PHYSICAL REVIEW FLUIDS **2**, 012201(R) (2017)

Phoretic drag reduction of chemically active homogeneous spheres under force fields and shear flows

Ehud Yariv* and Uri Kaynan
Department of Mathematics, Technion–Israel Institute of Technology, Haifa 32000, Israel
(Received 12 December 2016; published 31 January 2017)

Surrounded by a spherically symmetric solute cloud, chemically active homogeneous spheres do not undergo conventional autophoresis when suspended in an unbounded liquid domain. When exposed to external flows, solute advection deforms that cloud, resulting in a generally asymmetric distribution of diffusio-osmotic slip which, in turn, modifies particle motion. Inspired by classical forced-convection analyses [Acrivos and Taylor, Phys. Fluids **5**, 387 (1962); Frankel and Acrivos, Phys. Fluids **11**, 1913 (1968)] we illustrate this phoretic phenomenon using two prototypic configurations, one where the particle sediments under a uniform force field and one where it is subject to a simple shear flow. In addition to the Péclet number Pe associated with the imposed flow, the governing nonlinear problem also depends upon α, the intrinsic Péclet number associated with the chemical activity of the particle. As in the forced-convection problems, the small-Péclet-number limit is nonuniform, breaking down at large distances away from the particle. Calculation of the leading-order autophoretic effects thus requires use of matched asymptotic expansions, the outer region being at distances that scale inversely with Pe and $\mathrm{Pe}^{1/2}$ in the respective sedimentation and shear problems. In the sedimentation problem we find an effective drag reduction of fractional amount $\alpha/8$; in the shear problem we find that the magnitude of the stresslet is decreased by a fractional amount $\alpha/4$. For a dilute particle suspension the latter result is manifested by a reduction of the effective viscosity.

DOI: 10.1103/PhysRevFluids.2.012201

I. INTRODUCTION

There is presently a great interest in autophoretic phenomena involving catalytically active colloids. These are commonly analyzed using a macroscale description wherein an interfacial chemical reaction is modeled via a prescribed distribution of solute flux [1]. Prevailing analyses of autophoresis tend to consider Janus-type spherical particles where an asymmetric prescribed-flux distribution sets the self-propulsion direction.

The interaction of phoretic swimmers with externally imposed flows result in rich phenomena such as gravitaxis and polar order under gravity [2] and upstream swimming under shear [3]. In this paper we consider a different mode of interaction, driven by solute advection, between imposed flows and *isotropic* particles, which do not undergo conventional self-propulsion [4]. In particular, our interest lies in the effective drag reduction (in the broad sense) of chemically homogeneous spheres. At zero Péclet numbers, an imposed flow does not affect the spherically symmetric solute cloud surrounding such particles; conversely, that cloud does not result in diffusio-osmosis. At nonzero Péclet numbers, however, solute advection deforms the cloud; with the resulting distribution of diffusio-osmosis being generally asymmetric, the engendered phoretic flow modifies particle motion.

The mechanism considered herein resembles classical forced convection in the context of heat and mass transport. Indeed, following the classical work of Acrivos and coworkers [5,6] we consider two prototypic problems: one where the particle sediments under a uniform force field and one where it rotates under simple shear. There is a fundamental difference between the problems considered herein, where solute gradients generate fluid motion, and those of forced convection, where the

*udi@technion.ac.il

transported quantity (heat, mass) typically does not affect the flow. The mutual coupling between solute transport and flow in the present context has two consequences. The first is that the problem is inherently nonlinear. The second is that solute convection leads to mechanical effects, absent in the classical work of Acrivos and coworkers. (In that sense, the present contribution also differs from recent analyses of nutrient uptake by microorganisms [7].) Thus, while the forced-convection literature is typically concerned with the calculation of the Nusselt number, our interest here is in the effect of convection upon the settling velocity in the sedimentation problem and the rotation velocity and stresslet in the shear problem.

II. GENERIC FORMULATION

A. Physical problem

A chemically active spherical particle of radius a is suspended in an unbounded liquid domain. The chemical activity is assumed homogeneous, described by a uniform solute flux $\mathcal{A}$ (defined positively outwards) at the particle boundary. This prescribed flux results in the formation of a diffuse solute cloud in the vicinity of the particle with concentration of order $\mathcal{C} = a\mathcal{A}/D$, D being the solute diffusivity. We assume that the range λ of the solute-solid interaction is small compared with a. In the standard macroscale description [8] this interaction is manifested as diffusio-osmosis. The liquid thus effectively slips along the solid surface with a velocity which is proportional to the surface gradient of solute concentration, where the proportionality coefficient, $\mathcal{M}$, is of order $kT\lambda^2/\eta$ (kT being the Boltzmann temperature and η the solvent viscosity); this condition provides the phoretic velocity scale $\mathcal{U} = \mathcal{M}\mathcal{C}/a$.

When freely suspended in a free solution, and in the absence of any external loads, the particle remains stationary; chemical reaction at its surface merely maintains the spherically symmetric solute cloud. Our interest is in the modification to particle motion resulting from forced convection. We denote the characteristic magnitude of the imposed flow by $\mathcal{W}$. The relative importance of convection is accordingly represented by the Péclet number $\mathrm{Pe} = a\mathcal{W}/D$. We note that an additional Péclet number, $\alpha = a\mathcal{U}/D$, may be formed using the phoretic velocity scale. Unlike Pe, α is an intrinsic property of the particle-solution system.

B. Dimensionless formulation

Normalizing length variables by a, we employ both Cartesian coordinates (x,y,z) and spherical-polar coordinates (r,θ,ϕ), with the origin at the particle center and the latitudinal angle θ measured from the z axis. The solute concentration c (normalized by $\mathcal{C}$) is governed by (1) the advection-diffusion equation,

$$\nabla^2 c = \mathrm{Pe}\,\mathbf{u}\cdot\nabla c, \tag{1}$$

where $\mathbf{u}$ is the fluid velocity (normalized by $\mathcal{W}$); (2) the imposed activity condition,

$$\frac{\partial c}{\partial r} = -1 \quad \text{at} \quad r = 1; \tag{2}$$

and (3) far-field decay,

$$c \to 0 \quad \text{as} \quad r \to \infty. \tag{3}$$

The flow is governed by (1) the continuity and Stokes equations,

$$\nabla\cdot\mathbf{u} = 0, \quad \nabla^2\mathbf{u} = \nabla p, \tag{4}$$

where the pressure p is normalized by $\eta\mathcal{W}/a$; (2) the imposed velocity slip $(\mathcal{U}/\mathcal{W})\nabla_s c$ relative to the particle boundary, where ∇_s is the surface-gradient operator; and (3) appropriate far-field and integral conditions, which represent the specific mode of forced convection considered.

The solute concentration at zero Péclet number, c_0, is unaffected by the flow. It accordingly possesses the spherically symmetric profile

$$c_0 = \frac{1}{r}. \tag{5}$$

Conversely, this profile does not engender any slip; the fluid velocity is therefore unaffected by the chemical activity. At nonzero values of Pe, however, the solute-transport and flow problems are nonlinearly coupled. This is illustrated below using two concrete problems.

III. SEDIMENTATION

Consider the case where the particle is subject to an external force (e.g., gravity) of magnitude F. For Pe $= 0$, where the solute-transport and flow problems are uncoupled, the particle simply sediments with the dimensional speed $\mathcal{W} = F/6\pi\eta a$ in the force direction. Our interest lies in the settling velocity at nonzero Pe.

We here choose the z axis in the applied-force direction. Because of the obvious axial symmetry the fluid velocity adopts the form $\mathbf{u} = u\hat{\mathbf{e}}_r + v\hat{\mathbf{e}}_\theta$. All pertinent scalar variables, as well as the velocity components u and v, are independent of the azimuthal angle ϕ. The requirement that the particle is torque-free is trivially satisfied provided it does not rotate. The particle is accordingly stationary in the comoving reference frame to which the coordinates are attached. In that frame the slip condition reads

$$\mathbf{u} = \frac{\mathcal{U}}{\mathcal{W}}\nabla_s c \quad \text{at} \quad r = 1. \tag{6}$$

This condition is supplemented by the far-field approach to a uniform stream,

$$\mathbf{u} \to -W\hat{\mathbf{e}}_z, \tag{7}$$

wherein W is the particle velocity (normalized by $\mathcal{W}$) relative to the fluid. This quantity is set by the integral condition,

$$\text{hydrodynamic force on the particle in } z \text{ direction} = -6\pi, \tag{8}$$

the force being normalized by $\eta a\mathcal{W}$.

Roughly speaking, the streaming flow is expected to deflect the solute cloud in the negative z direction relative to the particle. With diffusio-osmosis acting along solute-concentration gradients, this deflection is accompanied by a slip distribution which is biased in the same direction. Such a distribution, in turn, imparts the freely suspended particle with a velocity in the positive z direction, increasing W. Solute convection is accordingly expected to effectively reduce the particle drag.

There are two parameters in the governing dimensionless problem, namely, Pe and $\mathcal{U}/\mathcal{W}$. In what follows we affirm the above drag-reduction conjecture by considering the asymptotic limit of small Péclet numbers, assuming $\mathcal{U}/\mathcal{W} = O(1)$.

A. Small Pe

At zero Péclet numbers the solute concentration is given by (5). With the flow problem being uncoupled to solute transport, constraint (8) implies that the particle moves at a unit velocity in the z direction relative to otherwise quiescent fluid, $W_0 = 1$. The associated velocity field in the body-fixed reference system is well known [5], namely,

$$u_0 = -\left(1 - \frac{3}{2r} + \frac{1}{2r^3}\right)\mu, \quad v_0 = \left(1 - \frac{3}{4r} - \frac{1}{4r^3}\right)(1-\mu^2)^{1/2}, \tag{9}$$

where $\mu = \cos\theta$.

We consider the asymptotic limit $\mathrm{Pe} \ll 1$, perturbing about the above. Toward that end, we employ the generic expansion

$$f \sim f_0 + \mathrm{Pe}\, f_1 + \cdots, \tag{10}$$

our goal being the calculation of the velocity perturbation W_1.

We consider first the concentration perturbation c_1. It is governed by the Poisson equation

$$\nabla^2 c_1 = \mathbf{u}_0 \cdot \nabla c_0, \tag{11}$$

the Neumann condition [see (2)],

$$\frac{\partial c_1}{\partial r} = 0 \quad \text{at} \quad r = 1, \tag{12}$$

and the requirement of far-field decay,

$$c_1 \to 0 \quad \text{as} \quad r \to \infty. \tag{13}$$

It is well known [5] that the preceding problem has no solution. Indeed, the particular integral of (11)

$$c_{1p} = -\left(\frac{1}{2} - \frac{3}{4r} - \frac{1}{8r^3}\right)\mu \tag{14}$$

includes a term that does not vanish at large r. This failure to satisfy (13) cannot be remedied by any choice of the complementary solution of (11). As explained by Acrivos and Taylor [5], the source of this incompatibility is the slow algebraic decay, as $1/r$, of c. Thus, while at large r the diffusive term in Eq. (1) is $O(1/r^3)$, the advective term there is $O(\mathrm{Pe}/r^2)$, entering the leading-order balance at $r = O(1/\mathrm{Pe})$. This nonuniformity calls for the use of matched asymptotic expansions, with the inner and outer region describing $O(1)$ and $O(1/\mathrm{Pe})$ distances from the origin, respectively.

We note that the preceding expressions for c_0 and $\mathbf{u}_0$, obtained by what is now viewed as a naïve calculation, are still valid. On the other hand, expression (14) is now understood to hold strictly at the inner region, where the decay condition (14) is abandoned in favor of asymptotic matching with the outer solution, considered next.

B. Outer region

It is convenient to respectively employ the inner and outer positions vectors, $\mathbf{x} = (x, y, z)$ and $\mathbf{X} = (X, Y, Z)$, the latter defined as $\mathbf{X} = \mathrm{Pe}\,\mathbf{x}$. As it is evident from (5) that c is $O(\mathrm{Pe})$ in the outer region, we define the outer concentration $C = \mathrm{Pe}^{-1} c$, which presumably possesses the asymptotic expansion (10). With the fluid velocity being $O(1)$ in the outer region we further define the outer flow fields $\mathbf{U} = \mathbf{u}$ and $P = \mathrm{Pe}^{-1} p$. Defining the outer gradient operator as $\nabla_{\mathbf{X}} = \mathrm{Pe}^{-1}\nabla$, the outer variables satisfy the advection-diffusion equation

$$\nabla_{\mathbf{X}}^2 C = \mathbf{U} \cdot \nabla_{\mathbf{X}} C, \tag{15}$$

as well as the Stokes equations,

$$\nabla_{\mathbf{X}} \cdot \mathbf{U} = 0, \quad \nabla_{\mathbf{X}}^2 \mathbf{U} = \nabla_{\mathbf{X}} P. \tag{16}$$

The far-field conditions now apply to the outer fields, giving

$$C \to 0, \quad \mathbf{U} \to -W\hat{\mathbf{e}}_z \quad \text{as} \quad R = |\mathbf{X}| \to \infty. \tag{17}$$

C. Analysis and matching

Since $W_0 = 1$ it is evident that $\mathbf{U}_0 \equiv -\hat{\mathbf{e}}_z$. The leading-order concentration is accordingly governed by

$$\nabla_{\mathbf{X}}^2 C_0 = -\frac{\partial C_0}{\partial Z}. \tag{18}$$

This equation has been solved in Ref. [5]. Thus, the transformation $C_0 = e^{-\frac{1}{2}Z} H$ yields the Helmholtz-type equation, $\nabla_{\mathbf{X}}^2 H = \frac{1}{4} H$, whose fundamental solution is the screened source $e^{-R/2}/R$. Asymptotic matching thus readily gives $C_0 = \exp[-\frac{1}{2}R(1+\mu)]/R$.

We now recalculate c_1, writing it as $c_{1p} + c_{1h}$ where c_{1p} is given by (14). The most general complementary solution that results in satisfaction of (12) is

$$c_{1h} = -\frac{9\mu}{16r^2} + \sum_{n=0}^{\infty} a^{(n)}[(n+1)r^n + nr^{-(n+1)}]L^{(n)}(\mu), \tag{19}$$

where $L^{(n)}$ is the Legendre polynomial of degree n. Applying van Dyke matching [9] gives $a^{(0)} = -1/2$ and $a^{(n)} = 0$ for all $n > 0$.

Consider now the $O(\mathrm{Pe})$ inner flow. It is governed by the Stokes equations, the slip condition, $\mathbf{u}_1 = (\mathcal{U}/\mathcal{W})\boldsymbol{\nabla}_s c_1$ at $r = 1$, the far-field condition, $\mathbf{u}_1 \to -W_1\hat{\mathbf{e}}_z$, and a force-free constraint. Making use of the reciprocal theorem [10] we circumvent the need for calculating the flow field, obtaining W_1 directly as a quadrature of the slip distribution:

$$W_1 = -\frac{\mathcal{U}}{4\pi\mathcal{W}}\hat{\mathbf{e}}_z \cdot \oint_{r=1} dA\, \boldsymbol{\nabla}_s c_1. \tag{20}$$

Integration by parts [11] gives here $W_1 = -(\mathcal{U}/\mathcal{W})\int_{-1}^{1} \mu c_1\, d\mu$, whereupon substitution of c_1 yields the requisite correction, $W_1 = \mathcal{U}/8\mathcal{W}$. Note that the product of $\mathcal{U}/\mathcal{W}$ and Pe furnishes the phoretic Péclet number α. We accordingly obtain the following simple approximation for the particle speed:

$$W \sim 1 + \frac{\alpha}{8} + \cdots, \tag{21}$$

which is independent of Pe. At leading order, this velocity enhancement is equivalent to a fractional drag reduction of magnitude $\alpha/8$. The dependence upon α is somewhat counterintuitive, since the phoretic flow does not affect the solute distribution at the asymptotic order considered.

IV. SIMPLE SHEAR

We now address the problem where the particle is freely suspended in a simple shear flow of magnitude G. Consider first the situation at zero Péclet, where the hydrodynamic problem is identical to that about an inert particle. To maintain itself force free, the particle translates with the velocity of the hypothetical streamline of the ambient flow that passes through its center; to maintain itself torque free, the particle rotates with the angular velocity of the ambient flow.

When advection is present the flow differs from that about an inert particle. It is evident by symmetry that the particle does not experience any hydrodynamic force when translating with the local ambient velocity and would accordingly retain that rectilinear velocity (see below). We now choose a reference frame which translates with the particle center and does not rotate relative to the imposed shear. In that frame the flow is steady. We place the x axis in the flow direction and the y axis in the shear direction; choosing here the velocity scale $\mathcal{W}$ as aG, the hydrodynamic far-field condition becomes [cf. (7)]

$$\mathbf{u} \sim \hat{\mathbf{e}}_x y \quad \text{as} \quad r \to \infty. \tag{22}$$

We note that the only fixed vectors in the problem are $\hat{\mathbf{e}}_x$ and $\hat{\mathbf{e}}_y$. Moreover, these vectors are specified only through the tensor product $\hat{\mathbf{e}}_x\hat{\mathbf{e}}_y$, from which no true vector can be extracted. This

reconfirms the lack of modification to the particle rectilinear velocity, as anticipated above. On the other hand, the tensor product $\hat{\mathbf{e}}_x\hat{\mathbf{e}}_y$ does provide a pseudovector in the z direction, allowing for particle rotation about it. The slip condition (6) is accordingly replaced by

$$\mathbf{u} = \hat{\mathbf{e}}_z\Omega \times \mathbf{x} + \frac{\mathcal{U}}{\mathcal{W}}\nabla_s c \quad \text{at} \quad r = 1, \tag{23}$$

wherein Ω is the angular velocity of the particle (normalized by G). This quantity is determined by the torque-free constraint [cf. (8)].

A. Stresslet

With the particle being both force- and torque-free and the flow governed by the Stokes equations, it is known [12] that the far-field disturbance to the imposed shear is

$$\mathbf{u} - \hat{\mathbf{e}}_x y \sim -3\frac{\mathbf{x}\cdot\mathsf{S}\cdot\mathbf{x}}{8\pi r^5}\mathbf{x}, \tag{24}$$

where the symmetric traceless second-order tensor S is the stresslet (normalized by $\eta a^2\mathcal{W}$). In the present problem tensorial symmetry necessitates that S is proportional to the rate-of-strain tensor, hence

$$\mathsf{S} = \tfrac{1}{2}(\hat{\mathbf{e}}_x\hat{\mathbf{e}}_y + \hat{\mathbf{e}}_y\hat{\mathbf{e}}_x)S. \tag{25}$$

The importance of S lies in the effective viscosity of particle suspensions [12]. Since a suspension of homogeneous spheres is macroscopically isotropic, it possesses a quasi-Newtonian relation between the shear stress and ambient rate of strain; in particular, for a dilute suspension the fractional increase of the effective viscosity (relative to the liquid viscosity) is proportional to the particle volume fraction, the constant of proportionality being $3S/8\pi$.

Our main interest is accordingly in the dependence of S upon Pe. As in the previous section we consider the asymptotic limit $\mathrm{Pe} \ll 1$ with $\mathcal{U}/\mathcal{W}$ fixed.

B. Small Pe

For $\mathrm{Pe} = 0$ the solute concentration is given by (5). As the flow field is uncoupled to the solute concentration, it is provided by the well-known [6] profile

$$\mathbf{u}_0 = y(1 - r^{-5})\hat{\mathbf{e}}_x - \frac{5xy}{2}(r^{-5} - r^{-7})\mathbf{x}, \tag{26}$$

which is consistent with a torque-free particle. The associated angular velocity, $\Omega_0 = -1/2$, is that of the ambient shear; the associated stresslet, $S_0 = 20\pi/3$, immediately follows from (24) and (25). Corresponding to that stresslet is the Einstein $5/2$ value of the proportionality coefficient mentioned after (25).

As in the sedimentation problem, a naïve attempt to employ a regular expansion of the form (10) results in the failure to obtain c_1, the source of incompatibility being again the r^{-1} decay rate of c at large r. Here the nonuniformity is stronger due to the associated $O(r)$ velocity at these distances [6]: thus, with advection being $O(\mathrm{Pe}\, r^{-1})$ in Eq. (1) it enters the leading-order balance already at distances $O(\mathrm{Pe}^{-1/2})$. This suggests replacing the generic expansion (10) by

$$f \sim f_0 + \mathrm{Pe}^{1/2} f_{1/2} + \mathrm{Pe}\, f_1 + \cdots. \tag{27}$$

The outer coordinates $\mathbf{X} = (X, Y, Z)$ are now defined as $\mathbf{X} = \mathrm{Pe}^{1/2}\mathbf{x}$. In the outer region we accordingly define the fields $C = \mathrm{Pe}^{-1/2}c$, $\mathbf{U} = \mathrm{Pe}^{1/2}\mathbf{u}$, and $P = p$; these $O(1)$ fields presumably possess expansion (27). With these definitions the outer differential equations (15) and (16) retain their validity, and the far-field conditions (17) are replaced by

$$C \to 0, \quad \mathbf{U} \sim Y\hat{\mathbf{e}}_x \quad \text{as} \quad R \to \infty. \tag{28}$$

C. Analysis and matching

It is evident that in the present problem $\mathbf{U}_0 = Y\hat{\mathbf{e}}_x$. The leading-order outer concentration C_0 is accordingly governed by

$$\nabla_{\mathbf{X}}^2 C_0 = Y\frac{\partial C_0}{\partial X}. \tag{29}$$

The fundamental solution of this equation was calculated by Elrick [13]:

$$\tilde{C} = \frac{1}{2\pi^{1/2}}\int_0^\infty \frac{ds}{s^{3/2}\left(1+\frac{1}{12}s^2\right)^{1/2}}\exp\left[-\frac{\left(X-\frac{1}{2}sY\right)^2}{4s\left(1+\frac{1}{12}s^2\right)} - \frac{Y^2+Z^2}{4s}\right]. \tag{30}$$

In what follows it turns out that we need three terms in the small-R approximation of (30). Asymptotic evaluation of the integral yields (see the Appendix)

$$\tilde{C} \sim \frac{1}{R} + \frac{\Lambda}{2\pi^{1/2}} + \frac{R}{8}\sin^2\theta\sin 2\phi + \cdots, \tag{31}$$

wherein

$$\Lambda = \int_0^\infty s^{-3/2}\left[\left(1+\frac{1}{12}s^2\right)^{-1/2} - 1\right]ds = -\frac{2^{1/2}[\Gamma(3/4)]^2}{3^{1/4}\Gamma(1/2)}. \tag{32}$$

Asymptotic matching with (5) thus gives $C_0 = \tilde{C}$.

Consider now the inner concentration $c_{1/2}$. It is unaffected by convection, $\nabla^2 c_{1/2} = 0$, and satisfies the homogenous Neumann condition $\partial c_{1/2}/\partial r = 0$ at $r = 1$. Application of van Dýke matching gives, in light of (31), the uniform concentration $c_{1/2} \equiv \Lambda/2\pi^{1/2}$. In the absence of $O(\mathrm{Pe}^{1/2})$ slip it is readily seen that $\mathbf{u}_{1/2}$ trivially vanishes, as do $\Omega_{1/2}$ and $S_{1/2}$. To find the leading-order effect of convection we accordingly need to calculate c_1, which in turn requires the next term, $C_{1/2}$, in the outer region [14].

To obtain the equation governing $C_{1/2}$ we need $\mathbf{U}_{1/2}$, the leading-order outer correction to the shear $\mathbf{U}_0$. Considering (26) we note that the disturbance in $\mathbf{u}_0$ relative to the imposed shear decays as r^{-2}. In the outer region, that disturbance is $O(\mathrm{Pe}^{3/2})$ smaller than the leading-order shear. It follows that $\mathbf{U}_{1/2}$ trivially vanishes [15], whereby $C_{1/2}$ satisfies an equation identical to (29). Performing van Dyke matching we find that $C_{1/2}$ too trivially vanishes.

We can now calculate c_1, which is governed by (11) and (12) together with the far-field behavior

$$c_1 \sim \frac{r}{8}\sin^2\theta\sin 2\phi, \tag{33}$$

which follows from van Dyke matching in conjunction with (31). We seek a solution proportional to a surface harmonic of degree two and order two:

$$c_1 = f(r)\sin^2\theta\sin 2\phi. \tag{34}$$

Substitution into (11) furnishes an ordinary differential equation governing f. In addition, we find from (12) and (33) that f satisfies $f'(1) = 0$ and the large-r matching condition $f \sim r/8$. The solution of this problem is $f(r) = \frac{r}{8} - \frac{5}{16r^2} + \frac{5}{12r^3} - \frac{1}{8r^4}$.

We can now consider the flow $\mathbf{u}_1$, driven by the slip condition:

$$\mathbf{u}_1 = \hat{\mathbf{e}}_z\Omega_1\times\mathbf{x} + \frac{\mathcal{U}}{\mathcal{W}}\nabla_s c_1. \tag{35}$$

As in the sedimentation problem, no need arises to solve the detailed flow problem when interest lies in the particle rigid-body motion. Rather, Ω_1 may be determined using (35) in conjunction with the reciprocal theorem [10]. It is then readily verified that the angular dependence in Eq. (34) results in a vanishing Ω_1. This was to be expected, since the imposed shear may be decomposed into rigid-body rotation about the z axis and pure strain with principal directions at 45° relative to the xy axes. The

former does not affect the leading-order spherically symmetric solute cloud, and the latter merely stretches it along one principal axis and compresses it along the other; given the symmetry of the modified solute cloud, no particle rotation results from the associated diffusio-osmotic slip.

On the other hand, the symmetrically deformed cloud *does* affect the stresslet at $O(\mathrm{Pe})$. It was recently shown by Lauga and Michelin [16] that use of the reciprocal theorem allows the stresslet magnitude to be expressed in terms of the velocity distribution along a particle boundary, thus eliminating the need to solve for the detailed flow field. In the present context, where $\mathbf{u}_1$ is tangential to the particle at $r = 1$, we obtain from Ref. [16] $\mathsf{S}_1 = -\frac{5}{2}\oint dA\,(\hat{\mathbf{n}}\mathbf{u}_1 + \mathbf{u}_1\hat{\mathbf{n}})$. Substitution (34) into (35) and making use of (25) gives $S_1 = -5\pi\mathcal{U}/3\mathcal{W}$. Again, we find that the leading-order correction is independent of Pe:

$$S \sim \frac{20\pi}{3} - \frac{5\pi}{3}\alpha + \cdots . \tag{36}$$

For a dilute particle suspension, approximation (36) corresponds to an increase in the effective viscosity of magnitude $\frac{5}{2}\eta(1 - \frac{1}{4}\alpha + \cdots)$ in the volume fraction.

V. CONCLUDING REMARKS

While isotropic chemically active particles lack the intrinsic asymmetry required for self-propulsion, they are known to exhibit a variety of autophoretic phenomena due to both particle-particle [17] and particle-boundary [18] interactions. The present paper has addressed a different autophoretic mechanism, animated by imposed solute convection. This mechanism has been illustrated in two prototypic problems, sedimentation under a force field and rotation under an imposed shear. In the sedimentation problem, our asymptotic analysis has predicted a fractional enhancement of the particle velocity, of magnitude $\alpha/8$. In the shear problem, it has predicted a fractional decrement of the stresslet, of magnitude $\alpha/4$. Recalling that $\alpha = a\mathcal{M}\mathcal{A}/D^2$ is a *signed* quantity, our "drag reduction" interpretation holds for $\alpha > 0$, as in the commonly considered case of solute production ($\mathcal{A} > 0$) and repulsive solute-solid interactions ($\mathcal{M} > 0$).

To the best of our knowledge, the only similar analysis in the literature is that of Frankel and Khair [19], who considered the dynamics of Janus-type particles in shear flows. Frankel and Khair assumed an antisymmetric activity distribution whose zero mean implies a dipole-like far-field solute concentration. This is in sharp contrast with the problems considered herein, where the isotropic activity results in monopole-like concentration. This contrast results in a fundamental qualitative difference between the analysis of Ref. [19] and the shear-problem analysis of Sec. IV.

There are two obvious extensions of our paper. The first involves a more realistic description of the interfacial chemical reaction [20]. Thus, upon making use of a first-order kinetic model the Neumann condition (2) is replaced by a Robin condition [21]. Inspection reveals that the associated generalization of the present analysis is rather technical, and that the asymptotic scaling remains intact.

The second extension, to non-small Péclet number, is more interesting. In the present leading-order analysis the convection-induced mechanical modifications, velocity enhancement in the sedimentation problem and stresslet decrement in the shear problem, are independent of the extrinsic Péclet number Pe. This independence, as well as the invariance of the particle angular velocity Ω in the shear problem, does not hold in the general case of finite Péclet numbers, which is accordingly much richer [21]. In particular, a dependence of the stresslet magnitude upon Pe implies a nonconventional mode of non-Newtonian rheology, where the effective viscosity of a dilute particle suspension depends upon the shear-flow magnitude.

ACKNOWLEDGMENT

This work was supported by the Israel Science Foundation (Grant No. 1081/16).

APPENDIX: DERIVATION OF EQ. (31)

It is convenient to rewrite (30) using polar coordinates

$$\tilde{C} = \frac{1}{2\pi^{1/2}} \int_0^\infty \frac{ds}{s^{3/2}\left(1+\frac{1}{12}s^2\right)^{1/2}} e^{-\sigma R^2/4s}, \tag{A1}$$

wherein

$$\sigma = \frac{\sin^2\theta\left(\cos\phi - \frac{1}{2}s\sin\phi\right)^2}{1+\frac{1}{12}s^2} + \sin^2\theta\sin^2\phi + \cos^2\theta. \tag{A2}$$

Note that

$$\sigma \sim 1 - \tfrac{1}{2}s\sin^2\theta\sin 2\phi + O(s^2) \quad \text{for} \quad s \ll 1. \tag{A3}$$

For small R it is evident that to leading order the integral is dominated by a "local" contribution [9] arising from an $O(R^2)$-wide interval about $s = 0$:

$$\tilde{C} \sim \frac{1}{2\pi^{1/2}} \int_0^\infty \frac{ds}{s^{3/2}} e^{-R^2/4s} = \frac{1}{R}. \tag{A4}$$

To obtain the next correction we subtract off the singularity:

$$2\pi^{1/2}\left(\tilde{C} - \frac{1}{R}\right) = \int_0^\infty \frac{ds}{s^{3/2}}\left[\frac{e^{-\sigma R^2/4s}}{\left(1+\frac{1}{12}s^2\right)^{1/2}} - e^{-R^2/4s}\right]. \tag{A5}$$

At leading order the integral is dominated by a "global" contribution from the entire integration range, namely,

$$\Lambda = \int_0^\infty \frac{ds}{s^{3/2}}\left[\left(1+\frac{1}{12}s^2\right)^{-1/2} - 1\right] = -\frac{2^{1/2}[\Gamma(3/4)]^2}{3^{1/4}\Gamma(1/2)} \cong -0.91039. \tag{A6}$$

Consider now

$$2\pi^{1/2}\left(\tilde{C} - \frac{1}{R} - \frac{\Lambda}{2\pi^{1/2}}\right) = \int_0^\infty \frac{ds}{s^{3/2}}\left[\frac{e^{-\sigma R^2/4s}}{\left(1+\frac{1}{12}s^2\right)^{1/2}} - e^{-R^2/4s} - \frac{1}{\left(1+\frac{1}{12}s^2\right)^{1/2}} + 1\right]. \tag{A7}$$

This integral is again dominated by a "local" contribution from an $O(R^2)$-wide interval about $s = 0$. Making use of (A3) we readily obtain at leading order

$$2\pi^{1/2}\left(\tilde{C} - \frac{1}{R} - \frac{\Lambda}{2\pi^{1/2}}\right) \sim \frac{R^2}{8}\sin^2\theta\sin 2\phi \int_0^\infty \frac{ds}{s^{3/2}} e^{-R^2/4s}, \tag{A8}$$

which, upon making use of (A4), finally furnishes (31). The first two terms in that expansion are provided in Ref. [6], with what appears like a sign typo: see Eq. (18) and the discussion there.

[1] R. Golestanian, T. B. Liverpool, and A. Ajdari, Designing phoretic micro-and nano-swimmers, New J. Phys. **9**, 126 (2007).

[2] M. Enculescu and H. Stark, Active Colloidal Suspensions Exhibit Polar Order Under Gravity, Phys. Rev. Lett. **107**, 058301 (2011); B. ten Hagen, F. Kümmel, R. Wittkowski, D. Takagi, L. Hartmut, and C. Bechinger, Gravitaxis of asymmetric self-propelled colloidal particles, Nat. Commun. **5**, 1 (2014).

[3] Y. G. Tao and R. Kapral, Swimming upstream: Self-propelled nanodimer motors in a flow, Soft Matter **6**, 756 (2010).

[4] At sufficiently large Péclet numbers such particles may undergo spontaneous self-propulsion in an arbitrary direction in space [11].

[5] A. Acrivos and T. D. Taylor, Heat and mass transfer from single spheres in Stokes flow, Phys. Fluids **5**, 387 (1962).

[6] N. A. Frankel and A. Acrivos, Heat and mass transfer from small spheres and cylinders freely suspended in shear flow, Phys. Fluids **11**, 1913 (1968).

[7] V. Magar, T. Goto, and T. J. Pedley, Nutrient uptake by a self-propelled steady squirmer, Q. J. Mech. Appl. Math. **56**, 65 (2003); S. Michelin and E. Lauga, Optimal feeding is optimal swimming for all Péclet numbers, Phys. Fluids **23**, 101901 (2011).

[8] J. L. Anderson, M. E. Lowell, and D. C. Prieve, Motion of a particle generated by chemical gradients. Part 1. Non-electrolytes, J. Fluid Mech. **117**, 107 (1982).

[9] E. J. Hinch, *Perturbation Methods* (Cambridge University Press, Cambridge, 1991).

[10] H. Brenner, The Stokes resistance of an arbitrary particle. IV. Arbitrary fields of flow, Chem. Eng. Sci. **19**, 703 (1964); H. A. Stone and A. D. T. Samuel, Propulsion of Microorganisms by Surface Distortions, Phys. Rev. Lett. **77**, 4102 (1996).

[11] S. Michelin, E. Lauga, and D. Bartolo, Spontaneous autophoretic motion of isotropic particles, Phys. Fluids **25**, 061701 (2013).

[12] G. K. Batchelor, The stress system in a suspension of force-free particles, J. Fluid Mech. **41**, 545 (1970).

[13] D. E. Elrick, Source functions for diffusion in uniform shear flow, Aust. J. Phys. **15**, 283 (1962).

[14] The present need for this higher-order calculation, which necessitates the calculation of three terms in Eq. (31), has no counterpart in the classical forced-convection analysis [6], where the spherically symmetric $O(\mathrm{Pe}^{1/2})$ solute concentration results in a nonzero contribution to the Nusselt number.

[15] Since higher-order inner corrections to $\mathbf{u}$ cannot diverge at large r, and given the force-free condition, these corrections decay at least as rapidly as r^{-2}.

[16] E. Lauga and S. Michelin, Stresslets Induced by Active Swimmers, Phys. Rev. Lett. **117**, 148001 (2016).

[17] R. Soto and R. Golestanian, Self-Assembly of Catalytically Active Colloidal Molecules: Tailoring Activity Through Surface Chemistry, Phys. Rev. Lett. **112**, 068301 (2014); S. Michelin and E. Lauga, Autophoretic locomotion from geometric asymmetry, Eur. Phys. J. E Soft Matter **38**, 1 (2015).

[18] A. Domínguez, P. Malgaretti, M. N. Popescu, and S. Dietrich, Effective Interaction Between Active Colloids and Fluid Interfaces Induced by Marangoni Flows, Phys. Rev. Lett. **116**, 078301 (2016); E. Yariv, Wall-induced self-diffusiophoresis of active isotropic colloids, Phys. Rev. Fluids **1**, 032101 (2016); Boundary-induced autophoresis of isotropic colloids: Anomalous repulsion in the lubrication limit, J. Fluid Mech. **812**, 26 (2017).

[19] A. E. Frankel and A. S. Khair, Dynamics of a self-diffusiophoretic particle in shear flow, Phys. Rev. E **90**, 013030 (2014).

[20] U. M. Córdova-Figueroa and J. F. Brady, Osmotic Propulsion: The Osmotic Motor, Phys. Rev. Lett. **100**, 158303 (2008); S. Ebbens, M.-H. Tu, J. R. Howse, and R. Golestanian, Size dependence of the propulsion velocity for catalytic Janus-sphere swimmers, Phys. Rev. E **85**, 020401 (2012).

[21] S. Michelin and E. Lauga, Phoretic self-propulsion at finite Péclet numbers, J. Fluid Mech. **747**, 572 (2014); E. Yariv and S. Michelin, Phoretic self-propulsion at large Péclet numbers, *ibid.* **768**, R1 (2015).