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Microscopic Derivation of Non-Markovian Thermalization of a Brownian Particle

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In this paper, the first microscopic approach to Brownian motion is developed in the case where the mass density of the suspending bath is of the same order of magnitude as that of the Brownian (B) particle. Starting from an extended Boltzmann equation, which describes correctly the interaction with the fluid, we derive systematically via multiple-time-scale analysis a reduced equation controlling the thermalization of the B particle, i.e., the relaxation toward the Maxwell distribution in velocity space. In contradistinction to the Fokker–Planck equation, the derived new evolution equation is nonlocal both in time and in velocity space, owing to correlated recollision events between the fluid and particle B. In the long-time limit, it describes a non-Markovian generalized Ornstein–Uhlenbeck process. However, in spite of this complex dynamical behavior, the Stokes–Einstein law relating the friction and diffusion coefficients is shown to remain valid. A microscopic expression for the friction coefficient is derived, which acquires the form of the Stokes law in the limit where the mean-free path in the gas is small compared to the radius of particle B.

KEY WORDS: Brownian motion; Fokker–Planck equation; non-Markovian process.

1. INTRODUCTION

This paper is concerned with a microscopic theory of the Brownian motion performed by a massive particle suspended in a gas of much lighter particles. Because of the mass and length-scale difference between the two components, one expects to eliminate the fluid variables from the description

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Knowing the interest of Matthieu Ernst in the subtle and fundamental problems of kinetic theory, we have the pleasure to dedicate this study to him.

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of the system and obtain a closed equation for the Brownian particle.[1-3] Traditionally, this task is done by assuming that the dynamical properties of the Brownian particle (B) evolve on a time scale much longer than the characteristic time scale of the fluid. This approach leads to the well-known Fokker–Planck equation, which governs the time evolution of the distribution function $f(\mathbf{R}, \mathbf{V}; t)$ of the position $\mathbf{R}$ and velocity $\mathbf{V}$ of particle B:

$$\left(\frac{\partial}{\partial t} + \mathbf{V} \cdot \frac{\partial}{\partial \mathbf{R}}\right) f(\mathbf{R}, \mathbf{V}; t) = \zeta \frac{\partial}{\partial \mathbf{V}} \cdot \left(\mathbf{V} + \frac{k_B T}{M} \frac{\partial}{\partial \mathbf{V}}\right) f(\mathbf{R}, \mathbf{V}; t) \tag{1}$$

M denotes the mass. In this equation, the fluid enters only through the friction coefficient ζ and the temperature T.

An equivalent description can be obtained starting from the stochastic Langevin equation

$$M \frac{d\mathbf{V}}{dt} = -M\zeta \mathbf{V}(t) + \tilde{\mathbf{F}}(t) \tag{2}$$

where $\mathbf{V}(t)$ is the velocity of B and $\tilde{\mathbf{F}}(t)$ is the fluctuating part of the force exerted by the fluid, assumed to have a white spectrum. This equation leads to an exponential decay of the velocity autocorrelation function with a relaxation time given by ζ^{-1}.[1,2]

On the other hand, some attempts at a fully microscopic description of the dynamics have been made.[4-9] It has indeed been possible to get rid of the stochastic assumption and obtain the Fokker–Planck equation by a systematic expansion of the dynamics of the complete system, bath + B particle, in powers of the square root of the mass ratio m/M, where m is the fluid particle mass. Such derivations lead in particular to a microscopic formula for the friction coefficient in terms of the autocorrelation function of the force acting on the B particle.

However, as pointed out by many authors,[10-13] but already by Lorentz in 1911,[14] the description based on Eqs. (1) and (2) can only be valid if the ratio ρ/ρ_B of the mass density of the fluid ρ and that of the B particle ρ_B is very small (ρ_B is defined as the mass M of particle B divided by its own volume, so that $\rho_B \sim M/\Sigma^3$, where Σ is the B-particle diameter). Indeed, as noticed above, the velocity of particle B relaxes on a time scale $\tau_V \sim \zeta^{-1}$. If Stokes' law is assumed, this leads to $\tau_V \sim M/\eta\Sigma$, where η is the viscosity of the fluid and Σ the B-particle diameter. On the other hand, a typical hydrodynamic time of the fluid is of the order $\tau_f \sim \Sigma^2/(\eta/\rho)$, which is the time for a shear flow to propagate over the distance Σ. These rough estimates give $\tau_f/\tau_V \sim \rho/\rho_B$, so that the assumption of a wide time scale separation is only justified if this ratio is small. This condition is unfortunately far from the experimental situation, where the mass density ratio ρ/ρ_B is taken rather close to unity to avoid sedimentation of particle B. In this case, the fluid dynamics must contain slowly decaying modes, which relax on the same time scale as the velocity of the B particle. In other words, non-Markovian effects are expected and the validity of the Fokker–Planck or Langevin equations becomes doubtful.[12] [3]

Several attempts have been made to overcome these difficulties and determine the dynamical evolution in the general case, that is, for any (finite) ρ/ρ_B.[10, 11, 15] The main idea underlying all these works is that the slowly decaying fluid modes result from the momentum conservation law for the fluid particles. A correct description should therefore treat both fluid and B-particle variables on the same level. This can be done, for example by using fluctuating hydrodynamics for the fluid motion, with appropriate boundary conditions on the surface of the suspended B particle.[10] These approaches lead to a non-Markovian Langevin equation, involving the time-dependent friction coefficient $\zeta(t)$:

$$M\frac{d\mathbf{V}}{dt} = -M\int_0^t d\tau\, \zeta(t-\tau)\, \mathbf{V}(\tau) + \tilde{\mathbf{F}}(t) \tag{3}$$

On the other hand, only simplified microscopic versions of this problem have been considered, including one B particle interacting with a gas of point particles, showing that in such cases, the velocity process is an Orstein–Uhlenbeck one.[16]

We present here the first systematic, microscopic approach which treats the B particle and the fluid at the same level, clarifying and providing the fundamental basis for the phenomenological descriptions. The problem is reconsidered within the kinetic theory of gases. The system consists of a single large, heavy hard sphere (mass M, diameter Σ) suspended in a fluid of small, light hard spheres (mass m, diameter σ). The dynamics of the system will be assumed to be governed by an extended Boltzmann equation which correctly takes the gas–B-particle collisions into account. The same type of equation was used by van Beijeren and Dorfman to develop the kinetic theory of hydrodynamic flows and in particular to clarify the dynamical origin of the hydrodynamic Stokes law for the friction coefficient.[17] The extended Boltzmann equation is expected to be correct in the Grad limit, defined for the gas with number density n by $n \to \infty$, $\sigma \to 0$,

[3] Let us note, however, that the Smoluchowski equation describing the spatial evolution of the B particle is not affected by these arguments, since the position of the B particle relaxes on a time scale much longer than the velocity (or fluid) relaxation times.

the mean free path $l = (n\sigma^2)^{-1} = \text{const.}$[18] We choose the diameter Σ of the B particle to fix the length scale. The condition of a constant mean free path thus imposes

$$\frac{\Sigma}{l} = n\sigma^2\Sigma = \text{const} \tag{4}$$

Moreover, and unlike all previous work, we require from the beginning the condition of essentially equal mass density of the gas and the B particle. In other words, the ratio ρ/ρ_B will be kept finite even in the small-mass-ratio limit, which amounts to assuming

$$\frac{M}{\Sigma^3} = \text{const} \times mn \tag{5}$$

Finally, we define the small parameter corresponding to the Brownian limit

$$\varepsilon = \left(\frac{m}{M}\right)^{1/2} \ll 1 \tag{6}$$

Introducing the diameter ratio $\kappa = \sigma/\Sigma$, we characterize the asymptotic regime (4)–(6) by the limit

$$\begin{cases} \varepsilon \to 0 \\ \kappa \sim \varepsilon, \qquad n\Sigma^3 \sim \varepsilon^{-2} \end{cases} \tag{7}$$

To simplify the calculations and without loss of generality, we will assume in the following that $\varepsilon = \kappa$ and $n\Sigma^3 = \varepsilon^{-2}$. This can be looked upon as redefining the parameters of the system (Σ, M, n,...), to set the two constants introduced in Eqs. (4) and (5) equal to one. The correct scaling will, however, be recovered in the final results.

Starting from the extended Boltzmann equation, we will perform a systematic expansion of the dynamical properties of the system in powers of ε, in order to obtain a closed equation for the distribution function of particle B.

The following results will be obtained:

(i) On the shortest time scale, $t \sim \tau_V \sim \tau_f$, the B particle "does not move" (in position space), whereas its velocity distribution relaxes in a thermalization process.

(ii) The equation (65) characterizing the relaxation of the velocity distribution is not a Fokker–Planck equation. It exhibits a complex non-Markovian character, which stems from building up the dynamical friction force by recollision events between the gas and the B particle.

(iii) We will be able to express the autocorrelation function (ACF) of the velocity of the B particle in terms of the time-dependent friction coefficient. A microscopic formula for this coefficient follows from our analysis. The final result justifies the phenomenological expression for the velocity ACF, based on fluctuating hydrodynamics.

(iv) In spite of non-Markovian effects, the Stokes–Einstein relation, expressing the diffusion constant of the B particle in terms of the friction coefficient, is explicitly derived.

2. KINETIC EQUATIONS

In the study of the dynamics of the B particle immersed in a bath of N small spheres, the short-hand notation

$$B \equiv (\mathbf{R}, \mathbf{V}); \qquad i \equiv (\mathbf{r}_i, \mathbf{v}_i), \qquad i = 1, 2, ..., N \tag{8}$$

will be used for the positions and velocities of the B particle and the N gas particles.

The temperature T of the gas introduces characteristic measures, $\sqrt{K_B T/M}$ and $\sqrt{k_B T/m}$ of the velocities of the B and fluid particles, respectively. On the other hand, the characteristic length scale in the system is given by the diameter Σ of the B particle. In order to study the asymptotic regime (7), it is convenient to use dimensionless variables defined by

$$\begin{aligned} \mathbf{V} &= \sqrt{\frac{k_B T}{M}}\, \mathbf{U}, \qquad & \mathbf{v}_j &= \sqrt{\frac{K_B T}{m}}\, \mathbf{u}_j \\ \mathbf{R} &= \Sigma \mathbf{X}, \qquad & \mathbf{r}_j &= \Sigma \mathbf{x}_j \end{aligned} \tag{9}$$

The shortest time scale in the system is fixed by the gas-B-particle collision frequency, and we introduce accordingly a dimensionless time variable τ by

$$t = \frac{1}{n\Sigma^2} \sqrt{\frac{m}{k_B T}}\, \tau \tag{10}$$

Our aim is to determine the dynamical evolution of the B-particle distribution function $f_B(B; t)$. To this end, we need to introduce the joint distribution, $f_{B1}(B, 1; t)$, representing at time t the number density of fluid particles in state 1 when at the same time particle B is in state B. Finally, we define the conditional distribution function of the gas $f_{1|B}$ by

$$f_{B1}(B, 1; t) = f_B(B; t)\, f_{1|B}(1 \mid B; t) \tag{11}$$

Under the dimensional scaling (9), the dimensionless distribution functions F_B and $\tilde{F}_{1|B}$ are defined by the relations

$$F_B(B, \tau) = \Sigma^3(k_B T/M)^{3/2} f_B(B; t) \tag{12}$$

and

$$\tilde{F}_{1|B}(1 \mid B; \tau) = \Sigma^3(k_B T/m)^{3/2} f_{1|B}(1 \mid B; t) \tag{13}$$

However, whereas $F_B(B; \tau)$ is essentially of order unity, $\tilde{F}_{1|B}$ does diverge as $\varepsilon \to 0$. Indeed, far from the suspended sphere B, the conditional distribution function $f_{1|B}$ is of the order of the density n of the gas, which implies [according to condition (7)]

$$\tilde{F}_{1|B} \sim n\Sigma^3 \sim \varepsilon^{-2} \tag{14}$$

Having in view a perturbation expansion, we thus introduce rescaled distribution functions $F_{1|B}(1 \mid B; \tau)$ and $F_{B1}(B, 1; \tau)$ defined as

$$\begin{aligned} F_{1|B}(1 \mid B; \tau) &= \varepsilon^2 \tilde{F}_{1|B}(1 \mid B; \tau) \\ F_{B1}(B, 1; \tau) &= \varepsilon^2 F_B(B; \tau)\, \tilde{F}_{1|B}(1 \mid B; \tau) \end{aligned} \tag{15}$$

which are of order unity when the small parameter ε goes to zero.

The scaled distribution functions evolve in time according to the coupled equations

$$\left(\frac{\partial}{\partial \tau} + \varepsilon^3 \mathbf{U} \cdot \frac{\partial}{\partial \mathbf{X}}\right) F_B(B; \tau) = \int d\mathbf{1}\, \bar{T}^{\varepsilon}_{-}(B, 1)\, F_{B1}(B, \mathbf{1}; \tau) \tag{16}$$

$$\begin{aligned} &\left\{\frac{\partial}{\partial \tau} + \varepsilon^3 \mathbf{U} \cdot \frac{\partial}{\partial \mathbf{X}} + \varepsilon^2 \left(\mathbf{u}_1 \cdot \frac{\partial}{\partial x_1} - \bar{T}^{\varepsilon}_{-}(B, 1)\right)\right\} F_{B1}(B, 1; \tau) \\ &\quad = \int d\mathbf{2}\, \left\{\bar{T}^{\varepsilon}_{-}(B, 2) + \varepsilon^2 \bar{T}_{-}(1, 2)\right\} F_{B12}(B, 1, 2; \tau) \end{aligned} \tag{17}$$

where the factorization of the three-particle distribution

$$F_{B12}(B, 1, 2; t) \simeq F_B(B; t)\, F_{1|B}(1 \mid B; t)\, F_{1|B}(2 \mid B; t) \tag{18}$$

is expected to hold in the Grad limit [see the discussion leading to (4)].

Equation (16) expresses the fact that the B-particle state changes in time through free motion (left-hand side, l.h.s, of the equation) and through collisions with gas particles (right-hand side, r.h.s., of the equation). The second equation (17) states that the two-particle distribution,

F_{B1}, evolves in time owing to free motion and collisions between the pair $(B, 1)$ (l.h.s. of the equation), and also owing to collisions of this pair with the surrounding gas particles (r.h.s. of the equation). Roughly speaking, Eq. (16) characterizes the time evolution of particle B due to collisions with the fluid, while the dynamics of the fluid, reacting to the B-particles' motion is contained in Eq. (17).

These evolution equations involve two hard-sphere collision operators, $\bar{T}^{\varepsilon}_{-}(B, 1)$ and $\bar{T}_{-}(1, 2)$. The first of them characterizes the effect of binary collisions between the B particle and a gas particle. It reads

$$\bar{T}^{\varepsilon}_{-}(B, 1) = \xi^2 \int d\hat{\sigma}\, [(\varepsilon\mathbf{U} - \mathbf{u}_1)\cdot\hat{\sigma}]\, \theta[(\varepsilon\mathbf{U} - \mathbf{u}_1)\cdot\hat{\sigma}] \times \{\delta(\mathbf{X} - \xi\hat{\sigma} - x_1)\, b^{\varepsilon}_{\hat{\sigma}}(B, 1) - \delta(\mathbf{X} + \xi\hat{\sigma} - \mathbf{x}_1)\} \tag{19}$$

where $\xi = 1/2$ is the dimensionless radius of the B particle; the operator $b^{\varepsilon}_{\hat{\sigma}}$ transforms the precollisional velocities into postcollisional velocities:

$$[b^{\varepsilon}_{\hat{\sigma}}(B, 1)\,\chi](\mathbf{U}, \mathbf{u}_1) = \chi\left(\mathbf{U} - \frac{2\varepsilon}{1+\varepsilon^2}[(\varepsilon\mathbf{U} - \mathbf{u}_1)\cdot\hat{\sigma}]\,\hat{\sigma}, \mathbf{u}_1 + \frac{2}{1+\varepsilon^2}[(\varepsilon\mathbf{U} - \mathbf{u}_1)\cdot\hat{\sigma}]\,\hat{\sigma}\right) \tag{20}$$

where $\hat{\sigma}$ is the unit vector along the axis joining the centers of the two spheres at contact and χ is any function of velocities $\mathbf{U}$ and $\mathbf{u}_1$.

On the other hand, $\bar{T}_{-}(1, 2)$ characterizes the effect of binary collisions between the fluid particles. It is given by

$$\bar{T}_{-}(1, 2) = \int d\hat{\sigma}(\mathbf{u}_{12}\cdot\hat{\sigma})\, \theta(\mathbf{u}_{12}\cdot\hat{\sigma})\, \delta(\mathbf{x}_{12})\{b_{\hat{\sigma}}(1, 2) - 1\} \tag{21}$$

where $\mathbf{u}_{12} = \mathbf{u}_1 - \mathbf{u}_2$, $\mathbf{x}_{12} = \mathbf{x}_1 - \mathbf{x}_2$, and $b_{\hat{\sigma}}$ acts as $b^{\varepsilon}_{\hat{\sigma}}$ in Eq. (20), with $\varepsilon = 1$ (collision between equal mass particles). Note that the dimensionless cross section of the gas, $(\sigma/\Sigma)^2 = \varepsilon^2$ [see Eq. (7)], has been extracted from the expression of $\bar{T}_{-}$ and displayed as a prefactor in Eq. (17).

In Eqs. (19) and (21), the fluid part of the collisional transfer is not present since we consider the Grad limit: at encounters, the positions of the particles coincide.[19]

To solve the system of integrodifferential Eqs. (16)–(17), we have to define the initial state of the system. As in our previous work,[8, 9] we shall assume that the fluid is initially in conditional equilibrium in the presence of particle B:

$$F^{\varepsilon}_{B1}(B, 1; \tau = 0) = F^{\varepsilon}_{B}(B; \tau = 0)\, F^{eq}(1 \mid \mathbf{X}) \tag{22}$$

with

$$F^{eq}(1 \mid \mathbf{X}) = \Theta(|\mathbf{X} - \mathbf{x}_1| - \xi)\, \phi(\mathbf{u}_1) \tag{23}$$

where

$$\phi(\mathbf{u}) = (2\pi)^{-3/2} \exp(-u^2/2) \tag{24}$$

is the Maxwell distribution and Θ is the Heaviside step function.

Before closing this section, let us recall some results concerning the ε expansion of the collision operator $\bar{T}^{\varepsilon}_{-}(B, 1)$, which will be used in the course of the analysis. The calculations leading to them, though technical, are quite straightforward and have been presented in our previous paper (ref. 8, cited henceforth as I).

The B-gas collision operator can be formally expanded in powers of $\varepsilon = (m/M)^{1/2}$ as

$$\bar{T}^{\varepsilon}_{-}(B, 1) = \bar{T}^{(0)}_{-}(B, 1) + \varepsilon \bar{T}^{(1)}_{-}(B, 1) + \varepsilon^2 \bar{T}^{(2)}_{-}(B, 1) + \cdots \tag{25}$$

The zeroth-order term $\bar{T}^{(0)}_{-}(B, 1)$ characterizes collisions of the gas particles with the immobile B particle, acting as an external field. One finds

$$\bar{T}^{(0)}_{-}(B, 1) = \xi^2 \int d\hat{\sigma}(-\mathbf{u}_1 \cdot \hat{\sigma})\, \theta(-\mathbf{u}_1 \cdot \hat{\sigma}) \times \{\delta(\mathbf{X} - \xi\hat{\sigma} - \mathbf{x}_1)\, b^{(0)}_{\hat{\sigma}}(B, 1) - \delta(\mathbf{X} + \xi\hat{\sigma} - \mathbf{x}_1)\} \tag{26}$$

where $b^{(0)}_{\hat{\sigma}}$ represents the change of velocity of a gas particle undergoing a specular collision with the B particle fixed at point $\mathbf{X}$ [see Eq. (20), with $\varepsilon = 0$].

The first-order correction has a much more complicated form (see Appendix of I for the full expression), but we will only need the action of $\bar{T}^{(1)}_{-}(B, 1)$ on a state in which the gas is in conditional equilibrium (23). In this case, one finds the following formula:

$$\bar{T}^{(1)}_{-}(B, 1)\, F(B)\, F^{eq}(1 \mid \mathbf{X}) = F(b)\, \mathbf{U} \cdot \frac{\partial}{\partial \mathbf{X}} F^{eq}(1 \mid \mathbf{X}) - \mathscr{F}_{-}(1) \cdot \left(\frac{\partial}{\partial \mathbf{U}} + \mathbf{U}\right) F(B)\, F^{eq}(1 \mid \mathbf{X}) \tag{27}$$

where the notation $\mathscr{F}_{\mp}(1)$ has been introduced to denote a dimensionless microscopic "force"

$$\mathscr{F}_{\mp}(1) = \xi^2 \int d\hat{\sigma}\, [2(\mathbf{u}_1 \cdot \hat{\sigma})^2\, \theta(\mp \mathbf{u}_1 \cdot \hat{\sigma})]\, \hat{\sigma}\, \delta(\mathbf{X} - \mathbf{x}_1 - \xi\hat{\sigma}) \tag{28}$$

We shall also need some formulas involving integration over the fluid degrees of freedom. First, one can verify that the integrated zeroth-order term identically vanishes:

$$\int d\mathbf{1}\, \bar{T}_{-}^{(0)}(B, 1)\, F_{B1}(B, 1) = 0 \tag{29}$$

The first-order term is given by

$$\int d\mathbf{1}\, \bar{T}_{-}^{(1)}(B, 1)\, F_{B1}(B, 1) = -\int d\mathbf{1}\, \mathscr{F}_{+}(1) \cdot \frac{\partial}{\partial \mathbf{U}} F_{B1}(B, 1) \tag{30}$$

with $\mathscr{F}_{+}(1)$ defined in Eq. (28).

The general expression for the integrated second-order term is more involved [see Eq. (31) of I for the complete formula], but when the fluid is in conditional equilibrium, it reduces to

$$\int d\mathbf{1}\, \bar{T}_{-}^{(2)}(B, 1)\, F_B(B)\, F_1^{\text{eq}}(1 \mid \mathbf{X}) = \xi^2 \frac{8}{3} \sqrt{2\pi}\, \frac{\partial}{\partial \mathbf{U}} \cdot \left(\frac{\partial}{\partial \mathbf{U}} + \mathbf{U} \right) F_B(B) \tag{31}$$

3. THE MULTIPLE-TIME-SCALE ANALYSIS

The system of coupled equations (16), (17) will be studied in the Brownian limit where $\varepsilon \ll 1$. As in our previous work, this will be achieved systematically by using a multiple-time-scale analysis, which leads to a uniformly convergent ε-expansion, avoiding secular divergences as time goes to infinity. We just recall here the spirit of the method. In the $\varepsilon \to 0$ limit, different time scales separate out in the system. Accordingly, we replace the distribution functions F_B, F_{B1} by auxiliary functions $F_B^{\varepsilon}(B; \tau_0, \tau_1, \tau_2,...)$, $F_{B1}^{\varepsilon}(B, 1; \tau_0, \tau_1, \tau_2,...)$ which now depend on many time arguments. The time derivative is replaced accordingly by the operator

$$\frac{\partial}{\partial \tau_0} + \varepsilon \frac{\partial}{\partial \tau_1} + \varepsilon^2 \frac{\partial}{\partial \tau_2} + \cdots \tag{32}$$

The auxiliary functions are also expanded in powers of ε:

$$\begin{aligned} F_B^{\varepsilon} &= F_B^{(0)} + \varepsilon F_B^{(1)} + \varepsilon^2 F_B^{(2)} + \cdots \\ F_{B1}^{\varepsilon} &= F_{B1}^{(0)} + \varepsilon F_{B1}^{(1)} + \varepsilon^2 F_{B1}^{(2)} + \cdots \end{aligned} \tag{33}$$

The expansions (32), (33) are substituted into the evolution equations (16) and (17), supplemented by (18), and terms of the same order in ε are identified. The determination of successive corrections $F_B^{(k)}$, $F_{B1}^{(k)}$ $(k = 0, 1, 2,...)$

is then achieved by combining the chosen initial condition with the requirement that the expansion in ε be uniform with respect to time, which amounts to eliminating secular divergences. The physically relevant solution of Eqs. (16) and (17) is then obtained by restricting the originally independent multiple time variables τ_0, τ_1, τ_2 to the "physical line":

$$\tau_0=\tau, \qquad \tau_1=\varepsilon\tau, \qquad \tau_2=\varepsilon^2\tau,... \tag{34}$$

on which the operator (32) reduces back to $\partial/\partial\tau$. The dependence of the distribution functions on the variable τ_j essentially defines the dynamical evolution on the time scale $\tau\simeq\varepsilon^{-j}$, $j=0, 1, 2,...$.

Finally, we shall assume that the initial condition is entirely contained in the zeroth-order terms:

$$\begin{aligned} F_B^{(0)}(B;\tau_0=0,\tau_1=0,\tau_2=0,...)&=F_B^{\varepsilon}(B;\tau=0)\\ F_{B1}^{(0)}(B,1;\tau_0=0,\tau_1=0,\tau_2=0,...)&=F_{B1}^{\varepsilon}(B,1;\tau=0)\end{aligned} \tag{35}$$

4. ε-EXPANSION AND DERIVATION OF THE REDUCED EQUATION FOR THE B PARTICLE

We first consider the zeroth-order terms in the coupled kinetic equations (16)–(17). Since at this order the integrated gas–B collision operator vanishes [see Eq. (29)], we are left with

$$\begin{aligned} \frac{\partial}{\partial\tau_0}F_B^{(0)}(B;\tau_0,\tau_1,\tau_2,...)&=0\\ \frac{\partial}{\partial\tau_0}F_{B1}^{(0)}(B,1;\tau_0,\tau_1,\tau_2,...)&=0\end{aligned} \tag{36}$$

So both distributions, $F_B^{(0)}$ and $F_{B1}^{(0)}$ do not depend on the time variable τ_0.

To first order in ε, the kinetic equations reduce to

$$\frac{\partial}{\partial\tau_0}F_B^{(1)}+\frac{\partial}{\partial\tau_1}F_B^{(0)}=\int d1\;\bar{T}_-^{(1)}(B,1)\,F_B^{(0)}F_{1|B}^{(0)} \tag{37a}$$

$$\frac{\partial}{\partial\tau_0}F_{B1}^{(1)}+\frac{\partial}{\partial\tau_1}F_{B1}^{(0)}=\int d2\;\bar{T}_-^{(1)}(B,2)\,F_B^{(0)}F_{1|B}^{(0)}F_{2|B}^{(0)} \tag{37b}$$

The action of the collision operator $\bar{T}_-^{(1)}$ is given in Eq. (30). Since $F_B^{(0)}$ and $F_{B1}^{(0)}$ and the r.h.s. of Eqs. (37) are independent of τ_0, we must impose

$$\frac{\partial}{\partial \tau_0} F_B^{(1)}(B; \tau_0, \tau_1, \tau_2,...) = 0$$
$$\frac{\partial}{\partial \tau_0} F_{B1}^{(1)}(B, 1; \tau_0, \tau_1, \tau_2,...) = 0 \tag{38}$$

to eliminate secular divergences. On the other hand, one can verify that Eq. (30) implies

$$\int d\mathbf{1}\, \bar{T}_-^{(1)}(B, 1)\, F_B(B)\, F^{\rm eq}(1 \mid \mathbf{X}) = 0 \tag{39}$$

The integrated operator $\bar{T}_-^{(1)}$ applied to a state of the system in which the fluid is in conditional equilibrium vanishes after integration over the fluid variables, whatever the state of particle B. Hence, the solution of Eqs. (37) consistent with the initial condition (22) has the form

$$\frac{\partial}{\partial \tau_1} F_B^{(0)}(B; \tau_1, \tau_2,...) = 0$$
$$F_{1 \mid B}^{(0)}(1 \mid B) = F^{\rm eq}(1 \mid \mathbf{X}) \tag{40}$$

The evolution of the system takes thus place on a longer timescale, corresponding to variable τ_2. The second order terms in the kinetic equations (16)–(17) yield the relations

$$\frac{\partial}{\partial \tau_0} F_B^{(2)} + \frac{\partial}{\partial \tau_1} F_B^{(1)} + \frac{\partial}{\partial \tau_2} F_B^{(0)}$$
$$= \int d\mathbf{1}\, \bar{T}_-^{(2)}(B, 1)\, F_B^{(0)} F^{\rm eq}(1 \mid \mathbf{X}) + \int d\mathbf{1}\, \bar{T}_-^{(1)}(B, 1)\, F_B^{(0)} F_{1 \mid B}^{(1)} \tag{41a}$$

$$\frac{\partial}{\partial \tau_0} F_{B1}^{(2)} + \frac{\partial}{\partial \tau_1} F_{B1}^{(1)} + \frac{\partial}{\partial \tau_2} F_B^{(0)} F^{\rm eq}(1 \mid \mathbf{X})$$
$$+ \left(\mathbf{u}_1 \cdot \frac{\partial}{\partial \mathbf{x}_1} - \bar{T}_-^{(0)}(B, 1) \right) F_B^{(0)} F^{\rm eq}(1 \mid \mathbf{X})$$
$$= \int d\mathbf{2}\, \bar{T}_-^{(2)}(B, 2)\, F_B^{(0)} F^{\rm eq}(1 \mid \mathbf{X})\, F^{\rm eq}(2 \mid \mathbf{X})$$
$$+ \int d\mathbf{2}\, \bar{T}_-^{(1)}(B, 2)\, F_B^{(0)} F^{\rm eq}(1 \mid \mathbf{X})\, F_{2 \mid B}^{(1)}$$
$$+ \int d\mathbf{2}\, \bar{T}_-(1, 2)\, F_B^{(0)} F^{\rm eq}(1 \mid \mathbf{X})\, F^{\rm eq}(2 \mid \mathbf{X}) \tag{41b}$$

In obtaining these equations, the equality $F^{(0)}_{1|B} = F^{eq}(1\,|\,\mathbf{X})$ and the relation (39) have been taken into account.

Since $F^{(0)}_B$ and $F^{(0)}_{B1}$ are independent of τ_0 and τ_1, and $F^{(1)}_B$ and $F^{(1)}_{B1}$ of τ_0, we conclude from (41) that in order to eliminate secular divergences one has to impose

$$\frac{\partial}{\partial\tau_0} F^{(2)}_B(B; \tau_0, \tau_1, \tau_2,...) = \frac{\partial}{\partial\tau_0} F^{(2)}_{B1}(B, 1; \tau_0, \tau_1, \tau_2,...) = 0$$

$$\frac{\partial}{\partial\tau_1} F^{(1)}_B(B; \tau_1, \tau_2,...) = \frac{\partial}{\partial\tau_1} F^{(1)}_{B1}(B, 1; \tau_1, \tau_2,...)$$

$$= \frac{\partial}{\partial\tau_1} F^{(1)}_{1|B}(1\,|\,B; \tau_1, \tau_2,...) = 0 \tag{42}$$

Now, since $F^{eq}(1\,|\,\mathbf{X})$ is the conditional equilibrium of the gas in the presence of the fixed B particle, the following relations are satisfied:

$$\left(\mathbf{u}_1 \cdot \frac{\partial}{\partial \mathbf{x}_1} - \bar{T}^{(0)}_-(B, 1)\right) F^{eq}(1\,|\,\mathbf{X}) = 0 \tag{43a}$$

$$\int d\mathbf{1}\; \bar{T}_-(1, 2)\, F^{eq}(1\,|\,\mathbf{X})\, F^{eq}(2\,|\,\mathbf{X}) = 0 \tag{43b}$$

In view of these equalities, both evolution equations (41a) and (41b) are found to reduce to

$$\frac{\partial}{\partial\tau_2} F^{(0)}_B = \zeta_B \frac{\partial}{\partial \mathbf{U}} \cdot \left(\frac{\partial}{\partial \mathbf{U}} + \mathbf{U}\right) F^{(0)}_B + \int d\mathbf{1}\; \bar{T}^{(1)}_-(B, 1)\, F^{(0)}_B F^{(1)}_{1|B} \tag{44}$$

where formula (31) for $\bar{T}^{(2)}_-(B, 1)$ has been used and we defined the Boltzmann friction coefficient ζ_B by

$$\zeta_B = \xi^2\, \tfrac{8}{3} \sqrt{2\pi} \tag{45}$$

The first term on the r.h.s. of (44) characterizes the instantaneous "static" friction force induced by collisions of the gas particles with particle B. By analogy with the hydrodynamic expression for the friction force ($\zeta \times$ velocity), the role of the velocity is played here by $(\partial/\partial\mathbf{U} + \mathbf{U})\, F^{(0)}_B(B)$. On the other hand, the physical meaning of the second term on the r.h.s.

of (44) can be clarified by introducing a mean dynamic friction force $\bar{\mathscr{F}}_+$ defined by

$$\bar{\mathscr{F}}_+(B; \tau_2) = \int d\mathbf{1}\, \mathscr{F}_+(1)\, F^{(1)}_{1|B}(1 \mid B; \tau_2) \tag{46}$$

This time-dependent force characterizes the "dynamical" part of the drag on the B particle, induced by the correlations building up inside the gas, in response to the motion of particle B. Using this definition, Eq. (44) can be rewritten

$$\frac{\partial}{\partial \tau_2} F^{(0)}_B(B; \tau_2,...)$$
$$= \frac{\partial}{\partial \mathbf{U}} \cdot \left\{ \zeta_B \left(\frac{\partial}{\partial \mathbf{U}} + \mathbf{U} \right) F^{(0)}_B(B; \tau_2,...) - \bar{\mathscr{F}}_+(B; \tau_2)\, F^{(0)}_B(B; \tau_2,...) \right\} \tag{47}$$

To close this equation, we have to evaluate the friction force (46). To this end we must consider the third-order terms in the kinetic equations (16), (17).

We first note that the reasoning already presented concerning the secular divergences [see Eq. (42)], when applied to the third-order equations, yields

$$\frac{\partial}{\partial \tau_0} F^{(3)}_B(B; \tau_0, \tau_1, \tau_2,...) = \frac{\partial}{\partial \tau_0} F^{(3)}_{B1}(B, 1; \tau_0, \tau_1, \tau_2,...) = 0$$

$$\frac{\partial}{\partial \tau_1} F^{(2)}_B(B; \tau_1, \tau_2,...) = \frac{\partial}{\partial \tau_1} F^{(2)}_{B1}(B, 1; \tau_1, \tau_2,...)$$
$$= \frac{\partial}{\partial \tau_1} F^{(2)}_{1|B}(1 \mid B; \tau_1, \tau_2,...) = 0 \tag{48}$$

The evolution equations to third order in ε thus take the form

$$\frac{\partial}{\partial \tau_2} F^{(1)}_B + \frac{\partial}{\partial \tau_3} F^{(0)}_B + \mathbf{U} \cdot \frac{\partial}{\partial \mathbf{X}} F^{(0)}_B$$
$$= \int d\mathbf{1}\, \bar{T}^{(1)}_-(B, 1)\, F^{(2)}_{B1} + \int d\mathbf{1}\, \bar{T}^{(2)}_-(B, 1)\, F^{(1)}_{B1} + \int d\mathbf{1}\, \bar{T}^{(3)}_-(B, 1)\, F^{(0)}_{B1} \tag{49a}$$

$$\frac{\partial}{\partial \tau_2}\{F_B^{(0)}F_{1|B}^{(1)}+F_B^{(1)}F^{\rm eq}(1\,|\,\mathbf{X})\}+\frac{\partial}{\partial \tau_3}F_B^{(0)}F^{\rm eq}(1\,|\,\mathbf{X})$$

$$+\left(\mathbf{u}_1\cdot\frac{\partial}{\partial \mathbf{x}_1}-\bar{T}_-^{(0)}(B,1)\right)F_B^{(0)}F_{1|B}^{(1)}$$

$$+\left\{\mathbf{U}\cdot\frac{\partial}{\partial \mathbf{X}}-\bar{T}_-^{(1)}(B,1)\right\}F_B^{(0)}F^{\rm eq}(1\,|\,\mathbf{X})$$

$$=\int d2\,\bar{T}_-^{(1)}(B,2)\,F_{B12}^{(2)}+\int d2\,\bar{T}_-^{(2)}(B,2)\,F_{B12}^{(1)}+\int d2\,\bar{T}_-^{(3)}(B,2)\,F_{B12}^{(0)}$$

$$+\int d2\,\bar{T}_-(1,2)\,F_{B12}^{(1)} \tag{49b}$$

where the relation (43) has already been taken into account. The functions $F_{B1}^{(k)}=\{F_BF_{1|B}\}^{(k)}$ and $F_{B12}^{(k)}=\{F_BF_{1|B}F_{2|B}\}^{(k)}$ in the collision terms represent the kth-order contributions $k=0, 1, 2$. For instance,

$$\begin{aligned}F_{B12}^{(2)}(B,1,2)&=\{F_BF_{1|B}F_{2|B}\}^{(2)}\\&=F_B^{(2)}F^{\rm eq}(1\,|\,\mathbf{X})\,F^{\rm eq}(2\,|\,\mathbf{X})\\&\quad+F_B^{(1)}\{F_{1|B}^{(1)}F^{\rm eq}(2\,|\,\mathbf{X})+F^{\rm eq}(1\,|\,\mathbf{X})\,F_{2|B}^{(1)}\}\\&\quad+F_B^{(0)}\{F_{1|B}^{(2)}F^{\rm eq}(2\,|\,\mathbf{X})+F^{\rm eq}(1\,|\,\mathbf{X})\,F_{2|b}^{(2)}+F_{1|B}^{(1)}F_{2|B}^{(1)}\}\end{aligned} \tag{50}$$

Our concern is now to obtain a closed equation for the conditional distribution of the gas $F_{1|B}^{(1)}(1\,|\,B)$. To this end, we have to eliminate the terms containing the τ_3-variable, which can be achieved by subtracting from (49b) the first equation (49a) multiplied by $F^{\rm eq}(1\,|\,\mathbf{X})$. One finds

$$\frac{\partial}{\partial \tau_2}\left(F_B^{(0)}F_{1|B}^{(1)}\right)+\left(\mathbf{u}_1\cdot\frac{\partial}{\partial \mathbf{x}_1}-\bar{T}_-^{(0)}(B,1)\right)F_B^{(0)}F_{1|B}^{(1)}$$

$$=\bar{T}_-^{(1)}(B,1)\,F_B^{(0)}F^{\rm eq}(1\,|\,\mathbf{X})-F_B^{(0)}\mathbf{U}\cdot\frac{\partial}{\partial \mathbf{X}}F^{\rm eq}(1\,|\,\mathbf{X})$$

$$+\int d2\,\bar{T}_-^{(1)}(B,2)\,F_B^{(0)}F_{1|B}^{(1)}F_{2|B}^{(1)}$$

$$+\int d2\,\bar{T}_-^{(2)}(B,2)\,F_B^{(0)}F_{1|B}^{(1)}F^{\rm eq}(2\,|\,\mathbf{X})$$

$$+\int d2\,\bar{T}_-(1,2)\,F_B^{(0)}\{F_{1|B}^{(1)}F^{\rm eq}(2\,|\,\mathbf{X})+F^{\rm eq}(1\,|\,\mathbf{X})\,F_{2|B}^{(1)}\} \tag{51}$$

The last term of (51) introduces the (dimensionless) linearized Boltzmann operator $\Lambda_B(1)$, defined as[19]

$$\Lambda_B(1)\,\Psi(1\,|\,B)=\int d2\,F^{\text{eq}}(1\,|\,\mathbf{X})\,\phi(\mathbf{u}_2)\,\bar{T}_-(1,2)\left\{\left(\frac{\Psi(1\,|\,B)}{\phi(\mathbf{u}_1)}\right)+\left(\frac{\Psi(2\,|\,B)}{\phi(\mathbf{u}_2)}\right)\right\} \tag{52}$$

where $\phi(\mathbf{u})$ is the Maxwell distribution. Note that $\Lambda_B(1)$ acts only on the fluid variables.

With the use of relations (27), (30), and (31), Eq. (51) reduces to

$$\begin{aligned}&\left\{\frac{\partial}{\partial\tau_2}+\mathbf{u}_1\cdot\frac{\partial}{\partial\mathbf{x}_1}-\bar{T}_-^{(0)}(B,1)-\Lambda_B(1)\right.\\&\quad\left.+\frac{\partial}{\partial\mathbf{U}}\cdot\left(\bar{\bar{\mathscr{F}}}_+(B)-\zeta_B\left(\frac{\partial}{\partial\mathbf{U}}+\mathbf{U}\right)\right)\right\}F_B^{(0)}(B)\,F_{1|B}^{(1)}(1\,|\,B)\\&=-\mathscr{F}_-(1)\cdot\left(\frac{\partial}{\partial\mathbf{U}}+\mathbf{U}\right)F_B^{(0)}(B)\,F^{\text{eq}}(1\,|\,\mathbf{X})\end{aligned} \tag{53}$$

The notation

$$\begin{aligned}\mathscr{L}_f&=\mathbf{u}_1\cdot\frac{\partial}{\partial\mathbf{x}_1}-\bar{T}_-^{(0)}(B,1)-\Lambda_B(1)\\\mathscr{L}_B&=\frac{\partial}{\partial\mathbf{U}}\cdot\left\{\bar{\bar{\mathscr{F}}}_+(B)-\zeta_B\left(\frac{\partial}{\partial\mathbf{U}}+\mathbf{U}\right)\right\}\equiv\frac{\partial}{\partial\mathbf{U}}\cdot\bar{\bar{\mathscr{F}}}_f\end{aligned} \tag{54}$$

and

$$F^{\text{corr}}(B,1;\tau_2)=F_B^{(0)}(B)\,F_{1|B}^{(1)}(1\,|\,B) \tag{55}$$

allows us to cast (53) in a more transparent form

$$\begin{aligned}&\left\{\frac{\partial}{\partial\tau_2}+\mathscr{L}_f+\mathscr{L}_B\right\}F^{\text{corr}}(B,1;\tau_2)\\&=-\mathscr{F}_-(1)\cdot\left(\frac{\partial}{\partial\mathbf{U}}+\mathbf{U}\right)F_B^{(0)}(B;\tau_2)\,F^{\text{eq}}(1\,|\,\mathbf{X})\end{aligned} \tag{56}$$

The force $\bar{\bar{\mathscr{F}}}_f$ appearing in the definition (54) of $\mathscr{L}_B$, can be interpreted as the total friction force acting on particle B. The system of equations (47), (56) is closed and characterizes the dynamical evolution of the distribution functions on the τ_2 time scale.

The formal solution of Eq. (56) can be written as

$$F^{\mathrm{corr}}(B, 1; \tau_2) = -\int_0^{\tau_2} ds \exp\left\{-\int_s^{\tau_2} ds' \, (\mathscr{L}_f + \mathscr{L}_B)(s')\right\} \mathscr{F}_-(1)$$
$$\times\left(\frac{\partial}{\partial \mathbf{U}} + \mathbf{U}\right) F_B^{(0)}(B; s) \, F^{\mathrm{eq}}(1 \mid \mathbf{X}) \tag{57}$$

However, as can be verified from the definitions (54) combined with relations (26), (52), and (46), $\mathscr{L}_f$ acts only on the fluid variables, while $\mathscr{L}_B$ acts only on the velocity of the B particle. The two operators thus commute and the solution for $F^{\mathrm{corr}}(B, 1)$ reads

$$F^{\mathrm{corr}}(B, 1; \tau_2) = -\int_0^{\tau_2} ds \, \mathscr{F}_-(1; -(\tau_2 - s)) \, F^{\mathrm{eq}}(1 \mid \mathbf{X})$$
$$\times \exp\left\{-\int_s^{\tau_2} ds' \, \mathscr{L}_B(s')\right\}\left(\frac{\partial}{\partial \mathbf{U}} + \mathbf{U}\right) F_B^{(0)}(B; s) \tag{58}$$

where $\mathscr{F}_-(1; -\tau)$ denotes the force $\mathscr{F}_-(1; 0)$ on particle B propagated by the intrinsic fluid dynamics in the presence of the B particle, fixed at point $\mathbf{X}$ [see the definition of $\bar{T}_-^{(0)}$, (26)], backward in time to the instant $-\tau$.

The dynamical part of the mean friction force, $\bar{\mathscr{F}}_+(B)$, defined in (46), can now be obtained by multiplying (58) by $\mathscr{F}_+(1)$ and averaging over the fluid variables. This yields

$$\bar{\mathscr{F}}_+(B; \tau_2) \, F_B^{(0)}(B; \tau_2)$$
$$\equiv \int d\mathbf{1} \, \mathscr{F}_+(1) \, F^{\mathrm{corr}}(B, 1; \tau_2)$$
$$= \int_0^{\tau_2} ds \, \mathscr{C}^{\mathrm{dyn}}(\tau_2 - s) \exp\left\{-\int_s^{\tau_2} ds' \, \mathscr{L}_B(s')\right\}\left(\frac{\partial}{\partial \mathbf{U}} + \mathbf{U}\right) F_B^{(0)}(B; s) \tag{59}$$

where we have introduced $\mathscr{C}^{\mathrm{dyn}}(\tau)$, a force–force correlation function, defined by

$$\mathscr{C}^{\mathrm{dyn}}(\tau) = \tfrac{1}{3}\int d\mathbf{1} \, \mathscr{F}_-(1; -\tau) \cdot \mathscr{F}_+(1; 0) \, F^{\mathrm{eq}}(1 \mid \mathbf{X})$$
$$\equiv \tfrac{1}{3}\langle \mathscr{F}_-(1; -\tau) \cdot \mathscr{F}_+(1; 0)\rangle_{(\mathrm{eq} \mid \mathbf{X})} \tag{60}$$

By using the notation $\langle ... \rangle_{(\text{eq}\,|\,\mathbf{X})}$ we stress the fact that the average is taken over the *equilibrium* ensemble of the fluid in the presence of particle B *fixed* at point **X**. The prefactor 1/3 stems from the isotropy of the system around the B particle.

Moreover, Eq. (59) can be seen as a self-consistency relation for the (time-dependent) mean force $\bar{\mathscr{F}}_+$, because the propagator $\mathscr{L}_B$ defined in (54) itself depends on the friction force $\bar{\mathscr{F}}_+$. Substituting this expression for $\bar{\mathscr{F}}_+$ in Eq. (47), one arrives at a reduced equation for the distribution of the B particle

$$\frac{\partial}{\partial \tau_2} F_B^{(0)}(B; \tau_2) = \zeta_B \frac{\partial}{\partial \mathbf{U}} \cdot \left(\frac{\partial}{\partial \mathbf{U}} + \mathbf{U} \right) F_B^{(0)}(B; \tau_2) + \int_0^{\tau_2} ds \, \mathscr{C}^{\text{dyn}}(\tau_2 - s) \frac{\partial}{\partial \mathbf{U}} \times \exp\left\{ -\int_s^{\tau_2} ds' \, \mathscr{L}_B(s') \right\} \left(\frac{\partial}{\partial \mathbf{U}} + \mathbf{U} \right) F_B^{(0)}(B; s) \tag{61}$$

where $\mathscr{L}_B$ is defined in (54) and $\mathscr{C}^{\text{dyn}}$ in (60).

At this stage, we can return to the original variables **R**, **V** and t, using relations (9), (10), (12), and (13). For the sake of simplicity, we keep the same notations for ζ_B, $\mathscr{C}_{\text{dyn}}$, and for different forces and propagators involved in the evolution equations. Their full expressions are now given by

$$\zeta_B = \frac{1}{M} \left(\frac{\Sigma}{2} \right)^2 \frac{8}{3} n (2\pi m k_B T)^{1/2} \tag{62}$$

$$\mathscr{C}^{\text{dyn}}(t) = \frac{1}{3 M k_B T} \langle \mathscr{F}_-(1; -t) \cdot \mathscr{F}_+(1; 0) \rangle_{(\text{eq}\,|\,\mathbf{R})} \tag{63}$$

where the formula for the microscopic forces $\mathscr{F}_\pm$ now reads

$$\mathscr{F}_\mp(1) = \left(\frac{\Sigma}{2} \right)^2 \int d\hat{\sigma} \, 2m(\mathbf{v}_1 \cdot \hat{\sigma})^2 \, \theta(\mp \mathbf{v}_1 \cdot \hat{\sigma}) \, \hat{\sigma} \, \delta\left(\mathbf{R} - \left(\frac{\Sigma}{2} \right) \hat{\sigma} - \mathbf{r}_1 \right) \tag{64}$$

In (63), the notation (eq|**R**) stands for an average over the equilibrium ensemble of the gas, in the presence of particle B, *fixed* at **R**. The dynamics is characterized by the propagator $\mathscr{L}_f$, given in the Appendix together with $\mathscr{L}_B$ [for the dimensionless form, see Eq. (54)].

Keeping in mind that we are only interested in the dynamics of the system on the time scale characterized by the variable τ_2, i.e., $\tau \sim \varepsilon^{-2}$ [τ is the dimensionless time variable defined in Eq. (10)], we obtain the following equation for the distribution function f_B of the B particle:

$$\frac{\partial}{\partial t} f_B(B; t) = \zeta_B \frac{\partial}{\partial \mathbf{V}} \cdot \left(\mathbf{V} + \frac{k_B T}{M} \frac{\partial}{\partial \mathbf{V}}\right) f_B(B; t) + \int_0^t ds\, \mathscr{C}^{\mathrm{dyn}}(t-s) \frac{\partial}{\partial \mathbf{V}} \cdot \exp\left\{-\int_s^t ds'\, \mathscr{L}_B(s')\right\} \times \left(\mathbf{V} + \frac{k_B T}{M} \frac{\partial}{\partial \mathbf{V}}\right) f_B(B; s) \tag{65}$$

This should be completed by the self-consistency equation for the dynamical part $\bar{\mathscr{F}}_+$ of the friction force

$$\bar{\mathscr{F}}_+(B; t)\, f_B(B; t) = \int_0^t ds\, \mathscr{C}^{\mathrm{dyn}}(t-s) \exp\left\{-\int_s^t ds'\, \mathscr{L}_B(s')\right\} \times \left(\mathbf{V} + \frac{k_B T}{M} \frac{\partial}{\partial \mathbf{V}}\right) f_B(B; s) \tag{66}$$

The system of equations (65)–(66) is closed and represents the dynamical evolution of the state of the B particle on the $\tau \sim \varepsilon^{-2}$ time scale. This condition can be rewritten $\varepsilon^3 \tau \ll 1$, so that using Eq. (10), we conclude that Eq. (65) applies for times t satisfying

$$t \ll \frac{\Sigma}{\sqrt{k_B T/M}} \times \frac{\rho_B}{\rho} \sim \frac{\Sigma}{\sqrt{k_B T/M}} \tag{67}$$

The spatial diffusion process has not yet started at this time scale, since time t is very short compared to the time needed to cover the B particle radius with the thermal velocity. However the fluid hydrodynamics is already at work. The time scale (67) thus characterizes the relaxation of the velocity of the B particle, while its spatial state is not yet affected and will relax only on a longer time scale.

Clearly, the derived equation is *not* of a Fokker–Planck type. The main reason is that the friction force due to the bath builds up on the same time scale as that characterizing the gas dynamics, which leads to memory effects in the relaxation of particle B. The friction force $\bar{\mathscr{F}}_f$ thus has to be constructed in a self-consistent way and depends on the whole history of the Brownian motion.

Equation (65) and its systematic derivation are the main results of the present work.

5. LONG-TIME LIMIT OF THE REDUCED EQUATION

In this section, we discuss briefly the long-time limit of the system (65)–(66). Note, however that we stay in the time window $\tau \sim \varepsilon^{-2}$, since it is only there that these equations represent the evolution of the system. On longer time scales (e.g., $\tau \sim \varepsilon^{-3}$, $\tau \sim \varepsilon^{-4}$), we expect the spatial relaxation to occur through the Smoluchowski equation (see footnote 3 in the Introduction).[12]

In the long-time limit, the B-particle velocity distribution will relax toward the stationary solution of (65), that is, toward the Maxwellian distribution. In the final stage, we can then write

$$\left(\mathbf{V} + \frac{k_B T}{M} \frac{\partial}{\partial \mathbf{V}}\right) f_B(B; t) \simeq 0 \tag{68}$$

and the friction force $\bar{\mathscr{F}}_f$ will accordingly decay to zero, too. Then to first order in this quantity, we can put $\mathscr{L}_B \simeq 0$ in the expression (66) for $\bar{\mathscr{F}}_+$ and obtain

$$\bar{\mathscr{F}}_+(B; t)\, f_B(B; t) \simeq \int_0^t ds\, \mathscr{C}^{\text{dyn}}(t-s) \left(\mathbf{V} + \frac{k_B T}{M} \frac{\partial}{\partial \mathbf{V}}\right) f_B(B; s) \tag{69}$$

The limiting form of the reduced equation for $f_B(B; t)$ then simplifies to

$$\frac{\partial}{\partial t} f_B(B; t) \simeq \int_0^t ds\, \zeta(t-s) \frac{\partial}{\partial \mathbf{V}} \cdot \left(\mathbf{V} + \frac{k_B T}{M} \frac{\partial}{\partial \mathbf{V}}\right) f_B(B; s) \tag{70}$$

where the time-dependent friction coefficient

$$\zeta(t) = \zeta_B\, \delta(t) + \mathscr{C}^{\text{dyn}}(t) \tag{71}$$

has been introduced. $\delta(t)$ is the Dirac distribution.

This asymptotic form for the reduced equation of the B distribution calls for several comments. First, though much simpler than the complete equation (65) [together with (66)], it still exhibits a non-markovian nature. However, the nonlocality of (65) in velocity space—hidden in the self-consistent definition of $\bar{\mathscr{F}}_+$ in (66)—in is now removed and the simplified form (70) only involves the Fokker–Planck operator

$$\frac{\partial}{\partial \mathbf{V}} \cdot \left(\mathbf{V} + \frac{k_B T}{M} \frac{\partial}{\partial V}\right)$$

In this sense, the velocity can be seen as following a "generalized Ornstein–Uhlenbeck process," still characterized by a transition probability sharply peaked around the mean value (i.e., only small jumps occur), but now keeping the memory of its whole previous history.

In particular, whereas for the Ornstein-Uhlenbeck process the B velocity relaxes in an exponential way,[1,3] the long-time behavior of the friction coefficient $\zeta(t)$ modifies the nature of the thermalization process. In other words, the *way* $\zeta(t)$ decays with time does affect the thermalization process itself, i.e., the relaxation of $f_B(B; t)$ toward the Maxwell distribution. This is a crucial difference with the standard Fokker–Planck (or Langevin) equation, where only the time-integrated friction coefficient $\int_0^\infty dt\, \zeta(t)$ plays a role. These equations lead in particular to an *exponential* decay of the velocity autocorrelation function, i.e., an exponentially fast thermalization. This differs substantially from our case. Indeed, $\zeta(t)$ is known from hydrodynamic arguments to exhibit a $t^{-3/2}$ long-time tail.[20] We thus expect $f_B(B; t)$ from Eq. (70) to decay in an *algebraic* way toward the Maxwell distribution. This behavior can be verified on the first moments of $f_B(B; t)$, i.e., the mean velocity (for a full discussion, see next section), mean squared fluctuations of the velocity, etc. A closed equation for these moments can be obtained by multiplying Eq. (70) by **V**, **VV**,... and then integrating over the velocity. However, an exact solution of Eq. (70) can be found in Refs. 21 and 22, where all these results can be directly checked.

This nonexponential relaxation and its implications will be discussed in the conclusions of the paper (Sect. 8).

6. VELOCITY AUTOCORRELATION FUNCTION AND THE STOKES–EINSTEIN RELATION

As we discussed above, the reduced equation (65) [together with (66)] characterizes the relaxation of the velocity of the B particle, while the spatial relaxation will occur on a longer time scale. As we shall see in this section, this time scale separation allows one to compute explicitly the velocity ACF of the B particle. In spite of the nonlocal nature of the complete system (65)–(66), we will eventually recover the Stokes–Einstein relation between the diffusion and the friction coefficient.[2]

Let us study the dynamical evolution of the mean velocity $\bar{\mathbf{V}}_B$ of the B particle, defined as

$$\bar{\mathbf{V}}_B(t) = \int d\mathbf{V}\, \mathbf{V} f_B(B; t) \tag{72}$$

Since $\bar{\mathbf{V}}_B(t)$ relaxes on the time scale $\tau \sim \varepsilon^{-2}$, f_B will be assumed to evolve according to Eq. (65). The evolution equation for $\bar{\mathbf{V}}_B(t)$ can then be obtained by multiplying (65) by $\mathbf{V}$ and integrating over the velocity. This yields

$$\frac{\partial}{\partial t}\bar{\mathbf{V}}_B(t) = \int d\mathbf{V}\,\zeta_B \mathbf{V}\,\frac{\partial}{\partial \mathbf{V}}\cdot\left(\mathbf{V} + \frac{k_B T}{M}\frac{\partial}{\partial \mathbf{V}}\right) f_B(B; t)$$
$$+ \int_0^t ds\,\mathscr{C}^{\text{dyn}}(t-s)\int d\mathbf{V}\,\mathbf{V}\,\frac{\partial}{\partial \mathbf{V}}\cdot \exp\left\{-\int_s^t du\,\mathscr{L}_B(u)\right\}$$
$$\times\left(\mathbf{V} + \frac{k_B T}{M}\frac{\partial}{\partial \mathbf{V}}\right) f_B(B; s) \tag{73}$$

The first term is calculated by an integration by parts, yielding

$$\int d\mathbf{V}\,\mathbf{V}\,\frac{\partial}{\partial \mathbf{V}}\cdot\left(\mathbf{V} + \frac{k_B T}{M}\frac{\partial}{\partial \mathbf{V}}\right) f_B(B) = -\bar{\mathbf{V}}_B(t) \tag{74}$$

The second term needs a more careful analysis. Let us introduce the notation

$$\gamma(\mathbf{V}; t\,|\,s) = \exp\left\{-\int_s^t du\,\mathscr{L}_B(u)\right\}\left(\mathbf{V} + \frac{k_B T}{M}\frac{\partial}{\partial \mathbf{V}}\right) f_B(B; s) \tag{75}$$

We thus have to compute

$$\int d\mathbf{V}\,\mathbf{V}\,\frac{\partial}{\partial \mathbf{V}}\cdot \gamma(\mathbf{V}; t\,|\,s) = -\int d\mathbf{V}\,\gamma(\mathbf{V}; t\,|\,s) \tag{76}$$

where an integration by parts has been performed. According to (75), $\gamma(\mathbf{V}; t\,|\,s)$ solves the following initial value problem:

$$\begin{cases} \left(\dfrac{\partial}{\partial t} + \mathscr{L}_B(t)\right)\gamma(\mathbf{V}; t\,|\,s) = 0, & t \geqslant s \\ \gamma(\mathbf{V}; t = s\,|\,s) = \left(\mathbf{V} + \dfrac{k_B T}{M}\dfrac{\partial}{\partial \mathbf{V}}\right) f_B(B; s), & t = s \end{cases} \tag{77}$$

Integrating Eq. (77) over $\mathbf{V}$, one obtains

$$\frac{\partial}{\partial t}\int d\mathbf{V}\,\gamma(\mathbf{V}; t\,|\,s) + \int d\mathbf{V}\,\mathscr{L}_B(t)\,\gamma(\mathbf{V}; t\,|\,s) = 0 \tag{78}$$

But according to Eq. (A.1) in the Appendix, the action of the operator $\mathscr{L}_B$ involves multiplication by the total friction force $\bar{\bar{\mathscr{F}}}_f$ and then derivation with respect to $\mathbf{V}$. The second term in (78) thus vanishes, since

$$\int d\mathbf{V}\, \mathscr{L}_B(t)\, \gamma(\mathbf{V}; t\,|\,s) = \int d\mathbf{V}\, \frac{\partial}{\partial \mathbf{V}} \cdot \{\bar{\bar{\mathscr{F}}}_f \gamma(\mathbf{V}; t\,|\,s)\} = 0 \tag{79}$$

and we are left with

$$\frac{\partial}{\partial t} \int d\mathbf{V}\, \gamma(\mathbf{V}; t\,|\,s) = 0 \tag{80}$$

We thus eventually find

$$\begin{aligned}\int d\mathbf{V}\, \gamma(\mathbf{V}; t\,|\,s) &= \int d\mathbf{V}\, \gamma(\mathbf{V}; t = s\,|\,s)\\ &= \int d\mathbf{V} \left(\mathbf{V} + \frac{k_B T}{M} \frac{\partial}{\partial \mathbf{V}}\right) f_B(B; s)\\ &= \bar{\mathbf{V}}_B(s)\end{aligned} \tag{81}$$

Combining Eqs. (73), (74), (76), and (81), we obtain a closed equation for $\bar{\mathbf{V}}_B$ of the form

$$\frac{\partial}{\partial t} \bar{\mathbf{V}}_B(t) = -\int_0^t ds\, \zeta(t-s)\, \bar{\mathbf{V}}_B(s) \tag{82}$$

where $\zeta(t) = \zeta_B\, \delta(t) + \mathscr{C}^{\text{dyn}}(t)$ is the time-dependent friction coefficient.

Introducing the Laplace transform

$$\tilde{\bar{\mathbf{V}}}_B(z) = \int_0^{+\infty} dt\, \exp(-z \cdot t)\, \bar{\mathbf{V}}_B(t) \tag{83}$$

one obtains the explicit solution for $\tilde{\bar{\mathbf{V}}}_B(z)$ as

$$\tilde{\bar{\mathbf{V}}}_B(z) = \frac{\mathbf{V}_B(t=0)}{z + \tilde{\zeta}(z)} \tag{84}$$

where $\tilde{\zeta}(z)$ denotes the Laplace transform of the time-dependent friction coefficient.

Equation (84) characterizes the relaxation of the mean velocity of the suspended sphere in a situation in which the B particle is initially out of equilibrium. But according to Onsager's principle of regression of fluctuations, this equation should as well describe the relaxation of a *fluctuation* of the velocity of the B particle *at equilibrium*.[23] This can be explicitly verified here.

At equilibrium, the B-particle velocity autocorrelation function is defined as

$$\langle \mathbf{V}(t) \cdot \mathbf{V}(0) \rangle_{\text{eq}} = \int d\mathbf{B}\, d\mathbf{1} \cdots d\mathbf{N}\, \mathbf{V}(t) \cdot \mathbf{V}(0)\, \rho^{\text{eq}}(\mathbf{B}, \mathbf{1} \cdots \mathbf{N}) \tag{85}$$

where $\rho^{\text{eq}}(\mathbf{B}, \mathbf{1} \cdots \mathbf{N})$ is the canonical equilibrium probability density of the system, and $\mathbf{V}(t)$ denotes the velocity of the B particle propagated in time through the dynamics of the complete system. Formally, this relation can be rewritten

$$\begin{aligned} \langle \mathbf{V}(t) \cdot \mathbf{V}(0) \rangle_{\text{eq}} &= \int d\mathbf{B}\, \mathbf{V}(0) \cdot \int d\mathbf{1} \cdots d\mathbf{N}\, \mathbf{V}(t)\, \rho^{\text{eq}}(\mathbf{B}, \mathbf{1} \cdots \mathbf{N}) \\ &= \int d\mathbf{B}\, \mathbf{V}(0) \cdot \langle \mathbf{V}(t) \rangle_{\text{n.e.}}\, F^{\text{eq}}(\mathbf{V}(0)) \end{aligned} \tag{86}$$

where $F^{\text{eq}}(\mathbf{V})$ is the equilibrium distribution function of particle B alone, and $\langle \mathbf{V}(t) \rangle_{\text{n.e.}}$ denotes the time-dependent mean velocity of the B particle, averaged over the fluid variables, for a given initial out-of-equilibrium state of velocity $\mathbf{V}(0)$.

But this mean velocity will, satisfy the evolution equation (82) derived previously by eliminating the fluid variables. This yields for the Laplace transform of the velocity ACF

$$\langle \mathbf{V}(z) \cdot \mathbf{V}(0) \rangle_{\text{eq}} = \frac{k_B T/M}{z + \tilde{\zeta}(z)} \tag{87}$$

The diffusion coefficient can be directly deduced from this relation, using

$$\begin{aligned} D &\equiv \int_0^\infty dt\, \langle \mathbf{V}(t) \cdot \mathbf{V}(0) \rangle_{\text{eq}} \\ &= \langle \tilde{\mathbf{V}}(z=0) \cdot \mathbf{V}(0) \rangle_{\text{eq}} \end{aligned} \tag{88}$$

This yields

$$D = \frac{k_B T/M}{\tilde{\zeta}(z=0)} = \frac{k_B T}{M\zeta_f} \tag{89}$$

where, according to (71), the microscopic expression for the (integrated) friction coefficient ζ_f is given by

$$\begin{aligned} \zeta_f &\equiv \int_0^{+\infty} d\tau\, \zeta(\tau) \\ &= \zeta_B + \frac{1}{3Mk_B T} \int_0^{\infty} dt\, \langle \mathscr{F}_-(1; -t) \cdot \mathscr{F}_-(1; 0) \rangle_{(\mathrm{eq}\,|\,\mathbf{R})} \end{aligned} \tag{90}$$

with ζ_B given in (62), and the microscopic forces defined in (64).

Equation (89) is precisely the Stokes–Einstein relation for the friction coefficient.

7. FRICTION COEFFICIENT AND THE HYDRODYNAMIC LIMIT

In this section, we discuss the microscopic formula (90) obtained for the friction coefficient. Our aim is twofold. First, we would like to connect these results to the friction coefficient in the response of the fluid to an imposed motion of the B particle. Second, we would like to obtain, at least in some limits, explicit expressions for the friction coefficient as a function of the microscopic characteristics of the system (B-particle diameter, transport coefficients of the fluid,...). This can be achieved by using the results of van Beijeren and Dorfman (DVB), concerning the kinetic theory of hydrodynamic flows.[17]

In accordance with (90), the friction coefficient is the sum of two terms. The first term, ζ_B, characterizes the static effect of instantaneous collisions between the B particle and the gas. The remaining part of the friction coefficient involves a time integral of a force–force correlation function, reflecting the dynamical correlations induced by time-displaced collisions between the gas and the B particle. As we discussed in our previous work (Ref. 24, henceforth cited as II), formula (90) is equivalent to the Kirkwood formula for smooth potentials, relating the friction coefficient to the time integral of the force autocorrelation function. In the case of hard-sphere interaction, however, the force is replaced by the momentum transferred to the B particle during instantaneous collisions, and the separation into a static term and a dynamical part then occurs (see II for further details). Moreover, it is interesting to note that the same microscopic expression for the friction coefficient was obtained in the limit considered in our previous work, $\varepsilon \to 0$, all other parameters kept constant. As discussed in the introduction, this limit leads to the Fokker–Planck equation for the B particle, so that its evolution exhibits a Markovian nature. We thus see that the same properties of the fluid are involved in both limits, but they enter the dynamical laws at different levels. They are intrinsic hydrodynamic properties of the fluid.

Let us consider the case in which the B particle has an imposed velocity $\mathbf{U}(t)$. This amounts to representing the B-particle distribution by a Dirac δ-function in velocity space, centered on the mean velocity $\bar{\mathbf{V}}(t) = U(t)$, with no thermal fluctuations around this value. The mean friction force $\bar{\mathscr{F}}_f$ acting on B can be then obtained from Eq. (82), determining evolution of the mean velocity of the B particle,

$$\bar{\mathscr{F}}_f(t) = -\int_0^t ds\, M\zeta(t-s)\, \mathbf{U}(s) \tag{91}$$

which identifies $\zeta(t)$ as the time-dependent friction coefficient in the hydrodynamic sense. We would like to stress the fact that this result holds (as shown in Sect. 6) although the reduced equation (65) is nonlocal in velocity space!

Let us now consider the dynamical part of the friction coefficient, which we defined in (71) and (90), as

$$\zeta_{\text{dyn}}(t) = \frac{1}{3Mk_B T} \int d\mathbf{1} \int_0^t ds\, \mathscr{F}_-(1; -s) \cdot \mathscr{F}_+(1; 0)\, f^{\text{eq}}(1 \mid \mathbf{R}) \tag{92}$$

Our aim now is to connect this expression to the calculations of DVB, who have computed the drag on a macroscopic sphere moving with constant velocity. The surrounding gas was assumed to obey an extended Boltzmann equation.

To this end, we first introduce an auxiliary function $f^c(1 \mid \mathbf{R})$ defined as

$$\begin{aligned} f^c(1 \mid \mathbf{R}; t) &= \int_0^t ds\, \mathscr{F}_-(1; -s) \cdot \hat{\mathbf{V}} f^{\text{eq}}(1 \mid \mathbf{R}) \\ &= \int_0^t ds \exp(-s\mathscr{L}_f)\, \mathscr{F}_-(1) \cdot \hat{\mathbf{V}} f^{\text{eq}}(1 \mid \mathbf{R}) \end{aligned} \tag{93}$$

where $\mathscr{L}_f$ is the fluid propagator [see Eq. (A.2) in the appendix]. $\hat{\mathbf{V}}$ is a unit vector introduced to establish the connections with the DVB analysis. According to (93), $f^c(1\,|\,\mathbf{R};t)$ satisfies the following differential equation:

$$\left(\frac{\partial}{\partial t}+\mathscr{L}_f\right) f^c(1\,|\,\mathbf{R};t)=\mathscr{F}_-(1)\, f^{\text{eq}}(1\,|\,\mathbf{R})\cdot\hat{\mathbf{V}} \tag{94}$$

Now if we define $\Psi(1\,|\,\mathbf{R};t)$ through the relation

$$f^c(1\,|\,\mathbf{R};t)=f^{\text{eq}}(1\,|\,\mathbf{R})\,\Psi(1\,|\,\mathbf{R};t) \tag{95}$$

then Eq. (94) can be rewritten

$$\left(\frac{\partial}{\partial t}+\mathscr{L}_f\right)\Psi(1\,|\,\mathbf{R};t)=T'(B,1)\frac{m}{k_B T}\mathbf{v}_1\cdot\hat{\mathbf{V}} \tag{96}$$

where $\mathbf{v}_1$ is the velocity of a gas particle, and the operator $T'(B,1)$ is that defined in DVB:

$$T'(B,1)=\left(\frac{\Sigma}{2}\right)^2\int d\hat{\sigma}(-\mathbf{v}_1\cdot\hat{\sigma})\,\theta(-\mathbf{v}_1\cdot\hat{\sigma})\,\delta\left(\mathbf{R}-\frac{\Sigma}{2}\hat{\sigma}-\mathbf{r}_1\right)[b_{\hat{\sigma}}^{(0)}(B,1)-1] \tag{97}$$

the operator $b_{\hat{\sigma}}^{(0)}$ changes the velocity $\mathbf{v}_1$ of the gas particle to $\mathbf{v}_1-2(\mathbf{v}_1\cdot\hat{\sigma})\,\hat{\sigma}$.

Equation (96) is equivalent to the inhomogeneous Boltzmann equation (4.4) in DVB. In their work, $\Psi(1\,|\,\mathbf{R};t)$ represents the dynamical correction to the distribution of the gas, induced by recollision events between the gas particles and the suspended sphere, which moves with velocity $\hat{\mathbf{V}}$. The authors obtained approximate solutions of this equation in the limit where the mean-free path ℓ is small compared to the radius $\Sigma/2$ of the suspended sphere. This solution was obtained by decomposing the distribution functions into a hydrodynamic part and a boundary layer part. We refer to their paper for further details.[17] Their solution may be used to compute the dynamical part of ζ_{dyn}. Indeed, with the aid of (92), (93), and (95), the dynamical contribution can be rewritten as

$$Mk_B T\zeta_{\text{dyn}}(t)=\int d\mathbf{1}\,\mathscr{F}_+(1;0)\,\Psi(1\,|\,R;t)\, f^{\text{eq}}(1\,|\,\mathbf{R})\cdot\hat{\mathbf{V}} \tag{98}$$

where we used the isotropy of the system when introducing $\hat{\mathbf{V}}$. In DVB, the r.h.s. of (98) is interpreted as the dynamical part of the *drag force* exerted by the fluid on the suspended sphere moving with velocity $\hat{\mathbf{V}}$. The established connection between both approaches reflects the linear-response theory, and can be physically interpreted through Onsager's principle of regression of fluctuations.

In the limit of long times, we can then use the results (6.43) and (6.44) of DVB to obtain to lowest order in l/Σ

$$\zeta_{\mathrm{dyn}}(t \to \infty) = \frac{2\pi\eta\Sigma}{M} - \zeta_B \tag{99}$$

where ζ_B is the Boltzmann friction coefficient, defined in (62), and 71 the shear viscosity of the gas, estimated within the Boltzmann approximation. Collecting both contributions ζ_B and $\zeta_{\mathrm{dyn}}(t \to \infty)$, we then recover, as expected, the Stokes law for the friction coefficient

$$\zeta_f = \int_0^\infty dt\, \zeta(t) = \frac{2\pi\eta\Sigma}{M} \tag{100}$$

In this limit, the diffusion coefficient acquires its Stokes–Einstein form

$$D \equiv \frac{k_{\mathrm{B}} T}{M\zeta_f} = \frac{k_{\mathrm{B}} T}{2\pi\eta\Sigma} \tag{101}$$

8. CONCLUSIONS

In this paper, we considered the Brownian motion of a single heavy particle moving in a bath of light particles. Our study started with a microscopic description of the system, in terms of the coupled dynamical evolution equations for the distribution functions of the B particle and of the host fluid. The latter was assumed to evolve according to an extended Boltzmann equation which correctly describes the effects of collisions between the gas particles and the suspended sphere.

Our aim was to derive a reduced equation for the B particle by eliminating the gas degrees of freedom in the limit of small mass ratio $\varepsilon = (m/M)^{1/2}$. However, even in the $\varepsilon \to 0$ limit, we kept the mass densities of both components to be of the same order of magnitude. The multiple-time-scale analysis has been used to construct a uniform expansion in ε. We derived in this way a new reduced equation (65) governing the evolution of the velocity distribution of particle B. This equation turned out to be nonlocal in time and in velocity space. The corresponding memory terms result from building up of the friction force by the reaction of the suspending gas to the motion of B. However, in spite of its non-Markovian character, this reduced equation allowed us to compute the velocity autocorrelation function of the B particle (87). Moreover, we recovered explicitly the Stokes–Einstein law relating the (spatial) diffusion coefficient to the friction coefficient (89). Finally, the derivation yielded a microscopic expression for the friction coefficient, reducing to the Stokes law in the hydrodynamic limit, where the mean-free path of the gas is small compared to the radius of the suspended sphere (100).

Our work is not the first attempt to obtain the reduced equation for the distribution function of the B particle from a microscopic point of view.[4-10] However, all previous approaches derive a (local) Fokker–Planck equation governing the dynamical evolution of the B particle by taking the $m/M \to 0$ limit with all other parameters kept constant. Such a limit implies an asymptotically vanishing mass density ratio, while in the present work, we maintained this ratio at a constant value.

This crucial difference is the source of the non-Markovian character of the reduced equation for the velocity distribution of the B particle, Eq. (65). Indeed, as discussed in the introduction, when $m/M \to 0$, while the mass density ratio is kept constant, the velocity of particle B is expected to decay on a hydrodynamic time scale of the suspending fluid. Therefore, the reaction of the fluid to the motion of the Brownian particle takes a finite time to occur (compared to the relaxation time of the velocity of particle B), and the friction force due to the fluid is accordingly displaced in time and velocity space [see Eq. (66)]. As shown in Sect. 5, this non-markovian effect leads to a "slow" thermalization, algebraic in time, in contrast to the exponentially decay predicted by the Langevin equation. This nonexponential behavior is in complete agreement with the predictions of the fluctuating hydrodynamics approaches,[15, 10] which lead to the generalized Langevin equation, Eq. (3). Numerical simulations of colloidal suspensions, based on fluctuating Lattice Bolztmann techniques,[25] confirm the presence of the so-called "long-time tails" in the velocity autocorrelation function of the Brownian particles. Moreover these algebraic decays has been observed experimentally in the "short-time" dynamics (i.e., on the scale of the relaxation of the velocity of the Brownian particles) of colloidal suspensions, the most recent experiments using diffusing wave spectroscopy (DWS) techniques.[26] [4]

The results derived here suggest some directions for future work. First, the extended Boltzmann equation was taken as a starting point, which can

[4] Note that the colloidal *suspensions* exactly meet the two crucial prescriptions of the studies limit, namely large mass ratio M/m between colloids and fluid particles, together with a mass density of colloids of the same order as the fluid mass density (prescribed to avoid sedimention of the suspended particles).

be expected to be valid in the Grad limit. This conjecture could perhaps be verified explicitly by starting from the BBGKY hierarchy for hard spheres,[19] and applying directly to it the present limit defined in (7). In particular, we could in principle compute the ε expansion of correlations induced in the bath by the presence of particle B. It would be then possible to check whether the results obtained in this work become indeed *exact* in the limit (7).

On the other hand, it would be interesting to generalize the analysis to an arbitrary number of suspended particles, in order to obtain the generalized reduced equation for the N-particle suspension. In this case, we expect the hydrodynamic interactions between the different suspended particles to be nonlocal in time, too.

The reduced equation we obtained here characterizes the relaxation of the velocity distribution of B, while the spatial distribution evolves on a much longer time scale. Because of this time scale separation, one then expects the spatial distribution to relax according to the local Smoluchowski equation. In principle, one could recover this equation from the same microscopic approach by pursuing the expansion up to the corresponding time scale, $t \sim \Sigma^2/D$, which corresponds to the τ_4 variable. Because of the complexity of the analysis, we have performed up to now the corresponding calculations only for the case of an ideal (collisionless) suspending gas. Even in this case, in spite of the absence of hydrodynamic modes in the bath, memory effects are still present and they show once two or more Brownian particles are present. Work along these lines is being carried out.

APPENDIX

In this appendix, we give the full expressions for the propagators $\mathscr{L}_B$ and $\mathscr{L}_f$. With proper dimensions, $\mathscr{L}_B$ can be written

$$\mathscr{L}_B(t) = \frac{\delta}{\delta \mathbf{V}} \cdot \left(\zeta_B \left(\mathbf{V} + \frac{k_B T}{M} \frac{\partial}{\partial \mathbf{V}} \right) - \bar{\mathscr{F}}_+(B; t) \right) \tag{A.1}$$

where the mean force $\bar{\mathscr{F}}_+(B; t)$ is defined self-consistently by Eq. (66). On the other hand, the fluid propagator $\mathscr{L}_f$ reads

$$\mathscr{L}_f(1) = \mathbf{v}_1 \cdot \frac{\partial}{\partial \mathbf{r}_1} - \bar{T}_-^{(0)}(B, 1) - \Lambda_B(1) \tag{A.2}$$

where $\Lambda_B(1)$ is the linearized Boltzmann operator, and $\bar{T}_-^{(0)}(B, 1)$ characterizes the effect of collisions between and the gas and the B particle fixed at point $\mathbf{R}$. The expression for $\Lambda_B(1)$ is given by[19]

$$\Lambda_B(1)\,\Psi(1\,|\,B) = f^{\rm eq}(1\,|\,\mathbf{R}) \int d2 \int d\hat{\sigma}\,[\mathbf{v}_1 - \mathbf{v}_2)\cdot\hat{\sigma}]$$
$$\times\,\theta[(\mathbf{v}_1 - \mathbf{v}_2)\cdot\hat{\sigma}]\,\delta(\mathbf{r}_1 - \mathbf{r}_2)\,\phi(\mathbf{v}_2)$$
$$\times\,\phi(\mathbf{v}_2)[b_{\hat{\sigma}}(1,2) - 1]\left\{\left(\frac{\Psi(1\,|\,B)}{\phi(\mathbf{v}_1)}\right) + \left(\frac{\Psi(2\,|\,B)}{\phi(\mathbf{v}_2)}\right)\right\} \tag{A.3}$$

where $\Psi(1\,|\,B)$ is a function of the fluid variables $1 \equiv \{\mathbf{r}_1, \mathbf{v}_1\}$, and $\phi(\mathbf{v})$ is the Maxwell distribution

$$\phi(\mathbf{v}) = \left(\frac{m}{2\pi k_{\rm B} T}\right)^{3/2} \exp\left(-\frac{mv^2}{2k_{\rm B}T}\right) \tag{A.4}$$

The operator $b_{\hat{\sigma}}(1,2)$ acting on a function $\chi(\mathbf{v}_1, \mathbf{v}_2)$ replaces the velocities $(\mathbf{v}_1, \mathbf{v}_2)$ by their postcollisional values

$$[b_{\hat{\sigma}}(1,2)\,\chi](\mathbf{v}_1, \mathbf{v}_2) = \chi(\mathbf{v}_1 - [(\mathbf{v}_1 - \mathbf{v}_2)\cdot\hat{\sigma}]\,\hat{\sigma}, \mathbf{v}_2 + [(\mathbf{v}_1 - \mathbf{v}_2)\cdot\hat{\sigma}]\,\hat{\sigma}) \tag{A.5}$$

Finally, $\bar{T}^{(0)}_{-}(B, 1)$ can be written as

$$\bar{T}^{(0)}_{-}(B, 1) = \left(\frac{\Sigma}{2}\right)^2 \int d\hat{\sigma}\,[-\mathbf{v}_1\cdot\hat{\sigma}]\,\theta[-\mathbf{v}_1\cdot\hat{\sigma}]$$
$$\times\left\{\delta\left(\mathbf{R} - \frac{\Sigma}{2}\hat{\sigma} - \mathbf{r}_1\right) b^{(0)}_{\hat{\sigma}}(B, 1) - \delta\left(\mathbf{R} + \frac{\Sigma}{2}\hat{\sigma} - \mathbf{r}_1\right)\right\} \tag{A.6}$$

where $b^{(0)}_{\hat{\sigma}}(B, 1)$ changes the velocity $\mathbf{v}_1$ of the gas particle into $\mathbf{v}'_1 = \mathbf{v}_1 - 2[\mathbf{v}_1\cdot\hat{\sigma}]\,\hat{\sigma}$.

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