Everlasting effect of initial conditions on single-file diffusion

N. Leibovich and E. Barkai
Department of Physics, Institute of Nanotechnology and Advanced Materials, Bar Ilan University, Ramat-Gan 52900, Israel
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We study the dynamics of a tagged particle in an environment of point Brownian particles with hard-core interactions in an infinite one-dimensional channel (a single-file model). In particular, we examine the influence of initial conditions on the dynamics of the tagged particle. We compare two initial conditions: equal distances between particles and uniform density distribution. The effect is shown by the differences of mean-square-displacement and correlation function for the two ensembles of initial conditions. We discuss the violation of Einstein relation, and its dependence on the initial condition, and the difference between time and ensemble averaging. More specifically, using the Jepsen line, we will discuss how transport coefficients, like diffusivity, depend on the initial state. Our work shows that initial conditions determine the long time limit of the dynamics, and in this sense the system never forgets its initial state in complete contrast with thermal systems (i.e., a closed system that attains equilibrium independent of the initial state).

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I. INTRODUCTION

Particles diffusion in one-dimensional systems, with hard-core interactions, have been studied for many years [1–16]. One aspect of this problem is the motion of a tagged particle. This kind of system can be used as a model for the motion of a single molecule in a crowded one-dimensional environment, such as a biological pore or channel [17,18], and experimental studies of physical systems such as zeolites [19] and colloid particles in confined topology [20] or optical tweezers [21].

In a system of interacting Brownian particles, the motion of a tagged particle was thoroughly investigated. The initial state of the system of particles is in most previous works taken from equilibrium. That means particles are initially uniformly distributed with density ρ. One of the known results is that the tracer particle subdiffuses, i.e., $\langle x_T^2\rangle \sim 2D_{1/2}t^{1/2}$. Harris was the first to provide a theoretical derivation for this phenomena by using statistical arguments [1]. For a finite system, e.g., single-file diffusion in a box, the tagged particle's mean-square-displacement reaches equilibrium, $\lim_{t\to\infty}\langle x_T^2\rangle = \text{const.}$ [22–24] (see also Ref. [25] for periodic boundary condition).

The goal of this paper is to investigate the sensitivity of single-file diffusion to the initial condition. Van Beijeren discusses the influence of initial condition in the context of an asymmetric simple exclusion process [26]. He compares two cases: a fixed initial state, drawn from equilibrium state, versus a stationary random initial state, where average over initial configuration was taken. The comparison was made for different properties of the motion, such as mass fluctuations. His model is related to other works [27–29], where effect of initial conditions in many particle driven system was investigated (see further discussion in the summary).

A treasure is found in an Appendix of Lizana *et al.* [30]. They note by passing that the generalized diffusion coefficient $D_{1/2}$ is sensitive to the way the system is prepared. That is an interesting result since naively we expect diffusivities of interacting systems not to be sensitive to the initial conditions. Here we confirm this prediction using the Jepsen line, showing that the prediction in Ref. [30] is correct. To show that $D_{1/2}$ is sensitive to the initial state of the system we consider two ensembles of initial conditions: an initial state that is taken from equilibrium versus particles initially situated in equal distances between each other. We show that $D_{1/2}$ for both ensembles differs by a prefactor of $\sqrt{2}$.

Since transport coefficient like $D_{1/2}$, for the many-body interacting system, is sensitive to initial preparation, we must challenge basic concepts in nonequilibrium statistical mechanics. For example, in Sec. V we investigate the response of the system to external field. Will that response depend on initial condition? How do we formulate the Einstein relation, if at least diffusivity $D_{1/2}$ is sensitive to the initial condition? Further, we investigate the correlation function $\langle x(t+\Delta)x(t)\rangle$ of the process showing clear differences between the ensembles of initial conditions (see Secs. III and IV). These are found also in long time limit $\Delta\to\infty$, where the correlation function is markably different from the mean-square-displacement (MSD).

Finally, we investigate the time average MSD (Sec. VI). Will that time average depend on the initial condition (like the ensemble average)? This becomes an interesting question. Further, is the MSD ergodic, in the sense that for two identical initial conditions do the corresponding time and ensemble average MSD coincide?

II. THE MODEL

In our model we have $2N+1$ point identical Brownian particles, with hard-core interactions, in a one-dimensional system. D represents the diffusion coefficient of a free Brownian particle. We tag the central particle, so there is an equal number of particles on its right and on its left. Initially, the tagged particle is situated at the origin $x_T(t=0)=0$. The system is stretched from $-L$ to L. The size of the system and the number of particles is infinite ($N,L\longrightarrow\infty$), but $N/L=\rho$ is fixed, where ρ represents the particles density. ρ^{-1} is the mean distance between nearest neighbors. We consider two types of initial conditions. The first is the case of particles distributed with fixed density; namely, the mean distance between particles, which are exponentially distributed, is ρ^{-1}. This case was treated previously. The second case we consider

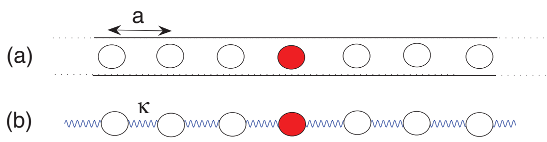

FIG. 1. (Color online) An illustration of the model of particles in a narrow channel so particles cannot pass each other with a marked tagged particle. The system can be mapped into a chain of beads, which are interconnected with springs [30] (see the Harmonization method below). This schematic diagram presents the initial state of the system with equally spaced particles.

is the case where particles are initially situated on a lattice, with equal distances between particles $L/N = a$, where a is the lattice constant. We labeled the tagged particle as $n = 0$ and the particles to its right are labeled $n = 1,2\ldots$, according to their order. Similarly, the particles to the left of the tagged particle are labeled $n = -1,-2\ldots$. Then, the initial position of each particle is represented by

$$x_n(t)|_{t=0} = na, \tag{1}$$

where n is the label of the particle, $n \in \{-N \ldots N\}$ (see Fig. 1).

III. THE JEPSEN LINE

A. Mean-square-displacement

In this section, we find the MSD with a rigorous method by treating the problem with a theoretical tool called the Jepsen line (see details below) [2,3,31,32]. We find the MSD of the tagged particle:

$$\begin{aligned} \langle x_T^2(t)\rangle_{\text{lat}} &= a\sqrt{2}\sqrt{\frac{D}{\pi}}\sqrt{t} \\ \langle x_T^2(t)\rangle_{\text{uni}} &= 2\rho^{-1}\sqrt{\frac{D}{\pi}}\sqrt{t}, \end{aligned} \tag{2}$$

which is in agreement with the statement in Ref. [30]. Here $\langle\ldots\rangle_{\text{lat}}$ refers to ensemble average when the system is initially on a lattice (nonequilibrium case), and $\langle\ldots\rangle_{\text{uni}}$ refers to an equilibrium state (uniform distribution). Notice that the difference between the MSD of the two initial conditions is a prefactor of $\sqrt{2}$ (see Fig. 2). More specifically, we compared between the two initial conditions by taking $a = \rho^{-1}$, since that is the average separation between particles on the lattice. This is an interesting effect, since we find that the influence of the initial conditions on the diffusion of the tagged particle in infinite system is lasting forever.

B. Jepsen line method

The motion of a single particle without interactions with other particles is given by the Green function $g(x,x_0,t)$, where $g(x,x_0,t)dx$ is the probability of the noninteracting particle, which started at x_0 to be in $(x,x+dx)$ at time t. For an infinite system, the Green function for a free Brownian particle is simply a Gaussian

$$g(x,x_0;t) = \frac{1}{\sqrt{4\pi Dt}}e^{-\frac{(x-x_0)^2}{4Dt}}. \tag{3}$$

We consider initial positions of particles given by $x_n(t)|_{t=0} = an$, where $n\epsilon\{-N\ldots N\}$ is the label of the interacting particles

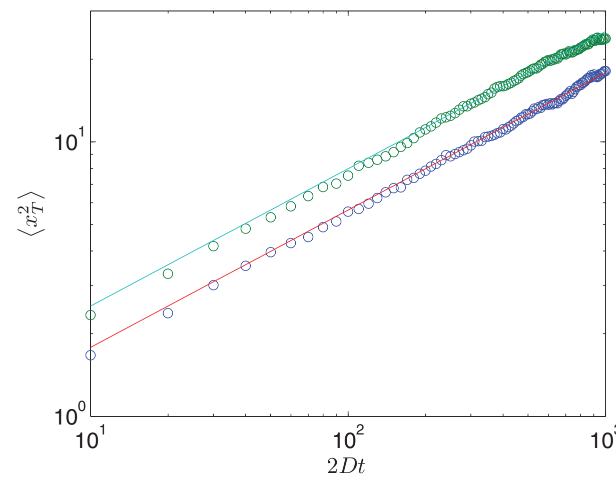

FIG. 2. (Color online) The MSD of the tagged particle in two cases: when the particles are initially in thermodynamic equilibrium (upper line) and when the particles are equally spaced (see simulations details in Appendix A). Comparing the two results gives the factor of $\sqrt{2}$ [see Eq. (2)]. The averaged distance between particles is $a = \rho^{-1}$.

and a is the lattice constant. A straight line that is initially at $x = 0$ and follows $x = vt$ (v is a "test" velocity) is called the Jepsen line (see Fig. 3). Initially, there are $N + 1$ particles, including the tagged particle, to the right of the line, and N particles to the left of the line.

Let α be the label of the first particle that is situated to the right of the Jepsen line; therefore, initially $\alpha(t)|_{t=0} = 0$. The random variable α increases or decreases by $+1$ or -1, according to the following rules: if a particle crosses the Jepsen line from left to right α decreases, $\alpha \to \alpha - 1$; and if a particle crosses the line from right to left α increases, $\alpha \to \alpha + 1$. Thus, α is a random walk decreasing or increasing its value $+1$ or -1 at random time (see Fig. 4).

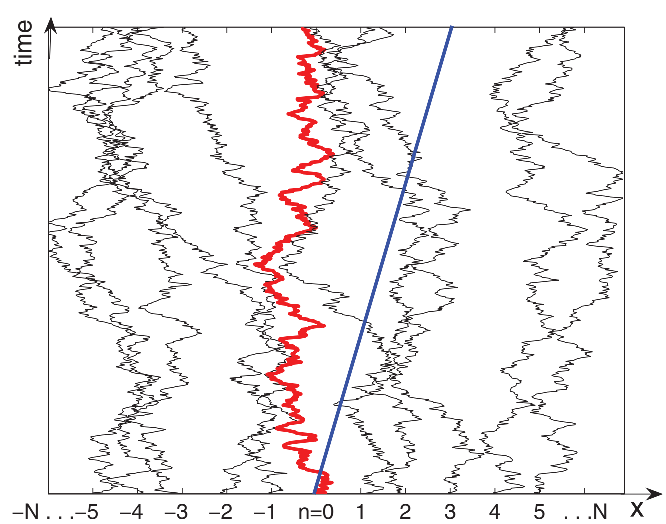

FIG. 3. (Color online) A schematic example of the trajectories of particles. The straight solid line that starts at (0,0) and follows $x = vt$ is called the Jepsen line. We label the particles so that the tagged particle is $n = 0$ and the particles to its right are labeled $n = 1,2\ldots$, according to their order, while the particles to its left are labeled similarly with $n = -1,-2,\ldots$.

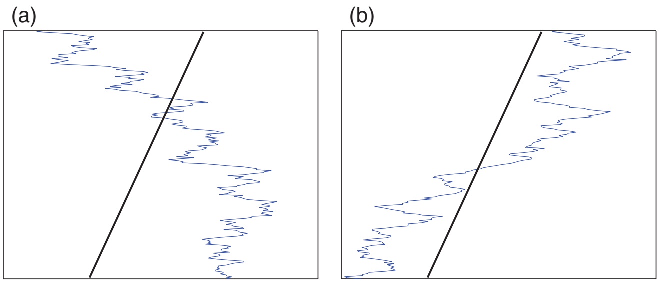

FIG. 4. (Color online) When a particle crosses the Jepsen line from right to left (or left to right), α increases, $\alpha \to \alpha + 1$ (or decreases, $\alpha \to \alpha - 1$).

In a one-dimension, hard-core elastic collision event, the result of two identical particles (same mass) colliding is that they switch their velocities. For an over-damped Brownian particle, this is equivalent to two particles that pass through each other, and after the particles cross each other, the labels of the two particles are switched. Instead of relabeling the particles after every collision, we let particles pass through each other, and then at time t, we label our particles. So, in fact, in the interval $(0,t)$ we view the particles as noninteracting (see Fig. 5).

We define

$$\alpha = \sum_{n=1}^{N} \delta\alpha_n, \tag{4}$$

where $\delta\alpha_n = \alpha_n^R + \alpha_{-n}^L$ and α_n^R is the number of times that nth particle crossed the Jepsen line from right to left, minus the number of times that it crossed the line from left to right, and

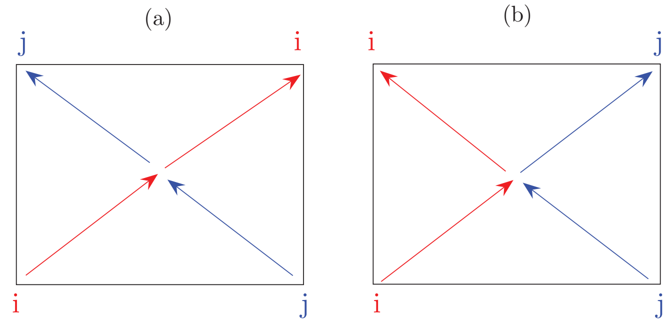

FIG. 5. (Color online) Illustration of collision event: since the system in unidimensional and the particles are identical, when particles collide the velocities are switched (b). Instead of switching their velocities, we can switch their labels and treat each particle as a free particle without interaction (a). The interactions come into account in sorting the particles and finding the central one.

α_{-n}^L is defined similarly. For example, $\alpha(t) = 0$ means that all the particles stay on their original side of the Jepsen line, so the first particle that is situated to the right of the line is the tagged one. The variable α is determined by $2N + 1$ random variables, and since $N \gg 1$, we can neglect the contribution from $\delta\alpha_0$.

For calculating the probability density function $P_N(\alpha)$, we mark $P_{ij}({x_0}^n)$ as the probability that the nth particle starts at x_0 to the i side of the Jepsen line and ends at the j side of the line $(i,j \in \{R,L\})$ (R represents the right side of the Jepsen line, and L stands for left). On a lattice, ${x_0}^n = an$, where a is an equal distance between particles. For one step of the random walk, $\delta\alpha_n$ can get the values $-1,0,1$ with the probabilities

$$\delta\alpha_n = \begin{cases} 1 & P(\delta\alpha_n = 1) = P_{RL}(an)P_{LL}(-an) \\ 0 & P(\delta\alpha_n = 0) = P_{RL}(an)P_{LR}(-an) + P_{RR}(an)P_{LL}(-an) \\ -1 & P(\delta\alpha_n = -1) = P_{RR}(an)P_{LR}(-an). \end{cases} \tag{5}$$

Notice that $\delta\alpha_n$ depends on the motion of two particles initially at an and at $-an$. For example, if one particle starting on an, i.e., right side of the Jepsen line (R), switches to the left (L) of the line while corresponding particle on $-an$ (L) remains in L, we have $\delta\alpha_n = 1$. The probability $P_{ij}(an)$ is given by Green function and initial condition

$$\begin{aligned} P_{RL}(an) &= \int_{-L}^{vt} \frac{1}{\sqrt{4\pi Dt}} e^{-\frac{(x-an)^2}{4Dt}} dx \\ P_{RR}(an) &= \int_{vt}^{L} \frac{1}{\sqrt{4\pi Dt}} e^{-\frac{(x-an)^2}{4Dt}} dx \\ P_{LR}(-an) &= \int_{vt}^{L} \frac{1}{\sqrt{4\pi Dt}} e^{-\frac{(x+an)^2}{4Dt}} dx \\ P_{LL}(-an) &= \int_{-L}^{vt} \frac{1}{\sqrt{4\pi Dt}} e^{-\frac{(x+an)^2}{4Dt}} dx, \end{aligned} \tag{6}$$

where system size $L \to \infty$, so $P_{RL}(an) + P_{RR}(an) = 1$.

C. The motion of the tagged particle

When $N \to \infty$, α is normally distributed, according to the central limit theorem for the random variable α (see Appendix A); hence,

$$P_N(\alpha) \sim \frac{1}{\sqrt{2\pi\sigma^2}} e^{-\frac{(\alpha - \langle\alpha\rangle)^2}{2\sigma^2}}, \tag{7}$$

where $\langle\alpha\rangle$ is the mean of α and $\sigma^2 = \langle\alpha^2\rangle - \langle\alpha\rangle^2$ is its variance.

The probability of finding the tagged particle to the left of the Jepsen line is

$$P(x_T < vt) = \int_{-\infty}^{vt} P_t(x_T) dx_T, \tag{8}$$

and our goal is to find the probability density function (PDF) $P_t(x_T)$ of finding the tagged particle in $(x_T, x_T + dx_T)$. The event $x_T < vt$ is equivalent to the case that the first particle that is situated to the right of the Jepsen line is labeled $\alpha > 0$;

hence,

$$P(x_T < vt) \sim \int_0^\infty \frac{1}{\sqrt{2\pi\sigma^2}} e^{-\frac{(\alpha-\langle\alpha\rangle)^2}{2\sigma^2}} d\alpha. \qquad (9)$$

Taking the derivative of Eq. (9), following Eq. (8), and replacing $vt \to x_T$, we find the PDF of the tagged particle position

$$P(x_T) \sim \partial_{x_T} \int_0^\infty \frac{1}{\sqrt{2\pi\sigma^2}} e^{-\frac{(\alpha-\langle\alpha\rangle)^2}{2\sigma^2}} d\alpha, \qquad (10)$$

where we will soon take the large N limit. By definition, the average of variable $\delta\alpha_n$ is

$$\langle \delta\alpha_n \rangle = \sum_{\delta\alpha_n=-1,0,1} \delta\alpha_n P(\delta\alpha_n). \qquad (11)$$

By using $P_{LL}(-an) = 1 - P_{LR}(-an)$ and $P_{RR}(an) = 1 - P_{RL}(an)$, it is easy to see

$$\langle \delta\alpha_n \rangle = P_{RL}(an) - P_{LR}(-an). \qquad (12)$$

Notice that here P_{RR}, P_{LR}, etc. [Eq. (6)] is now calculated with x_T as the upper or lower integration bound, i.e., replacing $vt \to x_T$ in Eq. (6). Similarly, we can find

$$\left\langle \delta\alpha_n^2 \right\rangle = P_{RR}(an)P_{LR}(-an) + P_{LL}(-an)P_{RL}(an). \qquad (13)$$

For the average, we clearly have

$$\langle \alpha \rangle = \sum_{n=1}^{N} \langle \delta\alpha_n \rangle. \qquad (14)$$

For the second moment,

$$\langle \alpha^2 \rangle = \sum_{n=1}^{N} \left\langle \delta\alpha_n^2 \right\rangle + \sum_{n=1}^{N} \sum_{m\neq n}^{N} \langle \delta\alpha_n \delta\alpha_m \rangle . \qquad (15)$$

Therefore, the variance is

$$\sigma_\alpha^2 = \langle \alpha^2 \rangle - \langle \alpha \rangle^2 = \sum_{n=1}^{N} \left\langle \delta\alpha_n^2 \right\rangle - \langle \delta\alpha_n \rangle^2. \qquad (16)$$

Note the random variables $\{\delta\alpha_n\}$ are independent, i.e., $\langle \delta\alpha_n \delta\alpha_m \rangle = \langle \delta\alpha_n \rangle \langle \delta\alpha_m \rangle$; therefore, the covariance term was canceled. Since $N \to \infty$, we can approximate $P_{t,N}(x_T)$ near its saddle point. The equation for finding the saddle point x_s is

$$\langle \alpha \rangle_{x_s} = \left[\sum_n P_{RL}(an) - P_{LR}(-an) \right] \Bigg|_{x_s} = 0; \qquad (17)$$

the particles are unbiased, therefore, $x_s = 0$. Approximating $\langle \alpha \rangle$ near the saddle point and using Eq. (6) in the limit $L \to \infty$ and $vt \to x_T \to 0$ and Eq. (12) gives (the first nonzero term)

$$\langle \alpha \rangle \approx \partial_x \langle \alpha \rangle |_{x_T=0} x_T = \sum_{n=1}^{N} \frac{2}{\sqrt{4\pi Dt}} e^{-\frac{(an)^2}{4Dt}} x_T. \qquad (18)$$

Similar approximation for σ^2 by using Eqs. (12), (13), and (16) gives

$$\sigma^2|_{x_T=0} = \sum_{n=1}^{N} [P_{RL}(an)P_{RR}(an) + P_{LR}(-an)P_{LL}(-an)]_{x_T=0}; \qquad (19)$$

hence, by using Eq. (6), we find

$$\sigma^2|_{x_T=0} \approx 2 \sum_{n=1}^{N} \int_0^\infty \frac{1}{\sqrt{4\pi Dt}} e^{-\frac{(x-na)^2}{4Dt}} dx \times \int_0^\infty \frac{1}{\sqrt{4\pi Dt}} e^{-\frac{(x+na)^2}{4Dt}} dx. \qquad (20)$$

By substituting Eqs. (18) and (20) into Eq. (10), we find

$$P(x_T) \sim \partial_{x_T} \int_0^\infty \frac{d\alpha}{\sqrt{2\pi\sigma^2(0)}} \exp\left[-\frac{(\alpha - \langle\alpha\rangle'(0)x_T)^2}{2\sigma^2(0)} \right], \qquad (21)$$

where $\sigma^2(0) = \sigma^2|_{x_T=0}$ and $\langle\alpha\rangle'(0) = \partial_{x_T} \langle\alpha\rangle|_{x_T=0}$. Changing the integration variable, according to $\overline{\alpha} = \alpha - \langle\alpha\rangle'(0)x_T$, and taking the derivative with respect to x_T gives

$$P(x_T) \sim \frac{1}{\sqrt{2\pi\sigma^2(0)}} \exp\left[-\frac{(\langle\alpha\rangle'(0)x_T)^2}{2\sigma^2(0)} \right] \langle\alpha\rangle'(0). \qquad (22)$$

We see that x_T is normally distributed with mean

$$\langle x_T \rangle = 0, \qquad (23)$$

which is clear from symmetry, and variance

$$\left\langle {x_T}^2 \right\rangle = \frac{\sum_{n=1}^{N} \int_0^\infty \frac{1}{\sqrt{4\pi Dt}} e^{-\frac{(x-na)^2}{4Dt}} dx \int_0^\infty \frac{1}{\sqrt{4\pi Dt}} e^{-\frac{(x+na)^2}{4Dt}} dx}{2\left(\sum_{n=1}^{N} \frac{1}{\sqrt{4\pi Dt}} e^{-\frac{(an)^2}{4Dt}} \right)^2}. \qquad (24)$$

D. Mean-square-displacement evaluation

We evaluate the MSD of the tagged particle that is given in Eq. (24):

$$\lim_{N\to\infty} \sum_{n=1}^{N} \frac{1}{\sqrt{4\pi Dt}} e^{-\frac{(an)^2}{4Dt}} \sim \frac{1}{2a} - \frac{1}{2\sqrt{4\pi Dt}}, \qquad (25)$$

where we used the Euler-Maclouren formula:

$$\sum_{k=1}^{N-1} f_k = \int_0^N f(k)dk - \frac{1}{2}[f(0) + f(N)]. \qquad (26)$$

For the time when particles interact with each other, i.e., $a^2/D \ll t$,

$$\sum_{n=1}^{\infty} \frac{1}{\sqrt{4\pi Dt}} e^{-\frac{(an)^2}{4Dt}} \sim \frac{1}{2a}. \qquad (27)$$

Similarly, we find the numerator:

$$\frac{1}{4} \sum_{n=1}^{\infty} Erfc\left(\frac{an}{\sqrt{4Dt}} \right) Erfc\left(-\frac{an}{\sqrt{4Dt}} \right) \sim \frac{1}{4} \left(\sqrt{\frac{2}{\pi}} \frac{\sqrt{4Dt}}{a} - \frac{1}{2} \right). \qquad (28)$$

Therefore, when particles interact with each other,

$$\frac{a^2}{D} = \tau_{\text{int}} \ll t, \qquad (29)$$

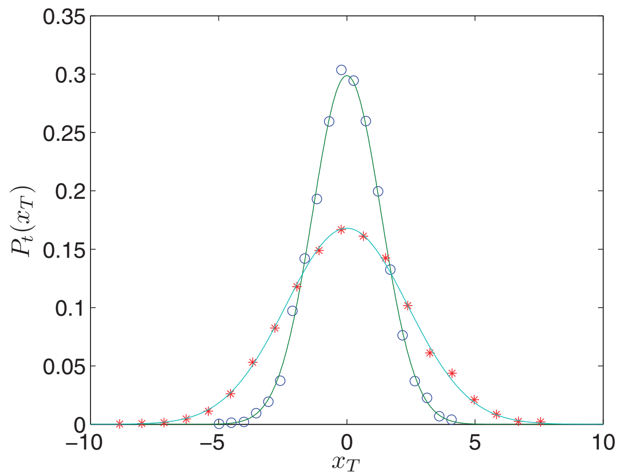

FIG. 6. (Color online) The tagged particle location with lattice initial condition is distributed normally when the system contains large numbers of particles. The solid curve is the normal distribution with mean zero and variance [Eq. (31)]. We simulated two different times: $t = 10$ (○) and $t = 100$ (∗).

the numerator in Eq. (24) is

$$\frac{1}{4}\sum_{n=1}^{\infty} Erfc\left(\frac{an}{\sqrt{4Dt}}\right)Erfc\left(-\frac{an}{\sqrt{4Dt}}\right) \longrightarrow \frac{1}{4}\sqrt{\frac{2}{\pi}}\frac{\sqrt{4Dt}}{a}; \tag{30}$$

hence, we find

$$\langle {x_T}^2\rangle_{\rm lat} = \sqrt{\frac{2}{\pi}}a\sqrt{Dt}. \tag{31}$$

For particles that are initially uniformally distributed, it is well known that [3]

$$\langle x_T^2\rangle_{\rm uni} = \frac{2}{\sqrt{\pi}}\rho^{-1}\sqrt{Dt}. \tag{32}$$

Comparing Eqs. (31) and (32), we see that if we assign $a = \rho^{-1}$, the two results differ by a prefactor $\sqrt{2}$. To summarize, in a long time limit the PDF for the tagged particle interacting with a bath initially on a lattice is

$$P_t(x_T) \sim \frac{1}{\sqrt{a\sqrt{8\pi}\,Dt}}\exp\left(-\frac{\sqrt{\pi}x_T^2}{a\sqrt{8Dt}}\right); \tag{33}$$

see Fig. 6.

IV. CORRELATION FUNCTION

A. Correlation function

The correlation function between the location of the tagged particle at time t to its location at time $t+\Delta$ is defined as: $\langle x_T(t+\Delta)x_T(t)\rangle$. We will soon show that the correlation function for particles initially on a lattice is

$$\langle x_T(t+\Delta)x_T(t)\rangle_{\rm lat} = a\sqrt{\frac{D}{\pi}}(\sqrt{2t+\Delta}-\sqrt{\Delta}), \tag{34}$$

while for particles initially in an equilibrium state

$$\langle x_T(t+\Delta)x_T(t)\rangle_{\rm uni} = \rho^{-1}\sqrt{\frac{D}{\pi}}(\sqrt{t+\Delta}+\sqrt{t}-\sqrt{\Delta}), \tag{35}$$

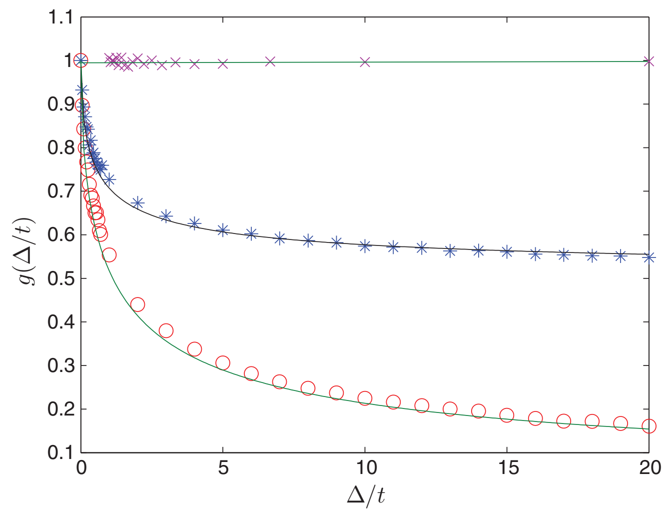

FIG. 7. (Color online) The normalized correlation function shows a significant difference between three cases: free particle (upper curve), uniform initial condition (middle curve), and lattice initial condition (lower curve). When $\Delta \gg t$, the normalized correlation function in thermal equilibrium goes to $1/2$ and in the nonthermal case it decays to zero. The circles and stars represent the simulations, and the solid lines represent the theory Eq. (37).

which agrees with Eqs. (31) and (32) when $\Delta = 0$. Equation (35) has the structure of the correlation function of fractional Brownian motion [33].

We define normalized correlation function:

$$g\left(\frac{\Delta}{t}\right) = \frac{\langle x_T(t+\Delta)x_T(t)\rangle}{\langle x_T^2(t)\rangle}. \tag{36}$$

Hence, using Eqs. (31), (32), (34), and (35), we find

$$\begin{aligned} g^{\rm lat}\left(\frac{\Delta}{t}\right) &= \sqrt{1+\frac{\Delta}{2t}}-\sqrt{\frac{\Delta}{2t}},\\ g^{\rm uni}\left(\frac{\Delta}{t}\right) &= \frac{1}{2}\left(1+\sqrt{1+\frac{\Delta}{t}}-\sqrt{\frac{\Delta}{t}}\right). \end{aligned} \tag{37}$$

For free Brownian particle, the normalized correlation function is $g^{\rm free} = 1$, since $\langle x(t+\Delta)x(t)\rangle_{\rm free} = \langle x^2(t)\rangle_{\rm free}$. When $\Delta/t = 0$, the correlation function is normalized $g^{\rm lat} = g^{\rm uni} = g^{\rm free} = 1$ (see Fig. 7). When $\Delta/t \to \infty$, we get

$$\begin{aligned} g^{\rm lat} &\to 0\\ g^{\rm uni} &\to \tfrac{1}{2}. \end{aligned} \tag{38}$$

This result emphasizes the strong dependence of initial conditions at long time limit.

B. The harmonization method

Up until now, we considered hard-core interactions between particles and used the Jepsen line to find the MSD. To find the two times correlation function, we follow Lizana *et al.* [30] and approximate general interactions into harmonic interactions between particles.

The Langevin equation that describes the motion of the nth particle is

$$\xi\frac{dx_n(t)}{dt} = \sum_{n'} F[x_n(t) - x_{n'}(t)] + \eta_n(t) + f_n(t), \quad (39)$$

where $x_n(t)$ is the position of the nth particle at time t and ξ is the friction constant

$$D = \frac{k_B T}{\xi}. \quad (40)$$

$F = -\frac{\partial V}{\partial x_n}$ is the force due to the interaction between particles, where the interaction potential between the nth particle and the n'th particle is $V(|x_n(t) - x_{n'}(t)|)$, and the potential V has singularity when $x_n(t) = x_{n'}(t)$, so particles cannot pass each other. η_n is white Gaussian noise with mean zero and covariance $\langle\eta_n(t)\eta_{n'}(t')\rangle = 2\xi k_B T\delta(t - t')\delta_{nn'}$, and f_n is an external force.

The main idea of the method is mapping the many-body problem into a solvable harmonic chain, by mapping the system into a system with beads interconnecting with harmonic springs (see Fig. 1). We convert the general interactions term in Eq. (39) to forces from the nearest-neighbors spring coupling, i.e.,

$$\xi\frac{dx_n(t)}{dt} = \kappa[x_{n+1}(t) + x_{n-1}(t) - 2x_n(t)] + \eta_n(t) + f_n(t), \quad (41)$$

where the effective spring constant κ for one-dimensional point-like particles with hard-core interactions is [30]

$$\kappa = \frac{N^2 k_B T}{L^2}. \quad (42)$$

In Ref. [30], κ is given in terms of the compressibility, which means that we can treat a general type of interaction (beyond hard-core). Equation (41) is the Edwards-Wilkinson equation, whose relation to single-file diffusion was uncovered in Refs. [34,35]. Under the assumption that the particles interact with others,

$$t \gg \tau_{\rm int} = \frac{L^2}{DN^2}, \quad (43)$$

we can take the continuum limit and turn $x_n(t)$ into a field $x(n,t)$ with the equation

$$\xi\frac{\partial x(n,t)}{\partial t} = \kappa\frac{\partial^2 x(n,t)}{\partial n^2} + \eta(n,t) + f(n,t). \quad (44)$$

Taking the Fourier ($n \to q$) and Laplace ($t \to s$) transforms,

$$x(q,s) = \int_{-\infty}^{\infty} dn \int_0^{\infty} dt e^{-iqn-st} x(n,t), \quad (45)$$

of Eq. (44) gives

$$x(q,s) = \frac{\xi x(q,t=0) + \eta(q,s) + f(q,s)}{\xi s + \kappa q^2}. \quad (46)$$

Equation (44) is the Rouse chain model, which describes polymer dynamics. In that well-known model, each monomer end is treated as a bead while the interaction between the beads is harmonic [36].

C. Evaluation of the correlation function

We now calculate the correlation function $\langle x_T(t+\Delta)x_T(t)\rangle$ by calculating $\langle x(q,s)x(q',s')\rangle$ first and then using inverse Fourier and Laplace transforms. We assume that there is no external force, $f(n,t) = 0$, and the initial condition is $x(n,t=0) = na$, so initially particles are on a lattice.

Using Eq. (46) gives

$$\langle x(q,s)x(q',s')\rangle = A_{\rm noise}(q,q',s,s') + A_{\rm init}(q,q',s,s'), \quad (47)$$

where the dependence on the initial condition is

$$A_{\rm init}(q,q',s,s') = \frac{\xi\langle x(q,t=0)x(q',t'=0)\rangle}{(\xi s + \kappa q^2)(\xi s' + \kappa q'^2)}, \quad (48)$$

and the noise term is

$$A_{\rm noise}(q,q',s,s') = \frac{\langle\eta(q,s)\eta(q',s')\rangle}{(\xi s + \kappa q^2)(\xi s' + \kappa q'^2)}. \quad (49)$$

Using the inverse Fourier transform $\mathcal{F}^{-1}\{\frac{2a}{a^2+q^2}\} = e^{-a|n|}$ and the convolution theorem,

$$A_{\rm init}(n,n',s,s') = \frac{\xi}{\kappa\sqrt{ss'}}\int_{-\infty}^{\infty} dm \int_{-\infty}^{\infty} dm' e^{-\sqrt{\frac{s\xi}{\kappa}}|n-m|} \times e^{-\sqrt{\frac{s'\xi}{\kappa}}|n'-m'|}\langle x(m,t=0)x(m',t=0)\rangle. \quad (50)$$

When particles are initially on a lattice,

$$\langle x(m,t=0)x(m',t=0)\rangle_{\rm lat} = a^2 mm' \quad (51)$$

(m and m' are integration variables in the spatial domain). We are interested in the tagged particle, so we integrate Eq. (50) for $n = n' = 0$. Therefore,

$$A_{\rm init}(n = n' = 0,s,s') = 0, \quad (52)$$

since $\int_{-\infty}^{\infty} e^{-|n-m|} m dm = 0$ for $n = 0$.

The variance of the Gaussian noise in q and s space is

$$\langle\eta(q,s)\eta(q',s')\rangle = \frac{4\pi\xi k_B T\delta(q+q')}{s+s'}; \quad (53)$$

thus, we can write

$$A_{\rm noise}(q,q',s,s') = \frac{4\pi\xi k_B T\delta(q+q')}{(\xi s + \kappa q^2)(\xi s' + \kappa q'^2)(s+s')}. \quad (54)$$

The inverse Laplace transform gives

$$A_{\rm noise}(q,q',t,t') = \frac{2\pi k_B T\delta(q+q')}{\kappa q^2}\left(-e^{-\frac{\kappa q^2}{\xi}(t+t')} + e^{-\frac{\kappa q^2}{\xi}|t-t'|}\right). \quad (55)$$

Inverse Fourier transform of Eq. (55) gives the covariance of the location of the tagged particle, i.e., $n = n' = 0$,

$$\langle x_T(t+\Delta)x_T(t)\rangle_{\rm lat} = \frac{k_B T}{\sqrt{\kappa\xi\pi}}(\sqrt{2t+\Delta} - \sqrt{\Delta}). \quad (56)$$

Returning to the original parameters using Eq. (42) and the Einstein relation Eq. (40) gives Eq. (34):

$$\langle x_T(t+\Delta)x_T(t)\rangle_{\text{lat}} = a\sqrt{\frac{D}{\pi}}(\sqrt{2t+\Delta}-\sqrt{\Delta}). \tag{57}$$

The derivation of the correlation function for the uniform case, Eq. (35), is done similarly, and we do not include it here since it is straightforward.

V. VIOLATION OF THE EINSTEIN RELATION

We would like to examine if the mean displacement of the tagged particle $\langle x_T(t)\rangle_F$ with the presence of constant external force F and the MSD without external force $\langle x_T^2(t)\rangle_{F=0}$ obey the generalized Einstein relation [37]:

$$\langle x_T(t)\rangle_F = F\frac{\langle x_T^2(t)\rangle_{F=0}}{2k_BT}. \tag{58}$$

This is a general relation valid within linear response theory. However, now that we find sensitivity to initial preparation of the system, the generalized Einstein relation must be checked.

For constant weak external force $f(t)=F_0$, the mean displacement is (see derivation later)

$$\langle x_T(t)\rangle_{F_0}^{\text{lat}} = \frac{aF_0\sqrt{Dt}}{k_BT\sqrt{\pi}} \tag{59}$$

and

$$\langle x_T(t)\rangle_{F_0}^{\text{uni}} = \frac{F_0\sqrt{Dt}}{\rho k_BT\sqrt{\pi}}. \tag{60}$$

The uniform case was treated previously in Refs. [30,38]. Unlike the MSD, here we do not see any impact of the initial condition on the response $\langle x_T(t)\rangle$. Namely, here we get the expected result: if we set $a=\rho^{-1}$, both results are identical (see Fig. 8).

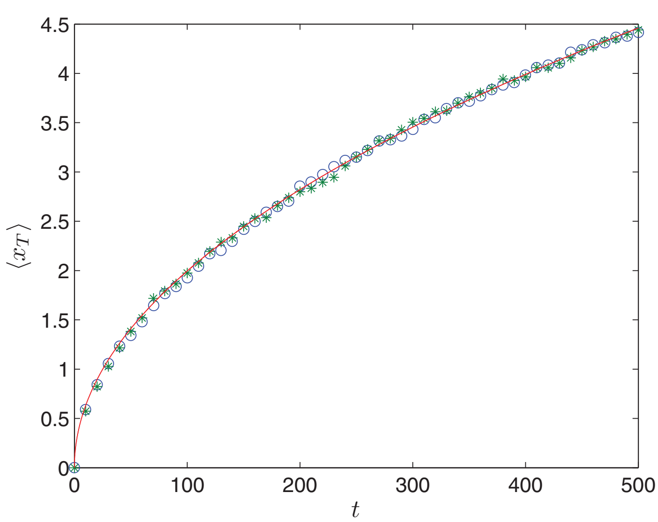

FIG. 8. (Color online) Simulations of the mean-displacement of the tagged particle when $F_0=0.5$ for the two cases: the uniform distribution ($*$) and the lattice initial conditions ($\circ$). The solid line represents the theory Eq. (71). The mean-displacement behaves similarly for the two types of initial conditions as is explained in the text.

Comparing the results with Eq. (58) gives an adequacy of Einstein relation for thermal equilibrium initial condition (uniform distribution) and violation for the nonthermal initial condition (lattice):

$$\begin{aligned}\langle x_T(t)\rangle_{F_0}^{\text{uni}} &= F_0\frac{\langle x_T^2(t)\rangle_{F_0=0}^{\text{uni}}}{2k_BT}\\ \langle x_T(t)\rangle_{F_0}^{\text{lat}} &= F_0\frac{\langle x_T^2(t)\rangle_{F_0=0}^{\text{lat}}}{\sqrt{2}k_BT};\end{aligned} \tag{61}$$

notice the factor $\sqrt{2}$. In the derivation of the mean-displacement, it was assumed that the external force is weak, $F_0 \ll \frac{k_BT}{a}$, since we assumed the system is near equilibrium. For strong force, our theory will fail.

A. The mean-displacement in presence of external force

We now treat the case where a constant force F acts on the tagged particle, using the Harmonization method of Lizana *et al.* [30]. Taking the ensemble average over Eq. (46) and using the fact that $\langle\eta(q,s)\rangle=0$ for white Gaussian noise,

$$\langle x(q,s)\rangle_f = B_{\text{init}}(q,s)+B_f(q,s), \tag{62}$$

where the dependence of the initial state is

$$B_{\text{init}}(q,s) = \frac{\xi\langle x(q,t=0)\rangle}{\xi s+\kappa q^2}, \tag{63}$$

and the force dependence is

$$B_f(q,s) = \frac{\langle f(q,s)\rangle}{\xi s+\kappa q^2}. \tag{64}$$

First we evaluate the initial conditions term $B_{\text{init}}(q,s)$. The inverse Fourier transformation $\mathcal{F}^{-1}\{\frac{2a}{a^2+q^2}\}=e^{-a|n|}$ and the convolution theorem give

$$B_{\text{init}}(n,s) = \frac{1}{2}\sqrt{\frac{\xi}{\kappa s}}\int_{-\infty}^{\infty}dm e^{-\sqrt{\frac{\xi s}{\kappa}}|n-m|}\langle x(m,t=0)\rangle. \tag{65}$$

The mean initial position is the same in both initial conditions, the equilibrium state and the lattice state, i.e., $\langle x(n,t=0)\rangle_{\text{uni}} = \langle x(n,t=0)\rangle_{\text{lat}} = na$ (note $a=\rho^{-1}$):

$$B_{\text{init}}^{\text{lat}}(n,s) = B_{\text{init}}^{\text{uni}}(n,s) = \sqrt{\frac{\xi}{4\kappa s}}\int_{-\infty}^{\infty}dm e^{-\sqrt{\frac{\xi s}{\kappa}}|n-m|}ma, \tag{66}$$

for the tagged particle $n=0$; therefore, the contribution from initial state term will vanish:

$$B_{\text{init}}^{\text{lat}}(0,s) = B_{\text{init}}^{\text{uni}}(0,s) = \sqrt{\frac{\xi}{4\kappa s}}\int_{-\infty}^{\infty}dm e^{-\sqrt{\frac{\xi s}{\kappa}}|m|}ma = 0. \tag{67}$$

We see that the response is not sensitive to initial conditions, while the MSD is.

For the force term $B_f(q,s)$, the inverse Fourier transform is

$$B_f(n,s) = \frac{1}{2\sqrt{\kappa\xi s}}\int_{-\infty}^{\infty}dm e^{-\sqrt{\frac{\xi s}{\kappa}}|n-m|}\langle f(m,s)\rangle, \tag{68}$$

since the external force acting on the tagged particle only, $f(m,s) = \delta(m) f(s)$ (notice that the external force generally depends on the time), we find

$$B_f(n=0,s) = \frac{1}{2\sqrt{\kappa\xi s}} f(s). \tag{69}$$

Taking inverse Laplace transform ($s \to t$) gives

$$\langle x_T(t)\rangle = \frac{1}{2(\kappa\xi\pi)^{1/2}} \int_0^t \frac{1}{\tau^{1/2}} f(t-\tau)d\tau. \tag{70}$$

For constant external force, $f(t) = F_0$, the mean displacement is

$$\langle x_T(t)\rangle_{F_0}^{\text{uni}} = \langle x_T(t)\rangle_{F_0}^{\text{lat}} = \frac{F_0\sqrt{t}}{\sqrt{\pi\xi\kappa}}. \tag{71}$$

Returning to the original parameters using Eqs. (40) and (42) gives

$$\langle x_T(t)\rangle_{F_0}^{\text{uni}} = \langle x_T(t)\rangle_{F_0}^{\text{lat}} = \frac{aF_0\sqrt{Dt}}{k_B T\sqrt{\pi}}. \tag{72}$$

Again, here $a = \rho^{-1}$ is the mean spacing between particles.

VI. TIME AVERAGE MSD

In experiments in many cases we measure an average over time [39]. Hence, we investigate the time average behavior of the tagged particle's location:

$$\overline{\delta^2(\Delta)} = \frac{1}{t-\Delta}\int_0^{t-\Delta} dt'[x_T(t'+\Delta) - x_T(t')]^2, \tag{73}$$

for the unbiased case, $F_0 = 0$. For some anomalous processes $\overline{\delta^2} \neq \langle x^2\rangle$ (see, e.g., Ref. [40]). If the time average $\overline{\delta^2}$ is equal to the ensemble average $\langle x_T^2\rangle$ in infinite measurement times, the system is called ergodic in the MSD sense. First, we find $\langle\overline{\delta^2(\Delta)}\rangle$. There is a relation between the correlation function we obtained in the previous sections and $\langle\overline{\delta^2(\Delta)}\rangle$:

$$\langle\overline{\delta^2(\Delta)}\rangle = \frac{1}{t-\Delta}\int_0^{t-\Delta} dt'\big[\langle x_T^2(t'+\Delta)\rangle + \langle x_T^2(t')\rangle - 2\langle x_T(t'+\Delta)x_T(t')\rangle\big]. \tag{74}$$

Using Eqs. (2) and (34) for $\langle x_T(t+\Delta)x_T(t)\rangle$ and $\langle x_T^2\rangle$ gives

$$\langle\overline{\delta^2(\Delta)}\rangle_{\text{lat}} = a\sqrt{\frac{D}{\pi}}\frac{2^{3/2}}{3}\left[\frac{t^{3/2}}{t-\Delta} + (t-\Delta)^{1/2} - \frac{(2t-\Delta)^{3/2}}{2^{1/2}(t-\Delta)} + \frac{3}{2^{1/2}}\Delta^{1/2} + (2^{-1/2}-1)\frac{\Delta^{3/2}}{t-\Delta}\right] \tag{75}$$

for lattice initial condition. When $\Delta \ll t$,

$$\langle\overline{\delta^2(\Delta)}\rangle_{\text{lat}} = 2a\sqrt{\frac{D}{\pi}}\sqrt{\Delta}\left\{1 + \frac{1-\sqrt{2}}{3}\frac{\Delta}{t} + o\left[\left(\frac{\Delta}{t}\right)^{3/2}\right]\right\}. \tag{76}$$

When the system is in equilibrium state,

$$\langle\overline{\delta^2(\Delta)}\rangle_{\text{uni}} = 2\rho^{-1}\sqrt{\frac{D}{\pi}}\sqrt{\Delta}. \tag{77}$$

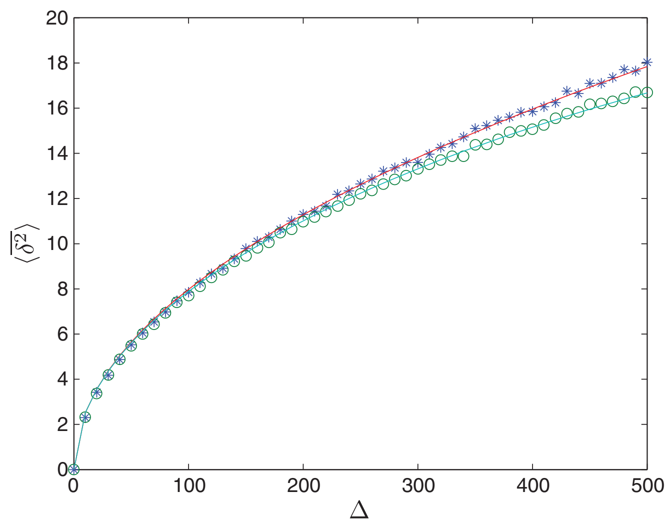

FIG. 9. (Color online) The time average $\langle\overline{\delta^2}\rangle$ when particles are in equilibrium state (upper curve) is compared with the time average when particles are initially situated with equal distances without randomness (lower curve). The simulations sampling is represented by the $\circ$ (lattice case) and $*$ (uniform case), and the theory by solid lines [Eqs. (75) and (77)]. The time measurement is $t = 1000$. When $\Delta/t \ll 1$, the two ensembles give identical results, namely there is no sensitivity to the initial condition.

Hence, when $\Delta \ll t$, we find $\langle\overline{\delta^2}\rangle_{\text{lat}} = \langle\overline{\delta^2}\rangle_{\text{uni}}$, where $\rho^{-1} = a$ is the average spacing between particles. We see that the ensemble averaged time average MSD is not sensitive to the way the system was prepared, when $\Delta \ll t$ (see Fig. 9). On the other hand, we find $\langle\overline{\delta^2}\rangle_{\text{lat}} \neq \langle x_T^2\rangle_{\text{lat}}$.

A. Relation to fractional Brownian motion

In equilibrium initial condition, it was shown that the problem can be mapped into fractional Brownian motion (fBm) with Hurst parameter $H = 1/4$ [30,41–46]. Hence, the process is ergodic in the MSD sense, i.e., $\langle\overline{\delta^2}\rangle$ is equal to the ensemble average $\langle x_T^2\rangle$ and the variance of $\overline{\delta^2}$ decays to zero when the measurement time is long [33]. Therefore, in an experiment with equilibrium initial condition, the time average $\overline{\delta^2}$ equals ensemble average. Note that the universality of fBm is restricted, since if the initial condition is not equilibrium state, we get a different behavior for the MSD.

VII. SUMMARY AND CONCLUSIONS

The generalized diffusion coefficient $D_{1/2}$ defined in $\langle x_T(t)^2\rangle \sim 2D_{1/2}t^{1/2}$ is sensitive to the initial preparation of the system. Hence, the assumption that in the long time limit initial conditions do not influence the asymptotic behavior, and that diffusivity is not sensitive to the method of preparation of the bath, is wrong. Moreover, we find a sensitivity to the initial conditions for two-time correlation function. On the other hand, in the presence on weak external force we find that the two initial conditions give the same result for the mean-displacement of the tagged particle since it depends on the mean initial conditions. We can conclude that the long-time behavior of the tagged particle is determined by the initial condition, in the absence of a force field, in the sense that the system always "remembers" its initial state. Similarly, the time-averaged MSD when $\Delta \ll t$ is not sensitive to the initial preparation. It might be worthwhile investigating the *fluctuations* of $\overline{\delta^2}$ and x_T in presence of a field, for the two types of initial conditions. A comparison between thermal and nonthermal initial condition was discussed recently [47,48]. The elastic model in Ref. [47] and the mathematical details are different than those considered here, but the main conclusion, that initial conditions have ever lasting effect, is shared. It seems that nonequilibrium quantities of many-body interacting system, like $D_{1/2}$, may depend on initial conditions, at least in some cases, so it would be nice to find more examples to this effect, e.g., the dynamics of a tagged particle in one dimension evolving according to Hamilton's laws without stochastic assumptions beyond the random initial condition [49].

How important are the initial condition for determining statistical properties of many-body systems? From equilibrium we know that Gibbs measure is not sensitive to initial preparation except for the total energy of the system.

Consider a symmetric simple exclusion process with step initial condition, where the occupation probability to the left of the origin, ρ_L, is different from the occupation probability to its right, ρ_R. Interestingly, the total flux of particles through the origin depends on ρ_L and ρ_R, i.e., the initial state affects the dynamics even for long times [29]. Notice that in this case, the initial conditions are not translation invariant. Thus, the choice of initial condition in this case induces a breaking of symmetry, which does not vanish, even in the long time limit. We call this effect an everlasting nontranslation invariant initial condition.

As is mentioned in the introduction, the sensitivity to the fluctuations of the initial preparation of a system was investigated by van Beijeren for a model of asymmetric simple exclusion process [26]. He compared between a random initial condition versus a fixed initial condition, where the same initial configuration was taken without randomness. This model is related to other models such as random matrices theory (see, e.g., Refs. [27,28]).

Therefore, we can classify two types of sensitivities to the initial condition. The first is revealed by comparison between different initial configurations, which depend on ρ_L and ρ_R, as is considered in the work of Derrida and Gerschenfeld [29]. The second type is the one discussed by van Beijeren [26], where he compares between different initial states with the same average and discusses how the *fluctuations* of the initial condition affect the dynamic properties. We call this type of effect fluctuation-induced sensitivity to initial conditions. Our work provides a quantative estimate to the effect, a factor of $\sqrt{2}$, which in principle can be checked in the laboratory.

An open question remains as to whether the results of Derrida and Gerschenfeld will be different in the following two cases: (a) The particles are initially positioned on a lattice, where the distance between particles is $1/\rho_L$ when $x < 0$ and $1/\rho_R$ otherwise; (b) the particles have densities ρ_L and ρ_R (exponential distribution of inter particles distances). In these two cases, the initial conditions *on average* are the same. As far as we know, no one has compared these two types of scenarios.

ACKNOWLEDGMENT

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APPENDIX A: SIMULATIONS DETAILS

As mentioned in the text, in one dimension, a hard-core collision event is equivalent to two identical particles that pass through each other, and after the particles cross each other, the labels of the two particles are switched. Instead of relabeling the particles after every collision, we let particles pass through each other, and then at time t, we relabel our particles. So, in fact, in the interval $(0,t)$ we view the particles as noninteracting. This fact simplifies the simulation process by ignoring the collisions during $(0,t)$, and sorting the particles labels at time t, and not after each collision.

We choose the system to be continuum in space. Each sampling time we move each particle with its probability density function:

$$P[x_n(t+\Delta)|x_n(t)] = \frac{1}{\sqrt{4\pi D\Delta}} e^{-\frac{[x_n(t+\Delta)-x_n(t)]^2}{4D\Delta}}, \quad \text{(A1)}$$

where n is the particle's label ($n \in \{-N \ldots N\}$), and $x_n(t)$ is the nth particle's location at the previous step. For each sampling time, we sort the particles and find the tagged one. For simulations we took 10^5 identical systems. Each system contains 10 001 identical particles. Notice that the system is finite, therefore, we need to consider times that will be short enough so that the particle will not reach the edges, $\sqrt{Dt} \ll L$, and long enough that particles interact with each other $a \ll \sqrt{Dt}$. For all the simulations, the distance between particles (or average distance for the uniform distribution initial state) was taken to be $L/N = 1$. The free particle diffusion coefficient was $D = 1/2$. For simulations we used MATLAB programming and used its standard sort and random numbers generator functions.

For the biased case, the tagged particle (the central particle) has probability density function

$$P[x_T(t+\Delta)|x_T(t)] = \frac{1}{\sqrt{4\pi D\Delta}} e^{-\frac{[x_T(t+\Delta)-x_T(t)-\frac{F_0 D\Delta}{k_B T}]^2}{4D\Delta}} \quad \text{(A2)}$$

instead of the PDF at Eq. (A1). The thermal energy is taken to be $k_B T = 1$.

APPENDIX B: NORMAL DISTRIBUTION OF α

To prove that $\alpha = \sum_{i=1}^{N} \delta\alpha_i$ is normally distributed when $N \longrightarrow \infty$, we define the kurtosis:

$$\gamma_2 = \frac{k_4}{(k_2)^2} = \frac{\mu_4 - 3\mu_2^2}{(\mu_2)^2}, \quad \text{(B1)}$$

where the μ_k is the kth central moment of α and is defined as

$$\mu_k = \langle (\alpha - \langle \alpha \rangle)^k \rangle. \quad \text{(B2)}$$

For normal distribution we expect that $\gamma_2 \longrightarrow 0$. First we find the 4th central moment μ_4. Using the multinomial formula and the fact that the first moment of $\delta\alpha_n$ is zero, i.e., $\langle\delta\alpha_n - \langle\delta\alpha_n\rangle\rangle = 0$, gives

$$\mu_4 = \sum_{n=1}^{N}\langle(\delta\alpha_n - \langle\delta\alpha_n\rangle)^4\rangle + 6\sum_{n<m}\langle(\delta\alpha_n - \langle\delta\alpha_n\rangle)^2\rangle\langle(\delta\alpha_m - \langle\delta\alpha_m\rangle)^2\rangle. \quad \text{(B3)}$$

For the 2nd central moment we find

$$\mu_2 = \sum_{n=1}^{N}\left\langle\delta\alpha_n^2\right\rangle - \langle\delta\alpha_n\rangle^2; \quad \text{(B4)}$$

therefore, the numerator in Eq. (B1) is

$$k_4 = \sum_{n=1}^{N}\langle(\delta\alpha_n - \langle\delta\alpha_n\rangle)^4\rangle - 3\sum_{n=1}^{N}\left(\left\langle\delta\alpha_n^2\right\rangle - \langle\delta\alpha_n\rangle^2\right)^2. \quad \text{(B5)}$$

Equation (5) gives

$$\delta\alpha_n = \begin{cases} 1 & P_{RL}(an)P_{LL}(-an) \\ 0 & P_{RL}(an)P_{LR}(-an) + P_{RR}(an)P_{LL}(-an). \\ -1 & P_{RR}(an)P_{LR}(-an) \end{cases} \quad \text{(B6)}$$

Therefore,

$$\langle\delta\alpha_n\rangle = \left\langle\delta\alpha_n^3\right\rangle = P_{RL}(na) - P_{LR}(-na)$$
$$\left\langle\delta\alpha_n^2\right\rangle = \left\langle\delta\alpha_n^4\right\rangle = P_{RL}(na)P_{LL}(-na) + P_{RR}(na)P_{LR}(-na). \quad \text{(B7)}$$

To evaluate k_4, we start with

$$\sum_{n=1}^{N} P_{RL}(na) = \sum_{n=1}^{N} Erf\left(\frac{vt - na}{\sqrt{2Dt}}\right) \propto N \quad \text{(B8)}$$

(generally, this term depends on the time t, we take $N \to \infty$ and t finite).

Similarly, we can prove that $\sum P_{LR} \propto N$, $\sum P_{LR}^2 \propto N$, etc.; hence, $k_4 \propto N$. we use the same derivation for the denominator and we get $k_2^2 \propto N^2$. Finally, we get

$$\gamma_2 \sim \frac{N}{N^2} \overset{N\to\infty}{\longrightarrow} 0. \quad \text{(B9)}$$

Therefore, α is normally distributed:

$$P(\alpha) = \frac{1}{\sqrt{2\pi\sigma_\alpha^2}} e^{-\frac{(\alpha-\langle\alpha\rangle)^2}{2\sigma_\alpha}}, \quad \text{(B10)}$$

where $\sigma_\alpha = \langle\alpha^2\rangle - \langle\alpha\rangle^2$.

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