Chapter 9

Half- and Full-Integer Power Law for Distance Fluctuations: Langevin Dynamics in One- and Two-Dimensional Systems

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Half- and full-integer power law for distance fluctuations: Langevin dynamics in one- and two-dimensional systems

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Abstract

Langevin dynamics of one- and two-dimensional systems with the nearest neighbor couplings is examined to derive the autocorrelation function (ACF) of the distance fluctuations. Understanding of the dynamics of pairwise distance correlation is essential in the studies using single-molecule spectroscopy. For both 1-D cases of an open chain and a closed loop, a power law of $t^{-1/2}$, $t^{-3/2}$ and $t^{-5/2}$ for the ACF are obtained, and for 2-D systems of a sheet and a tube, a power law of t^{-1}, t^{-2} and t^{-3} are found. The different exponent of the power law is shown to depend on the location of the pairwise beads and their topography.

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Keywords: Power laws; Langevin dynamics; Single molecule; Rouse model

1. Introduction

Decays in physical systems are usually characterized by pure-exponential or by non-exponential decays. Power-law decays have often been associated with self-similar processes involving fractals [1–7]. For example, in a recent work by Granek and Klafter [1] they showed that power-law decays in distance autocorrelation function (ACF) could arise due to vibrational excitation of fractals. Recently power-law behavior in fluorescence intermittency observed in single quantum dots [8–12] and other organic molecules [13] have generated some interest. It has been shown that diffusion-controlled electron transfer in a Debye and non-Debye dielectric media could lead to the power-law blinking statistics [14,15].

The recent single-molecule experiments by Xie and coworkers [16–18] on fluctuating fluorescence lifetimes were attributed to fluctuations in the distance between a donor and an acceptor attached to a protein chain. The observed $t^{-1/2}$ power law in the lifetime fluctuations and its relationship to chain dynamics have been investigated recently [19–21] using a Rouse model [22]. There are also many other theoretical studies of reaction controlled by barrier fluctuations and diffuse dynamics [1,23–27]. In electron transfer (ET) and fluorescence resonances energy transfer (FRET), the transfer rate depends on the distance R between donor and an acceptor (exponentially for ET and $1/R^6$ for FRET) [28]. If donor–acceptor distance is subjected to fluctuations, the rate constant would also fluctuate in time. In addition to single-molecule techniques, many other experimental techniques have been employed to study these conformational changes, such as NMR [29], neutron scattering [30,31], optical absorption [32], electrophoresis [33], optical tweezers [34] etc. Chain dynamics has been used in these studies.

In this work, we extend previous studies of Rouse model [22] to a long chain and a closed loop, shown in

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the schematic diagrams Fig. 1(a) and (b), respectively, as well as a two-dimensional system of a sheet or a tube, and examine the distance ACF between any pair of beads in such a 1-D or 2-D system. We will show that Langevin dynamics for 1-D chain/loop under some conditions could lead to the less well-know $t^{-3/2}$ and $t^{-5/2}$ power law, in addition to the more familiar $t^{-1/2}$ behavior. In addition to the half-integer power law for the 1-D system, we will also show that for a 2-D system, the ACF decays with a full-integer law (t^{-1}, t^{-2} and t^{-3}), depending on the topological conditions of such a pairwise beads. A ladder system, seen in Fig. 1(c), is also studied and has similar time behaviors of ACF in chain a system. Besides the half-integer laws, the ACF in a ladder also has only exponential (no power laws) decays at certain topological conditions. The main purpose of this work is to show these half- and full-integer power laws can arise and to explain their causes.

2. Theory

We will derive in this section the analytic results for ACF of the distance fluctuation for four different topological categories, *i.e.* an open chain, a closed loop, a sheet, a tube, and a ladder. Their corresponding long time behaviors are also summarized and illustrated.

2.1. An open chain

An ideal chain of N beads with equal couplings to their nearest neighbors and other vibrations have been studied widely [22]. The Langevin equation of over-damped oscillators in such a N-unit Rouse chain can be expressed by [35,36]

$$\zeta \frac{\mathrm{d}}{\mathrm{d}t} Q(t) + \omega^2 \mathbf{R} Q(t) = F(t)/m, \tag{1}$$

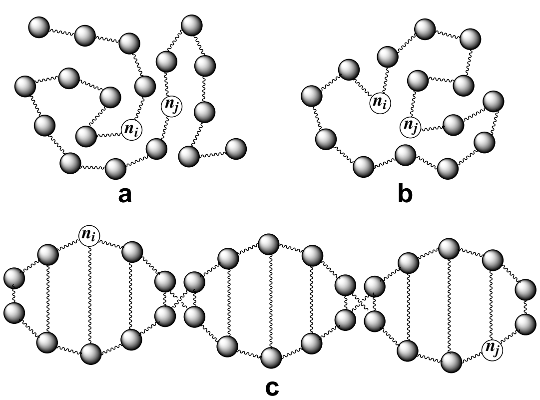

Fig. 1. The schematic diagrams of a Rouse model with a open chain (a), a closed loop (b), and a ladder (c) of beads. A bead interacts with others through the connected springs. The dynamics between n_ith and n_jth beads at various positions are studied in this work.

where ζ is a constant friction coefficient. The $Q(t)$ is a column super vector $(\vec{q}_1, \vec{q}_2, \vec{q}_3 \ldots \vec{q}_N)$, where $\vec{q}_i$ denotes the displacement vector of the ith bead from its equilibrium position. $\mathbf{R}$ is the Rouse coupling matrix. $F(t)$ represents white noise with zero mean and no correlation with $Q(t)$. Therefore, one has the following equations: $\langle F_i^{\mu}(t)\rangle = 0$, $\langle F_i^{\mu}(t) \cdot q_j^{\nu}(\tau)\rangle = 0$, and $\langle F_i^{\mu}(t) \cdot F_j^{\nu}(\tau)\rangle = 2mk_{\mathrm{B}} \; T\zeta\delta(t-\tau)$ $\delta_{\mu,\nu}\delta_{i,j}$, where μ and ν represent x, y, or z, whereas i and j represent the bead index. The pairwise correlation of the displacement vectors can be written as [20,21]

$$\begin{aligned}
&\frac{\mathrm{d}}{\mathrm{d}t}\langle \vec{q}_1(t)\cdot\vec{q}_i(0)\rangle + \frac{\omega^2}{\zeta}(\langle \vec{q}_1(t)\cdot\vec{q}_i(0)\rangle - \langle \vec{q}_2(t)\cdot\vec{q}_i(0)\rangle) = 0,\\
&\frac{\mathrm{d}}{\mathrm{d}t}\langle \vec{q}_n(t)\cdot\vec{q}_i(0)\rangle + \frac{\omega^2}{\zeta}(-\langle \vec{q}_{n-1}(t)\cdot\vec{q}_i(0)\rangle + 2\langle \vec{q}_n(t)\cdot\vec{q}_i(0)\rangle\\
&\quad - \langle \vec{q}_{n+1}(t)\cdot\vec{q}_i(0)\rangle) = 0,\\
&\frac{\mathrm{d}}{\mathrm{d}t}\langle \vec{q}_N(t)\cdot\vec{q}_i(0)\rangle + \frac{\omega^2}{\zeta}(-\langle \vec{q}_{N-1}(t)\cdot\vec{q}_i(0)\rangle\\
&\quad + \langle \vec{q}_N(t)\cdot\vec{q}_i(0)\rangle) = 0,
\end{aligned} \tag{2}$$

where $n = 2, 3, \cdots N-1$; $i = 1, 2, \cdots N$; and $\langle \vec{q}_j(t)\cdot\vec{q}_i(0)\rangle$ is the ensemble average of the inner product of $\vec{q}_j(t)$ and $\vec{q}_i(0)$. Using the method described in Appendix C of Ref. [37] the pairwise correlation functions can be simplified as

$$\begin{aligned}
\langle \vec{q}_n(t)\cdot\vec{q}_i(0)\rangle &= \sum_{j=1}^{N} \frac{\langle \vec{q}_j(0)\cdot\vec{q}_i(0)\rangle}{N}\\
&\quad \times \left\{1 + 2\sum_{k=1}^{N-1}\cos\left(\frac{\pi k(n-\frac{1}{2})}{N}\right)\right.\\
&\quad \left. \times \cos\left(\frac{\pi k\left(j-\frac{1}{2}\right)}{N}\right)\exp\left[-4\frac{\omega^2 t}{\zeta}\sin^2\left(\frac{\pi k}{2N}\right)\right]\right\}\\
&= \frac{\langle |\vec{q}_i(0)|^2\rangle}{N}\left\{1 + 2\sum_{k=1}^{N-1}\cos\left(\frac{\pi k(n-\frac{1}{2})}{N}\right)\right.\\
&\quad \left. \times \cos\left(\frac{\pi k\left(i-\frac{1}{2}\right)}{N}\right)\exp\left[-4\frac{\omega^2 t}{\zeta}\sin^2\left(\frac{\pi k}{2N}\right)\right]\right\},
\end{aligned} \tag{3}$$

where $\langle \vec{q}_j(0)\vec{q}_i(0)\rangle = \delta_{i,j}\langle |\vec{q}_i(0)|^2\rangle$ was used. By assuming the distance fluctuation of each bead from its equilibrium position is small, the ACF of the distance deviation, $C_Q(t)$, between the n_1th and n_2th beads from their equilibrium position equals $\langle [\vec{q}_{n_1}(t) - \vec{q}_{n_2}(t)]\cdot[\vec{q}_{n_1}(0) - \vec{q}_{n_2}(0)]\rangle/3$. Using Eq. (3), one obtains [20,21]

$$\frac{C_Q(t)}{C_0} = \sum_{k=1}^{N-1}\frac{\mathrm{e}^{\frac{-4\omega^2 t}{\zeta}\sin^2\left(\frac{\pi k}{2N}\right)}}{N}\left[\cos\left(\frac{\pi k\left(n_1-\frac{1}{2}\right)}{N}\right) - \cos\left(\frac{\pi k\left(n_2-\frac{1}{2}\right)}{N}\right)\right]^2, \tag{4}$$

where C_0 is defined as $C_Q(0)$. The power-law behaviors at long times are depending on the location and separation of the pairwise beads. In a long chain the long time behaviors of the above equation can be classified as

$$\frac{C_Q(t)}{C_0} \propto \begin{cases} t^{-1/2} & \text{as } |n_1 - n_2| \approx N, \\ t^{-3/2} & \text{as } |n_1 - n_2| \ll N, \text{and both } n_1 \,\&\, n_2 \\ & \quad \text{away from ends}, \\ t^{-5/2} & \text{otherwise}. \end{cases} \tag{5}$$

The numerical results are illustrated in Fig. 2(a). The power-laws behaviors are derived in A.1 using asymptotic approximation. More detailed discussion about the causes of different power laws will be discussed in Section 3.

2.2. A closed loop

The pairwise correlation function of displacement vectors in a N-unit loop satisfies

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \vec{q}_n(t)\cdot\vec{q}_i(0)\rangle + \frac{\omega^2}{\zeta}(-\langle \vec{q}_{n-1}(t)\cdot\vec{q}_i(0)\rangle + 2\langle \vec{q}_n(t)\cdot\vec{q}_i(0)\rangle - \langle \vec{q}_{n+1}(t)\cdot\vec{q}_i(0)\rangle) = 0, \tag{6}$$

where n, $i = 1, \ldots N$. One has the Nth bead connects with the first bead to form a loop, *i.e.* $\vec{q}_0 = \vec{q}_N$ and $\vec{q}_1 = \vec{q}_{N+1}$. Using the method described in Appendix A of Ref. [37], the correlation functions are simplified as

$$\langle \vec{q}_n(t)\cdot\vec{q}_i(0)\rangle = \frac{\langle |\vec{q}_n(0)|^2\rangle}{N}\sum_{k=1}^{N}\cos\left(\frac{2\pi k(n-i)}{N}\right) \times \exp\left[\frac{-4\omega^2 t}{\zeta}\sin^2\left(\frac{\pi k}{N}\right)\right]. \tag{7}$$

With the above equation, the ACF of the distance deviation between the n_1th and n_2th beads from their equilibrium positions can be expressed as

$$\frac{C_Q(t)}{C_0} = \frac{2}{N}\sum_{k=1}^{N}\sin^2\left(\frac{\pi k(n_1-n_2)}{N}\right)\exp\left[\frac{-4\omega^2 t}{\zeta}\sin^2\left(\frac{\pi k}{N}\right)\right]. \tag{8}$$

In a long chain the time power-law behaviors of C_Q can be grouped as

$$\frac{C_Q(t)}{C_0} \propto \begin{cases} t^{-1/2} & \text{when } |n_1 - n_2| \approx N/2, \\ t^{-3/2} & \text{otherwise}. \end{cases} \tag{9}$$

These behaviors of C_Q are also shown in Fig. 2(b). The mathematical asymptotic approximation of the power-law behaviors in Eq. (8) are given in A.1. More detailed discussion about the reasons of change from $t^{-1/2}$ to $t^{-3/2}$ are discussed in Section 3.

2.3. A sheet

Assuming a two-dimensional sheet with $N \times M$ beads where each bead is coupled to the nearest beads, the pairwise correlation of the displacement vectors can be obtained straight forward by introducing another independent dimension to Eq. (2). Extending Eq. (3) to two independent variables along N and M directions, the solution of the correlation of the displacement is given by

$$\begin{aligned}\langle \vec{q}_{n,m}(t)\cdot\vec{q}_{i,j}(0)\rangle = \frac{\langle |q_{n,m}(0)|^2\rangle}{NM}&\left\{1 + 2\sum_{k_x=1}^{N-1}\cos\left(\frac{\pi k_x(n-\frac{1}{2})}{N}\right)\right.\\ &\left.\times\cos\left(\frac{\pi k_x(i-\frac{1}{2})}{N}\right)\times\exp\left[-4\frac{\omega_x^2 t}{\zeta}\sin^2\left(\frac{\pi k_x}{2N}\right)\right]\right\}\\ &\times\left\{1 + 2\sum_{k_y=1}^{M-1}\cos\left(\frac{\pi k_y(m-\frac{1}{2})}{M}\right)\right.\\ &\left.\times\cos\left(\frac{\pi k_y(j-\frac{1}{2})}{M}\right)\exp\left[\frac{-4\omega_y^2 t}{\zeta}\sin^2\left(\frac{\pi k_y}{2M}\right)\right]\right\}.\end{aligned} \tag{10}$$

One can derive the ACF of the distance deviation between the beads at the (n_1, m_1)- and (n_2, m_2)-sites as

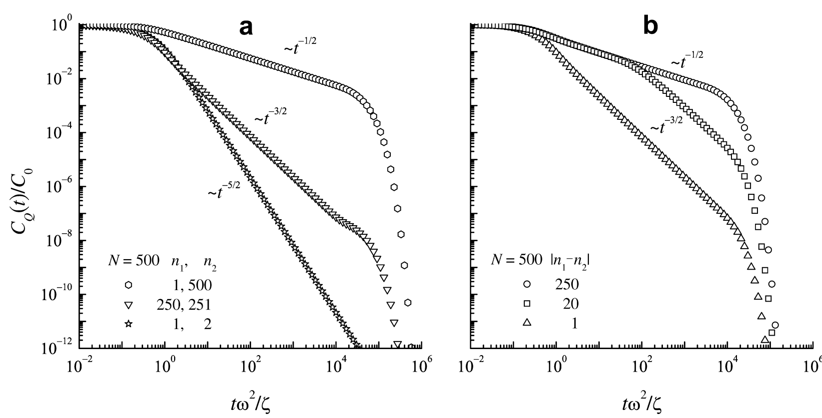

Fig. 2. The calculated $C_Q(t)/C_0$ for the various separations between the pairwise beads for a chain (a) and a loop (b) with $N = 500$. Here a dimensionless $t\omega^2/\zeta$ is used for the x-axis. n_1 and n_2 are the site indices of the pairwise beads. The exponent of the power law, $-1/2$, $-3/2$, and $-5/2$, depends on $|n_1 - n_2|$ and its location. At much longer times, $C_Q(t)/C_0$ becomes a single exponential decay.

$$
\begin{aligned}
\frac{C_Q(t)}{C_0} = \frac{1}{NM}\Bigg\{ & \sum_{k_x=1}^{N-1} \mathrm{e}^{\frac{-4\omega_x^2 t}{\zeta}\sin^2\left(\frac{\pi k_x}{2N}\right)} \left[\cos\left(\frac{\pi k_x\left(n_1-\frac{1}{2}\right)}{N}\right)\right. \\
& \left. - \cos\left(\frac{\pi k_x\left(n_2-\frac{1}{2}\right)}{N}\right)\right]^2 + \sum_{k_y=1}^{M-1} \mathrm{e}^{\frac{-4\omega_y^2 t}{\zeta}\sin^2\left(\frac{\pi k_y}{2M}\right)} \\
& \times \left[\cos\left(\frac{\pi k_y\left(m_1-\frac{1}{2}\right)}{M}\right) - \cos\left(\frac{\pi k_y\left(m_2-\frac{1}{2}\right)}{M}\right)\right]^2 \\
& + 2\sum_{k_x=1}^{N-1}\sum_{k_y=1}^{M-1} \mathrm{e}^{\frac{-4t}{\zeta}\left[\omega_x^2\sin^2\left(\frac{\pi k_x}{2N}\right)+\omega_y^2\sin^2\left(\frac{\pi k_y}{2M}\right)\right]} \\
& \times \left[\cos\left(\frac{\pi k_x\left(n_1-\frac{1}{2}\right)}{N}\right)\times\cos\left(\frac{\pi k_y\left(m_1-\frac{1}{2}\right)}{M}\right)\right. \\
& \left. - \cos\left(\frac{\pi k_x\left(n_2-\frac{1}{2}\right)}{N}\right)\cos\left(\frac{\pi k_y\left(m_2-\frac{1}{2}\right)}{M}\right)\right]^2\Bigg\}. \qquad (11)
\end{aligned}
$$

At various locations of the pairwise beads, its long time property in a large sheet gives the following asymptotic behaviors.

$$
\frac{C_Q(t)}{C_0} \propto \begin{cases} t^{-1} & \text{as } |n_1-n_2| \approx N \text{ and } |m_1-m_2| \approx M \\ t^{-2} & \text{as } |n_1-n_2| \ll N,\ |m_1-m_2| \ll M \text{ and } (n_1,m_1)\text{-} \\ & \&\ (n_2,m_2)\text{-sites are parallel to or far away from} \\ & \text{the boundaries,} \\ t^{-3} & \text{otherwise.} \end{cases} \qquad (12)
$$

The numerical results are illustrated in Fig. 3(a). More detailed discussions of the behaviors and the analytical asymptotic approximation to obtained the power laws will be presented in Section 3 and A.2, respectively.

2.4. A tube

The differential equations of the pairwise correlation of the displacement vector of a $N \times M$ hollow tube with N-bead in length can be obtained by combining Eq. (2) with N beads and Eq. (6) with M beads. Using the results in Eqs. (3) and (7), the correlation of the displacement $\langle q_{n,m}(t)q_{i,j}(0)\rangle$ and the ACF $C_Q(t)$ between the beats at (n_1, m_1)- and (n_2, m_2)-sites can be derived as

$$
\begin{aligned}
\langle \vec{q}_{n,m}(t)\cdot\vec{q}_{i,j}(0)\rangle = \frac{\langle |q_{n,m}(0)|^2\rangle}{NM}\Bigg\{ & 1 + 2\sum_{k_x=1}^{N-1} \mathrm{e}^{\frac{-4\omega_x^2 t}{\zeta}\sin^2\left(\frac{\pi k_x}{2N}\right)} \\
& \times \cos\left(\frac{\pi k_x\left(n-\frac{1}{2}\right)}{N}\right)\cos\left(\frac{\pi k_x\left(i-\frac{1}{2}\right)}{N}\right)\Bigg\} \\
& \times \sum_{k_y=1}^{M} \mathrm{e}^{\frac{-4\omega_y^2 t}{\zeta}\sin^2\left(\frac{\pi k_y}{M}\right)}\cos\left(\frac{2\pi k_y(m-j)}{M}\right). \qquad (13)
\end{aligned}
$$

$$
\begin{aligned}
\frac{C_Q(t)}{C_0} = \frac{1}{NM}\Bigg\{ & 2\sum_{k_y=1}^{M} \mathrm{e}^{\frac{-4\omega_y^2 t}{\zeta}\sin^2\left(\frac{\pi k_y}{M}\right)}\sin^2\left(\frac{\pi k_y(m_1-m_2)}{M}\right) \\
& + \sum_{k_x=1}^{N-1}\sum_{k_y=1}^{M} \mathrm{e}^{\frac{-4t}{\zeta}\left[\omega_x^2\sin^2\left(\frac{\pi k_x}{2N}\right)+\omega_y^2\sin^2\left(\frac{\pi k_y}{M}\right)\right]} \\
& \times \left[\cos^2\left(\frac{\pi k_x\left(n_1-\frac{1}{2}\right)}{N}\right) + \cos^2\left(\frac{\pi k_x\left(n_2-\frac{1}{2}\right)}{N}\right)\right. \\
& - 2\cos\left(\frac{\pi k_x\left(n_1-\frac{1}{2}\right)}{N}\right)\times\cos\left(\frac{\pi k_x\left(n_2-\frac{1}{2}\right)}{N}\right) \\
& \left.\times\cos\left(\frac{2\pi k_y(m_1-m_2)}{M}\right)\right]\Bigg\}. \qquad (14)
\end{aligned}
$$

The long time property of the AFC, shown in Fig. 3(b), similar to the case in a sheet and the asymptotic behaviors are classified as

$$
\frac{C_Q(t)}{C_0} \propto \begin{cases} t^{-1} & \text{as } |n_1-n_2| \approx N \text{ and } |m_1-m_2| \approx M/2 \\ t^{-2} & \text{as } |n_1-n_2| \ll N,\ |m_1-m_2| \ll M/2 \text{ and} (n_1,m_1)\text{-} \\ & \&\ (n_2,m_2)\text{-sites are parallel to or far away from} \\ & \text{the boundaries,} \\ t^{-3} & \text{otherwise.} \end{cases} \qquad (15)
$$

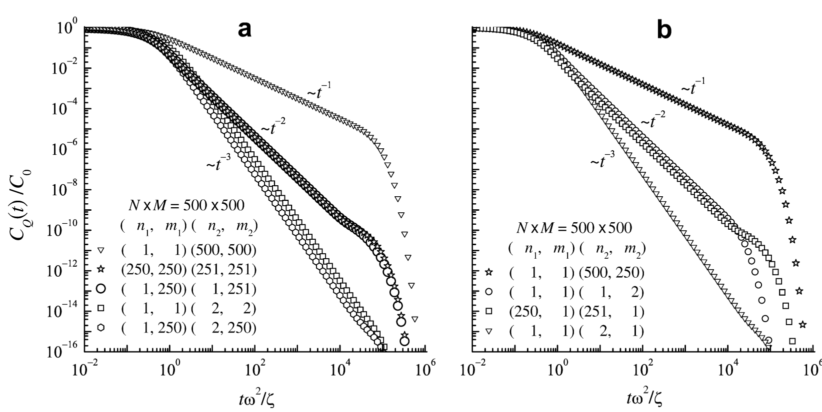

Fig. 3. The calculated $C_Q(t)/C_0$ for the various separations in a sheet (a) and a hallow tube (b) with $\omega = \omega_x = \omega_y$ and $N = M = 500$. The decay follows an full-integer power law, and becomes single exponential decays at much longer time.

The analytical derivations of the asymptotic power-law behaviors can be found in A.2.

2.5. A ladder

Although C_Q of a ladder can be formed by modifying Eq. (11) to an $N \times 2$ sheet, it behaves more like a chain. The numerical results and the analytical asymptotic approximation of Eq. (11) as $N \times 2$ in the long time are shown in Fig. 4 and A.3, respectively. The ACF formed the following behaviors at certain topological conditions:

$$\frac{C_Q(t)}{C_0} \propto \begin{cases} e^{-\alpha t} & \text{as two beads are at the same level } (i.e.\ n_1 = n_2), \\ t^{-1/2} & \text{as } |n_1 - n_2| \approx N, \\ t^{-3/2} & \text{as } |n_1 - n_2| \ll N, \text{and both } n_1 \ \&\ n_2 \text{ away} \\ & \text{from ends,} \\ t^{-5/2} & \text{otherwise.} \end{cases} \tag{16}$$

At the same level of a ladder, the exponential time behavior implies that these two beads have very fast synchronized motions due to the link between them. It may also contribute to the stability in molecules with a ladder structure, such as DNA.

3. Origins for different exponents of the power law

In the one-dimensional cases, the physical origin of the different asymptotic behaviors are given as following: The ACF of distance deviation is proportional to $\langle \vec{q}_{n_1}(t) \cdot \vec{q}_{n_1}(0) + \vec{q}_{n_2}(t) \cdot \vec{q}_{n_2}(0) - \vec{q}_{n_1}(t) \cdot \vec{q}_{n_2}(0) - \vec{q}_{n_2}(t) \cdot \vec{q}_{n_1}(0) \rangle$. The first two terms represent the ACF of distance deviation at the n_1th and n_2th sites, respectively. The last two terms are the correlation function of the displacement vector between the n_1th and n_2th beads. They also correspond to the displacement in a diffusion-like process from one bead to another since the interaction between beads are described by the Langevin equation, Eq. (1). These terms are much smaller than the first two because there is a long separation between beads. Therefore, the C_Q is dominated by the change of the displacement amplitude in itself by diffusion, which results in the usual $t^{-1/2}$ power-law behavior in the 1-D system. By rearranging terms, the C_Q is also proportional to $\langle [\vec{q}_{n_1}(t) - \vec{q}_{n_2}(t)] \cdot \vec{q}_{n_1}(0) + [\vec{q}_{n_2}(t) - \vec{q}_{n_1}(t)] \quad \cdot \vec{q}_{n_2}(0) \rangle$. The $\langle [\vec{q}_{n_1}(t) - \vec{q}_{n_2}(t)] \cdot \vec{q}_{n_1}(0) \rangle$ term represents the difference of the diffused displacement vector (originated from the n_1th bead) between the n_1th and n_2th beads at a time t. If the separation between them is comparable to or smaller than the diffusion distance of the displacement vector, the magnitude of $\langle \vec{q}_{n_2}(t) \cdot \vec{q}_{n_1}(0) \rangle$ is similar to that of $\langle \vec{q}_{n_1}(t) \cdot \vec{q}_{n_1}(0) \rangle$. A difference of these two terms corresponds to a time differential of the displacement vector, *i.e.* $\mathrm{d}\langle \vec{q}_{n_1}(t) \cdot \vec{q}_{n_1}(0) \rangle / \mathrm{d}t$, if the separation between the n_1th and n_2th beads is much smaller than the diffusion distance. Thus, it results in the $t^{-3/2}$ asymptotic behavior for the ACF for a small separation of beads. As the separation of the beads increases, the asymptotic behavior is shown in Fig. 2(b), having $t^{-1/2}$ initially and then becomes $t^{-3/2}$ at later time.

If two beads are close and near an end in a long chain, the ACF yields a $t^{-5/2}$ asymptotic behavior, shown in Fig. 2(a), instead of $t^{-3/2}$ as two close beads in a loop or far from ends in a chain. It is due to boundary conditions at the ends of a chain. Because of the reflection of distance deviation at the end of a chain, the value of $\langle [\vec{q}_{n_1}(t) - \vec{q}_{n_2}(t)] \cdot \vec{q}_{n_1}(0) \rangle$ is similar to taking a second time derivative at $\langle \vec{q}_{n_1}(t) \vec{q}_{n_1}(0) \rangle$ if n_1 and n_2 are close and near the end in an open chain.

Due to the same reason in one 1-D systems, the ACF in 2-D systems has one power smaller in large separation than in short one, shown in Fig. 3. It also means that the interference between the pairwise beads that are close is faster and stronger, which is similar to the 1-D cases. The long-time behavior for close pairwise beads near a corner in a large sheet is t^{-3}, instead of t^{-2} for beads near the center. It is due to the reflection at the boundaries. If the two beads are near the edge but far away from the corner, the ACF of distance deviation can have different long-time behavior, which depends on the orientation of the beads with the edge. If the beads are parallel to the edge and not close to a corner, the reflection of diffusion at the boundary does not affect the ACF of displacement. Therefore, the long-time behavior, shown in Fig. 3, has the t^{-2} character, similar to the case of two beads that are close but both far away from boundaries. If the pairwise beads are not parallel to an edge, the reflection at the boundary does affect the C_Q values. Therefore, the long-time behavior has t^{-3} asymptotic behavior, as seen in Fig. 3(a) and (b) for sheet and tube cases, respectively.

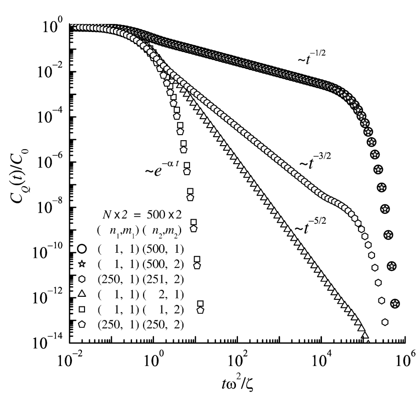

Fig. 4. The calculated $C_Q(t)/C_0$ for the various separations in a ladder with $\omega = \omega_x = \omega_y$ and $N = 500$. The decay is a single exponential if the pairwise beads are at the same level, and follows half-integer power law otherwise. All become single exponential decays eventually.

4. Discussion and conclusions

In this work, we have studied Langevin dynamics for an ideal 1-D and 2-D systems. Several kinds of power-law behavior are obtained and their exponent is found to be dependent on the distance of the pairwise beads such as a donor and an acceptor in electron or energy transfer. Studies of pairwise distance correlation using single-molecule spectroscopy are important in elucidating molecular dynamics. As shown in Figs. 2–4, the power-law behavior lasts for about six decades. Using the typical van der Waals couplings and the experimental values of the friction constant [32,38], the time range to observe such power-law behavior is estimated to cover picosecond to microsecond range.

This work shows that the distance ACF of a distant donor–acceptor pair follows a power law of $t^{-d/2}$ for a simple Rouse model with a d-dimension coupling network. This power-law behavior is very similar to d-dimension diffusion based on the scale invariance argument. The similarity is due to mathematical analogy between the Langevin equation of a Rouse model to a d-dimensional random walk process. However, for a close pair, our work predicts an interesting but different power law than the usual $t^{-d/2}$ dependence. For example, for an adjacent donor-acceptor pair of a 1-D Rouse chain, the power law does not follows the ordinary $t^{-1/2}$ behavior but follow $t^{-3/2}$ or $t^{-5/2}$ behavior, depending on the location of the donor–acceptor pair.

The half-integer power law ($t^{-1/2}$, $t^{-3/2}$ and $t^{-5/2}$) are obtained for the 1-D systems; and the full-integer power law (t^{-1}, t^{-2} and t^{-3}) for the 2-D systems. In 1-D (or 2-D) system, the $t^{-1/2}$ (or t^{-1}) power law is obtained if the pairwise beads have a large separation and very weak interference. The $t^{-3/2}$ (or t^{-2}) power-law behavior is found if the pairwise beads are near but far away from the boundaries. It is also found that if the pairwise beads in a 2-D system are near and parallel the boundaries, the reflection of the interference from the boundaries has no effects on the dynamics. The $t^{-5/2}$ (or t^{-3}) power law is obtained if the close pairwise beads are close to but not parallel to the boundaries.

The Langevin dynamics in a ladder is more special, and it yields both pure exponential and power-law ($t^{-1/2}$, $t^{-3/2}$ and $t^{-5/2}$) behaviors. The conditions and reasons for different power laws are similar to the cases of an open chain. The pure exponential behavior happens if two beads are at the same level of a ladder. It also implies that the distance fluctuation between two beads at the same level decays very fast as compared to those at different ladder levels.

In our treatment we focused on ACF of the distance fluctuations between a donor–acceptor pair. It can be directly applied in electron transfer reactions which have simple exponential distance dependence for the electronic coupling. The temporal behavior of the distance ACF is sensitive to the specific dynamics of the donor–acceptor pair and their interactions with surrounding atoms or molecules. For other processes such as FRET which has different distance dependence ($\sim 1/R^6$) and other dynamics with orientational dependence as occurs in a 2-D rotor [28], a characteristically different temporal behavior for the ACF could arise. In some special cases, power laws may happen for the temporal behavior of the ACF in FRET. With anomalous diffusion in the subdiffusion regime ($\langle x^2 \rangle \propto t^{\alpha}$, $0 < \alpha < 1$), the temporal behavior of the lifetime fluctuation correlation at long time could be written as a summation of six terms with different powers in time [23].

For native proteins and other macromolecules with heavy cross links the simple Rouse model treatment is insufficient. The presence of heavy cross links in native folded proteins could lead to deviation from the simple power law [21]. However, in polypeptides one can treat aperiodic chain by assigning a distribution of non-identical force constants among those springs between beads, except that for such a case with various spring constants a simple analytic solution is no longer available and numerical approaches need to be used. Based on our numerical simulations for such a situation [20], we have found that the power law still applies so long as the root-mean-square deviation for the distribution for the force constant is not too broad. Otherwise, these simple power-law behaviors for a system with a more complicate coupling network could break down.

Here we suggested some experiments to test the power laws of various exponents. If an electron/energy donor and an acceptor are attached to a 1-D system such as polypeptides and polymers, or a 2-D system such as a membrane, or a ladder structure such as DNA, studies of the distance fluctuations between the donor–acceptor pair could provide information about the conformational dynamics of these low-dimensional structures as beautifully illustrated by single-molecule experiments of proteins by Xie's group [16–18]. It would be interesting if the predicted power law of various exponent can be observed with a donor–acceptor pair at different topological conditions. By varying the location and the separation between the donor and the acceptor, one could explore the power law of $t^{-3/2}$, $t^{-5/2}$, in addition to the more well-known $t^{-1/2}$ law.

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Appendix A. The Asymptotic solutions of the power-law behavior

A.1. One-dimensional system

In the limit of a loop with a very large N and a long separation between the n_1th and n_2th beads (*i.e.* $N \to \infty$ and $|n_1 - n_2| \approx N/2$), Eq. (8) can be simplified by an integral [39] as

$$\frac{C_Q(t)}{C_0} \approx \frac{\exp\left(\frac{-2\omega^2 t}{\zeta}\right)}{\pi}\int_0^{\pi} \mathrm{d}x \exp\left(\frac{-2\omega^2 t}{\zeta}\cos x\right) = \mathrm{e}^{\frac{-2\omega^2 t}{\zeta}} I_0\left(\frac{2\omega^2 t}{\zeta}\right), \tag{A.1}$$

where $I_0(z)$ is the modified Bessel function of the first kind. Because of $I_\nu(x) \approx \exp(x)/(2\pi x)^{-1/2}$ at large x [40], Eq. (A.1) has an asymptotic power law, $(4\pi\omega^2 t/\zeta)^{-1/2}$, as shown in Fig. 2(b). The Rouse equation, Eq. (2), is mathematically equivalent to a 1-D diffusion equation. The $t^{-1/2}$ power behavior is expected since the solution of a diffusion equation in 1-D system has a pre-exponential factor proportional to $t^{-1/2}$.

If N of a loop is very large but the separation between the n_1th and n_2th beads is much smaller than $N/2$ (*i.e.* $N \to \infty$ and $|n_1 - n_2| \ll N/2$ or $|n_1 - n_2 - N| \ll N/2$), Eq. (8) can be approximated by an integral that can be simplified as [39]

$$\begin{aligned}\frac{C_Q(t)}{C_0} &\approx \frac{\mathrm{e}^{\frac{-2\omega^2 t}{\zeta}}}{2\pi}\int_0^{2\pi} \mathrm{d}x\{1 - \cos[x(n_1 - n_2)]\}\mathrm{e}^{\frac{2\omega^2 t \cos x}{\zeta}} \\ &= \mathrm{e}^{\frac{-2\omega^2 t}{\zeta}}\left[I_0\left(\frac{2\omega^2 t}{\zeta}\right) - I_{|n_1-n_2|}\left(\frac{2\omega^2 t}{\zeta}\right)\right].\end{aligned} \tag{A.2}$$

If $|z|$ and $|\arg z| \leqslant \pi/2 - \delta$, one has [40]

$$I_\nu(z) = \frac{e^z}{\sqrt{2\pi z}}\left[\sum_{k=0}^{n}(\nu,k)\left(\frac{-1}{2z}\right)^k + \mathrm{O}(|z|^{-n-1})\right],$$
$$\text{where } (\nu,k) = \frac{(4\nu^2 - 1^2)(4\nu^2 - 3^2)\cdots(4\nu^2 - (2k-1)^2)}{2^{2k}k!} \text{ and } (\nu,0) = 1. \tag{A.3}$$

Therefore, at a large t the leading term of Eq. (A.2) is proportional to $t^{-3/2}$. In the loop case, the asymptotic behavior of the distance ACF for a small separation is $t^{-3/2}$, which is different from the $t^{-1/2}$ behavior in a large separation, as seen in Fig. 2(b).

If two beads in a long chain have a short separation and are not close to the ends, the C_Q exhibits $t^{-3/2}$ behavior, as shown in Fig. 2(a). The behavior can also be expressed as Eq. (A.2) in a large loop. If two beads have a long distance separation (*i.e.* $n_1, n_2' \ll N$; $n_2 = N - n_2'$; and $N \to \infty$), the ACF of distance deviation can be written as

$$\begin{aligned}\frac{C_Q(t)}{C_0} &\approx \frac{\mathrm{e}^{\frac{-2\omega^2 t}{\zeta}}}{\pi}\int_0^{\pi} \mathrm{d}x\left\{\cos\left[x\left(n_1 + \frac{1}{2}\right)\right] - \cos\left[x\left(n_2 + \frac{1}{2}\right)\right]\right\}^2 \mathrm{e}^{\frac{2\omega^2 t\cos x}{\zeta}} \\ &= \mathrm{e}^{\frac{-2\omega^2 t}{\zeta}}\left[I_0\left(\frac{2\omega^2 t}{\zeta}\right) + \frac{1}{2}I_{2n_1+1}\left(\frac{2\omega^2 t}{\zeta}\right) + \frac{1}{2}I_{2n_2'-1}\left(\frac{2\omega^2 t}{\zeta}\right)\right].\end{aligned} \tag{A.4}$$

Using Eq. (A.3) the above equation at a large t yields a $t^{-1/2}$ behavior, which is similar to the behavior of two beads having a large separation for a loop case. Eq. (4) can be expressed by integral if N is very large and the beads are close to one end of the open chain (*i.e.* $N \to \infty$ and n_1, $n_2 \ll N$). It can be further simplified [39] as

$$\begin{aligned}\frac{C_Q(t)}{C_0} \approx \mathrm{e}^{\frac{-2\omega^2 t}{\zeta}}\Bigg[&I_0\left(\frac{2\omega^2 t}{\zeta}\right) - I_{n_1+n_2+1}\left(\frac{2\omega^2 t}{\zeta}\right) - I_{|n_1-n_2|}\left(\frac{2\omega^2 t}{\zeta}\right) \\ &+ \frac{1}{2}I_{2n_1+1}\left(\frac{2\omega^2 t}{\zeta}\right) + \frac{1}{2}I_{2n_2+1}\left(\frac{2\omega^2 t}{\zeta}\right)\Bigg].\end{aligned} \tag{A.5}$$

At a large t the leading term of Eq. (A.5) is proportional to $t^{-5/2}$, which is the same as the asymptotic behavior shown in Fig. 2(a).

A.2. Two-dimensional system

The C_Q of a sheet, Eq. (11), between the pairwise beads at (n_1, m_1) and (n_2, m_2) sites with a very large separation in the limit of a very large $N \times M$ (*i.e.* $N \to \infty$, $M \to \infty$, and $|m_1 - m_2| \approx M$) can be simplified as

$$\begin{aligned}\frac{C_Q(t)}{C_0} \approx \mathrm{e}^{-\frac{2t}{\zeta}(\omega_x^2+\omega_y^2)} I_0\left(\frac{2\omega_y^2 t}{\zeta}\right)\Bigg[&I_0\left(\frac{2\omega_x^2 t}{\zeta}\right) + \frac{1}{2}I_{2n_1-1}\left(\frac{2\omega_x^2 t}{\zeta}\right) \\ &+ \frac{1}{2}I_{2n_2-1}\left(\frac{2\omega_x^2 t}{\zeta}\right)\Bigg].\end{aligned} \tag{A.6}$$

Using Eq. (A.3), the leading term of the equation has t^{-1} dependence. The asymptotic behavior at large t is shown in Fig. 3(a). It is similar to the long-time behavior for two largely-separated beads in a very large and long tube, shown in Fig. 3(b), since Eq. (14) can also be approximate as Eq. (A.6). Therefore, the ACF of the distance deviation between widely separated beads in two-dimensional cases yields a t^{-1} asymptotic behavior.

If the separation of beads on a sheet is close and near the center, and $N \times M$ is very large, (*i.e.* n_1, $n_2 \approx N/2$ and m_1, $m_2 \approx M/2$) Eq. (10) can be simplified by an integral [39] as

$$\begin{aligned}\frac{C_Q(t)}{C_0} \approx \mathrm{e}^{\left[-\frac{2t}{\zeta}(\omega_x^2+\omega_y^2)\right]}\Bigg\{&I_0\left(\frac{2\omega_x^2 t}{\zeta}\right)I_0\left(\frac{2\omega_y^2 t}{\zeta}\right) \\ &- I_{|n_1-n_2|}\left(\frac{2\omega_x^2 t}{\zeta}\right)I_{|m_1-m_2|}\left(\frac{2\omega_y^2 t}{\zeta}\right)\Bigg\}.\end{aligned} \tag{A.7}$$

The equation can be expended by using Eq. (A.3), and has a leading term in t^{-2}, which is consistent with the asymptotic behavior shown in Fig. 3(a). This physical origin of one power smaller than in large separation is due to the interaction of the pairwise beads of the ACF through fewer beads, which is similar to the one-dimensional cases. If two beads are near a corner on a large sheet (*i.e.* $n_1, n_2 \ll N$ and $m_1,\ m_2 \ll M$). The ACF of distance deviation can be rewritten as

$$\begin{aligned}\frac{C_Q(t)}{C_0} \approx\ & \mathrm{e}^{\left[-\frac{2t}{\zeta}(\omega_x^2+\omega_y^2)\right]}\left\{\frac{1}{2}\left[I_0\left(\frac{2\omega_x^2 t}{\zeta}\right)+I_{2n_1-1}\left(\frac{2\omega_x^2 t}{\zeta}\right)\right]\right.\\ & \times\left[I_0\left(\frac{2\omega_y^2 t}{\zeta}\right)+I_{2m_1-1}\left(\frac{2\omega_y^2 t}{\zeta}\right)\right]\\ & +\frac{1}{2}\left[I_0\left(\frac{2\omega_x^2 t}{\zeta}\right)+I_{2n_2-1}\left(\frac{2\omega_x^2 t}{\zeta}\right)\right]\\ & \times\left[I_0\left(\frac{2\omega_y^2 t}{\zeta}\right)+I_{2m_2-1}\left(\frac{2\omega_y^2 t}{\zeta}\right)\right]\\ & -\left[I_{n_1+n_2-1}\left(\frac{2\omega_x^2 t}{\zeta}\right)+I_{|n_1-n_2|}\left(\frac{2\omega_x^2 t}{\zeta}\right)\right]\\ & \left.\times\left[I_{m_1+m_2-1}\left(\frac{2\omega_y^2 t}{\zeta}\right)+I_{|m_1-m_2|}\left(\frac{2\omega_y^2 t}{\zeta}\right)\right]\right\}. \end{aligned}\tag{A.8}$$

As expected, because of the reflection distance deviations at the boundaries, the long-time behavior is t^{-3}, instead of t^{-2} for beads near the center.

If the pairwise beads on a very long and large tube are close and far away from the edges, the ACF of distance deviation in Eq. (14) can also be expressed as Eq. (A.7). Thus, it also exhibits t^{-2} asymptotic behavior, as seen in Fig. 3(b). If the close pairwise beads are parallel and near an edge (*i.e.* $n_1 = n_2 = n \ll N$, $|m_1 - m_2| \ll M/2$, and $N,M \to \infty$), C_Q can be written as

$$\begin{aligned}\frac{C_Q(t)}{C_0} \approx\ & \mathrm{e}^{\left[-\frac{2t}{\zeta}(\omega_x^2+\omega_y^2)\right]}\left[I_0\left(\frac{2\omega_x^2 t}{\zeta}\right)+I_{2n-1}\left(\frac{2\omega_x^2 t}{\zeta}\right)\right]\\ & \times\left[I_0\left(\frac{2\omega_y^2 t}{\zeta}\right)-I_{|m_1-m_2|}\left(\frac{2\omega_y^2 t}{\zeta}\right)\right]. \end{aligned}\tag{A.9}$$

According to Eq. (A.3), it yields t^{-2} behavior in Fig. 3(b), which is similar to the results of the pairwise beads parallel to the edge and far away from a corner of a sheet. If the pairwise beads are close and unparallel the edges, the Eq. (14) can be expressed as

$$\begin{aligned}\frac{C_Q(t)}{C_0} \approx\ & \mathrm{e}^{\left[-\frac{2t}{\zeta}(\omega_x^2+\omega_y^2)\right]}\left\{I_0\left(\frac{2\omega_y^2 t}{\zeta}\right)\left[I_0\left(\frac{2\omega_x^2 t}{\zeta}\right)+\frac{1}{2}I_{2n_1-1}\left(\frac{2\omega_x^2 t}{\zeta}\right)\right.\right.\\ & \left.+\frac{1}{2}I_{2n_2-1}\left(\frac{2\omega_x^2 t}{\zeta}\right)\right]-I_{|m_1-m_2|}\left(\frac{2\omega_y^2 t}{\zeta}\right)\left[I_{n_1+n_2-1}\left(\frac{2\omega_x^2 t}{\zeta}\right)\right.\\ & \left.\left.+I_{|n_1-n_2|}\left(\frac{2\omega_x^2 t}{\zeta}\right)\right]\right\}, \end{aligned}\tag{A.10}$$

when $|n_1 - n_2| \ll N$ and $|m_1 - m_2| \ll M/2$ in a very large and long tube. The time behavior in Fig. 3 becomes t^{-3} at long time. The power law changes if the pairwise beads change the orientation with respect to the edges. The change is due to the reflection at the boundary having different effects at the different orientation of the pairwise beads. The effects are similar to the results on a sheet.

A.3. A ladder

The analytical expression at the limit of a very large N can be derived by using the integral in Ref. [39]. If $n_1 = n_2 = n$, it can be written as

$$\frac{C_Q(t)}{C_0} \approx \exp\left[-\frac{2t}{\zeta}(\omega_x^2+\omega_y^2)\right]\left[I_0\left(\frac{2\omega_x^2 t}{\zeta}\right)+I_{2n-1}\left(\frac{2\omega_x^2 t}{\zeta}\right)\right]. \tag{A.11}$$

Using Eq. (A.3), at a large t the asymptotic behavior is $(4\pi\omega_x^2 t/\zeta)^{-1/2}\exp(-2\omega_y^2 t/\zeta)$, as shown in Fig. 4, which is very different from the power-law results in the usual one- or two-dimensional systems. If the pairwise beads on a very long ladder are at a large separation (*i.e.* $|n_1 - n_2| \approx N$, $N \to \infty$), the ACF becomes

$$\frac{C_Q(t)}{C_0} \approx \frac{1}{2}\exp\left(-\frac{2\omega_x^2 t}{\zeta}\right)\left[1+\exp\left(-\frac{2\omega_y^2 t}{\zeta}\right)\right]I_0\left(\frac{2\omega_x^2 t}{\zeta}\right), \tag{A.12}$$

which exhibits $t^{-1/2}$ in Fig. 4 at longer times. The result is the same as that obtained in both chain and loop cases with a large separation. When the pairwise beads have a short separation and far away from the ends of the very long ladder, C_Q can be simplified by

$$\begin{aligned}\frac{C_Q(t)}{C_0} \approx \frac{\mathrm{e}^{-\frac{2\omega_x^2 t}{\zeta}}}{2}&\left\{\left[1+\mathrm{e}^{-\frac{2\omega_y^2 t}{\zeta}}\right]I_0\left(\frac{2\omega_x^2 t}{\zeta}\right)-\left[1\pm \mathrm{e}^{-\frac{2\omega_y^2 t}{\zeta}}\right]\right.\\ & \left.\times I_{|n_2-n_1|}\left(\frac{2\omega_y^2 t}{\zeta}\right)\right\}(\text{“}+\text{” if } m_1 = m_2;\ \text{“}-\text{” if } m_1 \neq m_2). \end{aligned}\tag{A.13}$$

As curve with the open hexagon symbols in Fig. 4, it yields $t^{-3/2}$ power-law behavior at longer times. It is the same as the 1-D case if the pairwise beads are close and far away from the boundaries.

When the two beads are close and near the ends of the very long ladder, (*i.e.* $n_1 \neq n_2$ and $n_1,n_2 \ll N$), the ACF is expressed as

$$\begin{aligned}\frac{C_Q(t)}{C_0} \approx \frac{\mathrm{e}^{-\frac{2\omega_x^2 t}{\zeta}}}{2}&\left\{\left[1+\exp\left(-\frac{2\omega_y^2 t}{\zeta}\right)\right]\left[I_0\left(\frac{2\omega_x^2 t}{\zeta}\right)\right.\right.\\ & \left.+\frac{1}{2}I_{2n_1-1}\left(\frac{2\omega_x^2 t}{\zeta}\right)+\frac{1}{2}I_{2n_2-1}\left(\frac{2\omega_x^2 t}{\zeta}\right)\right]\\ & \left.-\left[1\pm\exp\left(-\frac{2\omega_y^2 t}{\zeta}\right)\right]\left[I_{n_1+n_2+1}\left(\frac{2\omega_x^2 t}{\zeta}\right)+I_{|n_2-n_1|}\left(\frac{2\omega_x^2 t}{\zeta}\right)\right]\right\}\\ & (\text{“}+\text{” if } m_1 = m_2;\ \text{“}-\text{” if } m_1 \neq m_2). \end{aligned}\tag{A.14}$$

Its leading asymptotic term has a power law of $t^{-5/2}$, represented by the open-triangle-curve in Fig. 4, which is the same as obtained by the close pairwise beads near an end of a very long open chain.

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