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Mean-squared displacement of a hard-core tracer in a periodic system

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We show analytically and numerically that logarithmic contributions to the mean-squared displacement of a hard-core tracer particle in a finite two-dimensional periodic lattice can be considered as a transient, and only survive in the infinite system.

Memory effects in various fluid and fluidlike systems, resulting in asymptotic algebraic decay (long-time tails) of velocity correlation functions and in numerically measurable corrections to transport coefficients, have been the object of analytical and numerical study for more than 20 years.[1–6] The purpose of this work is to show that in the two-dimensional hard-core tracer problem[7] the long-time tail and the associated logarithmic correction to the mean-squared displacement (MSD or $\langle R^2\rangle$), only survive in a truly infinite system. In contrast, in finite systems the asymptotic behavior of the MSD is strictly linear.

Most of the analytical results of kinetic theory pertain to infinite systems. However, finite-system studies are important for two reasons: real physical systems are finite and the supporting numerical simulations are necessarily performed in finite systems. The few available results for finite lattices point to significant differences between finite and infinite systems. In standard fluids (continuum and lattice models), finite-system periodic boundary conditions have been shown to produce strong interference effects through sound modes.[3] In the one-dimensional tracer problem,[6] the asymptotic behavior of the MSD crosses over from $\sim t^{1/2}$, the infinite-system result, to $\sim t$. In what follows we examine finite-size effects in the two-dimensional tracer problem.

We begin by defining the model. A tagged random walker (the tracer) moves in a medium of similar particles, with a hard-core exclusion principle. We concentrate for simplicity on the case of isotropic jumping probability, with equal rates J for the tracer and the other walkers. It has been shown that, in the infinite-system limit, the velocity correlation function has an asymptotic t^{-2} behavior, leading to logarithmic corrections[4,5] to the linear dependence in time of the MSD of the tracer. This was supported by Monte Carlo (MC) simulations[5] done in fairly large systems (600×600). Great care was taken to ensure that the simulation times were smaller than the expected time needed for diffusion across the system. As boundary effects were avoided, the result of these simulations supports the existence of logarithmic corrections in an "infinite" system. We note that the logarithmic correction is of the same form as that predicted for another purely diffusive system, the Lorentz gas.[2]

The behavior of the model is governed by the master equation

$$\frac{dP_a(t)}{dt}=\sum_b W_{ab}P_b(t)-\sum_b W_{ba}P_a(t), \tag{1}$$

where $P_a(t)$ is the probability of the system being in configuration a at time t, and W_{ba} is the probability per unit time of a transition from configuration a to configuration b. The continuous-time process described by (1) is equivalent to a discrete-time process[8] which has a Poisson distribution in time with transition rate ϵ_0 and is governed by a transition matrix $\mathbf{A}$. We rewrite (1) in matrix form

$$\frac{d\mathbf{P}(t)}{dt}=\Omega\mathbf{P}, \tag{2}$$

where

$$\Omega_{ab}=W_{ab}, \tag{3}$$

$$\Omega_{aa}=-\sum_b W_{ba}. \tag{4}$$

The probabilities that at time t the system is in any of the possible configurations are completely specified by the elements of the propagator matrix $\mathbf{G}(t)=\exp(\Omega t)$. In terms of the discrete process this matrix assumes the form

$$\mathbf{G}(\mathbf{t})=\sum_{N=0}^{\infty}\mathbf{A}^N\frac{(\epsilon_0 t)^N}{N!}e^{-\epsilon_0 t}. \tag{5}$$

The relation between the matrices specifying the discrete and continuous-time processes is

$$\mathbf{A}=\frac{1}{\epsilon_0}(\Omega+\epsilon_0\mathbf{I}), \tag{6}$$

where $\mathbf{I}$ is the identity matrix. The only condition that has to be imposed on the transition rate ϵ_0 is that it should be greater than the largest diagonal element of Ω. It is convenient to fix ϵ_0 to be the maximum transition rate out of the possible configurations, i.e., $\epsilon_0=N_TZJ$, where N_T is the total number of particles and Z is the coordination number of the lattice. It is readily checked

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that the transition matrix $\mathbf{A}$ describes the discrete process simulated in the usual MC simulations of tracer diffusion.

From the Laplace transform of (5), the following relation is obtained:

$$(s+\epsilon_0)G(s)=\sum_{N=0}^{\infty}\mathbf{A}^N\left(\frac{\epsilon_0}{s+\epsilon_0}\right)^N. \qquad (7)$$

From (7) we see that the Laplace transform of the propagator matrix $\mathbf{G}$ is related to the generating function of the discrete process,

$$U(\lambda)=\sum_{N=0}^{\infty}\mathbf{A}^N\lambda^N, \qquad (8)$$

by the relations

$$(s+\epsilon_0)G(s)=U(\lambda), \qquad (9)$$

$$\frac{\epsilon_0}{s+\epsilon_0}=\lambda. \qquad (10)$$

In what follows we will find it more convenient to study the discrete-time process and find time-related quantities by use of (9) and (10).

We now consider the motion of a tracer in a finite square lattice with N_S sites, of which N_S-2 are occupied by background particles, one by the tracer, and one is vacant. As seen in Ref. 9, characteristics of the tracer motion can be obtained by considering a closed random walk of the whole system in a part of configuration space which is constituted by the possible configurations that the system can assume with the tracer fixed at a given position in the real-space lattice. This is accomplished by associating a factor $\exp(\pm i\theta_j)$ to each transition which involves a displacement of the tracer in a direction $\pm j$ in real space. In the case considered here, the configurations can be labeled by the position of the vacancy. The random walk performed by the system is, therefore, equivalent to a random walk performed by the vacancy in a frame in which the tracer is at rest. Since each tracer-vacancy exchange in this frame corresponds to a jump of the vacancy by two lattice units and the site occupied by the tracer is not available to the vacancy, the vacancy performs a free-particle random walk in a regular lattice with a single defect. By considering the properties of this random walk it is possible to show that the Laplace transform of the tracer MSD is given by

$$R^2(s)=\frac{ZJ}{(N_S-1)s^2}\left(\frac{s+ZJ[1+\cos\theta(s)]}{s+ZJ[1-\cos\theta(s)]}\right), \qquad (11)$$

where

$$\cos\theta(s)=\frac{-1}{ZN_S}\sum_{k_1=0}^{L-1}\sum_{k_2=0}^{L-1}\frac{1-e^{-i4\pi k_1/L}}{1-\frac{ZJ}{2(s+ZJ)}(\cos\frac{2\pi k_1}{L}+\cos\frac{2\pi k_2}{L})}, \qquad (12)$$

where $L=N_S^{1/2}$ and the lattice constant is taken to be one.

In the limit $s\to 0^+$, (11) assumes the form

$$R^2(s)\sim\frac{JZf_0}{N_S-1}\frac{1}{s^2}+\frac{k}{s}, \qquad (13)$$

where

$$f_0=(1+\cos\theta_0)/(1-\cos\theta_0), \qquad (14)$$

$$k=-\frac{f_0}{N_S-1}\left(\frac{1}{1+\cos\theta_0}+\frac{1}{1-\cos\theta_0}\right)\left(\cos\theta_0-\frac{s_e}{2}\right), \qquad (15)$$

$$s_e=\frac{-1}{ZN_S}\sum_{k_1=0}^{L-1}\sum_{k_2=0}^{L-1}\frac{(1-e^{-i4\pi k_1/L})(\cos\frac{2\pi k_1}{L}+\cos\frac{2\pi k_2}{L})}{[1-\frac{1}{2}(\cos\frac{2\pi k_1}{L}+\cos\frac{2\pi k_2}{L})]^2}, \qquad (16)$$

with $\cos\theta_0=\cos\theta(0)$. Inversion of (13) immediately yields the form $\langle R^2\rangle\sim k+4Dt$, with k as given in (15) and $D=JZf_0/4(N_S-1)$. Hence, in a finite lattice with periodic boundary conditions the MSD of the tracer grows linearly with t as $t\to\infty$. The argument leading to (11)–(16) also applies to the single background particle case and to low concentrations of background particles or vacancies. The values obtained for the constant k and for the diffusion coefficient are confirmed essentially to machine precision by the exact-enumeration simulations described below.

In the limit $N_S\to\infty$, with a suitably small but finite concentration of vacancies, (11) and (12) become

$$R^2(s)=(1-c)\frac{ZJ}{s^2}\left(\frac{s+ZJ[1+\cos\theta(s)]}{s+ZJ[1-\cos\theta(s)]}\right), \qquad (17)$$

$$\cos\theta(s)=-\frac{1}{(2\pi)^2Z}\times\int\int\frac{1-e^{-i2\theta_1}}{1-\frac{ZJ}{2(s+ZJ)}(\cos\theta_1+\cos\theta_2)}d\theta_1 d\theta_2. \qquad (18)$$

It was shown in Ref. 5 that in this case the limit $s\to 0^+$ gives rise to a logarithmic correction, that is (11) assumes the form

$$R^2(s)=(1-c)ZJf_0\frac{1}{s^2}+\frac{B}{s}+E\frac{\ln s}{s}, \qquad (19)$$

where B and E are constants. We conclude that the logarithmic term in $\langle R^2(t)\rangle$ is only present in the infinite system. This term can be thought of as emerging from transient behavior in the finite lattice. Indeed, it gets longer with increasing N_S and ceases to be a transient in the limit $N_S\to\infty$. The emergence of new long-time behavior is indicated by the divergence of (16) in this limit.

Next, we describe the result of simulations for small $L\times L$ lattices, with $L\leq 12$. We have developed an exact

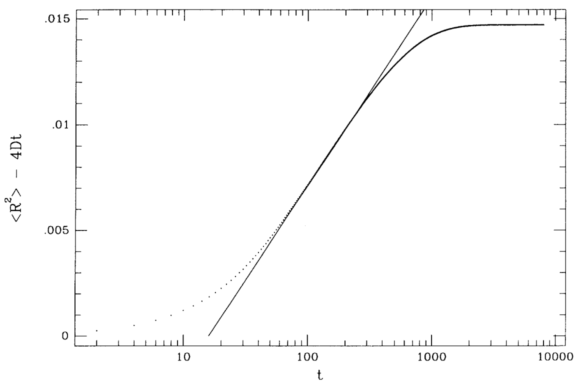

FIG. 1. Mean-squared displacement (minus linear part) vs time (logarithmic scale) for a single-vacancy 8×8 system, showing the logarithmiclike contribution to be a transient. The asymptotic expression for the MSD is $\langle R^2 \rangle \sim 0.0146968 + (1.22047772 \times 10^{-4})t$.

TABLE I. System size, onset time in trial steps, and Monte Carlo steps per particle (t_0, t_0') for linear behavior of the mean-squared displacement of the tracer particle, and onset time (MC steps) divided by the fourth power of system length, for single-vacancy tracer diffusion.

L	t_0 (MC steps)	t_0' (MC steps/particle)	t_0/L^4
3	60	8.6	0.74
4	150	10.0	0.59
5	400	17.4	0.64
6	700	20.6	0.54
8	2000	32.3	0.49
10	6000	61.2	0.60

enumeration technique[9,10] for measuring the MSD taking into account *all* possible histories of the system. As the tracer problem involves dynamical disorder, the method requires mapping the system into a single-particle walk in *configuration* space and is very memory-consuming. The advantage is that the results are free of statistical error and only limited by numerical error. Because of the memory requirements, we have concentrated on 3×3 periodic lattices (for arbitrary concentration of background particles) and on the single-vacancy and single background particle limits for larger lattices.

A typical plot of $\langle R^2 \rangle - 4Dt$ vs $\ln t$ is shown in Fig. 1. The system is an 8×8 periodic lattice containing the tracer, 62 background particles, and a single vacancy. In this case D can be calculated from (13), and the linear contribution $4Dt$ can be subtracted exactly. We see that a logarithmiclike contribution appears at intermediate times (corresponding to the straight line in the plot), only to disappear at even longer times, indicating that the MSD is asymptotically of the form $\langle R^2 \rangle \sim k + 4Dt$. It is instructive to compare this figure with Fig. 1 of Ref. 5, corresponding to a much larger system: the initial development of the logarithmic contribution is very similar in both cases.

An advantage of simulation over theory[11] is that we can see the time scale for crossover to linear behavior of the MSD. For constant density of background particles, one would expect that deviations from infinite-system behavior happen at times comparable to the diffusion time of the tracer across the system, given by L^2/D. However, for the case we considered (the single-vacancy problem), $D \sim 1/L^2$, and therefore finite-size effects should appear for $t \sim L^4$. We see from Table I that the times for the onset of linear behavior, obtained from visual estimation of plots similar to Fig. 1, appear to obey the scaling indicated by conventional wisdom.

In summary, we have provided analytical and numerical evidence indicating that the asymptotic dependence of the mean-squared displacement of a hard-core tracer particle in a periodic lattice is strictly linear in time. While we have only shown this explicitly in two dimensions, we expect that the expressions equivalent to (11)–(16) in higher dimensions should be similar in form.

This new result should be taken into account in two ways when doing numerical simulations of the tracer problem. First, if one is interested in measuring D, one should either simulate the system for times much shorter than the diffusion time of the tracer across the system, and fit the MSD with a logarithmic term, or simulate for times larger than the crossover to linear behavior and perform a purely linear fit. Plotting a figure similar to our Fig. 1 is recommended, but this may be impractical in the cases when D is not exactly known. Second, if one is interested in measuring the amplitude of the long-time tails, the times of measurement should be short enough to avoid both the linear and the crossover regimes. Again, this may take a considerable amount of trial and error. Finally, we conjecture that a similar crossover to purely linear diffusion may be observable in finite-system simulations of Lorentz gas models. In these the crossover may be more difficult to observe, as the onset of t^{-2} tails takes considerably longer than in the tracer problem.

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