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Erik Kalz, Hidde D. Vuijk, Iman Abdoli, Jens-Uwe Sommer, Hartmut Löwen, Abhinav Sharma

Angaben zur Veröffentlichung / Publication details:

Kalz, Erik, Hidde D. Vuijk, Iman Abdoli, Jens-Uwe Sommer, Hartmut Löwen, and Abhinav Sharma. 2022. "Collisions enhance self-diffusion in odd-diffusive systems." *Physical Review Letters* 129 (9): 090601. https://doi.org/10.1103/physrevlett.129.090601.

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Editors' Suggestion

Collisions Enhance Self-Diffusion in Odd-Diffusive Systems

Erik Kalz,[1,2,*] Hidde Derk Vuijk,[1,*] Iman Abdoli,[1] Jens-Uwe Sommer,[1,2]
Hartmut Löwen,[3] and Abhinav Sharma[1,2,†]

[1]Leibniz-Institut für Polymerforschung Dresden, Institut Theorie der Polymere, 01069 Dresden, Deutschland
[2]Technische Universität Dresden, Institut für Theoretische Physik, 01069 Dresden, Deutschland
[3]Heinrich Heine-Universität Düsseldorf, Institut für Theoretische Physik II: Weiche Materie, 40225 Düsseldorf, Deutschland

(Received 11 January 2022; revised 15 June 2022; accepted 29 July 2022; published 25 August 2022)

It is generally believed that collisions of particles reduce the self-diffusion coefficient. Here we show that in odd-diffusive systems, which are characterized by diffusion tensors with antisymmetric elements, collisions surprisingly can enhance the self-diffusion. In these systems, due to an inherent curving effect, the motion of particles is facilitated, instead of hindered by collisions leading to a mutual rolling effect. Using a geometric model, we analytically predict the enhancement of the self-diffusion coefficient with increasing density. This counterintuitive behavior is demonstrated in the archetypal odd-diffusive system of Brownian particles under Lorentz force. We validate our findings by many-body Brownian dynamics simulations in dilute systems.

DOI: 10.1103/PhysRevLett.129.090601

Self-diffusion is related to the dynamics of a single particle, commonly referred to as the *tracer particle*, in a homogeneous system of *host particles* [1–4]. For purely repulsive interaction potentials, it is quite intuitive that the tracer particle is hindered in its motion by the host particles giving rise to a slowdown in the dynamics. The long-time self-diffusion coefficient D_s for a system of Brownian particles can be calculated exactly in the low-density limit as $D_s = D_0(1 - \alpha\phi)$, where ϕ is the area fraction, D_0 is the diffusion coefficient at infinite dilution, and α is a numerical factor that depends on the nature of interactions ($\alpha = 2$ for hard-core interactions) [5–8]. The reduction in the self-diffusion coefficient of the tracer particle with increasing density of host particles has been thoroughly demonstrated in experimental and computational studies [9–12].

In this Letter, we study the self-diffusion coefficient in systems which are characterized by probability fluxes that are perpendicular to the density gradients. Analogous to odd-viscosity [13–15], recently such diffusive behavior has been aptly termed as odd diffusive [16] and has attracted considerable attention [17–24]. We show that in odd-diffusive systems, collisions, instead of hindering the motion of the tracer particle, facilitate it resulting in an enhancement of the dynamics. Specifically, we demonstrate that in the low-density limit, increasing the density of host particles leads to an increase in the self-diffusion coefficient of the tracer particle. Moreover, by tuning the odd diffusivity, particles can be rendered dynamically invisible such that the tracer particle diffuses as a free particle.

Odd-diffusive behavior emerges naturally in systems with broken time-reversal and parity symmetry. A charged Brownian particle in a magnetic field is a classic example of a system with broken time-reversal symmetry [17]. Other prominent examples of odd-diffusive systems are strongly damped particles subjected to Magnus [15], or Coriolis [25,26] forces or active chiral fluids [27–30]. In the limit of low persistence length, an active chiral particle follows curved trajectories, similar to the Brownian motion of a charged particle under a magnetic field.

The Fokker-Planck equation (FPE) for the probability density $P(\boldsymbol{r}, t)$ for an odd-diffusive particle reads as

$$\frac{\partial P(\boldsymbol{r}, t)}{\partial t} = \nabla \cdot [\mathbf{D}\nabla P(\boldsymbol{r}, t)], \tag{1}$$

where the diffusion tensor for two-dimensional isotropic systems can be written in the general form [16,17]

$$\mathbf{D} = D_0(\mathbf{1} + \kappa\boldsymbol{\epsilon}), \tag{2}$$

where $\mathbf{1} = \left(\begin{smallmatrix}1 & 0\\ 0 & 1\end{smallmatrix}\right)$ is the identity tensor and $\boldsymbol{\epsilon} = \left(\begin{smallmatrix}0 & 1\\ -1 & 0\end{smallmatrix}\right)$ is the antisymmetric Levi-Civita symbol in two dimensions. D_0 is the normal diffusion coefficient of a particle which governs the usual diffusive fluxes parallel to density gradients. The odd-diffusive behavior is characterized by the parameter κ which governs fluxes in the direction perpendicular to the density gradients [20]. These fluxes have been called Lorentz fluxes in the context of a Brownian particle diffusing under the effect of Lorentz force. Since these fluxes are divergence free, they do not affect the density distribution of a single particle. However, in the presence of boundaries, the Lorentz fluxes play an important role in the time evolution of density distributions [16].

The surface of a fixed particle, for example, can be regarded as an impenetrable boundary for other particles.

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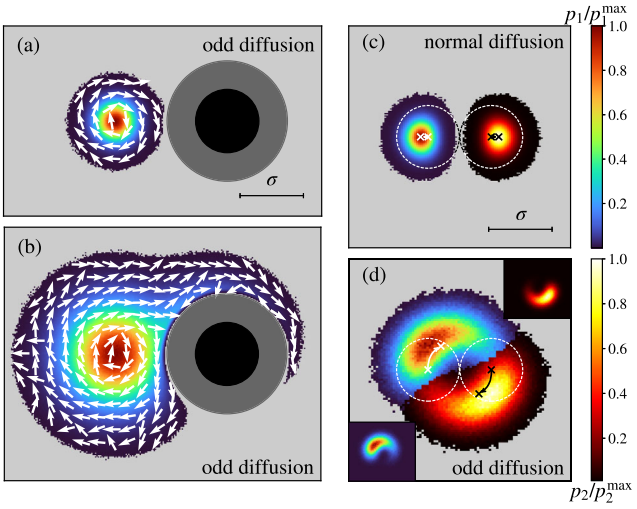

FIG. 1. Scaled density distribution $(p_1/p_1^{\max})$ of an odd-diffusive particle near a fixed particle of same diameter σ at short (a) and long times (b). The curved probability fluxes (arrows) flow around the fixed particle in a preferred direction which can be flipped by reversing the odd-diffusivity parameter κ. Density distribution after a collision between two identical diffusing particles: whereas a collision results in a probability flow away from each other for normal particles (c) for odd-diffusive particles the probability flows around each other (d). This "mutual rolling" of the particles around each other facilitates the motion resulting in an enhanced self-diffusion. The dashed circles represent the initial configuration, and the crosses indicate the displacements of particles' centers. Insets in (d) show individual probability distributions of the two particles. The results are obtained from Brownian dynamics simulations of charged Brownian particles under Lorentz force with $\kappa = 5$.

In Figs. 1(a) and 1(b), we show the probability distribution and fluxes of an odd-diffusive particle near a fixed particle, which are obtained from Brownian dynamics simulations of charged Brownian particles under Lorentz force [31]. At short times, the particle undergoes free diffusion, similar to a normal diffusive particle ($\kappa = 0$); see Fig. 1(a). In contrast, the distribution of the particle at later times is strongly affected by the fixed particle, shown in Fig. 1(b), where the effect of odd diffusivity is strikingly evident. The curved probability fluxes flow around the fixed particle in a preferred direction. This phenomenon has no counterpart in a normal diffusive system. In fact, as we show below, the origin of the enhanced self-diffusion in odd-diffusive systems is fundamentally related to these curved fluxes.

We now consider two diffusing particles. From the perspective of the tracer particle, a host particle represents a moving boundary at which the flux of the tracer must vanish at all times. For particles performing normal diffusion ($\kappa = 0$), a collision is followed by the two particles moving away from each other. It is apparent that collisions hinder the diffusive exploration of space [see Fig. 1(c)]. Odd-diffusive particles, on the contrary, diffuse in a strikingly different way: when two particles collide, rather than blocking each other, they move around each other [see Fig. 1(d)]. In fact, for sufficiently large κ, a collision effectively facilitates the motion of particles around each other, an effect which we refer to as the "mutual rolling effect."

We consider a model system of two distinguishable hard-core Brownian particles of diameter σ in two dimensions. Ignoring hydrodynamic interactions, the FPE corresponding to the probability density of this system is

$$\frac{\partial P(t)}{\partial t} = \nabla_1 \cdot [\mathbf{D}_1 \nabla_1 P(t)] + \nabla_2 \cdot [\mathbf{D}_2 \nabla_2 P(t)], \tag{3}$$

where $P(t) \equiv P(\boldsymbol{r}_1, \boldsymbol{r}_2, t)$. $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$ are the position vectors of particle one and two, respectively. $\mathbf{D}_1$ and $\mathbf{D}_2$ are the corresponding odd-diffusion tensors [Eq. (2)]. Since the particles cannot overlap, the FPE is defined only in the region $\Omega = \mathbb{R}^2 \times \mathbb{R}^2 \backslash \mathcal{B}$ where $\mathcal{B} = \{(\boldsymbol{r}_1, \boldsymbol{r}_2) \in \mathbb{R}^2 \times \mathbb{R}^2; \|\boldsymbol{r}_1 - \boldsymbol{r}_2\| \leq \sigma\}$ is the forbidden area due to an overlap. The hard-core interactions impose a no-flux boundary condition on the moving boundary $\partial\mathcal{B}$

$$\mathbf{D}_1[\nabla_1 P] \cdot \boldsymbol{n}_1 + \mathbf{D}_2[\nabla_2 P] \cdot \boldsymbol{n}_2 = 0, \tag{4}$$

where $\boldsymbol{n}_1$ and $\boldsymbol{n}_2$ are outward unit normal vectors of the two particles such that $\boldsymbol{n}_1 = -\boldsymbol{n}_2$ on $\partial\mathcal{B}$.

Our goal is to obtain an effective description for the marginal densities $p_1 \equiv \int_{\Omega(\boldsymbol{r}_1)} d\boldsymbol{r}_2 P(\boldsymbol{r}_1, \boldsymbol{r}_2, t)$ and $p_2 \equiv \int_{\Omega(\boldsymbol{r}_2)} d\boldsymbol{r}_1 P(\boldsymbol{r}_1, \boldsymbol{r}_2, t)$ for particles one and two, respectively. Here $\Omega(\boldsymbol{r}_i) = \mathbb{R}^2 \backslash B_\sigma(\boldsymbol{r}_i)$, where $B_\sigma(\boldsymbol{r}_i)$ is the disk of radius σ centered at $\boldsymbol{r}_i$, $i \in \{1, 2\}$. In the dilute regime, where only two-body collisions are relevant, we use an asymptotic method adapted from Bruna and Chapman [33–35] which treats the effect of density perturbatively. A crucial step is the inclusion of the zero-flux boundary condition in Eq. (4), which differs from the usual Neumann condition in normal diffusive systems. The detailed calculations are shown in the Supplemental Material [32].

We generalize our description to include an arbitrary number of particles of each species in the low-density limit. The effective FPEs for p_i read as

$$\begin{aligned}\frac{\partial p_i}{\partial t} = \nabla \cdot \mathbf{D}_i [&\nabla p_i + (N_i - 1)\sigma^2 \pi p_i \nabla p_i \\ &+ N_j \pi \sigma^2 (\mathbf{\Lambda}_i p_i \nabla p_j - \mathbf{\Gamma}_i p_j \nabla p_i)],\end{aligned} \tag{5}$$

where N_i and N_j denote the number of particles of species i and j with $(i, j) = (1, 2)$ and (2,1) and

$$\mathbf{\Lambda}_i \equiv \mathbf{1} + \frac{\mathbf{D}_1 + \mathbf{D}_2}{\det(\mathbf{D}_1 + \mathbf{D}_2)} \mathbf{D}_j, \tag{6}$$

$$\mathbf{\Gamma}_i \equiv \frac{\mathbf{D}_1 + \mathbf{D}_2}{\det(\mathbf{D}_1 + \mathbf{D}_2)} \mathbf{D}_i. \tag{7}$$

In our model, the two species can be differentiated via their parameter κ_i and their diffusivity $D_0^{(i)}$. This makes the model well suited for studying diffusion of a tagged particle

in the presence of host particles as well as the collective diffusion of identical particles.

Let us consider that the two species have identical κ and D_0, i.e., $\mathbf{D}_1 = \mathbf{D}_2 = \mathbf{D}$. To obtain the self-diffusion coefficient, we set $N_1 = 1$ (tagged particle), $N_2 = N$, and define $\phi = N\pi\sigma^2 p_2/4$ as the area fraction of host particles in two dimensions. The equation for the tagged particle reduces to

$$\frac{\partial p_1(\boldsymbol{r}, t)}{\partial t} = \nabla \cdot \mathbf{D}[(\mathbf{1} - 4\phi\boldsymbol{\Gamma})\nabla p_1], \tag{8}$$

where $\boldsymbol{\Gamma} = \boldsymbol{\Gamma}_1 = \boldsymbol{\Gamma}_2$ due to the identical diffusion matrices. Note that the probabilistic flux due to the odd part of the self-diffusion tensor $\mathbf{D}(\mathbf{1} - 4\phi\boldsymbol{\Gamma})$ in Eq. (8) is divergence free and hence does not contribute to the mean-squared displacement of the particle. Hence, the self-diffusion coefficient is determined by the symmetric part alone and reads as

$$D_s = D_0\left(1 - 2\phi\frac{1 - 3\kappa^2}{1 + \kappa^2}\right). \tag{9}$$

Equation (9) is the main result of this Letter. It reduces to the well-known expression for normal diffusive systems ($\kappa = 0$) with hard-core interactions $D_s = D_0(1 - 2\phi)$ [6–8]. Our model generalizes this result. It predicts that for $\kappa < \kappa_c = 1/\sqrt{3}$, collisions with the host particles reduce the self-diffusion coefficient with respect to D_0. For $\kappa = \kappa_c$, the host particles become effectively invisible to the tagged particle, which diffuses with $D_s = D_0$. The most interesting prediction of our model is that for $\kappa > \kappa_c$ the self-diffusion coefficient increases with increasing density of host particles [see Fig. 2(a)]. In this regime, instead of hindering, collisions with the host particles facilitate the motion of the tagged particle due to the mutual rolling effect [see Fig. 1(d)]. This suggests that in this regime the mutual rolling effect significantly compensates for the slowing down due to the many-body effects.

In Fig. 2(b), we show the variation of D_s with κ for a fixed density of host particles. Increasing κ results in a crossover from a reduced to an enhanced self-diffusion. Simulations reveal that odd diffusivity enhances the self-diffusion coefficient relative to a normal diffusing system ($\kappa = 0$) at all densities (see Supplemental Material [32]). Furthermore, in the Supplemental Material [32], we numerically show that for soft repulsive potentials our findings remain unaffected.

We now consider the general case in which the tracer particle and the host particles are characterized by different values of the odd-diffusivity parameter κ. Specifically, one can obtain the diffusion coefficient of a tracer particle diffusing in the presence of host particles of different species. For the tracer and the host particles, both with

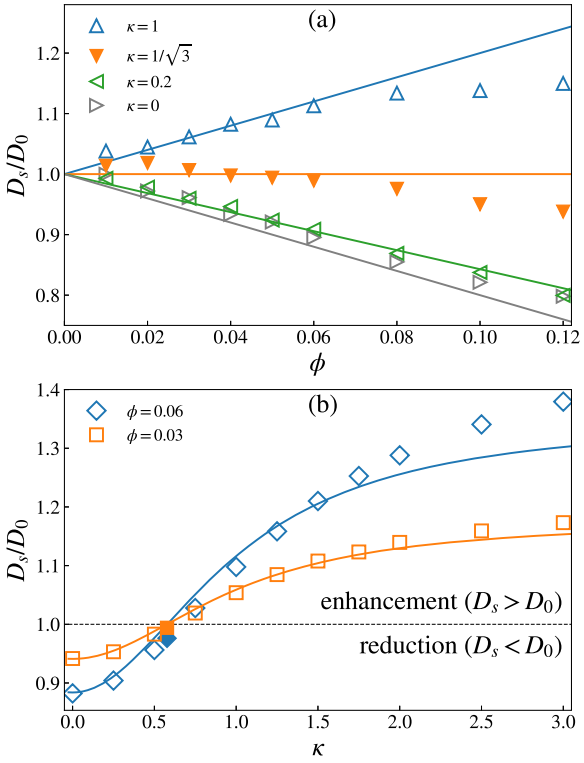

FIG. 2. (a) Reduced self-diffusion coefficient D_s/D_0 as a function of the area fraction ϕ for different values of the odd-diffusivity parameter κ. Symbols represent data from Brownian dynamics simulations of hard-core particles diffusing under Lorentz force. Error bars are smaller than the symbols. Solid lines correspond to the analytical prediction of Eq. (9). For the critical value $\kappa_c = 1/\sqrt{3}$ (filled symbols) particles are effectively invisible to each other. For $\kappa = 1 (> \kappa_c)$ the self-diffusion coefficient increases with increasing ϕ, and for $\kappa = 0.2 (< \kappa_c)$ it decreases with increasing ϕ. (b) κ-governed crossover from a reduced to an enhanced self-diffusion for two different area fractions. Self-diffusion coefficient for $\kappa = \kappa_c$ is shown as filled symbols.

same D_0, the self-diffusion coefficient of the tracer particle reads as

$$D_s = D_0\left(1 - 8\phi\frac{1 - \kappa_1\kappa_2 - 2\kappa_1^2}{(\kappa_1 + \kappa_2)^2 + 4}\right), \tag{10}$$

where κ_1 corresponds to the tagged particle and κ_2 to the host particles. Note that the result of the self-diffusion for identical particles [Eq. (9)] is a special case corresponding to $\kappa_1 = \kappa_2$. Figure 3(a) depicts the scenario of identical odd-diffusive particles ($\kappa_1 = \kappa_2 = \kappa$), for which the self-diffusion coefficient was shown in Fig. 2. Figure 3(b) shows D_s of an odd-diffusive tracer particle ($\kappa_1 = \kappa$) in the presence of normal diffusing host particles ($\kappa_2 = 0$). The self-diffusion coefficient $D_s = D_0\{1 - 8\phi[(1 - 2\kappa^2)/(4 + \kappa^2)]\}$ shows a crossover from reduction to enhancement at $\kappa_c = 1/\sqrt{2}$ due to collisions. For an odd-diffusive tracer particle for both

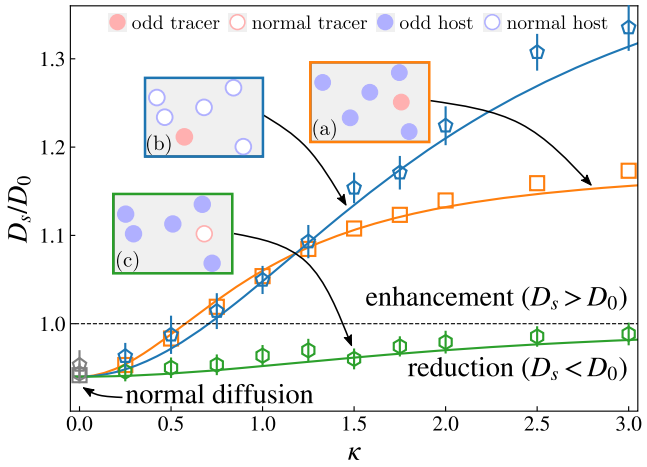

FIG. 3. Self-diffusion of tracer particle (red) in the presence of host particles (blue) of area fraction $\phi = 0.03$. Symbols represent data from Brownian dynamics simulations of hard-core Brownian particles diffusing under Lorentz force. (a) The tagged particle and the host particles are odd diffusive. In this case, there is a crossover at a critical value of $\kappa_c = 1/\sqrt{3}$ from reduction to enhancement of D_s/D_0. (b) The tagged particle is odd diffusive, and the host particles are normal. There is a crossover at $\kappa_c = 1/\sqrt{2}$ again from reduction to enhancement. (c) For a normal tagged particle and odd-diffusive host particles, there is no enhancement of the self-diffusion. With increasing κ, the self-diffusion coefficient asymptotically approaches the ideal diffusivity, $D_s = D_0$. The self-diffusion of normal diffusing particles ($\kappa = 0$) is shown in gray.

Figs. 3(a) and 3(b), a collision with the host particle facilitates its motion giving rise to an enhancement of the diffusion coefficient. Moreover, that Fig. 3(b) overtakes Fig. 3(a) for $\kappa > \sqrt{7/5}$ raises the question whether in this regime the diffusion of an odd particle is most efficient within (normal) obstacles. In contrast to the enhancement, Fig. 3(c) shows the case of a normal diffusing tracer particle ($\kappa_1 = 0$) in the presence of odd-diffusive host particles ($\kappa_2 = \kappa$) for which $D_s = D_0\{1 - [8\phi/(4 + \kappa^2)]\}$ does not show an enhancement of the self-diffusion coefficient. In this case, D_s approaches D_0 asymptotically. Nevertheless, odd diffusivity of the host particles gives rise to a faster diffusion of the tracer in comparison to normal host particles. The theoretical predictions are validated by Brownian dynamics simulations of hard-core interacting particles diffusing under the effect of the Lorentz force [31].

The two scenarios in Figs. 3(a) and 3(b) highlight that odd-diffusive tracer particles can enhance their diffusion coefficient beyond D_0 via collisions with the host particles in agreement with the physical mechanism suggested in Fig. 1. The mutual rolling effect furthermore gives rise to unusual force autocorrelation as we show in the Supplemental Material [32] in the low-density limit. The effect of collisions on the self-diffusion coefficient is quantified by the integral of the force autocorrelation function. While the autocorrelation is a positive monotonically decaying function of time for normal diffusing ($\kappa = 0$) particles [36], it turns negative in time for odd-diffusing particles. In fact for $\kappa > \kappa_c$, the integral of the autocorrelation function is negative implying an enhancement of the self-diffusion coefficient in odd-diffusive systems in agreement with Eq. (9).

We now consider how odd diffusion affects the collective dynamics. A collision helps the tracer particle escape local caging by the host particles resulting in enhanced exploration of space. From the perspective of the whole species, however, nothing has changed; only two particles have interchanged positions due to a collision. This suggests that the collective diffusion coefficient is unaffected by odd diffusivity. By untagging the tracer particle in Eq. (8), the interspecies terms drop out, and we obtain a single-species system, with $N + 1$ identical particles characterized by the collective diffusion coefficient

$$D_c = D_0(1 + 4\phi), \tag{11}$$

which is indeed independent of κ and has the same expression as in normal diffusive systems [34,37]. We note that $D_s > D_c$ for large κ which might have interesting implications for density relaxation in odd-diffusive fluids.

How can these theoretical predictions be tested in experiments? Mesoscopic overdamped systems of colloidal spinners (magnetic dipoles), when exposed to a viscoelastic solvent exhibit a Magnus force which can be tuned by the spinning frequency [38]. With κ of order 1 and above possible, this could be a promising realization for odd-diffusive systems. Another possible realization is millimeter-sized granules, which beget high charges when exposed to a vibrating substrate due to triboelectric effects [39,40]. For a millimeter-sized sphere where inertia can almost be ignored, with the surface charge density $\sigma = 1e$ nm^{-2}, where e is the electronic charge, the viscosity $\eta \approx 10^{-4}$ Pa s (Propylene at room temperature), and $B = 1$ T, one obtains $\kappa \approx 1$. A plausible experimental setup can also be realized in dusty plasma which can be almost overdamped for the high density of the ambient gas. Large magnetic fields exceeding 10^4 T can be effectively realized using noninertial rotating frames [25,26] at which κ of order 1 is in reach for millimeter-sized dust particles. Furthermore the mutual rolling effect should be verifiable in self-spinning granules [41–43], in chiral colloidal microswimmers [27–30,44], even in rotating molecular motors [45,46] and vortex fluids [47,48].

Self-diffusion is affected by both direct and hydrodynamic interactions [49,50]. Whereas hydrodynamic interactions can enhance the self-diffusion coefficient [51,52], direct interactions always reduce the self-diffusion in ordinarily diffusing systems. In this Letter, we ignored the hydrodynamic interactions and showed that in

odd-diffusive systems, interactions can enhance the self-diffusion coefficient.

Our findings might be relevant to other soft matter systems. The enhancement of self-diffusion is reminiscent of Taylor dispersion [53] in which flow along a direction affects diffusion along the orthogonal direction. This is phenomenologically similar to odd-diffusivity induced mutual rolling of two colliding particles. It would be interesting to investigate Taylor dispersion in a dilute suspension of odd-diffusive particles. Our findings might also be applicable to systems with Magnus forces where dragging a probe particle was found to speed up with increasing system density [15,54]. Recently wiggling nanopores have been shown to enhance the diffusion coefficient of a particle [55]. Surprisingly, the enhancement has exactly the same functional form as in Eq. (9). Additional work is needed to investigate whether the similarity extends beyond the mathematical formalism employed in our Letter and fluctuating nanopores.

A. S. and H. L. acknowledge support by the Deutsche Forschungsgemeinschaft (DFG) within the Projects No. SH 1275/3-1 and No. LO 418/25-1. J.-U. S. thanks the cluster of excellence "Physics of Life" at TU Dresden for its support.

*These authors contributed equally to this work.
†Corresponding author.
sharma@ipfdd.de

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and γ is the friction of the particle. T is the temperature in units such that the Boltzmann constant is unity. Note that $\gamma\kappa\boldsymbol{\epsilon}\boldsymbol{v} = q\boldsymbol{v} \times \mathbf{B}$, where q is the charge of the particle and $\mathbf{B} = B\hat{\mathbf{e}}_z$ is the magnetic field. In the small mass limit, the overdamped motion of the particle is characterized by the odd-diffusive tensor $\mathbf{D} = D_0(\mathbf{1} + \kappa\boldsymbol{\epsilon})$, where $D_0 = T/[\gamma(1+\kappa^2)]$. Hard-core interactions are included approximately via a steep, repulsive interaction potential. The Langevin equation is integrated in time with a small mass. Details of simulations are given in the Supplemental Material [32].

[32] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.129.090601 which includes Refs. [33,34] for a detailed description of the calculations and Refs. [8,36] for an outline of an alternative theoretical approach. Furthermore, we describe the simulation method and give additional numerical data for interacting particles and high densities.

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