Diffusion of active particles in a complex environment: role of surface scattering

Theresa Jakuszeit, Ottavio A. Croze, and Samuel Bell
Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, U.K.
(Dated: July 12, 2018)

Experiments have shown that self-propelled particles can slide along the surface of a circular obstacle without becoming trapped. Using simulations and theory, we study the impact of boundary conditions on diffusive transport in an obstacle lattice. We find that sliding can maintain large diffusive transport even at high obstacle density, unlike classical specular reflection. These dynamics are described by a model based on Run-and-Tumble particles with microscopically derived reorientation functions arising from obstacle-induced tumbles. Our results show that non-classical surface scattering introduces a dependence on the lattice geometry at high densities, which can guide particles.

The field of active matter covers the broad spectrum of particles which move by consuming energy from their environment [1]. These range from flocks of birds and insect swarms [2, 3], to cell tissues [4], microswimmers [5], microtubuli [6, 7], and enzymes [8]. Microswimmers such as bacteria and Janus particles self-propel at low Reynolds numbers, the latter being directly powered by an asymmetric chemical reaction on the particle surface, the former by rotating helical filaments. The propulsion's mechanisms set up complicated hydrodynamic flows, which determine the characteristics of interactions, both with other microswimmers, and with the boundaries of their environment. These boundary interactions may perform an essential function in nature. Surface-induced accumulation is an important step in the formation of biofilms, which are involved in many chronic diseases and pathogen spread [9, 10]. Blood pathogens are adapted to swimming in crowded environments [11], sperm cells follow the wall of the genital tract to reach the egg cell [12–14], and artificial Janus particles have been guided through microfluidic edges [15] and through obstacle arrays [16–18].

The nature of particle-surface interactions relies on a microswimmer’s propulsion mechanism, including steric and hydrodynamic effects. Microalgae, which are “puller” type swimmers, are scattered off surfaces [19–21], leading to billiard-like motion in polygon structures [22]. In contrast, “pusher” type swimmers, such as bacteria or Janus particles, are trapped by hydrodynamic effects near flat surfaces, where they accumulate [23, 24]. When the surface is instead convex, this trapping time can be reduced [25]. In particular, bacteria trace along convex surfaces such as microfluidic pillars before escaping with a small angle [25].

The modelling of these scenarios typically follows one of two approaches: hydrodynamic models, or random walk models. With a full hydrodynamic approach, the particle-surface interactions can be studied by modelling the active particle as a hard sphere with defined tangential surface velocity [28]. A recent study explored the migration of active particles through a body-centered cubic lattice of spheres of the same size as the particle [29].

Depending on the swimmer type and packing density, the authors found trapped, random walk and straight trajectories. The computational demands of the simulations, however, prevented study of long-time behavior. Random walk models can be used to study the diffusive behavior of active particles. Diffusion in complex media has been studied for several boundary interactions: for model particles that evade obstacles [30], particles that are trapped before being randomly reorientated [31], and particles that interact with obstacles via an excluded volume potential [32]. Hydrodynamic boundary interactions have been shown to play an important role in active systems, e.g. in the control of flow-induced phase separation [33]. Similarly, pusher-type boundary interactions may guide microswimmers through their environment [17, 34], which would facilitate diffusion.

In this Letter, we combine the knowledge of pusher-like hydrodynamic boundary conditions with a persistent random walk model. Through simulations, we show that these boundary conditions allow for large diffusive transport even at high obstacle densities. This is in contrast to classical specular reflection, as in the Lorentz gas model [35]. We develop a theoretical framework based on run-and-tumble particles [36, 37] that includes the microscopic details of obstacle-induced reorientation. This highlights the potential of guiding and sorting active particles by geometric restriction in regular obstacle arrays.

Model. We consider $N_P$ active particles in a two-dimensional space in which obstacles are placed in a hexagonal lattice. The centers of the obstacles are fixed with distance $d$, and the obstacle radius $R$ is varied. The equations of motion for the $i$-th particle are given by

$$\dot{x}_i = v \mathbf{p}(\phi_i)$$

$$\dot{\phi}_i = \sqrt{2D_R} \xi_i(t)$$

where dot denotes the time derivative, $v$ is the particle speed, $x_i$ and $\phi_i$ correspond to the position and moving direction of the $i$-th particle, respectively, and the unit vector $\mathbf{p} = [\cos \phi, \sin \phi]$. The white noise in Eq. (2) obeys $\langle \xi(t) \rangle = 0$ and $\langle \xi_i(t) \xi_j(t') \rangle = \delta_{ij} \delta(t - t')$. Thus, the moving direction undergoes rotational diffu-
sion with $\langle \varphi(t)^2 \rangle = 2D_R t$. As a result, the particle performs a persistent random walk with persistence length $l_p = v/(2D_R)$.

To capture the non-classical particle-surface interaction, we introduce a sliding boundary condition as illustrated in Fig. 1(a) inset [24]. Upon collision with an obstacle, the collision angle $\beta$ is defined as the angle between the tangent at the collision point and the orientation $p$. If $\beta < \pi/2$, the particle travels clockwise around the obstacle; if $\beta \geq \pi/2$, the particle travels counter-clockwise. The particle moves along the obstacle to traverse a central angle $\alpha$. For bacteria, the trapping time depends on obstacle radius, properties of the propulsion mechanism which can be characterized by the force dipole strength in Stokes equation [25], and diffusion [27]. For Janus particles, the trapping time is a function of obstacle size, diffusion and fuel concentration [15]. Moreover, the collision angle $\beta$ will affect the time that it takes the particle to reorientate on the obstacle before it can escape [27], but a theoretical description covering the entire range of possible $\beta$ is missing. Therefore, we explore the effect of boundary conditions assuming a fixed central angle $\alpha$. When the particle leaves the obstacle, its orientation $p$ makes a tangent to the obstacle’s surface. We also consider a reflecting boundary condition, where a particle is reflected with an angle equal to the incident angle, as illustrated in Fig. 1(b).

The system of Eqs. (1) and (2) is solved numerically, and example particle tracks are shown in Fig. 1. We derive the diffusion coefficient from $N_P$ simulated particle tracks by fitting the mean square displacement as $\langle \delta x(t)^2 \rangle = 4D_{\text{eff}} + 4D_{\text{eff}}\eta[\exp(-t/\eta) - 1]$, where $\eta$ is the time scale of ballistic motion.

Reflecting boundary condition. We first establish the diffusive properties of active particles with a reflective boundary condition. Here, we recognize an analogy to the Lorentz gas model, in which particles move ballistically between obstacles [35]. The Santaló formula is a well-known result for the mean-free path of a Lorentz gas [39] given by $\lambda = \pi A/P$, where $A$ and $P$ are the free area and obstacle perimeter in a unit cell, respectively. Since the active particles move diffusively at large time scales, we derive an active version of Santaló’s formula with a circle of radius $l_p$ as an additional boundary. This yields the mean-free path of an active particle as $\lambda_p = 2\pi N A/(NP + 2\pi l_p)$, where $N$ is the number of unit cells included in the circle of radius $l_p$. For a hexagonal lattice of circular obstacles, we obtain $\lambda_p = \sqrt{3}d^2/2 - \pi R^2$, $P = 2\pi R$ and $N = \pi l_p^2/(\sqrt{3}d^2/2)$ [40]. The inset in Fig. 2(a) plots the theoretical prediction and the diffusion coefficient fitted from simulations on a log-log scale, showing that at large $R/d$ the diffusion coefficient scales as $\ln(1/\rho)$, where obstacle density $\rho = 2\pi/\sqrt{3} (R/d)^2$.

Sliding boundary condition. By contrast, numerical solutions of Eqs. (1) and (2) with a sliding boundary condition reveal that diffusion depends both on the obstacle density $\rho$ and the central angle $\alpha$ [see Fig. 2(b)]. Surprisingly, a large diffusive transport can be sustained even at large obstacle density $\rho$ for certain values of $\alpha$. Despite frequent obstacle collisions, the reorientation is small because the sliding boundary condition conserves the major component of the velocity vector for small to intermediate values of $\alpha$. Large values of $\alpha$, on the other hand, cause a particle to retrace much of its track. The typical pusher surface interaction can, thus, lead to an increase in effective diffusion compared to the classical reflection.

Theoretical framework. We derive a theoretical description based on the model of run-and-tumble particles (RTP) [36, 37]. In this Letter, an effective ‘tumble’ is defined as an obstacle-induced reorientation of the particle, and the ‘run’ between obstacle collisions is influenced by rotational diffusion. The diffusion coefficient for an RTP also undergoing rotational diffusion is known to be

$$D = \frac{v^2}{2[D_R + (1 - \langle \cos \theta \rangle)/\tau]}$$

where $\tau$ is the mean run time and $\theta = \theta(\alpha, P(\beta))$ is the reorientation angle during a tumble [41, 42]. The reorientation angle is the combination of alignment upon collision with the obstacle, $\beta$, and sliding according to the central angle, $\alpha$: $\theta = \alpha - \beta$. The average $\langle \cos \theta \rangle$ is performed over the collision angle $\beta$, with probability distribution $P(\beta)$. To derive the distribution, we assume that a particle can start at any point in free space with uniform distribution of directions, and then travels in a straight line. The probability distribution of a collision angle $\beta$ at a given distance $x$, $P_x(\beta)$, can be written in terms of $P(\phi)$, where $\phi(\beta, x)$ is the angle between $x$ and the moving direction. Thus, $P_x(\beta) d\beta = P(\phi) d\phi$. For circular obstacles, $\phi(\beta, x)$ follows geometrically from the sine rule so that $\phi(\beta, x) = \sin^{-1}(R \cos \beta/x)$ [40]. Differentiation yields the Jacobian $|d\phi/d\beta| = |d\beta/d\phi|^{-1} = R \sin(\beta)(x \sqrt{1 - R^2 \cos^2 \beta/x^2})^{-1}$. Finally, we average over all initial positions

$$P(\beta) = \lim_{L \to \infty} \int_{-L}^{L} \int_{-L}^{L} \frac{\sin \beta}{2} d\beta = \frac{\sin \beta}{2}.$$
where $L$ is the system size. Despite using deterministic trajectories to calculate this distribution, it fits the observed collision angle distribution for simulations at low densities. Performing the average gives the reorientation function as:

$$\langle \cos \theta \rangle = 2 \int_0^{\pi/2} \cos(\alpha - \beta) P(\beta) d\beta = \frac{1}{4} (2 \cos \alpha + \pi \sin \alpha),$$

noting that $\cos \theta$ is even about $\beta = \pi/2$. For the reflecting boundary condition, $\theta = 2\beta$, and $\langle \cos \theta \rangle = -1/3$.

The second parameter in the RTP model (3) is the mean run time $\tau$, which corresponds to the time between obstacle collisions. Because the characteristic time between collisions is independent of the details of the random walk and depends purely on confinement [42], we use the mean collision time $\tau_c = \lambda/v$, where $\lambda$ is the mean free path given by Santalo’s formula. For the sliding boundary condition, the mean run time is adjusted by the time spent on an obstacle, i.e., $\tau = \tau_c + \tau_R$, with residence time $\tau_R = R\alpha/v$. Travelling on the obstacle causes an effective reduction in velocity. When the particle traces along the pillar, it travels a distance $l < v\tau_R$, which gives $v_{\text{obs}} = l/\tau_R$. By the cosine rule, $l = R\sqrt{2 - 2\cos \alpha}$. The effective speed in Eq. (5) is then $v_{\text{eff}} = v\tau_c/\tau + v_{\text{obs}}(\tau - \tau_c)/\tau$.

We first compare the RTP theory to simulations with reflecting boundary condition, using $\langle \cos \theta \rangle = -1/3$ and mean collision time $\tau_c$ ($\tau_R = 0$). As shown in Fig. 2(a), the RTP description yields a good approximation of the simulation results. Using instead a recently derived mean collision time based on Kac’s theorem [31], where $\tau_c^K = 1/\rho$, the RTP model approximates the simulations at low densities but diverges in the high density regime.

For the sliding boundary condition, the RTP framework reproduces the main features of the simulations, see Figs. 2(b) and 2(c): it maintains a large diffusion coefficient for small to intermediate $\alpha$. Since $\tau_c$ is independent of the boundary condition, this must stem from the reorientation function $\langle \cos \theta \rangle$ in Eq. (5), which has a maximum at $\alpha \approx \pi/3$ and a minimum at $\alpha \approx 4\pi/3$. These extrema coincide with the predicted maximum and minimum of the diffusion coefficient observed for small to intermediate $R/d$ in Fig. 2(c). Beyond $\alpha = 4\pi/3$, any increase in the diffusion coefficient due to the reorientation function is suppressed by the increase in residence time $\tau_R$ at large $R$ and $\alpha$.

High density geometrical effects. While the RTP model accounts for the diffusion coefficient $D_{\text{slid}}$ at low to intermediate obstacle densities, it fails to completely describe it at high density. Figures 2(b) and 2(c) show that at high density, the diffusion coefficient in hexagonal lattice simulations splits into a double peak structure with respect to $\alpha$, whereas the RTP model displays a single peak.

Simulations show that particle trajectories in the peaks are made up of long flights along the channels of the lattice, punctuated by turns into the next long flight. This type of trajectory is only possible at higher densities. At lower densities, the particle is not constrained by the lattice geometry, and rotational diffusion will reorient the particle between collisions. However, when $d - 2R \ll l_p$, orientation due to rotational diffusion will be small, and the trajectories are dominated by the interplay between the lattice geometry and the central angle $\alpha$. Therefore, we can use a deterministic model ($D_R = 0$) with only a single channel, as in Fig. 3(a), to explain the peaks in the high density diffusion coefficient. As the particle moves along this channel, it moves from one pillar to the
There are two stable regions of allowed region. Trajectories in the deterministic model too small (the flight. A flight will terminate if the angle becomes θ remain in the region illustrated in Fig. 3(a). Stable flights are trajectories that will be deflected out of the channel on its next collision, at θ next contact will be on the other side of the channel, and if θ > θ*, the next contact will be on the same side of the channel. This means the map g_{θ}(θ) is discontinuous at θ = θ*.

A flight is defined by the sequence of leaving angles θ_n, where the length of the sequence gives the length of the flight. A flight will terminate if the angle becomes too small (θ_n < θ_{min}) or too large (θ_n > θ_{max}) as it will be deflected out of the channel on its next collision, illustrated in Fig. 3(a). Stable flights are trajectories that remain in the region θ_{min} ≤ θ_n ≤ θ_{max} indefinitely. This can happen in two ways: (i) a stable fixed point may exist (a point θ such that g_{θ}(θ) = θ, and |g_{θ}′(θ)| < 1), so that long trajectories have a single repeated leaving angle; (ii) the map g_{θ}(θ_n) is bounded within the allowed region of leaving angles: θ_{min} ≤ g_{θ}(θ_n) ≤ θ_{max} for all θ_{min} ≤ θ_n ≤ θ_{max}, so that no trajectory may leave the allowed region. Trajectories in the deterministic model are ballistic, so that (x^2(t)) ∝ t^2.

The map g_{θ}(θ_n) is plotted for R/d = 0.47 in Fig. 3(b). There are two stable regions of α, shown as the shaded regions α_1 ≤ α ≤ α_2, and α_3 ≤ α < π/2. In the lower region, a stable fixed point for θ_n < θ* develops at α = α_1, and as α increases the map becomes bounded. Flights in this lower shaded region bounce from one side of the channel to the other. At α = α_2 the map again becomes unbounded, as θ_{n+1} > θ_{max}, and since the fixed point has also disappeared, the system loses stability. The upper region has a stable fixed point for θ_n > θ*, so that stable flights run along only one side of the channel in this region. Figure 3(c) matches these stable regions of the deterministic model against the diffusion coefficient found in simulations of the persistent random walk model Eqs. (1) and (2). The deterministic stable regions match well with the observed double peak of overshoots relative to the RTP model. These regions are not ballistic in the non-deterministic model because rotational diffusion will eventually cause long runs to terminate, so the particle still undergoes reorientation, and the MSD is diffusive. Likewise, the noise smears out the peaks relative to the deterministic model. Between these stable regions, the diffusion coefficient collapses back onto the RTP model, which does not consider geometry. The subsidiary peaks seen in the diffusion coefficient are more complicated steady state paths relying on the symmetry of the hexagonal lattice. As a comparison, the diffusion coefficient as a function of α is shown for a square lattice in Fig. 3(c) inset, which reveals only a single peak.

To conclude, we find that non-classical surface interactions have a significant impact on diffusive properties in complex environments, and microscopically derived rules should be included in active particle models. Further, for certain boundary interactions particles are geometrically guided through dense obstacle lattices resulting in large diffusive transport. The time that bacteria spend on an obstacle is influenced by the curvature of the surface as
well as the force dipole strength of the bacterium \cite{26, 27}.
The dipole strength is an intrinsic property of the bacterium depending on the body shape and the propulsion mechanism. Thus, we may speculate that certain hydrodynamic properties of, e.g., soil bacteria could be tailored towards guided transport in complex environments \cite{34}.
Microfluidic studies with post-like structures could explore the effect of those factors on diffusion in complex environments, and test for any long-range hydrodynamic effects.

We thank Eugene Terentjev, Mark Warner and Mike Cates for helpful discussions and feedback on the manuscript. This work has been funded by EPSRC EP/M508007/1 (S.B.), EP/L504920/1 and EP/N509620/1 (T.J.), and the Winton Programme for the Physics of Sustainability (T.J., O.C.).

\[\text{\cite{42956@cam.ac.uk}, sb855@cam.ac.uk}\]
Supplemental Materials: Diffusion of active particles in a complex environment: role of surface scattering

SLIDING BOUNDARY CONDITION

Collision angle distribution

To find the orientation function \( \langle \cos \theta \rangle \), we must first determine the collision angle distribution. At low densities, the collision angle distribution can be approximated by the problem set up in Fig. S1 and described in the main text. Since there is radial symmetry, we do not have to consider a specific direction, \( x \), relative to the circle. The function

\[
\phi(\beta, x) \text{ defined in the main text can be easily worked out through the sine rule:}
\]

\[
\frac{x}{\sin(\beta + \pi/2)} = \frac{R}{\sin \phi},
\]  

(S1)

Deriving the reorientation function

By construction (see Fig. S2), angle FCH must be a right angle. Since the particle leaves at a tangent, OCD is also a right angle. By the alternate angle theorem, OCF=\( \alpha \), and so DCF=\( \pi/2 - \alpha \). Therefore, \( \theta = \pi/2 - \beta - (\pi/2 - \alpha) = \alpha - \beta \).
DIFFUSION COEFFICIENT: REFLECTING BOUNDARY CONDITION

To calculate the diffusion coefficient of reflecting particles, we first considered the deterministic case \((D_R = 0)\). In this case, there is a formula from integral geometry, known as Santalo’s formula, which provides the mean free path length of a billiard moving in an array of obstacles. If the free area available to the billiards in the lattice is given by \(|Q|\), and the constraining boundary is given by \(|\partial Q|\), then the mean free path is

\[
\lambda = \frac{\pi |Q|}{|\partial Q|},
\]

(S2)

For our hexagonal lattice, we know that the ratio between a unit cell’s free area, \(A\) to its constraining surface, \(P\) will be the same as for the whole lattice. Choosing the unit cell to be a rhombus, \(A = \sqrt{3}d^2/2 - \pi R^2\), and \(P = 2\pi R\) (see Fig. S3a) for an illustration), and so the mean free path length then follows immediately as

\[
\lambda = \frac{(\sqrt{3}d^2 - 2\pi R^2)}{4R}.
\]

(S3)

This formula should hold for active particles at high obstacle density, where the persistence length \(l_p = v/D_R\) is much larger than the separation between pillars, \(d - 2R\). The trajectories will then almost look deterministic.

We found the diffusion coefficient \(D = v\lambda/2\) only fits the simulation data when we divide the mean free path length by a factor \(\pi/2\):

\[
D = \frac{v (\sqrt{3}d^2 - 2\pi R^2)}{2\pi R},
\]

(S4)

where \(d\) is the lattice spacing, and \(R\) the radius of the circular pillars. We believe that this is due to the choice of averaging conditions made in the earlier works. As an example, if you wish to calculate the mean free path length in a confining circle, you must average over all possible chords within that circle. Depending on how you construct that average, you get different answers. So, we think that the averaging process used in the works of integral geometry are not appropriate to our case.

Although this result is good for high obstacle densities, we would like a model that works at low densities as well. At low obstacle densities, we can no longer ignore rotational diffusion. The deterministic formula must obviously diverge as \(R \to 0\), as some rays will travel infinitely far before their next collision.

For active particles, the persistence length defines a length over which directional persistence is lost. This is analogous to the mean free path length in a gas. Guided by this, we include a circle with radius of the persistence length into the lattice. This acts as an extra piece of obstacle boundary, and constrains the particle according to its rotational diffusion coefficient. For high obstacle densities, this will play only a minor role; a particle is much more likely to hit an obstacle than the surrounding boundary, as illustrated in Fig. S3b). However, when the obstacle density becomes low enough, then the circle becomes important, as in Fig. S3c). Now, the mean free path becomes (dividing the Santalo formula by the same factor of \(\pi/2\) as above):

\[
\lambda = \frac{2NA}{NP + 2\pi l_p},
\]

(S5)

where \(N = \pi l_p^2/(\sqrt{3}d^2/2)\) is the number of rhombus unit cells within the confining circle. Note that for \(R = 0\), \(A = \sqrt{3}d^2/2\), and so \(\lambda = l_p\) as required.
FIG. S3. An illustration of the lattice geometry in the reflecting boundary condition calculation. A hexagonal lattice can be patterned by rhombi of side length equal to the lattice spacing, as shown in a). The shaded area is the area available to particles per unit cell, $A$, while the blue arcs highlight the obstacle surface per unit cell $P = 2\pi R$. In b) and c), an extra circular boundary is added, with a radius of the persistence length, to account for reorientation via rotational diffusion. In c) this effect will be more marked as the obstacle density is lower.