Solution of quantum Langevin equation: Approximations, theoretical and numerical aspects

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Based on a coherent state representation of noise operator and an ensemble averaging procedure using Wigner canonical thermal distribution for harmonic oscillators, a generalized quantum Langevin equation has been recently developed [Phys. Rev. E 65, 021109 (2002); 66, 051106 (2002)] to derive the equations of motion for probability distribution functions in $c$-number phase-space. We extend the treatment to explore several systematic approximation schemes for the solutions of the Langevin equation for nonlinear potentials for a wide range of noise correlation, strength and temperature down to the vacuum limit. The method is exemplified by an analytic application to harmonic oscillator for arbitrary memory kernel and with the help of a numerical calculation of barrier crossing, in a cubic potential to demonstrate the quantum Kramers’ turnover and the quantum Arrhenius plot. © 2004 American Institute of Physics.

I. INTRODUCTION

A system coupled to its environment is the standard paradigm for quantum theory of Brownian motion. Its overwhelming success in the treatment of various phenomena in quantum optics, transport processes in Josephson junction, coherence effects and macroscopic quantum tunneling in condensed matter physics, electron transfer in large molecules, thermal activation processes in chemical reactions is now well known and forms a large body of current literature. While the early development of quantum optics initiated in the sixties and seventies was based on density operator, semigroup, noise operator or master equation approximations, path integral approach to quantum Brownian motion attracted wide attention in the early eighties. Although this development had widened the scope of condensed matter and chemical physics significantly, so far as the large coupling between the system and the heat bath and large correlation times of the noise processes are concerned several problems still need to be addressed. First, a search for quantum analogue of Kramers’ equation for a nonlinear potential which describes quantum Brownian motion in phase space had remained elusive and at best resulted in equation of motion which contains higher (than second) derivatives of probability distribution functions whose positive definiteness is never guaranteed. As a result these quasiprobability distribution functions often become singular or negative in the full quantum domain. Second, although long correlation times and large coupling constants are treated nonperturbatively formally in an exact manner by path integrals, their analytic evaluation beyond harmonic oscillator is generally completed within semiclassical schemes. This results in situations where the theory fails to retain its validity in the vacuum limit. Third, although the numerical techniques based on the path integral Monte Carlo are very successful in the treatment of equilibrium properties it is often difficult to implement in a dynamical scheme because of the well-known sign problem associated with varying phases in a quantum evolution over paths due to the oscillating nature of the real time propagator. Fourth, the treatment of non-Markovian dynamics within a quantum formulation as retardation effect in a memory functional is very difficult, if not impossible, to handle even numerically.

Keeping in view of these difficulties it is worthwhile to ask how to extend classical theory of Brownian motion to quantum domain for arbitrary friction and temperature. Based on a coherent state representation of the noise operator and an ensemble averaging procedure using Wigner canonical thermal distribution for harmonic oscillator we have recently developed a scheme for quantum Brownian motion in terms of a $c$-number generalized Langevin equation. The approach allows us to use classical methods of non-Markovian dynamics to derive the generalized quantum Kramers’ equation and the other relevant quantum analogs of classical diffusion, Fokker–Planck and Smoluchowski equations and of the classical expression for reactive flux. The object of the present analysis is to explore several approximation schemes for solving $c$-number generalized quantum Langevin equation coupled to quantum dispersion equations.
II. QUANTUM BROWNIAN MOTION

A. Coherent state representation of quantum noise and quantum Langevin equation in c-numbers

We consider a particle of unit mass coupled to a medium comprised of a set of harmonic oscillators with frequency \( \omega_j \). This is described by the following Hamiltonian:

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

Here \( \hat{x} \) and \( \hat{p} \) are 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 \( \kappa_j \). 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 relations \( [\hat{x}, \hat{p}] = i\hbar \) and \( [\hat{q}_k, \hat{p}_j] = i\hbar \delta_{kj} \). Note that in writing down the Hamiltonian no rotating wave approximation has been used.

Eliminating the reservoir degrees of freedom in the usual way we obtain the operator Langevin equation for the particle,

\[
\dot{\hat{x}}(t) + \int_0^t d\tau \gamma(t - \tau) \dot{\hat{x}}(\tau) + V'(\hat{x}) = \hat{F}(t), \tag{2}
\]

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

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

and

\[
\gamma(t) = \sum_j \kappa_j \cos \omega_j t, \tag{3}
\]

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

Equation (2) is the well known exact quantized operator Langevin equation\(^7\) for which the noise properties of \( \hat{F}(t) \) can be derived using a suitable initial canonical distribution of the bath coordinate and momentum operators at \( t = 0 \) as follows:

\[
\langle \hat{F}(t) \rangle_{\text{QS}} = 0, \tag{5}
\]

\[
\frac{1}{2} \langle \hat{F}(t) \hat{F}(t') \rangle_{\text{QS}} + \langle \hat{F}(t') \hat{F}(t) \rangle_{\text{QS}} = \frac{1}{2} \sum_j \kappa_j \hbar \omega_j \left( \coth \frac{\hbar \omega_j}{2k_B T} \right) \cos \omega_j (t - t'), \tag{6}
\]

where \( \langle \cdots \rangle_{\text{QS}} \) refers to quantum statistical average on bath degrees of freedom and is defined as

\[
\langle \hat{F} \rangle_{\text{QS}} = \frac{\text{Tr} \exp(-\hat{H}_{\text{bath}}/k_B T)}{\text{Tr} \exp(-\hat{H}_{\text{bath}}/k_B T)} \tag{7}
\]

for any operator \( \hat{F}(\delta \hat{q}_j - \hat{x}) \), \( \{\hat{p}_j\} \), where \( \hat{H}_{\text{bath}} = \sum_j \left( \hat{p}_j^2/2 + 1/2 \kappa_j (\hat{q}_j - \hat{x})^2 \right) \) at \( t = 0 \). By Trace we mean the usual quantum statistical average. Equation (6) is the fluctuation-dissipation relation with the noise operators ordered appropriately in the quantum mechanical sense.

To construct a c-number Langevin equation we proceed from Eq. (2). We first carry out the quantum mechanical average of Eq. (2),

\[
\langle \ddot{x}(t) \rangle + \int_0^t dt' \gamma(t - t') \langle \ddot{x}(t') \rangle + \langle V'(\hat{x}) \rangle = \langle \hat{F}(t) \rangle, \tag{8}
\]

where the quantum mechanical average \( \langle \cdots \rangle \) is taken over the initial product separable quantum states of the particle and the bath oscillators at \( t = 0 \). \(|\phi\rangle\langle\alpha|\rangle (|\alpha|\rangle = |\alpha_j\rangle \cdots |\alpha_N\rangle) \) refers to the initial coherent state of the \( i \)th bath oscillator. \( |\alpha_i\rangle \) is given by \( |\alpha_i\rangle = \exp(-|\alpha_i|^2/2) \sum_n \frac{1}{\sqrt{n!}} |\alpha_i|^n \), \( \alpha_i \) being expressed in terms of the mean values of the shifted coordinate and momentum of the \( i \)th oscillator, \( \langle \hat{x}(0) \rangle - \langle \hat{x}(0) \rangle = (\sqrt{\hbar/2} \omega_j) \times (\alpha_j + \alpha_j^*) \) and \( \langle \hat{p}(0) \rangle = i\sqrt{\hbar} \omega_j/2 (\alpha_j^* - \alpha_j) \), 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 and is given by (\( \langle \hat{F}(t) \rangle = f(t) \),

\[
f(t) = \sum_j \left[ \left\{ \langle \hat{q}_j(0) \rangle - \langle \hat{x}(0) \rangle \right\} \kappa_j \cos \omega_j t \right. \]

\[
\left. + \langle \hat{p}_j(0) \rangle \kappa_j^{1/2} \sin \omega_j t \right]
\]

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

\[
\langle \ddot{x}(t) \rangle + \int_0^t dt' \gamma(t - t') \langle \ddot{x}(t') \rangle + \langle V'(\hat{x}) \rangle = f(t). \tag{10}
\]

To realize \( f(t) \) as an effective c-number noise we now assume the ansatz 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 form as

\[
\mathcal{P}_j = N \exp \left\{ -\frac{[\langle \hat{p}_j(0) \rangle]^2 + \kappa_j (\langle \hat{q}_j(0) \rangle - \langle \hat{x}(0) \rangle)^2]}{2 \hbar \omega_j (\alpha_j^* + \frac{1}{2})} \right\} \tag{11}
\]

so that for any quantum mechanical mean value \( O_j \langle \hat{p}_j(0) \rangle \), \( \langle (\hat{q}_j(0) - \langle \hat{x}(0) \rangle) \rangle \) the statistical average \( \langle \cdots \rangle \) is
\[ \langle O_j \rangle_S = \int O_j(\{\hat{\phi}_j(0)\},\{\hat{\varphi}_j(0)\} - \langle \hat{X}(0) \rangle) \times \mathcal{P}_j(\{\hat{\phi}_j(0)\},\{\hat{\varphi}_j(0)\} - \langle \hat{X}(0) \rangle) \times d\{\hat{\phi}_j(0)\}d\{\hat{\varphi}_j(0)\} - \langle \hat{X}(0) \rangle. \]  

(12)

Here \( \bar{n}_j \) indicates the average thermal photon number of the \( j \)th oscillator at temperature \( T \) and \( \bar{n}_j = 1 + \exp(\hbar \omega_j / k_B T) - 1 \) and \( \mathcal{N} \) is the normalization constant.

The distribution (11) and the definition of statistical average (12) imply that \( f(t) \) must satisfy
\[ \langle f(t) \rangle_S = 0 \]  

(13)

and
\[ \langle f(t) f(t') \rangle_S = \frac{1}{2} \sum_j \kappa_j \hbar \omega_j \left( \coth \frac{\hbar \omega_j}{2k_B T} \cos \omega_j (t-t') \right). \]  

(14)

That is, \( c \)-number noise \( f(t) \) is such that it is zero-centered and satisfies the standard fluctuation-dissipation relation (FDR) as expressed in Eq. (6). It is important to emphasize that the ansatz (11) is a canonical Wigner distribution for a shifted harmonic oscillator which remains always a positive definite function. A special advantage of using this distribution is that it remains valid as pure state nonsingular distribution function at \( T=0 \). Furthermore, this procedure allows us to bypass the operator ordering prescription of Eq. (6) for deriving the noise properties of the bath in terms of fluctuation-dissipation relation and to identify \( f(t) \) as a classical looking noise with quantum mechanical content. We now return to Eq. (10) to add the force term \( V'(\langle \hat{X} \rangle) \) on both sides of Eq. (10) and rearrange it to obtain
\[ \hat{X} = \hat{P}, \]  

(15)

\[ \hat{P} = -\int_0^t dt' \gamma(t-t') \hat{P}(t') - \dot{V}'(X) + f(t) + Q(t), \]  

(16)

where we put \( \langle \hat{X}(t) \rangle = X(t) \) for notational convenience and
\[ Q(t) = V'(\langle \hat{X} \rangle) - \langle \dot{V}'(\hat{X}) \rangle \]  

(17)

represents the quantum correction due to the system degrees of freedom. Equation (16) offers a simple interpretation. This implies that the QGLE is governed by a \( c \)-number quantum noise \( f(t) \) originating from the heat bath characterized by the properties (13) and (14) and a quantum fluctuation term \( Q(t) \) characteristic of the nonlinearity of the potential. To proceed further we need a recipe for the calculation of \( Q(t) \).

Referring to the quantum nature of the system in the Heisenberg picture, one may write
\[ \hat{X}(t) = X + \delta \hat{X}, \]  

(18)

\[ \hat{P}(t) = P + \delta \hat{P}, \]  

(19)

where \( \langle \delta \hat{X} \rangle = X \) and \( \langle \delta \hat{P} \rangle = P \) are the quantum-mechanical averages and \( \delta \hat{X}, \delta \hat{P} \) are the operators. By construction \( \langle \delta \hat{X} \rangle \) and \( \langle \delta \hat{P} \rangle \) are zero and \( [\delta \hat{X}, \delta \hat{P}] = i\hbar \). Using Eqs. (18) and (19) in \( \langle \dot{V}'(\hat{X}) \rangle \) and a Taylor series expansion around \( \hat{X} \) it is possible to express \( Q(t) \) as
\[ Q(t) = \sum_{n=2} \frac{1}{n!} V^{(n+1)}(X)(\delta \hat{X}^n(t)). \]  

(20)

Here \( V^{(n)}(X) \) is the \( n \)th derivative of the potential \( V(X) \). For example, the second order \( Q(t) \) is given by \( Q(t) = -\frac{1}{2} V''(X)(\delta \hat{X}^2) \). The calculation of \( Q(t) \) therefore rests on quantum correction terms, \( \langle \delta \hat{X}^n(t) \rangle \) which are determined by solving the quantum correction equations as discussed in the following section.

A digression on initial overall density matrix as well as on \( c \)-number Langevin equation may be now in order. We note that in the standard derivation of operator Langevin equation one assumes that the initial (system + reservoir) density matrix is separable into that of the system and that of the bath with the latter being an equilibrium density matrix at temperature \( T \). In contrast we assume that initial bath density matrix is diagonal in the coherent state basis. It appears as an \( \textit{ad hoc} \) assumption although it gives correct classical limit, fluctuation-dissipation relation and results in the deep tunneling region.

The choice of such initial conditions and how the present Langevin equation differs from the standard approaches can be clarified in the following way.

Since the additive noise operators (3) are one photon excitation-de-excitation bath operators, the quantum mechanical average with Fock states \( \{ |n_j \rangle \} \) of the harmonic oscillators of the bath yields zero \( \langle |n_j \rangle | \hat{F}_j(t) | n_j \rangle = 0 \) making a further statistical average redundant. The statistical property of the thermal bath only makes its presence felt through two-time correlation functions of the noise operators appropriately ordered. It is therefore difficult to make a one-to-one correspondence between a numerically computable \( c \)-number noise [as \( f(t) \) in our case] and the corresponding noise operator \( \hat{F}(t) \) in a nondiagonal basis of the bath states. A diagonal basis serves this purpose in the present context. Second, one may note that the choice of an initial nondiagonal basis generally leads to the time evolution of off-diagonal matrix elements. In the present approach we have avoided the appearance of these elements by expressing the system operator at any time as a quantum mechanical average plus a fluctuation operator [Eqs. (18) and (19)] in a nonlinear function of operators. The procedure, however, generates a set of quantum correction equations which are to be solved order by order. Thus the present approach differs from the standard one in the following way. The initial bath density in the diagonal basis allows us to construct the equation of motion for quantum mechanical averages and dispersions for the system driven by a classical looking noise whose correlation is quantum mechanical in its content. In the usual approach (e.g., Ref. 3), on the other hand, one sets up a correspondence between \( c \)-numbers and operators via a quasiclassical phase space distribution function whose evolution equation [which takes care of nonlinearity of the product of operators in terms of higher (than second) derivatives of the distribution function] can be expressed equivalently in terms of Langevin equation for \( c \)-numbers within the usual approximation schemes. Thus the quantum correction equations do not appear in this case. It is however important to note that the system equilibrates after a while so that the difference in initial conditions becomes immaterial.
Finally a note on noise correlation is relevant. As the theory of Brownian motion at low temperatures is largely stimulated by investigation on resistively shunted Josephson junctions where it has been confirmed by experiments that the voltage noise observed at low temperatures has a power spectrum containing a product of frequency dependent damping $\gamma(\omega)$ and an energy factor, $\hbar \omega \coth (\hbar \omega / 2 k_B T)$ which goes over to classical limit of white noise $\hbar \omega \ll k_B T$ for small frequencies where $\hbar \omega \ll k_B T$ and to $\hbar \omega$ for high frequency limit where $\hbar \omega \gg k_B T$, where $\Gamma$ is the dissipation constant in the classical Markovian limit. It is thus easy to guess that such an energy factor must have to appear as an integral part of $c$-number noise correlation of the thermal bath. From a theoretical point of view on the other hand, this energy factor can be recovered as a width of Wigner canonical thermal distribution function of $c$-numbers obtained as an exact solution of Wigner equation for the harmonic oscillator. The choice of initial conditions and ensemble averaging with a positive definite Wigner distribution can therefore be rationalized both from experimental and theoretical standpoint.

B. Quantum correction equations

We now return to operator equation (2) and put (18) and (19) to obtain

$$\delta \hat{x} = \delta \hat{p},$$

$$\delta \hat{p} + \int_0^t \gamma(t-t') \delta \hat{p}(t') dt' + V''(X) \delta \hat{x}$$

$$+ \sum_{n \geq 2} \frac{1}{n!} V^{(n+1)}(X) \delta \hat{x}^n = \hat{F}(t) - f(t).$$

Equations (21)–(22) form the key element for calculation of the quantum mechanical correction Eq. (20) due to nonlinearity of the system potential $V(x)$. Note that $\langle \delta \hat{x}^n(t) \rangle$ in $Q(t)$ does not include any statistical averaging and is a dispersion term obtained by pure quantum mechanical averaging which is not to be confused with $\langle \delta \hat{x}^n(t) \rangle$. Thus, to obtain $\langle \delta \hat{x}^n(t) \rangle$ we need to perform only a quantum mechanical averaging over the initial product separable bath states $\Pi_{n=1}^\infty \{ | \alpha_0(0) \rangle \}$ to get rid of the term $\hat{F}(t) - f(t)$. Depending on the nonlinearity of the potential and the memory kernel we consider the following cases separately.

1. Arbitrary memory kernel and harmonic potential

We consider the friction kernel to be arbitrary (but decaying) and the potential as harmonic for which the derivatives higher than second in $V(X)$ in Eq. (22) vanish. The above set of equations may be put in the form (after quantum mechanical averaging over bath states has been performed),

$$\delta \hat{x} = \delta \hat{p},$$

$$\delta \hat{p} = - \int_0^t \gamma(t-t') \delta \hat{p}(t') dt' - \omega^2 \delta \hat{x},$$

where $\omega^2 = V''(X)$, is a constant.

The operator equations (23) and (24) can be solved exactly by Laplace transform technique to obtain

$$\delta \hat{x}(t) = \delta \hat{x}(0) \left[ 1 - \omega^2 \int_0^t G(t') dt' \right] + \delta \hat{p}(0) G(t),$$

where $G(t)$ is identified as the inverse Laplace transform of $\tilde{G}(s)$ given by

$$\tilde{G}(s) = \frac{1}{s^2 + \gamma(s) + \omega^2}$$

with $\gamma(s)$ as

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

the Laplace transform of the friction kernel $\gamma(t)$. Equation (25) yields the relevant quantum correction $\langle \delta \hat{x}^2(t) \rangle$ after squaring and quantum mechanical averaging as,

$$\langle \delta \hat{x}^2(t) \rangle = (\langle \delta \hat{x}^2(0) \rangle) G^2(t) + (\langle \delta \hat{p}^2(0) \rangle) G^2(t) + G(t) \Gamma G(t)$$

$$\times (\langle \delta \hat{x}(0) \delta \hat{p}(0) + \delta \hat{p}(0) \delta \hat{x}(0) \rangle).$$

We shall return to this issue in more detail when we discuss the quantum Brownian motion of a damped harmonic oscillator, in Sec. III.

2. Exponential memory kernel and arbitrary potential

For a Lorentzian distribution of bath modes characterized by a density function $\rho(\omega)$,

$$\kappa(\omega) \rho(\omega) = \frac{2}{\pi} \frac{\Gamma}{\omega^2 + \Gamma^2},$$

where $\Gamma$ is the dissipation constant in the Markovian limit and $\tau_c$ refers to the correlation time, the memory kernel is exponential and $\gamma(t)$ is given by

$$\gamma(t) = \frac{\Gamma}{\tau_c} e^{-|t|/\tau_c}.$$ 

In this case the quantum correction Eqs. (21)–(22), after quantum mechanical averaging over bath states, may be rewritten as

$$\delta \hat{x} = \delta \hat{p},$$

$$\delta \hat{p} = - V''(X) \delta \hat{x} - \sum_{n \geq 2} \frac{1}{n!} V^{(n+1)}(X) \delta \hat{x}^n + \delta \hat{z},$$

$$\delta \hat{z} = - \frac{\Gamma}{\tau_c} \delta \hat{p} - \frac{1}{\tau_c} \delta \hat{z},$$

where we have introduced an auxiliary operator $\delta \hat{z}$ to bypass the convolution integral in Eq. (22). Making use of Eqs. (31)–(33) one can derive the quantum correction equations up to second order to obtain

$$\langle \delta \hat{x}^2 \rangle = \langle \delta \hat{x} \delta \hat{p} + \delta \hat{p} \delta \hat{x} \rangle,$$

$$\langle \delta \hat{x} \delta \hat{p} + \delta \hat{p} \delta \hat{x} \rangle = 2 \langle \delta \hat{p}^2 \rangle - 2 V''(X) \langle \delta \hat{x}^2 \rangle + \langle \delta \hat{x} \delta \hat{x} \rangle$$

$$+ \langle \delta \hat{z} \delta \hat{z} \rangle - V''(X) \langle \delta \hat{z}^2 \rangle,$$

$$\langle \delta \hat{p}^2 \rangle = - V''(X) \langle \delta \hat{x} \delta \hat{p} + \delta \hat{p} \delta \hat{x} \rangle + \langle \delta \hat{p} \delta \hat{z} + \delta \hat{z} \delta \hat{p} \rangle$$

$$- \frac{1}{2} \langle \delta \hat{x}^2 \delta \hat{z}^2 + \delta \hat{z}^2 \delta \hat{p} \rangle,$$
\[ \langle \delta p \delta \dot{z} + \delta z \delta \dot{p} \rangle = -V''(X)\langle \delta \dot{z} \delta z \rangle - \frac{1}{2} V''(X) \times \langle \delta \dot{z}^2 \delta z + \delta z \delta \dot{z}^2 \rangle - 2\Gamma \langle \delta p^2 \rangle \]
\[ - \frac{1}{\tau_c} \langle \delta \dot{z} \delta p + \delta z \delta \dot{p} \rangle + 2\langle \delta z^2 \rangle, \tag{37} \]
\[ \langle \delta \dot{p} \delta \dot{z} + \delta z \delta \dot{p} \rangle = \langle \delta \dot{p} \delta z + \delta z \delta \dot{p} \rangle - \frac{\Gamma}{\tau_c} \langle \delta \dot{z} \delta p + \delta z \delta \dot{p} \rangle \]
\[ - \frac{1}{\tau_c} \langle \delta \dot{z} \delta p + \delta z \delta \dot{p} \rangle, \tag{38} \]
\[ \langle \delta z^2 \rangle = - \frac{\Gamma}{\tau_c} \langle \delta \dot{z} \delta p + \delta z \delta \dot{p} \rangle - \frac{2}{\tau_c} \langle \delta z^2 \rangle. \tag{39} \]

Discarding the third order terms in Eqs. (34)–(39) we obtain a set of closed equations which can be solved numerically along with Eqs. (15) and (16), using suitable initial conditions for calculation of the leading (second) order contribution to quantum correction terms in \( Q(t) \) of Eq. (20), i.e., \(- (1/2) V''(X) \delta \dot{z}^2(t)\). A standard choice of initial conditions corresponding to minimum uncertainty states is \( \langle \delta z^2 \rangle_{z=0} = 1/2, \langle \delta \dot{p}^2 \rangle_{p=0} = 1/2 \) and \( \langle \delta \dot{z} \delta p + \delta z \delta \dot{p} \rangle_{z=0} = 1.0 \) with the other moments [Eqs. (37)–(39)] being set at zero. The procedure may be extended to include higher order quantum effects without difficulty and may be easily adopted for numerical simulation of quantum Brownian motion as shown in Sec. IV.

3. Overdamped limit

In the quantum overdamped limit we discard the inertial term \( \delta \dot{z} \) compared to the damping term. The operator equations (21) and (22) then assume a simple form, after quantum mechanical averaging over bath states, as
\[ \gamma_0 \delta \dot{z} + V''(X) \delta \dot{z} + \sum_{n=1}^{\infty} \frac{V'(n-1)(X)}{n!} \delta \ddot{z}^n = 0, \tag{40} \]
where \( \gamma_0 \) is the dissipation constant. Equation (40) yields the quantum correction equations order by order to a very high degree. The equations up to third order are shown below in which terms of the order \( \langle \delta \dot{z}^3 \rangle \) are to be neglected for closing the system of equations:

Second order \[ \langle \delta \dot{z}^2 \rangle = - \frac{2V''(X)}{\gamma_0} \langle \delta \dot{z}^2 \rangle - \frac{2V'''(X)}{2! \gamma_0} \langle \delta \dot{z}^3 \rangle; \tag{41} \]

Third order \[ \langle \delta \dot{z}^3 \rangle = - \frac{3V''(X)}{\gamma_0} \langle \delta \dot{z}^3 \rangle - \frac{3V'''(X)}{2! \gamma_0} \langle \delta \dot{z}^4 \rangle. \tag{42} \]

We have made use of the above scheme in our recent treatment of quantum Smoluchowskii equation as discussed in Ref. 18.

4. Markovian limit

In this limit \( \tau_c \rightarrow 0 \) so that \( \gamma(t) \) in Eq. (22) reduces to a delta function \( \gamma(t) = 2\Gamma \delta(t) \), where \( \Gamma \) is the Markovian limit of dissipation. The operator equations (21) and (22), after quantum mechanical averaging over bath states, then reduce to
\[ \delta \dot{z} = \delta \dot{p}, \tag{43} \]
\[ \delta \dot{p} = - \Gamma \delta \dot{p} - V''(X) \delta \dot{z} - \sum_{n=2}^{\infty} \frac{1}{n!} V'(n-1)(X) \delta \ddot{z}^n = 0. \tag{44} \]

Making use of the above equations we derive the quantum correction equations up to the third order as
\[ \langle \delta \dot{z}^2 \rangle = \langle \delta \dot{z} \delta \dot{p} + \delta \dot{p} \delta \dot{z} \rangle, \tag{45} \]
\[ \langle \delta \dot{z} \delta \dot{p} + \delta \dot{p} \delta \dot{z} \rangle = - 2V''(X) \langle \delta \dot{z}^2 \rangle - V'''(X) \langle \delta \dot{z}^3 \rangle, \tag{46} \]
\[ \langle \delta \dot{p}^3 \rangle = - 2\Gamma \langle \delta \dot{p}^2 \rangle - V''(X) \langle \delta \dot{z} \delta \dot{p} + \delta \dot{p} \delta \dot{z} \rangle \]
\[ - V'''(X) \langle \delta \dot{z}^3 \rangle, \tag{47} \]
\[ \langle \delta \dot{p} \delta \dot{z} \delta \dot{p} \rangle = - 3\langle \delta \dot{p}^3 \rangle - 3V''(X) \langle \delta \dot{z} \delta \dot{p} \delta \dot{p} \rangle + V'''(X) h^2, \tag{48} \]
\[ \langle \delta \dot{p} \delta \dot{z} \delta \dot{p} \rangle = - \Gamma \langle \delta \dot{p} \delta \dot{z} \delta \dot{p} \rangle + 2\langle \delta \dot{p} \delta \dot{z} \delta \dot{p} \rangle \]
\[ - V'''(X) \langle \delta \dot{z}^3 \rangle, \tag{49} \]
\[ \langle \delta \dot{p} \delta \dot{z} \delta \dot{p} \rangle = - 2\Gamma \langle \delta \dot{p} \delta \dot{z} \delta \dot{p} \rangle + \langle \delta \dot{p}^3 \rangle \]
\[ - 2V''(X) \langle \delta \dot{z} \delta \dot{p} \delta \dot{z} \rangle. \tag{51} \]

The equations of higher order can be similarly derived. Summarizing the discussions in the last two sections we now see that the quantum Langevin dynamics in \( c \)-numbers can be calculated by solving the Langevin equations (15)–(16) for quantum mechanical mean values simultaneously with the quantum correction equations [e.g., Eqs. (34)–(39) or Eqs. (45)–(51)] which describe quantum fluctuations (due to nonlinearity of the system) around the quantum mechanical mean values. The choice of quantum correction equations rests on the requirement demanded by the nature of the memory kernel and nonlinearity of the potential.

C. The canonical Wigner thermal distribution function for harmonic oscillator and fluctuation-dissipation relation

The realization of the stochastic process in a \( c \)-number formulation in terms of Eqs. (15)–(16) essentially depends on the ensemble averaging over the quantum mean values of the coordinates and momenta of the bath oscillators. This is based on a key point of the present analysis—the ansatz (11), the canonical thermal Wigner distribution for shifted harmonic oscillators, which reproduces the exact first and second moments of the \( c \)-number noise \( f(t) \). To make the perspective more clear in the context of noise properties of the bath we now briefly outline the derivation of the fluctuation-dissipation relation in three different ways.
1. Quantum statistical averaging

To make the discussion self-contained we first establish that the quantum-statistical average value of a two-time correlation function of the random force operator $\hat{F}(t)$ using standard usage of quantum-statistical averaging $\langle \cdots \rangle_{\text{QS}}$ can be written in the form (6)

$$\frac{1}{2} \langle \hat{F}(t) \hat{F}(t') + \hat{F}(t') \hat{F}(t) \rangle_{\text{QS}}$$

$$= \sum_j \frac{1}{2} \kappa_j \hbar \omega_j \left( \coth \frac{\hbar \omega_j}{2k_B T} \right) \cos \omega_j (t-t'),$$

(52)

where $\hat{F}(t)$ is defined by Eq. (3). Starting from a canonical distribution of bath oscillators at $t=0$ with respect to the free bath oscillator Hamiltonian $\hat{H}_0$, with shifted oscillator coordinates $\hat{q}_j(0) = \hat{q}_j(0) - \hat{F}(0)$ as stated earlier in Sec. II, we construct the expectation value of an arbitrary operator according to Eq. (7). As usual Trace implies both quantum mechanical averaging with respect to number states of the oscillators $\Pi_{n_1=0}^{\infty} |n_1\rangle \Pi_{n_2=0}^{\infty} |n_2\rangle \cdots \Pi_{n_N=0}^{\infty} |n_N\rangle$ and the system state $|\phi\rangle$ followed by Boltzmann statistical averaging. Making use of the definition of the quantum statistical average Eq. (7) we first derive

$$\langle \hat{q}_j'(0) \hat{q}_k'(0) \rangle_{\text{QS}} = \frac{\langle \hat{p}_j(0) \hat{p}_k(0) \rangle_{\text{QS}}}{\omega_j^2}$$

$$= \delta_{jk} \frac{\hbar \coth \left( \frac{\omega_j \hbar / 2k_B T}{2} \right)}{2 \omega_j},$$

(53)

and

$$\langle \hat{q}_j'(0) \hat{p}_k(0) \rangle_{\text{QS}} = -\langle \hat{p}_k(0) \hat{q}_j'(0) \rangle_{\text{QS}} = \frac{i}{2} \hbar \delta_{jk}.$$  

(54)

Since the expectation value of an odd number of factors of $\hat{q}_j'(0)$ and $\hat{p}_j(0)$ vanishes and the expectation value of an even number of factors is the sum of products of the pairwise expectation values with the order of the factors preserved we note that Gaussian property of the distribution of $\hat{q}_j'(0)$ and $\hat{p}_j(0)$ is implied.

Relations (53) and (54) directly yield (52). On the other hand quantum mechanical averaging with respect to the number states results in

$$\langle \hat{F}(t) \rangle_{\text{QS}} = 0.$$  

(55)

2. The thermal Glauber–Sudarshan P-function

In this method we first calculate the quantum mechanical average of $1/2[\hat{F}(t) \hat{F}(t') + \hat{F}(t') \hat{F}(t)]$ with respect to the coherent states of the bath oscillators $|\alpha_1\rangle |\alpha_2\rangle \cdots |\alpha_N\rangle$ and system state $|\phi\rangle$ at $t=0$, where $|\alpha_i\rangle$ has been defined in the text. $|\alpha_i\rangle$ is the eigenstate of the annihilation operator $\hat{a}_i$ such that $\hat{a}_i |\alpha_i\rangle = \alpha_i |\alpha_i\rangle$, where $\hat{q}_j'(0)$ and $\hat{p}_j(0)$ are expressed as usual as $\hat{q}_j'(0) = \sqrt{\hbar / 2 \omega_j} [\hat{a}_j'(0) + \hat{a}_j'(0)]$ and $\hat{p}_j(0) = i \sqrt{\hbar \omega_j / 2} [\hat{a}_j'(0) - \hat{a}_j'(0)]$. We thus note that the coherent state averaging $\langle \cdots \rangle$ leads to (we have discarded the cross terms like $\alpha_1^* \alpha_2^*$ since they will vanish after the statistical averaging that follows in the next step)

$$\frac{1}{2} \langle \hat{F}(t) \hat{F}(t') + \hat{F}(t') \hat{F}(t) \rangle$$

$$= \sum_j \left[ \frac{\hbar}{2 \omega_j} (\alpha_j^* \alpha_j + \alpha_j^* \alpha_j + 1 + \alpha_j^* \alpha_j) \cos \omega_j t \cos \omega_j t' - \frac{\kappa_j \hbar \omega_j}{2} \left( \alpha_j^* \alpha_j - 2 \alpha_j^* \alpha_j - 1 + \alpha_j^* \alpha_j \right) \sin \omega_j t \sin \omega_j t' 
+ \frac{i \hbar}{4} \kappa_j \omega_j (2 \alpha_j^* \alpha_j - 2 \alpha_j^* \alpha_j \sin \omega_j (t-t')) \right] .$$

(56)

We now introduce thermal Glauber–Sudarshan distribution $P_{\text{GS}}$ for the statistical averaging of the quantum mechanical expectation values,

$$P_{\text{GS}} (|\alpha_j(0), \alpha_j^*(0)|)$$

$$= N \exp \left[ - \frac{|\alpha_j|^2}{\bar{n}_j} \right]$$

$$= N \exp \left[ - \frac{\langle \hat{p}_j(0)^2 + \omega_j^2 \langle \hat{q}_j'(0)^2 \rangle \rangle_{\text{QS}}}{2 \hbar \omega_j \bar{n}_j} \right] ,$$

(57)

where $N$ is the normalization constant. Noting that with the distribution (57) the normalized statistical averages $\langle \cdots \rangle_{\text{GS}}$ are given by

$$\langle \alpha_j^2 \rangle_{\text{GS}} = \langle \alpha_j^* \alpha_j^* \rangle_{\text{GS}} = 0 \quad \text{and} \quad \langle \alpha_j \alpha_j^* \rangle_{\text{GS}} = \bar{n}_j \delta_{jk} .$$

(58)

With relations (58), the statistical average (with respect to the $P$-function) of the quantum mechanical mean value (56) leads us to

$$\frac{1}{2} \langle \hat{F}(t) \hat{F}(t') + \hat{F}(t') \hat{F}(t) \rangle_{\text{GS}}$$

$$= \frac{1}{2} \sum_j \kappa_j \hbar \omega_j \left( \coth \frac{\hbar \omega_j}{2k_B T} \right) \cos \omega_j (t-t')$$

(59)

and

$$\langle \hat{F}(t) \rangle_{\text{GS}} = 0.$$  

(60)
3. The canonical thermal Wigner distribution function

In this method we bypass the operator ordering of \( \hat{F}(t) \)s. Instead, we start from coherent state average of the noise operator \( \hat{F}(t) \), as given by Eq. (9), i.e., the \( c \)-number noise

\[
f(t) = \sum_j \left[ \langle \{ \hat{q}_j(t) \} \rangle - \langle \hat{q}(0) \rangle \right] \kappa_j \cos \omega_j t
+ \langle \hat{p}_j(0) \rangle \kappa_j^{1/2} \sin \omega_j t
\]  

(61)

and then calculate the product of two-time \( c \)-numbers \( f(t) f(t') \).

Expressing \( \langle \hat{q}_j^\dagger(0) \rangle \) and \( \langle \hat{p}_j(0) \rangle \) in terms of \( \alpha_j \) and \( \alpha_j^* \) as stated earlier, we may write

\[
f(t) f(t') = \sum_j \left[ \kappa_j \sqrt{\frac{\hbar}{2 \omega_j}} \left( \alpha_j^* + \alpha_j \right) \cos \omega_j t
+ i \sqrt{\frac{\hbar \omega_j}{2}} \omega_j (\alpha_j^* - \alpha_j) \sin \omega_j t \right]
\]

\[
\times \left[ \sum_k \left( \kappa_k \sqrt{\frac{\hbar}{2 \omega_k}} \left( \alpha_k^* + \alpha_k \right) \cos \omega_k t'
+ i \sqrt{\frac{\hbar \omega_k}{2}} \omega_k (\alpha_k^* - \alpha_k) \sin \omega_k t' \right) \right].
\]  

(62)

We now introduce the canonical thermal Wigner distribution function (11) for ensemble averaging \( \langle \cdots \rangle_S \) over the product \( f(t) f(t') \)

\[
P_{\alpha_j(0), \alpha_j^*(0)} = N \exp \left[ -\left( \frac{\alpha_j^2}{\bar{n}_j + 1/2} \right) \right]
\]

(63)

Noting that

\[
\langle \alpha_j^2 \rangle_S = \langle \alpha_j^2 \rangle_S = 0 \quad \text{and} \quad \langle \alpha_j \alpha_k^* \rangle_S = (\bar{n}_j + 1/2) \delta_{jk},
\]

we obtain on statistical averaging over \( f(t) f(t') \),

\[
\langle f(t) f(t') \rangle_S = \frac{1}{2} \sum_j \kappa_j \hbar \omega_j \left( \coth \frac{\hbar \omega_j}{2 k_B T} \right) \cos \omega_j (t-t').
\]

(65)

Several points are now in order. First, we note that the fluctuation-dissipation relation has been expressed in several different but equivalent forms in Eqs. (52), (59), and (65). Because of the prescription of operator ordering, the left-hand sides of Eqs. (52) and (59) do not permit us to identify a \( c \)-number noise term. Clearly Eq. (65) suits this purpose and quantum noise as a number \( f(t) \) can be identified in Eq. (65). The ensemble averaging procedure with Wigner thermal distribution function is therefore followed in this work. Second, the Wigner distribution (11) is distinctly different from the thermal Glauber–Sudarshan distribution (57) because of a vacuum term \( 1/2 \) in the width of the distribution (11). Third, the origin of distinction of the methods II and III lies in the fact that in the latter case we need not carry out an operator ordering of noise as in the former case since we are essentially dealing with a correlation of numbers rather than operators. The uncertainty principle is taken care of through the vacuum term \( 1/2 \) in the width of the distribution (11) rather than through operator ordering as in methods I and II. Fourth, the Wigner distribution (11) as well as the thermal Glauber–Sudarshan distribution (57) reduces to classical Maxwell–Boltzmann distribution in the limit \( \hbar \omega_j \ll k_B T \).

Equation (57) becomes singular in the vacuum limit \( \bar{n} \rightarrow 0 \). The distribution (11) on the other hand remains a valid non-singular distribution of quantum mean values whose width is simply the natural width due to nonthermal vacuum fluctuations. The thermal Wigner distribution then goes over to pure state Wigner distribution for the ground state in the form,

\[
P(q_j, p_j) = N_1 \exp \left[ -\frac{\langle \hat{p}_j(0) \rangle^2 + \omega^2 \langle \hat{q}_j^2(0) \rangle}{2 \hbar \omega_j} \right].
\]  

(66)

\( N_1 \) is the normalization constant which is always a positive definite function in contrast to its excited state counterparts. This is because, according to Wigner et al., “the incoherence induced by a finite temperature leads to a much smoother distribution function.”

III. APPLICATION TO HARMONIC OSCILLATOR

In order to make a consistency check of the above formulation of quantum Brownian motion in \( c \)-numbers, it is pertinent to consider the case of damped harmonic oscillator. The problem has been treated over many years by various group of workers using several methods, notably by path integrals. For a review we refer toGrabert et al. Although exact, we emphasize that because of using different kinds of initial conditions and frequency dependent friction and cutoff frequencies the results obtained in several cases tend to vary. Our aim here is to obtain the result for arbitrary (decaying) memory kernel given that the initial states of the system and the bath are product separable and to show that it corresponds with the standard results known in the literature.

To proceed further we return to our basic equations (15) and (16), which for the case of harmonic oscillator reduce to the following form:

\[
\dot{X} = P,
\]

(67)

\[
\dot{P} = -i \int_0^t dt' \gamma(t-t') P(t') - \omega^2 X + f(t).
\]  

(68)

Here \( \omega \) is the frequency of the harmonic oscillator. Evidently we have \( Q = 0 \). The \( c \)-number noise \( f(t) \) obeys the usual properties given by Eqs. (13) and (14). The solution \( X(t) \) of Eqs. (67) and (68) using the Laplace transform technique is given by

\[
X(t) = P(0) G(t) + X(0) \tilde{G}(t) + \int_0^t dt' G(t-t') f(t'),
\]

(69)
where $\dot{G}(t) = 1 - \omega^2 \int_0^t G(t') dt'$; $G(t)$ is defined as the Laplace transform of $G(s)$ as given earlier in Eq. (26). On squaring and statistical averaging $\langle \cdot \rangle_S$ we obtain,

$$\langle X^2(t) \rangle_S = P^2(0)G^2(t) + X^2(0)G^2(t)$$
$$+ 2P(0)X(0)G(t)\dot{G}(t)$$
$$+ 2\int_0^t dt' \int_0^{t'} dt'' G(t-t')$$
$$\times G(t' - t'')c(t'-t'').$$

Equation (70) Here the above expression is exactly equivalent to the standard result, for example,5 can be shown by an analysis of the Markovian limit of Eq. (56). The above expression may be put further in the following form after a little bit of algebra,

$$\langle \dot{x}^2(t) \rangle_S = 2(\dot{x}^2(0))G^2(t) + (\dot{x}^2(0))G^2(t) + G(t)\dot{G}(t)(\dot{\delta}(0)\delta(0) + \delta(0)\dot{\delta}(0))$$
$$+ 2P(0)X(0)G(t)\dot{G}(t)$$
$$+ 2\int_0^t dt' \int_0^{t'} dt'' G(t-t')G(t' - t'')c(t'-t'').$$

(71)

Equation (72) is the general expression for quantum statistical average $\langle \dot{x}^2(t) \rangle_S$. In the standard notation the above expression can be written as

$$\langle \dot{x}^2(t) \rangle_S = (\dot{\rho}^2(0))G^2(t) + (\dot{\delta}^2(0))G^2(t)$$
$$+ G(t)\dot{G}(t)(\dot{\rho}(0)\delta(0)$$
$$+ \dot{\delta}(0)\rho(0))$$
$$+ 2\int_0^t dt' \int_0^{t'} dt'' G(t-t')G(t' - t'')c(t'-t'').$$

(73)

Equation (72) or Eq. (73) is the central result of this section. This expresses the quantum statistical average of $\dot{x}^2(t)$ for a damped harmonic oscillator in the non-Markovian regime. That the above expression is exactly equivalent to the standard result, for example,5 can be shown by an analysis of Markovian limit of Eq. (73). Since in this limit the noise is $\delta$-correlated and we have,

$$\langle f(t)f(t') \rangle_S = c(t-t') = \Gamma \hbar \omega \coth \frac{\hbar \omega}{2k_B T} \delta(t-t')$$

(74)

and

$G(t) = \frac{1}{\xi} \sin(\xi t)e^{-\Gamma t/2}$

(75)

with $\xi = \sqrt{\omega^2 - \Gamma^2/4}$. With these simplifications the last term in Eq. (73) results in

$$2\int_0^t dt' \int_0^{t'} dt'' G(t-t')G(t' - t'')c(t'-t'')$$
$$= \Gamma \hbar \omega \coth \frac{\hbar \omega}{2k_B T} \int_0^t G^2(t') dt'$$
$$= I_0 \quad \text{(say)}.$$
We now take care of the following limits for a consistency check. As \( t \to 0 \) we have
\[
\dot{G} = 1, \quad \dot{G} = -\Gamma, \quad G = 0, \quad \text{and} \quad S(0) = \langle \hat{x}^2 \rangle_{QS}, \quad S_i(0) = 0, \quad \text{as} \quad t \to \infty \quad \text{we have} \quad G = \dot{G} = \dot{G} = 0. \tag{83}
\]
From Eqs. (79) and (85) it follows immediately that the thermal equilibrium averages of \( \hat{x}^2 \) and \( \hat{p}^2 \) are given by Eqs. (81) and (82) derived earlier by others, e.g., Caldeira and Leggett.\(^5\) Equation (79) is exactly identical to Eq. (8.29) of Grabert et al.\(^5\) in the Markovian limit for product separable initial states of the system and the heat bath. Furthermore, Eq. (84) also satisfies their Eqs. (6.65) and (6.66).\(^5\)

Similarly one can calculate \( \langle \hat{p}^2(t) \rangle_{QS} \) as
\[
\langle \hat{p}^2(t) \rangle_{QS} = \langle \hat{p}^2 \rangle_{S} + \langle \delta \hat{p}^2 \rangle. \tag{86}
\]
Repeating the procedure in a similar way we can show that the above expression may be written as a sum of contributions from thermal average value and quantum dispersion. We quote the final results as below:
\[
\langle \hat{p}^2(t) \rangle_{QS} = \sum_{i,j=1}^{2} \langle \hat{x}_i(0) \hat{x}_j(0) \rangle + \langle \hat{p}^2 \rangle_{QS} \bar{G}^2(t) + 1 \bigg] - \frac{S^2(t)}{\langle \hat{x}^2 \rangle_{QS}} + 2 \dot{G}, \tag{87}
\]
where
\[
\hat{x}_1 = G \hat{x}, \quad \hat{x}_2 = \hat{G} \hat{p}.
\]
Equation (87) corresponds to Eq. (8.26) of Grabert et al.\(^5\) for factorized initial condition in the Markovian limit.

IV. A NUMERICAL SCHEME FOR SOLUTION OF \( c \)-NUMBER LANGEVIN EQUATION

Equation (14), the fluctuation-dissipation relation, is the key element for generation of quantum noise. The left-hand side of this equation suggests that \( f(t) \) is a “classical” random number and \( (f(t)f(t'))_S \) is a correlation function which is classical in form but quantum-mechanical in its content. Since one of the most commonly occurring noise processes is exponentially correlated color noise, we develop the scheme first for this process. This is based on a simple idea that \( c \)-number quantum noise may be viewed as a superposition of several Ornstein–Uhlenbeck noise processes. It may be noted that Imamoğlu,\(^33\) a few years ago, has produced a scheme for approximating a class of reservoir spectral functions by a finite superposition of Lorentzian functions with positive coefficients corresponding to a set of fictitious harmonic oscillator modes to incorporate non-Markovian effects into the dynamics. The method is an extension of the stochastic wave function method in quantum optics. The present superposition method is reminiscent of this approach in the time domain. We however emphasize two important points. Because of rotating wave and Born approximations Imamoğlu’s scheme based on master equation is valid for weak system-reservoir coupling, i.e., weak damping regime. The present method on the other hand uses a superposition of noises in the time domain and the quantum Langevin equation is valid for arbitrary coupling since no weak coupling approximation or rotating wave approximation has been used in the treatment. Second, while the stochastic wave function approach as well as the density operator equation have no classical analog, the present scheme makes use of \( c \)-numbers to implement the classical numerical method quite freely to solve quantum Langevin equations (15)–(16).

A. Superposition method

We proceed in several steps as follows:
We first denote the correlation function \( c(t-t')(= \langle f(t)f(t') \rangle_S) \) in the continuum limit as
\[
c(t-t') = \frac{1}{2} \int_0^\infty d\omega K(\omega) \rho(\omega) \hbar \omega \coth \left( \frac{\hbar \omega}{2 k_B T} \right) \times \cos \omega(t-t'), \tag{88}
\]
where we have introduced the density function \( \rho(\omega) \) for the bath modes. In principle an \( a \) priori knowledge of \( K(\omega)\rho(\omega) \) is essential and a primary factor for determining the evolution of stochastic dynamics governed by Eqs. (15) and (16). We assume a Lorentzian distribution of modes given by Eq. (29) where \( \Gamma \) and \( \tau_c \) are the dissipation in the Markovian limit and correlation time, respectively. These two quantities and the temperature \( T \) are the three input parameters for our problem. Equation (29) when used in Eq. (4) in the continuum limit yields an exponential memory kernel (30). For a given set of these parameters we first numerically evaluate the integral (88) as a function of the time. A typical profile of such a correlation is shown in Fig. 1 for three different temperatures (solid lines) and for \( \Gamma = 1 \) and \( \tau_c = 1 \).

In the next step we numerically fit the correlation function with a superposition of several exponentials as
\[
c(t-t') = \sum_{i=1}^{3} \frac{D_i}{\tau_i} \exp \left( \frac{-|t-t'|}{\tau_i} \right) \quad i=1,2,3,... \tag{89}
\]

The set \( D_i \) and \( \tau_i \), the constant parameters are thus known. In Fig. 1 we exhibit these fitted curves (dotted lines) against the corresponding solid curves calculated by numerical integration of Eq. (88). It may be checked that at high temperature the required fitting curve is, in general, a single exponential, whereas at low temperatures down to absolute zero biexponential fitting is an excellent description. At the intermediate temperature one has to use even triexponential fitting, in general.

The numerical agreement between the two sets of curves based on Eqs. (88) and (89) in Fig. 1 suggests that one may generate a set of exponentially correlated color noise variables \( \eta_i \) according to
\[
\eta_i = -\frac{\eta_i}{\tau_i} + \frac{1}{\tau_i} \xi_i(t), \tag{90}
\]
where
\[
\langle \xi_i(t) \rangle = 0. \tag{91}
\]
\[
\langle \xi_i(0) \xi_j(\tau) \rangle = 2D_i \delta_{ij} \delta(\tau) \quad (i = 1, 2, 3, \ldots),
\]

in which \( \xi_i(t) \) is a Gaussian white noise obeying Eqs. (91) and (92); \( \tau_i \) and \( D_i \) are determined from numerical fit. The quantum noise \( \eta_i \) is thus an Ornstein–Ulhenbeck process with properties

\[
\langle \eta_i(t) \rangle = 0, \quad \langle \eta_i(t) \eta_j(t') \rangle = \delta_{ij} \frac{D_i}{\tau_i} \exp \left( -\frac{|t-t'|}{\tau_i} \right).
\]

Clearly \( \tau_i \) and \( D_i \) are the correlation time and the strength of color noise variables \( \eta_i \), respectively. It is also important to point out that they have nothing to do with true correlation time \( \tau_c \) and strength \( \Gamma \) of the noise \( f(t) \).

The \( c \)-number quantum noise \( f(t) \) due to the heat bath is therefore given by

\[
f(t) = \sum_{i=1,\ldots,n} \eta_i.
\]

Equation (95) implies that \( f(t) \) can be realized as a sum of several Ornstein–Ulhenbeck noises \( \eta_i \) satisfying

\[
\langle f(t)f(t') \rangle_S = \sum_i \langle \eta_i(t) \eta_i(t') \rangle.
\]

Having obtained the scheme for generation of \( c \)-number quantum noise \( f(t) \) we now proceed to solve the quantum Langevin equations (15) and (16) along with the quantum correction equations (34)–(39). To this end the first step is the integration of the stochastic differential equation (90). We note that to generate Gaussian white noise \( \xi_i(t) \) of Eq. (90) from two random numbers which are uniformly distributed on the unit interval, Box–Muller algorithm may be used. To be specific we start the simulation with integration of Eq. (90) to generate an initial value of \( \eta_i \) as follows:

\[
m = \text{random number},
\]

\[
n = \text{random number},
\]

\[
\eta_i = \left[ -\frac{2D_i}{\tau_i} \ln(m) \right]^{1/2} \cos(2\pi n).
\]

We then set \( E_i = \exp(-\Delta t/\tau_i) \), where \( \Delta t \) is the integration step length for Eq. (90). Following the integral algorithm of Fox et al.,\textsuperscript{33} we now generate the next step \( \eta_i \) from \( \eta_i \) according to the following lines:

\[
a = \text{random number},
\]

\[
b = \text{random number},
\]

\[
h_i = \left[ -\frac{2D_i}{\tau_i} (1 - E_i^2) \ln(a) \right]^{1/2} \cos(2\pi b)
\]

and

\[
\eta_i |_{t+\Delta t} = \eta_i E_i + h_i.
\]

After Eq. (102) the algorithm is repeated back to Eq. (99) and continued for a sufficient time evolution.

In the next step we couple the above-mentioned integral algorithm for solving Eq. (90) to standard fourth order Runge–Kutta (or other) algorithm for the solution of Eqs. (15), (16), and (34)–(39). For an accurate and fast algorithm \( \eta \)-integration (102) may be carried out at every \( \Delta t/2 \) step, while for Runge–Kutta integration of Eqs. (15)–(16) and (34)–(39) step size \( \Delta t \) is a good choice. With \( \Delta t \) in the range \(-0.001\sim-0.01\) the overall method gives excellent results for the solution of quantum Langevin equation.

**B. Spectral method**

The method of superposition of Ornstein–Ulhenbeck noises for generation of quantum noise as discussed above primarily pertains to situations where the correlation function can be described by a superposition of several exponentials. Although in overwhelming cases of applications in condensed matter and chemical physics at low-temperature where in general non-Ohmic conditions prevail we encounter such situations, there are cases where the noise correlation exhibits power law behavior or is Gaussian in nature. To extend the present scheme we now show how the spectral method can be implemented for generation of quantum noise. The method had been used earlier by several groups\textsuperscript{25–28} in the context of classical long and short range temporally correlated noises. We give here a brief outline for its extension to quantum noise.

The starting point of our analysis is the knowledge of quantum correlation function \( c(t\rangle t') \) as given in Eq. (88). Again this depends on \( \kappa(\omega) \rho(\omega) \), which \[ for example, for short range Gaussian correlated noise may be taken as \( 2G \exp(-\omega^2 \tau_c^2/4), \Gamma \) and \( \tau_c \) being the strength and the correlation time, respectively \] must be known \textit{a priori}. Having known \( c(t\rangle t') \) as a function of time we first carry out its Fourier transform

\[
\langle f(t)f(t') \rangle_S = c(t\rangle t'),
\]

\[
C(\omega) = \int \exp(-i\omega t)c(t)dt
\]

by discretizing the time in \( N=2^n \) intervals of size \( \Delta t \) (such that \( \Delta t \) is shorter than any other characteristic time of the system). The result is a string of discrete numbers \( C(\omega_n) \). In discrete Fourier space the noise correlation is given by
\[
\langle f(\omega_n)f(\omega_m') \rangle = N \Delta t C(\omega_n) \delta_{m+n,0}.
\]
(104)

Therefore the noise in the Fourier domain can be constructed simply as

\[
f(\omega_n) = \sqrt{N \Delta t C(\omega_n)} \alpha(\omega_n),
\]
(105)

where

\[
i = 0, 1, 2, ..., N; \quad f(\omega_0) = f(\omega_n); \quad \omega_n = \frac{2\pi n}{N \Delta t}.
\]

Here \(\alpha_n\) are the Gaussian complex random numbers with zero mean and anti-delta correlated noise, such that \(\alpha_n = a_n + ib_n\); \(a_n\) and \(b_n\) (the real and imaginary parts of \(\alpha_n\)) are Gaussian random numbers with zero mean value and variance \(1/2\).

\[
\langle a_n \rangle = \langle b_n \rangle = 0; \quad (106)
\]

\[
\langle a_n^2 \rangle = \langle b_n^2 \rangle = \frac{1}{2}, \quad \text{where} \ n \neq 0;
\]
(107)

and

\[
a_0^2 = 1. \quad (108)
\]

The Box–Mueller algorithm may again be used to generate the set of random numbers \(a_n, b_n\) (and consequently \(\alpha_n\)) according to the lines,

\[
p_n = \text{random number}, \quad (109)
\]

\[
q_n = \text{random number}, \quad (110)
\]

\[
a_n = (- \ln p_n)^{1/2} \cos 2\pi q_n, \quad (111)
\]

and similarly for \(b_n\). The discrete Fourier transform of the string \(f(\omega_n)\) is then numerically calculated by the fast Fourier transform algorithm to generate the quantum random numbers \(f(t_n)\). The recipe can be implemented for solving Eqs. (15)–(16) and (34)–(39) as discussed in the earlier subsection.

**V. NUMERICAL APPLICATION TO NONLINEAR OSCILLATOR: BARRIER CROSSING DYNAMICS**

We now present the numerical results on \(c\)-number quantum Langevin equation for a cubic potential. Our concern here is to solve Eqs. (15)–(16) for quantum mean values \(X(=\langle \hat{x} \rangle)\) and \(P(=\langle \hat{p} \rangle)\). The quantum dispersion around the mean path are taken into account by following the equations of quantum corrections, e.g., Eqs. (34)–(39) or Eqs. (45)–(51) etc., depending on the situation. Since the system is coupled to heat bath it is also necessary to take statistical mean over several thousands of realizations of trajectories of the quantum mechanical mean values. One of the major aims of this numerical simulation is to show that the method described in Sec. IV A can be implemented very easily to describe both thermally activated processes \(^{1,2,11,14–19,29–32}\) and tunneling within a unified scheme.

![FIG. 2. Plot of quantum turnover at two different temperatures.](image)

To begin with we consider a potential of the form

\[
V(X) = -\frac{1}{2}AX^3 + BX^2,
\]
(112)

where \(A\) and \(B\) are the constants to be chosen for a fixed set of barrier height \(V_0\) and the frequencies in the well \(\omega_0\) and at the barrier top \(\omega_b\). The other input parameters for our calculations are temperature \(T\), strength of noise correlation \(\Gamma\) and correlation time \(\tau_c\). For the given potential, \(Q(t)\) is given by a single quantum correction term \(Q = A\langle \delta \hat{\delta}^2 \rangle\) according to Eq. (20). Theoretical consideration demands that the activation barrier \(V_0\) must be much larger than the thermal noise, i.e., \(V_0 \gg k_B T\). In order to ensure the stability of the algorithm we have kept \(\Delta t/\tau_c \ll 1\), where \(\Delta t\) is the integration step size. For a given parameter set we have computed the time for the quantum Brownian particle in the cubic potential starting from the bottom of the metastable well at \(x = 0\) to reach the maximum at \(x_0 = 2B/A\) for a single realization of a stochastic path. The statistical average time to reach the barrier top is calculated by averaging over 5000 trajectories. We have used \(h = k_b = 1\) for the entire treatment. \(V_0\) is expressible as \(V_0 = 8B^2/3A^2\), or writing \(\omega_0^2 = 2B\), we have \(V_0 = \omega_0^2/3A^2\).

To test the efficacy of the method, we now present and compare with others the results obtained by the simulation of the quantum damped cubic potential (112) driven by quantum noise using Eqs. (15), (16), (45)–(51), and (90). Our results for quantum Kramers’ turnover for the cubic potential is shown in Fig. 2 for Ohmic dissipation on a logarithmic scale. We choose \(V_0 = 10k_B T\) and plot (dotted lines) the barrier crossing rate \(k\) (reciprocal of the time for crossing) divided by the transition state theory result \(k_{\text{TST}}\) at the two different temperatures for the values \(\hbar \omega_0/k_B T = 2.5\) (upper dotted line) and \(\hbar \omega_0/k_B T = 1.0\) (lower dotted line) against the scaled dissipation parameter \(\log(\Gamma/\omega_0)\). The results (normalized with respect to Ref. 21) have been compared with the analytical results of Rips et al. (Fig. 4 of Ref. 21) obtained by a completely different scheme.\(^{33,34}\) The agreement is found to be quite satisfactory. An important advantage of the present method is that it is equipped to deal with vacuum field assisted tunneling as well as the activated tunneling.
within an unified description since the Wigner distribution remains a valid nonsingular distribution even at absolute zero and quantum correction to nonlinear potential can be calculated significantly order by order. In Fig. 3 we exhibit (dotted lines) quantum Arrhenius rate in terms of \( \log(k/\omega_0) \) as a function of \( \hbar \omega_0 / k_B T \) for the cubic potential with barrier height \( V_0 = 5\hbar \omega_0 / \omega_0 = 2 \) and frequency independent damping \( \Gamma = 2\omega_0 \alpha \) for two \( \alpha \)-values, \( \alpha = 0.5 \) and \( \alpha = 1.0 \). The quantum corrections have been calculated by employing Eqs. (45)–(51). The normalized results have been compared with those of Grabert et al. (Fig. 10 of Ref. 35). It is apparent that at high temperature, the behavior is linear corresponding to the century-old Arrhenius classical result, significant quantum enhancement occurs at low temperature which has the usual form of \( 1/T^2 \). We thus observe that quantum Kramers’ turnover and quantum Arrhenius rate are in close conformity with the earlier results. The numerical method for solving quantum Langevin equation as worked out here is thus capable with dealing with the problem of dynamical theory of reaction rate in terms of a simple algorithm which mimics its classical counterpart and is independent of path integral approaches.

VI. CONCLUSION

In this paper we have proposed a simple numerical method within several approximation schemes for solution of the generalized quantum c-number Langevin equation. The method is based primarily on two aspects. First, it has been shown that it is possible to realize quantum noise (in a quantum Langevin dynamics) which satisfies quantum fluctuation-dissipation relation as c-numbers rather than as operators. Second, the quantum correction terms which appear due to the nonlinearity of the potential of the system, can be generated systematically order by order, so that the coupled set of equations for the stochastic dynamics of the system and the correction terms can be solved easily with sufficient degree of accuracy. The method of simulation is equipped to deal with arbitrary correlation time, temperature and damping strength. The mapping of quantum stochastic dynamics as a problem of classical simulation as presented here allows us to implement classical methods in quantum simulations. The method is much simpler to handle compared to path integral quantum Monte Carlo as employed earlier in the problem of barrier crossing dynamics.\(^{11} \)

We have shown that the exact agreement of our results on damped harmonic oscillators for arbitrary memory kernel and damping strength with the standard results has served as consistency check of the present theoretical framework. Second, our simulation results on quantum barrier crossing dynamics for a particle in a cubic metastable potential agree well with the results obtained earlier. We hope that with the prescription given, many problems with nonlinear potentials where the time correlation may not follow a typical prescribed dynamics can be simplified within the framework of this simulation.

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