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Nonperturbative approach to quantum Brownian motion

Subhasis Sinha and P. A. Sreeram
Indian Institute of Science Education and Research Kolkata, Mohanpur Campus, Mohanpur, 741252, India

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Starting from the Caldeira-Leggett model, we derive the equation describing the quantum Brownian motion, which has been originally proposed by Dekker purely from phenomenological basis containing extra anomalous diffusion terms. This nonperturbative approach yields explicit analytical expressions for the temperature dependence of the diffusion constants. At high temperatures, additional momentum diffusion terms are suppressed and classical Langevin equation can be recovered and at the same time positivity of the density matrix is satisfied. At low temperatures, the diffusion constants have a finite positive value. However, below a certain critical temperature, the master equation does not satisfy the positivity condition as proposed by Dekker.

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The problem of quantum Brownian motion is a longstanding and challenging problem [1–3], as it forms the underlying basis of nonequilibrium phenomena such as dissipative and relaxation dynamics of quantum systems [4,5]. Quantum dissipative dynamics has application in a wide variety of problems, starting from quantum cosmological models [6,7] to reaction-rate theory [8]. Unlike the classical nonequilibrium problem, quantum dynamics has additional complexity due to the Heisenberg uncertainty principle. Various attempts have been made to address this problem, which include semiclassical approaches using Wigner distribution function [9], phenomenological models [10–12], and Boltzmann's collision terms [13]. On the other hand, there have been attempts to obtain the master equation describing the time evolution of the density matrix of open quantum systems through toy microscopic models [6,14,15], where the diffusion constants have explicit time dependence [6,16]. Independently, a class of master equations has been put forward [17], purely from mathematical consideration, which guarantees the positivity of the density matrix, where the time evolution of the system takes place only through the physical states, and the master equation is commonly known to belong to the Lindblad class. Most of the master equations describing the nonequilibrium quantum dynamics suffer from the fact that they either do not belong to the Lindblad class or the correct classical limit cannot be recovered at high temperatures.

Dekker [12] proposed a phenomenological model of quantum Brownian dynamics, which contains extra diffusion terms in order to preserve Heisenberg's uncertainty condition as well as positivity of the density matrix [18,19]. However, the microscopic origin of the extra diffusion constants in Dekker's model is not clear and can only be justified in the weak-coupling limit. Moreover, most of the derivations of the master equation rely on the facts that the autocorrelation function of the random force on the Brownian particle is short ranged at high temperature and that the system is assumed to be Markovian. But in practice, the effective action obtained has a memory kernel which can decay as a power law at low temperatures [20]. In fact, the random force in quantum Brownian motion shows clear deviation from the Markovian limit [21]. This nonlocality makes the problem very complex.

In this paper, we propose a mechanism of deriving the master equation from a microscopic theory, which can be used to study complex problems in both the strong- and weak-coupling limits. The technique involves resummation of memory effects along the classical paths, which is both intuitively transparent and mathematically simpler. For simplicity and clarity we take the example of the Brownian particle for illustrating this technique.

We show that over a wide range of temperatures, the master equation satisfies the positivity condition and at the same time reproduces the classical Langevin dynamics at high temperatures. Although, the diffusion constants are finite and well behaved even at sufficiently low temperatures, the positivity of density matrix breaks down below a certain temperature depending on the damping rate of the system. Below this temperature, the transient behavior dominates the dynamics and the diffusion constants may become time dependent.

A very well-known description for the dissipative phenomena and relaxation dynamics of classical system is given by the Langevin equation

$$\ddot{q}(t) = -2\gamma\dot{q}(t) + \theta(t), \tag{1}$$

where $q(t)$ is the position of the Brownian particle, the dot denotes derivative with respect to time, γ is the damping constant, and $\theta(t)$ is related to the fluctuating force acting on the particle, whose autocorrelation function is given by $\langle\theta(t)\theta(t')\rangle = \Gamma\delta(t-t')$, where $\Gamma = 2k_BTM\gamma$; T is the temperature of the bath and M is the mass of the Brownian particle. However, in most cases, the microscopic details behind this dissipation are not well known.

Many microscopic models have been proposed which describe the dissipative dynamics of a heavy particle in the presence of a heat bath [22–26]. One of these is a model, where the heat bath consists of a collection of harmonic oscillator and is in recent times known as the Caldeira-Leggett model [14]. In this model, the full Hamiltonian H is given by $H = H_A + H_B + H_I$, where H_A, H_B, and H_I are the system, bath, and interaction Hamiltonians, respectively, and are given by

$$H_A = \frac{p^2}{2M} + V(q), \tag{2}$$

$$H_B = \sum_i \left(\frac{P_i^2}{2m_i} + \frac{1}{2}m_i\omega_i^2 Q_i^2\right), \tag{3}$$

$$H_I = -q\sum_i C_i Q_i + q^2\sum_i \frac{1}{2}\frac{C_i^2}{m_i\omega_i^2}, \tag{4}$$

where q and p are the position and momentum operators of the heavy particle having mass M, $V(q)$ is the potential in which the heavy particle is moving, Q_i and P_i are the position and momentum operators of the bath oscillators whose mass and frequency of oscillation are given by m_i and ω_i, respectively, and C_i is the coupling strength between the system and the ith oscillator in the bath. Following the work in [25], the time evolution of the position operator can be written as

$$M\ddot{q} + \int_0^t d\tau\alpha(t-\tau)\dot{q}(\tau) + V'(q) = F(t), \tag{5}$$

where the prime denotes the derivative with respect to the position variable. The operator valued random force, $F(t)$, is related to the statistical distribution of the bath variables Q_i and P_i. In order to recover the classical Brownian dynamics with short-time memory effect at high temperatures, the frequency dependence of the coupling is typically taken to be $\Sigma_i C_i^2/(m_i\omega_i^2) \propto M\gamma$, where γ is the damping constant. According to the Ehrenfest theorem, if the position and momentum operators are replaced by their classical values, then Eq. (5) translates to the classical Langevin equation in the limit of $\hbar \rightarrow 0$, where the force autocorrelation function takes the form of a delta function obeying classical fluctuation-dissipation theorem [4].

The classical Langevin equation and the quantum master equation differ in the sense that while the classical fluctuations in the canonical coordinates are controlled only by the scales set by the temperature and the dissipation constant, the quantum fluctuations have an additional scale given by $\hbar$ which appears due the Heisenberg uncertainty principle. Moreover, the evolution of the operators themselves has to be unitary. Apart from the semiclassical techniques, new methods have been proposed to take into account the constraints imposed by "uncertainty principle" in the quantum case [23]. Among these methods, the master equation of the density matrix ρ is more suitable to describe dissipative dynamics of quantum systems. The quantum dynamics which is governed by the time-dependent Schrödinger equation describes the pure state, whereas the dissipative dynamics introduces the concept of mixed state, where trace of ρ^2 is less than unity. The evolution equation of the reduced density matrix of the system can be obtained by tracing out the bath degrees of freedom.

Using the Feynman-Vernon method [27], the dissipative dynamics of the reduced density matrix can be written in terms of the influence functional, which is obtained after integrating out the bath degrees of freedom Q_i's. The main assumption of this method is that the subsystem is uncorrelated with the bath at the initial time. Hence the total density matrix at time $t=0$ is given by $\rho_T(0)=\rho_A(0)\otimes\rho_B(0)$, where ρ_A, ρ_B, and ρ_T are the density matrix of the subsystem, bath, and the total system, respectively. However, several authors have considered a correlated initial state, where the subsystem and bath density matrix cannot be factorized. For example, Hakim and Ambegaokar [28] compared the two cases of uncorrelated and correlated initial conditions and showed that, for the correlated initial condition, different transient behaviors can be obtained at time scales larger than the inverse cutoff frequency of the bath, in contrast to the uncorrelated initial conditions. This was further extended by Grabert and co-workers [29,30]. In this paper, we study the long-time behavior beyond the transient behavior. It would, of course, be very interesting to study the transient behavior. We follow the usual procedure of Feynman and Vernon [27] and show finally that we can obtain a consistent master equation, describing the time evolution of the density matrix. The time evolution of the reduced density matrix is given by

$$\rho(q_1,q_2,t) = \int dq_1' dq_2' J(q_1,q_2,t;q_1',q_2',0)\rho(q_1',q_2',0), \tag{6}$$

where the propagator $J(q_1,q_2,t;q_1',q_2',0)$ is given by

$$J(q_1,q_2,t;q_1',q_2',0) = \int\int \mathcal{D}q_1\mathcal{D}q_2 \exp\left(\frac{\imath}{\hbar}S_{\text{eff}}[q_1,q_2]\right). \tag{7}$$

After integrating out the bath degrees of freedom, one obtains the nonlocal effective action corresponding to the dissipative system,

$$\frac{\imath}{\hbar}S_{\text{eff}} = \frac{\imath}{\hbar}(S_A[q_1] - S_A[q_2]) + \int_0^t (\Sigma_R + \Sigma_I)d\tau, \tag{8}$$

where S_A is the action corresponding to the noninteracting system and

$$\Sigma_R = -\frac{1}{\hbar}\int_0^\tau [q_-(\tau)\alpha_R(\tau-s)q_-(s)]ds,$$

$$\Sigma_I = -\frac{\imath}{\hbar}\int_0^\tau [q_-(\tau)\alpha_I(\tau-s)q_+(s)]ds, \tag{9}$$

where $q_\pm = q_1 \pm q_2$ and the memory kernels are

$$\alpha_R(\tau) = \sum_i \frac{C_i^2}{2m\omega_i}\coth\left(\frac{\hbar\omega_i}{2k_BT}\right)\cos(\omega_i\tau),$$

$$\alpha_I(\tau) = -\sum_i \frac{C_i^2}{2m\omega_i}\sin(\omega_i\tau). \tag{10}$$

It is assumed that the oscillator frequencies are continuously distributed from zero to a maximum frequency ω_c, a value of which depends on specific physical system. Cutoff frequency of the heat bath is chosen in such way that the characteristic time scale of the dynamics of heavy particle is much larger than the collision time scale $1/\omega_c$. Thus, the summations in Eq. (11) can be replaced by integrals by introducing an appropriate density of state, which will make the memory function analytically tractable. Hence, Σ_i can be replaced by $\int d\omega F(\omega)$, where $F(\omega)$ is the density of states of the bath. We choose a smooth Drude form for the density of states given by $F(\omega)=\omega_c^2/(\omega^2+\omega_c^2)$ and $F(\omega)C^2(\omega)/(2m\omega^2)=(2M\gamma/\pi)[\omega_c^2/(\omega^2+\omega_c^2)]$. At any nonzero temperature, using the Drude form of density of states, the memory kernels can be evaluated analytically by using contour integrals in complex ω plane and are given by

$$\alpha_R(\tau)=M\gamma\omega_c^2\Bigg[\cot(\chi)\exp(-\omega_c\tau) + \frac{2}{\chi}\sum_{n=1}^{\infty}\frac{n\pi/\chi}{(n\pi/\chi)^2-1}\exp[-(n\pi/\chi)\omega_c\tau]\Bigg], \qquad (11)$$

$$\alpha_I(\tau)=M\gamma\omega_c\frac{\partial}{\partial\tau}\exp[-\omega_c\tau], \qquad (12)$$

where $\chi=\hbar\omega_c/2k_BT$. From the above expression of α_R we can clearly see the emergence of two-time scales—the first being the microscopic collision time given by $1/\omega_c$ and the other time scale given by temperature, i.e., $\hbar/k_BT$. It is interesting to note that in the high-temperature regime, α_R is short ranged over the thermal time scale while the time scale over which α_I decays is given only by the collision time scale, which is independent of temperature. However, as the temperature is reduced, at some point, the thermal time scale dominates over the collision time scale, giving rise to a nonlocal memory kernel which has a power-law decay.

We now proceed to simplify the nonlocal action, taking advantage of the above mentioned short-ranged kernel. The dynamics over a time scale larger than $\hbar/k_BT$ can be studied by assuming smooth classical trajectories and expanding the dynamical variables in a Taylor series,

$$q_{\pm}(s)=\sum_{l=0}^{\infty}\frac{q_{\pm}^{(l)}(\tau)}{l!}(s-\tau)^l, \qquad (13)$$

where $q^{(l)}$ denotes the lth derivative of q with respect to time. We now insert Eqs. (11) and (13) into Eq. (9). Neglecting the total derivative terms which generate the boundary terms in the action, thereby leaving the Lagrangian invariant, we obtain

$$\Sigma_R(\tau)=-\frac{1}{\hbar}\sum_{l=0}^{\infty}\frac{(-1)^l}{2l!}(q_-^{(l)}(\tau))^2\int_0^{\tau}\tilde{\tau}^{2l}\alpha_R(\tilde{\tau})d\tilde{\tau}. \qquad (14)$$

The transient term in the integral comes in the form of $\exp[-(k_BT/\hbar)\tau]$. Thus for $\tau\gg\hbar/k_BT$, we obtain

$$\Sigma_R(\tau)=-\frac{M\gamma\omega_c}{\hbar\chi}\Bigg[q_-^2(\tau) - 2\sum_{l=1}^{\infty}(q_-^{(l)})^2\frac{(-1)^l}{\omega_c^{2l}}\sum_{m=0}^{l}\left(\frac{\chi}{\pi}\right)^{2m}\zeta(2m)\Bigg]. \qquad (15)$$

This could be thought of as an effective "high-temperature" expansion of the nonlocal effective action. Similarly, one can evaluate Σ_I which gives

$$\Sigma_I(\tau)=-\frac{\imath\gamma M}{\hbar}q_-(\tau)\dot{q}_+(\tau), \qquad (16)$$

where the higher order terms in the expansion of $q_+(s)$ have been neglected since they fall off as $1/\omega_c$.

We have thus transformed a highly nonlocal action into a local action containing higher derivative terms. While in general this may seem to be a very complicated expression to work with, we show below that in at least the case of a Brownian particle there is considerable simplification of the expression which allows us to obtain exact results.

For the free particle, the general dynamical equation for any order of time derivative of the position coordinate can be written as

$$q^{(n)}=\frac{(-2\gamma)^{n-1}}{M}p. \qquad (17)$$

Substituting Eq. (17) in Eq. (15), we obtain

$$\Sigma_R(\tau)=-\frac{M\gamma\omega_c}{\hbar\chi}q_-^2(\tau) - \frac{M\gamma\omega_c}{\hbar\chi}\left[\frac{\dot{q}_-^2(\tau)}{4\gamma^2}\left\{\frac{\left(\frac{2\gamma\chi}{\omega_c}\right)\coth\left(\frac{2\gamma\chi}{\omega_c}\right)}{1+\left(\frac{2\gamma}{\omega_c}\right)^2}-1\right\}\right]. \qquad (18)$$

Thus, the effective action is given by

$$\frac{\imath}{\hbar}S_{\text{eff}}=\frac{\imath}{\hbar}\int_0^t d\tau\left[\frac{M}{2}\dot{q}_+\dot{q}_- - \gamma M q_-\dot{q}_+\right] - \frac{2k_BT\gamma M}{\hbar^2}\int_0^t d\tau[q_-^2+\alpha\dot{q}_-^2], \qquad (19)$$

where

$$\alpha=\frac{\frac{\hbar\gamma}{k_BT}\coth\left(\frac{\hbar\gamma}{k_BT}\right)-1}{4\gamma^2}, \qquad (20)$$

assuming $\gamma\ll\omega_c$. It is important to note that by using the dynamical equation of canonical coordinates and the resummation method we have converted the nonlocal action into an effective action which is local and quadratic in the canonical coordinates as well as independent of the cutoff frequency ω_c of bath oscillators. Inserting the above form of the effective action into Eq. (7) the time evolution of the density matrix can be evaluated.

Following the prescription of Caldeira and Leggett [14], we consider the change in density matrix from t to $t+\epsilon$ within a small time interval ϵ in order to obtain the master equation in a differential form. To do so we expand both side of Eq. (6) up to leading order in ϵ. Within the small time ϵ, we approximate $\dot{q}_1=(x-x')/\epsilon=\beta_1/\epsilon$ and $\dot{q}_2=(y-y')/\epsilon=\beta_2/\epsilon$. Now Eq. (6) reads

$$\rho + \epsilon\frac{\partial\rho}{\partial t} = \int\int d\beta_+ d\beta_- \exp\left[\frac{i}{\hbar\epsilon}M\beta_+(\beta_- - 2\gamma x_-\epsilon) - \frac{2k_BT\gamma M}{\hbar^2}\left(\epsilon x_-^2 + \alpha\frac{\beta_-^2}{\epsilon}\right)\rho(x-\beta_1, y-\beta_2, t)\right], \tag{21}$$

where $\beta_+ = (\beta_1 + \beta_2)/2$ and $\beta_- = \beta_1 - \beta_2$. We expand ρ up to second order in β, which is equivalent to expanding up to first order in ϵ. After doing some algebra and performing the Gaussian integrals we obtain the master equation describing the time evolution of the density matrix,

$$\frac{\partial\rho}{\partial t} = \frac{i\hbar}{2M}\left[\frac{\partial^2\rho}{\partial x^2} - \frac{\partial^2\rho}{\partial y^2}\right] - \gamma(x-y)\left(\frac{\partial\rho}{\partial x} - \frac{\partial\rho}{\partial y}\right) + \frac{2\imath}{\hbar}D_{pq}(x-y)\left(\frac{\partial\rho}{\partial x} + \frac{\partial\rho}{\partial y}\right) + D_{qq}\left(\frac{\partial}{\partial x} + \frac{\partial}{\partial y}\right)^2\rho - \frac{D_{pp}}{\hbar^2}(x-y)^2\rho, \tag{22}$$

where different diffusion constants are given in terms of α by

$$D_{qq} = \frac{2k_BT\gamma\alpha}{m}, \tag{23}$$

$$D_{pq} = 4\gamma^2 k_BT\alpha, \tag{24}$$

$$D_{pp} = 2k_BTM\gamma(1 + 4\gamma^2\alpha), \tag{25}$$

where α is given by Eq. (20). The above form of the master equation has been proposed earlier phenomenologically [12] in the context of a Brownian particle in a harmonic trap. It was shown that in order to satisfy the Heisenberg uncertainty principle in quantum dissipative systems, the diffusion constants must satisfy the following relation [18]:

$$\Delta = D_{pp}D_{qq} - D_{pq}^2 - \frac{\hbar^2\gamma^2}{4} \geq 0. \tag{26}$$

This is also known as the "positivity" condition since the master equation in Dekker's form satisfying the above condition can be recast into the Lindblad form [19]. It is relevant to note here that the phenomenological form of the diffusion constants obtained in [12] can be justified only in the weak-coupling limit. In contrast, the problem we are addressing is to obtain the diffusion constants for a free Brownian particle, which is intrinsically a strong-coupling problem, from a microscopic theory.

The mean-square momentum and position are given, respectively, by

$$\langle p^2\rangle = \frac{D_{pp}}{2\gamma} = M\hbar\gamma\coth\left(\frac{\hbar\gamma}{k_BT}\right) \tag{27}$$

and

$$\langle x^2\rangle \sim \frac{D_{pp} + 4M^2\gamma^2 D_{qq} - 4M\gamma D_{pq}}{2M^2\gamma^2}t = \frac{k_BT}{M\gamma}t. \tag{28}$$

The value of $\langle p^2\rangle$ recovers the equipartition results at high temperatures. The exact result is known to have an ultraviolet divergence [5], which can be regulated by the parameter $\hbar\omega_c/k_BT$, which is taken to be small at high temperature. In contrast, in our calculation, the results are independent of $\hbar\omega_c$ and depend only on the dimensionless parameter $\hbar\gamma/k_BT$. However, for reasonable values of the cutoff frequency, the above result matches quite well with the known results [5].

It is interesting to note that the long-time behavior $\langle x^2\rangle$ corresponds to the exact result and matches the classical behavior.

At high temperatures, the diffusion constants behave as, $D_{pp} = 2M\gamma k_BT[1 + \frac{1}{3}(\frac{\hbar\gamma}{k_BT})^2]$, $D_{qq} = \frac{\hbar^2\gamma}{6Mk_BT}$, and $D_{pq} = \frac{\hbar^2\gamma^2}{3k_BT}$. The forms of these diffusion constants are very similar to those obtained earlier [10]. However, we would like to point out that the diffusion constant D_{pq}, in our case, is independent of the cutoff parameter ω_c unlike the earlier result. It is also interesting to note that the diffusion constants D_{qq} and D_{pp} both originate from quantum effects and vanish at high temperatures as $1/k_BT$. However, at high temperatures, Δ approaches a value $\hbar^2\gamma^2/12$ which preserves the positivity condition of the master equation. At zero temperature, the diffusion constants are finite, positive, and proportional to $\hbar$, similar to the form proposed by Dekker [12]. However, Δ becomes negative at zero temperature, violating the positivity criterion. We estimate a critical temperature $T_0 \sim 0.4\hbar\gamma$, below which the above form of the master equation is not valid and transient behavior as well as long-range memory effects become crucial.

To summarize, within our scheme of calculation, we have shown that the master equation obtained from the microscopic Caldeira and Leggett [14] model satisfies the positivity condition and belongs to the Lindblad class for wide range of temperatures, above the critical temperature T_0. Analytic form for the diffusion constants has been obtained for any arbitrary temperature and is *independent of the cutoff frequency of the heat bath*. It is interesting to note that at high temperatures all the anomalous diffusion constants vanish as $1/T$, which preserves the structure of classical Brownian motion. At the same time, the diffusion constants conspire in such a way that they satisfy the Dekker criterion $\Delta > 0$ and hence the time evolution of the open quantum system takes place only through physical states. At low temperature, the diffusion constants have their origin from purely quantum effects. However, below the temperature $\simeq\hbar\gamma$, Dekker's positivity condition is violated, indicating that the long-range memory effect may become important in the time evolution of the system. The novelty of this method is the fact that it requires only the knowledge of the classical paths and hence can be applied to address more complex physical problems in a consistent manner.

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