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Generalized quantum Fokker-Planck, diffusion, and Smoluchowski equations with true probability distribution functions

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Traditionally, quantum Brownian motion is described by Fokker-Planck or diffusion equations in terms of quasiprobability distribution functions, e.g., Wigner functions. These often become singular or negative in the full quantum regime. In this paper a simple approach to non-Markovian theory of quantum Brownian motion using true probability distribution functions is presented. Based on an initial coherent state representation of the bath oscillators and an equilibrium canonical distribution of the quantum mechanical mean values of their coordinates and momenta, we derive a generalized quantum Langevin equation in *c* numbers and show that the latter is amenable to a theoretical analysis in terms of the classical theory of non-Markovian dynamics. The corresponding Fokker-Planck, diffusion, and Smoluchowski equations are the exact quantum analogs of their classical counterparts. The present work is independent of path integral techniques. The theory as developed here is a natural extension of its classical version and is valid for arbitrary temperature and friction (the Smoluchowski equation being considered in the overdamped limit).

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I. INTRODUCTION

A model quantum system coupled to its environment forms the standard paradigm of quantum Brownian motion. The initiation of early development of this stochastic process took place around the middle of the 20th century [1–3]. A major impetus was the discovery of lasers in the 1960s followed by significant advances in the fields of quantum optics and laser physics in the 1970s, where extensive applications of nonequilibrium quantum statistical methods were made. Various nonlinear optical processes/phenomena were described with the help of operator Langevin equations, density operator methods, and the associated quasi-classical distribution functions of Wigner, Glauber, Sudarshan, and others centering around the quantum Markov processes [1–6]. Subsequent to this early development the quantum theory of Brownian motion again emerged as a subject of immense interest in the early 1980s when the problem of macroscopic quantum tunneling was addressed by Leggett and others [7–11], and almost simultaneously the quantum Kramers problem attracted the serious attention of a number of workers [12–15]. The method that received major attention in the 1980s and 1990s in a wide community of physicists and chemists in these studies is the real time functional integral [16,17]. This method has been shown to be an effective tool for treatment of quantum transition states [18], and dissipative quantum coherence effects [8,19] as well as incoherent quantum tunneling processes [13,14,20] and many related problems [21].

In spite of this phenomenal success it may, however, be noted that compared to the classical theory the quantum theory of Brownian motion based on functional integrals rests on a fundamentally different footing. While the classical theory is based on the differential equations for evolution of the *true* probability density functions of the particle executing Brownian motion, the path integral methods rely on a noncanonical quantization procedure and the evaluation of the quantum partition function of the particle interacting with the heat bath, and one is, in general, led to the time evolution equations of quasiprobability distribution functions such as Wigner functions [15,22–26]. The question is whether there is any natural extension of the classical method to the quantum domain in terms of true probability distribution functions. It is therefore worthwhile to seek for a natural extension of the classical theory of Brownian motion to the quantum domain in the non-Markovian regime for arbitrary friction and temperature within the framework of a well-behaved true probabilistic description. Our aim in this paper is thus twofold: (i) To enquire whether there exists a quantum generalized Langevin equation (QGLE) in *c* numbers whose noise correlation satisfies the quantum fluctuation-dissipation relation (FDR) but which (the QGLE) at the same time is a natural analog of its classical counterpart; (ii) to formulate exact quantum Fokker-Planck and diffusion equations which are valid for arbitrary temperature and friction. We also intend to look for the overdamped limit to obtain the exact quantum analog of the classical Smoluchowski equation.

Before proceeding further it is important to stress the motivation for the present scheme.

(1) As we have already pointed out, the traditional theories of quantum Brownian motion in optics [1–5] and condensed matter physics [7] are based on quasiprobability functions. Apart from the usual shortcomings that they may become negative or singular [27] in the full quantum regime when the potential is nonlinear, the quasiprobability functions are, in general, not valid for non-Markovian processes with arbitrary noise correlation. While in the majority of quantum optical situations a Markovian description is sufficient, non-Markovian effects of noise correlation are strongly

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felt in the problems of quantum dissipation in condensed matter and chemical physics at low temperature. To include these effects even in the case of a free particle (see, for example, Ref. [11]) one has to use a suitable cutoff frequency of the heat bath to avoid intrinsic low frequency divergence. Clearly this poses serious difficulties for studying transient behavior for arbitrary noise correlation and temperature. In what follows we show that the present treatment is free from such difficulties.

(2) Our second motivation is to understand the quantum-classical correspondence in the problem of Brownian motion in a transparent way. To this end we note that in the classical theory the Fokker-Planck equation with nonlinear potential contains derivatives of probability distribution functions up to second order. The equations in terms of Wigner functions, on the other hand, involve higher (than two) order derivatives of distribution functions in the corresponding quantum formulation [28]. The higher derivative terms contain powers of $\hbar$ and derivatives of the potential signifying purely quantum diffusion in which quantum corrections and nonlinearity of the potential get entangled in the description of the system. Because of the occurrence of higher derivatives, the positivity of the distribution function is never ensured and the equation cannot be treated as a quantum analog of the classical Fokker-Planck equation. Any attempt to reduce the order of the derivatives to two amounts to a semiclassical approximation. Again, there exists no systematic procedure for this reduction. Keeping in view these problems we intend to derive exact quantum analog [Eqs. (42), (47), and (50)] of the classical Fokker-Planck, diffusion, and Smoluchowski equations, respectively, in terms of *true probability distribution functions*, where the equations contain derivatives of distribution functions up to second order only, for which the diffusion coefficients are positive definite. Since the equations are classical looking in form but quantum mechanical in their content, one can read the quantum drift and diffusion coefficients and also construct the quantum corrections due to the nonlinearity of the system systematically order by order in a straightforward way, so that the quantum-classical correspondence can be checked simply by taking the limit $\hbar\to 0$ both in the Markovian and in the non-Markovian description. We mention in passing that, in contrast to a recent treatment [29] of the large friction limit in a similar context, the quantum Smoluchowski equation as discussed here retains its validity in the full quantum regime as $T\to 0$.

(3) Since over the last two decades classical non-Markovian theories [30,31] and numerical methods of generating classical noise processes have made significant progress [32–34], the mapping of the quantum theory of Brownian motion into a classical form, as achieved here, suggests that the classical treatment can be extended to the quantum domain without much difficulty. Since the present scheme describes the generation of quantum noise [Eqs. (10) and (11)] as classical numbers which follow the quantum fluctuation-dissipation relation, it is easy to comprehend that the classical numerical techniques for the generation of noise and solving the stochastic Langevin equation [32–34] can be utilized in the present case in a straightforward way to solve the quantum Langevin equation [35]. The procedure is therefore much easier to implement compared to other methods, like path integral Monte Carlo techniques [36].

In what follows we consider the standard system-reservoir model and make use of the coherent state representation of the bath oscillators to derive a GLE for the quantum mechanical mean value of position of a particle in contact with a thermal bath whose quantum mechanical properties can be defined in terms of a classical-looking noise term and a canonical distribution of initial quantum mechanical mean values of the coordinates and momenta of the bath. This simple approach allows us to show that, although the equation is essentially quantum mechanical it is amenable to a theoretical analysis in terms of the classical theory of non-Markovian dynamics [30,31].

The rest of the paper is organized as follows. The system-reservoir model, the associated QGLE, and the canonical distribution for the bath oscillators are introduced in Sec. II. This is followed by a general analysis of the QGLE in the Sec. III and an illustration with an exponential memory kernel in Sec. IV to calculate the variances required for setting up a quantum Fokker-Planck equation and a quantum diffusion equation in Secs. V and VI, repectively. Section VII is devoted to the quantum overdamped limit and the Smoluchowski equation. The paper is summarized and concluded in Sec. VIII.

II. THE QUANTUM GENERALIZED LANGEVIN EQUATION IN *c* NUMBERS

We consider a particle in a medium. The latter is modeled as a set of harmonic oscillators with frequency $\{\omega_i\}$. The evolution of such a quantum open system has been studied over the last several decades under a variety of reasonable assumptions. Specifically, our interest here is to develop an exact description of quantum Brownian motion within the purview of this model described by the following Hamiltonian [37]:

$$\hat{H}=\frac{\hat{p}^2}{2}+V(\hat{x})+\sum_j\left[\frac{\hat{p}_j^2}{2}+\frac{1}{2}\kappa_j(\hat{q}_j-\hat{x})^2\right]. \tag{1}$$

Here $\hat{x}$ and $\hat{p}$ are the coordinate and momentum operators of the particle and the set $\{\hat{q}_j,\hat{p}_j\}$ is the set of coordinate and momentum operators for the reservoir oscillators coupled linearly to the system through their coupling coefficients κ_j. The potential $V(\hat{x})$ is due to the external force field for the Brownian particle. The coordinate and momentum operators follow the usual commutation relation $[\hat{x},\hat{p}]=i\hbar$ and $[\hat{q}_j,\hat{p}_j]=i\hbar\delta_{ij}$. Note that in writing down the Hamiltonian no rotating wave approximation has been used.

Eliminating the reservoir degrees of freedom in the usual way [1,38–40], we obtain the operator Langevin equation for the particle,

$$\ddot{\hat{x}}(t)+\int_0^t dt'\ \gamma(t-t')\dot{\hat{x}}(t')+V'(\hat{x})=\hat{F}(t), \tag{2}$$

where the noise operator $\hat{F}(t)$ and the memory kernel $\gamma(t)$ are given by

$$\hat{F}(t)=\sum_j [\{\hat{q}_j(0)-\hat{x}(0)\}\kappa_j \cos\omega_j t + \hat{p}_j(0)\kappa_j^{1/2}\sin\omega_j t] \tag{3}$$

and

$$\gamma(t)=\sum_j \kappa_j \cos\omega_j t, \tag{4}$$

with $\kappa_j=\omega_j^2$ (the masses have been assumed to be unity).

Equation (2) is an exact quantized operator Langevin equation which is now standard textbook material [1,4] and for which the noise properties of $\hat{F}(t)$ can be defined using a suitable initial canonical distribution of the bath coordinates and momenta. Our aim here is to replace it by an equivalent QGLE in c numbers. Again, this is not a new problem so long as one is restricted to standard quasiprobabilistic methods using, for example, Wigner functions [15,22–26]. To address the problem of quantum non-Markovian dynamics in terms of a true probabilistic description, however, we follow a different procedure. We first carry out the quantum mechanical average of Eq. (2):

$$\langle\ddot{\hat{x}}(t)\rangle+\int_0^t dt'\ \gamma(t-t')\langle\dot{\hat{x}}(t')\rangle+\langle V'(\hat{x})\rangle=\langle\hat{F}(t)\rangle, \tag{5}$$

where the average $\langle\cdots\rangle$ is taken over the initial product separable quantum states of the particle and the bath oscillators at $t=0$, $|\phi\rangle\{|\alpha_1\rangle|\alpha_2\rangle\cdots|\alpha_N\rangle\}$. Here $|\phi\rangle$ denotes any arbitrary initial state of the particle and $|\alpha_i\rangle$ corresponds to the initial coherent state of the ith bath oscillator. $|\alpha_i\rangle$ is given by $|\alpha_i\rangle=\exp(-|\alpha_i|^2/2)\Sigma_{n_i=0}^{\infty}(\alpha_i^{n_i}/\sqrt{n_i!})|n_i\rangle$, α_i being expressed in terms of the mean values of the coordinate and momentum of the ith oscillator, $\langle\hat{q}_i(0)\rangle=(\sqrt{\hbar}/2\omega_i)(\alpha_i+\alpha_i^\star)$ and $\langle\hat{p}_i(0)\rangle=i\sqrt{\hbar\omega_i/2}(\alpha_i^\star-\alpha_i)$, respectively. It is important to note that $\langle\hat{F}(t)\rangle$ of Eq. (5) is a classical-like noise term which, in general, is a nonzero number because of the quantum mechanical averaging over the coordinate and momentum operators of the bath oscillators with respect to the initial coherent states and arbitrary initial state of the particle, and is given by

$$\langle\hat{F}(t)\rangle=\sum_j [\{\langle\hat{q}_j(0)\rangle-\langle\hat{x}(0)\rangle\}\kappa_j\cos\omega_j t + \langle\hat{p}_j(0)\rangle\kappa_j^{1/2}\sin\omega_j t]. \tag{6}$$

It is convenient to rewrite the c-number equation (5) as follows:

$$\langle\ddot{\hat{x}}(t)\rangle+\int_0^t dt'\ \gamma(t-t')\langle\dot{\hat{x}}(t')\rangle+\langle V'(\hat{x})\rangle=F(t) \tag{7}$$

where we let the quantum mechanical mean value $\langle\hat{F}(t)\rangle=F(t)$. We now turn to the second averaging. To realize $F(t)$ as an effective c-number noise we now assume that the momenta $\langle\hat{p}_j(0)\rangle$ and the shifted coordinates $\{\langle\hat{q}_j(0)\rangle-\langle\hat{x}(0)\rangle\}$ of the bath oscillators are distributed according to a canonical distribution of Gaussian forms as

$$\mathcal{P}_j=\mathcal{N}\exp\left\{\frac{-[\langle\hat{p}_j(0)\rangle^2+\kappa_j\{\langle\hat{q}_j(0)\rangle-\langle\hat{x}(0)\rangle\}^2]}{2\hbar\omega_j\left(\overline{n}_j+\frac{1}{2}\right)}\right\} \tag{8}$$

so that for any quantum mechanical mean value $O_j(\langle\hat{p}_j(0)\rangle,\{\langle\hat{q}_j(0)\rangle-\langle\hat{x}(0)\rangle\})$ the statistical average $\langle\cdots\rangle_S$ is

$$\langle O_j\rangle_S=\int O_j(\langle\hat{p}_j(0)\rangle,\{\langle\hat{q}_j(0)\rangle-\langle\hat{x}(0)\rangle\}) \times\mathcal{P}_j(\langle\hat{p}_j(0)\rangle,\{\langle\hat{q}_j(0)\rangle-\langle\hat{x}(0)\rangle\}) \times d\langle\hat{p}_j(0)\rangle d\{\langle\hat{q}_j(0)\rangle-\langle\hat{x}(0)\rangle\}. \tag{9}$$

Here $\overline{n}_j$ indicates the average thermal photon number of the jth oscillator at temperature T, $\overline{n}_j=1/[\exp(\hbar\omega_j/k_BT)-1]$, and $\mathcal{N}$ is the normalization constant.

The distribution (8) and the definition of the statistical average (9) imply that $F(t)$ must satisfy

$$\langle F(t)\rangle_S=0 \tag{10}$$

and

$$\langle F(t)F(t')\rangle_S=\frac{1}{2}\sum_j \kappa_j\hbar\omega_j\left(\coth\frac{\hbar\omega_j}{2k_BT}\right) \times\cos\omega_j(t-t'). \tag{11}$$

That is, the c-number noise $F(t)$ is such that it is zero centered and satisfies the standard quantum fluctuation-dissipation relation as known in the literature [38] in terms of the quantum statistical average of the noise operators.

To proceed further we now add the force term $V'(\langle\hat{x}\rangle)$ on both sides of Eq. (7) and rearrange it to obtain formally

$$\ddot{X}(t)+\int_0^t dt'\ \gamma(t-t')\dot{X}(t')+V'(X)=F(t)+Q(X,t), \tag{12}$$

where we let $\langle\hat{x}(t)\rangle=X(t)$ for simple notational convenience and

$$Q(X,t)=V'(\langle\hat{x}\rangle)-\langle V'(\hat{x})\rangle \tag{13}$$

represents the quantum mechanical dispersion of the force operator $V'(\hat{x})$ due to the system degree of freedom. Since $Q(t)$ is a quantum fluctuation term, Eq. (12) offers a simple interpretation. This implies that the classical-looking QGLE is governed by a c-number quantum noise $F(t)$ which originates from the quantum mechanical heat bath characterized by the properties (10) and (11) and a quantum fluctuation term $Q(t)$ due to the quantum nature of the system characteristic of the nonlinearity of the potential. In Sec. VII we give a recipe for calculation of $Q(t)$.

Summarizing the above discussion, we point out that it is possible to formulate a QGLE (12) of the quantum mechanical mean value of position of a particle in a medium, provided the classical-like noise term $F(t)$ satisfies Eqs. (10) and (11), where the ensemble average has to be carried out with the distribution (8). It is thus apparent that to realize $F(t)$ as a noise term we have split up the standard quantum statistical averaging procedure into a quantum mechanical mean $\langle \cdots \rangle$ by explicitly using an initial coherent state representation of the bath oscillators and then a statistical average $\langle \cdots \rangle_S$ of the quantum mechanical mean values. Two pertinent points are to be noted: First, it may be easily verified that the distribution of quantum mechanical mean values of the bath oscillators (8) reduces to the classical Maxwell-Boltzmann distribution in the thermal limit $\hbar\omega_j \ll k_B T$. Second, the vacuum term in the distribution (8) prevents the distribution of quantum mechanical mean values from being singular at $T=0$; or in other words the width of the distribution remains finite even at absolute zero, which is a simple consequence of the uncertainty principle.

III. GENERAL ANALYSIS: DAMPED FREE PARTICLE

It is now convenient to rewrite the QGLE (12) of the quantum mechanical mean value of position of a particle in the absence of any external force field in the form

$$\ddot{X}(t) + \int_0^t \gamma(t-t')\dot{X}(t')dt' = F(t). \tag{14}$$

$\gamma(t)$ is the dissipative memory kernel as given by Eq. (4) and $F(t)$ is the zero centered stationary noise, i.e.,

$$\langle F(t)\rangle_S = 0 \text{ and } \langle F(t)F(t')\rangle_S = C(|t-t'|) = C(\tau), \tag{15}$$

where $C(t)$ is the correlation function which in the equilibrium state is connected to the memory kernel $\gamma(t)$ through an FDR of the form [7]

$$C(t-t') = \frac{1}{2}\int_0^\infty d\omega\, \kappa(\omega)\varrho(\omega)\hbar\omega \times \left(\coth\frac{\hbar\omega}{2k_B T}\right)\cos\omega(t-t'). \tag{16}$$

Equation (16) is the continuum version of Eq. (11). $\varrho(\omega)$ denotes the density of modes of the bath oscillators. Here it is important to note that Eq. (16) is the generalized FDR valid at any arbitrary temperature T. $\gamma(t-t')$ is the continuum version of Eq. (4) and is given by

$$\gamma(t-t') = \int_0^\infty d\omega\, \kappa(\omega)\varrho(\omega)\cos\omega(t-t'). \tag{17}$$

In the high temperature limit, i.e., for $\hbar\omega \ll k_B T$, we arrive at the well-known classical FDR of the second kind [41],

$$C(t-t') = k_B T\, \gamma(t-t'). \tag{18}$$

The general solution of Eq. (14) is given by

$$X(t) = \langle X(t)\rangle_S + \int_0^t H(t-\tau)F(\tau)d\tau \tag{19}$$

where

$$\langle X(t)\rangle_S = X_0 + V_0 H(t) \tag{20}$$

with $X_0 = X(0)$ and $V_0 = \dot{X}(0)$ being the initial quantum mechanical mean values of position and velocity of the particle, respectively. $H(t)$ is the inverse form of the Laplace transform

$$\tilde{H}(s) = \frac{1}{s^2 + s\tilde{\gamma}(s)} \tag{21}$$

with

$$\tilde{\gamma}(s) = \int_0^\infty \gamma(t)e^{-st}dt \tag{22}$$

the Laplace transform of the dissipative memory kernel $\gamma(t)$. The time derivative of Eq. (19) gives

$$V(t) = \langle V(t)\rangle_S + \int_0^t h(t-\tau)F(\tau)d\tau \tag{23}$$

where

$$\langle V(t)\rangle_S = V_0 h(t) \tag{24}$$

and

$$h(t) = \frac{dH(t)}{dt}. \tag{25}$$

Hence

$$\tilde{h}(s) = \frac{1}{s + \tilde{\gamma}(s)}. \tag{26}$$

Before proceeding further it is important to recall the physical significance of the two functions $H(t)$ and $h(t)$. It has already been assumed that the initial quantum mechanical velocity V_0 is independent of the random force $F(t)$,

$$\langle V_0 F(t)\rangle_S = 0. \tag{27}$$

Thus multiplying Eqs. (19) and (23) by V_0 and using relation (27) we obtain

$$\langle V_0 V(t)\rangle_S / \langle V_0^2\rangle_S = h(t), \tag{28}$$

$$\langle V_0 (X(t) - X_0)\rangle_S / \langle V_0^2\rangle_S = H(t). \tag{29}$$

Hence $H(t)$ and $h(t)$ are the two relaxation functions; $h(t)$ measures how the quantum mechanical mean velocity forgets its initial value and $H(t)$ measures how the quantum mechanical mean displacement forgets the initial velocity. As a result, the quantum mechanical mean velocity of the particle relaxes to a stationary state with zero statistical average of the quantum mechanical mean velocity.

Now, using the symmetry property of the correlation function

$$\langle F(t)F(t')\rangle_S = C(t-t') = C(t'-t)$$

and using the solution for $X(t)$ and $V(t)$ we obtain the following expressions for the variances:

$$\sigma_{XX}^2(t) \equiv \langle [X(t) - \langle X(t)\rangle_S]^2 \rangle_S = 2\int_0^t H(t_1)dt_1 \int_0^{t_1} H(t_2) C(t_1 - t_2) dt_2, \tag{30a}$$

$$\sigma_{VV}^2(t) \equiv \langle [V(t) - \langle V(t)\rangle_S]^2 \rangle_S = 2\int_0^t h(t_1)dt_1 \int_0^{t_1} h(t_2) C(t_1 - t_2) dt_2, \tag{30b}$$

and

$$\sigma_{XV}^2(t) \equiv \langle [X(t) - \langle X(t)\rangle_S][V(t) - \langle V(t)\rangle_S] \rangle_S = \frac{1}{2}\dot{\sigma}_{XX}^2(t) = \int_0^t H(t_1)dt_1 \int_0^{t} h(t_2) C(t_1 - t_2) dt_2. \tag{30c}$$

The above three expressions are valid for arbitrary temperature and friction and include quantum effects. However, in the high temperature classical limit (i.e., $\hbar\omega \ll k_B T$) one can derive simplified versions of the variances:

$$\sigma_{XX}^2(t) = k_B T\left[2\int_0^t H(t')dt' - H^2(t)\right], \tag{31a}$$

$$\sigma_{VV}^2(t) = k_B T[1 - h^2(t)], \tag{31b}$$

and

$$\sigma_{XV}^2(t) = k_B T\, H(t)[1 - h(t)]. \tag{31c}$$

Before closing this section we emphasize a pertinent point at this stage. Equations (30a)–(30c) are the expressions for the statistical variances of the quantum mechanical mean values X and V. These are not to be confused with the standard quantum mechanical variances, which are connected through uncertainty relations.

IV. A SPECIFIC EXAMPLE: EXPONENTIALLY CORRELATED MEMORY KERNEL

The very structure of $\gamma(t)$ given in Eq. (17) suggests that it is quite general and a further calculation requires prior knowledge of the density of modes $\varrho(\omega)$ of the bath oscillators. As a specific case we consider in the continuum limit

$$\kappa(\omega)\varrho(\omega) = \frac{2}{\pi}\frac{\gamma_0}{1+\omega^2\tau_c^2} \tag{32}$$

so that $\gamma(t)$ takes the well-known form

$$\gamma(t) = \frac{\gamma_0}{\tau_c} e^{-|t|/\tau_c}, \tag{33}$$

where γ_0 is the damping constant and τ_c refers to the correlation time of the noise. Once we get an explicit expression for $\gamma(t)$ in closed form and its Laplace transform, it is possible to make use of Eq. (21) to calculate the relaxation function $H(t)$, which for the present case is given by

$$H(t) = \frac{1}{\gamma_0}[1 - \mathcal{A}e^{-t/2\tau_c}\sin(\lambda t + \alpha)] \tag{34}$$

where

$$\mathcal{A} = \frac{\gamma_0}{\lambda},$$

$$\lambda = \left(\frac{\gamma_0}{\tau_c} - \frac{1}{4\tau_c^2}\right)^{1/2}, \quad \text{and} \quad \alpha = \tan^{-1}\left(\frac{2\lambda\tau_c}{1 - 2\gamma_0\tau_c}\right). \tag{35}$$

Now making use of the expressions for $H(t)$ and the correlation function $C(t)$ in Eqs. (30a)–(30c) we calculate explicitly after lengthy but straightforward algebra the time dependent expressions for the variances of the quantum mechanical mean value of position and momentum of the particle,

$$\sigma_{XX}^2(t) = \frac{2\hbar}{\pi}\int_0^\infty \frac{\omega}{1+\omega^2\tau_c^2}\left(\coth\frac{\hbar\omega}{2k_BT}\right) \times \mathcal{F}_X(\omega,t)d\omega, \tag{36}$$

$$\sigma_{VV}^2(t) = \frac{2\gamma_0\hbar}{\pi\lambda^2}\int_0^\infty \frac{\omega}{1+\omega^2\tau_c^2}\left(\coth\frac{\hbar\omega}{2k_BT}\right) \times \mathcal{F}_V(\omega,t)d\omega, \tag{37}$$

and

$$\sigma_{XV}^2(t) = \frac{1}{2}\dot{\sigma}_{XX}^2(t). \tag{38}$$

In the Appendix we provide the explicit structures of $\mathcal{F}_X(\omega,t)$ and $\mathcal{F}_V(\omega,t)$.

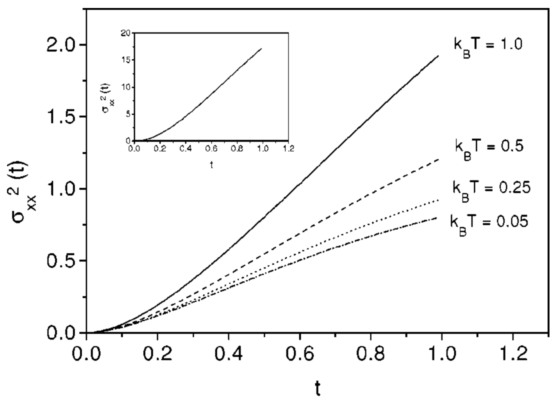

FIG. 1. Plot of $\sigma_{XX}^2(t)$ against time to show the short time behavior of the variances for different temperatures with fixed parameters $\gamma_0 = 1.0$ and $\tau_c = 1.0$. Inset: The same as in the main figure but for a higher temperature $k_B T = 10.0$ (units are arbitrary).

To examine the consistency of our calculation we check the long time behavior of the classical high temperature Ohmic limit of the variances $\sigma_{XX}^2(t)$ and $\sigma_{VV}^2(t)$. In this limit we have

$$\sigma_{XX}^2(t) = \frac{4k_B T}{\pi}\int_0^\infty d\omega \frac{1}{1+\omega^2\tau_c^2}\mathcal{F}_X(\omega,t).$$

Only the first term of $\mathcal{F}_X(\omega,t)$ gives the long time behavior of $\sigma_{XX}^2(t)$ in the Markovian limit, the contributions of the rest of the terms being zero. Taking this leading order contribution we have

$$\sigma_{XX}^2(t) = \frac{4k_B T}{\pi\gamma_0}\int_0^\infty d\omega \frac{1}{1+\omega^2\tau_c^2}\,\frac{1}{\omega^2}(1-\cos\omega t)$$

$$= \frac{8k_B T}{\pi\gamma_0}\left(\frac{1}{1+\omega^2\tau_c^2}\bigg|_{\omega=0}\right)\int_0^\infty d\omega \frac{\sin^2\frac{1}{2}\omega t}{\omega^2}$$

which gives

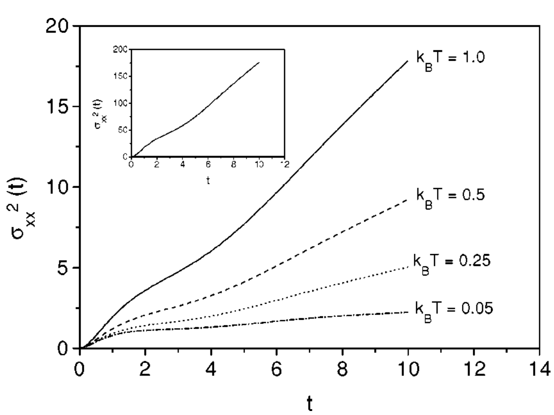

FIG. 2. Plot of $\sigma_{XX}^2(t)$ against time to show long time behavior of the variances for different temperatures. Other parameters are same as in Fig. 1. Inset: The same as in the main figure but for a higher temperature $k_B T = 10.0$ (units are arbitrary).

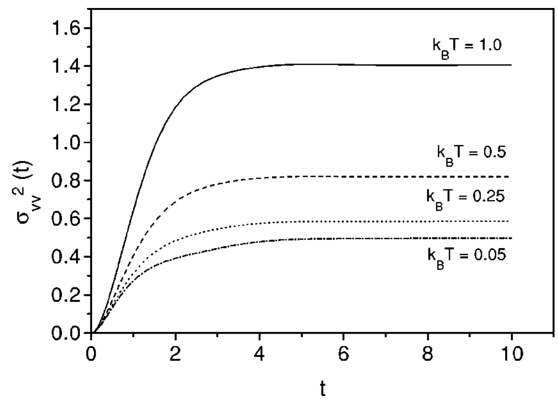

FIG. 3. Plot of $\sigma_{VV}^2(t)$ against time to show long time behavior of the variances for different temperatures. Other parameters are same as in Fig. 1 (units are arbitrary).

$$\sigma_{XX}^2(t) = \frac{2k_B T}{\gamma_0}t \quad \text{for} \quad t\to\infty. \tag{39}$$

Similarly one can show that for the classical high temperature Markovian limit

$$\sigma_{VV}^2(t) = k_B T \quad \text{for} \quad t\to\infty. \tag{40}$$

Since we are unable to further evaluate analytically the explicit time dependent structures of the variances in the general case, we resort to numerical integration of Eqs. (36) and (37). In Figs. 1 and 2 we show the short time and long time behavior of the variances $\sigma_{XX}^2(t)$ as functions of time for different values of temperature but for a fixed value of correlation time τ_c. It is apparent that, while the short time dynamics has a simple t^2 behavior, the asymptotic dependence is linear in t with a clear crossover around some intermediate time. Figure 3 exhibits the asymptotic constancy of $\sigma_{VV}^2(t)$ as a function of time for different temperatures. The effect of the correlation time τ_c on the variance $\sigma_{XX}^2(t)$ is examined in Fig. 4 for a fixed high temperature $k_B T = 10.0$. It is interesting to note that the crossover region gets longer for larger correlation times.

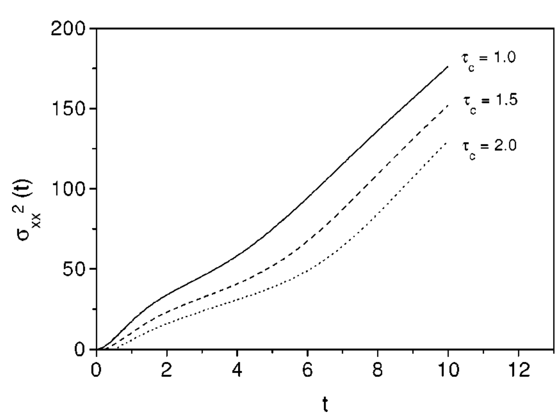

FIG. 4. Plot of $\sigma_{XX}^2(t)$ against time for different correlation times τ_c with fixed parameters $\gamma_0 = 1.0$ and $k_B T = 10.0$ (units are arbitrary).

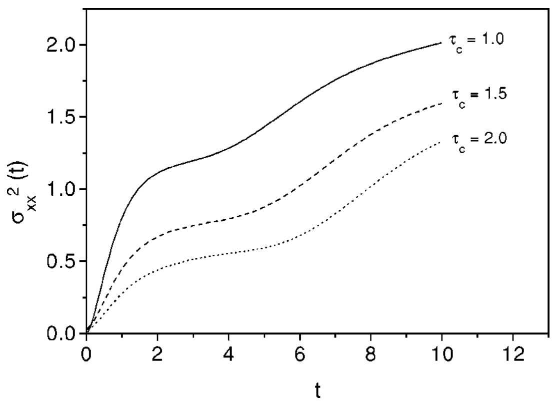

FIG. 5. Same as in Fig. 4 but for $k_BT=0.0$ (units are arbitrary).

Figures 5 and 6 illustrate the zero temperature situation. In this regime non-Markovian effects are strong, which is evident from the vacuum fluctuations growing in time in an oscillatory fashion at early stages for different values of the correlation time as shown in Fig. 5. In Fig. 6 we show how the initial growth of the variance $\sigma^2_{VV}(t)$ finally settles down to a constant nonthermal energy value.

V. THE GENERALIZED QUANTUM FOKKER-PLANCK EQUATION

We now return to our general analysis as carried out in Sec. III. To write down the Fokker-Planck description for the evolution of the probability density function of the quantum mechanical mean values of the coordinate and momentum of the particle, it is necessary to consider the statistical distribution of noise, which we assume here to be Gaussian. For Gaussian noise processes we define the joint characteristic function in terms of the standard mean values and variances as follows:

$$\tilde{P}(\mu,\rho,t)=\exp\left[i\mu\langle X(t)\rangle_S+i\rho\langle V(t)\rangle_S -\frac{1}{2}\{\sigma^2_{XX}(t)\mu^2+2\sigma^2_{XV}(t)\mu\rho+\sigma^2_{VV}(t)\rho^2\}\right]. \tag{41}$$

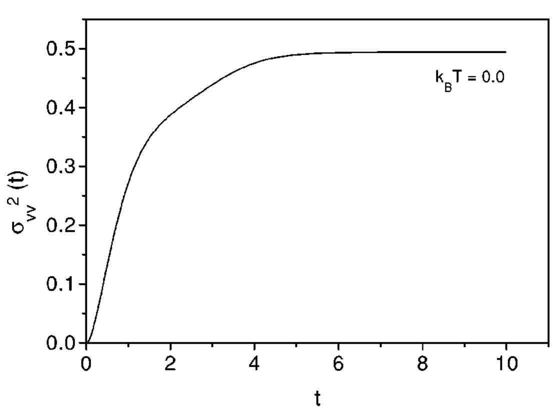

FIG. 6. Plot of $\sigma^2_{VV}(t)$ against time to show long time behavior due to vacuum fluctuations. Other parameters are same as in Fig. 1 (units are arbitrary).

Using the standard procedure [30,31] we write down below the Fokker-Planck equation (FPE) obeyed by the joint probability density function $P(X,V,t)$, which is the inverse Fourier transform of the characteristic function:

$$\left(\frac{\partial}{\partial t}+V\frac{\partial}{\partial X}\right)P(X,V,t) =\xi(t)\frac{\partial}{\partial V}VP(X,V,t)+\varphi(t)\frac{\partial^2}{\partial V^2}P(X,V,t) +\psi(t)\frac{\partial^2}{\partial X\partial V}P(X,V,t) \tag{42}$$

where

$$\xi(t)=-\dot{h}(t)/h(t), \tag{43a}$$

$$\varphi(t)=\xi(t)\sigma^2_{VV}(t)+\frac{1}{2}\dot{\sigma}^2_{VV}(t), \tag{43b}$$

and

$$\psi(t)=-\sigma^2_{VV}(t)+\xi(t)\sigma^2_{XV}(t)+\dot{\sigma}^2_{XV}(t). \tag{43c}$$

The above FPE is the exact quantum mechanical version of the classical non-Markovian FPE and is valid at any arbitrary temperature and friction.

The decisive advantage of the present approach is again noteworthy. We have mapped the operator generalized Langevin equation into a generalized Langevin equation in c numbers [Eq. (14)] and its equivalent Fokker-Planck equation [Eq. (42)]. The present approach bypasses the earlier methods of quasiprobabilistic distribution functions employed widely in quantum optics over the decades [1–5] in a number of ways. First, unlike the quasiprobabilistic distribution functions, the probability distribution function $P(X,V,t)$ is valid for non-Markov processes. Second, while the corresponding characteristic functions for quasiprobabilistic distribution functions are operators, we make use of characteristic functions which are numbers. Third, as pointed out earlier the quasidistribution functions often become negative or singular in the strong quantum domain and pose serious problems. The present approach is free from such shortcomings since the probability density function $P(X,V,t)$ behaves here as a true probability function rather than a quasiprobability function.

VI. GENERALIZED QUANTUM DIFFUSION EQUATION

In their landmark paper on classical Brownian motion, Uhlenbeck and Ornstein [42] solved the classical Markovian FPE to find $P(X,V,t)$ and then in a bid to obtain Einstein's diffusion equation tried to evaluate $p(X,t)$, the probability density function in configuration space, by integrating over V. It was shown that it is difficult if not impossible to obtain a differential equation for $P(X,V_0,t)$ from the classical Markovian FPE which for $t\gg 1/\gamma_0$ would become a diffusion equation. However, for the classical non-Markovian case

Mazo [30] in the late 1970s addressed this problem by considering an initial Maxwellian distribution $\Phi(V_0)$ of the initial velocity V_0, and then derived the exact differential equation satisfied by $p(X,t)$ where

$$p(X,t)=\int P(X,V_0,t)\Phi(V_0)dV_0.$$

The resulting equation thus reduces to the diffusion equation for $t\gg 1/\gamma$. We follow Mazo's procedure to derive an exact quantum mechanical version of the classical non-Markovian case, a differential equation which for $t\gg 1/\gamma$ goes over into a quantum diffusion equation. To this end we proceed as follows. From Eq. (41) for $\rho=0$ we have

$$\begin{aligned}\tilde{p}(\mu,t)&=\int \tilde{p}(\mu,t)\Phi(V_0)dV_0\\&=\exp\left(-\frac{1}{2}\mu^2\sigma_{XX}^2(t)\right)\exp(i\mu X_0)\\&\quad\times\int \exp[i\mu V_0 H(t)]\Phi(V_0)dV_0. \qquad (44)\end{aligned}$$

Here we take the initial Gaussian distribution of the quantum mechanical mean values of the velocity of the particle,

$$\Phi(V_0)=\left(\frac{1}{2\pi\Delta_0}\right)^{1/2}\exp\left(-\frac{V_0^2}{2\Delta_0}\right) \qquad (45)$$

where

$$\Delta_0=\varphi(\infty)/\xi(\infty). \qquad (46)$$

It is not difficult to note that the above choice is dictated by the stationary solution of the QFPE (42), i.e., Eq. (45) satisfies Eq. (42) at equilibrium. The explicit time dependent expressions for $\varphi(t)$ and $\xi(t)$ have been given in Eqs. (43a) and (43b). Inserting Eq. (45) in Eq. (44) and then performing the inverse Fourier transform after integration over V_0, we arrive at the following equation after a little algebra:

$$\frac{\partial p(X,t)}{\partial t}=D_q(t)\frac{\partial^2 p(X,t)}{\partial X^2}. \qquad (47)$$

This is the quantum analog of Einstein's diffusion equation where the explicit structure of the time dependent quantum diffusion coefficient $D_q(t)$ is given by

$$D_q(t)=\sigma_{XV}^2(t)+\Delta_0 H(t)h(t). \qquad (48)$$

The required variances, the relaxation functions, and other related quantities in Eq. (48) are given in Eqs. (30c), (25), (21), and (46). We now discuss the limiting cases. For the classical Markovian limit the variance $\sigma_{XV}^2(t)$ gives k_BT/γ_0 for $t\gg 1/\gamma_0$ and the second term in $D_q(t)$ vanishes in the long time limit, so that we recover Einstein's diffusion coefficient in configuration space. In the low temperature limit, however, the quantum effects begin to dominate. It is interesting to note that based on the Feynman-Vernon path integral technique [16,17], Hakim and Ambegaokar [11] considered explicit quantum corrections to classical diffusion to examine the differential behavior of high and low temperature dependence in the dynamics for Leggett-Caldeira initial conditions. The non-Markovian nature of the dynamics is taken into account by considering the frequency dependence of the bath with a suitable low frequency cutoff. The transient behavior in the quantum correction to classical diffusion is therefore only observable on time scales longer than the inverse cutoff frequency. The present treatment, being exact, equipped to deal with arbitrary noise correlation at all temperatures, and free from divergences, does not require any such cutoff. The quantum diffusion coefficient can be followed arbitrarily from the transient to the asymptotic regions. To explore the associated non-Markovian nature of the dynamics in the present case it is necessary to go over to numerical evaluation of $D_q(t)$. In Fig. 7 (compare with Fig. 1 of Ref. [11]) we plot the variation of quantum diffusion coefficient $D_q(t)$ for several values of the temperature as a function of time for the exponential memory kernel considered in our example in Sec. IV. It is apparent that, while the short time behavior is characterized by a sharp increase followed by a maximum, the diffusion coefficient settles down to a constant value in the asymptotic limit. The short time behavior is dominated by the second term in Eq. (48) due to the relaxation functions $H(t)$ and $h(t)$, of which the latter vanishes in the long time limit. Again, the first term in Eq. (48) offers no contribution to the diffusion coefficient from its classical part in the vacuum limit at $T=0$. The solid curve in Fig. 7 thus shows the evolution of a nonthermal diffusion coefficient of pure quantum origin.

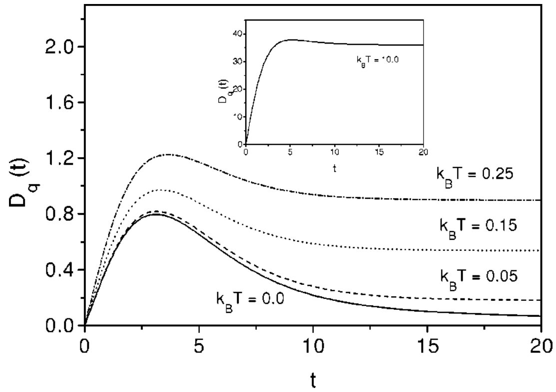

FIG. 7. Plot of quantum diffusion coefficient $D_q(t)$ against time for different temperatures and for $\gamma_0=0.275$ and $\tau_c=1.0$. Inset: Same as in the main figure but for a higher temperature $k_BT=10.0$ (units are arbitrary).

VII. QUANTUM SMOLUCHOWSKI EQUATION

We now consider the diffusion of a particle in an external potential $V(X)$ as described by the QGLE (12). In the overdamped limit we drop the inertial term $\ddot{X}(t)$ and the damping kernel $\gamma(t-t')$ is reduced to $\gamma_0\delta(t-t')$ for vanishing τ_c in Eq. (33). γ_0 is the Markovian limit of dissipation. Equation (12) then assumes the following form:

$$\dot{X}+\frac{1}{\gamma_0}[V'(X)-Q(X,t)]=\frac{F(t)}{\gamma_0}. \tag{49}$$

Expressing $V'(X)-Q(X,t)$ as a derivative of an effective quantum potential $V_{quant}(X,t)$ with respect to X, the equivalent description in terms of the true probability distribution function $p(X,t)$ is given by

$$\frac{\partial p(X,t)}{\partial t}=\frac{1}{\gamma_0}\frac{\partial}{\partial X}[V'_{quant}(X,t)p(X,t)]+D_{qo}\frac{\partial^2 p}{\partial X^2}, \tag{50a}$$

with

$$V'_{quant}(X,t)=V'(X)-Q(X,t) \tag{50b}$$

where $Q(X,t)$ is defined in Eq. (13). Here D_{qo} is the quantum diffusion coefficient in the overdamped limit, which can be obtained with the help of the following definition [1]:

$$2D_{qo}=\frac{1}{\Delta t}\int_t^{t+\Delta t}dt_1\int_t^{t+\Delta t}dt_2\frac{1}{\gamma_0^2}\langle F(t_1)F(t_2)\rangle_S. \tag{51}$$

Here the correlation function $\langle F(t_1)F(t_2)\rangle_S/\gamma_0^2$ of the c-number quantum noise is given by Eq. (16) in the continuum limit. We then make use of Eq. (32) for vanishing τ_c in Eq. (51) to obtain after explicit integration

$$D_{qo}=\frac{1}{2\gamma_0}\hbar\tilde{\omega}[2\overline{n}(\tilde{\omega})+1] \tag{52}$$

where the frequency $\tilde{\omega}$ in Eq. (52) refers to the linearized frequency of the nonlinear system [1]. We now discuss the classical and vacuum limits of the quantum Smoluchowski equation (50a). It is easy to check that in the limit $\hbar\tilde{\omega}\ll k_BT$ D_{qo} reduces to Einstein's classical diffusion coefficient k_BT/γ_0. At the same time $Q(X,t)$ vanishes so that $V'_{quant}(X,t)$ goes over to $V'(X)$ and one recovers the usual classical Smoluchowski equation. In the opposite limit as $T\to 0$, however, both quantum noise due to the nonlinearity of the system and vacuum fluctuations orginating from the heat bath make significant contributions. D_{qo} in this limit assumes the form $\hbar\tilde{\omega}/2\gamma_0$. In this context we refer to a recent treatment on the large friction limit in quantum dissipative dynamics [29] to point out that the latter theory does not retain its full validity as $T\to 0$ since the quantum noise of the heat bath disappears in the vacuum limit.

The second noteworthy feature about the quantum Smoluchowski equation (50a) is that, unlike Wigner function based equations [28], it does not contain higher order (higher than second) derivatives of $p(X,t)$. The positive definiteness of the probability distribution function is thus ensured.

It is important to emphasize at this juncture that, so far as the general formulation of the theory is concerned, Eq. (50a) contains quantum corrections to all orders. In this sense Eq. (50a) is formally an exact quantum analog of the classical Smoluchowski equation. To make it more explicit we return to the quantum mechanics of the system in the Heisenberg picture to write the operators $\hat{x}$ and $\hat{p}$ as

$$\hat{x}(t)=\langle\hat{x}(t)\rangle+\delta\hat{x}\quad\text{and}\quad\hat{p}(t)=\langle\hat{p}(t)\rangle+\delta\hat{p}. \tag{53}$$

$\langle\hat{x}(t)\rangle$ and $\langle\hat{p}(t)\rangle$ are quantities signifying quantum mechanical averages and $\delta\hat{x}$ and $\delta\hat{p}$ are quantum corrections. By construction $\langle\delta\hat{x}\rangle$ and $\langle\delta\hat{p}\rangle$ are zero and they obey the commutation relation $[\delta\hat{x},\delta\hat{p}]=i\hbar$. Using Eq. (53) in $\langle V'(\hat{x})\rangle$ and a Taylor expansion around $\langle\hat{x}\rangle$ it is possible to express $Q(X,t)$ as [see Eq. (13)]

$$Q(X,t)=-\sum_{n\geq 2}\frac{1}{n!}V_{n+1}(X)\langle\delta\hat{x}^n(t)\rangle \tag{54a}$$

where $V_n(X)$ is the nth derivative of the potential at X $(\equiv\langle\hat{x}\rangle)$. Equation (54a) suggests a simple expression for the effective potential $V_{quant}(X,t)$ as

$$V_{quant}(X,t)=V(X)+\sum_{n\geq 2}\frac{1}{n!}V_n(X)\langle\delta\hat{x}^n(t)\rangle \tag{54b}$$

where the classical potential $V(X)$ is modified by the quantum corrections to all orders. To solve the quantum Smoluchowski equation it is therefore necessary to calculate $\langle\delta\hat{x}^2(t)\rangle$, $\langle\delta\hat{x}^3(t)\rangle$, etc. To the lowest order $\langle\hat{x}\rangle$ and $\langle\delta\hat{x}^2\rangle$ follow a coupled set of equations as given below:

$$\frac{d}{dt}\langle\hat{x}\rangle=\langle\hat{p}\rangle, \tag{55a}$$

$$\frac{d}{dt}\langle\hat{p}\rangle=-V'(\langle\hat{x}\rangle)-\frac{1}{2}V'''(\langle\hat{x}\rangle)\langle\delta\hat{x}^2\rangle, \tag{55b}$$

$$\frac{d}{dt}\langle\delta\hat{x}^2\rangle=\langle\delta\hat{x}\,\delta\hat{p}+\delta\hat{p}\,\delta\hat{x}\rangle, \tag{55c}$$

$$\frac{d}{dt}\langle\delta\hat{x}\,\delta\hat{p}+\delta\hat{p}\,\delta\hat{x}\rangle=2\langle\delta\hat{p}^2\rangle-2V''(\langle\hat{x}\rangle)\langle\delta\hat{x}^2\rangle, \tag{55d}$$

$$\frac{d}{dt}\langle\delta\hat{p}^2\rangle=-V''(\langle\hat{x}\rangle)\langle\delta\hat{x}\,\delta\hat{p}+\delta\hat{p}\,\delta\hat{x}\rangle. \tag{55e}$$

The above set of equations can be derived [43] from Heisenberg's equation of motion. If one is interested in the local dynamics around a point (say, at the bottom or top of the potential well), the set of equations becomes decoupled and it is easy to obtain simple analytic solutions of Eqs. (55a)–(55e) for $\langle\hat{x}\rangle$ and $\langle\delta\hat{x}^2\rangle$ for Eq. (54a). The higher order estimates (e.g., fourth order) of the quantum corrections can be obtained from the solutions of the equations of successively higher order derived earlier by Sundaram and Milonni [43] or otherwise [44]. Since the quantum corrections due to the system are calculated by different sets of equations for successive orders, the measure of accuracy of truncation can be understood easily. It is therefore obvious that the present scheme provides a simple, systematic, and quantitative estimate of the mean field and other decorrelation methods on the basis of quantum-classical correspondence.

VIII. CONCLUSIONS

The main purpose of this paper is to enquire whether a stochastic differential equation in c numbers in the form of a generalized Langevin equation and its corresponding Fokker-Planck, diffusion, and Smoluchowski equations in terms of true probability functions are viable for description of non-Markovian quantum Brownian motion. Based on an initial coherent state representation of the bath oscillators and an equilibrium distribution of the quantum mechanical mean values of their coordinates and momenta, which satisfy the essential properties of the associated noise of the bath degrees of freedom, we derive a QGLE for the quantum mechanical mean value of the position of the particle. The main conclusions of this study are the following.

(i) Our QGLE (14) is amenable to analysis in terms of the methods developed earlier for the treatment of classical non-Markovian theory of Brownian motion.

(ii) The generalized Langevin equation (12), the corresponding Fokker-Planck equation (42), and the diffusion equation (47), and also the Smoluchowski equation (50a), are the exact quantum analog of their classical versions [30,31]. The probability distribution functions as employed here bear the true notion of statistical probability rather than that of quasiprobability.

(iii) The theory of quantum Brownian motion developed here is valid for arbitrary noise correlation and temperature and is free from divergences.

(iv) The realization of noise as a classical-looking entity which satisfies the quantum fluctuation-dissipation relationship (11) allows us to envisage quantum Brownian motion as a natural extension of its classical conterpart. The method is based on a canonical quantization procedure and makes no reference to path integral formulations.

We conclude by mentioning that the method discussed here is promising for simple differential equation based approaches [15] to quantum activated processes, tunneling problems as shown elsewhere [45], the quantum ratchet [46–48], and problems relating to motion in periodic fields [49–52] and allied issues.

ACKNOWLEDGMENT

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APPENDIX: THE EXPLICIT FORMS OF $\mathcal{F}_X(\omega,T)$ AND $\mathcal{F}_V(\omega,T)$

$\mathcal{F}_X(\omega,t)$ consists of eleven terms which are given below:

$$\mathcal{F}_X(\omega,t)=\mathcal{F}_X^{(1)}(\omega,t)+\mathcal{F}_X^{(2)}(\omega,t)+\mathcal{F}_X^{(3)}(\omega,t)+\mathcal{F}_X^{(4)}(\omega,t)+\mathcal{F}_X^{(5)}(\omega,t)+\mathcal{F}_X^{(6)}(\omega,t)+\mathcal{F}_X^{(7)}(\omega,t)+\mathcal{F}_X^{(8)}(\omega,t)+\mathcal{F}_X^{(9)}(\omega,t)+\mathcal{F}_X^{(10)}(\omega,t)+\mathcal{F}_X^{(11)}(\omega,t). \tag{A1}$$

The explicit structures of $\mathcal{F}_X^{(i)}(\omega,t)$ $(i=1,\ldots,11)$ are given by

$$\mathcal{F}_X^{(1)}(\omega,t)=(1/\gamma_0\omega^2)\,(1-\cos\omega t), \tag{A2}$$

$$\mathcal{F}_X^{(2)}(\omega,t)=\frac{\mathcal{A}A_3^{(\omega)}}{\gamma_0\omega}[\cos(\alpha+\omega t)-\cos\alpha]-\frac{\mathcal{A}A_4^{(\omega)}}{\gamma_0\omega}[\cos(\alpha-\omega t)-\cos\alpha]-\frac{\mathcal{A}A_5^{(\omega)}}{\gamma_0\omega}[\sin(\alpha+\omega t)-\sin\alpha]+\frac{\mathcal{A}A_6^{(\omega)}}{\gamma_0\omega}[\sin(\alpha-\omega t)-\sin\alpha], \tag{A3}$$

$$\mathcal{F}_X^{(3)}(\omega,t)=-\frac{\mathcal{A}A_1^{(\omega)}}{2\gamma_0^2}[e^{-t/2\tau_c}\{\sin(\lambda t+\alpha)+2\lambda\tau_c\cos(\lambda t+\alpha)\}-\{\sin\alpha+2\lambda\tau_c\cos\alpha\}], \tag{A4}$$

$$\mathcal{F}_X^{(4)}(\omega,t)=-\frac{\mathcal{A}A_2^{(\omega)}}{2\gamma_0^2}[e^{-t/2\tau_c}\{\cos(\lambda t+\alpha)-2\lambda\tau_c\sin(\lambda t+\alpha)\}-\{\cos\alpha-2\lambda\tau_c\sin\alpha\}], \tag{A5}$$

$$\mathcal{F}_X^{(5)}(\omega,t)=\frac{\mathcal{A}^2A_2^{(\omega)}}{8\gamma_0^2}[e^{-t/\tau_c}\{\sin 2(\lambda t+\alpha)+2\lambda\tau_c\cos 2(\lambda t+\alpha)\}-\{\sin 2\alpha+2\lambda\tau_c\cos 2\alpha\}], \tag{A6}$$

$$\mathcal{F}_X^{(6)}(\omega,t)=\mathcal{A}^2A_1^{(\omega)}\left(\frac{\tau_c}{2\gamma_0}\right)\left[e^{-t/\tau_c}+\frac{e^{-t/\tau_c}}{4\gamma_0\tau_c}\{2\lambda\tau_c\sin 2(\lambda t+\alpha)-\cos 2(\lambda t+\alpha)\}-\left\{1+\frac{1}{4\gamma_0\tau_c}(2\lambda\tau_c\sin 2\alpha-\cos 2\alpha)\right\}\right], \tag{A7}$$

$$\mathcal{F}_X^{(7)}(\omega,t) = -\frac{\mathcal{A}}{\gamma_0 \omega}[A_3^{(\omega)}(e^{-t/2\tau_c}\{2\tau_c(\lambda-\omega)\sin[\alpha+(\lambda-\omega)t]-\cos[\alpha+(\lambda-\omega)t]\}-2\tau_c(\lambda-\omega)\sin\alpha+\cos\alpha)$$
$$-A_4^{(\omega)}(e^{-t/2\tau_c}\{2\tau_c(\lambda+\omega)\sin[\alpha+(\lambda+\omega)t]-\cos[\alpha+(\lambda+\omega)t]\}-2\tau_c(\lambda+\omega)\sin\alpha+\cos\alpha)], \tag{A8}$$

$$\mathcal{F}_X^{(8)}(\omega,t) = \frac{\mathcal{A}^2 A_3^{(\omega)}}{\gamma_0}[A_3^{(\omega)}\{e^{-t/2\tau_c}[2\tau_c(\lambda-\omega)\sin(\lambda-\omega)t-\cos(\lambda-\omega)t]+1\}-A_4^{(\omega)}(e^{-t/2\tau_c}\{2\tau_c(\lambda+\omega)\sin[2\alpha+(\lambda+\omega)t]$$
$$-\cos[2\alpha+(\lambda+\omega)t]\}-[2\tau_c(\lambda+\omega)\sin 2\alpha-\cos 2\alpha])], \tag{A9}$$

$$\mathcal{F}_X^{(9)}(\omega,t) = \frac{\mathcal{A}^2 A_4^{(\omega)}}{\gamma_0}[A_4^{(\omega)}\{e^{-t/2\tau_c}[2\tau_c(\lambda+\omega)\sin(\lambda+\omega)t-\cos(\lambda+\omega)t]+1\}-A_3^{(\omega)}(e^{-t/2\tau_c}\{2\tau_c(\lambda-\omega)\sin[2\alpha+(\lambda-\omega)t]$$
$$-\cos[2\alpha+(\lambda-\omega)t]\}-[2\tau_c(\lambda-\omega)\sin 2\alpha-\cos 2\alpha])], \tag{A10}$$

$$\mathcal{F}_X^{(10)}(\omega,t) = -\frac{\mathcal{A}^2 A_5^{(\omega)}}{\gamma_0}[A_4^{(\omega)}(e^{-t/2\tau_c}\{\sin[2\alpha+(\lambda+\omega)t]+2\tau_c(\lambda+\omega)\cos[2\alpha+(\lambda+\omega)t]\}-[\sin 2\alpha+2\tau_c(\lambda+\omega)\cos 2\alpha])$$
$$+A_3^{(\omega)}\{e^{-t/2\tau_c}[\sin(\lambda-\omega)t+2\tau_c(\lambda-\omega)\cos(\lambda-\omega)t]-2\tau_c(\lambda-\omega)\}], \tag{A11}$$

and

$$\mathcal{F}_X^{(11)}(\omega,t) = -\frac{\mathcal{A}^2 A_6^{(\omega)}}{\gamma_0}[A_3^{(\omega)}(e^{-t/2\tau_c}\{\sin[2\alpha+(\lambda-\omega)t]+2\tau_c(\lambda-\omega)\cos[2\alpha+(\lambda-\omega)t]\}-[\sin 2\alpha+2\tau_c(\lambda-\omega)\cos 2\alpha])$$
$$+A_4^{(\omega)}\{e^{-t/2\tau_c}[\sin(\lambda+\omega)t+2\tau_c(\lambda+\omega)\cos(\lambda+\omega)t]-2\tau_c(\lambda+\omega)\}], \tag{A12}$$

where

$$A_1^{(\omega)} = \tau_c\left[\frac{1}{1+4\tau_c^2(\lambda-\omega)^2}+\frac{1}{1+4\tau_c^2(\lambda+\omega)^2}\right],$$

$$A_2^{(\omega)} = 2\tau_c^2\left[\frac{\lambda-\omega}{1+4\tau_c^2(\lambda-\omega)^2}+\frac{\lambda-\omega}{1+4\tau_c^2(\lambda+\omega)^2}\right],$$

$$A_3^{(\omega)} = \frac{\tau_c}{1+4\tau_c^2(\lambda-\omega)^2}, \quad A_4^{(\omega)} = \frac{\tau_c}{1+4\tau_c^2(\lambda+\omega)^2},$$

$$A_5^{(\omega)} = \frac{2\tau_c^2(\lambda-\omega)}{1+4\tau_c^2(\lambda-\omega)^2}, \quad \text{and} \quad A_6^{(\omega)} = \frac{2\tau_c^2(\lambda+\omega)}{1+4\tau_c^2(\lambda+\omega)^2}. \tag{A13}$$

Similarly, we have

$$\mathcal{F}_V(\omega,t) = \mathcal{F}_V^{(1)}(\omega,t)+\mathcal{F}_V^{(2)}(\omega,t)+\mathcal{F}_V^{(3)}(\omega,t)+\mathcal{F}_V^{(4)}(\omega,t)+\mathcal{F}_V^{(5)}(\omega,t)+\mathcal{F}_V^{(6)}(\omega,t)+\mathcal{F}_V^{(7)}(\omega,t) \tag{A14}$$

with

$$\mathcal{F}_V^{(1)}(\omega,t) = \frac{1}{4}\left(\frac{A_1^{(\omega)}}{2\tau_c}+\lambda A_2^{(\omega)}\right)\left[e^{-t/\tau_c}+\frac{e^{-t/\tau_c}}{4\gamma_0\tau_c}\{2\lambda\tau_c\sin 2(\lambda t+\alpha)-\cos 2(\lambda t+\alpha)\}-\left\{1+\frac{1}{4\gamma_0\tau_c}(2\lambda\tau_c\sin 2\alpha-\cos 2\alpha)\right\}\right], \tag{A15}$$

$$\mathcal{F}_V^{(2)}(\omega,t) = \frac{\lambda\tau_c}{2}\left(\lambda A_1^{(\omega)}-\frac{A_2^{(\omega)}}{2\tau_c}\right)\left[e^{-t/\tau_c}-\frac{e^{-t/\tau_c}}{4\gamma_0\tau_c}\{2\lambda\tau_c\sin 2(\lambda t+\alpha)-\cos 2(\lambda t+\alpha)\}-\left\{1-\frac{1}{4\gamma_0\tau_c}(2\lambda\tau_c\sin 2\alpha-\cos 2\alpha)\right\}\right], \tag{A16}$$

$$\mathcal{F}_V^{(3)}(\omega,t)=\frac{-1}{8\gamma_0}\left(\frac{\lambda A_1^{(\omega)}}{\tau_c}+\lambda^2A_2^{(\omega)}-\frac{A_2^{(\omega)}}{4\tau_c^2}\right)\times[e^{-t/\tau_c}\{\sin 2(\lambda t+\alpha)+2\lambda\tau_c\cos 2(\lambda t+\alpha)\}-\{\sin 2\alpha+2\lambda\tau_c\cos 2\alpha\}], \tag{A17}$$

$$\begin{aligned}\mathcal{F}_V^{(4)}(\omega,t)=&\left(\frac{A_3^{(\omega)}}{2\tau_c}+\lambda A_5^{(\omega)}\right)\Bigg[\frac{A_3^{(\omega)}}{2\tau_c}\{e^{-t/2\tau_c}[2\tau_c(\lambda-\omega)\sin(\lambda-\omega)t-\cos(\lambda-\omega)t]+1\}+\lambda A_3^{(\omega)}(e^{-t/2\tau_c}\{\sin[2\alpha+(\lambda-\omega)t]\\&+2\tau_c(\lambda-\omega)\cos[2\alpha+(\lambda-\omega)t]\}-e^{-t/2\tau_c}[\sin(\lambda-\omega)t+2\tau_c(\lambda-\omega)\cos(\lambda-\omega)t]-[\sin 2\alpha+2\tau_c(\lambda-\omega)\cos 2\alpha]\\&+2\tau_c(\lambda-\omega))-\frac{A_4^{(\omega)}}{2\tau_c}(e^{-t/2\tau_c}\{2\tau_c(\lambda+\omega)\sin[2\alpha+(\lambda+\omega)t]-\cos[2\alpha+(\lambda+\omega)t]\}-[2\tau_c(\lambda+\omega)\sin 2\alpha\\&-\cos 2\alpha])\Bigg],\end{aligned} \tag{A18}$$

$$\begin{aligned}\mathcal{F}_V^{(5)}(\omega,t)=&\left(\frac{A_4^{(\omega)}}{2\tau_c}+\lambda A_6^{(\omega)}\right)\Bigg[\frac{A_4^{(\omega)}}{2\tau_c}[e^{-t/2\tau_c}[2\tau_c(\lambda+\omega)\sin(\lambda+\omega)t-\cos(\lambda+\omega)t]+1]\\&-\frac{A_3^{(\omega)}}{2\tau_c}(e^{-t/2\tau_c}\{2\tau_c(\lambda-\omega)\sin[2\alpha+(\lambda-\omega)t]-\cos[2\alpha+(\lambda-\omega)t]\}-[2\tau_c(\lambda-\omega)\sin 2\alpha-\cos 2\alpha])\\&+\lambda A_3^{(\omega)}(e^{-t/2\tau_c}\{\sin[2\alpha+(\lambda-\omega)t]+2\tau_c(\lambda-\omega)\cos[2\alpha+(\lambda-\omega)t]\}-[\sin 2\alpha+2\tau_c(\lambda-\omega)\cos 2\alpha])\\&-\lambda A_4^{(\omega)}\{e^{-t/2\tau_c}[\sin(\lambda+\omega)t+2\tau_c(\lambda+\omega)\cos(\lambda+\omega)t]-2\tau_c(\lambda+\omega)\}\Bigg],\end{aligned} \tag{A19}$$

$$\begin{aligned}\mathcal{F}_V^{(6)}(\omega,t)=&\left(\lambda A_3^{(\omega)}-\frac{A_5^{(\omega)}}{2\tau_c}\right)\Bigg[\frac{A_3^{(\omega)}}{2\tau_c}\{e^{-t/2\tau_c}[\sin(\lambda-\omega)t+2\tau_c(\lambda-\omega)\cos(\lambda-\omega)t]-2\tau_c(\lambda-\omega)\}\\&+\frac{A_4^{(\omega)}}{2\tau_c}(e^{-t/2\tau_c}\{\sin[2\alpha+(\lambda+\omega)t]+2\tau_c(\lambda+\omega)\cos[2\alpha+(\lambda+\omega)t]\}-(\sin 2\alpha+2\tau_c(\lambda+\omega)\cos 2\alpha])\\&+\lambda A_4^{(\omega)}(e^{-t/2\tau_c}\{2\tau_c(\lambda+\omega)\sin[2\alpha+(\lambda+\omega)t]-\cos[2\alpha+(\lambda+\omega)t]\}-[2\tau_c(\lambda+\omega)\sin 2\alpha-\cos 2\alpha])\\&+\lambda A_3^{(\omega)}\{e^{-t/2\tau_c}[2\tau_c(\lambda-\omega)\sin(\lambda-\omega)t-\cos(\lambda-\omega)t]+1\}\Bigg],\end{aligned} \tag{A20}$$

and

$$\begin{aligned}\mathcal{F}_V^{(7)}(\omega,t)=&\left(\lambda A_4^{(\omega)}-\frac{A_6^{(\omega)}}{2\tau_c}\right)\Bigg[\frac{A_3^{(\omega)}}{\tau_c}(e^{-t/2\tau_c}\{\sin[2\alpha+(\lambda-\omega)t]+2\tau_c(\lambda-\omega)\cos[2\alpha+(\lambda-\omega)t]\}\\&-[\sin 2\alpha+2\tau_c(\lambda-\omega)\cos 2\alpha])+\frac{A_4^{(\omega)}}{\tau_c}\{e^{-t/2\tau_c}[\sin(\lambda+\omega)t+2\tau_c(\lambda+\omega)\cos(\lambda+\omega)t]-2\tau_c(\lambda+\omega)\}\\&+\lambda A_3^{(\omega)}(e^{-t/2\tau_c}\{2\tau_c(\lambda-\omega)\sin[2\alpha+(\lambda-\omega)t]-\cos[2\alpha+(\lambda-\omega)t]\}-[2\tau_c(\lambda-\omega)\sin 2\alpha-\cos 2\alpha])\\&+\lambda A_4^{(\omega)}\{e^{-t/2\tau_c}[2\tau_c(\lambda+\omega)\sin(\lambda+\omega)t-\cos(\lambda+\omega)t]+1\}\Bigg].\end{aligned} \tag{A21}$$

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