Time dependent current in a nonstationary environment: A microscopic approach
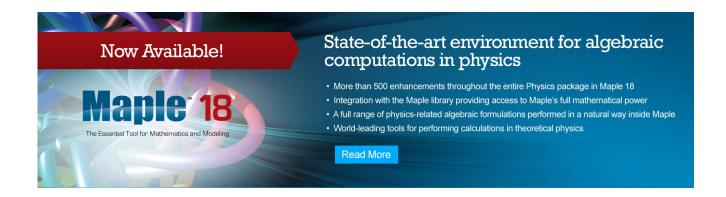
Satyabrata Bhattacharya, Suman Kumar Banik, Sudip Chattopadhyay, and Jyotipratim Ray Chaudhuri

Time dependent current in a nonstationary environment: A microscopic approach

Satyabrata Bhattacharya,[1] Suman Kumar Banik,[2,a]
Sudip Chattopadhyay,[1,b] and Jyotipratim Ray Chaudhuri[3,b]

[1]*Department of Chemistry, Bengal Engineering and Science University, Shibpur, Howrah 711103, India*
[2]*Department of Biological Sciences, Virginia Polytechnic Institute and State University, Blacksburg, Virginia 24061-0406, USA*
[3]*Department of Physics, Katwa College, Katwa, Burdwan 713130, India*

Based on a microscopic system reservoir model, where the associated bath is not in thermal equilibrium, we simulate the nonstationary Langevin dynamics and obtain the generalized nonstationary fluctuation dissipation relation (FDR) which asymptotically reduces to the traditional form. Our Langevin dynamics incorporates non-Markovian process also, the origin of which lies in the decaying term of the nonstationary FDR. We then follow the stochastic dynamics of the Langevin particle based on the Fokker–Planck–Smoluchowski description in ratchet potential to obtain the steady and time dependent current in an analytic form. We also examine the influence of initial excitation and subsequent relaxation of bath modes on the transport of the Langevin particle to show that the nonequilibrium nature of the bath leads to both strong nonexponential dynamics as well as nonstationary current. © *2008 American Institute of Physics.* [DOI: 10.1063/1.2942416]

I. INTRODUCTION

The traditional theory of noise induced transport deals with a Langevin equation describing the motion of a model Brownian particle in an external periodic potential, spatially symmetric or asymmetric.[1–5] The nature of asymmetry of the external force field, in which the Brownian particle is moving, is crucial in generating biased directed motion. While moving in a symmetric potential, the Brownian particle is unable to generate motion in a preferred direction due to the detailed balance principle which can be broken easily by applying an external time dependent perturbation, either deterministic or random. The correlation time of the external perturbation needs to be greater than the correlation time of the fluctuations which the system experiences from its immediate surroundings, the heat bath. A general approach in this direction involves the application of a time periodic deterministic field or the application of a colored noise to the system of interest.[1,3,4] Adopting a different approach, one can create directed motion by putting the Brownian particle in a biased asymmetric periodic potential from the very beginning. The spatial bias in the potential is able to overcome the detailed balance principle and hence can generate motion in a preferred direction.[2–4]

The theory of directed motion has gained wide interdisciplinary attention to model the phenomena of noise induced transport, where the interplay of fluctuations and nonlinearity of the system plays an important role.[1–5] Exploitation of the nonequilibrium fluctuations present in the medium helps generate directed motion of the Brownian particle. The presence of spatial anisotropy in the potential together with nonequilibrium perturbation enables one to extract useful work

---

[a]Electronic mail: skbanik@vt.edu.
[b]Authors to whom correspondence should be addressed. Electronic addresses: sudip_chattopadhyay@rediffmail.com and jprc_8@yahoo.com.

from random fluctuations without violating the second law of thermodynamics.[1,3] This leads to its wide applicability in explaining the mechanism of molecular motors,[2,3,6] tunneling in a Josephson junction,[3] rotation of dipoles in a constant field,[3] phase locked loop,[3] directed transport in photovoltaic and photoreflective materials,[7] and efficiency of a tiny molecular machine in a highly stochastic environment.[4,5,8] Motor proteins such as kinesins, dyenins, and myosins are versatile biomolecular shuttle cargo encapsulated in vesicles and are present in the different parts of the cell. In living cells, transport occurs via the cytoskeletal filaments and motor proteins.[6,9] Motor proteins are also important ingredients of the mechanism of muscle contraction and cell division.[9] The search for physical principles that enable such tiny molecular machines to function efficiently in a highly Brownian regime and construction of artificial molecular rotors which produce controlled directional motion mimicking molecular motor proteins[10] are the subject of ongoing interest.

During the past two decades, several theoretical models have been proposed using the idea of a Brownian particle moving in a ratchet potential[1–4] to explain the transport mechanism under various nonequilibrium situations. The ratchet model and its many variants such as rocking ratchet,[1] diffusion ratchet,[11] correlation ratchet,[12] flashing ratchet,[13] etc., have found wide attention in recent days.[3] To get a unidirectional current, either spatially asymmetric periodic potentials or time asymmetric external forces are necessary in these models. In explaining the above mentioned directional transport phenomena, most of the theoretical approaches adopt phenomenological models. The first self-consistent microscopic attempt was made by Millonas[14] in the context of construction of a Maxwell demon-like information engine that extracts work from a heat bath. In this microscopic construction, the Hamiltonian for the whole system includes a subsystem, a thermal bath, and a nonequilibrium bath that represents an information source or sink.[14]

In this article, we consider a simple variant of the system reservoir Hamiltonian[14] to model the directional transport processes where the associated bath is in a nonequilibrium state. The model incorporates some of the features of Langevin dynamics with a fluctuating barrier[15] and the kinetics due to space dependent friction along with the presence of local hot spots.[16–18] Since the theories of transport processes traditionally deal with stationary bath, the nonstationary transport processes have remained largely overlooked so far. We specifically address this issue and examine the influence of initial excitation and subsequent relaxation of bath modes[14,19–23] on the transport of system particle. We show that relaxation of the nonequilibrium bath modes may result in strong nonexponential kinetics and nonstationary current. The physical situation that has been addressed is that at $t=0_-$, the time just before the system and the bath are subjected to an external excitation, the system is appropriately thermalized. At $t=0$, the excitation is switched on and the bath is thrown into a nonstationary state which behaves as a nonequilibrium reservoir. We follow the stochastic dynamics of the system mode after $t>0$. The separation of the time scales of the fluctuations of the nonequilibrium bath and the thermal bath to which it relaxes is such that the former effectively remains stationary on the fast correlation of the thermal noise.[19]

The organization of the paper is as follows: We discuss in Sec. II a microscopic model necessary to compute the transient transport process where the system in question is not initially thermalized and the associated bath is thrown into a nonequilibrium and nonstationary situation by sudden initial excitation of some of the bath modes. Appropriate elimination of the reservoir degrees of freedom leads to a non-Markovian Langevin equation, stochasticity being contributed by both additive thermal noise and the multiplicative noise due to relaxing nonequilibrium modes. In Sec. III, following the prescription of Ref. 24, the Fokker–Planck description is provided in position space which is valid for state dependent dissipation. We then derive the time dependent solution of the associated Smoluchowski equation for probability density function (PDF). As an application of our development, in Sec. IV, we consider the motion of a Langevin particle in a periodic ratchet potential and obtain the stationary and time dependent average velocity of the Langevin particle and show that for symmetric periodic potential, the direction of average velocity depends on the initial excitation of intermediate bath modes. Summarizing remarks are presented in Sec. V.

II. THE BACKGROUND AND THE MODEL

To make the paper self-contained, we first discuss the essential features of the traditional theory of system reservoir dynamics in this section and then describe the model we adopt in the present work. This shows how our model deviates from the usual system reservoir theory and brings up the new features of our model.

A. The traditional system reservoir model

In the traditional system reservoir model,[25–27] the reservoir is assumed to be in equilibrium at $t=0$ in the presence of the system, and the appropriate distribution of the initial state of the heat bath is governed by the Hamiltonian

$$H_B + H_{SB} = \sum_\nu \left[\frac{p_\nu^2}{2m_\nu} + \frac{m_\nu \omega_\nu^2}{2} \left(q_\nu - \frac{g_\nu x}{m_\nu \omega_\nu^2} \right)^2 \right], \tag{1}$$

which includes the static interaction part H_{SB} between the system and the reservoir. The total Hamiltonian of the system plus bath is then usually written as

$$H = \frac{p^2}{2} + V(x) + H_B + H_{SB}. \tag{2}$$

In Eqs. (1) and (2), the system (mass weighted) is described by the coordinate x and the conjugate momentum p, and the heat bath, composed of a set of linear harmonic oscillators, is described by the coordinate q_ν and the conjugate momenta p_ν, $\nu = 1, 2, \ldots, N$. m_ν is the mass of the νth oscillator and ω_ν is the corresponding frequency. The system bath interaction is generally taken to be linear in both the system and the bath coordinates through the coupling constant g_ν. $V(x)$ represents the external force field in which the Brownian particle is executing random motion. The bath is assumed to be in thermal equilibrium at temperature T and the initial distribution is considered to be a canonical one:[25–27]

$$W[\mathbf{q}(0), \mathbf{p}(0)] = \frac{1}{Z} \exp\left(-\frac{H_B + H_{SB}}{k_B T} \right), \tag{3}$$

where Z is the normalization constant and k_B is the Boltzmann constant. To derive the dynamical equations for the system in terms of x and p, one usually eliminates the bath degrees of freedom from the equations of motion of the system variable[25] and obtains

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

$$\dot{p} = -V'(x) - \int_0^t d\tau \gamma(t-\tau) p(\tau) + \xi(t), \tag{4}$$

where $\gamma(t)$ is the memory kernel,

$$\gamma(t) = \sum_\nu \frac{g_\nu^2}{m_\nu \omega_\nu^2} \cos \omega_\nu t,$$

and $\xi(t)$ is the forcing function,

$$\xi(t) = \sum_\nu g_\nu \left[\left\{ q_\nu(0) - \frac{g_\nu}{m_\nu \omega_\nu^2} x(0) \right\} \cos \omega_\nu t + \frac{p_\nu(0)}{m_\nu \omega_\nu} \sin \omega_\nu t \right]. \tag{5}$$

Having chosen a distribution for the initial state of the bath, given by Eq. (3), the fluctuating force $\xi(t)$ becomes zero centered, and the correlation function of $\xi(t)$ gives the celebrated fluctuation dissipation relation[25–27] (FDR)

$$\langle \xi(t)\xi(t') \rangle = k_B T \gamma(t - t'). \tag{6}$$

To complete the identification of Eq. (4) as a generalized Langevin equation, one must establish the conditions on the coupling coefficients g_ν on the bath frequency ω_ν and on the number N of the bath oscillators which ensure that $\gamma(t)$ is indeed dissipative. Sufficient conditions for $\gamma(t)$ to be dissipative are that it is positive definite and decreases monotonically with time. Both the conditions are achieved if $N \to \infty$ and if $g_\nu/m_\nu \omega_\nu^2$ and ω_ν are sufficiently smooth functions of ν.[28] As $N \to \infty$, one replaces the sum by an integral over ω weighted by a density of states $D(\omega)$ to get

$$\gamma(t) = \int d\omega D(\omega)c(\omega)\cos(\omega t), \tag{7}$$

with $(g_\nu/m_\nu \omega_\nu^2) \to c(\omega)$. For

$$D(\omega)c(\omega) = \frac{\gamma/\tau_c}{1 + \tau_c^2 \omega^2}, \tag{8}$$

which can be achieved by a variety of combinations of the density of states $D(\omega)$ and the coupling function $c(\omega)$ and which broadly resembles the behavior of hydrodynamic model in a macroscopic system,[26] the dissipation kernel $\gamma(t)$ becomes

$$\gamma(t) = \frac{\gamma}{\tau_c}\exp(-|t|/\tau_c). \tag{9}$$

τ_c in the above expression is the cutoff frequency and is characterized as the correlation time of the bath. In the limit $\tau_c \to 0$, $\gamma(t) \to 2\gamma\delta(t)$ and one obtains the traditional Langevin equation in the Markovian domain,

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

$$\dot{p} = -V'(x) - \gamma p + \xi(t), \tag{10}$$

where $\langle \xi(t) \rangle = 0$ and $\langle \xi(t)\xi(t') \rangle = 2\gamma k_B T \delta(t - t')$. If one considers the dynamics of the Brownian particle in a periodic potential $V(x) = V(x+L)$, whose spatial symmetry can be broken by an external load (force), thereby creating a biased force field, then the system's dynamics is governed by

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

$$\dot{p} = -V'(x) - \gamma p + \xi(t) + F, \tag{11}$$

where F is the external force. The sum of the periodic potential $V(x)$ and the potential $-Fx$ due to the external force F, i.e., $U(x) = V(x) - Fx$, is a corrugated plane whose average slope (a measurement of the bias) is determined by the external force F.[29]

Equation (11) is the standard Langevin equation of a particle moving in an external potential under an external load force and is Markovian in nature. In addition to that, the dissipation term γ is constant due to the linear system reservoir coupling g_ν and the noise term $\xi(t)$ is Gaussian, additive in nature, reflecting the Markovian kinetics of the Brownian particle. In Sec. II B, we show how this Markovian kinetics changes to a non-Markovian one due to the sudden excitation of the few bath modes and splits the noise term $\xi(t)$ into two parts.

B. The nonstationary system reservoir model

We consider a Brownian particle of unit mass, described by the coordinate x and the conjugate momentum p, moving in a periodic potential of periodicity L, i.e., $V(x+L) = V(x)$. It is acted upon by an external force F, which for the present study is assumed to be constant and time indepen-

dent. The system mode is coupled to a set of relaxing modes considered as a semi-infinite dimensional system ($\{q_k\}$-subsystem) which effectively constitutes a nonequilibrium bath.[14,19,22] These $\{q_k\}$ modes are in contact with a thermally equilibrated reservoir. Both the reservoirs are composed of two sets of harmonic oscillators of unit mass characterized by the frequency sets $\{\omega_k\}$ and $\{\Omega_j\}$ for the nonequilibrium and the equilibrium bath, respectively. The system reservoir combination evolves under the total Hamiltonian

$$H = \frac{p^2}{2} + V(x) - Fx + \frac{1}{2}\sum_j (P_j^2 + \Omega_j^2 Q_j^2) + \frac{1}{2}\sum_k (p_k^2 + \omega_k^2 q_k^2) - x\sum_k \kappa_j Q_j - g(x)\sum_k q_k - \sum_{j,k} \alpha_{jk} q_k Q_j.$$

(12)

In Eq. (12), κ_j is the coupling constant describing the coupling of the system with the equilibrium bath modes and $g(x)$ is the coupling function. The term $g(x)\Sigma_j q_j$ indicates the coupling of the nonequilibrium bath to the system and the last term describes the coupling between the nonequilibrium bath and the thermal bath with coupling constant α_{jk}. The equilibrium bath is assumed to be in thermal equilibrium at a temperature T and the initial distribution of equilibrium bath variables is assumed to Gaussian. The form of the nonequilibrium bath, that of a set of phonons or photons, is chosen for both simplicity and because of its generic relationship to many condensed-matter-type systems.[27]

By eliminating the equilibrium bath variables $\{Q_j, P_j\}$ in the traditional way,[25–27] one may show that the nonequilibrium bath modes obey the dynamic equations

$$\dot{q}_k = p_k,$$

$$\dot{p}_k = -\gamma p_k - \omega_k^2 q_k - g(x) + \eta_k(t).$$

(13)

Equation (13) takes into account the average dissipation γ of the nonequilibrium reservoir modes q_k due to their coupling to the thermal reservoir which induces fluctuations $\eta_k(t)$ characterized by the usual FDR $\langle \eta_k(t)\eta_k(0)\rangle = 2\gamma k_B T\delta(t)$.[19,25] In general, $\langle \eta_k(t)\rangle$ is a nonzero constant quantity which, without loss of any generality, may be chosen as zero by shifting the origin of our coordinate system as we are dealing with a periodic potential. In passing we mention that in deriving Eq. (13) from Eq. (12), the cross terms for $\Sigma_j \gamma_{kj} q_j$ have been neglected.

Proceeding similarly to eliminate the thermal reservoir variables from the equations of motion of the system, we obtain

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

$$\dot{p} = -\gamma_e p - V'(x) + F + \xi_e(t) + g'(x)\sum_k q_k,$$

(14)

where γ_e refers to the dissipation coefficient of the system mode due to its direct coupling to the thermal bath providing fluctuations $\xi_e(t)$. The statistical properties of $\xi_e(t)$ are $\langle \xi_e(t)\rangle = 0$ and $\langle \xi_e(t)\xi_e(t')\rangle = 2\gamma_e k_B T\delta(t-t')$. Comparing with Eq. (11), it is easy to see that the dissipation term γ_e and the noise term $\xi_e(t)$ are basically γ and $\xi(t)$, respectively, that arise due to the direct linear system reservoir coupling. Now making use of the formal solution of Eq. (13), which takes into account the relaxation of the nonequilibrium modes, and integrating over the nonequilibrium bath with a Debye-type frequency distribution of the form[19]

$$\rho(\omega) = \frac{3\omega^2}{2\omega_c^3} \quad \text{for } |\omega| \leq \omega_c$$

$$=0 \ \text{for} \quad |\omega| > \omega_c, \tag{15}$$

where ω_c is the high frequency Debye cutoff, one finally obtains the following Langevin equation for the system mode from Eq. (14):

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

$$\dot{p} = -\Gamma(x)p - \tilde{V}'(x) + F + \xi_e(t) + g'(x)\xi_n(t). \tag{16}$$

In the above equation,

$$\Gamma(x) = \gamma_e + \gamma_n[g'(x)]^2 \tag{17}$$

is the state dependent dissipation constant comprising of γ_n and γ_e. ξ_n refers to the fluctuations of the nonequilibrium bath modes which effectively cause damping of the system mode. This damping is also state dependent due to the nonlinear coupling function $g(x)$ as is given by $\gamma_n[g'(x)]^2$. In Eq. (16), the potential $V(x)$ in which the particle moves has been modified to

$$\tilde{V}(x) = V(x) - \frac{\omega_c}{\pi}\gamma_n g^2(x). \tag{18}$$

The fluctuation $\xi_n(t)$ due to the presence of nonequilibrium bath is also assumed to be Gaussian with zero mean $\langle\xi_n(t)\rangle=0$. Also, the essential properties of $\xi_n(t)$ explicitly depend on the nonequilibrium state of the intermediate oscillator modes $\{q_j\}$ through $u(\omega,t)$, the energy density distribution function at time t in terms of the following FDR for the nonequilibrium bath:[19]

$$u(\omega,t) = \frac{1}{4\gamma_n}\int_{-\infty}^{+\infty}d\tau\langle\xi_n(t)\xi_n(t+\tau)\rangle e^{i\omega\tau} = \frac{1}{2}k_BT + e^{-\gamma t/2}\left[u(\omega,0) - \frac{1}{2}k_BT\right]. \tag{19}$$

$[u(\omega,0)-(k_BT/2)]$ is a measure of the departure of energy density from thermal average at $t=0$. The exponential term $\exp(-\gamma t/2)$ implies that this deviation, due to the initial excitation, decays asymptotically to zero as $t\rightarrow\infty$, so that one recovers the usual FDR for the thermal bath.[19,22] Equation (19) thus attributes the nonstationary character of the $\{q_k\}$-subsystem. At this point it is pertinent to note that the above derivation is based on the assumption that $\xi_n(t)$ is effectively stationary on the fast correlation time scale of the equilibrium bath modes. This is necessary for the systematic separation of the time scales involved in the dynamics.

Equation (16) is the required Langevin equation for the particle moving in a modified potential $\tilde{V}(x)$ and is acted upon by a uniform force F. The motion of the particle is damped by a state dependent friction $\Gamma(x)$. Depending on the coupling function $g(x)$, both $\tilde{V}(x)$ and $\Gamma(x)$ are, in general, nonlinear in nature. As a result, the stochastic differential equation (16) becomes nonlinear. The fluctuating part in Eq. (16) is comprised of two quantities: $\xi_e(t)$, an additive noise due to thermal bath, and $\xi_n(t)$, a multiplicative noise due to nonlinear coupling to the $\{q_k\}$-subsystem. The Langevin equation (16) describes a non-Markovian process as well, where the non-Markovian nature is characterized by the decaying term in Eq. (19), describing the initial nonequilibrium nature of the $\{q_k\}$-subsystem created by applying sudden excitation at $t=0$.[19,22]

III. STOCHASTIC DYNAMICS IN THE OVERDAMPED REGIME AND THE TIME DEPENDENT DISTRIBUTION

For large dissipation, i.e., in the overdamped limit, one usually eliminates the fast variable p adiabatically by omitting the inertial term dp/dt from the dynamical equations of motion to get a simpler description of the system in position space. The approach of adiabatically eliminating fast variables is valid on a much slower time scale and is a zero order approximation. For constant large dissipation, this adiabatic elimination of the fast variables leads to the correct description of the system's dynamics. However, in the presence of hydrodynamic interactions, i.e., when the

dissipation is state dependent, the traditional adiabatic reduction in fast variables does not work properly and gives an incorrect description of the system's dynamics. For state dependent dissipation, an alternative approach was proposed in Ref. 24. Using the method given in Ref. 24 and using Eq. (16), one may carry out a systematic expansion of the relevant variable in powers of $1/\gamma_e$ by neglecting terms smaller than $O(1/\gamma_e)$. Then, by the Stratonovich interpretation, it is possible to obtain the appropriate Langevin equation corresponding to a Fokker–Planck equation (FPE) in position space. Thus, following Ref. 24, the formal FPE for the PDF $P(x,t)$ corresponding to the process described by Eq. (16) can be obtained as

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial x}\left\{\frac{\widetilde{V}'(x) - F}{\Gamma(x)}P\right\} + \gamma_e k_B T \frac{\partial}{\partial x}\left\{\frac{1}{\Gamma(x)}\frac{\partial}{\partial x}\frac{1}{\Gamma(x)}P\right\} + \gamma_n k_B T(1 + re^{-\gamma t/2})\frac{\partial}{\partial x}\left\{\frac{g'(x)}{\Gamma(x)}\frac{\partial}{\partial x}\frac{g'(x)}{\Gamma(x)}P\right\}$$
$$+ \gamma_n k_B T(1 + re^{-\gamma t/2})\frac{\partial}{\partial x}\left\{\frac{g'(x)g''(x)}{\Gamma^2(x)}P\right\}, \tag{20}$$

where $r = \{[u(\omega \to 0,0)/2k_B T] - 1\}$ and is a measure of the deviation from equilibrium at $t=0$. Under the steady state condition (at $t \to \infty$), $\partial P/\partial t = 0$ and the stationary distribution obeys the following relation:

$$k_B T \frac{dP_S(x)}{dt} + (\widetilde{V}'(x) - F)P_S(x) = 0, \tag{21}$$

which has the solution

$$P_s(x) = N \exp\left[-\frac{1}{k_B T}\int^x (\widetilde{V}'(x') - F)dx'\right], \tag{22}$$

where N is the normalization constant. In the Stratonovich description, the Langevin equation corresponding to the FPE given by Eq. (21) is

$$\dot{x} = -\frac{(\widetilde{V}'(x) - F)}{\Gamma(x)} - \frac{\widetilde{D}(t)g'(x)g''(x)}{\Gamma^2(x)} + \frac{1}{\Gamma(x)}\xi_e(t) + \frac{g'(x)}{\Gamma(x)}\xi_n(t), \tag{23}$$

with $\widetilde{D} = \gamma_n k_B T(1 + r\exp(-\gamma t/2))$ being the time dependent diffusion constant due to the relaxation of nonequilibrium bath modes.[19] Let us consider that the time dependent solution of Eq. (20) is given by[20]

$$P(x,t) = P_S(x)\exp(-\phi(t)), \tag{24}$$

where ϕ is a function of time only and $\lim_{t\to\infty}\phi(t) = 0$. $P_S(x)$ is the steady state solution of Eq. (20),

$$\frac{d}{dx}\left\{\frac{(\widetilde{V}'(x) - F)}{\Gamma(x)}P_S(x)\right\} + \gamma_e k_B T\frac{d}{dx}\left\{\frac{1}{\Gamma(x)}\frac{d}{dx}\frac{1}{\Gamma(x)}P_S(x)\right\} + \gamma_n k_B T\frac{d}{dx}\left\{\frac{g'(x)}{\Gamma(x)}\frac{d}{dx}\frac{g'(x)}{\Gamma(x)}P_S(x)\right\}$$
$$+ \gamma_n k_B T\frac{d}{dx}\left\{\frac{g'(x)g''(x)}{\Gamma^2(x)}P_S(x)\right\} = 0. \tag{25}$$

Substitution of Eq. (24) in Eq. (20) separates the space and time parts and we have the dynamic equation for $\phi(t)$,

$$-\frac{d\phi}{dt}\exp(\gamma t/2) = \text{const} = \alpha(\text{say}).$$

On integration over time, we get

$$\phi(t) = \frac{2\alpha}{\gamma}\exp(-\gamma t/2), \tag{26}$$

where α can be determined from the initial condition. The time dependent solution of Eq. (20) thus reads as

$$P(x,t) = P_S(x)\exp\left[-\frac{2\alpha}{\gamma}\exp(-\gamma t/2)\right]. \tag{27}$$

To determine α, we now demand that just at the moment the system (and the nonthermal bath) is subjected to external excitation at $t=0$, the distribution must coincide with the usual Boltzmann distribution where the energy term in the Boltzmann factor, in addition to the usual kinetic and potential energy terms, contains the initial fluctuation of energy density $\Delta u[=u(\omega,0)-(k_BT/2)]$. This demands that

$$\alpha = \frac{\gamma\Delta u}{2k_BT}, \tag{28}$$

α is thus determined in terms of relaxing mode parameters and fluctuations of the energy density distribution at $t=0$.

IV. STATIONARY AND TRANSIENT CURRENT

In the overdamped limit, the stationary current from Eq. (25) can be represented as

$$J_S = -\frac{1}{\Gamma(x)}\left[\tilde{V}'(x) - F + k_BT\frac{d}{dx}\right]P_S(x). \tag{29}$$

Integrating Eq. (29) we have the expression for stationary probability distribution in terms of stationary current as

$$P_S(x) = e^{-U(x)}h(x)\left[\frac{P_S(0)}{h(0)} - \frac{J_S\gamma_e}{k_BT}\int_0^x h(x')e^{U(x')}dx'\right] \tag{30}$$

where $h(x)=1+(\gamma_n/\gamma_e)[g'(x)]^2$, $\Gamma(x)=\gamma_e h(x)$, and $U(x)=\gamma_e\int_0^x dx'h(x')[\tilde{V}'(x')-F]/k_BT$. We now consider a symmetric periodic potential with periodicity L, i.e., $V(x)=V(x+L)$, as well as the periodic derivative of coupling function with the same periodicity as that of the potential, i.e., $g'(x)=g'(x+L)$. As a consequence of this choice, $U(x)$ is also a periodic function of x with the period L. If we impose the condition that $P_S(x)$ is bounded for large enough x, it follows from the above mentioned conditions of periodicity that $P_S(x+L)=P_S(x)$ i.e., $P_S(x)$ must be periodic with the same period L.[29] Now applying the periodicity condition of $P_S(x)$, we have from Eq. (30)

$$\frac{P_S(0)}{h(0)} = J_S\frac{\gamma_e/k_BT}{1-e^{U(L)}}\int_0^L h(x)e^{U(x)}dx. \tag{31}$$

Because of the periodicity, we normalize the steady state PDF in the periodic interval,

$$\int_0^L P_S(x)dx = 1, \tag{32}$$

to get

$$\int_0^L h(x)e^{-U(x)}\left[\frac{P_S(0)}{h(0)} - \frac{J_S\gamma_e}{k_BT}\int_0^x h(x')e^{U(x')}dx'\right]dx = 1. \tag{33}$$

Now eliminating $P_S(0)/h(0)$ from Eqs. (31) and (33), one obtains the steady state current

$$J_S = \frac{k_B T}{\gamma_e}[1 - e^{U(L)}]\left[\int_0^L h(x)e^{-U(x)}dx \int_0^L h(x')e^{U(x')}dx'\right.$$

$$\left. - [1 - e^{U(L)}]\int_0^L (h(x)e^{-U(x)}\int_0^x h(x')e^{U(x')}dx')dx\right]^{-1}. \tag{34}$$

From Eq. (34) it is clear that in the absence of any external bias F, the steady current vanishes. We thus observe that there is no occurrence of current for a periodic potential and for periodic derivative of the coupling function with the same periodicity for $F=0$. At the macroscopic level, this confirms that there is no generation of perpetual motion of the second kind, i.e., no violation of the second law of thermodynamics. In passing, we note that in the absence of $\{q_k\}$-subsystem, i.e., when $\gamma_n=0$, Eq. (34) reduces to the standard form[29]

$$J_S = L\gamma_e k_B T[1 - e^{LF/k_B T}]\left[\int_0^L e^{V(x)/k_B T}dx \int_0^L e^{-V(x)/k_B T}dx - [1 - e^{-2LF/k_B T}]\right.$$

$$\left. \times \left\{\left(\int_0^L e^{-V(x)/k_B T}\int_0^x e^{V(x')/k_B T}dx'\right)dx\right\}\right]^{-1}. \tag{35}$$

Next, to find the time dependent current $J(x,t)$, we resort to Eq. (20) and observe that

$$J(x,t) = -\frac{\partial}{\partial x}\left\{\frac{\widetilde{V}(x')-F}{\Gamma(x)}P\right\} + \gamma_e k_B T\left\{\frac{1}{\Gamma(x)}\frac{\partial}{\partial x}\frac{1}{\Gamma(x)}P\right\} + \gamma_n k_B T(1 + re^{-\gamma t/2})\left\{\frac{g'(x)}{\Gamma(x)}\frac{\partial}{\partial x}\frac{g'(x)}{\Gamma(x)}P\right\}$$

$$+ \gamma_n k_B T(1 + re^{-\gamma t/2})\left\{\frac{g'(x)g''(x)}{\Gamma^2(x)}P\right\}. \tag{36}$$

Now substituting Eq. (24) in Eq. (36) and making use of Eq. (25), we find that $J(x,t)$ can be expressed in a much simpler form,

$$J(x,t) = J_S e^{-\phi(t)} - \mathcal{D}(t)\frac{1}{\Gamma(x)}\frac{d}{dx}\frac{1}{\Gamma(x)}[g'(x)]^2 P_S(x), \tag{37}$$

where $P_S(x)$ is the steady state PDF and J_S is the steady state current given by Eq. (35) and

$$\mathcal{D}(t) = r\gamma_n k_B T e^{-\gamma t/2}e^{-\phi(t)}. \tag{38}$$

The steady state current J_S thus can be obtained from

$$J_S = -\frac{(\widetilde{V}'(x)-F)}{\Gamma(x)}P_S(x) - \gamma_e k_B T\frac{1}{\Gamma(x)}\frac{d}{dx}\frac{1}{\Gamma(x)}P_S(x) - \gamma_n k_B T\frac{g'(x)}{\Gamma(x)}\frac{d}{dx}\frac{g'(x)}{\Gamma(x)}P_S(x)$$

$$- \gamma_n k_B T\frac{g'(x)g''(x)}{\Gamma^2(x)}P_S(x), \tag{39}$$

from which we have

$$\frac{1}{\Gamma(x)}\frac{d}{dx}\frac{1}{\Gamma(x)}[g'(x)]^2 P_S(x) = -\frac{J_S}{\gamma_n k_B T} - \frac{(\widetilde{V}'(x)-F)}{\gamma_n k_B T\Gamma(x)} - \frac{\gamma_e}{\gamma_n}\frac{1}{\Gamma(x)}\frac{d}{dx}\frac{1}{\Gamma(x)}P_S(x). \tag{40}$$

Using Eq. (40) we then obtain from Eq. (37)

$$J(x,t) = J_S\left[e^{-\phi(t)} + \frac{\mathcal{D}(t)}{\gamma_n k_B T}\right] + \frac{\mathcal{D}(t)}{\gamma_n k_B T}\left[\frac{(\widetilde{V}'(x)-F)}{\Gamma(x)}P_S(x) + \gamma_e k_B T\frac{1}{\Gamma(x)}\frac{d}{dx}\frac{1}{\Gamma(x)}P_S(x)\right]. \tag{41}$$

Defining the space dependent part on the right hand side of Eq. (41) as $M(x)$, we obtain

$$J(x,t) = J_S\left[e^{-\phi(t)} + \frac{\mathcal{D}(t)}{\gamma_n k_B T}\right] + \frac{\mathcal{D}(t)}{\gamma_n k_B T}M(x), \tag{42}$$

where

$$M(x) = \left[\frac{(\widetilde{V}'(x)-F)}{\Gamma(x)}P_S(x) + \gamma_e k_B T\frac{1}{\Gamma(x)}\frac{d}{dx}\frac{1}{\Gamma(x)}P_S(x)\right]. \tag{43}$$

From Eq. (42) we observe that the current $J(x,t)$ can be written as a sum of two terms. The first term is space independent and only a function of time. The second term is product separable in the form of time and space parts. As $t\rightarrow\infty$, right hand side of $\mathcal{D}(t)\rightarrow 0$ and asymptotically $J(x,t)$ reduces to the steady state current J_S. Now using the continuity equation

$$\frac{\partial P(x,t)}{\partial t} = -\frac{\partial J(x,t)}{\partial x},$$

along with $P(x,t) = P_S(x)e^{-\phi(t)}$, we get from Eq. (42)

$$\frac{dM(x)}{dx} = -\frac{\alpha}{r}P_S(x) \tag{44}$$

or equivalently,

$$M(x) = -\frac{\alpha}{r}\int^x P_S(x)dx. \tag{45}$$

As we are dealing with periodic functions, the constant of integration is chosen to be zero. Now integrating Eq. (29) for $P_S(x)$ and using the normalization condition [Eq. (32)], we have the expression for steady state PDF as

$$P_S(x) = e^{-(\widetilde{V}(x)-Fx)/k_B T}\left[\frac{1 + \frac{J_S}{k_B T}\int_0^L e^{-(\widetilde{V}(x)-Fx)/k_B T}\left\{\int_0^x \Gamma(x')e^{(\widetilde{V}(x')-Fx')/k_B T}dx'\right\}dx}{\int_0^L e^{-(\widetilde{V}(x)-Fx)/k_B T}dx}\right]. \tag{46}$$

Using Eq. (46) along with Eq. (45), one obtains from Eq. (42) the expression for the time dependent current $J(x,t)$ as

$$J(x,t) = J_S[e^{-\phi(t)} + re^{-\gamma t/2}] - \frac{\alpha e^{-\gamma t/2}\int^x dx'\{e^{-(\widetilde{V}(x')-Fx')/k_B T}\}}{\int_0^L e^{-(\widetilde{V}(x)-Fx)/k_B T}dx}\left[1 + \frac{J_S}{k_B T}\int_0^L e^{-(\widetilde{V}(x'')-Fx'')/k_B T}\right.$$

$$\left.\times\left\{\int_0^{x'}\Gamma(x''')\{e^{(\widetilde{V}(x''')-Fx''')/k_B T}dx'''\}dx''\right\}\right], \tag{47}$$

where J_S is given by Eq. (34). Since the potential possesses spatial periodicity, one has $J(x,t) = J(x+L,t)$. Hence the net time dependent current is given by

$$j(t) = \frac{1}{L}\int_0^L J(x,t)dx. \tag{48}$$

It should be noted that for symmetric potential with $F=0$, $J_S=0$. However, in our development, transient current exists and the direction of current depends on the sign of α. What is immediately

apparent is that for symmetric potential, the sign of $\Delta u[=u(\omega,0)-(1/2k_BT)]$ determines the direction of the initial current,

$$j(t) = -\frac{\alpha e^{-\gamma t/2}}{L}\frac{\displaystyle\int_0^L dx \int^x dx'\,\exp[-\widetilde{V}(x')/k_BT]}{\displaystyle\int_0^L dx\,\exp[-\widetilde{V}(x)/k_BT]}. \tag{49}$$

It is also clear from Eq. (47) that the time dependent current reduces to the steady state current J_S in the asymptotic limit. The presence of the term $\exp[-\phi(t)]$ in the expression of $J(x,t)$ makes the transient current strongly nonexponential in nature. The transient behavior of growth or decay of charge and current in *L-R*, *C-R*, or *L-C-R* circuit is important in construction of many electrical and electronic devices where there is the mechanism of storage of energy. In construction of molecular motor or nanoswitch, the transient behavior of the devices may be worth studying. In our development, the preparation of intermediate relaxing bath plays a key role in generating the time dependent current. Nevertheless, our methodology will also be applicable in the case when any arbitrarily prepared bath is approaching equilibrium. In passing, we mention that the model considered in the present paper may be realized in a guest-host system embedded in a lattice where the immediate neighborhood of the guest comprises intermediate oscillatory modes, while the lattice acts as a thermal bath.

V. CONCLUSION

We have hereby proposed a simple microscopic system nonequilibrium bath model to simulate nonstationary Langevin dynamics. The nonequilibrium bath is effectively realized in terms of a semi-infinite dimensional reservoir which is subsequently kept in contact with a thermal reservoir which allows the nonthermal bath to relax with characteristic time. The frequency spectrum of the relaxing bath is assumed to be of Debye type. By an appropriate separation of time scale, we then construct the Langevin equation for a particle in which the dissipation is state dependent and the stochastic forces appearing are both additive and multiplicative. The underlying stochastic dynamics is found to be nonstationary and non-Markovian. Based on the strategy of Sancho *et al.*,[24] we then show that this Langevin equation can be recast into the form of the generalized nonstationary Smoluchowski equation which reduces to its standard form asymptotically. We then solve the expression for time dependent PDF. As an immediate application of our recent development, we consider the dynamics of a Langevin particle in a ratchet potential and obtain the analytic expression for both stationary and nonstationary transient average velocities, which is followed by an immediate observation that in a periodic potential, the direction of nonstationary current depends on the preparation of the nonequilibrium bath.

[1] A. Ajdari and J. Prost, C. R. Acad. Sci., Ser. II: Mec., Phys., Chim., Sci. Terre Univers **315**, 1635 (1992); M. O. Magnasco, Phys. Rev. Lett. **71**, 1477 (1993); A. Ajdari, D. Mukamel, L. Peliti, and J. Prost, J. Phys. I **4**, 1551 (1994); R. Bartussek, P. Hänggi, and J. G. Kissner, Europhys. Lett. **28**, 459 (1994); M. M. Millonas and M. I. Dykman, Phys. Lett. A **185**, 65 (1994).
[2] F. Jülicher, A. Ajdari, and J. Prost, Rev. Mod. Phys. **69**, 1269 (1997).
[3] R. D. Astumian, Science **276**, 917 (1997); P. Reimann, Phys. Rep. **361**, 57 (2002).
[4] R. D. Astumian and P. Hänggi, Phys. Today **55**(11), 33 (2002).
[5] See *Ratchet and Brownian Motors: Basic Experiments and Applications*, edited by H. Linke, special issue of Appl. Phys. A: Mater. Sci. Process. 75, 167 (2002).
[6] J. Howard, *Mechanics of Motor Proteins and the Cytoskeletons* (Sinauer, Sunderland, 2001).

[7] P. J. Struman, *Photovoltaic and Photorefractive Effects in Nanocentrosymmetric Materials* (Gordon and Breach, Philadelphia, 1992).

[8] H. Linke, T. E. Humphery, A. Löfgren, A. O. Sushkov, R. Newbury, R. P. Taylor, and P. Omling, Science **286**, 2314 (1999).

[9] B. Alberts, A. Johnson, J. Lewis, M. Raff, K. Roberts, and P. Walter, *Molecular Biology of the Cell*, 4th ed. (Garland Science, New York, 2002).

[10] V. Balzani, A. Credi, F. M. Raymo, and J. F. Stoddart, Angew. Chem., Int. Ed. **39**, 3348 (2000); D. A. Leigh, J. K. Y. Wong, F. Dehez, and F. Zerbetto, Nature (London) **424**, 174 (2003).

[11] P. Reimann, R. Bartussek, R. Häussler, and P. Hänggi, Phys. Lett. A **215**, 26 (1996).

[12] C. R. Doering, W. Horsthemke, and J. Riordan, Phys. Rev. Lett. **72**, 2984 (1994).

[13] J.-F. Chauwin, A. Ajdari and J. Prost, Europhys. Lett. **32**, 373 (1995).

[14] M. M. Millonas, Phys. Rev. Lett. **74**, 10 (1995).

[15] P. Reimann and T. C. Elston, Phys. Rev. Lett. **77**, 5328 (1996).

[16] R. Landauer, J. Appl. Phys. **33**, 2209 (1962); J. Stat. Phys. **9**, 351 (1973).

[17] M. Porto, M. Urbakh, and J. Klafter, Phys. Rev. Lett. **85**, 491 (2000); G. Oshanin, J. Klafter, and M. Urbakh, Europhys. Lett. **68**, 26 (2004); G. Oshanin, J. Klafter, and M. Urbakh, J. Phys.: Condens. Matter **17**, S3697 (2005).

[18] J. R. Chaudhuri, D. Barik, and S. K. Banik, J. Phys. A **40**, 14715 (2007); J. R. Chaudhuri, S. Chattopadhyay, and S. K. Banik, J. Chem. Phys. **127**, 224508 (2007).

[19] J. R. Chaudhuri, G. Gangopadhyay, and D. S. Ray, J. Chem. Phys. **109**, 5565 (1998).

[20] J. RayChaudhuri, D. Barik, and S. K. Banik, Eur. Phys. J. B **55**, 333 (2007).

[21] R. Hernandez, J. Chem. Phys. **111**, 7701 (1999); R. Hernandez and F. L. Somer, J. Phys. Chem. B **103**, 1064 (1999).

[22] A. V. Popov and R. Hernandez, J. Chem. Phys. **126**, 244506 (2007).

[23] A. O. Bolivar, J. Math. Phys. **49**, 013301 (2008).

[24] J. M. Sancho, M. San Miguel, and D. Dürr, J. Stat. Phys. **28**, 291 (1982).

[25] R. Zwanzig, J. Stat. Phys. **9**, 215 (1973); K. Lindenberg and V. Seshadri, Physica A **109**, 483 (1981); M. I. Dykman and M. A. Krivoglaz, Phys. Status Solidi B **48**, 497 (1971).

[26] K. Lindenberg and B. J. West, *The Nonequilibrium Statistical Mechanics of Open and Closed Systems* (VCH, New York, 1990).

[27] A. Nitzan, *Chemical Dynamics in Condensed Phases* (Oxford University Press, Oxford, 2006).

[28] G. W. Ford, M. Kac, and P. Majur, J. Math. Phys. **6**, 504 (1965).

[29] H. Risken, *The Fokker-Planck Equation* (Springer, Berlin, 1989).