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Langevin formulation for single-file diffusion

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We introduce a stochastic equation for the microscopic motion of a tagged particle in the single-file model. This equation provides a compact representation of several of the system's properties such as fluctuation-dissipation and linear-response relations, achieved by means of a diffusion noise approach. Most importantly, the proposed Langevin equation reproduces quantitatively the three temporal regimes and the corresponding time scales: ballistic, diffusive, and subdiffusive.

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I. INTRODUCTION

Since its introduction in 1965 due to Harris' pioneering work [1], the single-file (SF) model has attracted more and more interest among the scientific community. Introduced first in the mathematical physics literature as an interesting though somewhat exotic topic, it has inspired over the last 40 years a large body of profound theoretical studies [2] and detailed numerical investigations, including extensive Monte Carlo and molecular dynamics simulations [3].

The motivations for the long-standing interest in this topic reside, on the one hand, in its analytical tractability and, on the other hand, in its effectiveness as a description of diffusion phenomena in real *quasi*-one-dimensional systems. As a matter of fact, since the direct observation and manipulation of nanoscopic systems have exponentially evolved in the last decade, models suitable to account for the single-particle diffusional mechanisms in constrained flow geometries have been the subject of increasing attention. Remarkably, among these, the SF model holds a preeminent position, since it correctly reproduces transport properties in a large category of quasi-one-dimensional systems, where each particle is free to diffuse against its neighbors, but is forbidden to overcome them [4]. Transport processes of this type may be observed in nanoporous materials [5] and in collective motion of ions through biological channels and membranes [6] as well as in nanodevices and cellular flows [7]

Along a mathematical line, the SF model is perhaps the simplest interacting one-dimensional gas one can consider: it consists of N unit-mass particles constrained to move along a line following a given dynamics. As the particles' interaction is purely hard core, no mutual exchanges of the diffusants are allowed; i.e., they retain their ordering over time (*single-filing condition*). In spite of the intricacies of its mathematical derivation, the long-time behavior of the single-file dispersion relation can be cast in the following suggestive form [2]:

$$\delta x^2(t) = \frac{\langle |X| \rangle}{\rho}, \tag{1}$$

where $\rho = \frac{N}{L}$ $(N \to \infty, L \to \infty)$ is the file's density and $\langle |X| \rangle$ is the absolute displacement of a noninteracting particle. If the free-particle dynamics is characterized by a diffusivity $D = k_B T/\gamma$, then relation (1) takes the form [8]

$$\delta x^2(t) = 2\sqrt{\frac{Dt}{\pi \rho^2}}. \tag{2}$$

The predicted subdiffusive behavior has been reproduced experimentally in colloidal particles systems [9] and observed in molecular sieves (zeolites) [10]. We remark that the subdiffusive behavior represented by Eq. (2) will eventually be replaced by regular diffusion if, for instance, the particles are allowed to overtake each other [11], if there is only a finite number of particles [12], or if the particles move in a ring [13].

In Ref. [14] it was pointed out that the subdiffusive regime of a SF tagged particle occurs on the score of long-ranged anticorrelations of its velocity and/or of the jump's statistics of the collisional mechanism underlying its dynamics. Beside these persistent memory effects, different mathematical derivations agree with the fact that asymptotically the tagged particle's probability distribution must be Gaussian with a variance growing in time according to (2) [2]. Together, these properties contrast with the continuous-time random-walk (CTRW) scheme and its corresponding Fokker-Planck representation, for which a stretched Gaussian solution has to be expected (see Ref. [15] and references therein). Furthermore, several subdiffusive systems in nature share the property of Gaussianicity with the SF model—e.g., a monomer in a one-dimensional phantom polymer [16], the "translocation coordinate" of a two-dimensional Rouse chain through a hole [17], a tagged monomer in an Edward-Wilkinson chain [18], de Gennes' defects along a polymer during its reptation [19], and solitons in the sine-Gordon chain [20].

In this paper we address the question of the microscopic effective description of the stochastic, anomalous motion of

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the tagged particle. Our aim is to extend the valuable Langevin approach, valid for the case of a diffusive Brownian walker, to the subdiffusive dynamics of a SF particle. We anticipate here that the generalized Langevin equation (GLE) [21,22] provides the ideal theoretical tool for such a goal, incorporating all the statistical properties enjoyed by the particle. Within this framework the non-Markovian memory effects are achieved by means of a power-law damping kernel [23], which is simply added algebraically to the instantaneous friction of the surroundings. We note here that the GLE has recently been successfully used to describe several physical phenomena and market flows [24].

The article is organized as follows: in Sec. II we study the density profile dynamics by means of a diffusion-noise approach and we connect the file's density fluctuations to the motion of a tagged particle. In Sec. III we introduce the GLE and we show the accuracy of the Langevin description by means of extensive molecular dynamic simulations.

II. DIFFUSION-NOISE APPROACH

We start by considering the file density dynamics. As stated in Sec. I, the system is composed of N Brownian-point-like particles, all of unit mass, moving along a ring of length L and performing a stochastic motion according to the Langevin equation (LE):

$$\dot{x}_i(t) = v_i(t),$$

$$\dot{v}_i(t) = -\gamma v_i(t) + \xi_i(t), \tag{3}$$

where $i \in [1,N]$ denotes the particle's index and the damping coefficient γ and the random noise source $\xi_i(t)$ satisfy the well-known *fluctuation-dissipation relation* (often called the second Kubo theorem [22]) $\langle \xi_i(t) \rangle \xi_j(t') \rangle = 2k_B T \gamma \delta_{i,j} \delta(t-t')$. The single-filing condition turns out to be merely the interchange of two particle labels, whenever these suffer an (elastic) collision. However, as pointed out in [25], all system properties which do not depend on particle labeling remain unchanged from those of an ideal gas—i.e., of N independent Brownian walkers.

Let us first define the file density at a point x of the line at time t as

$$\rho(x,t) = \frac{n(x,t)}{dx}, \tag{4}$$

where $n(x,t)$ refers to the number of particles in the bin $[x-\frac{dx}{2}, x+\frac{dx}{2}]$ at time t. It is straightforward to note that the quantity $\rho(x,t)$ is a *local* property of the file, independent of the relabeling of the particles due to collisions. A direct consequence of this is that the time evolution of the file profile density can be described by the diffusion-noise equation for a one-dimensional gas of N noninteracting Brownian particles [26]

$$\frac{\partial}{\partial t}\rho(x,t) = -\frac{\partial}{\partial x}J(x,t), \tag{5}$$

having a recourse to the definition of a stochastic flux $J(x,t) = -D\frac{\partial}{\partial x}\rho(x,t) + \eta(x,t)$. The noise term $\eta(x,t)$ can be shown to be Gaussian and satisfy the following properties [27]:

$$\eta(L,t) = \eta(0,t),$$

$$\langle \eta(x,t) \rangle = 0,$$

$$\langle \eta(x,t)\eta(x',t') \rangle = 2D\delta(x-x')\delta(t-t')\langle \rho(x,t) \rangle, \tag{6}$$

the first one of which refers to the conservation of the particle's number (*conserved noise*) along the segment $[0,L]$ with periodic boundary conditions. The remaining properties in (6) are required to fulfill the equations for the first two moments of $\rho(x,t)$:

$$\frac{\partial}{\partial t}\langle \rho(x,t) \rangle = D\frac{\partial^2}{\partial x^2}\langle \rho(x,t) \rangle \left[\frac{\partial}{\partial t} - D\frac{\partial^2}{\partial x_1^2} - D\frac{\partial^2}{\partial x_2^2} \right] \times \langle \rho(x_1,t)\rho(x_2,t) \rangle_C = 2D\frac{\partial}{\partial x_1}\frac{\partial}{\partial x_2}\delta(x_1-x_2) \times \langle \rho(x,t) \rangle, \tag{7}$$

where we made use of the short notation $\langle \rho(x_1,t)\rho(x_2,t) \rangle_C = \langle \rho(x_1,t)\rho(x_2,t) \rangle - \langle \rho(x_1,t) \rangle \langle \rho(x_2,t) \rangle$. As is apparent from (6), the correlation function of the noise depends upon the particular solution of the diffusion equation (7): in the following, as well as in the numerical simulations performed, we consider the case of a uniformly distributed file: namely, $\langle \rho(x,0) \rangle \equiv \langle \rho(x,t) \rangle \equiv \rho$.

The connection between the dynamics of the density over $[0,L]$ and the the motion of a tagged particle in the single-file system is achieved in the following way. Given two particle trajectories $x_1(t)$ and $x_2(t)$, the number of particles between these has to remain constant in time because of the nonoverlapping condition; this implies

$$\frac{d}{dt}\int_{x_1(t)}^{x_2(t)} \rho(x,t)dx = 0. \tag{8}$$

Performing the derivative and making use of (5), the previous relation then reads

$$v_2(t)\rho(x_2(t),t) - J(x_2(t),t) - v_1(t)\rho(x_1(t),t) + J(x_1(t),t) = 0;$$

now, both the terms must set to zero irrespective of the particle labels: we can thus write down the equation for the single-file particle as

$$v(t)\rho(x(t),t) = J(x(t),t). \tag{9}$$

We notice that, although the relation (9) just introduced is exact, it is highly nonlinear. We are thus compelled to fall back upon two approximations in order to solve it. The first approximation is to assume that the density surrounding the particle position is essentially constant: $\rho(0,t) \equiv \langle \rho(0,t) \rangle \equiv \rho$. The second approximation we put forward is to assume that the particle movements are correlated over a range equal to the displacement of the tagged particle, such that we can take the current $J(x(t),t)$ to be equal to the current at the particle initial position. Taking this to be at $x=0$, we thus have $J(x(t),t) = J(0,t)$. Notice that similar assumptions have been superimposed by Alexander and Pincus in a previous treatment of the single-file subdiffusive dynamics on a lattice [28].

With these approximations Eq. (9) gets the form

$$v(t) \simeq \frac{J(0,t)}{\rho}. \tag{10}$$

Moreover, defining the Fourier transform (and its inverse) in the space and time domains as

$$f(q,\omega) = \int_{-\infty}^{+\infty} dx\, dt f(x,t) e^{-i(qx-\omega t)},$$

$$f(x,t) = \int_{-\infty}^{+\infty} \frac{dq\, d\omega}{(2\pi)^2} f(q,\omega) e^{i(qx-\omega t)}, \tag{11}$$

Eq. (10) can be cast as

$$v(\omega) \simeq \frac{1}{\rho}\int_{-\infty}^{+\infty} \frac{dq}{2\pi} J(q,\omega),$$

which, by means of (5) and of the definition of the stochastic flux, reads

$$v(\omega) \simeq \frac{1}{\rho}\int_{-\infty}^{+\infty} \frac{dq}{2\pi} \frac{-i\omega}{q^2 D - i\omega} \eta(q,\omega). \tag{12}$$

Let us now define the quantity

$$\mu(\omega) = \frac{1}{\rho\gamma}\int_{-\infty}^{+\infty} \frac{dq}{2\pi} \frac{-i\omega}{q^2 D - i\omega}; \tag{13}$$

using (12) and the noise properties in (6), it is readily verified that the following equality holds:

$$\langle v(\omega) v(\omega')\rangle = 2k_B T\, \mathrm{Re}[\mu(\omega)] 2\pi\delta(\omega+\omega'). \tag{14}$$

Furthermore, a direct calculation of (13) gives

$$\mu(\omega) = \sqrt{\frac{2}{k_B T \gamma}} \frac{\sqrt{\omega}}{4\rho}[1-i], \tag{15}$$

which, substituted into (14) and consistent with the Wiener-Khintchine theorem, yields the asymptotic form of the velocity autocorrelation function (VAF) of a single-file particle [14]:

$$\langle v(t) v(0)\rangle = -\sqrt{\frac{k_B T}{\gamma\pi}} \frac{1}{4\rho} \frac{1}{t^{3/2}}. \tag{16}$$

In passing from Eq. (14)–(16) we implicitly adopted the convention $\langle v(t)v(0)\rangle = \langle v(-t)v(0)\rangle$, which uniquely determines the mobility $\mu(\omega)$ to be the Fourier-Laplace transform of the VAF—i.e.,

$$\mu(\omega) = \frac{\int_0^{+\infty}\langle v(t)v(0)\rangle e^{i\omega t}}{k_B T}. \tag{17}$$

Relation (17) is known as the first fluctuation-dissipation theorem or Green-Kubo relation [22]. However, for linear systems the connection between correlation functions and mobility hinges on the domain of transport processes [29]: within this framework Eqs. (12) and (13) on the one hand and their statistical counterpart (17) on the other encouraged us to explore the validity of a linear-response relation of the type

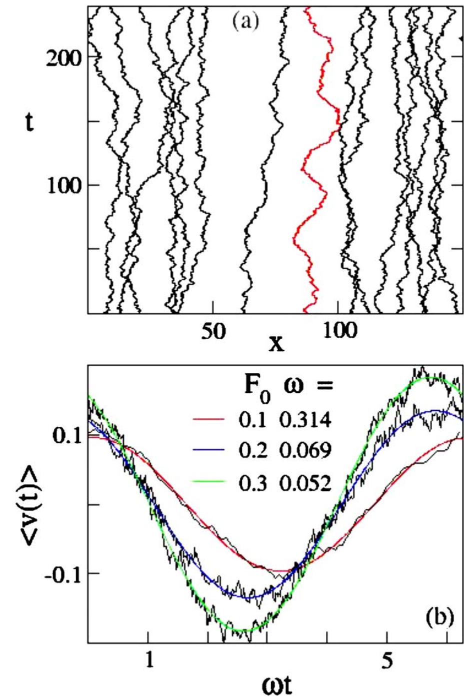

FIG. 1. (Color online) (a) Typical SF dynamics. The tagged particle (red) is subjected to the external periodic force $F(t) = F_0 \cos \omega t$, while the other ones perform the usual stochastic dynamics. (b) Linear-response relation (18). Average velocity $\langle v(t)\rangle$ vs ωt for $k_B T = 1.0$, $\gamma = 0.5$, $\rho = 0.25$, and different values of F_0 and ω. Three typical numerical curves (black lines) are fitted through relation (18), showing a linear dependence on the amplitude of the applied sinusoidal force. Averages have been taken over more than 2000 periods of each realization, performing at least 10 realizations corresponding to each ω.

$$\langle v(t)\rangle = \mathrm{Re}[\tilde{\mu}(\omega) F_0 e^{i\omega t}]. \tag{18}$$

Indeed, applying a periodic external force $F(t) = F_0 \cos \omega t$ to a tagged particle and leaving the remaining surrounding ones unaffected [see Fig. 1(a)], the response (18) has been directly measured by meaning of extensive numerical simulations. The results are shown in Figs. 1(b) and 2, where the real, the imaginary part, and the relative phase ϕ_ω of the quantity $\tilde{\mu}(\omega)$ are displayed. At first, we note as the average velocity depends linearly upon the force amplitude F_0 according to Eq. (18). Most important, the low-frequency behavior of $\tilde{\mu}(\omega)$ fully agrees with the analytical formula for the mobility $\mu(\omega)$ given in (15) (dashed line). We can interpret this result, recalling that a particle undergoes a normal diffusive behavior up to a time scale $\tau_d = \frac{1}{D\rho^2}$, which is the time needed by a couple of particles to collide with one of the neighboring diffusants [14]; correspondingly, we can assume that the tagged driven particle will *feel* the presence of the surrounding ones over frequencies smaller than the threshold $\omega_d = \frac{2\pi}{\tau_d}$ (dotted line). Conversely, for $\omega \gg \omega_d$, $\tilde{\mu}(\omega)$ coincide with the mobility of a free Brownian walker: $1/(\gamma - i\omega)$. The numeri-

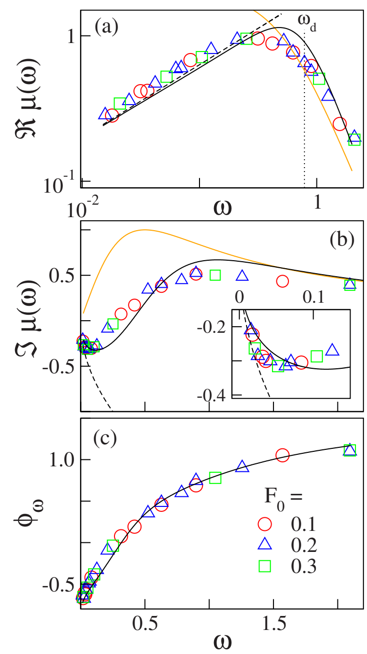

FIG. 2. (Color online). Real (a) and imaginary (b) parts and phase (c) of $\mu(\omega)$ obtained through relation (18), for different F_0 and ω. Simulation parameters are the same as in Fig. 1. The dashed lines represent the formula (15) and are seen to fit quite well the asymptotic subdiffusive behavior of the mobility [see the inset of panel (b) where the low-frequency behavior of Im $\mu(\omega)$ is blown up]. The initial diffusive behavior (orange lines) is responsible for the high-frequency regime $\omega \gg \omega_d$ [dotted line in (a)]. The GLE expression for the mobility, Eq. (45), furnishes a very good description of both regimes: solid black lines. The nonperfect agreement between theory and data is due to the small value of the ratio $\tau_d/\tau_b=4$ (see text and Table I).

cal results in Fig. 2 can thus be summarized by writing, besides relation (18), the mobility $\tilde{\mu}(\omega)$ as

$$\tilde{\mu}(\omega) \simeq \begin{cases} \dfrac{\gamma+i\omega}{\gamma^2+\omega^2}, & \omega \gg \omega_d, \\ \sqrt{\dfrac{2}{k_BT\gamma}}\dfrac{\sqrt{\omega}}{4\rho}[1-i], & \omega \ll \omega_d. \end{cases} \quad (19)$$

The numerical evidence of the effectiveness of a linear response relation allows us to rewrite Eq. (12) as

$$v(\omega) = \tilde{\mu}(\omega)\tilde{\xi}(\omega), \quad (20)$$

where the introduced noise satisfies $\langle \tilde{\xi}(t)\rangle = 0$. On the other hand, its spectrum $S_\xi(\omega)$ exhibits two different regimes according to (19):

$$S_\xi(\omega) = \int_{-\infty}^{+\infty} \langle \xi(t)\xi(0)\rangle e^{i\omega t} \simeq \begin{cases} 2k_BT\gamma, & \omega \gg \omega_d, \\ \dfrac{(2k_BT)^{3/2}\sqrt{\gamma\rho}}{\sqrt{\omega}}, & \omega \ll \omega_d. \end{cases} \quad (21)$$

The file particles surrounding the tagged one thus act as an additional bath responsible for the onset of subdiffusional behavior. The nature of this long-ranged correlations in the noise source can be easily understood in terms of the collisional interaction between the file components. Note in fact that expression (21) leads to a slowly decaying positive correlated noise $\langle \tilde{\xi}(t)\tilde{\xi}(0)\rangle \propto \frac{1}{\sqrt{t}}$: this nontrivial finding is the signature of constrained geometry systems. Indeed, although the collisions tend to tie back the particle motion, leading to the negative velocity correlations (16), the noise provides the way to maintain $\langle v(t)\rangle=0$ on time scale of the order $1/\gamma$. Another way to say this is that in a collision a particle exchanges velocity *and* noise. Anyway, this is a manifestation of the fluctuation-dissipation theorem. Furthermore, the non-Markovian power-law nature of the noise spectrum characterizes the asymptotic fractional Brownian motion (FBM) of the tagged particle [32].

We end this section stressing that, using the formalism so far developed, we can calculate the other asymptotic statistical properties of the system. In fact, it is straightforward to write down an expression for the particle's position similarly to what we did in (12) for its velocity:

$$x(\omega) \simeq \frac{1}{\rho}\int_{-\infty}^{+\infty} \frac{dq}{2\pi}\frac{\eta(q,\omega)}{q^2D - i\omega}. \quad (22)$$

For instance, making use of (22) and (12) we get

$$\langle x(\omega)v(\omega')\rangle = \sqrt{\frac{k_BT}{2\gamma}}\frac{1}{\rho}\frac{i}{\sqrt{\omega}}2\pi\delta(\omega+\omega'). \quad (23)$$

III. GENERALIZED LANGEVIN DESCRIPTION

In this section we will collect the results outlined in the previous one and will put them in a consistent compact formulation. We emphasize three fundamental properties that such a representation must incorporate.

(i) The linear-response (LR) relation must hold [Eq. (18)].

(ii) The fluctuation-dissipation theorem (FDT) is also valid [Eq. (21)].

(iii) All the statistical properties exhibit two different behaviors, Brownian or subdiffusive motion, depending on whether time is smaller or larger than τ_d. We will in the following assume that the time scale $\tau_b=1/\gamma$ is smaller than τ_d such that the particles are moving diffusively (in contrast with ballistically) before colliding with each other.

Our aim is thus to write down an effective equation for the microscopic dynamics of a tagged particle in single-file systems, according to (i), (ii), and (iii). Such an equation turns out to be

$$\dot{x}(t) = v(t),$$

$$\dot{v}(t) = -[\gamma + 2n_d^2\gamma_{1/2}{}_0D_t^{-1/2}]v(t) + \tilde{\xi}(t). \quad (24)$$

Several new symbols have been introduced in the previous expression deserving an explanation. First, the quantity $n_d = \frac{\gamma}{\rho\sqrt{k_BT}}$ plays the same role as τ_d in the collisional representation of the particle's motion [14]: it accounts for the number of collisions a particle suffers before attaining subdiffusive behavior. Second, the quantity $\gamma_{1/2}$, which has the dimension of $[1/t^{3/2}]$, is the *generalized damping coefficient* and is equal to $(\frac{1}{\tau_d})^{3/2}$ since τ_d is the unique relevant time scale of the system. The third symbol in (24) is the Riemann-Liouville fractional operator [30]

$$ {}_0D_t^{-1/2}f(t) = \frac{1}{\Gamma\left(\frac{1}{2}\right)}\int_0^t \frac{f(t')}{|t-t'|^{1/2}}dt'. \tag{25}$$

Note that if we use the definition of the Caputo fractional derivative [31],

$$\frac{d^{1/2}f(t)}{dt^{1/2}} = \frac{1}{\Gamma\left(\frac{1}{2}\right)}\int_0^t \frac{df(t')/dt'}{|t-t'|^{1/2}}dt',$$

then (24) takes the form

$$\frac{d^2x(t)}{dt^2} + \gamma\frac{dx(t)}{dt} + 2n_d^2\gamma_{1/2}\frac{d^{1/2}x(t)}{dt^{1/2}} = \tilde{\xi}(t). \tag{26}$$

The noise appearing in (24) satisfies the properties (21). Indeed, we recall that the diffusive microscopic time scale $\tau_b = \frac{1}{\gamma}$ can be expressed as $\tau_b = \frac{\tau_d}{n_d^2}$ [14] so that Eq. (24) can be recast as

$$\dot{v}(t) = -\int_0^t \tilde{\gamma}(t-t')v(t')dt' + \tilde{\xi}(t), \tag{27}$$

introducing the generalized damping and defining the δ function to contribute half at the end point of an integral:

$$\tilde{\gamma}(t) = 2\gamma\left[\delta(t) + \frac{1}{\sqrt{\pi\tau_d t}}\right]. \tag{28}$$

Such a definition allows us to express the properties in (21) through the compact notation of the *generalized FDT* [22]

$$\langle\tilde{\xi}(t)\tilde{\xi}(t')\rangle = k_BT\tilde{\gamma}(|t-t'|). \tag{29}$$

The solution of Eq. (24) can be easily achieved by means of the Laplace transform

$$x(s) = \frac{x(0)}{s} + v(0)\tilde{\psi}(s) + \tilde{\xi}(s)\tilde{\psi}(s),$$

$$v(s) = v(0)\tilde{\mu}(s) + \tilde{\xi}(s)\tilde{\mu}(s), \tag{30}$$

where the mobility $\tilde{\mu}(t)$ in the s domain is given by

$$\tilde{\mu}(s) = \frac{1}{s+\tilde{\gamma}(s)} = \frac{1}{s+\gamma+2n_d^2\gamma_{1/2}s^{-1/2}} \tag{31}$$

and

$$\tilde{\psi}(s) = \frac{\tilde{\mu}(s)}{s} = \frac{1}{s^2+\gamma s+2n_d^2\gamma_{1/2}s^{1/2}}. \tag{32}$$

It is immediate to verify that the expression in (31) matches the two predicted regimes in (19). In the time domain the solution of the GLE (24) takes the simple form

$$x(t) = x(0) + v(0)\tilde{\psi}(t) + \int_0^t \tilde{\xi}(t')\tilde{\psi}(t-t')dt',$$

$$v(t) = v(0)\tilde{\mu}(t) + \int_0^t \tilde{\xi}(t')\tilde{\mu}(t-t')dt', \tag{33}$$

from which it is apparent that the joint probability distribution for x and v is a Gaussian in this formulation. The position and velocity are in fact linear functionals of $\tilde{\xi}(t)$, which is a (non-Markovian) Gaussian random process. Such a property corroborates all the previous analytical derivations for the probability distribution of a tagged particle moving subdiffusively in a single-file system (see, for example, Ref. [2] and references therein), and it is clearly at odds with a fractional Fokker-Planck description of the corresponding subdiffusive process, which leads to a stretched Gaussian solution [15].

Furthermore, we point out that the GLE (24) correctly describes the time behavior of all the observable moments of position and velocity, not only in the long-time asymptotic regime, but also at the initial diffusive stage. To see this it is sufficient to start from the general solutions (33) and put in some physical assumptions [33]. For instance, the first moments of velocity and position will read

$$\langle x(t)\rangle = \langle x(0)\rangle + \langle v(0)\rangle\tilde{\psi}(t),$$

$$\langle v(t)\rangle = \langle v(0)\rangle\tilde{\mu}(t). \tag{34}$$

The second moments, however, are more interesting quantities: for the VAF, assuming $\langle v^2(0)\rangle = k_BT$, it is straightforward to prove

$$C(t) \equiv \langle v(t)v(0)\rangle = k_BT\tilde{\mu}(t), \tag{35}$$

which is the *generalized first Kubo theorem*. The numerical evidence of this is given in Fig. 3(a), where several rescaled curves are plotted against the analytic function in (35) after numerical inversion of the mobility in (31).

The excellent agreement between the analytical description yielded by (24) and the numerical data is even more apparent looking at the mean-square displacement of the tagged particle (Fig. 4). Indeed from (33) one obtains

$$\delta x^2(t) \equiv \langle[x(t)-x(0)]^2\rangle = 2\langle v^2(0)\rangle\int_0^t \tilde{\psi}(t')dt', \tag{36}$$

where it is possible to recognize the theorem stated in Ref. [14] provided that

$$\frac{d}{dt}\tilde{\psi}(t) = \tilde{\mu}(t). \tag{37}$$

The exact expression given in (36) quantitatively reproduces the *three* stages of the $\delta x^2(t)$ curves in Fig. 4 (ballistic, diffusive, subdiffusive) as well as the characteristic time scales (τ_b and τ_d) on which the crossovers between them take place:

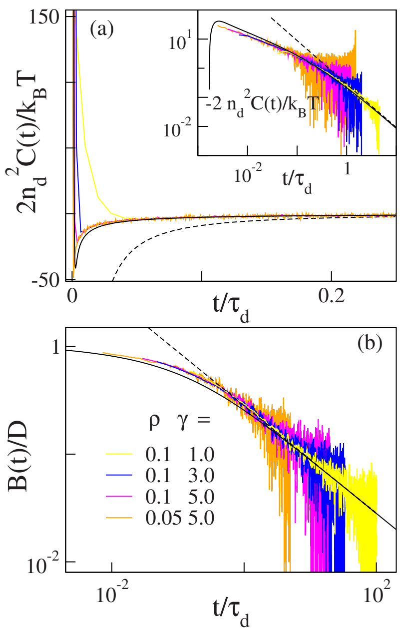

FIG. 3. (Color online) Second moments of observables: rescaled VAF $\langle v(t)v(0)\rangle$ (a) and position-velocity correlation function $\langle [x(t)-x(0)]v(t)\rangle$ (b) vs t/τ_d for several densities ρ and damping γ. $k_BT=1.0$. Dashed lines are the asymptotic formulas (16) and (23), respectively, whereas the solid ones represent the GLE predictions (35) and (40) for $\rho=0.05$ and $\gamma=5.0$. To improve our statistics we averaged over 3000 different realizations.

$$\delta x^2(t) = \begin{cases} k_BTt^2, & t \ll \tau_b, \\ 2Dt, & \tau_b \ll t \ll \tau_d, \\ 2\sqrt{\dfrac{D}{\pi\rho^2}}\sqrt{t}, & t \gg \tau_d. \end{cases} \tag{38}$$

An analytical inversion of the formula in (32) including the crossover to the subdiffusive regime can be achieved by neglecting the inertial (ballistic) term, leading to

$$\langle [x(t)-x(0)]^2\rangle \simeq \frac{1}{\rho^2}\left[\sqrt{\frac{4t}{\pi\tau_d}} + \frac{e^{4t/\tau_d}}{2}\operatorname{erfc}\left(\sqrt{\frac{4t}{\tau_d}}\right) - \frac{1}{2}\right]. \tag{39}$$

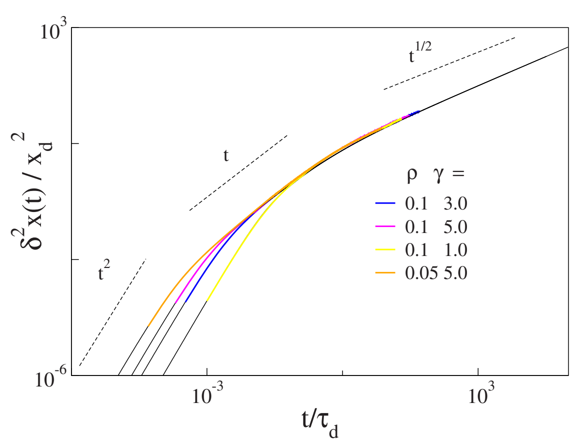

FIG. 4. (Color online) Mean-square displacement $\delta x^2(t) = \Sigma_{i=1}^N [x_i(t)-x_i(0)]^2/N$ for different values of ρ and γ with $k_BT = 1.0$ plotted versus t/τ_d. Data have been rescaled by $x_d^2 = 2/(\rho^2\sqrt{\pi})$ on the y axis [14]. The black solid lines represent the theoretical predictions as given by the GLE (36): they provide an excellent description of the three diffusive regimes. Data have been averaged over 30 different realizations for N of the order of 10^4.

TABLE I. Values of τ_b and τ_d and their ratio for the values of ρ and γ used in Figs. 2–5.

ρ	γ	τ_b	τ_d	τ_d/τ_b
0.25	0.5	2.0	8.0	4
0.1	1.0	1.0	100	100
0.1	3.0	~0.334	300	900
0.1	5.0	0.2	500	2500
0.05	5.0	0.2	2000	10000

In general, we believe the GLE (24) provides a very good description of the tagged particle's stochastic motion only when the condition $\tau_b \ll \tau_d$ is fulfilled (see Table I). In other words, the particle must attain a truly diffusive regime before getting a collision with one of its nearest neighbors. One can see that the proposed GLE cannot work well in the opposite case where $\tau_b > \tau_d$ since all interactions with neighboring particles vanish according to Eq. (24) as $\gamma \to 0$. This cannot be true, since the particle will still collide and exchange momentum with its neighbors even though the rest of the friction with the surroundings vanish. We have not found a GLE that works well in the case $\tau_b > \tau_d$.

The close comparison between theory and numerics when $\tau_b \ll \tau_d$ is also displayed by the position-velocity correlation function $\langle x(t)v(t)\rangle$ for which the following expression holds:

$$B(t) \equiv \langle [x(t)-x(0)]v(t)\rangle = \langle v^2(0)\rangle \tilde{\psi}(t). \tag{40}$$

In Fig. 3(b) we compare the numerical data with both (40) and (23), which is expected to work well only in the asymptotic regime.

The relaxation of the second moment of the velocity $\langle v^2(t)\rangle$ to the asymptotic value k_BT is given by

$$\langle v^2(t)\rangle = \langle v^2(0)\rangle \mu^2(t) + k_BT[1-\mu^2(t)] \tag{41}$$

and deserves particular attention. In Ref. [23] it has been shown that the fractional Brownian process [32] generated by a fractional Langevin equation and the stochastic process corresponding to the relative fractional Kramer equation [34] are *on average* the same, except for the second moment of the velocity. We point out that such a discrepancy is a common problem in nonequilibrium statistical mechanics whenever one is concerned to pass from a generalized Langevin dynamical description of a non-Markovian process [21] to the corresponding Fokker-Planck equation for the probability distribution [35] (see, for instance, the discussion in Ref. [37] on the inconsistency of a retarded Fokker-Planck equation of the Rubin model [36]). In the GLE (24) the $\langle v^2(t)\rangle$

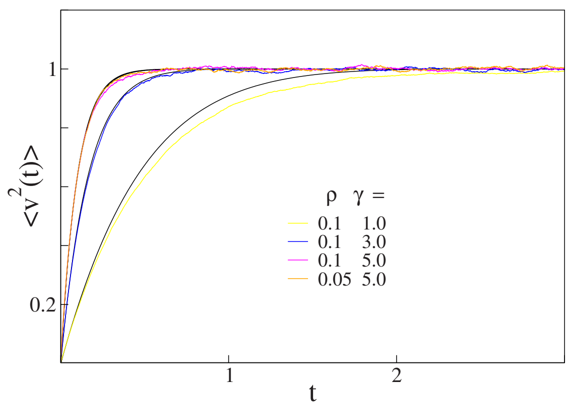

FIG. 5. (Color online). Second moment of the velocity $\langle v^2(t)\rangle$ vs t, displayed for several densities ρ and damping γ. $k_BT=1.0$. The initial tagged particle velocity $v(0)$ has been set to 0. Expression (41) (solid black lines) shows the typical Brownian exponential relaxation to k_BT: for all the displayed curves, $\tau_b/\tau_d \ll 1$. To improve our statistics we averaged the numerical curves over 3000 realizations.

relaxation process is dictated by the Brownian dynamics (see Fig. 5) as long as $\tau_b \ll \tau_d$. The fractional Kramer equation corresponding to (24) would read

$$\frac{\partial P(x,v,t)}{\partial t} + v\frac{\partial P(x,v,t)}{\partial x} = [\gamma + \gamma_{1/2}\,{}_0D_t^{-1/2}]\hat{L}_{FP}P(x,v,t), \tag{42}$$

where $\hat{L}_{FP}=\frac{\partial}{\partial v}v + k_BT\frac{\partial^2}{\partial v^2}$. In the $\tau_b \ll \tau_d$ limit the $\langle v^2(t)\rangle_{FKE}$ expression yielded by (42) is the one provided by the usual Fokker-Planck equation—i.e., $\langle v^2(t)\rangle_{FKE} \simeq k_BTe^{-2\gamma t}$—once $v(0)=0$, in agreement with the exponential saturation shown in Fig. 5. This could lead to believe that the GLE description (24) and the Kramer equation (42) are *de facto* equivalent in the range $\tau_b \ll \tau_d$. Nevertheless, we expect that the $P(x,t)$ solution of (42) is still a stretched exponential instead of a Gaussian: this fundamental difference casts some general doubt on the possibility to determine a Fokker-Planck equation (with time independent coefficients) for systems whose microscopic dynamics is represented by a FLE.

The last part of this section is devoted to property (i): namely, the validity of the LR relation. In the presence of an external force acting from time equal to zero and onwards, Eqs. (24)–(27) take the form

$$\dot{x}(t) = v(t),$$

$$\dot{v}(t) = -\int_0^t \tilde{\gamma}(t-t')v(t')dt' + F(t) + \tilde{\xi}(t). \tag{43}$$

The LR relation (18) in s space can thus expressed by using (31) as

$$\langle v(s)\rangle = \tilde{\mu}(s)F(s), \tag{44}$$

so that, thanks to expression (31), the real and imaginary parts of the mobility $\tilde{\mu}(t)$ read

$$\mathrm{Re}[\tilde{\mu}(\omega)] = \frac{\gamma + \sqrt{2}n_d^2\gamma_{1/2}\omega^{-1/2}}{(\gamma + \sqrt{2}n_d^2\gamma_{1/2}\omega^{-1/2})^2 + (\sqrt{2}n_d^2\gamma_{1/2}\omega^{-1/2} - \omega)^2},$$

$$\mathrm{Im}[\tilde{\mu}(\omega)] = \frac{\omega - \sqrt{2}n_d^2\gamma_{1/2}\omega^{-1/2}}{(\gamma + \sqrt{2}n_d^2\gamma_{1/2}\omega^{-1/2})^2 + (\sqrt{2}n_d^2\gamma_{1/2}\omega^{-1/2} - \omega)^2}. \tag{45}$$

Remarkably, in Fig. 2 both functions (45) (solid black lines) are shown to fit quite well the outcome of our numerics.

Finally in the case of a constant external force—i.e., $F(t)=F$ for $t \geq 0$ in (43)—the LR relation provides that the drift $\langle x(t)\rangle_F$ satisfies the *generalized Einstein relation*

$$\langle x(t)\rangle_F = \langle x(0)\rangle + F\frac{\delta x^2(t)}{2k_BT}, \tag{46}$$

which has been shown to be valid on file systems moving on a ring [38] and on an infinite one-dimensional lattice [39].

IV. CONCLUSION

In this paper we introduced the effective equation ruling the microscopic stochastic motion of a SF tagged particle. Starting from a diffusionlike formalism for the file density dynamics, we were led to several properties for which the GLEs (24)–(27) can be regarded as a representation. Indeed the GLE formalism provides an elegant representation of the generalized fluctuation-dissipation theorems (29)–(35), linear response relation (44), and generalized Einstein relation (46), all remarkable properties satisfied by the particle, both in its diffusive and in subdiffusive phase.

Nevertheless, we want to stress that the GLE is just a description, though very good, of the stochastic motion of the SF particle and, in this perspective, is valid within certain limits. However, along the same line, within certain approximations the Langevin equation provides an excellent effective equation for the motion of a Brownian particle immersed in a thermal bath; i.e., it describes well the ballistic and diffusive regimes of the particle, but one would not expect it to be exact in the region of crossover between the two regimes. In SF systems the tagged particle is subjected to two types of thermal baths, as is apparent from (24): the first is mimicked by the usual Markovian uncorrelated noise, whereas the second, physically embodied by the surrounding file's particles, is achieved by the introduction of an additional non-Markovian term, responsible for the strong memory effects. Surprisingly this "sum of thermal baths" turns out to be well described (in the same manner as for the Brownian particle and LE) by simply the algebraic sum of two independent terms in the stochastic equation of motion. Alternatively one could view (24) as an usual LE (3) where the relabeling-collisional symmetry accounts for the fractional term.

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