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Polarization of active Janus particles

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We theoretically study the motion of surface-active Janus particles, driven by an effective slip velocity due to a nonuniform temperature or concentration field ψ. With parameters realized in thermal traps, we find that the torque exerted by the gradient $\nabla\psi$ inhibits rotational diffusion and favors alignment of the particle axes. In a swarm of active particles, this polarization adds a novel term to the drift velocity and modifies the collective behavior. Self-polarization in a nonuniform laser beam could be used for guiding hot particles along a given trajectory.

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Swarms of self-propelling birds, fish, or insects show dynamical patterns that arise from the fact that each individual adapts to the state of its neighbors [1]. Similar phenomena occur for liquid dispersions of active objects. Thus cells of *Escherichia coli* move along a chemical gradient generated by their neighbors and form stable spatial structures [2]. More recently, self-driven Janus particles (JPs) have been studied as a model active system [3,4]. It has been shown that the interplay of self-propulsion and rotational diffusion leads to Brownian motion with an enhanced diffusion coefficient [5–10], and that the particles' chemical activity results in cluster formation [11–13] and oriented motion [14]. Guiding a single microswimmer along a given trajectory can be achieved by dynamical feedback [15].

Active colloids have been realized by partly coating silica or polystyrene particles with a metal or carbon layer; self-propulsion arises from nonuniform surface properties such as temperature or chemical activity. An excess temperature $\psi = T$ is induced by heating the metal or carbon cap through absorption of laser light [7–9,13,15] or magnetic fields [10]. Chemical signaling with a molecular solute, $\psi = c$, is achieved by electrocatalysis of hydrogen peroxide at a metal cap [3,5,11,12].

A minimal model for self-driven systems consists in a drift-diffusion equation that was originally designed for motile bacteria with chemotactic interactions [16], and that describes complex spatial structures observed in cell cultures [17]. More recently this model was adapted to JPs that aggregate due to the chemical gradient generated by their electrocatalytic activity [11]. For thermally active colloids, similar results were derived from the Smoluchowski equation [18,19]: Self-propulsion strongly enhances the diffusion term, whereas the drift velocity arises from the gradient field $\nabla\psi$ generated by the neighbor particles; a sufficiently strong attractive drift term may even cause the implosion of a swarm of JPs.

In the present paper we show that active colloids are polarized by their chemical or thermal interactions [20]. A nonuniform field ψ exerts a viscous torque on the JP, which in turn aligns its symmetry axis on the gradient $\nabla\psi$. This polarization affects both single-particle and collective motion. In particular, it adds a contribution to the effective velocity of the drift-diffusion equation, which is dominant for strong driving or large Péclet number and significantly modifes the collective behavior and the phase diagram [18]. It turns out that oriented self-propulsion is close to the usual model for bacteria motility.

Polarization of Janus particles. Consider a JP interacting with a concentration or temperature field ψ. Within a thin boundary layer, the parallel component of the gradient of the local field $\bar{\psi}$ induces an effective slip velocity along the particle surface [21,22],

$$\mathbf{v}_s(\mathbf{r}) = \mu(\mathbf{r})\nabla_\parallel\bar{\psi}(\mathbf{r}). \tag{1}$$

The main result of this paper arises from the material-dependent nonuniform mobility constant μ and from the properties of the local field $\bar{\psi}$. We consider the case where two values μ and μ' occur on the two half spheres of an otherwise homogeneous JP, as indicated in Fig. 1. The resulting effective slip velocities are indicated as solid and dotted lines; their sign and magnitude depend on the mobility values and on the local gradient $\nabla_\parallel\bar{\psi}$.

The effective slip velocity (1) constitutes the boundary condition for the velocity field $\mathbf{v}(\mathbf{r})$ of the surrounding fluid, $\mathbf{v}|_B = \mathbf{u} + \boldsymbol{\Omega}\times\mathbf{r}_B + \mathbf{v}_s$, where B indicates the outer limit of the interaction layer, typically at a few nanometers from the particle surface. This relation determines the linear and angular velocities of the JP. The former is given by the surface average $\mathbf{u} = -S^{-1}\int dS\mathbf{v}_s$, and reads for a spherical particle [23]

$$\mathbf{u} = -\xi_1\frac{\mu+\mu'}{3}\nabla\psi(\mathbf{r}). \tag{2}$$

Depending on the sign of $\mu+\mu'$, the particles move along or opposite to the field gradient. For uniform surface properties ($\mu = \mu'$) one recovers the usual phoretic velocity of particles in an external field $\nabla\psi$ [21].

The factor ξ_1 in (2) accounts for the deformation of the field due to the presence of the JP. The local gradient is given by the projection on the tangential plane, $\nabla_\parallel\bar{\psi} = \xi_1(1-\hat{\mathbf{r}}\hat{\mathbf{r}})\cdot\nabla\psi$, with the surface normal $\hat{\mathbf{r}}$. In the case of temperature, the correction factor $\xi_1 = 3\kappa_s/(2\kappa_s+\kappa_p)$ is given by the heat conductivities of solvant and particle, κ_s and κ_p [23]. The resulting local gradient $\nabla_\parallel\bar{T}$ is valid for a sufficiently thin cap, such that the metal layer does not modify the heat flow pattern. In contrast, a thick metal layer results in a spatially varying $\xi(\mathbf{r})$ that reduces the slip velocity on the cap and enhances it on the insulating hemisphere [24]; the limiting case of an isothermal cap is accounted for by putting $\mu = 0$ and augmenting μ' by a factor that depends on the orientation of the JP. In the case

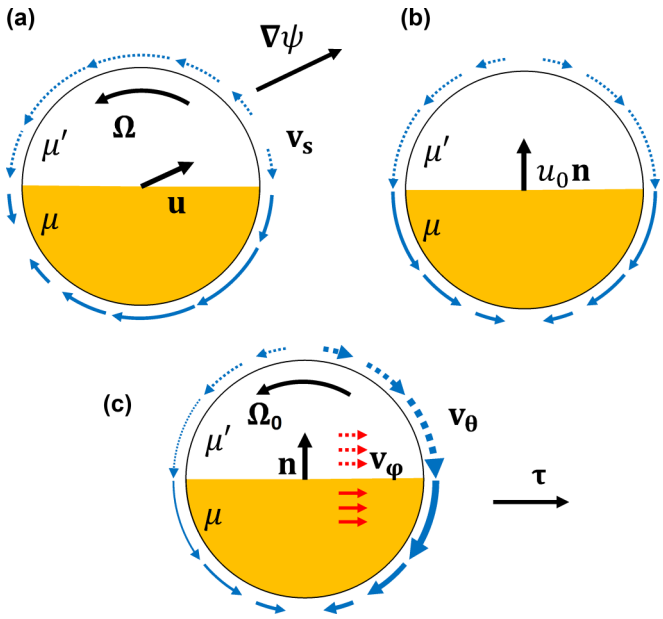

FIG. 1. (Color online) (a) Motion of a Janus particle resulting from an external field gradient $\nabla\psi$. The particle moves at velocity $u \propto \mu' + \mu$ along the field gradient and rotates at rate $\Omega \propto \mu' - \mu$. The mobility takes the values μ' on the insulating part and μ on the metal cap; the quasislip velocities are indicated by full and dotted arrows. (b) Self-propulsion of an active JP. The particle's own field ψ_S leads to a quasislip velocity $\mathbf{v}_s = \mu\nabla_\parallel\psi_S$ that is symmetric with respect to the particle's axis. The particle self-propels at a velocity $u_0 \propto \mu' + \mu$. (c) Nonuniform source field with gradient $\boldsymbol{\tau}$. The quasislip velocity at the particle surface has contributions in polar and azimuthal directions; both the magnitude of $\mathbf{v}_\theta$ and the orientation of $\mathbf{v}_\varphi$ are sensitive to the intensity gradient $\boldsymbol{\tau}$. The resulting angular velocity $\boldsymbol{\Omega}_0$ contains terms proportional to $\mu' \pm \mu$, as given in (8).

of an applied concentration gradient the κ_i are the diffusion coefficients of the solute; if the latter does not penetrate the particle, one has $\xi_1 = \frac{3}{2}$. For strong accumulation, additional corrections are required due to tangential diffusive flux within the interaction layer.

A nonuniform mobility factor $\mu(\mathbf{r})$ gives rise to a rotational component of the effective slip velocity [23,25]. For example, for $\mu' = 0$ the dotted arrows in Fig. 1(a) vanish, and the remaining $\mathbf{v}_s$ results in a clockwise motion of the surrounding fluid; the particle turns in the opposite direction until its axis is parallel to $\nabla\psi$. Equilibrating the surface and viscous forces one obtains the angular frequency

$$\boldsymbol{\Omega} = -\frac{3}{2a}\oint \frac{d\mathbf{S}\times\mathbf{v}_s}{4\pi a^2}, \tag{3}$$

where $d\mathbf{S}$ is the oriented surface element and a is the particle radius. Performing the integral and introducing the unit vector $\mathbf{n}$ along the particle axis, one finds

$$\boldsymbol{\Omega} = \mathbf{n}\times\mathbf{A}, \quad \mathbf{A} = -\frac{3\xi_1(\mu'-\mu)}{8a}\nabla\psi(\mathbf{r}). \tag{4}$$

Note that the angular velocity is proportional to the mobility difference $\mu - \mu'$ of the two hemispheres and thus vanishes for a homogeneous surface. Yet in general μ and μ' are quite different from each other. If the heated metal cap forms an isotherm, its thermophoretic mobility μ is zero [24], whereas μ' may take either sign depending on the precise driving mechanism [26,27].

The viscous stress underlying (3), tends to orient the JP along the external field, whereas rotational diffusion with coefficient D_r favors dispersion. The resulting kinetics are described by the Smoluchowski equation for the distribution function $f(\mathbf{n})$,

$$\partial_t f = -\mathcal{R}\cdot(\boldsymbol{\Omega} - D_r\mathcal{R})f \equiv \mathcal{L}_\mathbf{n} f, \tag{5}$$

with the rotation operator $\mathcal{R} = \mathbf{n}\times\nabla_\mathbf{n}$, and the gradient $\nabla_\mathbf{n}$ with respect to the orientation of the JP [18]. This equation is readily solved in terms of the angle θ between the particle axis and the field gradient, resulting in $\Omega = A\sin\theta$. The corresponding equation for the steady state $(A\sin\theta + D_r\partial_\theta)f = 0$, is readily solved, $f(\theta) \propto e^{(A/D_r)\cos\theta}$. This effective rotational potential aligns the JP axis on the field gradient, with the mean orientation

$$\mathbf{n}_{\rm eq} = \left(\coth\frac{A}{D_r} - \frac{D_r}{A}\right)\frac{\mathbf{A}}{A}. \tag{6}$$

For an estimate of the polarization amplitude, we use $A \approx u/a$ and note measured drift velocities u exceed 1 μm/s [28]. For a micron-size particle we then have $A \approx 1\ \mathrm{s}^{-1}$ and $D_r \approx 0.1\ \mathrm{s}^{-1}$, resulting in $n_{\rm eq} \approx 1$. In other words, for experimental conditions as realized in colloidal traps and thin films, we predict a strong polarization of Janus particles, as illustrated in Fig. 2(a).

Self-propelling Janus particles. Now we consider a swarm of active JPs as shown in Fig. 2(b). Their motion consists of single-particle and interaction contributions: Each particle self-propels in its own nonuniform field ψ_S, whereas that of the neighbors, $\psi(\mathbf{r}) = \sum_j \psi_j(\mathbf{r} - \mathbf{r}_j)$, results in the linear and angular velocities (2) and (4).

The self-generated term ψ_S arises from the active surface property Q; for example, the surface temperature T_S is modified by laser heating at power $Q = \beta I(\mathbf{r}_0)$, where I is the beam intensity at the particle position $\mathbf{r}_0$, and β the absorption coefficient per unit area of the cap. For constant power Q_0, the effective slip velocity is symmetric about the particle axis, as illustrated in Fig. 1(b), and results in self-propulsion at a speed $\mathbf{n}u_0 = S^{-1}\oint dS\mu(\mathbf{r})\nabla_\parallel\psi_S$. Solving the diffusion equation for ψ_S one finds [23]

$$u_0 = \xi_1\frac{\mu+\mu'}{3}\frac{Q_0}{8\kappa_s}, \tag{7}$$

where Q_0/κ_s gives the mean gradient of ψ_S.

For a spatially varying laser intensity $I(\mathbf{r})$ (or concentration of a catalytic agent [25]), the source field breaks the axial symmetry. The resulting slip velocity has both polar and azimuthal components, as illustrated in Fig. 1(c), and exerts a viscous torque

$$\boldsymbol{\Omega}_0 = \mathbf{n}\times\mathbf{A}_0, \tag{8a}$$

which is perpendicular on the particle axis $\mathbf{n}$ and on the gradient of the source, $\boldsymbol{\tau} = \nabla\ln I(\mathbf{r}_0)$,

$$\mathbf{A}_0 = -\boldsymbol{\tau}\left((\mu'-\mu)\frac{5}{18}\xi_1 + (\mu'+\mu)\tilde{\xi}\right)\frac{3Q_0}{8\kappa_s}. \tag{8b}$$

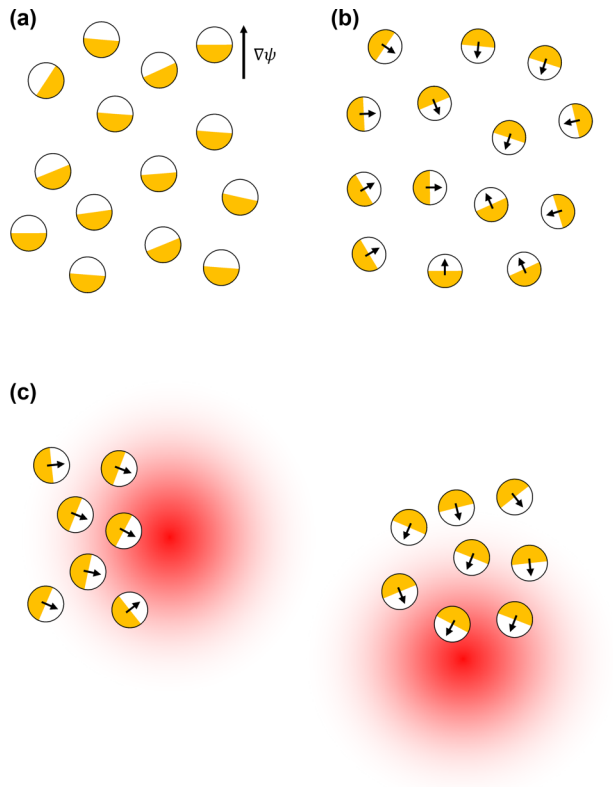

FIG. 2. (Color online) Polarization of Janus particles in a field gradient $\nabla\psi$. (a) Case of an externally applied field ψ; the orientation of the particles is given by Eq. (6). There is no self-propulsion, $u_0 = 0$; the small translational velocity $\mathbf{u}$ is not indicated. (b) Self-confinement of a swarm of Janus particles. With an appropriate choice of the mobilities μ and μ', the drift velocity (13) points toward the center of the swarm, thus favoring cluster formation or even implosion [18]. The polarization is due to the interaction field ψ which in the simplest case has radial symmetry and its related to the density by $\nabla^2\psi + k\rho = 0$. (c) Guided self-propulsion. Self-driven hot JPs are polarized either by the beam intensity gradient $\boldsymbol{\tau}$ of the heating laser, or by an external temperature gradient $\nabla T_{\rm ext}$ that results from local heating of the solvant with an IR laser. The JPs follow the heated spot, first to the right and then downward in the figure.

The first term involves the mobility difference and the dipolar deformation factor ξ_1. The second one, proportional to $\mu' + \mu$, arises from even-order multipoles of ψ_S and carries a correction factor $\widetilde{\xi}$ which is given by the weighted average of the diffusivity contrast factors $\xi_n = (2n+1)/(n+1+n\kappa_p/\kappa_s)$ [23]. For $\mu' > \mu$ both terms result in counterclockwise rotation as shown in Fig. 1(c). The case of catalytic activity is discussed in [25].

Comparing self-propulsed motion with driving due to neighbors in a swarm of JPs, we find that for a colloidal volume fraction of at most a few percent, the velocity u_0 is much larger than that due to an external field u. The torques exerted by the field of an active neighbor at distance R and by a intensity gradient vary as $\Omega \sim u_0 a/R^2$ and $\Omega_0 \sim u_0\tau$, respectively; depending on the system parameters, one or the other may dominate.

The state of a given JP is described by its position $\mathbf{r}$ and the orientation of its axis $\mathbf{n}$. The distribution function $P(\mathbf{r},\mathbf{n})$ obeys the equation

$$\partial_t P = -\nabla\cdot(u_0\mathbf{n} + \mathbf{u} - D\nabla)P + \mathcal{L}_{\mathbf{n}}P. \tag{9}$$

The first term on the right-hand side describes translational motion with velocity $u_0\mathbf{n} + \mathbf{u}$ and gradient diffusion with Einstein coefficient D. The second term accounts for rotational motion according to (5); the diffusion coefficients are related through $D = \frac{4}{3}a^2 D_r$.

The relaxation time $1/D_r$ and particle radius a are small compared to the time and length scales of collective motion [18]. Thus an approximate solution of (9) is obtained by inserting the moment expansion $P(\mathbf{r},\mathbf{n}) = \rho(\mathbf{r}) + \mathbf{n}\cdot\mathbf{p}(\mathbf{r}) + \ldots$, integrating over $\mathbf{n}$, and truncating the resulting hierarchy at finite order. Neglecting quadrupolar contributions and other small terms in the equation for the polarization vector $\mathbf{p}(\mathbf{r}) = (1/4\pi)\int d\mathbf{n}\mathbf{n}P$, we find [23]

$$\mathbf{p} = -\frac{u_0}{6D_r}\nabla\rho + \mathbf{n}_{\rm eq}\rho, \quad \mathbf{n}_{\rm eq} = \frac{\mathbf{A} + \mathbf{A}_0}{3D_r}. \tag{10}$$

The first term, which has been derived in previous work [18,29], accounts for the diffusive transport of polarization in a nonuniform density; the prefactor u_0/D_r gives the distance over which the particle self-propels during its rotational relaxation time; with $u_0 \sim 10\ \mu$m/sec and $1/D_r \sim 1$ sec one finds about 10 microns. The second term $\mathbf{n}_{\rm eq}\rho$ accounts for active polarization of JPs. Figure 2(b) illustrates the alignment on the field gradient (4) created by nearby JPs. The self-polarization amplitude of the JPs along the intensity gradient τ is given by $n_{\rm eq} \sim A_0/D_r$; noting $A_0 \sim \tau u_0$ and that τ is of the order of the inverse beam width w, we have $n_{\rm eq} \sim u_0/wD_r$. With typical values we find that Janus particles align on the intensity gradient of the laser beam, as illustrated in Fig. 2(c).

With the polarization $\mathbf{p}$ one obtains the drift-diffusion equation for the density ρ [23],

$$\partial_t\rho = -\nabla\cdot(\mathbf{u}_{\rm eff}\rho - D_{\rm eff}\nabla\rho), \tag{11}$$

where $D_{\rm eff} = D(1 + \frac{2}{9}{\rm Pe}^2)$ is the effective diffusion coefficient and ${\rm Pe} = u_0 a/D$ the Péclet number [4,5]. The effective velocity

$$\mathbf{u}_{\rm eff} = \mathbf{u} + u_0\mathbf{n}_{\rm eq} \tag{12}$$

consists of the interaction-driven drift (2) and oriented self-propulsion with the equilibrium polarization $\mathbf{n}_{\rm eq} = (\mathbf{A} + \mathbf{A}_0)/3D_r$.

In a constant source field Q there is no self-polarization, $\mathbf{A}_0 = 0$, and the drift velocity can be cast in the form

$$\mathbf{u}_{\rm eff} = \xi\left(-\frac{\mu+\mu'}{3} + {\rm Pe}\frac{\mu-\mu'}{6}\right)\nabla\psi(\mathbf{r}). \tag{13}$$

The first term in parentheses, which has been derived previously [11,18], is independent of the particle orientation. The second one arises from self-propulsion of polarized JP and dominates at large Péclet number. Since ${\rm Pe} \propto \mu + \mu'$, the two terms in (13) carry opposite signs for $\mu > \mu'$.

Temperature and concentration fields generated by the JPs' heat absorption or chemical activity satisfy $\nabla^2\psi + k\rho = 0$ with the particle density as source term. Then the sign of the prefactor of $\mathbf{u}_{\rm eff}$ determines whether self-propulsion disperses

or confines a cloud of JPs. A sufficiently large negative drift velocity results in clustering as illustrated in Fig. 2(b) [11–13] and may even drive implosion of the swarm [18]. This latter scenario has been discussed in detail for $\mathbf{u}_{\rm eff} = \mathbf{u}$ and $\mu = \mu' < 0$, corresponding to a negative Soret coefficient [18]. The correction term $u_0\mathbf{n}_{\rm eq}$ derived here, is dominant for $|{\rm Pe}| > 0$ and, according to (13) results in attraction $\mu'^2 > \mu^2$, independently of the sign of the mobilities. Thus polarization enhances $\mathbf{u}_{\rm eff}$ by a factor Pe and, at large Péclet number, even modifies the dynamical phase diagram. Experiments on cluster formation [11–13] and oriented motion [14] support the qualitative features of the drift-diffusion model, yet available data are not sufficiently precise for a quantitative comparison.

Guided self-propulsion. So far we discussed polarization along the field gradient $\nabla\psi$ generated by the heat absorption or chemical activity of neighbor JPs. Here we discuss the case where both propulsion and polarization result from the particle's self-generated temperature field T_S. With the linear velocity u_0 and the order parameter $\mathbf{n}_{\rm eq} = \mathbf{A}_0/3D_r$, we obtain oriented motion along the intensity gradient of the laser beam,

$$\mathbf{u}_{\rm eff} = u_0\mathbf{n}_{\rm eq} = \tfrac{4}{9}{\rm Pe}a\mathbf{A}_0. \tag{14}$$

Note that this a single-particle property and varies with the square of the laser intensity. A physical realization is sketched in Fig. 2(c), where a focused laser beam illuminates a swarm of JPs. Since the particles move towards the center of the beam according to (3), they could be guided by a mobile laser beam along a given trajectory.

Chemotaxis of bacteria. We compare the motion of polarized JPs with bacteria that are guided by chemotactic signaling. *E. coli* self-propels through flagella rotating in the "run" mode at a velocity u_0 along its axis $\mathbf{n}$ [17]. After a period of $\tau \sim 1$ sec, they switch to the "tumble" mode, which randomly changes the orientation and thus plays the role of rotational diffusion. The cell performs a random walk with diffusion coefficient $D_{\rm eff} \sim u_0^2\tau$.

Bacteria are not able to actively reorient in a field gradient, contrary to JPs according to (3). Yet they are sensitive to the concentration of certain solutes. If a cell detects a favorable change of ψ along its trajectory, it augments the time τ; on the other hand, if it feels it goes the wrong direction, it switches more rapidly to the tumble mode. As a consequence, the bacterium spends more time in an orientation toward the source [17]. Assuming a linear variation with the concentration gradient, one has $\tau = \tau_0 + \alpha\mathbf{n}\cdot\nabla\psi$, where α describes the strength of the response to chemical signalling. The resulting polarization $\mathbf{n}_{\rm eq} = \frac{1}{3}\alpha\nabla\psi$ results in the drift velocity

$$\mathbf{u}_{\rm eff} = u_0\mathbf{n}_{\rm eq} = \frac{u_0}{3}\alpha\nabla\psi. \tag{15}$$

Comparison with the drift velocity of JPs shows that bacteria motion corresponds to the second term in Eq. (12), that is, to self-propulsion along the field gradient $\nabla\psi$.

In view of Eqs. (12) and (15) one expects for swarms of JPs a dynamical behavior very similar to that observed in bacteria cultures. Fine-tuning of the surface parameters μ and μ' would allow to separate the effects of phoretic motion $\mathbf{u}$ and of oriented self-propulsion $u_0\mathbf{n}_{\rm eq}$. Since only the latter is present in (15), the relative weight of these terms is an important parameter when comparing the motion of JPs and bacteria.

We conclude with a remark on hydrodynamic interactions which have been neglected in the present paper. The interactions considered here are mediated by thermal or concentration gradients $\nabla\psi$ which in three dimensions vary with the square of the inverse distance, $\nabla\psi \propto \mathbf{r}^{-2}$. Depending on the symmetry of effective slip velocity, hydrodynamic interactions decay as r^{-3} or r^{-2} [24,30,31]; the latter term may attain values comparable to the interaction contribution $\mathbf{u}$ in (12). Yet at large Péclet number, it is small as compared to the self-propulsion contribution $u_0\mathbf{n}_{\rm eq}$.

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