Diffusion dynamics in external noise-activated non-equilibrium open system–reservoir coupling environment*

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The diffusion process in an external noise-activated non-equilibrium open system–reservoir coupling environment is studied by analytically solving the generalized Langevin equation. The dynamical property of the system near the barrier top is investigated in detail by numerically calculating the quantities such as mean diffusion path, invariance, barrier passing probability, and so on. It is found that, comparing with the unfavorable effect of internal fluctuations, the external noise activation is sometimes beneficial to the diffusion process. An optimal strength of external activation or correlation time of the internal fluctuation is expected for the diffusing particle to have a maximal probability to escape from the potential well.

Keywords: external noise, diffusion, non-equilibrium open environment

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1. Introduction

The barrier escaping models of diffusion passing over the saddle point of a metastable potential well is ubiquitous in almost all scientific areas of physics and chemistry. A great amount of chemical events, such as chemical reactions, molecular diffusion, or collision of molecular systems, can be modeled by a single barrier escaping process within the framework of standard Brownian motion.[1–3] Great progress has been witnessed in the study of dynamical properties of these diffusion processes since the foundation of Kramers' reaction rate formula.[4–9] However, an explicit character can be found in reviewing the previous studies that the particle is always considered to tracing in a closed system plus reservoir environment where an equilibrium Boltzmann distribution is satisfied all the while. Therefore problems rise from this point that what a different diffusion process the particle will experience if the whole environment is somewhat non-equilibrium or non-thermal? Is it possible for a particle to escape from the well more easily or difficultly? The answers to these problems are of great interest to be pursued.

Recently, a particular kind of system–reservoir coupling environment is investigated where the heat bath is modulated by an external noise.[10,11] This system is thermodynamically open, so no Boltzmann distribution is satisfied now as well as some classical theories such as the fluctuation–dissipation theorem (FDT).[12,13] The single last straw one may rely on is a Boltzmann-like steady state distribution.[10] This leads great difficulty to the study of dynamical diffusion processes in this non-equilibrium environment. Fortunately, the kinetic information can still be obtained via the generalized Langevin equation (GLE) by assuming an independent prerequisite between the internal fluctuation and the external noise disturbance without any loss of generality. Therefore, in this paper we make our effort to present a detailed investigation on the kinetic barrier escaping process of the diffusing particle in an external noise-activated open environment. We aim to gather useful information as more as possible by calculating its mean diffusion path, invariance, barrier passing probability, and so on.

The paper is organized as follows. In Section 2, the influence of the external noise activation resulted in a barrier escaping process is unfolded before our eyes by analytically solving the relevant GLE. In Section 3, diffusion dynamical properties of the system is investigated in the neighborhood of the saddle point of an inverse harmonic potential by numerically calculating the mean diffusion path, invariance, and barrier passing probability. Section 4 serves as a summary of our conclusion where some further applications of this study are also prospected.

2. Role of the external noise activation

The non-equilibrium, non-thermal systems is full of physical scenarios in variant different contexts, e.g., it is familiar to us in examining the role of color noise in obtaining stationary probabilities,[14,15] the property of nonlinear systems,[16] the nature of crossover phenomena,[17] the effect of monochromatic noises,[18] the rate of diffusion-limited coagulation processes,[19] and so on. While these treatments

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concern a direct driving on the system by an external noise, the present consideration is based on a straight modulation on the heat bath.[16,20] Given the required stationarity of this non-equilibrium environment being maintained, the barrier crossing dynamics of the system will evolve amenable to the succeeding theoretical analysis that follows.

Let us begin with the Hamiltonian describing a system of particles with unit mass that is coupled in a bilinear form to an external noise modulated harmonic heat bath. Mathematically it reads[21]

$$H = \frac{1}{2}p^2 + U(x) + \sum_{j=1}^{N}\left[\frac{1}{2}p_j^2 + \frac{1}{2}\omega_j^2(q_j - c_j x)^2\right] + \sum_{j=1}^{N}\lambda_j q_j \varepsilon(t), \tag{1}$$

where $\{x, p\}$ and $\{q_j, p_j\}$ are the sets of coordinate and momentum variables of system and bath oscillators, respectively; $c_j x$ measures the system–bath coupling strength, the external noise $\varepsilon(t)$ modulation on the reservoir is represented by the last part on the right-hand side; $U(x)$ is the potential.

The coupling between the system and heat bath is usually believed to be quadriform resulting in three kinds of quasi-monochromatic structured noises.[22,23] However, in order to highlight the role of external noise modulation, we consider only the simplest case here, namely the coordinate (system) and coordinate (bath) coupling. This will lead to a harmonic quasi-monochromatic noise that is related to the friction kernel function via the FDT formula. Meanwhile, without any loss of generality, the external noise $\varepsilon(t)$ is also assumed to be stationary and Gaussian with zero mean and decaying second order correlation function.

By eliminating the bath variables in the usual way,[24–26] a generalized Langevin equation (GLE) can be obtained as

$$\ddot{x} + \int_0^t \mathrm{d}t'\gamma(t-t')\dot{x}(t') + \partial_x U(x) = \xi(t), \tag{2}$$

where $\xi(t)$ $(= \zeta(t) + \varepsilon(t))$ is an effective fluctuation containing two random forces of different originations. For explicitness, $\zeta(t) = \sum_j c_j\{[q_j(0) - c_j x_0]\omega_j^2 \cos\omega_j t + p_j(0)\omega_j \sin\omega_j t\}$ is the fluctuation force generated from internal system–reservoir coupling while $\varepsilon(t) = -\int_0^t \varphi(t-t')\varepsilon(t')\mathrm{d}t'$ with $\varphi(t) = \sum_j c_j\lambda_j\omega_j \sin\omega_j t$ is resulted from the modulation of external noise $\varepsilon(t)$. It seems at first sight that $\zeta(t)$ and $\varepsilon(t)$ in the GLE are equal in status with each other, but noticing they originate from different sources, the total fluctuation $\xi(t)$ in the GLE therefore cannot be directly connected to the friction kernel function $\gamma(t)$ now. But the FDT relation lies still between the internal noise $\zeta(t)$ and $\gamma(t)$.

Supposing $\zeta(t)$ is independent of $\varepsilon(t)$ so that we have $\langle\zeta(t)\varepsilon(t')\rangle = 0$, the correlation of $\xi(t)$ can then be yielded from

$$\langle\xi(t)\xi(t')\rangle = \langle\zeta(t)\zeta(t')\rangle + \langle\varepsilon(t)\varepsilon(t')\rangle_{\mathrm{e}}, \tag{3}$$

where $\langle\zeta(t)\zeta(t')\rangle = k_{\mathrm{B}}T\gamma(t-t')$ is resulted from the well-known FDT relation,[12,13] while $\langle\varepsilon(t)\varepsilon(t')\rangle_{\mathrm{e}} = 2D\psi(t-t')$ is a thermodynamic consistency condition instead which is reminiscent of the familiar FDT formula due to the appearance of the external noise intensity. Here $\langle\cdots\rangle_{\mathrm{e}}$ implies the averaging is taken independently over all the realizations of $\varepsilon(t)$ with strength D and $\psi(t)$ is a relevant memory kernel. The anomalous form of Eq. (2) suggests that the system is under the combining government of two forcing functions. This will lead to some nontrivial results as the changing of time.

Due to the Gaussian properties of the internal and external noises as well as the linearity of the GLE, the joint probability density function $W(x, v, t; x_0, v_0)$ of the system can still be written in a Gaussian form[27]

$$W(x, v, t; x_0, v_0) = \frac{1}{2\pi|\boldsymbol{A}(t)|^{1/2}}\,\mathrm{e}^{-\left[y^{\dagger}(t)\boldsymbol{A}^{-1}(t)y(t)\right]/2}, \tag{4}$$

where $y(t)$ is the vector $[x - \langle x(t)\rangle, v - \langle v(t)\rangle]$ and $\boldsymbol{A}(t)$ is the matrix of second moments with each component as

$$A_{11}(t) = \langle[x - \langle x(t)\rangle]^2\rangle, \tag{5a}$$

$$A_{12}(t) = A_{21}(t) = \langle[x - \langle x(t)\rangle][v - \langle v(t)\rangle]\rangle, \tag{5b}$$

$$A_{22}(t) = \langle[v - \langle v(t)\rangle]^2\rangle. \tag{5c}$$

The reduced distribution function can then be yielded by integrating Eq. (4) over v as

$$W(x, t; x_0, v_0) = \frac{1}{\sqrt{2\pi A_{11}(t)}}\exp\left[-\frac{(x - \langle x(t)\rangle)^2}{2A_{11}(t)}\right], \tag{6}$$

in which the average position $\langle x(t)\rangle$ and variance $A_{11}(t)$ can be obtained by Laplace solving the GLE. In the case of an inverse harmonic potential $U(x) = -m\omega_b^2 x^2/2$, it reads

$$\langle x(t)\rangle = \left[1 + \omega_b^2\int_0^t H(t')\mathrm{d}t'\right]x_0 + H(t)v_0, \tag{7a}$$

$$A_{11}(t) = \int_0^t \mathrm{d}t_1 H(t-t_1)\int_0^{t_1}\mathrm{d}t_2\langle\xi(t_1)\xi(t_2)\rangle H(t-t_2), \tag{7b}$$

where $H(t)$ namely the response function can be yielded from inverse Laplace transforming $\hat{H}(s) = [s^2 + s\hat{\gamma}(s) - \omega_b^2]^{-1}$ with the residue theorem.[28,29] The probability of passing over the saddle point, namely also the characteristic function which is crucial for the activated barrier crossing process, can then be determined mathematically by integrating Eq. (6) over x from zero to infinity. This results in

$$P(x_0, v_0; t) = \int_0^{\infty} W(x, t; x_0, v_0)\mathrm{d}x = \frac{1}{2}\mathrm{erfc}\left(-\frac{\langle x(t)\rangle}{\sqrt{2A_{11}(t)}}\right). \tag{8}$$

It reveals to be in a trivial form with no difference from the previous results. However the nontrivial kinetic information has actually been contained in all the time-dependent quantities such as $\langle x(t)\rangle$ and variance $A_{11}(t)$. In the following sections we will give a thorough investigation on this point by some detailed numerical calculation.

3. Diffusion dynamics near the barrier top

We consider concretely a particular case for example where the internal is an Ornstein–Uhlenbeck (OU) process[30,31] while the external noise is δ-correlated, i.e.,

$$\langle \zeta(t)\zeta(t')\rangle = k_{\rm B}T\frac{G}{\tau}\,{\rm e}^{-|t-t'|/\tau}$$

and

$$\langle \varepsilon(t)\varepsilon(t')\rangle_{\rm e} = 2\mathscr{D}\delta(t-t'), \tag{9}$$

both symmetric with respect to the time argument and assumed to be uncorrelated with each other. Consequently, the dissipative friction kernel resulting only from the internal fluctuation can be derived from the FDT relation as

$$\gamma(t-t') = \frac{G}{\tau}\,{\rm e}^{-|t-t'|/\tau}. \tag{10}$$

It is noted that for $\tau \to 0$, the internal noise process becomes also δ-correlated. The Laplace transform of $\gamma(t)$, as given above, can then be obtained as

$$\tilde{\gamma}(s) = \frac{G}{s\tau+1}. \tag{11}$$

Subsequently, we have for $\tau \neq 0$,

$$\tilde{H}(s) = \frac{s\tau+1}{(s^2-\omega_b^2)(s\tau+1)+Gs}, \tag{12}$$

and inverse Laplace transform Eq. (12) in succeeding according to the residue theorem[28,29]

$$H(t) = \frac{\mu\tau+1}{3\mu^2\tau+2\mu+G-\tau\omega_b^2}\,{\rm e}^{\mu t}, \tag{13}$$

where μ is the largest positive root of

$$s^3\tau+s^2+(G-\tau\omega_b^2)s-\omega_b^2 = 0. \tag{14}$$

Deriving from the abbreviations heretofore, all the kinetic quantities can be obtained till now.

In the following, we calculate numerically some of the crucial dynamic quantities to pursue a relatively thorough comprehension of the noise-activated diffusion process. Firstly in Fig. 1 instantaneous values of $\langle x(t)\rangle$ and $A_{11}(t)$ for various strengths of pure internal fluctuation ($\mathscr{D}=0$) are plotted. From which we can see that both $\langle x(t)\rangle$ and $A_{11}(t)$ diverge quickly as the time goes on. This is a trivial phenomenon from the point of view of diffusing across the saddle point. In a diffusing process, the center of the wave packet (distribution) moves forward and the width of which broadens, but in the long time this result in a steady probability for the particle to escape from the potential well as is explicitly shown in Fig. 2 where the instantaneous values of $P(x_0,v_0;t)$ at various strengths of pure internal fluctuation is plotted. Further, we can also find in Fig. 2 that the steady value of the barrier passing probability $P(x_0,v_0;t)$ decreases as the increasing of intensity G of the internal noise. This reveals that the fierceness of the internal fluctuation is always harmful to the diffusion process. Thus it leads to a low barrier passing probability.

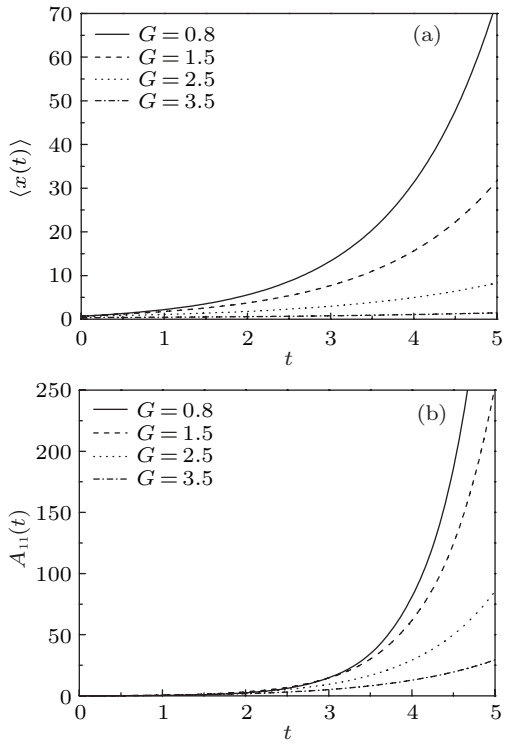


Fig. 1. Instantaneous values of $\langle x(t)\rangle$ (a) and variance $A_{11}(t)$ (b) for various strengths of pure internal fluctuation ($\mathscr{D}=0$). Dimensionless parameters used are $\omega_{\rm b}=1.0$ and $\tau=1.5$. Particles are assumed to start from $x_0=-1.0$ position with initial velocity $v_0=3.0$.

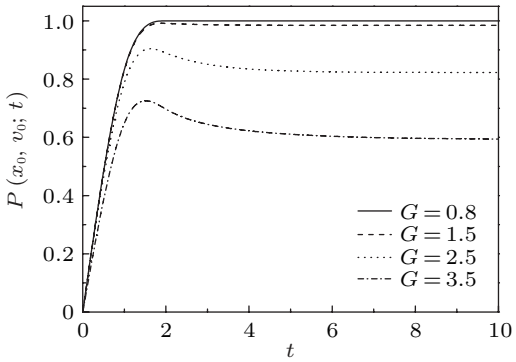


Fig. 2. Instantaneous values of $P(x_0,v_0;t)$ at various strengths of pure internal fluctuation. Identical parameters are used as those in Fig. 1.

However, the most important point that we concern in this study is actually the activation of external noise. Therefore in Fig. 3 we plot the instantaneous values of $A_{11}(t)$ and $P(x_0,v_0;t)$ at various intensities of external noise but left over that of $\langle x(t)\rangle$. This is because the noises are assumed to be initially uncorrelated. So the external fluctuation has no influence on the response function $H(t)$ as well as on $\langle x(t)\rangle$ in the sequel. The effect of the external noise settles only in $A_{11}(t)$ and thus results in a different $P(x_0,v_0;t)$. Seen from Fig. 3,

$A_{11}(t)$ diverges even more quickly in comparing with the pure internal case if the activation of an external noise is set on; however, the steady value of $P(x_0, v_0; t)$ is not always reduced (or enlarged) comparing to that of $\mathscr{D} = 0$ (pure internal case) as the increase of $\mathscr{D}$. It evolves as a non-monotonic function of $\mathscr{D}$ as is shown in Fig. 4 where $P_{\rm st}$ denotes the steady value of $P(x_0, v_0; t)$. This reveals that the external activation is not always harmful to the diffusing process, sometimes it is even beneficial to the diffusion. An optimal intensity of external noise activation is expected for the diffusing particle to have a maximal probability to escape from the barrier. In the particular case that is considered here this optimal value is about $\mathscr{D} = 3.5$ in dimensionless unit.

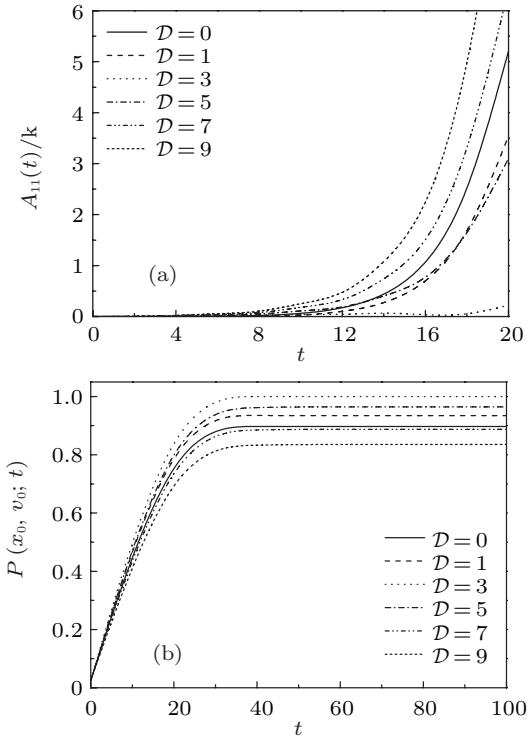


Fig. 3. Instantaneous values of $A_{11}(t)$ (a) and $P(x_0, v_0; t)$ (b) at various strengths of external noise. Dimensionless parameters used are $\omega_{\rm b} = 1.2$, $G = 3.5$, and $\tau = 1.0$. Particles are assumed to start from $x_0 = -1.0$ position with initial velocity $v_0 = 1.5$.

The noise-activated barrier escaping process is ubiquitously used to model many crucial physical scenarios such as the fusion of massive nuclei[32] where correlation time τ of the internal colored noise is an important factor in determining the fusion probability. Therefore the τ-dependent variation of the stationary barrier passing probability $P_{\rm st}$ is also calculated in this study as is plotted in Fig. 4(b). Seen from this figure, $P_{\rm st}$ emerges also as a non-monotonic function of τ. This is reminiscent of the non-Ohmic damping systems where $P_{\rm st}$ always evolves non-monotonically with the anomalous diffusion exponent.[8,9] It reveals that there also exists an optimal correlation time of the internal fluctuation for the diffusion particle to obtain a maximal barrier passing probability when the external activation is constantly conserved. This is easily explicable in the lights of stochastic resonance theory[33,34] and the previous results showing heretofore.

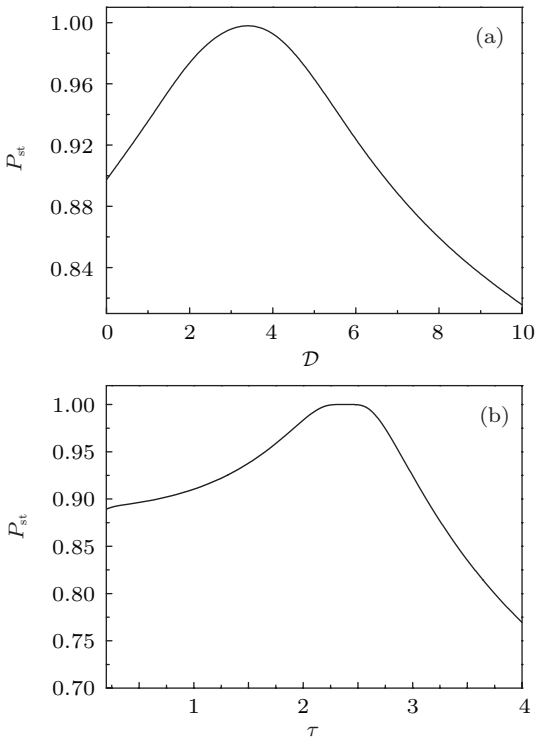


Fig. 4. Stationary values of $P(x_0, v_0; t)$ (i.e. $P_{\rm st}$) as a function of the intensity $\mathscr{D}$ (a) or correlation time τ (b). Where identical parameters are used as those in Fig. 3 except $\tau = 1.0$ in panel (a) and $\mathscr{D} = 9.0$ in panel (b).

4. Summary and discussion

In summary, we have studied in this paper the near barrier top diffusion mechanisms in an external noise-activated non-equilibrium open system–reservoir coupling environment. The dynamical properties is investigated in detail by analytically solving the generalized Langevin equation and numerically calculating the crucial relevant quantities. Contrary to the harmful effect of internal fluctuations, the external noise activation is found sometimes to be beneficial to the diffusion process. An optimal intensity of external activation or correlation time of the internal fluctuation is expected for the diffusing particle to have a maximal probability to escape from the barrier.

The external noise is generally believed to modify the dynamics of diffusion in two ways.[11] One is to influence the dynamics in the region around the barrier top so that the effective escaping rate across it gets modified. The other way is to disturb the equilibrium distribution of the initial environment. The reason for the occurrence of anomalous diffusion dynamics presented in this study obviously lies in the first one. Therefore, the results obtained heretofore may serve as a basis for studying processes such as those occurring within irreversibly driven environments[35,36] and for thermal ratchet

problems[37] as well as in further synthesis of some other non-equilibrium (or non-thermal) open systems.

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