Correlated anomalous diffusion: Random walk and Langevin equation

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A random walk model is formulated and examined which gives the correlated anomalous diffusion found in molecular dynamics simulations. The mean square displacement (MSD) shows a logarithmic behavior in one dimension. Corresponding Langevin equation is constructed by solving the inverse problem which gives a procedure to derive random impulse correlation from MSD function. © *2010 American Institute of Physics.* [doi:10.1063/1.3309329]

I. INTRODUCTION

Recently a new molecular dynamics (MD) method was introduced by one of the authors (Aoki[1]). This MD method, which utilizes a symplectic integrator, has an advantage that all the thermodynamic states are obtained through constant pressure and constant temperature processes and can even produce nonequilibrium steady states including some glass states.

Such a method was applied to the study of single-component soft-core repulsive particle system.[2] Some features and results of Ref. 2 are summarized as follows.

(1) A soft-core potential of finite radius is used with only repulsive part of Lennard-Jones-type potential.
(2) Simulation scheme employed is that of the Nosé–Poincaré Hamiltonian.
(3) Symplectic method is used to integrate the equations of motion with constant pressure and constant temperature.
(4) Three branches of nonequilibrium steady states are found. They are shown in the phase diagram Fig. 1, where (a), (b), and (c) are the newly found glass states, while (d) is the solid phase and (e) is the liquid phase.
(5) The properties of these states are analyzed in detail, such as specific volume, lattice structure, mean square displacement (MSD), and dynamical distribution functions (van Hove self-correlation function).

These results show some of the specific properties of glass state: the dynamical heterogeneities and the intermittent diffusivity. Especially in the obtained movies particles are observed to stay on the preconstructed lattice sites for a mean time, and suddenly jump onto the neighboring sites, showing a behavior of anomalous diffusion.

Anomalous diffusion observed in such single-component soft spherical particles MD could be

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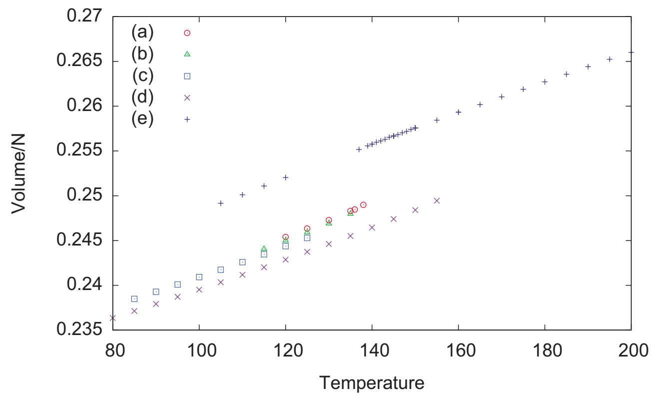

FIG. 1. (Color online) The phase diagram for the large ($N=2048$) system. Nonequilibrium steady states identified as the glass states are found [(a)–(c)].

examined by two models. The purpose of the present paper is to construct a random walk model simulating such anomalous diffusion and to analyze a Langevin equation model corresponding to it.

The first one is to construct a stochastic model, which will be discussed in Sec. II. Since particles move in a correlated manner each other, let us call this stochastic model *the correlated diffusion model*. The second model is to consider a Langevin equation which imitates a correlation effect and to solve such equation, which will be discussed in Sec. III.

II. RANDOM WALK MODEL OF ANOMALOUS DIFFUSION

A. Stochastic model of correlated diffusion

The model considered here is composed of many particles on a regular lattice (each site is empty or occupied by one particle). Additionally, the system proceeds under stochastic dynamics with the following rules.

(1) Particle always jumps to the neighboring site if it is empty.
(2) If there are many sites to jump, it chooses one of them with an equal probability.
(3) The order of jumps for particles is determined randomly for the molecular democracy.

In the simulations the algorithm called MERSENNE TWISTER (MT19937) of GNU Scientific Library (GSL) is used to generate quasirandom numbers.

B. Results of simulations

1. One dimensional case

While numerical simulations, the MSD function $M(t)$ and the van Hove functions $F(k,t)$ are computed. These quantities are defined as follows:

$$M(t) = \left\langle \frac{1}{N}\sum_{n=1}^{N} (x_n(t) - x_n(0))^2 \right\rangle, \tag{2.1}$$

$$F(k,t) = \left\langle \frac{1}{N}\sum_{n=1}^{N} e^{\mathrm{i}k(x_n(t)-x_n(0))} \right\rangle, \tag{2.2}$$

where $\langle \cdots \rangle$ implies the sample average. The number of sampling is taken as 10^5 typically.

For the particle number $N=1$ case, the problem can be analyzed rigorously, since the probability distribution function $P(x,t)$ obeys the difference equation

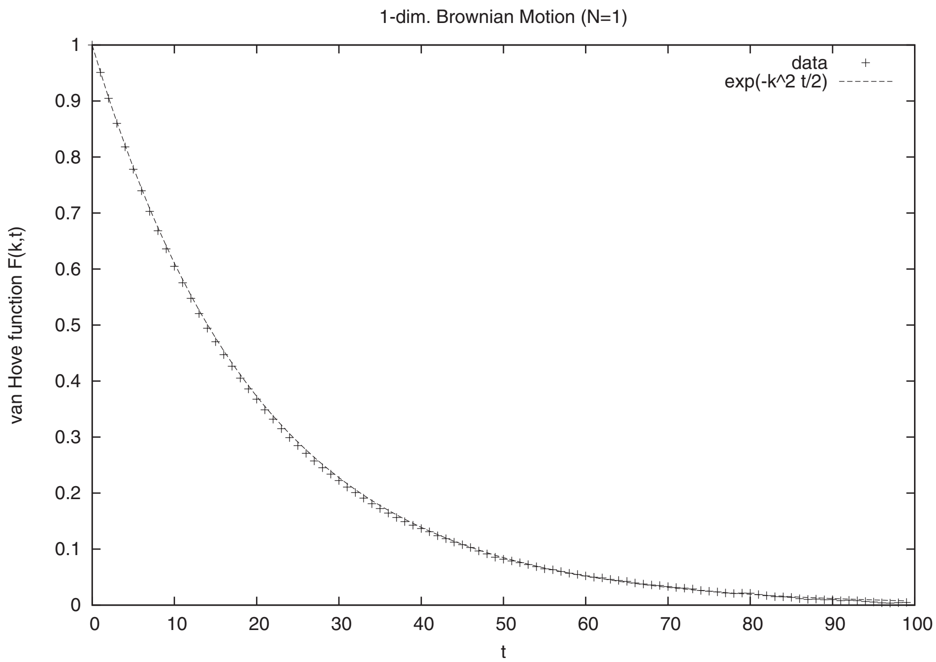

FIG. 2. The van Hove function $F(k,t)$ of $k=\pi/10$ for $N=1$.

$$P(x,t+1)=\tfrac{1}{2}(P(x+1,t)+P(x-1,t)), \tag{2.3}$$

which can be solved exactly. For example, the MSD function is given by

$$M(t)=\langle (x(t)-x(0))^2\rangle = t, \tag{2.4}$$

and the van Hove function $F(k,t)$ is computed as

$$F(k,t)=\exp\left(-\frac{k^2}{2}t\right) \quad (N=1 \ \text{case}). \tag{2.5}$$

Figure 2 shows the van Hove function $F(k,t)$ for $N=1$ and the theoretical result (2.5). The agreement is very well.

As the particle number N increases, the van Hove function $F(k,t)$ decays slowly as shown in Fig. 3.

The decay property of $F(k,t)$ is not exponential but power law such as $t^{-\gamma}$ for large t. This can be more directly understood by observing MSD, which is shown in Fig. 4. Since the motion of a particle is interrupted by other particles, the MSD becomes smaller than that of $N=1$.

Since the behavior of MSD in Fig. 4 as a function of t is estimated as

$$M(t)=\begin{cases}\alpha \log t & (t \gg t_0)\\ \beta t & (t \ll t_0),\end{cases} \tag{2.6}$$

with some t_0, the numerical results can be fitted by an interpolating function,

$$M(t)=a \log\left(1+\frac{t}{b}\right) \tag{2.7}$$

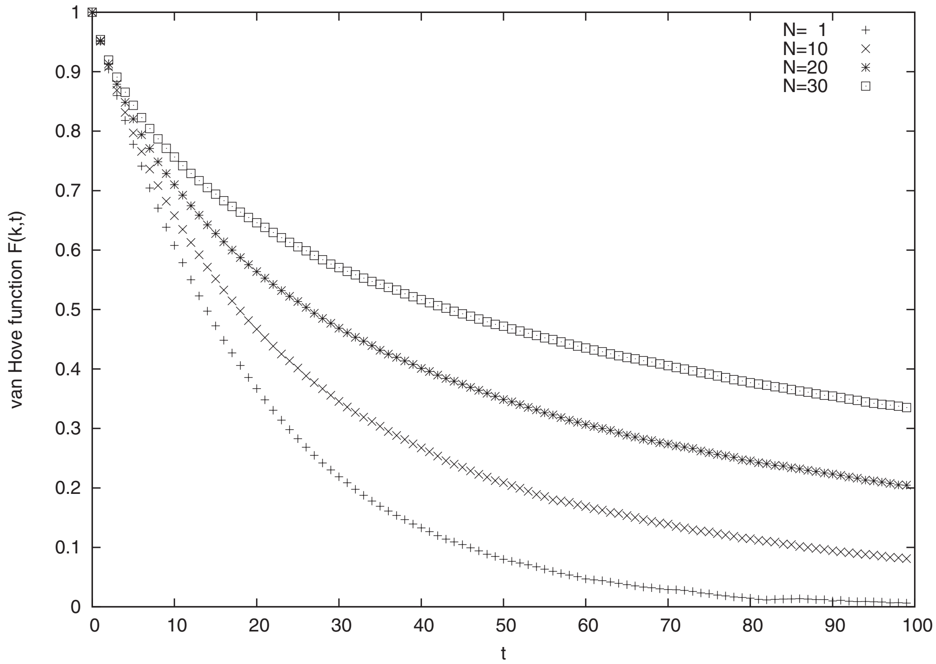

FIG. 3. The van Hove functions $F(k,t)$ of $k=\pi/10$ for several particle numbers N.

$$=\begin{cases} a\log(t/b) & (t \gg b) \\ (a/b)t & (t \ll b), \end{cases} \tag{2.8}$$

which is shown in Fig. 5.

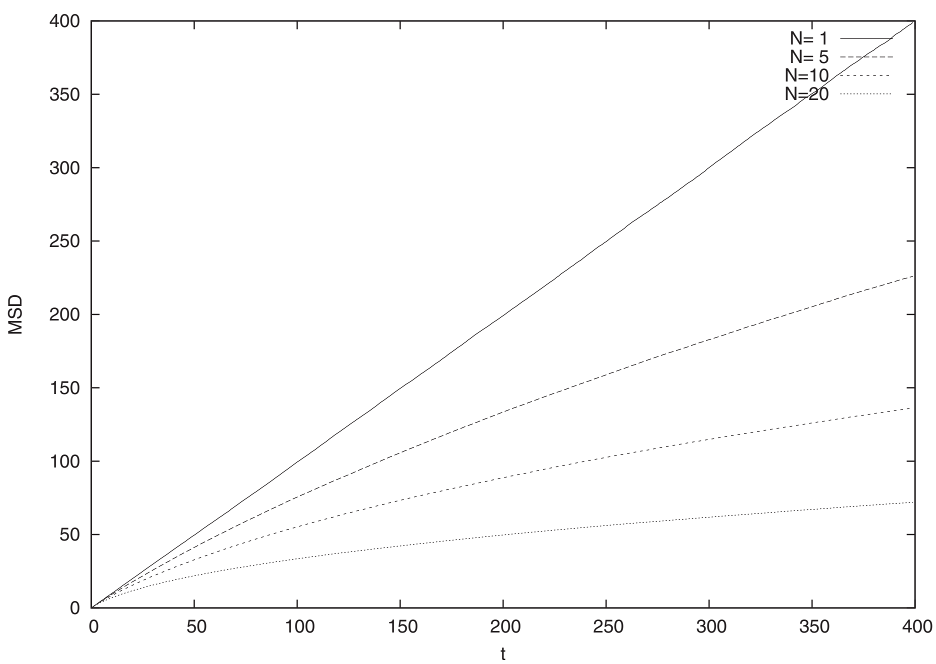

FIG. 4. MSD function for several particle numbers N.

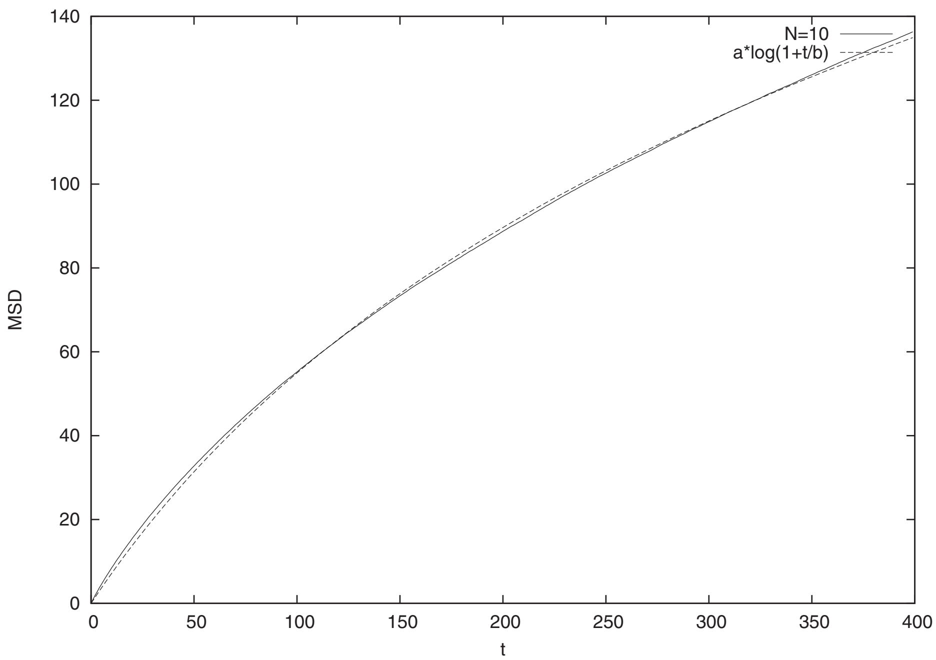

FIG. 5. Numerical results can be fitted by the function $a \log(1+t/b)$.

The coefficient a of fitting function $a \log(1+t/b)$ is shown for several values of the concentration $c=N/L$ (L=the number of sites) in Fig. 6, where the data are fitted well by the curve $a=\beta\cdot(1-c)/c$ with $\beta=10.2602$.

Now the van Hove function $F(k,t)$ is, in general, related with $M(t)$ as follows:

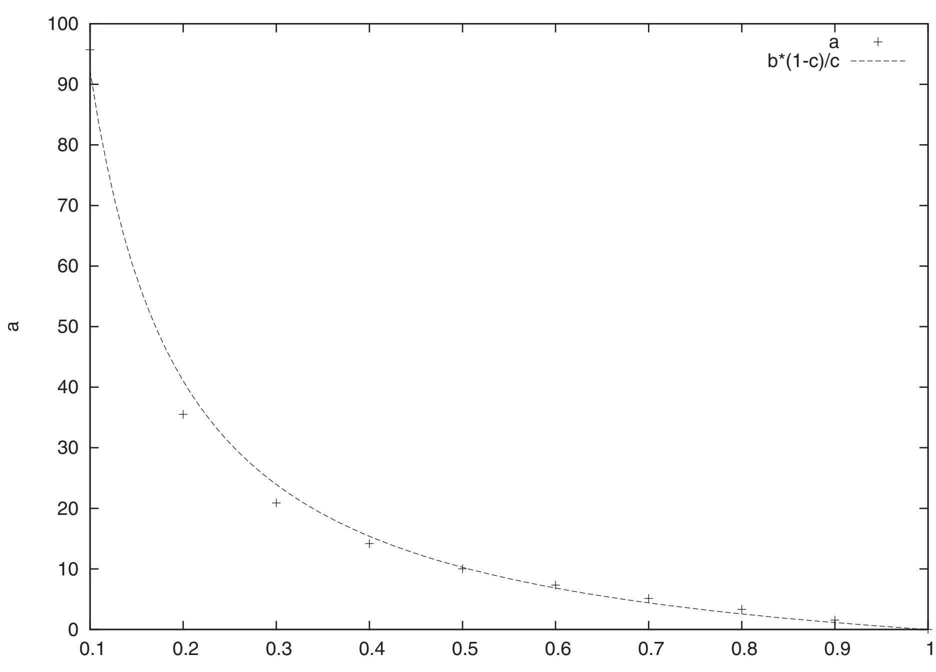

FIG. 6. Coefficient a of $M(t)=a \log(1+t/b)$ vs the concentration $c=N/L$. The data can be fitted well by the curve $a=\beta\cdot(1-c)/c$ with $\beta=10.2602$.

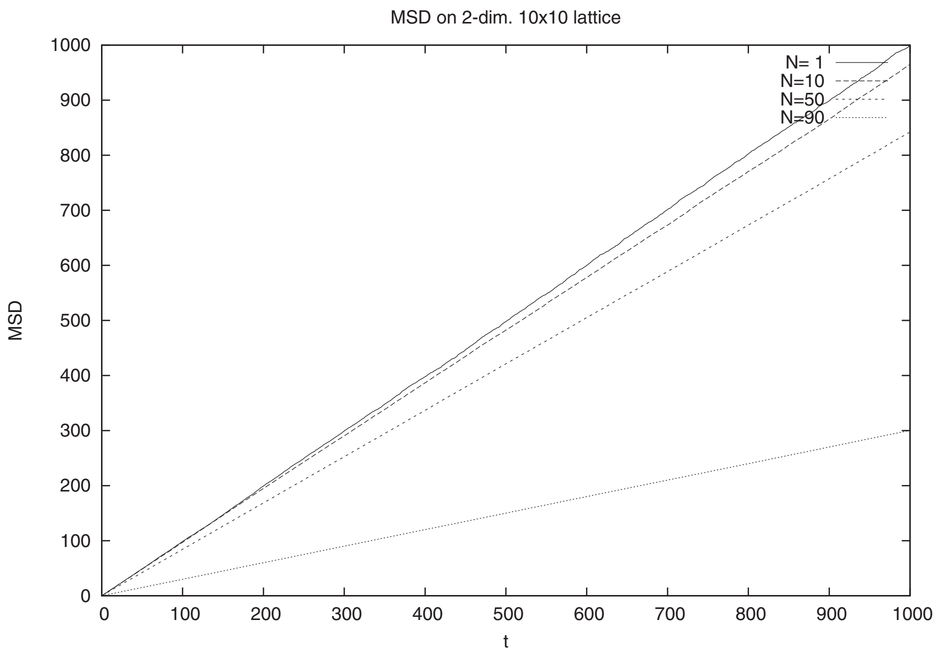

FIG. 7. MSD function in two dimension is of usual Brownian motion type.

$$F(k,t) = \langle \exp(\mathrm{i}k(x(t)-x(0)))\rangle = \exp\left(-\frac{k^2}{2}\langle (x(t)-x(0))^2\rangle\right) = \exp\left(-\frac{k^2}{2}\cdot M(t)\right), \tag{2.9}$$

where the second equality holds if our stochastic model is a Gaussian process.

Whether the process is Gaussian or not can be examined by checking whether the kurtosis defined by

$$\text{kurtosis} = \frac{m_4}{m_2^2} = \frac{\langle (x(t)-x(0))^4\rangle}{\langle (x(t)-x(0))^2\rangle^2} \tag{2.10}$$

is equal to 3 or not. Since typical value of this quantity is $3.0933\cdots$, for $N=10$, for example, our model can be considered as a Gaussian random process. In fact, the both sides of Eq. (2.9) agree well when we use the numerical results for both quantities. The van Hove function $F(k,t)$ decays as $t^{-\gamma}$ when $M(t)$ behaves as $\alpha \log t$ for large t, where the power is given by $\gamma=(k^2/2)\cdot\alpha$.

2. Two dimensional case

In two dimensional case the MSD function obtained is, surprisingly, different from that of one dimensional case. It has t linear behavior, as shown in Fig. 7. As before, the case of $N=1$ gives the rigorous $M(t)=t$ result as expected. Moreover, the slope decreases as the particle number N increases, which is a natural behavior due to mutual interruptions. The reason for this dimensionality dependent difference may be interpreted such that a particle in two dimensions can go around the other particles to diffuse.

Let us consider here a simple mean field theory by constructing an approximate Fokker–Planck equation. We set the particle number N, the number of sites L^2, and the particle concentration $c=N/L^2$, then the probability q that four neighboring sites are all occupied is estimated as $q=c^4$. Then we have

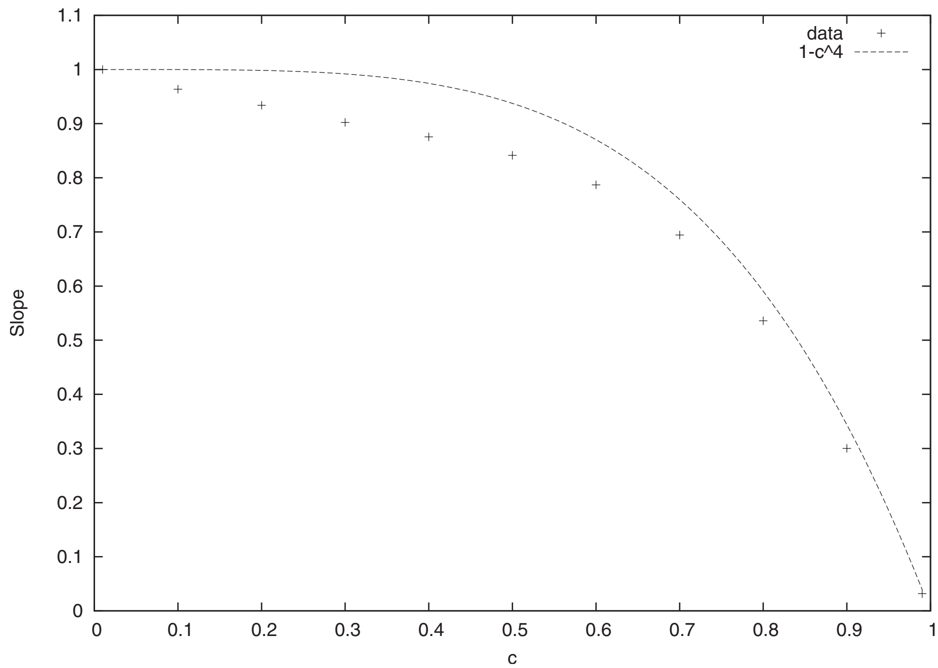

FIG. 8. The slope can be well approximated by $1-c^4$ of the mean field theory.

$$P(x,y,t+1) = p \cdot (P(x+1,y,t) + P(x-1,y,t) + P(x,y+1,t) + P(x,y-1,t)) + q \cdot P(x,y,t), \tag{2.11}$$

where the probability $p=(1-q)/4$. Then after some calculations we have

$$M(t) \equiv \langle (x(t)-x(0))^2 + (y(t)-y(0))^2 \rangle = (1-c^4) \cdot t, \quad (c = N/L^2). \tag{2.12}$$

This formula is compared with the simulations in Fig. 8, and the agreement is rather good.

C. Diffusion of a vacancy

It may be interesting to consider the case that the number of particles is less than the number of sites by 1. Then our model can be viewed as a diffusion of a vacancy. The result is given in Fig. 9, which shows an ordinary Brownian motion but with a larger slope. This is because that the vacancy can move to farther positions in a single time step.

III. LANGEVIN EQUATION FOR ANOMALOUS DIFFUSION

The random walk model discussed in Sec. II suggests that there might be an analytical theory by Langevin equation method. Let us consider a Langevin equation for *single* particle for which unknown random impulse is applied. In other words, we suppose that the correlated force from other particles can be treated to produce such random impulse. Moreover, we ask ourselves what kind of impulse correlation can give the observed MSD property. We formulate below such question as an inverse problem of MSD and solve the derived equation for several cases.

A. Langevin equation and inverse problem of MSD

Let us consider a Langevin equation

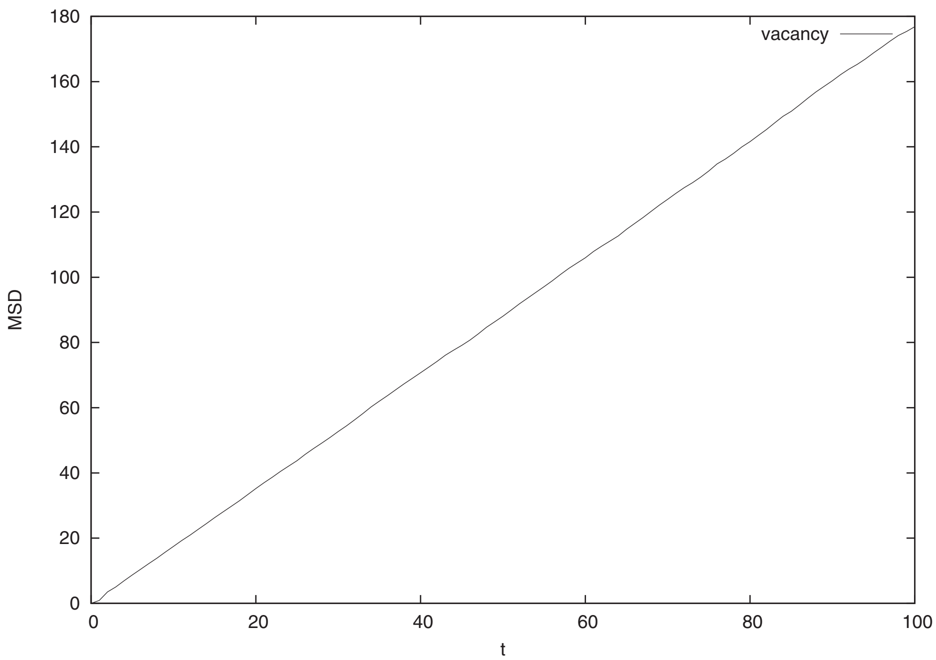

FIG. 9. MSD function of the vacancy is t linear with a larger slope.

$$\frac{\mathrm{d}x}{\mathrm{d}t} = f(t), \tag{3.1}$$

where the random function $f(t)$, a velocity produced by random impulsive force, is assumed to be a Gaussian process with

$$\langle f(t)\rangle = 0, \quad \langle f(t)f(t')\rangle = \psi(t-t'). \tag{3.2}$$

The impulse correlation $\psi(t)$ is assumed to be an even function: $\psi(-t)=\psi(t)$.

Since the differential equation (3.1) is integrated as

$$x(t) = x(0) + \int_0^t f(t')\mathrm{d}t',$$

the MSD function $M(t)$ is given by

$$M(t) = \langle (x(t)-x(0))^2\rangle = \int_0^t \mathrm{d}t_1 \int_0^t \mathrm{d}t_2 \langle f(t_1)f(t_2)\rangle = \int_0^t \mathrm{d}t_1 \int_0^t \mathrm{d}t_2 \psi(t_1-t_2). \tag{3.3}$$

Since $\psi(-t)=\psi(t)$, we have

$$M(t) = 2\int_0^t \mathrm{d}t_1 \int_0^{t_1} \mathrm{d}t_2 \psi(t_1-t_2),$$

which is written as a convolution integral,

$$M(t) = 2\int_0^t \mathrm{d}t_1 \Phi(t_1), \quad \Phi(t_1) \equiv \int_0^{t_1} \mathrm{d}t_2 1 \cdot \psi(t_1-t_2). \tag{3.4}$$

This relation is expressed in terms of Laplace transformation by

$$\Phi(t) = \mathcal{L}^{-1}\left(\frac{1}{s}\cdot\hat{\psi}(s)\right), \quad \hat{\psi}(s) = \mathcal{L}(\psi(t)), \tag{3.5}$$

where we have used the formula $\mathcal{L}(1)=1/s$.

Therefore, the derivative of MSD function is given by

$$\dot{M}(t) = 2\Phi(t) = 2\mathcal{L}^{-1}\left(\frac{1}{s}\cdot\hat{\psi}(s)\right) = 2\mathcal{L}^{-1}\left(\frac{1}{s}\cdot\mathcal{L}(\psi(t))\right), \tag{3.6}$$

which gives the MSD function $M(t)$ from the random impulse correlation function $\psi(t)$. Let us call this *the proper problem* of MSD.

For example, if $\psi(t)=e^{-\gamma|t|}$, we have for $t>0$

$$\dot{M}(t) = 2\mathcal{L}^{-1}\left(\frac{1}{s}\cdot\mathcal{L}(\psi(t))\right) = 2\mathcal{L}^{-1}\left(\frac{1}{s(s+\gamma)}\right) = \frac{2}{\gamma}(1-e^{-\gamma t}) \Rightarrow M(t) = \frac{2}{\gamma^2}(e^{-\gamma t}-1+\gamma t). \tag{3.7}$$

Now the relation (3.6) can be inverted as

$$\psi(t) = \tfrac{1}{2}\mathcal{L}^{-1}(s\cdot\mathcal{L}(\dot{M}(t))), \tag{3.8}$$

which we call *the inverse problem* of MSD to derive the impulse correlation $\psi(t)$ from MSD function $M(t)$.

B. Solution of the inverse problem

Let us consider solutions of inverse problem for some typical MSD functions.

The case of $M(t)=t$. This is the case of ordinary Brownian motion,

$$\psi(t) = \tfrac{1}{2}\mathcal{L}^{-1}(s\cdot\mathcal{L}(\dot{M}(t))) = \tfrac{1}{2}\mathcal{L}^{-1}(s\cdot\mathcal{L}(1)) = \delta(t), \tag{3.9}$$

where we used $\mathcal{L}(1)=1/s$ and $\mathcal{L}^{-1}(1)=2\delta(t)$. This is the well known result.

The case of $M(t)=t^{\alpha}$. This is the case of fractional diffusion,[3]

$$\psi(t) = \frac{1}{2}\mathcal{L}^{-1}(s\cdot\mathcal{L}(\dot{M}(t))) = \frac{\alpha}{2}\mathcal{L}^{-1}(s\cdot\mathcal{L}(t^{\alpha-1})) = \frac{1}{2}\Gamma(\alpha+1)\mathcal{L}^{-1}\left(\frac{1}{s^{\alpha-1}}\right) = \frac{\alpha(\alpha-1)}{2}t^{\alpha-2}, \tag{3.10}$$

where the Laplace transformation formula,

$$\mathcal{L}(t^{\beta-1}) = \Gamma(\beta)\cdot\frac{1}{s^{\beta}} \quad (\beta>0), \tag{3.11}$$

and its inverse formula is used. In the above result the parameter α should satisfy $1<\alpha<2$ by the physical requirement that the correlation $\psi(t)$ decays when $t\to\infty$. The limit $\alpha=1$ is the previous case.

The case of $M(t)=\log(1+t/\tau)$. This is the case of logarithmic MSD function observed in the previous section,

$$M(t) = \log\left(1+\frac{t}{\tau}\right). \tag{3.12}$$

Since its derivative is

$$\Phi(t) = \frac{1}{2}\dot{M}(t) = \frac{1}{2}\cdot\frac{1}{t+\tau}, \tag{3.13}$$

we have

$$\psi(t) = \frac{1}{2}\mathcal{L}^{-1}\left(s\cdot\mathcal{L}\left(\frac{1}{t+\tau}\right)\right) = \dot{\Phi}(t) + \Phi(0)\cdot 2\delta(t) = \frac{1}{2}\left(-\frac{1}{(t+\tau)^2} + \frac{2}{\tau}\cdot\delta(t)\right), \tag{3.14}$$

which have a long range negative correlation. However, it should be noted that this $\psi(t)$ satisfies

$$\int_0^\infty \psi(t)\mathrm{d}t = 0. \tag{3.15}$$

If the sign of such integration is negative, the system shows unphysical behavior: it does not diffuse. All physical diffusion processes should have non-negative values.[4] Above result (3.15) implies that our logarithmic MSD is indeed the marginal case.

IV. SUMMARY AND DISCUSSION

A stochastic model of the correlated diffusion is introduced and analyzed. By numerical simulations the MSD and the van Hove self-correlation functions are computed. Their behaviors for one dimensional case are similar to those observed in the glass phase of our MD simulations. Then the inverse problem of Langevin equation is studied so that the random impulse correlation function is determined to provide the given MSD behavior. Especially the impulse correlation is determined which gives the logarithmic MSD function found in the simulations.

As far we have considered for brevity the simplest Langevin equation. Moreover, the anomalous diffusion has been attributed to the long range impulse correlation. We may, however, extend the Langevin equation to more general ones. For example, one may consider equations for the velocity $v(t)$ with a memory term[5–7]

$$\frac{\mathrm{d}v}{\mathrm{d}t} + \int_0^t \varphi(t-t')v(t')\mathrm{d}t' = f(t). \tag{4.1}$$

Or one may even consider a Langevin equation with a fractional derivative.[3] Furthermore, the intermittent diffusion observed in the MD simulation might be explained by considering a waiting time probability discussed by Scher and Montroll.[8] Such possibilities will be discussed in future works.

ACKNOWLEDGMENTS

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