Editors' Suggestion

Tracer Diffusion beyond Gaussian Behavior: Explicit Results for General Single-File Systems

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Single-file systems, in which particles diffuse in narrow channels while not overtaking each other, is a fundamental model for the tracer subdiffusion observed in confined geometries, such as in zeolites or carbon nanotubes. Twenty years ago, the mean squared displacement of a tracer was determined at large times, for any diffusive single-file system. Since then, for a general single-file system, even the determination of the fourth cumulant, which probes the deviation from Gaussianity, has remained an open question. Here, we fill this gap and provide an explicit formula for the fourth cumulant of an arbitrary single-file system. Our approach also allows us to quantify the perturbation induced by the tracer on its environment, encoded in the correlation profiles. These explicit results constitute a first step towards obtaining a closed equation for the correlation profiles for arbitrary single-file systems.

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Introduction.—The investigation of the dynamic properties of interacting particle systems in nonequilibrium settings has been a prominent area of research in the last decades [1–5]. Among them, single-file diffusion, where particles diffuse in narrow channels and cannot overtake each other, plays an important role. Such geometrical constraint results in a subdiffusive behavior of the mean square displacement (MSD) of a tracer particle $\langle X_T^2 \rangle \propto T^{1/2}$ [6–8]. This theoretical prediction has been verified across various scales, ranging from the diffusion of molecules within zeolites [9] to the movement of colloids in confined narrow trenches [10,11].

Beyond the scaling behavior of the MSD, the prefactor, which contains the dependence on the mean density $\bar{\rho}$ of surrounding particles, has first been computed explicitly for specific models: for instance, for reflecting Brownian particles [6], and later for the simple exclusion process (SEP) [8]. Twenty years ago, Kollmann extended the result to any single-file system and showed that the MSD of a tracer can be written at large times in terms of macroscopic properties of the system as [12]

$$\langle X_T^2 \rangle \underset{T\to\infty}{\simeq} \frac{\sigma(\bar{\rho})}{\bar{\rho}^2 \sqrt{\pi D(\bar{\rho})}} \sqrt{T}. \tag{1}$$

In this expression, D is the collective diffusion coefficient, which controls the relaxation of the density, and σ the mobility, which governs the fluctuations of current [13]. Note that in all these results, as well as throughout this article, annealed (equilibrium) initial conditions have been adopted.

Recently, there has been a growing interest in the characterization of the statistical properties of various observables, and in particular the position of a tracer beyond the MSD (also known as second cumulant) [14–28]. This is typically done by studying higher order cumulants or, equivalently, the atypical fluctuations using a large deviation framework. These methods give access to finer properties of these observables, beyond the typical Gaussian behavior encoded in the MSD.

More precisely, the higher order cumulants, or large deviations, of the position of the tracer have only been determined for a few specific models. For reflecting Brownian particles the cumulants are known [14,18–20]. For the SEP, all the cumulants have first been determined in the high density limit [29]. At arbitrary density, the computation of the fourth cumulant was first achieved [14,19] and later all the cumulants have been determined [21,22]. However, for a general single-file system, even the determination of the fourth cumulant, which probes the deviation from Gaussianity, has remained an open question since the work of Kollmann [12].

Here, we fill this gap and provide an explicit formula for the fourth cumulant for an *arbitrary* single-file system. We stress that, unlike previous results, which were obtained for integrable models (essentially the SEP and those mappable on it [30]), using tools like Bethe ansatz or inverse scattering technique [14,21,22,25–28], our expression holds for any model, whether integrable or not. Furthermore, beyond quantifying the deviation from Gaussian behavior, our approach also allows us to quantify the perturbation induced by the tracer on its environment, encoded in the correlation profiles [30]. We show that these profiles exhibit a nonanalytic behavior for nonintegrable models.

Macroscopic fluctuation theory.—Our starting point to study the position of a tracer in a single-file system relies on the macroscopic fluctuation theory (MFT) [5]. At large scales (long times and large distances), the MFT gives the probability to observe a fluctuation of the density profile $\rho(x,t)$ of a diffusive system in terms of the two transport coefficients $D(\rho)$ and $\sigma(\rho)$ [3,5,31], for which explicit expressions have been obtained for several models. For instance, for the SEP, $D(\rho)=1$ and $\sigma(\rho)=2\rho(1-\rho)$. Other paradigmatic models include zero range processes (ZRP) [2,32], the Kipnis-Marchioro-Presutti (KMP) model [33], the Katz-Lebowitz-Spohn (KLS) model [34,35], and models with more realistic pairwise interactions such as Brownian particles with Weeks-Chandler-Anderson (WCA) potential or dipole-dipole interactions, as involved in experimental realisations of colloids confined in 1D [10]. The MFT is a powerful approach, in which all the microscopic details of the model are replaced by the two transport coefficients D and σ only. Note, however, that one typically ends up with nonlinear partial differential equations for the time evolution of the density. Solving these equations is a challenging task of current intense activity that has recently led to important achievements [25–28,36–38].

MFT has proved to be useful to study a wide range of observables, including even a microscopic observable such as the position X_T of a single tracer, for which different approaches have been devised. (i) The first one relies on expressing X_T as a functional of the density of particles $X_T=X_T[\rho]$ [14,19]. This can be done since the tracer effectively "cuts" the system into two parts, in which the number of particles is conserved due to the noncrossing condition. In practice, this is, however, tricky due to the emergence of discontinuities in the density profiles at the position of the tracer [14,19]. (ii) A second approach that circumvents this issue consists of introducing a generalized current [21,22] defined as the number of particles crossing a fictitious moving wall. The tracer is then located at the position where this current vanishes, again due to the noncrossing condition. (iii) An alternative method, which we apply here, consists of using a mapping between different single-file systems, in which the position X_T of the tracer in the original model is mapped onto (the opposite of) the integrated current $\tilde{Q}_T$ through the origin in a dual model (see Fig. 1). More precisely, the current is defined from the density $\tilde{\rho}(x,t)$ in the dual model as

$$\tilde{Q}_T=\int_0^\infty[\tilde{\rho}(x,T)-\tilde{\rho}(x,0)]\mathrm{d}x, \tag{2}$$

and the transport coefficients $\tilde{D}$ and $\tilde{\sigma}$ of the dual model are written in terms of those of the original model as [30]

$$\tilde{D}(\rho)=\frac{1}{\rho^2}D\left(\frac{1}{\rho}\right),\qquad \tilde{\sigma}(\rho)=\rho\,\sigma\left(\frac{1}{\rho}\right). \tag{3}$$

The main benefit of this approach is that, with MFT, the study of the current $\tilde{Q}_T$ is generally simpler than that of X_T [16]. However, this is often at the cost of handling more

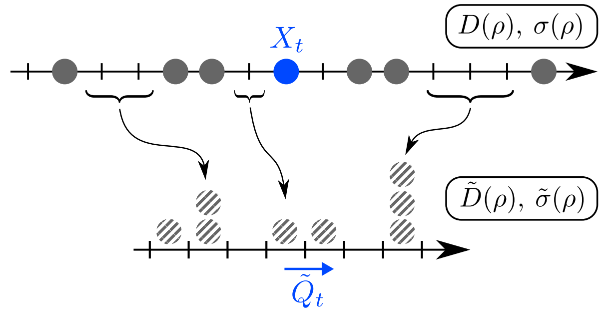

FIG. 1. An example of mapping between two single-file systems. The SEP (top) is mapped onto a zero range process (below). This well-known mapping holds at the microscopic level: the empty sites of the SEP becomes the particles of the ZRP [2], while the position X_t of the tracer in the SEP is mapped onto the integrated current through the origin $\tilde{Q}_t$ in the ZRP. At the macroscopic level, such a mapping holds for any single-file system [30]: the tracer in a system with transport coefficient $D(\rho)$ and $\sigma(\rho)$ is mapped onto the current in a system with $\tilde{D}(\rho)$ and $\tilde{\sigma}(\rho)$ given by (3).

complex transport coefficients [for instance, the constant $D(\rho)=1$ of the SEP is mapped onto the nonconstant $\tilde{D}(\rho)=1/\rho^2$]. Here, since we aim to study general single-file systems, and thus arbitrary D and σ, this is not a limitation and we use this latter approach.

The main steps of the computation of the fourth cumulant of X_T for general D and σ are as follows (see Supplemental Material for details [39]). First, we use the mapping described above that allows us to obtain the cumulants of X_T from those of $\tilde{Q}_T$ in the dual model, with $\tilde{D}$ and $\tilde{\sigma}$ given by (3). Explicitly, the cumulant generating functions are related by [30]

$$\hat{\psi}(\lambda)=\lim_{T\to\infty}\frac{1}{\sqrt{T}}\ln\langle e^{\lambda X_T}\rangle=\lim_{T\to\infty}\frac{1}{\sqrt{T}}\ln\langle e^{-\lambda\tilde{Q}_T}\rangle =\kappa_2\frac{\lambda^2}{2}+\kappa_4\frac{\lambda^4}{4!}+\cdots, \tag{4}$$

with κ_n the nth cumulant of the position of the tracer. Note that the odd order cumulants vanish by symmetry.

Second, we determine the first cumulants of $\tilde{Q}_T$ using the standard MFT formalism [5,16]. Explicitly, this requires solving the MFT equations [16]

$$\partial_t\tilde{q}=\partial_x[\tilde{D}(\tilde{q})\partial_x\tilde{q}]-\partial_x[\tilde{\sigma}(\tilde{q})\partial_x\tilde{p}], \tag{5}$$

$$\partial_t\tilde{p}=-\tilde{D}(\tilde{q})\partial_x^2\tilde{p}-\frac{1}{2}\tilde{\sigma}'(\tilde{q})(\partial_x\tilde{p})^2, \tag{6}$$

$$\tilde{p}(x,T)=-\lambda\Theta(x), \tag{7}$$

$$\tilde{p}(x,0)=-\lambda\Theta(x)+\int_{\tilde{\bar{\rho}}}^{\tilde{q}(x,0)}\frac{2\tilde{D}(r)}{\tilde{\sigma}(r)}\mathrm{d}r, \tag{8}$$

where $\tilde{\bar{\rho}} = 1/\bar{\rho}$ is the mean density in the dual model. The function $\tilde{q}(x,t)$ is the typical realization of the time evolution of the density $\tilde{\rho}(x,t)$ that yields a given value of the current $\tilde{Q}_T$ and fully controls the dynamics at large times T. $\tilde{p}(x,t)$ is a Lagrange multiplier that ensures the conservation of the number of particles at every point in space and time. The cumulants are then deduced from the solution of these equations by $\mathrm{d}\hat{\psi}/\mathrm{d}\lambda = -\tilde{Q}_T/\sqrt{T}$, where here $\tilde{Q}_T$ is given by (2) with $\tilde{\rho}(x,t)$ replaced by its typical fluctuation $\tilde{q}(x,t)$. Third, we expand $\tilde{q}$ and $\tilde{p}$ in powers of λ and solve (5)–(8) order by order, up to order 3 included to compute κ_4. The practical resolution requires us to solve diffusion equations with source terms of increasing complexity with the order in λ. Explicit results can be obtained by isolating the dependence of the source terms on $\tilde{D}(\rho)$, $\tilde{\sigma}(\rho)$ and their derivatives, and then relying on a combination of changes of functions and successive integrations by parts.

Results.—Lengthy calculations, given in Supplemental Material [39], finally provide an explicit formula for the fourth cumulant of the position X_t of a tracer for *any* $D(\rho)$ and $\sigma(\rho)$,

$$\kappa_4 = \frac{3\sigma(\bar{\rho})^3(\bar{\rho}D'(\bar{\rho}) + D(\bar{\rho}))}{\pi^{3/2}\bar{\rho}^6 D(\bar{\rho})^{7/2}} - \frac{\sigma(\bar{\rho})\big(\sigma(\bar{\rho})\sigma'(\bar{\rho})(\bar{\rho}D'(\bar{\rho}) + 4D(\bar{\rho})) + 2\sigma(\bar{\rho})^2 D'(\bar{\rho}) - \bar{\rho}D(\bar{\rho})\sigma'(\bar{\rho})^2\big)}{4\sqrt{\pi}\bar{\rho}^5 D(\bar{\rho})^{7/2}}$$
$$+ \frac{3\sigma(\bar{\rho})^3\big(D'(\bar{\rho})^2 - D(\bar{\rho})D''(\bar{\rho})\big)}{8\sqrt{\pi}\bar{\rho}^4 D(\bar{\rho})^{9/2}} + \frac{3\sigma(\bar{\rho})^3\big(2D(\bar{\rho})D''(\bar{\rho}) - D'(\bar{\rho})^2\big)}{8\pi^{3/2}\bar{\rho}^4 D(\bar{\rho})^{9/2}} + \frac{(3\sqrt{2}-4)\sigma(\bar{\rho})^2\sigma''(\bar{\rho})}{8\sqrt{\pi}\bar{\rho}^4 D(\bar{\rho})^{5/2}}$$
$$+ \frac{3\big(\sqrt{2}\pi - 2\sqrt{3}\big)\sigma(\bar{\rho})^3\big(2D(\bar{\rho})D''(\bar{\rho}) - 3D'(\bar{\rho})^2\big)}{16\pi^{3/2}\bar{\rho}^4 D(\bar{\rho})^{9/2}}. \tag{9}$$

This result constitutes the first step beyond the second cumulant (1) for any single-file system and provides a quantitative measure of the deviation from Gaussian behavior.

Several comments are in order. (i) The expression (9) encompasses all previously known results on fourth cumulants for specific single-file systems, for instance for reflecting Brownian particles [14,18–20], for the SEP [14,19] and models that can be related to the SEP, such as the KMP model, or the random average process [30]. These previous results were obtained for models that can be mapped, at least at the macroscopic level, to the SEP [30]. For all these models, the last term in (9) vanishes. (ii) More precisely, the last term in Eq. (9) vanishes if and only if $D(\rho) = 1/(a+b\rho)^2$, where a and b are constants. This is the class of diffusion coefficients corresponding to models that can be mapped onto a constant diffusion coefficient (see Supplemental Material [39] for details). In the general case of a model that cannot be mapped onto a constant $D(\rho)$, as for paradigmatic models like the KLS model or ZRP, or models with more realistic interactions (like Brownian particles with WCA or dipole-dipole interaction) this last term matters. Note that this term is the only one that involves a $\sqrt{3}$. (iii) Finally, the result (9), also gives the fourth cumulant of the current $\tilde{Q}_T$, in the dual model with $\tilde{D}$ and $\tilde{\sigma}$. Writing this expression in terms of these dual transport coefficients thanks to (3), gives this fourth cumulant of $\tilde{Q}_T$ for a general single-file system [see Eq. (S60) of the Supplemental Material].

Beyond the cumulants: Correlation profiles.—On top of the cumulants, our approach gives access to the response of the bath of surrounding particles to the perturbation induced by the displacement of the tracer. This response is described by the bath-tracer correlation profile introduced in [30], defined as

$$w(x,T) \equiv \frac{\langle \rho(X_T + x, T)e^{\lambda X_T}\rangle}{\langle e^{\lambda X_T}\rangle} = \sum_{n=0}^{\infty} \frac{\lambda^n}{n!}\langle \rho(X_T + x, T)X_T^n\rangle_c, \tag{10}$$

which generates all the connected correlation functions $\langle \rho(X_T + x, T)X_T^n\rangle_c$ between the density field and the displacement of the tracer. At large times T, these profiles display a diffusive scaling behavior $w(x,T) \simeq \Phi(z = x/\sqrt{T})$ [30,45,46]. The scaling function Φ, which thus contains the full spatial structure of the bath-tracer correlations in the long time limit, has been determined explicitly for the SEP and for models that can be related to it [30,45,46]. Here, for arbitrary $D(\rho)$ and $\sigma(\rho)$, Φ is derived from the solution of the MFT equation $\tilde{q}(x,T)$ at final time (in the dual model with $\tilde{D}$ and $\tilde{\sigma}$) and mapped back to the original model with D and σ. The details of this mapping and the expressions of the correlation profiles up to order 3 are given explicitly in Supplemental Material [39], Eqs. (S67)–(S69).

In parallel of this explicit calculation, an important question concerns the existence of a closed equation satisfied by Φ. Indeed, in the case of the SEP, these profiles have been shown to satisfy a simple exact closed equation [45,46]. This result has allowed the determination of all the correlation profiles (10). Since the publication of this equation [45], several works have obtained exact results for different observables for specific models of single-file systems [25–28], which can all be recast into a similar closed equation, making it a promising tool to investigate various questions in single-file diffusion and beyond.

We investigate the possibility to obtain such an equation for Φ by following the approach of [45,46]. It is shown in Supplemental Material [39] that in fact,

$$\begin{aligned}\partial_z(D(\Phi)\partial_z\Phi)+\frac{1}{2}(z+\xi)\partial_z\Phi=&\frac{\lambda\sigma''(\bar\rho)}{4\bar\rho}\int_0^\infty\Phi'(z+u)\Phi''(-u)\mathrm{d}u\\&+\left(\frac{\lambda\sigma(\bar\rho)D'(\bar\rho)}{8\sqrt{\pi}\bar\rho D(\bar\rho)^{3/2}}-\frac{\lambda^2\sigma(\bar\rho)D'(\bar\rho)(\bar\rho\sigma'(\bar\rho)-2\sigma(\bar\rho))}{32\sqrt{\pi}\bar\rho^3 D(\bar\rho)^{5/2}}+\frac{\lambda^2\sigma(\bar\rho)^2D'(\bar\rho)^2}{64\sqrt{\pi}\bar\rho^2D(\bar\rho)^{7/2}}\right)\Phi'(z)\\&-\frac{\lambda^3\sigma(\bar\rho)^3(2D(\bar\rho)D''(\bar\rho)-3D'(\bar\rho)^2)}{512\bar\rho^3D(\bar\rho)^5}\left(ye^{-\frac{y^2}{2}}\sqrt{\frac{2}{\pi}}\mathrm{erfc}\left(\frac{y}{\sqrt{2}}\right)+\frac{2}{\pi^{3/2}}\partial_y\int_0^1\frac{\mathrm{d}t}{\sqrt{1+2t}}e^{-\frac{(1+t)y^2}{(1-t)(1+2t)}}\right)\\&+\mathcal{O}(\lambda^4),\end{aligned}\tag{11}$$

where $\xi=\mathrm{d}\hat\psi/\mathrm{d}\lambda$, and we have denoted $y=z/[2\sqrt{D(\bar\rho)}]$ to simplify the notations. In the case of the SEP, corresponding to constant D, only the first term on the right-hand side of Eq. (11) remains. We have written this term as a convolution, instead of its explicit expression, because it was the key step in [45,46] that allowed us to find a closed form for the equation. Similarly, we have realized that the second term in (11) can be expressed in terms of Φ' only. The only remaining task to obtain a closed equation is to rewrite the last term in (11) in terms of Φ. Anyhow, Eq. (11) constitutes a first step towards obtaining a closed equation for Φ for arbitrary $D(\rho)$ and $\sigma(\rho)$.

On top of its intrinsic interest, Eq. (11) allows us, as we now discuss, to provide (i) a signature of the nonintegrable nature of general single-file models and (ii) a shortcut to obtain the cumulants of X_t.

Relation with integrability.—First, it can be shown that the last term in (11) is directly associated to the $\sqrt{3}$ in the expression of κ_4 (9), as discussed above. In particular, both vanish for the specific choice $D(\rho)=1/(a+b\rho)^2$, which is the class of diffusion coefficients for which the nonlinear heat equation is integrable [47]. Second, this term displays a nonanalytic behavior with respect to the distance to the tracer, with a logarithmic singularity $\sim y\ln y$ as $y\to 0$. It shows that this term introduces a completely new class of functions, compared to the case of the SEP (and related models) in which only analytic functions were present [45,46]. Note that such behavior was also observed in the correlation profile of a *driven* tracer in the SEP [48], a model which is expected to be not integrable. All these points indicate that the presence of the last term in (11) is a signature of the nonintegrability of a single-file model with arbitrary $D(\rho)$ and $\sigma(\rho)$.

A conjecture for a shortcut to the cumulants.—First of all, note that in the case of the SEP, boundary conditions for $\Phi(0^\pm)$ and $\Phi'(0^\pm)$ have been obtained from microscopic considerations [30,45,46]. These relations are very useful, since together with the bulk equation (11) written in the specific case of the SEP, they allow us to fully determine the profiles and the cumulants without solving the MFT equations (5)–(8). Several of these relations have recently been extended to any single-file system, and take a simple physical form [38]

$$P[\Phi(0^+)]-P[\Phi(0^-)]=\lambda,\qquad [\partial_z\mu(\Phi)]_{0^-}^{0^+}=0,\tag{12}$$

where $P(\rho)$ is the pressure, and $\mu(\rho)$ the chemical potential, given by $P'(\rho)=\rho\mu'(\rho)$ and $\mu'(\rho)=2D(\rho)/\sigma(\rho)$. We have used the notation $[f]_a^b=f(b)-f(a)$. The remaining boundary condition, obtained for the SEP in [30,45,46], which has not yet been generalized to an arbitrary single-file system [38], is a key relation allowing to obtain $\hat\psi$ directly from $\Phi(0^\pm)$ and $\Phi'(0^\pm)$ [which are fully determined by the bulk equation (11) and the boundary conditions (12) completed by $\Phi(\pm\infty)=\bar\rho$], instead of computing the integral (2), which is usually a difficult task. We conjecture that, for arbitrary D and σ, this last relation takes the form

$$\hat\psi=-2\partial_z\mu(\Phi)|_{z=0}\int_{\Phi(0^-)}^{\Phi(0^+)}D(r)\mathrm{d}r.\tag{13}$$

This conjecture is supported by the following points. (i) For $D(\rho)=1$ and $\sigma(\rho)=2\rho(1-\rho)$, it reduces to the expression obtained for the SEP [30,45,46]. (ii) Furthermore, from our above results on the profiles Φ and the fourth cumulant κ_4, we can check that this relation holds up to order 4 in λ included. (iii) Finally, Eq. (13) is invariant under the duality mapping (3) (see Supplemental Material [39]).

Conclusion.—We have considered tracer diffusion (as well as the current of particles) in general single-file systems at large times. We have determined an explicit expression for the fourth cumulant of X_t, which constitutes the first extension of the result of Kollmann on the second cumulant [12], for any $D(\rho)$ and $\sigma(\rho)$ and provides a quantitative measure of the deviation from Gaussian behavior. On top of the cumulants of X_t, we have obtained the response of the bath of surrounding particles to the displacement of the tracer by determining the full spatial structure of the bath-tracer correlation profiles (up to order 4). These explicit results, which hold for any system, allowed us to pinpoint the effect of nonintegrability, both on the cumulants and the correlation profiles. This work constitutes a first step towards obtaining a closed equation for the correlation profiles for arbitrary $D(\rho)$ and $\sigma(\rho)$.

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