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Slender-body approximations for electro-phoresis and electro-rotation of polarizable particles

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Slender-body asymptotic theory is used to evaluate the translational and rotational electrophoretic velocities of initially uncharged polarizable bodies of revolution. These velocities are obtained as asymptotic expansions in the small particle slenderness. Conducting particles which lack fore–aft symmetry translate parallel to the applied field direction, regardless of their orientation relative to it. Both conducting and dielectric particles tend to align with the field. The translational and rotational velocities of dielectric particles are asymptotically smaller than those of comparable conducting particles.

1. Introduction

Traditionally, the electrokinetic literature has focused upon flows around non-polarizable surfaces (Anderson 1989) in which the zeta potential is considered a physicochemical property of the solid–liquid interface. Following Levich (1962), however, a substantial body of work in the Soviet and post-Soviet literature has been dealing with flows around polarizable surfaces, in which the zeta potential is affected by the externally applied field. The prevailing mathematical models for such flows (Squires & Bazant 2004) employ the thin-Debye-layer limit and the small-zeta-potential model, and are typically applied for ideally polarizable (i.e. conducting) surfaces. The assumption of a thin Debye layer was relaxed by Yariv & Miloh (2008), and that of small-zeta-potentials by Yariv (2008).

The basic configuration which exhibits the essential features of induced-charge flows consists of an uncharged sphere in an unbounded fluid domain in which a uniform electric field is imposed at large distances (Gamayunov, Murtsovkin & Dukhin 1986). Since the highly symmetric flow pattern in that configuration does not result in particle motion, subsequent investigations considered more complicated 'symmetry-broken' geometries (Bazant & Squires 2004; Squires & Bazant 2004, 2006; Zhao & Bau 2007; Gangwal *et al.* 2008). Of special interest are non-spherical particles. Using qualitative arguments, Bazant & Squires (2004) suggested that such particles may undergo electrophoretic motion despite their net zero charge. The general problem of non-spherical particles was analysed by Yariv (2005) using tensorial symmetry arguments. It was shown that the electrical force and torque on the particle, obtained via integration of Maxwell stresses, are of the same scaling in the applied field as those produced by the electro-osmotic slip. (The existence of an electrical force on a zero-net-charge particle is discussed in Rivette & Baygents 1996 and Yariv 2006.) The first explicit solutions for non-spherical particles were provided by Squires & Bazant (2006) who employed regular perturbations to investigate slightly perturbed cylinders and spheres. It was found that such particles indeed undergo electrophoresis. Specifically, when the electric field coincides with the particle symmetry axis, the electrophoretic velocity is directed towards the 'bluff' end.

Subsequent papers addressed spheroidal particles. Using slender-body theory, Saintillan, Darve & Shaqfeh (2006*a*) analysed the electro-rotation of conducting spheroids, with the objective of modelling hydrodynamic interactions between rod-like particles (Saintillan, Shaqfeh & Darve 2006*b*). A general analysis of spheroidal particles was carried out by Yossifon, Frankel & Miloh (2007), making use of a Robin-type boundary condition which enabled the analysis of flows about dielectric non-spherical particles, the case of conductors emerging as a special case.

From the symmetry arguments of Yariv (2005) it follows that axisymmetric bodies which possess fore–aft symmetry – such as spheroids – cannot undergo induced-charge electrophoresis and therefore do not exhibit the full richness of symmetry-broken geometries (Bazant & Squires 2004). In this paper we use slender-body theory to investigate the general class of prolate axisymmetric particles (which may lack fore–aft symmetry), thereby extending the work of Saintillan *et al.* (2006*a*). Slender-body theory for arbitrary particle shapes has already been employed in the analysis of fixed-charge electrophoresis of high-aspect-ratio colloidal particles (Solomentsev & Anderson 1994); the present contribution presents a comparable investigation of induced-charge flows, wherein the newly derived Robin-type boundary condition (Yossifon *et al.* 2007) allows the analysis of both conducting and dielectric particles.

2. Problem formulation

Consider an initially uncharged particle of length $2a$ which may be either dielectric (permittivity ϵ_p) or conducting. It is suspended in an electrolyte solution (permittivity ϵ_f, viscosity η) and is exposed to a uniformly imposed electric field $\boldsymbol{E}_\infty = E_\infty \hat{\boldsymbol{E}}$, $\hat{\boldsymbol{E}}$ being a unit vector in the applied field direction. We assume the particle to be inert, with no chemical reactions occurring on its boundary s. Our interest lies in the hydrodynamic and electrical loads exerted upon the particle. For simplicity, we formulate below a stationary-particle problem; once the loads on such a particle are evaluated, we employ standard resistance relations (Happel & Brenner 1965) to extract the corresponding rectilinear and angular velocities of a comparable freely suspended particle.

We employ a dimensionless formulation in which length variables are normalized by a and the electric field by E_∞. The stress unit is chosen as $\epsilon_f E_\infty^2$; accordingly, forces and torques are respectively normalized by $\epsilon_f E_\infty^2 a^2$ and $\epsilon_f E_\infty^2 a^3$, while velocities and angular velocities are respectively normalized by $\epsilon_f E_\infty^2 a/\eta$ and $\epsilon_f E_\infty^2/\eta$. We focus upon the thin-Debye-layer limit, where the Debye thickness λ is small compared with a; the outer edge S of the Debye layer then coincides with s on the bulk scale. The electric potential in the bulk fluid, φ_f, is governed by Laplace's equation,

$$\nabla^2 \varphi_f = 0, \tag{2.1}$$

the no-flux condition on S,

$$\hat{\boldsymbol{n}} \cdot \nabla \varphi_f = 0 \tag{2.2}$$

(in which $\hat{\boldsymbol{n}}$ is a unit normal pointing into the fluid), and the far-field condition,

$$\nabla \varphi_f \to -\hat{\boldsymbol{E}}. \tag{2.3}$$

The asymptotic behaviour of φ_f at large distances from the particle can be written as a series of spherical harmonics (Batchelor 1967). In view of (2.2) the monopole term vanishes, whence the leading-order perturbation relative to the uniform field is represented by a dipole of magnitude $\boldsymbol{p}$:

$$\varphi_f \sim -\hat{\boldsymbol{E}} \cdot \boldsymbol{x} + \frac{1}{4\pi} \frac{\boldsymbol{p} \cdot \boldsymbol{x}}{|\boldsymbol{x}|^3} + \cdots . \tag{2.4}$$

The slip velocity on S is provided by the Helmholtz–Smoluchowski formula,

$$\boldsymbol{v} = \zeta \nabla \varphi_f, \tag{2.5}$$

in which the dimensionless 'zeta potential'

$$\zeta = \varphi_p|_s - \varphi_f|_S \tag{2.6}$$

depends upon the electric potential φ_p within the particle.

To evaluate φ_p we employ the common linear capacitor model for the Debye layer, valid for small zeta potentials (Squires & Bazant 2004). When the particle is dielectric, φ_p is also governed by Laplace's equation. The requisite matching between φ_p and φ_f is expressed via the Robin condition (Yossifon *et al.* 2007)

$$\varphi_p|_s + \alpha \hat{\boldsymbol{n}} \cdot \nabla \varphi_p|_s = \varphi_f|_S \tag{2.7}$$

wherein $\alpha = (\epsilon_p/\epsilon_f)(\lambda/a)$. In the limit $\alpha \to \infty$ of an ideally polarizable (i.e. conducting) particle, condition (2.7) yields $\hat{\boldsymbol{n}} \cdot \nabla \varphi_p|_s = 0$, which in conjunction with Laplace's equation implies the familiar requirement of a uniform particle potential. Superficially, the uniform value of φ_p may appear arbitrary, unrelated to the distribution of φ_f on S. The indeterminacy in the zeta potential (2.6) is resolved by imposing a global charge conservation constraint (Yariv 2005). Within the framework of a linear capacitor model, in which the surface charge is proportional to ζ, this condition is

$$\oint \mathrm{d}A\, \zeta = 0. \tag{2.8}$$

Note that when evaluated using (2.7) and (2.6), the divergence theorem readily shows that (2.8) is automatically satisfied for all finite α values.

The slip distribution (2.5), together with the Stokes equations and the requirement for velocity attenuation at large distances, serves to uniquely determine the flow field about a stationary particle. This field, in turn, exerts a force $\boldsymbol{F}$ and a torque $\boldsymbol{G}$ on the particle. Following the tensorial arguments of Yariv (2005), these hydrodynamic loads must possess the form

$$\boldsymbol{F} = \mathsf{\boldsymbol{F}} : \hat{\boldsymbol{E}} \hat{\boldsymbol{E}}, \quad \boldsymbol{G} = \mathsf{\boldsymbol{G}} : \hat{\boldsymbol{E}} \hat{\boldsymbol{E}}, \tag{2.9}$$

in which $\mathsf{\boldsymbol{F}}$ is a third-order tensor and $\mathsf{\boldsymbol{G}}$ a third-order pseudo-tensor. These dimensionless coefficients may depend upon α, the only physical parameter appearing in the dimensionless problem formulation, as well as the particle geometry.

In addition to the hydrodynamic loads, the particle also experiences electrical loads, which are obtained in principle as surface quadratures of Maxwell stresses. For a particle in an unbounded fluid domain, the far-field behaviour (2.4) implies (Rivette & Baygents 1996) that the electric force vanishes and the electric torque is

$$\boldsymbol{G}_E = \boldsymbol{p} \times \hat{\boldsymbol{E}}. \tag{2.10}$$

If the particle is freely suspended, it will acquire rectilinear and angular velocities which ensure it remains force- and torque-free. As in (2.9), these velocities have the

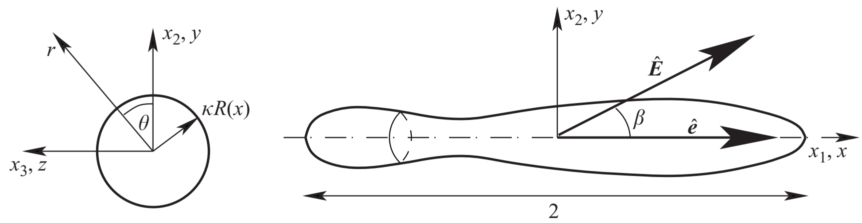

FIGURE 1. Schematic.

form

$$\boldsymbol{U} = \mathsf{U} : \hat{\boldsymbol{E}}\hat{\boldsymbol{E}}, \quad \boldsymbol{\Omega} = \mathsf{W} : \hat{\boldsymbol{E}}\hat{\boldsymbol{E}}, \tag{2.11}$$

in which the third-order tensor U and the third-order pseudo-tensor W are functions of α and the particle geometry.

3. Bodies of revolution

We focus upon bodies of revolution whose symmetry axis is inclined at an angle β relative to $\hat{\boldsymbol{E}}$, see figure 1. In a particle-fixed cylindrical coordinate system (r, θ, x) the particle boundary is given by

$$r = \kappa R(x), \quad -1 < x < 1, \tag{3.1}$$

in which the shape function $R(x)$ satisfies

$$R(\pm 1) = 0. \tag{3.2}$$

With no loss of generality we also assume $R = O(1)$. The parameter κ then represents the particle slenderness; it is uniquely set by imposing a normalization condition, say

$$\int_{-1}^{1} R(x)\,\mathrm{d}x = 2. \tag{3.3}$$

The only fixed vector appearing in the geometric specification of the particle is $\hat{\boldsymbol{e}}$, a particle-fixed unit vector attached to the symmetry axis. In view of their contraction with $\hat{\boldsymbol{E}}\hat{\boldsymbol{E}}$, the most general form of the tensors F and G is (Yariv 2005)

$$\mathsf{F} = f\mathsf{I}\hat{\boldsymbol{e}} + f'\hat{\boldsymbol{e}}\hat{\boldsymbol{e}}\hat{\boldsymbol{e}} + f''\hat{\boldsymbol{e}}\mathsf{I}, \quad \mathsf{G} = -g\boldsymbol{\epsilon}\cdot\hat{\boldsymbol{e}}\hat{\boldsymbol{e}}. \tag{3.4}$$

Here, I is the idemfactor and $\boldsymbol{\epsilon}$ is the alternating third-order pseudo-tensor. The four scalar coefficients appearing in (3.4) may depend upon κ and the function $R(x)$, but not upon β. Equations (2.9) and (3.4) imply that the vector $\boldsymbol{F}$ is spanned by $\hat{\boldsymbol{E}}$ and $\hat{\boldsymbol{e}}$, whereas the pseudo-vector

$$\boldsymbol{G} = g(\hat{\boldsymbol{e}} \times \hat{\boldsymbol{E}})(\hat{\boldsymbol{e}} \cdot \hat{\boldsymbol{E}}) \tag{3.5}$$

is perpendicular to both. It is convenient to define a particle-fixed Cartesian system, $(x_1, x_2, x_3) = (x, y, z)$, where the x_2-axis lies in the $\hat{\boldsymbol{E}}$–$\hat{\boldsymbol{e}}$ plane (see figure 1). The corresponding unit vectors are

$$\hat{\boldsymbol{e}}_1 = \hat{\boldsymbol{e}}, \quad \hat{\boldsymbol{e}}_2 = (\hat{\boldsymbol{E}} - \hat{\boldsymbol{e}}\cos\beta)/\sin\beta, \quad \hat{\boldsymbol{e}}_3 = \hat{\boldsymbol{e}} \times \hat{\boldsymbol{E}}/\sin\beta. \tag{3.6}$$

Substitution into (3.5) then yields $\boldsymbol{G} = \hat{\boldsymbol{e}}_3 G$, in which

$$G = g\sin\beta\cos\beta. \tag{3.7}$$

The same symmetry relations hold for the electrical torque, which is therefore of the form $\boldsymbol{G}_E = \hat{\boldsymbol{e}}_3 G_E$.

Similar argument also apply for the velocities of a freely suspended particle. Thus, the tensorial coefficients in (2.11) adopt the form

$$\boldsymbol{U} = u\boldsymbol{I}\hat{\boldsymbol{e}} + u'\hat{\boldsymbol{e}}\hat{\boldsymbol{e}}\hat{\boldsymbol{e}} + u''\hat{\boldsymbol{e}}\boldsymbol{I}, \quad \boldsymbol{W} = -\omega\boldsymbol{\epsilon}\cdot\hat{\boldsymbol{e}}\hat{\boldsymbol{e}}, \tag{3.8}$$

where the four scalar coefficients are independent of β. The particle velocity $\boldsymbol{U}$ is spanned by $\hat{\boldsymbol{E}}$ and $\hat{\boldsymbol{e}}$, $\boldsymbol{U} = \hat{\boldsymbol{e}}_1 U_1 + \hat{\boldsymbol{e}}_2 U_2$, and the angular velocity is perpendicular to both vectors, $\boldsymbol{\varOmega} = \hat{\boldsymbol{e}}_3 \varOmega$, wherein

$$\varOmega = \omega \sin\beta \cos\beta. \tag{3.9}$$

Note that positive ω values represent angular motion which tends to align the particle with the field.

4. Slender bodies

We analyse here the asymptotic limit of a slender particle, $\kappa \ll 1$. The focus upon this limit implies an important difference between the present analysis and that of Yossifon *et al.* (2007), where spheroids of arbitrary aspect ratio were analysed: since an asymptotic expansion in κ implicitly assumes that all other parameters are $O(1)$, it is not *a priori* guaranteed that the case of a conducting particle can be obtained by extracting the $\alpha \to \infty$ limit of the resulting expressions. It will become evident (see e.g. (5.25)) that the limits $\kappa \to 0$ and $\alpha \to \infty$ do not commute. Accordingly, the case of a conducting particle must be analysed separately (using the consistency condition (2.8)).

The slender limit is handled by matched asymptotic expansions (Cole 1968; Cox 1970). In principle, all equations in the fluid domain are solved separately in an 'outer' region, characterized by the particle length, and an 'inner' region, characterized by the $O(\kappa)$ cross-sectional dimension. The latter region is naturally handled using the stretched radial coordinate $\rho = r/\kappa$.

The solution of the Neumann-type boundary-value problem (2.1)–(2.3) governing φ_f is available in the literature (Cole 1968). The outer and inner expansions are

$$\varphi_f \sim -x\cos\beta - r\sin\beta\cos\theta + O(\kappa^2) \quad \text{for} \quad \kappa \to 0, \quad r \text{ fixed}, \tag{4.1}$$

$$\varphi_f \sim -x\cos\beta - \kappa(\rho + R^2/\rho)\sin\beta\cos\theta + O(\kappa^2\log\kappa) \quad \text{for} \quad \kappa \to 0, \quad \rho \text{ fixed}. \tag{4.2}$$

The latter is used to evaluate the electric field on S:

$$\nabla\varphi_f|_S \approx 2\hat{\boldsymbol{e}}_\theta \sin\beta\sin\theta - \hat{\boldsymbol{e}}_x\cos\beta. \tag{4.3}$$

Hereafter, the approximation symbol '$\approx$' implies that the asymptotic error term is of an algebraically small relative magnitude.

The electric potential within the particle, φ_p, is naturally evaluated using the inner variables. For a conducting particle, φ_p is uniform and set by the integral condition (2.8). Substitution of (4.2) into (2.8) in conjunction with (3.3) yields $\varphi_p \approx \mathscr{K}\cos\beta$, in which

$$\mathscr{K} = -\frac{1}{2}\int_{-1}^{1} xR(x)\,\mathrm{d}x \tag{4.4}$$

is a geometric first moment (which vanishes for fore–aft symmetric particles). Combining with (4.2) we find

$$\zeta \approx (x + \mathscr{K})\cos\beta. \tag{4.5}$$

For a dielectric particle, it is necessary to solve a boundary-value problem which consists of the differential equation,

$$\frac{1}{\rho}\frac{\partial}{\partial\rho}\left(\rho\frac{\partial\varphi_p}{\partial\rho}\right)+\frac{1}{\rho^2}\frac{\partial^2\varphi_p}{\partial\theta^2}+\kappa^2\frac{\partial^2\varphi_p}{\partial x^2}=0, \tag{4.6}$$

the Robin condition (cf. (2.7))

$$\kappa\varphi_p+\alpha\left[\frac{\partial\varphi_p}{\partial\rho}-\kappa^2\frac{\mathrm{d}R}{\mathrm{d}x}\frac{\partial\varphi_p}{\partial x}\right][1+O(\kappa^2)]=\kappa\varphi_f \quad \text{at} \quad \rho=R(x), \tag{4.7}$$

and the requirement of regularity on the symmetry axis $\rho=0$. Straightforward calculation yields

$$\varphi_p\sim -x\cos\beta-\kappa\alpha\frac{\mathrm{d}R}{\mathrm{d}x}\cos\beta+O(\kappa^2). \tag{4.8}$$

(Note that this expansion breaks down near rounded particle ends, where $\mathrm{d}R/\mathrm{d}x$ diverges. Our interest here, however, is in global quantities such as forces and torques, obtained from integration over the entire x-domain; it is easily verified (see (5.24)–(5.25)) that such end singularities are integrable.) Combining (4.8) with (4.2) we therefore obtain

$$\zeta\approx\kappa\left[2R(x)\sin\beta\cos\theta-\alpha\frac{\mathrm{d}R}{\mathrm{d}x}\cos\beta\right]. \tag{4.9}$$

Note the order of magnitude difference in the zeta potential (and consequent velocity slip) between the conducting (ideal polarizabilty) case (4.5) and the dielectric (finite polarizabilty) case (4.9).

5. The electrophoretic motion of a freely suspended particle

With the slip distribution provided by (2.5), it is possible in principle to evaluate the velocity field $\boldsymbol{v}$ and the corresponding stress field $\boldsymbol{\sigma}$, which can be integrated to yield the hydrodynamic loads acting on a stationary particle. Here, following Yariv (2005) and Saintillan *et al.* (2006*a*), we avoid this procedure by exploiting a method due to Brenner (1964) which utilizes the Lorentz reciprocal relation (Happel & Brenner 1965),

$$\oint_S \mathrm{d}A\,\hat{\boldsymbol{n}}\cdot\boldsymbol{\sigma}\cdot\bar{\boldsymbol{v}}=\oint_S \mathrm{d}A\,\hat{\boldsymbol{n}}\cdot\bar{\boldsymbol{\sigma}}\cdot\boldsymbol{v}, \tag{5.1}$$

in which $\bar{\boldsymbol{v}}$ is any flow field that satisfies the Stokes equations and $\bar{\boldsymbol{\sigma}}$ the corresponding stress field. The requisite loads are obtained by choosing $\bar{\boldsymbol{v}}$ as the velocity field which corresponds to a fictitious translation or rotation of the particle. This method is natural for slender bodies (Solomentsev & Anderson 1994; Saintillan *et al.* 2006*a*) because solutions of the Stokes equations which correspond to their rigid-body motion are already available (Cox 1970).

To find the ith component F_i $(i=1,2)$ of the hydrodynamic force we choose $\bar{\boldsymbol{v}}$ as the velocity field due to a translation of a particle with a unit velocity in the x_i-direction, obtaining

$$F_i=\oint_S \mathrm{d}A\,\hat{\boldsymbol{n}}\cdot\bar{\boldsymbol{\sigma}}\cdot\zeta\nabla\varphi_f. \tag{5.2}$$

The solution of the Stokes equations for particle translation appear in Cox (1970). For a longitudinal translation along the x_1-axis the results of Cox (1970) yield the traction

$$\hat{\boldsymbol{n}} \cdot \bar{\boldsymbol{\sigma}} \approx \frac{\bar{\mathscr{F}}_1}{2\pi\kappa R}\hat{\boldsymbol{e}}_x. \tag{5.3}$$

Here, $\bar{\mathscr{F}}_1$ is the force per unit length in the x_1-direction which is exerted upon the translating particle. It was evaluated by Cox (1970) as an asymptotic series in $1/\log\kappa$:

$$\bar{\mathscr{F}}_1(x)/2\pi \sim \frac{1}{\log\kappa} + \frac{1}{\log^2\kappa}\left(\log 2 - \frac{1}{2} + \frac{1}{2}\log\frac{1-x^2}{R^2(x)}\right) + O(\log^{-3}\kappa). \tag{5.4}$$

For a lateral translation along the x_2-axis the results of Cox (1970) yield the traction

$$\hat{\boldsymbol{n}} \cdot \bar{\boldsymbol{\sigma}} \approx \frac{\bar{\mathscr{F}}_2}{2\pi\kappa R}(\hat{\boldsymbol{e}}_\rho\cos\theta - \hat{\boldsymbol{e}}_\theta\sin\theta). \tag{5.5}$$

Here, $\bar{\mathscr{F}}_2$ is the force per unit length in the x_2-direction which is exerted upon the translating particle:

$$\bar{\mathscr{F}}_2(x)/2\pi \sim \frac{2}{\log\kappa} + \frac{2}{\log^2\kappa}\left(\log 2 + \frac{1}{2} + \frac{1}{2}\log\frac{1-x^2}{R^2(x)}\right) + O(\log^{-3}\kappa). \tag{5.6}$$

To obtain G we choose $\bar{\boldsymbol{v}}$ as the velocity field due to a fictitious rotation of a comparable particle with a unit angular velocity in the x_3-direction. Substituting $\bar{\boldsymbol{v}}|_S = \hat{\boldsymbol{e}}_3 \times \boldsymbol{x}$ into (5.1) yields

$$G = \oint_S \mathrm{d}A\,\hat{\boldsymbol{n}} \cdot \bar{\boldsymbol{\sigma}} \cdot \zeta\nabla\varphi_f. \tag{5.7}$$

The field $\bar{\boldsymbol{v}}$ which corresponds to rigid-body rotation was calculated using the method of Cox (1970). The traction is of the form (5.5),

$$\hat{\boldsymbol{n}} \cdot \bar{\boldsymbol{\sigma}} \approx \frac{\bar{\mathscr{F}}_2^{\mathrm{rot}}}{2\pi\kappa R}(\hat{\boldsymbol{e}}_\rho\cos\theta - \hat{\boldsymbol{e}}_\theta\sin\theta), \tag{5.8}$$

but now with the force density

$$\bar{\mathscr{F}}_2^{\mathrm{rot}}(x)/2\pi \sim \frac{2x}{\log\kappa} + \frac{2x}{\log^2\kappa}\left(\log 2 - \frac{1}{2} + \frac{1}{2}\log\frac{1-x^2}{R^2(x)}\right) + O\left(\log^{-3}\kappa\right). \tag{5.9}$$

Once the loads are calculated, the rectilinear and angular velocities of a comparable freely suspended particle are obtained from the force- and torque-free conditions:

$$F_1 + U_1\bar{F}_1 = 0, \quad F_2 + U_2\bar{F}_2 + \Omega\bar{C} = 0, \quad G + \Omega\bar{G} + U_2\bar{C} + G_E = 0. \tag{5.10a,b,c}$$

Here, $\bar{F}_1$ and $\bar{F}_2$ are the forces associated with a unit-velocity translation in the x_1- and x_2-directions, respectively; $\bar{G}$ is the torque associated with a unit-velocity rotation in the x_3-direction; and the coupling term $\bar{C}$ represents the torque in the x_3-direction due to a unit-velocity translation in the x_2-direction, or, equivalently (Happel & Brenner 1965), the force in the x_2-direction due to a unit-velocity rotation in the x_3-direction. Note that (2.4), (2.10), and (4.1) imply that G_E is $O(\kappa^2)$; in what follows it will become evident that it is algebraically small compared with G for both conducting and dielectric particles.

The forces $\bar{F}_1$ and $\bar{F}_2$ are respectively obtained from integration of (5.4) and (5.6). To leading order:

$$\bar{F}_1 \sim \frac{4\pi}{\log\kappa} + O(\log^{-2}\kappa), \quad \bar{F}_2 \sim \frac{8\pi}{\log\kappa} + O(\log^{-2}\kappa). \tag{5.11}$$

By definition, the torque $\bar{G}$ is provided by the integral

$$\bar{G} = \int_{-1}^{1} x \bar{\mathscr{F}}_2^{\mathrm{rot}}(x)\,\mathrm{d}x; \tag{5.12}$$

substitution of (5.9) yields:

$$\frac{3\bar{G}}{8\pi} \sim \frac{1}{\log\kappa} + \frac{1}{\log^2\kappa}\left[\frac{3}{4}\int_{-1}^{1} x^2 \log\frac{1-x^2}{R^2(x)}\,\mathrm{d}x - \frac{1}{2} + \log 2\right] + O(\log^{-3}\kappa). \tag{5.13}$$

It is evident from (5.6) (or, alternatively, from (5.9)) that, to leading-order, $\bar{C} \sim 2\pi\mathscr{L}/\log^2\kappa$, wherein

$$\mathscr{L} = \int_{-1}^{1} x \log\frac{1-x^2}{R^2(x)}\,\mathrm{d}x \tag{5.14}$$

is a geometry-specific coefficient that vanishes for fore–aft symmetric particles.

5.1. *Conducting particles*

Consider first the case of a conducting particle, in which the zeta potential is provided by (4.5). Substitution into (5.2) of (5.3)–(5.4) yields F_1, whereas substitution of (5.5)–(5.6) yields F_2. We therefore obtain

$$F_1 \sim -\frac{4\pi}{\log\kappa}\mathscr{K}\cos^2\beta, \quad F_2 \sim -\frac{8\pi}{\log\kappa}\mathscr{K}\sin\beta\cos\beta, \tag{5.15}$$

with an $O(1/\log\kappa)$ relative asymptotic error. As required, both expressions vanish for fore–aft symmetric particles.

Substitution of (5.8) into (5.7) yields

$$G \approx -\sin\beta\cos\beta \int_{-1}^{1} (x+\mathscr{K})\bar{\mathscr{F}}_2^{\mathrm{rot}}(x)\,\mathrm{d}x. \tag{5.16}$$

Use of (5.9) then furnishes an asymptotic series in $1/\log\kappa$. To leading order:

$$G \sim -\frac{8\pi}{3\log\kappa}\sin\beta\cos\beta + O(\log^{-2}\kappa). \tag{5.17}$$

Consider now a freely suspended particle. When expressing the particle velocities as asymptotic series in $1/\log\kappa$, it is evident from (5.10) that they are $O(1)$. To leading-order, the coupling term does not affect the force- and torque-free balances, and we find, with an $O(\log^{-1}\kappa)$ asymptotic error, that

$$U_1 \sim \mathscr{K}\cos^2\beta, \quad U_2 \sim \mathscr{K}\sin\beta\cos\beta, \tag{5.18}$$

$$\Omega \sim \sin\beta\cos\beta, \quad \text{or, equivalently,} \quad \omega \sim 1. \tag{5.19}$$

The result (5.19) was obtained, to the same accuracy, for a slender ellipsoid by Saintillan *et al.* (2006*a*). Here it is shown to hold for any slender shape. Moreover, if the particle is also fore–aft symmetric, whereby $\mathscr{K}=0$, (5.16) reads $G \approx -\bar{G}\sin\beta\cos\beta$ (cf. (5.12)); With $\bar{C}$ vanishing for such bodies (Happel & Brenner 1965) we find that (5.19) holds to any order in the $1/\log\kappa$ expansion.

Use of (3.6) shows that (5.18) is equivalent to the invariant representation

$$\boldsymbol{U} \sim \mathscr{K}(\hat{\boldsymbol{e}}\cdot\hat{\boldsymbol{E}})\hat{\boldsymbol{E}} + O(\log^{-1}\kappa). \tag{5.20}$$

Remarkably, the particle translates along the straight field-lines of the applied field regardless of its orientation relative to them (the speed, however, is orientation dependent). In terms of the representation (3.8), we find that u' and u'' vanish to leading order, while $u \sim \mathscr{K}$. From (4.4) is it readily seen that $\mathscr{K}$ is positive for particles which 'point' in the positive x-direction. For example, for a cone† with $\hat{\boldsymbol{e}}$ pointing toward the apex, $R(x) = 1 - x$, we obtain from (4.4) $\mathscr{K} = 1/3$. The positive sign of u agrees with the qualitative prediction of Bazant & Squires (2004). It is easily explained for the case of a particle that points into the field ($\beta = 0$). Then, the flow about it is mathematically equivalent to that in fixed-charge electrophoresis with constant zeta potential (namely $\mathscr{K}$) under the action of a uniform slip in the negative-x direction. Such a leading-order mechanism is absent for non-slender shapes; indeed, Squires & Bazant (2006) predicted negative values of u for near spheres.

5.2. *Dielectric particles*

The preceding analysis can be repeated for a dielectric particle, where (4.9) replaces (4.5). When using (5.2) to evaluate the hydrodynamic forces, (3.2) now implies that the $O(1/\log\kappa)$ contributions from (5.4) and (5.6) vanish upon integration. Thus, we find that

$$F_1 \sim \pi\alpha\mathscr{M}\frac{\kappa}{\log^2\kappa}\cos^2\beta, \quad F_2 \sim 2\pi\alpha\mathscr{M}\frac{\kappa}{\log^2\kappa}\sin\beta\cos\beta \tag{5.21}$$

with an $O(1/\log\kappa)$ relative asymptotic error. Here

$$\mathscr{M} = \int_{-1}^{1}\frac{\mathrm{d}R}{\mathrm{d}x}\log\frac{1-x^2}{R^2(x)}\,\mathrm{d}x \tag{5.22}$$

is a shape-dependent coefficient (that, as required, vanishes for fore–aft symmetric particles). When using (5.7)–(5.8) to evaluate the hydrodynamic torque, we find

$$G \approx \kappa\alpha\sin\beta\cos\beta\int_{-1}^{1}\frac{\mathrm{d}R}{\mathrm{d}x}\bar{\mathscr{F}}_2^{\mathrm{rot}}(x)\,\mathrm{d}x. \tag{5.23}$$

Substitution of (5.9) followed by integration by parts in conjunction with (3.3) provides an asymptotic expansion of G. Use of the representation (3.7) then yields:

$$\frac{g}{8\pi\kappa\alpha} \sim -\frac{1}{\log\kappa} + \frac{1}{\log^2\kappa}\left[\frac{1}{4}\int_{-1}^{1}x\frac{\mathrm{d}R}{\mathrm{d}x}\log\frac{1-x^2}{R^2(x)}\,\mathrm{d}x + \frac{1}{2} - \log 2\right] + O(\log^{-3}\kappa). \tag{5.24}$$

Considering again the force- and torque-free conditions (5.10), we find that the angular velocity is $O(\kappa)$, whereas the logarithmically small forces render the rectilinear velocities $O(\kappa/\log\kappa)$. Thus, the angular velocity can be evaluated from (5.10 *c*), (5.13), and (5.24) to two-term accuracy in the $1/\log\kappa$ expansion without any coupling effect. Upon making use of the representation (3.9) we find:

$$\omega \sim 3\alpha\kappa\left[1 + \frac{1}{4\log(1/\kappa)}\int_{-1}^{1}\left[3x^2 + x\frac{\mathrm{d}R}{\mathrm{d}x}\right]\log\frac{1-x^2}{R^2}\mathrm{d}x + O(\log^{-2}\kappa)\right]. \tag{5.25}$$

Note that the integral in the above formula vanishes for spheroids, where $R(x) = (4/\pi)\sqrt{1-x^2}$ (cf. (3.3)).

From (5.21) we see that F_2 is twice as large as F_1, just as in the conducting case (cf. (5.15)). It is however evident from (5.10 *b*) that the leading-order lateral motion is coupled to the particle rotation; thus, the velocity of a freely suspended dielectric

† Superficially, it may appear that (3.2) renders the cone an inappropriate shape for the present formulation; it is easily verified, however, that the discontinuity in R at $x = -1$ constitutes a weak singularity.

particle is not directed parallel to the field. Indeed, straightforward calculation yields here (cf. (5.20))

$$\boldsymbol{U} \sim \frac{\alpha\kappa}{4\log(1/\kappa)}[(3\mathscr{L}+\mathscr{M})(\hat{\boldsymbol{e}}\cdot\hat{\boldsymbol{E}})\hat{\boldsymbol{E}} - 3\mathscr{L}(\hat{\boldsymbol{e}}\cdot\hat{\boldsymbol{E}})^2\hat{\boldsymbol{e}}] + O\left(\frac{\kappa}{\log^2\kappa}\right). \tag{5.26}$$

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