Quantum effect in the diffusion along a potential barrier: Comments on the synthesis of superheavy elements

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We discuss a quantum effect in the diffusion process by developing a theory, which takes the finite curvature of the potential field into account. The transport coefficients of our theory satisfy the well-known fluctuation-dissipation theorem in the limit of Markovian approximation in the cases of diffusion in a flat potential and in a potential well. For the diffusion along a potential barrier, the diffusion coefficient can be related to the friction coefficient by an analytic continuation of the fluctuation-dissipation theorem for the case of diffusion along a potential well in the asymptotic time, but contains strong non-Markovian effects at short times. By applying our theory to the case of realistic values of the temperature, the barrier curvature, and the friction coefficient, we show that the quantum effects will play significant roles in describing the synthesis of superheavy elements, i.e., the evolution from the fusion barrier to the conditional saddle, in terms of a diffusion process. We especially point out the importance of the memory effect, which increases at lower temperatures. It makes the net quantum effects enhance the probability of crossing the conditional saddle.

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I. INTRODUCTION

Diffusion processes take place in a variety of problems [1–6]. The concept has recently been applied to theoretically describing the synthesis of superheavy elements [7,8]. It is now well accepted that it is not sufficient for the two nuclei in heavy-ion collisions to overcome the fusion (i.e., the Coulomb) barrier in order to form a heavy compound nucleus such as superheavy elements. Since the conditional saddle is located inside the fusion barrier for heavy-ion collisions between two heavy nuclei, two nuclei have to further progress inwards to approach inside the conditional saddle. The idea of Refs. [7,8], called fluctuation-dissipation dynamics, is to describe the time evolution from the fusion barrier to the conditional saddle as a diffusion process.

Though this approach is very attractive and is offering much useful information, one needs to examine the applicability of one of the basic assumptions made so far, i.e., the use of the standard fluctuation-dissipation theorem which holds at high temperatures to relate the diffusion coefficients to the friction coefficients. Since superheavy elements are stabilized by shell correction energies, one has to synthesize them at reasonably low energies, as low as 1 MeV or below. On the other hand, the curvature of the conditional saddle is also of the order of 1 MeV. It is thus required to carefully study quantum effects. An interesting issue is to explore the connection between the diffusion and friction coefficients in the diffusion process along a potential barrier under such circumstances. Though the generalization of the Einstein relation to the case of diffusion along a potential well is well known, the modification in the case of a diffusion along a potential barrier has so far been discussed only in a limited number of papers [5,6,9].

The aim of this paper is to examine this quantum effect caused by the finite curvature of the potential barrier and by the low temperature aspect of the diffusion process. To this end, we first develop a novel quantum diffusion theory which leads to a Fokker-Planck equation with non-Markovian transport coefficients. We then apply it to the situation relevant to the synthesis of superheavy elements. We will show that the quantum effects, especially the non-Markovian effects, are very important.

In Sec. II, we briefly sketch the derivation of the Fokker-Planck equation with non-Markovian transport coefficients, which include quantum effects. In Sec. III we discuss the quantum effects on the diffusion coefficient. In Sec. IV we apply the formalism to analyze the quantum effects by assuming the parameters which are relevant to describe the diffusion process in the synthesis of superheavy elements. We summarize the paper in Sec. V.

II. QUANTUM DIFFUSION EQUATION WITH NON-MARKOVIAN TRANSPORT COEFFICIENTS

We derive the required diffusion equation by extending the quasilinear response theory developed in Ref. [10] so as to include the curvature of the potential barrier. Details of the derivation will be published elsewhere [11]. Here we sketch the main steps: (1) We start form the von Neumann equation for the system consisting of space A for macroscopic degrees of freedom and space B for microscopic degrees of freedom. (2) We introduce the classical trajectory given by $(q(t), p(t))$, t being the time and $\hat{q}$ and $\hat{p}$ being the coordinate and the conjugate momentum operators of the macroscopic degrees

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of freedom. (3) We move to the Galilei transformed coordinate system specified by $q(t)$ and $p(t)$. (4) We keep only up to the second order terms of the potential and the coupling Hamiltonian in the expansion with respect to the fluctuations around the classical trajectory. (5) We solve the coupled equations describing the A and B spaces in the lowest order approximation concerning the fluctuating force. (6) We make a Wigner transform of the resultant extended von Neumann equation for subspace A.

Denoting the Wigner transform of the density operator of the subspace A by D_{AW}, we finally obtain

$$\frac{\partial}{\partial t}D_{AW}(p,q,t) = \Bigg(-\frac{1}{M}p_\alpha \frac{\partial}{\partial q_\alpha} + Cq_\alpha \frac{\partial}{\partial p_\alpha} - \chi^{(-E)}_{\alpha\beta} q_\beta \frac{\partial}{\partial p_\alpha} + \chi^{(-O)}_{\alpha\beta}\frac{\partial}{\partial p_\alpha}p_\beta + \chi^{(+O)}_{\alpha\beta}\frac{\partial^2}{\partial p_\alpha \partial q_\beta} + \chi^{(+E)}_{\alpha\beta}\frac{\partial^2}{\partial p_\alpha \partial p_\beta}\Bigg) D_{AW}(p,q,t). \tag{1}$$

The non-Markovian property of the diffusion process due to quantum effects, more specifically the effects of the curvature of the potential barrier, is hidden in the transport coefficients, which are generalized from the first and zeroth moments of the response and correlation functions, $\chi^{(-)}$ and $\chi^{(+)}$, e.g., as

$$\chi^{(-O)}_{\alpha\beta}(t) = \int_{t_0}^{t} dt_1 \mathcal{S}(t,t_1)\chi^{(-)}_{\alpha\beta}(t,t_1), \tag{2}$$

$$\chi^{(+E)}_{\alpha\beta}(t) = \int_{t_0}^{t} dt_1 \mathcal{C}(t,t_1)\chi^{(+)}_{\alpha\beta}(t,t_1). \tag{3}$$

The $\mathcal{C}$ and $\mathcal{S}$ are defined by using the curvature of the potential C as

$$\mathcal{C}(t,t_1) = \cos[\Omega(t-t_1)], \tag{4}$$

$$\mathcal{S}(t,t_1) = \frac{1}{\sqrt{MC}}\sin[\Omega(t-t_1)], \tag{5}$$

with $\Omega=\sqrt{C/M}$ when $C \geqslant 0$, i.e., for the motion in a potential well, and

$$\mathcal{C}(t,t_1) = \cosh[\Omega(t-t_1)], \tag{6}$$

$$\mathcal{S}(t,t_1) = \frac{1}{\sqrt{M|C|}}\sinh[\Omega(t-t_1)], \tag{7}$$

with $\Omega=\sqrt{|C|/M}$ when $C<0$, i.e., for a motion along a potential barrier. The response and the correlation functions, $\chi^{(-)}_{\alpha\beta}(t,t_1)$ and $\chi^{(+)}_{\alpha\beta}(t,t_1)$, are given by

$$\chi^{(-)}_{\alpha\beta}(t,t_1) = \frac{i}{\hbar}\mathrm{Tr}_B([\hat{f}_\alpha(t),\hat{f}_\beta(t_1)]\hat{D}_B(t_1)), \tag{8}$$

$$\chi^{(+)}_{\alpha\beta}(t,t_1) = \frac{1}{2}\mathrm{Tr}_B([\hat{f}_\alpha(t),\hat{f}_\beta(t_1)]_+\hat{D}_B(t_1)), \tag{9}$$

in terms of the fluctuation force $\hat{f}_\alpha(t)$ defined by

$$\hat{f}_\alpha(t) = \hat{u}_B^\dagger(t,t_0)\hat{F}_\alpha \hat{u}_B(t,t_0), \tag{10}$$

with

$$\hat{F}_\alpha \equiv \frac{\partial V_c(q(t),\hat{x})}{\partial q_\alpha} - \mathrm{Tr}\left(\frac{\partial V_c(q(t),\hat{x})}{\partial q_\alpha}\hat{\rho}_G(t)\right). \tag{11}$$

In Eq. (11), $\hat{\rho}_G(t)$ is the density operator of the total system in the Galilei transformed coordinate system. The $\hat{u}_B(t,t_0)$ is the time evolution operator of the subspace B, which satisfies the partial differential equation

$$i\hbar\frac{\partial}{\partial t}\hat{u}_B(t,t_0) = \hat{h}_B(t)\hat{u}_B(t,t_0), \tag{12}$$

with the initial condition $\hat{u}_B(t_0,t_0)=1$. In Eq. (12), the effective Hamiltonian is given by

$$\hat{h}_B(t) = \hat{H}_B(\hat{x}) + V_c(q(t),\hat{x}), \tag{13}$$

where $\hat{H}_B(\hat{x})$ is the unperturbed Hamiltonian of space B and V_c the coupling Hamiltonian. The density operator $\hat{D}(t)$ describing our basic equations [(1), (8), and (9)] is defined from the density operator in the Galilei transformed coordinate space $\hat{\rho}_G(t)$ by

$$\hat{\rho}_G(t) = \hat{u}_B(t,t_0)\hat{D}(t)\hat{u}_B^\dagger(t,t_0). \tag{14}$$

The $\hat{D}_B(t)$ is defined by $\hat{D}_B(t)=\mathrm{Tr}_A(\hat{D}(t))$.

III. QUANTUM EFFECTS ON THE FLUCTUATION-DISSIPATION THEOREM

The time evolution of the subspace B should in principle be determined by solving Eq. (12) or the corresponding reduced von Neumann equation [10,12]. Here we approximate the density operator by the canonical distribution,

$$\hat{\rho}_B(t) \approx \exp\{\beta(t)[\mathcal{F}-\hat{h}_B(t)]\}. \tag{15}$$

We further replace $\hat{h}_B(t)$ by $\hat{H}_B$ in Eq. (15) to be consistent with the linear response theory. One can then easily show by introducing spectral function [1,10] that the following well-known generalized fluctuation-dissipation theorem follows in the limit of Markovian approximation for the motion along a potential well,

$$\frac{\chi^{(+E)}_{\alpha\alpha}(t)}{\chi^{(-O)}_{\alpha\alpha}(t)} = MT^*, \tag{16}$$

with the effective temperature given by

$$T^* = \frac{1}{2}\hbar\Omega \coth\left[\tfrac{1}{2}\beta(t)\hbar\Omega\right] \tag{17}$$

$$= \begin{cases} T & (T \gg \hbar\Omega) \\ \frac{1}{2}\hbar\Omega & (T \ll \hbar\Omega). \end{cases} \tag{18}$$

Equation (1) is then nothing but the Kramers diffusion equation [13] postulated in Refs. [7,8], though there exist some modifications such as the temperature being replaced by the effective temperature.

As declared in the Introduction, our interest in connection with the synthesis of superheavy elements is the diffusion process along a potential barrier instead of along a potential well. In this case, one needs to specify a model in order to further discuss the properties of the diffusion coefficients. We assume the Feynmann-Vernon model [14], which has been used also by Caldeira and Leggett [15] to discuss macroscopic quantum tunneling, assume the Ohmic dissipation, and use the Drude regularization [3] by introducing the following cutoff function for the spectral density of the environment, i.e., the subspace B:

$$g(\omega) = \frac{1}{1 + (\omega/\omega_c)^2}. \tag{19}$$

We define

$$Y(t) \equiv \frac{\chi^{(+E)}(t)}{\chi_\infty^{(-O)}}\frac{1}{M}, \tag{20}$$

where $\chi_\infty^{(-O)}$ is the expression which the odd-moment of the response function takes in the limit of the Markovian approximation or in the asymptotic time. The $Y(t)$ consists of three terms, two of which strongly depend on time,

$$Y_1(t) = -\frac{\hbar}{4}\omega_c^2 e^{-\omega_c(t-t_0)}\cot\left[\tfrac{1}{2}\beta(t)\hbar\omega_c\right] \times\left\{\frac{1}{\omega_c-\Omega}e^{\Omega(t-t_0)} + \frac{1}{\omega_c+\Omega}e^{-\Omega(t-t_0)}\right\}, \tag{21}$$

$$Y_2(t) = \frac{1}{\beta(t)}\sum_{n=1,2,\ldots} e^{-[\pi\cdot 2n/\hbar\beta(t)](t-t_0)}\frac{\pi 2n}{\hbar\beta(t)}\frac{\omega_c^2}{\omega_c^2-[\pi 2n/\hbar\beta(t)]^2} \times\left\{\frac{1}{\pi 2n/\hbar\beta(t)-\Omega}e^{\Omega(t-t_0)} + \frac{1}{\pi 2n/\hbar\beta(t)+\Omega}e^{-\Omega(t-t_0)}\right\}. \tag{22}$$

The third one, which gives the asymptotic value, reads

$$Y_3(t) = \frac{\omega_c^2}{\omega_c^2-\Omega^2}\frac{1}{2}\hbar\Omega \cot\left[\tfrac{1}{2}\beta(t)\hbar\Omega\right]. \tag{23}$$

Note that the right-hand side (r.h.s.) of Eq. (23) is the analytic continuation of the r.h.s. of Eq. (17) concerning the sign of the barrier curvature if one ignores the minor change due to the first factor. Such analytic continuation formula of the r.h.s. of Eq. (17) has been argued in Ref. [6]. Our theory contains additionally non-Markovian effects. We note that $Y(t)$ reduces to the temperature at very high temperatures.

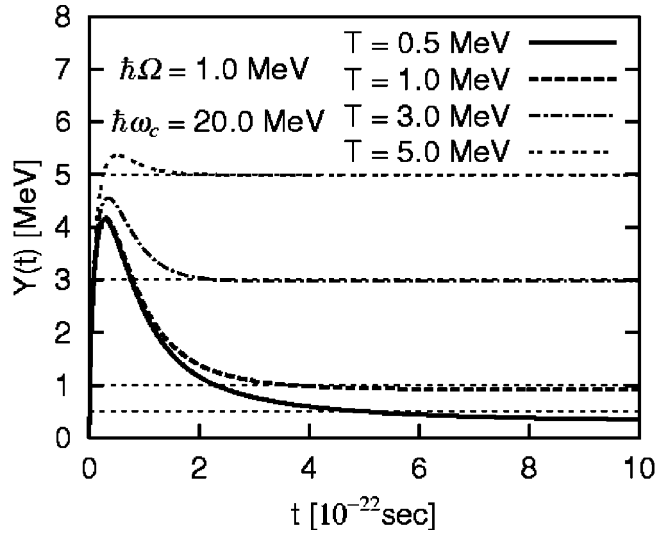

FIG. 1. Ratio of the diffusion to friction coefficients.

Figure 1 shows the time dependence of $Y(t)$, i.e., the ratio of the diffusion to the asymptotic friction coefficients, for four different values of the temperature. The barrier curvature Ω and the cutoff frequency ω_c in the spectral density in the Drude regularization have been fixed to be $\hbar\Omega = 1.0$ MeV and $\hbar\omega_c = 20.0$ MeV. The horizontal lines show the position of each temperature. The figure shows that the classical fluctuation-dissipation theorem postulated in the original Kramers paper [13] and that also in Refs. [7,8] holds only at very high temperatures. The more striking result is the non-Markovian effect, which appears as the strong variation of $Y(t)$ with time. We remark that the span where $Y(t)$ shows this strong variation gets longer roughly in proportional to the inverse of the temperature as long as $T < \hbar\omega_c$.

IV. BARRIER CROSSING PROBABILITY

We now apply our formalism to discuss quantum effects in the diffusion process from the fusion barrier to the conditional saddle in the synthesis of superheavy elements.

We first note that the average values of q and p are zero in the Galilei transformed space, and that the solution of Eq. (1) is a Gaussian. Therefore, one can set

$$D_{AW}(p,q,t) = \frac{1}{2\pi\Delta^{1/2}}\exp\left[-\frac{1}{2\Delta}\sum_{i,j} y_i y_j \tilde{\sigma}_{i,j}\right], \tag{24}$$

$$\Delta = \sigma_{qq}\sigma_{pp} - \sigma_{qp}^2, \tag{25}$$

where $y_1 = q$ and $y_2 = p$ and the 2×2 matrix $\tilde{\sigma}$ is the inverse matrix of the 2×2 matrix

$$\begin{pmatrix} \sigma_{qq} & \sigma_{qp} \\ \sigma_{qp} & \sigma_{pp} \end{pmatrix} \tag{26}$$

which determines the fluctuations, i.e., the mean square deviations from the average values. The values of σ_{ij} are obtained by solving the following coupled equations given by Eq. (1):

$$\frac{d}{dt}\begin{pmatrix}\sigma_{qq}(t)\\ \sigma_{qp}(t)\\ \sigma_{pp}(t)\end{pmatrix} = \begin{pmatrix} 0 & \frac{2}{M} & 0 \\ -(C-\chi^{(-E)}) & -\chi^{(-O)} & \frac{1}{M} \\ 0 & -2(C-\chi^{(-E)}) & -2\chi^{(-O)}\end{pmatrix} \times \begin{pmatrix}\sigma_{qq}(t)\\ \sigma_{qp}(t)\\ \sigma_{pp}(t)\end{pmatrix} + \begin{pmatrix}0\\ \chi^{(+O)}\\ 2\chi^{(+E)}\end{pmatrix}. \quad (27)$$

The Wigner distribution function for the macroscopic motion in the original space fixed frame is given by

$$\rho_{AW}(q,p,t) = D_{AW}(q-q(t),p-p(t),t), \quad (28)$$

once the Wigner distribution function in the Galilei transformed space D_{AW} is obtained.

We represent the conditional saddle by a parabola,

$$V_{cs}(q) = -\frac{1}{2}M\Omega^2 q^2, \quad (29)$$

and calculate the probability to cross the conditional saddle in order to form a compound nucleus by

$$P(t) = \int_0^\infty dq \frac{1}{\sqrt{2\pi\sigma_{qq}(t)}} \exp\left(-\frac{[q-q(t)]^2}{2\sigma_{qq}(t)}\right) = \frac{1}{2}\mathrm{erfc}\left(-\frac{q(t)}{\sqrt{2\sigma_{qq}(t)}}\right). \quad (30)$$

We ignore the radial dependence of the friction tensor. Denoting the initial position and momentum of the classical trajectory of the macroscopic variable as (q_0, p_0), the position at time t is given by

$$q(t) = e^{-\beta t/2}\left[q_0\left(\cosh\frac{\beta'}{2}t + \frac{\beta}{\beta'}\sinh\frac{\beta'}{2}t\right) + 2\frac{p_0}{\beta'}\sinh\frac{\beta'}{2}t\right], \quad (31)$$

with $\beta' = \sqrt{\beta^2 + 4\Omega^2}$. We adopt the value of the reduced friction parameter β from previous studies of fluctuation-dissipation dynamics using Langevin equation [7] as $\beta = 5\times 10^{21}\ \mathrm{s}^{-1}$. The curvature of the potential barrier is assumed to be $\hbar\Omega = 1$ MeV, which is relevant to heavy nuclei. The initial position q_0 is chosen to make the height of the conditional saddle be 4.0 MeV, and the mass parameter to correspond to the reduced mass in the collision of the mass number 48 and 238 nuclei. We defer the study of the effects of purely non-Markovian terms $\chi^{(-E)}$ and $\chi^{(+O)}$ and leave them in determining the $\sigma_{qq}(t)$ in the following analyses.

Figure 2 shows the probability to cross the conditional saddle as a function of the initial kinetic energy K, which is measured relative to the height of the conditional saddle V_B. We remark that the ratio $q(t)/\sqrt{2\sigma_{qq}(t)}$ in Eq. (30) converges to an asymptotic value. It was used to evaluate the probability to cross the conditional saddle P. We choose three values for the temperature. The solid lines are the results of our theory, while the dot-dashed lines are the results when the classical fluctuation-dissipation theorem has been assumed by ignoring the quantum effects due to the finite curvature of the conditional saddle. The figure clearly shows that the quantum effect is important at low temperatures, which are relevant to the synthesis of superheavy elements.

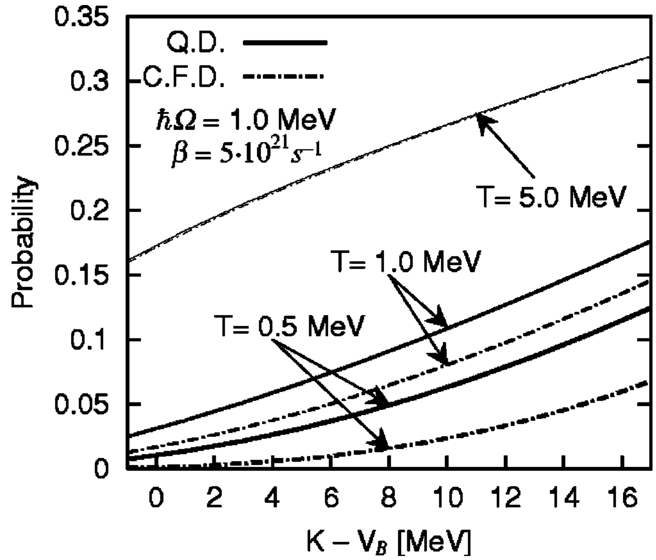

FIG. 2. Comparison of the probability to cross the conditional saddle calculated by quantum diffusion equation and by assuming the classical fluctuation-dissipation theorem.

Our theory contains a memory effect. In order to discuss the connection to a previous work [6], we artificially isolate the memory effect by calculating the probability to cross the conditional saddle by using the asymptotic value of the diffusion coefficient. The result is added in Fig. 3 by the dotted line. We observe that the probability to cross the conditional saddle is reduced by the quantum effect if one ignores the memory effect. This is because the asymptotic diffusion coefficient in the quantum theory is smaller than that obtained from the classical fluctuation-dissipation theorem. A similar

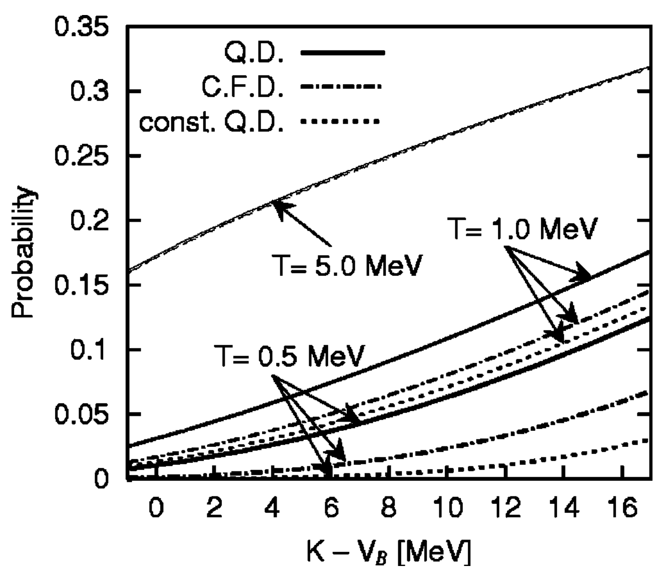

FIG. 3. Analysis of the memory effects.

effect has been shown in Ref. [9]. Our study shows in addition that the memory effect overcomes this effect and finally the net quantum effects enhance the transmission probability of the conditional saddle. In other words, the net quantum effects reduce the fusion hindrance.

In passing, we wish to mention that quantum effects on diffusion process are also discussed in Ref. [16] following a different approach. However, there are some important differences in the expressions of transport coefficients. For example, the diffusion coefficient given by Eq. (8) in Ref. [16] does not seem to match with our asymptotic formula (23), as well as those presented in Refs. [6,9]. We also wish to refer to Ref. [17], which discusses the dynamics of barrier penetration in a thermal medium for the inverted harmonic oscillator by using the influence functional formalism of the path integral method.

V. SUMMARY AND FUTURE DEVELOPMENTS

We have presented a diffusion theory which takes the finite curvature of the potential field into account. The theory is then applied to the case where the temperature, barrier curvature, and the friction coefficient are taken to represent a realistic situation of the diffusion process from the fusion barrier to the conditional saddle in the synthesis of superheavy elements. We have thus shown that the quantum effects will play an important role. We especially pointed out the importance of the memory effect which has been omitted in any previous works. It makes the net quantum effects enhance the probability of crossing the conditional saddle, while the quantum effect reduces it if the memory effect is ignored, as has been shown in Ref. [9].

We have artificially left out some of the genuine non-Markovian terms, i.e., the odd moment of the correlation function and the even moment of the response function. Also, we have assumed a sharp distribution at the initial time and left out the effects of spreading of the initial distribution. We will discuss these effects as well as the dependence of the quantum effects on the strength of the dissipative force in forthcoming papers. One of the interesting problems is to clarify whether our conclusion concerning the role of non-Markovian effect is special to our specific choice of the Caldeira-Leggett model, especially to the Ohmic dissipation, or holds in general. This is another issue which we will explore in the near future.

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