

Oscillatory Force Autocorrelations in Equilibrium Odd-Diffusive Systems

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The force autocorrelation function (FACF), a concept of fundamental interest in statistical mechanics, encodes the effect of interactions on the dynamics of a tagged particle. In equilibrium, the FACF is believed to decay monotonically in time, which is a signature of slowing down of the dynamics of the tagged particle due to interactions. Here, we analytically show that in odd-diffusive systems, which are characterized by a diffusion tensor with antisymmetric elements, the FACF can become negative and even exhibit temporal oscillations. We also demonstrate that, despite the isotropy, the knowledge of FACF alone is not sufficient to describe the dynamics: the full autocorrelation tensor is required and contains an antisymmetric part. These unusual properties translate into enhanced dynamics of the tagged particle quantified via the self-diffusion coefficient that, remarkably, increases due to particle interactions.

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Introduction.—The self-diffusion coefficient is a transport coefficient, that characterizes the average displacement of a tagged particle in an interacting system. It can be determined from the time integral of the force autocorrelation function (FACF), which is an example of the more general Green-Kubo relations between transport coefficients and correlation functions [1,2]. The effect of interparticle interactions on the self-diffusion is encoded in the FACF, and hence, understanding its dynamical properties is of central interest in many-body statistical mechanics. It has been conjectured that the FACF decays monotonically in overdamped equilibrium systems [3,18–20], independent of the nature of the interaction and for all densities [21–24]. This is consistent with the notion that interparticle interactions typically reduce the self-diffusion [24,25]. Nonmonotonicity in the FACF, and hence a more complex behavior of the self-diffusion, however, emerges when considering underdamped dynamics [26], memory [27], or nonequilibrium systems, as due to activity [20], or driving [28], features that are absent in overdamped equilibrium systems.

Here, we analytically show that contrary to the long-held belief, the FACF can be nonmonotonic and even oscillatory in *overdamped equilibrium systems* [3]. Systems showing this behavior are characterized by probability fluxes, which are perpendicular to concentration gradients and are referred to as odd-diffusive systems [29]. While these systems exhibit fluctuations in accordance with the fluctuation-dissipation theorem [30–32] and their dynamics are overdamped, they are fundamentally distinct from the usual

overdamped systems in that their time evolution is governed by a non-Hermitian operator. We further demonstrate that a nonmonotonic FACF provides a rationale for the atypical trend observed in the self-diffusion coefficient, i.e., it increases with increasing concentration [33].

The transverse response to the perturbation is the fundamental property of *odd* systems, which have received much interest lately [34]. In addition to odd-diffusive systems, there are odd systems characterized by odd viscosity [35–40], odd elasticity [41,42], and odd viscoelasticity [43,44]. With the advent of experimental odd systems such as spinning biological organisms [45], chiral fluids [46,47], and colloidal spinners [48], the interest in odd systems has increased rapidly.

The odd-diffusion tensor for a two-dimensional isotropic system can be written as

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

where $\mathbf{1}$ is the identity matrix, $\boldsymbol{\epsilon}$ is the antisymmetric Levi-Civita symbol in two dimensions ($\epsilon_{xy} = -\epsilon_{yx} = 1$ and $\epsilon_{xx} = \epsilon_{yy} = 0$), D_0 is the diffusivity, and κ is the odd-diffusion parameter. A nonzero κ results in probability fluxes perpendicular to concentration gradients. Examples of odd-diffusive systems are Brownian particles diffusing under the effect of Lorentz force [49–54], and diffusing skyrmions [55–60]; see also the Supplemental Material (SM) [4]. Although these are equilibrium odd-diffusive systems [30], there exist also driven odd-diffusive systems such as active chiral particles (also called circle

swimmers) [61–64] and strongly damped particles subjected to Magnus [65] or Coriolis force [66]. In contrast to equilibrium systems, which are invariant under time reversal, the odd-diffusive behavior in nonequilibrium systems is a consequence of broken time-reversal and parity symmetries [40].

While an exact calculation of the FACF is a formidable task, near-exact analytical results can be obtained in the dilute limit in which the dynamics are dominated by two-body effects. To this end, we generalize the first-principles approach developed by Hanna, Hess, and Klein [21,22] to calculate the FACF in a dilute odd-diffusive system of hard-core interacting particles. We show analytically that odd diffusion qualitatively alters the time correlations: the correlation function becomes negative for finite κ indicating the anticorrelated nature of the force experienced by an odd-diffusive particle due to collisions with other particles. Moreover, the correlation function exhibits temporal oscillations for certain values of κ ; specifically, it crosses zero twice. We further show that for sufficiently large κ , the integral of the correlation function becomes negative, which gives rise to the increase in the self-diffusion coefficient. Using the Green-Kubo relation, we derive exactly the same expression for the self-diffusion coefficient as in Ref. [33], which was obtained using an alternative approach.

Theoretical background.—We consider a two-dimensional system of two interacting, odd-diffusive hard disks with coordinates $\vec{\mathbf{x}} = (\mathbf{x}_1, \mathbf{x}_2)$. The two-particle conditional probability density function for the particles to evolve from $\vec{\mathbf{x}}'$ at time $t' \leq t$ to $\vec{\mathbf{x}}$ at time t , $P = P(\vec{\mathbf{x}}, t | \vec{\mathbf{x}}', t')$, satisfies the Fokker-Planck equation

$$\begin{aligned} \frac{\partial}{\partial t} P &= \nabla_1 \cdot \mathbf{D}[\nabla_1 + \beta \nabla_1 U(r)] P \\ &+ \nabla_2 \cdot \mathbf{D}[\nabla_2 + \beta \nabla_2 U(r)] P, \end{aligned} \quad (2)$$

with the odd-diffusion tensor (1) and ∇_1, ∇_2 as the partial differential operator with respect to the coordinates of particle one and two, respectively. $U(r)$ is the potential energy with $r = |\mathbf{x}_1 - \mathbf{x}_2|$ as the relative distance between the particles and $\beta = 1/k_B T$, where k_B is the Boltzmann constant and T is the temperature. We assume hard-core interactions between the two disks of diameter σ , which can be written as $U(r) = \begin{cases} \infty, & r \leq \sigma \\ 0, & r > \sigma \end{cases}$. The analytical solution to the two-particle Fokker-Planck equation was obtained for normal-diffusing particles, i.e., $\mathbf{D} = D_0 \mathbf{1}$ [21,22]. Note that for $\kappa \neq 0$, the time-evolution operator is non-Hermitian. While the hard-core interactions are modeled via Neumann boundary conditions in normal-diffusing systems, they are modeled as oblique boundary conditions in odd-diffusive systems due to the transverse fluxes [33,67]. This has profound consequences for the solution and therefore for the application of our theory.

We solve the two-particle problem (2) for odd-diffusive hard disks exactly in the SM [4].

Force autocorrelation tensor.—The force autocorrelation tensor (FACT), which is defined as $\mathbf{C}_F(\tau) = \langle \mathbf{F}(\tau) \otimes \mathbf{F}(0) \rangle$, can be written as [25]

$$\begin{aligned} \mathbf{C}_F(\tau) &= \int d\vec{\mathbf{x}} \int d\vec{\mathbf{x}}_0 \mathbf{F}(\vec{\mathbf{x}}) \otimes \mathbf{F}(\vec{\mathbf{x}}_0) \\ &\times P(\vec{\mathbf{x}}, \tau | \vec{\mathbf{x}}_0, 0) P_{\text{eq}}(\vec{\mathbf{x}}_0), \end{aligned} \quad (3)$$

for $\tau > 0$. Here, \mathbf{F} is the interaction force acting on a tagged particle due to other particles, $\langle \cdot \rangle$ denotes an ensemble average with the equilibrium distribution $P_{\text{eq}}(\vec{\mathbf{x}}_0)$, and the outer product is defined as $[\mathbf{A} \otimes \mathbf{B}]_{\alpha\beta} = A_\alpha B_\beta$. Throughout this Letter, time is measured in units of $\tau_0 = \sigma^2 / (2D_0)$, which is the characteristic timescale of a particle diffusing over a distance of diameter σ , i.e., $\tau = t / \tau_0$. The FACT can be calculated from Eq. (3) to first order in the concentration, details of which are shown in SM [4]. Similar to the diffusion tensor, the FACT can be split in a diagonal and an antisymmetric off-diagonal part:

$$\mathbf{C}_F(\tau) = C_F^{\text{diag}}(\tau) \mathbf{1} + C_F^{\text{off}}(\tau) \mathbf{e}, \quad (4)$$

for $\tau > 0$, where $C_F^{\text{diag}}(\tau)$ and $C_F^{\text{off}}(\tau)$ are the diagonal and antisymmetric off-diagonal elements of the FACT. In Laplace domain they read

$$\tilde{C}_F^{\text{diag}}(s) = \frac{2\phi}{\beta^2 D_0} \frac{K_1[\sqrt{s}K_0 + K_1]}{[\sqrt{s}K_0 + K_1]^2 + [\kappa K_1]^2}, \quad (5)$$

$$\tilde{C}_F^{\text{off}}(s) = \frac{2\phi}{\beta^2 D_0} \frac{\kappa [K_1]^2}{[\sqrt{s}K_0 + K_1]^2 + [\kappa K_1]^2}, \quad (6)$$

where $K_n = K_n(\sqrt{s})$ is the modified Bessel function of the second kind of order n , $\phi = \pi(N/V)(\sigma/2)^2$ is the area fraction for N particles of diameter σ in an area V , and $(\tilde{\cdot})$ denotes the Laplace transform with s as the Laplace variable conjugate to τ . Note that the off-diagonal elements C_F^{off} are proportional to the odd-diffusion parameter κ and therefore vanish in the case of normal diffusion ($\kappa = 0$). In this case the FACT reduces to $\mathbf{C}_F(\tau) = C_F^{\text{diag}}(\tau) \mathbf{1} = \frac{1}{2} \langle \mathbf{F}(\tau) \cdot \mathbf{F}(0) \rangle \mathbf{1}$, which is the usual FACF in normal systems.

The diagonal and off-diagonal elements of the FACT are plotted in Fig. 1 as a function of time. We first consider the behavior of the diagonal elements of the tensor in Fig. 1(a), which correspond to the usual FACF for odd-diffusive systems. For small values of κ , the FACF is a positive, monotonically decaying function of time, qualitatively similar to a normal diffusive system. For larger values of κ , however, a new feature appears in the FACF: it crosses through zero and hence becomes negative, indicating an anticorrelation of the force. The timescale of the force reversal on a tracer particle, i.e., when the FACF becomes

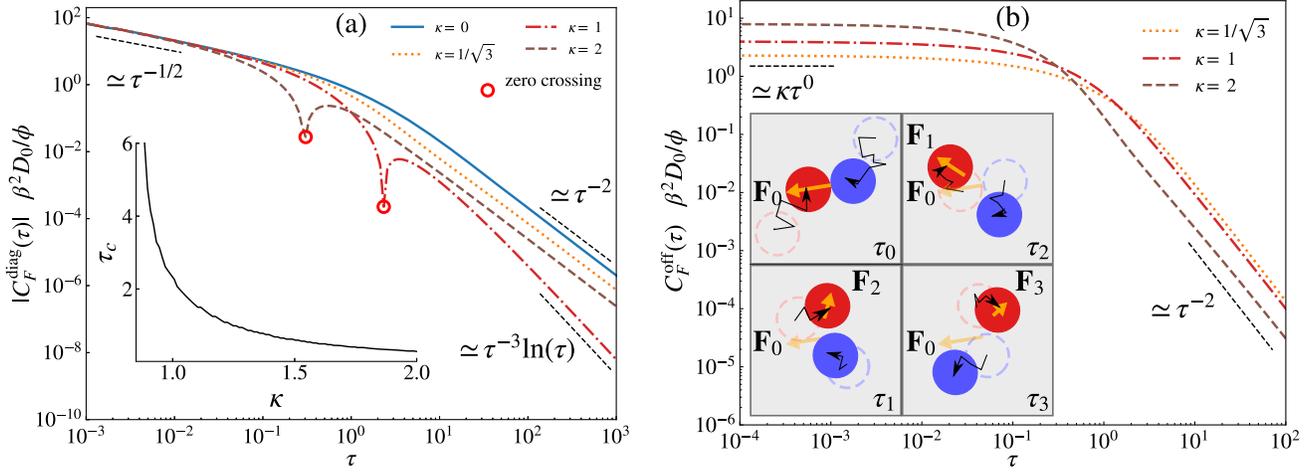


FIG. 1. Double-logarithmic plot of the diagonal and off-diagonal elements of the force autocorrelation tensor (FACT) of interacting hard disks as a function of reduced time $\tau = t/\tau_0$, where $\tau_0 = \sigma^2/(2D_0)$. (a) The diagonal elements of the FACT $C_F^{\text{diag}}(\tau)$, corresponding to the force autocorrelation function (FACF), can turn negative. The FACF diverges in the limit $\tau \rightarrow 0$ as $C_F^{\text{diag}}(\tau) \simeq \tau^{-1/2}$. At long times the FACF scales as $C_F^{\text{diag}}(\tau) \simeq \tau^{-2}$. For $\kappa = 1$ we find an exceptional long-time behavior, where $C_F^{\text{diag}}(\tau) \simeq \tau^{-3} \ln(\tau)$. The inset shows the zero-crossing time τ_c of $C_F^{\text{diag}}(\tau)$ as a function of κ , which in the main figure is marked by red circles. The onset of the anticorrelation corresponds to $\kappa > \kappa_{\text{th}} \approx 0.88$. (b) The off-diagonal elements of the FACT $C_F^{\text{off}}(\tau)$ are independent of time in the short-time limit $C_F^{\text{off}}(\tau) \simeq \kappa \tau^0$ and are directly proportional to κ . In the long-time limit, they scale similarly to the diagonal elements as $C_F^{\text{off}}(\tau) \simeq \tau^{-2}$ for all κ . The inset in (b) shows typical configurations after a collision of particles, where the orientational change of the force (orange arrow) $\mathbf{F}_i = \mathbf{F}(\tau_i)$, $i \in \{0, 1, 2, 3\}$ of the tagged particle (red) is indicated. The black arrows thereby indicate a possible trajectory from one frame to the next.

negative, depends strongly on κ , as can be seen in the inset of Fig. 1(a). There exists a numerically obtained threshold $\kappa_{\text{th}} \approx 0.88$ below which the FACF is strictly positive. The off-diagonal elements of the FACT are shown in Fig. 1(b). Unlike the diagonal elements, which diverge as $t \rightarrow 0$, the off-diagonal elements remain finite. Specifically they remain positive for all κ and decay monotonically in time.

It is interesting to investigate the short- and long-time behavior of the elements of the FACT. Using the asymptotic behavior of the modified Bessel functions K_0 and K_1 , see SM [4] for details, from Eqs. (5) and (6) we have analytical access to the behavior on timescales $t \ll \tau_0$ and $t \gg \tau_0$, i.e., $s \gg 1$ and $s \ll 1$ in the Laplace domain, respectively. At short times, the FACF behaves like $C_F^{\text{diag}}(\tau) \simeq \tau^{-1/2}$, as shown in Fig. 1(a), and is independent of κ . Here, \simeq is used to denote asymptotic proportionality. The long-time behavior of the FACF can be obtained from the $s \ll 1$ expansion and behaves asymptotically as

$$\tilde{C}_F^{\text{diag}}(s) \sim \frac{2\phi}{\beta^2 D_0} \frac{1}{1+\kappa^2} \left(1 + \frac{1-\kappa^2}{1+\kappa^2} \left(\gamma - \ln(2) + \frac{\ln(s)}{2} \right) s + \frac{1-6\kappa^2+\kappa^4}{8(\kappa^2+1)^3} s^2 \ln^2(s) \right), \quad (7)$$

for $s \rightarrow 0$ and where $\gamma = 0.5772$ is the Euler-Mascheroni constant. For $\kappa = 0$, the asymptotic behavior of $\tilde{C}_F^{\text{diag}}$ coincides with the form reported for related 2D Lorentz

gas systems [68]. Furthermore, from Eq. (7) it can be seen that the long-time behavior of $C_F^{\text{diag}}(\tau)$ strongly depends on κ . The FACF decays as τ^{-2} for all κ except for $\kappa = 1$, at which the leading order contribution vanishes in Eq. (7) and $C_F^{\text{diag}}(\tau) \simeq \tau^{-3} \ln(\tau)$, as shown in Fig. 1(a) [69,70]. The ordinary algebraic long-time decay $\simeq \tau^{-2}$ ($\kappa \neq 1$) is consistent with the general prediction of a decay $\simeq \tau^{-(d/2+1)}$, $d = 1, 2, 3$, for correlation functions in systems, which do not conserve momentum [71,72]. This universal behavior was theoretically and numerically exhaustively demonstrated specifically for the 2D Lorentz gas model [68,73–76]. In three dimensions, the decay of the correlation functions $\simeq \tau^{-5/2}$ [21,77–80] could recently be demonstrated computationally [24]. In contrast, the short-time behavior $\simeq \tau^{-1/2}$ is independent of dimensionality and attributed to the hard interactions between the particles [21,78].

The asymptotic short-time behavior of $C_F^{\text{off}}(\tau)$ turns out to be independent of time but depends linearly on κ , $C_F^{\text{off}}(\tau) \simeq \kappa \tau^0$, as can be seen in Fig. 1(b). Such a scaling of the off-diagonal elements with κ at short times has been recently derived by Yasuda *et al.* in Ref. [81] for odd Langevin systems. The authors also pointed out that this could be useful for estimating the odd-diffusion parameter in experiments. The asymptotic long-time behavior of $C_F^{\text{off}}(\tau)$ shows a monotonic decay in time and also depends on κ , $C_F^{\text{off}}(\tau) \simeq \kappa \tau^{-2}/(\kappa^2 + 1)^2$, as can be seen in Fig. 1(b).

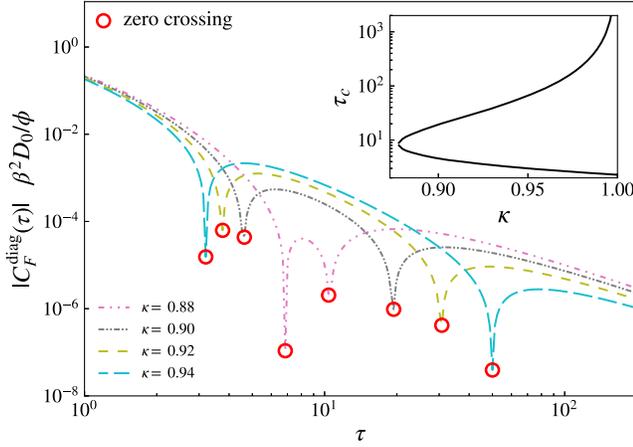


FIG. 2. Double-logarithmic plot of the absolute value of the diagonal elements of the force autocorrelation tensor $C_F^{\text{diag}}(\tau)$ of interacting hard disks as a function of reduced time $\tau = t/\tau_0$, where $\tau_0 = \sigma^2/(2D_0)$. Investigating the regime $\kappa \in [0.88, 1.0]$, we find oscillatory behavior of $C_F^{\text{diag}}(\tau)$. At short times $C_F^{\text{diag}}(\tau)$ starts as a positive function, turns negative, and after a second zero crossing becomes positive again. The inset shows the zero-crossing times τ_c of $C_F^{\text{diag}}(\tau)$ as a function of κ in a linear-logarithmic plot, which in the main figure are marked as red circles. The oscillatory behavior starts at $\kappa \geq \kappa_{\text{th}} = 0.88$, whereas the second zero crossing drifts to infinity as $\kappa \rightarrow 1$. For $\kappa > 1$, $C_F^{\text{diag}}(\tau)$ only shows one zero crossing and remains anticorrelated for the remaining $\tau \rightarrow \infty$ [see also inset in Fig. 1(a)].

In the low-concentration system studied here, only two-body correlations are important. Despite this, the FACF can turn negative as shown in Fig. 1. Furthermore there even exists a range of $\kappa \in (\kappa_{\text{th}}, 1)$ for which the FACF exhibits not one but two zero crossings, as shown in Fig. 2. It appears that for κ slightly larger than $\kappa_{\text{th}} \approx 0.88$, which is obtained from numerical inversion of Eq. (5), the FACF first becomes anticorrelated (first zero crossing) in time before it crosses the time axis again (second and last zero crossing). Here, at long times, the FACF decays to zero from above. We have numerically inverted the Laplace transform over much longer times than shown here and did not find more than two zero crossings. This “temporal oscillation” in the FACF ceases to exist for $\kappa \geq 1$. For $\kappa > 1$, the asymptotic expansion in Eq. (7), transformed back into time domain, is strictly negative and therefore the FACF decays to zero from below, i.e., the second zero crossing vanishes (see also inset in Fig. 2).

Green-Kubo relation for the self-diffusion coefficient.— The self-diffusion coefficient D_s can be obtained from the velocity autocorrelation function (VACF) $C_v(\tau) = \langle \mathbf{v}(\tau) \cdot \mathbf{v}(0) \rangle / 2$, where $\mathbf{v}(\tau) = d\mathbf{x}/d\tau$ and \mathbf{x} is the position of the tagged particle as the time integral

$$D_s = \int_0^\infty d\tau C_v(\tau), \quad (8)$$

a Green-Kubo relation between an equilibrium autocorrelation function ($C_v(\tau)$) and a transport coefficient (D_s) [1].

In normal diffusive systems, the VACF is related to the FACF. In contrast, in an odd-diffusive system, the knowledge of the FACF alone is not sufficient to calculate the VACF. This is despite the fact that the system is isotropic. In fact, one requires the entire FACT to calculate the VACF. We show in SM [4] that in odd-diffusive systems, the VACF can be written as

$$C_v(\tau) = D_0(\delta_+(\tau) - D_0\beta^2 C_F(\tau)), \quad (9)$$

where

$$C_F(\tau) = \frac{1}{2} \frac{1}{D_0^2} (\mathbf{D}^2)^T : \mathbf{C}_F(\tau), \quad (10)$$

and where the double contraction is defined as $\mathbf{A} : \mathbf{B} = \sum_{\alpha, \beta=1}^2 A_{\alpha\beta} B_{\beta\alpha}$. $\delta_+(\cdot)$ is the one-sided delta distribution; see also SM [4]. We refer to $C_F(\tau)$ as the “generalized” force autocorrelation function (gFACF), which reads

$$C_F(\tau) = (1 - \kappa^2) C_F^{\text{diag}}(\tau) - 2\kappa C_F^{\text{off}}(\tau). \quad (11)$$

For normal diffusive systems (i.e., $\kappa = 0$), C_F reduces to the ordinary FACF. Note that even though the gFACF is diverging for all $\kappa \neq 1$ in $\tau \rightarrow 0$ in the hard-disk system, the function remains integrable. This is of physical significance since the integral of the gFACF captures the effect of collisions on the self-diffusion as we see from the Green-Kubo relation Eq. (8) together with Eq. (9).

The self-diffusion coefficient D_s can be obtained from the time integral of Eq. (9) or by using the limit theorem $\int_0^\infty f(t) dt = \lim_{s \rightarrow 0} \tilde{f}(s)$ in Eq. (7) for $\tilde{C}_F^{\text{diag}}$ and similarly for \tilde{C}_F^{off} , which yields

$$\lim_{s \rightarrow 0} \tilde{C}_F^{\text{diag}}(s) = \frac{1}{\kappa} \lim_{s \rightarrow 0} \tilde{C}_F^{\text{off}}(s) = \frac{2\phi}{\beta^2 D_0} \frac{1}{1 + \kappa^2}. \quad (12)$$

Together with Eqs. (9) and (11), this gives the self-diffusion coefficient in an odd-diffusive system,

$$D_s = D_0 \left(1 - 2\phi \frac{1 - 3\kappa^2}{1 + \kappa^2} \right), \quad (13)$$

valid up to first order in area concentration ϕ for a system of hard disks. This result was previously derived by us in Refs. [33,82] by a different method.

For $\kappa = 0$ the expression for D_s reproduces the known result of normal diffusive systems of hard disks in two dimensions $D_s = D_0(1 - 2\phi)$ [22,78]. The surprising result of D_s in Eq. (13) is that the prefactor of ϕ can change sign. This shows that odd diffusivity ($\kappa > 0$) results in a cancellation of the ordinary collision-induced reduction

of the self-diffusion. For $\kappa = \kappa_c = 1/\sqrt{3}$, up to first order in the area fraction, the effect of the collisions on the self-diffusion vanishes ($D_s = D_0$), meaning that on long time and length scales hard disks appear to diffuse as non-interacting particles. For $\kappa > \kappa_c$, collisions surprisingly increase the self-diffusion coefficient: the system mixes more efficiently.

It is natural to ask whether our findings can be extended to three dimensions. However, in three dimensions, odd systems cannot be isotropic because the plane in which the rotation takes place breaks isotropy [29,34,83]. We investigated the self-diffusion in such a system via Brownian dynamics simulations and found that the in-plane odd diffusivity has no effect on the diffusion along the axes of rotation, which turns out to be exactly the same as that of a normal-diffusive system of hard spheres. The in-plane diffusivity, however, shows the same κ -dependent behavior as in a two-dimensional odd-diffusive system.

Discussion.—We analytically demonstrated that equilibrium correlation functions can be nonmonotonic and even oscillatory in overdamped systems. This finding is at odds with the statement that in an equilibrium system the correlation function and all its derivatives decay monotonically [19,20]. While the latter holds in systems where the time evolution is described by a Hermitian Fokker-Planck operator [3], for odd systems this is not applicable due to their intrinsic antisymmetric off-diagonal elements in the diffusion tensor (1).

Our work shows that rich physics is to be explored in equilibrium, odd-diffusive systems. In normal-diffusive systems, for instance, there exists a crossover between two diffusive regimes: short-time diffusion with diffusivity D_0 and long-time diffusion with $D_s < D_0$ [25]. That the long-time self-diffusion coefficient is smaller than the short-time is indicative of the slowing down of the dynamics of the tracer particle in the crossover. In odd-diffusive systems, in contrast, the dynamics can be enhanced, which is reflected in the anticorrelated force autocorrelations. The anticorrelation can be physically interpreted in terms of reversal of the force experienced by a tagged particle such that rather than impeding, collisions with other odd-diffusive particles enhance the motion of the tagged particle; see also the inset in Fig. 1(b). Even though qualitatively this mutual rolling of particles explains the enhancement of self-diffusion with collisions in an odd-diffusive system through the reversal of force [33], a detailed mechanism is still elusive. To this end, we believe it will be interesting to investigate the structural rearrangements that occur in an odd-diffusive system and contrast them with those in a normal-diffusive system. We further expect that the unusual behavior could also have implications for the rheological properties of odd fluids, such as viscosity.

With increasing experimental interest in systems such as spinning biological organisms [45], chiral fluids [46,47],

and colloidal spinners [48], our work will contribute to the broadening interest of the physics community in these systems, especially in the novel and interesting way interactions modify the particle dynamics here. Lastly, since exact analytical results are rather rare in interacting systems, our work may serve as a reference to validate approximate theories for dense systems or computer simulations.

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 - [4] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevLett.132.057102>, in which we provide detailed information about exemplarily odd-diffusive equilibrium systems in Secs. I and II, detailed calculations in Secs. III–VI and debate in Sec. VII whether existing theorems on autocorrelation functions can be applied. The two-particle Smoluchowski equation for interacting odd-diffusive hard disks in two dimensions is solved in Sec. IV, and we establish the connection from the velocity to the force autocorrelation function and explicitly solve for the elements of the force autocorrelation tensor in the Laplace domain in Secs. V and VI, respectively. The Supplemental Material includes Refs. [5–17], which are not in the Letter.
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