*J. Fluid Mech.* (1983), *vol.* 137, *pp.* 217–220
*Printed in Great Britain*



# The long-time tail of the angular-velocity autocorrelation function for a rigid Brownian particle of arbitrary centrally symmetric shape

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(Received 14 June 1983)

The long-time $t^{-\frac{5}{2}}$ behaviour of the angular-velocity autocorrelation function is determined by the diffusion of fluid motion to large distances, from where the particle appears a point singularity. From an examination of this flow, the coefficient of $t^{-\frac{5}{2}}$ can be related to some effective aspect ratios which describe how the particle rotates in a simple shear flow.

## 1. Introduction

The autocorrelation function of the velocity of a spherical particle in Brownian motion has a $t^{-\frac{3}{2}}$ asymptotic decay at long times in translational motion (Alder & Wainwright 1970), and a $t^{-\frac{5}{2}}$ asymptotic decay in rotational motion (Ailawadi & Berne 1971). The translational result for a sphere is precisely true for a particle of arbitrary shape, whereas the coefficient of $t^{-\frac{5}{2}}$ in the rotational result depends on the shape of the particle. This coefficient has been calculated for ellipsoids of revolution by Hocquart (1977), who obtained the Laplace transform of the entire angular-velocity autocorrelation function by solving the time-dependent linearized equations fôr the fluid motion in spheroidal coordinates.

In this paper we shall find the coefficient of $t^{-\frac{5}{2}}$ in the long-time asymptotic form of the angular-velocity autocorrelation function for a particle of general shape, restricted only so that it has a mirror symmetry about three mutually orthogonal planes, i.e. a so-called centrally symmetric particle. This restriction is necessary to avoid a coupling between translation and rotation, because the rotational $t^{-\frac{5}{2}}$ term found here would be changed by a small $t^{-1}$ correction in the translational $t^{-\frac{3}{2}}$ term, a correction which we have been unable to calculate.

We shall determine the angular velocity autocorrelation function in §4 as the decay of the angular velocity after an impulsively applied couple of magnitude $kT$ (see e.g. Hinch 1975). The long-time asymptotic behaviour is governed by the diffusion of fluid motion to large distances, at which the particle appears at leading order as a force dipole. We derive this motion in §3, having previously introduced some useful properties of the steady motion of the particle in §2.

## 2. A grand resistance matrix and the effect aspect ratios

We consider centrally symmetric particles, i.e. those with a mirror symmetry about three mutually orthogonal planes, because such particles have no coupling between their translational and rotational motion in slow viscous flows (low Reynolds number). Thus we can ignore the translation and just consider the particle rotating steadily with angular velocity $\boldsymbol{\omega}$ in a steady linear shear flow:

$$\boldsymbol{\Omega} \wedge \boldsymbol{x}+\boldsymbol{E}\cdot\boldsymbol{x},$$

where we have split the shear into a rotational part $\boldsymbol{\Omega}$ and a pure straining motion $\boldsymbol{E} = \boldsymbol{E}^{\mathrm{T}}$. This viscous flow relative to the spinning particle will exert a steady couple $\boldsymbol{G}$ and a steady symmetric force dipole $\boldsymbol{S}$ on the particle given by

$$\tfrac{1}{2}\epsilon_{ijk} G_k + S_{ij} = \oint x_i \sigma_{jk} n_k \,\mathrm{d}A.$$

By the linearity of Stokes flow, $\boldsymbol{G}$ and $\boldsymbol{S}$ are linearly related to $\boldsymbol{\Omega}-\boldsymbol{\omega}$ and $\boldsymbol{E}$, i.e.

$$\begin{pmatrix}\boldsymbol{G}\\ \boldsymbol{S}\end{pmatrix} = \begin{pmatrix}\boldsymbol{A} & \boldsymbol{B}\\ \boldsymbol{B}^{\mathrm{T}} & \boldsymbol{C}\end{pmatrix}\begin{pmatrix}\boldsymbol{\Omega}-\boldsymbol{\omega}\\ \boldsymbol{E}\end{pmatrix}, \tag{1}$$

where the tensors $\boldsymbol{A}$, $\boldsymbol{B}$ and $\boldsymbol{C}$ depend on the size, shape and orientation of the particle, and are known for some particular particles such as ellipsoids and long slender particles. Using a reciprocal theorem it is possible to show that the grand resistance matrix is symmetric (Hinch 1972).

The restriction above to steady motion is really a restriction to changes taking place over times long compared with the time vorticity takes to diffuse the length $l$ of the particle, i.e. $l^2/\nu$. For more rapid changes in time it is possible to make a frequency-dependent generalization of the grand resistance relation (1).

In § 4 we shall find that it is not necessary to know the tensors $\boldsymbol{A}$, $\boldsymbol{B}$ and $\boldsymbol{C}$ in full detail, because the final answer only depends on the combination

$$\boldsymbol{D} = \boldsymbol{A}^{-1}\cdot\boldsymbol{B}.$$

Now Bretherton (1962) has shown that this third-order tensor for a centrally symmetric particle has only three independent components with respect to axes that coincide with the symmetry planes of the particle. These independent components are

$$D_{312} = D_{321}, \quad D_{123} = D_{132}, \quad D_{231} = D_{213}.$$

The remaining components are zero. The non-zero components are often expressed in terms of three effective aspect ratios $r_{12}$, $r_{23}$ and $r_{31}$ by

$$D_{312} = \frac{1}{2}\frac{r_{12}^2-1}{r_{12}^2+1}. \tag{2}$$

For an ellipsoid $r_{12} = a_1/a_2$, where $a_i$ are the semimajor axes. Note for a general particle it is not necessary for the product $r_{12}r_{23}r_{31}$ to be equal to unity, as in the case for an ellipsoid.

The effective aspect ratios of a centrally symmetric particle can be measured experimentally from the period of rotation of a particle of the same shape placed in a simple shear flow (see e.g. Harris, Nawaz & Pittman 1979). In the simple shear $\boldsymbol{u} = (\gamma x_2, 0, 0)$ the particle will rotate about its 3-axis with a period $2\pi\,(r_{12}^2+1)/\gamma r_{12}$.

## 3. Flow due to point singularities

We first calculate the flow due to an impulsive point force, i.e. the flow governed by

$$\nabla\cdot\boldsymbol{u} = 0,$$

$$\rho\frac{\partial \boldsymbol{u}}{\partial t} = -\nabla p+\mu\nabla^2\boldsymbol{u}+\boldsymbol{F}\delta(\boldsymbol{x})\,\delta(t).$$

Fourier-transforming in space, we have, in $t \geqslant 0$,

$$\boldsymbol{u}(\boldsymbol{x},t) = \frac{1}{\rho(2\pi)^3}\int\left(\boldsymbol{F}-\boldsymbol{k}\frac{\boldsymbol{k}\cdot\boldsymbol{F}}{k^2}\right)\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x}-\nu k^2 t}\,\mathrm{d}^3k.$$

Later we shall need the behaviour at $x$ small compared with the diffusion distance $(\nu t)^{\frac{1}{2}}$, which comes from $k$ small $O((\nu t)^{\frac{1}{2}})$. Expanding $\exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x})$ for small $\boldsymbol{k}\cdot\boldsymbol{x}$, we obtain

$$\boldsymbol{u} \sim \frac{1}{\rho(4\pi\nu t)^{\frac{3}{2}}}\left\{\tfrac{2}{3}\boldsymbol{F}+\frac{1}{10\nu t}[\boldsymbol{x}(\boldsymbol{F}\cdot\boldsymbol{x})-2\boldsymbol{F}x^2]+O\left(\frac{x^4}{\nu^2t^2}\right)\right\}.$$

From this fundamental solution we can calculate the response to an impulsive point dipole $(\frac{1}{2}\epsilon_{ijk}G_k+\delta_{ij})$ applied to the fluid, i.e. the flow governed by

$$\nabla\cdot\boldsymbol{u} = 0,$$

$$\rho\frac{\partial u_j}{\partial t} = \frac{\partial p}{\partial x_j}+\rho\nabla^2 u_j+(\tfrac{1}{2}\epsilon_{ijk}G_k+S_{ij})\frac{\partial}{\partial x_i}\delta(\boldsymbol{x})\,\delta(t).$$

We find, in $t \geqslant 0$,

$$\boldsymbol{u} \sim \frac{1}{\rho(4\pi\nu t)^{\frac{3}{2}}\,10\nu t}\left[\tfrac{5}{2}\boldsymbol{G}\wedge\boldsymbol{x}+3\boldsymbol{S}\cdot\boldsymbol{x}-\boldsymbol{x}(\boldsymbol{S}\cdot\boldsymbol{I})+O\left(\frac{x^3}{\nu t}\right)\right]. \quad (3)$$

## 4. Long-time behaviour after an impulsively applied couple

Now when a couple $\boldsymbol{G}$ is impulsively applied to a centrally symmetric particle it will rotate in a complicated time-dependent way. In order to remain rigid and not deform, it will exert stresses on the fluid which have a symmetric force dipole, as well as force octopoles and higher multipoles. (The central symmetry of the particle rules out any coupling to the force pole, quadrupole and higher even multipoles.)

At large times the fluid motion is dominated by the force dipole, with small corrections from the force octopole and higher multipoles. The overall force dipole which enters the long-time behaviour is the integral over time of the instantaneous value. The antisymmetric part of the overall force dipole is just the given impulsive couple $\boldsymbol{G}$. The symmetric part of the overall force dipole is given by solving the zero-frequency grand resistance problem (1) with no flow given at infinity ($\boldsymbol{\Omega} = 0 = \boldsymbol{E}$), i.e.

$$\boldsymbol{S} = \boldsymbol{B}^{\mathrm{T}}\cdot\boldsymbol{A}^{-1}\cdot\boldsymbol{G}. \quad (4)$$

Thus at large times the particle sees the flow (3) with $\boldsymbol{S}$ given by (4). This is a quasi-steady shearing flow with

$$\boldsymbol{\Omega} = \frac{1}{\rho(4\pi\nu t)^{\frac{3}{2}}\,10\nu t}\boldsymbol{G},\quad \boldsymbol{E} = \frac{3}{\rho(4\pi\nu t)^{\frac{3}{2}}\,10\nu t}\boldsymbol{S}. \quad (5)$$

The free response of the particle to the flow (3) is a quasi-steady rotation $\boldsymbol{\omega}$ given by solving another zero-frequency grand resistance problem (1), this time with $\boldsymbol{G} = 0$ corresponding to no couple being applied after the initial impulse, i.e.

$$\boldsymbol{\omega} = \boldsymbol{\Omega} + \boldsymbol{A}^{-1} \cdot \boldsymbol{B} : \boldsymbol{E}.$$

Substituting (4) and (5), we have

$$\boldsymbol{\omega} \sim \frac{1}{\rho(4\pi\nu t)^{\frac{3}{2}}\,\nu t}\left[\tfrac{1}{4}\boldsymbol{G} + \tfrac{3}{10}\boldsymbol{A}^{-1} \cdot \boldsymbol{B} : \boldsymbol{B}^{\mathrm{T}} \cdot \boldsymbol{A}^{-1} \cdot \boldsymbol{G}\right]. \qquad (6)$$

Finally we recall that the angular-velocity autocorrelation function for Brownian motion is the decay of the angular velocity after an impulsively applied couple of magnitude $kT$. Thus combining result (6) with the representation (2), we have in the principle axes of the centrally symmetric particle at long times

$$\langle \omega_i(t+\tau)\,\omega_j(t)\rangle \sim \frac{\pi kT}{\rho(4\pi\nu t)^{\frac{5}{2}}}\left[1 + \frac{3}{5}\left(\frac{r_{23}^2 - 1}{r_{23}^2 + 1}\right)^2\right] \quad (i = j = 1),$$

with similar results for $i = j = 2$ and 3, and zero for $i \neq j$. This long-time asymptotic decay agrees with that of Hocquart (1977) in the case of ellipsoids which have $r_{23} = a_2/a_3$.

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