Self-motility of an active particle induced by correlations in the surrounding solution

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Current models of phoretic transport rely on molecular forces creating a “diffuse” particle–fluid interface. We investigate theoretically an alternative mechanism, in which a diffuse interface emerges solely due to a non-vanishing correlation length of the surrounding solution. This mechanism can drive self-motility of a chemically active particle. Numerical estimates indicate that the velocity can reach micrometers per second. The predicted phenomenology includes a bilinear dependence of the velocity on the activity and a possible double velocity reversal upon varying the correlation length.

Out-of-equilibrium behavior has been a research field of sustained interest, relevant to understanding the emergence of complexity [1–4]. The last decade witnessed a rapidly growing engagement with self-phoretic, chemically active particles as a new paradigm thereof, which exhibits a wealth of phenomena, such as micro-phase separation [5], self-assembly of super-structures [6], and self-organized patterns of collective motion [7, 8]. Additionally, phoresis has proven to be an effective, versatile transport mechanism at the microscale [9–11], leading to a significant resurgence of interest also from a technological perspective.

Following the landmark review by Anderson [12], the phenomenon of phoresis of particles suspended in a fluid is characterized by the presence of a driving thermodynamic field and by a transition region (also called “diffuse interface”) around the particle. The forces relevant for the phoretic displacement act within this region: its nonvanishing width gives rise to a response of the fluid medium even though the net sum of the forces and torques vanishes. Thus the particle is transported by the driving field while the system (particle plus fluid) is mechanically isolated, which sets this effect fundamentally apart from that of transport by an external force. The thermodynamic driving field usually comprises an externally imposed gradient in one of the state-parameters (such as composition, temperature, etc.) of the fluid, or of an electric field in the case of a charged particle in an electrolyte. The alternative case, in which the particle itself generates the driving field — a scenario also envisioned by Anderson [12] — has recently received much attention under the generic name of “self-phoresis” [13–16]. Experimentally, self-propelled particles have been realized most often in the form of so-called Janus particles. In a typical scenario, such particles are immersed in a multi-component fluid solution and promote catalytic reactions which vary across their surfaces, leading to self-electrophoresis [17, 18] or self-chemiophoresis [5, 13, 19–21]. Alternatively, the particles behave as heat sources, leading to self-thermophoresis through a single-component fluid [22] or self-chemiophoresis via demixing of a critical binary liquid mixture [23].

A common ingredient in the models, which have so far addressed phoretic [12] and self-phoretic [15, 16, 22, 24–27] phenomena, is a direct physical interaction between the particle and the molecules of the surrounding fluid [28]. The corresponding non-vanishing interaction range provides the aforementioned diffuse interfacial region, where the internal forces and torques act and induce motion. Here, we propose an alternative transport mechanism which does not require this direct, extended interaction: instead, the interfacial region is diffuse because the fluid exhibits a non-vanishing correlation length. Although the phenomenology driven by this mechanism could also be called “phoresis” (in accordance with the definition proposed by Anderson [12]), we coin the notion of “correlation–induced transport” in order to distinguish it from the usual one, which necessarily invokes a direct, spatially extended interaction.

Here we focus on the example of chemiophoresis, for which physically insightful analytical expressions can be derived, in order to illustrate the correlation–induced transport. However, the conceptual framework discussed below is expected to exhibit a very general applicability for other types of phoresis — albeit significantly more technically involved. For clarity and simplicity, we consider a rigid, spherical particle (radius $R$), immersed in a solution, i.e., a solvent plus a single solute species (called “the chemical” in the following). The particle is active, i.e., it is a source or sink of the chemical. The two main approximations are a spatially and temporally constant temperature and mass density of the solution, and a slow motion of the particle [29]. Consequently, the state of the solution can be characterized [1] by the instantaneous stationary profiles of, e.g., the mass density of the chemical or, equivalently, of its number density $n(r)$ valid for
small Péclet numbers, and of the velocity field $u(r)$ of the solution valid for small Reynolds and Mach numbers. Thus, $n(r)$ is determined by the constraint of mass conservation for the chemical:

$$\frac{\partial n}{\partial t} = -\nabla \cdot J_{\text{diff}} = 0, \quad J_{\text{diff}} = \Gamma f. \quad (1)$$

Here $\Gamma$ is the mobility of the chemical within the solution, while $f$ is the body force density acting on the chemical and which drives its diffusion. Likewise, $u(r)$ follows from the balance between this body force and the fluid stresses, as expressed by the Stokes equation for incompressible flow,

$$\nabla \cdot \Omega + f = 0, \quad \nabla \cdot u = 0, \quad (2)$$

in terms of the stress tensor $\Omega = \eta \nabla u + (\nabla u)\dagger - p I$ ($\eta$ is the viscosity of the solution, $p$ the pressure field enforcing incompressibility, and $I$ the identity tensor). Complementarily to the slow particle dynamics, one assumes local equilibrium for the chemical [12], so that

$$f(r) = -n(r)\nabla \mu(r), \quad (3)$$

in terms of its chemical potential $\mu$. The latter is modelled by means of a free energy functional [30] (we take spherical coordinates $(r, \theta, \varphi)$ with the origin at the center of the particle; $dV$ denotes the volume element):

$$\mathcal{H}[n] = \int_{|r| > R} dV \left[ h(n) + \frac{1}{2} \lambda^2 |\nabla n|^2 + n W(r) \right], \quad (4)$$

so that

$$\mu(r) = \frac{\delta \mathcal{H}[n]}{\delta n(r)} = h'(n(r)) - \lambda^2 \nabla^2 n(r) + W(r). \quad (5)$$

Here, $h(n)$ is a local free energy density, and $W(r)$ is a potential energy (which has a non-zero and finite range) generated by the particle [31]. In Eq. (4), the term $\propto \lambda^2$ accounts for the range of the molecular interactions between the dissolved molecules of the chemical [32]. The relevant control parameters of this model for the chemical are $\lambda$ and the temperature (implicit in the definition of $\mathcal{H}$). However, it turns out (see, e.g., Eq. (12)) that the results depend only on $\lambda$ and the correlation length $\xi := \lambda/\sqrt{\mathcal{H}'(n_0)}$ for a reference density $n_0$ [33], without the need to specify the dependence to infinity.

Finally, one has to specify boundary conditions: at infinity, a homogeneous equilibrium state is recovered, i.e., a vanishing velocity of the fluid solution and a constant chemical potential due to a reservoir (thereby also fixing the density at a value $n_0$ such that $h'(n_0) = \mu_0$):

$$u(r) \rightarrow 0, \quad \mu(r) \rightarrow \mu_0, \quad \text{as } |r| \rightarrow \infty. \quad (6)$$

On the surface of the particle, we impose a no-slip boundary condition in terms of the translational $(V)$ and angular $(\Omega)$ velocities of the particle,

$$u(r) = V + \Omega \times r, \quad \text{at } |r| = R, \quad (7)$$

the boundary condition for the density, which follows from the surface term of the variation of the free energy functional [34],

$$\lambda^2 e_r \cdot \nabla n(r) = 0, \quad \text{at } |r| = R, \quad (8)$$

and a prescribed current of the chemical, modelling the activity of the particle as a source of the chemical:

$$e_r \cdot J_{\text{diff}}(r) = q\lambda(\theta, \varphi), \quad \text{at } |r| = R. \quad (9)$$

The positive constant $q$ represents the production rate of the chemical per area of the particle, and $\lambda(\theta, \varphi)$ is the dimensionless activity, which can vary along the surface of the particle.

Equations (1–9), together with the condition that the particle does not experience any external force or torque, constitute a complete description for obtaining the velocities $V$ and $\Omega$ of the particle for a prescribed activity function $\lambda$. If $\lambda = 0$, the equilibrium state $\mu(r) = \mu_0$ is a solution of the equations, which implies $V = 0$ and $\Omega = 0$. Thus, particle transport requires the non-equilibrium imbalance introduced by the chemical activity. We address two important aspects for solving this model. First, mechanical isolation of the whole system “particle + solution” holds because, according to Eq. (2), the medium transmitting forces from the particle to infinity is in local mechanical balance [29]. Second, according to Eq. (2), the incompressibility constraint implies that the motion is driven by the spatially extended field $\nabla \times f$, which (because of mechanical isolation) is the only source of flow vorticity [35]. Combining Eqs. (3) and (5), one obtains

$$\nabla \times f = \nabla \mu \times \nabla n - \lambda^2 \nabla \times \{ \nabla \cdot [\mu W(\nabla n)] \}. \quad (10)$$

Therefore, the transport, i.e., $\nabla \times f \neq 0$, requires either an explicit interaction between the particle and the chemical, i.e., $W \neq 0$, or a deviation of the solution from ideality, i.e., $\lambda \neq 0$, which is associated with a non-vanishing correlation length in the solution. To the best of our knowledge, all previous studies of phoresis have analyzed and identified the case $W \neq 0$ as the primary origin of particle transport. Here we address the opposite case ($W = 0, \lambda \neq 0$), for which the model [Eqs. (1–9)] implies that the spatially homogeneous equilibrium state $(n(r) = n_0, u(r) = 0)$ would not be perturbed by the presence of a passive particle, i.e., $q = 0$. This case isolates the effect of correlations and already provides distinctive observable predictions.

A complete analytical solution for the correlation–induced transport velocity can be obtained if the deviations from homogeneity are small, i.e., for small values (weak activity) of the Damköhler number $Da = qR\beta/(n_0\Gamma)$, where $\beta$ is the inverse thermal energy and $\Gamma/\beta$ is the diffusivity of the chemical: the dimensionless ratio $Da$ quantifies the activity–induced “chemical crowding” near the particle against the opposing effect of diffusion. In this case, the spatial variation of $\eta$ and $\Gamma$ (via a
possible dependence on the density \( n \) can be neglected. Consequently, the “diffusion problem” for the density profile \( n(r) \), posed by Eqs. (1, 3, 5, 6, 8, 9), is decoupled from the velocity field and can be solved first on its own. This can be carried out analytically, after linearizing in the small quantity \( n(r) - n_0 \). Subsequently, one solves the “hydrodynamical problem”, stated by Eqs. (2, 6, 7), with the body force in Eq. (3) determined by the profile \( n(r) \) obtained in the first step. Conveniently, the Lorentz reciprocal theorem allows one to sidestep the full solution of the “hydrodynamical problem” and to obtain expressions for \( V \) and \( \Omega \) directly in terms of \( n(r) \). Details of this procedure are collected in the Supplemental Material \[29\]. By expanding the activity in terms of spherical harmonics,

\[
A(\theta, \varphi) = \sum_{\ell=0}^{\infty} \sum_{m=-\ell}^{\ell} a_{\ell m} Y_{\ell m}(\theta, \varphi),
\]

one obtains a compact expression for the correlation-induced transport velocity:

\[
V = V_0 \sum_{\ell, m} \sum_{\ell', m'} a_{\ell m} a_{\ell' m'} \times \left[ g^{\perp}(\xi/R) G^{\perp}_{\ell m, \ell' m'} + g^{\parallel}(\xi/R) G^{\parallel}_{\ell m, \ell' m'} \right]
\]

with the velocity scale \( V_0 := D a^2 R^3/(6\pi \eta^2 \lambda^2) \). In the Supplemental Material we provide \[36\] the rather lengthy expressions for the purely geometrical factors \( G \), which depend only on the mathematical properties of the spherical harmonics, and for the dimensionless coefficients \( g \), which fully incorporate the dependence on the correlation length \( \xi \) brought about by the physical model. The superscripts \( \perp \) and \( \parallel \) denote the contributions by the radial and tangential components of the body force \( f \), respectively. The angular velocity \( \Omega \) turns out to vanish identically (see below).

Several general conclusions can be extracted already from this expression. Since Eq. (12) is bilinear in the activity, the velocity is invariant under the transformation \( a_{\ell m} \rightarrow -a_{\ell m}, \forall \ell, m \). This means that the direction of translation is insensitive to whether the particle acts as a source or a sink of the exchanged chemical. As shown by Eq. (10), a nonvanishing velocity requires a deviation from equilibrium \((\nabla n \neq 0)\), as well as a deviation from homogeneity \((\nabla n \neq 0)\). In the model of correlation-induced transport, both requirements are enforced by the activity, which explains the bilinear dependence. This is at variance with the case of standard phoresis, which gives rise to a velocity linear in the activity \[29\] because \( W \neq 0 \) usually suffices to bring about a relevant inhomogeneity. Another consequence of the symmetric form of the term \( \propto \lambda^2 \) in Eq. (10) is the absence of chirality, so that \( \Omega = 0 \) \[37\]. In the standard model, on the contrary, the relative orientation of \( \nabla n \) and \( \nabla W \) will provide, in general, a preferred direction and sense of rotation, thus inducing a non-vanishing angular velocity.

The geometrical factors \( G_{\ell m, \ell' m'}^{\perp} \) and \( G_{\ell m, \ell' m'}^{\parallel} \) can be expressed in terms of the Wigner 3j symbols \[29\], which include “selection rules”: these factors will vanish unless \( |\ell - \ell'| = 1 \) and \( |m + m'| \leq 1 \). Therefore, the sum in Eq. (12) explicitly couples only pairs of “neighboring multipoles”. This result captures a major difference as compared to standard phoresis. For instance, an activity pattern lacking a dipole in Eq. (11) generically yields \( V \neq 0 \), while the same pattern of activity, together with a spherically symmetric potential \( W(r) \), does not yield phoresis in the standard model \[38\].

Another interesting feature of the transport velocity is revealed by considering the limit \( \xi \ll R \): the deviations from homogeneity are then localized within a thin layer (of thickness \( \propto \xi \)) at the surface of the particle. This case, the velocity \( V \) can be expressed as an integral over the surface of the particle \[29\]:

\[
V \approx -\frac{1}{4\pi} \int_0^\pi d\theta \sin \theta \int_0^{2\pi} d\varphi v_{\text{slip}}(\theta, \varphi).
\]

The integrand can be interpreted as a slip velocity for the “hydrodynamical problem” \[12\], which is proportional to the tangential component of the density gradient,

\[
v_{\text{slip}}(r) := L(r) \nabla \cdot n(r), \quad \text{at } |r| = R,
\]

with a local coefficient of phoretic mobility:

\[
L(\theta, \varphi) := \frac{D a^3}{\eta^2 R} \, \kappa(\theta, \varphi).
\]

Due to the explicit dependence of \( L \) on the activity (via \( D a \)), the present mechanism of self-propulsion cannot be interpreted as “passive” transport in an external (albeit self-induced) gradient, which is another significant deviation from standard phoresis \[39\]. Finally, we note that \( V \propto \xi^3 \) if \( \xi \ll R \) because the tangential gradient vanishes \( \propto \xi^2 \) \[29\]. In the opposite limit \( \xi \gg R \), the velocity given by Eq. (12) approaches a finite value \[40\], which depends only on \( V_0 \) and \( a_{\ell m} \).

As an illustration \[29\], we study the simplest activity distribution with polar symmetry by retaining only monopolar and dipolar contributions: \( a_{00} = 1 \), \( a_{1m} = a_{10} \delta_{m,0} \), and \( a_{2m} = 0 \) in Eq. (11). For this case, Eq. (12) renders \( V = e_1 V_z \), \( V_z = a_{10} V_0 f_1(\xi/R) \), with the function \( f_1(\xi/R) \) shown in Figure 1. The velocity depends linearly on the dipole strength \( a_{10} \) and vanishes in the spherically symmetric limit \( a_{10} \rightarrow 0 \). The asymptotic behaviors for \( \xi \ll R \) or \( \xi \gg R \) are given by the general results just discussed. If an additional quadrupolar activity contribution \( a_{20} \neq 0 \) is included, Eq. (12) gives \( V_z = a_{10} V_0 [f_1(\xi/R) + a_{20} f_2(\xi/R)] \), where \( f_1 \) and \( f_2 \) are monotonic functions of \( \xi \) with opposite signs. Strikingly, this implies that the sign of \( V_z \) can be altered by merely
changing the correlation length $\xi$ (e.g., through temperature), while keeping the particle properties (i.e., the coefficients $a_{\ell m}$) fixed. This rich behavior is illustrated in Fig. 2. For certain values of $a_{20}$, such as $a_{20} = 7$, one indeed observes a reentrant regime where $V_z$ is negative at very small and very large $\xi$, but positive in an intermediate range of correlation lengths.

These results allow one to estimate the magnitude of the velocity under realistic conditions. From Fig. 1, upon taking $a_{10} = 1$, one obtains $V_z \approx V_0 (\xi/R)^3$ in the regime $\xi/R < 1$. In order to estimate the velocity scale $V_0$ (see below Eq. (12)), we approximate the local free energy of the chemical by the ideal gas expression $\beta h(n) = n (\ln n - 1)$, so that $\xi^2 = \beta n_0 \lambda^2$, and

$$V_0 = (Da R/\xi)^2 n_0 R/(6\pi\eta\beta).$$

With the typical values $R = 1 \mu m$, $n_0 = 1 \text{ mM}$, $\eta = 10^{-3}$ Pas (viscosity of water), and $Da = 10^{-4}$ (as for, e.g., the Pt catalyzed decomposition of hydrogen peroxide reported in Refs. [17, 26]), one obtains $V_z \approx (10 \xi/R)^3 \mu m/s$ at room temperature. For $\xi/R = 10^{-1}$, the predicted velocity would be easily measurable; actually, the strong dependence on $\xi/R$ provides a broad range of variation covering the values reported from experiments for distinct types of systems [19–21, 41, 42].

In summary, we have studied a mechanism of phoresis driven by correlations in the solution. Significant differences with respect to the standard mechanism of phoresis become evident: (i) The correlation–induced self-propulsion is bilinear in the activity, so that for the same activity pattern the two mechanisms predict distinct observable velocities. (ii) There is no self-rotation, regardless of the activity pattern. (iii) The correlation–induced phoresis cannot be understood as “passive” phoresis in an external (albeit self-induced) driving field. Already for simple activity patterns, the self-propulsion velocity exhibits remarkable features, including reentrant changes of sign obtained upon varying the correlation length (see Fig. 2). Numerical estimates for realistic conditions give values comparable to those observed experimentally, which are usually interpreted within the framework of the standard model of phoresis; correlation–induced phoresis provides an additional, plausible mechanism for addressing these observations.

Thus, our formalism opens the door to studying the role which correlations play in phoretic phenomena for a broad class of systems and geometries. In the physically relevant situation in which $\xi \neq 0$ and $W \neq 0$, both correlation–induced transport and common phoresis would occur, including a coupling between correlations within the fluid and the inhomogeneities induced by $W(r)$. An extension of our analytical model could address their relative importance [43]. In this regard, the present study constitutes, inter alia, an important addition to the theoretical machinery which is relevant for sorting and interpreting recent results pertaining to self-propulsion due to demixing in a binary liquid mixture (its order parameter playing the role of $n(r)$). For example, this encompasses the numerical analysis in Ref. [25], which incorporates nonlinear couplings (due to a nonvanishing Péclet number) in order to explain the emergence of self-motility, and the experimental observation of velocity reversals for certain self-propelled particles [44] following a change of illumination intensity (which changes the temperature distribution in the fluid and, correspondingly, the correlation length).

To conclude, we have identified and characterized a novel mechanism for self-phoresis of an active particle, which features potentially observable differences in comparison with the mechanism considered so far in the literature, and which can be controlled, e.g., via varying...
the temperature of the bath. A.D. acknowledges support by the Spanish Government through Grant FIS2017-87117-P (partially financed by FEDER funds).

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29. For full details see the Supplemental Material at [URL link], which also cites Refs. [45–54].
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31. For an incompressible solution, $W$ is actually the difference in interaction of the particle with a molecule of solute and with a molecule of solvent, respectively [1].
32. More precisely, $A$ can be directly related to the second moment of the attractive part of the pair potential between the molecules of the chemical.
33. The free energy in Eq. (4), with $W = 0$ and linearized in $\delta n = n - n_0$, provides the equilibrium correlation function $\langle \delta n(r) \delta n(0) \rangle \sim e^{-r/\xi}.\lambda$.
34. In the limit $\lambda \to 0$, this boundary condition becomes actually irrelevant.
35. The force can be decomposed into potential (i.e., rotational) and solenoidal (i.e., divergence-free) components, $f = f_{\text{pot}} + f_{\text{sol}}$. The potential component, $f_{\text{pot}} = -\nabla P_t$, can be absorbed by the (auxiliary) hydrodynamic pressure field, $p \rightarrow p + P_t$, that enforces incompressibility; see also Eqs. (III.19, III.22) in Ref. [29].
36. See Eqs. (III.39–III.42) in Ref. [29].
See the discussion of Eq. (IV.16) in Ref. [29].

See Ref. [15]. There is also the complementary case: a purely dipolar pattern (i.e., the simplest model of a Janus particle with no net production) renders $V = 0$ [29], but in the standard model it yields a non-vanishing phoresis even for a spherically symmetric potential.

Correlation–induced phoresis is also predicted for a passive particle in an external density gradient, yielding a transport velocity which is at least cubic in this gradient [43]. This might explain why it has not received attention, being overshadowed by the mechanism based on an extended interaction.

This conclusion is not trivial because the density field $n(\mathbf{r})$ does require a finite $\xi$ in order to satisfy the boundary condition $n \to n_0$ at infinity [29].

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Self-motility of an active particle induced by correlations in the surrounding solution.

Supplemental Material

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In this Supplemental Material, equations in the main text are referenced by those same numbers. Equations in the Supplemental Material are cited by a combination of the section number (in Roman numerals) and the equation number within the section (in Arabic numerals).
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I. GENERAL CONSIDERATIONS CONCERNING THE BALANCE OF FORCES

The model system, which is analyzed in detail in the present study, consists of the particle and the fluid solution in which it is immersed; the latter comprises a solvent species and a solute species (denoted as “chemical”). The chemical is involved in the activity of the particle (which changes the local composition of the solution). Here we discuss in more detail the conceptual framework of the “force- and torque-free” motility of the active particle, with an emphasis on the identification of the relevant forces. We first consider the force balance.

1. The active particle: The particle is assumed to drift at a constant velocity $V$ and to rotate with a constant angular velocity $\Omega$ relative to the fluid at infinity. We impose no-slip of the fluid solution at the impenetrable surface $\partial D_{\text{part}}$ of the particle (see Eq. (7) in the main text). Since the particle is not acted upon by any external forces, the total force experienced by the particle consists only of the interaction with the fluid,

$$\mathbf{F}^{(\text{part})} = \oint_{\partial D_{\text{part}}} dS \mathbf{e}_n \cdot \mathbf{\Pi}(\mathbf{r}) + \mathbf{F}^{(\text{part})}_{\text{chem}},$$

(I.1)

where we take $\mathbf{e}_n$ to be the unit vector normal to $\partial D_{\text{part}}$ and pointing into the fluid. The first term involves the hydrodynamic stress tensor $\mathbf{\Pi}$ [1], and the second term is the explicit force that the chemicals exert on the particle. In the overdamped regime, for which our model is formulated, the accelerations are negligible, so that the total force on the particle will vanish:

$$\mathbf{F}^{(\text{part})} = 0.$$  

(I.2)

2. The fluid solution: In the regime of low Reynolds and Mach numbers, and assuming a steady state, the dynamics of the solution is described by the Stokes equations for incompressible creeping flow (see Eq. (2)). The integral version of this equation,

$$\int_D dV \nabla \cdot \mathbf{\Pi}(\mathbf{r}) + \int_D dV \mathbf{f}(\mathbf{r}) = 0,$$

(I.3)

where $D$ denotes the volume of the solution, describes the mechanical balance in bulk between the hydrodynamic forces and the force density field $\mathbf{f}(\mathbf{r})$, which represents the force acting on the molecules of the chemical in a volume element located at $\mathbf{r}$. The force $\mathbf{f}(\mathbf{r})$ arises from interactions with other molecules of the chemical and from any external field created by the particle.

3. Boundary condition at infinity and mechanical isolation: At infinity we impose (see Eq. (6) in the main text) that the fluid solution is at rest and that the perturbation of the density of the chemical also vanishes. This ensures that any local effect of the particle on its environment does not “propagate too far”; actually, since the total system “particle + fluid solution” is mechanically isolated, the combination of Eqs. (I.1–I.3) renders

$$0 = \mathbf{F}^{(\text{total})} = \text{force on particle} + \text{force on fluid solution}$$

$$= \mathbf{F}^{(\text{part})} + \int_D dV \nabla \cdot \mathbf{\Pi}(\mathbf{r}) + \int_D dV \mathbf{f}(\mathbf{r})$$

$$= \mathbf{F}^{(\text{part})}_{\text{chem}} + \oint_{\partial D_{\infty}} dS (-\mathbf{e}_n) \cdot \mathbf{\Pi}(\mathbf{r}) + \int_D dV \mathbf{f}(\mathbf{r}),$$

(I.4)

where the volume $D$ is bounded by the surface $\partial D_{\text{part}}$ of the particle and the surface $\partial D_{\infty}$ at infinity, with the unit normal $\mathbf{e}_n$ oriented towards the fluid. Due to the principle of action–reaction, the “chemical” force experienced by the particle is balanced by the total force experienced by the chemical in the fluid solution:

$$0 = \mathbf{F}^{(\text{part})}_{\text{chem}} + \int_D dV \mathbf{f}(\mathbf{r}).$$

(I.5)

Thus one concludes from Eq. (I.4) that

$$\oint_{\partial D_{\infty}} dS \mathbf{e}_n \cdot \mathbf{\Pi}(\mathbf{r}) = 0.$$  

(I.6)

Therefore, at infinity the flow must decay sufficiently fast so that the integrated hydrodynamic stress vanishes far from the particle. This would not be the case if the particle would experience an external force.
The argument can be repeated, without conceptual changes, in order to address the torque balance. In particular, it is found that the total torque exerted on the particle can be written as the sum of hydrodynamical and chemical contributions:

\[ \tau^{(\text{part})} = \int d\mathbf{r} \times (\nabla \cdot \Pi(\mathbf{r})) + \int d\mathbf{r} \times f(\mathbf{r}) = 0, \]  

(I.7)

which vanishes due to the balance condition expressed in Eq. (I.3). Additionally, the integrated hydrodynamic torque at infinity vanishes:

\[ \oint_{\partial D_{\infty}} d\mathbf{s} \times (e_n \cdot \Pi(\mathbf{r})) = 0. \]  

(I.8)

Recalling Eqs. (3, 5), the force density can be written as

\[ f = -n \nabla \mu = -n \nabla W - nh''(n) \nabla n + \lambda^2 n \nabla (\nabla^2 n) = -n \nabla W + \nabla \cdot \Pi_{\text{chem}}, \]  

(I.9)

in terms of a chemical (or osmotic) stress tensor (sometimes called the Korteweg stress tensor) \[ \Pi_{\text{chem}} : = \lambda^2 \left[ n \nabla \nabla n - \frac{1}{2} \nabla |n|^2 I \right] - 1 p_{\text{chem}}(n), \]  

(I.10)

where the function

\[ p_{\text{chem}}(n) : = nh'(n) - h(n) = \int_0^n d\hat{n} \hat{n} h''(\hat{n}) \]  

(I.11)

represents the osmotic pressure by the chemical in the limit \( \lambda \to 0 \). (Note that for \( n \to 0 \) (ideal gas) one has \( h(n) \to k_B T \ln n \) and \( p_{\text{chem}} \to nk_B T \).) Accordingly, the force balance arguments above can be formulated in an equivalent manner in terms of the chemical stress tensor. The total force on the particle, as given by Eqs. (I.1) and (I.5), turns into

\[ F^{(\text{part})} = \int d\mathbf{r} n \nabla W + \oint_{\partial D_{\text{part}}} d\mathbf{s} e_n \cdot (\Pi + \Pi_{\text{chem}}) \]

\[ = \int d\mathbf{r} n \nabla W - \oint_{\partial D_{\text{part}}} d\mathbf{s} e_n \left[ p + p_{\text{chem}} + \lambda^2 \left( \frac{1}{3} n \nabla^2 n \right) \right] \]

\[ + \oint_{\partial D_{\text{part}}} d\mathbf{s} e_n \cdot \left\{ \eta \left[ \nabla \mathbf{u} + (\nabla \mathbf{u})^\dagger \right] + \lambda^2 n \left[ \nabla \nabla n - \frac{1}{3} \nabla^2 n \right] \right\}, \]

(I.12)

after using the explicit expressions in Eqs. (I.10) and (III.2) for the chemical and the hydrodynamical stress tensor, respectively. We note that the total osmotic pressure, represented by the term \( p_{\text{chem}} \), and the trace of the correlation-induced contribution, is actually irrelevant for the dynamics, as it can be absorbed into the anyhow unspecified dynamic pressure \( p \), which enforces the incompressibility of the fluid flow. Thus the only forces being effective in driving transport are the distant interaction represented by \( W \), and the local, correlation-induced osmotic stress, which is proportional to \( \lambda^2 \). This is a restatement of the conclusions already extracted from Eq. (10) in the main text.

**II. THE DIFFUSION PROBLEM**

The “diffusion problem” for the density profile \( n(\mathbf{r}) \) is given by Eqs. (1, 3, 5, 6, 8, 9) with \( W = 0 \). This set of equations was derived under the assumption of slow particle motion so that, on the time scale of the particle motion, the distribution of chemicals can be considered to be in the stationary state associated with the instantaneous particle position. More specifically, if the particle moves with velocity \( V \), changes in the configuration will be noticeable after a time of the order \( t_{\text{part}} \sim R/V \) of particle displacement by a sizeable fraction of itself. On the other hand, the
diffusive dynamics of the chemicals is characterized by a diffusion coefficient
\( D_{\text{chem}} = \Gamma / \beta \), in terms of the chemical mobility \( \Gamma \) and the inverse thermal energy \( \beta \), so that the time it takes the chemical to smooth out perturbations over a length scale \( R \) is of the order of \( t_{\text{chem}} \sim R^2 / D_{\text{chem}} \). Consequently, the dimensionless ratio

\[
\frac{t_{\text{chem}}}{t_{\text{part}}} \sim \frac{VR}{D_{\text{chem}}}
\]

quantifies what it means to be “slow” from the perspective of the dynamics of the chemical. With the typical numbers \( D_{\text{chem}} \approx 10^{-9} \text{m}^2/\text{s} \) for molecular diffusion and the observed self-phoretic velocity \( V \approx \mu \text{m/s} \) for micron-sized particles, this gives a ratio \( \approx 10^{-3} \). Thus, there is a clear separation of time scales and the diffusion problem may be treated to be stationary. An analogous reasoning allows one to quantify the relevance of the convective transport by the hydrodynamic flow, characterized also by the velocity scale \( V \) or the spatial range of the interaction potential \( W \). The diffusion is able to smooth out the distortions induced by the hydrodynamic flow, characterized also by the velocity scale \( V \) of the particle motion, provided the time ratio \( VL / D_{\text{chem}} \) (which is actually the Péclet number) is small. An estimate of the length \( L \) can be either the particle radius \( R \) or an intrinsic length related to the chemical distribution, e.g., the correlation length \( \xi \) (see, c.f., Eq. (II.4)) or the spatial range of the interaction W(\( r \)). For the numbers quoted above, the Péclet number is small as long as the scale \( L \) lies below the millimeter (\( \sim 10^3 R \)).

The diffusion problem can be solved analytically in the limit that the deviations \( \delta n(r) = n(r) - n_0 \) from homogeneity are small, with \( n_0 \) being fixed according to \( \mu_0 = h'(n_0) \). As it will be shown, these deviations can be quantified in terms of the dimensionless ratio

\[
\text{Da} := \frac{qR \beta}{n_0 \Gamma} = \frac{qR}{n_0 D_{\text{chem}}},
\]

which, in the language of chemical kinetics, is the Damköhler number of the chemical. According to the boundary condition in Eq. (9), which models the effect of activity, the ratio \( q / n_0 \) can be interpreted as the effective velocity of influx current of chemicals due to activity. On the other hand, as we have seen, \( t_{\text{chem}} \sim R^2 / D_{\text{chem}} \) is the characteristic time for diffusion to smear out inhomogeneities on the length scale \( R \). Correspondingly, the Damköhler number quantifies to which extent a sizable inhomogeneity can be induced by the activity near the particle against the opposing effect of diffusion. A small value implies that the chemicals which are produced by the activity do not “crowd” near the particle (i.e., the kinetics of the activity is in the so-called “reaction-limited” regime), and the perturbations \( \delta n \) will be small. The numerical estimate of \( \text{Da} \) requires knowledge of the reaction rate \( q \); for example, the Pt catalyzed decomposition of hydrogen peroxide reported in Refs. [3, 4] leads to \( \text{Da} \approx 10^{-1} \).

Therefore, we consider the linearized chemical potential of the chemical species,

\[
\mu \approx \mu_0 + \lambda^2 \left[ \frac{\delta n}{\xi^2} - \nabla^2 \delta n \right],
\]

with the correlation length defined as

\[
\xi := \frac{\lambda}{\sqrt{h''(n_0)}},
\]

Both quantities \( \lambda \) and \( h''(n_0) \) depend on temperature. In particular, \( 1/h''(n_0) \) represents a susceptibility, and a possible critical point of the model for the chemical solute can be identified by the condition \( h''(n_0) = 0 \). The temperature dependence is model-specific; for instance, \( h(n) \) could be approximated by, e.g., the ideal gas model (as done in the main text for the purpose of quantitative estimates), the van der Waals model, or a mean-field Ginzburg–Landau model. However, the formalism presented in the manuscript does not require the explicit form of \( h(n) \) to be specified, as only its general properties are used. It is actually more advantageous to parametrize the state by \( \xi \) explicitly instead of by the temperature. Consequently, in our formulation the two relevant independent parameters of the model for the chemical are \( \lambda \) and \( \xi \), while the activity is parametrized by the Damköhler number \( \text{Da} \).

In this linear approximation, Eq. (1) reduces to

\[
\nabla^2 \mu(r) = 0.
\]

By choosing the origin of the coordinates to coincide with the center of the spherical particle, the boundary conditions can be written as (see Eqs. (6) and (8) in the main text)

\[
\frac{\partial \mu}{\partial r}(r = R_{\text{er}}) = - \frac{q}{n_0 \Gamma} \delta \mu_{\text{er}}, \quad \mu(r \to \infty) \to \mu_0
\]

\( \text{(II.6)} \)
for the chemical potential and (see Eqs. (6) and (9) in the main text)
\[ \lambda^2 \frac{\partial \delta n}{\partial r} (r = Re_r) = 0, \quad \delta n(r \to \infty) \to 0 \]  
(II.7)

for the density of the chemical.

In order to proceed, one first solves the Laplace equation (II.5), with the activity field in Eq. (11) entering through the boundary condition in Eq. (II.6). This gives
\[ \mu(r) = \mu_0 + \frac{D_a}{\beta} \sum_{\ell m} a_{\ell m} \left( \frac{R}{r} \right)^{\ell+1} Y_{\ell m}(\theta, \varphi), \]  
(II.8)
in terms of the spherical harmonics \( Y_{\ell m}(\theta, \varphi) \) (see Eq. (IV.1)). Next one solves the inhomogeneous Helmholtz equation (II.3) for \( \delta n(r) \) with the chemical potential \( \mu(r) \) found above (Eq. (II.8)). In view of Eq. (II.5), one can introduce the auxiliary dimensionless field \( \Psi(r) \) via
\[ \delta n(r) = \left( \frac{\xi}{\lambda} \right)^2 [\mu(r) - \mu_0] + n_0 \Psi(r), \]  
(II.9)
so as to obtain a homogeneous Helmholtz equation,
\[ \nabla^2 \Psi - \frac{\Psi}{\xi^2} = 0, \]  
(II.10)
together with the boundary conditions following from Eq. (II.7):
\[ \Psi(r) \to 0, \quad \text{for } |r| \to \infty, \]  
(II.11)
\[ \frac{\partial \Psi}{\partial r}(r) = -\frac{1}{n_0} \left( \frac{\xi}{\lambda} \right)^2 \frac{\partial \mu}{\partial r}(r) = \frac{D_a}{R\beta n_0} \left( \frac{\xi}{\lambda} \right)^2 \sum_{\ell m} a_{\ell m} Y_{\ell m}(\theta, \varphi), \quad \text{for } |r| = R. \]  
(II.12)
The solution of this boundary problem is given straightforwardly by
\[ \Psi(r) = \frac{D_a R^2}{\beta n_0 \lambda^2} \sum_{\ell m} c_\ell \left( s = \frac{r}{R}, \tilde{\xi} = \frac{\xi}{R} \right) a_{\ell m} Y_{\ell m}(\theta, \varphi), \]  
(II.13)
where we have introduced the coefficients
\[ c_\ell(s, \tilde{\xi}) := \psi_\ell(\tilde{\xi}) \frac{K_{\ell+1/2}(s\tilde{\xi})}{\sqrt{s}} \]  
(II.14)
and
\[ \psi_\ell(\tilde{\xi}) := -\frac{\tilde{\xi}^2}{(\ell + 1) K_{\ell+1/2}(\xi^{-1}) + \tilde{\xi}^{-1} K_{\ell-1/2}(\xi^{-1})} \]  
(\leq 0)  
(II.15)
in terms of the modified Bessel function of the second kind \( K_p(z) \) [5, 6]. For future reference, we also quote the explicit solution for the field \( \delta n(r) \) given by Eq. (II.9):
\[ \delta n(r) = \frac{D_a R^2}{\beta \lambda^2} \sum_{\ell m} \hat{c}_\ell \left( s = \frac{r}{R}, \tilde{\xi} = \frac{\xi}{R} \right) a_{\ell m} Y_{\ell m}(\theta, \varphi), \]  
(II.16)
with the coefficients
\[ \hat{c}_\ell(s, \tilde{\xi}) := c_\ell(s, \tilde{\xi}) + \frac{\tilde{\xi}^2}{(\ell + 1)s^{\ell+1}}. \]  
(II.17)

As anticipated, the typical magnitude of the perturbation in the chemical potential is set by the Damköhler number (see Eq. (II.8)) whereas the magnitude of the field \( \Psi \) (see Eq. (II.13)) is also fixed by \( D_a \) but it is modulated by the...
ratio of the particle size and a characteristic length $\sqrt{\beta n_0 \lambda^2}$ associated with the term $\alpha \lambda^2$ in the chemical potential (see Eq. (5)). (If the local free energy is approximated by the ideal-gas form, $\beta h(n) = n(\ln n - 1)$, this characteristic length is actually the correlation length defined by Eq. (II.4).)

As a final remark, we note that those changes in the boundary condition for $\delta n(r)$ at the surface of the particle which preserve its homogeneous character, do not alter the structure of the solution given by Eq. (II.13), but only the value of the factors $\psi_\ell(\xi)$ given by Eq. (II.15). For instance, a Dirichlet boundary condition (which is the other option ensuring a vanishing surface term in the functional derivative in Eq. (5)), i.e.,

$$\delta n(r) = 0, \quad \text{for } |r| = R,$$

would translate into the boundary condition

$$\Psi(r) = -\frac{1}{n_0} \left(\frac{\xi}{\lambda}\right)^2 [\mu(r) - \mu_0] = -\frac{D}{\beta n_0} \left(\frac{\xi}{\lambda}\right)^2 \sum_{\ell m} a_{\ell m} Y_{\ell m}(\theta, \varphi), \quad \text{for } |r| = R.$$  

This leads to a solution like in Eq. (II.13), but with the factors

$$\psi_\ell(\xi) \rightarrow \psi_\ell(D)(\xi) = -\frac{\xi^2}{(\ell + 1)K_{\ell+1/2}(\xi^{-1})}$$

in the coefficients defined by Eq. (II.14).

### III. THE HYDRODYNAMICAL PROBLEM

The hydrodynamical problem for the velocity field $u(r)$ is given by Eqs. (2, 3) with the boundary conditions given by Eqs. (6, 7) and with the fields $n(r)$ and $\mu(r)$ obtained as solutions of the diffusion problem discussed in Sec. II. The assumption of slow particle motion allows one to neglect the effects of compressibility (i.e., small Mach number), convection (i.e., small Reynolds number), and time–dependence on the flow. For small Mach and Reynolds numbers, the flow dynamics reduces to the diffusion of vorticity (time–dependent Stokes equation [1]), which is characterized by the diffusion coefficient $D_{\text{vort}} = \eta/\rho$, in terms of the viscosity $\eta$ and the mass density $\rho$ of the fluid. Thus, one only needs to repeat the reasoning presented in Sec. II for the chemical diffusion, but with the replacement $D_{\text{chem}} \rightarrow D_{\text{vort}}$. For water one has $\eta = 10^{-3}$ Pa·s and $\rho = 10^3$ kg/m³, resulting in a diffusion coefficient $D_{\text{vort}} = 10^{-6}$ m²/s, i.e., three orders of magnitude larger than the estimate of $D_{\text{chem}}$. One can therefore conclude that, concerning the hydrodynamical flow, the slow–particle approximation is even more justified.

The ultimate goal of solving the hydrodynamical problem for the field $u(r)$ is to obtain the velocities of translation, $V$, and of rotation, $\Omega$, of the particle. There is, however, an approach, based on the generalized Lorentz reciprocal theorem [7–10], which allows one to sidestep the explicit computation of the hydrodynamic flow field $u(r)$, and instead to express $V$ and $\Omega$ explicitly in terms of the body force $f$ only.

#### A. The generalized Lorentz reciprocal theorem

This theorem states the following relation between any two Newtonian fluid flows $\{\eta, u, \Pi\}$ and $\{\eta', u', \Pi'\}$ (the notation refers to (viscosity, flow field, hydrodynamic stress tensor)) which are solutions, in the same domain $\mathcal{D}$, of the incompressible Stokes equations (Eq. (2)) with (sufficiently well behaved) force densities $f$ and $f'$:

$$\eta' \left[ \int_{\partial \mathcal{D}} u' \cdot \Pi' \cdot dS - \int_{\mathcal{D}} d\mathcal{V} \cdot u' \cdot f' \right] = \eta \left[ \int_{\partial \mathcal{D}} u \cdot \Pi' \cdot dS - \int_{\mathcal{D}} d\mathcal{V} \cdot u \cdot f \right].$$  

(III.1)

Here the orientation of the surface element $dS := e_n \, dS$ is chosen to be the one given by the inner normal (i.e., pointing into the fluid domain $\mathcal{D}$), in agreement with the convention used in Sec. I. For the present system, the fluid domain $\mathcal{D}$ is the volume of the solution outside the particle; thus $\partial \mathcal{D}$ is composed of the surface $\partial \mathcal{D}_{\text{part}}$ of the particle and a distant (at infinity) surface $\partial \mathcal{D}_{\infty}$. The hydrodynamic stress tensor is related to the velocity field as follows:

$$\Pi = \eta \left[ \nabla u + (\nabla u)^\dagger \right] - Ip.$$  

(III.2)

For convenience, a brief derivation of the theorem is included here (see the textbook by Kim and Karrila [10]). By applying Gauss’ theorem [1] for the (sufficiently well behaved, but otherwise arbitrary) tensor and vector fields $\Pi$,
\( u \), and \( f \), as well as for the primed fields, respectively, one can re-write the left-hand side of Eq. (III.1) as (note the use of Einstein’s convention of summation over repeated indices and of the inner normal \( e_n \) for the oriented surface element):

\[
A := \eta' \left[ \int_{\partial D} u' \cdot d\mathcal{S} - \int_{D} dV u' \cdot f \right] = \eta' \int_{D} dV \left[ -\nabla \cdot (u' \cdot \Pi) - u' \cdot f \right]
\]

\[= \eta' \int_{D} dV \left[ -\left( \partial_j u'_j \right) \Pi_{ij} - u' \cdot (\nabla \cdot \Pi + f) \right] \quad \text{Eq. (2)} \]

\[\equiv \left\{ \begin{array}{l}
\frac{1}{2} \eta' \int_{D} dV \left[ \left( \partial_j u'_j \right) \Pi_{ij} + (\partial_i u'_i) \Pi_{ij} \right] \nabla u' = \nabla f, -1 \int_{D} dV \left[ \eta' \left( \partial_j u'_j + \partial_i u'_i \right) \right] \Pi_{ij}
\end{array} \right. \]

\[\text{Eq. (III.2)} \]

\[\equiv \frac{1}{2} \int_{D} dV \left[ \nabla \cdot \Pi \cdot \Pi - \eta \int_{D} dV p' (\nabla \cdot u') + \frac{3}{2} \int_{D} dV p' p \right] \quad \text{Eq. (2)} \]

\[\equiv \frac{1}{2} \int_{D} dV \left[ -\Pi' \cdot \Pi + 3p' p \right]. \quad \text{(III.3)}
\]

A similar sequence of transformations can be applied to the right-hand side of Eq. (III.1) (because the fields involved are defined in the same domain), rendering as a result the last line above (up to an irrelevant swapping of the primed and unprimed quantities). Accordingly, one concludes that both sides of Eq. (III.1) are equal.

**B. Translational and rotational velocities**

We employ the Lorentz theorem by choosing for the unprimed system the fluid motion driven by the body force distribution \( f(r) \) and corresponding to presence of the force- and torque-free self-propelled particle, which translates with velocity \( V \) and rotates with angular velocity \( \Omega \) (with respect to the center of mass) through an otherwise quiescent fluid. For the primed system, the choice is that of the flow of a fluid of the same viscosity (i.e., \( \eta' = \eta \)), which is induced by a no-slip particle of the same shape moving with constant velocities \( \mathbf{V}' \) and \( \mathbf{\Omega}' \) (through an otherwise quiescent fluid) due to being acted upon by an external force \( \mathbf{F}' \) and an external torque \( \mathbf{\tau}' \), in the absence of body forces acting on the fluid, i.e., \( \mathbf{f}' \equiv 0 \).

1. **Particle of arbitrary shape**

By taking the origin of the system of coordinates to coincide with the center of mass of the particle, the corresponding boundary conditions turn into

\[
\mathbf{u}(r) \in \partial D_{\text{part}} = \mathbf{V} + \mathbf{\Omega} \times r, \quad \mathbf{u}(r) \to \infty = 0, \quad \text{[see Eqs. (6, 7)]}
\]

and similarly for the primed system. The field \( u' \) decays as \( 1/r \), corresponding to the Stokeslet induced by the localized force \( \mathbf{F}' \) pulling the particle. The field \( u' \) can also be assumed to decay at least as \( 1/r \), provided that the external potential \( W(r) \), and thus the body force \( f(r) \) (see Eq. (I.9)), decays sufficiently fast (for a finite correlation length, the specific contribution to \( f(r) \) by the chemical decays exponentially, see Sec. II). Therefore, the surface integrals over \( \partial D_{\infty} \) vanish, and in Eq. (III.1) only the integrals over the surface \( \partial D_{\text{part}} \) of the particle contribute. One can insert the expressions for the flows \( u \) and \( u' \) from the boundary conditions (Eq. (III.4)) into the surface integrals over the surface of the particle. The expression can be simplified by noting the following points:

(i) \( \mathbf{V}, \mathbf{\Omega}, \mathbf{V}', \) and \( \mathbf{\Omega}' \) are constant vectors over the surface of the particle.

(ii) \( (\mathbf{\Omega}' \times r) \cdot (\mathbf{\Pi} \cdot e_n) \equiv \mathbf{\Omega}' \cdot [r \times (\mathbf{\Pi} \cdot e_n)] \) (and similarly for \( \mathbf{\Omega} \) and \( \mathbf{\Pi}' \)).

(iii) The integrals of \( \mathbf{\Pi}' \cdot e_n \) and \( r \times (\mathbf{\Pi}' \cdot e_n) \) over the surface of the particle represent the force and torque, respectively, by the fluid on the particle (thus they are equal in magnitude and opposite in sign to the external force \( \mathbf{F}' \) and torque \( \mathbf{\tau}' \), respectively).
(iv) Due to Gauss’ theorem, the integral of $\Pi \cdot e_n$ over the surface of the particle is equal to the volume integral of $f(r)$ (see Eqs. (I.3) and (I.6)), and similarly the integral of $r \times (\Pi \cdot e_n)$ over the surface of the particle can be written as a volume integral of $r \times f(r)$ (see Eqs. (I.7) and (I.8)). This is the step at which the mechanical isolation of the system is imposed upon solving the hydrodynamical problem. If the particle would experience a force or torque from an external source, there would be a nonzero contribution from the integrated stresses at infinity.

In this manner, one arrives at

$$\mathbf{V}' \cdot \int_D dV \, f + \mathbf{\Omega}' \cdot \int_D dV \, \mathbf{r} \times f - \int_D dV \, \mathbf{u}' \cdot \mathbf{f} = -\mathbf{V} \cdot \mathbf{F}' - \mathbf{\Omega} \cdot \tau'. \quad (III.5)$$

Due to the linearity of the Stokes equations, the flow $\mathbf{u}'(\mathbf{r})$ (i.e., the solution of the incompressible Stokes equations without body forces or torques) depends linearly on the boundary condition at the surface of the particle, i.e., on $\mathbf{V}'$ and $\mathbf{\Omega}'$, and thus can be written as [1]

$$\mathbf{u}'(\mathbf{r}) = \mathbf{V}' \cdot \mathbf{T}(\mathbf{r}) + \mathbf{\Omega}' \cdot \mathbf{K}(\mathbf{r}), \quad (III.6)$$

where the second–rank tensors $\mathbf{T}(\mathbf{r})$ and $\mathbf{K}(\mathbf{r})$ depend solely on the shape of the particle and on the boundary conditions, and can thus be considered to be known. Likewise, the forces and velocities are also related linearly. This can be expressed in a compact manner by introducing the supervector notation

$$\mathbf{V} := \begin{pmatrix} \mathbf{V} \\ \mathbf{\Omega} \end{pmatrix}, \quad \mathbf{F} := \begin{pmatrix} \mathbf{F} \\ \mathbf{\tau} \end{pmatrix}, \quad (III.7)$$

(and likewise for the primed system). This is a formal procedure in the sense that the components do not have the same units, and both true vectors (velocity, force) and pseudovectors (angular velocity, torque) are mixed. This allows one to write [1]

$$\mathbf{F} = \mathbf{R} \cdot \mathbf{V} \quad \leftrightarrow \quad \mathbf{V} = \mathbf{M} \cdot \mathbf{F} \quad (III.8)$$

in terms of the resistance supertensor $\mathbf{R}$ and its inverse, the mobility supertensor $\mathbf{M}$, which both also depend only on the shape of the particle and the boundary conditions.

By inserting Eq. (III.6) into Eq. (III.5) and defining the supervector

$$\mathbf{T}(\mathbf{r}) := \begin{pmatrix} \mathbf{T}(\mathbf{r}) - I \cdot f(\mathbf{r}) \\ \mathbf{K}(\mathbf{r}) \cdot f(\mathbf{r}) - \mathbf{r} \times f(\mathbf{r}) \end{pmatrix}, \quad (III.9)$$

the Lorentz theorem takes the compact form

$$\mathbf{V} \cdot \mathbf{F}' = \int_D dV \, \mathbf{T}(\mathbf{r}) \cdot \mathbf{V}' = \int_D dV \, \mathbf{T}(\mathbf{r}) \cdot \mathbf{M} \cdot \mathbf{F}', \quad (III.10)$$

where the second equality follows from Eq. (III.8). Since this relation holds for arbitrary choices of the force $\mathbf{F}'$ and of the torque $\mathbf{\tau}'$ in the primed problem, one straightforwardly obtains the translational and rotational velocities of the active particle as

$$\mathbf{V} = \int_D dV \, \mathbf{T}(\mathbf{r}) \cdot \mathbf{M}. \quad (III.11)$$

2. The case of a spherical particle

We now particularize the expression above for the case of a sphere of radius $R$, for which the rotation and the translation are decoupled. In this case one has [1]

$$\mathbf{M} = \begin{pmatrix} \frac{1}{6\pi \eta R} & 0 \\ 0 & \frac{1}{8\pi \eta R^3} \end{pmatrix}, \quad (III.12)$$
for the mobility supertensor. The tensors $T(r)$ and $K(r)$, which relate the flow at $r = re_r$ (within the domain occupied by the fluid, i.e., $r > R$) with the boundary condition on the surface of the spherical particle (see Eq. (III.6)), are given by the expressions [1]

$$T(r) := \frac{3R}{4} \left[ O(r) + \frac{R^2}{6} \nabla^2 O(r) \right] = \frac{3R}{4r} \left( 1 + \frac{R^2}{3r^2} \right) l + \frac{3R}{4r} \left( 1 - \frac{R^2}{r^2} \right) e_r e_r,$$

where

$$O(r) = \frac{1}{r} \left[ l + e_r e_r \right]$$

is the Oseen tensor (i.e., Green’s function for the unbounded Stokes equation), and

$$K(r) := -\left( \frac{R}{r} \right)^3 r e_r \cdot \epsilon,$$

in terms of the tensor $\epsilon = -I \times I = e_i e_j e_k \epsilon_{ijk}$ involving the three-dimensional permutation (Levi-Civita) tensor. (We note that the constraint of incompressible flow implies $\nabla \cdot O = 0$, $\nabla \cdot T = 0$, and $\nabla \cdot K = 0$.) Accordingly, from Eq. (III.11) it follows that

$$V = \frac{1}{6\pi \eta R} \int_{r > R} d\mathbf{V} \left[ T(r) - I \right] \cdot f(r)$$

and

$$\Omega = \frac{1}{8\pi \eta R^3} \int_{r > R} d\mathbf{V} \left[ K(r) \cdot f(r) - r \times f(r) \right] = \frac{1}{8\pi \eta R^3} \int_{r > R} d\mathbf{V} \left[ \left( \frac{R}{r} \right)^3 - 1 \right] r e_r \times f(r).$$

3. Alternative expressions for the drift and rotational velocities

There are alternative ways of expressing Eqs. (III.16) and (III.17) which provide further physical insight. As remarked above, the source of the motion is the curl of the force field $\mathbf{f}$, and indeed it is possible to express $V$ and $\Omega$ as explicit functionals of $\nabla \times \mathbf{f}$. Since the kernel in the integral in Eq. (III.17) is a potential field, one can establish the following relations:

$$\left[ \left( \frac{R}{r} \right)^3 - 1 \right] r e_r = -\nabla Q(r), \quad \text{with} \quad Q(r) = -\frac{R^2}{10} \left[ 3 - 5 \left( \frac{r}{R} \right)^2 + 2 \left( \frac{r}{R} \right)^5 \right],$$

and Eq. (III.17) can be recast as

$$\Omega = \frac{1}{8\pi \eta R^3} \int_{r > R} d\mathbf{V} Q(r) \nabla \times f(r),$$

after integration by parts for a field $\mathbf{f}$ that decays sufficiently fast at infinity. The contribution due to the boundary integral at $r = R$ vanishes, because $Q(r = R) = 0$ by construction. In a similar fashion, the incompressibility constraint $\partial_j (T_{ij} - \delta_{ij}) = 0$ implies the existence, for each value of the index $i$, of a vector potential field $\mathbf{A}_i$ in the domain $r > R$, which can be obtained by solving three boundary-value problems (one for each value of the index $i$):

$$\nabla \times \mathbf{A}_i = \mathbf{e}_i \cdot (T - I), \quad \nabla \cdot \mathbf{A}_i = 0, \quad \mathbf{e}_r \times \mathbf{A}_i = 0 \text{ at } |r| = R, \quad \mathbf{A}_i \to \frac{r \times \mathbf{e}_i}{2} \text{ as } |r| \to \infty.$$

The choice of gauge ($\nabla \cdot \mathbf{A}_i = 0$) and of the boundary condition for the tangential component at $r = R$ is arbitrary; the boundary condition at infinity is determined by the inhomogeneous term $I$. The solution of this problem$^1$ is given by

$$\mathbf{A}_i(r) = \frac{3R}{4} \left[ 1 - \frac{2r}{3R} - \frac{R^2}{3r^2} \right] e_i \times e_r.$$

$^1$ This follows from expanding the fields in terms of spherical harmonics and by noting that $\nabla \times (\mathbf{e}_i \cdot T) = (3R/4) \nabla \times (\mathbf{e}_i \cdot O) = -(3R/2) e_i \times \nabla (1/r)$ (see Eq. (III.14)). The solution is determined up to the gradient of a harmonic function.
Consequently, Eq. (III.16) can be rearranged also as
\[ {V_i} = \frac{1}{{6\pi \eta R}} \int_{r > R} {dV} [\nabla \times A_i(r)] \cdot f(r) = \frac{1}{{6\pi \eta R}} \int_{r > R} {dV} A_i \cdot (\nabla \times f), \] (III.22)
after integration by parts for a field \( f \) that decays sufficiently fast at infinity (the contribution by the boundary integral at \( r = R \) vanishes because \( A_i(r = R e_r) = 0 \) by construction).

Further simplification is possible in the particular case that the force is due solely to the chemical stress tensor (see Eq. (I.10)),
\[ f = \nabla \cdot \Pi_{\text{chem}}, \quad \Pi_{\text{chem}} = \lambda^2 \left[ \frac{1}{2} \nabla \nabla (n^2) - (\nabla n)(\nabla n) \right] - \frac{1}{2} \left[ \frac{\lambda^2}{2} |\nabla n|^2 + p_{\text{chem}}(n) \right]. \] (III.23)
In this manner, Eq. (III.16) turns into
\[ \mathbf{V} = -\frac{1}{{6\pi \eta R}} \int_{r > R} {dV} \Pi_{\text{chem}} : \nabla \mathbf{U} = \frac{\lambda^2}{6\pi \eta R} \int_{r > R} {dV} (\nabla n)(\nabla n) : \nabla \mathbf{U}, \] (III.24)
after integration by parts and use of the incompressibility constraint. (We note that \( \nabla \mathbf{U}(r = R e_r) = (3/2R)e_r(e_r, e_r, -1) \) (see Eq. (V.2)), so that the contribution by the term \( \propto \nabla \nabla (n^2) \) to the boundary integral at \( r = R \) does vanish.) Likewise, Eq. (III.17) can be written as (summation over repeated indices is assumed)
\[ \mathbf{\Omega} = \frac{1}{{8\pi \eta R^3}} \int_{r > R} {dV} e_i \varepsilon_{ijk} (\Pi_{\text{chem}})_{jm} (e_k \cdot \nabla)(-e_m \cdot \nabla Q) = \frac{3\lambda^2}{8\pi \eta R} \int_{r > R} {dV} \frac{1}{r} e_r \cdot (\nabla n)(\nabla n) \times e_r. \] (III.25)
Here the term \( \propto \nabla \nabla (n^2) \) does not contribute because the operator \( e_r \times \nabla \) is essentially the derivative tangential to spheres \( r = \text{constant} \), so that the angular integral over the whole spherical surface renders zero. Equations (III.24) and (III.25) show explicitly the symmetric manner in which the inhomogeneity induced by the activity enters the expressions for \( \mathbf{V} \) and \( \mathbf{\Omega} \) (compare also with Eq. (10)).

4. Faxén’s law

Finally, one can derive a suggestive interpretation of the velocities of translation and rotation. To this end, we introduce the velocity field
\[ \mathbf{u}^\infty(r) = \frac{1}{{8\pi \eta}} \int_{r' > R} {dV} \mathcal{O}(r' - r) \cdot f(r'), \] (III.26)
in terms of the Oseen tensor (Eq. (III.14)). Physically, \( \mathbf{u}^\infty(r) \) is the velocity field that would be induced by the chemical bulk force \( f \) alone, i.e., without regard to the boundary conditions imposed by the particle. (In other words, \( \mathbf{u}^\infty(r) \) is the flow in an unbounded fluid, as if the particle would not be present; correspondingly, it is also defined in the region \( |r| < R \).) In this sense, \( \mathbf{u}^\infty(r) \) plays the role of an ambient flow for the particle. If Eq. (III.16) is evaluated by using the definition in Eq. (III.13) and the balance of chemical forces (Eq. (I.5)), one arrives at
\[ \mathbf{V} = \left[ \mathbf{u}^\infty(r) + \frac{R^2}{6} \nabla^2 \mathbf{u}^\infty(r) \right]_{r=0} + \frac{1}{{6\pi \eta R}} \mathbf{F}_{\text{chem}}^{(\text{part})}, \] (III.27)
i.e., the velocity of the particle is the sum of a drag by the ambient flow and of a direct pull on the particle by the interaction with the molecules of the chemical. This expression can be recast in the form
\[ 6\pi \eta R \mathbf{V} - 6\pi \eta R \left[ \mathbf{u}^\infty(r) + \frac{R^2}{6} \nabla^2 \mathbf{u}^\infty(r) \right]_{r=0} = \mathbf{F}_{\text{chem}}^{(\text{part})}. \] (III.28)
This is Faxén’s law [10], which simply expresses the force balance for the particle: the hydrodynamical force exerted by the fluid on the particle (left hand side of the equation) is equal to the resultant of any other forces (right hand side). From this perspective, Eq. (III.28) is just a disguised form of Eqs. (I.1) and (I.2), but with an explicit expression for the integral of the hydrodynamic stress \( \Pi \) over the surface of the particle. Likewise, there is also Faxén’s law for the torque:
\[ 8\pi \eta R^3 \left[ \mathbf{\Omega} - \frac{1}{2} \nabla \times \mathbf{u}^\infty(r = 0) \right] = \mathbf{\tau}_{\text{chem}}^{(\text{part})}. \] (III.29)
C. Velocity of a spherical particle as a function of activity

By inserting the definition of \( f \) (Eq. (3) in the main text) into Eq. (III.16) one arrives at

\[
\mathbf{V} = \frac{1}{6\pi\eta R} \int_{r>R} \, dV \left[ \mathbf{T}(\mathbf{r}) - \mathbf{l} \right] \cdot \left[ -\delta n(\mathbf{r}) \nabla \mu(\mathbf{r}) \right],
\]

(III.30)

after disregarding the gradient term \( n_0 \nabla \mu \) which does not contribute because it can be absorbed in the dynamic pressure. (Alternatively, integration by parts in Eq. (III.30) gives zero due to the incompressibility constraint \( \nabla \cdot \mathbf{T} = 0 \).) The expression in Eq. (III.30) actually does not rely on any specific mechanism of phoresis. (It follows solely from the Stokes equation and the local equilibrium hypothesis embodied in Eq. (3).) It shows explicitly that a nonvanishing velocity \( \mathbf{V} \) requires both a deviation from equilibrium, so that \( \nabla \mu \neq 0 \), and a deviation from a homogeneous state, so that \( \delta n \neq 0 \) (compare with Eq. (10)). In the model of correlation–induced transport we are considering here, both requirements are enforced by the activity. This explains why the final result (given by Eq. (12) in the main text) is bilinear in the activity. In the scenario of standard phoresis, the activity is only needed to impose a deviation from equilibrium, because the extended interaction \( W \) is usually already sufficient to bring about the necessary inhomogeneity (see Eq. (VII.8), linear in the activity, as well as the comparative analysis in Sec. VII).

For the particular model of correlation–induced transport, one can insert the relationship given by Eq. (II.9) into Eq. (III.30) and neglect a perfect gradient term \( \propto \nabla \cdot \delta n \) after disregarding the gradient term \( n_0 \nabla \mu \), so that

\[
\mathbf{V} = \frac{1}{6\pi\eta R} \int_{r<R} \, dV \left[ \mathbf{T}(\mathbf{r}) - \mathbf{l} \right] \cdot \left[ -n_0 \Psi(\mathbf{r}) \nabla \mu(\mathbf{r}) \right].
\]

(III.31)

One can split the integrand into tangential and radial components as follows. The driving force is written as

\[
-n_0 \Psi(\mathbf{r}) \nabla \mu(\mathbf{r}) = -n_0 \Psi \left[ \frac{1}{r} \mathbf{e}_\parallel + \mathbf{e}_r \frac{\partial \mu}{\partial r} \right],
\]

(III.32)
in terms of the gradient operator defined on the unit sphere:

\[
\mathbf{\nabla}_\parallel := \mathbf{e}_\theta \frac{\partial}{\partial \theta} + \mathbf{e}_\varphi \frac{\sin \theta}{\sin \theta} \frac{\partial}{\partial \varphi}.
\]

(III.33)

The hydrodynamic tensor multiplying the driving force is given by (see Eq. (III.13))

\[
\mathbf{T}(\mathbf{r}) - \mathbf{l} = \left( 3R \frac{4r}{4r^3} + R \frac{3r^3}{4r^3} - 1 \right) \left( \mathbf{l} - \mathbf{e}_r \mathbf{e}_r \right) + \left( \frac{3R^3}{2r^3} - R \frac{3r^3}{2r^3} - 1 \right) \mathbf{e}_r \mathbf{e}_r,
\]

(III.34)

where in the first summand one recognizes the tangential projector \( \mathbf{l} - \mathbf{e}_r \mathbf{e}_r \) onto the spherical surfaces centered at the origin, while the second summand involves the radial projector \( \mathbf{e}_r \mathbf{e}_r \) along the normal to these surfaces. Therefore, one can write Eq. (III.31) as

\[
\mathbf{V} = \frac{-n_0}{6\pi\eta R} \int_{r<R} \, dV \Psi(\mathbf{r}) \left[ \frac{1}{r} \left( 3R \frac{4r}{4r^3} + R \frac{3r^3}{4r^3} - 1 \right) \mathbf{\nabla}_\parallel \mu(\mathbf{r}) + \left( \frac{3R^3}{2r^3} - R \frac{3r^3}{2r^3} - 1 \right) \mathbf{e}_r \frac{\partial \mu}{\partial r}(\mathbf{r}) \right].
\]

(III.35)

Introducing the dimensionless position vector \( \mathbf{s} := \mathbf{r}/R = (r/R)\mathbf{e}_r(\theta, \varphi) \) and the element of solid angle \( d\Omega := \sin \theta \, d\theta \, d\varphi \), we find

\[
\mathbf{V} = \frac{-n_0 R}{6\pi\eta} \int_{s<1} \, ds \, s^2 \int_{d\Omega} \, d\Omega \Psi(Rs_\mathbf{e}_r) \left[ \frac{1}{s} \left( \frac{3}{4s} + \frac{1}{4s^3} - 1 \right) \mathbf{\nabla}_\parallel \mu(Rs_\mathbf{e}_r) \right.
\]

\[
\left. + \left( \frac{3}{2s} - \frac{1}{2s^3} - 1 \right) \mathbf{e}_r \frac{\partial \mu}{\partial s}(Rs_\mathbf{e}_r) \right].
\]

(III.36)

The integral over the solid angle represents the scalar product in the Hilbert space of the functions defined on the unit sphere, with respect to which the spherical harmonics form an orthonormal basis. Therefore, if the expansions in Eqs. (II.8) and (II.13) in terms of spherical harmonics for \( \mu \) and \( \Psi \) are inserted, one can exchange the angular integral with the sums over the indices \( l \) and \( m \). Rearranging terms in this manner, one arrives at the expression (see Eq. (12) in the main text)

\[
\mathbf{V} = V_0 \sum_{\ell m} \sum_{\ell' m'} a_{\ell m} a_{\ell' m'} \left[ g_{\ell\ell'}^\perp \left( \frac{\xi}{R} \right) \mathbf{c}_{\ell m;\ell' m'}^\perp + g_{\ell\ell'}^\parallel \left( \frac{\xi}{R} \right) \mathbf{c}_{\ell m;\ell' m'}^\parallel \right],
\]

(III.37)
with the following definitions:

\[ V_0 := \frac{Da^2 R^3}{6\pi\eta\beta^2\lambda^2} \]  

(III.38)

for the velocity scale,

\[ G^\perp_{\ell m,\ell' m'} := \int d\Omega \ e_r(\theta, \varphi) Y_{\ell m}(\theta, \varphi) Y_{\ell' m'}(\theta, \varphi) \]  

(III.39)

and

\[ G^\parallel_{\ell m,\ell' m'} := -\frac{1}{\ell' + 1} \int d\Omega \ Y_{\ell m}(\theta, \varphi) \tilde{\nabla}_r Y_{\ell' m'}(\theta, \varphi) \]  

(III.40)

for the geometrical factors (with \( \tilde{\nabla}_r \) defined by Eq. (III.33)), and

\[ g^\perp_{\ell r}(\xi) := \int_1^\infty ds \left( -\frac{1}{2s^3} + \frac{3}{2s} - 1 \right) s^{-\ell} c_\ell(s, \xi) \]  

(III.41)

and

\[ g^\parallel_{\ell r}(\xi) := \int_1^\infty ds \left( \frac{1}{4s^3} + \frac{3}{4s} - 1 \right) s^{-\ell} c_\ell(s, \xi) \]  

(III.42)

for the dimensionless functions \( g \) that encode the dependence on the correlation length brought about by the solutions of the diffusion and the hydrodynamical problems (see Eq. (II.14) for the definition of the coefficients \( c_\ell \)). These integrals can be expressed in terms of the exponential integral function (see Sec. VIII for an example). The superscripts \( \parallel \) and \( \perp \) pertain to the contribution by the tangential and the radial component of the driving force, respectively. A glimpse at Eq. (III.31) allows one to rationalize the velocity scale \( V_0 \), defined by Eq. (III.38), in terms of the characteristic scale \( Da^2 R/(\beta \lambda)^2 \) of the source term \( n_0 \Psi \nabla \mu \) (already discussed in connection with Eqs. (II.8) and (II.13)) and in terms of the scale \( R^2/(6\pi\eta) \) associated with the hydrodynamic kernel.

One can employ the same procedure in order to compute the angular velocity: if Eq. (II.9) is inserted into the expression for the angular velocity given by Eq. (III.17), after dropping the perfect gradient terms one obtains

\[ \Omega = \frac{1}{8\pi\eta R^3} \int_{r > R} dV \left[ \left( \frac{R}{r} \right)^3 - 1 \right] r \times [-\delta n(r) \nabla \mu(r)] \]

\[ = \frac{1}{8\pi\eta R^3} \int_{r > R} dV \left[ \left( \frac{R}{r} \right)^3 - 1 \right] r \times [-n_0 \Psi(r) \nabla \mu(r)]. \]  

(III.43)

Inserting the decomposition (III.32) into this expression, one likewise arrives at

\[ \Omega = \Omega_0 \sum_{\ell m} \sum_{\ell' m'} a_{\ell m} a_{\ell' m'} g^\perp_{\ell r} \left( \frac{\xi}{R} \right) G^\perp_{\ell m,\ell' m'} \quad \Omega_0 := \frac{3V_0}{4R}, \]  

(III.44)

with the geometrical factor

\[ G^\perp_{\ell m,\ell' m'} := -\frac{1}{\ell' + 1} \int d\Omega \ Y_{\ell m}(\theta, \varphi) e_r(\theta, \varphi) \times \tilde{\nabla}_r Y_{\ell' m'}(\theta, \varphi) \]  

(III.45)

and the dimensionless function

\[ g^\perp_{\ell r}(\xi) := \int_1^\infty ds \left( \frac{1}{s^3} - 1 \right) s^{-\ell} c_\ell(s, \xi) \]  

(III.46)

where the superscript \( \tau \) reminds that these factors arise from the driving torque \( \mathbf{r} \times \mathbf{f}(r) \). As will be shown in Sec. IV below, it turns out that \( \Omega \equiv 0 \), regardless of the form of the activity pattern described by the amplitudes \( a_{\ell m} \).
IV. THE GEOMETRICAL CONSTRAINTS

We define the spherical harmonics following the standard convention employed in quantum mechanics [11]:

\[
Y_{\ell m}(\theta, \varphi) = (-1)^m \sqrt{\frac{2\ell + 1}{4\pi} \frac{(\ell - m)!}{(\ell + m)!}} P^m_\ell (\cos \theta) e^{im\varphi}, \quad m \geq 0,
\]

(IV.1)

and

\[
Y_{\ell,-m}(\theta, \varphi) = (-1)^m Y^*_\ell m(\theta, \varphi)
\]

(IV.2)
in terms of the associated Legendre functions [11]

\[
P^m_\ell (u) = (1 - u^2)^{m/2} \frac{d^m}{du^m} P_\ell (u)
\]

and

\[
P_\ell (u) = \frac{\ell!}{2^\ell \ell!} (1 - u^2)^m \frac{d^m}{du^m} (u^2 - 1)^\ell.
\]

(IV.3)

We introduce the orbital angular momentum operator \( \mathbf{L} \), which satisfies (upon setting the Planck constant \( \hbar = 1 \))

\[
\mathbf{L} = e_z \mathbf{L}_z + e_x - i e_y \mathbf{L}_+ + e_x + i e_y \mathbf{L}_-
\]

(IV.4)

with \( \mathbf{L}_z = -i \partial_\varphi \) and \( \mathbf{L}_\pm = e^{\pm i \varphi} (\pm \partial_\theta + i \cot \theta \partial_\varphi) \), so that

\[
\mathbf{L}_z Y_{\ell m} = m Y_{\ell m}
\]

(IV.5)

and

\[
\mathbf{L}_\pm Y_{\ell m} = \sqrt{\ell (\ell + 1) - m(m \pm 1)} Y_{\ell m \pm 1}.
\]

(IV.6)

In order to evaluate the integrals in Eqs. (III.39) and (III.40), we make use of the following relations:

\[
\mathbf{L} = \mathbf{r} \times (-i \nabla) = -i \mathbf{r} \times \hat{\nabla}_\parallel
\]

(IV.7)

and

\[
\hat{\nabla}_\parallel = -i \mathbf{r} \times \mathbf{L},
\]

(IV.8)

with the operator \( \hat{\nabla}_\parallel \) given by Eq. (III.33). In order to evaluate Eq. (III.39), we employ the following representation of the unit radial vector:

\[
\mathbf{e}_r = \sqrt{\frac{4\pi}{3}} \left[ e_z Y_{1,0} - \frac{e_x - i e_y}{\sqrt{2}} Y_{1,1} + \frac{e_x + i e_y}{\sqrt{2}} Y_{1,-1} \right],
\]

(IV.9)

and we use the identity [11]

\[
\int d\Omega \ Y_{1,M} Y_{\ell m} Y_{\ell' m'} = \sqrt{\frac{3(2\ell + 1)(2\ell' + 1)}{4\pi}} \begin{pmatrix} 1 & \ell & \ell' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} 1 & \ell & \ell' \\ M & m & m' \end{pmatrix},
\]

(IV.10)
in terms of the Wigner 3j symbols. It is then straightforward to obtain

\[
G^\perp_{\ell m,\ell' m'} = \sqrt{(2\ell + 1)(2\ell' + 1)} \begin{pmatrix} 1 & \ell & \ell' \\ 0 & 0 & 0 \end{pmatrix} \times \left[ e_z \begin{pmatrix} 1 & \ell & \ell' \\ 0 & m & m' \end{pmatrix} - \frac{e_x - i e_y}{\sqrt{2}} \begin{pmatrix} 1 & \ell & \ell' \\ 1 & m & m' \end{pmatrix} + \frac{e_x + i e_y}{\sqrt{2}} \begin{pmatrix} 1 & \ell & \ell' \\ -1 & m & m' \end{pmatrix} \right].
\]

(IV.11)

The properties of the 3j symbols lead to the conclusion that \( G^\perp_{\ell m,\ell' m'} \) can be non-zero only if \( |\ell - \ell'| \leq 1 \) (excluding the case \( \ell = \ell' = 0 \)) and if \( |m + m'| \leq 1 \). Furthermore, the 3j symbol which has been factored out vanishes if \( \ell = \ell' \), and thus the case \( |\ell - \ell'| = 0 \) actually does not arise.
Combining Eqs. (IV.4, IV.8, IV.9), one finds
\[
\tilde{\nabla} \parallel = \sqrt{\frac{4\pi}{3}} \left[ \frac{e_x - ie_y}{\sqrt{2}} Y_{1,1} + \frac{e_x + ie_y}{\sqrt{2}} Y_{1,-1} \right] L_z + \frac{2\pi}{3} \left[ \frac{e_x - ie_y}{\sqrt{2}} Y_{1,0} - e_z Y_{1,-1} \right] L_+ - \frac{2\pi}{3} \left[ \frac{e_x + ie_y}{\sqrt{2}} Y_{1,0} + e_z Y_{1,1} \right] L_-.
\] (IV.12)

Inserting this expression into Eq. (III.40) and evaluating it as above, one finally arrives at
\[
G_{\ell m,\ell' m'} = -\sqrt{(2\ell + 1)(2\ell' + 1)} \frac{\ell + 1}{\ell' + 1} \begin{pmatrix} 1 & \ell & \ell' \\ 0 & 0 & 0 \end{pmatrix} \times \left\{ \begin{pmatrix} e_x + ie_y & 1 & \ell \\ \frac{1}{\sqrt{2}} & -1 & m m' \end{pmatrix} + \begin{pmatrix} e_x - ie_y & 1 & \ell \\ \frac{1}{\sqrt{2}} & 1 & m m' \end{pmatrix} \right\}
+ \frac{\ell' - \ell + m' - m}{\sqrt{2}} \left[ \begin{pmatrix} e_x - ie_y & 1 & \ell \\ \frac{1}{\sqrt{2}} & 0 & m m' + 1 \end{pmatrix} - e_z \begin{pmatrix} 1 & \ell \\ 1 & -m m' + 1 \end{pmatrix} \right]
+ \frac{\ell' - \ell + m' - m-1}{\sqrt{2}} \left[ -e_x + ie_y \begin{pmatrix} 1 & \ell \\ 0 & m m' - 1 \end{pmatrix} - e_z \begin{pmatrix} 1 & \ell \\ 1 & m m' - 1 \end{pmatrix} \right].
\] (IV.13)

The same conditions for the indices as in Eq. (IV.11) also hold for this expression.

Concerning the geometrical factors entering into the expression of the angular velocity, Eq. (III.45) can be written as
\[
G^\tau_{\ell m,\ell' m'} = -\frac{i(-1)^m}{\ell' + 1} \int d\Omega \ Y_\ell_{\ell,-m} L Y_{\ell' m'}
= \frac{i(-1)^m}{\ell' + 1} \delta_{\ell,\ell'} \left[ e_x m' \delta_{m+m',0} \right.
+ \left. \sqrt{\ell' + 1 + mm'} \left( \frac{e_x - ie_y}{2} \delta_{m+m',-1} + \frac{e_x + ie_y}{2} \delta_{m+m',1} \right) \right].
\] (IV.14)

This result implies that the angular velocity given by Eq. (III.44) vanishes identically: the activity associated with a given multipole \( \ell \) is
\[
\mathcal{A}_\ell(\theta, \varphi) := \sum_{m=-\ell}^{\ell} a_{\ell m} Y_{\ell m}(\theta, \varphi),
\] (IV.15)
while the constraint \( \delta_{\ell,\ell'} \) in Eq. (IV.14) implies that Eq. (III.44) can be written as
\[
\Omega = \Omega_0 \sum_{\ell} \frac{g_{\ell,\ell}}{\ell + 1} \omega_\ell,
\] (IV.16)
with the vector
\[
\omega_\ell := \int d\Omega \mathcal{A}_\ell^* (iL) \mathcal{A}_\ell,
\] (IV.17)

after taking into account the identity \( a_{\ell m}^* = (-1)^m a_{\ell,-m} \) for the real function \( \mathcal{A}(\theta, \varphi) \). Written in this form, it is clear that each component of the vector \( \omega_\ell \) is actually the diagonal matrix element of the corresponding component of the anti-Hermitian operator \( iL \) (i.e., the generator of rotations), and therefore it must be imaginary. But the angular velocity \( \Omega \) as given by Eq. (IV.16) must be real. Consequently, the only possibility is that it is zero. We emphasize that the symmetric dependence on the activity–induced inhomogeneity is crucial for the conclusion, as illustrated by Eqs. (10) or (III.25). This differs from the standard model of phoresis, in which the expression equivalent to Eq. (IV.17) is an off-diagonal matrix element, involving on one hand the angular dependence of the activity pattern, and on the other hand the angular dependence of the interaction potential.
V. THE LIMIT OF A SHORT CORRELATION LENGTH ($\xi \ll R$)

In order to evaluate the transport velocity in this limit, it is physically more insightful and mathematically more straightforward to start with the general expression in Eq. (III.31) rather than with the expansion in Eq. (III.37). In the limit $\xi/R \ll 1$, one infers from the explicit solution in Eq. (II.13) that the field $\Psi(r)$ is exponentially confined to the radial region of extension $\sim \xi$ around the particle (because the Bessel functions appearing in the expansion decay exponentially for large values of their argument [5, 6]), while the chemical potential in Eq. (II.8) is insensitive to this limit. Therefore, the integral in Eq. (III.31) is exponentially dominated by the behavior of the integrand near the lower limit $r = R$. One can apply Watson’s lemma [12] in order to estimate the integral by approximating the whole non-exponential dependence of the integrand by the expansion around the lower limit $r = R$, which represents the tangential gradient on the sphere of radius $R$. Therefore, the transport velocity can be approximated as

$$\mathbf{v} = -v_{0} \frac{\lambda}{6\pi\eta R} \int_{r>0} d\Omega \int_{R}^{\infty} dr R^2 \Psi(r) \frac{3}{2} \left(1 - \frac{r}{R}\right) \nabla_{\parallel} n(Re_{r}),$$

(V.1)

The hydrodynamic tensor in the integrand is approximated by a Taylor expansion as

$$\mathbf{T}(\mathbf{r}) - I \approx (r - R)\mathbf{e}_{r} \cdot \nabla \mathbf{T}(Re_{r}) = \frac{3}{2} \left(1 - \frac{r}{R}\right) \left(1 - \mathbf{e}_{r} \cdot \mathbf{e}_{r}\right),$$

(V.2)

because the difference $\mathbf{T}(\mathbf{r}) - I$ vanishes at $r = R$ due to the no-slip boundary condition (7). This result implies the approximation

$$\mathbf{T}(\mathbf{r}) - I \cdot \nabla n(\mathbf{r}) \approx \frac{3}{2} \left(1 - \frac{r}{R}\right) \nabla_{\parallel} n(Re_{r}),$$

(V.3)

where we have introduced the operator

$$\nabla_{\parallel} := (1 - \mathbf{e}_{r} \cdot \mathbf{e}_{r}) \cdot \nabla|_{r=R} = \frac{1}{R} \nabla_{\parallel}$$

(V.4)

(in terms of the operator introduced in Eq. (III.33)), which represents the tangential gradient on the sphere of radius $R$. Therefore, the transport velocity can be approximated as

$$\mathbf{v} \approx -v_{0} \frac{\lambda}{6\pi\eta R} \int d\Omega \int_{R}^{\infty} dr R^2 \Psi(r) \frac{3}{2} \left(1 - \frac{r}{R}\right) \nabla_{\parallel} n(Re_{r}) = -\frac{1}{4\pi} \int d\Omega \mathbf{v}_{\text{slip}}(\theta, \varphi)$$

(V.5)

with (see Eq. (14) in the main text)

$$\mathbf{v}_{\text{slip}}(\theta, \varphi) := \mathbf{L}(\theta, \varphi) \nabla_{\parallel} n(Re_{r})$$

(V.6)

and

$$\mathbf{L}(\theta, \varphi) := \frac{v_{0}}{\eta} \frac{\lambda}{\xi} \frac{2}{1} \int_{R}^{\infty} dr (R - r) \Psi(\theta, \varphi).$$

(V.7)

An interpretation of this result is that the hydrodynamical effect of the activity can be modeled as a local boundary condition at each point of the surface of the particle in terms of a slip velocity given by $\mathbf{v}_{\text{slip}}$ with a phoretic coefficient $\mathbf{L}(\theta, \varphi)$. One can now evaluate the expression for the phoretic coefficient by inserting the explicit solution (see Eqs. (II.13–II.15)) and taking the limit $\xi := \xi/R \to 0$:

$$\mathbf{L}(\theta, \varphi) = -Da \frac{R^4}{\eta \xi^3} \sum_{\ell m} a_{\ell m} Y_{\ell m}(\theta, \varphi) \int_{1}^{\infty} ds \frac{1}{\sqrt{s}} \frac{\xi^2 K_{\ell+1/2}(s \xi^{-1})}{(\ell + 1) K_{\ell+1/2}(\xi^{-1}) + \xi^{-1} K_{\ell-1/2}(\xi^{-1})}$$

$$= Da \frac{R^2 \xi^3}{\eta \beta} \sum_{\ell m} a_{\ell m} Y_{\ell m}(\theta, \varphi) \int_{1}^{\infty} ds \frac{1}{s} e^{(1-s)/\xi}$$

$$= Da \frac{R^2 \xi^3}{\eta \beta} \sum_{\ell m} a_{\ell m} Y_{\ell m}(\theta, \varphi) \int_{1}^{\infty} ds (1 - s) e^{(1-s)/\xi}$$

(V.8)
where for $\tilde{\xi} \to 0$ the Bessel functions have been expanded uniformly inside the integral, and Watson’s lemma [12] was used to approximate the non-exponential contributions in the integrand around $s = 1$. If the last line is evaluated by using the definition for the activity function (Eq. (11) in the main text), one arrives at the final result for the phoretic coefficient (see Eq. (15) in the main text),

$$\mathcal{L}(\theta, \varphi) := \frac{\beta n \xi^3}{\eta^2 R^3} \mathcal{A}(\theta, \varphi). \quad (V.9)$$

In order to complete the analysis, we address the limit $\xi \to 0$ of the field $\nabla || n$. It is clear from Eq. (III.31) that one could have derived a similar expression for the slip velocity given by Eq. (V.6) but with $\nabla || n$ replaced by $(\xi/\lambda)^2 \nabla || \mu$. Since the distribution of the chemical potential induced by the activity is actually independent of the correlation length (see Eq. (II.8)), the phoretic velocity will scale as $V \sim \xi^4$ in the limit $\xi/R \to 0$ (see Eq. (V.9)).

Although the angular velocity $\Omega$ has been shown to vanish identically, it is instructive and useful for future reference to compute also the approximation for $\Omega$ in this limit. The procedure is identical: we use Eq. (II.9) in Eq. (III.43) and drop an irrelevant gradient term, so as to obtain

$$\Omega = -\frac{n_0}{8\pi^2 R^2} \left(\frac{\lambda}{\xi}\right)^2 \int_{r > R} dV \Psi(r) \left[\left(\frac{R}{r}\right)^3 - 1\right] r \times \nabla n(r). \quad (V.10)$$

Approximating the integrand as before, one arrives at

$$\Omega = -\frac{1}{4\pi} \int d\Omega \omega_{\text{slip}}(\theta, \varphi), \quad (V.11)$$

with a “slip angular velocity”

$$\omega_{\text{slip}}(\theta, \varphi) = \frac{3}{2R} \mathcal{L}(\theta, \varphi) \hat{e}_r \times \nabla || n(Re_r). \quad (V.12)$$

Although $\mathcal{L} \neq 0$ in general, and the relationship between $\omega_{\text{slip}}$ and $v_{\text{slip}}$ (see Eq. (14)) agrees with the standard result for phoresis [13], the angular integral in Eq. (V.11) over $\omega_{\text{slip}}$ is identically zero because it was demonstrated in Sec. IV that $\Omega \equiv 0$.

VI. THE LIMIT OF A LARGE CORREATION LENGTH ($\xi \gg R$)

The behavior of the transport velocity in the limit $\tilde{\xi} := \xi/R \gg 1$ can be addressed by considering the behavior of the radial integrals (see Eqs. (III.41) and (III.42)). It turns out that they exhibit a finite limit as $\tilde{\xi} \to \infty$, except for the terms representing the monopole–dipole coupling in Eq. (III.37): they require a separate analysis because the diffusion problem is ill-posed for these modes in the limit $\tilde{\xi} \to \infty$.

Instead of dealing with the expressions in Eqs. (III.41) and (III.42), it is actually more useful to consider the expression analogous to Eq. (III.37), but starting from Eq. (III.30) rather than from Eq. (III.31): although both expressions agree, the alternative one is mathematically less involved upon considering the limit $\tilde{\xi} \to \infty$. If this alternative is adopted, one obtains an expansion similar to Eq. (III.37), but with new radial integrals:

$$\tilde{g}_{l\ell}^{1}(\tilde{\xi}) := \int_{1}^{\infty} ds \left( -\frac{1}{2s^3} + \frac{3}{2s} - 1 \right) s^{-\ell} \hat{e}_\ell(s, \tilde{\xi}) \quad (VI.1)$$

and

$$\tilde{g}_{l\ell}^{||}(\tilde{\xi}) := \int_{1}^{\infty} ds \left( \frac{1}{4s^3} + \frac{3}{4s} - 1 \right) s^{-\ell} \hat{e}_\ell(s, \tilde{\xi}), \quad (VI.2)$$

with the coefficients $\hat{e}_\ell$ stemming from the expansion for the field $\delta n(r)$ (see Eq. (II.17)) rather than from the field $\Psi(r)$ (see Eq. (II.14)). One has to consider only those integrals for which the pair $(\ell, \ell')$ satisfies the geometrical constraint $|\ell - \ell'| = 1$ (because these are the only terms contributing to the sum in Eq. (III.37), see Sec. IV). In this case the new integrals do converge at the upper limit of integration in spite of the algebraic decay of the new factor $\hat{e}_\ell(s, \tilde{\xi})$ as $s \to \infty$, with the worst case corresponding to the pairs $(\ell, \ell') = (0, 1)$ or $(1, 0)$, which are associated with the monopole–dipole coupling in the sum in Eq. (III.37).
It can be proven that these integrals converge \textit{uniformly} as functions of $\hat{\xi}$ (see the subsection below), so that the limit $\xi \to \infty$ can be taken inside the integrals. In this limit one has

$$\hat{c}_\ell(s, \hat{\xi}) = \hat{d}_\ell(s) + \begin{cases} 
\hat{\xi} + a\hat{\xi}^{-1}, & \ell = 0, \\
o\hat{\xi}^{-1}, & \ell = 1, \\
o\hat{\xi}^{-2}, & \ell \geq 2, 
\end{cases} \quad \hat{d}_\ell(s) := \frac{s^{1-\ell}}{2(2\ell - 1)(\ell + 1)} \left( 1 - \frac{\ell - 1}{\ell + 1} s^{-2} \right). \quad (VI.3)$$

Thus one obtains

$$\lim_{\hat{\xi} \to \infty} g^{\hat{c}_\ell}(\hat{\xi}) = \int_1^{\infty} ds \left( -\frac{1}{2s^3} + \frac{3}{2s} - 1 \right) s^{-\ell} \hat{d}_\ell(s)$$

$$= \frac{3(\ell - 1) - 3(\ell + \ell')(4\ell + \ell')}{(2\ell - 1)(\ell + 1)^2(\ell + \ell')(\ell + \ell' - 1)(\ell + \ell' + 1)(\ell + \ell' - 2)(\ell + \ell' + 3)}, \quad (VI.4)$$

and

$$\lim_{\hat{\xi} \to \infty} g^{\hat{d}_{\ell'}}(\hat{\xi}) = \int_1^{\infty} ds \left( \frac{1}{4s^5} + \frac{3}{4s} - 1 \right) s^{-\ell} \hat{d}_\ell(s)$$

$$= \frac{6(\ell + 1) + 3(3\ell + \ell')(\ell + \ell' - 1)(\ell + \ell' + 2)}{2(2\ell - 1)(\ell + 1)^2(\ell + \ell')(\ell + \ell' - 1)(\ell + \ell' + 1)(\ell + \ell' - 2)(\ell + \ell' + 3)}, \quad (VI.5)$$

for all pairs $(\ell, \ell')$ that satisfy $|\ell - \ell'| = 1$, except for the pairs $(0, 1)$ and $(1, 0)$ — the latter does occur because the asymptotic behaviors of $\hat{d}_0(s)$ and $\hat{d}_1(s)$, respectively, do not decay with increasing $s$ and the corresponding integrals diverge. The resulting expressions (VI.4) and (VI.5) decay so quickly upon increasing $\ell$, that they yield a convergent contribution to the sum in Eq. (III.37) which determines the phoretic velocity of the particle.

At this stage, one can understand the rationale behind the redefinitions in Eqs. (VI.1) and (VI.2): the term that accounts for the difference between $c_\ell$ and $\hat{c}_\ell$ (see Eq. (II.17)), and which is related to the chemical potential, eliminates the dominant divergent contributions (of the order of $\hat{\xi}$) to the radial integrals. Those terms, however, do not play any role because they correspond to a purely potential force in the Stokes equation and cancel each other if the sum in Eq. (III.37) is carried out.

This kind of global cancellation is actually what occurs for the monopole–dipole coupling, the analysis of which has been pending: one can explicitly carry out the integrals in Eqs. (VI.1) and (VI.2) for the specific choices $(\ell, \ell') = (0, 1)$ and $(1, 0)$ and one finds that, in spite of the redefinition, they still exhibit diverging terms proportional to $\xi$ or $\ln \xi$. These divergences, however, drop out in the combination

$$\sum_m a_{00}a_{1m} \left[ g^{\hat{c}_\ell}(\hat{\xi}) \mathbf{G}_{1m,00} + g^{\hat{d}_{\ell'}}(\hat{\xi}) \mathbf{G}_{1m,00}^\dagger + g^{\hat{c}_\ell}(\hat{\xi}) \mathbf{G}_{00,1m}^\dagger + g^{\hat{d}_{\ell'}}(\hat{\xi}) \mathbf{G}_{00,1m}^\dagger \right],$$

which contributes to the sum in Eq. (III.37) and is finite and nonzero in the limit $\hat{\xi} \to \infty$, as illustrated explicitly by the example worked out in Sec. VIII. In order to understand this particularity of the monopolar and the dipolar terms, one can consider the diffusion problem posed by Eqs. (II.3), (II.7), and (II.8) for the field $\delta n(r)$: the coefficients $\hat{c}_\ell(r/R, \xi/R)$ given by Eq. (II.17) satisfy the boundary value problem

$$\frac{d^2 \hat{c}_\ell}{dr^2} + \frac{2}{r} \frac{d \hat{c}_\ell}{dr} - \frac{\ell(\ell + 1)}{r^2} \hat{c}_\ell - \frac{\hat{\xi} \hat{c}_\ell}{\xi^2} = -\frac{1}{\ell + 1} \left( \frac{R}{r} \right)^{\ell + 1}, \quad \frac{d \hat{c}_\ell}{dr} (r = R) = 0, \quad \lim_{r \to \infty} \hat{c}_\ell = 0. \quad (VI.6)$$

For a coefficient with $\ell \geq 2$, in the limit $\xi \to \infty$ one can drop the term $\hat{c}_\ell/\xi^2$ from the equation, and still obtain a well–posed problem, the solution of which is precisely the function $\hat{d}_\ell(r/R)$ (Eq. (VI.3)). This is not the case for the monopole $(\ell = 0)$ and the dipole $(\ell = 1)$, because the inhomogeneity of the differential equation, stemming from the spatial distribution (Eq. (II.8)) of the chemical potential induced by the activity, decays too slowly with separation, so that the boundary condition at infinity cannot be fulfilled. Therefore, the monopolar and dipolar modes of the density field require a large–distance cutoff $\xi$ in order to be well defined. Ultimately, however, the nontrivial conclusion is that this cutoff is nevertheless irrelevant for the phoretic velocity of the particle, which attains a finite and nonzero value in the limit $\xi \to \infty$, as has been shown above.
Uniform convergence of the integrals in Eqs. (VI.1) and (VI.2)

In order to study the limit $\tilde{\xi} \to \infty$ of the integrals, we prove uniform convergence. To this end, we derive bounds on the coefficients $\hat{c}_\ell$: one can rearrange the coefficients according to (see Eqs. (II.14) and (II.17))

$$\hat{c}_\ell(s, \tilde{\xi}) = \frac{\tilde{\xi}^2}{(\ell + 1)s^{\ell + 1}} \left[ 1 - \chi_\ell(\tilde{\xi}) \kappa_\ell \left( \frac{\ell}{\tilde{\xi}} \right) \right],$$

with $\chi_\ell(\tilde{\xi}) := -(\ell + 1)\tilde{\xi}^{\ell-3/2} \psi_\ell(\tilde{\xi}) \geq 0$ (see Eq. (II.15)) and

$$\kappa_\ell(z) := z^{\ell+1/2} K_{\ell+1/2}(z), \quad \kappa_\ell'(z) = -z^{\ell+1/2} K_{\ell-1/2}(z).$$

(The last result follows from the recursion relation $[z^p K_p(z)]' = -z^p K_{p-1}(z)$ [5, 6].) Since $K_p(z)$ is a positive function that approaches zero exponentially, one concludes that $\kappa_\ell(z)$ is a positive, monotonically decreasing function that approaches zero exponentially; furthermore, we find

$$\chi_\ell(\tilde{\xi}) \kappa_\ell(\tilde{\xi}^{-1}) = \frac{(\ell + 1)K_{\ell+1/2}(\tilde{\xi}^{-1})}{(\ell + 1)K_{\ell+1/2}(\tilde{\xi}^{-1}) + \tilde{\xi}^{-1}K_{\ell-1/2}(\tilde{\xi}^{-1})} \leq 1.$$  

Consequently, one has

$$0 \leq \frac{\tilde{\xi}^2}{(\ell + 1)s^{\ell + 1}} \left[ 1 - \chi_\ell(\tilde{\xi}) \kappa_\ell(\tilde{\xi}^{-1}) \right] \leq \hat{c}_\ell(s, \tilde{\xi}) \leq \frac{\tilde{\xi}^2}{(\ell + 1)s^{\ell + 1}}$$

within the relevant integration range $1 \leq s < \infty$; accordingly, the radial integrals in Eqs. (VI.1) and (VI.2) are bounded as

$$\tilde{\xi}^{-2} \left| g_{\ell r}(\tilde{\xi}) \right| \leq \frac{1}{(\ell + 1)} \int_1^{\infty} ds \left| -\frac{1}{2s^3} + \frac{3}{2s} - 1 \right| s^{-\ell - \ell - 1}$$

and

$$\tilde{\xi}^{-2} \left| g_{\ell \ell}^{\parallel}(\tilde{\xi}) \right| \leq \frac{1}{(\ell + 1)} \int_1^{\infty} ds \left| \frac{1}{4s^3} + \frac{3}{4s} - 1 \right| s^{-\ell - \ell - 1}.$$  

Both bounding integrals converge for any pair $(\ell, \ell')$ which satisfies $|\ell - \ell'| = 1$. Therefore, the integrals defining the functions $\tilde{\xi}^{-2} g_{\ell r}(\tilde{\xi})$ and $\tilde{\xi}^{-2} g_{\ell \ell}^{\parallel}(\tilde{\xi})$ converge uniformly as functions of $\tilde{\xi}$. This allows one to take the limit $\tilde{\xi} \to \infty$ inside the integrals for the functions $g_{\ell r}(\tilde{\xi})$ and $g_{\ell \ell}^{\parallel}(\tilde{\xi})$. The limit of these integrals either exists or diverges not faster than $\tilde{\xi}^2$.

VII. PHORESIS WITH $\lambda = 0$ AND $W \neq 0$

In the absence of correlations (i.e., $\lambda = 0$) but with a spatially extended interaction between the particle and the molecules of the chemical (i.e., $W \neq 0$), the chemical potential is given by Eq. (5) as

$$\mu(r) = h'(n(r)) + W(r).$$

The phoretic velocity is still given by Eq. (III.30), but now the deviation $\delta n = n - n_0$ from homogeneity is determined both by the activity $A$ and the spatially extended interaction $W$. (We note that a non-vanishing drift still requires a non-zero activity, so that $\nabla \mu \neq 0$ in Eq. (III.30)).

We follow the same strategy as in the case $\lambda \neq 0$ and $W = 0$: first the diffusion problem is solved by linearization in terms of the deviations $\delta n$, and then one evaluates Eq. (III.30) with the solutions which have been found. In this case, Eq. (II.3) is replaced by the linearized form of Eq. (VII.1), i.e.,

$$\mu \approx \mu_0 + h''(n_0) \delta n + W,$$

assuming that $W$ vanishes at infinity. The chemical potential is obtained as the solution for the same boundary-value problem (Eqs. (II.5) and (II.6)) as before, i.e., Eq. (II.8) holds, and the density field follows from Eq. (VII.2):

$$\delta n \approx \frac{1}{h''(n_0)} (\mu - \mu_0 - W).$$
(We note that the boundary conditions in Eq. (II.7) are irrelevant if \( \lambda = 0 \).) Inserting this expression into Eq. (III.30), and noting that the term involving \( \mu - \mu_0 \) becomes a perfect gradient that does not contribute, one finally arrives at

\[
\mathbf{V}^{(W)} = \frac{1}{6\pi \eta R} \int_{r > R} dV \left[ T(r) - \mathbf{I} \right] \cdot \left[ \frac{1}{h''(n_0)} W(r) \nabla \mu(r) \right].
\]  

(VII.4)

(The superscript \((W)\) is a reminder that the expression corresponds to the case \( W \neq 0 \) and \( \lambda = 0 \).) This expression has precisely the same form as the one in Eq. (III.31) with the identification

\[
n_0 \Psi(r) \leftrightarrow -\frac{1}{h''(n_0)} W(r).
\]  

(VII.5)

Therefore, by expanding the interaction field \( W(r) \) in terms of the spherical harmonics,

\[
W(r) = \sum_{\ell=0}^{\infty} \sum_{m=\ell} \omega_{\ell m}(r) Y_{\ell m}(\theta, \varphi),
\]  

(VII.6)

and by comparison with the expansion in Eq. (II.13), one can infer the mapping

\[
\frac{Da R^2}{\beta \lambda^2} (\frac{r}{R}) \ell \ell \leftrightarrow \frac{1}{h''(n_0)} \omega_{\ell m}(r)
\]  

(VII.7)

and borrow the final expression in Eqs. (III.37–III.42). In this manner, one arrives at the expression

\[
\mathbf{V}^{(W)} = V_0^{(W)} \sum_{\ell m \ell' m'} a_{\ell' m'} \left[ \gamma_{\ell m, \ell'} G_{\ell m, \ell' m'}^{\perp} + \gamma_{\ell m, \ell'} G_{\ell m, \ell' m'}^{\parallel} \right]
\]  

(VII.8)

with the velocity scale

\[
V_0^{(W)} = \frac{Da R}{6\pi \eta \beta^2 h''(n_0)}
\]  

(VII.9)

and the dimensionless radial integrals

\[
\gamma_{\ell m, \ell'}^{\perp} := - \int_1^{\infty} ds \left( -\frac{1}{2s^3} + \frac{3}{2s} - 1 \right) s^{-\ell'} \beta \omega_{\ell m} (Rs).
\]  

(VII.10)

and

\[
\gamma_{\ell m, \ell'}^{\parallel} := - \int_1^{\infty} ds \left( \frac{1}{4s^3} + \frac{3}{4s} - 1 \right) s^{-\ell'} \beta \omega_{\ell m} (Rs).
\]  

(VII.11)

The velocity scale (Eq. (VII.9)) is related to the corresponding one in the correlation–induced model as \( V_0 = V_0^{(W)} (R/\xi)^2 Da \) (compare with Eqs. (III.38) and (II.4)). The dimensionless functions \( \gamma_{\ell m, \ell'} \) encode the “shape” of the interaction \( W(r) \), whereby any length scale that characterizes the spatial extent of \( W \) would formally play the role of a cutoff and replace \( \xi \) in the mathematical expressions. The geometrical constraints derived from the functions \( G_{\ell m, \ell' m'}^{\perp}, G_{\ell m, \ell' m'}^{\parallel} \) also affect the conclusions following from Eq. (VII.8). In particular, a spherically symmetric external potential \( W(r) \) gives rise only to a monopolar contribution, i.e., \( \omega_{\ell m} = 0 \) if \( \ell \neq 0 \) in Eq. (VII.6). Therefore any activity pattern lacking a dipole, i.e., \( a_{\ell m} = 0 \) if \( \ell = 1 \) in the expansion in Eq. (11), cannot lead to phoresis: \( V^{(W)} = 0 \) according to Eq. (VII.8). Likewise, in the complementary case of a purely dipolar activity pattern (\( a_{\ell m} = 0 \) if \( \ell \neq 1 \)), the correlation–induced velocity vanishes, but \( V^{(W)} \neq 0 \) generically for a spherically symmetric potential (it is \( G_{00,10} = -G_{10,0} = e_2/\sqrt{3} \), while \( \gamma_{00,10}^{\perp} - \gamma_{10,0}^{\perp} \neq 0 \) in general.)

The same approach can be applied to obtain the phoretic angular velocity: inserting Eq. (VII.3) into Eq. (III.43) allows one to make again use of the identification in Eq. (VII.7), so that starting from the solution given in Eqs. (III.44)–(III.46) one can write

\[
\Omega^{(W)} = \Omega_0^{(W)} \sum_{\ell m \ell' m'} a_{\ell' m'} \gamma_{\ell m, \ell'} G_{\ell m, \ell' m'}^{\tau}, \quad \Omega_0^{(W)} := \frac{3V_0^{(W)}}{4R},
\]  

(VII.12)
with the same geometrical factor as in Eq. (III.45) and with the dimensionless radial integral
\[
\gamma_{\ell m, \ell' m'} = -\int_1^\infty ds \left( \frac{1}{s^3} - 1 \right) s^{1 - \ell'} \beta w_{\ell m}(Rs).
\]  
(VII.13)

(As above, the superscript \( W \) is a reminder that the expression corresponds to the case \( W \neq 0 \) and \( \lambda = 0 \).) In this case, however, in general the angular velocity is non-zero. This is the case because the argument presented in Sec. IV, which relies fundamentally on the symmetric bilinear dependence on the activity (see Eq. (IV.17)), is not applicable here.

Finally, for comparison with the case of a vanishing correlation length addressed in Sec. V, we consider the limit in which the spatial extension of the potential \( W(r) \) vanishes, so that the interaction between the particle and the molecules of the chemical is localized at the surface \( r = R \). The identification in Eq. (VII.5) still holds; if \( W(r) \) becomes exponentially small for \( r \neq R \), Watson’s lemma can be applied again, so that the expressions in Eqs. (V.5) and (V.11) in terms of the slip velocities, Eq. (14) and Eq. (V.12), are valid. The phoretic coefficient is given by
\[
\mathcal{L}^{(W)}(\theta, \varphi) := \frac{1}{\eta} \int_0^\infty dR \int_0^{2\pi} \left[ 1 - e^{-\beta W(R + x, \theta, \varphi)} \right],
\]
(VII.14)
as derived from Eq. (V.7). This coefficient agrees with the limit of small \( W \) for the expression derived in the case of a very dilute chemical (so that the ideal gas approximation holds, see, e.g., Ref. [14]). Indeed, Eq. (VII.14) can be written also as
\[
\mathcal{L}^{(W)}(\theta, \varphi) := \frac{1}{\beta \eta} \int_0^\infty dx \int_0^{2\pi} \left[ 1 - e^{-\beta W(R + x, \theta, \varphi)} \right],
\]
(VII.15)
up to corrections of order \( (\beta W)^2 \), which are neglected in the approximation of small \( W \) employed in our calculations.

### VIII. DETAILS OF THE ILLUSTRATIVE EXAMPLE IN FIGS. 1 AND 2

Here we collect the specific details for the example introduced in the main text consisting of a particle with an activity which comprises mono-, di-, and quadrupolar contributions:
\[
\lambda(\theta, \phi) = a_{00}Y_{00}(\theta, \phi) + a_{10}Y_{10}(\theta, \phi) + a_{20}Y_{20}(\theta, \phi).
\]  
(VIII.1)

(The values \( a_{1, \pm 1} = 0 \) can be realized via a convenient choice of the orientation of the \( z \)-axis, and thus it does not represent a loss of generality.) In this case, Eq. (III.37) renders (with \( \xi := \xi/R \))
\[
\frac{V_z}{V_0} = \frac{a_{00} a_{10}}{\sqrt{3}} \left[ g_{01}(\hat{\xi}) (G_{00,10} || \cdot \mathbf{e}_z) + g_{01}(\hat{\xi}) (G_{00,10} || \cdot \mathbf{e}_z) + g_{01}(\hat{\xi}) (G_{00,10} || \cdot \mathbf{e}_z) \right]
+ a_{10} a_{20} \left[ g_{12}(\hat{\xi}) (G_{10,20} || \cdot \mathbf{e}_z) + g_{12}(\hat{\xi}) (G_{10,20} || \cdot \mathbf{e}_z) + g_{12}(\hat{\xi}) (G_{10,20} || \cdot \mathbf{e}_z) \right]
= \frac{a_{00} a_{10}}{\sqrt{3}} \left[ g_{01}(\hat{\xi}) - g_{01}(\hat{\xi}) + g_{01}(\hat{\xi}) \right]
+ \frac{a_{10} a_{20}}{\sqrt{15}} \left[ 2g_{12}(\hat{\xi}) - 2g_{12}(\hat{\xi}) + 2g_{12}(\hat{\xi}) + g_{12}(\hat{\xi}) \right]
:= a_{10} \left[ a_{00} f_1(\hat{\xi}) + a_{20} f_2(\hat{\xi}) \right].
\]  
(VIII.2)

Using Eqs. (III.39)-(III.42) and (II.15), one arrives at
\[
f_1(\hat{\xi}) = \frac{e^{\hat{\xi} - 1} (12\hat{\xi}^2 - 1)}{32\sqrt{3} (\xi + 1) [2\xi (\xi + 1) + 1]},
\]  
(VIII.3)

where \( \text{Ei}(x) \) denotes the exponential integral function [5, 6], and
\[
f_2(\hat{\xi}) = \frac{\hat{\xi} \{ \xi \{ 4\xi (\xi (\xi (7\xi - 5) - 11) - 25) + 3 \} + 2 - e^{\hat{\xi} - 1} [ \xi (6\xi (\xi (24\xi (\xi + 1) + 7) + 4) - 5) - 2] \text{Ei}(\hat{\xi} - 1) \}}{192\sqrt{15} \xi^2 [2\xi (\xi + 1) + 1] \{ \xi (9\xi (\xi + 1) + 4) + 1 \}}.
\]  
(VIII.4)
These functions are used to obtain Figs. 1 and 2, and feature in the related discussions in the main text. These explicit results allow one to study the asymptotic limits $\xi \to 0$ and $\xi \to \infty$, respectively, and to compare with the general results obtained previously. By using the asymptotic behavior of the exponential integral function in Eqs. (VIII.3) and (VIII.4), one obtains $V \sim \xi^5$ when $\xi \ll 1$ (in agreement with the conclusions from Sec. V), and $V \sim \xi^0$ when $\xi \gg 1$ (in agreement with the conclusions from Sec. VI).

Acknowledgments

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