Journal of Statistical Physics, Vol. 87, Nos. 5/6, 1997

Long-Time Translational and Rotational Brownian Motion in Two Dimensions

B. Cichocki[1] **and B. U. Felderhof**[2]

Received January 30, 1996; final September 20, 1996

The long-time translational and rotational motion of a Brownian particle in two dimensions is studied on the basis of the fluctuation-dissipation theorem and linearized hydrodynamics. The long-time motion follows from the low frequency behavior of the mobility matrix. The coefficient of the long-time tail for the translational motion turns out to be independent of shape and size of the body, in agreement with mode-coupling theory. For rotational Brownian motion the coefficient of the long-time tail is found to depend on the shape of the body. This result is in conflict with a recent prediction from mode-coupling theory, and indicates that the mode-coupling calculation should be revised.

KEY WORDS: Brownian motion; long-time tails.

1. INTRODUCTION

The long-time tail of the translational velocity autocorrelation function was first discovered for a tagged particle in a hard sphere fluid in a computer simulation by Alder and Wainwright.(1) The long-time behavior was understood on the basis of an extension of kinetic theory(2) and of mode-coupling theory.(3) It was soon realized that for a Brownian particle the same behavior follows from the fluctuation-dissipation theorem and linearized hydrodynamics.(4-8) On this basis we have shown(9) that the coefficient of the long-time tail does not depend on shape or size of the Brownian particle, in agreement with mode-coupling theory. Experimental evidence(10-13) and computer simulations(14,15) are in accord with the theoretical predictions.

This article is dedicated in friendship to Prof. Matthieu Ernst on the occasion of his 60th birthday.

[1] Institute of Theoretical Physics, Warsaw University, Hoza 69, 00-618 Warsaw, Poland.
[2] Institut für Theoretische Physik A, R.W.T.H. Aachen, 52056 Aachen, Germany.

0022-4715/97/0600-0989$12.50/0

The situation is less simple for the long-time tail in rotational Brownian motion. Ladd[15] has calculated the angular velocity correlation function for a sphere in computer simulation and found good agreement with the theoretical prediction from mode-coupling theory and from the fluctuation-dissipation theorem. Masters and Keyes[16] and Garisto and Kapral[17] argued on the basis of mode-coupling theory that the coefficient of the long-time tail does not depend on the shape of the particle. A hydrodynamic argument by Hocquart and Hinch[18] for a centrally symmetric body showed that the coefficient does depend on shape. We have provided a rigorous proof,[19] based on linear hydrodynamics and the fluctuation-dissipation theorem, that for an arbitrary rigid body the coefficient of the long-time tail depends on shape. The coefficient we found reduces to that of Hocquart and Hinch for a centrally symmetric particle. A recent computer simulation for a finite cylinder immersed in a lattice Boltzmann fluid[20] shows good agreement with the theoretical prediction for an ellipsoid of approximately the same shape.[21]

The computer simulation was also performed for a rectangle immersed in a two-dimensional lattice Boltzmann fluid.[20] In the following we calculate the coefficient of the long-time tail of the angular velocity correlation function from the fluctuation-dissipation theorem and two-dimensional hydrodynamics. We find that the coefficient depends on shape in a manner similar to that in three dimensions. This is in conflict with the prediction of Lowe *et al.*[20] based on mode-coupling theory. These authors found a coefficient that does not depend on shape. We calculate the coefficient explicitly for an ellipse and find good agreement with the computer simulation result for a rectangle if the axes of the ellipse are taken equal to the sides of the rectangle.

No doubt for a sufficiently large Brownian particle the fluctuation-dissipation theorem and linearized hydrodynamics can be trusted to yield the correct result. The disagreement with mode-coupling theory in both two and three dimensions indicates that the mode-coupling calculation should be revised.

2. EQUATIONS OF MOTION

We consider a solid cylinder of infinite length, immersed in a viscous incompressible fluid of shear viscosity η and mass density ρ. The fluid extends to infinity and obeys stick boundary conditions at the surface of the cylinder. The z axis of the Cartesian coordinate system is taken along the axis of the cylinder. The cross section of the cylinder in the xy plane is simply connected, but otherwise arbitrary. We denote the cross-sectional area of the cylinder at rest by V_0 and its circumference by S_0. We consider only motions transverse to the axis and fluid flow independent of the z coordinate. Thus we deal with a two-dimensional flow problem.

For small-amplitude motion the flow velocity $\mathbf{v}(\mathbf{r}, t)$ and pressure $p(\mathbf{r}, t)$ are governed by the linearized Navier–Stokes equations

$$\rho \frac{\partial \mathbf{v}}{\partial t} = \eta \nabla^2 \mathbf{v} - \nabla p, \qquad \nabla \cdot \mathbf{v} = 0 \tag{2.1}$$

The pressure field p is determined from the condition of incompressibility. The rigid body motion is described by the velocity field

$$\mathbf{w}(\mathbf{r}, t) = \mathbf{U}(t) + \mathbf{\Omega}(t) \times \mathbf{r}, \qquad \mathbf{r} \in V_0 \tag{2.2}$$

Here and in the following all vectors have only x and y components, except for the angular velocity $\mathbf{\Omega}(t)$ and the torque, which are directed along the z axis. We choose the origin at the center of mass of the cross section. The stick boundary condition requires

$$\mathbf{v}(\mathbf{s}, t) = \mathbf{w}(\mathbf{s}, t), \qquad \mathbf{s} \in S_0 \tag{2.3}$$

On account of our assumption of small amplitude motion the boundary condition may be applied at the undisplaced surface.

Fourier analyzing in time we find from Eq. (2.1) that the fluid equations of motion for the Fourier components with time factor $\exp(-i\omega t)$ are

$$-i\omega\rho \mathbf{v}_\omega = \eta \nabla^2 \mathbf{v}_\omega - \nabla p_\omega, \qquad \nabla \cdot \mathbf{v}_\omega = 0 \tag{2.4}$$

It is convenient to read this as an equation in two dimensions. The equation holds everywhere in the whole outer space $\bar{V} = R_2 - V_0$. It is convenient to extend the equations to be valid in the whole space R_2, with the boundary condition accounted for by a force density $\mathbf{F}_\omega(\mathbf{r})$ acting on the imagined fluid for $\mathbf{r} \in V_0$. This gives rise to the inhomogeneous equations[22,23]

$$\eta[\nabla^2 \mathbf{v}_\omega - \alpha^2 \mathbf{v}_\omega] - \nabla p_\omega = -\mathbf{F}_\omega(\mathbf{r}), \qquad \nabla \cdot \mathbf{v}_\omega = 0 \tag{2.5}$$

where $\alpha = (-i\omega\rho/\eta)^{1/2}$ with $\operatorname{Re} \alpha > 0$. The induced force density $\mathbf{F}_\omega(\mathbf{r})$ is made unique by the requirement that for $\mathbf{r} \in V_0$ the flow velocity and pressure are given by[22,24]

$$\mathbf{v}_\omega(\mathbf{r}) = \mathbf{w}_\omega(\mathbf{r}), \qquad p_\omega(\mathbf{r}) = -\eta\alpha^2 \mathbf{U}_\omega \cdot \mathbf{r}, \qquad \mathbf{r} \in V_0 \tag{2.6}$$

Then

$$\mathbf{F}_\omega(\mathbf{r}) = \mathbf{f}_\omega(\mathbf{s})\, \delta(\mathbf{r} - \mathbf{s}) + \eta\alpha^2(\mathbf{\Omega}_\omega \times \mathbf{r})\, \Theta_0(\mathbf{r}) \tag{2.7}$$

where $\mathbf{f}_\omega(\mathbf{s})$ is the surface force density, and $\Theta_0(\mathbf{r})$ is the characteristic function for the cross section V_0. The surface force density follows from the requirement that the flow $(\mathbf{v}_\omega, p_\omega)$ in the outer space $\bar{V}$ be identical with that of the original problem.

The solution of Eq. (2.5) reads in integral form

$$\mathbf{v}_\omega(\mathbf{r}) = \mathbf{v}_{0\omega}(\mathbf{r}) + \int \mathbf{G}(\mathbf{r}-\mathbf{r}'; \omega) \cdot \mathbf{F}_\omega(\mathbf{r}')\, d\mathbf{r}' \tag{2.8}$$

$$p_\omega(\mathbf{r}) = p_{0\omega}(\mathbf{r}) + \frac{1}{2\pi} \int \mathbf{Q}(\mathbf{r}-\mathbf{r}') \cdot \mathbf{F}_\omega(\mathbf{r}')\, d\mathbf{r}' \tag{2.9}$$

where $(\mathbf{v}_{0\omega}, p_{0\omega})$ represents an incident flow in the absence of the cylinder, restricted by the conditions that $v_{0\omega z} = 0$, and that $\mathbf{v}_{0\omega}$ and $p_{0\omega}$ depend only on the x, y coordinates. The incident flow $(\mathbf{v}_{0\omega}, p_{0\omega})$ satisfies the homogeneous equations (2.4) everywhere in space. The Green function is given explicitly by[25]

$$\begin{aligned} G(\mathbf{r}; \omega) &= \frac{1}{2\pi\eta} \{\mathbf{1} G(\mathbf{r}; \omega) + \alpha^{-2} \nabla\nabla[G_0(\mathbf{r}) - G(\mathbf{r}; \omega)]\} \\ G(\mathbf{r}; \omega) &= K_0(\alpha r), \qquad G_0(\mathbf{r}) = -\ln r \end{aligned} \tag{2.10}$$

where $K_0(\alpha r)$ is a modified Bessel function.[26] Note that Eq. (A.3) of ref. 25 has a sign error. The Green function for the pressure is

$$\mathbf{Q}(\mathbf{r}) = \frac{\mathbf{r}}{r^2} = -\nabla G_0(\mathbf{r}) \tag{2.11}$$

The peculiarity of hydrodynamics in two dimensions becomes apparent if we consider the low-frequency expansion of the Green function. The first few terms of the expansion are

$$\mathbf{G}(\mathbf{r}, \omega) = \mathbf{G}^{(l)}(\mathbf{r}) \ln \alpha + \mathbf{G}^{(0)}(\mathbf{r}) + \mathbf{G}^{(1)}(\mathbf{r})\, \alpha^2 \ln \alpha + O(\alpha^2) \tag{2.12}$$

where the first coefficient function is given by

$$\mathbf{G}^{(l)}(\mathbf{r}) = \frac{-\mathbf{1}}{4\pi\eta} \tag{2.13}$$

and the second by

$$\mathbf{G}^{(0)}(\mathbf{r}) = \frac{1}{4\pi\eta} \left[-\mathbf{1} \ln r + \hat{\mathbf{r}}\hat{\mathbf{r}} + \left(\ln 2 - \gamma - \frac{1}{2} \right) \mathbf{1} \right] \tag{2.14}$$

where γ is Euler's constant. The third coefficient function in Eq. (2.12) is

$$\mathbf{G}^{(1)}(\mathbf{r})=\frac{1}{32\pi\eta}\left[-3r^2\mathbf{1}+2\mathbf{rr}\right] \tag{2.15}$$

The first term in Eq. (2.12) shows that the Green function diverges in the zero-frequency limit. The corresponding flow pattern is spatially uniform. The second term in Eq. (2.12) differs from the steady-state Green function proposed by Oseen[27] by a constant multiplying the unit tensor. Such a term corresponds to a solution of the homogeneous steady-state equations.[28]

3. LOW-FREQUENCY MOBILITY MATRIX

The low-frequency behavior of the Green function, shown in Eq. (2.12), leads to corresponding behavior of the mobility matrix and the friction matrix. The low-frequency behavior of the mobility matrix is closely related to the long-time motion of the cylinder after an initial translational or rotational impulse, and to the long-time behavior of the velocity autocorrelation function in the case of Brownian motion.

The 3×3 mobility matrix $\boldsymbol{\mu}(\omega)$ is defined from the relations

$$\begin{aligned}\mathbf{U}_\omega&=\boldsymbol{\mu}^{tt}(\omega)\cdot\mathscr{F}_\omega+\boldsymbol{\mu}^{tr}(\omega)\,\mathscr{T}_\omega\\ \Omega_\omega&=\boldsymbol{\mu}^{rt}(\omega)\cdot\mathscr{F}_\omega+\mu^{rr}(\omega)\,\mathscr{T}_\omega\end{aligned} \tag{3.1}$$

where $\mathscr{F}_\omega$ is the two-dimensional force exerted on the fluid,

$$\mathscr{F}_\omega=\int \boldsymbol{F}_\omega\,d\mathbf{r} \tag{3.2}$$

and $\mathscr{T}_\omega$ is the z component of the torque,

$$\mathscr{T}_\omega=\int\left[\mathbf{r}\times\mathbf{F}_\omega\right]_z d\mathbf{r} \tag{3.3}$$

It follows from a generalization of Lorentz' reciprocity theorem[29-31] that the mobility matrix is symmetric. Hence its inverse, the 3×3 friction matrix $\boldsymbol{\zeta}(\omega)$ defined by

$$\boldsymbol{\mu}(\omega)\,\boldsymbol{\zeta}(\omega)=\mathbf{I} \tag{3.4}$$

is also symmetric.

The first few terms in the low-frequency expansion of the mobility matrix can be found from a study of Eq. (2.8). We consider a situation where the force and torque exerted on the fluid take fixed values

$$\mathscr{F}^{(0)} = \int \mathbf{F}_{\omega}(\mathbf{r})\, d\mathbf{r}, \qquad \mathscr{T}^{(0)} = \int \mathbf{r} \times \mathbf{F}_{\omega}(\mathbf{r})\, d\mathbf{r} \tag{3.5}$$

with $\mathscr{F}^{(0)}$ a vector in the xy plane, and $\mathscr{T}^{(0)}$ directed along the z axis. The cylinder is freely moving with translational velocity $\mathbf{U}_{\omega}$ and rotational velocity $\mathbf{\Omega}_{\omega}$. The incident flow $(\mathbf{v}_{0\omega}, p_{0\omega})$ is identically zero. We write Eq. (2.8) in the abbreviated form

$$\mathbf{v}_{\omega} = \mathsf{G}(\omega)\, \mathbf{F}_{\omega} \tag{3.6}$$

The stick boundary condition implies

$$\mathbf{w}_{\omega}|_{S_0} = [\mathsf{G}(\omega)\, \mathbf{F}_{\omega}]_{S_0} \tag{3.7}$$

Corresponding to the expansion in Eq. (2.12), we write the first few terms of the expansion of the solid-body motion $\mathbf{w}_{\omega}(\mathbf{r})$ as

$$\mathbf{w}_{\omega} = \mathbf{w}^{(l)} \ln \alpha + \mathbf{w}^{(0)} + \mathbf{w}^{(1)} \alpha^2 \ln \alpha + \mathbf{w}^{(2)} \alpha^2 + O(\alpha^4 \ln \alpha) \tag{3.8}$$

and of the force density $\mathbf{F}_{\omega}(\mathbf{r})$ as

$$\mathbf{F}_{\omega} = \mathbf{F}^{(0)} + \mathbf{F}^{(1)} \alpha^2 \ln \alpha + \mathbf{F}^{(2)} \alpha^2 + O(\alpha^4 \ln \alpha) \tag{3.9}$$

Substituting in Eq. (3.7) and comparing terms, we find from the terms proportional to $\ln \alpha$

$$\mathbf{w}^{(l)}|_{S_0} = [\mathsf{G}^{(l)} \mathbf{F}^0]_{S_0} \tag{3.10}$$

From Eqs. (2.2) and (2.13) we find

$$\mathbf{U}^{(l)} = -\frac{1}{4\pi\eta} \mathscr{F}^{(0)}, \qquad \mathbf{\Omega}^{(l)} = 0 \tag{3.11}$$

This shows that the lowest order term of the mobility matrix is universal and independent of shape or size of the cylinder.

From the terms of order unity in Eq. (3.7) we find

$$\mathbf{w}^{(0)}|_{S_0} = [\mathsf{G}^{(0)} \mathbf{F}^{(0)}]_{S_0} \tag{3.12}$$

This corresponds to a solution of the steady-state Stokes problem for prescribed force $\mathscr{F}^{(0)}$ and torque $\mathscr{T}^{(0)}$ exerted on the fluid. Since Eq. (2.14) provides a well-defined Green function, the solution of the problem is unique. The solution yields a definite translational velocity $\mathbf{U}^{(0)}$, rotational velocity $\boldsymbol{\Omega}^{(0)}$, flow pattern $(\mathbf{v}^{(0)}, p^{(0)})$, and induced force density $\mathbf{F}^{(0)}(\mathbf{r})$. Since $\mathbf{U}^{(0)}$ and $\boldsymbol{\Omega}^{(0)}$ are linear in $\mathscr{F}^{(0)}$ and $\mathscr{T}^{(0)}$, we can read off the corresponding term $\boldsymbol{\mu}^{(0)}$ in the expansion of the mobility matrix.

It follows from Eq. (3.11) that the lowest order contribution to the translational mobility matrix is given by

$$\boldsymbol{\mu}^{tt}(\omega) = -\frac{1}{4\pi\eta}\mathbf{1}\ln\alpha + O(1) \tag{3.13}$$

independent of shape or size of the body. The remaining elements $\boldsymbol{\mu}^{tr}(\omega)$, $\boldsymbol{\mu}^{rt}(\omega)$, and $\mu^{rr}(\omega)$ are of order unity at low frequency. Hence the tensor $\boldsymbol{\zeta}^{tt}(\omega)$ and the vectors $\boldsymbol{\zeta}^{tr}(\omega)$ and $\boldsymbol{\zeta}^{rt}(\omega)$ vanish as $1/\ln\alpha$ as $\omega\to 0$. The rotational element $\mu^{rr}(\omega)$ behaves at low frequency as

$$\mu^{rr}(\omega) = \mu^{rr}(0) + \mu^{rr(1)}\alpha^2\ln\alpha + O(\alpha^2) \tag{3.14}$$

The first term is the steady-state rotational mobility. We evaluate the coefficient $\mu^{rr(1)}$ in the next section. The form of the translational mobility matrix shown in Eq. (3.13) leads to universal long-time behavior of the translational velocity autocorrelation function. The coefficient $\mu^{rr(1)}$ depends on the shape of the body. As a consequence the coefficient of the long-time tail of the rotational velocity autocorrelation function is not universal.

4. ROTATIONAL MOBILITY

In this section we show that the coefficient $\mu^{rr(1)}$ in Eq. (3.14) can be expressed in terms of elements of the steady-state grand mobility matrix. From the terms of order $\alpha^2\ln\alpha$ in Eq. (3.7) we find

$$\mathbf{w}^{(1)}|_{S_0} = [\mathsf{G}^{(l)}\mathbf{F}^{(2)} + \mathsf{G}^{(0)}\mathbf{F}^{(1)} + \mathsf{G}^{(1)}\mathbf{F}^{(0)}]_{S_0} \tag{4.1}$$

The first term on the right vanishes on account of Eq. (2.13) and the fact that $\mathscr{F}^{(2)} = 0$, as follows from Eq. (3.5). The remaining terms in Eq. (4.1) can be related to the solution of a steady-state Stokes problem. To this order we need only the rotational–rotational part $\mu^{rr}(\omega)$ of the mobility matrix, so that we can put $\mathscr{F}^{(0)} = 0$. It follows from Eq. (2.15) that the kernel $\mathbf{G}^{(1)}(\mathbf{r}-\mathbf{r}')$ equals

$$\mathbf{G}^{(1)}(\mathbf{r}-\mathbf{r}') = \frac{1}{32\pi\eta}[-3(r^2 + r'^2 - 2\mathbf{r}\cdot\mathbf{r}')\,\mathbf{1} + 2(\mathbf{r}-\mathbf{r}')(\mathbf{r}-\mathbf{r}')] \tag{4.2}$$

For $\mathscr{F}^{(0)}=0$ the terms with $r^2\mathbf{1}$ and $\mathbf{rr}$ can be omitted from this expression. The remaining terms in the velocity field $\mathsf{G}^{(1)}\mathbf{F}^{(0)}$ are constant or linear in $\mathbf{r}$. Hence this velocity field satisfies the homogeneous Stokes equations with zero pressure. We recall that the first term on the right in Eq. (4.1) is zero. For $\mathscr{F}^{(0)}=0$ the remaining terms can be taken to be the limiting values at the circumference S_0 of a steady-state Stokes velocity field $\mathbf{v}^{(1)}(\mathbf{r})$ that satisfies the equation

$$\mathbf{v}^{(1)}=\mathsf{G}^{(0)}\mathbf{F}^{(1)}+\mathsf{G}^{(1)}\mathbf{F}^{(0)} \tag{4.3}$$

with the conditions $\mathscr{F}^{(1)}=0$, $\mathscr{T}^{(1)}=0$. As remarked following Eq. (4.2), for $\mathscr{F}^{(0)}=0$ the flow field

$$\mathbf{v}_0^{(1)}(\mathbf{r})=\int \mathsf{G}^{(1)}(\mathbf{r}-\mathbf{r}')\cdot\mathbf{F}^{(0)}(\mathbf{r}')\,d\mathbf{r}' \tag{4.4}$$

satisfies the homogeneous Stokes equations at zero pressure, and is a superposition of a uniform and a linear flow

$$\mathbf{v}_0^{(1)}=\mathbf{v}_{0U}^{(1)}+\mathbf{v}_{0L}^{(1)} \tag{4.5}$$

The uniform flow is

$$\mathbf{v}_{0U}^{(1)}=\frac{1}{32\pi\eta}\int [-3r'^2\mathbf{1}+2\mathbf{r}'\mathbf{r}']\cdot\mathbf{F}^{(0)}(\mathbf{r}')\,d\mathbf{r}' \tag{4.6}$$

This corresponds to a translational velocity $\mathbf{U}_{0U}^{(1)}=\mathbf{v}_{0U}^{(1)}$ and a vanishing contribution to the force density $\mathbf{F}^{(1)}(\mathbf{r})$. The linear flow is

$$\mathbf{v}_{0L}^{(1)}(\mathbf{r})=\frac{1}{16\pi\eta}\int [(3\mathbf{r}\cdot\mathbf{r}')\,\mathbf{1}-\mathbf{r}\mathbf{r}'-\mathbf{r}'\mathbf{r}]\cdot\mathbf{F}^{(0)}(\mathbf{r}')\,d\mathbf{r}' \tag{4.7}$$

This can be expressed as

$$\mathbf{v}_{0L}^{(1)}(\mathbf{r})=\mathbf{\Omega}_L\times\mathbf{r}+\mathbf{g}_L\cdot\mathbf{r} \tag{4.8}$$

with rotational velocity $\mathbf{\Omega}_L=\Omega_L\,\mathbf{e}_z$ given by

$$\Omega_L=\frac{1}{8\pi\eta}\mathscr{T}^{(0)} \tag{4.9}$$

and traceless symmetric tensor $\mathbf{g}_L$ given by

$$\mathbf{g}_L = \frac{1}{8\pi\eta} \overline{\mathsf{F}}^{(0)} \tag{4.10}$$

where $\overline{\mathsf{F}}^{(0)}$ is the traceless symmetric force dipole moment

$$\overline{\mathsf{F}}^{(0)} = \tfrac{1}{2} \int [\mathbf{r}\mathbf{F}^{(0)} + \mathbf{F}^{(0)}\mathbf{r} - (\mathbf{r} \cdot \mathbf{F}^{(0)})\, \mathbf{1}]\, d\mathbf{r} \tag{4.11}$$

From Eq. (4.3) one finds for the resulting rotational velocity

$$\Omega^{(1)} = \Omega_L + \boldsymbol{\mu}^{rd}(0) : \mathbf{g}_L \tag{4.12}$$

where the tensor $\boldsymbol{\mu}^{rd}(0)$ is part of the steady-state grand mobility matrix.[32] The symmetric force dipole moment is related to the torque $\mathscr{T}^{(0)} = \mathscr{T}^{(0)}\mathbf{e}_z$ by

$$\overline{\mathsf{F}}^{(0)} = -\boldsymbol{\mu}^{dr}(0) \cdot \mathscr{T}^{(0)} \tag{4.13}$$

From Eqs. (4.9)–(4.13) we therefore find for the contribution of order $\alpha^2 \ln \alpha$ to the rotational mobility

$$\mu^{rr(1)} = \frac{1}{8\pi\eta} [1 - \boldsymbol{\mu}^{rd}(0) : \boldsymbol{\mu}^{dr}(0)] \tag{4.14}$$

This relation is similar to a corresponding result in three dimensions.[19] The coefficient depends on the shape of the body. From symmetry it follows that the scalar product in Eq. (4.14) is negative. For a circular cylinder the product vanishes.

5. LONG-TIME TAILS

The low-frequency results embodied by Eqs. (3.13) and (3.14) can be used to derive the long-time tails of translational and rotational motion. We consider first deterministic motion in the absence of stochastic forces and torques. Thus we assume that the body is accelerated from rest by a sudden translational and rotational impulse. The applied force and torque per unit length of cylinder are given by

$$\mathbf{E}(t) = \mathbf{S}_T \delta(t), \qquad \mathbf{N}(t) = S_R \mathbf{e}_z \delta(t) \tag{5.1}$$

where the vector $\mathbf{S}_T$ has vanishing z component. The subsequent motion of the body follows from

$$\begin{pmatrix} \mathbf{U}_\omega \\ \Omega_\omega \end{pmatrix} = \frac{1}{2\pi} \mathcal{Y}(\omega) \begin{pmatrix} \mathbf{S}_T \\ S_R \end{pmatrix} \tag{5.2}$$

with the 3×3 admittance matrix $\mathcal{Y}(\omega)$ given by

$$\mathcal{Y}(\omega) = [-i\omega \mathbf{m} + \zeta(\omega)]^{-1} \tag{5.3}$$

The 3×3 effective mass matrix $\mathbf{m}$ follows from the behavior at high frequency. For the low-frequency behavior under consideration the effective mass matrix is not relevant. From Eq. (3.13) we find at low frequency

$$\mathbf{U}_\omega = -\frac{\ln \alpha}{8\pi^2 \eta} \mathbf{S}_T + O(1) \tag{5.4}$$

This corresponds to a slow long-time decay[33]

$$\mathbf{U}(\mathrm{t}) \approx \frac{1}{8\pi\eta t} \mathbf{S}_T, \qquad \text{as} \quad t \to \infty \tag{5.5}$$

independent of shape and size of the body.

Similarly we find from Eq. (3.14) at low frequency

$$\Omega_\omega = \frac{1}{2\pi} [\mu^{rr(0)} + \mu^{rr(1)} \alpha^2 \ln \alpha] \, S_R + O(\alpha^2) \tag{5.6}$$

This corresponds to the long-time decay[33]

$$\Omega_\omega \approx \mu^{rr(1)} \frac{\rho}{2\eta t^2} S_R, \qquad \text{as} \quad t \to \infty \tag{5.7}$$

In discussing Brownian motion we take a strictly two-dimensional point of view, corresponding to computer simulations.[14,20] Thus the mean-square translational velocity in a thermal equilibrium ensemble is

$$\langle \mathbf{U}^2 \rangle = \frac{k_\mathrm{B} T}{m_p} \tag{5.8}$$

where the angle brackets indicate an equilibrium average in the thermodynamic limit, and m_p is the mass of the two-dimensional body. The translational velocity autocorrelation function characterizing the Brownian motion is defined as

$$\mathsf{C}_{UU}(t) = \langle \mathbf{U}(t)\, \mathbf{U}(0) \rangle \tag{5.9}$$

More generally one considers the 3×3 time-correlation matrix

$$\mathsf{C}(t) = \begin{pmatrix} \mathsf{C}_{UU}(t) & \mathsf{C}_{U\Omega}(t) \\ \mathsf{C}_{\Omega U}(t) & \mathsf{C}_{\Omega\Omega}(t) \end{pmatrix} \tag{5.10}$$

This has the one-sided Fourier transform

$$\hat{\mathsf{C}}(\omega) = \int_0^\infty e^{i\omega t} \mathsf{C}(t)\, dt \tag{5.11}$$

According to the fluctuation-dissipation theorem,[6,34,35] it is given by

$$\hat{\mathsf{C}}(\omega) = k_B T \mathscr{Y}(\omega) \tag{5.12}$$

From Eqs. (3.13) and (5.3) we therefore find for the long-time behavior of the translational velocity autocorrelation function

$$\mathsf{C}_{UU}(t) \approx \frac{k_B T}{8\pi\eta t} \mathbf{1} \qquad \text{as} \quad t \to \infty \tag{5.13}$$

independent of shape, size, or mass of the body. This result agrees with the prediction of mode-coupling theory.[3]

Similarly we find for the long-time tail of the angular velocity autocorrelation function

$$\mathsf{C}_{\Omega\Omega}(t) \approx k_B T \mu^{rr(1)} \frac{\rho}{2\eta t^2} \qquad \text{as} \quad t \to \infty \tag{5.14}$$

with coefficient $\mu^{rr(1)}$ given by Eq. (4.14). A mode-coupling calculation by Lowe *et al.*[20] yielded a coefficient independent of the shape of the body. The two results agree only for a circular disk. We have suggested that the mode-coupling calculation should be revised.[21] The coefficient $\mu^{rr(1)}$ is independent of the orientation of the body. Hence an additional configurational averaging has no effect.

6. DUMBBELL AND ELLIPSE

We again take a strictly two-dimensional point of view. For a circular disk the tensors $\boldsymbol{\mu}^{rd}(0)$ and $\boldsymbol{\mu}^{dr}(0)$ vanish identically. In order to get an estimate of the importance of the second term in brackets in Eq. (4.14) for a shape different from a circle, we consider in particular a dumbbell with neglect of hydrodynamic interactions, and an ellipse.

For a dumbbell with points of friction $\mathbf{R}_1$ and $\mathbf{R}_2$ in the xy plane one finds the two tensors[19]

$$\begin{aligned} \mu^{rd}_{z\alpha\beta}(0) &= -\tfrac{1}{2}[\varepsilon_{z\alpha\mu}u_\mu u_\beta + \varepsilon_{z\beta\mu}u_\mu u_\alpha] \\ \mu^{dr}_{\alpha\beta z}(0) &= -\tfrac{1}{2}[u_\alpha u_\mu \varepsilon_{\mu\beta z} + u_\beta u_\mu \varepsilon_{\mu\alpha z}] \end{aligned} \tag{6.1}$$

where $\mathbf{u}$ is the unit vector along the bond from $\mathbf{R}_2$ to $\mathbf{R}_1$. Hence the scalar product is

$$\mu^{rd}(0):\mu^{dr}(0) = -1/2 \tag{6.2}$$

Next we consider an ellipse with semiaxes a and b. We choose a coordinate system with axes x, y along the axes of the ellipse. A solution of the steady-state flow problem for a rotating elliptic cylinder in terms of elliptic cylinder coordinates (ref. 30, p. 495) has been provided by Edwardes.[36] Alternatively one may use Cartesian coordinates and solve the problem in terms of functions of the elliptical coordinate $\lambda(x, y)$ defined as the positive root of

$$\frac{x^2}{a^2+\lambda} + \frac{y^2}{b^2+\lambda} = 1 \tag{6.3}$$

following the method demonstrated by Oseen (ref. 27, p. 136) for the ellipsoid. For the components of the tensor $\boldsymbol{\mu}^{rd}(0)$ we find

$$\begin{aligned} &\mu^{rd}_{zxx}(0) = \mu^{rd}_{zyy}(0) = 0 \\ &\mu^{rd}_{zxy}(0) = \mu^{rd}_{zyx}(0) = \frac{1}{2}\frac{a^2-b^2}{a^2+b^2} \end{aligned} \tag{6.4}$$

The tensor $\boldsymbol{\mu}^{dr}(0)$ can be found from the symmetry relation

$$\mu^{rd}_{\gamma\alpha\beta}(0) = -\mu^{dr}_{\alpha\beta\gamma}(0) \tag{6.5}$$

which follows from Lorentz' reciprocity theorem.[29,30] The relation can be verified from the explicit solution. The scalar tensor product is therefore given by

$$\boldsymbol{\mu}^{rd}(0):\boldsymbol{\mu}^{dr}(0) = -\frac{1}{2}\left(\frac{a^2-b^2}{a^2+b^2}\right)^2 \tag{6.6}$$

For a long ellipse this result tends to that in Eq. (6.2).

For an ellipse with semiaxes $a=11$, $b=3$ one finds for the coefficient in square brackets in Eq. (4.14) the value 1.371. This agrees well with the result of computer simulation for a rectangle of length 11 and width 3 immersed in a two-dimensional lattice Boltzmann fluid.[20] Lowe *et al.*[20] found for the coefficient of the long-time tail of the rotational velocity autocorrelation function 1.31 ± 0.01, in the same units.

In the computer simulation the center of mass of the body was held fixed. For a centrally symmetric body the elements $\boldsymbol{\mu}^{tr}(0)$ and $\boldsymbol{\mu}^{rt}(0)$ of the grand mobility matrix vanish identically. Hence for such a body the predictions of the coefficient of the long-time tail according to Eqs. (4.14) and (5.14) are the same, whether or not the center of mass is held fixed.

In conclusion we note that for the circular disk, or equivalently the circular cylinder in three dimensions, the complete admittance matrix $\mathcal{Y}(\omega)$ can be calculated. By symmetry there is no coupling between translation and rotation, and it suffices to consider the scalar translational admittance $\mathcal{Y}_t(\omega)$ and the rotational admittance $\mathcal{Y}_r(\omega)$. The translational admittance for a circle of radius a is

$$\mathcal{Y}_t(\omega) = [-i\omega(m_p + m_f) + \zeta_t(\omega)]^{-1} \tag{6.7}$$

where $m_f = \pi a^2 \rho$ is the added mass, and the friction coefficient $\zeta_t(\omega)$ is given by[25]

$$\zeta_t(\omega) = 4\pi\eta \frac{\alpha a K_1(\alpha a)}{K_0(\alpha a)} \tag{6.8}$$

where $K_0(z)$ and $K_1(z)$ are modified Bessel functions.[26] The rotational admittance is

$$\mathcal{Y}_r(\omega) = [-i\omega I_p + \zeta_r(\omega)]^{-1} \tag{6.9}$$

where I_p is the moment of inertia, and the rotational friction coefficient is given by

$$\zeta_r(\omega) = 2\pi\eta a^2 \left[1 + \frac{1}{2} \frac{\alpha a K_0(\alpha a)}{K_1(\alpha a)}\right] \tag{6.10}$$

The fluctuation-dissipation theorem, Eq. (5.12), provides a prediction for the complete time dependence of the translational and rotational velocity autocorrelation functions. It is easy to check that the coefficients of the long-time tails agree with the general theorems (5.13) and (5.14). For an ellipsoid of revolution the entire angular velocity correlation function has been calculated by Hocquart.[37] No doubt a similar solution could be found for the ellipse.

7. DISCUSSION

We have calculated the long-time behavior of the translational and rotational velocity autocorrelation function for a Brownian particle in two dimensions by use of the fluctuation-dissipation theorem and linearized hydrodynamics. Because of the wide separation of time scales of macroscopic and molecular motion there seems no reason to doubt the validity of the procedure. The situation is different for a particle of size comparable to the molecular size, and for a tagged particle in a one-component fluid. Mode-coupling theory shows that for a particle of finite size the kinematic viscosity in the coefficient of the long-time tail of the translational velocity autocorrelation function should be replaced by the sum of diffusion coefficient and kinematic viscosity. Since in two dimensions the steady-state diffusion coefficient is not well defined, because of the long-time tail in the velocity correlation function, a self-consistent mode-coupling theory has been proposed.[38] This theory suggests that the $1/t$ long-time tail is replaced by $1/t\sqrt{\ln t}$ behavior. Although faster than $1/t$ decay was observed in computer simulation,[14,39] the possibility of verifying the $1/t\sqrt{\ln t}$ behavior seemed questionable.[39,40] However, recently such super long-time decay has been found.[41]

Mode-coupling theory suggests that for a small particle the coefficient of the long-time tail of the rotational velocity autocorrelation function will also depend on size. The theory as presented[16,20] does not lead to the correct coefficient for a large particle, and hence cannot be trusted for a small one. The derivation of Hocquart and Hinch[18] for a large particle in three dimensions is close to the mode-coupling theory. The latter should be revised along these lines in both two and in three dimensions. The near quantitative agreement of our prediction for the coefficient of the rotational long-time tail with computer simulation for a particle of similar shape in both two and three dimensions gives confidence in the applicability of the procedure even on a fairly small length scale.

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