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State-Dependent Diffusion: Thermodynamic Consistency and its Path Integral Formulation

Andy W.C. Lau
Florida Atlantic University

Tom C. Lubensky
University of Pennsylvania, tom@physics.upenn.edu

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State-Dependent Diffusion: Thermodynamic Consistency and its Path Integral Formulation

Abstract

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Disciplines

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State-dependent diffusion: Thermodynamic consistency and its path integral formulation

A. W. C. Lau

Department of Physics, Florida Atlantic University, Boca Raton, Florida 33431, USA

T. C. Lubensky

Department of Physics and Astronomy, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA

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The friction coefficient of a particle can depend on its position, as it does when the particle is near a wall. We formulate the dynamics of particles with such state-dependent friction coefficients in terms of a general Langevin equation with multiplicative noise, whose evaluation requires the introduction of specific rules. Two common conventions, the Ito and the Stratonovich, provide alternative rules for evaluation of the noise, but other conventions are possible. We show that the requirement that a particle's distribution function approach the Boltzmann distribution at long times dictates that a drift term must be added to the Langevin equation. This drift term is proportional to the derivative of the diffusion coefficient times a factor that depends on the convention used to define the multiplicative noise. We explore the consequences of this result in a number of examples with spatially varying diffusion coefficients. We also derive a path integral representation for arbitrary interpretation of the noise, and use it in a perturbative study of correlations in a simple system.

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I. INTRODUCTION

Brownian motion provides a paradigm for exploring the dynamics of nonequilibrium systems, especially those that are not driven too far from equilibrium [1–4]. In particular, the Langevin formulation of Brownian motion finds applications that go beyond its original purpose of describing a micrometer-sized particle diffusing in water. It has been extended to treat problems in dynamics of critical phenomena [5], in glassy systems [6], and even in evolutionary biology [7]. Brownian motion is important for soft-matter and biological systems because they are particularly prone to thermal fluctuations [1,8], and Langevin theory is an important tool for describing their properties, such as the dynamics of molecular motors [9] and the viscoelasticity of a polymer network [10].

In most applications, the diffusion coefficient is assumed to be independent of the state of the system. Yet, there are many soft-matter systems in which the diffusion coefficient is state dependent. A simple example of such a system is a particle in suspension near a wall: its friction coefficient, and hence its diffusion coefficient, depends because of hydrodynamic interactions on its distance from the wall [11], a phenomenon that affects interpretation of certain single-molecule force-extension measurements [12] and that plays a crucial role in experimental verification of the fluctuation theorem in a dilute colloidal suspension near a wall [13]. Similarly, the mutual diffusion coefficient of two particles in suspension depends on their separation [14]. Other examples with state-dependent diffusion include a particle diffusing in a reversible chemical polymer gel [15] and the dynamics of fluid membranes [16]. In spite of the recent advances in digital imaging methods to probe equilibrium properties of soft matter [17], there have been relatively few experimental studies of the dynamical properties of a physical system in which the diffusion coefficient is state dependent. This is clearly an area for further experimental exploration. Although the mathematical problem of how to treat systems

with state-dependent diffusion has been studied for some time [2,10,18–20], the results of these studies have not been collected in one place to provide a clear and concise guide to both theorists and experimentalists who might use them.

In this largely expository paper, we develop a Langevin theory and its associated path integral representation for systems with state-dependent diffusion and explore its use in systems of physical interest. In accord with previous treatments [2,3,10,11,18–20], we show that a position-dependent diffusion coefficient leads naturally to multiplicative noise. This noise is the product of a state-dependent prefactor proportional to the square root of the diffusion coefficient and a state-independent Gaussian white noise function, and it is meaningless without a prescription for the temporal order in which the two terms are evaluated. There are two common prescriptions or conventions for dealing with multiplicative noise: the Ito convention, in which the prefactor is evaluated before the Gaussian noise, and the Stratonovich convention, which results when the δ -correlated white noise is obtained as a limit of a noise with a nonzero correlation time [2–4]. There are, however, other conventions, as we will discuss. Using general thermodynamic arguments, we show that, in order for Boltzmann equilibrium to be reached, a drift term proportional to the derivative of the diffusion coefficient times a factor depending on the convention for the evaluation of multiplicative noise must be added to the Langevin equation. Though this drift term has been noted before [10,18–20], we have found only one (recent) reference [10] that specifically associates the form of the drift term with the convention for evaluating multiplicative noise. On the other hand, others claim that it is the choice of the convention that is dictated by physics [11]. In particular, the authors of Ref. [11], without allowing for the possibility of the drift term, argued that neither the Ito nor Stratonovich convention properly describes the dynamics of a Brownian particle with a spatially varying friction coefficient, but a third convention—what the authors called the isothermal convention—does. Incidentally, for this third convention, the

drift term in our formalism vanishes. Therefore, the necessity of the drift term for enforcing thermal equilibrium is not widely known, and it is often incorrectly ignored [12]. Here, we aim to provide a clear exposition for clarifying the technical issues that might have been a source of confusion in the literature.

This paper is organized as follows. In Sec. II, we first review the case of a uniform diffusion coefficient and extend it to the case of spatially varying diffusion coefficient. We discuss in depth the stochastic interpretation issues associated with multiplicative noise, we derive the Fokker-Planck equation, and we show that, depending on the stochastic interpretation, an additional drift term must be added to the standard friction term in order for the system to relax to equilibrium. We also discuss how measurements of the eigenvalues and eigenfunctions of the probability that a particle is at position x' at time $t + \delta t$, given that it was at position x at time t , can be used to obtain information about whether the diffusion coefficient is state dependent or not. In Sec. III, we present some exactly solvable toy models that clearly illustrate the consequences of spatially varying diffusion, and suggest some experimental techniques which may elucidate its role in colloidal tracking experiments. We also give numerical confirmation that the extra drift term is needed to produce equilibrium distribution. In Sec. IV, we derive and discuss the path integral formulation for a Langevin equation with a multiplicative noise, correlation functions, and perturbation theory. In Sec. V, we briefly summarize the results for multicomponent systems. Technical details are presented in the Appendixes.

II. FORMALISM IN ONE DIMENSION

A. A review of the case of a uniform diffusion coefficient

Let us first briefly review the simplest case in which a Brownian particle diffuses in space with a uniform diffusion constant [2]. In the Langevin formulation of Brownian motion, the stochastic equation of motion for the particle's position [8] is

$$\partial_t x = -\Gamma \frac{\partial \mathcal{H}}{\partial x} + g \eta(t), \quad (2.1)$$

where x denotes the position, Γ is the dissipative coefficient (inverse mobility), \mathcal{H} is the Hamiltonian, and $g \eta(t)$ models the stochastic force arising from the rapid collisions of the water molecules with the particle. The strength of this force is set by g , and $\eta(t)$ is a Gaussian white noise with zero mean, $\langle \eta(t) \rangle = 0$, and variance $\langle \eta(t) \eta(t') \rangle = \delta(t - t')$, δ correlated in time. The first term on the right-hand side of Eq. (2.1) describes a dissipative process. Thus, Eq. (2.1) can be viewed as a balancing equation in which the first term drains the energy of the particle while the random noise pumps it back. Equation (2.1) neglects an inertial term that is important only at short times, typically less than 10^{-7} s in soft-matter systems [8]. Thus, Eq. (2.1) tacitly assumes that there is a separation of time scales in which the time scale of the fast processes reflecting microscopic degrees of freedom is much shorter than the typical time scale for the random vari-

able $x(t)$. Hence the white noise assumption in Eq. (2.1).

The Fokker-Planck equation [2–4],

$$\partial_t P(x, t) = \partial_x \left(\Gamma \frac{\partial \mathcal{H}}{\partial x} + \frac{1}{2} g^2 \partial_x \right) P(x, t), \quad (2.2)$$

which can be derived for the Langevin equation, for the probability density $P(x, t)$ that a particle is at position x at time t , provides an alternative to the Langevin equation for describing the motion of Brownian particles. It is easy to see that Eq. (2.2) has a steady state solution $P_s(x) \propto \exp[-(\frac{2\Gamma}{g^2})\mathcal{H}]$. If a particle is in equilibrium with a heat bath at temperature T , then $P_s(x) \propto \exp(-\beta\mathcal{H})$, from which we conclude that $g^2 = 2\Gamma k_B T$. If $\mathcal{H} = 0$, Eq. (2.2) reduces to a diffusion equation with diffusion constant $D = g^2/2$. Hence, for systems in equilibrium at temperature T , the diffusion constant obeys the Einstein relation $D = k_B T \Gamma$.

B. Extension to the case of a state-dependent diffusion coefficient

How must the Langevin equation for a Brownian particle be modified when the friction coefficient Γ depends on position $x(t)$, i.e., when Γ depends on the state of the system? Although it is generally understood [2, 10, 18–21] that an x -dependent Γ leads to an x -dependent g and thus to multiplicative noise $g[x(t)]\eta(t)$, it is less well known that the requirements of long-time thermal equilibrium require an additional specific modification to the Langevin equation—the addition of a convention-dependent drift term. Although there are discussions in the literature of this drift term [10, 11, 18, 20], they are not very detailed, and they generally treat only a specific convention for dealing with multiplicative noise. Here we show that constraints of equilibrium require a unique drift term with each noise convention, and resolve any ambiguities [22] arising from the fact that multiplicative noise can be interpreted in many ways.

Using the argument that the stochastic force is balanced by the dissipative term as in the case of a uniform dissipative coefficient above, we may reasonably postulate a Langevin equation, which trivially generalizes Eq. (2.1) to the case of spatially varying dissipative coefficient, to take the following form:

$$\partial_t x = -\Gamma(x) \frac{\partial \mathcal{H}}{\partial x} + g(x) \eta(t), \quad (2.3)$$

where $g(x) = \sqrt{2k_B T \Gamma(x)}$. But we must first confront the issue of interpreting the multiplicative noise $g(x)\eta(t)$, which by itself is not defined [2, 21]. This is because of the stochastic nature of $\eta(t)$, which in general consists of a series of δ -function spikes of random sign. The value of $g[x(t)]\eta(t)$ depends on whether $g[x(t)]$ is to be evaluated before a given spike, after it, or according to some other rule. It turns out, as we will show shortly, that this naive generalization of Eq. (2.1) to Eq. (2.3) is valid only for a particular interpretation of the noise.

There are a number of approaches to assigning meaning to the multiplicative noise, but they all boil down to providing rules for the evaluation of the integral

$$\mathcal{J}(t, \Delta t) = \int_t^{t+\Delta t} ds g[x(s)] \eta(s), \quad (2.4)$$

in the limit of small Δt . If $g(x)$ and $\eta(s)$ are both continuous functions, this integral could, for arbitrary Δt , be expressed via the first integral mean-value theorem as

$$\mathcal{J}_{\text{cont}}(t, \Delta t) = g[x(t_i)] \int_t^{t+\Delta t} ds \eta(s), \quad (2.5)$$

where t_i is a uniquely determined time in the interval $[t, t + \Delta t]$. In the limit of small Δt , this expression, Eq. (2.5), reduces trivially to $g[x(t)]\eta(t)\Delta t$, to lowest order in Δt . The noise $\eta(s)$ is, however, not continuous and Eq. (2.5) with a uniquely determined time does not apply. One can, however, use Eq. (2.5) to motivate a definition of $\mathcal{J}(t, \Delta t)$ for a stochastic $\eta(s)$. There are two commonly used conventions for defining $\mathcal{J}(t, \Delta t)$: the Stratonovich convention

$$\mathcal{J}_S(t) = g\{[x(t) + x(t + \Delta t)]/2\} \int_t^{t+\Delta t} ds \eta(s), \quad (2.6)$$

in which $g[x(t)]$ is evaluated at the midpoint of the interval $[x(t), x(t + \Delta t)]$, and the Ito convention

$$\mathcal{J}_I(t) = g[x(t)] \int_t^{t+\Delta t} ds \eta(s), \quad (2.7)$$

in which $g[x(t)]$ is evaluated before any noise in the interval $(t, t + \Delta t)$ occurs. We will use a generalized definition:

$$\mathcal{J}_\alpha(t, \Delta t) = g[\alpha x(t + \Delta t) + (1 - \alpha)x(t)] \int_t^{t+\Delta t} ds \eta(s), \quad (2.8)$$

which is parametrized by a continuous variable $\alpha \in [0, 1]$, that reduces to the Ito convention when $\alpha=0$, to the Stratonovich convention when $\alpha=1/2$, and to the isothermal convention of Ref. [11] when $\alpha=1$.

We note in passing that, in the mathematics community, the Ito calculus is most commonly used. Perhaps this is because of the conceptual simplicity arising from the property that the noise increment $\int_t^{t+\Delta t} ds \eta(s)$ and $x(t)$ are statistically independent as implied in Eq. (2.7), i.e., $\langle g(x) \eta(t) \rangle = 0$ [23]. On the other hand, in the physics community, the Stratonovich interpretation is favored. In addition to the advantage that it gives rise to the ordinary rules of calculus, the Stratonovich convention also has a deeper physical origin. Since the noise term in Eq. (2.3) models, in a coarse-grained sense, the effects of microscopic degrees of freedom that have finite (albeit short) correlation times, this term should be physically interpreted as the limit in which these correlation times go to zero. By the Wong-Zakai theorem, this limit corresponds to a white noise that must be interpreted using the Stratonovich convention [23]. However, Eq. (2.3) does not provide a correct description for systems in contact with a thermal bath at temperature T for either interpretation: their associated Fokker-Planck equations do not have long-time thermal-equilibrium solutions.

To return to our main discussion, it is clear that $\mathcal{J}_\alpha(t, \Delta t)$ depends on the value of α . Integration of Eq. (2.3) yields

$$\Delta x(t + \Delta t) \equiv x(t + \Delta t) - x(t) = \mathcal{J}_\alpha(t, \Delta t), \quad (2.9)$$

when $\mathcal{H}=0$. The integral $\int_t^{t+\Delta t} ds \eta(s)$ is statistically of the order of $\sqrt{\Delta t}$, implying that $\Delta x(t + \Delta t)$ is also of the order of $\sqrt{\Delta t}$. Thus, $\alpha x(t + \Delta t) + (1 - \alpha)x(t) = x(t) + \alpha \Delta x(t + \Delta t)$ has a term of order $\sqrt{\Delta t}$ proportional to α , and the order- Δt term in $\mathcal{J}_\alpha(t, \Delta t)$ depends on α . An alternative approach to defining $\mathcal{J}(t, \Delta t)$ is simply to expand $g[x(s)]$ in the integrand as $g[x(s)] = g[x(t)] + [x(s) - x(t)]g'[x(t)] + \dots$. In this approach, which we outline in Appendix A, ambiguities in the interpretation of $\mathcal{J}(t, \Delta t)$ are resolved by specifying the value of the Heaviside unit step function $\theta(t)$ at $t=0$. Setting $\theta(0) = \alpha$ is equivalent to using Eq. (2.8) for $\mathcal{J}(t, \Delta t)$.

The stochastic integral $\mathcal{J}_\alpha(t, \Delta t)$ depends on our convention for evaluating it, i.e., on α . Thus, different values of α define different dynamics. But the requirements of thermal equilibrium should imply a unique dynamics. What is missing? To resolve this dilemma, we consider the general stochastic equation

$$\partial_t x = f(x) + g(x) \eta(t), \quad (2.10)$$

where

$$f(x) = -\Gamma(x) \frac{\partial \mathcal{H}}{\partial x} + f_1(x), \quad (2.11)$$

in which we leave $f_1(x)$ unspecified for the moment. Equation (2.10) is easily integrated using the rules we just outlined to yield

$$\begin{aligned} x(t + \Delta t) - x(t) &= \int_t^{t+\Delta t} ds \{f[x(s)] + g[x(s)]\eta(s)\} \\ &= f[x(t) + \alpha \Delta x] \Delta t + g[x(t) + \alpha \Delta x] \\ &\quad \times \int_t^{t+\Delta t} ds \eta(s), \end{aligned} \quad (2.12)$$

from which we obtain, to the first order in Δt ,

$$\langle \Delta x \rangle = f(x_0) \Delta t + \alpha g(x_0) g'(x_0) \Delta t, \quad (2.13)$$

$$\langle (\Delta x)^2 \rangle = g^2(x_0) \Delta t, \quad (2.14)$$

where we set $x(t) = x_0$. Thus, there is a stochastic contribution $\alpha g g' \Delta t$ to $\langle \Delta x \rangle$ arising from the x dependence of g and depending on the convention for evaluating $\mathcal{J}(t, \Delta t)$. In equilibrium, $\langle \Delta x \rangle$ should be independent of α . Thus, it is apparently necessary to include a contribution to $f(x)$ depending on α .

C. Derivation of the Fokker-Planck equation and equilibrium conditions

To determine the appropriate form of $f(x)$ and $g(x)$ to describe equilibrium systems with a spatially varying friction coefficient $\Gamma(x)$, we derive the Fokker-Planck equation for the probability density $P(x, t)$. The Fokker-Planck equation is most easily derived using the identity

$$P(x, t + \Delta t) = \int dx_0 P(x, t + \Delta t | x_0 t) P(x_0 t), \quad (2.15)$$

where $P(x, t + \Delta t | x_0 t)$ is the conditional probability distribution of x at time $t + \Delta t$, given that it was x_0 at time t . It is defined by

$$P(x, t + \Delta t | x_0 t) = \langle \delta[x - x(t + \Delta t)] \rangle_{x_0 t}, \quad (2.16)$$

where the average is over the random noise $\eta(s)$, and $x(t + \Delta t)$ is determined by Eq. (2.12) with $x(t) = x_0$. Taylor expanding the conditional probability around x_0 yields

$$\begin{aligned} P(x, t + \Delta t | x_0 t) &= \delta(x - x_0) - \langle \Delta x \rangle \frac{\partial}{\partial x} \delta(x - x_0) \\ &\quad + \frac{1}{2} \langle (\Delta x)^2 \rangle \frac{\partial^2}{\partial x^2} \delta(x - x_0) + \dots \end{aligned}$$

Then, using this in Eq. (2.15), we obtain

$$\partial_t P(x, t) = \frac{\partial}{\partial x} [-f(x) - \alpha g(x)g'(x)] P(x, t) + \frac{1}{2} \frac{\partial^2}{\partial x^2} [g^2(x) P(x, t)] \quad (2.17)$$

$$\begin{aligned} &= \frac{\partial}{\partial x} \left(\Gamma(x) \frac{\partial \mathcal{H}}{\partial x} - f_1(x) + (1 - \alpha)g(x)g'(x) \right) \\ &\quad + \frac{1}{2} g^2(x) \frac{\partial}{\partial x} P(x, t). \end{aligned} \quad (2.18)$$

For an equilibrium system, this equation must have a steady-state solution with the canonical form

$$P(x, t) \sim e^{-\mathcal{H}/(k_B T)} \quad (2.19)$$

that is always approached at long times. Such a solution is guaranteed if

$$g^2(x) = 2k_B T \Gamma(x), \quad (2.20)$$

$$f_1(x) = (1 - \alpha)g(x)g'(x) = 2(1 - \alpha)k_B T \Gamma'(x). \quad (2.21)$$

Thus, an additional drift term $f_1(x)$, which depends on the convention for evaluating $\mathcal{J}(t, \Delta t)$, must be added to the standard friction term $-\Gamma(x)\partial_x \mathcal{H}$ in the equation for $\partial_t P$ in order for the system to evolve to the Boltzmann distribution at long times, i.e., be consistent with thermodynamics. Note that $f_1(x)$ is proportional to the temperature T , indicating that its origin arises from random fluctuations rather than from forces identified with a potential. It is clear now from Eq. (2.21) that if we insist on using the Langevin equation in the form of Eq. (2.3), we are forced to take $\alpha = 1$ [11].

It is customary to express the Fokker-Planck equation in terms of the diffusion constant rather than the friction coefficient. From Eq. (2.14) for $\langle (\Delta x)^2 \rangle$, we can identify $g^2(x)$ with the short-time diffusion constant $D(x) = 2k_B T \Gamma(x)$. With this definition of $D(x)$ and $f_1(x)$ given by Eq. (2.21), the Fokker-Planck equation becomes

$$\partial_t P(x, t) = \frac{\partial}{\partial x} D(x) \left(\beta \frac{\partial \mathcal{H}}{\partial x} + \frac{\partial}{\partial x} \right) P(x, t), \quad (2.22)$$

where $\beta = 1/(k_B T)$. As required, this equation is independent of α : different conventions now give the same equilibrium condition, as they should. For a free particle diffusing in spatially varying $D(x)$, $\mathcal{H} = 0$ and Eq. (2.22) becomes

$$\partial_t P(x, t) = \frac{\partial}{\partial x} D(x) \frac{\partial}{\partial x} P(x, t). \quad (2.23)$$

This implies that the correct generalization of Fick's law for equilibrium systems with a spatially varying diffusion coefficient is given by

$$J(x, t) = -D(x) \partial_x P(x, t). \quad (2.24)$$

Historically, the generalization of Fick's law has long been debated [24]. It is commonly acknowledged that Eq. (2.24) is right even though many derivations of the right-hand side of Eq. (2.24) seem not to be as transparent as the one given above.

D. Experimental probes of $D(x)$

One interesting property of Eq. (2.22) is that it necessarily has an eigenstate with eigenvalue zero and eigenfunction given by the equilibrium distribution $P_{eq}(x) \propto e^{-\beta \mathcal{H}(x)}$. This fact is exploited by Crocker and Grier [17] to measure directly the interaction between an isolated pair of colloidal particles. In these experiments, the data from tracking the motion of the particles are used to compute the conditional probability $P(x, t + \delta t | x' t)$, which may be viewed as the Green's function or the inverse of the Fokker-Planck equation, Eq. (2.22). The equilibrium distribution is then the solution to

$$P_{eq}(x) = \int dr' P(x, t + \delta t | x' t) P_{eq}(x'), \quad (2.25)$$

from which the interaction potential can be constructed via $U(x) = -k_B T \ln P_{eq}(x)$.

Since the conditional probability contains all the dynamical information of the system, one could, in principle, characterize how the system relaxes to equilibrium by extracting the nonzero eigenvalues of Eq. (2.22). In particular, the Fokker-Planck equation describing a system with a state-dependent diffusion coefficient would have eigenvalues and eigenfunctions that are, in general, different from those of a system with a uniform diffusion coefficient, even though the two systems have the same Hamiltonian. Thus, in principle, one could extract the Hamiltonian from image analysis following the procedures of Crocker and Grier [17], and solve Eq. (2.22) with uniform diffusion constant to obtain a set of eigenvalues (probably numerically) and compare it with experimentally measured eigenvalues, which can be extracted from the measured conditional probability. If they are different, then the diffusion coefficient is state dependent, and one needs to model the diffusion coefficient to understand the dynamical behaviors of the system. We suggest this procedure as a possible general method for experimentalists to

explore the dynamics and measure the state-dependent friction coefficient in, for example, hydrodynamic interactions between two spheres [25], diffusion of particles in polymer solutions [26], and rods in a nematic environment [27].

III. ILLUSTRATIVE EXAMPLES

In this section, we consider some exactly solvable toy models to illustrate some central ideas presented in the last section. In particular, we address the effects of spatial dependence in the diffusion coefficient and use numerical solution of the Langevin equation to show that the equilibrium distribution is obtained only if $f_1(x)$, Eq. (2.21), is added to the standard friction term.

A. Diffusion of a particle near a wall

How does a diffusion coefficient acquire a spatial dependence? The simplest example is a Brownian particle diffusing near a wall located at $z=0$. Brenner [28] has shown that, for $z>0$, the diffusion coefficient acquires a spatial dependence in which it is zero at the wall, rises linearly in z , and approaches a uniform bulk value of D_0 at large z as

$$\frac{D(z)}{D_0} = 1 - \frac{9a}{8z} + \dots \quad (3.1)$$

Note the long-range component of $D(z)$ in Eq. (3.1), which reflects the long-ranged nature of the hydrodynamic interaction. Recently, it has been pointed out that, in single-molecule experiments, it is crucial to take the spatial dependence in the diffusion coefficient properly into account [12].

Rather than treating the system with the above $D(z)$, we consider a toy model, which correctly describes diffusion close to a wall, in which $D(z)=\Lambda z$. This diffusion coefficient has another experimental realization: diffusion of a colloidal particle bounded by two parallel walls, with one of the walls slightly tilted [11]. Then, the diffusion coefficient acquires a spatial dependence, approximately given by $D(z)\sim z$, for the motion of the particle parallel to the walls. In this case, the Fokker-Planck equation becomes

$$\partial_t P(z,t) = \Lambda \frac{\partial}{\partial z} z \frac{\partial}{\partial z} P(z,t), \quad (3.2)$$

which can be solved exactly. Let $P(z,t)=\sum_n c_n e^{-\lambda_n t} \psi_n(z)$, where λ_n are a set of eigenvalues. With the transformation $y=\sqrt{z}$, Eq. (3.2) can be written as

$$\psi_n''(y) + \frac{1}{y} \psi_n'(y) + \frac{4\lambda_n}{\Lambda} \psi_n(y) = 0, \quad (3.3)$$

whose solution is the Bessel function $\psi(y)=J_0(ky)$, and whose eigenvalues form a continuous spectrum given by $\lambda = \Lambda k^2/4$. The probability distribution as a function of time can be written as

$$P(z,t) = \int_0^\infty dk c(k) e^{-\Lambda k^2 t/4} J_0(k\sqrt{z}). \quad (3.4)$$

The probability distribution for a particle at $z=z_0$ at time $t=0$ evolves as

$$P(z,t) = \frac{1}{\Lambda t} e^{-[(z+z_0)/(\Lambda t)]} I_0[2\sqrt{z z_0}/(\Lambda t)]. \quad (3.5)$$

Unlike its counterpart for a uniform diffusion coefficient, this probability distribution is non-Gaussian. It is straightforward to calculate the moments

$$\langle z(t) \rangle = z_0 + \Lambda t,$$

$$\langle [z(t) - z_0]^2 \rangle = 2\Lambda t(z_0 + \Lambda t).$$

These behaviors are very different from those of a constant diffusion. In particular, the mean-squared displacement exhibits ballistic behavior. It is interesting to observe that the second moment can be written as $\langle [z(t) - z_0]^2 \rangle = 2\Lambda \langle z(t) \rangle t$. This suggests that in order to extract the diffusion coefficient for this simple problem, we need to know not only the second moment $\langle [z(t) - z_0]^2 \rangle$, but also the first moment $\langle z(t) \rangle$. Only for short times does the second moment reduce to $\langle [z(t) - z_0]^2 \rangle \sim 2\Lambda z_0 t = 2D(z_0)t$, which is the formula commonly used to extract the diffusion coefficient. It is clearly incorrect to use this formula for times greater than z_0/Λ . The method we suggested at the end of the last section complements this approach. Note also that the $\langle z(t) \rangle \sim t$ behavior has been measured in Ref. [11].

If the particle is subject to a constant force F , like gravity, in the $-z$ direction, then the Fokker-Planck equation is

$$\partial_t P(z,t) = \Lambda \frac{\partial}{\partial z} z \left(\beta F + \frac{\partial}{\partial z} \right) P(z,t). \quad (3.6)$$

This problem can also be solved exactly. Let $P(z,t) = e^{-\beta F z} \sum_n c_n e^{-\lambda_n t} \psi_n(z)$, we find that the eigenfunctions satisfy the Laguerre equation

$$x\psi'' + (1-x)\psi' + \frac{\lambda_n}{\Lambda\beta F} \psi = 0, \quad (3.7)$$

with eigenvalues $\lambda_n = n\Lambda\beta F$. The eigenvalue spectrum is discrete rather than continuous, as it is in the case of a constant diffusion coefficient. If the particle is initially at z_0 , the distribution evolves as

$$P(z,t) = \frac{\beta F}{1 - e^{-\Lambda\beta Ft}} \exp - \left(\frac{\beta F(z + z_0 e^{-\Lambda\beta Ft})}{1 - e^{-\Lambda\beta Ft}} \right) \times I_0 \left(\frac{2\beta F \sqrt{z_0 z} e^{-\Lambda\beta Ft}}{1 - e^{-\Lambda\beta Ft}} \right). \quad (3.8)$$

Note that, at $t \rightarrow \infty$, this distribution reaches the equilibrium distribution $P_{eq} \sim e^{-\beta F z}$. The first two moments of $z(t)$ are

$$\langle z(t) \rangle = z_0 e^{-\Lambda\beta Ft} + \frac{1}{\beta F} (1 - e^{-\Lambda\beta Ft}),$$

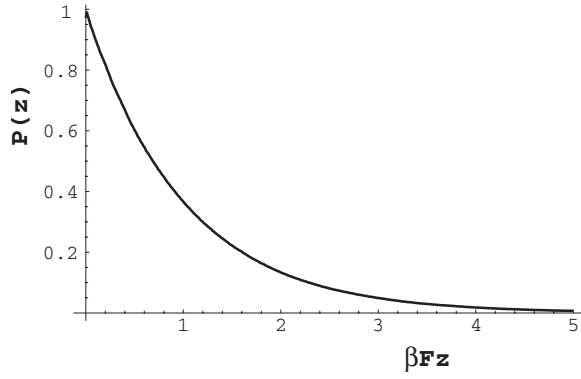


FIG. 1. Stationary distribution for a particle diffusing in a diffusion coefficient $D(z)=\Lambda z$ subject to a constant force $\mathbf{F}=-F\hat{z}$. What is shown here is the numerical simulation of the Langevin equation Eq. (3.10); it is of the form $e^{-\beta Fz}$, as expected.

$$\begin{aligned} \langle z^2(t) \rangle &= z_0^2 e^{-2\Lambda\beta Ft} + \frac{4z_0 e^{-\Lambda\beta Ft}}{\beta F} (1 - e^{-\Lambda\beta Ft}) \\ &+ \frac{2}{(\beta F)^2} (1 - e^{-\Lambda\beta Ft})^2. \end{aligned} \quad (3.9)$$

Note that, at long time, $\langle z(t) \rangle = k_B T / F$ as thermal equilibrium dictates.

We numerically solve the Langevin equation corresponding to this problem,

$$\partial_t z(t) = \Lambda + \Lambda\beta Fz + \sqrt{2\Lambda z} \eta(t), \quad (3.10)$$

in the Ito convention [29]. Note that the first term in the right-hand side arises from the additional drift $f_1(x)$ given by Eq. (2.21). The result for the stationary distribution is plotted in Fig. 1. Obviously, it agrees with the equilibrium distribution $P_{eq} \sim e^{-\beta Fz}$.

B. Diffusion of a particle bounded by two parallel walls

Next, we consider the diffusion of a particle bounded by two walls, which was studied experimentally in Ref. [30] and more recently in Ref. [31]. We approximate the spatially varying diffusion coefficient of this system by $D(x)=D_0[1-(x/L)^2]$. The resulting Fokker-Planck equation is

$$\partial_t P(x,t) = D_0 \frac{\partial}{\partial x} [1 - (x/L)^2] \frac{\partial}{\partial x} P(x,t), \quad (3.11)$$

with boundary conditions that particles cannot penetrate the walls, i.e., that the flux at both walls be zero:

$$J(x,t) = D_0 [1 - (x/L)^2] \frac{\partial}{\partial x} P(x,t) = 0 \quad \text{at } x = \pm L. \quad (3.12)$$

Again, the solution to this problem differs considerably from that with a spatially uniform diffusion coefficient. The spectrum is discrete rather than continuous, with eigenvalues $\lambda_n = n(n+1)D/L^2$ ($n=1, 2, \dots$), and the associated eigenfunctions are Legendre polynomials rather than linear combinations of plane waves. The first two moments of $x(t)$ are

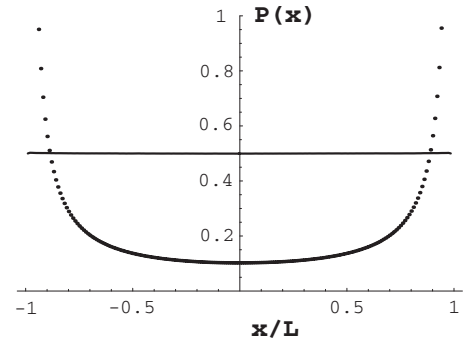


FIG. 2. Stationary distribution for a particle diffusing between two walls with a diffusion coefficient $D(x)=D_0[1-(x/L)^2]$. The solid line is the numerical simulation of the Langevin equation in Eq. (3.14), and the dotted line is the numerical simulation of the Langevin equation without the extra gg' term.

$$\langle x(t) \rangle = x_0 e^{-2D_0 t/L^2},$$

$$\langle x(t)^2 \rangle = x_0^2 e^{-6D_0 t/L^2} + \frac{L^2}{3} (1 - e^{-6D_0 t/L^2}). \quad (3.13)$$

These moments again are different from those in the case in which the diffusion is uniform.

We performed numerical simulation of the Langevin equation,

$$\partial_t x = -2D_0 x/L^2 + \sqrt{2D_0(1-x^2/L^2)} \eta(t), \quad (3.14)$$

where the first term arises from the gg' term. In Fig. 2, we plot the long time distribution (solid line) which is uniform, as it should be. We also show the numerical results for the case in which we did not add gg' (dotted line). Clearly, we get the wrong answer if we do not add the gg' term.

C. Diffusion constant: $D(x)=D_0(1+\gamma x^2)$

As a final example, let us consider a free particle diffusing with $D(x)=D_0(1+\gamma x^2)$ in the bulk. The Fokker-Planck equation is given by

$$\partial_t P(x,t) = D_0 \frac{\partial}{\partial x} (1 + \gamma x^2) \frac{\partial}{\partial x} P(x,t). \quad (3.15)$$

Multiplying both sides by x^2 and integrating, we find

$$\partial_t \langle x^2(t) \rangle = 2D_0 + 6D_0 \gamma \langle x^2(t) \rangle, \quad (3.16)$$

whose solution is

$$\langle x^2(t) \rangle = \frac{1}{3\gamma} [\exp(6D_0 \gamma t) - 1]. \quad (3.17)$$

Thus, the second moment grows exponentially with time; this peculiar behavior illustrates the dramatic effects of the noise in problems with a spatial-dependent diffusion coefficient.

IV. PATH INTEGRAL FORMULATION

Path integral formalisms provide an alternative to the Fokker-Planck and Langevin equations for the description of

stochastic dynamics. They have the advantage that well-established perturbative and nonperturbative field-theoretic techniques [5,32] can be used to calculate the effects of nonlinearities. They also provide a convenient treatment of correlation and response functions. The path integral for a state-dependent dissipation coefficient has been derived previously in either the Stratonovich or Ito convention [5,22,33,34]. In this section, we derive the path integral for the general α convention, use it along with the detailed balance, a condition that any thermal system must satisfy, to give further insight into the additional drift term derived in Sec. II C. We also discuss the equilibrium correlation and response functions and prove the fluctuation-dissipative theorem for a state-dependent diffusion coefficient. We then set up perturbation theory for systems with a coordinate-dependent friction coefficient.

The path integral is based on the statistics of a path $x(t)$. We discretize the path into segments $x_i=x(t_i)$ with $t_0 < t_1 < \dots < t_{N-1} < t_f$ and $\Delta t \equiv t_n - t_{n-1}$ small. The joint probability distribution $P(x_N t_N; x_{N-1} t_{N-1}; \dots; x_1 t_1 | x_0 t_0)$ that $x(t)$ takes on values of x_1 at time t_1 , x_2 at time t_2 , and so on, given that it has value of x_0 at time t_0 , is then

$$P(x_f t_f; x_{N-1} t_{N-1}; \dots; x_1 t_1 | x_0 t_0) = \langle \delta[x_N - \phi(t_N; x_0, t_0)] \dots \delta[x_1 - \phi(t_1; x_0, t_0)] \rangle,$$

where the average is taken with respect to the noise and $\phi(t_i; x_{i-1}, t_{i-1})$ is the solution to the Langevin equation, Eq. (2.10), for $x(t_i)$, given that $x(t_{i-1})=x_{i-1}$. Since the noise in Eq. (2.10) is δ -correlated in time, the noise in different time intervals is not correlated, and x_i depends only on x_{i-1} . Thus, we can write

$$P(x_N t_N; \dots; x_1 t_1 | x_0 t_0) = \prod_{i=1}^N \langle \delta[x_i - \phi(t_i; x_{i-1}, t_{i-1})] \rangle.$$

The function

$$P(x_i t_i | x_{i-1} t_{i-1}) = \langle \delta[x_i - \phi(t_i; x_{i-1}, t_{i-1})] \rangle \quad (4.1)$$

gives the conditional probability that the random variable $x(t)$ has the value x_i at time t_i given that it had a value x_{i-1} at t_{i-1} . Using Eq. (4.1) and the identity $P(xt) = \int dx' P(xt; x't')$, it is easy to see that

$$P(x_i t_i | x_{i-2} t_{i-2}) = \int dx_{i-1} P(x_i t_i | x_{i-1} t_{i-1}) P(x_{i-1} t_{i-1} | x_{i-2} t_{i-2}), \quad (4.2)$$

$$P(x_i t_i) = \int dx_{i-1} P(x_i t_i | x_{i-1} t_{i-1}) P(x_{i-1} t_{i-1}). \quad (4.3)$$

Equation (4.2) is the Chapman-Kolmogorov equation, which defines a Markov process [2], while Eq. (4.3) is just an identity, true for all stochastic processes. Note that a Markov process is completely specified if we know $P(x_i t_i)$ and $P(x_i t_i | x_{i-1} t_{i-1})$, but they are not arbitrary because they are linked through Eqs. (4.2) and (4.3). Using Eq. (4.2), the conditional probability for the particle to go from x_0 at time t_0 to x_f at time t_f is

$$P(x_f t_f | x_0 t_0) = \int dx_{N-1} \dots \int dx_1 \langle \delta[x_f - \phi(t_f; x_{N-1}, t_{N-1})] \rangle \dots \times \langle \delta[x_1 - \phi(t_1; x_0, t_0)] \rangle. \quad (4.4)$$

This is the basic construct for the path integral. First, we have to evaluate $\langle \delta[x_i - \phi(t_i; x_{i-1}, t_{i-1})] \rangle$. We discretize Eq. (2.10) as follows:

$$x_i = x_{i-1} + \Delta t f_i + g_i \int_{t_{i-1}}^{t_i} dt' \eta(t'), \quad (4.5)$$

where $\Delta t = t_i - t_{i-1}$, $f_i = f[\alpha x_i + (1-\alpha)x_{i-1}]$, and $g_i = g[\alpha x_i + (1-\alpha)x_{i-1}]$. We next introduce the function $h(x_i, x_{i-1})$:

$$h(x_i, x_{i-1}) = \frac{x_i - x_{i-1} - \Delta t f_i}{g_i} - \int_{t_{i-1}}^{t_i} dt' \eta(t'), \quad (4.6)$$

which vanishes when x_i is the unique solution to Eq. (4.5), $\phi(t_i; x_{i-1}, t_{i-1})$, i.e., $h[\phi(t_i; x_{i-1}, t_{i-1}), x_{i-1}] = 0$. Using the property of the δ function

$$\delta[h(x_i, x_{i-1})] = \left| \frac{\partial h}{\partial x_i} \right|_{x_i=\phi(t_i)}^{-1} \delta[x_i - \phi(t_i)],$$

and noting that $|\partial h / \partial x_i|_{x_i=\phi(t_i)}^{-1}$ depends only on x_i and x_{i-1} , which are set by the δ function, and not explicitly on the noise, we have

$$\begin{aligned} \langle \delta[h(x_i, x_{i-1})] \rangle &= \left\langle \left| \frac{\partial h}{\partial x_i} \right|_{x_i=\phi(t_i)}^{-1} \delta[x_i - \phi(t_i)] \right\rangle \\ &= \left| \frac{\partial h}{\partial x_i} \right|^{-1} \langle \delta[x_i - \phi(t_i)] \rangle, \end{aligned}$$

since for any function $q[\phi(t_i)]$, $\langle q[\phi(t_i)] \delta[x_i - \phi(t_i)] \rangle = q(x_i) \times \langle \delta[x_i - \phi(t_i)] \rangle$. We can, therefore, write the conditional probability as

$$\begin{aligned} P(x_i t_i | x_{i-1} t_{i-1}) &= \langle \delta[x_i - \phi(t_i; x_{i-1}, t_{i-1})] \rangle \\ &= \left| \frac{\partial h(x_i, x_{i-1})}{\partial x_i} \right| \langle \delta[h(x_i, x_{i-1})] \rangle, \end{aligned}$$

with

$$\frac{\partial h}{\partial x_i} = \frac{1}{g_i} \left(1 - \alpha \Delta t f'_i - \alpha \frac{g'_i}{g_i} (x_i - x_{i-1} - \Delta t f_i) \right),$$

where the prime denotes the derivative. The average over noise can be easily done with the aid of the Fourier representation of the δ function:

$$\begin{aligned} \langle \delta[h(x_i, x_{i-1})] \rangle &= \int \frac{dk_i}{2\pi} e^{+i(k_i/g_i)(x_i - x_{i-1} - \Delta t f_i)} \\ &\quad \times \left\langle \exp \left(-ik_i \int_{t_{i-1}}^{t_i} dt' \eta(t') \right) \right\rangle \\ &= \int \frac{dk_i}{2\pi} e^{+i(k_i/g_i)(x_i - x_{i-1} - \Delta t f_i) - k_i^2 \Delta t / 2}, \end{aligned}$$

where we have made use of the fact that $\int_{t_{i-1}}^{t_i} dt' \eta(t')$ is a zero-mean Gaussian random variable with variance Δt . Putting these results together, we can express $P(x_i t_i | x_{i-1} t_{i-1})$ as

$$P(x_i t_i | x_{i-1} t_{i-1}) = \int \frac{dk_i}{2\pi g_i} e^{+(k_i/g_i)(x_i - x_{i-1} - \Delta t f_i) - k_i^2 \Delta t/2} \times \left(1 - \alpha \Delta t f_i' - \alpha \frac{g_i'}{g_i} (x_i - x_{i-1} - \Delta t f_i) \right). \quad (4.7)$$

Next, in order to derive the path integral, which is of the form $\sim e^{-S}$, we need to “exponentiate” the term in parentheses in Eq. (4.7) and keep all the terms that are of order of Δt in the exponential. However, we cannot simply exponentiate the third term in the parentheses because this term contains $\Delta x_i \equiv x_i - x_{i-1}$, which is of the order of $\sqrt{\Delta t}$. This is noted in Ref. [22], where the author derives the path integral for the Stratonovich convention, and circumvents this difficulty by keeping the second-order term in Δx in the exponential and replacing this term with its average value. Although the final expression is correct, that derivation might be inconsistent with the concept of the path integral, since the derivation is valid only in the mean-squared sense instead of for all paths, as required by the path integral. Here, we provide an alternative derivation that is valid for each path. First, we note that the last term in the parentheses can be written as

$$\begin{aligned} & - \int \frac{dk_i}{2\pi g_i} \left(\alpha \frac{g_i'}{g_i} (\Delta x_i - \Delta t f_i) \right) e^{+(k_i/g_i)(\Delta x_i - \Delta t f_i) - k_i^2 \Delta t/2} \\ & = - \alpha g_i' \int \frac{dk_i}{2\pi g_i} e^{-k_i^2 \Delta t/2} \left(-i \frac{\partial}{\partial k_i} \right) e^{+(k_i/g_i)(\Delta x_i - \Delta t f_i)} \\ & = + \alpha g_i' \int \frac{dk_i}{2\pi g_i} (i k_i \Delta t) e^{+(k_i/g_i)(\Delta x_i - \Delta t f_i) - k_i^2 \Delta t/2}, \end{aligned} \quad (4.8)$$

where the last line is explicitly of order of Δt and can, therefore, be exponentiated without incurring any error to the first order in Δt . Returning to the conditional probability, we have

$$P(x_i t_i | x_{i-1} t_{i-1}) = \int \frac{dk_i}{2\pi g_i} e^{+(k_i/g_i)(\Delta x_i - \Delta t f_i) - k_i^2 \Delta t/2} (1 - \alpha \Delta t f_i' + i \alpha \Delta t k_i g_i') \quad (4.9)$$

$$= \int \frac{dk_i}{2\pi g_i} e^{+(k_i/g_i)(\Delta x_i - \Delta t f_i + \alpha \Delta t g_i g_i') - k_i^2 \Delta t/2 - \alpha \Delta t f_i'} \quad (4.10)$$

$$= \frac{1}{\sqrt{2\pi \Delta t g_i}} e^{-(\Delta t/2g_i^2)(\Delta x_i/\Delta t - f_i + \alpha g_i g_i')^2 - \alpha \Delta t f_i'}, \quad (4.11)$$

where the last line is valid to the first order in Δt . It should be noted that the Fokker-Planck equation, Eq. (2.17), can also be derived using Eq. (4.11) and the identity of Eq. (4.3). This is done in Sec. IV A. Returning to Eq. (4.4), we have

$$P(x_f t_f | x_0 t_0) = \int \frac{dx_1}{\sqrt{2\pi \Delta t g_1}} \cdots \int \frac{dx_{N-1}}{\sqrt{2\pi \Delta t g_{N-1}}} \frac{1}{\sqrt{2\pi \Delta t g_N}} \times \exp \left[- \sum_i \frac{\Delta t}{2g_i^2} \left(\frac{x_i - x_{i-1}}{\Delta t} - f_i + \alpha g_i' g_i \right)^2 - \sum_i \alpha \Delta t f_i' \right] \quad (4.12)$$

$$\begin{aligned} & = \int_{x_0}^{x_f} \mathcal{D}x \exp \left[- \int_{t_0}^{t_f} dt \left(\frac{1}{2g(x)^2} [\partial_t x - f(x) + \alpha g(x)g'(x)]^2 + \alpha f'(x) \right) \right] \\ & = \int_{x_0}^{x_f} \mathcal{D}x e^{-S}, \end{aligned} \quad (4.13)$$

with $\mathcal{D}x \equiv \prod_{i=1}^N dx_i / \sqrt{2\pi \Delta t g_i}$, and the action given by

$$S = \int_{t_0}^{t_f} dt \left(\frac{1}{2g(x)^2} [\partial_t x - f(x) + \alpha g(x)g'(x)]^2 + \alpha f'(x) \right), \quad (4.14)$$

where we have taken the formal limit by letting $N \rightarrow \infty$ and $\Delta t \rightarrow 0$. Note the extra terms in S coming from the Jacobian $|\partial h / \partial x_i|$; they are needed in order to ensure that $\int dx_f P(x_f t_f | x_0 t_0) = 1$. This can be demonstrated by explicit, but tedious, calculation for general α (see Appendix B). From Eq. (4.14), it is clear that the Ito convention with $\alpha = 0$ is the simplest to deal with. Another particularly useful form of the path integral is obtained using the Hubbard-Stratonovich transformation, which linearizes the quadratic term,

$$P(x_f t_f | x_0 t_0) = \int \mathcal{D}y \int_{x_0}^{x_f} \mathcal{D}x \exp - \int_{t_0}^{t_f} dt \left(\frac{g(x)^2}{2} y^2(t) - iy(t) \times [\partial_t x - f(x) + \alpha g(x)g'(x)] + \alpha f'(x) \right), \quad (4.15)$$

where the measure now is $\int \mathcal{D}y \int_{x_0}^{x_f} \mathcal{D}x = \int dy_N / 2\pi \cdots \int (dy_1 / 2\pi) \int dx_{N-1} \cdots \int dx_1$. This result could, of course, also have been obtained directly by substituting $k_i \equiv g_i y_i$ in Eq. (4.10) and taking the continuum limit. Note that, in the discretized version of Eq. (4.15), y_n is associated with time $t_n^* = \alpha t_n + (1 + \alpha)t_{n-1}$. This form of the path integral is closely related to the Martin-Siggia-Rose (MSR) formalism [32] to calculate the response and correlation functions. This will be explored in Sec. IV B.

It is interesting to see how the additional drift term $f_1(x)$ in the Langevin equation [Eq. (2.11)] arises from the constraints that equilibrium statistical mechanics impose on the path integral formulation [35]. Thermal systems must obey detailed balance, which states that

$$P(x_f t_f | x_0 t_0) P_{eq}(x_0) = P(x_0 t_f | x_f t_0) P_{eq}(x_f). \quad (4.16)$$

The equilibrium distribution has the form $P_{eq}(x) = \exp[-\beta \mathcal{H}(x)]$, and $P(x_0 t_f | x_f t_0)$ is the conditional probability

ity for the reversed path, i.e., for $\bar{x}(t)=x(-t)$. It turns out that the Stratonovich convention is simplest for the discussion of time-reversal properties not only because it obeys the ordinary rule of differential calculus, but also because it has the property that the forward and backward paths are evaluated at the same points. We will employ the Stratonovich convention below. First, we note that

$$\begin{aligned} \frac{P_{eq}(x_f)}{P_{eq}(x_0)} &= \exp[-\beta\mathcal{H}(x_f) + \beta\mathcal{H}(x_0)] \\ &= \exp\left(-\int_{t_0}^{t_f} dt (\partial_t x) \frac{\partial\beta\mathcal{H}(x)}{\partial x}\right), \end{aligned} \quad (4.17)$$

and that $P(x_0 t_f | x_f t_0)$ can be obtained simply by noting that the path associated with this distribution is the time-reversal path of $P(x_f t_f | x_0 t_0)$, which can be written as

$$\begin{aligned} P(x_0 t_f | x_f t_0) &= \int \mathcal{D}\bar{y} \int_{x_0}^{x_f} \mathcal{D}x \exp\left\{-\int_{t_0}^{t_f} dt \left[\frac{g^2}{2}\bar{y}^2 + i\bar{y}\left(\partial_t x + f\right.\right.\right. \\ &\quad \left.\left.\left. - \frac{1}{2}g g'\right) + \frac{1}{2}f'\right]\right\}. \end{aligned} \quad (4.18)$$

Now, using Eqs. (4.16)–(4.18) and comparing them term by term with the exponential in Eq. (4.15) (in the Stratonovich convention $\alpha=1/2$), we see that

$$i\bar{y}(t) = -\left(iy(t) + \frac{\partial\beta\mathcal{H}(x)}{\partial x}\right), \quad (4.19)$$

$$f(x) = \frac{1}{2}g(x)g'(x) - \frac{g^2(x)}{2} \frac{\partial\beta\mathcal{H}(x)}{\partial x}. \quad (4.20)$$

The first term in Eq. (4.20) is identical to Eq. (2.21) in the Stratonovich interpretation. The second term is the standard frictional term, from which we identify the dissipation coefficient as $\Gamma(x) = \beta g^2(x)/2$, which is the Einstein relation. This derivation again demonstrates that equilibrium distribution is the only physics needed to fix $f(x)$ for a given stochastic interpretation.

A. Derivation of the Fokker-Planck equation from the path integral

In this section, we derive the Fokker-Planck equation directly from the conditional probability Eq. (4.11), thereby establishing the equivalence of the path integral formulation and the Fokker-Planck equation for general α . Let us rewrite the conditional probability, Eq. (4.10), where we set $x=x_i$, $t=t_i$, $x'=x_{i-1}$, $t'=t_{i-1}$, and $k_i \equiv g_i y_i$:

$$\begin{aligned} P(xt|x't') &= \int \frac{dy_i}{2\pi} e^{-\Delta t[(g_i^2/2)y_i^2 - iy_i(\Delta x_i/\Delta t - f_i + \alpha g_i g'_i) + \alpha f'_i]} \\ &= \int \frac{dy_i}{2\pi} e^{-\Delta t A_i + iy_i \Delta x_i}, \end{aligned} \quad (4.21)$$

where

$$A_i[x_i, x_{i-1}; y_i] \equiv \frac{g_i^2}{2} y_i^2 + iy_i(f_i - \alpha g_i g'_i) + \alpha f'_i, \quad (4.22)$$

$\Delta x_i = x - x'$, and $\Delta t = t - t'$. Our aim is to calculate

$$P(x, t) = \int dx' P(xt|x't') P(x', t'), \quad (4.23)$$

to first order in Δt . Expanding $P(x', t)$,

$$\begin{aligned} P(x', t') &= P(x - \Delta x_i, t') \\ &= P(x, t') - \Delta x_i \frac{\partial}{\partial x} P(x, t') + \frac{(\Delta x_i)^2}{2} \frac{\partial^2}{\partial x^2} P(x, t'), \end{aligned}$$

and putting this back in Eq. (4.23), we find that it can be cast in the form

$$\begin{aligned} \frac{P(x, t) - P(x, t')}{\Delta t} &= \alpha(x) P(x, t') + \beta(x) \frac{\partial}{\partial x} P(x, t') \\ &\quad + \gamma(x) \frac{\partial^2}{\partial x^2} P(x, t'), \end{aligned} \quad (4.24)$$

where $\alpha(x) \equiv \lim_{\Delta t \rightarrow 0} [\mathcal{I}_0(x) - 1]/\Delta t$, $\beta(x) \equiv -\lim_{\Delta t \rightarrow 0} \mathcal{I}_1(x)/\Delta t$, and $\gamma(x) \equiv \lim_{\Delta t \rightarrow 0} \mathcal{I}_2(x)/(2\Delta t)$. The main task is to evaluate integrals of the form

$$\mathcal{I}_m(x) \equiv \int \frac{dy_i}{2\pi} \int d\Delta x_i (\Delta x_i)^m Q(x, \Delta x_i; y_i) e^{iy_i \Delta x_i}, \quad (4.25)$$

where

$$\begin{aligned} Q(x, \Delta x_i; y_i) &= e^{-\Delta t A_i[x, x'; y_i]}, \\ &= \sum_k \frac{(\Delta x_i)^k}{k!} \frac{\partial^k}{\partial \Delta x_i^k} Q(x, \Delta x_i; y_i) \Big|_{\Delta x_i=0}, \end{aligned} \quad (4.26)$$

where in the last line, we have Taylor expanded the function $Q(x, \Delta x_i; y_i)$. It is easy to see that

$$\int d\Delta x_i (\Delta x_i)^m e^{iy_i \Delta x_i} = 2\pi (-i)^m \frac{\partial^m}{\partial y_i^m} \delta(y_i), \quad (4.27)$$

and therefore

$$\mathcal{I}_m(x) = \sum_k \frac{(i)^{k+m}}{k!} \frac{\partial^{m+k}}{\partial y_i^{m+k}} Q^{(k)}(x, 0; y_i) \Big|_{y_i=0}, \quad (4.28)$$

where $Q^{(k)}(x, 0; y_i) \equiv \partial^k Q(x, \Delta x_i; y_i)/\partial \Delta x_i^k|_{\Delta x_i=0}$. Using Eq. (4.28), it is straightforward to compute $\mathcal{I}_m(x)$ to the first order in Δt ; we obtain

$$\mathcal{I}_0(x) = 1 - f'(x)\Delta t + (1 - \alpha)\{[g'(x)]^2 + g(x)g''(x)\}\Delta t, \quad (4.29)$$

$$\mathcal{I}_1(x) = [f(x) - (2 - \alpha)g(x)g'(x)]\Delta t, \quad (4.30)$$

$$\mathcal{I}_2(x) = g^2(x)\Delta t, \quad (4.31)$$

with vanishing higher-order terms, i.e., $\mathcal{I}_n(x) = 0$ for $n \geq 3$. Therefore, we have

$$\alpha(x) = -f'(x) + (1 - \alpha)[g'(x)^2 + g(x)g''(x)], \quad (4.32)$$

$$\beta(x) = -f(x) + (2 - \alpha)g(x)g'(x), \quad (4.33)$$

$$\gamma(x) = \frac{1}{2}g(x)^2. \quad (4.34)$$

This is equivalent to the Mori expansion [3]. It is clear that with these coefficients Eq. (4.24) becomes Eq. (2.17), the Fokker-Planck equation.

B. Correlation, response functions, and fluctuation-dissipation theorem

One of the advantages of the particular form of the path integral in Eq. (4.15) is that correlation and response functions can be computed conveniently from it. The average of any functional $O[x(t), y(t)]$ of $x(t)$ and $y(t)$ at fixed x_0 is given by

$$\langle O[x(t), y(t)] \rangle_{x_0} = \int \mathcal{D}y \int_{x_0} \mathcal{D}x O[x(t), y(t)] e^{-\mathcal{S}}, \quad (4.35)$$

where $l_n(x)$ is the modified Bessel function. In particular, the two-point correlation function is

$$\langle x(t_1)x(t_2) \rangle_{x_0} = \int \mathcal{D}y \int_{x_0} \mathcal{D}x x(t_1)x(t_2) e^{-\mathcal{S}}, \quad (4.36)$$

and the propagator function

$$G(t_2, t_1) \equiv \langle x(t_2)[-iy(t_1)] \rangle_{x_0} = \int \mathcal{D}y \int_{x_0} \mathcal{D}x x(t_2)[-iy(t_1)] e^{-\mathcal{S}}. \quad (4.37)$$

Physically, the propagator describes the response of the system to a δ perturbation. One of the nice features of the propagator function, which is useful in perturbative expansions, is that causality is automatically built in, i.e., $G(t, t') = 0$ if $t < t'$. To see this, we go back to the discretized form of the path integral, and write $G(t, t')$ as

$$G(t_m, t_n) = \int \frac{dy_N}{2\pi} \cdots \int \frac{dy_1}{2\pi} \int dx_N \cdots \int dx_1 \times x_m[-iy_n] \exp\left(-\sum_i \Delta t \mathcal{A}_i + \sum_i iy_i \Delta x_i\right), \quad (4.38)$$

where $\mathcal{A}_i[x_i, x_{i-1}; y_i]$ is defined in Eq. (4.22). First, let us consider $t_n > t_m$; each pair of the integrals $\int (dy_i/2\pi) \int dx_i$ in Eq. (4.38) gives 1 for $i > n$. When integrating over x_n , we make use of the identity

$$\int dx_n \mathcal{Q}[x_{n-1}, \Delta x_n; y_n] e^{iy_n \Delta x_n} = \sum_k \frac{(iy_n)^k \mathcal{Q}^{(k)}[x_{n-1}, 0, y_n]}{k!} \times \frac{\partial^k}{\partial y_n^k} \delta(y_n), \quad (4.39)$$

which gives zero when integrating y_n . Thus, we have shown that $\langle x_m[-iy_n] \rangle_{x_0} = 0$ for all $n > m$. Now, suppose $m = n$, one can show that, using the above identity, $\langle x_n[-iy_n] \rangle_{x_0} = 1$. Clearly, $\langle x_m[-iy_n] \rangle_{x_0} \neq 0$, if $m > n$. Thus, we have shown how the path integral enforces causality, i.e., $G(t, t') = 0$ if $t < t'$, and $G(t, t') \neq 0$ if $t > t'$. However, there is a subtle point about the value of $G(t, t)$ in the continuum limit, which has to be consistent with the α convention. The simplest way to do this is to note that, since y_n is really associated with time at $t_n^* = \alpha t_n + (1 - \alpha)t_{n-1}$, we have to evaluate

$$G(t, t) \equiv \langle x(t_n^*)[-iy_n] \rangle_{x_0} = \langle [\alpha x_n + (1 - \alpha)x_{n-1}] [-iy_n] \rangle_{x_0} = \alpha.$$

Now, we specialize to a system near equilibrium, and we investigate how the path integral describes properties such as the fluctuation-dissipation theorem. The equilibrium average of any function $O[x(t), y(t)]$ of $x(t)$ and $y(t)$ is defined as

$$\langle O[x(t), y(t)] \rangle_{eq} = \int \mathcal{D}y \int \mathcal{D}x O[x(t), y(t)] e^{-\mathcal{S}} P_{eq}(x_0). \quad (4.40)$$

Note that equilibrium averages are independent of α , provided that we add the additional drift $f_1(x)$. When the system is under a time-dependent physical force $h(t)$, the total Hamiltonian is $\mathcal{H}_T = \mathcal{H}_0(x) - x(t)h(t)$, so that

$$\begin{aligned} f(x, t) &= (1 - \alpha)gg' - \Gamma(x) \frac{\partial \mathcal{H}_T}{\partial x} \\ &= f_0(x) + \Gamma(x)h(t), \\ f_0(x) &= (1 - \alpha)gg' - \Gamma(x) \frac{\partial \mathcal{H}_0}{\partial x}. \end{aligned}$$

Therefore, we have

$$\begin{aligned} \left. \frac{\delta \langle x(t) \rangle}{\delta h(t')} \right|_{h(t)=0} &= \frac{1}{2k_B T} \langle x(t) \{ -iy(t')g[x(t')]^2 \} \rangle_{eq} \\ &\quad - \frac{\alpha}{k_B T} \langle x(t)g[x(t')]g'[x(t')] \rangle_{eq} \\ &\equiv \chi_{xx}(t, t'). \end{aligned} \quad (4.41)$$

We observe that the response $\chi_{xx}(t, t')$ to a physical forces and the propagator $G(t, t')$ defined in Eq. (4.37) are different, although they are proportional to each other for the case of uniform diffusion constant. In particular, there is an additional term arising from the normalization factor f' in the action and it is absent when the diffusion constant is spatially uniform. By integration by parts, the first term in Eq. (4.41) can be evaluated to be

$$\begin{aligned} \langle x(t) \{ -iy(t')g[x(t')]^2 \} \rangle_{eq} &= \langle x(t) \{ \partial_t x(t') - f_0[x(t')] \\ &\quad + \alpha g[x(t')]g'[x(t')] \} \rangle_{eq}. \end{aligned}$$

Therefore, the physical response function is

$$\chi_{xx}(t, t') = \frac{1}{2k_B T} \langle x(t) \{ \partial_{t'} x(t') - \mathcal{B}[x(t')] \} \rangle_{eq}, \quad (4.42)$$

where $\mathcal{B}(x) \equiv gg' - \Gamma(x) \partial \mathcal{H}_0 / \partial x$. Note that the physical response function is independent of α , as it should be; note also that the drift proportional to gg' arises from a spatial varying diffusion constant. To proceed further, we note that, as a consequence of the detailed balance condition, Eq. (4.16), the equilibrium correlation function is symmetric with respect to exchange of $t \leftrightarrow t'$:

$$\langle O_1[x(t)] O_2[x(t')] \rangle_{eq} = \langle O_1[x(t')] O_2[x(t)] \rangle_{eq}.$$

Applying this to Eq. (4.42) and subtracting the results, we have

$$[\partial_t - \partial_{t'}] \langle x(t)x(t') \rangle_{eq} = 2k_B T [\chi_{xx}(t', t) - \chi_{xx}(t, t')].$$

Since the correlation function is time-translation invariant, we must have $\partial_{t'} \langle x(t)x(t') \rangle_{eq} = -\partial_t \langle x(t)x(t') \rangle_{eq}$. Thus,

$$\partial_t \langle x(t)x(t') \rangle_{eq} = -k_B T [\chi_{xx}(t, t') - \chi_{xx}(t', t)].$$

This is the fluctuation-dissipation theorem (FDT). To put it in a more traditional form, we note that $\chi_{xx}(t', t) = 0$ when $t > t'$, and we can write

$$\chi_{xx}(t - t') = -\frac{1}{k_B T} \partial_{t'} \langle x(t)x(t') \rangle_{eq} \theta(t - t'),$$

where $\theta(t)$ is the Heaviside unit step function. The Fourier transform of the response function is given by

$$\langle x(\omega)x(-\omega) \rangle_{eq} = \frac{2k_B T}{\omega} \text{Im} \chi_{xx}(\omega), \quad (4.43)$$

which is of the form that is commonly quoted in the literature.

C. Perturbation theory

One of the advantages of the path integral formulation of stochastic dynamics is that it is by construction a field theory that facilitates systematic perturbative calculation of correlation functions. In particular, for systems with state-dependent dissipative coefficients, the resulting Langevin equation is generally nonlinear, and perturbation theory is a convenient way to derive the mode-coupling theory [36]. Thus, in this section, we set up the perturbation theory for a systematic calculation of correlation and response functions in the deviation of the diffusion coefficient from spatial uniformity. First, we need to set up the generating functional. Note that $P_{eq}(x_0)$ satisfies

$$P_{eq}(x) = \int dx_s P(xt|x_s t_s) P_{eq}(x_s), \quad (4.44)$$

which implies that

$$P_{eq}(x) = \lim_{t_s \rightarrow -\infty} P(xt|x_s t_s). \quad (4.45)$$

Thus, the equilibrium averages can be written as

$$\begin{aligned} \langle O[x(t), y(t)] \rangle_{eq} &= \int \mathcal{D}y \int \mathcal{D}x O[x(t), y(t)] e^{-S} P_{eq}(x_0) \\ &= \int \mathcal{D}y \int \mathcal{D}x O[x(t), y(t)] e^{-S}, \end{aligned}$$

where, in the last line, the limit of the time integration in the action S is extended to $-\infty$ to ∞ . This allows us to define the generating function for equilibrium averages by

$$\mathcal{Z}[F, \tilde{F}] = \int \mathcal{D}y \int \mathcal{D}x \exp\left(-S + \int dt [x(t)F(t) - iy(t)\tilde{F}(t)]\right). \quad (4.46)$$

The correlation functions and the propagator are simply functional derivatives of \mathcal{Z} . This sets up the MSR perturbation scheme [32], which allows the immediate application of all of the powerful techniques of field theory, including the renormalization group, to nonlinear stochastic problems. It is customary to introduce the variables $\hat{x}(t) \equiv -iy(t)$. Note that, in the perturbation expansion, all the α -dependent terms cancel provided that we use $\langle x(t)\hat{x}(t) \rangle_0 = \alpha$ (see Appendix C). Therefore, it is convenient to use $\alpha=0$ at the outset.

As an informative model calculation, we explore the problem in which a particle diffuses with $D(x) = D_0(1 + \gamma x^2)$ confined in a harmonic potential $\mathcal{H} = kx^2/2$. If the confining potential is turned off, this problem is exactly solvable, as shown in Sec. III C. It can also be solved exactly when $\gamma = 0$ but not when $\gamma \neq 0$, and a perturbative expansion in γ is useful. The goal of this exercise is to compute the propagator $\langle x\hat{x} \rangle$ and the correlation function $\langle xx \rangle$ separately and to check that the fluctuation-dissipation theorem is satisfied. For simplicity, we work in the Ito convention (see Appendix C for general α), and set $k_B = 1$. According to the formalism, we have

$$f(x) = g(x)g'(x) - \Gamma(x) \frac{\partial \mathcal{H}}{\partial x} = -\Gamma_0(k - 2\gamma T)x - \Gamma_0 k \gamma x^3. \quad (4.47)$$

It should be pointed out that from Eq. (4.47) one might at first sight conclude that there is a broken-symmetry state, when $k < 2\gamma T$, with $\langle x \rangle \neq 0$. But we know that this cannot happen because the stationary distribution is in fact the Boltzmann distribution. Therefore, one could get the wrong physics if one only looked at the ‘‘classical’’ trajectory, i.e., the solution to $\partial_t x = f(x)$, which maximizes the action S in the Ito convention. This shows again the importance of noise in these problems.

The unperturbed and perturbing actions are

$$\mathcal{S}_0 = \int dt [-D_0 \dot{x}^2 + \hat{x}(\partial_t x + \Gamma_0 k' x)], \quad (4.48)$$

$$\mathcal{S}_I = \Gamma_0 \gamma \int dt (kx^3 \hat{x} - Tx^2 \hat{x}^2), \quad (4.49)$$

where $k' \equiv k - 2\gamma T$. Introducing the state vector $x_\alpha = (\hat{x}, x)$, we can write \mathcal{S}_0 as

$$\begin{aligned}
\text{wavy line} \begin{array}{l} \diagup \\ \diagdown \end{array} &= +\Gamma_0 \gamma k x^3 \hat{x} \\
\text{wavy line} \begin{array}{l} \diagdown \\ \diagup \end{array} &= -\Gamma_0 T \gamma x^2 \hat{x}^2
\end{aligned}$$

FIG. 3. The two vertices corresponding to Eq. (4.49) for a particle diffusing in a spatially varying diffusion coefficient given by $D(x)=D_0(1+\gamma x^2)$, and confined in a harmonic potential.

$$S_0 = \frac{1}{2} \int \frac{d\omega}{2\pi} x_\alpha(\omega) G_{\alpha\beta}^{-1}(\omega) x_\beta(-\omega), \quad (4.50)$$

where

$$G_{\alpha\beta}^{0-1}(\omega) = \begin{pmatrix} -2D_0 & -i\omega + \Gamma_0 k' \\ i\omega + \Gamma_0 k' & 0 \end{pmatrix},$$

and thus

$$G_{\alpha\beta}^0(\omega) = \begin{pmatrix} 0 & \frac{1}{i\omega + \Gamma_0 k'} \\ \frac{1}{-i\omega + \Gamma_0 k'} & \frac{2D_0}{|-i\omega + \Gamma_0 k'|^2} \end{pmatrix},$$

from which we can read off the bare propagator and the zeroth-order correlation function:

$$\langle \hat{x}(\omega) \hat{x}(\omega') \rangle_0 = 0,$$

$$\langle \hat{x}(\omega) x(\omega') \rangle_0 = \frac{1}{i\omega + \Gamma_0 k'} \delta(\omega + \omega'),$$

$$\langle x(\omega) \hat{x}(\omega') \rangle_0 = \frac{1}{-i\omega + \Gamma_0 k'} \delta(\omega + \omega'),$$

$$\langle x(\omega) x(\omega') \rangle_0 = \frac{2D_0}{|-i\omega + \Gamma_0 k'|^2} \delta(\omega + \omega').$$

The interacting \mathcal{S}_I consists of two vertices that are depicted in Fig. 3. To second order in γ , the inverse of the propagator, $G^{-1}(\omega)$, can be written in frequency space as

$$G^{-1}(\omega) = -i\omega + \Gamma_0(k + \gamma T) + \Sigma(\omega). \quad (4.51)$$

The self-energies $\Sigma(\omega)$ are computed from the diagrams listed in Fig. 4 and it is given by

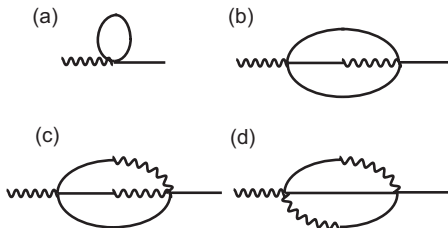


FIG. 4. Diagrams that contribute to the self-energy $\Sigma(\omega)$. Note that diagram (d) is identically zero.

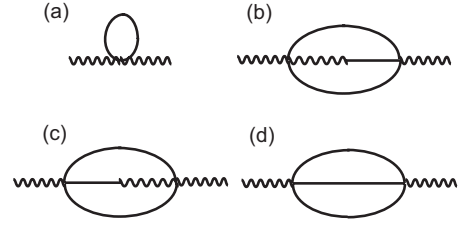


FIG. 5. Diagrams that contribute to the noise $D(\omega)$.

$$\Sigma(\omega) = 6k(\Gamma_0 \gamma)^2 [2TA(\omega) - 3kB(\omega)],$$

where

$$A(\omega) = \int \frac{d\omega_1}{2\pi} \int \frac{d\omega_2}{2\pi} G_0(\omega - \omega_1 - \omega_2) G_0(\omega_1) C_0(\omega_2),$$

$$B(\omega) = \int \frac{d\omega_1}{2\pi} \int \frac{d\omega_2}{2\pi} G_0(\omega - \omega_1 - \omega_2) C_0(\omega_1) C_0(\omega_2),$$

where $C_0(\omega) = \langle x(\omega)x(-\omega) \rangle_0$ is the zero-order correlation function. After some algebra, we obtain

$$G^{-1}(\omega) = -i\omega + \Gamma_0 k [1 + \gamma T/k - 6(\gamma T/k)^2 \mu(\omega/\Gamma_0 k)],$$

where $\mu(s) \equiv (-is + 3)^{-1}$.

Next, we turn to the correlation function $C(t, t') = \langle x(t)x(t') \rangle$, which can be written in the form $C(\omega) = 2D(\omega)|G(\omega)|^2$, with $D(\omega)$ computed from diagrams listed in Fig. 5 and it is given by

$$D(\omega) = D_0 \left(1 + \frac{\gamma T}{k} \right) + k\Gamma_0^2 \gamma^2 [3kE(\omega) - 8TH(\omega)],$$

$$E(\omega) = \int \frac{d\omega_1}{2\pi} \int \frac{d\omega_2}{2\pi} C_0(\omega - \omega_1 - \omega_2) C_0(\omega_1) C_0(\omega_2),$$

$$H(\omega) = \int \frac{d\omega_1}{2\pi} \int \frac{d\omega_2}{2\pi} C_0(\omega_1) C_0(\omega_2) [G_0(\omega - \omega_1 - \omega_2) + G_0(\omega_1 + \omega_2 - \omega)].$$

After some algebra, we find

$$D(\omega) = 2D_0 [1 + (\gamma T/k) - 6(\gamma T/k)^2 \text{Re } \mu(\omega/\Gamma_0 k)],$$

and we evaluate the correlation function

$$\begin{aligned}
C(t, 0) &= \frac{T}{k} \left(\frac{3\alpha_2 - \alpha_1(1 + \gamma T/k)}{\alpha_2^2 - \alpha_1^2} \right) e^{-\alpha_1 t} \\
&\quad + \frac{T}{k} \left(\frac{\alpha_2(1 + \gamma T/k) - 3\alpha_1}{\alpha_2^2 - \alpha_1^2} \right) e^{-\alpha_2 t}, \quad (4.52)
\end{aligned}$$

with decay rates

$$\alpha_1 \equiv \frac{\Gamma_0 k}{2} [4 + \gamma T/k - \sqrt{4 - 4\gamma T/k + 25(\gamma T/k)^2}],$$

$$\approx \Gamma_0 k [1 + \gamma T/k - 3(\gamma T/k)^2] \quad \text{for } \gamma T/k \ll 1,$$

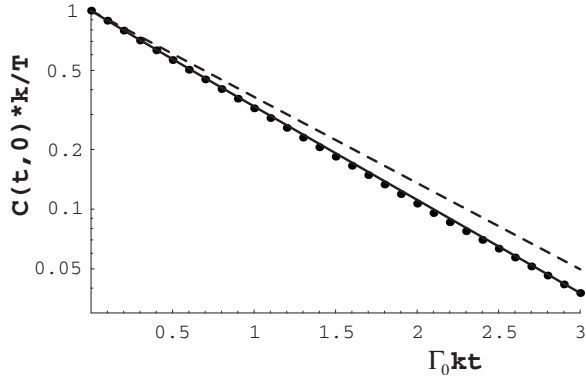


FIG. 6. Correlation function of x , $C(t,0)$, as a function of t for $\gamma T/k=0.15$. The solid line represents $C(t,0)$ as given by Eq. (4.52), which is calculated from the perturbation theory to second order in $\gamma T/k$. The data points are obtained from numerical simulation of the corresponding Langevin equation. Clearly, the result from the perturbation theory agrees excellently with the simulation. The dashed line represents the bare (zeroth-order) correlation function, which has a decaying rate that is different from the case of a spatially varying dissipative coefficient.

$$\alpha_2 \equiv \frac{\Gamma_0 k}{2} [4 + \gamma T/k + \sqrt{4 - 4\gamma T/k + 25(\gamma T/k)^2}],$$

$$\approx 3\Gamma_0 k [1 + (\gamma T/k)^2] \quad \text{for } \gamma T/k \ll 1.$$

Note that there are now two decaying modes with a fast mode α_2 and a slow mode α_1 in the system, in contrast to the case with uniform diffusion. Note also that $\langle x^2(0) \rangle = T/k$, as it should be. If we did not put in the extra drift term gg' in $f(x)$, this relation would not hold. In fact, it would have been $\langle x^2(0) \rangle = T/(k - \gamma T)$, which violates the equipartition theorem. This is yet another demonstration that this extra drift term gg' is needed to ensure the correct thermodynamic properties. In Fig. 6, we plot the correlation function in Eq. (4.52) and the numerical simulation of the Langevin equation describing this system for $\gamma T/k=0.15$. Clearly, the second-order perturbation theory agrees very well with the simulation. Note, however, that when $\gamma T/k \sim 1$, α_1 becomes negative, signaling the breakdown of perturbation theory.

Finally, we demonstrate the FDT to second order in perturbation theory. The physical response function is given by

$$\begin{aligned} \chi_{xx}(t,t') &= \Gamma_0 \langle x(t) \hat{x}(t') [1 + \gamma x^2(t')] e^{-S_I} \rangle_0 \\ &= \Gamma_0 G(t,t') + \Gamma_0 \gamma G_0(t,t') C_0(0) \\ &\quad - \Gamma_0 \gamma \langle x(t) \hat{x}(t') x^2(t') S_I \rangle_0, \end{aligned}$$

which corresponds to the diagrams in Fig. 7. We find

$$\chi_{xx}(\omega) = \Gamma_0 G(\omega) [1 + \gamma T/k - 6(\gamma T/k)^2 \mu(\omega/\Gamma_0 k)]. \quad (4.53)$$

Note that this clearly shows that the physical response function and the propagator are different. Taking the imaginary part of Eq. (4.53), it can be easily verified that the fluctuation-dissipation theorem Eq. (4.43) is indeed satisfied to second order in perturbation theory.

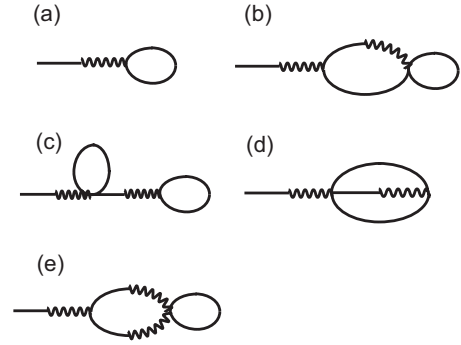


FIG. 7. Diagrams that contribute to the physical response function $\chi_{xx}(\omega)$.

V. N-COMPONENT LANGEVIN EQUATION

Many physical problems involve more than one variable, and some of the issues we have addressed so far may not apply to higher-dimensional systems. For example, the drift term $f(x)$ in one dimension (1D) can always be written as a derivative of another function, i.e., 1D systems are conservative; however, for higher-dimensional systems, this may not be true. A complete analysis of higher-dimensional systems requires a separate publication. Here, we briefly discuss the Fokker-Planck equation and the path integral in the α convention for a multidimensional Langevin equation of the form

$$\partial_t x_i = f_i(x_1, \dots, x_N) + g_{ij}(x_1, \dots, x_N) \eta_j(t), \quad (5.1)$$

where $\eta_j(t)$ are the noises, with zero mean and correlation given by

$$\langle \eta_i(t) \eta_j(t') \rangle = \delta_{ij} \delta(t - t'). \quad (5.2)$$

In Eq. (5.1) and the following, Einstein summation is assumed. We focus on the case where the system is near thermal equilibrium and address, as we did in the 1D case, how the Boltzmann distribution determines the form of f_i and g_{ij} in the α convention. The Fokker-Planck equation corresponding to Eq. (5.1) can be derived following the same procedure as outlined in Sec. II C. In the α convention, we find

$$\frac{\partial \mathcal{P}}{\partial t} = \frac{\partial}{\partial x_i} \left[- \left(f_i + \alpha \frac{\partial g_{il}}{\partial x_k} g_{kl} \right) \mathcal{P} + \frac{1}{2} \frac{\partial}{\partial x_j} (g_{il} g_{jl} \mathcal{P}) \right], \quad (5.3)$$

where $\mathcal{P}[\{x\}, t]$ is the joint probability distribution of x_i at time t . If there are only dissipative terms and no reactive terms in f_i , the constraint that \mathcal{P} reach a long-time state of thermal equilibrium value proportional to $\exp(-\beta \mathcal{H})$ requires that

$$f_i(\{x\}) = \frac{1}{2} \frac{\partial}{\partial x_j} (g_{ik} g_{jk}) - \alpha \frac{\partial g_{ij}}{\partial x_l} g_{lj} - \frac{1}{2} \beta g_{ik} g_{jk} \frac{\partial \mathcal{H}}{\partial x_j}, \quad (5.4)$$

in order that Eq. (5.3) reduce to

$$\frac{\partial \mathcal{P}}{\partial t} = \frac{\partial}{\partial x_i} \left(\frac{1}{2} g_{il} g_{jl} \right) \left(\beta \frac{\partial \mathcal{H}}{\partial x_j} \mathcal{P} + \frac{\partial \mathcal{P}}{\partial x_j} \right), \quad (5.5)$$

with the steady state solution $\mathcal{P} \propto \exp(-\beta \mathcal{H})$.

The diffusion matrix is defined as

$$D_{ij}(\{x\}) = \frac{1}{2} g_{il} g_{jl} = k_B T \Gamma_{ij}(\{x\}), \quad (5.6)$$

where $\Gamma_{ij}(\{x\})$ is the matrix of dissipative coefficients. Thus,

$$f_i(\{x\}) = \frac{\partial D_{ij}(\{x\})}{\partial x_j} - \alpha \frac{\partial g_{ij}}{\partial x_l} g_{lj} - \Gamma_{ij}(\{x\}) \frac{\partial \mathcal{H}}{\partial x_j}. \quad (5.7)$$

Note that, since the diffusion matrix $D_{ij}(\{x\})$ is symmetric with respect to $i \leftrightarrow j$, it has only $N(N+1)/2$ independent entries, and we may impose $N^2 - N(N+1)/2 = N(N-1)/2$ constraints on $g_{ij}(\{x\})$ without sacrificing the physical content. We could, for example chose g_{ij} to be symmetric, in which case it is simply the square root of $k_B T \Gamma_{ij}$.

To derive the path integral, we first discretize Eq. (5.1) as

$$x_i^{(n)} = x_i^{(n-1)} + \Delta t f_i^{(n)} + g_{ij}^{(n)} \int_{t_{n-1}}^{t_n} ds \eta_j(s), \quad (5.8)$$

and introduce

$$h_i[\{x_k^{(n)}\}, \{x_k^{(n-1)}\}] \equiv [g_{ij}^{(n)}]^{-1} (x_j^{(n)} - x_j^{(n-1)} - \Delta t f_j^{(n)}) - \int_{t_{n-1}}^{t_n} ds \eta_i(s), \quad (5.9)$$

where $f_i^{(n)} = f_i[\{\alpha x_i^{(n)} + (1-\alpha)x_i^{(n-1)}\}]$ and $g_{ij}^{(n)} = g_{ij}[\{\alpha x_i^{(n)} + (1-\alpha)x_i^{(n-1)}\}]$. Following the basic steps as outlined in Sec. IV, we can write the conditional probability as

$$\mathcal{P}(\{x_k^{(n)}\}_{t_n} | \{x_k^{(n-1)}\}_{t_{n-1}}) = \det \left| \frac{\partial h_i}{\partial x_k^{(n)}} \right| \times \left\langle \prod_{i=1}^N \delta[h_i(\{x_k^{(n)}\}, \{x_k^{(n-1)}\})] \right\rangle. \quad (5.10)$$

Taking the derivative of h_i explicitly, we find

$$\begin{aligned} \mathcal{P}(\{x_k^{(n)}\}_{t_n} | \{x_k^{(n-1)}\}_{t_{n-1}}) &= \prod_{i=1}^N \int \frac{dk_i}{2\pi \det g_{ij}} \left(1 - \alpha \Delta t \partial_l f_l + \alpha k_m \partial_l g_{lm} \Delta t + \frac{\alpha^2}{2} \Delta t (\partial_k g_{lm} \partial_l g_{km} - \partial_l g_{lm} \partial_k g_{km}) \right) e^{i k_i g_{ij}^{-1} (\Delta x_j - \Delta t f_j) - k_i^2 \Delta t / 2}, \\ &= \int \frac{dy_k}{2\pi} e^{+i y_k (\Delta x_k - f_k \Delta t + \alpha g_{kj} \partial_l g_{lj} \Delta t) - g_{lk} g_{kj} y_l y_j / 2 - \alpha \Delta t \partial_l f_l - (\alpha^2 / 2) (\partial_k g_{lm} \partial_l g_{km} - \partial_l g_{lm} \partial_k g_{km}) \Delta t}, \end{aligned} \quad (5.14)$$

where, in the last line, we have exponentiated the terms in the parentheses and substituted $y_i = g_{ij} k_j$. In the continuum limit, we have

$$\begin{aligned} \mathcal{P}(\{x_k^{(f)}\}_{t_f} | \{x_k^{(0)}\}_{t_0}) &= \int_{x_k^{(0)}}^{x_k^{(f)}} \mathcal{D}x_k \int \mathcal{D}y_k e^{-\mathcal{S}}, \\ \mathcal{S} &= \int dt \left(\frac{1}{2} g_{lk} g_{jk} y_l y_j - i y_k (\partial_t x_k - f_k + \alpha g_{kj} \partial_l g_{lj}) + \alpha \partial_l f_l + \frac{\alpha^2}{2} (\partial_k g_{lm} \partial_l g_{km} - \partial_l g_{lm} \partial_k g_{km}) \right). \end{aligned} \quad (5.15)$$

$$\frac{\partial h_i}{\partial x_k^{(n)}} = g_{il}^{-1} (\delta_{lk} - \mathcal{M}_{lk}),$$

where we have defined the matrix \mathcal{M}_{lk} by

$$\mathcal{M}_{lk} \equiv \alpha \Delta t \frac{\partial f_l}{\partial x_k} + \alpha \frac{\partial g_{lm}}{\partial x_k} g_{ij}^{-1} (x_j^{(n)} - x_j^{(n-1)} - \Delta t f_j^{(n)}). \quad (5.11)$$

Using the identity $\det \hat{X} = \exp \text{Tr} \hat{X}$, the determinant can be evaluated to give

$$\det \left| \frac{\partial h_i}{\partial x_k^{(n)}} \right| = \frac{1}{\det g_{ij}} \left(1 - \mathcal{M}_{ll} + \frac{1}{2} (\mathcal{M}_{ll} \mathcal{M}_{kk} - \mathcal{M}_{lk} \mathcal{M}_{kl}) + \dots \right). \quad (5.12)$$

Therefore, the conditional probability can be written as

$$\begin{aligned} \mathcal{P}(\{x_k^{(n)}\}_{t_n} | \{x_k^{(n-1)}\}_{t_{n-1}}) &= \prod_{i=1}^N \int \frac{dk_i}{2\pi \det g_{ij}} \left(1 - \mathcal{M}_{ll} \right. \\ &\quad \left. + \frac{1}{2} (\mathcal{M}_{ll} \mathcal{M}_{kk} - \mathcal{M}_{lk} \mathcal{M}_{kl}) \right) \\ &\quad \times e^{i k_i g_{ij}^{-1} (\Delta x_j - \Delta t f_j) - k_i^2 \Delta t / 2}, \end{aligned} \quad (5.13)$$

where we have only kept terms up to order Δt . Following a procedure similar to that leading to Eq. (4.11) for the 1D case, we find

Note that there is an extra term proportional to α^2 . This term is identically zero for a 1D system. If we start with Eq. (5.14) and follow the procedure as outlined in Sec. IV A, we can show that the path integral is equivalent to the Fokker-Planck equation in Eq. (5.3).

VI. CONCLUSION

In this paper, we have examined a thermodynamically consistent Langevin formulation of the Brownian motion with a diffusion coefficient that depends on space. We argue, in particular, that the requirement that the Boltzmann distribution be reached in equilibrium determines the interpretation of stochastic integrals arising from multiplicative noise in the Langevin equation. We hope that this paper clarifies some of the confusion over these stochastic issues that have persisted for some time. We have also constructed path integral representations of the Langevin equations with multiplicative noise, and we used this representation as a starting point for the development of a systematic perturbation theory. Such a formulation can be employed to treat nonlinear stochastic equations arising from a variety of problems. Future work includes generalizing this formulism to “fields” and examines how state-dependent dissipative coefficients may give rise to long-time tails and corrections to scaling in dynamic critical phenomena. Of course, one of the most interesting open questions is whether there is an equivalent criterion for systems that are driven far from equilibrium.

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APPENDIX A: CONNECTION BETWEEN α AND $\theta(0)$

In this appendix, we outline the connection between α and the Heaviside unit step function $\theta(t)$ evaluated at $t=0$. For simplicity, we set $f(x)=0$ in Eq. (2.10):

$$\partial_t x = g(x) \eta(t). \quad (\text{A1})$$

Using the α -convention rule Eq. (2.8), we have for $\Delta x(t) \equiv x(t+\Delta t) - x(t)$

$$\begin{aligned} \Delta x(t) &= g[x(t) + \alpha \Delta x] \int_t^{t+\Delta t} ds \eta(s) \\ &= g[x(t)] \int_t^{t+\Delta t} ds \eta(s) + \alpha g'[x(t)] \Delta x(t) \int_t^{t+\Delta t} ds \eta(s) \\ &\quad + \dots \\ &= g[x(t)] \int_t^{t+\Delta t} ds \eta(s) \\ &\quad + \alpha g g' \int_t^{t+\Delta t} ds \int_t^{t+\Delta t} ds' \eta(s) \eta(s') + \dots \end{aligned}$$

Therefore, the average $\Delta x(t)$ over the noise is

$$\langle \Delta x(t) \rangle = \alpha g g' \int_t^{t+\Delta t} ds \int_t^{t+\Delta t} ds' \delta(s-s'). \quad (\text{A2})$$

The integral

$$\int_{-\infty}^t ds' \delta(s-s') = \begin{cases} 0, & s > t \\ 1, & s < t \end{cases} \quad (\text{A3})$$

$$= \theta(t-s) \quad (\text{A4})$$

defines the Heaviside unit step function $\theta(t)$, and Eq. (A2) becomes

$$\langle \Delta x(t) \rangle = \alpha g g' \int_t^{t+\Delta t} ds [\theta(t+\Delta t-s) - \theta(t-s)] = \alpha g g' \Delta t. \quad (\text{A5})$$

On the other hand, we can directly integrate Eq. (A1) to obtain

$$\Delta x(t) = \int_t^{t+\Delta t} ds g[x(s)] \eta(s). \quad (\text{A6})$$

Expanding $g[x(s)]$ as

$$g[x(s)] = g[x(t) + \Delta x(s)] = g[x(t)] + g'[x(t)] \Delta x(s) + \dots, \quad (\text{A7})$$

we find

$$\Delta x(t) = g \int_t^{t+\Delta t} \eta(s) + g g' \int_t^{t+\Delta t} ds \int_t^s ds' \eta(s) \eta(s'). \quad (\text{A8})$$

Note that the upper limit of integration for s' is different from that in Eq. (A2). Therefore, we find

$$\langle \Delta x(t) \rangle = \theta(0) g g' \Delta t. \quad (\text{A9})$$

Comparing this with Eq. (A5), we conclude that $\theta(0) = \alpha$.

APPENDIX B: EQUIVALENCE BETWEEN EQ. (4.7) AND EQ. (4.11)

In this appendix, we demonstrate that the conditional probability given in Eq. (4.7) and its exponentiated form Eq. (4.11) are indeed equivalent. Note that the latter expression involves a subtle step which is required for the construction of the path integral in Sec. IV. Therefore, it is crucial to confirm that Eq. (4.11) is correct at least to order of Δt . We have already checked that the correct Fokker-Planck equation, Eq. (2.17), can be derived from Eq. (4.11) in Sec. IV A. Here, we check that the normalization condition

$$\int dx_i P(x_i t_i | x_{i-1} t_{i-1}) = 1 \quad (\text{B1})$$

for general α is satisfied by Eq. (4.11). First, let us check that Eq. (B1) is true for Eq. (4.7). For simplicity, we set $f(x)=0$. We have

$$\int dx_i P(x_i t_i | x_{i-1} t_{i-1}) = \int \frac{d\Delta x_i}{\sqrt{2\pi\Delta t g_i}} e^{-(\Delta t/2g_i^2)(\Delta x_i/\Delta t)^2} \left(1 - \alpha \frac{g'_i}{g_i} \Delta x_i\right) = \int \frac{dw}{\sqrt{2\pi g_i}} \left(1 - \alpha \frac{g'_i}{g_i} \sqrt{\Delta t} w\right) e^{-w^2/2g_i^2}, \quad (\text{B2})$$

where we have made a change of variable: $\Delta x_i = \sqrt{\Delta t} w$. Remembering that $g_i = g(x_{i-1} + \alpha\sqrt{\Delta t} w)$ and expanding them in Eq. (B2) in powers of Δt , we have

$$\int dx_i P(x_i t_i | x_{i-1} t_{i-1}) = \int \frac{dw}{\sqrt{2\pi g}} e^{-w^2/2g^2} \left[1 - \frac{2\alpha g'}{g} \sqrt{\Delta t} w + \left(\frac{3\alpha^2 g'^2}{g^2} - \frac{3\alpha^2 g''}{2g} \right) \Delta t w^2 + \frac{\alpha g'}{g^3} \sqrt{\Delta t} w^3 + \left(\frac{\alpha^2 g''}{2g^3} - \frac{7\alpha^2 g'^2}{2g^4} \right) w^4 + \frac{\alpha^2 g'^2}{2g^6} \Delta t w^6 + \dots \right] \quad (\text{B3})$$

$$= 1 + O(\Delta t^{3/2}), \quad (\text{B4})$$

where $g \equiv g(x_{i-1})$. Thus, Eq. (4.7) indeed satisfies the normalization condition, which is hardly surprising since it must be true by construction. Now, let us check the exponentiated form, Eq. (4.11). We have

$$\int dx_i P(x_i t_i | x_{i-1} t_{i-1}) = \int \frac{d\Delta x_i}{\sqrt{2\pi\Delta t g_i}} e^{-(\Delta t/2g_i^2)(\Delta x_i/\Delta t + \alpha g_i g'_i)^2} = \int \frac{dw}{\sqrt{2\pi g_i}} e^{-(\Delta t/2g_i^2)(w/\sqrt{\Delta t} + \alpha g_i g'_i)^2} = \int \frac{dw}{\sqrt{2\pi g}} e^{-w^2/2g^2} \left[1 - \frac{1}{2} \alpha^2 g'^2 - \frac{2\alpha g'}{g} \sqrt{\Delta t} w + \left(\frac{7\alpha^2 g'^2}{2g^2} - \frac{3\alpha^2 g''}{2g} \right) \Delta t w^2 + \frac{\alpha g'}{g^3} \sqrt{\Delta t} w^3 + \left(\frac{\alpha^2 g''}{2g^3} - \frac{7\alpha^2 g'^2}{2g^4} \right) w^4 + \frac{\alpha^2 g'^2}{2g^6} \Delta t w^6 + \dots \right] \quad (\text{B5})$$

$$= 1 + O(\Delta t^{3/2}). \quad (\text{B6})$$

Thus, Eq. (4.11) also satisfies the normalization condition. We note in passing that, although the expansions Eqs. (B3) and (B5) are different, they both give one the lowest order, and the next-order term is of the order $\Delta t^{3/2}$.

APPENDIX C: α -DEPENDENT PERTURBATION THEORY FOR THE MODEL SYSTEM WITH $D(x) = D_0(1 + \gamma x^2)$

In this appendix, we carry out a first-order perturbation calculation for general α of the model system studied in Sec. IV C in which $D(x) = D_0(1 + \gamma x^2)$, in order to clarify some subtle issues associated with the α convention. It is straightforward to work out the action in Eq. (4.15). Up to an irrelevant constant, we have

$$\mathcal{S}_0 = \int dt [-D_0 \dot{x}^2 + \hat{x}(\partial_t x + \Gamma_0 kx)]$$

$$\mathcal{S}_I = \Gamma_0 \gamma \int dt \left(2(2\alpha - 1)x\dot{x} + \frac{k}{T} x^3 \dot{x} - x^2 \dot{x}^2 - 3\alpha \frac{k}{T} x^2 \right).$$

Note that there are more diagrams to evaluate than there are for $\alpha=0$. Consider first the propagator $G(t, t') = \langle x(t) \hat{x}(t') \rangle$,



FIG. 8. Diagrams that contribute to the self-energy $\Sigma(\omega)$.

which can be written as $G^{-1}(\omega) = G_0^{-1}(\omega) + \Sigma(\omega)$, where the diagrams for the self-energy are displayed in Fig. 8. Note that the closed loop diagram (c) in Fig. 8 contains $G_0(t=0)$, which must be set to α as explained in Sec. IV B. We find

$$\Sigma(\omega) = 2(2\alpha - 1)D_0\gamma + 3D_0\gamma - 4D_0\gamma G_0(0) = D_0\gamma,$$

which agrees with the calculation for $\alpha=0$. Note that the final result is independent of α , as it should be. To first order, the noise $D(\omega)$ renormalizes exactly the same way as in the $\alpha=0$ calculation. However, the physical response function is different. It is given by Eq. (4.41) with an extra α -dependent term:

$$\begin{aligned} \chi_{xx}(t, t') &= \Gamma_0 \langle x(t) \hat{x}(t') [1 + \gamma x^2(t')] e^{-S_I} \rangle_0 \\ &\quad - \frac{\alpha}{T} \langle x(t) [2\gamma D_0 x(t')] e^{-S_I} \rangle_0 \\ &= \Gamma_0 \left(1 + \frac{\gamma T}{k} \right) G(t, t') + 2\alpha \Gamma_0 \gamma C_0(t, t') \\ &\quad - 2\alpha \Gamma_0 \gamma C_0(t, t') + O(\gamma^2) \\ &= \Gamma_0 \left(1 + \frac{\gamma T}{k} \right) G(t, t'). \end{aligned}$$

Without the cancellation of the α -dependent terms, $\chi_{xx}(t, t')$ would not have been causal.

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- $$x_{n+1} = x_n + f(x_n)h + g(x_n)\Delta W_n + \frac{1}{2}g(x_n)g'(x_n)[(\Delta W_n)^2 - h],$$
- where h is the time step, and ΔW_n is a Gaussian-distributed random variable with zero mean and variance of $\sigma^2=h$. See, for example, P. E. Kloeden and E. Platen, *Numerical Solution of Stochastic Differential Equations* (Springer, New York, 1995).
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