Non-Gaussian diffusion in static disordered media

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Non-Gaussian diffusion is commonly considered as a result of fluctuating diffusivity, which is correlated in time or in space or both. In this work, we investigate the non-Gaussian diffusion in static disordered media via a quenched trap model, where the diffusivity is spatially correlated. Several unique effects due to quenched disorder are reported. We analytically estimate the diffusion coefficient $D_{\text{lim}}$ and its fluctuation over samples of finite size. We show a sharp peak in the distribution of displacement $P(x, t)$ around $x = 0$, that has frequently been observed in experiments, but not previously explained theoretically. We examine the fidelity of the coarse-grained diffusion map, which is reconstructed from particle trajectories. Finally, we propose a procedure to estimate the correlation length in static disordered environments, where the information stored in the sample-to-sample fluctuation has been utilized.

I. INTRODUCTION

Modern imaging experiments provide a huge amount of dynamic details of diffusion in crowded intracellular environments\cite{1}, which has greatly deepen our understanding of the underlying stochastic motion\cite{2}. A novel class of anomalous diffusion, the non-Gaussian diffusion, has been frequently reported by experiments in a wide range of disordered systems, including soft matter systems\cite{3,4,5,6}, cytoplasm\cite{7,8}, cell membrane\cite{9,10}, and even in the heat transport problem\cite{11}.

In the non-Gaussian diffusion, the distribution of displacement is not Gaussian, while the mean squared displacement can be either linear\cite{3,4,5,12} or sub-linear\cite{6,13} to time. A simple interpretation suggests the dynamic heterogeneity plays a key role in this phenomenon\cite{3,4,14}. In the case that each particle diffuses with random instantaneous diffusivity $D^{(i)}$, the statistics over all the possible diffusivity introduces a convolution to the distribution of displacement by

$$P(x, t) = \int_0^\infty dD^{(i)} G(x, t | D^{(i)}) P(D^{(i)}). \quad (1)$$

In general, $P(x, t)$ is non-Gaussian even when $G(x, t | D^{(i)})$ is Gaussian. A theory of fluctuating diffusivity was formulated\cite{15,16,17,18}, where the diffusivity of each particle follows an independent stochastic process. The theory has been supported by direct observation of fluctuating diffusivity in experiments\cite{3,19} and simulation\cite{10}. Most recently, a comprehensive theoretical framework for the random walk of fluctuating diffusivity was constructed, based on the idea of subordination\cite{14}.

It is a general concern that the dynamic heterogeneity may be introduced by the quasi-static disordered environment\cite{6,10,13,20,21}, where the diffusivity is correlated in space instead of in time. Our understanding on non-Gaussian diffusion in a static environment, however, is still very limited. We note such anomalous diffusive processes can be described by the random walk on the lattices of quenched traps\cite{22}, which has been intensively studied in the context of sub-diffusion\cite{23,24,25,26,27}. The quenched trap model provides insights into the fluctuation among disordered static samples\cite{28,29}, which is essential in biology, known as “every cell is different”.

In the current work, we study a quenched trap model for non-Gaussian diffusion\cite{30}, where the landscape is locally correlated and the local diffusivity $D^{(i)}$ follows the exponential distribution $P(D^{(i)} = D) = D^{-1} \exp(-D/D_0)$.

II. QUENCHED TRAP MODEL

Let us begin with the simplest form of the quenched trap model, i.e., a particle hopping on a $d$-dimensional simple cubic lattice with a set of site-dependent transition rates $W_{i \rightarrow j} = n_i^{-1} \tau_i^{-1}$ from site $i$ to one of its nearest neighbors $j$. Here $n_i = 2d$ is the lattice coordination number. The hopping rate $k = \tau_i^{-1}$ out of site $i$ can be associated with a site energy $V_i(< 0)$ through the Arrhe-
distribution for large uncorrelated, one can see $P_i$ following the exponential distribution.

A two-step approach based on extreme statistics is hence introduced up to the basin size $\xi \approx 2r_c$. By setting $T \neq 1$, this approach can be generalized for the samples with $P(D) = D_0^{-1} T D^{-1} e^{-D^2/D_0}$.

Let us focus on the $T = 1$ case in this work.

III. UNIQUE EFFECTS DUE TO STATIC DISORDERED ENVIRONMENTS

A. Sample-dependent diffusion coefficient

For normal Brownian motion, the mean square displacement (MSD) $\langle x^2 \rangle = \langle |x(t) - x(0)|^2 \rangle$ is expected linear to time $t$ in long time limit. The diffusion coefficient is defined by $D_{\text{dis}} \equiv \lim_{t \to \infty} \langle x^2 \rangle / 4t$, where the average $\langle \cdot \rangle$ is performed over the trajectories. Following Kehr and Haus [22], the diffusion coefficient of trap model depends on the local diffusivity

$$D_{\text{dis}} = \left[ \frac{1}{N} \sum_{i=1}^N \langle D_i^{(l)} \rangle^{-1} \right]^{-1}.$$

It is confirmed by our simulation (not shown here). Its relation to the instantaneous diffusivity $D_i^{(l)}$ is shown later by Eq. (5).

One can find more details in the recent paper by Akimoto et al. [29]. Noting that the connection between the diffusion coefficient and the mean sojourn time of the given sample

$$\bar{\tau}_N \equiv \frac{1}{N} \sum_{i=1}^N \tau_i = \frac{a^2}{2dD_{\text{dis}}},$$

$D_{\text{dis}}$ and $\bar{\tau}$ depend on the specific configuration of the sample. We further consider the statistics over the ensemble of samples, first estimating the distribution of $\bar{\tau}$, then the mean value and higher moments of $D_{\text{dis}}$. Exact estimation is challenging due to the spatial correlation. Noting that the correlation among extremal basins is quite weak, we consider the coarse-grained lattice of basins instead. The typical sojourn time in basin $i$ is proportional to the inverse of the local diffusivity, $\tau_i \approx 1/D_i^{(l)}$. Noting that $D_i^{(l)}$ follows exponential distribution given by Eq. (6), we have

$$P(\tau_i = \tau) = \tau_0 \tau^{-2} \exp(-\tau_0/\tau),$$

where $\tau_0 \approx 1/D_0$. The distribution is with heavy tail $P(\tau) \sim \tau^{-2}$, which is mainly contributed by the frozen sites with $P(D_i^{(l)} = 0) = 1/D_0$. The expectation value of
\( \tau \) diverges. The generalized central limit theorem suggests the mean waiting time \( \bar{\tau}_N \) follows the one-sided Lévy stable distribution with \( \mu = 1 \), \( \gamma = 6.35 \), and \( \delta_N = \delta_0 + 2\pi \gamma \ln N \) with \( \delta_0 = -26.1 \).

\( \bar{\tau}_N = \frac{1}{N} \sum_{i=1}^{N} \tau_i \overset{d}{\to} z \sim S(\mu = 1, \beta = 1, \gamma, \delta_N; 1), \) for large \( N \),

where \( \overset{d}{\to} \) means converging in the sense of the probability distribution. Here we adopt the type-1 parameterization of the stable distribution, where \( \gamma \) is the scale parameter and \( \delta_N = \delta_0 + 2\pi \gamma \ln N \) is the position parameter \([31]\). The distribution is not strictly stable. As a consequence, the position of the distribution shifts to infinity by \( \ln N \), which is well confirmed by numerical sampling (see Figure 2).

Noting that \( D_{\text{dis}} = a^2/(2d\bar{\tau}_N) \), we see the moments of \( D_{\text{dis}} \) are indeed negative moments of \( \bar{\tau}_N \)

\[
\langle (D_{\text{dis}})^\alpha \rangle = \left( \frac{a^2}{2d} \right)^\alpha \langle \bar{\tau}_N^{-\alpha} \rangle.
\]

Noting the Laplace transform of the one-sided Lévy stable distribution \([33]\), it can be shown that

\[
\langle D_{\text{dis}} \rangle^\alpha \sim \left( \delta_0 + \frac{2}{\pi} \ln N \right)^{-\alpha}.
\]

(See appendix for details.) The analytical results are confirmed by numerical sampling for \( \alpha = 1, 2 \), as shown in Figure 3. One can read that the uncorrelated approximation works well for samples larger than the extremal basins, i.e. \( L > \xi = 2r_c \).

B. Non-Gaussian distribution of displacement with additional peak around origin

For more dynamic details, we performed kinetic Monte Carlo simulation \([32]\) for the random walk on two-dimensional quenched samples with periodic boundary. On each quenched sample, the initial sites of trajectories are chosen from Boltzmann distribution. Being specific, a trajectory starts from site \( i \) with the probability

\[
P_i = \frac{e^{-V_i/T}}{Z},
\]

where \( Z = \sum_{i=1}^{N} e^{-V_i/T} \) is the partition function over all \( N = L^2 \) sites. One can find simulation details in SM.

Figure 4 shows the distribution of displacement \( P(x, t) \) for various lag time \( t \). As can be seen in the figure, the tail of the distribution changes continuously for increasing \( t \), from the exponential tail towards Gaussian. Such behavior was also reported in previous studies \([11, 14, 19]\). It can be well explained by noticing the heterogeneity in the ensemble of the trajectories \([19]\). In the early stage of each trajectory, the random walk is dominated by the local diffusivity of the initial extremal basin. The exponential distributed local diffusivity introduces the non-Gaussian tail. At the later stage a large fraction of particles leave the original extremal basins. Self-averaging is gradually achieved along the long trajectories of these fast moving particles, which leads to the Gaussian tail.

An unexpected sharp peak of \( P(x, t) \) appears around \( x = 0 \) for all \( t \). Similar peaks were also observed in various systems of non-Gaussian diffusion, both by experiments \([4, 8, 9]\) and by molecular dynamics simulations \([10]\). The nontrivial peak can be explained in the annealed framework of fluctuating diffusivity \([14, 19]\), but at the cost of introducing an additional immobile state. In the quenched model, however, such sharp peak is a natural consequence of the coupling between the local
diffusivity and the sojourn time in the trap. The argument follows. The distribution of displacement $P(x,t)$ counts all the segments $\{x(t_0) \rightarrow x(t_0 + t)\}$ of the trajectories. For small $t$, we assume each segment is dominated by a fixed instantaneous diffusivity $D^{(t)}$, which is equal to the local diffusivity $D^{(t)}_i$ of the center of the segment. Noting that the segments sample the landscape with the Boltzmann weight (Eq. [11]) and also Eq. [2], we can get the probability that a segment centers at site $i$ in the condition of $D^{(t)}_i = D$ by

$$P(x = x_i | D^{(t)}_i = D) = \frac{D^{-1}}{\sum_{j=1}^{N} (D^{(t)}_j)^{-1}}.$$

(12)

The instantaneous diffusivities $D^{(t)}$ of the segments hence follow the distribution with additional weight

$$P(D^{(t)} = D) = \sum_{i=1}^{N} P(x = x_i | D^{(t)}_i = D) P(D^{(t)}_i = D)$$

$$= \frac{D_{\text{dis}}}{D_0} D^{-1} e^{-D/D_0},$$

(13)

where Eq. [8], Eq. [5] and Eq. [12] are used. Further assuming in each short segment the particle diffuses as normal Brownian motion dominated by the instantaneous diffusivity $D^{(t)}$, we have

$$G(x, t | D^{(t)}) = \frac{1}{\sqrt{4 \pi D^{(t)} t}} \exp \left( -\frac{x^2}{4D^{(t)} t} \right).$$

(14)

The distribution of displacement is obtained by counting all the segments, which gives

$$P(x, t) = \int_{0}^{\infty} dD \ G(x, t | D^{(t)}) P(D^{(t)} = D)$$

$$= \frac{D_{\text{dis}}}{D_0} x^{-1} \exp(-\frac{x}{\sqrt{D_0} t}).$$

(15)

A sharp peak appears around the origin as $P(x) \sim x^{-1}$, as shown in the inset of Fig. [4].

**FIG. 4.** The distribution of displacement $P(x,t)$ for a typical sample and various $t$. Eq. [15] is plotted by the dash line. The inset enlarges the peak of $P(x,t = 20)$ for a clearer view.

IV. TRAJECTORY-BASED DATA ANALYSIS

A. Reconstruct the diffusion map from trajectories

The structural information of the environment is often represented in the style of diffusion map (D-map), which is a map of local diffusivity retrieved from trajectories. D-map is generally constructed in the coarse-grained fashion, since the trajectories are sparse in most experiments. We say a map is of $s$-resolution, if it is composed of the grains of size $s \leq L$. The local diffusivity of each grain is estimated from all the segments centered in the grain $q$ and its nearest-neighbours by

$$\bar{\delta}_q^{(s)} = \frac{1}{2dM} \frac{1}{M} \sum_{k=1}^{M} \frac{|x_k(t + dt) - x_k(t)|^2}{dt},$$

(16)

where $dt$ is the time bin and $\{x_k(t) \rightarrow x_k(t + dt)\}$ is the $k$th segment of all the $M$ relevant ones. The fidelity of D-map can be evaluated from the mean value of the retrieved local diffusivities averaged over all the $K$ grains in the sample,

$$\delta^{(s)} = \frac{1}{K} \sum_{q=1}^{K} \delta_q^{(s)}.$$

(17)

The inset of Figure [5] shows $\delta^{(s)}$ retrieved for 10 disordered samples, where $10^4$ trajectories are simulated for each sample. As can be seen in the figure, the fidelity is merely guaranteed for $s < r_s$ that $\delta^{(s)} \approx D_0$, $\delta^{(s)}$ deviates from $D_0$ in the limit $s \rightarrow 1$, since $10^4$ trajectories are still not enough for reconstruction in the finest resolution.
resolutions. For coarse-grained maps with $s > r_c$, the deviation becomes more significant, accompanied by the rise of sample-to-sample fluctuation which is clearly presented in the fashion of $(\bar{\delta}^{(s)})^{-1}$ (see Fig. 5(a)). Noticing the segments sample the landscape by the Boltzmann weight, one can show $\delta^{(s)}_q$ equals the diffusion coefficient $D^{(i)}_{\text{dis}}$ of the of grain $q$, given by Eq. (8) with $N = s^2$ traps. In the most coarse-grained case $s = L$, $\delta^{(L)}$ is exactly the diffusion coefficient $D_{\text{dis}}$ estimated from ensemble-averaged MSD of all the trajectories. One can expect that $(\bar{\delta}^{(L)})^{-1}$ follows the stable distribution provided by Eq. (8) (shown in Fig. 5(b)). $\bar{\delta}^{(s)}$ introduces a path linking the local diffusivity and the instantaneous diffusivity, from determined $\bar{D}^{(i)} = D_0$ to random $\bar{D}^{(i)} = D_{\text{dis}}$, which is shown by color lines in Fig. 5(a).

B. Estimate the correlation length

It is worthy to notice the sample-to-sample fluctuation of $(\bar{\delta}^{(s)})^{-1}$ is depressed in D-map with fine resolutions. It leads to an approach to estimate the typical correlation length in quenched samples as follows:

1) Divide the whole sample into sub-samples of size $s$.
2) Measure diffusion coefficient $\delta^{(s)}_q$ of each sub-sample $q$ and calculate the mean value $\bar{\delta}^{(s)} = \frac{1}{K} \sum_{q=1}^{K} \delta^{(s)}_q$ over all the $K$ sub-samples.
3) Repeat 1) and 2) for various $s$, and record the $s$ dependence of $\bar{\delta}^{(s)} = (\bar{\delta}^{(s)})^{-1}$.
4) Repeat 1), 2) and 3) for various samples, calculate the sample-to-sample fluctuation $\Delta^2 \equiv \langle (\bar{\delta}^{(s)})^2 \rangle - \langle \bar{\delta}^{(s)} \rangle^2$.

In the case the $x^{-1}$-peak is observed in the non-Gaussian distribution of displacement, one may also expect that $\Delta^2$ falls for decreasing $s$ as that shown in Fig. 5(a). For the diffusion map with $s < r_c$, each sub-sample is dominated by a unique local diffusivity. One may expect $\Delta^2$ vanishes. The random samples are hence characterized by two universal parameters, the correlation length $r_c$ and the mean local diffusivity $\bar{\delta}^{(s)}|_{s<r_c} \simeq D_0$.

V. DISCUSSION AND SUMMARY

The main results in Sec. III and Sec. IV only require $P(\tau) \sim \tau^{-2}$ for large $\tau$, which is contributed by the most deeply trapped particles. The results are hence expected unchanged as long as $P(D^{(i)}_1) \sim 0$ is finite and continuous, while the exponential tail is not required.

In this work, the spatially correlated local diffusivity $\{D^{(i)}_1\}$ is generated from the extreme landscape $\{V_i\}$ by $D^{(i)}_1 = e^{V_i/T}/2d$. The $T = 1$ case leads to exponential distributed $\{D^{(i)}_1\}$, which was reported by experiments. In more general $T \neq 1$ cases, $P(D^{(i)}_1)$ could be stretched or compressed as $P(D^{(i)}_1) = D_0^{-1}T^{D-1}e^{-D/T}/D_0$. The whole class of models provide description of a wide range of non-Gaussian diffusion in static disordered environments.

In summary, our careful study on the quenched model represents several unique properties of non-Gaussian diffusion in static disordered environments. A sharp peak appears in the immobile part of the distribution of displacement, from which the cell would largely benefit since the biology functions of molecular machines are mostly carried out by the immobile ones. The size-dependence of the diffusion coefficient calls our attention to the fidelity of the coarse-grained diffusion map. On the other hand, it offers us an approach to estimate the correlation length from a large bunch of samples. It provides a new insight into the experiments on cells where the cell-to-cell fluctuation is always significant.

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Appendix: Negative moments of one-sided Lévy stable distribution with exponent $\mu = 1$

Let $x_N$ follow $S(x; 1, 1, \gamma, \delta_N)$ with $\delta_N = \delta_0 + \frac{2}{\gamma} \ln N$. The negative moments of $x_N$ is defined as

$$\langle x_N^{-\alpha} \rangle = \int_0^\infty dx \ x^{-\alpha} S(x; 1, 1, \gamma, \delta_N), \ \alpha > 0. \quad (A.1)$$

Because the concerned distribution is not strictly stable, the moments depend on $N$. In this section, we estimate the large-$N$ dependence of $\langle x_N^{-\alpha} \rangle$.

The approach is similar to that by Chechkin et al. [2], where they deal with the $\mu < 1$ case. Noting

$$x^{-\alpha} = \frac{1}{\Gamma(\alpha)} \int_0^\infty ds \ s^{\alpha-1}e^{-s x}, \ \alpha > 0, \quad (A.2)$$
we have
\[
\langle x_N^{-\alpha} \rangle = \frac{1}{\gamma^\alpha} \left( \left( \frac{x_N}{\gamma} \right)^{-\alpha} \right)
= \frac{1}{\gamma^\alpha} \int _0 ^\infty dx \ x^{-\alpha} f(x)
= \frac{1}{\gamma^\alpha} \Gamma (\alpha) \int _0 ^\infty ds \ s^{-\alpha - 1} \int _0 ^\infty dx \ e^{-sx} f(x)
= \frac{1}{\gamma^\alpha} \Gamma (\alpha) \int _0 ^\infty ds \ s^{-\alpha - 1} \overline{\mathcal{J}}(s),
\]
(A.3)
where \( f(x) \equiv P \left( \frac{x_N}{\gamma} = x \right) \) and its Laplace transform \( \overline{\mathcal{J}}(s) = \mathcal{L}(f(x)) \). Noting the Laplace transform of \( g(x) \equiv P \left( \frac{x_N}{\gamma} - \delta_N = x \right) = S(x; 1, 1, 1, 0) \) is given by \[34\]
\[
\overline{g}(s) = \mathcal{L} \left( S(x; 1, 1, 1, 0) \right) = e^{-s \ln s},
\]
(A.4)
we can see
\[
\overline{\mathcal{J}}(s) = \mathcal{L}(f(x)) = \mathcal{L}(g(x - \delta_N)) = e^{-s(\delta_N + \ln s)},
\]
(A.5)
For the large-\( N \) dependence of \( \langle x_N^{-\alpha} \rangle \), we estimate the integral in Eq.\( \text{A.3} \). The integral can be separated into three parts:
\[
I = I_1 + I_2
= \left( \int _0 ^1 ds \ + \int _1 ^\infty ds \right) \mathcal{Q}(s),
\]
(A.6)
where the integrand \( \mathcal{Q}(s) = s^{-\alpha - 1} e^{-s(\delta_N + \ln s)} \).
For the first part, we can see
\[
\int _0 ^1 ds \ s^{\alpha - 1} e^{-s\delta_N} < I_1 < \int _0 ^1 ds \ s^{\alpha - 1} e^{-s\delta_N - \min(s \ln s)},
\]
which gives
\[
\frac{\Gamma (\alpha) - \Gamma (\alpha, \delta_N)}{\delta_N^\alpha} < I_1 < e^{1/\delta} \Gamma (\alpha) - \Gamma (\alpha, \delta_N) ,
\]
(A.7)
where \( \Gamma (\alpha, x) = \int _0 ^x dt \ t^{\alpha - 1} e^{-t} \) is the incomplete gamma function. Since \( \Gamma (\alpha, x) \sim x^{1-\alpha} e^{-x} \), it can be neglected for large \( N \). We see \( I_1 \) is controlled by \( N \) as
\[
I_1 \sim \delta_N^{-\alpha}.
\]
(A.9)
For the second part, we have
\[
0 < I_2 < \int _1 ^\infty ds \ s^{\alpha - 1} e^{-s\delta_N} = \frac{\exp(-\delta_N)}{\delta_N} (1 + O(\delta_N^{-1})).
\]
(A.10)
For large \( N \), \( I_1 \) dominates the whole integral \( I \). The negative moments \( \langle x_N^{-\alpha} \rangle \) depends on \( N \) as
\[
\langle x_N^{-\alpha} \rangle \sim \left( \delta_0 + \frac{2}{\pi} \ln N \right)^{-\alpha}.
\]
(A.11)
It is confirmed by numerical sampling, as shown in Fig.3 of the main text.

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