

# Phase induced current in presence of nonequilibrium bath: A quantum approach

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Based on a system-reservoir nonlinear coupling model, where the associated bath is externally driven by a fluctuating force, we present a microscopic approach to quantum state-dependent diffusion and multiplicative noises in terms of a quantum (Markovian) Langevin equation in overdamped limit when the associated bath is in nonequilibrium state. We then explore the possibility of observing a quantum current when the bath is modulated by white noise, the phenomena which is absent in the classical regime. © 2008 American Institute of Physics.

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## I. INTRODUCTION

Thermal diffusion in a periodic potential has a prominent role in various systems such as Josephson's junction,<sup>1</sup> system for diffusion in crystal surfaces,<sup>2</sup> noisy limit oscillators,<sup>3</sup> etc. There has been a renewed interest in recent times in the study of transport properties of Brownian particles moving in a periodic potential<sup>4</sup> with special emphasis on coherent transport and giant diffusion.<sup>5</sup> These studies have been motivated in part by the attempt to understand the mechanism of movement of protein motors in biological system.<sup>6</sup> Several physical models have been proposed to understand the transport phenomena in such systems, such as vibrational ratchet,<sup>7</sup> rocking ratchet,<sup>8</sup> flashing ratchet,<sup>5</sup> diffusion ratchet,<sup>9,10</sup> correlation ratchet,<sup>11</sup> etc. Such ratchet models have a wide range of applications in biology and nanoscopic systems<sup>12</sup> because of its extraordinary success in exploring experimental observations on biochemical molecular motors, active in muscle contraction,<sup>13</sup> observation on directed transport in photovoltaic and photorefective materials,<sup>14</sup> etc. In all the above models the potential is taken to be asymmetric in space. One can also obtain a unidirectional current in the presence of spatially symmetric potential. For such nonequilibrium systems one requires time asymmetric random forces<sup>15</sup> or space-dependent diffusion.<sup>16–18</sup> Space dependent diffusion coefficient may arise either due to space-dependent friction coefficient<sup>18</sup> or space-dependent temperature. Frictional inhomogeneities are common in superlattice structure, semiconductors, or motion in porous media. Particles moving close to a surface<sup>19</sup> and molecular motor proteins moving along the periodic structures or microtubule also experience space-dependent friction.<sup>20</sup>

In 1987 Büttiker<sup>18</sup> had shown that in the case of space-dependent friction in the overdamped limit, a classical particle under a symmetric sinusoidal potential field and also in

the presence of a sinusoidally modulated space-dependent diffusion with some periodicity experiences a net drift force resulting in generation of current. This current is basically due to the phase difference between the symmetric periodic potential and the space-dependent diffusion. The current does vanish when the phase difference is either zero or integral multiples of  $\pi$ . van Kampen<sup>21</sup> in a latter work also ended up with a similar kind of conclusion for a system with space-dependent temperature under the overdamped condition. The result of van Kampen is a re-examination of the earlier observation due to Landaure<sup>22,23</sup> who explored the problem of characterizing nonequilibrium steady states in the transition kinetics between the two locally stable states in bistable systems.

In this paper, we address the problem of Langevin equation with multiplicative noise and state-dependent diffusion for a thermodynamically open quantum mechanical system in the context of transport phenomena. Although the quantum mechanical system—reservoir linear coupling model for microscopic description of additive noise and linear dissipation, which are related by fluctuation—dissipation relation, well known over many decades in several fields,<sup>24,25</sup> the nature of nonlinear coupling and its consequences have been explored with renewed interest only recently.<sup>26,27</sup> For example, Tanimura and co-workers<sup>28</sup> have extensively used nonlinear coupling in modeling elastic and inelastic relaxation mechanisms and their interplay in vibrational and Raman spectroscopy. Recently Ishizala and Tanimura<sup>29</sup> have proposed a quantum dissipative equation (with Gaussian Markovian noise) that has applicability to low-temperature systems strongly coupled to a harmonic bath without employing the rotating wave approximation for the system-bath coupling. Recently diffusive transport properties of a quantum Brownian particle moving in biased washboard potential have been explored by Machura *et al.*<sup>30</sup> where, apart from the average velocity, they have investigated the diffusive behavior by evaluating the effective diffusion coefficient together with the corresponding Peclet number. Based on the

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path integral formulation, quantum Brownian motion of a dissipative system has been studied in the large friction case by Ankerhold *et al.*<sup>31</sup> where they cast the nonlocal reduced dynamics into an effective equation of motion: quantum Smoluchowski equation. More recently, Ankerhold and Pollak<sup>32</sup> have suggested a mechanism by which they explored the fact that dissipative effect may enhance the transport of a quantum Brownian particle, a counterintuitive but very interesting observation.

In the present paper, we consider a system-reservoir model in quantum mechanical context where the reservoir is modulated by an external noise, and the system is nonlinearly coupled with the heat bath, thereby resulting in a nonlinear multiplicative quantum Langevin equation with state-dependent diffusion. When the reservoir is modulated by an external noise, it is likely that it induces fluctuations in the polarization of the reservoir<sup>33</sup> due to the external noise from a microscopic standpoint, and one may expect that the non-equilibrium situation created by modulating the bath may induce an asymmetry in the effective potential. It has been shown earlier by us<sup>33,34</sup> that the bath modulation by an external force creates an environment with an effective temperature that depends on the coupling constant and strength of the external noise. Here we are interested in the situation where the associated bath is modulated by an external random force to study the directed transport of a frictional ratchet system in the quantum mechanical context. The problem of particle motion in an inhomogeneous media in the presence of an external noise becomes equivalent to the problem in a space-dependent temperature and generation of unidirectional current which follows as a corollary to Landauer's blow-torch effect.<sup>22,23</sup>

A number of different situations depicting the modulation of the heat bath may be physically relevant. Although the dynamics of a Brownian particle in a uniform solvent is well known, it is not very clear when the response of the solvent will be time dependent, as in a liquid crystal,<sup>35</sup> or in diffusion and reaction in supercritical liquids,<sup>36</sup> growth in living polymerization,<sup>37</sup> etc. As another example, we may consider a simple unimolecular conversion  $A \rightarrow B$ , say, an isomerization reaction, carried out in a photochemically active solvent. Since the fluctuations in the light intensity result in fluctuations in the polarization of the solvent molecules, the effective reaction field around the reactant system gets modified. In the present work, we address the problem of quantum Langevin equation with multiplicative noise and state-dependent diffusion for a thermodynamically open system to explore the nature of nonlinear coupling and its consequences, specially the possibility of observing directed transport in a periodic potential as a consequence of state-dependent diffusion.

The organization of the paper is as follows. Following the recently developed methodology by Ray *et al.*,<sup>38</sup> in Sec. II, starting from a Hamiltonian picture of a quantum system nonlinearly coupled with a harmonic bath which is modulated by an external noise, we derive the  $C$ -number analog of quantum Langevin equation for the system mode, followed by a Smoluchowski description of the process valid for a state-dependent dissipation and multiplicative noise pro-

cesses. In Sec. III, as an application of our development, we derive the net current in a sinusoidal symmetric potential as a consequence of phase difference between the potential and the derivative of periodic coupling function. Various aspects of the current is discussed in length. The paper is concluded in Sec. IV.

## II. THE MODEL: LANGEVIN AND SMOLUCHOWSKI DESCRIPTION OF QUANTUM OPEN SYSTEM

We consider a particle of unit mass coupled to a reservoir comprised of a set of harmonic oscillators with characteristic frequencies  $\{\omega_j\}$ . Initially, i.e., at  $t=0$ , the system and reservoir is in thermal equilibrium at temperature  $T$ . At  $t=0_+$ , the bath is started to be modulated by an external noise. This is described by the Hamiltonian

$$\hat{H} = \frac{\hat{p}^2}{2} + V(\hat{q}) + \sum_{j=1}^N \left[ \frac{\hat{p}_j^2}{2} + \frac{1}{2} \omega_j^2 (\hat{x}_j - c_j f(\hat{q}))^2 \right] + \sum_j \kappa_j \hat{x}_j \epsilon(t), \quad (1)$$

where  $\hat{q}$  and  $\hat{p}$  are the coordinate and momentum operators of the system and  $\{\hat{x}_j, \hat{p}_j\}$  are the set of coordinate and momentum operators for the bath oscillators with unit mass. The system particle is coupled to the bath oscillators nonlinearly through the general coupling term  $c_j f(\hat{q})$ , where  $c_j$  is the coupling constant. The last term in (1),  $H_{\text{int}} = \sum_j \kappa_j \hat{x}_j \epsilon(t)$  represents the fact that the bath is driven by an external noise  $\epsilon(t)$  which is assumed to be stationary and Gaussian.  $\epsilon(t)$  is a classical noise with the following statistical property:

$$\langle \epsilon(t) \rangle = 0$$

and

$$\langle \epsilon(t) \epsilon(t') \rangle = 2D \Psi(t-t'). \quad (2)$$

In Eq. (2), the average is taken over each realization of  $\epsilon(t)$ .  $V(\hat{q})$  is the potential operator. The coordinates and momentum operators satisfy the standard commutation relations

$$[\hat{q}, \hat{p}] = i\hbar$$

and

$$[\hat{x}_i, \hat{p}_j] = i\hbar \delta_{ij}, \quad (3)$$

and  $D$  is the strength of the noise and  $\Psi(t)$  being some decaying correlation function.

Using the Heisenberg equation of motion for any operator  $\hat{O}$ , given by

$$\dot{\hat{O}} = \frac{i}{\hbar} [\hat{H}, \hat{O}],$$

we obtain the equations of motion for position and momentum operators as

$$\dot{\hat{q}} = \hat{p}, \quad (4)$$

$$\dot{\hat{p}} = -V'(\hat{q}) + f'(\hat{q}) \sum_j c_j \omega_j^2 [\hat{x}_j - c_j f(\hat{q})], \quad (5)$$

$$\dot{\hat{x}}_j = \hat{p}_j, \quad (6)$$

$$\dot{\hat{p}}_j = -\omega_j^2(\hat{x}_j - c_j f(\hat{q})) - \kappa_j \epsilon(t) \hat{I}, \quad (7)$$

where  $\hat{I}$  is the unit operator. In the above equations the dot ( $\dot{\cdot}$ ) denotes derivative with respect to time and the prime ( $\prime$ ) indicates derivative with respect to  $\hat{q}$ .

Now eliminating the bath variables in usual manner, we obtain the equations of motion for the system variables as

$$\begin{aligned} \dot{\hat{q}} &= \hat{p}, \\ \dot{\hat{p}} &= -V'(\hat{q}(t)) - f'(\hat{q}(t)) \int_0^t \gamma(t-t') f'(\hat{q}(t')) \hat{p}(t') dt' \\ &\quad + f'(\hat{q}(t)) \hat{\eta}(t) + f'(\hat{q}(t)) \pi(t) \hat{I}, \end{aligned} \quad (8)$$

where

$$\pi(t) = - \int_0^t dt' \varphi(t-t') \epsilon(t'), \quad (9)$$

$$\gamma(t) = \sum_{j=1}^N c_j^2 \omega_j^2 \cos \omega_j t, \quad (10)$$

$$\varphi(t) = \sum_{j=1}^N c_j \omega_j \kappa_j \sin \omega_j t, \quad (11)$$

and  $\hat{\eta}(t)$  is the internal thermal noise operator given by

$$\hat{\eta}(t) = \sum_{j=1}^N c_j \omega_j^2 \left\{ [\hat{x}(0) - c_j f(\hat{q}(0))] \cos \omega_j t + \frac{\hat{p}_j(0)}{\omega_j} \sin \omega_j t \right\}, \quad (12)$$

with the statistical properties

$$\langle \hat{\eta}(t) \rangle_{\text{QS}} = 0, \quad (13)$$

$$\begin{aligned} \frac{1}{2} \langle \hat{\eta}(t) \hat{\eta}(t') + \hat{\eta}(t') \hat{\eta}(t) \rangle_{\text{QS}} \\ = \frac{1}{2} \sum_{j=1}^N c_j^2 \omega_j^2 \hbar \omega_j \coth \left( \frac{\hbar \omega_j}{2k_B T} \right) \cos \omega_j (t-t'). \end{aligned} \quad (14)$$

Here  $\langle \cdots \rangle_{\text{QS}}$  implies quantum statistical average on bath degrees of freedom and is defined as

$$\langle \hat{O} \rangle_{\text{QS}} = \frac{\text{Tr} \left[ \hat{O} \exp \left( \frac{-H_B}{k_B T} \right) \right]}{\text{Tr} \left[ \exp \left( \frac{-H_B}{k_B T} \right) \right]} \quad (15)$$

for any bath operator  $\hat{O}(\hat{x}_j, \hat{p}_j)$ , where

$$H_B = \sum_{j=1}^N \left[ \frac{\hat{p}_j^2}{2} + \frac{1}{2} \omega_j^2 (\hat{x}_j - c_j f(\hat{q}))^2 \right] \text{ at } t=0. \quad (16)$$

Let us now carry out a quantum mechanical averaging of the operator equation (8) to get

$$\langle \hat{q} \rangle_{\text{Q}} = \langle \hat{p} \rangle_{\text{Q}}, \quad (17)$$

$$\begin{aligned} \langle \hat{p} \rangle_{\text{Q}} &= - \langle V'(\hat{q}(t)) \rangle_{\text{Q}} \\ &\quad - \left\langle f'(\hat{q}(t)) \int_0^t dt' \gamma(t-t') f'(\hat{q}(t')) \hat{p}(t') \right\rangle_{\text{Q}} \\ &\quad + \langle f'(\hat{q}) \hat{\eta}(t) \rangle_{\text{Q}} + \langle f'(\hat{q}) \rangle_{\text{Q}} \pi(t), \end{aligned} \quad (18)$$

where the quantum mechanical average  $\langle \cdots \rangle_{\text{Q}}$  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 system and  $\{|\alpha_j\rangle\}$  corresponds to the initial coherent state of the  $j$ th bath oscillator.  $\langle \hat{\eta}(t) \rangle_{\text{Q}}$  is now a classical-like noise term, which because of the quantum mechanical averaging, in general, is a nonzero number and is given by

$$\begin{aligned} \langle \hat{\eta}(t) \rangle_{\text{Q}} &= \sum_{j=1}^N \left[ c_j \omega_j^2 \left\{ [\langle \hat{x}_j(0) \rangle_{\text{Q}} - c_j \langle f(\hat{q}(0)) \rangle_{\text{Q}}] \cos \omega_j t \right. \right. \\ &\quad \left. \left. + \frac{\langle \hat{p}_j(0) \rangle_{\text{Q}}}{\omega_j} \sin \omega_j t \right\} \right]. \end{aligned} \quad (19)$$

To realize  $\langle \hat{\eta}(t) \rangle_{\text{Q}}$  as an effective  $C$ -number noise, we now introduce the ansatz<sup>38</sup> that the momenta  $\langle \hat{p}_j(0) \rangle_{\text{Q}}$  and the shifted coordinate  $(\langle \hat{x}_j(0) \rangle_{\text{Q}} - c_j \langle f(\hat{q}(0)) \rangle_{\text{Q}})$  of the bath oscillators are distributed according to the canonical distribution of Gaussian form,

$$P_j = N \exp \left\{ - \frac{\langle \hat{p}(0) \rangle_{\text{Q}}^2 + \omega_j^2 [\langle \hat{x}_j(0) \rangle_{\text{Q}} - c_j \langle f(\hat{q}(0)) \rangle_{\text{Q}}]^2}{2\hbar \omega_j (\bar{n}_j(\omega_j) + \frac{1}{2})} \right\} \quad (20)$$

so that for any quantum mechanical mean value of operator  $\langle \hat{O} \rangle_{\text{Q}}$  which is a function of bath variables, its statistical average  $\langle \cdots \rangle_{\text{S}}$  is

$$\langle \langle \hat{O} \rangle_{\text{Q}} \rangle_{\text{S}} = \int [\langle \hat{O} \rangle_{\text{Q}} P_j d\{\omega_j^2 [\langle \hat{x}_j(0) \rangle_{\text{Q}} - c_j \langle f(\hat{q}(0)) \rangle_{\text{Q}}]\} d\langle \hat{p}_j(0) \rangle_{\text{Q}}]. \quad (21)$$

In Eq. (20)  $\bar{n}_j(\omega_j)$  is the average thermal photon number of the  $j$ th bath oscillator at temperature  $T$  and is given by

$$\bar{n}_j(\omega_j) = \frac{1}{\exp \left( \frac{\hbar \omega_j}{k_B T} \right) - 1}. \quad (22)$$

The distribution  $P_j$  given by Eq. (20) and the definition of statistical average imply that the  $C$ -number noise  $\langle \hat{\eta}(t) \rangle_{\text{Q}}$  given by Eq. (19) must satisfy<sup>38</sup>

$$\langle \langle \hat{\eta}(t) \rangle_{\text{Q}} \rangle_{\text{S}} = 0, \quad (23)$$

$$\langle \langle \hat{\eta}(t) \hat{\eta}(t') \rangle_{\text{Q}} \rangle_{\text{S}} = \frac{1}{2} \sum_{j=1}^N c_j^2 \omega_j^2 \hbar \omega_j \coth \left( \frac{\hbar \omega_j}{2k_B T} \right) \cos \omega_j (t-t'). \quad (24)$$

Now to obtain a finite result in the continuum limit, the coupling functions  $c_j = c(\omega)$  and  $\kappa_j = \kappa(\omega)$  are chosen as  $c(\omega)$

$=c_0/\omega\sqrt{\tau_c}$  and  $\kappa(\omega)=\kappa_0\omega\sqrt{\tau_c}$ , respectively. Consequently  $\gamma(t)$  and  $\varphi(t)$  reduce to the following form:

$$\gamma(t) = \frac{c_0^2}{\tau_c} \int_0^\infty d\omega \mathcal{D}(\omega) \cos \omega t \quad (25)$$

and

$$\varphi(t) = c_0 \kappa_0 \int_0^\infty d\omega \omega \mathcal{D}(\omega) \sin \omega t, \quad (26)$$

where  $c_0$  and  $\kappa_0$  are constants and  $1/\tau_c$  is the cutoff frequency of the oscillators.  $\tau_c$  may be characterized as the correlation time of the bath and  $\mathcal{D}(\omega)$  is the density of modes of the heat bath which is assumed to be Lorentzian,

$$\mathcal{D}(\omega) = \frac{2}{\pi} \frac{1}{\tau_c(\omega^2 + \tau_c^{-2})}. \quad (27)$$

With these forms of  $\mathcal{D}(\omega)$ ,  $c(\omega)$ , and  $\kappa(\omega)$ ,  $\gamma(t)$  and  $\varphi(t)$  take the following form:

$$\gamma(t) = \frac{c_0^2}{\tau_c} \exp(-t/\tau_c) = \frac{\Gamma}{\tau_c} \exp(-t/\tau_c) \quad (28)$$

and

$$\varphi(t) = \frac{c_0 \kappa_0}{\tau_c} \exp(-t/\tau_c), \quad (29)$$

with  $\Gamma=c_0^2$ . For  $\tau_c \rightarrow 0$ , Eqs. (28) and (29) reduce to  $\gamma(t) = 2\Gamma \delta(t)$  and  $\varphi(t) = 2c_0 \kappa_0 \delta(t)$ , respectively. The noise correlation function (24) becomes

$$\begin{aligned} & \langle \langle \hat{\eta}(t) \hat{\eta}(t') \rangle \rangle_S \\ &= \frac{1}{2} \frac{c_0^2}{\tau_c} \int_0^\infty d\omega \hbar \omega \coth\left(\frac{\hbar \omega}{2k_B T}\right) \cos \omega(t-t') \mathcal{D}(\omega). \end{aligned} \quad (30)$$

It is important to mention the fact that until now Eq. (30) is an exact expression for quantum statistical average of  $\hat{\eta}(t) \hat{\eta}(t')$ , i.e., for the two time correlation. We now resort to the following approximation. As  $\hbar \omega \coth(\hbar \omega / 2k_B T)$  is a much more smooth function of  $\omega$  than the remaining part of integral in Eq. (30), at least for not very low temperature, the integral can be approximated as

$$\begin{aligned} & \langle \langle \hat{\eta}(t) \hat{\eta}(t') \rangle \rangle_S \\ & \approx \frac{1}{2} \frac{c_0^2}{\tau_c} \hbar \omega_0 \coth\left(\frac{\hbar \omega_0}{2k_B T}\right) \int_0^\infty d\omega \cos \omega(t-t') \mathcal{D}(\omega), \end{aligned} \quad (31)$$

$\omega_0$  is the average bath frequency. This approximation is well known and frequently used in quantum optics for weak coupling scheme.<sup>24</sup> Using Eq. (27) we thus have the expression for two time correlation as

$$\langle \langle \hat{\eta}(t) \hat{\eta}(t') \rangle \rangle_S = D_0 \delta(t-t') \quad (32)$$

where

$$D_0 = \frac{\Gamma}{2} \hbar \omega_0 \left( \bar{n}(\omega_0) + \frac{1}{2} \right). \quad (33)$$

Here it is pertinent to comment that *our above assumption is not valid at very low temperature*. In this regard, our devel-

opment cannot be claimed as a fully quantum treatment, rather, a quasiclassical one, although ansatz Eq. (20), which is the canonical thermal Wigner distribution function for a shifted harmonic oscillator and always remains a positive definite function contains the quantum information of the bath. A special advantage of using this distribution function is that it remains valid as a pure state nonsingular distribution function even, at  $T=0$ . Thus from the very mode of our development it is clear that barring the calculation of two time correlation function, Eq. (33), rest of our treatment is truly quantum mechanical in nature.

Writing  $q = \langle \hat{q} \rangle_Q$  and  $p = \langle \hat{p} \rangle_Q$  for brevity, we can now rewrite Eqs. (17) and (18) as

$$\dot{q} = p, \quad (34)$$

$$\dot{p} = -\langle V'(\hat{q}) \rangle_Q - \Gamma \langle [f'(\hat{q})]^2 \hat{p} \rangle_Q + \langle f'(\hat{q}) \rangle_Q \eta(t) + \langle f'(\hat{q}) \rangle_Q \pi(t), \quad (35)$$

where  $\eta(t) = \langle \eta(t) \rangle_Q$  and is a classical-like noise term. In writing Eq. (35) we have made use of the fact that the correlation time of the reservoir is very short, i.e.,  $\tau_c \rightarrow 0$ .

We now add  $V(q')$ ,  $\Gamma [f'(q)]^2 p$  and  $f'(q) \xi(t)$  on both sides of Eq. (35) and rearrange it to obtain

$$\dot{q} = p, \quad (36)$$

$$\dot{p} = -V'(q) + Q_V - \Gamma [f'(q)]^2 p + Q_1 + f'(q) \xi(t) + f'(q) + Q_2, \quad (37)$$

where  $\xi(t) = \eta(t) + \pi(t)$  being effective noise and

$$\begin{aligned} Q_V &= V'(q) - \langle V'(\hat{q}) \rangle_Q, \\ Q_1 &= \Gamma \langle [f'(q)]^2 p \rangle_Q - \langle [f'(\hat{q})]^2 \hat{p} \rangle_Q, \end{aligned} \quad (38)$$

and

$$Q_2 = \xi(t) [\langle [f'(\hat{q})] \rangle_Q - f'(q)].$$

Referring to the quantum nature of the system in the Heisenberg picture we now write the system operator  $\hat{q}$  and  $\hat{p}$  as

$$\begin{aligned} \hat{q} &= q + \delta \hat{q}, \\ \hat{p} &= p + \delta \hat{p}, \end{aligned} \quad (39)$$

where  $q = \langle \hat{q} \rangle_Q$  and  $p = \langle \hat{p} \rangle_Q$  are the quantum mechanical mean values and  $\delta \hat{q}$  and  $\delta \hat{p}$  are the operators and they are quantum fluctuations around their respective mean values. By construction  $\langle \delta \hat{q} \rangle_Q = \langle \delta \hat{p} \rangle_Q = 0$  and they also follow the usual commutation relation  $[\delta \hat{q}, \delta \hat{p}] = i\hbar$ . Using Eq. (39) in  $V'(\hat{q})$ ,  $[f'(\hat{q})]^2 \hat{p}$ , and  $f'(\hat{q})$  and a Taylor series expansion in  $\delta \hat{q}$  around  $q$ ,  $Q_V$ ,  $Q_1$ , and  $Q_2$  can be obtained as

$$Q_V = - \sum_{n \geq 2} \frac{1}{n!} V^{n+1}(q) \langle \delta \hat{q}^n \rangle_Q, \quad (40)$$

$$Q_1 = - \Gamma [2p f'(q) Q_f + p Q_3 + 2f'(q) Q_4 + Q_5], \quad (41)$$

and

$$Q_2 = \xi(t)Q_f, \quad (42)$$

where

$$Q_f = \sum_{n \geq 2} \frac{1}{n!} f^{n+1}(q) \langle \delta \hat{q}^n \rangle_Q, \quad (43)$$

$$Q_3 = \sum_{m \geq 1} \sum_{n \geq 1} \frac{1}{m!} \frac{1}{n!} f^{m+1}(q) f^{n+1}(q) \langle \delta \hat{q}^m \delta \hat{q}^n \rangle_Q,$$

$$Q_4 = \sum_{n \geq 1} \frac{1}{n!} f^{n+1}(q) \langle \delta \hat{q}^n \delta \hat{p} \rangle_Q,$$

and

$$Q_5 = \sum_{m \geq 1} \sum_{n \geq 1} \frac{1}{m!} \frac{1}{n!} f^{m+1}(q) f^{n+1}(q) \langle \delta \hat{q}^m \delta \hat{q}^n \delta \hat{p} \rangle_Q.$$

From the above expression it is evident that  $Q_V$  represents quantum correction due to nonlinearity of the system potential and  $Q_1$  and  $Q_2$  represent quantum corrections due to nonlinearity of the system-bath coupling function. Using Eqs. (40)–(42), we get the dynamical equations for system variable from Eqs. (35) and (8) as

$$\dot{q} = p,$$

$$\dot{p} = -V'(q) + Q_V - \Gamma[f'(q)]^2 p - 2\Gamma p f'(q) Q_f - \Gamma p Q_3 - 2\Gamma f'(q) Q_4 - \Gamma Q_5 + f'(q) \xi(t) + Q_f \xi(t). \quad (44)$$

The above equations contain a quantum multiplicative noise term  $Q_f \xi(t)$  in addition to the usual classical contribution  $f'(q) \xi(t)$ . The classical limit of the above equation, apart from the term  $\pi(t)$ , was obtained earlier by Lindenberg and Seshadri.<sup>39</sup> Moreover, quantum dispersion due to nonlinearity of the potential and of the coupling function in the Hamiltonian make their felt presence in Eq. (44).

It is well known and documented in literature<sup>40,41</sup> that when the fluctuation is state dependent or equivalently when the noise is multiplicative with respect to the system variable, which is a manifestation of nonlinear nature of system-bath coupling function, the traditional adiabatic elimination of fast variables in overdamped limit does not provide correct result. To obtain a correct equilibrium distribution Sancho *et al.*<sup>40</sup> had proposed an alternative approach in the case of multiplicative noise system. By carrying out a systematic expansion of the relevant variables in power of  $\Gamma^{-1}$  and neglecting terms smaller than  $O(\Gamma^{-1})$ , they obtained the dynamical equation of motion for position coordinate. We follow the same procedure in our context. In this limit the transient correction terms  $Q_4$  and  $Q_5$  do not affect the dynamics of the position<sup>38</sup> which varies in a much more slower time scale in the overdamped limit. So the equations governing the dynamics of the system variables are

$$\dot{q} = p,$$

$$\dot{p} = -V'(q) + Q_V - \Gamma h(q)p + g(q) \xi(t), \quad (45)$$

where

$$h(q) = [f'(q)]^2 + 2f'(q)Q_f + Q_3 \quad (46)$$

and

$$g(q) = f'(q) + Q_f. \quad (47)$$

The function  $g(q)$  arises due to nonlinearity of the system-bath coupling function  $f(q)$ , where  $Q_f$  is the quantum correction to the classical contribution  $f'(q)$ . For a linear coupling function  $g(q)$  reduces to a constant term and consequently, the noise in Eq. (45) appears additively.

One can now easily identify Eq. (45) as the  $C$ -number analog of the quantum Langevin equation, where  $\Gamma h(q)$  is the state-dependent damping,  $\eta(t)$  the thermal noise, arises due to the system-bath interaction, and  $\pi(t)$  arise due to the bath modulation by the external noise  $\epsilon(t)$ . The statistical properties of  $\pi(t)$  is

$$\langle \pi(t) \rangle = 0,$$

$$\langle \pi(t) \pi(t') \rangle = 2D \int_0^t dt'' \int_0^t dt''' \varphi(t-t'') \varphi(t'-t''') \Psi(t''-t'''), \quad (48)$$

where  $\langle \dots \rangle$  denotes the ensemble average over each realization of  $\epsilon(t)$ . At this point we assume that the external noise  $\epsilon(t)$  is  $\delta$  correlated,

$$\langle \epsilon(t) \epsilon(t') \rangle = 2D \delta(t-t').$$

Then the correlation function of  $\pi(t)$  becomes in the limit  $\tau_c \rightarrow 0$ ,

$$\langle \pi(t) \pi(t') \rangle = 2D \Gamma \kappa_0^2 \delta(t-t'), \quad (49)$$

and consequently the effective noise  $\xi(t)$  has correlation

$$\langle \xi(t) \xi(t') \rangle = 2D_R \delta(t-t'),$$

where  $D_R = (D_0 + D \kappa_0^2) \Gamma$ . Following the method of Sancho *et al.*<sup>40</sup> we obtain the Fokker–Planck equation in position space corresponding to Langevin equation (45),

$$\frac{\partial P(q,t)}{\partial t} = \frac{\partial}{\partial q} \left[ \frac{V'(q) - Q_V}{\Gamma h(q)} \right] P(q,t) + D_R \frac{\partial}{\partial q} \left[ \frac{g(q)g'(q)}{\Gamma(h(q))^2} \right] P(q,t) + D_R \frac{\partial}{\partial q} \left[ \frac{g(q)}{\Gamma h(q)} \frac{\partial}{\partial q} \frac{g(q)}{\Gamma h(q)} \right] P(q,t). \quad (50)$$

According to Stratonovich description,<sup>41</sup> the Langevin equation corresponding to the above Fokker–Planck equation is

$$\dot{q} = -\frac{V'(q) - Q_V}{\Gamma h(q)} - D_R \frac{g(q)g'(q)}{\Gamma[h(q)]^2} + \frac{g(q)}{\Gamma h(q)} \xi(t). \quad (51)$$

Equation (51) is the  $C$ -number quantum Langevin equation for multiplicative noise with state-dependent dissipation in the overdamped limit, when the associated thermal bath is modulated externally by a  $\delta$ -correlated Gaussian noise.

### III. PHASE INDUCED CURRENT

The Fokker–Planck equation (50) can be written in a more compact form as

$$\frac{\partial P(q,t)}{\partial t} = \frac{\partial}{\partial q} \frac{1}{\Gamma h(q)} \left[ V'(q) - Q_V + \frac{D_R}{\Gamma} \frac{\partial}{\partial q} \frac{g^2(q)}{h(q)} \right] P(q,t), \quad (52)$$

from which the stationary current can be obtained as

$$J = - \frac{1}{\Gamma h(q)} \left[ \frac{D_R}{\Gamma} \frac{d}{dq} \frac{g^2(q)}{h(q)} + (V'(q) - Q_V) \right] P_{st}(q). \quad (53)$$

Integrating Eq. (53) we obtain the expression of stationary probability density in terms of stationary current as

$$P_{st}(q) = \frac{e^{-\phi(q)} h(q)}{g^2(q)} \left[ \frac{g^2(0)}{h(0)} P_{st}(0) - J \frac{\Gamma^2}{D_R} \int_0^q h(q') e^{\phi(q')} dq' \right], \quad (54)$$

where

$$\phi(q) = \frac{\Gamma}{D_R} \int_0^q \frac{(V'(q) - Q_V) h(q')}{g^2(q')} dq'$$

is the effective potential. Any tilt of the effective potential  $\phi(q)$  makes the transition between the left to right and right to left of the Brownian particle unequal and thereby creates a directed mass motion. If  $[V'(q)/g^2(q)/h(q)]$  and  $[Q_V/g^2(q)/h(q)]$  are periodic with same period, there will be no symmetry breaking of initial potential and  $\phi(q)$  will remain symmetric periodic with same period. Nonintegrability of  $\phi(q)$  will create spatial symmetry breaking, and consequently, there will be a net mass flow in a particular direction.

We now consider a symmetric periodic potential with periodicity  $2\pi$  and periodic derivative of coupling function with the same periodicity, i.e.,  $V(q+2\pi)=V(q)$  and  $f'(q+2\pi)=f'(q)$ . Consequently,  $Q_V$ ,  $h(q)$ , and  $g(q)$  are also periodic function of  $q$  with period  $2\pi$ . Now applying the periodic boundary condition on  $P_{st}$ , i.e.,  $P_{st}(q+2\pi)=P_{st}(q)$  we have from Eq. (54)

$$\frac{g^2(0)}{h(0)} P_{st}(0) = \frac{\Gamma^2}{D_R} J \left[ \frac{1}{1 - e^{\phi(2\pi)}} \right] \int_0^{2\pi} h(q) e^{\phi(q)} dq. \quad (55)$$

By applying the normalization condition on stationary probability distribution given by  $\int_0^{2\pi} P_{st}(q) dq = 1$ , we get from Eq. (54)

$$\int_0^{2\pi} \frac{e^{-\phi(q)} h(q)}{g^2(q)} \left[ \frac{g^2(0)}{h(0)} P_{st}(0) - J \frac{\Gamma^2}{D_R} \int_0^q dq' h(q') \exp(\phi(q')) \right] dq = 1. \quad (56)$$

Now eliminating  $(D_R/\Gamma)g^2(0)/h(0)P_{st}(0)$  from Eqs. (55) and (56) we obtain the expression for stationary current as

$$J = \frac{D_R}{\Gamma^2} (1 - e^{\phi(2\pi)}) \left\{ \int_0^{2\pi} \frac{h(q)}{g^2(q)} e^{-\phi(q)} dq \int_0^{2\pi} h(q') e^{\phi(q')} dq' - [1 - e^{\phi(2\pi)}] \int_0^{2\pi} \left( \frac{h(q)}{g^2(q)} e^{-\phi(q)} \int_0^q h(q') e^{\phi(q')} dq' \right) dq \right\}^{-1}. \quad (57)$$

From the condition of periodicity of potential and different quantum correction terms it is apparent that for the periodic potential and the periodic derivative of coupling function with same period,  $\phi(2\pi)=0$ . Therefore, the numerator of the expression for current Eq. (57) reduces to zero. We thus conclude that there is no occurrence of current for a periodic potential and periodic derivative of coupling with same periodicity as there will be no symmetry breaking mechanism.

In this paper, we follow Barik *et al.*<sup>38</sup> approach to calculate the quantum correction terms (for details, see Appendix A). Instead of calculating quantum dispersion terms  $\langle \delta \hat{q}^n \rangle_Q$  via numerical simulation of the coupled Eq. (A3) using appropriate physically motivated boundary conditions, in this paper we have carried out direct analytical calculations to find approximated value for the same. Neglecting the term  $\delta \hat{p}$  term from Eq. (A2) in overdamped limit, we get

$$\frac{d}{dt} \delta \hat{q} = \frac{1}{\Gamma [f'(q)]^2} [-V''(q) \delta \hat{q} - 2\Gamma p f'(q) f''(q) \delta \hat{q} + \xi(t) f''(q) \delta \hat{q}] + O(\delta \hat{q}^2). \quad (58)$$

We then obtain the following equations for  $\langle \delta \hat{q}^n \rangle_Q$  in the lowest order, using Eq. (58):

$$\frac{d}{dt} \langle \delta \hat{q}^2 \rangle_Q = \frac{2}{\Gamma [f''(q)]^2} [-V''(q) \langle \delta \hat{q}^2 \rangle_Q - 2\Gamma p f'(q) f''(q) \langle \delta \hat{q}^2 \rangle_Q + \xi(t) f''(q) \langle \delta \hat{q}^2 \rangle_Q]. \quad (59)$$

At this point we want to mention the fact that we have neglected the terms  $O(\langle \delta \hat{q}^3 \rangle_Q)$ . One can compute the leading order quantum correction term  $\langle \delta \hat{q}^2 \rangle_Q$  by neglecting the higher-order coupling terms in the square bracket in Eq. (59) and rewrite it as

$$d \langle \delta \hat{q}^2 \rangle_Q = - \frac{2}{\Gamma [f''(q)]^2} V''(q) \langle \delta \hat{q}^2 \rangle_Q dt.$$

In the overdamped limit, we get the following equation from deterministic classical motion:

$$dq = - \frac{V'(q)}{\Gamma [f'(q)]^2} dt.$$

The last two expressions on integration gives

$$\langle \delta \hat{q}^2 \rangle_Q = \Delta_q [V'(q)]^2, \quad (60)$$

where

$$\Delta_q = \frac{\langle \delta \hat{q}^2 \rangle_Q^0}{[V'(q)]^2},$$

and  $q^0$  is a quantum mechanical mean position at which  $\langle \delta \hat{q}^2 \rangle_Q$  becomes minimum,

$$\langle \delta \hat{q}^2 \rangle_Q^0 = \frac{\hbar}{2\omega_0},$$

$\omega_0$  being defined earlier.

It had been show by Büttiker<sup>18</sup> that for an inhomogeneous medium where the friction is state dependent, a classical particle moving in a symmetric potential in the presence of a sinusoidally modulated space-dependent diffusion with same periodicity experiences a net drift force resulting in generation of current. This current is basically due to the phase difference between the potential and the state-dependent friction.

We are now in a position to discuss the numerical implementation of our development to establish the applicability and potentiality of our formalism. We explore, in the spirit of Büttiker, but in the present quantum mechanical context where the associated bath is modulated by an external Gaussian white noise, the possibility of generation of directed mass motion (due to the phase difference between the potential and dissipation which breaks the principle of detailed balanced). In the numerical applications of our key equation, Eq. (57), we consider the following sinusoidal periodic and symmetric potential:

$$V(q) = V_0[1 + \cos(q + \theta)]. \quad (61)$$

In the above expression  $V_0$  stands for the barrier height and  $\theta$  is the externally controlled phase factor. In our application, the coupling function  $f(q) = (q + \alpha \sin q)$  is chosen in such a way so that the derivative of it appears as  $f'(q) = (1 + \alpha \cos q)$ . Here, the term  $\alpha$  can be considered as a modulation parameter. As a result of this the second order quantum correction reduces to  $\langle \delta \hat{q}^2 \rangle_Q = -\Delta_q V_0^2 \sin^2(q + \theta)$ . It is now clear that the correction to the potential in the leading order can be written as follows [see Eqs. (40)–(43)]:

$$Q_V = -\frac{1}{2} \Delta_q V_0^3 \sin^3(q + \theta). \quad (62)$$

In the same order, the quantum corrections,  $Q_f$  and  $Q_3$ , can be expressed as

$$Q_f = -\frac{1}{2} \Delta_q \alpha V_0^2 \cos q \sin^2(q + \theta),$$

$$Q_3 = \Delta_q \alpha^2 V_0^2 \sin^2 q \sin^2(q + \theta). \quad (63)$$

The functions  $h(q)$  and  $g(q)$  can be estimated from Eqs. (46) and (47) using Eq. (63) as

$$h(q) = (1 + \alpha \cos q)^2 - \Delta_q \alpha V_0^2 \cos q \sin^2(q + \theta)(1 + \alpha \cos q) + \Delta_q \alpha^2 V_0^2 \sin^2 q \sin^2(q + \theta),$$

$$g(q) = (1 + \alpha \cos q) - \frac{1}{2} \Delta_q \alpha V_0^2 \cos q \sin^2(q + \theta). \quad (64)$$

In our numerical applications, we have used the parameters  $\langle \delta \hat{q}^2 \rangle_Q^0 = \frac{1}{2}$ , the minimum uncertainty value,  $\Delta q = 0.5$ ,  $V_0 = 1.0$ ,  $\omega_0 = 1.0$ ,  $\alpha = 1.0$ ,  $T = 0.5$ ,  $\Gamma = 1.0$ , and  $\kappa_0 = 0.2$ , and we set the unit using  $\hbar = k_B = 1$ . In Fig. 1, we have plotted the potential  $\phi(q)$  against spatial coordinate  $q$  to demonstrate the breakdown of the spatial symmetry which is mainly responsible in generation of current in our formalism. In Fig. 2, we have shown the variation in current corresponding to Eq. (57) for various values of  $D$  such as 0.5, 1.0, and 1.5.

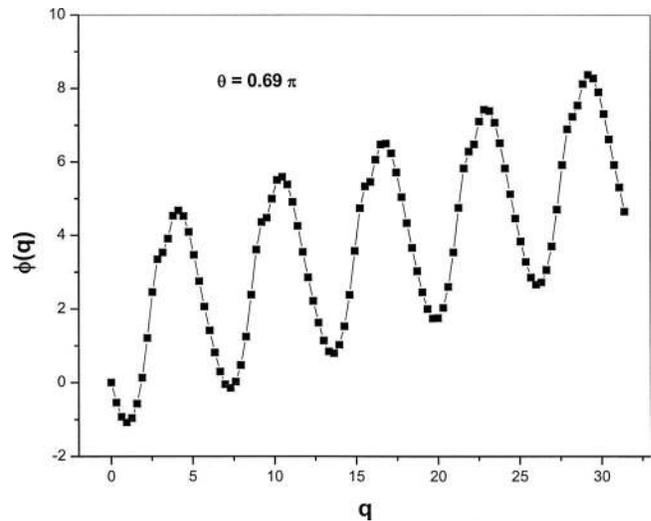


FIG. 1. Plot of effective potential as a function of  $q$  for  $\theta = 0.69\pi$  and  $D = 1.0$ .

From the Fig. 2, it is clear that the current for  $\theta \neq 0, n\pi$ , where  $n = \pm 1, \pm 2, \pm 3, \dots$  there exists a phase induced current. This current  $J$  is a periodic function of the phase difference between modulations of potential and diffusion. The amplitude of current increases with the increase in the strength  $D$  of the external noise. This is due the fact that the “effective temperature” of the bath has been increased from its equilibrium temperature, when the bath being modulated by external noise. This is apparent from the expression of  $D_R$ . The origin of the current is in contrast to classical one proposed by Büttiker.<sup>18</sup>

From the above numerical application of our formalism it is quite clear that nonlinear system-bath coupling gives rise to a state-dependent noise and diffusion in a quantum system, when the bath is modulated by external noise. In the classical system, modulation of bath by  $\delta$ -correlated noise will not create any directed net mass flow.<sup>42</sup> But for a quantum system, for a periodic potential and for a periodic derivative the state-dependent “effective noise” may lead to symmetry breaking in the presence of a phase bias, when the

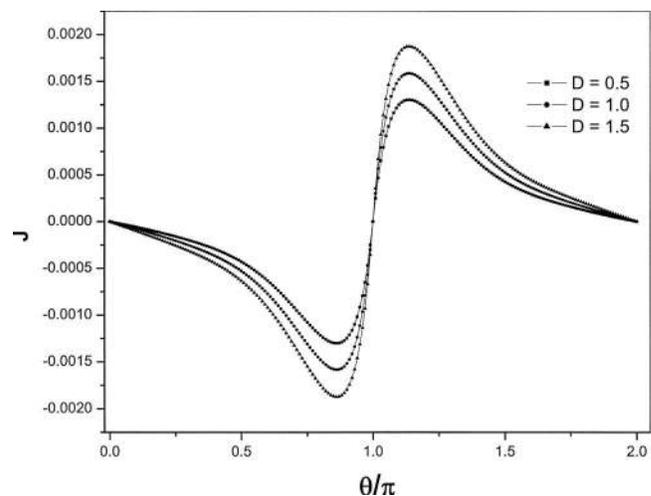


FIG. 2. Plot of current ( $J$ ) as a function of phase difference ( $\theta$ ) for different values of  $D$ .

associated bath is modulated by  $\delta$ -correlated noise. This generation of directional flow will vanish in the absence of the bias and the modulation of the bath by external noise. This phase induced phenomena are solely a quantum effect. This is the key issue of our present article.

#### IV. CONCLUDING REMARKS

We have developed a theory of diffusion of an open quantum system in inhomogeneous media. The quantum system is open in the sense that the associated bath is modulated by an external noise. Our approach is based on the system-reservoir model with nonlinear system-bath coupling. We then derive the quantum Langevin equation for multiplicative noises and a nonlinear dissipation. Then based on the methodology, developed by Barik *et al.*,<sup>38</sup> we obtain the C-number analog of quantum Langevin equation in the Markovian limit, which is coupled to a set of quantum correction equations developed order by order. A systematic expansion of the relevant variable in powers of inverse of the dissipation constant and use of large friction limit lead to a quantum analog of Smoluchowski equation for the state-dependent diffusion of a quantum open system. The openness is due to the modulation of associated quantum heat bath. It is apparent that the state dependence owes its origin to nonlinear coupling between the system and bath degrees of freedom. We have applied the formalism to the problem of diffusion of a quantum particle in a periodic potential where the derivative of coupling function is also periodic with same periodicity. We observe that a phase difference between these two spatially periodic modulations may give rise to a directed quantum current when the bath is modulated by  $\delta$ -correlated noise. This current vanishes in the classical limit if the modulation of bath is done by white noise. Phase induced current can only be obtained classically if the bath is modulated by colored noise. Of course, if the system is driven by any type of noise, there will be a generation of current always. But our point is that we are driving the bath, not the system, by the external noise. Thus the generation of current in this case and the nonlinearity in the effective potential is essentially a quantum effect. The extension of our formulation to the case when bath is modulated by colored noise is also worth pursuing. We hope to address this issue in near future.

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#### APPENDIX A: QUANTUM CORRECTION TERMS

In this part of the appendix, we present the calculation of the required quantum correction terms. For detail description, we refer to Ref. 38. Referring to the quantum nature of the system in the Heisenberg picture we now write the sys-

tem operators  $\hat{q}$  and  $\hat{p}$  as  $\hat{q}=q+\delta\hat{q}$  and  $\hat{p}=p+\delta\hat{p}$ , respectively.  $\delta\hat{q}$  and  $\delta\hat{p}$  represent quantum fluctuations around their respective mean values.

Following Barik *et al.*,<sup>38</sup> the time evolution of these correction terms can be computed using the following equations with the help of the operator Langevin equations (8) in the Markovian limit:

$$\begin{aligned}\dot{\hat{q}} &= \hat{p}, \\ \dot{\hat{p}} &= -V'(\hat{q}) - \Gamma[f'(\hat{q})]^2\hat{p} + f'(\hat{q})\eta(t) + f'(\hat{q})\pi(t),\end{aligned}\quad (\text{A1})$$

$$\begin{aligned}\delta\dot{\hat{q}} &= \delta\hat{p}, \\ \delta\dot{\hat{p}} &= -V''(q)\delta\hat{q} - \sum_{n\geq 2} \frac{1}{n!}V^{n+1}(q)[\delta\hat{q}^n - \langle\delta\hat{q}^n\rangle_Q] \\ &\quad - \gamma \left[ 2f'(q)f''(q)\delta\hat{q} + 2f'(q) \sum_{n\geq 2} \frac{1}{n!}f^{n+1}(q)[\delta\hat{q}^n - \langle\delta\hat{q}^n\rangle_Q] \right. \\ &\quad \left. + \sum_{m\geq 1} \sum_{n\geq 1} \frac{1}{m!} \frac{1}{n!} f^{m+1}(q)f^{n+1}(q)[\delta\hat{q}^m\delta\hat{q}^n - \langle\delta\hat{q}^m\delta\hat{q}^n\rangle_Q] \right] p \\ &\quad - \gamma \left[ [f'(q)]^2\delta\hat{p} + 2f'(q) \sum_{n\geq 1} \frac{1}{n!}f^{n+1}(q)[\delta\hat{q}^n\delta\hat{p} \right. \\ &\quad \left. - \langle\delta\hat{q}^n\delta\hat{p}\rangle_Q] + \sum_{m\geq 1} \sum_{n\geq 1} \frac{1}{m!} \frac{1}{n!} f^{m+1}(q)f^{n+1}(q)[\delta\hat{q}^m\delta\hat{q}^n\delta\hat{p} \right. \\ &\quad \left. - \langle\delta\hat{q}^m\delta\hat{q}^n\delta\hat{p}\rangle_Q] \right] + \eta(t) \left[ f''(q)\delta\hat{q} + \sum_{n\geq 2} \frac{1}{n!}f^{n+1}(q)[\delta\hat{q}^n \right. \\ &\quad \left. - \langle\delta\hat{q}^n\rangle_Q] \right].\end{aligned}\quad (\text{A2})$$

At this point it is pertinent to mention the fact that the above equations were derived by Ray and co-workers using quantum mechanical average over the initial product-separable coherent bath states.

From the very mode of derivation, it is quite evident that the operator correction equations can be used to yield an infinite hierarchy of equations. For example, up to third order one can construct the following set of equations:

$$\begin{aligned}\frac{d}{dt}\langle\delta\hat{q}^2\rangle_Q &= \langle\delta\hat{q}\delta\hat{p} + \delta\hat{p}\delta\hat{q}\rangle_Q, \\ \frac{d}{dt}\langle\delta\hat{q}\delta\hat{p} + \delta\hat{p}\delta\hat{q}\rangle_Q &= -2\chi(q,p)\langle\delta\hat{q}^2\rangle_Q + 2\langle\delta\hat{q}^2\rangle_Q - \gamma[f'(q)]^2\langle\delta\hat{q}\delta\hat{p} + \delta\hat{p}\delta\hat{q}\rangle_Q \\ &\quad - \zeta(q,p)\langle\delta\hat{q}^3\rangle_Q - 2\gamma f'(q)f''(q)\langle\delta\hat{q}^2\delta\hat{p} + \delta\hat{p}\delta\hat{q}^2\rangle_Q, \\ \frac{d}{dt}\langle\delta\hat{p}^2\rangle_Q &= -2\gamma[f'(q)]^2\langle\delta\hat{p}^2\rangle_Q - \chi(q,p)\langle\delta\hat{q}\delta\hat{p} + \delta\hat{p}\delta\hat{q}\rangle_Q \\ &\quad - \frac{1}{2}\zeta(p,q)\langle\delta\hat{q}^2\delta\hat{p} + \delta\hat{p}\delta\hat{q}^2\rangle_Q - 2\gamma f'(q)f''(q) \\ &\quad \times \langle\delta\hat{q}\delta\hat{p}^2 + \delta\hat{p}^2\delta\hat{q}\rangle_Q,\end{aligned}$$

$$\frac{d}{dt}\langle\delta\hat{q}^3\rangle_Q = \frac{3}{2}\langle\delta\hat{q}^2\delta\hat{p} + \delta\hat{p}\delta\hat{q}^2\rangle_Q,$$

$$\frac{d}{dt}\langle\delta\hat{p}^3\rangle_Q = -3\chi[f'(q)]^2\langle\delta\hat{p}^3\rangle_Q - \frac{3}{2}\chi(q,p)\langle\delta\hat{q}\delta\hat{p}^2 + \delta\hat{p}^2\delta\hat{q}\rangle_Q,$$

$$\begin{aligned} \frac{d}{dt}\langle\delta\hat{q}^2\delta\hat{p} + \delta\hat{p}\delta\hat{q}^2\rangle_Q &= -2\chi(q,p)\langle\delta\hat{p}^3\rangle_Q + 2\langle\delta\hat{q}\delta\hat{p}^2 + \delta\hat{p}^2\delta\hat{q}\rangle_Q \\ &\quad - \chi[f'(q)]^2\langle\delta\hat{q}^2\delta\hat{p} + \delta\hat{p}\delta\hat{q}^2\rangle_Q, \end{aligned}$$

$$\begin{aligned} \frac{d}{dt}\langle\delta\hat{q}\delta\hat{p}^2 + \delta\hat{p}^2\delta\hat{q}\rangle_Q &= 2\langle\delta\hat{p}^3\rangle_Q - 4\chi(q,p)\langle\delta\hat{q}^2\delta\hat{p} + \delta\hat{p}\delta\hat{q}^2\rangle_Q \\ &\quad - 2\chi[f'(q)]^2\langle\delta\hat{q}\delta\hat{p}^2 + \delta\hat{p}^2\delta\hat{q}\rangle_Q, \quad (\text{A3}) \end{aligned}$$

where

$$\chi(q,p) = V''(q) + 2\gamma p f'(q) f''(q) - \eta(t) f''(q),$$

$$\zeta(q,p) = V'''(q) + 2\gamma p f'(q) f'''(q) + 2\gamma p [f''(q)]^2 - \eta(t) f'''(q).$$

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