

Relaxation functions of the Ornstein-Uhlenbeck process with fluctuating diffusivity

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We study the relaxation behavior of the Ornstein-Uhlenbeck (OU) process with time-dependent and fluctuating diffusivity. In this process, the dynamics of the position vector is modeled by the Langevin equation with a linear restoring force and a fluctuating diffusivity (FD). This process can be interpreted as a simple model of relaxation dynamics with internal degrees of freedom or in a heterogeneous environment. By utilizing the functional integral expression and the transfer matrix method, we show that the relaxation function can be expressed in terms of the eigenvalues and eigenfunctions of the transfer matrix for general FD processes. We apply our general theory to two simple FD processes where the FD is described by the Markovian two-state model or an OU-type process. We show analytic expressions of the relaxation functions in these models and their asymptotic forms. We also show that the relaxation behavior of the OU process with an FD is qualitatively different from those obtained from conventional models such as the generalized Langevin equation.

DOI: [10.1103/PhysRevE.99.032127](https://doi.org/10.1103/PhysRevE.99.032127)**I. INTRODUCTION**

Recently, diffusion of Brownian particles with time-dependent and fluctuating diffusivity has been studied extensively in many contexts such as in heterogeneous environments and a simple model of normal yet non-Gaussian diffusion [1–11]. (The anomalous diffusion behavior of time-dependent diffusivity models has been also studied [12,13].) The Langevin equation with a time-dependent and fluctuating diffusivity exhibits nontrivial diffusion behavior. If the system is in equilibrium, the ensemble-averaged mean-square displacement (MSD) reduces to just a normal diffusion which is the same as the simple Brownian motion. However, this does not mean that the diffusion behavior can be expressed as a normal and Gaussian process. If we consider the (time-averaged) MSD data for individual realizations, the distribution of MSD data around their ensemble-average does not obey the Gaussian distribution. This is because the thermal noise becomes non-Gaussian due to the multiplicative coupling between the fluctuating diffusivity (FD) and the Gaussian noise. The non-Gaussian nature becomes evident if we consider the higher-order correlation functions. For example, we have shown that the relative standard deviation of the time-averaged MSD (the square of which is also known as the ergodicity breaking parameter [14,15]) can be related to the correlation function of an FD [7].

The concept of the FD seems to be useful in studying the microscopic dynamical behavior of molecules with an internal degrees of freedom and/or in heterogeneous environments. For example, protein dynamics has many internal degrees of freedom and the conformational dynamics exhibits

non-single-exponential-type relaxation behavior [5,16,17]. The diffusivity of the center of mass of an entangled polymer depends on the end-to-end vector of the polymer chain [7,18,19]. A molecule in a supercooled liquids exhibits heterogeneous and fluctuating diffusion behavior, which is known as dynamic heterogeneity [20–22]. We expect that other dynamical behavior, such as the relaxation behavior, in these systems reflects their FDs.

Unfortunately, from the viewpoint of experiments, the direct observations of MSDs of individual molecules are not easy in some systems. Instead of the direct observations of individual molecules, macroscopic measurements of relaxation functions (response functions) are useful. The macroscopic relaxation functions can be measured by imposing an external perturbation on the system and monitoring the response of the system. The linear-response theory gives the relation between the time-correlation function of microscopic variables and the macroscopic response function [23]. Thus we can study the microscopic molecular-level dynamics from the macroscopic response functions. For example, we can investigate the microscopic molecular dynamics of polymers from the experimental data of the dielectric relaxation function (the response function of the electric flux to the imposed electric field) and the relaxation modulus (the response function of the stress to the imposed strain) [24,25].

At the molecular level, we expect that the relaxation dynamics can be described by a Langevin equation with a restoring force. The Ornstein-Uhlenbeck (OU) process [26] is the simplest model to describe such dynamics. For example, in the harmonic dumbbell model for a polymer, the dynamics is described as an OU-type process. The dynamics of a

colloidal particle under an optical tweezer can also be modeled as an OU-type process. If a molecule or a particle is in a heterogeneous environment (such as a supercooled liquid), we expect that the relaxation behavior is affected by an FD arising from the heterogeneity. The relaxation behavior of a polymer or a colloidal particle in a supercooled environment will be affected by both a restoring force and a heterogeneous environment. The dynamics of a protein may also be interpreted as relaxation dynamics under a heterogeneous environment.

Thus we expect that the relaxation behavior of the systems with FDs would be informative in modeling and/or analyzing the microscopic molecular dynamics of polymers and supercooled liquids. However, the relaxation behavior of the Langevin equation with an FD has not been studied in detail. In this work we consider an OU-type process with a time-dependent and fluctuating diffusivity as a simple and analytically tractable relaxation model with an FD. We analyze relaxation functions theoretically and show that the relaxation functions are strongly affected by the dynamics of the diffusivity. We give a formal expression for the relaxation function for a general Markovian dynamics of diffusivity. We show that the form of the relaxation function is determined as a result of the competition between the relaxation (diffusion) dynamics in the OU process and the transition dynamics of the FD. Then we apply our result to two simple and solvable models. One is the Markovian two-state model where the diffusivity can take only two different values. The other is the OU-type model where the noise coefficient obeys an OU process. We discuss the properties of the relaxation functions by the OU process with an FD and possible applications of our theoretical results to the analyses of experimental data.

II. MODEL

We consider the dynamics of a position in a d -dimensional space \mathbf{r} . This position can be interpreted as the position of a tagged particle in the system or a bond vector which connects two particles (in the dumbbell model [27]). In both cases, at the mesoscopic scale, the dynamics of the position can be reasonably described by the Langevin-type equation. We consider the dynamics of a particle or a bond vector in a heterogeneous environment, where the diffusivity (or the mobility) fluctuates in time. The effect of the inertia term in a dynamic equation is generally small at the mesoscopic scale and thus we can safely ignore the inertial term (the overdamped limit). As a simple model to describe such systems, we employ the Langevin equation with an FD [7]

$$\frac{d\mathbf{r}(t)}{dt} = \frac{D(t)}{k_B T} \mathbf{F}(\mathbf{r}, t) + \sqrt{2D(t)} \mathbf{w}(t). \quad (1)$$

Here k_B is the Boltzmann constant, T is the temperature, $D(t)$ is a time-dependent and fluctuating diffusivity, $\mathbf{F}(\mathbf{r}, t)$ is the force for the position, and $\mathbf{w}(t)$ is the Gaussian white noise. We limited ourselves to the case where the diffusion coefficient is given as a scalar quantity. (In general, the diffusion coefficient is a tensor quantity [7,28].) From the fluctuation-dissipation relation of the second kind, the first and second moments of the Gaussian noise $\mathbf{w}(t)$ are $\langle \mathbf{w}(t) \rangle = 0$ and $\langle \mathbf{w}(t) \mathbf{w}(t') \rangle = \mathbf{1} \delta(t - t')$, where $\langle \cdots \rangle$ represents the statistical average and $\mathbf{1}$ is the unit tensor in d dimensions. We

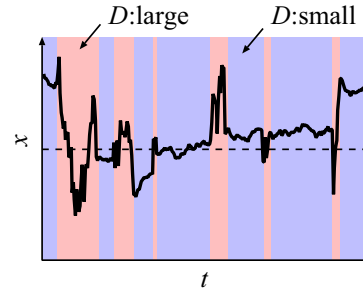


FIG. 1. Example of a realization of the stochastic process which obeys the OUFD in one dimension $x(t)$. The solid black curve represent the position $x(t)$ at time t and the background colors represents the diffusivity $D(t)$. The position is fluctuating around the origin $x = 0$ (dashed black line) due to the restoring force.

assume that the dynamics of $D(t)$ is statistically independent of the noise $\mathbf{w}(t)$. Namely, the dynamics of $D(t)$ is statistically not affected by the dynamics of $\mathbf{r}(t)$.

Equation (1) can be interpreted as a simplified model for the dynamics of supercooled polymer melts [29,30] or trapped Brownian particles in heterogeneous environments [31–33]. For the case of a polymer chain in a supercooled melt, we consider the end-to-end vector as the bond vector and the effective potential can be expressed as a harmonic potential [19]. For the case of a trapped Brownian particle, we consider the situation where the Brownian particle is trapped by an optical tweezer. The effective potential by an optical tweezer can be approximated well by a harmonic potential [34]. Thus we assume that the force in Eq. (1) is derived from a harmonic potential $U(\mathbf{r}) = \alpha k_B T r^2 / 2$, where α is constant. (If the force is purely entropic, α is independent of the temperature T [19]. However, in general, α depends on the temperature. Fortunately, the explicit form of α and its temperature dependence is not important for the analysis in this work.) The force becomes $\mathbf{F} = -\partial U(\mathbf{r}) / \partial \mathbf{r} = -\alpha k_B T \mathbf{r}$ and Eq. (1) reduces to the following OU process with an FD (OUFD):

$$\frac{d\mathbf{r}(t)}{dt} = -\alpha D(t) \mathbf{r}(t) + \sqrt{2D(t)} \mathbf{w}(t). \quad (2)$$

We show an example of the OUFD process in Fig. 1. If $D(t)$ is constant, Eq. (2) reduces to a usual OU process [26] and we can easily analyze it. (As clearly observed in Fig. 1, the fluctuation of the diffusivity qualitatively affects the stochastic process and thus generally the OUFD behaves in a different way from a usual OU process.)

To fully describe the OUFD, we need a dynamic equation for $D(t)$. We assume that the dynamics of $D(t)$ obeys a Markovian stochastic process and $D(t)$ is sampled from the equilibrium ensemble. (The effect of nonequilibrium initial ensembles to the OU process with a constant diffusivity was recently studied [35].) For example, we can employ a Markovian jump processes between several discrete states for the diffusivity or a Langevin equation for the diffusivity. In any case, as long as the process is Markovian, we can formally express the time-evolution equation for the probability distribution of D by a master equation as

$$\frac{\partial P(D, t)}{\partial t} = \hat{\mathcal{L}} P(D, t), \quad (3)$$

where $P(D, t)$ is the probability distribution of D at time t and $\hat{\mathcal{L}}$ is a linear operator (such as the Fokker-Planck operator and the transition matrix [26]). Alternatively, more generally, we assume that the diffusivity $D(t)$ is obtained from a Markovian variable $\xi(t)$ as $D(t) = D(\xi(t))$ and that its probability distribution follows a master equation

$$\frac{\partial P(\xi, t)}{\partial t} = \hat{\mathcal{L}}P(\xi, t). \quad (4)$$

Note that Eq. (3) is a special case for which $D(t) = \xi(t)$.

To study the relaxational behavior of the OU process, we consider the following relaxation function:

$$\Phi(t) \equiv \frac{\langle \mathbf{r}(t) \cdot \mathbf{r}(0) \rangle}{\langle \mathbf{r}^2(0) \rangle}. \quad (5)$$

Here it should be noticed that the statistical average $\langle \dots \rangle$ is taken for both the noise $\mathbf{w}(t)$ and the diffusion coefficient $D(t)$. In this work we take the statistical average of $\mathbf{w}(t)$ before that of $D(t)$. The order of two statistical averages does not affect the results in most cases, including the equilibrium systems. Equation (5) can be interpreted as the normalized relaxation function of $\mathbf{r}(t)$. If we interpret $\mathbf{r}(t)$ as the bond vector and it has an electric dipole, Eq. (5) can be related to the dielectric relaxation function [24]. If we interpret $\mathbf{r}(t)$ as the position of a trapped particle, Eq. (5) can be related to the ensemble-averaged MSD as

$$\langle [\mathbf{r}(t) - \mathbf{r}(0)]^2 \rangle = 2\langle \mathbf{r}^2(0) \rangle [1 - \Phi(t)]. \quad (6)$$

Therefore, if the relaxation function $\Phi(t)$ exhibits some non-trivial properties, we will observe them via the ensemble-averaged MSD. This is in contrast to the case without a trap potential, where the ensemble-averaged MSD exhibits just a normal diffusion.

We calculate the relaxation function $\Phi(t)$ for the OUFD, from Eqs. (2) and (5). By integrating Eq. (2) from time 0 to t , we have

$$\begin{aligned} \mathbf{r}(t) = & \exp\left[-\alpha \int_0^t dt' D(t')\right] \mathbf{r}(0) \\ & + \int_0^t dt' \exp\left[-\alpha \int_{t'}^t dt'' D(t'')\right] \sqrt{2D(t')} \mathbf{w}(t'). \end{aligned} \quad (7)$$

Because $D(t)$, $\mathbf{w}(t)$ (for $t > 0$), and $\mathbf{r}(0)$ are statistically independent of each other, we take the average over $\mathbf{w}(t)$ and $\mathbf{r}(0)$. Then we have

$$\langle \mathbf{r}(t) \cdot \mathbf{r}(0) \rangle = \left\langle \exp\left[-\alpha \int_0^t dt' D(t')\right] \right\rangle \langle \mathbf{r}^2(0) \rangle \quad (8)$$

and

$$\Phi(t) = \left\langle \exp\left[-\alpha \int_0^t dt' D(t')\right] \right\rangle. \quad (9)$$

For comparison, we also consider another relaxation function defined as

$$\Psi(t) \equiv \frac{\langle [r_x(t)r_y(t)][r_x(0)r_y(0)] \rangle}{\langle [r_x(0)r_y(0)]^2 \rangle}. \quad (10)$$

We have assumed that the dimension of the space is at least 2 ($d \geq 2$). Equation (10) can be interpreted as the normalized relaxation function of the off-diagonal component of a

second-rank tensor $\mathbf{r}(t)\mathbf{r}(t)$. If we interpret $\mathbf{r}(t)$ as a bond vector, the relaxation function $\Psi(t)$ can be related to the relaxation modulus (the viscoelastic relaxation function) [24]. The relaxation function $\Psi(t)$ can be calculated in a similar way to $\Phi(t)$. From Eq. (7) we have

$$\begin{aligned} \langle r_x(t)r_y(t)r_x(0)r_y(0) \rangle = & \left\langle \exp\left[-2\alpha \int_0^t dt' D(t')\right] \right\rangle \\ & \times \langle r_x^2(0)r_y^2(0) \rangle \end{aligned} \quad (11)$$

and thus

$$\Psi(t) = \left\langle \exp\left[-2\alpha \int_0^t dt' D(t')\right] \right\rangle. \quad (12)$$

If $D(t)$ is constant, $\Psi(t)$ can be related to $\Phi(t)$ simply as $\Psi(t) = \Phi(2t) = \Phi^2(t)$. Thus the two relaxation functions are essentially the same. However, in the case of the OUFD, the relation between $\Psi(t)$ and $\Phi(t)$ is generally not that simple.

So far, the calculation of the relaxation functions $\Phi(t)$ and $\Psi(t)$ was rather straightforward [Eqs. (9) and (12)]. However, to obtain the explicit expression of these relaxation functions, we need to evaluate the ensemble averages of the state-dependent relaxation functions, which seem not to be trivial. Because the relaxation functions $\Phi(t)$ and $\Psi(t)$ have almost the same form, in the following we mainly consider the relaxation function $\Phi(t)$ unless explicitly stated. Once we have the analytic expression for $\Phi(t)$, one for $\Psi(t)$ can be easily obtained by replacing α by 2α . From Eq. (9) the relaxation function can be calculated if the statistics of the integral $\int_0^t dt' D(t')$ is known. There are several different methods to evaluate Eq. (9). In this work we will utilize the transfer matrix method [36–38], but other methods can be employed as well. For example, from the viewpoint of the renewal theory, Eq. (9) can be related to the statistics of the occupation time or the magnetization [39,40]. Thus we can utilize the methods developed in the field of the renewal theory to analyze the relaxation function. The analysis based on the renewal theory will be discussed elsewhere [41]. Also, the propagator for a free Brownian particle with an FD has a similar form to Eq. (9) [10]. Thus the analysis for a free Brownian motion would also be utilized to analyze the relaxation function and vice versa.

III. THEORY

To evaluate the statistical average over an FD, we introduce the path probability [42] which gives the statistical weight for a certain realization of $D(t)$ or $\xi(t)$. We express the diffusion coefficient as $D(t) = D(\xi(t))$, where $\xi(t)$ is a Markovian stochastic process [Eq. (4)]. [The stochastic process $\xi(t)$ can be both a continuum stochastic process and a discrete jump process.] The path probability is expressed as a functional of $\xi(t)$ and we describe it as $\mathcal{P}[\xi]$. The relaxation function $\Phi(t)$ (9) can be rewritten as

$$\Phi(t) = \int \mathcal{D}\xi \exp\left[-\alpha \int_0^t dt' D(\xi(t'))\right] \mathcal{P}[\xi], \quad (13)$$

where $\int \mathcal{D}\xi$ represents the functional integral (or the path integral) over the stochastic variable $\xi(t)$ [42,43]. We

assume that the measure of the functional integral is determined appropriately so that the total probability becomes unity. Equation (13) has a form similar to the partition function for the Ginzburg-Landau (GL) model in a one-dimensional space [44]. (In the GL model, the free energy of a system is expressed as a functional of the order parameter field $\psi(x)$, as $\mathcal{F}[\psi]$. Under a constant external field which is conjugate to the order parameter h , an extra term $\int dx h\psi(x)$ is added to the free-energy functional. The partition function under the external field is expressed as $\mathcal{Z} = \int \mathcal{D}\psi \exp[-(h/k_B T) \int dx \psi(x) - \mathcal{F}[\psi]/k_B T]$, which has the same form as Eq. (13).) In analogy to the GL model, α and $D(\xi(t))$ can be interpreted as an external field and the order parameter which is conjugate to the applied external field, respectively. (This situation would be similar to the Martin-Siggia-Rose formalism [45].) This analogy leads us to employ techniques developed for the GL model, such as the transfer matrix method.

We rewrite the path probability as $\mathcal{P}[\xi] = \exp(-\mathcal{S}[\xi])$, where $\mathcal{S}[\xi]$ is the dimensionless action functional [46]. (In analogy to the GL model, this action functional can be interpreted as the free-energy functional without an applied external field.) For convenience, we consider the discretized process for the diffusivity. Namely, we discretize time t as $t_j = j\Delta$ (with Δ the time step size) and approximate the function $\xi(t)$ by the set of discrete points $\xi_j \equiv \xi(t_j)$. Due to the Markovian nature, the action functional can be rewritten as

$$\mathcal{S}[\xi] \approx \Delta \sum_j s(\xi_{j+1}, \xi_j), \quad (14)$$

where $s(\xi_{j+1}, \xi_j)$ is a function of ξ_{j+1} and ξ_j . The functional integral can also be rewritten as $\int \mathcal{D}\xi \cdots \approx \int \prod_j d\xi_j \cdots$. For simplicity, we assume that t/Δ is a positive integer. Then we can rewrite Eq. (13) as follows:

$$\begin{aligned} \Phi(t) &\approx \int \prod_{j=0}^{t/\Delta} d\xi_j \exp \left[-\Delta \sum_{j=0}^{t/\Delta-1} \alpha D(\xi_j) - \Delta \sum_{j=0}^{t/\Delta-1} s(\xi_{j+1}, \xi_j) \right] P(\xi_0) \\ &= \int \prod_{j=0}^{t/\Delta} d\xi_j \exp \left[-\Delta \sum_{j=0}^{t/\Delta-1} [s(\xi_{j+1}, \xi_j) + \alpha D(\xi_j)] \right] P(\xi_0). \end{aligned} \quad (15)$$

Here $P(\xi_0)$ is the probability distribution of ξ at the initial state and is given as the equilibrium probability distribution $P(\xi_0) = P_{\text{eq}}(\xi_0)$. To derive Eq. (15), we have performed the functional integration over $\xi(t')$ for $t' < 0$ and $t' > t$, since the relaxation function in Eq. (13) depends on $\xi(t')$ only in the time range $0 \leq t' \leq t$. The functional integral over $\xi(t')$ for $t' > t$ becomes unity and the functional integral over $\xi(t')$ for $t' < 0$ gives the initial probability distribution $P(\xi_0)$. The function $s(\xi, \xi')$ can be related to the linear operator $\hat{\mathcal{L}}$ in Eq. (4). The formal solution of Eq. (4) for the time interval $\Delta = t_{j+1} - t_j$ is

$$P(\xi, t_{j+1}) = e^{\Delta \hat{\mathcal{L}}} P(\xi, t_j). \quad (16)$$

On the other hand, the factor $e^{-\Delta s(\xi_{j+1}, \xi_j)}$ represents the transition probability from the state ξ_j to the state ξ_{j+1} during the time interval Δ :

$$P(\xi_{j+1}, t_{j+1}) = \int d\xi_j e^{-\Delta s(\xi_{j+1}, \xi_j)} P(\xi_j, t_j). \quad (17)$$

By comparing Eqs. (16) and (17), we have the simple relation between $s(\xi, \xi')$ and $\hat{\mathcal{L}}$,

$$\int d\xi' e^{-\Delta s(\xi, \xi')} P(\xi') = e^{\Delta \hat{\mathcal{L}}} P(\xi), \quad (18)$$

where $P(\xi)$ is an arbitrary function.

Equation (18) means that the action functional can be calculated from the linear operator $\hat{\mathcal{L}}$. Then Eq. (15) can be solved by utilizing the transfer matrix technique [36–38] in a similar way. It is worth mentioning that a similar method was employed by Bressloff and Newby [2,3] to analyze the diffusion properties of a model with a time-dependent and

fluctuating diffusivity. We introduce the transfer operator $\hat{\mathcal{W}}$,

$$\begin{aligned} e^{-\Delta \hat{\mathcal{W}}} P(\xi) &= \int d\xi' \exp\{-\Delta [s(\xi, \xi') + \alpha D(\xi')]\} P(\xi') \\ &= e^{\Delta \hat{\mathcal{L}}} [e^{-\Delta \alpha D(\xi)} P(\xi)], \end{aligned} \quad (19)$$

where we have utilized Eq. (18). Since Δ is small, the exponential functions can be expanded into the power series of Δ . By keeping only the leading-order terms, we have

$$-\hat{\mathcal{W}} P(\xi) \approx [\hat{\mathcal{L}} - \alpha D(\xi)] P(\xi). \quad (20)$$

Therefore, we find that the transfer operator $\hat{\mathcal{W}}$ consists of two contributions. One is the time-evolution operator for the diffusivity $\hat{\mathcal{L}}$ and the other is the diffusivity-dependent decay factor $-\alpha D$. We call the former the transition dynamics and the latter the relaxation dynamics. At the limit of $\Delta \rightarrow 0$, Eq. (20) becomes exact. Then, from Eqs. (15) and (19), we have the following simple expression for the relaxation function $\Phi(t)$:

$$\Phi(t) = \int d\xi e^{-t \hat{\mathcal{W}}} P_{\text{eq}}(\xi). \quad (21)$$

Equation (21) means that the relaxation function $\Phi(t)$ is determined by the transfer operator $\hat{\mathcal{W}}$ and the equilibrium probability distribution of ξ .

To continue the calculation, we introduce the n th eigenvalue and eigenfunction of $\hat{\mathcal{W}}$, λ_n , and $\psi_n(\xi)$:

$$\hat{\mathcal{W}} \psi_n(\xi) = \lambda_n \psi_n(\xi). \quad (22)$$

For simplicity, here we assume that the eigenvalues are in ascending order ($\lambda_n \leq \lambda_m$ if $n < m$). We construct the basis

set by the eigenfunctions as

$$\int d\xi \psi_n^\dagger(\xi) \psi_m(\xi) = \delta_{nm}, \quad (23)$$

where $\psi_n^\dagger(\xi)$ is the eigenfunction of the adjoint operator of $\hat{\mathcal{W}}$. [In general, the transfer operator is not self-adjoint and thus $\psi_n^\dagger(\xi)$ does not coincide with $\psi_n(\xi)$. The eigenfunctions $\psi_n(\xi)$ and $\psi_n^\dagger(\xi)$ form a biorthogonal basis set [47].]

Then Eq. (15) can be rewritten in terms of the eigenvalues and eigenfunctions. We can rewrite Eq. (21) with the eigenvalues and eigenfunctions as

$$\begin{aligned} \Phi(t) &= \int d\xi \sum_n e^{-t\lambda_n} \psi_n(\xi) \int d\xi' \psi_n^\dagger(\xi') P_{\text{eq}}(\xi') \\ &= \sum_n \phi_n e^{-t\lambda_n}, \end{aligned} \quad (24)$$

where

$$\phi_n \equiv \int d\xi \psi_n(\xi) \int d\xi' \psi_n^\dagger(\xi') P_{\text{eq}}(\xi'). \quad (25)$$

From Eq. (24) we conclude that the relaxation function of the OUF is given as the sum of single-exponential relaxations, in the case where the dynamics of the diffusivity is described by a Markovian stochastic process. The relaxation rate and intensity of the n th mode are λ_n and ϕ_n , respectively. The relaxation time of the n th mode is simply given as $\tau_n \equiv 1/\lambda_n$. At the long time region, only the eigenmode with the smallest eigenvalue is dominant and the relaxation function asymptotically approaches a single-exponential-type relaxation. The longest relaxation time is τ_n with the smallest n . Intuitively, the relaxation rate λ_n is determined by the competition between the relaxation dynamics of the OU process which is characterized by the operator $-\alpha D(\xi)$ and the transition dynamics of the diffusivity which is characterized by the operator $\hat{\mathcal{L}}$.

The relaxation function $\Psi(t)$ can be calculated in almost the same way. As we mentioned, the expression for $\Psi(t)$ is obtained by replacing α in one for $\Phi(t)$ by 2α [Eqs. (9) and (12)]. This can be done by employing the following transfer operator instead of Eq. (20): $-\hat{\mathcal{W}}' P(\xi) = [\hat{\mathcal{L}} - 2\alpha D(\xi)] P(\xi)$. Alternatively, if we have the analytical expressions for λ_n , $\psi_n(\xi)$, and $\psi_n^\dagger(\xi)$, we obtain the eigenvalue and eigenfunctions for $\hat{\mathcal{W}}'$ by simply replacing α in them by 2α .

The number of relaxation modes is finite if the diffusivity is a discrete variable and the number of states is finite. This corresponds to the case of the Markovian N -state model, where the dynamics of the diffusion coefficient is described by a stochastic jump process between states. In this case, the stochastic variable $\xi(t)$ can take only N values and the probability distribution function $P(\xi)$ reduces to the probability distributions for discrete states P_n (with $n = 1, 2, \dots, N$ the index for the discrete state). In the Markovian N -state model, the equilibrium probability distribution is expressed by an N -dimensional vector as $P_{\text{eq},n}$ and the transition matrix is an $N \times N$ matrix $L_{n,n'}$. Thus the transfer operator is also an $N \times N$ matrix and the number of eigenvalues is N . Then there are N relaxation modes (some modes may be degenerated and intensities of some modes may be zero) and the relaxation

function $\Phi(t)$ becomes

$$\Phi(t) = \sum_{n=1}^N \phi_n e^{-t\lambda_n}. \quad (26)$$

In the simplest Markovian two-state model ($N = 2$), we have only two relaxation modes. We show the detailed calculations for the Markovian two-state model in the next section.

IV. DISCUSSION

A. Two-state model

As a simple yet nontrivial example, we consider a simple model where the diffusivity obeys the Markovian two-state model [7,22] [where the stochastic variable $\xi(t)$ can take only two values]. As we mentioned, there are only two relaxation modes in this case [$N = 2$ in Eq. (26)]. Here we analyze the behavior of the two-state model in detail. We describe two states in the model as the fast (f) and slow (s) states and describe the probability distributions of the fast and slow states as $P_f(t)$ and $P_s(t)$. We describe the diffusivity at the fast and slow states as D_f and D_s ($D_f \geq D_s$). Equation (4) now reduces to the simple equation

$$\frac{d}{dt} \begin{bmatrix} P_f(t) \\ P_s(t) \end{bmatrix} = \begin{bmatrix} -k_f & k_s \\ k_f & -k_s \end{bmatrix} \begin{bmatrix} P_f(t) \\ P_s(t) \end{bmatrix}, \quad (27)$$

where k_f and k_s are the transition rate from the fast to slow states and from the slow to fast states, respectively. The equilibrium distribution is simply given as

$$P_{\text{eq},f} = \frac{k_s}{k_f + k_s}, \quad P_{\text{eq},s} = \frac{k_f}{k_f + k_s}. \quad (28)$$

As we showed in Sec. III, this model has only two relaxation modes. The explicit form of the relaxation function can be analytically calculated as

$$\Phi(t) = \phi_- e^{-t\lambda_-} + \phi_+ e^{-t\lambda_+}, \quad (29)$$

where λ_\pm and ϕ_\pm are, respectively, the relaxation rates and intensities of two relaxation modes. Their explicit forms are given as follows, with the relaxation rate defined as $\mu_h \equiv \alpha D_h$ ($h = f, s$):

$$\begin{aligned} \lambda_\pm &\equiv \frac{1}{2} [\mu_f + \mu_s + k_f + k_s \\ &\quad \pm \sqrt{(\mu_f + \mu_s + k_f + k_s)^2 - 4(k_f \mu_s + k_s \mu_f + \mu_f \mu_s)}], \end{aligned} \quad (30)$$

$$\phi_\pm \equiv \frac{1}{\lambda_+ - \lambda_-} \left[\pm \frac{k_f \mu_s + k_s \mu_f}{k_s + k_f} \mp \lambda_\mp \right]. \quad (31)$$

(See Appendix A for the derivation.) From Eq. (31) we have $\phi_+ + \phi_- = 1$. [This is trivial since the relaxation function $\Phi(t)$ is normalized.]

The relaxation times of two modes are given as $\tau_\pm \equiv 1/\lambda_\pm$. From Eq. (30), the explicit expressions of $1/\lambda_\pm$ contain both the relaxation and transition rates. This means that the relaxation times do not coincide with the relaxation times naively estimated as the inverse relaxation rates $1/\mu_f$ and $1/\mu_s$. Also, from Eq. (31), the intensities for two modes are also affected

by both the relaxation and transition rates. Therefore, we conclude that the behavior of the relaxation function $\Phi(t)$ is generally not simple, even for the simple Markovian two-state model.

Although the behavior of the relaxation function $\Phi(t)$ given by Eq. (29) is generally not simple, the relaxation function reduces to simple forms for some special cases. Here we consider two limiting cases. The first case is the case where the transition between the fast and slow states is sufficiently fast. In this case we assume that $k_f, k_s \gg \mu_f, \mu_s$. The relaxation rates reduce to $\lambda_- \approx (k_f \mu_s + k_s \mu_f)/(k_f + k_s)$ and $\lambda_+ \approx k_f + k_s$ and the intensities reduce to $\phi_- \approx 1$ and $\phi_+ \approx 0$. Therefore, in this case the second mode disappears and the single-exponential-type relaxation behavior is recovered:

$$\Phi(t) \approx \exp\left(-\frac{k_f \mu_s + k_s \mu_f}{k_f + k_s} t\right). \quad (32)$$

Equation (32) means that the relaxation time is given as the harmonic average of the relaxation times of the fast and slow states. Intuitively, this result can be understood as follows: Due to the fast transition between the fast and slow states, the diffusivity $D(t)$ can be replaced by the equilibrium average $\langle D \rangle$. Then the effective relaxation rate is estimated to be

$$\alpha \langle D \rangle = \alpha(D_f P_{\text{eq},f} + D_s P_{\text{eq},s}) = \frac{k_f \mu_s + k_s \mu_f}{k_f + k_s}, \quad (33)$$

which coincides with λ_- .

The second case is the case where the transition between fast and slow states is sufficiently slow. We assume that $k_f, k_s \ll \mu_f, \mu_s$ and then the relaxation rates become $\lambda_- \approx \mu_s$ and $\lambda_+ \approx \mu_f$ and the relaxation intensities are $\phi_- \approx k_f/(k_f + k_s)$ and $\phi_+ \approx k_s/(k_f + k_s)$. Therefore, the relaxation function $\Phi(t)$ consists of two modes with the relaxation times determined solely by the relaxation rates μ_f and μ_s :

$$\Phi(t) \approx \frac{k_f}{k_f + k_s} e^{-\mu_s t} + \frac{k_s}{k_f + k_s} e^{-\mu_f t}. \quad (34)$$

In this case we have two relaxation modes and thus non-single-exponential-type behavior is observed. The relaxation times coincide with the relaxation times of the pure fast and slow states and the intensities correspond to the equilibrium fractions $\phi_- \approx P_{\text{eq},s}$ and $\phi_+ \approx P_{\text{eq},f}$ [Eq. (28)]. Intuitively, this case corresponds to the mixture of two statistically independent relaxation processes. Because the transition rates are small, the system can fully relax before the transition occurs. Thus the relaxation times are not affected by the transition dynamics and the intensities are just given as the equilibrium probabilities.

We show the relaxation function $\Phi(t)$ for various transition and relaxation rates in Fig. 2. For simplicity, here we limit ourselves to the case where two transition rates are the same: $k_f = k_s$. In this case, we have essentially two freely tunable parameters $\kappa \equiv k_f/\mu_f = k_s/\mu_f$ and μ_s/μ_f . Figure 2(a) shows the relaxation function for $\mu_s/\mu_f = 10^{-2}$ and various values of κ . The asymptotic forms for the fast and slow transition limits [Eqs. (32) and (34)] are also shown for comparison. We can observe that even if the value of μ_s/μ_f is constant, the relaxation function largely changes if we change κ . For small- and large- κ cases, we observe that the asymptotic forms work

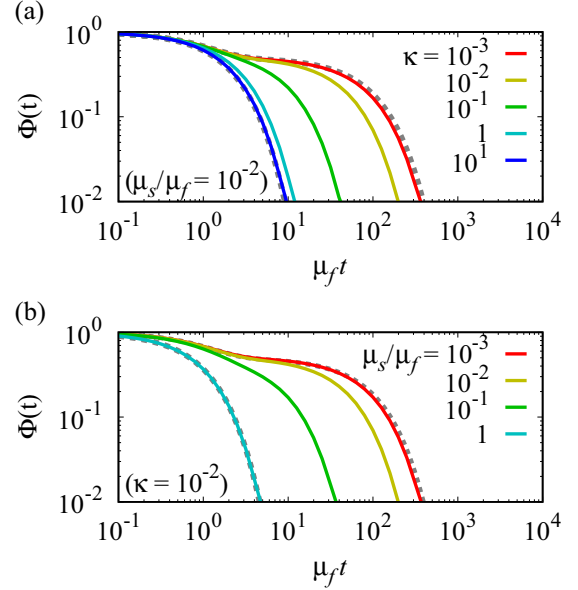


FIG. 2. Relaxation function $\Phi(t)$ for the OUF by the Markovian two-state model. (a) The relaxation rate is $\mu_s/\mu_f = 10^{-2}$ and the transition rate varies. The solid curves represent data for $\kappa = k_f/\mu_f = k_s/\mu_f = 10^{-2}, 10^{-1}, 1$, and 10^1 . The dotted gray curves represent asymptotic forms [Eqs. (32) and (34)]. (b) The transition rate is constant $\kappa = 10^{-2}$ and the relaxation rate varies. The solid curves represent data for $\mu_s/\mu_f = 1, 10^{-1}, 10^{-2}$, and 10^{-3} . The dotted gray curves represent the asymptotic forms.

as good approximations. Figure 2(b) shows the relaxation function for $\kappa = 10^{-2}$ and various values of μ_s/μ_f . For the case of $\mu_s/\mu_f = 1$, the relaxation function trivially reduces to a single-exponential form $\Phi(t) = \exp(-\mu_f t)$. For the case of small μ_s , we consider the condition $\mu_s/\mu_f \ll \kappa \ll 1$ and have $\lambda_+ \approx \mu_f$, $\lambda_- \approx \mu_f \kappa$, and $\phi_+ \approx \phi_- \approx 1/2$. We observe that the data for small and large μ_s/μ_f agree well with the asymptotic forms.

Except for the special cases examined above, in general, the two relaxation times cannot be simply related to the relaxation rates of the fast and slow states. In addition, we cannot determine the relaxation and transition rates solely from the relaxation function $\Phi(t)$, even if $\Phi(t)$ can be approximately expressed as the sum of two relaxation modes. To investigate whether the relaxation behavior is really affected by the FD or not, we can utilize another relaxation function $\Psi(t)$. The relaxation function $\Psi(t)$ can be obtained by replacing α in $\Phi(t)$ by 2α , as we mentioned. Thus, for the current case we have

$$\Psi(t) = \phi'_- e^{-t\lambda'_-} + \phi'_+ e^{-t\lambda'_+}, \quad (35)$$

with λ'_\pm and ϕ'_\pm defined as

$$\lambda'_\pm \equiv \frac{1}{2} [2\mu_f + 2\mu_s + k_f + k_s \pm \sqrt{(2\mu_f + 2\mu_s + k_f + k_s)^2 - 8(k_f \mu_s + k_s \mu_f + 2\mu_f \mu_s)}], \quad (36)$$

$$\phi'_\pm \equiv \frac{1}{\lambda'_+ - \lambda'_-} \left[\pm \frac{2(k_f \mu_s + k_s \mu_f)}{k_s + k_f} \mp \lambda'_\mp \right]. \quad (37)$$

We consider two limiting cases again. For the case where $k_f, k_s \gg \mu_f, \mu_s$, Eq. (35) reduces to a simple exponential form as

$$\Psi(t) \approx e^{-2\alpha(D)t} \approx \Phi(2t) \approx \Phi^2(t). \quad (38)$$

Thus we find that in this case the relaxation behavior is the same as the usual OU process with a constant diffusivity. On the other hand, for $k_f, k_s \ll \mu_f, \mu_s$, the relaxation function $\Psi(t)$ simply reduces as

$$\Psi(t) \approx P_{\text{eq},s} e^{-2\mu_s t} + P_{\text{eq},s} e^{-2\mu_f t} \approx \Phi(2t) \neq \Phi^2(t). \quad (39)$$

This means that the relation between $\Phi(t)$ and $\Psi(t)$ becomes different from one for the constant diffusivity case. In general, the relation between $\Phi(t)$ and $\Psi(t)$ is not simple. We conclude that by combining the two relaxation functions $\Phi(t)$ and $\Psi(t)$, we are able to extract some information on the FD such as the transition rates between states. For example, if we have four relaxation times $1/\lambda_{\pm}$ and $1/\lambda'_{\pm}$ from relaxation functions, we can determine relaxation and transition rates μ_f, μ_s, k_f , and k_s .

This analysis method will be useful to analyze relaxation functions obtained by experiments. The dielectric relaxation functions obtained by the dielectric measurements can be related to the relaxation function $\Phi(t)$. The relaxation moduli obtained by rheological measurements can be related to the relaxation function $\Psi(t)$. Matsumiya *et al.* [25] reported both the rheological and dielectric relaxation data for glassy polystyrene samples with various molecular weights. They showed that the storage modulus and the dielectric relaxation function of the same sample have almost the same form, but the relaxation times are different. Although their data cannot be simply expressed as two relaxation modes, analyses based on our results would be informative to understanding the nature of glassy dynamics in polymers.

B. Ornstein-Uhlenbeck-type model

We consider another simple case, where the dynamics of the diffusivity obeys an OU-type process. While the diffusivity is a discrete variable in the two-state model, the diffusivity is a continuum variable in this model. Thus the relaxation function consists of infinite relaxation modes (at least formally).

Since the diffusivity should be positive, we introduce the noise coefficient whose square gives the diffusion coefficient

$$D(t) = \bar{D}b^2(t), \quad (40)$$

where $b(t)$ is the noise coefficient, which can be both positive and negative, and \bar{D} is constant. We interpret $b(t)$ as the stochastic variable $\xi(t)$ in Eqs. (4), (21), and (24). For the dynamics of $b(t)$, we employ the following Langevin equation:

$$\frac{db(t)}{dt} = -kb(t) + \sqrt{2k}w'(t). \quad (41)$$

Here k is the rate constant and $w'(t)$ is the Gaussian white noise whose first and second moments are given as $\langle w'(t) \rangle = 0$ and $\langle w'(t)w'(t') \rangle = \delta(t-t')$. Equation (41) is an OU process. A similar model for the noise coefficient was employed to model the diffusion behavior in a heterogeneous medium

(the diffusing diffusivity model) [9,10,48]. In the diffusing diffusivity model, the diffusion coefficient is expressed as the square of a vector variable which obeys an OU process. Our model can be interpreted as a one-dimensional version of the diffusing diffusivity model.

Because the stochastic process for the diffusion coefficient is fully specified, now we can calculate the explicit form of the relaxation function $\Phi(t)$. Unlike the case of the two-state model, the OU-type model has infinite relaxation modes. The explicit expression for the relaxation function is

$$\Phi(t) = \sum_{n=0}^{\infty} \phi_n \exp\{-k[(2n+1)\gamma - 1/2]t\}, \quad (42)$$

where we have defined $\gamma \equiv \sqrt{\mu/k + 1/4}$, with $\mu \equiv \alpha\bar{D}$, and

$$\phi_n \equiv \begin{cases} \frac{\sqrt{2n!}}{2^n[(n/2)!]^2} \frac{\gamma^{1/2}(\gamma-1/2)^n}{(\gamma+1/2)^{n+1}} & \text{for } n \text{ even} \\ 0 & \text{for } n \text{ odd.} \end{cases} \quad (43)$$

(The detailed calculations are shown in Appendix B.) From Eq. (43), only the modes with even n survive. We set $n = 2m$ and rewrite Eq. (42) as

$$\Phi(t) = \sum_{m=0}^{\infty} \frac{\sqrt{2}(2m)! \gamma^{1/2}(\gamma-1/2)^{2m}}{2^{2m}(m!)^2 (\gamma+1/2)^{2m+1}} \times \exp\{-k[(4m+1)\gamma - 1/2]t\}. \quad (44)$$

The longest relaxation time τ_0 is the inverse of the relaxation rate for $n = 0$ by Eq. (42):

$$\tau_0 \equiv \frac{1}{\lambda_0} = \frac{1}{k(\sqrt{\mu/k + 1/4} - 1/2)}. \quad (45)$$

If the transition dynamics is much faster or slower than the relaxation dynamics, we have simple approximate forms for the longest relaxation time:

$$\tau_0 \approx \begin{cases} 1/\mu, & k \gg \mu \\ 1/\sqrt{k\mu}, & k \ll \mu. \end{cases} \quad (46)$$

Equation (46) means that the relaxation behavior of this model is largely affected by the transition rate if the transition dynamics is slow. We consider two limiting cases in detail, as the case of the Markovian two-state model. First, we assume that the transition dynamics is much faster than the relaxation dynamics and that $k \gg \mu$. In this case, we have $\gamma \approx 1/2 + \mu/k$ and the intensity becomes

$$\phi_n \approx \begin{cases} 1 & \text{for } n = 0 \\ 0 & \text{otherwise.} \end{cases} \quad (47)$$

Therefore, the relaxation function reduces to the single-exponential form

$$\Phi(t) \approx \exp(-\mu t). \quad (48)$$

From Eq. (48), the relaxation time $1/\mu$ is independent of the transition rate k . This is the same as the case of the Markovian two-state model. As before, this result can be intuitively understood by considering the equilibrium average of the relaxation rate

$$\alpha \langle D \rangle = \alpha \int_{-\infty}^{\infty} db \bar{D}b^2 P_{\text{eq}}(b) = \mu, \quad (49)$$

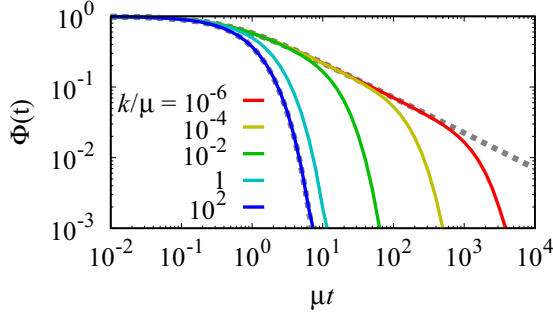


FIG. 3. Relaxation function $\Phi(t)$ for the OUFD. The noise coefficient obeys the OU model. The solid curves represent the relaxation functions for $k/\mu = 10^{-6}$, 10^{-4} , 10^{-2} , 1, and 10^2 , calculated by Eq. (44). The dotted gray curves represent the asymptotic forms for $k/\mu \gg 1$ [Eq. (48), left] and $k/\mu \ll 1$ [Eq. (50), right].

where $P_{\text{eq}}(b) = e^{-b^2/2}/\sqrt{2\pi}$ is the equilibrium distribution for the noise coefficient.

Second, we assume that the transition dynamics is much slower than the relaxation dynamics. In this case we have $k \ll \mu$ and $\gamma \gg 1$, but it is rather difficult to calculate the approximate form for the relaxation function from Eq. (44) under these conditions. Fortunately, we can calculate the approximate form starting from the dynamic equation. The result is

$$\Phi(t) \approx \frac{1}{\sqrt{1 + 2\mu t}}. \quad (50)$$

(See Appendix C for detailed calculations.)

Thus, in this case, we observe the power-law type behavior at the long time region, $\Phi(t) \propto t^{-1/2}$ ($t \gtrsim 1/2\mu$). Such power-law type relaxation behavior is also observed for polymers (the Rouse model) [19] and critical gels [49]. In most cases, the power-law-type relaxation is interpreted as the relaxation of fractal structures where the relaxation time distribution is given as a power-law-type distribution. Our result gives another interpretation: The power-law-type relaxation can also be attributed to the FD. As in the case of the two-state model, intuitively, the relaxation function is expressed as the sum of relaxation modes and their intensities are given as the equilibrium probability distribution. In the same way, Eq. (50) can be reproduced as the average of the relaxation function with respect to the equilibrium distribution of the noise coefficient:

$$\Phi(t) \approx \int_{-\infty}^{\infty} db e^{-\alpha \bar{D} b^2 t} P_{\text{eq}}(b) = \frac{1}{\sqrt{1 + 2\mu t}}. \quad (51)$$

The same expression as Eq. (51) can be obtained by substituting the approximate transfer operator [Eq. (C1)] directly into Eq. (21).

We show the relaxation function for various values of k/μ , directly calculated by Eq. (44), in Fig. 3. For comparison, the asymptotic forms for $k/\mu \gg 1$ and $k/\mu \ll 1$ [Eqs. (48) and (50)] are also shown in Fig. 3. We observe that for sufficiently large k/μ , the relaxation function $\Phi(t)$ is well approximated by the asymptotic form. On the other hand, for small k/μ such as $k/\mu = 10^{-6}$, we observe the deviation from the asymptotic form. This is because the longest relaxation

time is finite for finite k/μ , as shown in Eqs. (45) and (46). In the relatively short time region ($t \lesssim 1/\sqrt{k\mu}$), the asymptotic form works well.

It is straightforward to show that the relation between the two relaxation functions $\Phi(t)$ and $\Psi(t)$ becomes almost the same as the case of the Markovian two-state model [Eqs. (38) and (39)]. This implies that the OUFD satisfies the relations $\Psi(t) \approx \Phi^2(t) \approx \Phi(2t)$ and $\Psi(t) \approx \Phi(2t)$ for sufficiently fast and slow transition rates, respectively. These approximate relations are expected to be independent of the details of the dynamics for the diffusivity.

C. Comparison with other models

It would be informative to compare the results of the OUFD with other models. Here we consider two models. One is the generalized Langevin equation (GLE) model, which is obtained by the projection operator method [23] and widely utilized to describe the dynamics of coarse-grained variables. The other is the multimode OU process in which multiple OU processes are linearly combined. Currently, these conventional models are utilized as standard models to analyze experimental data. The GLE is widely utilized to analyze diffusion behavior of particles in viscoelastic environments [50,51]. The multimode OU process is also widely utilized to analyze and model non-single-exponential relaxation functions [52]. However, from the viewpoint of the FD, analyses based on these conventional models may not be physically reasonable for some systems. Thus it would be informative to compare the properties of relaxation functions in conventional models with those in the OUFD.

First, we consider the GLE with the memory function (the generalized OU process). In the generalized OU process, the dynamic equation for $\mathbf{r}(t)$ is described as a linear GLE. For simplicity, we assume that the dynamics is isotropic and the memory kernel can be expressed as a scalar quantity. We can describe the dynamic equation as

$$\frac{d\mathbf{r}(t)}{dt} = -\alpha \bar{D} \int_{-\infty}^t dt' K(t-t') \mathbf{r}(t') + \sqrt{\bar{D}} \boldsymbol{\eta}(t), \quad (52)$$

where \bar{D} is constant (the reference diffusion coefficient), $K(t)$ is the memory kernel, and $\boldsymbol{\eta}(t)$ is the Gaussian colored noise. The fluctuation-dissipation relation of the second kind requires the noise to satisfy the following relations: $\langle \boldsymbol{\eta}(t) \rangle = 0$ and $\langle \boldsymbol{\eta}(t) \boldsymbol{\eta}(t') \rangle = K(|t-t'|) \mathbf{1}$. The memory kernel can be simply related to the two-point-correlation function [which is proportional to the relaxation function $\Phi(t)$]. Fox [53] showed that the memory kernel satisfies the relation

$$\tilde{\Phi}(u) = \frac{1}{u + \alpha \bar{D} \tilde{K}(u)}, \quad (53)$$

where $\tilde{\Phi}(u)$ and $\tilde{K}(u)$ are the Laplace transforms of the relaxation function $\Phi(t)$ and the memory kernel $K(t)$: $\tilde{\Phi}(u) \equiv \int_0^{\infty} dt e^{-ut} \Phi(t)$ and $\tilde{K}(u) \equiv \int_0^{\infty} dt e^{-ut} K(t)$. From Eq. (53) we can tune the memory kernel and reproduce the relaxation function $\Phi(t)$ by the OUFD [Eq. (24)]. For example, in the case of the Markovian two-state model, the Laplace transforms of the relaxation function are calculated as follows,

from Eq. (29):

$$\tilde{\Phi}(u) = \frac{\phi_-}{\lambda_- + u} + \frac{\phi_+}{\lambda_+ + u}. \quad (54)$$

From Eqs. (53) and (54) we find that the following Laplace-transformed memory kernel reproduces the same relaxation function as the Markovian two-state model:

$$\tilde{K}(u) = \frac{1}{\alpha D} \left[(\phi_- \lambda_- + \phi_+ \lambda_+) - \frac{\phi_- \phi_+ (\lambda_+ - \lambda_-)^2}{u + (\phi_- \lambda_+ + \phi_+ \lambda_-)} \right]. \quad (55)$$

By performing the inverse Laplace transform, we have the following simple expression in the time domain:

$$K(t) = \frac{1}{\alpha D} [(\phi_- \lambda_- + \phi_+ \lambda_+) \delta(t) - \phi_- \phi_+ (\lambda_+ - \lambda_-)^2 \times e^{-(\phi_- \lambda_+ + \phi_+ \lambda_-)t}]. \quad (56)$$

However, the thus obtained GLE [Eq. (52) with (56)] cannot reproduce the relaxation function $\Psi(t)$ by the OUFDF. The linear GLE (52) gives $\mathbf{r}(t)$ as a Gaussian process. By utilizing Wick's theorem, the multipoint-correlation functions of $\mathbf{r}(t)$ can be decomposed into two-point-correlation functions. For the case of the four-point correlation, which appears in the relaxation function $\Psi(t)$, we have

$$\begin{aligned} & \langle [r_x(t)r_y(t)][r_x(0)r_y(0)] \rangle \\ &= \langle r_x(t)r_y(t) \rangle \langle r_x(0)r_y(0) \rangle + \langle r_x(t)r_x(0) \rangle \langle r_y(t)r_y(0) \rangle \\ &+ \langle r_x(t)r_y(0) \rangle \langle r_y(t)r_x(0) \rangle \\ &= \langle r_x(t)r_x(0) \rangle \langle r_y(t)r_y(0) \rangle. \end{aligned} \quad (57)$$

In the last line of Eq. (57), we have utilized the fact that the system is isotropic and there is no correlation between $r_x(t)$ and $r_y(t)$. From Eq. (57), the relaxation function $\Psi(t)$ is simply given as

$$\Psi(t) = \Phi^2(t). \quad (58)$$

Equation (58) means that the relaxation functions $\Phi(t)$ and $\Psi(t)$ contain essentially the same information. Here it should be stressed that Eq. (58) holds for any kernel functions. In the case of the Markovian N -state model, Eq. (58) generally consists of $N(N+1)/2$ relaxation modes. This is clearly different from the case of the OUFDF, where we have only N relaxation modes for $\Psi(t)$. Therefore, we conclude that the OUFDF cannot be expressed as the GLE. The effects of an FD seem to be clearly observed when we analyze higher-order correlation functions. This is consistent with the fact that the Langevin equation with an FD in the absence of the potential exhibits only the normal diffusion behavior on average and the effects of an FD are observed in the higher-order fluctuations [7].

Next we consider the multimode OU process. We express the position $\mathbf{r}(t)$ as the sum of N modes. If we express the k th mode as $\mathbf{r}_k(t)$, \mathbf{r} is expressed as the (weighted) sum of \mathbf{r}_k ,

$$\mathbf{r}(t) = \sum_{k=1}^N c_k \mathbf{r}_k(t), \quad (59)$$

where c_k represents the weight factor for the k th mode and c_k is normalized to satisfy $\sum_{k=1}^N c_k^2 = 1$. We assume that modes

are statistically independent and each mode obeys an OU process,

$$\frac{d\mathbf{r}_k(t)}{dt} = -\alpha D_k \mathbf{r}_k(t) + \sqrt{2D_k} \mathbf{w}_k(t), \quad (60)$$

where D_k is the diffusion coefficient of the k th mode (D_k is assumed to be constant) and $\mathbf{w}_k(t)$ is the Gaussian white noise. In addition, $\mathbf{w}_k(t)$ satisfies the following relations: $\langle \mathbf{w}_k(t) \rangle = 0$ and $\langle \mathbf{w}_k(t) \mathbf{w}_l(t') \rangle = \delta_{kl} \mathbf{1} \delta(t-t')$.

Equation (60) is just a usual OU process and thus the relaxation function $\Phi(t)$ can be calculated straightforwardly. The two-time-correlation function is calculated to be $\langle \mathbf{r}_k(t) \cdot \mathbf{r}_l(0) \rangle = \delta_{kl} \langle \mathbf{r}_k^2(0) \rangle e^{-\alpha D_k t}$ and thus the relaxation function $\Phi(t)$ becomes

$$\Phi(t) = \sum_{k=1}^N c_k^2 e^{-\alpha D_k t}. \quad (61)$$

To reproduce the correlation function $\Phi(t)$ in the Markovian two-state model, we simply set $N=2$ and then we have $\alpha D_1 = \lambda_-$, $\alpha D_2 = \lambda_+$, $c_1 = \phi_-^{1/2}$, and $c_2 = \phi_+^{1/2}$.

As in the case of the GLE, even if we employ the thus determined parameters, the multimode OU process cannot reproduce the correlation function $\Psi(t)$ correctly. Due to the Gaussian nature, the relaxation function $\Psi(t)$ in the multimode OU process can be calculated in a similar way to the case of the GLE. The result is the same as Eq. (58). Therefore, the situation is the same as the case of the generalized OU model with the memory kernel. (Actually, the multimode OU is a Gaussian process and it can also be expressed as the linear GLE.) The relaxation function $\Psi(t)$ has $N(N+1)/2$ relaxation modes in the multimode OU model, whereas there are N modes in the OUFDF. We conclude that the OUFDF cannot be expressed as the multimode OU process.

From the discussion above, we conclude that the OU process with an FD belongs to a different class of dynamic equations compared with widely utilized stochastic dynamic equation models such as the GLE. (This conclusion is rather trivial, since two independent stochastic processes are multiplicatively coupled in the OUFDF, whereas the couplings of stochastic processes in the GLE and the multimode OU process are additive.) The importance of an FD is especially observed via higher-order correlations. In analogy to the non-Gaussianity parameter for diffusion processes [54], we can introduce a simple yet useful quantity which distinguishes the OUFDF from the linear GLE and the multimode OU process:

$$A(t) = \frac{\Psi(t)}{\Phi^2(t)} - 1. \quad (62)$$

This quantity becomes zero if a relaxation process can be described by the linear GLE or the multimode OU process. Conversely, if it is nonzero, that process cannot be described by popular conventional models, whereas the OUFDF can successfully describe it. Although it would not be easy to experimentally observe two relaxation functions $\Phi(t)$ and $\Psi(t)$ for the same system, the combination of two relaxation functions [such as Eq. (62)] enables us to investigate heterogeneous dynamics of the systems. We consider that an FD will be especially useful to model and/or analyze dynamics

and relaxation behavior in heterogeneous environments such as supercooled liquids [20–22]. It will be also informative to apply the concept of the FD to analyze the single-molecule dynamics of proteins [16,17].

V. CONCLUSION

In this work we studied the relaxation behavior of the OUFD. We modeled the stochastic process with a linear restoring force and a thermal noise, both coupled to a time-dependent and fluctuating diffusivity. We showed that the relaxation functions $\Phi(t)$ and $\Psi(t)$ are expressed in terms of the integral of the diffusion coefficient over time [Eqs. (9) and (12)]. To calculate the explicit forms of a relaxation function, we utilized the functional integral expression with the action functional and the transfer matrix method. We derived the simple expression for the relaxation function as the sum of relaxation modes [Eq. (24)]. The relaxation rate and the intensity of each mode are calculated from the eigenvalue and eigenfunction of the transfer matrix.

As analytically solvable models we studied the Markovian two-state model and the OU-type model for the noise coefficient. The two-state model has only two relaxation modes, but the relaxation modes and relaxation intensities depend on both the relaxation rates and the transition rates. This is because the relaxation behavior of the OUFD is determined as a result of the competition between the relaxation dynamics and the transition dynamics. If the transition rates are sufficiently larger or smaller than the relaxation rates, the corresponding relaxation function reduces to a simple asymptotic form. The situation is similar to the case when the dynamics for the noise coefficient is described by the OU process. In this model, the noise coefficient is a continuum stochastic variable and we have infinite relaxation modes. We showed that if the transition rate is sufficiently smaller than the relaxation rate, the relaxation function exhibits a power-law-type behavior. Because the relation between two relaxation functions $\Phi(t)$ and $\Psi(t)$ is not simple as in the cases of the GLE and the multimode OU process, we conclude that the OUFD is qualitatively different from those conventional models. Thus, it is important and possible to unravel the underlying dynamics by analyzing the two relaxation functions.

We believe that our model and analysis would be useful to analyze some experimental data for supercooled liquids, polymers, and proteins. Work on the formulation of the relaxation function from the viewpoint of the renewal theory and the extension of this work are left for future study.

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APPENDIX A: RELAXATION MODES FOR THE TWO-STATE MODEL

In this Appendix we show the calculations for the two-state model in Sec. IV A. From Eq. (27), the transfer operator can be expressed in a 2×2 matrix form

$$\hat{\mathcal{W}} = \begin{bmatrix} \mu_f + k_f & -k_s \\ -k_f & \mu_s + k_s \end{bmatrix}, \quad (\text{A1})$$

where we have expressed the relaxation rate at each state as $\mu_h = \alpha D_h$ ($h = f, s$). The eigenvalues of the matrix in Eq. (A1) are obtained as

$$\lambda_{\pm} = \frac{1}{2} [\mu_f + \mu_s + k_f + k_s \pm \sqrt{(\mu_f + \mu_s + k_f + k_s)^2 - 4(k_f \mu_s + k_s \mu_f + \mu_f \mu_s)}]. \quad (\text{A2})$$

The first and second eigenvalues correspond to λ_- and λ_+ , respectively. We describe the second and first eigenvectors as $\psi_{\pm} = [\psi_{\pm,f}, \psi_{\pm,s}]^T$. The eigenvectors are calculated to be

$$\psi_{\pm} = \begin{bmatrix} \lambda_{\pm} - \mu_s \\ \mu_f - \lambda_{\pm} \end{bmatrix}. \quad (\text{A3})$$

From Eq. (A3) the relaxation function can be expressed as follows, with the thus obtained eigenvalues and eigenvectors [Eqs. (A2) and (A3)] and the equilibrium distribution [Eq. (28)]:

$$\Phi(t) = \begin{bmatrix} 1 & 1 \end{bmatrix} \begin{bmatrix} \psi_- & \psi_+ \end{bmatrix} \begin{bmatrix} e^{-t\lambda_-} & 0 \\ 0 & e^{-t\lambda_+} \end{bmatrix} \times \begin{bmatrix} \psi_- & \psi_+^{-1} \end{bmatrix} \begin{bmatrix} P_{\text{eq},f} \\ P_{\text{eq},s} \end{bmatrix}. \quad (\text{A4})$$

Equation (A4) can be rewritten in a simple form as $\Phi(t) = \phi_- e^{-\lambda_- t} + \phi_+ e^{-\lambda_+ t}$ if we introduce the relaxation intensities defined as

$$\phi_- = \frac{1}{\lambda_+ - \lambda_-} \left[\lambda_+ - \frac{k_f \mu_s + k_s \mu_f}{k_s + k_f} \right], \quad (\text{A5})$$

$$\phi_+ = \frac{1}{\lambda_+ - \lambda_-} \left[\frac{k_f \mu_s + k_s \mu_f}{k_s + k_f} - \lambda_- \right]. \quad (\text{A6})$$

Thus we have Eqs. (30) and (31) in the main text.

APPENDIX B: RELAXATION MODES FOR THE ORNSTEIN-UHLENBECK-TYPE MODEL

In this Appendix we show the calculations for the OU-type model in Sec. IV B. First we convert Eq. (41) into the Fokker-Planck equation. Following a standard procedure [26], we have the Fokker-Planck equation for the distribution function of b , $P(b, t)$,

$$\frac{\partial P(b, t)}{\partial t} = \hat{\mathcal{L}}P(b, t), \quad (\text{B1})$$

with the Fokker-Planck operator defined as

$$\hat{\mathcal{L}}P = k \frac{\partial}{\partial b} \left[bP + \frac{\partial P}{\partial b} \right]. \quad (\text{B2})$$

Obviously, the equilibrium distribution $P_{\text{eq}}(b)$ is a Gaussian: $P_{\text{eq}}(b) = e^{-b^2/2}/\sqrt{2\pi}$.

From Eqs. (20) and (B2), the transfer operator can be explicitly expressed as

$$-\hat{\mathcal{W}}\psi = -\mu b^2\psi + k \frac{\partial}{\partial b} \left[b\psi + \frac{\partial \psi}{\partial b} \right], \quad (\text{B3})$$

where $\mu \equiv \alpha\bar{D}$. The eigenvalue λ and the eigenfunction $\psi(b)$ satisfy the eigenvalue equation

$$\hat{\mathcal{W}}\psi(b) = \lambda\psi(b). \quad (\text{B4})$$

Here we introduce the variable transform to make the transfer operator self-adjoint [47]:

$$\tilde{\psi}(b) \equiv e^{b^2/4}\psi(b) = e^{-b^2/4}\psi^\dagger(b). \quad (\text{B5})$$

Then we have the eigenvalue equation

$$\gamma^2 b^2 \tilde{\psi} - \frac{d^2 \tilde{\psi}}{db^2} = \left(\frac{\lambda}{k} + \frac{1}{2} \right) \tilde{\psi}, \quad (\text{B6})$$

where $\gamma = \sqrt{\mu/k + 1/4}$. Roughly speaking, the parameter γ represents the competition between the transition and relaxation. If the transition becomes faster than the relaxation, γ decreases. If the relaxation becomes faster than the transition, γ increases. It should be noted that γ satisfies $\gamma > 1/2$. This eigenvalue equation has the same form as the Schrödinger equation for a one-dimensional harmonic potential [55] and thus we can calculate eigenfunctions and eigenvalues straightforwardly. The n th eigenvalue and eigenfunction ($n = 0, 1, 2, \dots$) are given as

$$\lambda_n = k[(2n+1)\gamma - 1/2], \quad (\text{B7})$$

$$\tilde{\psi}_n(b) = \left(\frac{\gamma^{1/2}}{\pi^{1/2} 2^n n!} \right)^{1/2} H_n(\sqrt{\gamma}b) e^{-\gamma b^2/2}, \quad (\text{B8})$$

where $H_n(x)$ is the n th-order Hermite polynomial [55,56].

The relaxation function can be expressed with the eigenvalues and eigenfunctions. From Eqs. (B5), (B7), and (B8) we have

$$\Phi(t) = \sum_{n=0}^{\infty} \phi_n \exp\{-k[(2n+1)\gamma - 1/2]t\}, \quad (\text{B9})$$

with

$$\phi_n = \frac{\gamma^{1/2}}{\pi 2^{n+1/2} n!} \left[\int_{-\infty}^{\infty} db H_n(\sqrt{\gamma}b) e^{-\gamma b^2/2 - b^2/4} \right]^2. \quad (\text{B10})$$

The integral in Eq. (B10) can be calculated analytically. We rewrite Eq. (B10) as

$$\phi_n = \frac{\gamma^{-1/2}}{\pi 2^{n+1/2} n!} I_n^2, \quad (\text{B11})$$

where I_n is the following integral:

$$I_n \equiv \int_{-\infty}^{\infty} ds H_n(s) e^{-s^2/2 - s^2/4\gamma}. \quad (\text{B12})$$

For $n = 0$, the integral I_0 can be easily calculated, because $H_0(s) = 1$. We have

$$I_0 = \int_{-\infty}^{\infty} ds e^{-s^2/2 - s^2/4\gamma} = \sqrt{\frac{2\pi\gamma}{\gamma + 1/2}}. \quad (\text{B13})$$

Also, the integral I_n can be calculated easily for odd n . In this case, from the symmetry of the Hermite polynomial $H_n(s) = -H_n(-s)$, the integrand in Eq. (B12) is an odd function of s . Then we simply have

$$I_n = 0 \quad \text{for } n \text{ odd} \quad (\text{B14})$$

and thus the odd n modes vanish:

$$\phi_n = 0 \quad \text{for } n \text{ odd}. \quad (\text{B15})$$

Thus now we need to calculate the integral I_n for even $n \geq 2$. By utilizing the recurrence relation for the Hermite polynomial [55,56]

$$H_{n+1}(s) = 2sH_n(s) - 2nH_{n-1}(s), \quad (\text{B16})$$

we have

$$\begin{aligned} I_n &= \int_{-\infty}^{\infty} ds [2sH_{n-1}(s) - 2(n-1)H_{n-2}(s)] e^{-s^2/2 - s^2/4\gamma} \\ &= 2 \int_{-\infty}^{\infty} ds s H_{n-1}(s) e^{-s^2/2 - s^2/4\gamma} - 2(n-1)I_{n-2} \\ &= \frac{2}{1 + 1/2\gamma} \int_{-\infty}^{\infty} ds \frac{dH_{n-1}(s)}{ds} e^{-s^2/2 - s^2/4\gamma} - 2(n-1)I_{n-2}, \end{aligned} \quad (\text{B17})$$

where in the last line we have utilized the partial integral. We utilize another recurrence relation for the Hermite polynomial [55,56]:

$$\frac{dH_n(s)}{ds} = 2nH_{n-1}(s). \quad (\text{B18})$$

Finally, we have the recursive relation for the integral I_n ,

$$\begin{aligned} I_n &= \frac{4(n-1)}{1 + 1/2\gamma} I_{n-2} - 2(n-1)I_{n-2} \\ &= 2(n-1) \frac{\gamma - 1/2}{\gamma + 1/2} I_{n-2}, \end{aligned} \quad (\text{B19})$$

and from Eqs. (B13) and (B19) the solution is

$$\begin{aligned} I_n &= 2^{n/2} (n-1)!! \left(\frac{\gamma - 1/2}{\gamma + 1/2} \right)^{n/2} I_0 \\ &= \sqrt{2\pi} 2^{n/2} (n-1)!! \frac{\gamma^{1/2} (\gamma - 1/2)^{n/2}}{(\gamma + 1/2)^{n/2 + 1/2}}. \end{aligned} \quad (\text{B20})$$

Here $n!!$ represents the double factorial of n [57].

By substituting Eq. (B20) into (B11) and utilizing some relations for the double factorial and the factorial [57], the intensity of the n th mode is explicitly expressed as

$$\phi_n = \frac{\gamma^{-1/2}}{\pi 2^{n+1/2} n!} I_n^2 = \frac{\sqrt{2n!}}{2^n [(n/2)!]^2} \frac{\gamma^{1/2} (\gamma - 1/2)^n}{(\gamma + 1/2)^{n+1}}. \quad (\text{B21})$$

Equations (B9), (B15), and (B21) give Eqs. (42) and (43) in the main text.

APPENDIX C: SLOW TRANSITION LIMIT OF THE ORNSTEIN-UHLENBECK-TYPE MODEL

In this Appendix we show the detailed calculation of the relaxation function of the OU-type model at the slow transition limit where $k \ll \mu$. As we mentioned in the main text, it is difficult to obtain an approximate form for (42). Instead, here we approximate the transfer operator and solve the eigenvalue equation with the approximate transfer operator. For $k \ll \mu$, the transfer operator [Eq. (B3)] can be approximated as

$$-\hat{\mathcal{W}}\psi \approx -\mu b^2 \psi. \quad (\text{C1})$$

From Eq. (C1), the eigenvalue equation [Eq. (B6)] can be simply approximated as

$$\lambda \psi(b) \approx \mu b^2 \psi(b). \quad (\text{C2})$$

Equation (C2) is not a differential equation, unlike Eq. (B6). Formally, we have the eigenvalue and the eigenfunction for Eq. (C2),

$$\psi(b, a) = \psi^\dagger(b, a) = \delta(b - a), \quad \lambda(a) = \mu a^2, \quad (\text{C3})$$

where a is the index of the eigenvalue and eigenfunction and b is a continuum variable. [The eigenvalue is not in ascending order in a , but here we do not need the eigenvalues to be ordered, thus we simply use Eq. (C3).] The sum over all the eigenmodes should be replaced by the integral over the index a . Therefore, we have the following approximate expression for the relaxation function $\Phi(t)$:

$$\begin{aligned} \Phi(t) &\approx \int_{-\infty}^{\infty} da \left[\int_{-\infty}^{\infty} db \delta(b - a) \right. \\ &\quad \left. \times \int_{-\infty}^{\infty} db' \delta(b' - a) \frac{1}{\sqrt{2\pi}} e^{-b'^2/2} \right] \\ e^{-\mu a^2 t} &\approx \frac{1}{\sqrt{1 + 2\mu t}}. \end{aligned} \quad (\text{C4})$$

Thus we have Eq. (43) in the main text.

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