Anomalous Diffusion of Symmetric and Asymmetric Active Colloids

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The stochastic dynamics of colloidal particles with surface activity—in the form of catalytic reaction or particle release—and self-phoretic effects are studied analytically. Three different time scales corresponding to inertial effects, solute redistribution, and rotational diffusion are identified and shown to lead to a plethora of different regimes involving inertial, propulsive, anomalous, and diffusive behaviors. For symmetric active colloids, a regime is found where the mean-squared displacement has a superdiffusive $t^{3/2}$ behavior. At the longest time scales, an effective diffusion coefficient is found which has a nonmonotonic dependence on the size of the colloid.

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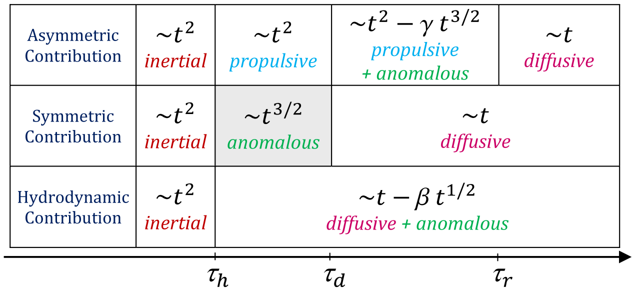
The development of biomimetic technology would significantly benefit from the ability to make synthetic components with desired motility properties. In recent years, there has been a range of developments along these lines, with functionalities that can be manipulated at length scales ranging from microns down to molecular scales [1]. These include experimental realization of actuated microswimmers [2] and theoretical proposals of simple model swimmers that can tackle the problems caused by low Reynolds number conditions [3]. Moreover, it has been recently demonstrated (both experimentally and theoretically) that interfacial phoretic effects (such as electrophoresis, electro-osmosis, and diffusiophoresis) could lead to self-propulsion of colloidal particles [4–6]. It has also been shown that phoretic effects can be used to steer both active and passive colloidal particles [7,8], which adds to the promise of these phenomena for designing functional self-motile vessels in the nanoscale.

A fundamental property of such small objects, even when equipped with a self-propulsion mechanism, is that their motion is stochastic due to the ambient fluctuations that could be of thermal origin or otherwise. This means that we cannot directly control the motion of self-motile objects, and any design characteristic needs to be incorporated into statistical average outcomes. For example, it has been shown that a self-propelled colloidal particle makes a crossover between ballistic and diffusive behaviors over a time scale that is set by the rotational diffusion of the colloid, when its orientation will be randomized [5]. A study of a two-dimensional model of self-propelled objects with fluctuations both in direction and magnitude of the velocity has shown the possibility of reentrant ballistic and diffusive behaviors [9]. Other examples include the effect of the activity of proteins on the dynamics of membranes [10], and collective behavior of mixtures of motors and filaments (or reorganizing living cells) and active particles such as swimming bacteria [11,12], where a host of qualitative and quantitative changes have been found to occur due to nonequilibrium fluctuations. In light of this inherent feature, it will be natural to ask how many distinct regimes of motion we could have for active colloidal particles, what the relevant time scales that differentiate between these regimes are, and how they can be tuned so that the desired type of motion could be achieved by choosing the right parameters.

Here, we aim to address some of these questions for a class of isolated self-motile active colloids. We study the velocity autocorrelation function and the mean-squared displacement of surface-active spherical colloidal particles that interact with their self-generated surrounding clouds of solute particles via interfacial phoretic effects. We identify the relevant time scales in the dynamics, namely, the hydrodynamic relaxation time τ_h that controls the crossover between inertial and viscous regimes, the diffusion time of the solute particles around the colloid τ_d, and the rotational diffusion time of the colloid τ_r. We calculate the contribution due to hydrodynamic fluctuations, as well as the self-phoretic contributions that depend on whether the particles are symmetric (in which case there is no net propulsion) or asymmetric (where the colloids are self-propelled). We find that these different contributions lead to a variety of different regimes, as summarized in Fig. 1.



Asymmetric Contribution	$\sim t^2$ inertial	$\sim t^2$ propulsive	$\sim t^2 - \gamma\, t^{3/2}$ propulsive + anomalous	$\sim t$ diffusive
Symmetric Contribution	$\sim t^2$ inertial	$\sim t^{3/2}$ anomalous	$\sim t$ diffusive	
Hydrodynamic Contribution	$\sim t^2$ inertial	$\sim t - \beta\, t^{1/2}$ diffusive + anomalous		

FIG. 1 (color online). Summary of results for the different contributions to the mean-squared displacement of active colloids. The total mean-squared displacement is obtained by summing all of the contributions for asymmetric colloids, and the bottom two rows (only) for symmetric colloids.

For symmetric surface activity, we find a regime corresponding to $\tau_h \ll t \ll \tau_d$, where the active colloid demonstrates a superdiffusive behavior with a mean-squared displacement $\sim t^{3/2}$ (shaded region in Fig. 1). This is similar and somewhat related to an anomalous superdiffusive regime found in active bacterial suspensions [12]. Other regimes include inertial t^2 or propulsive t^2 behaviors, diffusive behavior $\sim t$, and an anomalous correction of the form $-\gamma t^{3/2}$.

We consider a spherical colloidal particle of radius R with an axially symmetric pattern of surface activity as shown in Fig. 2(a), which in essence leads to the release of (excess) product particles $\mathcal{P}$ with diffusion coefficient D. This could correspond to a chemical reaction $\mathcal{S} \rightarrow \mathcal{S}' + \mathcal{P}$ catalyzed on the surface of the colloid, with the simplifying assumption that the produced $\mathcal{S}'$ will act almost like the consumed $\mathcal{S}$ [see Fig. 2(b)]. On the other hand, it could also correspond to a system that actively releases $\mathcal{P}$ particles from the interior of the sphere. The product particles will have a concentration profile $C(\mathbf{r}, t)$ that could interact with the fluid in the vicinity of the colloid due to surface phoretic effects, and cause relative motion with a slip velocity $\mathbf{v}_s(\theta, \phi, t) = \mu \nabla_{\|} C(R, \theta, \phi, t)$ [in standard spherical coordinates; see Fig. 2(a)], where μ is the surface mobility and $\nabla_{\|}$ denote the lateral gradient [13]. In diffusiophoresis, $\mu = k_B T \lambda^2/\eta$, where $k_B T$ is the thermal energy scale, η is the viscosity of water, and λ is the Derjaguin length [14], which is defined in terms of the interaction potential $W(z)$ between the diffusing particles and the surface of the colloid as $\lambda^2 = \int_0^\infty dz z[1 - e^{-W(z)/k_B T}]$. Note that, in our notation, λ^2 will be positive (negative) for repulsive (attractive) effective surface potentials. In the case of ionic systems λ will be set by the Debye length [8,13]. Averaging over the surface of the sphere (using integration over the solid angle Ω), we can obtain the instantaneous velocity of the colloid as $\mathbf{v}(t) = -\frac{1}{4\pi} \times \int d\Omega \, \mu \nabla_{\|} C(R, \theta, \phi, t)$.

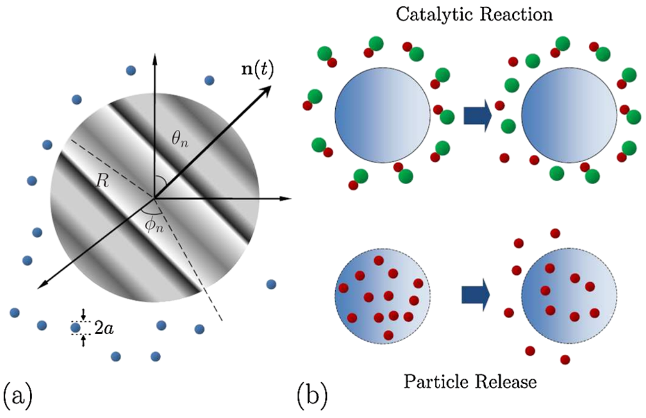


FIG. 2 (color online). (a) Schematics of an instantaneous configuration of an axially symmetric surface-active spherical colloid. (b) The model could correspond to a chemical reaction catalyzed on the surface with the simplifying assumption that one of the product particles is very similar to the substrate, or a container that releases particles through channels.

The axis of symmetry of the colloid, which points to the direction of propulsion for sufficiently asymmetric patterns [6], is defined by the unit vector $\mathbf{n}(t) = (\sin\theta_n(t) \times \cos\phi_n(t), \sin\theta_n(t) \sin\phi_n(t), \cos\theta_n(t))$ [see 2(a)]. The stochastic nature of $\mathbf{n}(t)$ due to rotational diffusion of the colloid causes the cloud of product particles to constantly redistribute, which will in turn make the velocity of the active colloid fluctuate. To get the instantaneous velocity of the colloid, we need to solve the diffusion equation for the concentration profile of the product particles, namely,

$$\partial_t C(\mathbf{r}, t) - D \nabla^2 C(\mathbf{r}, t) = \alpha(\theta, \phi, t) \delta(r - R), \qquad (1)$$

subject to the boundary condition of vanishing normal current on the surface of the sphere. In Eq. (1), $\alpha(\theta, \phi, t)$ is the surface activity function of the sphere, i.e., rate per unit area of the introduction of (excess) product particles. For axially symmetric surface activity, we can represent the function in terms of the spherical harmonics as $\alpha(\theta, \phi, t) = \sum_{\ell, m} (\frac{4\pi}{2\ell+1}) \alpha_\ell Y^*_{\ell m}(\theta_n(t), \phi_n(t)) Y_{\ell m}(\theta, \phi)$. Equation (1) only gives the average density, and the linear relation between the velocity and the concentration profile suggests that in order to calculate velocity correlations we need to incorporate the density fluctuations as well, which we do using the method outlined in Ref. [6].

Using the formulation described above we can calculate the velocity autocorrelation function for the active colloid $A^{vv}(t) \equiv \langle \mathbf{v}(t) \cdot \mathbf{v}(0) \rangle$ as well as the mean-squared displacement $\Delta L^2(t) \equiv \langle [\mathbf{r}(t) - \mathbf{r}(0)]^2 \rangle = \int_0^t dt_1 \int_0^t dt_2 \langle \mathbf{v}(t_1) \cdot \mathbf{v}(t_2) \rangle$. There are three important time scales in the problem. The characteristic diffusion time of the product particles around the sphere is $\tau_d = R^2/D$, where $D = k_B T/(6\pi\eta a)$ depends on the radius of the solute particles a. This time scale sets the relaxation time of the redistribution of the particles around the sphere when it changes orientation. The rotational diffusion time, $\tau_r = 4\pi\eta R^3/k_B T$, controls the changes in the orientation of the sphere, and is defined via the orientation autocorrelation function: $\langle \mathbf{n}(t) \cdot \mathbf{n}(0) \rangle = e^{-t/\tau_r}$ [15]. Finally, the hydrodynamic time that controls the crossover between the inertial and the viscous regimes is given as $\tau_h = R^2/\nu$, where $\nu = \eta/\rho$ is the kinematic viscosity of water that involves the mass density ρ. Practically speaking, we always have $\tau_h \ll \tau_d \ll \tau_r$, although this is not a fundamental requirement.

We can identify three distinct contributions to the velocity autocorrelation function: (1) a contribution from the density fluctuations, which turns out to be only sensitive to the overall symmetric component of the activity and is present even for nonpropelled active colloids, (2) a contribution from the asymmetric component of the activity, and (3) a hydrodynamic contribution that entails the passive diffusion of the colloid and the hydrodynamic long-time tail. These will lead to distinct contributions to the mean-squared displacement, which will add up to make the total mean-squared displacement, namely, $\Delta L^2(t) = \Delta L^2_{\rm sym}(t) + \Delta L^2_{\rm asym}(t) + \Delta L^2_{\rm hyd}(t)$. We will focus on each of these contributions separately below.

Symmetric contribution.—The density fluctuations that are accounted for by adding a noise term to Eq. (1) lead to a contribution to the velocity autocorrelation function that is proportional to α_0 [16]—the $\ell = 0$ coefficient in the expression for $\alpha(\theta, \phi, t)$ in terms of the spherical harmonics $[\alpha_0 = \frac{1}{4\pi}\int d\Omega\, \alpha(\theta, \phi, t)]$. This means that the contribution by density fluctuations is only sensitive to the mean overall surface activity of the colloid, and not the patterning structures on it. We find the asymptotic behaviors $A^{vv}_{\rm sym} \simeq (\alpha_0\mu^2/\pi^{3/2}DR^4)(t/\tau_d)^{-1/2}$ for $t \ll \tau_d$ and $A^{vv}_{\rm sym} \simeq (3\alpha_0\mu^2/32\pi^{3/2}DR^4)(t/\tau_d)^{-5/2}$ for $t \gg \tau_d$. We also calculate the contribution of density fluctuations to the mean-squared displacement, which has the asymptotic behaviors of

$$\Delta L^2_{\rm sym} \simeq \frac{8\alpha_0\mu^2}{3\pi^{3/2}D^{3/2}R^3}\, t^{3/2}; \qquad t \ll \tau_d, \tag{2}$$

at short times, and

$$\Delta L^2_{\rm sym} \simeq \frac{2c_1\alpha_0\mu^2}{\pi^2 D^2 R^2}\, t; \qquad t \gg \tau_d, \tag{3}$$

at long times, and a smooth crossover between them. Here, $c_1 = 1.178\,10$ is a numerical prefactor.

Asymmetric contribution.—Solving Eq. (1) without the noise term, we can calculate the propulsion velocity of the colloid as a function of time for a given time-dependent orientation trajectory. We find $\mathbf{v}(t) = \frac{v_0}{\tau_d}\int_{-\infty}^{t} dt'\,\mathcal{M}(t - t')\mathbf{n}(t')$, where $v_0 = -\alpha_1\mu/(3D)$ is the mean propulsion velocity [6], and the *memory* kernel is given as $\mathcal{M}(t) = \frac{2}{\pi}\int_0^\infty du\, \frac{u^{3/2}}{(u^2+4)}\, e^{-u(t/\tau_d)}$, with asymptotic behaviors $\mathcal{M}(t) \simeq \frac{2}{\sqrt{\pi}}(t/\tau_d)^{-1/2}$ for $t \ll \tau_d$ and $\mathcal{M}(t) \simeq \frac{3}{8\sqrt{\pi}}(t/\tau_d)^{-5/2}$ for $t \gg \tau_d$. Note that the propulsion velocity is controlled by the $\ell = 1$ term (α_1) in the surface activity profile.

Rotational diffusion of the colloid randomizes its orientation over the time scale τ_r, which leads to a contribution to the velocity autocorrelation function of the form of a convolution between two memory kernels and the orientation autocorrelation function. This leads to a velocity autocorrelation function [16], which has three different regimes, due to the presence of two characteristic time scales τ_d and τ_r. At short times, $t \ll \tau_d \ll \tau_r$, we find $A^{vv}_{\rm asym} \simeq v_0^2[1 - \frac{4c_2}{\pi}\frac{\tau_d}{\tau_r} - \frac{1}{2}\frac{\tau_d^{3/2}}{\tau_r^{5/2}}t + \frac{4}{\pi}\frac{t^2}{\tau_d\tau_r}\ln(\frac{t}{\tau_d})]$ where $c_2 = 0.642\,699$ is a numerical prefactor, which implies that the autocorrelation function will be rounded off at small t. For intermediate time, $\tau_d \ll t \ll \tau_r$, we find $A^{vv}_{\rm asym} \simeq v_0^2[1 - \frac{t}{\tau_r} - \frac{1}{\sqrt{\pi}}\frac{\tau_d^{3/2}}{\tau_r}t^{-1/2}]$, and for long times, $\tau_d \ll \tau_r \ll t$, we find $A^{vv}_{\rm asym} \simeq v_0^2[e^{-t/\tau_r} + \frac{3}{4\sqrt{\pi}}\tau_r\tau_d^{3/2}t^{-5/2}]$, which means that the decay at long times is primarily algebraic and not exponential. Consequently, the mean-squared displacement will have three different regimes. We find the asymptotic form of

$$\Delta L^2_{\rm asym} \simeq v_0^2 t^2\left[1 - \frac{4c_2}{\pi}\left(\frac{\tau_d}{\tau_r}\right)\right]; \qquad t \ll \tau_d \ll \tau_r, \tag{4}$$

at short times,

$$\Delta L^2_{\rm asym} \simeq v_0^2 t^2 - \left(\frac{8}{3\sqrt{\pi}}\right)\frac{v_0^2\tau_d^{3/2}}{\tau_r}\, t^{3/2}; \qquad \tau_d \ll t \ll \tau_r, \tag{5}$$

at intermediate times, and

$$\Delta L^2_{\rm asym} \simeq 2v_0^2\tau_r t; \qquad \tau_d \ll \tau_r \ll t, \tag{6}$$

at long times, with a smooth crossover between them.

Hydrodynamic contribution.—Thermal fluctuations of the solvent fluid velocity also contribute to the velocity autocorrelation function of the sphere, because of the no-slip boundary condition between the fluid and the colloid. Performing a similar calculation, one finds [17] $\Delta L^2_{\rm hyd} \simeq 6D_0 t - \frac{2k_BT\rho^{1/2}}{\pi^{3/2}\eta^{3/2}}t^{1/2}$ for $t \gg \tau_h$, where $D_0 = k_BT/(6\pi\eta R)$ is the bare diffusion coefficient of the colloid. The first term in the above equation describes the standard passive diffusion of the sphere while the second term corresponds to the hydrodynamic long-time tail [18,19]. At short times when $t \ll \tau_h$, one finds $\Delta L^2_{\rm hyd} \simeq 3(\frac{k_BT}{M_{\rm eff}})t^2$, where $M_{\rm eff}$ is the effective inertial mass of the colloid in water. The above results are summarized in Fig. 1.

Discussion.—At the longest time scales ($t > \tau_r$), all of the contributions are diffusive, leading to a total effective diffusion coefficient

$$D_{\rm eff} = \frac{k_BT}{6\pi\eta R} + \frac{4\pi\alpha_1^2\mu^2\eta R^3}{27D^2k_BT} + \frac{c_1\alpha_0\mu^2}{3\pi^2D^2R^2}. \tag{7}$$

The different terms in the above expression exhibit different R dependencies, which causes the asymmetric contribution to be dominant for $R \gtrsim [Dk_BT/(\alpha_1\mu\eta)]^{1/2}$, while the symmetric contribution takes over when $R \lesssim \alpha_0\mu^2\eta^2/(D^2k_BT)$. At the shortest time scales, on the other hand, the contribution due to phoretic effects will also be dominated by inertial effects that should lead to ballistic contributions (see Fig. 1).

In the intermediate times, we observe a number of anomalous behaviors. For symmetric active colloids, the superdiffusive $t^{3/2}$ behavior [Eq. (2)] for $\tau_h < t < \tau_d$ is a new regime (for isolated self-propelled particles) where the motion is neither ballistic nor diffusive. The reason a symmetric particle can move at all is because density fluctuations of the cloud of solute particles can instantaneously produce an asymmetric distribution and therefore a net propulsion in some direction. This motion, however, will be decorrelated via density fluctuations themselves, leading to fluctuations without symmetry breaking. We can understand the form of Eq. (2) as follows: using $\Delta L^2 \sim v^2t^2$, and putting $v \sim \mu\nabla C \sim \mu\delta C/R$, we find $\Delta L^2 \sim \mu^2\langle\delta C(t)\delta C(0)\rangle t^2/R^2$. The density autocorrelation function can be written as $\langle \delta C(t) \delta C(0) \rangle = \langle \delta C^2 \rangle k(t)$, involving the density fluctuations $\langle \delta C^2 \rangle$ and the kernel $k(t)$ that controls the relevant relaxation mode [20]. Here, relaxation is controlled by diffusion, hence $k(t) \sim 1/(Dt)^{3/2}$, and the number fluctuations are controlled by the average number of particles ($\langle \delta N^2 \rangle \sim N_{\rm av}$)—as inherent to any Poisson process—that yield $\langle \delta C^2 \rangle \sim C_{\rm av}$. On the other hand, the average density is controlled by the average particle production rate (per unit area) α_0 as $C_{\rm av} \sim (\alpha_0 R^2 t)/R^3$. Putting these all together, we find Eq. (2). This shows that the active velocity fluctuations are controlled by two mechanisms: particle production (that controls the density fluctuations) and diffusion of the produced particles. Interestingly, a similar $t^{3/2}$ power law has been observed in the motion of passive tracer particles in a bath of bacteria whose flagella stir up the fluid (and theoretically accounted for using a phenomenological continuum active medium theory) [12], and in ion channel gating [21], both of which cases are also governed by some sort of density fluctuations [22].

For asymmetric particles when $\tau_h < t < \tau_d$, the $t^{3/2}$ contribution is added (with a positive coefficient) to the t^2 propulsive term. On the other hand, for $\tau_d < t < \tau_r$ the memory effect that exists for self-propelled asymmetric colloids introduces an anomalous *anticorrelation* (i.e., contribution with negative sign) in the velocity autocorrelation function and the mean-squared displacement [Eq. (5)]. Such anomalous corrections, which have also been observed in continuum theories of interacting active self-propelled particles [11], are reminiscent of the effect of the hydrodynamic long-time tail. Note, however, that the anomalous $-\gamma t^{3/2}$ correction in Eq. (5) corresponds to much longer time scales and should be more easily observable than the hydrodynamic long-time tail.

To get a better feel for the working domain of each regime, we can write the time scales (for water at room temperature and using a typical value of $a = 1$ Å) in the following convenient form: $\tau_h = 10^{-6}(R/1\ \mu\mathrm{m})^2$ s, $\tau_d = 10^{-3}(R/1\ \mu\mathrm{m})^2$ s, and $\tau_r = 3(R/1\ \mu\mathrm{m})^3$ s. This shows that while rapid cameras or scattering techniques [12,19] could in principle resolve all the three domains for micron-sized beads, using $R = 20\ \mu$m (which yields $\tau_d = 0.4$ s and $\tau_r = 2.4 \times 10^4$ s) should provide a comfortable working range for an experiment that aims to resolve the anomalous components of the motion. Finally, we note that here we have only focused on the phoretic contributions to velocity fluctuations, and in practice other sources of fluctuations might also be present [10], which need to be taken into account.

In summary, we have shown that active colloidal particles that interact with their self-generated cloud of solute particles can have a range of different types of stochastic motions. Using parameters such as surface activity, surface mobility, and size, we can tune the behavior of active colloidal particles, and this could provide new possibilities in designing functional motile agents for applications in micro- and nanofluidics and targeted delivery.

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