \qquad \text{for} \quad t < t'$$

with,

$$\mathscr{D} \equiv \begin{pmatrix} 0 & 0 \\ \hline 0 & \overset{\leftrightarrow}{D} \end{pmatrix} \qquad (3.5)$$

The abbreviated notation here is such that $\overset{\leftrightarrow}{I}$, $\overset{\leftrightarrow}{a}$, and $\overset{\leftrightarrow}{D}$ are the 3×3 matrices whose elements are $\delta_{ij}, a_{ij}, D_{ij}$, respectively. The solution to the

Langevin equations is therefore

$$z(t) = e^{-t\mathscr{L}}z(0) + \int_0^t ds\, e^{-\mathscr{L}(t-s)}\Gamma(s) \tag{3.6}$$

The matrix of correlations between positions and velocities,

$$G(t,\tau) = \langle z(t+\tau)z(\tau)\rangle \tag{3.7}$$

is then easily calculated from the properties (3.4). The results agree with those from the Boltzmann–Lorentz equation in Ref. 1. In particular the source term in the equation for equal time fluctuations [Eq. (4.17) of Ref. 1) is the same matrix $\mathscr{D}$ as defined in Eq. (3.5), so its interpretation there as the noise amplitude is confirmed.

The conditional probability density, or Green's function for the Fokker–Planck equation, is defined by

$$P(x,t\,|\,x_0,t_0) = \langle\delta(x-z(t))\delta(x_0-z(t_0))\rangle/\langle\delta(x_0-z(t_0))\rangle \tag{3.8}$$

The distribution function and joint probability distribution are given in terms of P by

$$h(x,t) = \int dx_0 h(x,t;x_0,t_0)$$

$$h(x,t;x_0,t_0) = P(x,t\,|\,x_0,t_0)h(x_0,t_0) \tag{3.9}$$

Since $P(x,t\,|\,x_0,t_0)$ is Gaussian it is entirely determined by the correlation matrix G, Eq. (3.7). Equivalently, a direct solution to the Fokker–Planck equation with initial conditions appropriate for (3.8) leads to

$$P(x,t+\tau\,|\,x_0,\tau) = \frac{1}{(2\pi)^3\|M\|^{1/2}}\exp\left[-\tfrac{1}{2}(YM^{-1}Y)\right] \tag{3.10}$$

with

$$Y = x - e^{-t\mathscr{L}}x_0$$

$$M = \int_0^t ds\, e^{-s\mathscr{L}}\mathscr{D}(t+\tau-s)e^{-s\mathscr{L}^T} \tag{3.11}$$

Also, M^{-1} denotes the inverse of M and $\|M\|$ its determinant. It is straightforward to evaluate the time integral and matrix products in Eq. (3.10). However, the general results are quite complex functions of the shear rate and not particularly instructive. Instead, the corresponding reduced distribution function in coordinate space is considered in more detail in the next section.

4. DIFFUSION IN SHEAR FLOW

The coordinate space distribution function for a tagged particle in an equilibrium fluid obeys a diffusion equation for $\nu_1 t \gg 1$. To see the effect of

uniform shear flow on this diffusion process it is useful to separate motion due primarily to convection of the particle by the fluid from that associated with collisions. The convective flow can be eliminated by considering the motion relative to the Lagrangian coordinate system of the fluid. For example, the solution to the coordinate part of the Langevin equation

$$\mathbf{r} = \mathbf{r}_0 + \int_0^t ds\, \mathbf{v}(s)$$

may be written as

$$\mathbf{q} = \int_0^t ds\, \mathbf{v}'(s) \tag{4.1}$$

where $\mathbf{q}$ is the Lagrangian coordinate of the system and $\mathbf{v}'$ is the relative velocity,

$$\mathbf{q} = \mathbf{r} - \mathbf{r}_0 - \int_0^t ds\, \mathbf{U}(r(s)), \qquad \mathbf{v}' = \mathbf{v} - \mathbf{U}(r) \tag{4.2}$$

Then, since $\mathbf{v}'$ is Gaussian distributed, the conditional probability distribution for the Lagrangian coordinate of the tagged particle is

$$P(\mathbf{q}, t \,|\, \mathbf{0}, 0) = (2\pi)^{-3} \int d\mathbf{k}\, e^{i\mathbf{q}\cdot\mathbf{k}} \exp\left[-\tfrac{1}{2} k_i k_j \int_0^t d\tau \int_0^t ds\, R_{ij}(\tau - s, s) \right] \tag{4.3}$$

where $R_{ij}(t, \tau)$ is the velocity autocorrelation function in the rest frame,

$$R_{ij}(t, \tau) = \langle v_i'(t + \tau) v_j'(\tau) \rangle \tag{4.4}$$

It is easily verified by direct differentiation that $P(q, t \,|\, 0, 0)$ satisfies the exact equation,

$$\frac{\partial P}{\partial t} = L_{ij}(t) \frac{\partial^2}{\partial q_i \partial q_j} P \tag{4.5}$$

with $L_{ij}(t)$ given by

$$L_{ij}(t) = \int_0^t ds\, R_{ij}(t - s, s) \tag{4.6}$$

The rest frame velocity autocorrelation function, $R_{ij}(t - s, s)$, has been calculated in Ref. 1 [Eq. (5.17)] with the result

$$R_{ij}(t - s, s) = e^{-\nu_1 t}(\delta_{ik} - a_{ik}t) R_{kj}(0, s) \tag{4.7}$$

Equation (4.6) then gives

$$L_{ij}(t) + \int_0^t ds\, e^{-\nu_1(t - s)}\left[\delta_{ik} - a_{ik}(t - s) \right] R_{kj}(0, s) \tag{4.8}$$

This may be rewritten in the suggestive form

$$L_{ij}(t) = D(T(t)) \alpha_{ij}(\nu_1 t) \tag{4.9}$$

where $D(T(t))$ is the diffusion coefficient for a tagged particle in an equilibrium field, as a function of the temperature at the time of interest,

$$D(T(t)) = \frac{k_B T(t)}{M\nu_1} \tag{4.10}$$

and $\alpha_{ij}(\nu_1 t)$ is defined by

$$\alpha_{ij}(\nu_1 t) = \int_0^{\nu_1 t} ds\, e^{-s} (\delta_{ik} - a_{ik}s/\nu_1)\rho R_{kj}(0, t - s/\nu_1)/p(t) \tag{4.11}$$

Here, $p(t)$ is the pressure.

In Ref. 1 the equal time correlation function, $R_{ij}(0, s)$ was shown to be simply proportional to the pressure tensor. Such a relationship was valid as a consequence of the mass ratio being equal to 1, and consequently does not hold here. However, $R_{ij}(0, s)$ is readily calculated from the Langevin equations, (2.12), expressed in terms of the relative velocity v'. The calculation of $\alpha_{ij}(\nu, t)$ for large $\nu_1 t$ (the diffusion limit) is outlined in Appendix B, with the result

$$\lim_{\nu_1 t \gg 1} \alpha_{ij}(t) \equiv \alpha_{ij} = A_1\delta_{ij} + A_2 a_{ij} + A_3 a_{ji} + A_4 a_{ik}a_{jk} \tag{4.12}$$

The constants, A_i, are scalar functions of the magnitude of the shear rate $(a_{ij}a_{ij})^{1/2}$ and are also given in Appendix B. The probability density therefore obeys the diffusion equation [the Smoluchowski equation associated with Eq. (2.8) in Lagrangian coordinates]

$$\frac{\partial P}{\partial t} = L_{ij}(T(t)) \frac{\partial^2}{\partial q_i \partial q_j} P \tag{4.13}$$

with the diffusion coefficient given by

$$L_{ij}(T(t)) = \alpha_{ij} D(T(t)) \tag{4.14}$$

This equation must be supplemented with the equation for the temperature, $T(t)$, which for $\nu_1 t \gg 1$ is simply Eq. (A.35) of Ref. 1,

$$\frac{\partial T(t)}{\partial t} = z_1(a)T(t) \tag{4.15}$$

where $z_1(a)$ is a positive function of the shear rate given by Eq. (A.33) of Ref. 1.

There are essentially two modifications of the diffusion coefficient due to shear flow. The first is an anisotropy due to the fluid anisotropy as reflected through the pressure tensor and an inertial force on the particle in the Lagrangian frame [see Eq. (B.3)]. The second modification is due to viscous heating which appears both in the time dependence of the temperature and in the shear rate dependence of the coefficients in Eq. (4.12). The effect of the heating can be formally suppressed by a nonlinear transforma-

tion to a new time variable,

$$\tau \equiv \frac{1}{z_1}(e^{z_1 t} - 1) \tag{4.16}$$

In terms of τ the diffusion equation becomes

$$\frac{\partial}{\partial \tau} P = L_{ij}(T(0)) \frac{\partial^2 P}{\partial q_i \partial q_j} \tag{4.17}$$

The solution to this equation is

$$P(\mathbf{q}_1 \tau \,|\, \mathbf{0}, 0) = \frac{1}{\left[(2\pi\tau)^3 \|L\| \right]^{1/2}} \exp\left\{ -q_i q_j L_{ij}^{-1}/4\tau \right\} \tag{4.18}$$

Thus, in the Lagrangian space coordinates of the fluid and with respect to a time variable adjusted to the heating rate, the diffusion process differs from that in equilibrium only by the anisotropy of the diffusion coefficient. The effect of the heating depends on both the time of interest and the shear rate. Figure 1 illustrates the deviation of τ from t over the range $0 \leqslant \nu_1 t \leqslant 2$ for several values of the shear rate, and $\epsilon = 0.1$. The time dependence of the temperature can lead to qualitative changes in the solution to the bilinear diffusion equation. For example, the heating here is exponential so that the mean square displacement of the tagged particle is linear in τ, but exponential in t.

The effect of the factor α_{ij} on the diffusion coefficient is to decrease it relative to its "local equilibrium" value, $D(T(t))$. This is illustrated in
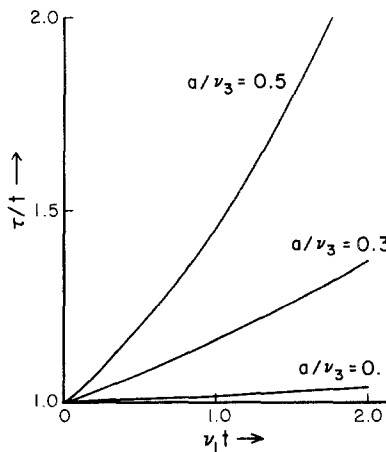

Fig. 1.   Relationship between the times t and τ as given by Eq. (4.16) for several values of the shear rate and $\epsilon = 0.1$.
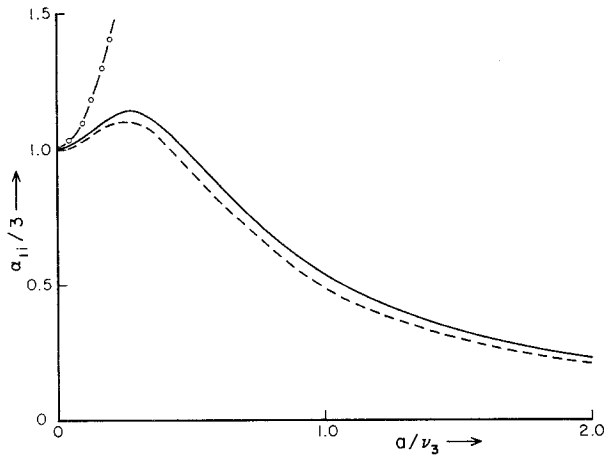
Fig. 2. Trace of $\alpha_{ij}(t)/3$ from Eqs. (4.12) and (B6) for $\epsilon = 0.1$, (————); same quantity neglecting the irreversible part of the stress tensor, t_{ji}^*, (---); same quantity neglecting viscous heating and t_{ij}^*, (- ○ - ○ -).

Figure 2 where the trace of $\alpha_{ij}/3$ is plotted as a function of reduced shear rate, a/ν_3. Since the shear viscosity of the fluid decreases with shear rate (shear thinning) it might be expected from a Stokes–Einstein relation that the diffusion coefficient should increase. However, the Stokes–Einstein relation would apply only for Brownian particles, whose size is comparable to or larger than the mean free path. Here the opposite case has been considered, a particle small compared to the mean free path, and the friction constant is not related to the shear viscosity as in Stokes law.

5. DISCUSSION

The Fokker–Planck and Langevin equations discussed here have been obtained directly from the Boltzmann–Lorentz equations for nonequilibrium fluctuations. These results follow from well-understood approximations in nonequilibrium statistical mechanics, and therefore serve as a test case for phenomenological or stochastic theories of nonequilibrium fluctuations. The dependence of the diffusion tensor on the irreversible stress tensor is an illustration of the need for caution in extending equilibrium theories to nonequilibrium states. In general, the diffusion tensor cannot be characterized entirely by the thermodynamic temperature. This is sometimes emphasized by defining a nonequilibrium "noise temperature," or for anisotropic systems, a "noise temperature tensor."[10] One such definition is

$$\theta_{ij}(t) = MD_{ij}(t)/2k_B\nu_1 \tag{5.1}$$

In equilibrium, the noise and thermodynamic temperatures are the same. For nonequilibrium states it has been conjectured[10] that the noise temperature is always greater than or equal to the thermodynamic temperature in the sense $\theta_{ii}(t)/3 \geqslant T(t)$. This conjecture is valid for the uniform shear flow considered here, because the contribution from the irreversible stress tensor is traceless.

The Fokker–Planck equation for a general nonequilibrium state, Eq. (2.2), depends on the irreversible momentum flux but not the irreversible heat flux. This is an anomaly of the Maxwell potential. For other potential models there is an additional contribution to the drift vector that would depend on any temperature gradients in the fluid. The mass ratio expansion for a general central potential has been discussed recently by Fernandez de la Mora and Mercer.[4] Equation (A.14) here agrees with their Eqs. (31) and (32) after the indicated Mach number expansion of the latter is carried out. The primary differences occur in the application of these results. Here, attention is restricted to the Maxwell potential so that states far from equilibrium can be discussed; in Ref. 4 the general potential model is retained and attention is limited to states near equilibrium (the Chapman–Enskog expansion). In particular, attention is focused on the thermal diffusion process in Ref. 4 and the contributions for gradients of the velocity field are considered to be negligibly small. To clarify this latter point the case of uniform shear flow can be specialized to states near equilibrium. To first order in the shear rate the solutions to the Boltzmann equation for the fluid distribution function, f, and the Fokker–Planck equation for the tagged particle distribution, F, are

$$f = f_L \left\{ 1 - \gamma_{ij}(v)(a_{ij}/v_3) \right\}$$
$$F = F_L \left\{ 1 - \gamma_{ij}(v)\left[(a_{ij}/v_1) - 0.44(a_{ij}/v_3) \right] \right\}$$

$$(5.2)$$

where $\gamma_{ij}(v)$ is defined by

$$\gamma_{ij}(v) \equiv \tfrac{1}{2}\left(v_i^* v_j^* - \tfrac{1}{3}\delta_{ij}v^{*2} \right) \qquad (5.3)$$

and v_i^* is the velocity of the fluid or tagged particle reduced by the appropriate thermal velocity. The distributions, f_L and F_L, are the local equilibrium distributions for the fluid and tagged particle, respectively. The deviations of the fluid distribution from local equilibrium are characterized by the single dimensionless quantity a_{ij}/v_3, where v_3 is an eigenvalue of the Boltzmann operator [Eq. (A.14) of Ref. 1]. In contrast, there are *two* dimensionless quantities that determine the deviation of the tagged particle distribution from local equilibrium, (a_{ij}/v_3) and (a_{ij}/v_1). The terms of order a_{ij}/v_3 are due to the contributions of the irreversible stress tension to the diffusion tensor, whereas the term of order a/v_1 is due simply to the dependence of the drift vector on the relative velocity. Consequently, when

discussing nonequilibrium fluid properties (e.g., the diffusion tensor) terms of first order in a/ν_3 must be retained; however, for tagged particle properties the contribution of order a/ν_3 may be neglected compared to a/ν_1 since $\nu_1/\nu_3 \sim \epsilon$. More generally, though, both a_{ij}/ν_1 and a_{ij}/ν_3 terms must be considered beyond linear order.

These comments also indicate the relationship of the results here to those of San Miguel and Sancho,[3] who solve the Langevin equations (2.17) but with an equilibrium noise spectrum $\overline{\gamma_i(t)\gamma_j(t')} = (2\nu_1 k_B T/M)\delta_{ij}\delta(t-t')$. Thus they neglect the irreversible stress tensor contributions and the viscous heating. The above discussion shows that such assumptions are correct only to linear order in a_{ij}. In addition, there are secular terms of order $(a_i/\nu_3)^2$ due to the viscous heating that eventually become important even for small shear rates.

ACKNOWLEDGMENTS

APPENDIX A: MASS RATIO EXPANSION OF THE BOLTZMANN–LORENTZ OPERATOR

In this Appendix the Boltzmann–Lorentz operator occurring in the kinetic equations for the tagged particle distribution and the phase space fluctuations is expanded to lowest order in the mass ratio, $\epsilon \equiv m/M$, where m is the mass of the fluid particle and M is the mass of the tagged particle. It is convenient to perform the expansion in the local rest frame defined by transforming all velocities to those relative to the local fluid velocity (see Section II of Ref. 1). The operator is then

$$J[f', F'] = \int d\mathbf{v}_1 \int_0^\infty db\, bg \int_0^{2\pi} d\phi\, (f'(\mathbf{v}_1 + \Delta\mathbf{v}_1)F'(\mathbf{v} + \Delta\mathbf{v}) - f'(\mathbf{v}_1)F'(\mathbf{v}))$$

(A1)

where $\Delta\mathbf{v}_1$ and $\Delta\mathbf{v}$ are the change in the velocity of the fluid and tagged particles, respectively, due to a binary collision. Also, $\mathbf{g} = \mathbf{v} - \mathbf{v}_1$ is the relative velocity and the prime on the distribution functions, f' and F', is included as a reminder that they are the distributions in the local rest frame. The dependence of F' on position has been left implicit.

To identify all the dependence on the mass ratio it is necessary to express the collision operator in some appropriate dimensionless variables.

It will be required that the relevant velocities determined by the distribution functions f' and F' are characterized by the thermal velocities for the fluid and tagged particle, respectively:

$$v_f = (k_B T/m)^{1/2}, \qquad v_T = (k_B T/M)^{1/2} \qquad (A2)$$

This characterization precludes strongly nonthermal distributions, such as occur in particle beams, but otherwise accommodates a wide class of nonequilibrium states. Above, T is some constant temperature of the same order as the nonequilibrium temperature, $T(t)$. The characteristic length, σ, is taken to be that defining the interparticle potential assumed to have the form

$$V(r) = V_0 u(r/\sigma) \qquad (A3)$$

where V_0 is a constant with dimensions of energy. The dimensionless velocities and impact parameter are then defined by

$$\mathbf{v}^* = \mathbf{v}/v_T, \qquad \mathbf{v}_1^* = \mathbf{v}_1/v_f, \qquad \mathbf{g}^* = \mathbf{v}^* - (\epsilon)^{1/2}\mathbf{v}_1^* \qquad b^* = b/\sigma \quad (A4)$$

Also, the dimensionless distribution functions are defined by

$$f^*(\mathbf{v}_1^*) = (\sigma v_f)^3 f'(\mathbf{v}_1), \qquad F^*(\mathbf{v}^*) = v_T^3 F'(\mathbf{v}) \qquad (A5)$$

Finally the dimensionless collision operator is given by

$$J^*[f^*, F^*] = \sigma v_T^2 J[f', F']$$

$$= \int d\mathbf{v}_1 \int db^* \, b^* g^* \int d\phi \, (f^*(\mathbf{v}_1^* + \Delta\mathbf{v}_1^*)F^*(\mathbf{v} + \Delta\mathbf{v}^*)$$

$$- f^*(\mathbf{v}_1^*)F^*(\mathbf{v}^*)) \qquad (A6)$$

In the following the asterisk (*) will be deleted on the integration variables for notational simplicity but are understood to be dimensionless unless indicated otherwise. At this point it is easily verified that the operator J^* is a function of only two dimensionless constants, ϵ and $V_0/k_B T \equiv 1/T^*$.

The expansion of J^* in the small parameter, ϵ, is most easily performed by representing J^* in terms of its adjoint. Let $H(\mathbf{v})$ be a function that scales according to the thermal velocity of the tagged particle, but is otherwise arbitrary, and consider the integral,

$$\int d\mathbf{v}\, H^*(\mathbf{v})J^*(f^*, F^*)$$

$$= \int d\mathbf{v}\, d\mathbf{v}_1\, db\, bg\, d\phi\, H^*(\mathbf{v})\big[f^*(\mathbf{v}_1 + \Delta\mathbf{v}_1)F^*(\mathbf{v} + \Delta\mathbf{v}) - f^*(\mathbf{v}_1)F^*(\mathbf{v}) \big]$$

$$(A7)$$

A change of variables $(\mathbf{v}_1 + \Delta\mathbf{v}_1, \mathbf{v} + \Delta\mathbf{v}) \rightarrow (\mathbf{v}_1, \mathbf{v})$ in the first term of the brackets leads to (the Jacobian equals 1, from conservation of energy and

momentum),

$$\int dv\, H^*(v) J^*(f^*, F^*) = \int dv\, dv_1\, db\, bg\, d\phi\, f^*(v_1) F^*(v)(H^*(v + \Delta v) - H^*(v))$$

(A8)

The change in the tagged particle velocity, Δv, may be expressed in terms of the relative velocity and the scattering angle as

$$\Delta v = \frac{\epsilon g}{1 + \epsilon} \left[\hat{g}(\cos\theta - 1) + \hat{a}\sin\theta \right]$$

(A9)

$$\theta(b, g) = \pi - 2\int_{x_0}^{\infty} \frac{dx}{x\left(x^2\left\{1 - \left[2(1 + \epsilon)u(xb)/T^*g^2\epsilon\right]\right\} - 1\right)^{1/2}}$$

(A10)

where $\hat{a}$ is a unit vector orthogonal to g and x_0 is the minimum relative distance [the root of the radical in Eq. (A10)]. The ϵ dependence occurs explicitly as shown in Δv and θ, but also implicitly through g. A further simplification is possible if the potential function is a homogeneous function of x; for example, the r^{-n} potential gives $u(xb) = u(x)b^{-n}$. The dependence of θ on ϵ and g can then be extracted explicitly in Eq. (A8) by the change of variables $y = b^2(T^*\epsilon g^2/2(1 + \epsilon))^{2/n}$, in the b integration,

$$\int dv\, H^*(v) J^*(f^*, F^*) = \int dv\, dv_1\, f^*(v_1) F^*(v) \frac{1}{2} \left[\frac{(1 + \epsilon)2}{T^*\epsilon} \right]^{2/n} g^{(1 - 4/n)}$$

$$\times \int dy\, d\phi \left[H^*(v + \Delta v) - H^*(v) \right]$$

(A11)

The scattering angle $\theta(y)$ is now independent of all constants and g,

$$\theta(y) = \pi - 2\int_{x_0}^{\infty} \frac{dx}{x\left\{ x^2\left[1 - y^{-n/2}u(x)\right] - 1 \right\}^{1/2}}$$

(A12)

and the complete ϵ dependence of Δv is contained in the prefactor $g\epsilon/(1 + \epsilon)$. The dominant behavior for small ϵ is therefore $\epsilon^{1/2}$. With this in mind the ϵ expansion can be carried out in two steps: first an expansion in powers of Δv, followed by an expansion of each term of the first expansion in ϵ. The first expansion is known as the Kramers–Moyal expansion[11] and is often used to justify the Fokker–Planck equation under the assumption that the terms in Δv beyond second order are zero or negligible. However, the Kramers–Moyal expansion alone is not systematic in any small parameter (for example, it does not order the ϵ expansion) and its truncation at any order does not even preserve the stationary solution for an equilibrium fluid. It is only systematic if a subsequent expansion of each term in some small parameter is performed to the order considered. Here, the terms of order ϵ^0 and $\epsilon^{1/2}$ are calculated so it is sufficient to retain initially only the

first two terms of the Kramers–Moyal expansion,

$$\int d\mathbf{v}\, H^*(\mathbf{v})J^*[f^*, F^*]$$

$$\rightarrow \int d\mathbf{v}\, F^*(\mathbf{v})\left[\langle \Delta v_i \rangle \frac{\partial}{\partial v_i} + \frac{1}{2}\langle \Delta v_i \Delta v_j \rangle \frac{\partial^2}{\partial v_i \partial v_j}\right] H^*(\mathbf{v}) \quad \text{(A13)}$$

with

$$\langle \Delta v_i \rangle = \int d\mathbf{v}_1 \, f^*(\mathbf{v}_1) \frac{1}{2}\left[\frac{(1+\epsilon)2}{T^*\epsilon}\right]^{2/n} g^{(n-4)/n}\int dy\, d\phi \, \Delta v_i$$

$$\langle \Delta v_i \Delta v_j \rangle = \int d\mathbf{v}_1 \, f^*(\mathbf{v}_1) \frac{1}{2}\left[\frac{(1+\epsilon)2}{T^*\epsilon}\right]^{2/n} g^{(n-4)/n}\int dy\, d\phi \, \Delta v_i \Delta v_j \tag{A14}$$

The integration over ϕ can be performed using Eq. (A9) and the results,

$$\int_0^{2\pi} d\phi\, a_i = 0, \qquad \int_0^{2\pi} d\phi\, a_i a_j = \pi\left(\delta_{ij} - g_i g_j/g^2\right) \tag{A15}$$

to give

$$\langle \Delta v_i \rangle = -\nu_1^*\left(\frac{\epsilon}{1+\epsilon}\right)^{(n-2)/n}\int d\mathbf{v}_1 \, f^*(\mathbf{v}_1)g^{(n-4)/n}g_i$$

$$\langle \Delta v_i \Delta v_j \rangle = 2\nu_1^*\left(\frac{\epsilon}{1+\epsilon}\right)^{(2n-2)/n}\int d\mathbf{v}_1 \, f^*(\mathbf{v}_1)g^{(n-4)/n}g_i g_j \tag{A16}$$

$$- 2\nu_2^*\left(\frac{\epsilon}{1+\epsilon}\right)^{(2n-2)/n}\int d\mathbf{v}_1 \, f^*(\mathbf{v}_1)g^{(n-4)/n}\left(g_i g_j - \frac{1}{3}g^2\delta_{ij}\right)$$

The constants ν_1^* and ν_2^* are independent of ϵ and are functions only of the reduced temperature, T^*, and the power law of the potential,

$$\nu_1^* = \pi\left(\frac{2}{T^*}\right)^{2/n}\int_0^\infty dy\,[1 - \cos\theta(y)]$$

$$\nu_2^* = \frac{3\pi}{4}\left(\frac{2}{T^*}\right)^{2/n}\int_0^\infty dy\,\sin^2\theta(y) \tag{A17}$$

It remains to complete the ϵ expansion of the Kramers–Moyal coefficients in Eq. (A16). Use of the definition of **g** in Eq. (A4) and retaining terms only to order $\epsilon^{1/2}$ gives

$$\langle \Delta v_i \rangle \rightarrow \epsilon^0 \nu_1^* \int d\mathbf{v}_1 \, f^*(\mathbf{v}_1)v_{1i}v_1^{(n-4)/n}$$

$$- \epsilon^{1/2}\nu_1^* \int d\mathbf{v}_1 \, f^*(\mathbf{v}_1)v_1^{(n-4)/n}\left[v_i + v_{1i}(n-4)\mathbf{v}_1 \cdot \mathbf{v}/nv_1^2\right]$$

$$\langle \Delta v_i \Delta_j \rangle \rightarrow \epsilon^{1/2}\left[2\nu_1^* \int d\mathbf{v}_1 \, f^*(\mathbf{v}_1)v_1^{(n-4)/n}v_{1i}v_{1j} \right. \tag{A18}$$

$$\left. - 2\nu_2^* \int d\mathbf{v}_1 \, f^*(\mathbf{v}_1)v_1^{(n-4)/n}\left(v_{1i}v_{1j} - \tfrac{1}{3}v_1^2\delta_{ij}\right)\right]$$

The integrals over the fluid distribution in Eq. (A18) cannot be simplified further in general without specification of the nonequilibrium distribution itself. Since the latter is determined from the nonlinear Boltzmann equation, very little progress can be made except for states near equilibrium where the Chapman–Enskog solution can be used. An important exception is the case of Maxwell molecules for which the power law is $n = 4$. The results, (A18), then simplify considerably. More importantly the integrals over the fluid distribution function reduce to first- or second-order velocity moments of the distribution function and may be identified in terms of the hydrodynamic variables for the nonequilibrium state. This is significant because it means that the effects of the fluid on the tagged particle can be characterized completely in terms of the macroscopic state of the fluid rather than the experimentally less accessible details of the distribution function. In the following, attention will be restricted to a fluid of Maxwell molecules. Then, with the definition of the pressure tensor, $P_{ij}(\mathbf{r}, t)$, and number density, $n(\mathbf{r}, t)$ (in dimensionless form),

$$P_{ij}(\mathbf{r}, t) = \int d\mathbf{v}_1 \, f^*(\mathbf{v}_1) v_{1i} v_{1j}, \qquad n(\mathbf{r}, t) = \int d\mathbf{v}_1 \, f^*(\mathbf{v}_1) \tag{A19}$$

the results, (A18), become

$$\langle \Delta v_i \rangle = -\epsilon^{1/2} \nu_1^* n(\mathbf{r}, t) v_i \equiv -A_1^*(\mathbf{v}, \mathbf{r}, t)$$

$$\langle \Delta v_i \Delta v_j \rangle = \epsilon^{1/2} 2 \left[\nu_2^* \tfrac{1}{3} P_{kk} \delta_{ij} + (\nu_1^* - \nu_2^*) P_{ij} \right] \equiv D_{ij}^*(\mathbf{r}, t) \tag{A20}$$

The term of order ϵ^0 vanishes by definition of the average flow velocity. Integrating by parts in Eq. (A13) and use of Eqs. (A20) gives

$$\int d\mathbf{v} \, H^*(\mathbf{v}) \left\{ J^*[f^*, F^*] - \frac{\partial}{\partial v_i} \left[A_i^*(\mathbf{v}, \mathbf{r}, t) + \frac{1}{2} \frac{\partial}{\partial v_j} D_{ij}^*(\mathbf{r}, t) \right] F^* \right\}$$

$$= 0 + (\text{order } \epsilon) \tag{A21}$$

Finally then, since $H^*(\mathbf{v})$ is arbitrary, the Boltzmann–Lorentz operator to order $\epsilon^{1/2}$ can be represented as the Fokker–Planck operator,

$$J^*[f^*, F^*] \to \frac{\partial}{\partial v_i} \left(A_i^*(\mathbf{v}, \mathbf{r}, t) + \frac{1}{2} \frac{\partial}{\partial v_j} D_{ij}^*(\mathbf{r}, t) \right) F^* \tag{A22}$$

The corresponding results in the laboratory frame are obtained by the replacement, $\mathbf{v} \to \mathbf{v} - \mathbf{U}(r, t)$. Also, returning to the original variables with the appropriate units, the desired form of the Fokker–Planck operator is obtained:

$$J[f, F] \to \frac{\partial}{\partial v_i} \left[A_i(\mathbf{v}, \mathbf{r}, t) + \frac{1}{2} \frac{\partial}{\partial v_j} D_{ij}(\mathbf{r}, t) \right] F \tag{A23}$$

The drift vector, A_i, and the diffusion tensor, $D_{ij}(\mathbf{r}, t)$ are defined by

$$A_i(\mathbf{v}, \mathbf{r}, t) = \nu_1 n_0^{-1} n(\mathbf{r}, t) \left[v_i - U_i(\mathbf{r}, t) \right] \tag{A24}$$

$$D_{ij}(r, t) = 2\rho_0^{-1} \left[\nu_2 \tfrac{1}{3} P_{kk}(\mathbf{r}, t) \delta_{ij} + (\nu_1 - \nu_2) P_{ij}(\mathbf{r}, t) \right] \tag{A25}$$

with

$$\nu_1 = \epsilon^{1/2} \nu_1^* n_0 v_T \sigma^2, \qquad \nu_2 = \epsilon^{1/2} \nu_2^* n_0 v_T \sigma^2, \qquad \rho_0 = M n_0 \tag{A26}$$

and n_0 is a constant density of the order of $n(\mathbf{r}, t)$ introduced for the interpretation of ν_1 as a collision rate. The form used in the text, Eqs. (2.2)–(2.4), is obtained by dividing the pressure tensor into its diagonal and traceless parts,

$$P_{ij}(\mathbf{r}, t) = p(\mathbf{r}, t) \delta_{ij} + t_{ij}^*(\mathbf{r}, t) \tag{A27}$$

where $p(\mathbf{r}, t)$ is the local pressure and the traceless tensor, $t_{ij}^*(\mathbf{r}, t)$ is the irreversible part of the pressure tensor (the asterisk here does not indicate a dimensionless quantity as above, but is used to conform to the notation of Ref. 1).

The constants ν_1^* and ν_2^* have been evaluated numerically for Maxwell molecule with the results

$$\nu_1^* = 1.19\pi (2/T^*)^{1/2}$$

$$\nu_2^* = \frac{3\pi}{4} 1.23 (2/T^*)^{1/2}$$

APPENDIX B: LONG TIME LIMIT OF $\alpha_{ij}(t)$

The factor, $\alpha_{ij}(\nu_1 t)$, in the diffusion coefficient is defined by Eq. (4.11),

$$\alpha_{ij}(\nu_1 t) = \int_0^{\nu_1 t} ds\, e^{-s} \rho (\delta_{ik} - a_{ik} s / \nu_1) R_{kj}(0, t - s/\nu_1)/p(t) \tag{B1}$$

The velocity fluctuation $R_{kj}(0, t - s/\nu_1)$ is defined by

$$R_{kj}(0, t - s/\nu_1) = \langle v_i'(t - s/\nu_1) v_j'(t - s/\nu) \rangle \tag{B2}$$

To calculate $R_{rj}(0, t - s/\nu_1)$ it is convenient to express the Langevin equation (2.11) in terms of the relative velocity,

$$\left(\frac{\partial}{\partial t} + \nu_1 \right) v_i' + a_{ij} v_j' = \gamma_i \tag{B3}$$

Then, solving Eq. (B3) and using the properties (2.12) for γ gives

$$R_{kj}(0, t - s/\nu_1) = e^{-2(\nu_1 t - 2)} \Lambda_{kl}(\nu_1 t - s) \Lambda_{jm}(\nu_1 t - s) R_{lm}(0, 0)$$

$$+ \nu_1^{-1} \int_0^{\nu_1 t - s} d\tau\, e^{-\tau} \Lambda_{km}(\tau/\nu_1) \Lambda_{jl}(\tau/\nu_1)$$

$$\times D_{lm}((\nu_1 t - \tau - s)/\nu_1) \tag{B4}$$

$$\Lambda_{ij}(t) \equiv \delta_{ij} - a_{ij} t$$

The exponential factor in Eq. (B1) means that $(\nu_1 t - s) \gg 1$ if $\nu_1 t \gg 1$, for the relevant values of s. Similarly $(\nu_1 t - \tau - s) \gg 1$ for $\nu_1 t \gg 1$ in the integrand of (B4). Consequently $D_{ij}((\nu_1 t - s - \tau)/\nu_1)$ may be evaluated using the asymptotic forms of Eqs. (A35) and (A36) in Ref. 1,

$$P_{ij}(t) = p(0)B_{ij}e^{z_1 t}$$

$$p(t) = p(0)Ae^{z_1 t}$$

(B5)

where A, B_{ij}, and z_1 are functions of the shear rate and are given explicitly by Eqs. (A37) and (A38) of Ref. 1. Equation (B1) then becomes, for large $\nu_1 t$,

$$\alpha_{ij}(t) \to \alpha_{ij} = A_1 \delta_{ij} + A_2 a_{ij} + A_3 a_{ji} + A_4 a_{ik} a_{jk}$$

(B6)

with

$$A_1 = \frac{2\nu_1(\nu_1\nu_3 + z_1\nu_2)}{(z_1 + \nu_1)(z_1 + 2\nu_1)(z_1 + \nu_3)}$$

$$A_2 = \frac{-2\nu_1}{(z_1 + \nu_1)(z_1 + 2\nu_1)(z_1 + \nu_3)} \left[\frac{\nu_3(\nu_1 - \nu_2)}{z_1 + \nu_3} + \frac{\nu_1\nu_3 + z_1\nu_2}{z_1 + 2\nu_1} \right]$$

$$- \frac{A_1}{z_1 + \nu_1}$$

(B7)

$$A_3 = A_2 + \frac{A_1}{z_1 + \nu_1}$$

$$A_4 = \frac{4\nu_1}{(z_1 + \nu_1)(z_1 + 2\nu_1)(z_1 + \nu_3)}$$

$$\times \left[\frac{\nu_3(\nu_1 - \nu_2)}{(z_1 + 2\nu_1)(z_1 + \nu_3)} + \frac{\nu_3(\nu_1 - \nu_2)}{(z_1 + \nu_3)^2} + \frac{(\nu_1\nu_3 + z_1\nu_2)}{(z_1 + 2\nu_1)^2} \right] - \frac{A_3}{(z_1 + \nu_1)}$$

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