Dance of odd-diffusive particles: A Fourier approach

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Odd-diffusive systems are characterized by transverse responses and exhibit unconventional behaviors in interacting systems. To address the dynamical interparticle rearrangements in a minimal system, we here exactly solve the problem of two hard disklike interacting odd-diffusing particles. We calculate the probability density function (PDF) of the interacting particles in the Fourier-Laplace domain and find that oddness rotates all modes except the zeroth, resembling a mutual rolling of interacting odd particles. We show that only the first Fourier mode of the PDF, the polarization, enters the calculation of the force autocorrelation function (FACF) for generic systems with central-force interactions. An analysis of the polarization as a function of time reveals that the relative rotation angle between interacting particles overshoots before relaxation, thereby rationalizing the recently observed oscillating FACF in odd-diffusive systems [Kalz *et al.*, Phys. Rev. Lett. **132**, 057102 (2024)].

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I. INTRODUCTION

The description of dissipative systems with an inherent broken time-reversal or parity symmetry has recently attracted considerable attention [1–5]. Relevant systems can be found in various domains of statistical physics and include Brownian particles under the effect of Lorentz force [6], skyrmionic spin structures [7], and active chiral particles [1]. The transport coefficients of these systems show a characteristic transverse response to perturbations, which is encoded in antisymmetric off-diagonal elements in transport tensors. These characteristic elements behave odd under the transformation of the underlying (broken) symmetry, which serves as the namesake for these *odd* systems. Interestingly, a transverse response does not necessarily imply an anisotropic description of the system. In fact, in two spatial dimensions, odd transport coefficients represent the most general description of an isotropic physical system [8]. In this paper, we specifically consider odd-diffusive systems [1,2], which in two spatial dimensions are characterized by a diffusion tensor of the form

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

where D_0 is the bare diffusivity with physical dimensions $[D_0] = \mathrm{m}^2/\mathrm{s}$, and κ is the characteristic odd-diffusion parameter with $[\kappa] = 1$, encoding the transverse response. $\mathbf{1}$ is the identity tensor and $\boldsymbol{\epsilon}$ the fully antisymmetric Levi-Civita symbol in two dimensions ($\epsilon_{xy} = -\epsilon_{yx} = 1$ and $\epsilon_{xx} = \epsilon_{yy} = 0$).

While the oddness parameter enters explicitly in the diffusion tensor in Eq. (1), the mean-squared displacement of a freely diffusing particle is determined only by the symmetric part of the diffusion tensor. However, in a system of interacting particles, oddness qualitatively alters the diffusive behavior by affecting the self-diffusion [2,9]. The self-diffusion measures the dynamic of a tagged particle in a crowded system and explicitly accounts for interactions of particles [10]. Independent of the microscopic details of the interparticle interactions, the self-diffusion is usually reduced [11–21]. In odd-diffusive systems, however, it was recently shown that even purely repulsive interactions can enhance the self-diffusion [2]. Via expressing the self-diffusion as a time-integral of the (interaction) force autocorrelation function (FACF) [22], these findings could further be related to the unusual microscopic particle rearrangements in odd-diffusive systems [9].

For the overdamped equilibrium system under consideration here and consistent with the reduction of the self-diffusion [22], autocorrelation functions, in general, decay monotonically in time [23]. However, the enhancement of self-diffusion for odd systems can occur only if the FACF switches signs, i.e., becomes nonmonotonic in time. This apparent contradiction could be resolved by recognizing that the time-evolution operator in an odd-diffusive system becomes non-Hermitian when $\kappa \neq 0$ in Eq. (1), thereby breaking the monotonicity requirements on the FACF [9]. Surprisingly, in this work, it was further observed that the FACF even oscillates in time. While this is consistent with the observed enhancement of the self-diffusion in odd systems, a physical interpretation of this phenomenon remains elusive.

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In this paper, we substantially extend our previous work [9] and show how the nonmonotonic FACF originates in the unusual dynamics of interacting odd particles. When a pair of odd-diffusive particles interacts, their motion resembles that of two mutually rotating particles [2]. This is despite the fact that the interaction potential is central force, i.e., it acts along the vector that connects the centers of two particles. Our approach is based on the exact analytical derivation of the propagator of two interacting odd-diffusive particles. The joint probability density function (PDF) for the two particles separates into a center-of-mass PDF and a PDF of the relative coordinate, capturing the interplay of odd diffusion and interactions. We can express the relative PDF in a Fourier series to find that only certain modes enter the averages of observables, such as in the FACF. In particular, it is the polarization mode of the relative PDF, representing the positioning of the particles with respect to each other in time, which determines the full tensorial force autocorrelation behavior. By analyzing the polarization mode in time, we understand what originates the oscillating FACF. Interacting odd-diffusive particles mutually rotate further in time before they eventually relax. Figuratively, they perform a dance, reminiscent of the classical step of the Viennese waltz.

The remainder of this paper is organized as follows: In Sec. II, we set the problem of two interacting odd-diffusive particles, which is then exactly solved in Sec. II A. The propagator can be put into the form of a Fourier series, of which the numerical analysis of the modes is presented in Sec. II B. In Sec. II C, we show that only certain Fourier modes enter in averages of observables, in particular, into the FACF. Finally, in Sec. III we summarize and give an extensive overview of systems, which can be subsumed under the terminology of odd diffusion. In Appendix A, we present the detailed solution of the problem of interacting particles, in Appendix B we give details about the asymptotic behavior, and Appendix C lists the relevant integral relations.

II. INTERACTING ODD-DIFFUSIVE PARTICLES

Following the setup in Ref. [9], we study the dynamics of two odd-diffusive particles at positions $\mathbf{x}_1$ and $\mathbf{x}_2$ in two spatial dimensions. The particles are assumed to interact with the potential energy U, which we assume to be of the hard-disk type

$$U(\mathbf{x}_1, \mathbf{x}_2) = U(r) = \begin{cases} \infty, & r \leqslant 1 \\ 0, & r > 1, \end{cases} \tag{2}$$

where $r = |\mathbf{x}_1 - \mathbf{x}_2|/d$ is the rescaled relative distance between the particles and d is the particle diameter.

The conditional joint PDF, i.e., the propagator for the particles to be found at positions $(\mathbf{x}_1, \mathbf{x}_2)$ at time t given that they were at positions $(\mathbf{x}_{1,0}, \mathbf{x}_{2,0})$ at time t_0, $P(t) = P(\mathbf{x}_1, \mathbf{x}_2, t|\mathbf{x}_{1,0}, \mathbf{x}_{2,0}, t_0)$, evolves according to the time-evolution equation

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

where the initial condition is given as $P(t = t_0) = \delta(\mathbf{x}_1 - \mathbf{x}_{1,0})\,\delta(\mathbf{x}_2 - \mathbf{x}_{2,0})$. In Eq. (3), ∇_1, ∇_2 are the partial differential operators for the positions of the particles, and $\mathbf{D}$ is the odd-diffusion tensor of Eq. (1). $\boldsymbol{\mu}$ is the mobility tensor, and we assume the fluctuation-dissipation relation (FDR) to hold

$$\mathbf{D} = k_B T \boldsymbol{\mu}, \tag{4}$$

where k_B is the Boltzmann constant and T the temperature of the solvent. Note that even though Eq. (3) looks formally equivalent to a Fokker-Planck equation for the joint PDF $P(t)$, strictly spoken it is not, due to the antisymmetric (odd-diffusive) elements in the diffusion tensor [24]. However, based on the assumption of the FDR, Eq. (3) resembles equilibrium dynamics with a unique steady-state solution [9,25].

A. Analytical solution

We rescale space with the diameter of the particles d, $\mathbf{x}_i \to \mathbf{x}_i/d$, and time by the natural timescale of diffusing the radial distance of a particle diameter $\tau_d = d^2/(2D_0)$, $t \to \tau = t/\tau_d$. Given the radial symmetry of the interaction potential $U(r)$, the time-evolution equation (3) can be written in terms of a center-of-mass coordinate $\mathbf{x}_c = (\mathbf{x}_1 + \mathbf{x}_2)/2$ and a relative coordinate $\mathbf{x} = \mathbf{x}_1 - \mathbf{x}_2$ as

$$\frac{\partial}{\partial \tau}P(\tau) = \frac{1}{4}\nabla^2_{\mathbf{x}_c} P(\tau) + \nabla_{\mathbf{x}} \cdot (\mathbf{1} + \kappa\boldsymbol{\epsilon})[\nabla_{\mathbf{x}} + \beta\, \nabla_{\mathbf{x}} U(r)]\, P(\tau), \tag{5}$$

where $\beta = 1/k_B T$ and $\nabla_{\mathbf{x}_c}, \nabla_{\mathbf{x}}$ are the partial differential operators corresponding to the center-of-mass and relative coordinates. Note that $(\mathbf{1} + \kappa\boldsymbol{\epsilon}) = \mathbf{D}/D_0$ represents the dimensionless odd-diffusion tensor of Eq. (1). As Eq. (5) decouples the coordinates, the propagator can be written as $P(\mathbf{x}_1, \mathbf{x}_2, \tau|\mathbf{x}_{2,0}, \mathbf{x}_{1,0}, \tau_0) = p_c(\mathbf{x}_c, \tau|\mathbf{x}_{c,0}, \tau_0)\, p(\mathbf{x}, \tau|\mathbf{x}_0, \tau_0)$ and the center-of-mass problem can be solved straight forwardly in the form

$$p_c(\mathbf{x}_c, \tau|\mathbf{x}_{c,0}) = \frac{1}{\pi\tau} \exp\left(-\frac{|\mathbf{x}_c - \mathbf{x}_{c,0}|^2}{\tau}\right), \tag{6}$$

where we have set $\tau_0 = 0$ as the underlying stochastic process is time-translational invariant [26].

The relative PDF can be put into the form of a (Cartesian) multipole expansion [27–31], which in polar coordinates $\mathbf{x} = (r, \varphi)$, $\mathbf{x}_0 = (r_0, \varphi_0)$, is given as

$$p(\mathbf{x}, \tau|\mathbf{x}_0) = \Theta(r-1)\big[\varrho(r, \tau|r_0) + \boldsymbol{\sigma}(r, \tau|r_0) \cdot \mathbf{e}(\Delta\varphi) + \boldsymbol{Q}(r, \tau|r_0) : \big(\mathbf{e}(\Delta\varphi) \otimes \mathbf{e}(\Delta\varphi) - \tfrac{1}{2}\big) + \dots\big], \tag{7}$$

where $\mathbf{e}(\Delta\varphi) = (\cos(\Delta\varphi), \sin(\Delta\varphi))^{\mathrm{T}}$, $\Delta\varphi = \varphi - \varphi_0$ is the angular difference. $\mathbf{Q} : (\mathbf{e} \otimes \mathbf{e} - \mathbf{1}/2) = \sum_{\alpha,\beta=1}^{2} Q_{\alpha\beta}(e_\beta e_\alpha - \delta_{\beta\alpha}/2)$ denotes the full contraction, where $\mathbf{e} \otimes \mathbf{e}$ is the outer product. We again set $\tau_0 = 0$ here. Note the multiplicative Heaviside function $\Theta(r-1)$ in Eq. (7), which is defined as $\Theta(x) = 1$, if $x > 1$ and $\Theta(x) = 0$ otherwise. This ensures the no-overlap condition of the hard-disk interaction potential in

Eq. (2). The explicit Cartesian multipole expansion for the relative PDF in Eq. (7) constitutes the first terms of a Fourier expansion for $p(\mathbf{x}, \tau|\mathbf{x}_0)$ which reads

$$p(\mathbf{x}, \tau|\mathbf{x}_0) = \frac{\Theta(r-1)}{2\pi}\bigg[a_0(r, \tau|r_0) + 2\sum_{n=1}^{\infty}\begin{pmatrix} a_n(r, \tau|r_0) \\ b_n(r, \tau|r_0)\end{pmatrix}\cdot \mathbf{e}(n\,\Delta\varphi)\bigg]. \quad (8)$$

Here a_n and b_n are the Fourier coefficients of order n, $n \in \mathbb{N}_0$. We introduce the notation $p = \Theta(r-1)\,[p_0 + \sum_{n=1}^{\infty} p_n]$, where $p_0 = a_0/2\pi$ and $p_n = [a_n \cos(n\,\Delta\varphi) + b_n \sin(n\,\Delta\varphi)]/\pi$, $n \geqslant 1$ for our subsequent shorthand notation of the Fourier modes. The Cartesian modes in Eq. (7) are thus connected to the Fourier modes as

$$\varrho(r, \tau|r_0) = \frac{1}{2\pi}\,a_0(r, \tau|r_0), \quad (9)$$

which is the (scalar) mean positional PDF,

$$\boldsymbol{\sigma}(r, \tau|r_0) = \frac{1}{\pi}\begin{pmatrix} a_1(r, \tau|r_0) \\ b_1(r, \tau|r_0)\end{pmatrix}, \quad (10)$$

which is the (vectorial) polarization order PDF, and

$$\boldsymbol{Q}(r, \tau|r_0) = \frac{1}{\pi}\begin{pmatrix} a_2(r, \tau|r_0) & b_2(r, \tau|r_0) \\ b_2(r, \tau|r_0) & -a_2(r, \tau|r_0)\end{pmatrix}, \quad (11)$$

which is the (tensorial) nematic order PDF.

In Appendix A, we provide the full solution to the relative problem, thereby following the original derivations in Refs. [9,13], and show that the general Fourier coefficients a_n, b_n are given by

$$a_n(r, \tau|r_0) = \frac{e^{-\frac{r_0^2+r^2}{4\tau}}}{2\tau}\,I_n\Big(\frac{r_0 r}{2\tau}\Big) - \mathscr{L}^{-1}\bigg\{k_n(r, s|r_0)\bigg[s\,K_n'(\sqrt{s})\,I_n'(\sqrt{s}) + (n\kappa)^2 K_n(\sqrt{s})\,I_n(\sqrt{s}) - \delta(r_0-1)\,\frac{\sqrt{s}\,K_n'(\sqrt{s})}{2K_n(\sqrt{s})}\bigg]\bigg\}, \quad (12)$$

$$b_n(r, \tau|r_0) = -n\kappa\left(1 - \frac{\delta(r_0-1)}{2}\right)\mathscr{L}^{-1}\{k_n(r, s|r_0)\}, \quad (13)$$

where we used the abbreviation

$$k_n(r, s|r_0) = \frac{K_n(r\sqrt{s})\,K_n(r_0\sqrt{s})}{(\sqrt{s}\,K_n'(\sqrt{s}))^2 + (n\kappa\,K_n(\sqrt{s}))^2}. \quad (14)$$

Here s denotes the (dimensionless) Laplace variable. The Laplace transform for a function $f(\tau)$ is defined as $\mathscr{L}\{f\}(s) = \int_0^\infty d\tau\, \exp(-s\tau) f(\tau)$, using the rescaled time variable $\tau = t/\tau_d$. The prime denotes the derivative $g'(a) = dg(x)/dx|_{x=a}$ and $I_n(x)$, $K_n(x)$ are the modified Bessel functions of the first kind and second kind, respectively [32]. As apparent from Eqs. (12) and (13), the inverse Laplace transformations remain unfeasible at the moment, and we rely on established numerical Laplace inversion methods.

We stress that the problem of solving for the full propagator of interacting particles in Eq. (3) can be formulated in the language of scattering theory [33], thus allowing us to use well-developed tools from quantum field theory [34]. Odd diffusion here adds the perspective of non-Hermicity with potentially unique insights [9].

B. Numerical results and Fourier modes

In Fig. 1, we compare the positional mode $p_0 = \Theta(r-1)\,\varrho$, the polarization mode $p_1 = \Theta(r-1)\,[\boldsymbol{\sigma}\cdot\mathbf{e}]$, and the nematic mode $p_2 = \Theta(r-1)\,[\mathbf{Q} : (\mathbf{e}\otimes\mathbf{e} - \mathbf{1}/2)]$ for a normal ($\kappa = 0$) and an odd-diffusive ($\kappa = 1$) system of interacting particles. The modes are evaluated at fixed time $\tau = 1$ and for $(r_0, \varphi_0) = (1.01, 0)$, i.e., particles are initially placed at a distance of 1.01 times their diameter along the (arbitrarily) chosen x axis.

We observe that $\kappa \neq 0$ does not affect the interacting particles' mean positional distribution p_0, but rotates higher order modes in comparison to $\kappa = 0$. We can understand this by observing from Eq. (13) that $b_n \propto n\kappa$, such that p_0 is not affected by $\kappa \neq 0$—but for higher order modes b_n contributes for an odd-diffusive system. It is of interest to analyze the contribution of the higher-order modes to the time evolution of the relative PDF p. We define a measure of the significance of a mode of order $n \geqslant 1$ as

$$\delta_n(\tau) = \frac{\int d\mathbf{x}\,|p_n(\mathbf{x}, \tau|\mathbf{x}_0)|}{\int d\mathbf{x}\,p_0(\mathbf{x}, \tau|\mathbf{x}_0)}. \quad (15)$$

We take the absolute value $|p_n|$ in the definition of $\delta_n(\tau)$, as the full space integral for the polarization, nematic, and all higher order modes vanishes due to the orthogonality of the harmonic functions, $\int d\mathbf{x}\; p_n = 0$, $n \geqslant 1$. In contrast, $\int d\mathbf{x}\; p_0 = 1$ (see Appendix C), which we only include in Eq. (15) to avoid numerical discretization errors.

Figure 2 shows $\delta_n(\tau)$ for $n = 1, \ldots, 5$ and for different values of the odd-diffusion parameter $\kappa = 0, 1, 5$. When considering δ_n as a measure for the relevance of the nth order mode, we observe that the modes are ordered consecutively in their contribution to the relative PDF p and that considering higher order modes becomes important for $\tau \to 0$ as $p(\tau) \to p(0) \propto \delta(\mathbf{x} - \mathbf{x}_0)$, but higher order modes become less important for $\tau \gg 0$. As can be seen in Figs. 2(b) and 2(c), $\kappa > 0$ shifts the decay of $\delta_n(\tau)$ to even shorter times. By an $s \to 0$ expansion of the Fourier coefficients in Eqs. (12) and (13), we gain access to the $\tau \to \infty$ behavior of the modes $p_n(\tau)$ via term-by-term Laplace-inversion. We find that $p_n \simeq \tau^{-(n+1)}$ for $\tau \to \infty$ for all $n \geqslant 0$ (see Appendix B for details). The long-time scaling of $\delta_n(\tau)$ therefore is $\delta_n(\tau) \simeq \tau^{-n}$, independently of κ, as demonstrated in Figs. 2(d)–2(f).

In Fig. 3, we plot the full relative PDF $p(\tau)$ at times $\tau = 1, 5, 50$ for a normal ($\kappa = 0$) and an odd-diffusive ($\kappa = 1$) system. Again we choose the initial condition to be $(r_0, \varphi_0) = (1.01, 0)$ and, based on the observation in Fig. 2, we truncated

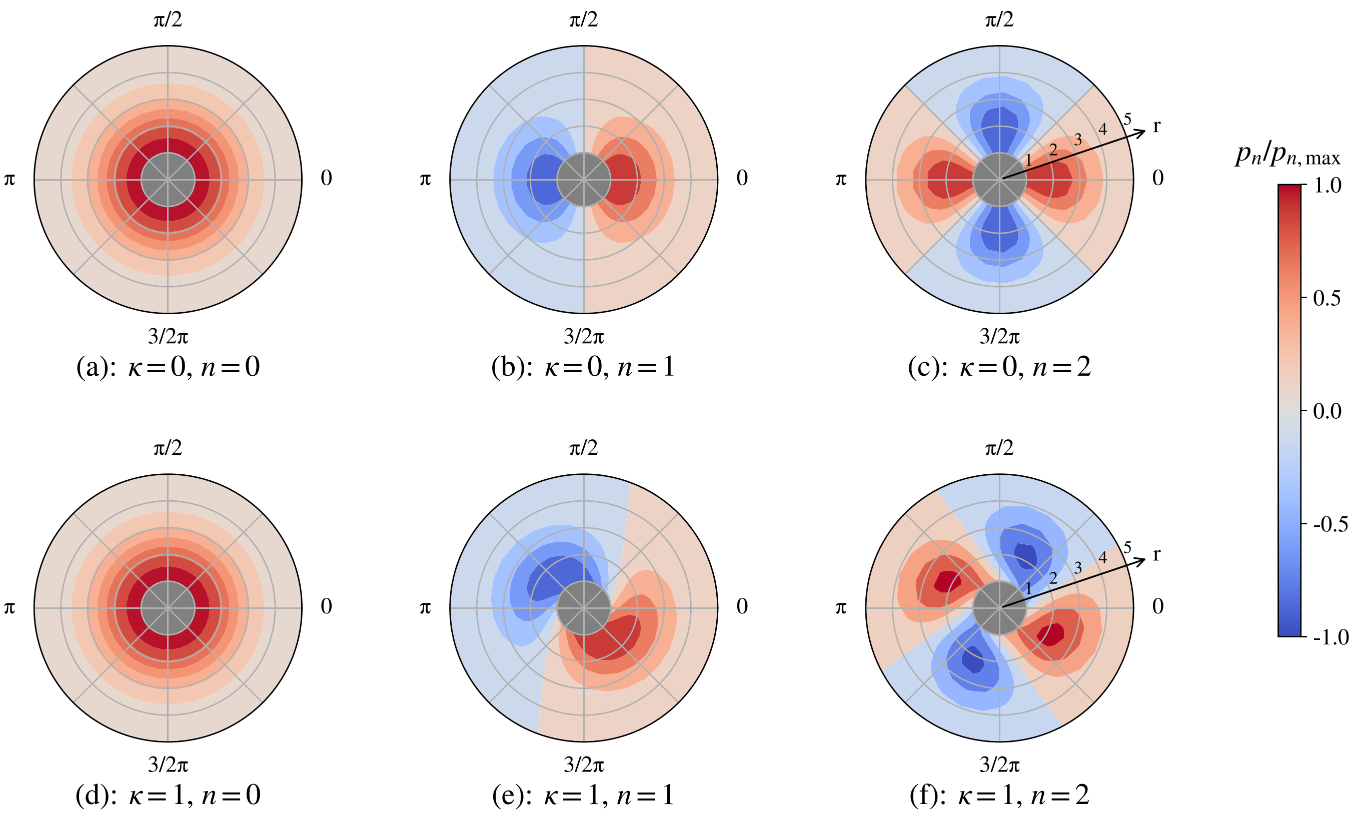


FIG. 1. Plots of the first three Fourier modes p_0, p_1, and p_2 of the relative PDF of interacting (odd-diffusive) particles $p(r, \tau|r_0) = \Theta(r-1)\sum_{n=0}^{\infty} p_n(r, \tau|r_0)$ at fixed time $\tau = 1$. (a)–(c) Modes for normal particles ($\kappa = 0$) and (d)–(f) for odd-diffusive particles ($\kappa = 1$). The initial positions are chosen as $(r_0, \varphi_0) = (1.01, 0)$, i.e., the particles are placed 1.01 times their diameter along an arbitrarily defined x axis. The zeroth-order mode in (a) and (d) equals the mean positional PDF for the relative coordinate r [see Eq. (9)] which is unaffected by odd diffusion. The first order mode in (b) and (e), which corresponds to the (contracted) polarizational order PDF [see Eq. (10)], and the second order mode in (c) and (f), which corresponds to the (contracted) nematic order PDF [see Eq. (11)], as well as all higher order modes are affected by odd diffusion such that the modes are rotated in time compared to the $\kappa = 0$ case; see also Fig. 4. Note that the gray circle in the middle of each plot is due to the excluded volume condition $\Theta(r-1)$ of the hard-disk interaction of the particles.

the Fourier series at $n = 10$. Comparing the normal and odd-diffusive relative PDF, we observe that the κ-induced rotation of every but the zeroth order Fourier mode persists into the full PDF. For $\kappa = 0$, $b_n = 0$ for all modes, and the Fourier representation of the relative PDF therefore only contains cosine terms. The relative PDF of a normal particle thus is symmetric around $\Delta\varphi = 0, \pi$ for all times. This symmetry implies that particles encounter the space from both sides after a collision with equal likelihood. However, this does not hold for odd-diffusive particles. $\kappa \neq 0$ introduces a handedness in the diffusive exploration of space. For interacting odd-diffusive particles, the additional rotational probability flux introduced via Eq. (1) results in a preferred direction after a collision depending on the sign of the odd-diffusion parameter κ. This effect was recently observed from Brownian dynamics simulations and termed mutual rolling [2]. The interaction-induced symmetry breaking has far-reaching consequences for observable transport coefficients such as the self-diffusion coefficient. In odd-diffusive systems, even though resembling equilibrium overdamped dynamics, the self-diffusion can be enhanced by interactions instead of being reduced as for a normal system [2]. Even though seemingly contradicting equilibrium statistical mechanics theorems [23,35,36], the interaction-enhanced self-diffusion could be rationalized by observing that the time-evolution in odd-diffusive systems [see Eq. (3)] becomes non-Hermitian for finite κ [9].

C. Relevance of the polarization mode for the force autocorrelation

The force autocorrelation tensor (FACT) $\mathbf{C}_F(\tau)$ encodes the most detailed microscopic information in an interacting system. Via a Taylor-Green-Kubo relation [37–39], it encodes the particle-particle interaction effects in the self-diffusion. For a stationary system, $\mathbf{C}_F(\tau)$ is defined as

$$\mathbf{C}_F(\tau) = \langle \mathbf{F}(\vec{\mathbf{x}}) \otimes \mathbf{F}(\vec{\mathbf{x}}_0)\rangle = \int \mathrm{d}\vec{\mathbf{x}} \int \mathrm{d}\vec{\mathbf{x}}_0\, \mathbf{F}(\vec{\mathbf{x}}) \otimes \mathbf{F}(\vec{\mathbf{x}}_0)\, P_N(\vec{\mathbf{x}}, \tau, \vec{\mathbf{x}}_0, \tau_0), \tag{16}$$

where $\vec{\mathbf{x}} = \{\mathbf{x}_1, \dots, \mathbf{x}_N\}$ and similarly $\vec{\mathbf{x}}_0$ for a system of, in general, N particles. P_N is the N-particle joint PDF, which can be rewritten as $P_N(\vec{\mathbf{x}}, \tau, \vec{\mathbf{x}}_0, \tau_0) = P_N(\vec{\mathbf{x}}, \tau|\vec{\mathbf{x}}_0, \tau_0)\, P_{\mathrm{eq}}(\vec{\mathbf{x}}_0, \tau_0)$, assuming the particles where in equilibrium at time τ_0, which we take again to be 0. The force on the tagged particle (particle one) is $\mathbf{F}(\vec{\mathbf{x}}) = -\nabla_1 U_N(\vec{\mathbf{x}})$, and we assume a pairwise additive and radially symmetric potential $U_N(\vec{\mathbf{x}}) = \sum_{i,j=1}^{N} U(r_{ij})/2$, where $r_{ij} = |\mathbf{x}_i - \mathbf{x}_j|, i \neq j$. In the dilute limit, we

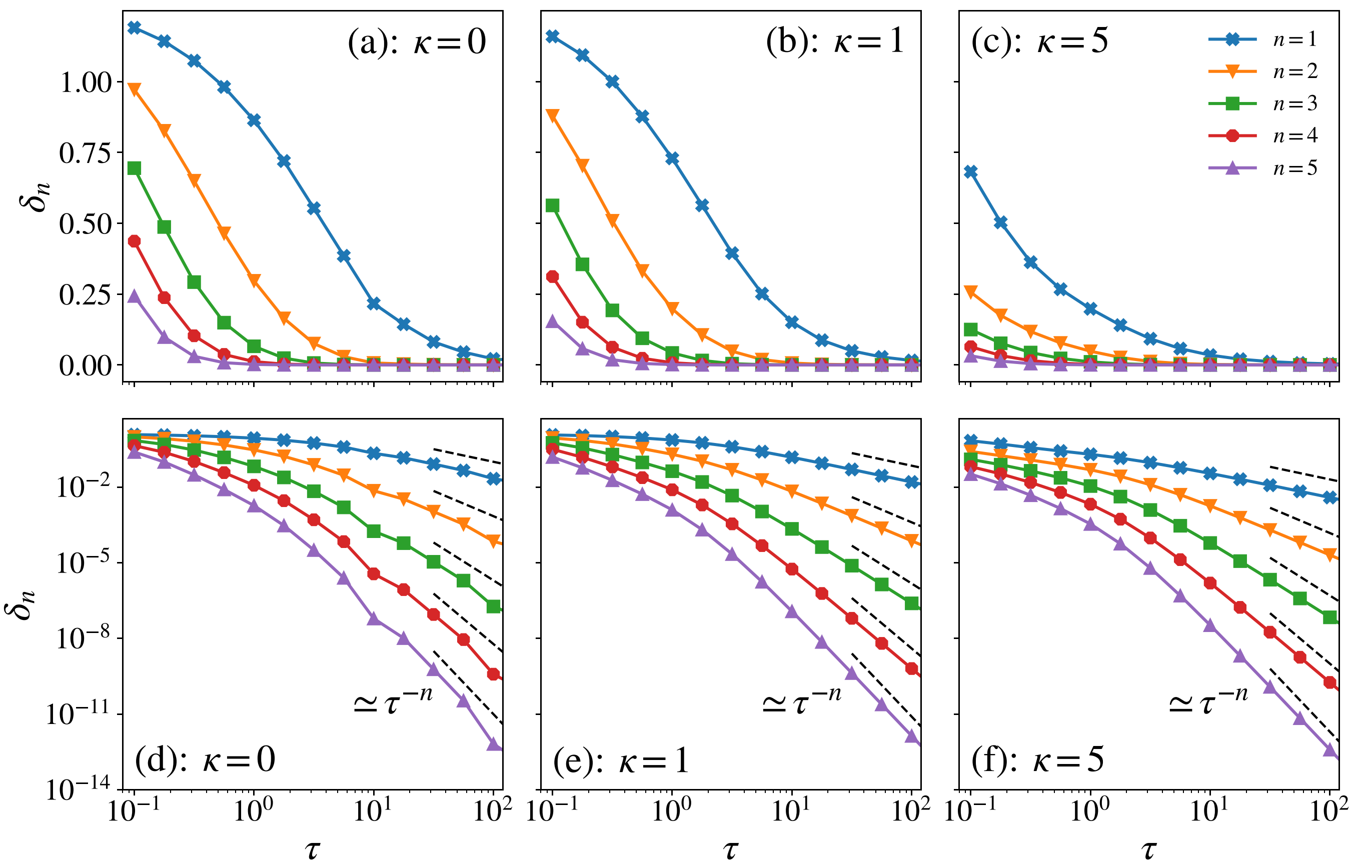


FIG. 2. Significance $\delta_n(\tau)$ of a mode n as defined in Eq. (15), which measures the contribution of a Fourier mode of order $n \geqslant 1$, $p_n(\tau)$, to the full relative PDF $p(\tau) = \Theta(r-1)\sum_{n=0}^{\infty} p_n(\tau)$ as a function of time τ for odd-diffusion parameter $\kappa = 0$, 1 and 5 in (a), (d); (b), (e); and (c), (f); respectively. We see that higher order modes are consecutively ordered in their contribution to p in time and become of comparable importance for $\tau \to 0$ as $p(\tau) \to p(0) \propto \delta(\mathbf{x} - \mathbf{x}_0)$. Comparing (a) with (c), we observe that $\kappa > 0$ shifts the characteristic decay to shorter times. Thus, for dynamics where one is not interested in the $\tau \to 0$ limit, one can truncate the Taylor expansion at finite n. (d)–(f) Algebraic decay of $\delta_n(\tau) \simeq \tau^{-n}$ for $\tau \to \infty$, independently of κ.

can safely assume that only two-body correlations are important and thus ignore correlations between the untagged particles. The equilibrium PDF can be approximated as $P_{\text{eq}}(\vec{\mathbf{x}}_0) = 1/(VZ_N)\prod_{i=2}^{N}\exp(-\beta\, U(r_{1i,0})/2)$, where $V \subset \mathbb{R}^2$ is the bounded space of diffusion and $Z_N = \int \mathrm{d}\vec{\mathbf{x}}_0 \quad \prod_{i=2}^{N}\exp(-\beta\, U(r_{1i,0})/2)$ is the N-particle partition function. The conditional PDF, i.e., the N-particle propagator can be approximated as $P_N(\vec{\mathbf{x}}, \tau|\vec{\mathbf{x}}_0) = (1/V)\prod_{i=2}^{N} p(\mathbf{x}_{1i}, \tau|\mathbf{x}_{1i,0})$, where $p(\mathbf{x}_{1i}, \tau|\mathbf{x}_{1i,0})$ is the PDF of the relative coordinate $\mathbf{x}_{1i} = \mathbf{x}_1 - \mathbf{x}_i$, similar to Eq. (7) but for a generic two-body interaction potential U. Following these approximations, all but one particle (particle two) can be integrated out in Eq. (16) and we denote, as before, $\mathbf{x} = \mathbf{x}_1 - \mathbf{x}_2$. The central interaction force between the particles can be written as $\mathbf{F}(\mathbf{x}) = F(r)\,\mathbf{e}(\varphi)$, where $\mathbf{e}(\varphi) = (\cos(\varphi), \sin(\varphi))^{\mathrm{T}}$, as before, and coincides with the radial unit vector. In the dilute limit, the FACT of Eq. (16) thus becomes

$$\mathbf{C}_F(\tau) = \frac{N-1}{V}\int \mathrm{d}\mathbf{x} \int \mathrm{d}\mathbf{x}_0\, F(r)\,F(r_0)\,\frac{\mathrm{e}^{-\beta U(r_0)}}{Z_2} \times\, p(\mathbf{x}, \tau|\mathbf{x}_0)\,\mathbf{e}(\varphi) \otimes \mathbf{e}(\varphi_0) \tag{17}$$

$$= \frac{N-1}{V}\pi \int \mathrm{d}r\, r \int \mathrm{d}r_0\, r_0\, F(r)\,F(r_0) \times\, \frac{\mathrm{e}^{-\beta U(r_0)}}{Z_2}\,[a_1(r, \tau|r_0)\,\mathbf{1} - b_1(r, \tau|r_0)\,\boldsymbol{\epsilon}]. \tag{18}$$

Here we used the orthogonality of the Fourier modes in the relative PDF $p(\tau)$ [analogously to Eq. (7)] from Eq. (17) to Eq. (18) to find that only the polarization mode $\boldsymbol{\sigma}(r, \tau|r_0) = (a_1(r, \tau|r_0), b_1(r, \tau|r_0))^{\mathrm{T}}/\pi$ [analogously to Eq. (10)] contributes to the FACT. Note that relations analogous to Eqs. (7) and (10) also hold in a system with generic radially symmetric interaction potential $U(r)$, specifically also for forces with transverse, odd components [7,40–42].

We can specify Eq. (18) for the hard-disk interaction potential of Eq. (2) by observing that $P_{\text{eq}}(\mathbf{x}_0) = \Theta(r_0 - 1)/V^2$ and that we can rewrite the singular interaction force via the trick $\beta\Theta(r-1)\,\mathbf{F}(r) = \delta(r-1)\,\mathbf{e}(\varphi)$ [43], obeying the same generic form of a central force as before. We can perform the radial integral of Eq. (18) and find that the FACT of a hard-disk system in the dilute limit is given by $\mathbf{C}_F(\tau) = \beta^{-2}\phi\,[a_1(1, \tau|1)\,\mathbf{1} - b_1(1, \tau|1)\,\boldsymbol{\epsilon}]$, where $\phi = (\pi d^2/4)(N/V)$ is the area fraction in dimensional form

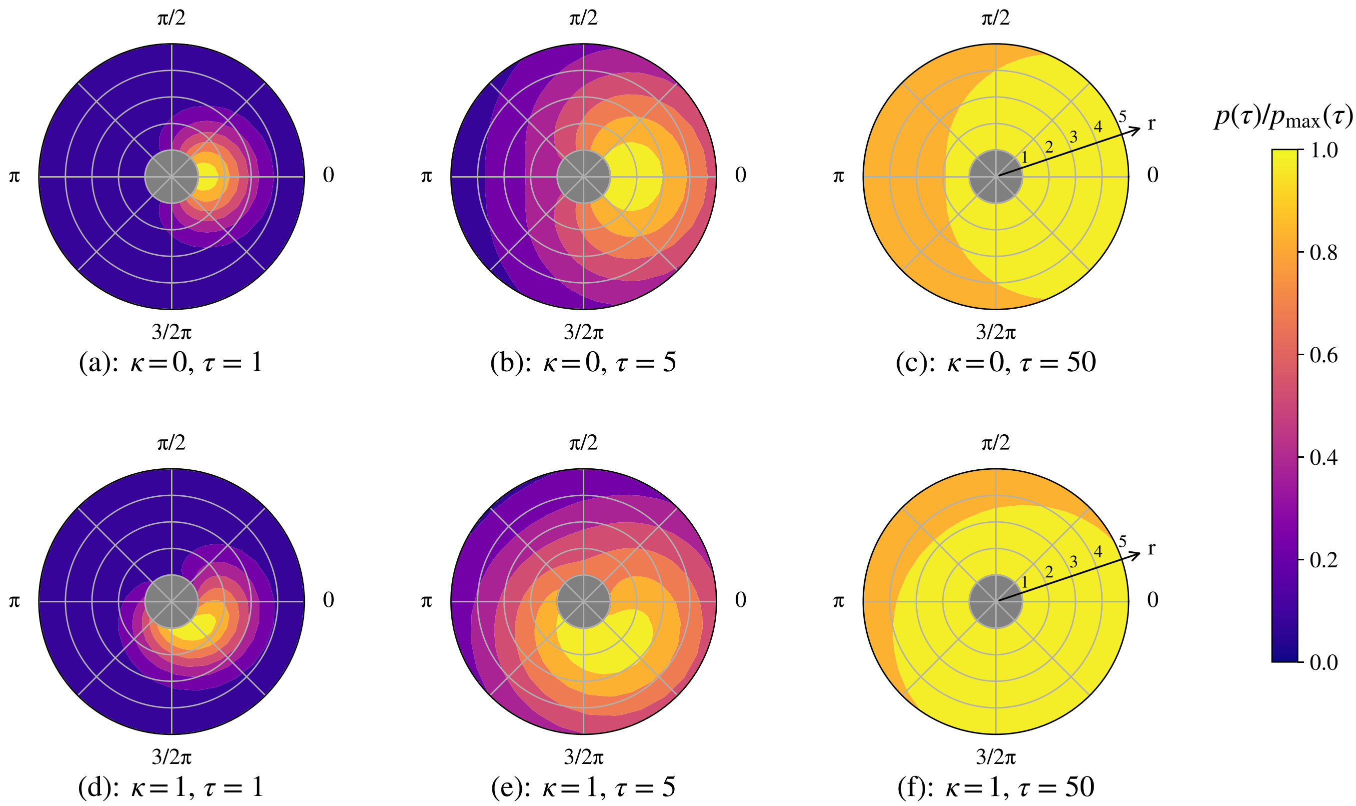


FIG. 3. PDF $p(r, \varphi, \tau|r_0, \varphi_0)$ for the relative coordinate of two hard interacting particles. (a)–(c) Normal particles ($\kappa = 0$) and (d)–(f) odd-diffusive particles ($\kappa = 1$) at times $\tau = 1$, 5, and 50. The initial positions are chosen as $(r_0, \varphi_0) = (1.01, 0)$, i.e., the particles are placed 1.01 times their diameter along an arbitrarily defined x axis. The Fourier expansion of $p(\tau)$ in Eq. (8) is truncated at $n = 10$ in the numerical evaluation for the plots; see also Fig. 2 for a rationale. (d)—(f) The mutual rolling effect, i.e., that odd-diffusive particles rotate around each other after a collision [2], which is in contrast to a symmetric back reflection for normal particles, visible in (a)–(c). Note the grey circle in the middle of each plot which is due to the excluded volume condition $\Theta(r-1)$ of the hard-disk interaction of the particles.

[9,22]. Thus we understand that the behavior of the polarization mode $\boldsymbol{\sigma}(\tau)$ in time governs the behavior of the FACT for interacting systems, in particular, for a hard-disk interaction. Note specifically here that the off-diagonal correlation ($\propto b_1\,\boldsymbol{\epsilon}$) is directly proportional to κ, and therefore might serve as a characteristic to odd diffusion [3,9].

To further analyze the polarization mode in the hard system, we define

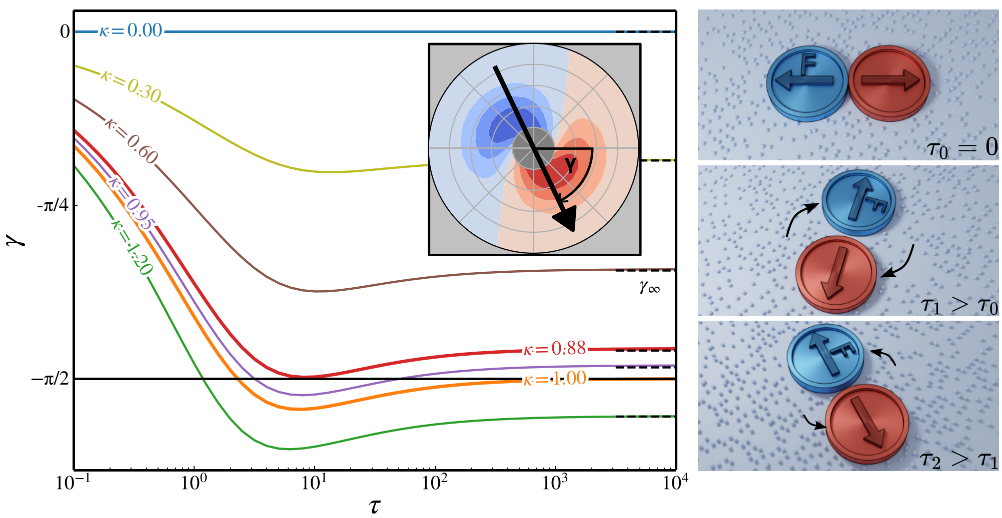
$$\gamma(\tau) = \arctan\left(\frac{b_1(1, \tau|1)}{a_1(1, \tau|1)}\right) \tag{19}$$

as the mean angle between the polarization vector $\boldsymbol{\sigma}(1, \tau|1)$ as a function of time and its initial direction, which defines the x axis in the system; see also inset in Fig. 4(a). We plot $\gamma(\tau)$ for different values of κ as a function of time in Fig. 4(a) and use the arctan2(·) function for numerical evaluation to obtain angles within the interval $(-\pi, \pi)$. We observe that for $\kappa = 0$ the initial direction of the polarization does not change with time and stays constant. Recalling the polarization mode in Fig. 1(b), the location of the extrema thus remains unchanged, meaning that the particles are symmetrically back-reflected from the center of the collision. For $\kappa \neq 0$, however, $\gamma(\tau)$ changes in time [see again Fig. 1(e)]. The mean direction of the polarization develops an extremum $\gamma_{\rm ext}$ and relaxes to a constant, nonzero value at long times:

$$\gamma_\infty = -\arctan\left(\frac{2\kappa}{1-\kappa^2}\right). \tag{20}$$

This result can be obtained from the long-time behavior of a_1 and b_1 (see Appendix B for details). For $\kappa > 0.88$, $\gamma_{\rm ext} < -\pi/2$ and for $\kappa > 1$ even $\gamma_\infty < -\pi/2$. The interval $\kappa \in (0.88, 1)$ in which $\gamma(\tau)$ oscillates around $-\pi/2$ thereby coincides with the range of the odd-diffusion parameter for which we reported an oscillating FACF earlier [9]. The sign switches in the FACF coincide with what we observe now for the direction of $\boldsymbol{\sigma}(\tau)$; see also inset in Fig. 4(a): $\gamma(\tau)$ crosses $-\pi/2$ for some time (negative FACF) and eventually relaxes to angles larger than $-\pi/2$ again (positive FACF).

We interpret this behavior as interacting odd-diffusive particles, which for $|\kappa| > 0.88$ rotate more than $\pi/2$ but relax to a steady state with a relative angle $|\gamma_\infty| < \pi/2$ as long as $|\kappa| < 1$; see Fig. 4(b) for a sketch. This behavior also rationalizes the oscillating FACF in the same interval, as $\langle \mathbf{F}(\tau)\cdot\mathbf{F}(0)\rangle = \langle F(\tau)F(0)\cos(\pi/2)\rangle = 0$ here and the particles oscillate around that angle. However, a steady-state angle $|\gamma_\infty|$ which is smaller than the maximal angle $|\gamma_{\rm ext}|$ is a generic observation from Fig. 4(a). On average, particles always rotate further in time, as they finally relax. Note that for $\kappa \to -\kappa$, we have that $\gamma(\tau) \to -\gamma(\tau)$, as a_n is even and



(a) (b)

FIG. 4. (a) Angle $\gamma(\tau)$ between the polarization mode $\boldsymbol{\sigma}(r=1,\tau|r_0=1)$, Eq. (10), and its initial direction as function of time τ for different odd-diffusion parameters κ; see Eq. (19) and the inset. The polarization mode constitutes the vectorial mode of the relative PDF $p(\tau)$ for interacting particles and represents relative particle rearrangements. In a normal system ($\kappa=0$), $\gamma(\tau)=0$ and is constant in time, whereas it develops an extremum $\gamma_{\rm ext}$ different from its asymptotic steady state value $\gamma_\infty = -\arctan(2\kappa/(1-\kappa^2))$ (dashed lines) for $\kappa \neq 0$ in time. We observe that $|\gamma_{\rm ext}| > |\gamma_\infty|$, indicating that particles on average rotate further in time than they finally relax to. Specifically for $\kappa \in (0.88, 1.0)$, $\gamma_{\rm ext} < -\pi/2$, but $\gamma_\infty > -\pi/2$ which quantitatively aligns with the oscillating FACF previously observed for this regime [9]. (b) The sketch qualitatively illustrates the scenario, that odd-diffusive particles with $0.88 < \kappa < 1$ collide at τ_0 with each other along the (arbitrary) x axis and rotate for $\tau_1 > \tau_0$ to $\gamma_{\rm ext} < -\pi/2$, and eventually relax at $\tau_2 > \tau_1$ to $\gamma_\infty > -\pi/2$.

b_n is an odd function of κ. Thus, $\kappa < 0$ results in the same phenomenology for the relaxation of $\gamma(\tau)$ and thus for the FACF. Further, if $|\kappa| \to \infty$, we find that $|\gamma_{\rm ext}| \to |\gamma_\infty| \to \pi$, which means that, at most, odd particles exchange positions but do not rotate any further.

III. DISCUSSION

We here presented the exact analytical solution and numerical evaluation of two hard disklike interacting odd-diffusing particles in two spatial dimensions. Odd diffusion thereby is characterized by antisymmetric elements $\propto \kappa$ in the diffusion tensor $\mathbf{D} = D_0(\mathbf{1} + \kappa\boldsymbol{\epsilon})$. Our analysis showed that the two-particle problem separates into a center of mass and a relative coordinate problem, of which the first can be solved straightforwardly, and the latter incorporates the nontrivial effects of interactions and odd diffusion. The relative PDF can be written as a Fourier series, and we observe that oddness rotates all apart from the zeroth order mode in time, which represents the unaffected positional distribution of the relative coordinate. The modes are consecutively ordered in their contribution to the relative PDF, and when summed up show a rotated form of the PDF. This effect is a characteristic of odd diffusion and has been termed as mutual rolling earlier [44] as the particles rotate around each other while interacting, in contrast to normal diffusive ($\kappa = 0$) particles, which are symmetrically back-reflected when interacting.

The representation of the relative PDF in its Fourier modes becomes useful in the analysis of microscopic correlation functions, specifically the FACF. For any central interaction force, only the polarization mode of the relative PDF determines the correlation function. We conjecture here that a similar phenomenology holds for other (radially symmetric) observables of arbitrary tensorial order in an interacting system; the corresponding mode of the relative PDF might determine the expectation value and even the correlation function. We used the analytical access to the polarization mode to understand the average configurations of the hard-disk-like interacting odd-diffusive particles. The maximal relative rotation angle overshoots the final relaxation on average, which quantitatively aligns with an oscillating FACF, recently reported for these systems [9].

Our work shines light on unique particle interactions in odd-diffusive systems. Thereby, it might serve as a reference case for interactions in the various odd-diffusive systems such as in equilibrium, e.g., Brownian particle under Lorentz force [6,44–56], with a long-lasting history in statistical mechanics [57–59], or skyrmionic spin structures [60–62]. Here numerical studies recently reproduced the interaction-enhanced self-diffusion [7], which originates in the mutual rolling effect [2]. But there also exist nonequilibrium odd-diffusive systems, such as systems under shear [63,64] or active chiral particles [1,30,42,65–69]. In general, for systems that break time-reversal symmetry, the relevance of off-diagonal correlation functions for transport properties has attracted considerable interest recently [1,3,5,42,67,70,71]. Odd diffusion further might serve as the unifying terminology for systems showing transverse responses such as systems with Magnus forces [72–74], Coriolis forces [75,76], in complex (porous) environments [77–79], or to describe nonconservative force fields which are found in optical tweezer experiments [80–83]. Vortex fluids, such as those found in the biological, low Reynolds number regime [84,85] but also in the high Reynolds number regime [86,87], further show a remarkable similarity to the phenomenology presented in this paper and might be considered in this framework. Systems with an artificial transverse interaction component [41,88] recently showed that odd interactions enhance the sampling of configurations in dense systems, originating again in the unique particle rearrangements in odd-diffusive systems. Transverse forces further have been the subject of interest in so-called linear-diffusive systems [89–93]. Odd diffusion is finally also relevant for strongly rotating [94,95] or magnetized plasmas, as can be found, e.g., in the realm of astrophysics, where antisymmetric transport is relevant to describe the movement of energetic particles moving through magnetized plasmas such as the interplanetary and interstellar medium [96–98].

ACKNOWLEDGMENTS

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APPENDIX A: ANALYTICAL SOLUTION OF THE RELATIVE PROBLEM

This Appendix closely follows Ref. [9] in its presentation of the analytical solution to the relative problem, which itself adapted the work of Hanna *et al.* [13] to odd-diffusive systems.

The equation for the relative PDF $p(\tau) = p(\mathbf{x}, \tau|\mathbf{x}_0)$, where $\mathbf{x} = \mathbf{x}_1 - \mathbf{x}_2$, follows from Eq. (5) as

$$\frac{\partial}{\partial \tau} p(\tau) = \nabla_{\mathbf{x}} \cdot (\mathbf{1} + \kappa \boldsymbol{\epsilon})[\nabla_{\mathbf{x}} + \beta\, \nabla_{\mathbf{x}} U(r)]\, p(\tau), \tag{A1}$$

and reads in (rescaled) polar coordinates $\mathbf{x} = (r, \varphi)$ as

$$\frac{\partial}{\partial \tau} p(\tau) = \frac{1}{r^2}\left(r\frac{\partial}{\partial r} r\frac{\partial}{\partial r} + r\frac{\partial}{\partial r}\, r\beta\, \frac{\partial U(r)}{\partial r} + \frac{\partial^2}{\partial \varphi^2} - \kappa\, r\beta\, \frac{\partial U(r)}{\partial r}\frac{\partial}{\partial \varphi}\right) p(\tau). \tag{A2}$$

Note that space is rescaled with the diameter of the particle d, $\mathbf{x} \to \mathbf{x}/d$, and time is rescaled by the natural timescale of diffusing the radial distance of a particle diameter $\tau_d = d^2/(2D_0)$, $t \to \tau = t/\tau_d$. The initial condition $p(\tau = 0) = \delta(\mathbf{x} - \mathbf{x}_0)\, \Theta(r - 1)$ in polar coordinates becomes

$$p(\tau = 0) = \frac{\delta(r - r_0)}{r_0}\, \delta(\varphi - \varphi_0)\, \Theta(r - 1). \tag{A3}$$

The angular part of the initial condition can be expanded into a Fourier series $\delta(\varphi - \varphi_0) = \sum_{n=-\infty}^{\infty} \exp(\mathrm{i}n(\varphi - \varphi_0))/2\pi$, which, following Refs. [9,13], we use as an ansatz to solve for $p(\tau)$ as

$$p(\mathbf{x}, \tau|\mathbf{x}_0) = \frac{\Theta(r - 1)}{2\pi} \sum_{n=-\infty}^{\infty} R_n(r, \tau|r_0)\, \mathrm{e}^{\mathrm{i}n(\varphi - \varphi_0)}. \tag{A4}$$

Note that except for the radial functions $R_n(r, \tau|r_0)$, all other parts of the ansatz are time independent.

Observing that for the hard-disk potential, see Eq. (2), we have that $\exp(-\beta U(r)) = \Theta(r - 1)$, we can replace the otherwise singular interaction force $\partial U(r)/\partial r$ in Eq. (A2) via

$$-\beta \Theta(r - 1)\, \frac{\partial U(r)}{\partial r} = \frac{\partial}{\partial r}\, \mathrm{e}^{-\beta U(r)} = \delta(r - 1). \tag{A5}$$

Rewriting Eq. (A2) for the radial functions $R_n(\tau) = R_n(r, \tau|r_0)$, we thus find

$$\frac{\partial}{\partial \tau} R_n(\tau) = \frac{1}{r^2}\left(r\frac{\partial}{\partial r} r\frac{\partial}{\partial r} - n^2\right) R_n(\tau) \tag{A6}$$

in the domain $r \geqslant 1$ and for each order $n \in \mathbb{Z}$, and

$$\frac{\partial}{\partial r} R_n(\tau) = -\frac{\mathrm{i}n\kappa}{r} R_n(\tau) \tag{A7}$$

to be satisfied additionally at $r = 1$. Equation (A7) can be viewed as an extension of an ordinary Neumann no-flux boundary condition which is recovered for $\kappa = 0$. This generalized condition can be found in the literature under the name of *oblique* boundary conditions; see, for example, Ref. [99]. Equation (A6) is equipped with a second boundary condition, which can be found from the natural boundary condition on $p(\tau)$, $\lim_{r\to\infty} p(\tau) = 0$, as $\lim_{r\to\infty} R_n(\tau) = 0$ and the initial condition on $R_n(\tau)$ translates from Eq. (A3) as $R_n(\tau = 0) = \delta(r - r_0)/r_0$. Note that Eq. (A6) together with the boundary condition (A7) also forms the basis of solving the problem by using the language of scattering theory [33], the only difference being the introduction of the the generalized Neumann boundary condition for interacting odd-diffusive particles.

For the particular solution of Eq. (A6), $R_n^{\text{part}}(\tau)$, which satsifies the initial condition, we make the ansatz

$$R_n^{\text{part}}(\tau) = \int_0^{\infty} \mathrm{d}u\, w_n(r, u|r_0)\, \mathrm{e}^{-\tau u^2}, \tag{A8}$$

which when inserted into Eq. (A6) shows that $w_n(r, u|r_0)$ satisfy the Bessel equation [32] with solutions $J_n(ur)$ and $Y_n(ur)$ as the Bessel functions of first and second kinds, respectively. Matching the particular solution with the initial condition $R_n(\tau = 0) = \delta(r - r_0)/r_0$ and noting that the delta-distribution $\delta(\cdot)$ can be expanded in Bessel functions of the first kind [see Eq. (C1)], we find that $Y_n(ur)$ does not contribute to the particular solution.

After a Laplace transformation with (dimensionless) Laplace variable s, the complementary part of Eq. (A6), with initial condition zero appears as the modified Bessel equation [32] with solutions $I_n(r\sqrt{s})$ and $K_n(r\sqrt{s})$, as the modified Bessel functions of the first and second kinds, respectively. The Laplace transformation for a function $f(\tau)$ thereby is defined as $\mathscr{L}\{f\}(s) = \int_0^{\infty} \mathrm{d}\tau\, \exp(-s\tau) f(\tau)$, noting that $\tau = t/\tau_d$. As the complementary solution has to satisfy the natural boundary condition on $R_n(\tau)$, $I_n(x)$ is no suitable solution as it diverges for $x \to \infty$ for every $n \in \mathbb{Z}$ [32, 9.7.1]. We conclude that the homogeneous part is solved by $\mathscr{L}\{R_n^{\text{comp}}\}(s) = A_n(s|r_0)\, K_n(r\sqrt{s})$ for some amplitude $A_n(s|r_0)$ to be determined by matching with the oblique boundary condition (A7).

The relative solution $p(\tau)$ in the Laplace domain thus is given by

$$\mathscr{L}\{p\}(\mathbf{x}, s|\mathbf{x}_0) = \Theta(r-1)\sum_{n=-\infty}^{\infty}\frac{\mathrm{e}^{\mathrm{i}n(\varphi-\varphi_0)}}{2\pi}\int_0^\infty \mathrm{d}u\,\frac{u\,J_n(ur_0)}{s+u^2}\left[J_n(ur) - K_n(r\sqrt{s})\,\frac{u\,J_n'(u)+\mathrm{i}n\kappa\,J_n(u)}{\sqrt{s}\,K_n'(\sqrt{s})+\mathrm{i}n\kappa\,K_n(\sqrt{s})}\right]. \tag{A9}$$

This solution was already found in Ref. [9]. There are two types of integrals appearing in Eq. (A9), which involve products of Bessel functions and which we list in Appendix C. The integrals $\int_0^\infty \mathrm{d}u\; u\,J_n(ur_0)J_n(ub)/(s+u^2)$ can be evaluated using Eq. (C2), where $b \in \{1, r\}$. Depending on whether $b = r > r_0$, $r = r_0$, or $r < r_0$, Eq. (C2) gives different results, but we only need to consider the latter two for the cases $b = 1 = r_0$ and $1 < r_0$. The second integral $\int_0^\infty \mathrm{d}u\; u^2\,J_n(ur_0)J_n'(u)/(s+u^2)$ involves the derivative of a Bessel function and requires more work. We list the integral in Eq. (C5) and again need to differentiate whether $r_0 = 1$ or $1 < r_0$. Note that the integrals are formally defined for $n > -1$ only. However, we can use them for $n \in \mathbb{Z}$ as we may easily extend them to negative n by using the symmetry relation of the Bessel function, $J_{-n}(x) = (-1)^n J_n(x)$ [32, 9.1.5]. Taking into account these different cases, we find for the Laplace transformed PDF of the relative coordinate

$$\begin{aligned}\mathscr{L}\{p\}(\mathbf{x}, s|\mathbf{x}_0) = \frac{\Theta(r-1)}{2\pi}\sum_{n=-\infty}^{\infty}\mathrm{e}^{\mathrm{i}n(\varphi-\varphi_0)}\Bigg[&\Theta(r_0-r)\,I_n(r\sqrt{s})\,K_n(r_0\sqrt{s}) + \Theta(r-r_0)\,I_n(r_0\sqrt{s})\,K_n(r\sqrt{s})\\ &-\frac{K_n(r\sqrt{s})\,K_n(r_0\sqrt{s})}{\sqrt{s}\,K_n'(\sqrt{s})+\mathrm{i}n\kappa\,K_n(\sqrt{s})}\left(\sqrt{s}\,I_n'(\sqrt{s})+\mathrm{i}n\kappa\,I_n(\sqrt{s})-\frac{\delta(r_0-1)}{2K_n(\sqrt{s})}\right)\Bigg].\end{aligned} \tag{A10}$$

We can Laplace invert some parts of Eq. (A10) as

$$\begin{aligned}\mathscr{L}^{-1}\{I_n(r_0\sqrt{s})\,K_n(r\sqrt{s})\} &= \mathscr{L}^{-1}\{I_n(r\sqrt{s})\,K_n(r_0\sqrt{s})\}\\ &= \frac{\exp\left(-\frac{r_0^2+r^2}{4\tau}\right)}{2\tau}I_n\left(\frac{r_0 r}{2\tau}\right),\end{aligned} \tag{A11}$$

[100, 13.96]. Together with decomposing $\mathrm{e}^{\mathrm{i}x} = \cos x + \mathrm{i}\sin x$ in Eq. (A10), this gives the Fourier expanded relative PDF as

$$\begin{aligned}p(\mathbf{x}, \tau|\mathbf{x}^0) = \frac{\Theta(r-1)}{2\pi}\Bigg[&a_0(r,\tau|r_0) + 2\sum_{n=1}^{\infty}\begin{pmatrix}a_n(r,\tau|r_0)\\ b_n(r,\tau|r_0)\end{pmatrix}\\ &\cdot\begin{pmatrix}\cos\left(n(\varphi-\varphi_0)\right)\\ \sin\left(n(\varphi-\varphi_0)\right)\end{pmatrix}\Bigg],\end{aligned} \tag{A12}$$

where a_n and b_n are given in Eqs. (12) and (13) in the main text. Note that by using the symmetry in the order of the modified Bessel functions, $I_{-n}(x) = I_n(x)$ and $K_{-n}(x) = K_n(x)$ [32, 9.6.6], we can restrict the sum on positive modes only.

APPENDIX B: LONG-TIME ASYMPTOTIC BEHAVIOR

The long-time behavior of p is governed by the long-time behavior of the modes p_n. We individually infer these from a $\tau \to \infty$ expansion of the analytically Laplace inverted part in Eq. (A11), which is given by

$$\begin{aligned}&\frac{\exp\left(-\frac{r_0^2+r^2}{4\tau}\right)}{2\tau}I_n\left(\frac{r_0 r}{2\tau}\right)\\ &\simeq \frac{(rr_0)^n}{n!\,2^{1+2n}}\,\tau^{-(n+1)} - \frac{(rr_0)^n\left(r^2+r_0^2\right)}{n!\,2^{3+2n}}\,\tau^{-(n+2)}\\ &\quad+\mathcal{O}(\tau^{-(n+3)}),\end{aligned} \tag{B1}$$

and an $s \to 0$ expansion with a piecewise Laplace inversion of the remainder of Eq. (A10), which we abbreviate as

$$\begin{aligned}f_n(s) = &\frac{K_n(r\sqrt{s})\,K_n(r_0\sqrt{s})}{\sqrt{s}\,K_n'(\sqrt{s})+\mathrm{i}n\kappa\,K_n(\sqrt{s})}\\ &\times\left(\sqrt{s}\,I_n'(\sqrt{s})+\mathrm{i}n\kappa\,I_n(\sqrt{s})-\frac{\delta(r_0-1)}{2K_n(\sqrt{s})}\right).\end{aligned} \tag{B2}$$

We cannot present the expansion in a closed form for an arbitrary mode n here, but instead find that for $n = 0$ as $s \to 0$:

$$\begin{aligned}f_0(s) \simeq\; &A_0^0(r,r_0) + B_0^0(r,r_0)\,\ln(s)\\ &+A_0^1(r,r_0)\,s^1 + B_0^1(r,r_0)\,s^1\ln(s) + C_0^1(r,r_0)\,s^1\ln^2(s)\\ &+A_0^2(r,r_0)\,s^2 + B_0^2(r,r_0)\,s^2\ln(s) + C_0^2(r,r_0)\,s^2\ln^2(s)\\ &+D_0^2(r,r_0)\,s^2\ln^3(s) + \mathcal{O}(s^3).\end{aligned} \tag{B3}$$

For $n = 1$, we find in the limit $s \to 0$:

$$\begin{aligned}f_1(s) \simeq\; &A_1^0(r,r_0,\kappa) + A_1^1(r,r_0,\kappa)\,s^1\\ &+B_1^1(r,r_0,\kappa)\,s^1\ln(s) + A_1^2(r,r_0,\kappa)\,s^2\\ &+B_1^2(r,r_0,\kappa)\,s^2\ln(s)\\ &+C_1^2(r,r_0,\kappa)\,s^2\ln^2(s) + \mathcal{O}(s^3).\end{aligned} \tag{B4}$$

For $n = 2$, we find in the limit $s \to 0$:

$$\begin{aligned}f_2(s) \simeq\; &A_2^0(r,r_0,\kappa) + A_2^1(r,r_0,\kappa)\,s^1 + A_2^2(r,r_0,\kappa)\,s^2\\ &+B_2^2(r,r_0,\kappa)\,s^2\ln(s) + \mathcal{O}(s^3).\end{aligned} \tag{B5}$$

The coefficients X_n^m follow the scheme that the subscript n indicates the mode and the superscript m corresponds to the power of the Laplace variable s. The alphabetical order of the coefficients indicates the power of the multiplying term $\ln(s)$, i.e., the coefficient A is used for $\ln^0(s)$, the coefficient B for $\ln^1(s)$, etc. Note that it is sufficient to consider $n \geqslant 0$ as the modified Bessel functions are symmetric under $n \to -n$. The exact analytical form of the coefficients is lengthy and not of direct importance for us.

From an iterative expansion for consecutive modes, we observe that the first appearance of the multiplicative term $\ln(s)$ in the expansion occurs at $m = n$. It is this term $\propto B_n^n(r, r_0, \kappa)\, s^n \ln(s)$ which gives rise to the leading order behavior of f_n, in time as $\mathscr{L}^{-1}(s^n \ln(s)) \simeq \tau^{-(n+1)}$ for $n \in \mathbb{N}_0$ [101]. The dominating behavior for the mode p_n as $\tau \to \infty$ is thus given by the scaling $\simeq \tau^{-(n+1)}$ in Eq. (B1) and in f_n.

The long-time behavior of the individual Fourier coefficients a_n and b_n follows the same scheme as $a_n = \mathrm{Re}\{p_n\}$ and $b_n = \mathrm{Im}\{p_n\}$. In the main text, we specifically investigate $a_1(r, \tau|r_0)$ and $b_1(r, \tau|r_0)$ as they define the polarization angle $\gamma(\tau) = \arctan(b_1(1, \tau|1)/a_1(1, \tau|1))$, according to Eq. (19). We find that for $s \to 0$:

$$\mathscr{L}\{a_1\}(1, s|1) \simeq A^0_{1,a}(\kappa) + A^1_{1,a}(\kappa)\, s + B^1_{1,a}(\kappa)\, s \ln(s) + \mathcal{O}(s^2), \tag{B6}$$

$$\mathscr{L}\{b_1\}(1, s|1) \simeq A^0_{1,b}(\kappa) + A^1_{1,b}(\kappa)\, s + B^1_{1,b}(\kappa)\, s \ln(s) + \mathcal{O}(s^2). \tag{B7}$$

Therefore, at $\tau \to \infty$, the leading order behavior is given by $a_1, b_1 \simeq \tau^{-2}$ [101]. The prefactors are given by $B^1_{1,a}(\kappa) = (1-\kappa^2)/(4(1+\kappa^2)^2)$ and $B^1_{1,b}(\kappa) = -\kappa/(2(1+\kappa^2)^2)$, which leads to the asymptotic angle:

$$\gamma_\infty = \lim_{\tau\to\infty} \gamma(\tau) = \arctan\left(\frac{B^1_{1,b}(\kappa)}{B^1_{1,a}(\kappa)}\right) = -\arctan\left(\frac{2\kappa}{1-\kappa^2}\right). \tag{B8}$$

Note that we use the arctan2$(\cdot)$-function for numerical evaluation as $\tan(\gamma_\infty) \to \infty$ as $\kappa \to 1$.

APPENDIX C: INTEGRAL EXPRESSIONS

To evaluate the integrals in Eq. (A9), we use the following tabulated integrals, listed in this Appendix.

Following Arfken and Weber's *Mathematical Methods for Physicists* [102], we find that Bessel functions of (integer) order n, J_n, obey the integral relation

$$\int_0^\infty \mathrm{d}u\, u\, J_n(au)\, J_n(bu) = \frac{\delta(a-b)}{a}, \tag{C1}$$

valid for $n > -1/2$ and a, b some real-valued constants [102, 11.59].

To evaluate the remaining integrals of Bessel functions, we draw on Gradshteyn and Ryzhik's *Table of Integrals, Series and Products* [103] and Abramowitz and Stegun's *Handbook of Mathematical Functions* [32]. The first relevant integral is

$$\int_0^\infty \mathrm{d}u\, \frac{u\, J_n(au)J_n(bu)}{u^2+c^2} = \begin{cases} I_n(ac)\, K_n(bc), & 0 < a < b \\ I_n(ac)\, K_n(ac), & 0 < a = b \\ I_n(bc)\, K_n(ac), & 0 < b < a, \end{cases} \tag{C2}$$

which is valid for $n > -1$ and $c > 0$ [103, 6.541, 6.535]. Here I_n, K_n are the modified Bessel functions of the first and second kinds, and a, b, c some (real-valued) constants. Together with the symmetry relations for the order of Bessel functions, $J_{-n}(x) = (-1)^n J_n(x)$, $I_{-n}(x) = I_n(x)$ and $K_{-n}(x) = K_n(x)$ [32, 9.1.5, 9.6.6], Eq. (C2) can be further extended to the cases of $n < 0$.

The second relevant integral is $\int_0^\infty \mathrm{d}u\;\; u^2 J_n(au) J_n'(bu)/(u^2+c^2)$ for some real-valued, positive constants a, b, c. Using $\mathrm{d}J_n(x)/\mathrm{d}x = (J_{n-1}(x) - J_{n+1}(x))/2$ [103], [8.471], the integral can be evaluated as

$$\int_0^\infty \mathrm{d}u\, \frac{u^2\, J_{n_1}(au)J_{n_2}(bu)}{u^2+c^2} = \begin{cases} (-1)^{\alpha_+}\, c\, I_{n_1}(ac)\, K_{n_2}(bc), & 0 < a < b \\ (-1)^{\alpha_-}\, c\, I_{n_2}(bc)\, K_{n_1}(ac), & 0 < b < a, \end{cases} \tag{C3}$$

which is valid for $n_1, n_2 > -1$ [103, 6.577]. The exponents are $\alpha_\pm = (1 \pm (n_1 - n_2))/2 \in \mathbb{N}_0$. We use Eq. (C3) for the case of $n_1 = n \pm 1$ and $n_2 = n$. By relying on the symmetry relations for the orders of the Bessel functions, we can extend Eq. (C3) again to the cases of $n_1 = -(n \pm 1)$ and $n_2 = -n$. Thus, together with [103, 8.471], we find

$$\int_0^\infty \mathrm{d}u\, \frac{u^2\, J_n(au)J_n'(bu)}{u^2+c^2} = \begin{cases} \frac{n}{b} K_n(bc)\, I_n(ac), & 0 < a < b \\ c\, K_n(ac)\, I_n'(bc), & 0 < b < a, \end{cases} \tag{C4}$$

valid for all $n \in \mathbb{Z}$, but limited to $b \neq a$.

To generalize Eq. (C4) to the case of $b = a > 0$, which cannot be found in Ref. [103], we generalize the relation, which was already derived in Ref. [9] for $n = 1$ to an arbitrary $n \in \mathbb{Z}$. By using the indefinite integral $\int \mathrm{d}u\, J_n(u) J_n'(u) = J_n^2(u)/2 + \text{const}$, we can partially integrate and find an integral of the form of Eq. (C2) which, using the Wronskian $\mathcal{W}[I_n(x), K_n(x)] = I_n(x)\, K_{n+1}(x) + I_{n+1}(x)\, K_n(x) = 1/x$ [32, 9.1.15], gives

$$\int_0^\infty \mathrm{d}u\, \frac{u^2\, J_n(au)J_n'(bu)}{u^2+c^2} = \begin{cases} \frac{n}{b} K_n(bc)\, I_n(ac), & 0 < a < b \\ c\, K_n(ac)\, I_n'(ac) - \frac{1}{2a}, & 0 < a = b \\ c\, K_n(ac)\, I_n'(bc), & 0 < b < a. \end{cases} \tag{C5}$$

To prove the normalization of the zeroth order mode $\int \mathrm{d}\mathbf{x}\;\; p_0 = 1$, Eq. (9) in the main text, we transform the integrals over I_0 and K_0 into higher order Bessel functions evaluated at the boundaries $r = 1$ and $r \to \infty$ by using the recursion relations [103, 8.486]

$$\left(\frac{1}{u}\frac{\mathrm{d}}{\mathrm{d}u}\right)^m (u^n\, I_n(u)) = u^{n-m}\, I_{n-m}(u), \tag{C6}$$

and

$$\left(\frac{1}{u}\frac{\mathrm{d}}{\mathrm{d}u}\right)^m (u^n\, K_n(u)) = (-1)^m\, u^{n-m}\, K_{n-m}(u), \tag{C7}$$

which specifically imply for $m = 1$ and $n = 1$: $u\, I_0(u) = \mathrm{d}/\mathrm{d}u\, (u\, I_1(u))$ and $u\, K_0(u) = -\mathrm{d}/\mathrm{d}u\, (u\, K_1(u))$. The integrals over $I_0(u)$ and $K_0(u)$ thus turn into evaluating $u\, I_1(u)$ and $u\, K_1(u)$ at the integration bounds and the upper bound vanishes by the asymptotic expansion of $\lim_{u\to\infty} uK_n(u) \sim \lim_{u\to\infty} u \exp(-u) = 0$ [103, 8.451]. The remaining expressions from the lower bounds are evaluated by using the Wronskian $\mathcal{W}[I_n(x), K_n(x)]$ to be exactly 1, thus proving the normalization of the PDF to be given solely by the zeroth-order mode $\int d\mathbf{x}\, p(\tau) = \int d\mathbf{x}\, p_0(\tau) = 1$.

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