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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 non-uniform 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 non-uniform 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 non-uniform 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 signalling 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 Letter we show that active colloids are polarized by their chemical or thermal interactions [20]. A non-uniform 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 novel contribu-

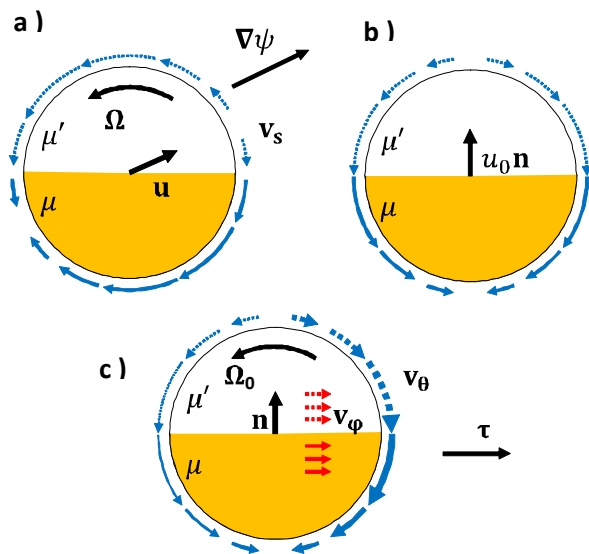


FIG. 1: a) Motion of a Janus particle resulting from an external field gradient $\nabla\psi$. The particle moves at velocity $u \propto \mu' + \mu$ along the field gradient and rotates at rate $\Omega \propto \mu' - \mu$. The mobility takes the values μ' on the insulating part and μ on the metal cap; the quasi-slip 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 $\mathbf{v}_s = \mu\nabla_{||}\psi_s$ that is symmetric with respect to the particle's axis. The particle self-propels at a velocity $u_0 \propto \mu' + \mu$. c) Non-uniform source field with gradient τ . The quasislip velocity at the particle surface has contributions in polar and azimuthal directions; both the magnitude of \mathbf{v}_θ and the orientation of \mathbf{v}_φ are sensitive to the intensity gradient τ . The resulting angular velocity Ω_0 contains terms proportional to $\mu' \pm \mu$, as given in (8).

tion to the effective velocity of the drift-diffusion equation, which is dominant for strong driving or large Péclet number and significantly modifies 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 in-

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 [22, 23]

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

The main result of this paper arise from the material-dependent non-uniform 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 hemispheres 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 [24]

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

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 $\nabla\psi$ [22].

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 solvent and particle, κ_s and κ_p [24]. The resulting local gradient $\nabla_{\parallel}\bar{\psi}$ 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 [25]; 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 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 non-uniform mobility factor $\mu(\mathbf{r})$ gives rise to a rotational component of the effective slip velocity [21, 24]. For example, for $\mu' = 0$ the dotted arrows in Fig. 1a 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

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

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

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

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 [25], 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 $f(\mathbf{n})$,

$$\partial_t f = -\mathcal{R} \cdot (\boldsymbol{\Omega} - D_r \mathcal{R}) f \equiv \mathcal{L}_n f, \quad (5)$$

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

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

For an estimate of the polarization amplitude, we use $A \approx u/a$ and note measured drift velocities u exceed 1 $\mu\text{m/s}$ [28]. For a micron-size particle we then have $A \approx 1 \text{ s}^{-1}$ and $D_r \approx 0.1 \text{ s}^{-1}$, resulting in $n_{\text{eq}} \approx 1$. 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. 2a).

Self-propelling Janus particles. Now we consider a swarm of active JPs as shown in Fig. 2b). Their motion consists of single-particle and interaction contributions: Each particle self-propels in its own non-uniform 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

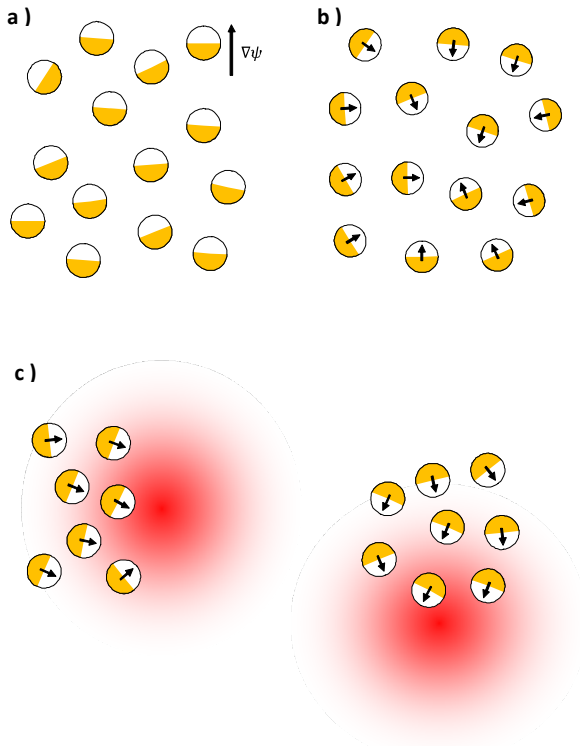


FIG. 2: Polarization of Janus particles in a field gradient $\nabla\psi$. a) Case of an externally applied field ψ ; the orientation of the particles is given by Eq. (6). There is no self-propulsion, $u_0 = 0$; the small translational velocity \mathbf{u} is not indicated. b) Self-confinement of a swarm 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 ∇T_{ext} that results from local heating of the solvent with an IR laser. The JPs follow the heated spot, first to the right and then downward in the figure.

in Fig. 1b, and results in self-propulsion at a speed $\mathbf{u}u_0 = S^{-1} \oint dS \mu(\mathbf{r}) \nabla_{\parallel} \psi_S$. Solving the diffusion equation for ψ_S one finds [24]

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

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 [21]), the source field breaks the axial symmetry. The resulting slip velocity has both polar and azimuthal components, as illustrated in Fig. 1c), and exerts a viscous torque

$$\boldsymbol{\Omega}_0 = \mathbf{n} \times \mathbf{A}_0 \quad (8a)$$

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

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

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 $\tilde{\xi}$ which is given by the weighted average of the diffusivity contrast factors $\xi_n = (2n + 1)/(n + 1 + n\kappa_p/\kappa_s)$ [24]. For $\mu' > \mu$ both terms result in counterclockwise rotation as shown in Fig. 1c.) The case of catalytic activity is discussed in [21].

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

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

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 $P(\mathbf{r}, \mathbf{n}) = \rho(\mathbf{r}) + \mathbf{n} \cdot \mathbf{p}(\mathbf{r}) + \dots$, 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 [24]

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

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

the intensity gradient of the laser beam, as illustrated in Fig. 2c).

With the polarization \mathbf{p} one obtains the drift-diffusion equation for the density ρ [24]

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

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

$$\mathbf{u}_{\text{eff}} = \mathbf{u} + u_0 \mathbf{n}_{\text{eq}} \quad (12)$$

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

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

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

The first term in parentheses, which has derived previously [11, 18], is independent of the particle orientation. The second one has not been considered so far; it arises from self-propulsion of polarized JP and dominates at large Péclet number. Since $\text{Pe} \propto \mu + \mu'$, 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}_{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. 2b [11–13] and may even drive implosion of the swarm [18]. This latter scenario has been discussed in detail for $\mathbf{u}_{\text{eff}} = \mathbf{u}$ and $\mu = \mu' < 0$, corresponding to a negative Soret coefficient [18]. The correction term $u_0 \mathbf{n}_{\text{eq}}$ derived here, is dominant for $|\text{Pe}| > 0$ and, according to (13) results in attraction $\mu'^2 > \mu^2$, independently of the sign of the mobilities. Thus polarization enhances \mathbf{u}_{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 $\mathbf{n}_{\text{eq}} = \mathbf{A}_0 / 3D_r$, we obtain oriented motion along the intensity gradient of the laser beam,

$$\mathbf{u}_{\text{eff}} = u_0 \mathbf{n}_{\text{eq}} = \frac{4}{9} \text{Pe} a \mathbf{A}_0. \quad (14)$$

Note that this a single-particle property and varies with the square of the laser intensity. A physical realization is sketched in Fig. 2c, where a focussed 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 signalling. *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 $D_{\text{eff}} \sim u_0^2 \tau$.

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 $\mathbf{n}_{\text{eq}} = \frac{1}{3} \alpha \nabla \psi$ results in the drift velocity

$$\mathbf{u}_{\text{eff}} = u_0 \mathbf{n}_{\text{eq}} = \frac{u_0}{3} \alpha \nabla \psi. \quad (15)$$

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 $u_0 \mathbf{n}_{\text{eq}}$. 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, $\nabla \psi \propto r^{-2}$. Depending on the symmetry of effective slip velocity, hydrodynamic interactions decay as r^{-3} or r^{-2} [25, 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 $u_0 \mathbf{n}_{\text{eq}}$.

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Supplementary information: Polarization of active Janus particles

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In these notes, we give some details regarding the derivation of the equations that are used in the main body of the paper. We first derive general results regarding the phoretic velocity of an active particle, and the deformation of the field ψ due to the presence of the JP. We then discuss the linear and angular velocity of a spherical Janus particle in the gradient of an external field. Self-propulsion of the active particle in its own field is discussed next. The distribution functions are considered in the last section.

I. GENERAL RESULTS

We consider an active spherical particle of radius a interacting with the solvent through a field ψ , that can be either the temperature or the concentration of some fuel molecules. Within a thin boundary layer, the gradient of the field exerts an osmotic stress on the fluid that results in a quasi-slip velocity parallel to the particle surface [1]

$$\mathbf{v}_s(\mathbf{r}) = \mu(\mathbf{r}) \nabla_{\parallel} \psi(\mathbf{r}), \quad \mathbf{r} \in B \quad (1)$$

where B indicates the outer limit of the interaction layer, typically a few tens of nanometers from the surface. The mobility coefficient $\mu(\mathbf{r})$ depends on the local material properties. The quasi-slip velocity contributes to the boundary condition for the velocity field \mathbf{v} of the surrounding fluid

$$\mathbf{v}(\mathbf{r}) = \mathbf{u} + \boldsymbol{\Omega} \times (\mathbf{r} - \mathbf{r}_0) + \mathbf{v}_s, \quad \mathbf{r} \in B \quad (2)$$

where \mathbf{u} and $\boldsymbol{\Omega}$ respectively stand for the linear and angular velocity of the sphere centered at \mathbf{r}_0 .

At low Reynolds number, the velocity field is solution of the Stokes equation. The corresponding Green's function is the Oseen tensor [2]

$$\mathcal{O}(\mathbf{r}) = \frac{1}{8\pi\eta r} \left(\mathbf{I} + \frac{\mathbf{r}\mathbf{r}}{r^2} \right), \quad (3)$$

with \mathbf{I} the 3×3 unit matrix and η the viscosity of the fluid. The velocity field is then given by

$$\mathbf{v}(\mathbf{r}) = \oint_B S' \mathcal{O}(\mathbf{r} - \mathbf{r}') \cdot \mathbf{f}(\mathbf{r}'), \quad (4)$$

where \mathbf{f} is the surface force density exerted by the particle on the fluid. It is related to the total force \mathbf{F} and torque \mathbf{T} acting on the particle respectively by

$$\mathbf{F} = - \oint_B S \mathbf{f}(\mathbf{r}), \quad \text{and} \quad \mathbf{T} = - \oint_B S \mathbf{r} \times \mathbf{f}(\mathbf{r}). \quad (5)$$

Our goal is to relate the linear and angular velocity to the gradient of the field ψ . To proceed, it is convenient to define the average over the particle surface

$$\langle \dots \rangle = \frac{1}{4\pi a^2} \oint_B S \dots \quad (6)$$

We first discuss the linear velocity; the angular velocity is considered in a second step.

A. Linear velocity

The angular velocity is constant over the surface of the particle. We thus have

$$\langle \boldsymbol{\Omega} \times (\mathbf{r} - \mathbf{r}_0) \rangle = \boldsymbol{\Omega} \times \langle \mathbf{r} - \mathbf{r}_0 \rangle = \mathbf{0}. \quad (7)$$

Averaging Eq. (2) then simply leads to

$$\langle \mathbf{v} \rangle = \mathbf{u} + \langle \mathbf{v}_s \rangle. \quad (8)$$

The average velocity can also be calculated from Eq. (4) (see for instance Ref. [2])

$$\begin{aligned} \langle \mathbf{v} \rangle &= \frac{1}{4\pi a^2} \oint_B dS' \left(\oint_B dS \mathcal{O}(\mathbf{r} - \mathbf{r}') \right) \cdot \mathbf{f}(\mathbf{r}') \\ &= \frac{1}{4\pi a^2} \oint_B dS' \left(\frac{2a}{3\eta} \mathbf{I} \right) \cdot \mathbf{f}(\mathbf{r}') = -\frac{1}{6\pi\eta a} \mathbf{F}. \end{aligned} \quad (9)$$

Comparing Eqs. (8) and (9), we find

$$\mathbf{u} = -\langle \mathbf{v}_s \rangle - \frac{1}{6\pi\eta a} \mathbf{F}. \quad (10)$$

This result reduces to the usual Stokes' law $\mathbf{F} = -6\pi\eta\mathbf{u}$ for the passive situation ($\mu(\mathbf{r}) = 0$). In the case of self-propulsion, there is no net force acting on the particle so that

$$\mathbf{u} = -\langle \mathbf{v}_s \rangle. \quad (11)$$

B. Angular velocity

Let us first define the local normal to the particle's surface $\mathbf{e}_r = (\mathbf{r} - \mathbf{r}_0)/a$, $\mathbf{r} \in B$. Taking the cross product with Eq. (2), one can write

$$\mathbf{e}_r \times \mathbf{v}(\mathbf{r}) = \mathbf{e}_r \times \mathbf{u} + \mathbf{e}_r \times (\boldsymbol{\Omega} \times a\mathbf{e}_r) + \mathbf{e}_r \times \mathbf{v}_s. \quad (12)$$

Given that $\langle \mathbf{e}_r \rangle = 0$ and $\langle \mathbf{e}_{r_i} \mathbf{e}_{r_j} \rangle = \delta_{ij}/3$ (with $i = x, y$ or z), we get

$$\begin{aligned} \langle \mathbf{e}_r \times \mathbf{v} \rangle &= a\boldsymbol{\Omega} - a \langle (\boldsymbol{\Omega} \cdot \mathbf{e}_r) \mathbf{e}_r \rangle + \langle \mathbf{e}_r \times \mathbf{v}_s \rangle \\ &= \frac{2a}{3} \boldsymbol{\Omega} + \langle \mathbf{e}_r \times \mathbf{v}_s \rangle. \end{aligned} \quad (13)$$

The term on the left-hand side is evaluated thanks to Eq. (4) (again, see Ref. [2])

$$\begin{aligned} \langle \mathbf{e}_r \times \mathbf{v} \rangle &= \frac{1}{4\pi a^2} \oint_B S' \left(\oint_B S \mathbf{e}_r \times \mathcal{O}(\mathbf{r} - \mathbf{r}') \cdot \mathbf{f}(\mathbf{r}') \right) \\ &= -\frac{1}{12\pi\eta a^2} \mathbf{T}, \end{aligned} \quad (14)$$

so that

$$\mathbf{\Omega} = -\frac{1}{8\pi\eta a^3} \mathbf{T} - \frac{3}{2a} \langle \mathbf{e}_r \times \mathbf{v}_s \rangle. \quad (15)$$

In the case of a passive sphere, we recover the Stokes' law $\mathbf{T} = -8\pi\eta a^3 \mathbf{\Omega}$ for rotational friction. On the other hand, we get for a torque-free active particle

$$\mathbf{\Omega} = -\frac{3}{2a} \langle \mathbf{e}_r \times \mathbf{v}_s \rangle. \quad (16)$$

II. DEFORMATION OF AN EXTERNAL FIELD ψ

According to (11) and (16), for the linear and angular velocities in an external field ψ we need to know the local gradient $\nabla_{\parallel} \bar{\psi}$. When calculating the latter, one has to account for the local deformation of the field ψ due to the presence of the JP. Thus temperature is sensitive to the heat conductivity contrast and concentration on whether the solute can diffuse into the particle.

As discussed in the main paper, we assume that the "active" properties of the JP are due to the surface only, and that the particle volume is homogeneous. In other words, we assume that the active surface does not contribute to bulk material properties such as the heat conductivity of the JP. In this section we show that the parallel component of the gradient of the local field $\bar{\psi}$ is given by

$$\nabla_{\parallel} \bar{\psi} = \xi_1 \nabla_{\parallel} \psi = \xi_1 (1 - \hat{\mathbf{r}}\hat{\mathbf{r}}) \cdot \nabla \psi, \quad (17)$$

where $\hat{\mathbf{r}}$ is the normal vector on the particle surface and ψ the undeformed applied field. We discuss the correction factor occurring in the translational and angular velocities, and derive the well-known expression for ξ_1 .

We consider fields that are harmonic functions, $\nabla^2 \psi = 0$, and obey well-known boundary conditions at the particle-solvent interface. We first discuss the case of temperature and then generalize our results to concentration fields. The temperature field and the heat flux $\kappa \nabla \bar{T}$ are continuous at the interface. The heat flux condition is expressed by the normal components of the gradient

$$\kappa_s \nabla_{\perp} \bar{T}_s = \kappa_p \nabla_{\perp} \bar{T}_p,$$

where κ_s and κ_p are the heat conductivities of solvent and particle.

We adopt polar coordinates r and θ , where $r = 0$ is the particle center and θ the polar angle with respect to the

external field gradient. Then the applied temperature reads $T = T_0 - \Delta T (r/a) \cos \theta$; its gradient is constant in space. In order to meet the above continuity condition for heat conduction, one has to add at both sides of the interface an additional solution of $\nabla^2 T = 0$ which needs to be proportional to $\cos \theta$. In the solvent ($r > a$), the additional term must vanish at infinity,

$$\bar{T} = T_0 - \Delta T \cos \theta \left(\frac{r}{a} + \alpha \frac{a^2}{r^2} \right), \quad (18a)$$

whereas inside the particle ($r < a$) we have

$$\bar{T} = T_0 - \Delta T \cos \theta \left(\frac{r}{a} + \alpha \frac{r}{a} \right). \quad (18b)$$

The coefficient α remains to be determined. Inserting the temperature in the heat-flux conservation law one finds

$$\alpha = \frac{\kappa_s - \kappa_p}{2\kappa_s + \kappa_p}; \quad (19)$$

Not surprisingly, the coefficient α vanishes for zero conductivity contrast; in this case the temperature field is not deformed by the JP.

Plugging the expression for α in the parallel gradient one calculates

$$\begin{aligned} \nabla_{\parallel} \bar{T} &= \hat{\theta} (1 + \alpha) \sin \theta \frac{\Delta T}{a} \\ &= \frac{3\kappa_s}{2\kappa_s + \kappa_p} (1 - \hat{\mathbf{r}}\hat{\mathbf{r}}) \cdot \nabla T. \end{aligned} \quad (20)$$

Comparing with (17) gives

$$\xi_1 = 1 + \alpha = \frac{3\kappa_s}{2\kappa_s + \kappa_p}. \quad (21)$$

The heat conductivity of various dielectric materials is not very different from that of water, $\kappa_s \approx \kappa_p$; thus for thermoosmotic effects many authors put $\alpha = 0$ and $\xi_1 = 1$.

Similar results are obtained for the concentration ψ of a molecular solute that interacts with the particle surface. If this solute does not penetrate the particle, the corresponding diffusivity vanishes, $\kappa_p = 0$, and we have $\xi_1 = \frac{3}{2}$. A more complex situation may arise if the solute is involved in a chemical reaction at the particle surface.

III. PHORETIC MOTION IN THE GRADIENT OF AN EXTERNAL FIELD ψ

We now focus on the motion of a Janus particle in an external field ψ . The linear and angular velocities are given by Eqs. (11) and (16) respectively, the slip velocity being proportional to the parallel projection of the local gradient. From Eqs. (1) and (17) we readily have

$$\mathbf{u} = -\xi_1 \langle \mu(\mathbf{r}) \nabla_{\parallel} \psi \rangle.$$

We also assume that the Janus particle consists in two hemispheres with constant phoretic mobility, and such that the orientation vector \mathbf{n} of the particle is along the z -direction: $\mathbf{n} = \mathbf{e}_z$. Defining the polar angle θ with respect to this axis, $\cos \theta = \hat{\mathbf{r}} \cdot \mathbf{n}$, the mobility reads

$$\mu(\mathbf{r}) = \begin{cases} \mu' & \text{for } 0 \leq \theta < \pi/2, \\ \mu & \text{for } \pi/2 < \theta \leq \pi. \end{cases}$$

Regarding the gradient of the field, it is assumed to be constant at the scale of the particle. In the particle-fixed cartesian coordinate systems it reads: $\nabla\psi = \partial_x\psi \mathbf{e}_x + \partial_y\psi \mathbf{e}_y + \partial_z\psi \mathbf{e}_z$, and

$$\nabla_{\parallel}\psi = \mathbf{e}_\theta (\mathbf{e}_\theta \cdot \nabla\psi) + \mathbf{e}_\varphi (\mathbf{e}_\varphi \cdot \nabla\psi). \quad (22)$$

The next step consists in writing the spherical unit vectors in cartesian coordinates and averaging over the surface of the sphere. The algebra is a little bit tedious but presents no difficulty. We end up with

$$\begin{aligned} \langle \mu(\mathbf{r}) \nabla_{\parallel}\psi \rangle \Big|_x &= \frac{\partial_x\psi}{4} \int_0^\pi d\theta \sin\theta \mu(\theta) (1 + \cos^2\theta) \\ &= \frac{\mu + \mu'}{3} \partial_x\psi, \end{aligned} \quad (23a)$$

$$\begin{aligned} \langle \mu(\mathbf{r}) \nabla_{\parallel}\psi \rangle \Big|_y &= \frac{\partial_y\psi}{4} \int_0^\pi d\theta \sin\theta \mu(\theta) (1 + \cos^2\theta) \\ &= \frac{\mu + \mu'}{3} \partial_y\psi, \end{aligned} \quad (23b)$$

$$\begin{aligned} \langle \mu(\mathbf{r}) \nabla_{\parallel}\psi \rangle \Big|_z &= \frac{\partial_z\psi}{2} \int_0^\pi d\theta \sin\theta \mu(\theta) (1 - \cos^2\theta) \\ &= \frac{\mu + \mu'}{3} \partial_z\psi, \end{aligned} \quad (23c)$$

so that the linear velocity is obtained according to Eqs. (1) and (11)

$$\mathbf{u} = -\xi_1 \frac{\mu + \mu'}{3} \nabla\psi. \quad (24)$$

We proceed in a similar manner for the angular velocity. We first note that $\mathbf{e}_r \times \nabla_{\parallel}\psi = \mathbf{e}_r \times \nabla\psi$. Averaging over the surface of the particle, we find

$$\begin{aligned} \langle \mu(\mathbf{r}) \mathbf{e}_r \times \nabla\psi \rangle \Big|_x &= -\frac{\partial_y\psi}{2} \int_0^\pi d\theta \sin\theta \cos\theta \mu(\theta) \\ &= \frac{\mu - \mu'}{4} \partial_y\psi, \end{aligned} \quad (25a)$$

$$\begin{aligned} \langle \mu(\mathbf{r}) \mathbf{e}_r \times \nabla\psi \rangle \Big|_y &= \frac{\partial_x\psi}{2} \int_0^\pi d\theta \sin\theta \cos\theta \mu(\theta) \\ &= -\frac{\mu - \mu'}{4} \partial_x\psi, \end{aligned} \quad (25b)$$

$$\langle \mu(\mathbf{r}) \mathbf{e}_r \times \nabla\psi \rangle \Big|_z = 0. \quad (25c)$$

Finally, given the fact that $\mathbf{n} \times \nabla\psi_{\text{ext}} = (-\partial_y\psi, \partial_x\psi, 0)$, we get

$$\mathbf{\Omega} = \xi_1 \frac{3(\mu - \mu')}{8a} \mathbf{n} \times \nabla\psi, \quad (26)$$

so that $\mathbf{\Omega} = \mathbf{n} \times \mathbf{A}$ with $\mathbf{A} = 3\xi_1(\mu - \mu')/(8a) \nabla\psi$.

IV. SELF-GENERATED FIELD ψ_S WITH UNIFORM SOURCE

Here we evaluate the field ψ_S generated due to the active surface of the JP. Its precise form depends on the permeability of the particle's material for ψ_S , and on the expression of the source at the active segment of the JP. We discuss the case of temperature, $\psi = T$, which is readily generalized to other mechanisms. In a second step we evaluate self-propulsion of the particle due its own field gradient $\nabla\psi_S$, and calculate its linear velocity u_0 . The angular velocity vanishes for a uniform source field.

In our analysis we assume that, beyond the active surface, the field ψ satisfies Laplace's equation $\nabla^2\psi = 0$. For the temperature field this is always the case; because of the large heat diffusivity, advection is irrelevant and the corresponding Péclet number is small. Deviations may occur, however, in the case of a concentration field. At moderate solution concentration, advective transport in the interaction layer is slower than diffusion. For a strong interaction potential, however, the Péclet number is not necessarily small, thus requiring corrections to the simple boundary layer picture used here.

A. Source field

We consider the case where the metal or carbon capped hemisphere is heated through plasmonic absorption of resonant laser light. Then the stationary field satisfies Fourier's diffusion equation

$$\kappa \nabla^2 T(\mathbf{r}) = Q(\mathbf{r}) \quad (27)$$

with heat conductivity κ . The absorbed power density Q is the product of the absorption coefficient β and the intensity of the laser beam $I(\mathbf{r})$.

The metal layer of a sufficiently thin cap can be replaced by a halfsphere of zero thickness [3]. Then the continuity of flux at the particle surface ($r = a$) reads

$$-[\kappa_s \partial_r T_s - \kappa_p \partial_r T_p]_{r=a} = Q(\theta), \quad (28)$$

with

$$Q(\theta) = \begin{cases} 0 & \text{for } 0 \leq \theta \leq \frac{\pi}{2}, \\ Q_0 & \text{for } \frac{\pi}{2} \leq \theta \leq \pi. \end{cases} \quad (29)$$

Expansion in Legendre polynomials P_n gives the series

$$Q(\theta) = \frac{Q_0}{2} \sum_{n=0}^{\infty} (2n+1) q_n P_n(\cos\theta). \quad (30)$$

The even coefficients vanish except for q_0 ,

$$q_{2n} = \delta_{n0}, \quad (31a)$$

whereas the odd ones are given by

$$q_{2n+1} = \int_{-1}^0 dc P_{2n+1}(c) = \frac{(-1)^{n+1} (2n)!}{2^{2n+1} n! (n+1)!} \quad (31b)$$

(The above definition of $Q(\theta)$ corresponds to $q(\pi - \theta)$ of our previous work [3], and the present $Q_0 \frac{2n+1}{2} q_n$ to $(-1)^n q_n$ of [3].)

B. Temperature field

In order to solve Fourier's equation (27) we expand the temperature field in the liquid phase,

$$T = T_0 + \sum_{n=0}^{\infty} \alpha_n \frac{a^{n+1}}{r^{n+1}} P_n(\cos \theta); \quad (32)$$

a similar series occurs within the particle, with the same coefficients α_n yet powers r^n/a^n . Inserting these expressions in the boundary condition (28) one finds

$$[(n+1)\kappa_s + n\kappa_p] \alpha_n = \frac{aQ_0}{2} (2n+1) q_n.$$

For later convenience we introduce the factors

$$\xi_n = \frac{(2n+1)\kappa_s}{(n+1)\kappa_s + n\kappa_p}. \quad (33)$$

and thus have the finite coefficients of the temperature field read as

$$\alpha_0 = \frac{aQ_0}{2\kappa_s}, \quad \alpha_{2n+1} = \xi_{2n+1} \frac{aQ_0}{2\kappa_s} \frac{(-1)^{n+1} (2n)!}{2^{2n+1} n! (n+1)!} \quad (34)$$

The parallel component of the gradient is obtained as the derivative with respect to the polar angle, $\partial_\theta = -\sin \theta \partial_c$, with $c = \cos \theta$ and $r = a$,

$$\nabla_{\parallel} T = \hat{\theta} \frac{\partial_\theta T}{a} = -\hat{\theta} \frac{\sin \theta}{a} \sum_{n=0}^{\infty} \alpha_n \partial_c P_n(c). \quad (35)$$

C. Particle velocity u_0

The self-generated temperature gradient $\nabla_{\parallel} T$ is symmetric with respect to the particle axis \mathbf{n} . Thus the surface integral in the average of the slip velocity, $\mathbf{n}u_0 = -\langle \mu(\hat{\mathbf{r}}) \nabla_{\parallel} T \rangle$, simplifies significantly. Eq. (29) defines polar coordinates such that $r = a$ and $\theta = 0$ indicates the pole of the insulating part of the JP; in other words

$$\mathbf{n} \cdot \hat{\mathbf{r}} = \cos \theta, \quad \mathbf{n} \cdot \hat{\theta} = -\sin \theta.$$

From (35) one obtains

$$u_0 = \left\langle \mu(\mathbf{r}) \sin \theta \frac{\partial_\theta T}{a} \right\rangle.$$

With the shorthand notation $c = \cos \theta$ the surface integral becomes

$$u_0 = -\frac{\mu'}{2a} \sum_{n=1}^{\infty} \alpha_n \int_0^1 dc (1-c^2) \partial_c P_n - \frac{\mu}{2a} \sum_{n=1}^{\infty} \alpha_n \int_{-1}^0 dc (1-c^2) \partial_c P_n. \quad (36)$$

From the relation

$$(1-c^2) \partial_c P_n = n(P_{n-1} - cP_n) \quad (37)$$

one readily finds that the integrals vanish for $n = 3, 5, 7, \dots$. Since, on the other hand, all even coefficients $\alpha_2, \alpha_4, \alpha_6, \dots$ are zero according to (31a) and (34), the only finite contribution is provided by the term $n = 1$. Both remaining integrals take the value $\frac{1}{3}$, resulting in

$$u_0 = \xi_1 \frac{\mu + \mu'}{3} \frac{Q_0}{8\kappa_s}. \quad (38)$$

The parameter $-Q_0/8\kappa_s$ gives the mean gradient of the self-generated temperature field. Since self-propulsion arises from the dipolar term ($n = 1$) only, its deformation factor is the same as that of the thermophoretic velocity (24). Positive u_0 means that the particle moves the insulating part ahead, and thus opposite to its self-generated temperature gradient.

In the case where ψ denotes the concentration of a molecular species involved in a chemical reaction at the particle surface, the parameter Q_0 is related to the activity, and κ_i are the diffusion coefficients. Since in general $\kappa_p = 0$, we have $\xi_1 = \frac{3}{2}$.

D. Angular velocity Ω_0

Finally we consider the viscous torque

$$\mathbf{\Omega}_0 = -\frac{3}{2a} \langle \mu(\hat{\mathbf{r}}) \hat{\mathbf{r}} \times \nabla_{\parallel} T \rangle \quad (39)$$

exerted by the self-generated temperature field. Since the argument of the surface average is of axial symmetry, the angular velocity vanishes,

$$\mathbf{\Omega}_0 = 0,$$

for a source field Q_0 in (29) that is constant over the cap of the JP.

V. SELF-GENERATED FIELD ψ_S WITH NON-UNIFORM SOURCE

Here we consider a self-driven particle with a non-uniform source field. In the case of a laser-heated metal-capped JP, a non-uniform source Q_0 occurs if the intensity of the laser beam $I(\mathbf{r})$ varies along the particle surface. The catalytic activity can vary over the particle surface by a concentration gradient or, in the case of photoactive driving, by a non-uniform light intensity.

We consider the case of temperature, $\psi = T$. First we evaluate the temperature gradient due to a non-uniform intensity of the laser beam, and then calculate the resulting angular velocity Ω_0 of the JP. The linear velocity u_0 calculated in the preceding section, is hardly affected the non-uniform external source field.

A. Non-uniform laser intensity

Expanding the laser intensity to linear order about the position \mathbf{r}_0 of the center of the JP, we find that the absorbed power density $Q(\mathbf{r})$ carries a correction factor

$$Q(\theta) (1 + a \hat{\mathbf{r}} \cdot \boldsymbol{\tau}), \quad (40)$$

where $Q(\theta)$ is defined in (29) with the constant $Q_0 = \beta I(\mathbf{r}_0)$, and where we have introduced the logarithmic derivative

$$\boldsymbol{\tau} = \nabla \ln I(\mathbf{r}_0).$$

For a Gaussian profile $I(\mathbf{r}) = I_0 e^{-r^2/2\omega^2}$ of width ω , one has $\boldsymbol{\tau} = \mathbf{r}_0/\omega^2$. Note that $\boldsymbol{\tau}$ has the dimension of an inverse length. The corrections due to non-uniform heating carry a small parameter $a\tau \sim a/\omega$ which is significantly smaller than unity.

It turns out convenient to use spherical coordinates r, θ, φ but to express vectors in the cartesian basis $\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z$, which is chosen such that the z -direction coincides with the axis of the JP, $\mathbf{e}_z = \mathbf{n}$, and that $\boldsymbol{\tau}$ lies in the $x-z$ -plane,

$$\boldsymbol{\tau} = \tau_x \mathbf{e}_x + \tau_z \mathbf{e}_z.$$

For later use we give

$$\begin{aligned} \hat{\mathbf{r}} &= \mathbf{e}_x \sin \theta \cos \varphi + \mathbf{e}_y \sin \theta \sin \varphi + \mathbf{e}_z \cos \theta, \\ \hat{\boldsymbol{\theta}} &= \mathbf{e}_x \cos \theta \cos \varphi + \mathbf{e}_y \cos \theta \sin \varphi - \mathbf{e}_z \sin \theta, \\ \hat{\boldsymbol{\varphi}} &= -\mathbf{e}_x \sin \varphi + \mathbf{e}_y \cos \varphi. \end{aligned}$$

Inserting the scalar product in the source field, we find for the modulation factor in (40)

$$1 + a \hat{\mathbf{r}} \cdot \boldsymbol{\tau} = 1 + a\tau_z \cos \theta + a\tau_x \sin \theta \cos \varphi.$$

The second term on the right-hand side is independent of the azimuthal angle φ and merely results in a slight modification of axisymmetric temperature field discussed in the previous section.

Thus in the following, we keep only the correction proportional to τ_x , which depends on φ and thus breaks the symmetry of the temperature field with respect to the particle axis,

$$Q(\theta) (1 + a \hat{\mathbf{r}} \cdot \boldsymbol{\tau}) = Q(\theta) [1 + a\tau_x \sin \theta \cos \varphi]. \quad (41)$$

Expanding the factor $Q(\theta) \sin \theta$ in terms of associated Legendre polynomials $P_{n1}(\cos \theta)$ results in

$$Q(\theta) \sin \theta = \frac{Q_0}{2} \sum_{n=1}^{\infty} (2n+1) \hat{q}_n P_{n1}(\cos \theta).$$

The coefficients are given by

$$\hat{q}_n = \frac{1}{n(n+1)} \int_{-1}^0 dc \sqrt{1-c^2} P_{n1}(c).$$

where we have used $\cos \theta = c$ and $\sin \theta = \sqrt{1-c^2}$, and the normalization

$$\int_{-1}^1 dc P_{n1}^2(c) = \frac{2n(n+1)}{(2n+1)}.$$

For further use we note the definition

$$P_{n1} = -\sqrt{1-c^2} \partial_c P_n \quad (42a)$$

and the relation

$$(1-c^2) \partial_c P_n = -n(cP_n - P_{n-1}), \quad (42b)$$

and thus find

$$\hat{q}_n = \frac{1}{(n+1)} \int_{-1}^0 dc (cP_n - P_{n-1})$$

From the symmetry properties of the Legendre polynomials P_n it is clear that the coefficients of odd order $n = 3, 5, 7, \dots$ vanish; the finite ones read

$$\hat{q}_1 = -\frac{1}{3}, \quad \hat{q}_{2m} = (-1)^{m+1} \frac{(2m-2)!}{4^m (m+1)! (m-1)!}. \quad (43)$$

B. Temperature field

The temperature field comprises an axisymmetric part T_{as} , which is essentially given by (32), and a second term that shows the same angular dependence as the correction to $Q(\theta)$,

$$T = T_{\text{as}} + a\tau_x \cos \varphi \sum_{n=1}^{\infty} \hat{\alpha}_n \frac{a^{n+1}}{r^{n+1}} P_{n1}(c),$$

Then the heat diffusion equation is readily solved. The continuity condition (28) for the radial heat flux at the particle surface, relates the $\hat{\alpha}_n$ and \hat{q}_n according to

$$[(n+1)\kappa_s + n\kappa_p] \hat{\alpha}_n = \frac{aQ_0}{2} (2n+1) \hat{q}_n.$$

Inserting the factors ξ_n defined in (33), one finds

$$\hat{\alpha}_n = \xi_n \hat{q}_n \frac{aQ_0}{2\kappa_s}, \quad (44)$$

The component of the temperature gradient parallel to the particle surface are given by the derivatives with respect to θ and φ . With $\partial_\varphi \cos \varphi = -\sin \varphi$ and $\partial_\theta P_{n1} = -\sin \theta \partial_c P_{n1}(c)$ one readily finds

$$\begin{aligned} \nabla_{\parallel} T &= \nabla_{\parallel} T_{\text{as}} - \tau_x \sum_{n=1}^{\infty} \hat{\alpha}_n \times \\ &\quad \left(\hat{\boldsymbol{\varphi}} \sin \varphi P_{n1} + \hat{\boldsymbol{\theta}} \cos \varphi \sin \theta \partial_c P_{n1} \right). \end{aligned} \quad (45)$$

C. Angular velocity Ω_0

Inserting the above temperature gradient in (39) and noting that the cross-product of the axisymmetric part vanishes, $\langle \hat{\mathbf{r}} \times \nabla_{\parallel} T_{\text{as}} \rangle = 0$, we have

$$\Omega_0 = \frac{3}{2a} \tau_x \sum_{n=1}^{\infty} \hat{\alpha}_n \times \left\langle \mu(c) \left(\hat{\mathbf{r}} \times \hat{\boldsymbol{\varphi}} \sin \varphi P_{n1} + \hat{\mathbf{r}} \times \hat{\boldsymbol{\theta}} \cos \varphi \sin \theta \partial_c P_{n1} \right) \right\rangle.$$

In a first step we evaluate the average of φ -dependent factors with respect to the azimuthal angle,

$$\begin{aligned} \langle \hat{\mathbf{r}} \times \hat{\boldsymbol{\varphi}} \sin \varphi \rangle_{\varphi} &= -\langle \hat{\boldsymbol{\theta}} \sin \varphi \rangle_{\varphi} = -\frac{\cos \theta}{2} \mathbf{e}_y, \\ \langle \hat{\mathbf{r}} \times \hat{\boldsymbol{\theta}} \cos \varphi \rangle_{\varphi} &= \langle \hat{\boldsymbol{\varphi}} \cos \varphi \rangle_{\varphi} = \frac{1}{2} \mathbf{e}_y, \end{aligned}$$

and obtain

$$\Omega_0 = -\frac{3}{4a} \tau_x \mathbf{e}_y \sum_{n=1}^{\infty} \hat{\alpha}_n \left\langle \mu(c) (\cos \theta P_{n1} - \sin \theta \partial_c P_{n1}) \right\rangle.$$

Since $\mu(c) = \mu'$ for $c > 0$ and $\mu(c) = \mu$ for $c < 0$, the integrals simplify to

$$\left\langle \mu(c) (\cos \theta P_{n1} - \sin \theta \partial_c P_{n1}) \right\rangle = (\mu' + (-1)^n \mu) k_n,$$

with

$$k_n = \int_0^1 dc \left(c P_{n1} - \sqrt{1-c^2} \partial_c P_{n1} \right).$$

In view of $\hat{\alpha}_n$ and \hat{q}_n , we only need to calculate k_1 and k_{2n} . Using (42a) we find

$$k_1 = -\frac{5}{6}, \quad k_{2n} = 1 - \frac{\pi}{2} \frac{(2n+2)!(2n-2)!}{[4^n(n+1)!(n-1)!]^2}. \quad (46)$$

Noting $\hat{\alpha}_{2n+1} = 0$ for $n > 1$, we have

$$\Omega_0 = -\frac{3}{4a} \tau_x \mathbf{e}_y \left((\mu' - \mu) \hat{\alpha}_1 k_1 + (\mu' + \mu) \sum_{n=1}^{\infty} \hat{\alpha}_{2n} k_{2n} \right).$$

Inserting $\hat{\alpha}_{2n}$ from (44), summing the even-order terms in

$$\tilde{\xi} = \sum_{m=1}^{\infty} \xi_{2m} \hat{q}_{2m} k_{2m}, \quad (47)$$

and introducing the cross product $\mathbf{n} \times \boldsymbol{\tau} = \tau_x \mathbf{e}_y$, we obtain the form

$$\Omega_0 = -(\mathbf{n} \times \boldsymbol{\tau}) \frac{3Q_0}{8\kappa_s} \left(\frac{5}{6} (\mu' - \mu) \xi_1 + (\mu' + \mu) \tilde{\xi} \right) \quad (48)$$

Note that Q_0/κ_s gives the self-generated temperature gradient along the particle surface. With the definition

of the intensity gradient $\boldsymbol{\tau}$, one finds the approximate relation between angular and linear velocities, $\Omega_0 \approx \tau u_0$.

For convenience we give the first terms of the series

$$\tilde{\xi} = \frac{\xi_2}{8} \left(1 - \frac{3\pi}{16} \right) - \frac{\xi_4}{48} \left(1 - \frac{5\pi}{64} \right) + \dots$$

For the case where the diffusivities in particle and solvent are the same, $\kappa_p = \kappa_s$, we have $\xi_n = 1$ and the numerical value $\tilde{\xi} \approx 0.040$. On the other hand, if the diffusivity in the particle vanishes, $\kappa_p = 0$, the factors $\xi_n = \frac{2n+1}{n+1}$ result in $\tilde{\xi} \approx 0.065$.

VI. PROBABILITY DISTRIBUTION FUNCTIONS

A. Angular distribution function

A Janus particle in an external field tends to align its director \mathbf{n} with the field according to Eq. (26), whereas rotational diffusion favors dispersion. As a result, the orientation of the particle follows a distribution $f(\mathbf{n}, t)$ that satisfies a continuity equation

$$\partial_t f = -\nabla_{\mathbf{n}} \cdot \mathcal{J}. \quad (49)$$

where the total probability current \mathcal{J} is the sum of a convective part with angular velocity $\boldsymbol{\Omega}$ and a diffusive part with rotational diffusion coefficient D_r . In Eq. (49), $\nabla_{\mathbf{n}}$ is the gradient with respect to the orientation of the Janus particle. Introducing the rotation operator $\mathcal{R} = \mathbf{n} \times \nabla_{\mathbf{n}}$, the Smoluchowski equation for the probability distribution becomes [4]

$$\partial_t f = -\mathcal{R} \cdot (\boldsymbol{\Omega} f) + D_r \mathcal{R}^2 f \equiv \mathcal{L}_{\mathbf{n}} f. \quad (50)$$

In spherical coordinates one has $\mathbf{n} = \mathbf{e}_r$ and $\nabla_{\mathbf{n}}(\dots) = \partial_{\theta}(\dots) \mathbf{e}_{\theta} + (\sin \theta)^{-1} \partial_{\varphi}(\dots) \mathbf{e}_{\varphi}$, with θ the angle between \mathbf{n} and \mathbf{e}_z . Without loss of generality it can be assumed that $\mathbf{A} = A \mathbf{e}_z$. (Here and in the following the vector \mathbf{A} comprises both contributions, \mathbf{A} and \mathbf{A}_0 , discussed in the main paper.) We then have $\boldsymbol{\Omega} = \mathbf{n} \times \mathbf{A} = -A \sin \theta \mathbf{e}_{\varphi}$, so that the rotational diffusion operator reads

$$\mathcal{L}_{\mathbf{n}} f = A \partial_{\theta} (\sin \theta f) + D_r \partial_{\theta}^2 f. \quad (51)$$

The steady-state solution of the Smoluchowski equation is readily obtained

$$f(\theta) = \frac{A}{2D_r \sinh(A/D_r)} e^{(A/D_r) \cos \theta}. \quad (52)$$

The (thermodynamic) average of the orientation vector is then $\overline{n_x} = \overline{n_y} = 0$, and

$$\overline{n_z} = \coth \frac{A}{D_r} - \frac{D_r}{A}. \quad (53)$$

B. Probability distribution function

Next, we consider a set of active Janus particles. Each particle self-propels in its own non-uniform field with velocity $u_0 \mathbf{n}$ — see Eq. (38). The field due to the neighbors plays the role of an external field and results in the translational and rotational velocities \mathbf{u} and $\boldsymbol{\Omega}$ given by Eqs. (24) and (26), respectively. The corresponding probability distribution function $P(\mathbf{r}, \mathbf{n}, t)$ obeys the equation

$$\partial_t P = -\nabla \cdot (u_0 \mathbf{n} + \mathbf{u} - D \nabla) P + \mathcal{L}_n P, \quad (54)$$

with \mathcal{L}_n defined in Eq. (50). Note that the translational and rotational diffusion coefficients are related through $D = \frac{4}{3} a^2 D_r$. We then decompose $P(\mathbf{r}, \mathbf{n}, t)$ as

$$P(\mathbf{r}, \mathbf{n}, t) = \rho(\mathbf{r}, t) + \mathbf{n} \cdot \mathbf{p}(\mathbf{r}, t) + (\mathbf{nn} - \mathbf{I}/3) : \mathbf{Q}(\mathbf{r}, t) + \dots, \quad (55)$$

where the density ρ , the polarization vector \mathbf{p} , the quadrupolar tensor \mathbf{Q} , and higher moments are functions of \mathbf{r} and t but not \mathbf{n} . Explicitly, the first moments read

$$\rho(\mathbf{r}, t) = \int d\hat{\mathbf{n}} P(\mathbf{r}, \mathbf{n}, t), \quad (56)$$

$$\mathbf{p}(\mathbf{r}, t) = \int d\hat{\mathbf{n}} \mathbf{n} P(\mathbf{r}, \mathbf{n}, t), \quad (57)$$

$$\mathbf{Q}(\mathbf{r}, t) = \int d\hat{\mathbf{n}} (\mathbf{nn} - \mathbf{I}/3) P(\mathbf{r}, \mathbf{n}, t), \quad (58)$$

with $\hat{\mathbf{n}} = \mathbf{n}/(4\pi)$.

1. Evolution equation for the density

We first focus on the particle density $\rho(\mathbf{r}, t)$. Integrating Eq. (54) over \mathbf{n} and using the fact that $\int d\hat{\mathbf{n}} \nabla \dots = \nabla \cdot (\int d\hat{\mathbf{n}} \dots)$, it is straightforward to get

$$\partial_t \rho = -\nabla \cdot (u_0 \mathbf{p} + \mathbf{u} \rho - D \nabla \rho). \quad (59)$$

Note that the contribution from rotational diffusion vanishes since

$$\int d\hat{\mathbf{n}} \nabla_{\mathbf{n}} \cdot \mathcal{J} = 0. \quad (60)$$

2. Evolution equation for the polarization

We now consider the polarization vector $\mathbf{p}(\mathbf{r}, t)$. Multiplying Eq. (54) by \mathbf{n} and integrating over \mathbf{n} yields

$$\begin{aligned} \partial_t \mathbf{p} = & -\nabla \cdot \left(u_0 \int d\hat{\mathbf{n}} P \mathbf{nn} \right) \\ & - \nabla \cdot (\mathbf{u} \mathbf{p} - D \nabla \mathbf{p}) + \int d\hat{\mathbf{n}} \mathbf{n} \mathcal{L}_n P. \end{aligned} \quad (61)$$

Let us discuss separately the first and the third terms on the RHS of Eq. (61). Writing $\mathbf{nn} = \mathbf{I}/3 + (\mathbf{nn} - \mathbf{I}/3)$, we simply get for the first term

$$\int d\hat{\mathbf{n}} P \mathbf{nn} = \frac{\mathbf{I}}{3} \rho + \mathbf{Q}. \quad (62)$$

The third term is the sum of two contributions

$$\begin{aligned} \int d\hat{\mathbf{n}} \mathbf{n} \mathcal{L}_n P &= - \int d\hat{\mathbf{n}} \mathbf{n} \mathcal{R} \cdot (\boldsymbol{\Omega} P) + D_r \int d\hat{\mathbf{n}} \mathbf{n} \mathcal{R}^2 P \\ &\equiv \mathbf{I}_1 + \mathbf{I}_2. \end{aligned} \quad (63)$$

The latter are evaluated thanks to the moment expansion (55). We get for the first integral

$$\begin{aligned} \mathbf{I}_1 &= - \int d\hat{\mathbf{n}} \mathbf{n} \mathcal{R} \cdot [\boldsymbol{\Omega} (\rho + \mathbf{n} \cdot \mathbf{p} + \dots)] \\ &= -\rho \int d\hat{\mathbf{n}} \mathbf{n} \mathcal{R} \cdot \boldsymbol{\Omega} - \mathbf{p} \cdot \int d\hat{\mathbf{n}} \mathbf{n} \mathcal{R} \cdot (\boldsymbol{\Omega} \mathbf{n}) + \dots, \end{aligned} \quad (64)$$

with $\boldsymbol{\Omega} = \mathbf{n} \times \mathbf{A}$, the vector \mathbf{A} being independent of the orientation. Since $\mathcal{R}_\alpha n_\beta = -e_{\alpha\beta\gamma} n_\gamma$, with $e_{\alpha\beta\gamma}$ the Levi-Civita symbol, we find that $\mathcal{R} \cdot \boldsymbol{\Omega} = -2\mathbf{n} \cdot \mathbf{A}$ and so

$$- \int d\hat{\mathbf{n}} \mathbf{n} \mathcal{R} \cdot \boldsymbol{\Omega} = 2 \left(\int d\hat{\mathbf{n}} \mathbf{nn} \right) \cdot \mathbf{A} = \frac{2}{3} \mathbf{A}. \quad (65)$$

Here, the last equality comes from $\int d\hat{\mathbf{n}} n_\alpha n_\beta = \frac{1}{3} \delta_{\alpha\beta}$. The second integral in Eq. (64) vanishes since it involves an odd number of components n_α . We then find

$$\mathbf{I}_1 = \frac{2}{3} \mathbf{A} \rho + \dots, \quad (66)$$

where the terms that are neglected are at least quadrupolar. Regarding the second integral, it reads

$$\begin{aligned} \mathbf{I}_2 &= D_r \int d\hat{\mathbf{n}} \mathbf{n} \mathcal{R}^2 (\rho + \mathbf{n} \cdot \mathbf{p} + \dots) \\ &= D_r \left(\int d\hat{\mathbf{n}} \mathbf{n} \mathcal{R}^2 \mathbf{n} \right) \cdot \mathbf{p} + \dots \end{aligned} \quad (67)$$

Since $\mathcal{R}^2 \mathbf{n} = -6\mathbf{n}$, it is straightforward to get $\mathbf{I}_2 = -2D_r \mathbf{p} + \dots$.

Closure of the above equations is achieved by neglecting quadrupolar and higher-order terms; this approximation is supported by its relation to a gradient expansion [7], which in turn is justified by a rather smooth spatial variation. We thus obtain the evolution equation for the polarization vector

$$\partial_t \mathbf{p} = -\frac{u_0}{3} \nabla \rho - \nabla \cdot (\mathbf{u} \mathbf{p} - D \nabla \mathbf{p}) + \frac{2}{3} \mathbf{A} \rho - 2D_r \mathbf{p}. \quad (68)$$

where the last two terms arise from the rotational operator \mathcal{L}_n . Following Golestanian [6] we assume moreover that both the explicit time derivative and the divergence terms are small. Solving the remainder for \mathbf{p} and replacing $\mathbf{A} \rightarrow \mathbf{A} + \mathbf{A}_0$, we readily obtain the steady-state polarization

$$\mathbf{p} = -\frac{u_0}{6D_r} \nabla \rho + \frac{\mathbf{A}}{3D_r} \rho + \frac{\mathbf{A}_0}{3D_r} \rho. \quad (69)$$

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