Effects of Fore–Aft Asymmetry on the Sedimentation and Dispersion of Axisymmetric Brownian Particles

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1. INTRODUCTION

Consider the sedimentation in a quiescent fluid of a dilute "cloud" of Brownian particles. The settling under the action of an external force field is always accompanied by dispersion resulting from the translational molecular diffusion of the particles. When the cloud consists of particles possessing internal degrees of freedom (e.g., orientational degrees of freedom for rigid, nonspherical particles, conformational degrees of freedom for flexible particles, etc.), dispersion is enhanced by the Taylor mechanism arising from the coupling between the dependence of the settling velocity upon the instantaneous internal configuration and the stochastic sampling of the configurational space via Brownian (rotary or conformational) diffusion.

Previous analyses have addressed the Taylor dispersion accompanying the sedimentation of both rigid nonspherical (1, 2) and flexible (3–5) particles. Explicit results have, however, been presented only for centrosymmetric homogeneous particles (rigid ellipsoids, symmetric flexible dumbbells, and flexible porous spheres). It is thus the goal of the present contribution to examine the effects of fore–aft asymmetry upon the transport process of Brownian axisymmetric homogeneous particles.

Rather than considering the whole multiparticle cloud, we focus, as is commonly accepted in the dilute limit, on the motion of but a single tracer particle. The state of such a particle is completely specified by the position vector \( \mathbf{R} = (x, y, z) \) in three-dimensional physical space of a body-fixed material point (its "locator point") together with its instantaneous orientation. It is most convenient to choose the locator point at the center of reaction of the particle, and to represent the instantaneous orientation by a unit vector \( \mathbf{e} \) attached to its symmetry axis. (See Fig. 1.) The full description of the motion of the particle is thus embodied in \( P(\mathbf{R}, \mathbf{e}, t; |\mathbf{e}|) \), the conditional probability density for finding the tracer particle at time \( t > 0 \) with its locator point situated at the physical-space position \( \mathbf{R} \) and possessing the orientation \( \mathbf{e} \), given that it was originally introduced at time \( t = 0 \) at the position \( \mathbf{R} = 0 \) with the orientation \( \mathbf{e}' \). This probability density satisfies the continuity equation

\[
\frac{\partial P}{\partial t} + \nabla_{\mathbf{R}} \cdot J + \nabla_{\mathbf{e}} \cdot \mathbf{j} = 0, \tag{1.1}
\]

in conjunction with the constitutive relations

\[
J = M(\mathbf{e}) \cdot \mathbf{FP} - D(\mathbf{e}) \cdot \nabla_{\mathbf{R}} P \tag{1.2}
\]

and

\[
\mathbf{j} = m \mathbf{T} \times \mathbf{e} P - \mathbf{d} \nabla_{\mathbf{e}} P \tag{1.3}
\]

for the respective physical- and orientational-flux density.
in which \( \mathbf{r} \) is the vector from the center of reaction to the center of mass of the particle (or, generally, from the locator point to the line of action of \( \mathbf{F} \)). Thus the axisymmetric particle experiences an orienting torque (which tends to align it with the external field) competing with the disorienting effect of rotary Brownian diffusion (which, in turn, tends to create a uniform orientational distribution). Furthermore,

\[
\nabla_\mathbf{R} = \frac{\partial}{\partial \mathbf{R}} = \left( \frac{\partial}{\partial x}, \frac{\partial}{\partial y}, \frac{\partial}{\partial z} \right)
\]

[1.8]

denotes the physical-space gradient operator, and

\[
\nabla_\mathbf{e} = i_\theta \frac{\partial}{\partial \theta} + i_\phi \frac{1}{\sin \theta} \frac{\partial}{\partial \phi}
\]

[1.9]
is the orientation-space gradient operator. [Herc, \( \mathbf{e}, i_\theta, i_\phi \)] is a particle-fixed right-handed triad of unit vectors and \( (\theta, \phi) \) are the spherical polar (Eulerian) angles parameterizing the orientation of the particle \( \mathbf{e} = \mathbf{e}(\theta, \phi) \) relative to a space-fixed Cartesian frame of reference, cf. Fig. 1.]

The above equations are supplemented by the boundary condition

\[
|\mathbf{R}|^m(P, \mathbf{J}, \mathbf{j}) = (0, 0, 0) \quad \text{as} \quad |\mathbf{R}| \to \infty
\]

\[
(m = 0, 1, 2, \cdots)
\]

[1.10]

assuring that \( P \) decays exponentially rapidly in physical space, and by the requirement that \( P \) be continuous and single-valued in the orientation-space variable \( \mathbf{e} \). To these we adjoin the initial condition

\[
P = \begin{cases} 
\delta(\mathbf{R})\delta(\mathbf{e} - \mathbf{e}') & (t = 0) \\
0 & (t < 0),
\end{cases}
\]

[1.11]
in which the Dirac distributions are, explicitly,

\[
\delta(\mathbf{R}) = \delta(x)\delta(y)\delta(z)
\]

[1.12a]

and

\[
\delta(\mathbf{e} - \mathbf{e}') = (\sin \theta)^{-1}\delta(\theta - \theta')\delta(\phi - \phi').
\]

[1.12b]
The foregoing initial- and boundary-value problem uniquely determines \( P \geq 0 \). Furthermore, it is readily verified that the solution satisfies the normalization condition

\[
\int_{\mathbb{R}^3} \int_{S_2} Pd^2\mathbf{e}d^3\mathbf{R} = 1 \quad (t \geq 0),
\]

[1.13]

wherein \( d^3\mathbf{R} = dx dy dz \) is a volume element of the three-
dimensional physical space and \( d^2e = (\sin \theta) \, \delta\phi \, d\theta \) is an areal element on the unit sphere, and the integration extends over the entire physical space \( \mathbf{R}^3 \), and the entire surface of the unit sphere \( S_2 \). Additionally, it can be easily demonstrated that the solution of the foregoing problem can serve as the appropriate Green’s function for the general initial-value problem where [1.11] is replaced by an arbitrary initial distribution.

The explicit solution of the above problem in the five-dimensional phase space \( \mathbf{R}^3 \otimes S_2 \) is a rather formidable task. Furthermore, one is often not interested in the exhaustively detailed information embodied in the exact solution \( \bar{P} \), but rather in the orientationally averaged density

\[
\bar{P}(\mathbf{R}, t | e') = \int_{S_2} P d^2e
\]

which provides the description of the transport process in physical space irrespective of the particle instantaneous orientation. While \( \bar{P} \) can, in principle, be calculated from [1.14], it is obviously desirable to obtain it (at least approximately) without the a priori knowledge of the exact solution.

In the next section we show how this can be achieved through the use of the generalized Taylor dispersion paradigm (7, 8, 1). In Section 3 we study the dependence of the macroscale phenomenological coefficients characterizing \( \bar{P} \) upon the relevant Langevin parameter representing the combined effect of particle asymmetry and the external field. Explicit results describing the influence of particle shape are presented in Section 4 for the case of an asymmetric dumbbell consisting of a pair of unequal homogeneous spheres.

2. GENERALIZED TAYLOR DISPERSION THEORY

Upon recognition of the equivalence relations

\[
\begin{align*}
\mathbf{R} & \rightarrow \mathbf{Q} \quad [2.1a] \\
e & \rightarrow q \quad [2.1b]
\end{align*}
\]

which identify the physical-space position and orientation of the particle with the respective “global” and “local” coordinates of the generalized Taylor dispersion theory, it is established that the above problem consisting of Eqs. [1.1]–[1.3], [1.10], and [1.11] is formally equivalent to the generic formulation of the initial- and boundary-value problem underlying the latter theory. Consequently, in the long-time limit \( d_t \gg 1 \), when the particles have (through Brownian rotations) sampled the orientation space sufficiently many times so as to achieve a steady-state orientational distribution, the averaged density \( \bar{P} \) is approximated by the solution of the model problem

\[
\frac{\partial \bar{P}}{\partial t} + \nabla_R \cdot \mathbf{J} = 0 \quad [2.2a]
\]

in conjunction with

\[
\begin{align*}
\mathbf{J} &= \mathbf{U}^* \bar{P} - \mathbf{D}^* \cdot \nabla_R \bar{P} \\
\mathbf{R}^m(\bar{P}, \mathbf{J}) &= (0, 0) \quad \text{as} \ |\mathbf{R}| \rightarrow \infty \quad [2.2c]
\end{align*}
\]

and

\[
\bar{P} = \begin{cases} 
\delta(\mathbf{R}) & (t = 0) \\
0 & (t < 0)
\end{cases} \quad [2.2d]
\]

i.e., a convection–diffusion problem formulated exclusively within the physical space and characterized by the pair of constant phenomenological coefficients \( \mathbf{U}^* \), the average settling velocity vector, and \( \mathbf{D}^* \), the dispersivity dyadic. These coefficients are related to the respective long-time limits of the time derivatives

\[
\begin{align*}
\mathbf{U}^* &= \lim_{t \rightarrow \infty} \frac{dM_1}{dt} \quad [2.3] \\
\mathbf{D}^* &= \lim_{t \rightarrow \infty} \frac{1}{2}(\mathbf{M}_2 - \mathbf{M}_1 \mathbf{M}_1) \quad [2.4]
\end{align*}
\]

of the polyadic statistical moments,

\[
M_m \overset{\text{def}}{=} \int_{\mathbf{R}^3} \int_{S_2} R^m P d^2e d^3R \quad (m = 0, 1, 2, \cdots) \quad [2.5]
\]

of the probability density \( P \). Alternatively, \( \mathbf{U}^* \) and \( \mathbf{D}^* \) are obtainable (cf. (7, 8)) via the following quadratures over the orientation space

\[
\mathbf{U}^* = \bar{\mathbf{M}} \cdot \mathbf{F}, \quad [2.6a]
\]

where

\[
\bar{\mathbf{M}} \overset{\text{def}}{=} \int_{S_2} P_0^{\bar{\mathbf{M}}} \mathbf{M}(e) d^2e, \quad [2.6b]
\]

and

\[
\mathbf{D}^* = \mathbf{D}^u + \mathbf{D}^c, \quad [2.7a]
\]

in which

\[
\mathbf{D}^u \overset{\text{def}}{=} \int_{S_2} P_0^{\bar{\mathbf{D}}} \mathbf{D}(e) d^2e = kT \bar{\mathbf{M}} \quad [2.7b]
\]

[cf. (1.5)], and

\[
\mathbf{D}^c \overset{\text{def}}{=} \int_{S_2} P_0^{\bar{\mathbf{D}}} \mathbf{B}(e) \Delta \mathbf{U}(e) d^2e, \quad [2.7c]
\]
\[ \Delta U(e) \overset{\text{def.}}{=} (\hat{M}(e) - \hat{M}) \cdot F \quad [2.7d] \]

are, respectively, the "molecular" and "Taylor" contributions to the dispersivity dyadic. In [2.7c], \[ \mathbf{F} \] denotes the symmetrization operator acting on the polyadic quantity within the brackets.

The scalar field \( P_0^\infty(e) \) has been obtained by Brenner and Condif (9) to be

\[ P_0^\infty = \frac{1}{4\pi \sinh \chi} e^{\chi e \cdot \hat{r}}, \quad [2.8] \]

in which \( \chi \) is given by

\[ \chi = \chi_0 \left( \frac{r}{\hat{e}} \right), \quad [2.9a] \]

\[ \chi_0 = \frac{F_c}{kT} \quad [2.9b] \]

is the appropriate Langevin parameter representing the effect of the orienting torque associated with the asymmetry of the particle relative to the disorienting influence of the rotary Brownian diffusion, \( F = |\mathbf{F}| \) is the magnitude of the external force, \( \hat{F} = F/F \), and \( c \) is a characteristic dimension of the particle.

Substitute [2.8] and [1.4] into [2.6b] to obtain the dimensionless, transversely isotropic average mobility (appropriately normalized relative to the average value pertaining to the symmetric case)

\[ \tilde{\hat{M}} = \frac{\hat{M}}{(M_{||} + 2 M_{\perp})/3} = \frac{M_{||}}{M_{||} + 2 M_{\perp})/3} \tilde{\hat{M}} \tilde{\hat{F}} \tilde{\hat{F}} + \frac{M_{||}}{M_{||} + 2 M_{\perp})/3} \tilde{\hat{M}} \frac{1 - \hat{F} \hat{F}}{3}, \quad [2.10a] \]

wherein

\[ \tilde{\hat{M}}_{||} = \frac{M_{||} - 2(M_{||} - M_{\perp})L(\chi)/\chi}{(M_{||} + 2 M_{\perp})/3}, \quad [2.10b] \]

\[ \tilde{\hat{M}}_{\perp} = \frac{M_{||} + (M_{||} - M_{\perp})L(\chi)/\chi}{(M_{||} + 2 M_{\perp})/3}, \quad [2.10c] \]

and

\[ L(\chi) = \coth \chi - 1/\chi \]

is the Langevin function. Use [2.10a] in [2.6a] to obtain the dimensionless average settling velocity

\[ \tilde{U}^* = \frac{U^*}{(M_{||} + 2 M_{\perp})/3} = \tilde{\hat{M}}_{||} \tilde{\hat{F}}. \quad [2.11] \]

The vector \( \mathbf{B}(e) \) field satisfies (cf. (10)) the equation

\[ \nabla_e \cdot [\chi(1 - e e) \cdot \hat{F} P_0^\infty(e) \mathbf{B} - \nabla_e(P_0^\infty(e) \mathbf{B})] = d_{\tau}^{-1} P_0^\infty \Delta U \quad [2.12] \]

in conjunction with the normalization condition

\[ \int_{S_2} P_0^\infty \mathbf{B} d^2e = 0 \quad [2.13] \]

and the requirement that \( \mathbf{B}(e) \) be a continuous and single-valued function of \( e \). Use [1.4], [2.7d], and [2.10], together with the definition of the dimensionless vector \( \tilde{\mathbf{B}}(e) \) field,

\[ \mathbf{B} \overset{\text{def.}}{=} \frac{(M_{||} - M_{\perp})F}{d_{\tau}} \tilde{\mathbf{B}}, \quad [2.14] \]

to obtain from [2.12]

\[ \nabla_e \cdot [\chi(1 - e e) \cdot \hat{F} P_0^\infty(e) \tilde{\mathbf{B}} - \nabla_e(P_0^\infty(e) \tilde{\mathbf{B}})] 
\]

\[ = P_0^\infty(e) \left[ e e - \left( 1 - \frac{2}{\chi} \frac{L(\chi)}{3} \right) \hat{F} \right]. \quad [2.15] \]

When \( \mathbf{D}^* \) is normalized relative to \( kT(M_{||} + 2 M_{\perp})/3 \), the value corresponding to the case \( F = 0 \), in which (in the absence of Taylor dispersion) \( \mathbf{D}^* \) is isotropic, we obtain from [1.4], [2.7], [2.10a], [2.13], and [2.14]

\[ \mathbf{D}^* \overset{\text{def.}}{=} \frac{\mathbf{D}^*}{kT(M_{||} + 2 M_{\perp})/3} = \mathbf{M} + \chi \hat{e} g \mathbf{D}^C, \quad [2.16] \]

wherein the dimensionless scalar factor

\[ g = \frac{3(M_{||} - M_{\perp})^2}{(M_{||} + 2 M_{\perp}) m_{\tau} c^2} \quad [2.17] \]

depends only upon the shape of the particle, and the dyadic

\[ \mathbf{D}^C(\chi) = \int_{S_2} P_0^\infty(e) \left[ \tilde{\mathbf{B}}(e) e e \cdot \hat{F} \right] d^2e \quad [2.18] \]

is functionally dependent only upon the Langevin parameter \( \chi \). In the next section we calculate the vector \( \mathbf{B}(e) \) field enabling the subsequent evaluation of \( \mathbf{D}^C \). Explicit results for the functions \( f = r/c \) (cf. [2.9a]) and \( g \) [2.16], representing the effect of particle geometry, are subsequently presented in Section 4 for the case of an asymmetric homogeneous dumbbell consisting of unequal spheres.

3. THE (DIMENSIONLESS) VECTOR \( \mathbf{B}(e) \) FIELD AND TAYLOR DISPERSIVITY DYADIC \( \mathbf{D}^C \)

Assume a solution in the form

\[ P_0^\infty \mathbf{B} = P_0^\infty \mathbf{A} + \frac{P_0^\infty}{\chi} e \quad [3.1] \]
and make use of the equation
\[\chi (1 - e_e) \cdot \mathbf{F} \mathbf{P}_0^\infty - \nabla_e \mathbf{P}_0^\infty = 0, \quad [3.2]\]
satisfied by \( \mathbf{P}_0^\infty \) in conjunction with the definition [1.9] of \( \nabla_e \) and a number of vector identities, to obtain from [2.15] the equation
\[\nabla_e^2 (\mathbf{P}_0^\infty \mathbf{A}) - \chi \mathbf{F} \cdot \nabla_e (\mathbf{P}_0^\infty \mathbf{A}) + 2\chi (e \cdot \hat{\mathbf{F}}) \mathbf{P}_0^\infty \mathbf{A} = \frac{2}{\chi} \mathbf{P}_0^\infty \{ e - L(x) \hat{\mathbf{F}} \} \quad [3.3]\]
satisfied by \( \mathbf{A} \). In the following derivation it is useful to express the various quantities relative to a Cartesian space-fixed frame of reference. Without loss of generality we choose the \( z \)-axis to be parallel to \( \hat{\mathbf{F}} \). Subject to a posteriori verification we postulate a solution of the form
\[\mathbf{P}_0^\infty \mathbf{A} = \mathbf{P}_0^\infty [\mathbf{A}_\parallel (\theta) (\cos \phi + j \sin \phi) + k \mathbf{A}_\perp (\theta)], \quad [3.4]\]
in which \((i, j, k)\) is a right-handed triad of unit vectors such that \( k = \hat{\mathbf{F}} \). (See Fig. 1.) Substitution of the latter equation into [3.3] yields a pair of equations satisfied by \( \mathbf{A}_\parallel \) and \( \mathbf{A}_\perp \), respectively,
\[\frac{1}{\sin \theta} \frac{d}{d\theta} \left( \frac{d}{d\theta} (\mathbf{P}_0^\infty \mathbf{A}_\parallel) \sin \theta \right) + \chi \frac{d}{d\theta} (\mathbf{P}_0^\infty \mathbf{A}_\parallel) \sin \theta + 2\chi \mathbf{P}_0^\infty \mathbf{A}_\parallel \cos \theta = \frac{2}{\chi} \mathbf{P}_0^\infty \{ \cos \theta - L(x) \} \quad [3.5]\]
and
\[\frac{1}{\sin \theta} \frac{d}{d\theta} \left( \frac{d}{d\theta} (\mathbf{P}_0^\infty \mathbf{A}_\perp) \sin \theta \right)
+ \chi \frac{d}{d\theta} (\mathbf{P}_0^\infty \mathbf{A}_\perp) \sin \theta + \left( 2\chi \cos \theta - \frac{1}{\sin^2 \theta} \right) \mathbf{P}_0^\infty \mathbf{A}_\perp = \frac{2}{\chi} \mathbf{P}_0^\infty \sin \theta. \quad [3.6]\]

Alternatively, upon making use of [2.8], Eqs. [3.5] and [3.6] can be recast in the respective forms
\[\frac{1}{\sin \theta} \frac{d}{d\theta} \left( \frac{d}{d\theta} \mathbf{A}_\parallel \sin \theta \right) - \chi \frac{d}{d\theta} \mathbf{A}_\parallel \sin \theta
= \frac{2}{\chi} \{ \cos \theta - L(x) \} \quad [3.7]\]
and
\[\frac{1}{\sin \theta} \frac{d}{d\theta} \left( \frac{d}{d\theta} \mathbf{A}_\perp \sin \theta \right) - \chi \frac{d}{d\theta} \mathbf{A}_\perp \sin \theta
- \frac{1}{\sin^2 \theta} \mathbf{A}_\perp = \frac{2}{\chi} \sin \theta. \quad [3.8]\]

Calculation of \( \mathbf{A}_\parallel \)

Equation [3.7] is a first-order equation for \((dA_\parallel /d\theta) \sin \theta\), which is readily solved to obtain
\[\frac{dA_\parallel}{d\theta} \sin \theta = \frac{2 e^{-x \cos \theta}}{x^2 \sinh x} - \frac{2}{x^2} (\cos \theta - \coth \chi),\]
satisfying the regularity conditions \((dA_\parallel /d\theta) \sin \theta = 0\) at \( \theta = 0, \pi \). Integrate the latter result to obtain
\[A_\parallel = -\frac{2}{x^2} \int_0^\pi e^{-x \cos \psi} + (\sinh \chi)(\cos \psi) - \cosh \chi \\frac{(\sin \chi)(\sin \psi)}{\sin \psi} d\psi + \bar{A}_\parallel, \quad [3.9]\]
wherein the constant \( \bar{A}_\parallel \) is obtainable from the normalization condition [2.13] when use is made of [2.8] and [3.1] as
\[\bar{A}_\parallel = \frac{1}{x^2 \sinh^2 x} \int_0^\pi e^{-x \cos \psi} + (\sinh \chi)(\cos \psi) - \cosh \chi \sin \psi \times (e^x \cos \psi - e^{-x}) d\psi + \frac{1}{x} \left( \frac{1}{\chi} - \coth \chi \right). \quad [3.10]\]

Calculation of \( \mathbf{A}_\perp \)

Expanding \( \mathbf{P}_0^\infty \mathbf{A}_\perp \) in a series of associated Legendre functions
\[\mathbf{P}_0^\infty \mathbf{A}_\perp = \sum_{n=1}^{\infty} A_n \mathbf{P}_n^0(\cos \theta), \quad [3.11]\]
one obtains from [3.6] the system of linear algebraic equations for the coefficients \( A_n \) \((n = 1, 2, \cdots)\)
\[2A_1 + \frac{2}{x} A_2 = b_1, \quad [3.12a]\]
\[-\chi \frac{(n-1)(n+1)}{2n-1} A_{n-1} + n(n+1)A_n
+ \frac{n(n+1)}{2n+3} A_{n+1} = b_n \quad (n = 2, 3, \cdots), \quad [3.12b]\]
in which the forcing terms are
\[b_n = -\frac{1}{x} \frac{2n+1}{n(n+1)} \int_0^\pi \mathbf{P}_0^\infty \mathbf{P}_n^0(\cos \theta) \sin^2 \theta d\theta,
(n = 1, 2, \cdots). \quad [3.13]\]
Truncation of the system beyond \( n = N \) yields the approximant \((P_{\theta}^{n}A_{\theta})^{(N)}\) which is orthogonal to all \( P_{n}^{\perp} \) \((n = 1, 2, \ldots, N)\).

**Calculation of \( \mathbf{D}^C \)**

Substitute [2.1], [3.1], [3.9], [3.10], and [3.11] into [2.18] and make use of the orthogonality properties of the functions \( P_{n}^{\perp} \) to obtain the transversely isotropic result

\[
\mathbf{D}^C = \mathbf{D}_{||}^C(x) \mathbf{\hat{F}} + \mathbf{D}_{\perp}^C(x)(1 - \mathbf{\hat{F}}),
\]

[3.14]

wherein

\[
\mathbf{D}_{||}^C = \frac{1}{x} \left( \coth x - \frac{3}{x} + \frac{6}{x^2} \coth x - \frac{6}{x^3} \right) \\
+ \frac{3}{x} \left( \frac{1}{x} \coth x + \frac{2}{x^2} \right) \\
- \frac{1}{x} \int_0^x e^{-x \cos \psi} + \left( \sinh x \right) (\cos \psi) - \cosh x \\
\times \left[ \frac{1}{x} (e^{x \cos \psi} \cos^2 \psi - e^{-x}) - \frac{2}{x^2} (e^{x \cos \psi} \cos \psi + e^{-x}) \\
+ \frac{2}{x^3} (e^{x \cos \psi} - e^{-x}) \right] d\psi
\]

[3.15]

\[
\mathbf{D}_{\perp}^C = \frac{1}{x^2} \left( 1 - \frac{3}{x} \coth x + \frac{3}{x^2} \right) - \frac{4\pi A_2}{x^2}.
\]

[3.16]

Before proceeding to present the results obtained from the numerical calculations of both \( P_{\theta}^{n}A_{\theta} \) and the integrals in [3.15], we pause to obtain asymptotic approximations for \( \mathbf{D}^C \) in the respective limits of weak \((x \ll 1)\) and strong \((x \gg 1)\) external fields.

### 3.1. The Weak Field \((x \ll 1)\) Limit

Expanding \( P_{\theta}^{n} \) in a power series of \( x \), we obtain from [2.8]

\[
P_{\theta}^{n} \simeq \frac{1}{4\pi} \left[ 1 + x e \cdot \mathbf{\hat{F}} + \frac{1}{2} x^2 P_2 \cdot \mathbf{\hat{F}} + O(x^3) \right]
\]

[3.17a]

\[
\simeq \frac{1}{4\pi} \left[ 1 + x \cos \theta + \frac{1}{2} x^2 (3 \cos^2 \theta - 1) + O(x^3) \right].
\]

[3.17b]

Similarly, from [2.7d] in conjunction with [1.4] and [2.10]

\[
\Delta U = (M_{\|} - M_{\perp})F \left\{ \frac{3}{2} P_2(e) \cdot \mathbf{\hat{F}} \\
- 2 \left[ \frac{1}{x} - \frac{1}{x} \left( x - \coth x \right) \right] \right\}
\]

[3.18a]

\[
\simeq (M_{\|} - M_{\perp})F \left\{ \frac{3}{2} P_2(e) \cdot \mathbf{\hat{F}} - \frac{2}{45} x^2 \mathbf{\hat{F}} + O(x^4) \right\}.
\]

[3.18b]

In the above, the dyadic

\[
P_2 = \frac{1}{3} (3ee - 1)
\]

[3.19]

is the polyadic surface spherical harmonic of degree 2 \((11, 12)\). Substitution of the foregoing into [2.15] yields the equation

\[
\nabla^2 P_{\theta}^{n} \mathbf{\hat{B}} = x \left[ \mathbf{\hat{F}} \cdot \nabla P_{\theta}^{n} \mathbf{\hat{B}} - 2 (e \cdot \mathbf{\hat{F}}) P_{\theta}^{n} \mathbf{\hat{B}} \right]
\]

\[
- \frac{1}{4\pi} \left( \frac{3}{2} (3ee - 1) \cdot \mathbf{\hat{F}} - \frac{1}{x} x (3eee - le) \cdot \mathbf{\hat{F}} \right)
\]

\[
- x^2 (\frac{3}{2} eee (\cdot) \mathbf{\hat{F}} \cdot \mathbf{\hat{F}} - \frac{1}{6} ee \cdot \mathbf{\hat{F}} + \frac{1}{10} \mathbf{\hat{F}}) + O(x^3) \right],
\]

[3.20]

where \((\cdot)^m\) denotes \( m \) successive scalar contractions. The latter equation suggests an expansion of the form

\[
P_{\theta}^{n} \mathbf{\hat{B}} = \frac{1}{4\pi} \sum_{n=0}^{\infty} x^n \mathbf{\hat{B}}_n(e).
\]

[3.21]

When this is substituted into [3.20] and use is made of the normalization conditions (cf. [2.13])

\[
\int_{S_2} \mathbf{\hat{B}}_n d^2e = 0,
\]

[3.22]

one obtains a sequence of boundary-value problems which can be solved recursively for the various \( \mathbf{\hat{B}}_n(e) \) fields.

*The \( O(1) \) vector \( \mathbf{\hat{B}}_0(e) \) field.* From the leading-order terms of [3.20] we have

\[
\nabla^2 \mathbf{\hat{B}}_0 = -\frac{1}{3} (3ee - 1) \cdot \mathbf{\hat{F}} = -\frac{2}{3} P_2 \cdot \mathbf{\hat{F}}.
\]

[3.23]

Utilizing the equation

\[
\nabla^2 P_n = -n(n + 1)P_n
\]

[3.24]

satisfied by the surface spherical harmonics \((11, 12), [3.23]\) is readily inverted to yield

\[
\mathbf{\hat{B}}_0 = \frac{1}{3} P_2 \cdot \mathbf{\hat{F}}
\]

[3.25]

which satisfies the normalization condition [3.22] as a consequence of the orthogonality property

\[
\int_{S_2} P_n d^2e = 0 \quad (n \neq m)
\]

[3.26]

upon noting that \( P_0 = 1 \).
The $O(x)$ vector $\mathbf{B}_1(e)$ field. Substitution of $\mathbf{B}_0$ into [3.20] yields for the $O(x)$ contribution the equation

$$\nabla_2^2 \mathbf{B}_1 = -\frac{5}{3} \mathbf{eee} : \mathbf{F} \mathbf{F} + \frac{11}{18} \mathbf{e} \cdot \mathbf{F} \mathbf{F} + \frac{1}{6} \mathbf{F}. $$

Expressing the right-hand side of the latter equation in terms of $\mathbf{P}_a$ and utilizing [3.24], we obtain

$$\mathbf{B}_1 = \frac{1}{18} \mathbf{P}_3 : \mathbf{F} \mathbf{F} + \frac{5}{3} \mathbf{P}_1 : \mathbf{F} \mathbf{F} + \frac{1}{12} \mathbf{P}_1, \quad [3.27]$$

in which

$$\mathbf{P}_1 = \mathbf{e} \quad \text{and} \quad \mathbf{P}_3 = \frac{1}{2} (5 \mathbf{eee} - 3 \mathbf{ef} \mathbf{f}). \quad [3.28a, b]$$

The $O(x^2)$ vector $\mathbf{B}_2(e)$ field. Substitute $\mathbf{B}_1$ [3.27] into the right-hand side of [3.20] to obtain the $O(x^2)$ balance

$$\nabla_2^2 \mathbf{B}_2 = -\frac{41}{30} \mathbf{eee} (\cdot)^3 \mathbf{F} \mathbf{F} + \frac{7}{4} \mathbf{eee} : \mathbf{F} \mathbf{F} + \frac{11}{18} \mathbf{e} \cdot \mathbf{F} + \frac{1}{360} \mathbf{F},$$

which is inverted by expressing the right-hand side in terms of $\mathbf{P}_a$ and utilizing [3.24] to yield

$$\mathbf{B}_2 = \frac{41}{30} \mathbf{P}_4 (\cdot)^3 \mathbf{F} \mathbf{F} + \frac{31}{2288} \mathbf{P}_2 : \mathbf{F} \mathbf{F} + \frac{29}{2288} \mathbf{P}_2 : \mathbf{F}, \quad [3.29]$$

wherein

$$\mathbf{P}_4 = \frac{1}{8} (35 \mathbf{eee} - 30 \mathbf{ef} \mathbf{f} \mathbf{f} + 3 \mathbf{f} \mathbf{f} \mathbf{f}). \quad [3.30]$$

The foregoing procedure can, in principle, be continued to higher orders. The calculations involved become, however, increasingly tedious and of doubtful utility. We therefore only point out the observation (which can be substantiated by invoking inductive arguments) that in general, $\mathbf{B}_2n$ will depend only upon $\mathbf{P}_{2k+2}(k = 0, 1, \ldots, n)$ while $\mathbf{B}_{2n+1}$ will involve only $\mathbf{P}_{2k+1}(k = 0, 1, \ldots, n + 1)$.

The approximate expression for $\mathbf{D}^C$. Substitute the expansion [3.21] in conjunction with the definition [3.19] of $\mathbf{P}_2$ and the normalization condition [3.22] into [2.18], to obtain

$$\mathbf{D}^C \equiv \frac{1}{6\pi} \int_{S_1} \left\{ \mathbf{B}_0(e) + x \mathbf{B}_1(e) + x^2 \mathbf{B}_2(e) \right\} \cdot \mathbf{F} d^2 e.$$

Employing the orthogonality property [3.26] as well as the result

$$\frac{1}{4\pi} \int_{S_1} \mathbf{P}_2 d^2 e = \frac{1}{36} \mathbf{f} \mathbf{f} \mathbf{f} - \frac{1}{3} \mathbf{f} \mathbf{f}$$

together with the expressions for $\mathbf{B}_i(i = 0, 1, 2)$ result in

$$\mathbf{D}^C = \mathbf{D}_0^C + x^2 \mathbf{D}_2^C + O(x^4), \quad [3.31a]$$

in which the leading-order term,

$$\mathbf{D}_0^C = \frac{1}{720} (\mathbf{F} \mathbf{F} + 3 \mathbf{I}), \quad [3.31b]$$

agrees with the result (1) for centrosymmetric particles, and

$$\mathbf{D}_2^C = \frac{1}{2288} (61 \mathbf{F} \mathbf{F} + 59 \mathbf{I}). \quad [3.31c]$$

Odd powers of $x$ are missing in the expansion [3.31a] in accordance with the above observation that $\mathbf{B}_{2n-1}(n = 0, 1, 2, \ldots)$ do not include a contribution of $\mathbf{P}_2$. (Thus the error term is $O(x^4)$ even though $\mathbf{B}_3$ has not been calculated explicitly.) Adopting the transversely isotropic form [3.14], one obtains

$$\mathbf{D}_2^C \approx \frac{2}{15} \mathbf{I} + \frac{1}{180} x^2 + O(x^4)$$

and

$$\mathbf{D}_2^C \approx \frac{1}{90} + \frac{59}{2288} x^2 + O(x^4). \quad [3.2a, b]$$

3.2. The Strong Field ($x \gg 1$) Limit

(i) Approximate expression for $\mathbf{D}^C$. The dominant contribution to the integral on the right-hand side of [3.10] comes from the neighborhood of $\psi = 0$. Defining the "inner" variable $\lambda$ by

$$\psi = \delta(x) \lambda, \quad \delta = x^{-1/2}. \quad [3.33a, b]$$

and applying Laplace's method (cf. (13)), we obtain

$$\frac{1}{x^2 \sin^2 x} \int_0^\pi e^{-x \cos \psi} + (\sinh x)(\cos \psi) - \cosh x \sin \psi$$

$$\times (e^{x \cos \psi} - e^{-x}) d\psi$$

$$\equiv - \frac{2\delta}{x} \int_0^\pi e^{-x \sin(\delta \lambda)} [1 - \cos(\delta \lambda)] d\lambda + O(e^{-2x})$$

$$\equiv - \frac{1}{x^3} \int_0^\infty \lambda e^{-(1/2)x^2} \left[ 1 + \frac{1}{12x} (\lambda^2 + \frac{1}{2} \lambda^4) + O(x^{-2}) \right]$$

$$\times d\lambda + O(e^{-2x})$$

$$\equiv - \frac{1}{x^3} - \frac{1}{2x^4} + O(x^{-5}). \quad [3.34]$$

hence, from [3.10],

$$\mathbf{A}_l \approx - \frac{1}{x} + \frac{1}{x^2} - \frac{1}{x^3} - \frac{1}{2x^4} + O(x^{-5}). \quad [3.35]$$

In a similar manner, neglecting exponentially small terms, we estimate the last term on the right-hand side of [3.15] to be


\[
\frac{2b}{\chi^2} \int_0^{\pi/\delta} e^{-x[1-\cos(\delta \lambda)]} \left[ 1 - \cos(\delta \lambda) \right] \sin(\delta \lambda) \left( \cos^2(\delta \lambda) - \frac{2}{\lambda} \cos(\delta \lambda) + \frac{2}{\chi^2} \right) d\lambda \\
\approx \frac{1}{\chi^3} - \frac{7}{2\chi^4} + O(\chi^{-5}).
\]

Substitution of this latter result together with \[3.35\] into \[3.15\] leads to

\[
\tilde{D}_0^C \approx \frac{2}{\chi^3} - \frac{6}{\chi^4} + O(\chi^{-5}). \quad [3.36]
\]

(ii) Approximate expression for \(\tilde{D}_C\). Upon making use of the definition of the coefficient \(A_2\) (cf. \[3.11\]), \(\tilde{D}_C\) \[3.16\] may be written in the form

\[
\tilde{D}_C^C = \frac{1}{\chi^2} \left( 1 - \frac{3}{\chi} \coth \chi + \frac{3}{\chi^2} \right) \\
+ \pi \int_0^{\pi} A_1(\theta) P_0^2(\theta) \sin^2 \theta \cos \theta d\theta, \quad [3.37]
\]

in which \(A_1\) satisfies \[3.8\] together with the regularity conditions\(^2\)

\[
A_1(\theta) = 0 \quad \text{for} \quad \theta = 0, \pi \quad [3.38]
\]

Assuming an asymptotic expansion of the form

\[
A_1 = \sum_{n=0}^{\infty} \delta_n(\chi) f_0(\theta),
\]

wherein \(\delta_n(n = 0, 1, 2, \ldots)\) constitute an asymptotic sequence for \(\chi \to \infty\), one obtains

\[
A_1 \approx -\frac{2\theta + a_0}{\chi^2} + O(\chi^{-3}), \quad [3.39a]
\]

which cannot simultaneously satisfy both boundary conditions \[3.38\]. It is established in Appendix A that one has to satisfy the condition at \(\theta = 0\); i.e.,

\[
A_1 \approx -\frac{2\theta}{\chi^2} + O(\chi^{-3}). \quad [3.39b]
\]

The resulting expansion is nonuniform in an \(O(\chi^{-1/2})\) vi-

\(^2\) These conditions are compatible with the kinematic interpretation (14) of the vector \(B(e)\) field as the long-time limit of the average physical space position of the particle, given that its orientation is \(e\), relative to its "total" (i.e., free of the latter conditioning) average physical-space position.

The results of the present calculation are summarized in Fig. 2 which describes the variation with \(\chi\) of \(\tilde{D}_0^C\) and \(\tilde{D}_C^C\), the respective longitudinal and transverse components of \(\tilde{D}_C\) (cf. \[3.14\]). Full lines denote the exact solution ([3.15] and [3.16], respectively); dashed lines represent the weak field \(\chi \ll 1\) approximation, Eq. [3.32]; and dotted lines correspond to the strong field \(\chi \gg 1\) asymptotes, Eqs. [3.36] and [3.40], respectively. Both "exact" curves start from the
values corresponding to sedimentation of centrosymmetric particles (\(1/3\) for \(D_\theta^C\) and \(1/6\) for \(D_\perp^C\)). For \(\chi > 0.1\) they start to rise, attain respective maxima (first \(D_\theta^C \approx 2.02 \times 10^{-2}\) at \(\chi \approx 1.66\) and then \(D_\perp^C \approx 1.58 \times 10^{-2}\) at \(\chi \approx 2.74\)), and finally descend monotonically. The curves intersect once near the peak of the \(D_\theta^C\) curve. Thus, while \(D_\theta^C > D_\perp^C\) in weak fields, the latter becomes the larger of the two in strong fields. The respective weak field approximations are accurate for \(\chi < \sim 0.6\) for both \(D_\theta^C\) and \(D_\perp^C\). The respective strong field asymptotes closely approximate \(D_\theta^C\) for \(\chi > \sim 6\) and \(D_\perp^C\) for \(\chi > \sim 14\).

The Taylor dispersion contribution to \(D^*\) arises from the fluctuations of the settling velocity about its mean value, originating from the Brownian rotations. The above trends are therefore rationalized in the following in terms of the respective variations with increasing \(\chi\) of \(P_0^O\), the orientational distribution, and \(\Delta U\), the settling velocity difference. Toward this end, it is useful to express \(\Delta U\) by the sum,

\[
\Delta U = \Delta U_\parallel + \Delta U_\perp,
\]

of the respective components parallel and perpendicular to the external field \(F\). Upon making use of (3.18a), we obtain the magnitudes of the latter components

\[
\Delta U_\parallel \equiv |\Delta U_\parallel| = (M_\parallel - M_\perp)F
\times \left[ \cos^2 \theta - \left[ 1 + \frac{2}{\chi} \left( \frac{1}{\chi} - \coth \chi \right) \right] \right],
\]

and

\[
\Delta U_\perp \equiv |\Delta U_\perp| = (M_\parallel - M_\perp)F \sin \theta \cos \theta,
\]

whose orientational gradients are, respectively,

\[
\nabla_\theta (\Delta U_\parallel) = -i_\theta (M_\parallel - M_\perp)F \sin 2\theta
\]

and

\[
\nabla_\theta (\Delta U_\perp) = -i_\theta (M_\parallel - M_\perp)F \cos 2\theta.
\]

With increasing value of \(\chi\), \(P_0^O\) which initially (for \(\chi = 0\)) represents a uniform orientational distribution becomes increasingly biased toward the preferred orientation \(e = \tilde{F}\) (\(\theta = 0\)). Indeed, from (2.8)

\[
\lim_{\chi \to \infty} P_0^O = \delta(e - \tilde{F});
\]

i.e., particles are aligned with the external field. The confinement of the particles to an increasingly narrower domain of orientation space effectively reduces (and eventually eliminates altogether) the aforementioned settling-velocity fluctuations which are the source of Taylor dispersion in the present context. As a result both \(D_\theta^C\) and \(D_\perp^C\) decrease (and ultimately vanish) with increasing \(\chi > 3\). Furthermore, it is worthwhile to note that in the vicinity of the aligned orientation, \(\theta = 0\), the orientational gradients of \(\Delta U_\parallel\), the longitudinal component of \(\Delta U\), vanish, cf. (3.43a), while the corresponding gradients of \(\Delta U_\perp\), the transverse component of \(\Delta U\), are maximized, cf. (3.43b). These explain the earlier observation that \(\tilde{D}_\theta^C\) decays faster (like \(O(\chi^{-3})\), cf. (3.31)) than \(\tilde{D}_\perp^C\) (only like \(O(\chi^{-2})\), cf. (3.40)). The present reduction of \(\tilde{D}_\perp^C\) is comparable in the context of the classical Taylor problem (15) to the decrease in longitudinal solute dispersion resulting from the restriction of the lateral motion of solute particles to but a narrow portion of the duct cross-sectional domain. Evidently, the longitudinal dispersion decays more rapidly when the latter cross-sectional portion is centered about a point of extremum in the longitudinal fluid velocity (where its lateral gradients vanish).

Following the preceding line of reasoning, one might expect \(\tilde{D}_\perp^C\) to decrease monotonically with \(\chi\). The occurrence of the maxima in Fig. 2 demonstrates that this is not the case, yet the initial growth of \(\tilde{D}_\perp^C\) (for \(\chi < 1\)) is a higher-order, \(O(\chi^4)\), effect and is accordingly less intuitive than the strong-field behavior outlined above. Inasmuch as Taylor dispersion arises from the orientational dependence of \(\Delta U\) and since the variable part of the latter (cf. (3.18a)) is explicitly independent of \(\chi\), it seems that the initial growth of \(\tilde{D}_\perp^C\) originates from the redistribution of orientations resulting from the action of the external force on the asymmetric particles. Focusing on the asymptotic expansion (3.17), we observe that the leading, \(O(\chi)\) correction represents a positive increment to \(P_0^O\) for \(0 < \theta < \pi/2\) accompanied by an equal deficit for the corresponding \(\pi - \theta\). Owing to the invariance of \(\Delta U\) under the transformation \(e \rightarrow -e\), this \(O(\chi)\) contribution to \(P_0^O\) cannot affect \(\tilde{D}_\perp^C\). Thus, in agreement with (3.32a, b), it is only the second-order, \(O(\chi^2)\) correction to \(P_0^O\) which can potentially account for the leading contribution to the initial enhancement of Taylor dispersion.

### 4. EFFECTS OF PARTICLE GEOMETRY: ASYMMETRIC DUMBBELLS

In the following we examine the influence of particle shape upon the Taylor dispersion accompanying its sedimentation. Specifically, we consider an asymmetrical dumbbell consisting of a pair of unequal spheres (of radii \(a_1\) and \(a_2\), respectively) whose center-to-center distance is \(\ell\) (cf. Fig. 1). Following (16), this geometry is effectively represented by the size ratio of the spheres

\[
\lambda = \frac{a_2}{a_1}
\]

which, without loss of generality, is restricted to the interval \(0 < \lambda \leq 1\), as well as the dimensionless separation between the centers.
The above parameters affect $D_C$ in two ways: (i) implicitly, via the magnitude of $\chi$, the Langevin parameter [2.9] representing the effect of the orienting torque which, for a given intensity of external field $F$, is determined (for a homogeneous particle) by the distance $r$ (cf. [1.7] et seq.) between the respective centers of mass and reaction (see Fig. 1), and (ii) explicitly, through the scalar factor $g$ appearing in the expression for $D_C$ (see [2.7], [2.16], and [2.17]). Both $r/a_1(s, \lambda)$ and $g(s, \lambda)$ presented in the following were calculated from hydrodynamic data available in the literature for a pair of free (17, cf. Appendix B) or touching (18) spheres.

Figure 3 describes the variation of $r/a_1$, with $\lambda$ for the indicated values of $s$. The dashed lines correspond to the leading-order term in the large-$s$ asymptotic expansion

$$
\frac{r}{a_1} \approx \frac{\lambda - \lambda^3}{1 + \lambda^3} + \frac{3}{4} \frac{\lambda - \lambda^2}{1 + \lambda^2} + O(s^{-1}). \quad [4.3]
$$

With decreasing value of $\lambda$, $r/a_1$ initially increases from $r/a_1 = 0$ at $\lambda = 1$ (i.e., a symmetric dumbbell), attains a maximal value (at $\lambda = \frac{1}{2}$), and then diminishes monotonically with $\lambda \to 0$. Thus, with the size of the smaller sphere becoming vanishingly small, the dumbbell is rendered hydrodynamically equivalent to the single (larger) sphere. It is interesting to note that, throughout the range of $s$ values displayed in the figure, the asymptotic approximation (which derives from the assumption $s \gg 1$) is remarkably accurate.\footnote{For the indicated values of $s$ the incorporation of the second term on the right-hand side of [4.3] does not improve the accuracy of the approximation. This may result from [4.3] being an asymptotic rather than a convergent series.}

Figure 4 shows the variation of $g$ with $\lambda$ for the indicated values of $s$. Also presented (by the dashed line) is the large-$s$ asymptote

$$
g(s, \lambda) \approx \frac{\lambda^3}{4(1 + \lambda)^3} + O(s^{-1}). \quad [4.4]
$$

All the curves descend monotonically to zero with $\lambda \to 0$, since (cf. [2.17]) $M_2 - M_1$ decreases while $M_0$ increases. With increasing $s$, $g$ grows monotonically and approaches the above $s$-independent limit [4.4]. In this limit hydrodynamic interactions between the spheres become negligible, hence $M_2 - M_1$ tends to zero. Nevertheless, $g$ does not vanish inasmuch as $M_0$, the rotary mobility, diminishes simultaneously.

**APPENDIX A: BOUNDARY-LAYER ANALYSIS OF $A_\perp$**

Define the inner variables

$$
\pi - \theta = x^{-1/2} \psi \quad \text{and} \quad A_\perp(\theta) = x^{-5/2} h(\psi) \quad [A.1a, b]
$$

and substitute into [3.8] to obtain the equation

$$
\frac{1}{\psi} \frac{d}{d\psi} \left( \frac{d}{d\psi} \psi \right) + \frac{d}{d\psi} \psi - \frac{h}{\psi^2} = 2\psi, \quad [A.2]
$$

which is supplemented by the boundary condition

$$
h(\psi) = 0 \quad \text{for} \quad \psi = 0, \quad [A.3]
$$

as well as the requirement of matching with the outer solution [3.39b]. Equation [A.2] possesses the particular solution

$$
h^{(0)} = 2\psi. \quad [A.4]
$$

Applying the additional transformation of variables

$$
h = x \psi, \quad x = -\frac{1}{2} \psi^2
$$
leads from the homogeneous equation associated with [A.2] to the confluent hypergeometric equation for $y(x)$

$$xy'' - xy' + \frac{1}{2}y = 0.$$  

We thus obtain (cf. (19)) the general solution of [A.2]

$$h = \psi M(\frac{1}{2}, 2, -\frac{1}{2} \psi^2)$$

$$\times \left[ A + B \int \psi \frac{e^{-\psi x^2} d\psi}{\chi^3 M(\frac{1}{2}, 2, -\frac{1}{2} \psi^2)} \right] + 2\psi, \quad \text{[A.5]}$$

in which $M(a, b, z)$ denotes Kummer's function. For small values of $\psi$, the second term within the brackets is $O(\psi^2)$. Satisfaction of (A.3) thus requires that $B = 0$. Making use of the asymptotic expression (19)

$$M(a, b, z) \approx \frac{\Gamma(b)}{\Gamma(a)} (-z)^{-a} [1 + O(|z|^{-1})]$$

as $|z| \to \infty (R_e \{ z \} < 0)$, \text{[A.6]}

(whence $\Gamma(z)$ denotes the gamma function and $R_e \{ \} \) represents the real part of the quantity within the braces). In the matching condition, we obtain $A = \left( \frac{1}{2} \right) \pi (2\pi \chi)^{1/2}$, thus the inner solution is

$$A_{\perp} \approx \frac{2\psi}{\chi^{1/2}} \frac{1}{2} \pi \left( 2\pi \chi \right)^{1/2} \psi \frac{\Gamma\left( \frac{1}{2}, 2, -\frac{1}{2} \psi^2 \right)}{\psi^2}, \quad \text{[A.7]}$$

Since it is not a priori obvious that the outer solution is nonuniform near $\theta = \pi$, one could attempt to locate the boundary layer at $\theta = 0$ by selecting $a_0 = -2\pi$ in [3.39a] and defining the inner variable

$$\theta = \chi^{-1/2} \psi. \quad \text{[A.8]}$$

Subsequent derivation is similar to the above, the most significant difference being the appearance of Kummer's function $M(\frac{1}{2}, 2, \frac{1}{2} \psi^2)$ rather than the prior $M(\frac{1}{2}, 2, -\frac{1}{2} \psi^2)$. Because of the exponential asymptotic divergence (cf. (19)) of

$$M(a, b, z) \approx \frac{\Gamma(b)}{\Gamma(a)} e^{z \alpha - b} [1 + O(|z|^{-1})]$$

as $|z| \to \infty (R_e \{ z \} > 0)$, the present inner solution cannot be matched to the outer one, which substantiates our earlier statement (cf. [3.39]).

\textbf{APPENDIX B: THE MOBILITY DYADICS OF A RIGID ASYMMETRICAL DUMBBELL}

In the following we outline the calculation of mobility dyadics of the asymmetric rigid dumbbell the hydrodynamic data (17) pertaining to a pair of (free) spheres.

The hydrodynamic force on sphere (I) (see Fig. 1), $F_I$, and the hydrodynamic torque about its center, $T_I$, are given by

$$F_I = -\mu (c^{\mathbf{K}}^{(11)} + c^{\mathbf{K}}^{(12)}) \cdot \mathbf{U}_0 + (c^{\mathbf{K}}^{(11)} + c^{\mathbf{K}}^{(21)})$$

$$- (c^{\mathbf{K}}^{(11)} \times r_1 - c^{\mathbf{K}}^{(12)} \times r_2) \cdot \Omega, \quad \text{[B.1]}$$

and

$$T_I = -\mu (c^{\mathbf{K}}^{(11)} + c^{\mathbf{K}}^{(12)}) \cdot \mathbf{U}_0 + (c^{\mathbf{K}}^{(11)} + c^{\mathbf{K}}^{(21)})$$

$$- (c^{\mathbf{K}}^{(12)} \times r_1 - c^{\mathbf{K}}^{(12)} \times r_2) \cdot \Omega, \quad \text{[B.2]}$$

respectively. In the above $\mu$ is the fluid viscosity, $\mathbf{U}_0$ is the velocity vector of the locator point $O$ of the dumbbell, $\Omega$ is the angular velocity vector, and $r_1, r_2$ are the position vectors from $O$ to the centers of spheres (I) and (II), respectively. The dyadics $c^{\mathbf{K}}^{(0)}, c^{\mathbf{K}}^{(0)}$, and $c^{\mathbf{K}}^{(0)}$, respectively, denote the hydrodynamic resistances to translation, rotation, and the coefficient of coupling between these two elementary motions. These dyadic coefficients are obtainable via the formulation and solution of the appropriate hydrodynamic boundary-value problems for the pair of spheres and are tabulated in (17). Interchanging the indices, 1 and 2, we obtain, similarly to [B.1] and [B.2], the expressions for $F_2$ and $T_2$, the respective force and torque on sphere (II).

The resultant hydrodynamic force and torque on the dumbbell are, respectively,

$$F = F_I + F_2 = c^{\mathbf{K}}_0 \cdot \mathbf{U}_0 + c^{\mathbf{K}}_o \cdot \Omega \quad \text{[B.3]}$$

and

$$T_0 = T_I + T_2 + r_1 \times F_I + r_2 \times F_2$$

$$= c^{\mathbf{K}}_o \cdot \mathbf{U}_0 + c^{\mathbf{K}}_o \cdot \Omega, \quad \text{[B.4]}$$

wherein $c^{\mathbf{K}}_0, c^{\mathbf{K}}_o$, and $c^{\mathbf{K}}_o$ denote the respective hydrodynamic resistances to translation and rotation and the coefficient of coupling between these two elementary modes of motion of the dumbbell. Substituting [B.1] and [B.2] into [B.3] and [B.4], we obtain

$$c^{\mathbf{K}}_0 = c^{\mathbf{K}}^{(11)} + c^{\mathbf{K}}^{(12)} + c^{\mathbf{K}}^{(21)} + c^{\mathbf{K}}^{(22)}, \quad \text{[B.5]}$$

$$c^{\mathbf{K}}_o = c^{\mathbf{K}}^{(11)} + c^{\mathbf{K}}^{(12)} + c^{\mathbf{K}}^{(21)} + c^{\mathbf{K}}^{(22)}$$

$$+ r_1 \times (c^{\mathbf{K}}^{(11)} + c^{\mathbf{K}}^{(12)}) + r_2 \times (c^{\mathbf{K}}^{(21)} + c^{\mathbf{K}}^{(22)}), \quad \text{[B.6]}$$

and

$$c^{\mathbf{K}}_o = c^{\mathbf{K}}^{(11)} + c^{\mathbf{K}}^{(12)} + c^{\mathbf{K}}^{(21)} + c^{\mathbf{K}}^{(22)}$$

$$- 2[r_1 \times (c^{\mathbf{K}}^{(11)} + c^{\mathbf{K}}^{(12)}) \times r_1] + 2[r_2 \times (c^{\mathbf{K}}^{(21)} + c^{\mathbf{K}}^{(22)}) \times r_2]$$

$$- r_1 \times (c^{\mathbf{K}}^{(11)} + c^{\mathbf{K}}^{(12)}) \times r_2 \times (c^{\mathbf{K}}^{(21)} + c^{\mathbf{K}}^{(22)}) \times r_2$$

$$- 2[r_1 \times (c^{\mathbf{K}}^{(11)} + c^{\mathbf{K}}^{(12)}) \times r_2 \times (c^{\mathbf{K}}^{(21)} + c^{\mathbf{K}}^{(22)}) \times r_2]. \quad \text{[B.7]}$$
Making use of the axial symmetry, the various resistance dyadics of the individual spheres as well as those of the dumbbell can be represented (20, 21) as

\[ ^rK^{(ij)} = ^rK^{(ij)}_\parallel ee + rK^{(ij)}_\perp (I - ee), \quad \text{(B.8a)} \]
\[ ^rK^{(ij)} = rK^{(ij)}_\parallel ee + rK^{(ij)}_\perp (I - ee). \quad \text{(B.8b)} \]

and

\[ ^cK^{(ij)} = ^cK^{(ij)}_e e, \quad \text{(B.8c)} \]

in which \(e\) denotes the unit alternating triadic. From the representations [B.8a, b], in conjunction with the symmetry of the grand resistance matrix (22), result the additional relations

\[ ^tK^{(ij)} = ^rK^{(ij)} \quad \text{and} \quad ^rK^{(ij)} = ^rK^{(ij)} \].

Substitute the foregoing expressions into [B.5]–[B.7] and write (cf. Fig. 1)

\[ r_1 = r_1 e, \quad r_2 = -r_2 e \quad \text{(B.9)} \]

to obtain

\[ ^tK^\parallel = ^tK^{(11)} + 2^tK^{(12)} + ^tK^{(22)}, \quad \text{(B.10a, b)} \]
\[ ^tK^\perp = ^tK^{(11)} + 2^tK^{(12)} + ^tK^{(22)}, \quad \text{(B.11a)} \]
\[ ^rK^\parallel = rK^{(11)} + 2rK^{(12)} + rK^{(22)}, \quad \text{(B.11b)} \]
\[ ^rK^{(o)} = rK^{(11)} + 2rK^{(12)} + rK^{(22)} + 2r_2(\ ^cK^{(12)} + ^cK^{(22)}) - 2r_1(\ ^cK^{(11)} + ^cK^{(21)}) + (r_1)^2\ ^tK^{(11)} + (r_2)^2\ ^tK^{(22)} - 2r_1r_2\ ^tK^{(12)}. \]

and

\[ ^cK^{(o)} = ^cK^{(11)} + ^cK^{(12)} + ^cK^{(21)} + ^cK^{(22)} + r_2(^tK^{(22)} + ^tK^{(21)}) - r_1(^tK^{(22)} + ^tK^{(21))}. \quad \text{(B.12)} \]

The mobility dyadics of the dumbbell are obtained by substitution of the above results into the relations (cf. 9)

\[ ^tM_o = \frac{1}{\mu} [^tK^{-1} + ^tK^{-1} \cdot ^cK_0 \cdot A^{-1} \cdot ^cK_0 \cdot ^tK^{-1}], \quad \text{(B.13a)} \]
\[ ^tM = \frac{1}{\mu} A^{-1}, \quad \text{and} \quad ^cM_o = -\frac{1}{\mu} A^{-1} \cdot ^cK_0 \cdot ^tK^{-1} \quad \text{(B.13b, c)} \]

wherein the dyadic \(A\) is defined by

\[ A^{\text{def}} = ^tK_o - ^cK_0 \cdot ^tK^{-1} \cdot ^cK_0 = A_\parallel ee + A_\perp (I - ee) \quad \text{(B.14a)} \]

in which

\[ A_\parallel = ^tK^\parallel \quad \text{and} \quad A_\perp = ^tK^{-1}_\perp (^tK^{(o)}_\perp K_\perp - [^cK^{(o)}]^2) \quad \text{(B.14b, c)} \]

REFERENCES