Whole fusion-fission process with Langevin approach and compared with analytical solution for barrier passage

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Abstract

We investigate time-dependent probability for a Brownian particle passing over the barrier to stay at a metastable potential pocket against escaping over the barrier. This is related to whole fusion-fission dynamical process and can be called the reverse Kramers problem. By the passing probability over the saddle point of inverse harmonic potential multiplying the exponential decay factor of a particle in the metastable potential, we present an approximate expression for the modified passing probability over the barrier, in which the effect of reflection boundary of potential is taken into account. Our analytical result and Langevin Monte-Carlo simulation show that the probability passing and against escaping over the barrier is a non-monotonous function of time and its maximal value is less than the stationary result of passing probability over the saddle point of inverse harmonic potential.

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I. INTRODUCTION

The metastable system decay can be applied widely to describe various science problems such as chemical reaction kinetics, phase transient, nuclear fission, and so on. The well-known Kramers problem is such a process that a Brownian particle subjected to thermal fluctuation escapes from the barrier of a metastable potential. As early as 1940, Kramers published his seminal paper “Brownian motion in force fields and chemical reaction diffusion model” [1], in which he proposed a formula for the reaction rate constant for a general-damped particle escaping from a metastable potential well and used this model to explain the mechanism of excited nuclear fission. Abe is the first researcher who used Langevin Monte-Carlo simulation to calculate numerically the nuclear fission rate [2]. In 1990, Hänggi et. al. [3] summarized the works fifty years after Kramers, including various improvements and extensions for the Kramers rate theory.

Now a reverse problem appears timely, i.e., a Brownian particle with initial velocity passes over the saddle point to enter into the well of metastable potential and escapes from the saddle point finally. In fact, molecular collision, atom cluster and heavy-ion fusion are such barrier passage problems [4–12]. In the pervious works, the fusion probability was obtained by the passing probability of a Brownian particle over the top of an inverse harmonic potential [13, 14], the latter has been generalized to include effects of quantum fluctuation [15], initial distribution [16], anomalous diffusion [17] as well as colored noise [18].

As one knows that the fusion probability has been estimated by the stationary value of time-dependent passing probability in terms of the fusion by diffusion model [14], it has a simple form of error function. There are no need for considering the shell correction of potential energy and neutron emission in the fusion phase. Actually, the transient process is very important for the asymptotical passing probability regarding as the fusion probability. The inverse harmonic potential approximation is suitable only for the near barrier fusion and high fission barrier cases. In this case, the fission life or the mean first passage time from the ground state to the barrier is much longer than the transient time of passing probability over the saddle point; however, the super-heavy element cases should be much carefully, because the component inside the barrier of time-dependent spatial distribution function (SDF) decays quickly and opposes to the process of passing-over barrier. Therefore,
it is necessary to consider the influence of metastable potential structure upon the passing probability over the barrier. Of course, competition between neutron emission and fission decay needs to be investigated, the former decreases the temperature of compound nucleus, but which occurs in the survival-evaporation phase. At present, we focus on time-dependent dynamical fusion probability modified by the effect of reflection boundary of metastable potential.

The paper is organized as follows. In Sec. II we describe the barrier passage dynamics and propose an approximate expression for the probability passing and against escaping over the barrier of metastable potential. In this section, we also analyze the error for the stationary passing probability over the saddle point of inverse harmonic potential regarded as the fusion probability. Finally, concluding remarks are given in Sec. III.

II. THE MODIFIED BARRIER PASSING PROBABILITY AND FUSION-FISSION DYNAMICS

The dynamics of a Brownian particle of mass \( m \) subjected to a fluctuation force \( \xi(t) \) in a potential \( U(x) \) is described by the following Langevin equation:

\[
m\ddot{x}(t) + \gamma \dot{x}(t) + U'(x) = \xi(t),
\]

where \( \xi(t) \) is the Gaussian white noise satisfying \( \langle \xi(t) \rangle = 0 \) and \( \langle \xi(t)\xi(t') \rangle = 2\gamma k_B T \delta(t-t') \), \( k_B \) is the Boltzmann constant, \( T \) is the temperature and \( \gamma \) is the damping coefficient. In order to present an approximate expression for time-dependent passing probability and against escaping over the barrier in a metastable potential, we consider an inverse harmonic potential linking smoothly with a harmonic potential, linking point of two potentials is determined by

\[
U_g(x_c) = U_s(x_c) \quad \text{and} \quad U'_g(x_c) = U'_s(x_c),
\]

where \( x_g \) denotes the coordinate of the ground state, \( \omega_g \) and \( \omega_s \) are the circular frequencies of potential at the ground state and the saddle point, respectively, the linking point of two potentials is determined by \( x_c = x_g \omega_g^2 / (\omega_g^2 + \omega_s^2) \) through \( U_g(x_c) = U_s(x_c) \) and \( U'_g(x_c) = U'_s(x_c) \), \( U_b \) is the barrier height given by \( U_b = \frac{1}{2} \omega_s^2 x_c x_g \). In the calculations, all the parameters are chosen to be dimensionless forms and \( m = k_B = 1.0 \). By the way we choose \( x_s = 0 \) to be the coordinate of saddle point.
FIG. 1: (color online). Time evolution of SDF of a particle. The black-solid and red-triangle lines are the SDFs of particle in the inverse harmonic and metastable potentials, respectively. Each inset shows the potential with dots representing the positions where the peak of the distributions locate. The parameters used are: $T = 0.4$, $\gamma = 1.0$, $U_b = 1.0$, $\bar{v}_0 = -5.0$ and $\bar{x}_0 = 0.2$. Note that each subgraph has a different scale.
Firstly, in Fig. 1, we use Langevin Monte-Carlo simulation to plot time evolution of SDF of the particle in the inverse harmonic potential and the metastable potential, respectively. It is seen that the two SDFs are the same at the beginning, because the metastable well does not bring effect; however, some test particles have come into the saddle point and then the both occur different, as time goes. Due to the reflection boundary of metastable potential, the SDF in the potential of this kind shows quasi-stationary Boltzmann distribution around the well and its right-tail escapes continually from the barrier, of course, all the test particles escape form the barrier in the long time limit. Nevertheless, the SDF in the inverse harmonic potential case retains Gaussian all along, but its center tends towards to the infinity after crossing over the potential top when the initial conditions are larger than the critical conditions [14]. On the other hand, we find that the descent time of the particle from the barrier to the bottom of well is enough fast, so that the influence of this process upon the modified passing probability is not important.

Let us reconsider the time-dependent process for passing over the saddle point of an inverse harmonic potential, in this case, the first equation in Eq. (2) is ignored. This model has been used widely in the calculations of fusion probability. The Brownian particle locals initially at the position \( x_0 > 0 \) and has a negative velocity \( v_0 < 0 \). The phase distribution function \( W(x, v, t) \) of the particle at time \( t \) is also a Gaussian one due to both linear equation and Gaussian noise, it is written as [16, 17, 21, 25, 26]:

\[
W(t; x, v) = \frac{1}{2\pi\sigma_x(t)\sigma_v(t)} \exp \left( -\frac{(x(t) - \langle x(t) \rangle)^2}{2\sigma_x^2(t)} \right) \exp \left( -\frac{(v(t) - \langle v(t) \rangle)^2}{2\sigma_v^2(t)} \right),
\]

(3)

where \( \langle x(t) \rangle \) is the average position of the particle and \( \sigma_x^2(t) \) is the coordinate variance, they are respectively [16]

\[
\langle x(t) \rangle = x_0 A(t) + v_0 B(t), \tag{4}
\]

\[
\sigma_x^2(t) = \frac{T}{m\omega^2} \left\{ \exp(-\gamma t) \left[ \frac{2\gamma^2}{4\omega^2 + \gamma^2} \sinh \left( \frac{t}{2} \sqrt{4\omega^2 + \gamma^2} \right) \right. \right.
\]

\[
+ \left. \frac{\gamma}{\sqrt{4\omega^2 + \gamma^2}} \sinh \left( t \sqrt{4\omega^2 + \gamma^2} \right) + 1 \right] - 1 \}, \tag{5}
\]
where $A(t)$ and $B(t)$ are given by

$$A(t) = \exp(-\gamma t) \left[ \cosh \left( \frac{t}{2} \sqrt{4\omega_s^2 + \gamma^2} \right) + \frac{\gamma}{\sqrt{4\omega_s^2 + \gamma^2}} \sinh \left( \frac{t}{2} \sqrt{4\omega_s^2 + \gamma^2} \right) \right],$$

$$B(t) = \frac{2}{m \sqrt{4\omega_s^2 + \gamma^2}} \exp(-\gamma t) \sinh \left( \frac{t}{2} \sqrt{4\omega_s^2 + \gamma^2} \right). \quad (6)$$

Time-dependent passing probability $P_{\text{pass}}(t, x_0, v_0)$ of the particle over the saddle point of inverse harmonic potential is determined by

$$P_{\text{pass}}(t; x_0, v_0) = \int_{-\infty}^{\infty} \int_{-\infty}^{0} W(t; x, v) dv dx = \frac{1}{2} \text{erfc} \left( \frac{\langle x(t) \rangle}{\sqrt{2}\sigma_x(t)} \right), \quad (7)$$

which depends on the initial preparations of coordinate and velocity of the particle.

In the case of heavy-ions fusion, a dispersion of the initial conditions should be considered with a different width, assuming a Gaussian distribution [16],

$$W_0(\bar{x}_0, \sigma_{x_0}, \bar{v}_0, T_0) = \frac{1}{2\pi \sigma_{x_0} \sqrt{mT_0}} \exp \left( -\frac{[x_0 - \bar{x}_0]^2}{2\sigma_{x_0}^2} \right) \exp \left( -\frac{[v_0 - \bar{v}_0]^2}{2mT_0} \right). \quad (8)$$

Thus time-dependent passing probability $\bar{P}_{\text{pass}}(t, x_0, v_0)$ over the saddle point of inverse harmonic potential is written as

$$\bar{P}_{\text{pass}}(t; \bar{x}_0, \sigma_{x_0}, \bar{v}_0, T_0) = \int_{-\infty}^{\infty} dx_0 \int_{-\infty}^{0} dv_0 P_{\text{pass}}(t; x_0, v_0) W_0(\bar{x}_0, \sigma_{x_0}, \bar{v}_0, T_0)$$

$$= \frac{1}{2} \text{erfc} \left( \frac{\langle \bar{x}(t) \rangle}{\sqrt{2}\sigma'_x(t)} \right), \quad (9)$$

where $\langle \bar{x}(t) \rangle$ is the same as in Eq. (5), provided that $x_0$ and $v_0$ are replaced by $\bar{x}_0$ and $\bar{v}_0$, respectively. The variance becomes

$$\sigma'^2_x(t) = \sigma^2_x(t) + \sigma^2_{x_0}(t)A^2(t) + mT_0B^2(t). \quad (10)$$

In these equations, $T_0$ is a parameter for the initial distribution that could be interpreted as the temperature of the nuclei at contact [16]. Naturally, the SDF of particle under fluctuation force becomes wider and wider, its center moves along the direction of initial velocity, as time goes. After the transient time, a part of the SDF has passed over the saddle point and then the passing probability converges to a finite value with $0 \leq \bar{P}_{\text{pass}} \leq 1$, because of $\lim_{t \to \infty} \langle \bar{x}(t) \rangle/\sigma'_x(t)=\text{constant}$ in Eq. (10).

In Fig. 2(a), we can see that the stable value of the time-dependent passing probability decreases with the increase of the initial temperature of thermalization. We also find that the
FIG. 2: (color online). (a) Time-dependent passing probability over the saddle point of inverse harmonic potential with three kinds of typical initial temperature of thermalization ($T_0 = 0.0$, $T_0 = 0.4$ and $T_0 = 2.0$) and (b) is the time-dependent passing probability with logarithmic of the time. Here, $T = 0.4$, $\gamma = 1.0$, $U_b = 1.0$, $\sigma_{x_0} = 0.0$, $\bar{v}_0 = -5.0$ and $\bar{x}_0 = 0.2$.

descent time of the particle from the barrier to the bottom of well increases with the increase of the initial temperature of thermalization, as shown in Fig. 2(b). As a consequence, the initial kinetic energy should be considered into account and this result is similar to the ref. [16] for a sharp initial condition.

We now address the modified passing probability taking into account the influence of reflection boundary of potential by a reasonable assumption. According to the Kramers rate theory, the particle subjected to thermal fluctuation in the metastable potential will decay over the barrier finally [25, 27]. We multiply the exponential decay factor into the passing probability which has been coupled the fusion and fission processes, so that the modified passing probability, namely, time-dependent probability of the particle staying inside the saddle point, is assumed to be

$$P_{m-pass}(t; x_0, v_0) = P_{pass}(t; \bar{x}_0, \sigma_{x_0}, \bar{v}_0, T_0) \exp(-r_e t) = \frac{1}{2} \text{erfc} \left( \frac{\langle x(t) \rangle}{\sqrt{2\sigma_x'(t)}} \right) \exp(-r_e t), \quad (11)$$

where $r_e$ is the steady escape rate [1, 3, 28, 32]. This approximation implies that once the particle passes over the barrier top at last time, it should escape over the barrier with the Karman's decay form. In Fig. 2(b), it is obviously that the transient time can be ignored in the calculation of the time-dependent modified passing probability. If $r_e \to 0$, $\exp(-r_e t) \simeq 1$ after a finite time, the modified passing probability [Eq. (11)] approaches the
FIG. 3: (color online). Time-dependent escape rate calculated by Langevin simulation and compared by the analytical formula of two kinds. (a) is the low-temperature case \((U_b = 1.0, T = 0.4)\) and (b) is the high-temperature case \((U_b = 0.25, T = 2.0)\).

passing probability [Eq. (9)] for the inverse harmonic potential.

The Kramers rate formula \([1, 3, 30]\) produces the better stationary result of time-dependent escape rate when the barrier height of metastable potential is larger than the temperature, as shown in Fig. 3(a). However, when the temperature is larger than the barrier height, the Kramers rate formula is not applicable, we use the inverse of the mean first passage time (MFPT) \([31]\) across an exit \(x_{ex}\) given by

\[
\tau_{\text{MFPT}}(x_0 \rightarrow x_{ex}) = \left(\frac{\sqrt{\frac{\gamma^2}{4} + \frac{\omega_s^2}{\omega_s^2} - \frac{\gamma^2}{2}}}{\omega_s^2} \frac{\omega_s}{\omega_s^2} \int_{x_0}^{x_{ex}} dy \exp \left[\frac{U(y)}{T}\right] \int_y^\infty dz \exp \left[-\frac{U(z)}{T}\right] \right) - 1.
\]  

(12)

to replace of the stationary escape rate of particle in a metastable potential well, i.e., \(r_e = (\tau_{\text{MFPT}})^{-1}\) \([3, 32]\). Noticed that we introduce here a correction factor of general damping to the pervious overdamped result, indeed, Eq. (12) is in agreement with the result of Refs. \([31, 33–37]\) in the overdamped case \((\gamma \gg \omega_s)\). At low temperature, Eq. (12) can be evaluated within the steepest-descent approximation \([3]\) as the following

\[
\tau_{\text{MFPT}}(x_0 \rightarrow x_{ex}) = \left(\frac{\sqrt{\frac{\gamma^2}{4} + \frac{\omega_s^2}{\omega_s^2} - \frac{\gamma^2}{2}}}{\omega_s^2} \frac{2\pi}{\omega_s} \exp \left(-\frac{U_b}{T}\right) \right),
\]

(13)

its inverse coincides with the Kramers rate formula \([38]\).

Furthermore, the advance of MFPT or the mean last passage time (MLPT) \([36]\) is not
FIG. 4: (color online). The time-dependent modified passing probability over the barrier of metastable potential and the passing probability over the saddle point of inverse harmonic potential. The parameters used are: $U_b = 1.0$, $T = 0.4$, $T_0 = 0.4$, $\sigma_{x_0} = 0$, $\gamma = 1.0$, $x_0 = 0.2$.

restricted to smooth metastable potentials, Eq. (12) is still suitably even if the nuclear shell correction is taken into account in the deformation potential energy of super-heavy elements. A statistical proof for the relation between the Karmers rate constant and the MFPT or the MLPT was presented in Ref. [37].

In Figs. 4 and 5, we compare the time-dependent modified passing probability [Eq. (11)] with the Langevin Monte-Carlo simulation for Eqs. (1) and (2) and the passing probability [Eq. (9)] over the saddle point of inverse harmonic potential, respectively, where three typical initial velocities are used. It is evident from Eq. (11), the modified passing probability over
FIG. 5: (color online). Comparison of time-dependent modified passing probability over the barrier of metastable potential and the passing probability over the saddle point of inverse harmonic potential. The parameters used are: $U_b = 1.0$, $T = 2.0$, $T_0 = 2.0$, $\gamma = 2.0$, $\sigma_{x_0} = 0$, $x_0 = 0.2$.

It is seen from Fig. 4 that the modified passing probability calculated by our theoretical formula is in agreement with the Langevin Monte-Carlo simulation when $U_b > T$. In particular, the maximal value of time-dependent modified passing probability is close to the stationary value of passing probability over the saddle point of inverse harmonic potential. This means that the influence of reflection boundary of metastable potential upon the transient part of time-dependent passing probability is weakly in the case of low temperature or high barrier. Figure 5 shows the calculated result at high temperature, in which the barrier
height of metastable potential is $U_b = 1.0$ and the temperature $T = 2.0$.

If the barrier height is low, the particle under influence of reflection boundary of potential is easier to escape over the saddle point, so that the time required for the modified passing probability arriving at the maximum is earlier than that of the passing probability approaching its stationary value. This concludes that the reflection boundary of metastable potential plays a decreasing role to the transient result of passing probability.

We have proposed the expression of time-dependent modified passing probability against escaping over the barrier of the metastable potential, i.e., Eq. (11), the time leading to $P_{\text{m-pass}}$ become the maximum is determined by the positive real root of following equation:

$$\frac{dP_{\text{m-pass}}}{dt} = \frac{1}{2} \exp(-r_\epsilon t)J(t) - \frac{1}{2} r_\epsilon \exp(-r_\epsilon t)\text{erfc}\left(\frac{\langle \bar{x}(t) \rangle}{\sqrt{2}\sigma'_x(t)}\right) = 0,$$

where $J(t)$ is the derivative of $\text{erfc}[\langle \bar{x}(t) \rangle/(\sqrt{2}\sigma'_x(t))]$ given by

$$J(t) = -\frac{2}{\sqrt{\pi}} \exp \left(-\frac{(\langle \bar{x}(t) \rangle)^2}{\sigma'_x(t)^2}\right) \left[\frac{M(t)}{\sqrt{2}\sigma'_x(t)} - \frac{T}{2\sqrt{2}m\omega_s^2} \langle \bar{x}(t) \rangle G(t)\right],$$

where $M(t)$ and $G(t)$ are

$$M(t) = \exp(-\gamma t) \left[\left(\frac{\bar{v}_0}{m} - \frac{\bar{x}_0}{2}\right) \cosh \left(\frac{1}{2}at\right) + \left(\frac{a\bar{x}_0}{2} + \frac{\gamma^2\bar{x}_0}{a} - \frac{2\gamma\bar{v}_0}{ma}\right) \sinh \left(\frac{1}{2}at\right)\right],$$

$$G(t) = 2\gamma \left(1 - \frac{\gamma^2}{a^2}\right) \exp(-\gamma t) \sinh^2 \left(\frac{1}{2}at\right),$$

where $a = \sqrt{4\omega_s^2 + \gamma^2}$. Hence the maximum of the time-dependent modified passing probability can be obtained by Eq. (11) through solving numerically Eqs. (14)-(16). Noticed that this quantity is a defined one depending on the model parameters. It is seen from Fig. 2 that the time corresponding to the maximal staying probability is equal approximately to the transient time of the passing probability only in the case of high barrier.

In Fig. 6 we show the maximal value of time-dependent modified passing probability over the saddle point of the metastable potential as a function of the barrier height, which is also compared with the stationary passing probability over the saddle point of inverse harmonic potential. It is seen that with the increase of the barrier height, the maximal value of time-dependent modified passing probability is close to the stationary value of the passing probability over the saddle point of inverse harmonic potential, so that one can approximately treat the asymptotical passing probability over the saddle point of the inverse harmonic potential as the fusion probability in massive nuclear fusion reaction. However,
FIG. 6: (color online). The maximum value of time-dependent modified passing probability (blue-circled-line) over the barrier of the metastable potential and the stationary passing probability (black-squared-line) over the saddle point of inverse harmonic potential, they are compared with the Langevin Monte-Carlo simulations (red-triangled-line).

when the fission barrier is low, this occurs in the super-heavy element cases, the stationary value of time-dependent passing probability over the saddle point of inverse harmonic potential is no longer applicable for the fusion probability. From the present work, we think that it is better to regard the maximal value of time-dependent modified passing probability over the saddle point of the metastable potential as the fusion probability, for that the modified passing probability is the result of whole fusion-fission process.

III. CONCLUSION

We have investigated whole fusion-fission process with Langevin approach, in which the influence of reflection boundary of the metastable potential is taken into account in the calculation of time-dependent passing probability over the saddle point. By the passing probability over the saddle point of inverse harmonic potential multiplying the exponential decay factor of the particle in the metastable potential, an approximate analytical expression of the modified time-dependent passing probability over the saddle point of metastable potential has been proposed. Our results have shown that only when the fission barrier of of fusing system is larger than the temperature, the stationary passing probability over the saddle point of an inverse harmonic potential can be regarded as the fusion probability of massive nuclei. Nevertheless, at low fission barrier, the reflection boundary plays a decreasing
role for the passing probability over the saddle point. It has been found that the time required for the modified time-dependent passing probability arriving at the maximal value is earlier than the transient time of the passing probability. This is due to the decaying probability against the passing probability.

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