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Long-time rotational motion of a set of rigid bodies immersed in a viscous fluid

B.U. Felderhof

Institut für Theoretische Physik A, RWTH Aachen, Templergraben 55, 52056 Aachen, Germany

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Abstract

The long-time rotational motion of a set of rigid bodies immersed in a viscous fluid is studied on the basis of the linearized Navier–Stokes equations. After an initial twist applied to one of the bodies the rotational motion of each of them decays at long times with a $t^{-5/2}$ long-time tail. The coefficient of the tail depends on shape and configuration of the bodies. It is shown that it can be expressed in terms of zero frequency mobility tensors that in principle can be calculated from the steady-state Stokes equations.

Keywords: Brownian motion; Linear hydrodynamics; Long-time tail of rotational motion

1. Introduction

The $t^{-3/2}$ long-time tail of the translational velocity autocorrelation function for a tagged particle in a fluid was first discovered in a computer simulation by Alder and Wainwright [1] for a system of hard spheres. It was soon realized that for a Brownian particle the same behavior follows from the fluctuation–dissipation theorem and linearized hydrodynamics [2–6]. The coefficient of the long-time tail does not depend on shape or size of the Brownian particle if it is sufficiently large. Even for a collection of rigid bodies of arbitrary shape one finds a long-time tail with the same coefficient [7]. The coefficient depends only on shear viscosity and mass density of the fluid.

The situation is less simple for the $t^{-5/2}$ long-time tail of rotational Brownian motion. Garisto and Kapral [8] and Masters and Keyes [9] 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 [10] for a centrally symmetric body showed that the coefficient does depend on shape. Cichocki and Felderhof provided a rigorous proof [11], based on linear hydrodynamics and the fluctuation–dissipation theorem, that for an arbitrary rigid body the coefficient does depend on shape. The coefficient they found reduces to that of Hocquart and Hinch for a centrally symmetric body. Similar results were found in two dimensions [12]. It should be emphasized that in using the results of linearized hydrodynamics in the theory of Brownian motion one is limited to the timescale of velocity relaxation. On a longer timescale the body diffuses in space and in

E-mail address: ufelder@physik.rwth-aachen.de.

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orientational space, as described by a Smoluchowski equation. It has been argued by Masters [13] that the results of Garisto and Kapral [8] and Masters and Keyes [9] are true asymptotic results that include diffusion.

In the following we consider the rotational velocity correlation function for a set of rigid bodies of arbitrary shape. Like for a single body the coefficient of the long-time tail is given by an expression that involves zero frequency mobility tensors that in principle can be calculated from the steady-state Stokes equations. The coefficient depends on shape and configuration.

The rotational velocity autocorrelation function of interacting Brownian particles has been determined by computer simulation for a suspension of spheres [14]. It was conjectured [14–16] that the coefficient of the long-time tail can be found from the single-sphere result by replacement of the viscosity and mass density of the fluid by the effective viscosity and mass density of the suspension. The present work was prompted by a calculation by Hermanns [17], who showed that the conjecture is incorrect. He found that in the pair virial term there are correction terms to the single-sphere result beyond those accounted for in the effective medium conjecture. The general theorem derived here allows simplification of the calculation of the pair virial term [18].

2. Admittance matrix

We consider a set of N rigid bodies of arbitrary shape immersed in a viscous incompressible fluid of shear viscosity η, mass density ρ. The fluid is assumed to fill the whole outer space. Stick boundary conditions are assumed to hold at the surface of the bodies. We consider disturbance of a situation where both the bodies and the fluid are at rest. The center of mass of body j in the rest situation is denoted by $\boldsymbol{R}_j$. The rest situation is perturbed by small time-dependent forces $(\boldsymbol{E}_1(t),\dots,\boldsymbol{E}_N(t))$ and torques $(\boldsymbol{N}_1(t),\dots,\boldsymbol{N}_N(t))$ applied to the bodies. For small amplitude motion the flow velocity $\boldsymbol{v}(\boldsymbol{r},t)$ and $p(\boldsymbol{r},t)$ are governed by the linearized Navier–Stokes equations. The rigid motion of body j is given by the velocity field

$$\boldsymbol{w}_j(\boldsymbol{r},t) = \theta_j(\boldsymbol{r})[\boldsymbol{U}_j(t) + \boldsymbol{\Omega}_j(t)\times(\boldsymbol{r}-\boldsymbol{R}_j)], \tag{2.1}$$

where $\theta_j(\boldsymbol{r})$ is the characteristic function for the body. To linear order the change of configuration need not be considered. For the equations of motion and for notation we refer to earlier articles [7,11].

The linear relation between velocities and applied forces and torques has memory character. Fourier analyzing in time we find for the Fourier components

$$\boldsymbol{U}_{j\omega} = \sum_{k=1}^{N}[\mathcal{Y}^{tt}_{jk}(\omega)\cdot\boldsymbol{E}_{k\omega} + \mathcal{Y}^{tr}_{jk}(\omega)\cdot\boldsymbol{N}_{k\omega}],$$

$$\boldsymbol{\Omega}_{j\omega} = \sum_{k=1}^{N}[\mathcal{Y}^{rt}_{jk}(\omega)\cdot\boldsymbol{E}_{k\omega} + \mathcal{Y}^{rr}_{jk}(\omega)\cdot\boldsymbol{N}_{k\omega}], \quad (j=1,\dots,N), \tag{2.2}$$

with admittance tensors $\mathcal{Y}^{\kappa\lambda}_{jk}(\omega)$. The tensors can be combined into a $6N\times 6N$ admittance matrix

$$\mathcal{Y}(\omega) = [-\mathrm{i}\omega\boldsymbol{m} + \boldsymbol{\zeta}(\omega)]^{-1}, \tag{2.3}$$

where $\boldsymbol{m}$ is the $6N\times 6N$ effective mass matrix and $\boldsymbol{\zeta}(\omega)$ is the $6N\times 6N$ friction matrix. The mass matrix follows from the response at high frequency. The $6N\times 6N$ mobility matrix $\boldsymbol{\mu}(\omega)$ is defined as the inverse of the friction matrix.

We shall be concerned in particular with the rotational tensors $\mathcal{Y}^{rr}_{jk}(\omega)$. These tensors are related to the rotational velocity correlation functions

$$\boldsymbol{C}^{rr}_{jk}(t) = \langle\boldsymbol{\Omega}_j(t)\boldsymbol{\Omega}_k(0)\rangle, \tag{2.4}$$

characterizing Brownian motion, by the fluctuation–dissipation theorem [4]. The angled brackets denote a thermal average at temperature T. The correlation function has the one-sided Fourier transform

$$\hat{\boldsymbol{C}}^{rr}_{jk}(\omega) = \int_0^\infty \mathrm{e}^{\mathrm{i}\omega t}\boldsymbol{C}^{rr}_{jk}(t)\,\mathrm{d}t. \tag{2.5}$$

According to the fluctuation–dissipation theorem this is given by

$$\hat{C}^{rr}_{jk}(\omega) = k_B T \mathcal{Y}^{rr}_{jk}(\omega). \tag{2.6}$$

We are interested in particular in the long-time behavior of the correlation functions. This is determined by the low-frequency behavior of the admittance.

Formal expressions for the admittance tensors can be derived in terms of a multiple scattering expansion. In earlier work we have derived such expressions for the zero frequency case [19]. The same formal work is valid at any frequency. Thus the translational and rotational velocity of body j can be expressed as

$$\boldsymbol{U}_j = \boldsymbol{U}^{St}_j + (\boldsymbol{t}_j|\boldsymbol{v}^a_j), \quad \boldsymbol{\Omega}_j = \boldsymbol{\Omega}^{St}_j + (\boldsymbol{r}_j|\boldsymbol{v}^a_j), \tag{2.7}$$

where the first "Stokes" terms $\boldsymbol{U}^{St}_j$ and $\boldsymbol{\Omega}^{St}_j$ are the velocities the body would acquire if it were by itself in infinite fluid and acted upon with force $\boldsymbol{E}_{j\omega}$ and torque $\boldsymbol{N}_{j\omega}$, and the second terms take account of the presence of the other bodies with their forces and torques. We have used the same bra–ket notation as in Ref. [19], but presently all quantities depend on frequency. In earlier work [7] the bras $(\boldsymbol{t}_j|$ and $(\boldsymbol{r}_j|$ were denoted as $(\boldsymbol{C}^t_{j\omega}|$ and $(\boldsymbol{C}^r_{j\omega}|$, with the frequency-dependence made explicit. The ket $|\boldsymbol{v}^a_j)$ in Eq. (2.7) represents the flow velocity acting on body j. It is given formally by

$$\boldsymbol{v}^a_j = \sum_k \hat{\mathsf{V}}_{jk}[|\boldsymbol{t}_k)\boldsymbol{E}_k + |\boldsymbol{r}_k)\boldsymbol{N}_k], \tag{2.8}$$

where $\hat{\mathsf{V}}_{jk}$ is a linear operator given by

$$\hat{\mathsf{V}}_{jk} = \theta_j \mathsf{G}(1-\delta_{jk})\theta_k - \theta_j \mathsf{G} \sum_{\substack{l\neq j\\ m\neq k}} \hat{\mathsf{Z}}_{lm}\mathsf{G}\theta_k, \tag{2.9}$$

where G is the Green operator for infinite space, and $\hat{\mathsf{Z}}_{lm}$ is given by the multiple scattering expansion

$$\hat{\mathsf{Z}}_{lm}(1,\ldots,N) = \hat{\mathsf{Z}}_j\delta_{lm} - \hat{\mathsf{Z}}_l\mathsf{G}\hat{\mathsf{Z}}_m(1-\delta_{lm}) + \sum_{n\neq l,m} \hat{\mathsf{Z}}_l\mathsf{G}\hat{\mathsf{Z}}_n\mathsf{G}\hat{\mathsf{Z}}_m + \cdots. \tag{2.10}$$

Substituting into Eq. (2.7) we find the expressions for the admittance tensors

$$\mathcal{Y}^{tt}_{jk}(\omega) = \mathcal{Y}^{tt}_j\delta_{jk} + (\boldsymbol{t}_j|\hat{\mathsf{V}}|\boldsymbol{t}_k), \quad \mathcal{Y}^{tr}_{jk}(\omega) = \mathcal{Y}^{tr}_j\delta_{jk} + (\boldsymbol{t}_j|\hat{\mathsf{V}}|\boldsymbol{r}_k),$$

$$\mathcal{Y}^{rt}_{jk}(\omega) = \mathcal{Y}^{rt}_j\delta_{jk} + (\boldsymbol{r}_j|\hat{\mathsf{V}}|\boldsymbol{t}_k), \quad \mathcal{Y}^{rr}_{jk}(\omega) = \mathcal{Y}^{rr}_j\delta_{jk} + (\boldsymbol{r}_j|\hat{\mathsf{V}}|\boldsymbol{r}_k), \tag{2.11}$$

where the first term on the right characterizes the single-body response. The linear operator $\hat{\mathsf{V}}$ is defined by

$$\hat{\mathsf{V}} = \sum_{j,k} \hat{\mathsf{V}}_{jk}. \tag{2.12}$$

In the matrix elements in Eq. (2.11) only the term $\hat{\mathsf{V}}_{jk}$ contributes. The notation differs slightly from that in Ref. [19] in that we write particle labels systematically as subscripts. When only a single label is present the symbol refers to a single body in infinite fluid.

3. Expansion to first order

In order to study long-time motion we consider expansion of the matrices $\mathcal{Y}(\omega)$, $\boldsymbol{\zeta}(\omega)$, and $\boldsymbol{\mu}(\omega)$ in powers of the variable α, defined by

$$\alpha = (-\mathrm{i}\omega\rho/\eta)^{1/2}, \quad \Re\alpha > 0. \tag{3.1}$$

The expansion of the admittance matrix is expressed as

$$\mathcal{Y}(\omega) = \mathcal{Y}^{(0)} + \mathcal{Y}^{(1)}\alpha + \mathcal{Y}^{(2)}\alpha^2 + \mathcal{Y}^{(3)}\alpha^3 + O(\alpha^4). \tag{3.2}$$

In this section we consider first the expansion of the admittance $\mathcal{Y}^{tt}_{jk}(\omega)$, as given by Eq. (2.11), to first order in α. We recover the result found earlier [7]. The calculation sets the stage for the similar expansion of the rotational admittance in the next sections.

In the expansion of the operator $\hat{\mathsf{V}}$ in powers of α we need the expansion of the Green operator. The Green function tensor depends only on the relative distance vector $\boldsymbol{r}-\boldsymbol{r}'$. The first two terms of the expansion of the Green function are given by [11]

$$\boldsymbol{G}^{(0)}(\boldsymbol{r}) = \frac{1}{8\pi\eta}\frac{\mathbf{1}+\hat{\boldsymbol{r}}\hat{\boldsymbol{r}}}{r}, \quad \boldsymbol{G}^{(1)}(\boldsymbol{r}) = -\frac{1}{6\pi\eta}\mathbf{1}. \tag{3.3}$$

The second order term is given by

$$\boldsymbol{G}^{(2)}(\boldsymbol{r}) = \frac{r}{32\pi\eta}(3\mathbf{1}-\hat{\boldsymbol{r}}\hat{\boldsymbol{r}}). \tag{3.4}$$

The third order term is

$$\boldsymbol{G}^{(3)}(\boldsymbol{r}) = \frac{1}{60\pi\eta}(\boldsymbol{r}\boldsymbol{r}-2r^2\mathbf{1}). \tag{3.5}$$

As a consequence of the simple expression for the first-order term and the projection property of the first-order friction kernels we find

$$\hat{\mathsf{Z}}_j^{(1)} = -\hat{\mathsf{Z}}_j^{(0)}\mathsf{G}^{(1)}\hat{\mathsf{Z}}_j^{(0)} = 0, \tag{3.6}$$

as well as

$$\hat{\mathsf{V}}_{jk}^{(1)} = \frac{-1}{6\pi\eta}(1-\delta_{jk})|\boldsymbol{\tau}_j)\cdot(\boldsymbol{\tau}_k|. \tag{3.7}$$

The bras $(\boldsymbol{\tau}_j|$ and $(\boldsymbol{\rho}_j|$ are given by simple expressions [19], and are independent of frequency. The bras $(\boldsymbol{t}_j|$ and $(\boldsymbol{r}_j|$ in Eq. (2.7) are defined by

$$(\boldsymbol{t}_j| = (\gamma_j^t|\mathsf{Z}_j, \quad (\boldsymbol{r}_j| = (\gamma_j^r|\mathsf{Z}_j, \tag{3.8}$$

with frequency-dependent single-body friction kernel Z_j and bra-tensors

$$(\gamma_j^t| = \mathscr{Y}_j^{tt}(\boldsymbol{\tau}_j| + \mathscr{Y}_j^{tr}(\boldsymbol{\rho}_j|,$$

$$(\gamma_j^r| = \mathscr{Y}_j^{rt}(\boldsymbol{\tau}_j| + \mathscr{Y}_j^{rr}(\boldsymbol{\rho}_j|. \tag{3.9}$$

The zeroth and first order terms of the single-body admittance matrix are

$$\mathscr{Y}_j^{(0)} = \boldsymbol{\mu}_j(0), \quad \mathscr{Y}_j^{(1)} = -\frac{1}{6\pi\eta}\begin{pmatrix}\mathbf{1} & \mathbf{0}\\ \mathbf{0} & \mathbf{0}\end{pmatrix}. \tag{3.10}$$

As a consequence the first order contribution to the bras in Eq. (3.9) is

$$(\gamma_j^{t(1)}| = \frac{-1}{6\pi\eta}(\boldsymbol{\tau}_j|, \quad (\gamma_j^{r(1)}| = 0. \tag{3.11}$$

The first order single-body friction kernel is

$$\mathsf{Z}_j^{(1)} = -\mathsf{Z}_j^{(0)}\mathsf{G}^{(1)}\mathsf{Z}_j^{(0)}. \tag{3.12}$$

In the following we often use the property that the steady state single-body friction and mobility matrices are each other's inverse,

$$\begin{pmatrix}\boldsymbol{\mu}_j^{tt}(0) & \boldsymbol{\mu}_j^{tr}(0)\\ \boldsymbol{\mu}_j^{rt}(0) & \boldsymbol{\mu}_j^{rr}(0)\end{pmatrix}\begin{pmatrix}\zeta_j^{tt}(0) & \zeta_j^{tr}(0)\\ \zeta_j^{rt}(0) & \zeta_j^{rr}(0)\end{pmatrix} = \begin{pmatrix}\zeta_j^{tt}(0) & \zeta_j^{tr}(0)\\ \zeta_j^{rt}(0) & \zeta_j^{rr}(0)\end{pmatrix}\begin{pmatrix}\boldsymbol{\mu}_j^{tt}(0) & \boldsymbol{\mu}_j^{tr}(0)\\ \boldsymbol{\mu}_j^{rt}(0) & \boldsymbol{\mu}_j^{rr}(0)\end{pmatrix} = \begin{pmatrix}\mathbf{1} & \mathbf{0}\\ \mathbf{0} & \mathbf{1}\end{pmatrix}. \tag{3.13}$$

Using this we find from Eqs. (3.8) and (3.11)

$$(\boldsymbol{t}_j^{(1)}| = 0, \quad (\boldsymbol{r}_j^{(1)}| = 0, \tag{3.14}$$

as derived elsewhere [7] in slightly different fashion.

The kets $|\boldsymbol{t}_j)$ and $|\boldsymbol{r}_j)$ in Eq. (2.8) are defined by

$$|\boldsymbol{t}_j) = \mathsf{Z}_j|\boldsymbol{\omega}_j^t), \quad |\boldsymbol{r}_j) = \mathsf{Z}_j|\boldsymbol{\omega}_j^r), \tag{3.15}$$

where

$$|\boldsymbol{\omega}_j^t) = |\boldsymbol{\tau}_j)\mathcal{Y}_j^{tt} - |\boldsymbol{\rho}_j)\mathcal{Y}_j^{rt},$$

$$|\boldsymbol{\omega}_j^r) = |\boldsymbol{\tau}_j)\mathcal{Y}_j^{tr} - |\boldsymbol{\rho}_j)\mathcal{Y}_j^{rr}. \tag{3.16}$$

Hence the first order contribution is

$$|\boldsymbol{\omega}_j^{t(1)}) = \frac{-1}{6\pi\eta}|\boldsymbol{\tau}_j), \quad |\boldsymbol{\omega}_j^{r(1)}) = 0. \tag{3.17}$$

Using Eq. (3.13) again we find

$$|\boldsymbol{t}_j^{(1)}) = 0, \quad |\boldsymbol{r}_j^{(1)}) = 0. \tag{3.18}$$

Expanding Eq. (2.11) we find by use of Eqs. (3.7), (3.14) and (3.18)

$$\mathcal{Y}_{jk}(\omega) = \boldsymbol{\mu}_{jk}(0) - \frac{\alpha}{6\pi\eta}\begin{pmatrix} \mathbf{1} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} \end{pmatrix} + O(\alpha^2), \tag{3.19}$$

as derived earlier by a different method [7]. This simple result leads to the generic amplitude of the $t^{-3/2}$ long-time tail of translational motion.

4. Expansion of single-body admittance matrix

In the following we shall need the second and third order terms $\mathcal{Y}_j^{(2)}$ and $\mathcal{Y}_j^{(3)}$ in the expansion of the 6×6 single-body admittance matrix. In the expressions (II.2.6) and (II.2.7) for these quantities we need in turn $\boldsymbol{\zeta}_j^{(2)}$ and $\boldsymbol{\zeta}_j^{(3)}$. These can be calculated by use of Eqs. (II.3.5) and (II.3.6). It is convenient to define the 6×6 matrices

$$\boldsymbol{M}_j^{(n)} = \begin{pmatrix} (\boldsymbol{\tau}_j|\mathsf{Z}_j^{(0)}\mathsf{G}^{(n)}\mathsf{Z}_j^{(0)}|\boldsymbol{\tau}_j) & -(\boldsymbol{\tau}_j|\mathsf{Z}_j^{(0)}\mathsf{G}^{(n)}\mathsf{Z}_j^{(0)}|\boldsymbol{\rho}_j) \\ (\boldsymbol{\rho}_j|\mathsf{Z}_j^{(0)}\mathsf{G}^{(n)}\mathsf{Z}_j^{(0)}|\boldsymbol{\tau}_j) & -(\boldsymbol{\rho}_j|\mathsf{Z}_j^{(0)}\mathsf{G}^{(n)}\mathsf{Z}_j^{(0)}|\boldsymbol{\rho}_j) \end{pmatrix}. \tag{4.1}$$

Then the matrix $\boldsymbol{\zeta}_j^{(2)}$ is given by

$$\boldsymbol{\zeta}_j^{(2)} = \frac{1}{(6\pi\eta)^2}\boldsymbol{\zeta}_j(0)\begin{pmatrix} \boldsymbol{\zeta}_j^{tt}(0) & \mathbf{0} \\ \mathbf{0} & \mathbf{0} \end{pmatrix}\boldsymbol{\zeta}_j(0) - \boldsymbol{M}_j^{(2)}, \tag{4.2}$$

and the matrix $\boldsymbol{\zeta}_j^{(3)}$ is given by

$$\boldsymbol{\zeta}_j^{(3)} = \frac{1}{(6\pi\eta)^3}\boldsymbol{\zeta}_j(0)\begin{pmatrix} \boldsymbol{\zeta}_j^{tt}(0)^2 & \mathbf{0} \\ \mathbf{0} & \mathbf{0} \end{pmatrix}\boldsymbol{\zeta}_j(0) - \frac{1}{6\pi\eta}(\boldsymbol{M}_j^{(2)}\boldsymbol{\zeta}_j^u + \boldsymbol{\zeta}_j^l\boldsymbol{M}_j^{(2)}) - \boldsymbol{M}_j^{(3)}, \tag{4.3}$$

with the matrices

$$\boldsymbol{\zeta}_j^u = \begin{pmatrix} \boldsymbol{\zeta}_j^{tt}(0) & \boldsymbol{\zeta}_j^{tr}(0) \\ \mathbf{0} & \mathbf{0} \end{pmatrix}, \quad \boldsymbol{\zeta}_j^l = \begin{pmatrix} \boldsymbol{\zeta}_j^{tt}(0) & \mathbf{0} \\ \boldsymbol{\zeta}_j^{rt}(0) & \mathbf{0} \end{pmatrix}. \tag{4.4}$$

Hence one finds for the second-order admittance matrix

$$\mathcal{Y}_j^{(2)} = \boldsymbol{\mu}_j(0)\boldsymbol{M}_j^{(2)}\boldsymbol{\mu}_j(0) - \frac{\eta}{\rho}\boldsymbol{\mu}_j(0)\boldsymbol{m}_j\boldsymbol{\mu}_j(0), \tag{4.5}$$

and for the third order matrix

$$\mathcal{Y}_j^{(3)} = \boldsymbol{\mu}_j(0)\boldsymbol{M}_j^{(3)}\boldsymbol{\mu}_j(0) + \frac{1}{6\pi\rho}\begin{pmatrix} \mathbf{1} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} \end{pmatrix}\boldsymbol{m}_j\boldsymbol{\mu}_j(0) + \frac{1}{6\pi\rho}\boldsymbol{\mu}_j(0)\boldsymbol{m}_j\begin{pmatrix} \mathbf{1} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} \end{pmatrix}. \tag{4.6}$$

It was shown earlier [11] that in particular the tensor $\mathcal{Y}_j^{(3)rr}$ is given by

$$\mathcal{Y}_j^{(3)rr} = \frac{1}{24\pi\eta}\mathbf{1} - \frac{1}{20\pi\eta}\boldsymbol{\mu}_j^{rd}(0) : \boldsymbol{\mu}_j^{dr}(0), \tag{4.7}$$

with zero-frequency mobility matrices that depend on the shape of the body j. As a consequence the amplitude of the $t^{-5/2}$ long-time tail for a single body depends on its shape, in contrast to the amplitude of the $t^{-3/2}$ long-time tail of translational motion.

5. Expansion of rotational admittance

It follows from Eq. (3.19) that the first odd power term in the expansion of the tensor $\mathcal{Y}_{jk}^{rr}(\omega)$ is of order α^3. Hence the rotational velocity correlation function $\boldsymbol{C}_{jk}^{rr}(t)$ decays at long times with a $t^{-5/2}$ long-time tail. The amplitude of the tail is determined by the tensor $\mathcal{Y}_{jk}^{(3)rr}$. We shall derive a result similar to Eq. (4.7) for this many-body tensor. We consider again the expression in Eq. (2.11).

From Eqs. (3.7) and (3.13) we find

$$(\boldsymbol{r}_j^{(0)}|\hat{\mathsf{V}}^{(1)} = 0, \quad \hat{\mathsf{V}}^{(1)}|\boldsymbol{r}_j^{(0)}) = 0. \tag{5.1}$$

By expansion of the rotational admittance in Eq. (2.11) to third order we therefore find

$$\mathcal{Y}_{jk}^{(3)rr} = \mathcal{Y}_j^{rr(3)}\delta_{jk} + (\boldsymbol{r}_j^{(0)}|\hat{\mathsf{V}}^{(3)}|\boldsymbol{r}_k^{(0)}) + (\boldsymbol{r}_j^{(3)}|\hat{\mathsf{V}}^{(0)}|\boldsymbol{r}_k^{(0)}) + (\boldsymbol{r}_j^{(0)}|\hat{\mathsf{V}}^{(0)}|\boldsymbol{r}_k^{(3)}). \tag{5.2}$$

It is shown straightforwardly from the multiple scattering expansion of the operator $\hat{\mathbf{V}}_{jk}$ by use of Eqs. (3.6) and (3.7) that

$$\hat{\mathsf{V}}_{jk}^{(3)} = \theta_j\mathsf{G}^{(3)}(1-\delta_{jk})\theta_k - \sum_l \hat{\mathsf{V}}_{jl}^{(0)}\hat{\mathsf{Z}}_l^{(3)}\hat{\mathsf{V}}_{lk}^{(0)} + \sum_{\substack{ln \\ l\neq n}} \hat{\mathsf{V}}_{jl}^{(0)}\hat{\mathsf{Z}}_l^{(0)}\mathsf{G}^{(3)}\hat{\mathsf{Z}}_n^{(0)}\hat{\mathsf{V}}_{nk}^{(0)}. \tag{5.3}$$

The second and third order terms in the expansion of the convective single-body friction kernel are given by

$$\hat{\mathsf{Z}}_j^{(2)} = -\hat{\mathsf{Z}}_j^{(0)}\mathsf{G}^{(2)}\hat{\mathsf{Z}}_j^{(0)}, \quad \hat{\mathsf{Z}}_j^{(3)} = -\hat{\mathsf{Z}}_j^{(0)}\mathsf{G}^{(3)}\hat{\mathsf{Z}}_j^{(0)}. \tag{5.4}$$

Hence Eq. (5.3) can be abbreviated as

$$\hat{\mathsf{V}}_{jk}^{(3)} = \theta_j\mathsf{G}^{(3)}(1-\delta_{jk})\theta_k + \sum_{ln} \hat{\mathsf{V}}_{jl}^{(0)}\hat{\mathsf{Z}}_l^{(0)}\mathsf{G}^{(3)}\hat{\mathsf{Z}}_n^{(0)}\hat{\mathsf{V}}_{nk}^{(0)}. \tag{5.5}$$

The third-order Green function in Eq. (3.5) can be expressed as

$$\boldsymbol{G}^{(3)}(\boldsymbol{r}-\boldsymbol{r}') = \frac{1}{60\pi\eta}[\boldsymbol{r}_j\boldsymbol{r}_k - \boldsymbol{r}_j\boldsymbol{r}'_k - \boldsymbol{r}'_j\boldsymbol{r}_k + \boldsymbol{r}'_j\boldsymbol{r}'_k - 2(\boldsymbol{r}_j\cdot\boldsymbol{r}_k - \boldsymbol{r}_j\cdot\boldsymbol{r}'_k - \boldsymbol{r}'_j\cdot\boldsymbol{r}_k + \boldsymbol{r}'_j\cdot\boldsymbol{r}'_k)\mathbf{1}], \tag{5.6}$$

with relative positions $\boldsymbol{r}_j = \boldsymbol{r} - \boldsymbol{R}_j$, $\boldsymbol{r}_k = \boldsymbol{r} - \boldsymbol{R}_k$, and $\boldsymbol{r}'_j = \boldsymbol{r}' - \boldsymbol{R}_j$, $\boldsymbol{r}'_k = \boldsymbol{r}' - \boldsymbol{R}_k$. This can be written in terms of bra and ket tensors as

$$\theta_j(\boldsymbol{r})\boldsymbol{G}^{(3)}(\boldsymbol{r}-\boldsymbol{r}')\theta_k(\boldsymbol{r}') = \frac{1}{60\pi\eta}[|\boldsymbol{q}_j)\cdot(\boldsymbol{\tau}_k| + |\boldsymbol{\tau}_j)\cdot(\boldsymbol{q}_k|] + \frac{1}{20\pi\eta}|\hat{\boldsymbol{\sigma}}_j) : (\hat{\boldsymbol{\sigma}}_k| - \frac{1}{24\pi\eta}|\boldsymbol{\rho}_j) : (\boldsymbol{\rho}_k|. \tag{5.7}$$

Here

$$|\boldsymbol{q}_j) = \theta_j(\boldsymbol{r})(\boldsymbol{r}_j\boldsymbol{r}_j - 2r_j^2\mathbf{1}), \quad (\boldsymbol{q}_k| = \theta_k(\boldsymbol{r}')(\boldsymbol{r}'_k\boldsymbol{r}'_k - 2r'^2_k\mathbf{1}). \tag{5.8}$$

The other bras and kets have been defined before [11,19]. Hence in the second term in Eq. (5.2) we have, using Eq. (3.13),

$$(\boldsymbol{r}_j^{(0)}|\hat{\mathbf{G}}^{(3)}|\boldsymbol{r}_k^{(0)}) = \frac{1}{20\pi\eta}[\boldsymbol{\mu}_j^{rt}(0)\boldsymbol{\zeta}_j^{td}(0) + \boldsymbol{\mu}_j^{rr}(0)\boldsymbol{\zeta}_j^{rd}(0)] : [\boldsymbol{\zeta}_k^{dt}(0)\boldsymbol{\mu}_k^{tr}(0) + \boldsymbol{\zeta}_k^{dr}(0)\boldsymbol{\mu}_k^{rr}(0)] + \frac{1}{24\pi\eta}\mathbf{1}. \tag{5.9}$$

This can be simplified to

$$(\boldsymbol{r}_j^{(0)}|\hat{\mathsf{G}}^{(3)}|\boldsymbol{r}_k^{(0)}) = \frac{1}{24\pi\eta}\mathbf{1} - \frac{1}{20\pi\eta}\mu_j^{rd}(0) : \boldsymbol{\mu}_k^{dr}(0). \tag{5.10}$$

Similarly, using the single-body result Eq. (4.7) and Eq. (5.5), we find for the sum of the first two terms in Eq. (5.2)

$$\mathscr{Y}_j^{(3)rr} + (\boldsymbol{r}_j^{(0)}|\hat{\mathsf{V}}^{(3)}|\boldsymbol{r}_k^{(0)}) = \frac{1}{24\pi\eta}\mathbf{1} - \frac{1}{20\pi\eta}\boldsymbol{\mu}_j^{rd}(0) : \boldsymbol{\mu}_k^{dr}(0) - \frac{1}{20\pi\eta}\sum_{ln}\Delta\boldsymbol{\mu}_{jl}^{rd}(0) : \Delta\boldsymbol{\mu}_{nk}^{dr}(0), \tag{5.11}$$

with the definition

$$\Delta\boldsymbol{\mu}_{ij}^{\kappa\lambda} = \boldsymbol{\mu}_{ij}^{\kappa\lambda} - \boldsymbol{\mu}_i^{\kappa\lambda}\delta_{ij}. \tag{5.12}$$

In order to find $\mathscr{Y}_{jk}^{rr(3)}$ we must add the third and fourth terms in Eq. (5.2).

By expansion in Eq. (3.15) we find

$$|\boldsymbol{r}_j^{(3)}) = \mathsf{Z}_j^{(0)}|\boldsymbol{\omega}_j^{r(3)}) + \mathsf{Z}_j^{(1)}|\boldsymbol{\omega}_j^{r(2)}) + \mathsf{Z}_j^{(3)}|\boldsymbol{\omega}_j^{r(0)}), \tag{5.13}$$

omitting a term with $|\boldsymbol{\omega}_j^{r(1)})$ on account of Eq. (3.17). The kets $|\boldsymbol{\omega}_j^{r(2)})$ and $|\boldsymbol{\omega}_j^{r(3)})$ follow by expansion in Eq. (3.16). In the sum the mass terms cancel between the first and second term, and the matrix $\boldsymbol{M}^{(2)}$ cancels between the second and third term. We are left with

$$|\boldsymbol{r}_j^{(3)}) = \mathsf{Z}_j^{(0)}|\boldsymbol{\tau}_j)[\boldsymbol{\mu}_j(0)\boldsymbol{M}_j^{(3)}\boldsymbol{\mu}_j(0)]^{tr} - \mathsf{Z}_j^{(0)}|\boldsymbol{\rho}_j)[\boldsymbol{\mu}_j(0)\boldsymbol{M}_j^{(3)}\boldsymbol{\mu}_j(0)]^{rr} - \mathsf{Z}_j^{(0)}\mathsf{G}^{(3)}\mathsf{Z}_j^{(0)}[|\boldsymbol{\tau}_j)\boldsymbol{\mu}_j(0)^{tr} - |\boldsymbol{\rho}_j)\boldsymbol{\mu}_j(0)^{rr}]. \tag{5.14}$$

Similarly we find by expansion in Eq. (3.8)

$$(\boldsymbol{r}_j^{(3)}| = [\boldsymbol{\mu}_j(0)\boldsymbol{M}_j^{(3)}\boldsymbol{\mu}_j(0)]^{rt}(\boldsymbol{\tau}_j|\mathsf{Z}_j^{(0)} + [\boldsymbol{\mu}_j(0)\boldsymbol{M}_j^{(3)}\boldsymbol{\mu}_j(0)]^{rr}(\boldsymbol{\rho}_j|\mathsf{Z}_j^{(0)} - [\boldsymbol{\mu}_j(0)^{rt}(\boldsymbol{\tau}_j| + \boldsymbol{\mu}_j(0)^{rr}(\boldsymbol{\rho}_j|]\mathsf{Z}_j^{(0)}\mathsf{G}^{(3)}\mathsf{Z}_j^{(0)}. \tag{5.15}$$

By use of Eqs. (4.1) and (5.6) we find

$$[\boldsymbol{\mu}_j(0)\boldsymbol{M}_j^{(3)}\boldsymbol{\mu}_j(0)]^{tr} = -\frac{1}{60\pi\eta}\boldsymbol{\mu}_j^{qr}(0) - \frac{1}{20\pi\eta}\boldsymbol{\mu}_j^{td}(0) : \boldsymbol{\mu}_j^{dr}(0),$$

$$[\boldsymbol{\mu}_j(0)\boldsymbol{M}_j^{(3)}\boldsymbol{\mu}_j(0)]^{rr} = \frac{1}{24\pi\eta}\mathbf{1} - \frac{1}{20\pi\eta}\boldsymbol{\mu}_j^{rd}(0) : \boldsymbol{\mu}_j^{dr}(0),$$

$$[\boldsymbol{\mu}_j(0)\boldsymbol{M}_j^{(3)}\boldsymbol{\mu}_j(0)]^{rt} = \frac{1}{60\pi\eta}\boldsymbol{\mu}_j^{rq}(0) - \frac{1}{20\pi\eta}\boldsymbol{\mu}_j^{rd}(0) : \boldsymbol{\mu}_j^{dt}(0). \tag{5.16}$$

The terms with $\mathbf{G}^{(3)}$ in Eqs. (5.14) and (5.15) can be evaluated by use of Eq. (5.7). Hence we find

$$|\boldsymbol{r}_j^{(3)}) = \frac{-1}{20\pi\eta}\mathbf{Z}_j^{(0)}[|\boldsymbol{\tau}_j)\boldsymbol{\mu}_j^{td}(0) : \boldsymbol{\mu}_j^{dr}(0) - |\boldsymbol{\rho}_j)\boldsymbol{\mu}_j^{rd}(0) : \boldsymbol{\mu}_j^{dr}(0) - |\hat{\boldsymbol{\sigma}}_j)\boldsymbol{\mu}_j^{dr}(0)],$$

$$(\boldsymbol{r}_j^{(3)}| = \frac{-1}{20\pi\eta}[\boldsymbol{\mu}_j^{rd}(0) : \boldsymbol{\mu}_j^{dt}(0)(\boldsymbol{\tau}_j| + \boldsymbol{\mu}_j^{rd}(0) : \boldsymbol{\mu}_j^{dr}(0)(\boldsymbol{\rho}_j| + \boldsymbol{\mu}_j^{rd}(0)(\hat{\boldsymbol{\sigma}}_j|]\mathbf{Z}_j^{(0)}, \tag{5.17}$$

or by use of Eqs. (I.4.21-24)

$$|\boldsymbol{r}_j^{(3)}) = \frac{-1}{20\pi\eta}|\hat{\mathbf{d}}_j)\boldsymbol{\mu}_j^{dr}(0), \quad (\boldsymbol{r}_j^{(3)}| = \frac{-1}{20\pi\eta}\boldsymbol{\mu}_j^{rd}(0)(\hat{\mathbf{d}}_j|. \tag{5.18}$$

Hence the third and fourth terms in Eq. (5.2) are given by

$$(\boldsymbol{r}_j^{(0)}|\hat{\mathbf{V}}^{(0)}|\boldsymbol{r}_k^{(3)}) = \frac{-1}{20\pi\eta}\Delta\boldsymbol{\mu}_{jk}^{rd}(0) : \boldsymbol{\mu}_k^{dr}(0),$$

$$(\boldsymbol{r}_j^{(3)}|\hat{\mathbf{V}}^{(0)}|\boldsymbol{r}_k^{(0)}) = \frac{-1}{20\pi\eta}\boldsymbol{\mu}_j^{rd}(0) : \Delta\boldsymbol{\mu}_{jk}^{dr}(0). \tag{5.19}$$

This yields finally by use in Eq. (5.2)

$$\mathcal{Y}_{jk}^{(3)rr} = \frac{1}{24\pi\eta}\mathbf{1} - \frac{1}{20\pi\eta}\left[\boldsymbol{\mu}_j^{rd}(0) : \boldsymbol{\mu}_k^{dr}(0) + \sum_{ln} \Delta\boldsymbol{\mu}_{jl}^{rd}(0) : \Delta\boldsymbol{\mu}_{nk}^{dr}(0) + \Delta\boldsymbol{\mu}_{jk}^{rd}(0) : \boldsymbol{\mu}_k^{dr}(0) + \boldsymbol{\mu}_j^{rd}(0) : \Delta\boldsymbol{\mu}_{jk}^{dr}(0)\right]. \tag{5.20}$$

For a sphere the tensors $\boldsymbol{\mu}_j^{rd}(0)$ and $\boldsymbol{\mu}_j^{dr}(0)$ vanish [11], so that for a collection of spheres the result simplifies to

$$\mathcal{Y}_{jk}^{(3)rr} = \frac{1}{24\pi\eta}\mathbf{1} - \frac{1}{20\pi\eta}\sum_{ln} \boldsymbol{\mu}_{jl}^{rd}(0) : \boldsymbol{\mu}_{nk}^{dr}(0) \quad \text{(spheres)}. \tag{5.21}$$

We have used traceless symmetric force dipole moments, see the remark following Eq. (II.4.11).

The result (5.20) can be used in the expression for the long-time rotational motion of an arbitrary set of rigid bodies. We consider an initial state where all bodies and the fluid are at rest. If body k is suddenly accelerated by a rotational impulse corresponding to the applied torque

$$\boldsymbol{N}_k(t) = \boldsymbol{S}_{Rk}\delta(t), \tag{5.22}$$

then all bodies will be set into motion, as given by the admittance matrix described in Eq. (2.2). By inverse Fourier transform one finds that the rotational velocity of body j decays asymptotically as

$$\boldsymbol{\Omega}_j(t) \approx \frac{3}{4\sqrt{\pi}}\left(\frac{\rho}{\eta}\right)^{3/2} \mathcal{Y}_{jk}^{(3)rr} \cdot \boldsymbol{S}_{Rk} t^{-5/2}, \quad \text{as } t \to \infty. \tag{5.23}$$

The applied torque is assumed so small that the change of configuration can be neglected.

In the theory of Brownian motion one considers the rotational velocity correlation function, as discussed in Section 2. By use of the fluctuation–dissipation theorem one finds for the long-time behavior

$$\boldsymbol{C}_{jk}^{rr}(t) \approx k_B T \frac{3}{4\sqrt{\pi}}\left(\frac{\rho}{\eta}\right)^{3/2} \mathcal{Y}_{jk}^{(3)rr} t^{-5/2} \quad \text{as } t \to \infty. \tag{5.24}$$

The tensor $\mathcal{Y}_{jk}^{(3)rr}$ depends on the initial configuration of all bodies. It is expressed in terms of zero frequency mobility tensors that in principle can be found from the steady-state Stokes equations. It is a purely geometrical quantity, independent of the shear viscosity and the mass density of bodies and fluid.

6. Discussion

We have derived a simple expression for the long-time behavior of the rotational motion of a set of rigid bodies immersed in a viscous fluid after an initial twist applied to one of the bodies. The coefficient of the $t^{-5/2}$ long-time tail for each of the bodies depends on shape and configuration of all bodies. This is in contrast to the corresponding long-time coefficient for the translational motion after an initial impulse, which is universal and independent of shape and configuration [7]. The rotational long-time coefficients are expressed in terms of zero frequency mobility tensors.

The theorem can be used in the study of rotational Brownian motion in a suspension. For a semi-dilute suspension one must find the motion of a pair of spheres. The calculation of the complete behavior in time is complicated [17]. For the long-time behavior the above theorem allows reduction to a steady-state problem.

For definiteness we have assumed rigid bodies with stick boundary conditions, but it is clear that the same derivation holds for mixed slip-stick boundary conditions, as well as for porous bodies. A similar derivation holds also in two dimensions.

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