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Escape of coupled Brownian particles across a fluctuating barrier

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The escape of two harmonically coupled Brownian particles across the fluctuating barrier of a bistable potential is investigated with correlated additive and multiplicative fluctuations. Positive correlations enhance the rate of escape across the barrier when the coupling is effective, whereas for weakly coupled particles, escape becomes difficult. It is found that the system exhibits the phenomenon of resonant activation when the rate of barrier fluctuations is comparable to the relaxation time in the bistable potential. Using a decoupling ansatz, we derive the Markovian limit of the problem in the steady state, under the constraint that the barriers fluctuate on a time scale faster than the relative oscillation of the two particles. Adiabatic elimination of the fast variable of the dynamical system is discussed in appropriate limits.

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I. INTRODUCTION

Thermally driven escape across potential barriers is a problem of general interest in domains varying from chemical kinetics to transport theory. The rate of thermal escape across a barrier was provided in the seminal work by Kramers [1] and has received several useful extensions since then [2–4]. However, in many cases of interest, the potential barrier confining the Brownian particle is itself randomly fluctuating with its own time scale [5], which may be at times comparable to the relevant time scales of the system [6]. A particular example is the escape of O_2 or CO ligands out of proteins after photodissociation [7]. Taking such fluctuations into account, Doering and Gadoua [8,9] showed in their pioneering work that the rate of escape across the fluctuating barrier of a bistable potential depends nonmonotonically on the rate of barrier fluctuations. This phenomenon of resonant activation was later experimentally investigated using an *RC* circuit with a tunnel diode [10], confirming the theoretical predictions. Later studies incorporated Gaussian fluctuations [11–13] to show a generic occurrence of the phenomenon [14–16], whenever the time scale of barrier fluctuations is comparable to the relaxation times in the system.

The above studies, however, have focused on independent fluctuations, which form a subset of a broader class of correlated fluctuations. Physically, such correlations arise naturally when the fluctuations have a common origin [17–19] and provide a new bifurcation branch in the dynamics of the system; e.g., a random dynamical system perturbed by two correlated additive noises can exhibit a purely deterministic behavior for the case of perfect anticorrelation, a nontrivial feature for a randomly perturbed dynamical system. Such correlations are known to affect both the transient and steady-state dynamical properties of a particle in a bistable potential [20–25]. This is because correlations control the relative distribution of power from the source to different fluctuations, e.g., additive and multiplicative, and lead to observed behaviors with varying correlation. Apart from being of theoretical interest, the dynamics in a bistable potential is also of technological relevance [26] wherein correlated noises are employed to achieve asymmetric confinement in one of the bistable states.

In many cases of interest, the system under investigation is not a single particle but a collection of particles, e.g., a dimer or a polymer. In addition, the escape of dimers and polymers [27] across potential barriers has been of considerable interest for independent noise sources. Recent studies have shown that the presence of correlations [28] significantly modifies the escape process. In particular, the rate of escape of a dimer vanishes for the case of strong anticorrelations, at any finite temperature. Motivated by these observations, we take up the study of escape properties of two harmonically coupled Brownian particles in a bistable potential with a fluctuating barrier. Such a study is directly relevant to the escape of O_2 like molecules across proteins, which contain additional vibrational degrees of freedom, and also has implications towards the understanding of two-headed molecular motors like kinesin [29]. In the present example, the barrier fluctuations associated with the two particles are colored Gaussian and correlated with each other, making the problem intrinsically non-Markovian. In addition, the heat baths associated with the two particles are chosen to be Gaussian white and correlated. However, in the present study we choose barrier fluctuations to arise independently of thermal fluctuations. We find that the system exhibits the phenomenon of resonant activation. We also study the Markovian limit [30] of the above problem by using a decoupling ansatz [31]. The Markovian approximation is constrained by the relative magnitude of the time scales of barrier fluctuations and vibrational degrees of freedom of the dimer. We also find that in appropriate limits, the vibrational degrees of freedom can be adiabatically eliminated. The paper is organized as follows. In the next section we study the dynamical properties of the non-Markovian system, followed by its Markovian limit in Sec. III. A discussion and summary are given in Sec. IV.

II. DYNAMICAL SYSTEM

Let us start with the equations of motion for the system of two coupled Brownian particles in a bistable potential U with a fluctuating barrier

$$\dot{x}_1 = -U'(x_1) + F_1\big(x_1,\eta_1^m\big) + F_{12}(x_1,x_2) + \eta_1^a(t), \tag{1a}$$

$$\dot{x}_2 = -U'(x_2) + F_2\big(x_2,\eta_2^m\big) + F_{21}(x_1,x_2) + \eta_2^a(t), \tag{1b}$$

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where η_i^a, $i = 1,2$, are Gaussian-white-noise processes with mean zero and correlations

$$\langle \eta_1^a(t)\eta_1^a(t')\rangle = \langle \eta_2^a(t')\eta_2^a(t)\rangle = 2D^a\delta(t-t'), \quad (2a)$$

$$\langle \eta_1^a(t)\eta_2^a(t')\rangle = \langle \eta_1^a(t')\eta_2^a(t)\rangle = 2D^a\rho^a\delta(t-t'), \quad (2b)$$

where D^a is the noise intensity and a measure of the dimensionless temperature of the associated heat bath and $\rho^a \in [-1,1]$ the correlation. The existence of such a correlation between the noise processes is natural as η_1^a and η_2^a have the same thermal origin. The potential $U(x) = -x^2/2 + x^4/4$ in Eq. (1) is a bistable potential with global minima at $x = \pm 1$ and a local maximum at $x = 0$, constituting the barrier separating the two wells. The fluctuations in the barrier are characterized by the fluctuating terms $F_i(x_i,\eta_i^m) = x_i\eta_i^m$, $i = 1,2$, where the noise processes η_i^m are colored Gaussian with mean zero and correlations,

$$\langle \eta_1^m(t)\eta_1^m(t')\rangle = \langle \eta_2^m(t)\eta_2^m(t')\rangle = (D^m/\tau)e^{-|t-t'|/\tau}, \quad (3a)$$

$$\langle \eta_1^m(t)\eta_2^m(t')\rangle = \langle \eta_1^m(t')\eta_2^m(t)\rangle = \rho^m(D^m/\tau)e^{-|t-t'|/\tau}, \quad (3b)$$

with τ the correlation time and D^m the intensity of barrier fluctuations in the limit of vanishing τ. For finite correlation times, the intensity of barrier fluctuations is $\sigma^2 = D^m/\tau$. In the above equations, $\rho^m \in [-1,1]$ is the correlation between the fluctuations of the potential barriers associated with the individual particles. In addition, we choose the barrier fluctuations to be independent of the thermal fluctuations. The two Brownian particles interact by the harmonic potential $U_{sh} = \frac{k}{2}(x_1 - x_2)^2$, with the corresponding forces $F_{ij}(x_1,x_2) = -\frac{\partial}{\partial x_i}U_{sh}(x_1,x_2)$, with $i \in \{1,2\}$ and $i \neq j$, and k being the spring constant. In Eqs. (1)–(3) the superscripts a and m correspond to additive and multiplicative, respectively.

In order to diagonalize the correlation matrices in Eqs. (2) and (3), let us transform the dynamical system to its center of mass $x_c = \frac{x_1+x_2}{2}$ and relative coordinates $x_r = \frac{x_1-x_2}{2}$, with the dynamical equations now reading

$$\dot{x}_c = f_c(x_c,x_r) + x_c\zeta_c^m(t) + x_r\zeta_r^m(t) + \zeta_c^a(t), \quad (4a)$$

$$\dot{x}_r = f_r(x_c,x_r) + x_c\zeta_r^m(t) + x_r\zeta_c^m(t) + \zeta_r^a(t), \quad (4b)$$

where $f_c(x_c,x_r) = -[U'(x_c + x_r) + U'(x_c - x_r)]/2$ and $f_r(x_c,x_r) = -[U'(x_c + x_r) - U'(x_c - x_r)]/2 - 2kx_r$. The additive noise processes in Eq. (4) are defined as $\zeta_c^a = \frac{\eta_1^a+\eta_2^a}{2}$ and $\zeta_r^a = \frac{\eta_1^a-\eta_2^a}{2}$ and are independent Gaussian-white-noise processes of mean zero and correlations

$$\langle \zeta_c^a(t)\zeta_c^a(t')\rangle = D^a(1+\rho^a)\delta(t-t'), \quad (5a)$$

$$\langle \zeta_r^a(t)\zeta_r^a(t')\rangle = D^a(1-\rho^a)\delta(t-t'). \quad (5b)$$

The multiplicative noise processes in Eq. (4) defined similarly to $\zeta_c^m = \frac{\eta_1^m+\eta_2^m}{2}$ and $\zeta_r^m = \frac{\eta_1^m-\eta_2^m}{2}$ are independent colored Gaussian noise processes with mean zero and correlations

$$\langle \zeta_c^m(t)\zeta_c^m(t')\rangle = (D^m/2\tau)(1+\rho^m)e^{-|t-t'|/\tau}, \quad (6a)$$

$$\langle \zeta_r^m(t)\zeta_r^m(t')\rangle = (D^m/2\tau)(1-\rho^m)e^{-|t-t'|/\tau}. \quad (6b)$$

Equations (4)–(6) define the dynamical system of two harmonically coupled Brownian particles in a bistable potential with a fluctuating barrier in contact with a heat bath in terms of the center of mass x_c and relative coordinates x_r. The advantage of transforming Eqs. (1)–(3) is the independence of the transformed additive and multiplicative noise processes. However, the transformed equations in terms of x_c and x_r are still non-Markovian due to the presence of colored noise sources ζ_c^m and ζ_r^m. In order to learn about the dynamical properties of the system under study, we numerically solve Eqs. (4)–(6) using the concept of Markovian embedding [5]. All the numerical calculations are performed using Heun's method [32] in the Stratonovich interpretation [33], with the initial conditions $(x_c,x_r) = (-1.0,0.02)$. In order to characterize the escape properties of the coupled Brownian particles across the fluctuating barrier, we calculate the escape times of the center of mass x_c to the other minima at $x_c = 1.0$. We also choose for simplicity the intensity of thermal fluctuations to be fixed at $D^a = 0.1$ and independent of each other, i.e., $\rho^a = 0$ in what follows, unless explicitly stated.

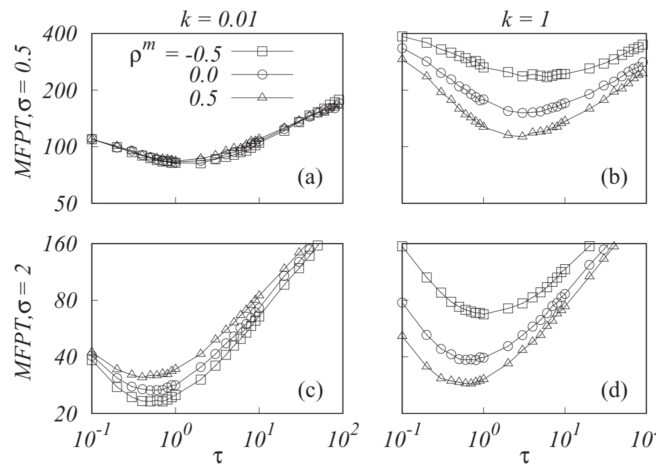

FIG. 1. Variation of the MFPT with correlation time τ of the barrier fluctuations. The nonmonotonic dependence of the MFPT on τ is evident from the figure and occurs for $\tau \approx 1$, which is the relaxation time in the bistable potential U. It should be noted that both axes are represented in logarithmic scale. The intensity of the barrier fluctuations $\sigma^2 = D^m/\tau$. The MFPT is calculated over 10 000 ensembles.

Figure 1 shows the dependence of the mean first-passage time (MFPT) on the correlation time τ of the barrier fluctuations. The nonmonotonic dependence of the MFPT on τ implies that the escape of the coupled Brownian particles across a fluctuating barrier exhibits the phenomenon of resonant activation. The existence of such a phenomenon for a system of two coupled Brownian particles results as an outcome of two competing events: (i) a decrease in the MFPT with increasing D^m for a fixed τ, which is seen, for example, when we compare the MFPT for different values of the intensity of fluctuation σ^2 for $\tau = 1$ [Figs. 1(a) and 1(c)], and (ii) an increase in the MFPT with increasing τ for a fixed D^m, which is evident from Eq. (6), as the intensity of barrier fluctuations decreases with increasing τ for fixed D^m, making the escape relatively difficult compared to low τ. The observed properties are similar to the observations reported for a single particle in a fluctuating bistable potential, with

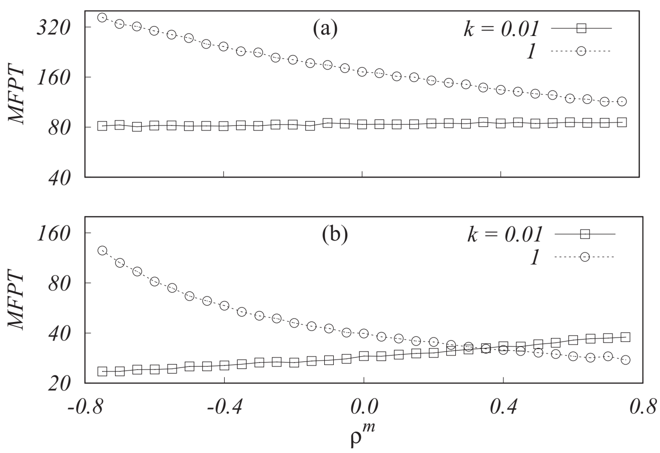

FIG. 2. Variation of the MFPT with noise correlation ρ^m for a fixed correlation time $\tau = 1$ for (a) $\sigma = 0.5$ and (b) $\sigma = 2$. It is shown in (b) that for a higher intensity of barrier fluctuations, the MFPT is lower when the coupling between the particles is large, e.g., $k = 1$ as compared to nearly independent particles, $k = 0.01$, for strongly correlated barrier fluctuations. The y axis is shown in logarithmic scale and the MFPT is calculated over 10 000 ensembles.

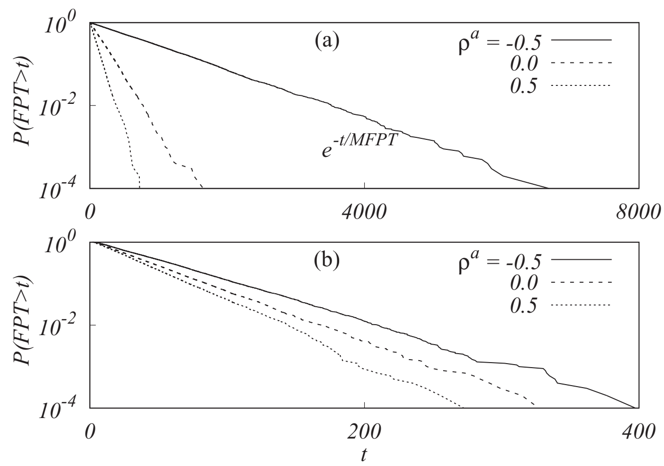

FIG. 3. Distribution of FPTs of the center of mass starting at $x_c = -1$ and reaching $x_c = 1$, for fixed temperature $D^a = 0.1$, correlation time $\tau = 1$, and independent barrier fluctuations $\rho^m = 0$ for spring constant $k = 1$ for (a) $\sigma = 0.5$ and (b) $\sigma = 2$. The FPT distributions are calculated using 10 000 data points.

the minima of the MFPT occurring at $\tau \approx 1$, which is the relaxation time of the center of mass in the bistable potential $U(x) = -x^2/2 + x^4/4$ [14–16]. Also evident from Fig. 1 is the effect of coupling between the two particles, i.e., large k leads to a lower rate of escape from one potential minimum to the other. In addition, the noise correlation ρ^m can have contrasting effects depending on the value of coupling constant k and the intensity of barrier fluctuations $\sigma^2 = D^m/\tau$, e.g., Figs. 1(c) and 1(d) for a fixed correlation time τ.

To better understand the effect of correlation ρ^m between the barrier fluctuations on the escape properties of the coupled particle system, we report in Fig. 2 the variation of the MFPT with ρ^m for different values of spring constant k and the intensity of barrier fluctuations σ^2, for a fixed correlation time $\tau = 1$. It is observed that the MFPT of the center of mass from $x_c = -1$ to $x_c = 1$ decreases monotonically with ρ^m for a strong coupling between the particles, e.g., $k = 1$, independently of the intensity of barrier fluctuations σ^2. On the other hand, when the harmonic coupling between the particles is weak, e.g., $k = 0.01$, the MFPT shows a tendency to monotonically increase with noise correlation ρ^m, which is particularly evident for strong fluctuations in the barrier, e.g., $\sigma = 2$. To understand the reason for such contrasting behaviors between limits of very strong to very weak coupling, let us first focus on the limit of strong coupling, i.e., large k. It is known that for a very strong coupling between two particles, the coupled-particle system behaves effectively as a single particle placed at the center of mass of the system. As a result the decrease in the MFPT with increasing ρ^m is expected in view of Eq. (6), which makes the multiplicative fluctuations in the center-of-mass motion (4a) relatively strong for a larger correlation for a fixed value of σ. As the magnitude of the fluctuations is linearly dependent on the value correlation ρ^m, the decrease in the MFPT with increasing ρ^m is expected in the light of single-particle dynamics in the strong-coupling limit. On the other hand, in the limit of weak coupling, $F_{ij}(x_1,x_2) \approx 0$ in Eq. (1) and the two particles are coupled only by the correlation ρ^m [34] between the barrier fluctuations as the associated heat baths are independent, $\rho^a = 0$. Now, for $\rho^m > 0$, the barriers associated with the two particles move up and down simultaneously, making the escape of the center of mass to $x_c = 1$ difficult because both particles should be present in the other well at the same time. In contrast, for $\rho^m < 0$, the barriers associated with the two particles fluctuate opposite to each other, making the barrier crossing for one of the particles easier relative to the other, hence making the escape of the center of mass to the absorbing boundary at $x_c = 1$ easier as compared to when $\rho^m > 0$. The above results imply that the presence of an additional vibrational degree of freedom leads to novel features in the escape dynamics across a fluctuating barrier. For example, compared with the case of independent barrier fluctuations, the correlated barrier fluctuations can result in a faster or slower escape rate depending on the coupling between the particles; i.e., for weak coupling positive correlations result in a decreased rate of escape of the center of mass across the fluctuating barrier, whereas for strong coupling the positive correlations enhance the same.

Let us now study the effects of correlation between the thermal baths, i.e., $\rho^a \neq 0$, on the transient properties of the coupled particle system, for fixed temperature $D^a = 0.1$. In Fig. 3 we report the distribution of first-passage times (FPTs) for the center of mass starting at $x_c = 1$ and reaching $x_c = -1$. It is observed that the distribution of FPTs has an exponentially decaying tail with the MFPT as the parameter. The dependence of the MFPT on the correlation between the heat baths associated with the two particles, i.e., $\rho^a \neq 0$, implies that the rate of escape of the coupled Brownian particles across the fluctuating barrier can be altered by varying the degree of correlation ρ^a, even when the temperature D^a of the heat bath is kept constant. This is because the presence of correlation distributes the power received from the heat bath between the translational and vibrational degrees of freedom, depending on the strength of correlation. For negatively correlated heat baths, the fluctuations in the vibrational degrees of freedom are enhanced at the cost of reducing the fluctuations associated with the translational degrees of freedom, thereby making the diffusion of the center of mass slower and hence making escape difficult, whereas for $\rho^a > 0$, the magnitude of the corresponding fluctuations are reversed. Also evident from Fig. 3 is the increase in the rate of escape with the increase in the intensity of barrier fluctuations σ^2 for a given correlation ρ^a at fixed temperature D^a of the associated heat baths.

The above results have focused on the escape properties of a system of coupled Brownian particles across a fluctuating barrier with finite rate of fluctuation $1/\tau$ for a fixed intensity $\sigma^2 = D^m/\tau$. For the purpose of completeness, it is interesting to cover the extreme limits of very slow ($\tau \to \infty$) and very fast ($\tau \to 0$) barrier fluctuations for a fixed D^m. First, let us consider the case of quasistatically fluctuating barriers. In the extreme limit of a vanishingly small rate, i.e., $1/\tau \to 0$, the barrier fluctuations are slower than all the time scales of the system, resulting in a nearly static bistable potential in contact with a heat bath [28]. On the other hand, for very fast barrier fluctuations, with $1/\tau \to \infty$, the Markovian limit of the problem is recovered for a fixed D^m. Such a limiting case is particularly interesting when the coupling k between the two Brownian particles is large. In this limit, the fast time scales of the relative fluctuations x_r and correlation times τ become comparable and put a natural constraint on the Markovian limit of the problem. This is because a reduction of the Markovian limit via elimination of the time scale of barrier fluctuations is possible only when the barriers fluctuate on the fastest time scale among all the relevant time scales of the system. In the next section, employing the decoupling ansatz proposed by Hänggi [31], we derive the Markovian limit of the above problem in a self-consistent approximation [35] along the lines of an approximate Fokker-Planck equation [36]. We also discuss the appropriate limits of the original non-Markovian problem (4)–(6) where further simplifications can be achieved by adiabatically eliminating [37] the fast vibrational degrees of freedom when coupling between the two particles is very strong.

III. MARKOVIAN APPROXIMATION AND ADIABATIC ELIMINATION

Given the random dynamical system defined by Eqs. (4)–(6), the equation of motion for the probability density $p = p(x_c,x_r,t)$ is given by

$$\frac{\partial}{\partial t}p = -\sum_{i\in\{c,r\}}\frac{\partial}{\partial x_i}f_i p - \sum_{i\in\{c,r\}}\frac{\partial}{\partial x_i}\left\langle\zeta_i^a(t)\delta(\mathbf{x}(t)-\mathbf{x})\right\rangle - \sum_{i\in\{c,r\}}\frac{\partial}{\partial x_c}x_i\left\langle\zeta_i^m(t)\delta(\mathbf{x}(t)-\mathbf{x})\right\rangle - \sum_{i\neq j\in\{c,r\}}\frac{\partial}{\partial x_r}x_i\left\langle\zeta_j^m(t)\delta(\mathbf{x}(t)-\mathbf{x})\right\rangle, \tag{7}$$

where, according to van Kampen's lemma [38], $p(x_c,x_r,t) = \langle\delta(\mathbf{x}(t)-\mathbf{x})\rangle$, the averaging being done over the noise processes. The averages in Eq. (7) can be evaluated according to Novikov's theorem for Gaussian fluctuations [39]. Application of Hänggi's decoupling ansatz [31] leads to an approximate Fokker-Planck equation in the Stratonovich sense

$$\frac{\partial}{\partial t}p = -\sum_{i\in\{c,r\}}\frac{\partial}{\partial x_i}f_i p + \sum_{i,j\in\{c,r\}}f_{(ij)}^{ij}\frac{\partial}{\partial x_{(i}}x_i\frac{\partial}{\partial x_{j)}}x_j p + \sum_{i\neq j\in\{c,r\}}f_{jj}^{ii}\frac{\partial}{\partial x_j}x_i\frac{\partial}{\partial x_j}x_i p + \sum_{i\in\{c,r\}}f^i\frac{\partial^2}{\partial x_i^2}p, \tag{8}$$

where $(ij) = (\{ij\}+\{ji\})/2$, with the coefficients

$$\begin{aligned}
f^c &= D^a(1+\rho^a)/2, \quad f^r = D^a(1-\rho^a)/2,\\
f_{rr}^{cc} &= D^m(1-\rho^m)/2[1+2\tau(1+k)],\\
f_{cc}^{rr} &= D^m(1-\rho^m)/2(1-2k\tau),\\
f_{cc}^{cc} &= D^m(1+\rho^m)/2(1+2\tau), \quad f_{rr}^{rr} = D^m(1+\rho^m)/2,\\
f_{cr}^{cr} &= 2f_{rr}^{rr}, \quad f_{cr}^{rc} = 2f_{rr}^{cc}, \quad f_{rc}^{cr} = 2f_{cc}^{rr}, \quad f_{rc}^{rc} = 2f_{cc}^{cc}.
\end{aligned}$$

The crucial step in the above derivation is decoupling of the functional derivatives and the probability density as

$$\left\langle\delta(\mathbf{x}(t)-\mathbf{x})\frac{\delta}{\delta\zeta_r^m(t')}x_c(t)\right\rangle \approx \langle\delta(\mathbf{x}(t)-\mathbf{x})\rangle\left\langle\frac{\delta}{\delta\zeta_r^m(t')}x_c(t)\right\rangle, \tag{9}$$

from which the averages in Eq. (7) are calculated self-consistently [35] by taking into account the stable steady state of the noise-free dynamical system corresponding to Eq. (4). The steps involved in the derivation follow [36] and are outlined in the Appendix. It should be noted that the decoupling ansatz is valid in the limit of relatively small magnitudes of barrier fluctuations and has been extended to moderate to strong noise intensities in a unified colored noise approximation, for both additive [40] and multiplicative [41] perturbations. The approximate theory has been in excellent agreement in predicting the steady-state solution of the non-Markovian problem. The decoupling ansatz has been justified by Fox [42,43] using functional calculus, wherein the uniform convergence to an effective Fokker-Planck equation for weakly colored noise has been shown for both additive and multiplicative perturbations. However, results based on path integrals [44] have shown that the smallness of the correlation time τ does not lead to an effective Markovian approximation. Notwithstanding such differences, explicit numerical calculations for a symmetric bistable potential perturbed by a multiplicative colored noise [36] verify the applicability of the decoupling ansatz [31] in predicting the steady states, particularly for the case of a bistable potential with the barrier fluctuating by exponentially correlated Gaussian noise. This justifies our use of the decoupling ansatz in deriving the Markovian approximation of the non-Markovian problem defined in Eqs. (4)–(6).

It is also noted from Eq. (8) that the approximate Fokker-Planck equation is constrained by the inequality $2k\tau < 1$. The physical understanding behind the existence of such an inequality is that the time scale of barrier fluctuations is required to be faster than the time scale of vibrational motion of the system of coupled Brownian particles, in order to be eliminated. For $\tau = 0$, the above equation reduces to an equivalent Markovian problem for two coupled Brownian particles in which the exponential time dependence in Eq. (6) is replaced by a δ function. For $\tau > 0$ but small compared to the relaxation time in the bistable potential, it is evident that the presence of finite τ modifies the effective magnitude of barrier fluctuations when compared to the equivalent Markovian problem. In addition, in light of the discussion in the preceding paragraph, the use of the approximate Fokker-Planck equation (8) is limited to the calculation of steady-state properties of the non-Markovian problem in the limit of weak noise color.

Let us now discuss a few cases of interest for the system of two coupled Brownian particles. In the limit when the barrier fluctuations associated with the two particles are very strongly correlated, i.e., $\rho^m \approx 1$, the multiplicative fluctuations associated with the relative coordinate x_r become insignificant in the dynamics of the center-of-mass motion x_c and vice versa, as can be observed from Eq. (4). This results in a considerable amount of simplification in the corresponding Fokker-Planck equation (8) wherein the third summation term vanishes in the limit $\rho^m \to 1$. As a result, f_{cr}^{rc} and f_{rc}^{cr} also vanish. Consequently, in the limit of strong positive correlations between ζ_c^m and ζ_r^m, the competition between the time scales of barrier fluctuations and the relaxation of the relative coordinates is also eliminated. The situation can be understood physically as follows: When the barrier fluctuations associated with the two Brownian particles are strongly correlated, the two particles almost always find a similar environment, independent of the rate of fluctuations of the barrier. As a result, the rate of relaxation of the relative coordinate x_r, which occurs at a time scale determined by the spring constant k, becomes uncoupled from the rate of barrier fluctuations. It is noted that this is not true for any arbitrary value of correlation ρ^m and was also seen in the derivation of the approximate Fokker-Planck equation (8), where the Markovian limit of the non-Markovian problem was derived under the constraint that the barriers fluctuate on the fastest time scale among the relevant time scales of the dynamical system (4)–(6).

However, the limit of strongly correlated barrier fluctuations allows us to adiabatically eliminate the fast degree of freedom, the relative coordinate x_r, in the limit of large spring constant k from the original non-Markovian problem. When the coupling between the particles is large, the two particles move very close to each other, $x_1 \approx x_2$, resulting in $f_r(x_c,x_r) \approx -2kx_r$, leading to

$$\begin{aligned}\dot{x}_r &\approx -2kx_r + x_r\zeta_c^m(t) + \zeta_r^a(t)\\ &= \left[-2k + \zeta_c^m(t)\right]x_r + \zeta_r^a(t).\end{aligned} \tag{10}$$

Now the intensity of barrier fluctuations is $\sigma^2 = D^m/\tau$; hence, in the limit of very strong coupling between the particles, the spring constant k can be chosen large enough to ignore the fluctuations due to $\zeta_c^m(t)$ without incurring much error. Consequently, in the limit of strongly correlated barrier fluctuations and large coupling between the particles, the dynamical equations (4)–(6) reduce to

$$\dot{x}_c \approx f_c(x_c,x_r) + x_c\zeta_c^m(t) + \zeta_c^a(t), \tag{11a}$$

$$\dot{x}_r \approx f_r(x_c,x_r) + \zeta_r^a(t), \tag{11b}$$

where

$$f_c(x_c,x_r) = -[U'(x_c + x_r) + U'(x_c - x_r)]/2,$$

$$f_r(x_c,x_r) \approx -2kx_r,$$

and the correlations $\langle\zeta_c^m(t)\zeta_c^m(t')\rangle = (D^m/\tau)e^{-|t-t'|/\tau}$, with the additive fluctuations following Eq. (5). The approximate Fokker-Planck equation associated with (11) is

$$\frac{\partial}{\partial t}p(x_c,x_r,t) = \left(L_{\mathrm{FP}}^c + L_{\mathrm{FP}}^r\right)p(x_c,x_r,t), \tag{12}$$

where

$$L_{\mathrm{FP}}^c = -\frac{\partial}{\partial x_c}f_c + \frac{D^m}{1+2\tau}\frac{\partial}{\partial x_c}x_c\frac{\partial}{\partial x_c}x_c + \frac{D^a(1+\rho^a)}{2}\frac{\partial^2}{\partial x_c^2},$$

$$L_{\mathrm{FP}}^r = -\frac{\partial}{\partial x_r}f_r + \frac{D^a(1-\rho^a)}{2}\frac{\partial^2}{\partial x_r^2}$$

are the Fokker-Planck operators associated with the center-of-mass (slow) and relative (fast) coordinates, respectively. Now the adiabatic elimination of the fast variable x_r requires marginalizing the probability density $p(x_c,x_r,t)$ with the stationary solution of the corresponding Fokker-Planck operator L_{FP}^r. The operator L_{FP}^r admits the Gaussian distribution of mean zero and variance $\langle x_r^2\rangle = \frac{D^a(1-\rho^a)}{4k}$ as its stationary solution $\psi_0(x_r)$. The only x_r dependence entering the dynamics of the slow center-of-mass motion is through the drift term $f_c(x_c,x_r) = x_c - x_c^3 - 3x_cx_r^2$. As a result, the drift term associated with the center-of-mass motion is modified to

$$V'(x_c) = \int dx_r\, f_c\psi_0 = x_c\left(1 - 3\langle x_r^2\rangle\right) - x_c^3. \tag{13}$$

This implies that the presence of an additional vibrational degree of freedom results in an increased barrier height on an average. This explains why the escape across the fluctuating barrier becomes difficult for the system of coupled Brownian particles when the coupling k between the particles is strong and the associated potential barriers fluctuate in a strongly correlated manner but with a low intensity σ^2 [see, e.g., Fig. 2(a)]. However, the modification is negligible due to large coupling k between the particles and it can be said without much error that in the strong-coupling limit, when the barriers associated with x_1 and x_2 fluctuate in a strongly correlated manner, the dynamics of the slow center-of-mass variable follows a single particle in the bistable potential $U(x)$ with the barrier fluctuating with Gaussian white noise. The equation of motion of such a particle is

$$\dot{x} = x - x^3 + x\zeta^m(t) + \zeta^a(t), \tag{14}$$

with ζ^m and ζ^a being Gaussian-white-noise processes of mean zero and correlations

$$\langle \zeta^m(t)\zeta^m(t')\rangle = \frac{2D^m}{1+2\tau}\delta(t-t'), \tag{15a}$$

$$\langle \zeta^a(t)\zeta^a(t')\rangle = D^a(1+\rho^a)\delta(t-t'). \tag{15b}$$

The transient and steady-state properties of a particle following Eq. (14) are well understood [20–25] and hence we do not repeat the calculations. However, it is worth noticing that the dynamics of a Brownian particle following Eq. (14) may not be directly identified with the Markovian limit of the corresponding colored multiplicative noise problem [36]. This is because Eq. (14) was derived by first taking the $\tau \to 0$ limit of the approximate Langevin equations (11) followed by the adiabatic elimination of the fast variable x_r and the correspondence would require the equation of motion for the probability density $p(x_c,x_r,t)$ for any arbitrary value of correlation τ.

IV. CONCLUSION

We have looked at the escape properties of two harmonically coupled Brownian particles across the fluctuating barrier of a bistable potential. It was found that the escape of the center of mass of the two particles exhibits the phenomenon of resonant activation, i.e., the nonmonotonic variation of the escape times with the rate of fluctuations of the potential barrier, with the minima of the MFPT occurring when the correlation time τ is comparable to the relaxation time in the bistable potential. Coupling between the particles generally tends to diminish the escape rates for weakly fluctuating barriers. It is also of interest to know the extreme limits when barriers fluctuate on time scales that are very slow and very fast when compared to the relaxation times in the bistable potential. In the limit of very slow fluctuations, $1/\tau \to 0$ and the barriers become nearly static. On the other hand, for very fast time scales of barrier fluctuations, when $1/\tau \to \infty$, the problem approaches an equivalent Markovian limit. This is particularly interesting for a finite coupling between the particles, due to competing time scales in the $k \to \infty$ and $\tau \to 0$ limits. We derive the Markovian limit of the problem under the constraint that the barriers fluctuate on the fastest of the relevant time scales of the system. For strongly correlated barrier fluctuations, the competition between $1/\tau$ and k can be eliminated, which is used to study the adiabatic limit of the problem. The present study has implications in noise-driven transport, particularly in biological systems, e.g., motor proteins like kinesin, and can also be generalized to polymers, which are of general interest. These results, however, are based on the special case in which the parametric fluctuations arise independently of the thermal fluctuations; nontrivial features could be expected to emerge when the barriers fluctuate anticorrelated to thermal fluctuations due to their competing effects.

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APPENDIX: APPROXIMATE FOKKER-PLANCK EQUATION

Novikov's theorem [39] allows the calculation of the averages involved in Eq. (7) as

$$\begin{aligned}
&\big\langle \zeta_r^m(t)\delta(\mathbf{x}(t)-\mathbf{x})\big\rangle \\
&= \int_0^t dt'\big\langle \zeta_r^m(t)\zeta_r^m(t')\big\rangle\bigg\langle \frac{\delta}{\delta\zeta_r^m(t')}\delta(\mathbf{x}(t)-\mathbf{x})\bigg\rangle \\
&= -\frac{\partial}{\partial x_c}\int_0^t dt'\big\langle \zeta_r^m(t)\zeta_r^m(t')\big\rangle\bigg\langle \delta(\mathbf{x}(t)-\mathbf{x})\frac{\delta}{\delta\zeta_r^m(t')}x_c(t)\bigg\rangle \\
&\quad -\frac{\partial}{\partial x_r}\int_0^t dt'\big\langle \zeta_r^m(t)\zeta_r^m(t')\big\rangle\bigg\langle \delta(\mathbf{x}(t)-\mathbf{x})\frac{\delta}{\delta\zeta_r^m(t')}x_r(t)\bigg\rangle.
\end{aligned} \tag{A1}$$

Similar expressions can be written for other averages in Eq. (7). However, all such averages involve functional derivatives of the sort $\delta x_c(t)/\delta\zeta_r^m(t')$ in order to evaluate the integrals like that in Eq. (A1).

Now, from Eq. (4a) we have

$$x_c(t) = x_c(0) + \int_0^t ds\big(f_c + x_c\zeta_c^m + x_r\zeta_r^m + \zeta_c^a\big), \tag{A2}$$

which leads to

$$\begin{aligned}
\frac{\delta x_c(t)}{\delta\zeta_r^m(t')} = x_c(t') + \int_{t'}^t ds\bigg(&\frac{\partial f_c}{\partial x_c}\frac{\delta x_c(s)}{\zeta_r^m(t')} + \frac{\partial f_c}{\partial x_r}\frac{\delta x_r(s)}{\zeta_r^m(t')} \\
&+ \zeta_c^m(s)\frac{\delta x_c(s)}{\zeta_r^m(t')} + \zeta_r^m(s)\frac{\delta x_r(s)}{\zeta_r^m(t')}\bigg),
\end{aligned} \tag{A3}$$

with the initial condition $\frac{\delta x_c(t)}{\delta\zeta_r^m(t')}|_{t'=t} = x_r(t')$. Also from Eq. (A3),

$$\begin{aligned}
\frac{\partial}{\partial t}\frac{\delta x_c(t)}{\delta\zeta_r^m(t')} = &\frac{\partial f_c}{\partial x_c}\frac{\delta x_c(t)}{\zeta_r^m(t')} + \frac{\partial f_c}{\partial x_r}\frac{\delta x_r(t)}{\zeta_r^m(t')} \\
&+ \zeta_c^m(t)\frac{\delta x_c(t)}{\zeta_r^m(t')} + \zeta_r^m(t)\frac{\delta x_r(t)}{\zeta_r^m(t')}.
\end{aligned} \tag{A4}$$

Proceeding along similar lines, the matrix differential equation for the functional derivatives can be written as

$$\frac{\partial}{\partial t}\frac{\delta}{\delta\boldsymbol{\zeta}(t')}\mathbf{X}(t) = \mathbf{F}[\mathbf{X}]\frac{\delta}{\delta\boldsymbol{\zeta}(t')}\mathbf{X}(t), \tag{A5}$$

where

$$\mathbf{F}[\mathbf{X}] = \begin{pmatrix} \mathbf{A} & \mathbf{B} \\ \mathbf{C} & \mathbf{D} \end{pmatrix}_{8\times 8}$$

and

$$\mathbf{A} = \big(\partial f_c/\partial x_c + \zeta_c^m\big)\mathbf{I}_4, \quad \mathbf{B} = \big(\partial f_c/\partial x_r + \zeta_r^m\big)\mathbf{I}_4,$$

$$\mathbf{C} = \big(\partial f_r/\partial x_c + \zeta_r^m\big)\mathbf{I}_4, \quad \mathbf{D} = \big(\partial f_r/\partial x_r + \zeta_c^m\big)\mathbf{I}_4,$$

with $\mathbf{I}_4$ being the 4×4 identity matrix. In addition,

$$\frac{\delta \mathbf{X}(t)}{\delta \boldsymbol{\zeta}(t')} = \left(\frac{\delta x_c(t)}{\delta \zeta_c^m(t')}, \frac{\delta x_c(t)}{\delta \zeta_r^m(t')}, \frac{\delta x_c(t)}{\delta \zeta_c^a(t')}, \frac{\delta x_c(t)}{\delta \zeta_r^a(t')}, \frac{\delta x_r(t)}{\delta \zeta_c^m(t')}, \frac{\delta x_r(t)}{\delta \zeta_r^m(t')}, \frac{\delta x_r(t)}{\delta \zeta_c^a(t')}, \frac{\delta x_r(t)}{\delta \zeta_r^a(t')} \right).$$

The initial condition associated with the matrix differential equation (A5) is

$$\left.\frac{\delta \mathbf{X}(t)}{\delta \boldsymbol{\zeta}(t')}\right|_{t'=t} = [x_c(t'), x_r(t'), 1, 0, x_r(t'), x_c(t'), 0, 1].$$

Now Eq. (A5) admits the solution

$$\frac{\delta}{\delta \boldsymbol{\zeta}(t')} \mathbf{X}(t) = \exp\left(\int_{t'}^{t} ds\, \mathbf{F}[\mathbf{X}(s)] \right) \left.\frac{\delta}{\delta \boldsymbol{\zeta}(t')} \mathbf{X}(t)\right|_{t'=t}. \tag{A6}$$

Evaluation of the right-hand side of (A6) requires the initial values of the functional derivatives and the calculation of the matrix $\mathbf{F}$. To achieve this, we observe that

$$x_c(t') = x_c(t) \exp\left(-\int_{t'}^{t} ds \frac{\dot{x}_c(s)}{x_c(s)} \right),$$

$$x_r(t') = x_r(t) \exp\left(-\int_{t'}^{t} ds \frac{\dot{x}_r(s)}{x_r(s)} \right). \tag{A7}$$

These integrals can be solved by invoking Hänggi's decoupling ansatz [31] according to which the long-time dynamics of a random dynamical system is determined according the stable fixed points of the corresponding deterministic dynamical system. In the absence of fluctuations, Eq. (4) admits $(x_c, x_r) = (\pm 1, 0)$ as the stable fixed points. In addition, we also ignore the random fluctuations in Eq. (A6) and the initial conditions as a self-consistent approximation [35,36], resulting in

$$\mathbf{F}[\mathbf{X}]_{(x_c,x_r)=(\pm 1,0)} = \begin{pmatrix} -2\mathbf{I}_4 & \mathbf{0} \\ \mathbf{0} & -2(1+k)\mathbf{I}_4 \end{pmatrix}_{8\times 8}, \tag{A8}$$

with $\lim_{(x_c,x_r)\to(\pm 1,0)} \frac{\dot{x}_c}{x_c} = 0$ and $\lim_{(x_c,x_r)\to(\pm 1,0)} \frac{\dot{x}_r}{x_r} = -2(1+k)$. This results in the initial conditions $x_c(t') \approx x_c(t)$ and $x_r(t') \approx x_r(t) \exp[2(1+k)(t-t')]$. Substituting the values of $\mathbf{F}$, $x_c(t')$, and $x_r(t')$ in Eq. (A6) leads to the values of the functional derivatives

$$\frac{\delta \mathbf{X}(t)}{\delta \boldsymbol{\zeta}(t')} \approx [x_c(t)e^{-2(t-t')}, x_r(t)e^{2k(t-t')}, e^{-2(t-t')}, 0, x_r(t), x_c(t)e^{-2(1+k)(t-t')}, 0, e^{-2(1+k)(t-t')}].$$

It is interesting to note that

$$\frac{\delta x_c(t)}{\delta \zeta_r^m(t')} = x_r(t)e^{2k(t-t')}, \tag{A9}$$

which arises due to the presence of ζ_r^m in Eq. (4a) and plays a decisive role in the evaluation of the first term in Eq. (A1) because of its exponentially growing character. To see this explicitly, let us calculate the first term in Eq. (A1),

$$\begin{aligned}
&\int_0^t dt' \langle \zeta_r^m(t)\zeta_r^m(t')\rangle \left\langle \delta(\mathbf{x}(t)-\mathbf{x}) \frac{\delta}{\delta \zeta_r^m(t')} x_c(t) \right\rangle \\
&= \int_0^t dt' \frac{D^m(1-\rho^m)}{2\tau} e^{-(t-t')/\tau} \langle \delta(\mathbf{x}(t)-\mathbf{x}) x_r(t) e^{2k(t-t')} \rangle \\
&= \frac{D^m(1-\rho^m)}{2\tau} x_r \int_0^t dt' e^{-(t-t')/\tau} e^{2k(t-t')} p \\
&= \frac{D^m(1-\rho^m)}{2(1-2k\tau)} x_r p,
\end{aligned} \tag{A10}$$

where we have used the decoupling ansatz in the second equality and the last step is evaluated under the constraint that $1/\tau - 2k > 0$ and approaches the final value in the long-time limit. Physically, this means that the rate of fluctuation of the potential barrier is rapid as compared to the time scale at which the relative vibration of the two coupled Brownian particles relaxes towards the origin. Evaluating the other integrals similarly leads to the steady-state Fokker-Planck equation (8).

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