Communication: Memory effects and active Brownian diffusion

Pulak K. Ghosh,1 Yunyun Li,2,3[a] Giampiero Marchegiani,3 and Fabio Marchesoni2,3
1Department of Chemistry, Presidency University, Kolkata 700073, India  
2Center for Phononics and Thermal Energy Science, Tongji University, Shanghai 200092, People’s Republic of China  
3Dipartimento di Fisica, Università di Camerino, I-62032 Camerino, Italy

(Received 9 October 2015; accepted 13 November 2015; published online 1 December 2015)

A self-propelled artificial microswimmer is often modeled as a ballistic Brownian particle moving with constant speed aligned along one of its axis, but changing direction due to random collisions with the environment. Similarly to thermal noise, its angular randomization is described as a memoryless stochastic process. Here, we speculate that finite-time correlations in the orientational dynamics can affect the swimmer’s diffusivity. To this purpose, we propose and solve two alternative models. In the first one, we simply assume that the environmental fluctuations governing the swimmer’s propulsion are exponentially correlated in time, whereas in the second one, we account for possible damped fluctuations of the propulsion velocity around the swimmer’s axis. The corresponding swimmer’s diffusion constants are predicted to get, respectively, enhanced or suppressed upon increasing the model memory time. Possible consequences of this effect on the interpretation of the experimental data are discussed. © 2015 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4936624]

An artificial microswimmer enhances its diffusivity by harvesting propulsion energy from its suspension fluid,1–3 as a result of some sort of functional asymmetry of its own.4–6 The particle thus propels itself with constant speed $v_0$ but keeps changing direction due to both environmental fluctuations (and/or spatial disorder) and the intrinsic randomness of the propulsion mechanism itself.

For simplicity, we restrict here our analysis to the two-dimensional case of an overdamped swimmer with coordinates $x$ and $y$ in a fixed Cartesian frame, then its spatial diffusion is described by the Langevin equations (LEs)

$$\dot{x} = v_0 \cos \phi + \sqrt{D_0} \xi_x(t), \quad \dot{y} = v_0 \sin \phi + \sqrt{D_0} \xi_y(t),$$

where $D_0$ is the intensity of the thermal noise and $\phi(t)$ denotes the instantaneous direction of the propulsion velocity with respect to the $x$ axis [see inset of Fig. 1(a)]. To model the angular dynamics of the swimmer, two assumptions are generally made in the current literature (with a few significant exceptions7–9 discussed below): (a) $v_0$ has a fixed direction in the particle’s reference frame, say, along some symmetry axis, so that a change in the direction of the propulsion velocity implies a rotation of the swimmer; (b) such rotations are assumed to be mostly of thermal nature and, therefore, uncorrelated in time. Accordingly, the swimmer’s angular dynamics is modeled by a third LE,

$$\dot{\phi} = \sqrt{D_\phi} \xi_\phi(t),$$

where $D_\phi$ is the intensity of the rotational fluctuations. The noises appearing in all three LEs are Gaussian, stationary, zero-mean valued, and delta-correlated, that is, $\langle \xi_i(t) \xi_j(0) \rangle = 2\delta_{ij} \delta(t)$, with $i, j = x, y, \phi$. The exact result,10

$$\langle \cos \phi(t) \cos \phi(0) \rangle = (1/2) \exp[-D_\phi t],$$

obtained by combining the general identity,

$$\langle \cos \phi(t) \cos \phi(0) \rangle = (1/2) \exp[-(\Delta \phi^2(t))/2],$$

valid for any Gaussian process $\phi(t)$, and the mean square displacement,11

$$\langle \Delta \phi^2(t) \rangle = 2D_\phi t,$$

from LE (1), suggests to interpret the reciprocal of $D_\phi$ as an angular diffusion time. Note that Eq. (1) still describes a memoryless process.

The corresponding swimmer’s spatial diffusion constant has also an exact analytical expression,12 that is,

$$D \equiv \lim_{t \to \infty} \langle x(t)x(0) \rangle = \lim_{t \to \infty} \langle y(t)y(0) \rangle = D_0 + D_s,$$

with $D_s = v_0^2/2D_\phi$. The quantities $v_0$ and $D_s$ are experimentally accessible, so that $D_\phi$ is usually estimated from the above model-dependent expression for $D_s$. Moreover, $D_0$ and $D_\phi$ are often compared to assess their relationship as, respectively, the translational and rotational constant associated with a unique underlying diffusive mechanism.13–16 In this communication, we show that releasing assumptions (a) and (b) itemized above impacts the estimate of $D_\phi$ and, as a consequence, the interpretation of its physical meaning.

(1) Finite angular time correlation. We expect that rotational fluctuations of the swimmer and, therefore, random changes in the direction of its velocity are caused to an appreciable extent by the noisy nature of the propulsion mechanism itself.4,5 The complex interactions between the swimmer and the active suspension fluid occur on finite spatio-temporal scales.3–5 The ensuing orientational fluctuations of the swimmer, contrary to assumption (b), are thus characterized by at least one finite relaxation rate, $\kappa_\phi$. Stated otherwise, $\phi$ is more appropriately described by the Ornstein-Uhlenbeck process,

$$\dot{\phi} = -\kappa_\phi \phi + \kappa_\phi \sqrt{D_\phi} \xi_\phi(t).$$

[a]Electronic mail: yunyunli@tongji.edu.cn
The idea of adding an extra time scale, $\kappa^{-1}$, in the angular relaxation mechanism of an active Brownian particle is corroborated by the simulation work of Peruani and Morelli.\textsuperscript{7} Second order LE (4) reproduces the standard first-order stochastic differential Eq. (1) only in the limit of zero-correlation time, i.e., for $\kappa_\phi \to \infty$. Accordingly, the angular diffusion law now reads\textsuperscript{11}

$$
\langle \Delta \phi^2(t) \rangle = 2D_\phi [t - (1 - e^{-\kappa_\phi t})/\kappa_\phi].
$$

The spatial diffusion constant follows immediately from Kubo’s formula,\textsuperscript{12}

$$
D = D_0 + \int_0^\infty C(t) dt,
$$

with $C(t) = v_0^2 \langle \cos \phi(t) \cos \phi(0) \rangle$. Since the process in Eq. (4) is Gaussian, $C(t)$ can be expressed in terms of $\langle \Delta \phi^2(t) \rangle$, Eq. (5), by making use of Eq. (2), which allows us to formally perform the integration in Eq. (6); hence

$$
D = D_0 + D_\phi \Gamma(D_\phi/\kappa_\phi) \sum_{m=0}^{\infty} \frac{(D_\phi/\kappa_\phi)^{m+1}}{\Gamma(m+1+D_\phi/\kappa_\phi)},
$$

where $\Gamma(x)$ denotes a gamma function. Two limits of this sum can be calculated analytically,

$$
D \approx D_0 + D_\phi (1 + D_\phi/\kappa_\phi),
$$

for $D_\phi/\kappa_\phi \ll 1$ and

$$
D \approx D_0 + D_\phi \sqrt{\pi/2} (D_\phi/\kappa_\phi - 1),
$$

for $D_\phi/\kappa_\phi \gg 1$. Our analytical predictions compare well with the simulation data obtained by numerically integrating the model LEs\textsuperscript{7} [see Fig. 1(a)].

(2) Velocity fluctuations in the body frame. Contrary to what stipulated in assumption (a), the instantaneous direction of the propulsion velocity can fluctuate around its mean, represented by the swimmer’s axis of angular coordinate $\phi$. The resulting swimmer’s dynamics is thus modeled through a set of four LEs, namely,

$$
\begin{align*}
\dot{x} &= v_0 \cos(\phi + \psi) + \sqrt{D_0} \xi_x(t), \\
\dot{y} &= v_0 \sin(\phi + \psi) + \sqrt{D_0} \xi_y(t), \\
\dot{\phi} &= \sqrt{D_\phi} \xi_\phi(t), \\
\dot{\psi} &= -\kappa_\phi \psi + \sqrt{D_\psi} \xi_\psi(t),
\end{align*}
$$

(10)

where all noises are defined as above and the auxiliary angle $\psi$ represents the misalignment between the instantaneous propulsion velocity and its mean [see inset of Fig. 1(a)]. Here, the restoring constant $\kappa_\phi$ plays the role of a relaxation rate, whereas the $\psi$ fluctuations have zero mean, $\langle \psi \rangle = 0$, and magnitude $\langle \phi^2 \rangle = D_\phi/\kappa_\phi$. For $D_\phi/\kappa_\phi \ll 1$, the velocity fluctuations in the body frame are suppressed and the standard model with $\psi = 0$ recovered. The model of Eq. (10) exhibits normal diffusion as always expected for realistic self-phoretic swimmers, as long as one assumes observation times larger than $D_\phi^{-1}$.\textsuperscript{9} In this regard, we stress that the memory effects postulated here are intrinsic to the propulsion mechanism and should not mistaken for the additional inertial and translational memory effects considered by Golestanian.\textsuperscript{9}

In order to apply Kubo’s formula, Eq. (6), to evaluate the diffusion constant, we need first to calculate the autocorrelation function $C(t) = v_0^2 \langle \cos(\phi(t) + \psi(t)) \cos(\phi(0) + \psi(0)) \rangle$. Simple algebraic manipulations yield

$$
\begin{align*}
C(t) &= \frac{v_0^2}{2} \left( e^{-\kappa_\phi t} e^{-\kappa_\psi t} + e^{-(\kappa_\phi + \kappa_\psi) t} \right),
\end{align*}
$$

(11)

with $\langle \Delta \phi^2(t) \rangle = 2D_\phi t$. Kubo’s integral can then be analytically calculated as a power series, i.e.,

$$
D = D_0 + D_\phi e^{-D_\phi/\kappa_\phi} \sum_{m=0}^{\infty} \frac{1}{m^m} \frac{(D_\phi/\kappa_\phi)^{m+1}}{m! (D_\psi/\kappa_\psi) + (D_\phi/\kappa_\phi)}. 
$$

(12)

This expression is plotted against the simulation data in Fig. 1(b) for different values of $D_\phi$.

In the realistic case when the velocity angular fluctuations in the body frame are small, $D_\phi/\kappa_\phi \ll 1$, and their relaxation time is much shorter than the body’s characteristic rotation time, i.e., for $\kappa_\phi/D_\phi \gg 1$, $D$ in Eq. (12) tends to

$$
D = D_0 + D_\phi e^{-D_\phi/\kappa_\phi}. 
$$

(13)

Vice versa, for a slowly relaxing $\psi(t)$, $\kappa_\phi/D_\phi \ll 1$, the spatial diffusion approaches the even smaller value

$$
D = D_0 + D_\phi/(1 + D_\psi/D_\phi). 
$$

(14)

Both limits closely reproduce the numerical data of Fig. 1(b) within the appropriate parameter range.

Discussion. We now compare models (1) and (2). In both cases, memory effects have been expressed in terms of a relaxation rate, respectively, $\kappa_\phi$ and $\kappa_\psi$. In model (1), memory
is deemed as intrinsic to the nonequilibrium microscopic processes responsible for the swimmer’s propulsion — for instance, a shot-noise like sequence of finite-time pulses, or power-strokes, associated with the chemical reactions catalyzed by the active tips of the swimmer. In model (2), we argued that the direction of the shifts associated with such power-strokes may fluctuate around its average orientation in the body frame as an effect of the extended geometry of the swimmer’s active. Of course, the above memory mechanisms might well operate simultaneously. This is the case, for instance, of the reaction-driven swimmers of Ref. 8, where two sources of velocity fluctuations are singled out, namely, the product particle density fluctuations and the randomness in the catalytic reaction that leads to the product particle release.

By inspecting Figs. 1(a) and 1(b), we immediately recognize that finite memory-time corrections to the spatial diffusion constant have opposite sign: \( D \) gets either enhanced or suppressed by decreasing the corresponding model relaxation rate, namely, \( \kappa_0 \) in Fig. 1(a) and \( \kappa_0 \) in Fig. 1(b). The physical interpretation of these opposite behaviors is straightforward. In model (1), lowering \( \kappa_0 \) means increasing the persistence time of the propulsion mechanism, which is known to cause excess diffusion. On the contrary, in model (2), weakening the restoring constant \( \kappa_0 \) favors the spatial reorientation of the swimmer’s kinematic velocity and, correspondingly, the suppression of its spatial diffusion.

These remarks have a practical consequence on the interpretation of the experimental data. Upon ignoring memory effects, one determines \( v_0 \) and \( D = D_0 \) by direct measurements and extracts \( D_0^{\text{exp}} \) from the identity, \( D = D_0 = v_0^2/2D_0^{\text{exp}} \), provided by the standard model, see Eq. (3). However, if we reconsider such a procedure in view of model (1), the computed \( D_0^{\text{exp}} \) must differ from \( D_0 \). For instance, from Eq. (8) for \( D_0/\kappa_0 \ll 1 \), \( (D_0^{\text{exp}})^{-1} = D_0^{-1} + \kappa_0^{-1} \). The same conclusion holds for model (2), where Eq. (13) for \( \kappa_0 \gg D_0, D_0 \) implies that \( D_0^{\text{exp}} = D_0 \kappa_0/D_0 \) and Eq. (14) for \( \kappa_0 \gg D_0 \) and \( \kappa_0 \ll D_0 \) yields \( D_0^{\text{exp}} = D_0 + \kappa_0^{-1} D_0 \). In other words, \( D_0^{\text{exp}} \) systematically under- or over-estimates \( D_0 \), depending on which model better reproduces the active swimmer’s dynamics.

In the regime of short memory times, the difference \( |D_0^{\text{exp}} - D_0| \) is proportional to the ratios \( D_0/\kappa_0 \) in model (1) and \( D_0/\kappa_0 \) in model (2). Being a measure of the spatio-temporal structure of the propulsion mechanism, the parameters introduced in models (1) and (2), \( \kappa_0, \kappa_0, \) and \( D_0 \), may vary with the physico-chemical properties of the active fluid, the fabrication specifics (and defects) of the active microswimmers, and their interactions with the surrounding fluid. As a consequence, the estimated values of \( D_0^{\text{exp}} \) also depend on all those factors. Therefore, the measured quantity \( D_0^{\text{exp}} \) for an active swimmer cannot be analyzed as the rotational counterpart of the translational diffusion constant, \( D_0 \), if not after correcting, case by case, for the memory effects peculiar of the propulsion mechanism actually at work. Variations of the \( D_0 \) to \( D_0 \) ratio under different swimmer’s operation conditions have already been reported in the earlier literature.

Concluding remarks. In this communication, we emphasized the dynamical role of rotational fluctuations. In a forthcoming publication, we will generalize both models (1) and (2) in various ways. In model (1), the constant propulsion speed, \( v_0 \), will be explicitly derived from a fluctuating “effective force” to mimic the microscopic discreteness of the propulsion mechanism. However, since the average values of the such a propulsion force are typically rather large in comparison to its standard deviation, no significant contribution to the swimmer’s diffusion is expected. Model (2), instead, can be conveniently improved to account for possible instability effects. Due to its functional asymmetry, the center of mass and the center of the propulsion force acting upon a swimmer, like the Janus particle in the inset of Fig. 1(a), may well rest a finite distance apart, say, along the symmetry axis. As a consequence, the angular fluctuations of the propulsion velocity vector are associated with an additional instantaneous torque. Although such a random torque has zero mean, it suffices to further suppress active diffusion. Finally, for more asymmetric geometries, where the fluctuating propulsion velocity points in average at an angle from the swimmer’s axis, \( \langle \phi \rangle = \psi_0 \), the ensuing nonzero average torque drives the swimmer along spiraling trajectories. These generalizations of model (2) will allow us to study the effects of chirality on active diffusion.16,19

We thank RIKEN’s RICC for computational resources. Y. Li is supported by the NSF China under Grant No. 11505128 and P.K.G. by SERB-Start Up Research Grant (Young Scientist) File No. YSS/2014/000853 and the UGC-BSR start-up Grant No. F.30-92/2015.