Tracer diffusion in active suspensions

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We study the diffusion of a Brownian probe particle of size $R$ in a dilute dispersion of active Brownian particles (ABPs), which move with a self-propulsive velocity $U_0$ and randomly reorient with a characteristic time scale $\tau_R$, of size $a$, characteristic swim speed $U_{sw}$, reorientation time $\tau_R$, and mechanical energy $k_BT = \zeta_aU_0^2\tau_R/6$, where $\zeta_a$ is the Stokes drag coefficient of a swimmer. The probe has a thermal diffusivity $D_T = k_BT/\zeta_T$, where $k_BT$ is the thermal energy of the solvent and $\zeta_T$ is the Stokes drag coefficient for the probe. When the swimmers are inactive, collisions between the probe and the swimmers sterically hinder the probe’s diffusive motion. In competition with this steric hindrance is an enhancement driven by the activity of the swimmers. The strength of swimming relative to thermal diffusion is set by $Pec = U_0a/D_T$. The active contribution to the diffusivity scales as $Pec^2$ for weak swimming and $Pec$ for strong swimming, but the transition between these two regimes is nonmonotonic. When fluctuations in the probe motion decay on the time scale $\tau_R$, the active diffusivity scales as $k_BT/\zeta_T$; the probe moves as if it were immersed in a solvent with energy $k_BT$ rather than $k_BT$. DOi: 10.1103/PhysRevE.95.052605

I. INTRODUCTION

Diffusive and rheological properties of active suspensions are important for understanding many biological systems and processes, such as transport within cells. Active Brownian particles (ABPs), which move with a self-propulsive velocity $U_0$ and randomly reorient with a characteristic time scale $\tau_R$, provide a minimal model for active suspensions; even the precise mechanism of their autonomous motion need not be specified. The motion of these active particles, or “swimmers,” affects not only material properties (e.g., viscosity) but also the motion of passive constituents, such as nutrients or signaling proteins that may be important for cell survival.

In a passive suspension where particles lack the ability to self-propel, it is well known that “collisions” between a probe and the bath particles sterically hinder the long-time diffusive motion of a probe; the effective long-time diffusivity is less than the isolated Stokes-Einstein-Sutherland (SES) value [1,2]. By contrast, experiments have confirmed that colloidal tracers (both Brownian and non-Brownian) in active bacterial suspensions undergo enhanced diffusive motion at long times due to bath activity. This is observed not only in liquid cultures, but also in porous media and on agar surfaces [3–5]. As a result, recent theoretical and experimental investigations have been motivated to understand the character of this enhanced diffusive motion and to provide models that describe this behavior [6–15]. For example, Kasyap et al. [10] developed a mean-field hydrodynamic theory to describe the effects of binary interactions between point tracers and ellipsoidal bacterial swimmers. This theory predicts a net enhancement of tracer diffusivity arising from the fluid flow induced by the swimming bacteria, which was shown to be a nonmonotonic function of a Péclet number relating the strength of bacterial advection to the Brownian motion of the tracer. Experiments have also observed a nonmonotonicity in the Péclet number when varying the size of the tracer particle [16]. Other theory and experiments propose that the enhancement to the diffusivity is linear in the “active flux” due to the swimmers’ autonomous motion [6–9], and that, in close contact, entrainment of tracers in the swimmers’ flow field is primarily responsible for this enhancement [13,14].

Here we show that these same qualitative features are recovered without considering hydrodynamic interactions (HI)—the enhanced diffusivity of passive particles may be understood as a result of the activity of the bath particles and excluded volume interactions alone. This does not mean the HI are not important, only that their effect is quantitative, not qualitative. We use a Smoluchowski-level analysis to model the active suspension and compute the long-time diffusivity of a passive probe using generalized Taylor dispersion theory and expansions in orientational tensor harmonics [2,17,18]. The derivation and complete expressions for the active diffusivity of the probe are given in the Appendix; here we focus on limiting behaviors. Additionally, we show that these excluded volume interactions have important implications for experimental measurements of activity-enhanced diffusion: steric hindrance to passive diffusion is in competition with active enhancement, and both effects must be considered when designing and analyzing experiments.

II. MECHANICAL MODEL

Consider a passive Brownian particle of size $R$ moving through a bath comprised of a Newtonian solvent of viscosity $\eta$, and a dispersion of ABPs of size $a$, swim speed $U_0$, and reorientation time $\tau_R$. This reorientation time may be the Brownian reorientation time, or a characteristic tumbling time of a swimmer. The origin of fluctuations in swimmer orientation is unimportant; at long times ($t \gg \tau_R$), it may be effectively modeled by a diffusive process. In the absence of the probe, the swimmers undergo both a thermal and an active random walk, where the thermal walk is characterized by the SES diffusivity $D_a$, and the random walk due to their self-propulsion is characterized by a swim diffusivity $D_{swim} = U_0^2\tau_R/6$. We define the mechanical activity of the bath as the Stokes drag times the swim diffusivity: $k_BT = \zeta_aU_0^2\tau_R$, just as $k_BT = \zeta_aD_a$ [19,20]. The volume fraction of swimmers is $\phi = 4\pi a^3 n_\infty/3$, where $n_\infty$ is the uniform number density of swimmers far from the probe. The probe has

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a thermal diffusivity \( D_p = k_B T / \zeta_p \), and the probe-swimmer pair has a relative thermal diffusivity \( D^{rel} = D_a + D_p \). The competition between swimming and Brownian motion is governed by the swim Péclet number: \( P_{\text{sw}} = U_0 R_c / D^{rel} = U_0 R / D_a = U_0 a / D_p \), and \( R_c = R + a \) is the center-to-center separation distance of the probe and swimmer upon contact.

In the absence of activity, the (passive) bath particles hinder the probe’s motion due to steric interactions [1]. For dilute suspensions, the active contribution to the diffusivity is \( (D^{act}) \equiv (D^{eff}) - D_p I (1 - \phi_{\text{act}}) \), where \( (D^{eff}) \) is the effective diffusivity of the probe, and \( \phi_{\text{act}} \equiv \phi(R_c / a)^2 / 2 \) measures the number of swimmers colliding with the probe (which can be much larger than the actual volume fraction \( \phi \) for large probes). The diffusivity of a probe in a suspension of inactive swimmers is \( D_p I (1 - \phi_{\text{act}}) \). When the probe and ABP are the same size, \( \phi_{\text{act}} = 2 \phi \), and the steric reduction is \( 1 - 2 \phi \), a well-known result in the absence of HI. (With full HI, the reduction is \( 1 - 2 \phi \).) Both the effective and active diffusivities are isotropic.

We can predict \( D^{act} \) with simple scaling arguments. The kinematic definition of the diffusivity is \( D^{act} = N U' \tau \), where \( U' \) is the magnitude of the probe’s velocity fluctuations due to collisions with the swimmers, \( \tau \) is the time scale over which these fluctuations become decorrelated, and \( N \) is the number of swimmers colliding with the probe. Upon collision, a swimmer pushes the probe with its propulsive force, \( \zeta_p \), and the solvent resists this motion via the probe’s Stokes drag. Thus, the magnitude of velocity fluctuations is \( U' \sim c_0 U_0 / \zeta_p \). (When the probe is small compared to the swimmers, the velocity fluctuations scale with the swim speed, \( U' \sim U_0 \).) On average the probe will experience \( N \sim n R^3 \) collisions, where \( R^3 \) is the volume occupied by a swimmer-probe pair. Hence, \( D^{act} \sim n R^3 \left( \frac{c_0 U_0}{\zeta_p} \right)^2 U_0^2 \tau \), \( R \gtrsim a \), \( n \sim R^3 U_0^2 \tau \), \( R \ll a \).

\[ \text{III. ACTIVE DIFFUSIVITY} \]

(i) When the decorrelation time \( \tau = \tau_D \equiv R^2 / D^{eff} \), the probe’s fluctuations are induced by the swimming bath particles, but the fluctuations are sufficiently weak (\( P_{\text{sw}} \ll 1 \)) that they decay on the time scale of Brownian diffusion. The scaling argument predicts \( D^{act} \sim D_p R^2 \phi_{\text{act}} \), and the detailed calculations give

\[ D^{act} = \frac{29}{52} D_p R^2 \phi_{\text{act}} \]  

as one would expect for Taylor dispersion: the linear-response diffusivity scales as \( \phi_{\text{act}} \), (or \( U_0^2 \)). Kasyp et al. [10] found that the hydrodynamically driven diffusivity of a point tracer scales as \( \phi_{\text{act}} U_0 / U_0 \tau_R / \sqrt{\zeta_p} \), which is also quadratic in \( U_0 \). We predict that \( D^{act} \sim \phi_{\text{act}} U_0 \) for all \( a / R \), but curiously we find no explicit dependence on \( \tau_R \), although such a dependence is evident in Fig. 1; we address this in point (ii) below.

(ii) When swimming is strong compared to Brownian motion, the appropriate time scale is \( \tau = \tau_{\text{adv}} \sim R_c / U_0 \). The swimmers are bombarding the probe so rapidly that the resulting fluctuations become decorrelated on the time it takes for swimmers to traverse the distance \( R_c \). The scaling analysis (1) predicts \( D^{act} \sim D_p R_c \phi_{\text{act}} \sim U_0 \phi_{\text{act}} \), and the detailed Smoluchowski approach gives

\[ D^{act} = \frac{1}{3} \frac{U_0 a}{\sqrt{\tau_D / \tau_R}} \left( \frac{2 + \sqrt{2 \tau_D / \tau_R}}{1 + \sqrt{2 \tau_D / \tau_R}} \right) \phi_{\text{act}} \].

The probe’s diffusivity is now linear in the swim speed \( U_0 \) (or linear in \( P_{\text{sw}} \)), as expected from Taylor dispersion theory. Kasyp et al. [10] find that \( D^{act} \sim n R^2 U_0 / a \) (because the tracers have no size in their analysis, the only geometric length scale is the swimmer size \( a \)), but their result is independent of \( \tau_R \). The transition from diffusive to advective behavior is shown in Fig. 1.

In this limit, the run length of a swimmer, \( \ell \equiv U_0 \tau_R \), is large compared to the pair size \( R_c \), and a swimmer collides with the probe before it is able to traverse its full run length. The swimmer pushes the probe with force \( \zeta_p U_0 \), but it is only able to move it a distance of \( O(a) \) on average. One might think that the swimmer should be able to push the probe the contact length \( R_c \), but the no-flux boundary condition allows the swimmer to slide along the probe’s surface, and thus the average distance of a push is only \( O(a) \). Just as in the diffusion-controlled regime, the result is insensitive to the swimmer-probe size ratio \( a / R \).
It manifests only in $\phi_{\text{act}}$, which simply becomes $\phi$ for point tracers. Finally, we note that the ratio of the other two time scales $\tau_D/\tau_R$ has no bearing on the scaling of the diffusivity in this limit—it can only change the result by a factor of 2.

However, $\tau_D/\tau_R$ significantly affects the behavior in the diffusion-dominated regime and the location of the transition from diffusive to advective behavior. When $\tau_D/\tau_R \ll 1$, reorientations are slow and the transition occurs for $\text{Pe}_c \sim O(1)$, as one would expect. However, as reorientations become faster ($\tau_D/\tau_R$ increases), the transition occurs at much higher values of $\text{Pe}_c$ (see Fig. 1). In the athermal limit of no translational diffusion ($\tau_D \to \infty$), the transition to strong swimming is governed by the reorientation Péclet number $\text{Pe}_c \equiv \tau_{\text{act}}/\tau_R = R_c/\ell \sim O(1)$ rather than the swim Péclet number $\text{Pe}_s$.

(iii) When Brownian motion is weak compared to the swimmers’ reorientations, the decorrelation time is set by the reorientation time: $\tau = \tau_R$. The scaling arguments predict $D^{\text{act}} \sim (k_sT_s/\xi_p)\phi_{\text{act}}$, or $D^{\text{act}} \sim D^{\text{swim}}\phi$ for small probes. The result of the Smoluchowski analysis is in agreement:

$$D^{\text{act}} = \left(\frac{k_sT_s}{\xi_p}\right)\frac{R}{R_c}\phi_{\text{act}}. \quad (4)$$

Note that there is no dependence on $k_BT$.

Suppose that the swimmers and probe are large enough so that Brownian motion is not important, but the swimmers’ reorientation time is relatively fast. The probe receives many small active kicks of size $k_sT_s$ from the swimmers, which are dissipated by the Stokes drag $\xi_p$. Thus, the diffusivity looks like what one would expect from a stochastic “Brownian” process, where the energy is $k_sT_s$ rather than $k_BT$. In the limit when the probe is very small, $(k_sT_s/\xi_p)(R/R_c) \to U_0^2\tau_R/6$, $\phi_{\text{act}} \to \phi$, and the active diffusivity is simply the swim diffusivity times the volume fraction of swimmers: $D^{\text{act}} = D^{\text{swim}}\phi$. As a swimmer hops in one direction, an equal volume of solvent is displaced in the opposite direction.

Because the probe receives many small kicks from the swimmers, its motion is governed by a Langevin equation, $0 = -\xi_pU + F^{\text{swim}}$, where $U$ is the probe velocity and the swimmers exert a fluctuating force with zero mean $\langle F^{\text{swim}} \rangle = 0$ and autocorrelation $\langle F^{\text{swim}}(t)F^{\text{swim}}(t') \rangle = 2k_sT_s\xi_p I \delta(t-t')$ for times long compared to $\tau_R$. The mean-squared displacement follows as $\langle (\Delta x(t))^2 \rangle = 2(k_sT_s/\xi_p)t I$ for the diffusivity of a particle immersed in such an active medium.

In this “continuum limit,” the probe acts as a thermometer that measures the swimmers’ activity $k_sT_s$. When $\ell/R_c \to 0$, active suspensions have a well-defined “temperature” through their activity $k_sT_s$ [21] because the motion looks like a stochastic Brownian process. When $\ell/R_c \gg 1$, as is the case in the strong swimming regime, the definition of temperature breaks down because the swimmers no longer move the probe a distance $\ell$, they only push it a distance $a$ between reorientations. Thus, the swimmers do not “share” their activity fully with the probe; the appropriate shared quantity in this limit is $\text{Pe}_c$.

Figure 2 shows $D^{\text{act}}$ as a function of $\tau_D/\tau_R$ for various values of $D^{\text{swim}}/D^{\text{rel}} = (\tau_D/\tau_R)\tau_{\text{adv}}^{\text{rel}} \sim k_sT_s/k_BT$. For $\tau_D/\tau_R \to \infty$, we recover the continuum-like scaling for any value of $k_sT_s/k_BT$. Even though intuition might suggest that the diffusivity should be dominated by thermal kicks when $k_sT_s \ll k_BT$, it is important to remember that it is the solvent, not the bath particles, that gives the probe thermal kicks. The swimmers can only give kicks of size $k_sT_s$. The finite size of the swimmers replaces a volume of solvent, thus reducing the number of thermal kicks the probe receives. The $O(\phi_{\text{act}})$ change in the probe diffusivity is actually negative when $k_sT_s < k_BT$ (see the inset of Fig. 3): steric hinderance exceeds active enhancement.

![FIG. 2. Active diffusivity of the probe nondimensionalized by $(k_sT_s/\xi_p)(R/R_c)\phi_{\text{act}}$ as a function of the ratio of the diffusion time to the swimmer reorientation time $\tau_D/\tau_R$. $k_sT_s/\xi_p(R/R_c)\phi_{\text{act}}$ for various values of the mechanical to thermal energy, $k_sT_s/k_BT$, where $k_sT_s = \xi_pU_0^2\tau_R/6$.](image-url)

![FIG. 3. Active diffusivity of the probe nondimensionalized by $U_0^2\phi_{\text{act}}$ as a function of $\text{Pe}_s = \tau_D/\tau_{\text{adv}} = U_0R_c/D^{\text{rel}}$. The ratio $\tau_D/\tau_{\text{adv}} = U_0R_c/\ell$ reflects the speed of reorientation relative to advection. The inset shows the total $O(\phi_{\text{act}})$ change in the probe’s diffusivity, nondimensionalized by $D^{\text{rel}}\phi_{\text{act}}$, where $D^{\text{rel}}$ is the bare diffusivity of the probe.](image-url)
An interesting feature predicted by the detailed theory is a nonmonotonic dependence of $D^{\text{act}}$ on both $\tau_D/\tau_R$ and $Pe_s$, as seen in Figs. 2 and 3, respectively. As $Pe_s$ increases, thermal diffusion slows and swimming becomes more important, so we transition from diffusive to advective behavior. This transition does not occur monotonically with $Pe_s$ because $Pe_R = \tau_{\text{adv}}/\tau_R$ also influences the dynamics. Imagine a scenario in which $\tau_D$ and $\tau_R$ are fixed and $R \gg a$, but we adjust the swimmers’ speed (perhaps by altering the amount of available fuel). When the swimmers move slowly, Brownian motion dominates: $D^{\text{act}} \equiv D^{\text{act}}/U_0a\phi_{\text{act}} \sim Pe_s$. When the swim speed is large, advection dominates and $D^{\text{act}}$ is constant. When $\tau_D \sim \tau_{\text{adv}}$, neither wins out and the reorientations are allowed to influence the dynamics. Finite Brownian motion keeps the swimmers close to the probe after a collision, and slow reorientation allows the swimmer to collide with the probe again rather than run off, thus the diffusivity is slightly higher than the advective scaling. When reorientations are too fast, this peak disappears. This is corroborated by Fig. 2, which reveals that $D^{\text{act}}$ is only nonmonotonic when $k_1T_s < k_BT$. The nonmonotonicity still occurs when $\tau_D \sim \tau_R$, but Brownian motion is only strong enough to compete with activity if the thermal energy of the solvent exceeds the activity of the bath.

Kasyp et al. [10] find the same phenomenon in their treatment. When the diffusion is hydrodynamic in origin and advection dominates, the tracer follows a straight trajectory along fluid streamlines. Weak Brownian motion allows the tracer to sample more trajectories, and the odd symmetry of the bacterium’s dipolar flow field results in an increased correlation in probe motion. When Brownian motion is strong, the probe’s motion decorrelates and the diffusivity decreases. Thus the diffusivity decreases nonmonotonically with increasing Brownian motion (i.e., as one moves from right to left in Fig. 3). Patteson et al. [16] see something similar in experiments by varying the probe size, which is equivalent to varying $Pe_s$ when all other parameters are fixed. They scale $D^{\text{act}}$ by $n^{-2}L^3U_0L$, where $L$ is the total bacterium length. They find that this scaled diffusivity first increases with probe size as approximately $R^2$ and then decreases to a plateau. Our scaling analysis predicts that $D^{\text{act}}$ is linear in probe size when diffusion dominates, and independent of probe size when advection dominates. In between, when the appropriate time scale is $\tau_R$, $D^{\text{act}}$ scales as $1/R$, thus capturing the nonmonotonicity. The peak in $D^{\text{act}}$ is predicted around $Pe_s \sim 5$ in our study and in [10], but it is found experimentally around $Pe_s \sim O(10^5)$; the source of such a large discrepancy is not known. Lastly, we note that the inset of Fig. 3 shows that this nonmonotonicity is obscured by the steric hindrance, reinforcing the importance of considering excluded-volume interactions in active suspensions.

Another common model, used by Miño et al. [7] to describe enhanced diffusion of tracers in bacterial suspensions, says that the active enhancement is proportional to the advective flux of the active particles: $D^{\text{eff}} = D_P(1 - \phi_{\text{act}}) + \beta J_a$. The steric hinderance is especially important when swimming is weak (Fig. 3). Experimentally, one should measure the bare diffusivity of a tracer, and then the change in diffusivity among nonmotile swimmers to recover the effective particle size $R_c$ from Batchelor’s theory [1]. Knowing $R_c$, the average swim speed, the reorientation time, and the bare particle diffusivities, one can calculate the active diffusivity from our theory and then compare to experimental measurements.

We presented a micromechanical model for the effective diffusivity of a passive particle embedded in a suspension of ABPs. Using a generalized Taylor dispersion approach, and employing an expansion in orientational tensor harmonics, we found an exact analytical expression for the effective diffusivity of a Brownian probe for arbitrary particle sizes, swimmer activity, and time scales. Our theory agrees qualitatively with previous experimental and theoretical investigations of enhanced diffusion in active suspensions, and it can be used to explore regimes of parameter space not typically considered in most experiments. It highlights several key features of diffusion in active suspensions: (i) the diffusion of a tracer is nonmonotonic in a Péclet number comparing swimming to thermal diffusion, (ii) steric hindrance of tracer motion is in competition with the enhancement due to bath activity, and (iii), when fluctuations of the tracer’s motion correlate on the same time scale as the swimmers’ reorientations, the bath mimics a homogeneous solvent with energy $k_BT_c$. 

For this $\beta$ model, our Smoluchowski theory predicts $\beta = (2\pi/9\sqrt{3})\beta_{Ja}(2 + \sqrt{2}\tau_D/\tau_R)/(1 + \sqrt{2}\tau_D/\tau_R)$. The ability of the swimmer to randomly reorient is not required for this enhancement to the diffusivity, as argued in [8]. In contrast to some of these experimental studies, our result depends on the size of the tracer particle. In the system of Jepson et al. [6], the tracers are nonmotile E. Coli in a suspension of motile E. Coli with equivalent spherical dimension $a = 1.4 \mu m$. From their experimental parameters, we predict $\beta = 3.22a^{-4} - 6.45a^{-4}$. To match the experimentally found value of $\beta = 7.1 \mu m^4$, our theory predicts that the E. Coli would have an equivalent spherical dimension of $a = 1.02 - 1.22 \mu m$. 

As previously proposed, this advective flux model ignores the steric hinderance of the passive suspension, which should be accounted for by $D^{\text{eff}} = D_P(1 - \phi_{\text{act}}) + \beta J_a$.
Fig. 4: Depiction of the model system. There is a Brownian probe of size $R$ immersed in a dispersion of ABPs with size $a$ at number density $n^\infty$. The ABPs swim in a direction $q$ with speed $U_0$, and they reorient with a characteristic time $\tau_R$.

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**APPENDIX A: AVERAGE PROBE MOTION**

The dynamics of the suspension are described by a Smoluchowski equation for $P((x_j),(q_j), t)$, the conditional probability of finding particle $j$ at position $x_j$, with orientation $q_j$ at time $t$. The braces indicate a dependence on the position and orientation of each particle in the suspension. When the suspension is sufficiently dilute, only the pairwise interactions between the probe and a single swimmer matter, and one may write a Smoluchowski equation for the joint probability distribution, $P_2(z,r,q,t)$, where $z$ is the position of the probe, $r$ is the position of the swimmer relative to the probe, and $q$ is the orientation of the swimmer relative to the probe (see Fig. 4):

$$\frac{\partial P_2(z,r,q,t)}{\partial t} + \nabla_z \cdot j_1^T + \nabla_r \cdot (j_2^T - j_3^T) + \nabla_k \cdot j_2^R = 0,$$

where

$$j_1^T = -D_p \nabla_z P_2 + D_p \nabla_r P_2,$$

$$j_2^T = U_0 q P_2 - D_p \nabla_r P_2,$$

$$j_3^T = -\tau^{-1}_R \nabla_k P_2,$$

and $\nabla_k = q \times \nabla_q$ is the rotational operator. Again, we have neglected HI in this study. When computing the effective diffusivity of the probe, we must track the average single-particle motion due to collisions with bath particles. To accomplish this, one averages the pair-level equation over all possible positions and orientations of the swimmer relative to the probe. This yields an equation for $P_1(z,t)$, the probe’s single-particle probability distribution:

$$\frac{\partial P_1}{\partial t} + \nabla_z \cdot \langle j_1^T \rangle = 0,$$

where we have made use of the divergence theorem and the condition that there is no relative translational flux at contact or infinity. The angular brackets denote an ensemble average over all possible relative configurations of the swimmer relative to the probe—over $dq$. It is easier to work in Fourier space, and as such we take a Fourier transform with respect to the coordinate $z$ to give

$$\hat{\frac{\partial \hat{P}_1}{\partial t}} + i \hat{k} \cdot \langle \hat{j}_1^T \rangle = 0,$$

where the carats denote the Fourier transform of their corresponding quantities in physical space, and $k$ is the Fourier wave vector, which has the interpretation of the inverse wavelength for the probe’s fluctuations. The ensemble-averaged flux of the probe in Fourier space is

$$\langle \hat{j}_1^T \rangle = -ik D_P \hat{P}_1 - D_P \int \nabla \cdot \hat{P}_1 dq.$$

The last term in the above expression shows that the collisional effects on probe motion are governed by the average gradient of the bath particles relative to the probe, it is useful to define a structure function, $g(k,r,q;t)$, such that

$$\hat{P}_1(r, q; t) = g(k, r, q; t) \hat{P}_1(k, t).$$

To find the long-time diffusivity of the probe, we take a small-wave vector expansion of the structure function,

$$g = g_0(r,q;t) + ik \cdot d(r,q;t) + O(k^2),$$

where $d = (\nabla g)_{k=0}$ is the fluctuation field.

The average flux of the probe is

$$\langle \hat{j}_1^T \rangle = \hat{P}_1 \left[ D_P \int \nabla \cdot g_0(r,q,t) dq - D_P ik \cdot \left[ I - \int \nabla \cdot d(r,q,t) dq \right] \right].$$

The first term gives the average probe speed as it moves through the suspension:

$$\langle U_1 \rangle = D_P \int_{r=R_c} \int n g_0(r,q,t) dq dS.$$

The $O(k)$ bracketed term represents the probe dispersion, and defines the effective diffusivity:

$$\langle D_{eff} \rangle = D_P \left[ I - \int_{r=R_c} \int n d(r,q,t) dq dS \right],$$

which follows from Eq. (A10) by the divergence theorem. The vector $n$ is the outward pointing unit normal vector of the probe. Because we are considering steric effects, the surface integral is computed over the exclusion region around the probe-swimmer contact distance: $r = R_c$.

**APPENDIX B: MICROSTRUCTURE GOVERNING EQUATIONS**

To determine the structure functions, we write the pair-level Smoluchowski equation in Fourier space:

$$\frac{\partial \hat{P}_2}{\partial t} + i \hat{k} \cdot \hat{j}_2^T + \nabla_r \cdot (\hat{j}_2^T - \hat{j}_1^T) + \nabla_k \cdot \hat{j}_2^R = 0,$$

where

$$\frac{\partial \hat{P}_1}{\partial t} + i \hat{k} \cdot \hat{j}_1^T = 0.$$
which can be simplified using the definition of $g$:

\[
\frac{\partial g}{\partial t} \hat{P}_1 + g \frac{\partial \hat{P}_1}{\partial t} + ik \cdot \left[ -i k D_P + D_P \int \nabla r g dr \right] \hat{P}_1 \\
+ \nabla r \cdot [U_0 q g - D_{\text{rel}} \nabla g] \hat{P}_1 \\
+ i k \cdot \nabla r g \hat{P}_1 - \tau_R^{-1} \nabla^2 g \hat{P}_1 = 0.
\] (B2)

Making use of the governing equation for $\hat{P}_1$ and taking the system to be at steady state, the Smoluchowski equation reduces to

\[
\nabla r \cdot [U_0 q g - D_{\text{rel}} \nabla g] + 2 D_P i k \cdot \nabla r g - \tau_R^{-1} \nabla^2 g = 0,
\] (B3)

with boundary conditions of no-flux at particle contact and uniform probability distribution at infinity:

\[
n \cdot [U_0 q g - D_{\text{rel}} \nabla g + D_P i k g] = 0, \quad r = R_c, \quad (B4)
\]

\[
g \sim n^\infty, \quad r \to \infty. \quad (B5)
\]

We expand the function $g_0$ and the fluctuation field $d$ in terms of orientational tensor harmonics,

\[
g_0(r, q; t) = G^{(0)}(r; t) + q \cdot G^{(1)}(r; t) \\
+ (qq - \frac{1}{2} I) : G^{(2)}(r; t) + \cdots, \quad (B6)
\]

\[
d(r, q; t) = d^{(0)}(r; t) + q \cdot d^{(1)}(r; t) \\
+ (qq - \frac{1}{2} I) : d^{(2)}(r; t) + \cdots, \quad (B7)
\]

and make the closures $G^{(2)} = 0, d^{(2)} = 0$. We take the zeroth and first orientational moments of Eq. (B3), scale the coordinate $r$ by $R_c$, and divide through by $n^\infty R_c^2 / D_{\text{rel}}$ to obtain four coupled PDEs:

\[
\nabla \cdot \left[ Pe_s G^{(1)} - \nabla G^{(0)} \right] = 0, \quad (B8)
\]

\[
n \cdot \left[ Pe_s G^{(1)} - \nabla G^{(0)} \right] = 0, \quad r = 1, \quad (B9)
\]

\[
G^{(0)} \sim 1, \quad r \to \infty, \quad (B10)
\]

\[
\nabla \cdot \left[ \frac{Pe_s}{3} I G^{(0)} - \nabla G^{(1)} \right] + \gamma^2 G^{(1)} = 0, \quad (B11)
\]

\[
n \cdot \left[ \frac{Pe_s}{3} I G^{(0)} - \nabla G^{(1)} \right] = 0, \quad r = 1, \quad (B12)
\]

\[
G^{(1)} \sim 0, \quad r \to \infty, \quad (B13)
\]

\[
\nabla \cdot \left[ Pe_s d^{(1)} - \nabla d^{(0)} - \epsilon I G^{(0)} \right] = 0, \quad (B14)
\]

\[
n \cdot \left[ Pe_s d^{(1)} - \nabla d^{(0)} - \frac{\epsilon}{2} I G^{(0)} \right] = 0, \quad r = 1, \quad (B15)
\]

\[
d^{(0)} \sim 0, \quad r \to \infty, \quad (B16)
\]

\[
\nabla \cdot \left[ \frac{Pe_s}{3} I d^{(0)} - \nabla d^{(1)} - \epsilon I G^{(1)} \right] + \gamma^2 d^{(1)} = 0, \quad (B17)
\]

\[
n \cdot \left[ \frac{Pe_s}{3} I d^{(0)} - \nabla d^{(1)} - \frac{\epsilon}{2} I G^{(1)} \right] = 0, \quad r = 1, \quad (B18)
\]

\[
d^{(1)} \sim 0, \quad r \to \infty, \quad (B19)
\]

where $\epsilon = 2a / R_c$. The natural dimensionless groups that arise in scaling the equations are the swim Péclet number $Pe_s = U_0 R_c / D_{\text{rel}}$ and the rotational parameter $\gamma^2 = 2 \tau_D / \tau_R$. The effective diffusivity will be determined entirely by $d^{(0)}$.

**APPENDIX C: SCREENED HARMONIC SOLUTION**

Equations (B8) and (B11) may be decoupled by taking the divergence of (B11):

\[
\nabla^2 (\nabla \cdot G^{(1)}) - \lambda^2 (\nabla \cdot G^{(1)}) = 0, \quad (C1)
\]

where $\lambda^2 = Pe_s^2 / 3 + \gamma^2$. This is a Helmholtz equation for the divergence of $G^{(1)}$, so we can expand it in gradients of the fundamental solution:

\[
\nabla \cdot G^{(1)} = C_0 e^{-\lambda r} + B_0 \nabla \left( e^{-\lambda r} \right) + \cdots, \quad (C2)
\]

where the constants $A_0, B_0, C_0, \ldots$. are determined from the boundary conditions. Because there is no tensorial order at the boundaries, $\nabla \cdot G^{(1)} = C_0 e^{-\lambda r} / r$ only. We then substitute into Eq. (B8) and obtain a Poisson equation for the function $G^{(0)}$:

\[
\nabla^2 G^{(0)} = C_0 Pe_s e^{-\lambda r}, \quad (C3)
\]

which has the solution

\[
G^{(0)} = C_1 + \frac{C_2}{r} + \frac{C_0 Pe_s}{\lambda^2} e^{-\lambda r} r. \quad (C4)
\]

This can be substituted into the governing equation for $G^{(1)}$:

\[
\nabla^2 G^{(1)} - \gamma^2 G^{(1)} = \frac{Pe_s}{3} \nabla G^{(0)}, \quad (C5)
\]

which has the solution

\[
G^{(1)} = -\frac{1}{\lambda^2} \nabla \left( C_0 e^{-\lambda r} + C_2 Pe_s n / 3 \gamma^2 r^2 \right), \quad (C6)
\]

The boundary condition at infinity requires $C_1 = 1$ and the contact conditions require $C_2 = 0$. Thus,

\[
G^{(0)} = 1 + \frac{Pe_s / 3}{2(\lambda + 1) + \gamma^2} \frac{e^{-\lambda(r - 1)}}{r}, \quad (C7)
\]

which is the same as the concentration profile of ABPs outside a sphere. From here, one can then solve the equations for the fluctuation field by the same method.
The fluctuation field $d^{(0)}$ in terms of two undetermined coefficients is

$$d^{(0)} = e^n \left[ \frac{B}{r^2} - \frac{\text{Pe}_e^4/9\gamma^4}{2(\lambda + 1) + \gamma^2} e^{-\lambda (r-1)} \left( \frac{\lambda}{r} + \frac{1}{r^2} \right) - A \frac{\text{Pe}_e}{\lambda^2} e^{-\lambda (r-1)} \left( \frac{\lambda}{r} + \frac{1}{r^2} \right) \right] + \frac{\text{Pe}_e^2/3\lambda^2}{2(\lambda + 1) + \gamma^2} \left( 1 - \frac{\text{Pe}_e^4/9\gamma^4}{2(\lambda + 1) + \gamma^2} \right) \left( \frac{\lambda}{r} + \frac{1}{r^2} \right) e^{-\lambda (r-1)} \left( \frac{3}{4\lambda^2} + \frac{3}{4\lambda} + \frac{1}{2} \right) \right],$$

where the coefficients $A$ and $B$ are coupled to the field $d^{(1)}$, which can be constructed with screened multipoles and linearity arguments:

$$d^{(1)} = e \left[ C e^{-\gamma (r-1)} \left( \frac{I_1}{r} - \frac{1}{r^2} \right) \right] + \gamma \left( \frac{3nn}{r^3} - \frac{I_1}{r^3} \right) + \gamma \frac{nn}{r^3} \right] \right]$$

$$- \frac{1}{\lambda^2 - \gamma^2} \left[ \lambda^2 \right] \left( \frac{3nn}{r^3} - \frac{I_1}{r^3} \right) + \gamma \left( \frac{3nn}{r^3} - \frac{I_1}{r^3} \right) + \lambda^2 \frac{nn}{r^3} \right]$$

$$+ \frac{1}{\lambda^2} \lambda^2 \left[ \frac{3nn}{r^3} - \frac{I_1}{r^3} \right) + \lambda \left( \frac{3nn}{r^3} - \frac{I_1}{r^3} \right) + \lambda^2 \frac{nn}{r^3} \right] + B \frac{1}{3} \frac{\text{Pe}_e}{\lambda^2} \left( \frac{3nn}{r^3} - \frac{I_1}{r^3} \right)$$

$$+ \left( \frac{2\lambda^2 - \gamma^2}{\lambda^2} \right) \frac{\text{Pe}_e}{\lambda^2} \left( \frac{3nn}{r^3} - \frac{I_1}{r^3} \right) + \lambda \left( \frac{3nn}{r^3} - \frac{I_1}{r^3} \right) + \lambda^2 \frac{nn}{r^3} \right]$$

$$+ \frac{\text{Pe}_e}{2(\lambda + 1) + \gamma^2} e^{-\lambda (r-1)} \left[ \frac{3nn}{r^3} - \frac{I_1}{r^3} \right) + \lambda \left( \frac{3nn}{r^3} - \frac{I_1}{r^3} \right) + \lambda^2 \frac{nn}{r^3} \right] \right].$$

The coefficients $A$, $B$, and $C$ are determined by the no-flux boundary condition at $r = 1$. We have a linear system of three equations that may be found algebraic expressions for $d^{(0)}$:

$$2B \left( 1 + \frac{\text{Pe}_e^2}{3\gamma^2} \right) - 2C \text{Pe}_e \left( \frac{1}{\gamma} + \frac{\gamma}{\gamma^2} \right) = \frac{1}{2} \left( 1 + C_0 \frac{\text{Pe}_e}{\lambda^2} \right) - C_0 \frac{\text{Pe}_e}{\lambda^2} \left( \frac{2 + 2\lambda + \gamma}{\lambda^2} \right),$$

$$B \frac{\text{Pe}_e}{\gamma^2} - C \left( \frac{2}{\gamma^2} + \frac{3}{\gamma} + \gamma \right) + \frac{C_0}{\lambda^2} \left( \frac{2\lambda^2 - \gamma^2}{\lambda^2} \right) \left( \frac{1}{2} + \frac{9}{4\lambda^2} + \frac{3}{2\lambda} + \frac{3(1 + \lambda)}{4\lambda} \right)$$

$$= \left( 3 + 3\lambda + \lambda^2 \right) \left( \frac{C_0}{\lambda^2} - \frac{A}{\lambda^2} + \frac{C_0}{\lambda^2} \left( \frac{2\lambda^2 - \gamma^2}{\lambda^2} \right) \left( \frac{1}{2} + \frac{9}{4\lambda^2} + \frac{3}{2\lambda} + \frac{3(1 + \lambda)}{4\lambda} \right) \right),$$

$$- 3B \frac{\text{Pe}_e}{\gamma^2} + C \left( \frac{2\lambda^2 - \gamma^2}{\lambda^2} \right) \left( \frac{1}{2} + \frac{9}{4\lambda^2} + \frac{3}{2\lambda} + \frac{3(1 + \lambda)}{4\lambda} \right)$$

$$= \left( 3 + 3\lambda + \lambda^2 \right) \left( \frac{C_0}{\lambda^2} + \frac{A}{\lambda^2} - \frac{C_0}{\lambda^2} \left( \frac{2\lambda^2 - \gamma^2}{\lambda^2} \right) \left( \frac{1}{2} + \frac{9}{4\lambda^2} + \frac{3}{2\lambda} + \frac{3(1 + \lambda)}{4\lambda} \right) \right),$$

$$= \frac{\text{Pe}_e}{3} \left( \frac{C_0}{\lambda^2} \left( 1 + \lambda \right) + \frac{A}{\lambda^2} \left( 1 + \lambda \right) \right) - B + \frac{C_0}{\lambda^2} \left( \frac{1}{2} + \frac{3(1 + \lambda)}{4\lambda^2} \right).$$

We solved this system of equations in MATHEMATICA to create the figures in this article. They may be solved by hand as well, but the expressions are long and it is difficult to elucidate important physics from the full expressions.

The effective diffusivity is given by

$$\langle D^{(\text{eff})} \rangle = D_p \left[ I - n \int_{r=R_i} nd^{(0)} dS \right],$$

which, in terms of the undetermined coefficients, is

$$\langle D^{(\text{eff})} \rangle = D_p I \left[ 1 - 3 \frac{4\pi}{3} e^n R_i^3 \left( B - \frac{\text{Pe}_e^4/9\gamma^4}{2(\lambda + 1) + \gamma^2} \left( \frac{\lambda}{r} + \frac{1}{r^2} \right) - A \frac{\text{Pe}_e}{\lambda^2} e^{-\lambda (r-1)} \left( \frac{\lambda}{r} + \frac{1}{r^2} \right) \right) \right]$$

$$- \frac{\text{Pe}_e^2/3\lambda^2}{2(\lambda + 1) + \gamma^2} \left( 1 - \frac{\text{Pe}_e^4/9\gamma^4}{2(\lambda + 1) + \gamma^2} \right) \left( \frac{\lambda}{r} + \frac{1}{r^2} \right) e^{-\lambda (r-1)} \left( \frac{3}{4\lambda^2} + \frac{3}{4\lambda} + \frac{1}{2} \right) \right].$$

When the bath particles are inactive, $\text{Pe}_e = 0$, $A = 0$, $B = 1/4$, and we recover the classic result: $\langle D^{(\text{eff})} \rangle = D_p I [1 - \frac{1}{2} \left( \frac{\lambda}{r} \right)^2].$
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