Generalized Einstein Relation in Tilted Periodic Potential: A Semiclassical Approach

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This paper concerns the investigation of the quantum motion of a system in a dissipative Ohmic heat bath in the presence of an external field using the traditional system-reservoir model. Using physically motivated initial conditions, we then obtain the *c*-number of the generalized quantum Langevin equation by which we calculate the quantum correction terms using a perturbation technique. As a result of this, one can apply a classical differential equation-based approach to consider quantum diffusion in a tilted periodic potential, and thus our approach is easy to use. We use our expression to calculate the Einstein relation for the quantum Brownian particle in a ratchet-type potential in a very simple closed analytical form using a suitable and convenient approximation. It is found that the diffusion rate is independent of the detailed form of the potential both in quantum and classical regimes, which is the main essence of this work.

I. Introduction

The Einstein relation[1](arguably the most famous linear response relation) is the simplest form of the *fluctuation−dissipation relation* in the context of Brownian dynamics. This relation bears the hallmark of a deep connection between the fluctuations and the resulting diffusion and dissipation that is responsible for friction expressed by a finite mobility. The Einstein relation, however, breaks down when the system is sufficiently out of equilibrium where linear response theories are not applicable. It is still challenging to explore whether Einstein's relations remain valid for more general nonequilibrium steady states. Motivated by this challenge, many recent efforts have come about that exploit the concepts of statistical physics to explore the away-from-equilibrium situations in various systems. In particular, the fluctuation−dissipation theorem in nonequilibrium systems has been studied by several authors in several contexts.[2–4] As reported in literature,[5–7] the diffusion coefficient for a Brownian particle may assume several forms depending on the nature of the coupling with the environment and also on the form of the system potential and external perturbation. For example, the diffusion term for a Brownian particle may have the forms $(\partial/\partial q)\mathscr{D}(q)[\partial P(q, t)/\partial q]$ or $(\partial^2/\partial q^2)\mathscr{D}(q)P(q, t)$, supplemented by the thermal potential or state dependent drift term that appears in the Fokker−Planck equation.[8] Here, $\mathscr{D}(q)$ refers to the diffusion coefficient, and $P(q, t)$ is the probability density function for the Brownian particle. As the phenomenological forms of the diffusion coefficient are different, they do not have a common microscopic Hamiltonian. Thus, the physics of diffusion is somewhat model dependent.[8] Also, the diverse forms notwithstanding, the generalization of the Boltzmann factor, i.e., the steady state assumes a common structure.

If one wants to calculate the diffusion of particles moving in an infinite periodic potential, it can be assumed that the particles, which were initially near $x = 0$, will diffuse to adjacent wells. For very large times they will be distributed over many wells. The particles hop from one well to an adjacent well for large damping, whereas for small damping, they move over quite a number of wells before they get trapped in one and are then excited again to move to other wells. The diffusion constant will be defined by either of the following two relations:

$$\mathscr{D} = \frac{1}{2}\lim_{t\to\infty}\frac{\mathrm{d}}{\mathrm{d}t}\langle[x(t) - x(0)]^2\rangle$$

or

$$\mathscr{D} = \frac{1}{2}\lim_{t\to\infty}\frac{1}{t}\langle[x(t) - x(0)]^2\rangle$$

which describes the diffusion in the infinite periodic potential for large times. The diffusion constant is connected to mobility μ in a simple fashion for free Brownian motion:

$$\mathscr{D} = \frac{k_B T}{m\gamma} = \mu k_B T = \frac{\gamma\mu k_B T}{\gamma}$$

which is known as the celebrated Einstein relation for the diffusion constant $\mathscr{D}$. For periodic potential, the diffusion constant is given by the diffusion constant for free Brownian motion multiplied by a factor that can be determined easily.[8] It is noteworthy to emphasize that, when there is a finite external

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force F, the diffusion constant of the Brownian particle in a periodic potential is no longer connected to the change of the stationary drift velocity, and one then usually calculates the mobility μ in linear response via the relation

$$\mu = \lim_{F \to 0} \frac{\langle v \rangle}{F}$$

and then the Einstein relation reads

$$\mathscr{D} = \mu k_{\mathrm{B}} T(F)$$

In the quantum domain, things get complicated due to the operator nature of the governing dynamical equation(s) and the definition of mobility and the fact that $T(F)$ is not straightforward. Consequently, the Einstein's relation demands a cautious definition. In passing, we mention that, despite the tremendous methodological developments, strategies capable of reliable computation of thermal diffusion still remain an active research area. Thermal diffusion in a tilted or slanted periodic potential has an important role when studying Josepson's junction,[9] oscillators with noisy limit cycles,[10] diffusion in crystals surfaces,[11] and many others.[12,13] More recently, it has been followed with a lot of interest while studying the transport properties of Brownian particles moving in a periodic potential[8,14] with special stress on coherent transport.[15] A lot of interest[16] has been seen in studying Brownian motion under the influence of tilted ratchet potential. This possibly stems from the fact that the tilted periodic potential or ratchet systems allow one to study the transport phenomena under the action of unbiased forces. In this context we want to mention that the diffusion of a colloidal particle in a rotating array of laser traps can be modeled as a one-dimensional Brownian motion in a tilted periodic potential.[17]

In contrast to the classical regime where the transport of Brownian particle is well elaborated in literature[18] and special interest has been directed to transport in a ratchet system,[19] the quantum properties of directed transport are only partially elaborated.[20–26] Challenges arise in the quantum regime because the transport or diffusion can strongly depend on the mutual interplay of pure quantum effects such as tunnelling and particle wave interference with dissipation process nonequilibrium fluctuation and external driving.[27] An important concept induced recently in classical and quantum Hamiltonian transport is that for spatially periodic systems in which, without a biasing force, a directed current of particles can be established. In classical type systems, a directed current of particles may arise under asymmetry conduction for a mixed phase space.[28] The corresponding quantized system may exhibit a significant ratchet behavior, even in a fully chaotic regime.[29] Such a behavior, which occurs in a variant of the kicked rotor and which can be related to the underlying classical dynamics, has been observed recently in experiments using an ultracold atom.[30] Very recently, an experimental realization associated with the quantum resonance of a kicked particle for arbitrary values of the quasi-momentum has been reported.[31] However, the theoretical study of the phenomena of quantum diffusion remains wide open.

In the present work, we address the problem of quantum Brownian motion in a ratchet potential to obtain the generalized form of the Einstein relation. In Section II, starting from a microscopic Hamiltonian picture, we obtain the quantum mechanical operator Langevin equation from a particle, which is coupled to a heat-bath, comprising a set of noninteresting harmonic oscillators. The potential is exposed to an external potential field V_e. Since the Langevin equation is an operator equation, it is not amenable to the classical differential equation-based approach. In this paper, we have derived a c-number-based generalized quantum Langevin equation (GQLE) from the operator form assuming a particular sort of initial condition following the formalism of Ray and co-workers.[32] In Section III, we present the calculation of quantum correction factors using a perturbation approach, which uses the first-order quantum correction term in closed analytical form, both in large friction and strong friction limits. Since the GQLE is not an operator but rather a c-number equation, the rest of our treatment relies on the differential equation-based approach. Calculating the relevant quantum correction factors in Section III, we then consider the diffusion of the particle in a tilted periodic potential in Section IV to obtain the generalized form of the Einstein relation, which is valid for any potential form and in the quantum regime. Finally, Section V is devoted to some conclusion and future work.

II. Theoretical Development

We allow the particle to interact with a dissipative thermal environment in an attempt to investigate quantum Brownian motion in a tilted periodic potential. Following the Zwanzig[33–35] form, we consider the Hamiltonian of the system−heat bath model in our development as follows (in a one-dimensional periodic potential):

$$\begin{aligned} \hat{H} &= \frac{\hat{p}^2}{2} + V(\hat{q}) + \sum_j \left[\frac{\hat{p}_j^2}{2} + \frac{1}{2}\left(\hat{q}_j - \frac{c_j}{\omega_j^2}\hat{q} \right)^2 \right] + V_e(\hat{q}) \\ &= \hat{H}_0 + H_{\mathrm{B}} + H_{\mathrm{SB}} + V_e(\hat{q}) \end{aligned} \tag{1}$$

where $H_0 = (\hat{p}^2/2) + V(\hat{q})$ is the bare system Hamiltonian, and $H_{\mathrm{B}} + H_{\mathrm{SB}} = \sum_j [(\hat{p}_j^2/2) + (1/2)(\hat{q}_j - (c_j/\omega_j^2)\hat{q})^2]$ is the bath Hamiltonian, which includes system−bath interaction. The sum contains the Hamiltonian for a set of N harmonic oscillators, which are bilinearly coupled with strength c_j to the system. In the above equation, $\hat{q}$ and $\hat{p}$ corresponds to the coordinate and momentum operators of the unit mass Brownian particle, respectively, $\{\hat{q}_j, \hat{p}_j\}$ is the set of coordinate and momentum operators for the mass weighted bath oscillators with characteristic frequency set $\{\omega_j\}$, and $V(\hat{q})$ is the external potential. The potential $V_e(\hat{q})$ emerges from the external force field acting on the unit mass Brownian particle. In our development, the coordinate and momentum operators mentioned above obey the following commutation relations:

$$[\hat{q}, \hat{p}] = i\hbar, \quad [\hat{q}_j, \hat{p}_k] = i\hbar\delta_{jk} \tag{2}$$

Using the traditional approach of eliminating the reservoir degrees of freedom,[5,36] we derive the following form of the quantum Langevin equation corresponding to the Hamiltonian given by eq 1:

$$\ddot{\hat{q}} + \int_0^t \mathrm{d}t' \gamma(t - t') \dot{\hat{q}}(t') + V'(\hat{q}) = \hat{\xi}(t) + F_{\mathrm{e}}(\hat{q}) \tag{3}$$

where the noise operator $\hat{\xi}(t)$, the memory kernel $\gamma(t)$, and $F_e(\hat{q})$ can be defined as

$$\hat{\xi}(t) = \sum_j c_j\left[\left(\hat{q}_j(0) - \frac{c_j}{\omega_j^2}\hat{q}(0)\right)\cos(\omega_j t) + \frac{\hat{p}_j(0)}{\omega_j}\sin(\omega_j t)\right] \tag{4}$$

$$\gamma(t) = \sum_j \frac{c_j^2}{\omega_j^2}\cos(\omega_j t) \tag{5}$$

$$F_e(\hat{q}) = -V_e'(\hat{q}) \tag{6}$$

In a true sense, eq 3 can be viewed as the GQLE in operator form. One can reach the Langevin equation via both classical as well as quantum mechanical routes. The variables of the system are transcribed to operators in the Heisenberg representation in the quantum mechanical treatment. The quantum Langevin equation, eq 3, is essentially the Heisenberg equation of motion for the (operator) coordinate of a Brownian particle coupled to a heat bath. The noise term $[\hat{\xi}(t)]$ in eq 3 obeys a nonvanishing commutator relation (see ref 37 for a detailed discussion) since it is an operator in the Hilbert space spanning the system as well as the environment. This property is imperative in order to stay consistent with Heisenberg uncertainty throughout the whole reduced dynamical evolution. It is worth noting that in the classical limit, the noise correlation reduces to the usual fluctuation−dissipation relation.

The temporal behavior of a quantum mechanical Brownian-like particle in the presence of dissipation is usually borne out by the GQLE. The time evolution governed by the quantum Langevin equation actually preserves the canonical commutation relation, i.e., eq 2, at one time, and this remains true for all other times. The GQLE, eq 3, traces its origin from the work of Magalinskiĭ[34] (in the absence of the potential renormalization term in the Hamiltonian eq 1). For a comprehensive review of this, we refer to the textbook by Weiss.[35]

As working with operator GQLE is a tedious job, we prefer to transfer it to its c-number analogue, cGQLE, which has been originally developed to simulate and analyze quantum dissipative dynamics.[32] To implement the formalism, we introduce product separable quantum states of the system and the bath oscillators at $t = 0$

$$|\psi\rangle = |\varphi\rangle\{|\alpha_j\rangle\} \tag{7}$$

In eq 7, $|\phi\rangle$ represents an arbitrary initial state of the unit mass Brownian particle, and $\{|\alpha_j\rangle\}$ describes the initial coherent state of the jth bath oscillator. In the present development $\{|\alpha_j\rangle\}$ can be constructed as

$$\{|\alpha_j\rangle\} = \exp\left(-\frac{1}{2}\left|\alpha_j\right|^2\right)\sum_{n_j=0}^{\infty}\left(\frac{\alpha_j^{n_j}}{\sqrt{n_j!}}\right)|n_j\rangle \tag{8}$$

We now define the shifted coordinate and momentum of the jth oscillator in terms of α_j as

$$\langle q_j(0)\rangle - \langle\hat{q}(0)\rangle = \sqrt{\frac{\hbar}{2\omega_j}}(\alpha_j + \alpha_j^*) \tag{9}$$

$$\langle\hat{p}_j(0)\rangle = -i\sqrt{\frac{\hbar\omega_j}{2}}(\alpha_j - \alpha_j^*) \tag{10}$$

To derive the desired cGQLE we then perform the quantum statistical averaging starting from an initial product separable quantum state[32] and obtain the following expression for cGQLE:

$$\ddot{q}(t) + \int_0^t \mathrm{d}t'\gamma(t - t')\dot{q}(t') + V(q') = \xi(t) + Q_0(q, t) + Q_e(q) \tag{11}$$

where

$$q(t) = \langle\hat{q}(t)\rangle \tag{12}$$

$$\xi(t) = \langle\hat{\xi}(t)\rangle = \sum_j\left[\{\langle\hat{q}_j(0)\rangle - \langle\hat{q}(0)\rangle\}\frac{c_j}{\omega_j^2}\cos(\omega_j t) + \sqrt{\frac{c_j}{\omega_j^2}}\langle\hat{p}_j(0)\rangle\sin(\omega_j t)\right] \tag{13}$$

In the above expression, Q_0 and Q_e are the quantum correction terms, given by

$$Q_0(q, t) = V'(q) - \langle V'(\hat{q})\rangle \tag{14}$$

$$Q_e(q) = V_e'(q) - \langle V_e'(\hat{q})\rangle \tag{15}$$

To describe $\xi(t)$ as an effective c-number noise, one must have properties of $\xi(t)$ such that it is centered around zero and satisfies the quantum fluctuation dissipation relation. This is possible if and only if the initial quantum mechanical mean values of momenta and coordinates of the bath oscillators have the following distribution:[32]

$$\boldsymbol{P}_j = N\exp\left[-\frac{\omega_j^2\{\langle\hat{q}_j(0)\rangle - \langle\hat{x}_j(0)\rangle\}^2 + \langle\hat{p}_j(0)\rangle^2}{2\hbar\omega_j\left(\bar{n}_j + \frac{1}{2}\right)}\right] \tag{16}$$

where N is the normalization constant and

$$\bar{n}_j = \left[\exp\left(\frac{\hbar\omega_j}{k_B T}\right) - 1\right]^{-1}$$

is the average photon number at temperature T. At this juncture, it is important to note that $\mathbf{P}_j$ can be viewed as a canonical Wigner distribution for a displaced harmonic oscillator, which always remains positive.[38] Thus, $\xi(t)$ satisfies the following properties:

$$\langle\xi(t)\rangle_S = 0 \tag{17}$$

$$\langle \xi(t)\xi(t')\rangle_S = \frac{1}{2}\sum_j \frac{c_j}{\omega_j^2}\hbar\omega_j \coth\left(\frac{\hbar\omega}{2k_BT}\right) \times \cos \omega_j(t - t') \tag{18}$$

with $\langle \cdots \rangle_S$ describing the statistical average taken over the initial distribution of the mean values of the momenta and coordinates of the bath oscillators. Thus, the statistical average of any observable $O_j(\langle \hat{p}_j(0)\rangle, \{\langle \hat{q}_j(0)\rangle - \langle \hat{q}(0)\rangle\})$ describing the quantum mechanical mean value is defined as

$$\langle O_j(0)\rangle_S = \int O_j(\langle \hat{p}_j(0)\rangle, \{\langle \hat{q}_j(0)\rangle - \langle \hat{q}(0)\rangle\}) \times \mathbf{P}_j d\langle \hat{p}_j(0)\rangle d\{\langle \hat{q}_j(0)\rangle - \langle \hat{q}(0)\rangle\} \tag{19}$$

Equation 11, which is our required cGQLE for the quantum Brownian particle in the continuum limit, is monitored by the c-number quantum noise, $\xi(t)$, emerging from the heat bath. It is important to note that the properties of $\xi(t)$ are controlled by eqs 17 and 18. Actually, one can easily verify the properties, eqs 17 and 18, of the c-number noise via eqs 12, 16, and 19. $Q_0(q, t)$ and $Q_e(q)$ are the two quantum fluctuation terms arising due to the nonlinearity of the system potential $V(\hat{q})$ and the external potential $V_e(\hat{q})$, respectively. To identify the c-GQLE, eq 11, we have to sweep to the continuum limit. Also, in the context of our present work, one must have detailed knowledge about the two quantum fluctuation terms, which will be discussed at length in the next section.

Now we impose some conditions on the coupling coefficients c_j, on the bath frequency ω_j and on the number N of the bath oscillators to ensure that $\gamma(t)$ is indeed dissipative. A sufficient condition for $\gamma(t)$ to be dissipative is that it is positively definite and decreases monotonically with time, which can be achieved if $N \to \infty$ and if both $c_j\omega_j^2$ and ω_j are sufficiently smooth functions of j. As $N \to \infty$, one replaces the sum by an integral over ω weighted by a density of states $\rho(\omega)$. Thus, to obtain a finite result in the continuum limit, the coupling function $c_j = c(\omega)$ is chosen as

$$c(\omega) = \frac{c_0}{\omega\sqrt{\tau_c}}$$

Consequently, $\gamma(t)$ reduces to the following form

$$\gamma(t) = \frac{c_0^2}{\tau_c}\int d\omega\rho(\omega)\cos(\omega t) \tag{20}$$

where c_0 is some constant and $1/\tau_c$ is the cutoff frequency of the bath oscillators. $\rho(\omega)$ is the density of modes of the heat bath and is assumed to be Lorentzian:

$$\rho(\omega) = \frac{2}{\pi\tau_c(1 + \omega^2\tau_c^2)} \tag{21}$$

Using the above forms of $\rho(\omega)$ and $c(\omega)$, we have the expressions for $\gamma(t)$ as

$$\gamma(t) = \frac{c_0^2}{\tau_c}\exp\left(-\frac{|t|}{\tau_c}\right) = \frac{\gamma}{\tau_c}\exp\left(-\frac{|t|}{\tau_c}\right) \tag{22}$$

with $\gamma = c_0^2$. For $\tau_c \to 0$, $\gamma(t) = 2\gamma\delta(t)$. On the other hand, the noise correlation function, eq 18, becomes

$$\langle \xi(t)\xi(t')\rangle_S = \frac{\gamma}{2\tau_c}\int_0^\infty d\omega\hbar\omega \coth\left(\frac{\hbar\omega}{2k_BT}\right) \times \rho(\omega)\cos\omega(t - t') \tag{23}$$

Equation 23 is an exact expression for the quantum statistical average of two time correlation functions of $\hat{\xi}(t)$. As $\{\hbar\omega \coth(\hbar\omega/2k_BT)\}$ is a smooth function of ω, at least for not too low temperatures, the integral can be approximated as

$$\langle \xi(t)\xi(t')\rangle_S \simeq \frac{\gamma}{2\tau_c}\coth\left(\frac{\hbar\omega_0}{2k_BT}\right) \times \int_0^\infty d\omega\hbar\omega\rho(\omega) \times \cos\omega(t - t') \tag{24}$$

This approximation is well-known and frequently used in quantum optics. Here, ω_0 is the average (or mean) frequency of the bath. Defining

$$D_0 = \frac{\gamma}{2}\hbar\omega_0\left[\bar{n}(\omega_0) + \frac{1}{2}\right] \tag{25}$$

the above expression, eq 24, in the limit $\tau_c \to 0$ reduces to

$$\langle \xi(t)\xi(t')\rangle_S = 2D_0\delta(t - t') \tag{26}$$

The analysis presented above cannot be claimed to be fully quantum, rather a quasi-quantum one where the system is treated quantum mechanically and the bath is treated quasi-quantum mechanically. We emphasize at this juncture that, in spite of the fact that the c-number noise $\xi(t)$ yields the same anticommutator as that of the operator noise term $\hat{\xi}(t)$, being a c-number term, the commutator of $\xi(t)$ vanishes. This semiclassical scheme helps us to deal with the complicated operator quantum Langevin equation in the same footing as that of the classical Langevin equation retaining the quantum effects in the leading orders. We point out that the projection operator method of Zwanzig[33] is in principle applicable to any system, but the operator is too complicated for practical applications.

At this point, we can now write the required generalized c-number analogue of GQLE, from eq 11, as

$$\ddot{q}(t) + \gamma\dot{q}(t) + V'(q) + V_e'(q) = \xi(t) + Q_0(q, t) + Q_e(q) \tag{27}$$

where the noise term $\xi(t)$ satisfies the relations

$$\langle \xi(t)\rangle_S = 0, \quad \langle \xi(t)\xi(t')\rangle_S = 2D_0\delta(t - t') \tag{28}$$

III. Quantum Correction

In this section, we present the calculation of the two quantum correction terms, $Q_0(q, t)$ and $Q_e(q)$. Considering the quantum nature of the system, one can write the system operators $\hat{q}$ and $\hat{p}$ as follows:

$$\hat{q}(t) = q(t) + \delta\hat{q}(t), \quad \hat{p}(t) = p(t) + \delta\hat{p}(t) \tag{29}$$

where q (= $\langle\hat{q}\rangle$) and p (= $\langle\hat{p}\rangle$) are the quantum mechanical mean values, and $\delta\hat{q}$ and $\delta\hat{p}$ are the operators, which follow

$$\langle\delta\hat{q}\rangle = \langle\delta\hat{p}\rangle = 0, \quad [\delta\hat{q}, \delta\hat{p}] = i\hbar \tag{30}$$

Now using eq 29 and a Taylor series expansion around q, one obtains

$$Q_0(q, t) = -\sum_{n\geq 2}\frac{1}{n!}V^{(n+1)}(q)\langle\delta\hat{q}^n(t)\rangle \tag{31}$$

$$Q_e(q) = -\sum_{n\geq 2}\frac{1}{n!}V_e^{(n+1)}(q)\langle\delta\hat{q}^n(t)\rangle \tag{32}$$

where $V^{(n+1)}(q)$ and $V_e^{(n+1)}(q)$ are the $(n + 1)$th derivative of the potential $V(q)$ and $V_e(q)$ respectively, and calculation of the two quantum terms $Q_0(q, t)$ and $Q_0(q, t)$ depends on the quantum correction factor $\langle\delta\hat{q}^n(t)\rangle$. The quantum correction can be evaluated by solving the corresponding equations that govern the quantum corrections. Using eq 3 and combining with eq 29 one can derive the following quantum correction equation:

$$\begin{aligned}\delta\hat{\xi}(t) = \delta\ddot{q}(t) + \int_0^t dt'\gamma(t - t')\delta\dot{q}(t') + V''(q)\delta\hat{q}(t) + \\ V_e''(q)\delta\hat{q}(t) + \sum_{n\geq 2}\frac{1}{n!}V^{(n+1)}(q)\langle\delta\hat{q}^n(t)\rangle + \\ \sum_{n\geq 2}\frac{1}{n!}V_e^{(n+1)}(q)\langle\delta\hat{q}^n(t)\rangle\end{aligned} \tag{33}$$

with $\delta\hat{\xi}(t) = \hat{\xi}(t) - \xi(t)$.

As we calculate $\langle\delta\hat{q}(t)\rangle$ in a perturbative way, we restrict our calculation to the leading order of the quantum correction factors, and consider that the confining potential is harmonic, i.e., $V(q) = (1/2)\Omega_0^2 q^2$, that yields the leading order quantum correction factors as

$$Q_0(q, t) = -\frac{1}{2}V'''(q)\langle\delta\hat{q}^2(t)\rangle \tag{34}$$

$$Q_e(q) = -\frac{1}{2}V_e'''(q)\langle\delta\hat{q}^2(t)\rangle \tag{35}$$

which are adequate to calculate the quantum correction factors including $\langle\delta\hat{q}^2(t)\rangle$ terms. With this approximation (in the absence of external forcing) the quantum correction equation becomes

$$\delta\ddot{q}(t) + \int_0^t \mathrm{d}t'\gamma(t - t')\delta\dot{q}(t') + \Omega_0^2\delta\hat{q}(t) = \delta\hat{\xi}(t) \tag{36}$$

The solution of eq 36 can be written as

$$\delta\hat{q}(t) = G(t)\delta\hat{q}(0) + H(t)\delta\dot{q}(0) + \int_0^t \mathrm{d}t'H(t - t')\delta\hat{\xi}(t'), \tag{37}$$

where $G(t)$ and $H(t)$ are the inverse Laplace transformation of $\tilde{G}(s)$ and $\tilde{H}(s)$, respectively, where

$$\tilde{G}(s) = \frac{s + \tilde{\gamma}(s)}{s + s\tilde{\gamma}(s) + \omega_0^2} \tag{38}$$

$$\tilde{H}(s) = \frac{1}{s + s\tilde{\gamma}(s) + \omega_0^2} \tag{39}$$

with

$$\tilde{\gamma}(s) = \int_0^\infty \gamma(t)\exp(-st)dt \tag{40}$$

being the Laplace transformation of the dissipation kernel $\gamma(t)$.

Squaring eq 37 and taking the quantum statistical average one can obtain the relevant quantum correction term $\langle\delta\hat{q}^2(t)\rangle$ as follows:

$$\begin{aligned}\langle\delta\hat{q}^2(t)\rangle = G^2(t)\langle\delta\hat{q}^2(0)\rangle + H^2\langle\delta\hat{p}^2(0)\rangle + \\ G(t)H(t)\langle(\delta\hat{q}(0)\delta\hat{p}(0) + \delta\hat{p}(0)\delta\hat{q}(0))\rangle + \\ 2\int_0^t dt'\int_0^{t'} dt''H(t - t')H(t - t'') \times \langle\delta\hat{\xi}(t')\delta\hat{\xi}(t'')\rangle\end{aligned} \tag{41}$$

The standard choices of initial conditions corresponding to the minimum uncertainty state are[32,39]

$$\begin{aligned}\langle\delta\hat{q}^2(0)\rangle = \frac{\hbar}{2\Omega_0}, \quad \langle\delta\hat{p}^2(0)\rangle = \frac{\hbar\Omega_0}{2}, \\ \langle(\delta\hat{q}(0)\delta\hat{p}(0) + \delta\hat{p}(0)\delta\hat{q}(0))\rangle = \hbar\end{aligned} \tag{42}$$

To calculate $\langle\delta\hat{q}^2(t)\rangle$, one needs to know the exact forms of the functions $H(t)$ and $G(t)$, which from the definitions of the same, can be written as

$$H(t) = \frac{1}{2\pi i}\int_{-i\infty+\varepsilon}^{i\infty+\varepsilon}\tilde{H}(s)\exp(st)\,\mathrm{d}s \tag{43a}$$

$$G(t) = \frac{1}{2\pi i}\int_{-i\infty+\varepsilon}^{i\infty+\varepsilon}\tilde{G}(s)\exp(st)\,\mathrm{d}s \tag{43b}$$

Using the residue theorem, it is easy to show that, for the Ohmic dissipative bath and for the underdamped case,

$$H(t) = \exp\left(-\frac{\gamma t}{2}\right)\left[\frac{1}{\Omega_1}\sin(\Omega_1 t)\right] \tag{44}$$

$$G(t) = \exp\left(-\frac{\gamma t}{2}\right)\left[\cos(\Omega_1 t) + \frac{\gamma}{2\Omega_1}\sin(\Omega_1 t)\right] \tag{45}$$

where $\Omega_1 = \pm[\Omega_0^2 - (\gamma^2/4)]^{1/2}$. For the overdamped case, Ω_1 becomes imaginary and both $H(t)$ and $G(t)$ modify to

$$H(t) = \exp\left(-\frac{\gamma t}{2}\right)\left[\frac{1}{\Omega_1'}\sin(\Omega_1' t)\right] \tag{46}$$

$$G(t) = \exp\left(-\frac{\gamma t}{2}\right)\left[\cos(\Omega_1' t) + \frac{\gamma}{2\Omega_1'}\sin(\Omega_1' t)\right] \quad (47)$$

where $\Omega_1' = \pm[(\gamma^2/4) - \Omega_0^2]^{1/2}$. Now for the Ohmic heat bath, the double integral in eq 41 yields

$$I = \frac{\gamma}{\pi}\int_0^\infty \mathrm{d}\omega\hbar\omega \coth\left(\frac{\hbar\omega}{2k_\mathrm{B}T}\right) \times \left|\frac{1 - \mathrm{e}^{-(\gamma/2 - i\omega)t}\left[\cos\Omega_1 t + (\gamma/2 - i\Omega_1)\frac{\sin\Omega_1 t}{\Omega_1}\right]}{\omega^2 - \Omega_0^2 + i\gamma\omega}\right|^2 \quad (48)$$

The time dependence of the mean fluctuation in displacement is complicated in eq 48, but it reduces to a simple form for large time compared to $1/\gamma$ and is given by

$$\langle\delta\hat{q}^2\rangle_\mathrm{eq} = \frac{\gamma}{\pi}\int_0^\infty \mathrm{d}\omega\left\{\hbar\omega\coth\left(\frac{\hbar\omega}{2k_\mathrm{B}T}\right) \times \frac{1}{(\omega^2 - \Omega_0^2)^2 + \gamma^2\omega^2}\right\} \quad (49)$$

In the strong damping regime ($\gamma \gg \omega$), eq 49 becomes

$$\langle\delta\hat{q}^2\rangle_\mathrm{eq} = \frac{\hbar}{\pi\gamma}\left[2\ln\left(\frac{\gamma}{\Omega_0}\right) + \ln\left(\frac{\hbar\Omega_0^2}{2\pi\gamma k_\mathrm{B}T}\right) + \left(\frac{\pi\gamma k_\mathrm{B}T}{\hbar\Omega_0^2}\right) - \left(\frac{\pi k_\mathrm{B}T}{\hbar\gamma}\right)^2 + \gamma_\mathrm{E}\right] \quad (50)$$

where $\gamma_\mathrm{E} = 0.57772$ is Euler's constant.

At this point we mention that we use eq 50, rather than the full-blown time-dependent version of $\langle\delta\hat{q}^2\rangle$, just in an attempt to visualize the diffusion effect in the steady-state situation. Here we also mention that the formulation developed by us in the present paper is valid under the conditions of $k_\mathrm{B}T \gg \hbar\omega$ (as the c-number Langevin equation is basically a type of high temperature approximation) and $\gamma \gg \omega$, the combined effect of which ultimately leads to the validity under $\gamma\hbar\beta \ll 1$. In that sense, eq 50 is a statement of the strong friction limit of the position variance. Here we reemphasize that, in the derivation of eq 49, we have explicitly implemented the high temperature approximation ($k_\mathrm{B}T \gg \hbar\omega$), followed by an inclusion of the condition that $\gamma \gg \omega$ to arrive at eq 50. In light of this we may say that eq 50 is a consequence of a two-step approximation ultimately leading to its validity in the regime of $\gamma\hbar\beta \ll 1$.

Now for static tilting force F and for harmonic oscillator, the above leading order correction factor requires no approximation as, in this case, Q_e becomes zero, and the expression given in eq 50 is exact (in the overdamped case). However, when the particle is moving in a sinusoidal potential with the same static tilting force F, $\langle\delta\hat{q}^2\rangle_\mathrm{eq}$ cannot be calculated exactly as the integro-differential eq 33 becomes strongly nonlinear, which requires some approximations in the calculation of $\langle\delta\hat{q}^2\rangle_\mathrm{eq}$. To illustrate this, we consider the dynamics of an underdamped oscillator:

$$\ddot{q} = -B^2\sin q$$

which can be replaced by the linear differential equation

$$\ddot{q} = -b^2 q$$

where b^2 depends on initial conditions, i.e., on the amplitudes. In the above linearization, we have replaced the nonlinear spring by a linear one such that the stored energies (both for linear and nonlinear oscillators) are equal for the same amplitudes, say C,

$$B^2\int_0^C \sin q\,dq = \frac{1}{2}b^2C^2$$

which results in

$$b^2 = 2B^2\frac{(1 - \cos C)}{C^2}$$

Let us define a quantity e, measuring the error due to the linearization

$$e = b^2q - B^2\sin q \quad (51)$$

which represents the difference between the linear and the nonlinear restoring terms. Here, 'e' is a function of b^2 and q or of b^2 and t if a solution for $q(t)$ is assumed of the form

$$q(t) = C\sin bt$$

The function $b^2(C)$ is now determined in such a way so that the mean square error

$$\frac{1}{T}\int_0^T e^2(b^2, t)\,\mathrm{d}t$$

is minimized for a fixed T. Consequently, one has the condition

$$\frac{\partial}{\partial b^2}\int_0^T e^2(b^2, t)\,\mathrm{d}t = \int_0^T 2e\frac{\partial e}{\partial b^2}\,\mathrm{d}t = 0 \quad (52)$$

Now use of eq 51 results in

$$\int_0^T 2(b^2q^2 - B^2q\sin q)\,\mathrm{d}t = 0$$

and hence

$$b^2 = B^2\frac{\int_0^T q\sin q\,\mathrm{d}t}{\int_0^T q^2\,\mathrm{d}t} \quad (53)$$

If the solution $q(t) = C\sin bt$ of the linear equation $\ddot{q} = -b^2q$ is substituted in the integral of eq 53, then it can be computed for $T = 2\pi/b$

$$\int_0^T C\sin bt\sin(C\sin bt)\,\mathrm{d}t = \frac{2\pi}{b}CJ_1(C)$$

which then results in

$$b^2 = B^2\left(\frac{2J_1(C)}{C}\right) \tag{54}$$

Here, J_k is the kth order Bessel function of the first kind. Now the quantity C is determined from the initial energy stored in the oscillator

$$B^2\int_0^C \sin q\, dq = B^2(1 - \cos C) = \frac{1}{2}b^2C^2 \tag{55}$$

Now use of eq 54 in 55 indicates that C can be calculated from the solution of the equation

$$CJ_1(C) + \cos C = 1 \tag{56}$$

Thus, for the system potential $V(q) = -B\cos q$, the harmonic frequency Ω_0, to the first order of approximation may be taken as

$$\Omega = \Omega_0\left(\frac{2J_1(C)}{C}\right) \tag{57}$$

where C can be determined using eq 56. Thus, to the first order of approximation, the quantum correction is given by

$$\langle\delta\hat{q}^2\rangle_{\mathrm{eq}} = \frac{\hbar}{\pi\gamma}\left[2\ln\left(\frac{\gamma}{\Omega}\right) + \ln\left(\frac{\hbar\Omega^2}{2\pi\gamma k_{\mathrm{B}}T}\right) + \left(\frac{\pi\gamma k_{\mathrm{B}}T}{\hbar\Omega^2}\right) - \left(\frac{\pi k_{\mathrm{B}}T}{\hbar\gamma}\right)^2 + \gamma_{\mathrm{E}}\right] \tag{58}$$

IV. Derivation of Generalized Einstein Relation Using Physically Motivated Approximation

We now consider the overdamped version of eq 27 with a static tilting force F

$$\dot{q} = -\tilde{B}\sin q + \tilde{F} + \tilde{\xi}(t) + \tilde{Q}_0 \tag{59}$$

where the quantities $\tilde{B}$, $\tilde{F}$, $\tilde{\xi}$, and $\tilde{Q}_0$ are scaled by γ. In the rest of the analysis, we remove the $\sim$ for notational simplicity. In addition to that, the above equation contains no correction due to static tilting force (see eq 31) but has a leading order quantum correction term Q_0

$$Q_0 = -B\sin q\langle\delta\hat{q}^2\rangle_{\mathrm{eq}} \tag{60}$$

and hence eq 59 reads as

$$\dot{q} = -A\sin q + F + \xi(t) \tag{61}$$

with

$$A = B(1 - \langle\delta\hat{q}^2\rangle_{\mathrm{eq}}) \tag{62}$$

The average velocity $v = \langle\dot{q}\rangle$ and the effective diffusion coefficient D can be explicitly calculated as

$$v = \frac{[1 - \exp(-2\pi F/D_0)]}{\int_0^{2\pi}(I_+(q)/2\pi)\, dq} \tag{63}$$

and

$$D = D_0\frac{\int_0^{2\pi}(I_+^2(q)I_-(q)/2\pi)\, dq}{[\int_0^{2\pi}(I_+(q)/2\pi)\, dq]^3} \tag{64}$$

with

$$I_\pm(q) = \frac{1}{D_0}\int_0^{2\pi} dy\, \times \exp\left[\frac{\{\mp A\cos q \pm A\cos(q \mp y) - yF\}}{D_0}\right] \tag{65}$$

It is important to mention here that, the quantity D_0 used in the above and in the following equations is a scaled quantity whose original value is D_0/γ^2. Now for $F = 0$, $I_\pm(q)$ becomes

$$I_\pm(q) = \exp\left\{\frac{\mp A\cos q}{D_0}\right\}I_{\pm 0} \tag{66}$$

with

$$I_{\pm 0} = \frac{1}{D_0}\int_0^{2\pi} dy\, \exp\left[\frac{\pm A\cos y}{D_0}\right] \tag{67}$$

For sufficiently small F, v and D are given by

$$v = \frac{(2\pi F)/D_0}{I_{+0}I_{-0}/(2\pi D_0)}$$

$$D = \frac{k_{\mathrm{B}}TI_{+0}^2I_{-0}^2/(2\pi D_0^3)}{[I_{+0}I_{-0}/(2\pi D_0)]^3} = \frac{4\pi^2 D_0}{I_{+0}I_{-0}}$$

which yields the famous Einstein relation given by D/v as

$$\frac{D}{v} = \frac{D_0}{F}$$

At this point we try to examine the nature of the average velocity and the effective diffusion coefficient given by eqs 63 and 64, respectively. This will also help us to gain better insight regarding the change of average velocity 63 and effective diffusion coefficient 64 with temperature. Therefore, one must resort to the numerical calculation for studying the effect of temperature on the average velocity and diffusion coefficient for which we have to find the profiles depicted in Figures 1 and 2. In order to check the sensitivity of the average velocity and diffusion coefficient with respect to the increase in tilting

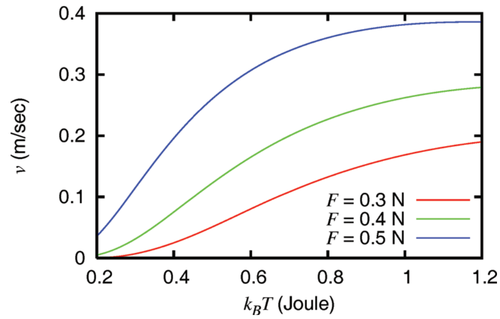

Figure 1. Plot of average velocity v as a function of temperature T for different values of tilting force F. The values of the parameters used are $\hbar = k_B = 1$, $\gamma_E = 0.57772$, $\gamma/\Omega = 10$.

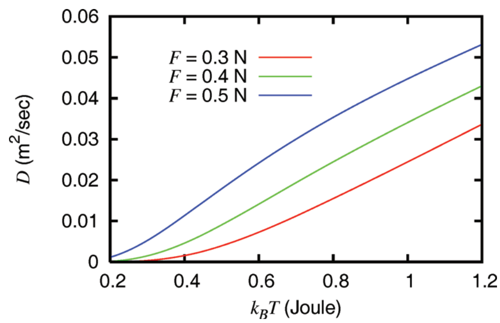

Figure 2. Plot of effective diffusion coefficient D as a function of temperature T for different values of tilting force F. The values of the parameters used are $\hbar = k_B = 1$, $\gamma_E = 0.57772$, $\gamma/\Omega = 10$.

force F, we have done calculations using three different F values. As can be seen clearly from the plots given in Figure 1, at the start the average velocity is zero and then increases with temperature and reaches the saturation limit. This behavior can be attributed to the very mode of definition of our model that noise-induced transport phenomena is physically meaningful only for $T < B$. As a consequence, it has been observed that the velocity increases with temperature within this region of temperature (i.e., when it is valid or is a good approximation) and moves consistently toward the saturation limit for $T > B$. Our results depicted in Figure 2 clearly indicate that the diffusion coefficient increases monotonically with increase in temperature for all F values. From our numerical analysis, presented in Figures 1 and 2, we have observed the enhancement of average velocity and diffusion coefficient with increase in F, as it should be. These outcomes are qualitatively consistent with our analytical analysis.

To underpin the intrinsic robustness of our formalism, it will be instructive to compare quantum feature associated with our model with the corresponding classical counterpart. To achieve this, we first redefine v as follows:

$$v = \frac{4\pi^2 F}{I_{0+}I_{0-}} \tag{68}$$

$$I_{0\pm} = \int_0^{2\pi} dy \exp\left[\pm\frac{A}{D_0}\cos y\right] = 2\pi I_0\left(\frac{A}{D_0}\right) \tag{69}$$

where I_0 is the modified Bessel function of the first kind. Hence,

$$v = \frac{F}{I_0^2\dfrac{A}{D_0}} \tag{70}$$

The argument of I_0 includes temperature. Classically, A will not depend on temperature and $D_0 \sim T$. Thus, the argument (A/D_0) of I_0 is equivalent to $1/T$. Graphically, if we plot v as a function of $1/T$, I_0^2 will increase for large $1/T$ as $I_0(x)$ increases monotonically with x, i.e., for small T. Thus for small T, v vanishes and v increases with T. Examining numerical analysis, we note that the mode of variation of the average velocity with temperature for the quantum and its classical counterpart is similar in nature.

When $F(F < A)$ is not so small and A/D_0 is sufficiently large, the Brownian particle is strongly trapped near the potential minimum, $q = q_0 + 2\pi n$, $n = 0, \pm1, \pm2, ...$ In this situation, the integral $I_+(q)$ can be written as

$$I_+(q) = \frac{1}{D_0}\exp\left[\frac{(-A\cos q - qF)}{D_0}\right]\times \int_{q-2\pi}^{q} dy \exp\left[\frac{(A\cos y + yF)}{D_0}\right]$$

where we have changed the variable $(q - y) \rightarrow y$. The integral of $I_+(q)$ is almost zero except near the potential minimum. If the integral of $I_+(q)$ is approximated using a Gaussian function near the potential minimum $y = q_0 - 2\pi$ (for $q < q_0$) or $y = q_0$ (for $q > q_0$), the integral $I_+(q)$ can be written as

$$I_+(q) \sim \sqrt{\frac{2\pi D_0}{A\cos q_0}}\left(\frac{1}{D_0}\right)\exp\left[\frac{(-A\cos q - qF)}{D_0}\right]\times \exp\left[\frac{[A\cos q_0 + (q_0 - 2\pi)F]}{D_0}\right]$$

for $q < q_0$ and

$$I_+(q) \sim \sqrt{\frac{2\pi D_0}{A\cos q_0}}\left(\frac{1}{D_0}\right)\exp\left[\frac{(-A\cos q - qF)}{D_0}\right]\times \exp\left[\frac{[A\cos q_0 + q_0F]}{D_0}\right]$$

for $q > q_0$. By similar induction,

$$I_-(q) \sim \sqrt{\frac{2\pi D_0}{A\cos q_0}}\left(\frac{1}{D_0}\right)\exp\left[\frac{(A\cos q + qF)}{D_0}\right]\times \exp\left[\frac{[A\cos q_0 + (q_0 - \pi)F]}{D_0}\right]$$

for $q < (\pi - q_0)$ and

$$I_-(q) \sim \sqrt{\frac{2\pi D_0}{A\cos q_0}}\left(\frac{1}{D_0}\right)\exp\left[\frac{(A\cos q + qF)}{D_0}\right]\times \exp\left[\frac{[A\cos q_0 + (q_0 - 3\pi)F]}{D_0}\right]$$

for $q > (\pi - q_0)$, as the integrand of $I_-(q)$ becomes a maximum at $y = (\pi - q_0)$, for $q < (\pi - q_0)$ or at $y = (3\pi - q_0)$, for $q > (\pi - q_0)$. Thus, integrals in eqs 63 and 64 can be written as

$$\int_0^{2\pi} I_+(q)\,\mathrm{d}q = \sqrt{\frac{2\pi D_0}{A\cos q_0}}\left(\frac{1}{2\pi D_0}\right)\times \left\{\int_0^{q_0} dq\,\exp\left[\frac{(-A\cos q - qF)}{D_0}\right]\times \exp\left[\frac{[A\cos q_0 + (q_0-2\pi)F]}{D_0}\right] + \int_0^{2\pi} dq\,\exp\left[\frac{(-A\cos q - qF)}{D_0}\right]\times\exp\left[\frac{[A\cos q_0 + q_0F]}{D_0}\right]\right\} \tag{71}$$

$$\sim\frac{1}{A\cos q_0}\exp\left[\frac{[2A\cos q_0 + (2q_0-\pi)F]}{D_0}\right] \tag{72}$$

and

$$\int_0^{2\pi} I_+^2(q)I_-(q)dq \sim \left(\frac{1}{A\cos q_0}\right)^2\left(\frac{\pi}{D_0}\right)\times \exp\left[\frac{[4A\cos q_0 + 4q_0F]}{D_0}\right]\times\left[\exp\left(-\frac{2\pi F}{D_0}\right)+\exp\left(-\frac{4\pi F}{D_0}\right)\right] \tag{73}$$

where the integral is evaluated at $q = (\pi - q_0)$. The ratio D_0/v using the above expression and eqs 63 and 64 is then given by

$$\frac{D_0}{v} = \frac{\pi}{\tanh(\pi F/D_0)} \tag{74}$$

Equation 74 can be viewed as a generalized Einstein relation.

In the classical regime, neglecting quantum fluctuation, eq 74 yields the classical version of the generalized Einstein relation.

$$\frac{D_0}{v} = \frac{\pi v}{\tanh(\pi F/k_BT)} = \gamma(F)T_{\mathrm{eff}} \tag{75}$$

where $\gamma(F) = v/F$ and the effective temperature T_{eff} is given by

$$T_{\mathrm{eff}} = \frac{\pi F}{\tanh(\pi F/k_BT)}$$

For sufficiently small F, $\gamma(F) = \lim_{F\to 0}(\mathrm{d}v/\mathrm{d}F)$ and $T_{\mathrm{eff}} = T$, and thus the usual Einstein relation $D = \gamma(0)k_BT$ is recovered. The generalized Einstein relation is the simplest form of the fluctuation dissipation relation and is satisfied even in the nonlinear regime, providing a relation in v and F. It does not depend on the detailed form of the potential, and, as $(x/\tanh x) > 1$, the effective temperature T_{eff} is larger than T.

It is pertinent to mention here that, although the generalized Einstein relation does not depend on the detailed form of the potential, the diffusion coefficient D indeed depends on it.

In the quantum regime (see eq 74), the diffusion coefficient D can be written as

$$D = \gamma(F)D_{\mathrm{eff}}$$

with D_{eff} given by

$$D_{\mathrm{eff}} = \frac{\pi F}{\tanh(\pi F/D_0)} \tag{76}$$

which reduces to its classical counterpart in the high temperature limit.

V. Conclusion

In this paper, we analyze the quantum motion of a system in a dissipative Ohmic heat-bath in the presence of external field. Starting from the usual system−reservoir model, we obtain the operator Langevin equation, and then, starting from a particular initial condition, and following the formalism of Ray and coworkers, we obtain the *c*-number GQLE. Consequently, we calculate the quantum correction terms using a perturbation technique. This enables us to apply a classical differential equation-based approach to consider quantum diffusion in a tilted periodic potential. With the help of a very general approximation, we derived the Einstein's relation for the quantum Brownian particle in a ratchet-type potential in a very simple closed analytical form. We observed that the diffusion rate is independent of the detailed form of the potential both in the quantum and classical regime, which is the key observation of our study. It is worth noting that, for a quantum system coupled to a heat bath environment, Ankerhold et al.[25] studied the strong friction limit from the exact path integral formulation and derived the generalized quantum Smoluchowski equation, which incorporates a multiplicative noise term associated with the curvature of the potential. In the near future, we would like to study the diffusion process using Ankerhold's equation both analytically and numerically to compare the same with our present formulation. In the near future, we also intend to study the diffusion in the presence of state-dependent dissipation, which may lead us to realize phase-induced quantum-directed motion in various ratchet systems.

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