Quantum Kramers model: Solution of the turnover problem

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The quantum-mechanical version of the Kramers turnover problem is considered. The multidimensional character of the problem is taken into account via transformation to normal modes. This eliminates the coupling to the bath near the barrier top allowing the use of a simple harmonic transmission coefficient for the barrier dynamics. The well dynamics is described by a continuum form of a master equation for the energy in the unstable normal mode. Within first-order perturbation theory, the equations of motion for the stable normal modes have the form of a forced oscillator. The transition probability kernel is found using the known solution for the quantum forced oscillator problem. An expression for the quantum escape rate is derived. It encompasses all previously known limiting results in the thermally activated tunneling regime. The depopulation factor, which accounts for the nonequilibrium energy distribution is evaluated. The quantum transition probability kernel is broader than the classical and is skewed towards lower energies. Interplay between these two effects, together with a positive tunneling contribution, determines the relative magnitude of the quantum rate compared to the classical one. The theory is valid for arbitrary dissipation. Its use is illustrated for the case of a cubic potential with Ohmic (Markovian) dissipation.

I. INTRODUCTION

Fifty years ago, Kramers formulated the problem of escape of a particle trapped in a potential energy well, separated from a continuum by a barrier.[1] The particle is coupled to a heat bath which exerts on it a friction and a random force. Kramers showed that the escape mechanism is qualitatively different in the weak and strong coupling limits. Strong coupling leads to a thermal equilibrium within the well. Consequently the passage over the barrier is the rate determining step. In the weak-coupling limit there is depopulation of particles with energies close to the top of the barrier, so that the rate is determined by the exchange of energy between the particle and the bath.[2,3] In the former limit the rate decreases with increasing coupling while in the latter case it increases with the coupling strength. Kramers solved the classical problem in both limits, noted the existence of a turnover region, but did not derive a uniform expression valid for all coupling strengths. In Kramers's work, the fundamental equation describing the dynamics was a Langevin equation. Recently it has been replaced by the generalized Langevin equation (GLE) which includes memory effects.[2,4–6]

During the last decade the quantum version of the Kramers problem, namely, the tunneling decay of a metastable state in the presence of coupling to a thermal bath (dissipation),[7] has become very popular. Interest was stimulated by an increasing amount of experimental data on the decay of the zero voltage state in current biased resistively shunted Josephson junctions[8] (RSJ) and on the transitions between the fluxon states in superconducting rings interrupted by low capacitance junctions[9] (SQUID). These experiments span a large temperature domain from a few mK, where quantum effects are extremely important, to higher temperatures, at which the system can be described classically. There is also a possibility of varying the damping in a controlled way, spanning a wide region, from the extremely underdamped limit to the overdamped limit.[10]

The quantum Kramers problem is formulated by replacing the GLE with an equivalent Hamiltonian in which the system is coupled linearly to a bath of harmonic oscillators.[11,12] Initial work on the quantum problem was implicitly based on the assumption of thermal equilibrium within the well, allowing the use of methods of equilibrium statistical mechanics. Caldeira and Leggett[12] have shown that at zero temperature dissipation leads to an exponential reduction of the tunneling rate. Note that at $T=0$ the equilibrium assumption is valid for arbitrary coupling strength. Coupling to the bath leads to an exponential enhancement of the rate at low temperatures compared with its zero temperature value as shown by Larkin and Ovchinnikov[13] and by Grabert, Weiss, and Hänggi.[14] The expression for the high-temperature escape rate derived by Wolynes[15] assuming parabolic barrier was divergent at the so-called crossover temperature T_0. This divergence originates from the extrapolation of the lower limit of integration to $-\infty$ in the calculation of the flux. Larkin and Ovchinnikov,[16] Grabert and Weiss,[17] and Riseborough, Hänggi, and Freidkin[18] have shown that the artificial divergence can be eliminated by taking into account the deviation of the barrier shape from a parabolic one. It should be noted that the harmonic approximation to the barrier is justified only at high temperatures, when the escape occurs close to the top of the barrier. At lower temperatures the actual shape of the barrier is important. Wolynes's result has been shown subsequently to be the quantum analog of the classical multidimensional transition state theory[19,20] (TST).

Within the harmonic approximation for the barrier, it is possible to eliminate the coupling to the bath by a normal mode transformation. This observation was used by Pollak and by Dakhnovskii and Ovchinnikov to show that the tunneling rate in the presence of dissipation can be derived from TST both below[21] and above[19,20(b)] T_0. Adapting the procedure of Affleck,[22] Hänggi and Hontscha have shown[23] that a unified expression for the rate, valid at all temperatures, may be derived from the Miller formulation[24] of multidimensional semiclassical TST.

If the coupling of the particle to the bath is weak, the thermal equilibrium assumption is no longer valid. This limit has been treated for the classical escape problem.[1,3,25,26] The first attempt to solve the quantum Kramers problem without the equilibrium assumption has been made by Mel'nikov.[27] He formulated and solved an integral equation for the steady-state energy distribution in the presence of tunneling. The dynamics in the energy space may also be approximated by the diffusion equation. This has been solved for a parabolic barrier in Ref. 27 within the WKB approximation and by Rips and Jortner[28] using the exact expression for the transmission coefficient. Mel'nikov's integral equation is a steady-state solution of the master equation, which describes the well dynamics. The diffusion equation can be derived from it as shown recently by Griff *et al.*[29]

The original description of the well dynamics[27] was classical, since it was based on a Gaussian transition probability kernel for the energy of the particle. Larkin and Ovchinnikov[30] generalized Mel'nikov's approach, and derived an approximate expression for the quantum transition probability for a system, coupled to a bath, whose spectrum is modeled by the Johnson-Nyquist quantum thermal noise. In the paper of Mel'nikov and Sütö[31] the method was applied to the case of the tilted cosine (washboard) potential. This is of practical importance as it describes the decay of the zero-voltage state in a RSJ.[8,10] The results in these papers were complementary to that based on the thermal equilibrium assumption as they were limited to a low damping region.

Mel'nikov's work was at the same time an important step towards the solution of the Kramers turnover problem (the early attempts[32,33] were based upon the mean first-passage time approximation and consequently unextendable to the quantum case). He derived an expression for the rate which went continuously from the extremely underdamped limit, to the TST formula for the particle in the absence of friction. Mel'nikov and Meshkov[34] have also written an *ad hoc* product form for the classical escape rate which leads to the multidimensional TST limit for large damping. The main elements missing from their theory are a derivation of their expression and its being limited to Ohmic (Markovian) dissipation. Memory effects,[35] which can be extremely important as demonstrated in the numerical simulations of Straub, Borkovec, and Berne[36] were not included in their treatment.

The first systematic solution of the classical Kramers turnover problem was recently given by Grabert[37] and by Pollak, Grabert, and Hänggi[38] (PGH). It was based upon two new elements. The first one was the observation that escape does not occur along the original system coordinate, but along the unstable normal mode of the combined system and bath.[20,21] The second was a systematic perturbative treatment[37] of the nonlinear part of the potential which couples the unstable mode with the bath of stable modes.

The purpose of the present paper is to provide a consistent solution of the quantum Kramers turnover problem. The method is a synthesis of the quantum theoretical treatment of the well and barrier dynamics of Mel'nikov and Larkin and Ovchinnikov, and the normal-mode approach to the classical Kramers turnover problem of PGH. Using the equations of motion for the stable normal modes we derive a new quantum-mechanical expression for the transition probability kernel. The latter is employed in the solution of the integral equation for the stationary energy distribution function for the unstable normal mode and derivation of the quantum escape rate. The main physical results of our analysis (which can be extended below T_0) is that the quantum transition probability kernel is asymmetric and broader than the classical one. The quantum broadening leads to an equilibration of the system at weaker damping values relative to the classical case and to an enhancement of the quantum rate. The skewedness, however, is biased towards lower energies and causes depopulation at energies close to the barrier top and reduction of the rate.

In Sec. II we review briefly the turnover theory[37,38] for the classical case. The expression for the quantum transition probability kernel is derived in Sec. III. It is used in Sec. IV to obtain a closed expression for the quantum escape rate. The leading quantum correction is analyzed in Sec. V via the high temperature expansion of the rate (depopulation factor). Finally, in Sec. VI we provide a detailed study of the particular case of a cubic potential with Ohmic (Markovian) dissipation. Limitations of the theory, its possible extensions and applications are discussed in Sec. VII.

II. TURNOVER THEORY FOR THE CLASSICAL KRAMERS PROBLEM

The general physical model to be studied in this paper is that of a particle with mass M moving along the system coordinate q in a potential $V(q)$. The interaction of the particle with a thermal bath is described by the time-dependent friction function $\gamma(t)$. The classical equation of motion for the particle is the GLE

$$\ddot{q}(t)+\frac{1}{M}\frac{\partial V(q)}{\partial q}+\int_0^t dt'\gamma(t-t')\dot{q}(t')=\frac{F_{\text{st}}(t)}{M} . \tag{2.1}$$

$F_{\text{st}}(t)$ is a stochastic Gaussian force whose autocorrelation function is related to the friction function by the classical fluctuation-dissipation theorem

$$\langle F_{st}(t)F_{st}(t')\rangle = Mk_BT\gamma(t-t') \equiv \frac{M}{\beta}\gamma(t-t') \ . \quad (2.2)$$

The brackets denote a thermal average at temperature T. The potential $V(q)$ is assumed to have a local minimum at $q=q_w$ with frequency Ω. This well is separated from a continuum by a barrier, located at $q=0$ with height V_0 (cf. Fig. 1). The problem to be considered is the determination of the quantum-mechanical escape rate of the particle, which is initially trapped in the well. Before proceeding to the quantum problem we review the theory for the classical limit derived in Refs. 37 and 38.

Zwanzig[11(b)] has shown that a GLE can be derived from the Hamiltonian

$$H = \frac{P_q^2}{2M} + V(q) + \sum_{i=1}^{N}\left[\frac{p_i^2}{2m_i} + \frac{1}{2}m_i\left[\omega_i x_i - \frac{c_i}{m_i\omega_i}q\right]^2\right] \quad (2.3)$$

in which the system coordinate q is coupled linearly via the coupling constant c_i to a bath of harmonic oscillators with frequencies ω_i, masses m_i, and coordinates x_i. The time-dependent friction of the GLE, which follows from the Hamiltonian is expressed in terms of the bath frequencies and the coupling constants

$$\gamma(t) = \frac{1}{M}\sum_{i=1}^{N}\frac{c_i^2}{m_i\omega_i^2}\cos(\omega_i t) \ . \quad (2.4)$$

An alternative description of dissipative properties of the bath is based on the spectral density $J(\omega)$ defined as[12,39]

$$J(\omega) = \frac{\pi}{2M}\sum_{i=1}^{N}\frac{c_i^2}{m_i\omega_i}[\delta(\omega-\omega_i)-\delta(\omega+\omega_i)] \ . \quad (2.5)$$

The latter is related to the friction function $\gamma(t)$ by

$$J(\omega) = \omega\int_0^\infty dt\, \gamma(t)\cos(\omega t) \ . \quad (2.6)$$

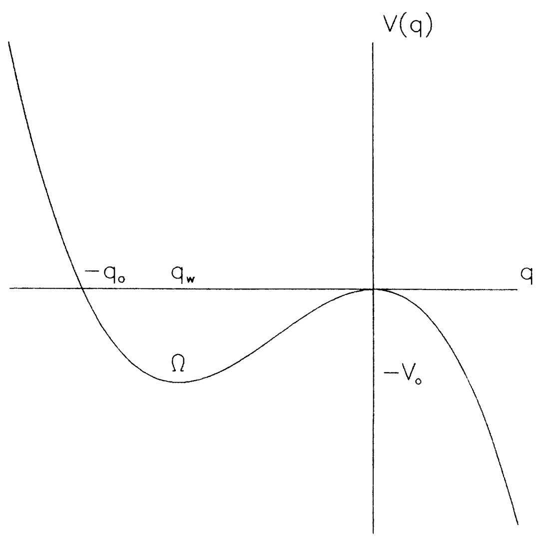

FIG. 1. Schematic representation of a metastable potential. The figure corresponds to a cubic potential $V(q) = -\frac{27}{4}V_0(q/q_0)^2(1+q/q_0)$. For the notations see text.

Equations (2.4)–(2.6) give the connection between the continuum GLE, Eq. (2.1), and the Hamiltonian (2.3).

In the following we shall mostly work in the frequency domain. The Laplace transform of a function $g(t)$ is defined as

$$\hat{g}(p) = \int_0^\infty dt\, g(t)\exp(-pt) \ . \quad (2.7)$$

In the vicinity of the barrier top ($q=0$) one can write the harmonic expansion of the potential $V(q)$ as

$$V(q) \simeq V_L(q) = -\tfrac{1}{2}M\omega_b^2 q^2 \quad (2.8)$$

where ω_b is the bare frequency of the barrier. The total potential is then separated into the linear part, Eq. (2.8), and the nonlinear term $V_{NL}(q)$:

$$V_{NL}(q) = V(q) + \tfrac{1}{2}M\omega_b^2 q^2 \quad (2.9)$$

Substitution of the linearized potential $V_L(q)$ into Eq. (2.3) leads to a quadratic Hamiltonian which can be diagonalized by an orthogonal transformation. The technical details of this normal mode transformation can be found in Refs. 20, 21, and 40. Here we give only the important relations required for the development of the quantum theory.

The linearized Hamiltonian H_L in the normal mode representation has the form

$$H_L = \tfrac{1}{2}\dot{\rho}^2 - \tfrac{1}{2}\lambda_0^2\rho^2 + \sum_{i=1}^{N}\tfrac{1}{2}(\dot{y}_i^2 + \lambda_i^2 y_i^2) \quad (2.10)$$

where ρ and $\{y_i\}_{i=1}^N$ are the mass-weighted coordinates of the unstable normal mode (with frequency λ_0) and the stable normal modes (with frequencies $\{\lambda_i\}_{i=1}^N$). Transformation to the normal mode coordinates is specified by the orthogonal matrix u_{ij}. The mass-weighted system coordinate q' is expressed in terms of the normal modes as follows:

$$q' \equiv M^{1/2}q = u_{oo}\rho + \sum_{i=1}^{N}u_{io}y_i \ . \quad (2.11)$$

The normal mode frequencies and matrix elements u_{ij} are readily expressed in terms of the parameters of the model, namely, the spectral density or the friction function. We shall be particularly interested in the expressions for the frequency of the unstable mode (the Grote-Hynes relation[41])

$$\lambda_0^2 = \frac{\omega_b^2}{1+\hat{\gamma}(\lambda_0)/\lambda_0} \quad (2.12)$$

and the diagonal matrix element u_{oo} which gives the projection of the system coordinate onto the unstable normal mode:

$$u_{oo} = \left[1 + \frac{1}{\pi}\int_{-\infty}^{\infty}d\omega\frac{J(\omega)\omega}{(\omega^2+\lambda_0^2)^2}\right]^{-1/2} \ . \quad (2.13)$$

Note that λ_0 is always smaller than the bare barrier frequency ω_b. The orthogonal transformation diagonalizes the Hamiltonian only locally (at the barrier top). The modes are coupled by the nonlinear part of the potential

$V_{NL}(q)$. However, if the matrix element u_{oo} is close to unity then the deviation of the unstable normal mode from the system coordinate may be taken as a small parameter of the problem[37]

$$\sigma \equiv \sum_{i=1}^{N} g_i^2 = \frac{1}{u_{oo}^2} - 1 \tag{2.14}$$

where $g_i \equiv u_{io}/u_{oo}$. The small parameter σ can be also expressed in terms of the friction function as[38]

$$\sigma = \frac{1}{2}\left[\frac{\hat{\gamma}(\lambda_0)}{\lambda_0} + \left.\frac{\partial \hat{\gamma}(p)}{\partial p}\right|_{p=\lambda_0}\right] . \tag{2.15}$$

The existence of the small parameter justifies a perturbative approach to the solution of the equations of motion for the normal modes.[37] Grabert and co-workers have shown that the zero-order equation of motion for the unstable normal mode has the form[37,38]

$$\ddot{\rho} - \lambda_0^2 \rho = F(t) . \tag{2.16}$$

The equation of motion for the ith stable mode to first order in the small parameter g_i is

$$\ddot{y}_i + \lambda_i^2 y_i = g_i F(t) . \tag{2.17}$$

Here $F(t)$ is the time-dependent (zero-order) force acting on the unstable normal mode

$$F(t) = -u_{oo} M^{-1/2} \left.\frac{\partial V_{NL}(q)}{\partial q}\right|_{q=M^{-1/2}u_{oo}\rho} . \tag{2.18}$$

The solution of the forced oscillator equation of motion for the ith stable normal mode, Eq. (2.17), depends on the initial conditions. It is assumed that at the initial time the stable modes are in thermal equilibrium. As a result, any physical observable which is a function of the initial conditions and, in particular, the energy E_i, acquired by the ith stable mode during one period of the unstable normal mode is a stochastic Gaussian variable. The same is true also for the energy E, lost by the unstable normal mode, which is simply given by the sum of all energies E_i. This leads to the conclusion[37] that the probability $P_{cl}(\varepsilon;\varepsilon')d\varepsilon$ that a system leaving the barrier region with energy ε' in the unstable mode, will return to the barrier with an energy between ε and $\varepsilon+d\varepsilon$ has a Gaussian form[27]

$$P_{cl}(\varepsilon;\varepsilon') = P_{cl}(\varepsilon-\varepsilon') = (4\pi\delta)^{-1/2}\exp\left[-\frac{(\varepsilon-\varepsilon'+\delta)^2}{4\delta}\right] . \tag{2.19}$$

Here and in the following ε is the dimensionless energy variable $\varepsilon \equiv \beta E$. Furthermore δ is the average energy loss per period of the unstable normal mode

$$\delta \equiv \beta\langle \Delta E\rangle_{cl} = \frac{\beta}{2}\int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 K(t_1-t_2)F(t_1)F(t_2) . \tag{2.20}$$

The force $F(t)$ [cf. Eq (2.18)] comes from the classical conservative trajectory for the unstable mode at the barrier energy ($\varepsilon=0$). $K(t)$ is the classical dissipation kernel:[37,38]

$$K(t) = \sum_{i=1}^{N} g_i^2 \cos(\lambda_i t) . \tag{2.21}$$

Its Laplace transform can also be expressed in terms of the friction function[38,40]

$$\hat{K}(p) = \frac{1}{u_{oo}^2}\frac{p}{p^2+p\hat{\gamma}(p)-\omega_b^2} - \frac{p}{(p^2-\lambda_0^2)} . \tag{2.22}$$

As shown in Refs. 37 and 38 the classical escape rate of a particle factorizes and can be written as

$$\Gamma_{cl} = \frac{\Omega}{2\pi}\frac{\lambda_0}{\omega_b}\Upsilon_{cl}\exp(-\beta V_0) . \tag{2.23}$$

The classical depopulation factor Υ_{cl} is given in terms of the average dissipated energy as[27,34]

$$\Upsilon_{cl} = \exp\left[\frac{1}{2\pi}\int_{-\infty}^{\infty} d\tau \frac{\ln(1-e^{-\delta(\tau^2+1/4)})}{\tau^2+\frac{1}{4}}\right] . \tag{2.24}$$

The information about the potential enters the final rate expression via the well (Ω) and the barrier (ω_b) frequency of the potential $V(q)$ as well as the barrier height V_0 (cf. Fig. 1).

III. THE QUANTUM TRANSITION PROBABILITY AND ENERGY LOSS

A. The transition probability

In the classical theory the depopulation factor is evaluated using the Gaussian transition probability $P_{cl}(\varepsilon;\varepsilon')$ that the energy of the unstable mode changes from ε' to ε during one period of motion. In this section we derive a quantum-mechanical expression for this transition probability. The basic assumption is that the motion of the unstable normal mode is classical and can be approximated by the zero-order equation of motion, Eq. (2.16). The stable normal modes are treated quantum mechanically. Their classical equation of motion is that of a forced oscillator. The general quantum expressions for the time-dependent transition probability from the initial oscillator state n to the final state m has been derived by Feynman[42] and by Schwinger.[43] The result for the ith stable mode is[43]

$$W_{m_i\leftarrow n_i}(t) = W_{n_i\leftarrow m_i}(t) = \frac{n_{i<}!}{n_{i>}!}\nu_i^{|n_i-m_i|}\exp(-\nu_i)[L_{n_{i<}}^{|n_i-m_i|}(\nu_i)]^2 , \tag{3.1}$$

where $n_{i>} \equiv \max(m_i,n_i)$ and $n_{i<} \equiv \min(m_i,n_i)$; $L_n^{\alpha}(x)$ is the generalized Laguerre polynomial and ν_i is

$$\nu_i \equiv \nu_i(t) = \frac{g_i^2}{2\hbar\lambda_i}\left|\int_{-\infty}^{t} dt_1 e^{i\lambda_i t_1}F(t_1)\right|^2 \tag{3.2}$$

where the time-dependent force starts acting upon the oscillator at $t=-\infty$ and is switched off at time t. [Note that the $M^{-1/2}$ term is incorporated in the definition of the force, Eq. (2.18), and therefore does not appear in Eq. (3.2)].

To obtain the quantum-mechanical transition probability $P(\varepsilon;\varepsilon')$ for the unstable mode we shall have to make some further assumptions.

(i) The stable normal modes are statistically independent. This assumption is implicit in the classical derivation[37,38] and is true in the first-order approximation with respect to the parameters g_i when there is no coupling between different stable modes.

(ii) Energy distribution in all of the stable modes at the initial time ($t \to -\infty$) is a thermal one, namely,

$$\rho_{n_i} = e^{-n_i \beta\hbar\lambda_i}(1-e^{-\beta\hbar\lambda_i}) \ . \tag{3.3}$$

Here the only difference with respect to the classical limit is the use of the exact quantum distribution function for the harmonic oscillator instead of the classical.

(iii) The unstable mode can accommodate any amount of energy that the bath of stable modes can supply and vice versa. This assumption is actually the most severe one. It implies neglect of quantization of the energy levels of the unstable normal mode and has been already introduced implicitly by the classical treatment of this mode. It is justified because we are interested in the transition probability at energies that are very close to the top of the barrier. This is the energy interval in which the density of (resonance) states of the unstable mode is highest. Classically the density of states diverges at $\varepsilon=0$.

(iv) Finally, the force $F(t)$ is independent of the energy in the unstable mode in an energy interval of $\sim k_B T$ around the barrier top ($\varepsilon=0$).[27,30,37,38] This assumption leads to the transition probability $P(\varepsilon;\varepsilon')$ which is a function of the energy difference $\varepsilon-\varepsilon'$ only, i.e., $P(\varepsilon;\varepsilon')=P(\varepsilon-\varepsilon';0)$. This property of the transition probability will be fully exploited in Sec. IV. Here we note that the assumption allows us to use the force for the conservative trajectory of the unstable mode at the barrier energy $\varepsilon=0$. This asymptotic trajectory starts at $t\to-\infty$ at the barrier top, reaches the turning point at $t=0$ and returns to the barrier at $t\to\infty$.

The transition probability $P(\varepsilon)\equiv P(\varepsilon;0)$ that the unstable mode acquires energy ε during the period of conservative motion is

$$P(\varepsilon) = \sum_{\{m_i\}=0}^{\infty} \sum_{\{n_i\}=0}^{\infty} \delta\left[\varepsilon + \sum_{i=1}^{N}(m_i-n_i)\beta\hbar\lambda_i\right] \times \prod_{i=1}^{N} W_{m_i\leftarrow n_i}\rho_{n_i} \ . \tag{3.4}$$

This expression is obtained by noting that the energy gained by the unstable mode is equal to the sum of energies lost by all the stable modes. One must then simply sum over transition probabilities for all states of the stable modes, weighted by their initial distribution. Note that the time dependence of $W_{m_i\leftarrow n_i}$ has been removed. The upper limit in Eq. (3.2) for the asymptotic trajectory tends to infinity.

The next step is to simplify Eq. (3.4) by performing the summation over all bath states. This is achieved by studying the Fourier transform of $P(\varepsilon)$

$$\tilde{P}(\tau) = \int_{-\infty}^{\infty} d\varepsilon \exp(i\tau\varepsilon)P(\varepsilon) \ . \tag{3.5}$$

Using Eq. (3.4) we find

$$\tilde{P}(\tau) = \sum_{\{m_i\}=0}^{\infty} \sum_{\{n_i\}=0}^{\infty} \prod_{i=1}^{N} e^{-i\tau(m_i-n_i)\beta\hbar\lambda_i} W_{m_i\leftarrow n_i}\rho_{n_i} \ . \tag{3.6}$$

Explicit evaluation of this expression is facilitated by defining a generating function

$$G(\xi,\nu,x) \equiv (1-x)\sum_{m=0}^{\infty}\sum_{n=0}^{\infty} e^{i\xi(m-n)}W_{m\leftarrow n}x^n \ . \tag{3.7}$$

This generating function is simplified by using Schwinger's result[43]

$$\sum_{m=0}^{\infty} e^{i\xi(m-n)}W_{m\leftarrow n} = \exp[(e^{i\xi}-1)\nu]L_n^0((e^{-i\xi}-1)(e^{i\xi}-1)\nu) \equiv \exp(y)L_n^0(z) \ , \tag{3.8}$$

where the last expression on the right-hand side defines the variables y and z. Substitution of Eq. (3.8) into Eq. (3.7) gives

$$G(\xi,\nu,x) = (1-x)\exp(y)\sum_{n=0}^{\infty} L_n^0(z)x^n = \exp\left[y+\frac{xz}{x-1}\right] , \tag{3.9}$$

where we have used the expression for the generating function of Laguerre polynomials.[44]

Inserting Eqs. (3.1)–(3.3) into Eq. (3.6), using Eq. (3.9) and straightforward manipulation we derive a compact expression for the Fourier transform of the transition probability function

$$\tilde{P}(\tau) = \prod_{i=1}^{N} G(\xi_i,\nu_i,x_i) \tag{3.10}$$

where

$$\xi_i \equiv -\tau\beta\hbar\lambda_i \tag{3.10a}$$

and

$$x_i \equiv \exp(-\beta\hbar\lambda_i) \ . \tag{3.10b}$$

Using the result for the generating function $G(\xi,\nu,x)$ we can obtain a closed expression for $\tilde{P}(\tau)$:

$$\tilde{P}(\tau) = \exp\left[-\sum_{i=1}^{N}\frac{\nu_i[1-e^{-i\tau\beta\hbar\lambda_i}][1-e^{-\beta\hbar\lambda_i(1-i\tau)}]}{[1-e^{-\beta\hbar\lambda_i}]}\right] .$$

However, as will be shown in Sec. IV, what is actually required for the calculation of the quantum-mechanical escape rate is the function $\tilde{P}(\tau-i/2)$. For the latter we derive the following expression

$$\tilde{P}(\tau-i/2)=\exp\left[-\sum_{i=1}^{N}\frac{\nu_i[\cosh(\beta\hbar\lambda_i/2)-\cos(\tau\beta\hbar\lambda_i)]}{\sinh(\beta\hbar\lambda_i/2)}\right]\equiv\exp[-R(\tau)] \ . \tag{3.11}$$

Equation (3.11) is the central result of this section. Later on we shall express it in terms of the macroscopic parameters of the model (i.e., the friction function). In Sec. IV it will be used to derive the quantum expression for the escape rate.

B. The quantum energy loss

The result for the generating function [cf. Eq. (3.9)] can be used to derive analytic expressions for the moments of the energy dissipated by the unstable mode. The (dimensionless) energy acquired by the ith stable normal mode per period of the asymptotic trajectory of the unstable mode is given by

$$\langle\Delta\varepsilon_i\rangle=\sum_{m_i=0}^{\infty}\sum_{n_i=0}^{\infty}\beta\hbar\lambda_i(m_i-n_i)W_{m_i\leftarrow n_i}\rho_{n_i} \ . \tag{3.12}$$

Using Eq. (3.9) one finds that

$$\langle\Delta\varepsilon_i\rangle=\beta\hbar\lambda_i\frac{\partial G(\xi_i,\nu_i,x_i)}{\partial(i\xi_i)}\bigg|_{\xi_i=0}=\beta\hbar\lambda_i\nu_i \ . \tag{3.13}$$

The average energy lost by the unstable mode is equal to the total energy acquired by the system of stable normal modes. Using the definition of ν_i, Eq. (3.2), we obtain

$$\langle\Delta\varepsilon\rangle=\frac{\beta}{2}\sum_{i=1}^{N}g_i^2\left|\int_{-\infty}^{\infty}dt\, e^{i\lambda_i t}F(t)\right|^2=\delta \ , \tag{3.14}$$

which coincides with the classical energy loss [cf. Eqs. (2.20) and (2.21)].

Following a similar reasoning it is straightforward to derive expressions for the moments $\langle\Delta\varepsilon_i^n\rangle$ and cumulants $\langle\delta^n\varepsilon_i\rangle$ of the energy acquired by the ith stable mode. Using the statistical independence of stable modes we obtain the cumulants[45] of the energy loss of the unstable mode. As an example we present below the expressions for the second- (dispersion) and the third-order cumulants

$$\langle\delta^2\varepsilon\rangle=\sum_{i=1}^{N}(\langle\Delta\varepsilon_i^2\rangle-\langle\Delta\varepsilon_i\rangle^2)$$
$$=\sum_{i=1}^{N}(\beta\hbar\lambda_i)^2\nu_i\coth(\beta\hbar\lambda_i2) \tag{3.15}$$

and

$$\langle\delta^3\varepsilon\rangle=\sum_{i=1}^{N}(\langle\Delta\varepsilon_i^3\rangle-3\langle\Delta\varepsilon_i^2\rangle\langle\Delta\varepsilon_i\rangle+\langle\Delta\varepsilon_i\rangle^2)$$
$$=\sum_{i=1}^{N}(\beta\hbar\lambda_i)^3\nu_i \ . \tag{3.16}$$

Note that the third-order cumulant as well as the leading quantum correction to the dispersion are of order $\hbar^2$. In the limit $\hbar\to 0$ the expression for the dispersion reduces to the classical result $\langle\delta^2\varepsilon\rangle_{\rm cl}=2\delta$. Furthermore, since $\coth(x)\geq 1$ the quantum dispersion is greater than the classical result. The fact that the third-order (and higher-order) cumulants do not vanish demonstrates that in the quantum case the transition probability function $P(\varepsilon)$ is no longer Gaussian.[30]

C. The continuum limit

Thus far, the transition probability, its Fourier transform, and the moments of dissipated energy have been given in terms of the parameters of the normal modes. These expressions are appropriate if the bath consists of a finite number of modes or, equivalently, that their spectrum is discrete. Below we derive closed expressions for these quantities in terms of parameters of the continuum model, namely, the Laplace transforms of the friction function $\hat{\gamma}(\omega)$ or, equivalently, the classical dissipation kernel $\hat{K}(\omega)$. For this purpose we introduce the spectral function $I(\lambda)$ of the bath of stable normal modes

$$I(\lambda)=\frac{\pi}{2}\sum_{i=1}^{N}\frac{g_i^2}{\lambda_i}[\delta(\lambda-\lambda_i)-\delta(\lambda+\lambda_i)] \ . \tag{3.17}$$

This function provides a complete description (within the first-order perturbation approximation) of the dissipative properties of the bath of stable modes, to which the unstable mode is linearly coupled. In this respect it is analogous to the spectral density $J(\omega)$ of the bath to which the system coordinate is coupled. As we shall see the dissipative properties of the two baths can differ considerably.

The spectral function $I(\lambda)$ can be expressed in terms of the parameters of the continuum model. For this purpose we use the integral representation of the δ function together with the definition of the classical dissipation kernel $K(t)$, Eq. (2.21), to recast Eq. (3.17) in the form

$$I(\lambda)=-\frac{i}{2}\int_{-\infty}^{\infty}dt\, e^{i\lambda t}\int_0^t dt' K(t') \ .$$

Since $K(t)$ is an even function of time, the relaxation function $\Psi(t)=\int_0^t dt' K(t')$ is an odd function of time. As a result we obtain

$$I(\lambda)=\mathrm{Im}\left[\int_0^{\infty}dt\, e^{i\lambda t}\Psi(t)\right]=\frac{1}{\lambda}\mathrm{Re}[\hat{K}(i\lambda)] \tag{3.18}$$

where Re and Im denote the real and imaginary part of the function, respectively. The expression for the $\hat{K}(p)$ function in terms of the parameters of the continuum model has been given in Sec. II [cf. Eq. (2.22)]. It now remains to express the relevant physical quantities (Fourier transform of the transition probability, the average energy loss, etc.) in terms of the spectral function $I(\lambda)$.

From Eq. (3.11) it follows that

$$R(\tau)=\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}d\lambda\frac{I(\lambda)f(\lambda)[\cosh(\beta\hbar\lambda/2)-\cos(\tau\beta\hbar\lambda)]}{\sinh(\beta\hbar\lambda/2)}$$
$$=\frac{\beta}{2\pi}\int_{-\infty}^{\infty}d\lambda\frac{\mathrm{Re}[\hat{K}(i\lambda)]f(\lambda)[\cosh(\beta\hbar\lambda/2)-\cos(\tau\beta\hbar\lambda)]}{(\beta\hbar\lambda)\sinh(\beta\hbar\lambda/2)} \quad (3.19)$$

where

$$f(\lambda)\equiv\left|\int_{-\infty}^{\infty}dt\, e^{i\lambda t}F(t)\right|^2=f(-\lambda)\ . \quad (3.19a)$$

In a similar fashion one derives results for the average energy loss of the unstable mode δ and for the second- (dispersion) and the third-order cumulants [cf. Eqs. (3.14)–(3.16)]

$$\delta=\frac{\beta}{2\pi}\int_{-\infty}^{\infty}d\lambda\,\mathrm{Re}[\hat{K}(i\lambda)]f(\lambda)\ , \quad (3.20a)$$

$$\langle\delta^2\varepsilon\rangle=\frac{\beta}{2\pi}\int_{-\infty}^{\infty}d\lambda(\beta\hbar\lambda)\coth(\beta\hbar\lambda/2)\mathrm{Re}[\hat{K}(i\lambda)]f(\lambda)\ , \quad (3.20b)$$

$$\langle\delta^3\varepsilon\rangle=\frac{\beta}{2\pi}\int_{-\infty}^{\infty}d\lambda(\beta\hbar\lambda)^2\mathrm{Re}[\hat{K}(i\lambda)]f(\lambda)\ . \quad (3.20c)$$

These results, combined with the expression for the Laplace transform of the classical dissipation kernel, Eq. (2.22), constitute the solution of the problem. It should be pointed out that the classical dissipation kernel depends only on the barrier frequency and the friction function. The detailed form of the potential enters the final results via the Fourier transform of the zero-order force, Eq. (3.19a). The force $F(t)$ depends explicitly upon the classical asymptotic trajectory. For certain potentials (e.g., the cubic potential to be studied in Sec. VI) the analytic form of this trajectory is known, and a closed expression for the $f(\lambda)$ function can be derived.

The third-order cumulant $\langle\delta^3\varepsilon\rangle$ is a measure of the skewedness (asymmetry) of the quantum transition probability kernel. This skewedness is a quantum effect. The classical transition probability is symmetric. To emphasize the quantum nature of the skewedness we shall write it in the form

$$\langle\delta^3\varepsilon\rangle=(\beta\hbar\lambda_0)^2\frac{\beta}{2\pi}\int_{-\infty}^{\infty}d\lambda(\lambda/\lambda_0)^2\mathrm{Re}[\hat{K}(i\lambda)]f(\lambda) \quad (3.21)$$

where λ_0 is a frequency scale, which we shall take to be the unstable normal mode frequency. The skewedness is closely related to the lowest-order quantum correction to the dispersion $\langle\delta^2\varepsilon\rangle$:

$$\langle\delta^2\varepsilon\rangle\simeq2\delta+\tfrac{1}{6}\langle\delta^3\varepsilon\rangle\ . \quad (3.22)$$

In a similar manner we obtain the lowest-order quantum correction to the Fourier transform of the transition probability kernel

$$\tilde{P}(\tau-i/2)\simeq\tilde{P}_{\mathrm{cl}}(\tau-i/2)\exp\left[\frac{1}{12}\langle\delta^3\varepsilon\rangle(\tau^2+\tfrac{1}{4})^2\right] \quad (3.23)$$

where $\tilde{P}_{\mathrm{cl}}(\tau-i/2)$ is the classical Gaussian kernel:[27]

$$\tilde{P}_{\mathrm{cl}}(\tau-i/2)=\exp[-\delta(\tau^2+\tfrac{1}{4})]\ . \quad (3.24)$$

Explicit results for these quantities for the particular case of a cubic potential with Ohmic dissipation are presented in Sec. VI.

IV. THE QUANTUM-MECHANICAL ESCAPE RATE

The starting point for the evaluation of the quantum-mechanical expression for the escape rate is a kinetic equation for time-dependent energy distribution function of the unstable mode $\tilde{n}(\varepsilon;t)$. As already mentioned, this mode is treated (semi) classically unlike the stable modes, which are treated quantum mechanically. We denote by $\tilde{n}(\varepsilon;t)$ the nonstationary probability to find the system with a (dimensionless) energy between ε and $\varepsilon+d\varepsilon$ in the unstable mode at the barrier turning point. The kinetic equation for $\tilde{n}(\varepsilon;t)$ is

$$\frac{d\tilde{n}(\varepsilon;t)}{dt}=\int_{-\infty}^{\infty}d\varepsilon'[w(\varepsilon;\varepsilon')R(\varepsilon')\tilde{n}(\varepsilon';t)-w(\varepsilon';\varepsilon)R(\varepsilon)\tilde{n}(\varepsilon;t)]-\frac{T(\varepsilon)}{2\pi\hbar\beta}\tilde{n}(\varepsilon;t)\ . \quad (4.1)$$

In this equation $w(\varepsilon;\varepsilon')$ is the transition rate between the states with energy ε' and ε. It is related to the transition probability $P(\varepsilon;\varepsilon')$ determined in Sec. III via $w(\varepsilon;\varepsilon')=(1/2\pi\hbar\beta)P(\varepsilon;\varepsilon')$. The transition rates satisfy the detailed balance condition. $R(\varepsilon)$ is the quantum-mechanical reflection coefficient for the parabolic barrier of the unstable mode

$$R(\varepsilon)=1-T(\varepsilon)=[1+\exp(\alpha\varepsilon)]^{-1} \quad (4.2)$$

with

$$\alpha\equiv2\pi/\beta\hbar\lambda_0\equiv\tilde{\omega}_1/\lambda_0$$

where $\tilde{\omega}_1$ is the first Matsubara frequency.

A particle at the barrier turning point may either be transmitted through the barrier with a rate $T(\varepsilon)/2\pi\hbar\beta$ or it may be reflected. The transmission is taken into account by the sink term in Eq. (4.1). The reflected particle undergoes an energy redistribution process which is described by the integral term in the kinetic equation. This term is simply the continuum form of the Pauli master equation and implies physically that the phase of the particle is destroyed during the excursion in the well. The use of a continuous energy which implies the neglect of energy quantization is consistent with the (semi) classical treatment of the unstable mode. Note that $(1/2\pi\hbar\beta)\tilde{n}(\varepsilon;t)\equiv\tilde{f}(\varepsilon;t)$ is the flux of particles per unit (dimensionless) energy incident on the barrier and, once again, implies semiclassical treatment of the unstable mode. The stationary energy distribution function for the unstable mode $n(\varepsilon)$ is given by the steady-state solution of the kinetic equation, Eq. (4.1):

$$n(\varepsilon)=\int_{-\infty}^{\infty} d\varepsilon' P(\varepsilon;\varepsilon')R(\varepsilon')n(\varepsilon') \ . \tag{4.3}$$

This integral equation is *formally* identical to the one suggested by Mel'nikov.[27] However, the physical meaning as well as the method of calculation of the transition probability $P(\varepsilon;\varepsilon')$ is different. Equation (4.3) refers to the *unstable mode,* while the Mel'nikov original equation was for the *system coordinate.* This has a number of implications, both qualitative and quantitative.

(i) The most important qualitative aspect is that in Mel'nikov's original theory[27] the use of the quantum-mechanical reflection (transmission) coefficient, Eq. (4.2) for the system coordinate is an approximation which neglects the effect of dissipation on the tunneling.[28] How good this approximation is is not clear *a priori.* In this treatment we are dealing with the dynamics of the unstable mode. Consequently, these coefficients are *exact* as long as the harmonic approximation for the barrier is valid.[21] In other words, we neglect the nonlinearity of the barrier but we account for the effect of the thermal bath upon the tunneling. From this discussion, it is clear that the treatment will break down when deviations from linearity become important. This is the case for temperatures substantially below the crossover temperature T_0, between tunneling and thermal activation.[16,17] However at such low temperatures the escape is dominated by tunneling close to the bottom of the well, where the deviation of the energy distribution function from the equilibrium one (depopulation) is not important.

(ii) The most important quantitative difference is that the barrier frequency for the unstable mode λ_0 may be substantially smaller than the bare barrier frequency for the system coordinate ω_b.[41] It is λ_0 which appears in the expression for the reflection (transmission) coefficient of the barrier, Eq. (4.2). This leads to reduction of the escape rate and tends to make the escape dynamics more classical.

(iii) Finally, one has to determine the transition probability $P(\varepsilon;\varepsilon')$. Mel'nikov[27] has employed a Gaussian kernel, which is appropriate only in the classical limit.[30] In Section III we derived an exact expression for the quantum transition probability kernel.

The quantum-mechanical escape rate is given by the steady-state total probability flux of particles out of the well:

$$\Gamma=\frac{1}{2\pi\hbar\beta}\int_{-\infty}^{\infty} d\varepsilon\, T(\varepsilon)n(\varepsilon)=\int_{-\infty}^{\infty} d\varepsilon\, T(\varepsilon)f(\varepsilon) \ . \tag{4.4}$$

Determination of the rate reduces to solving the integral equation, Eq. (4.3), for $n(\varepsilon)$. The appropriate boundary condition is obtained by noting that for energies below the barrier top the energy distribution function $n(\varepsilon)$ approaches the equilibrium one:

$$n_{\mathrm{eq}}(\varepsilon)=\frac{Z^*(\{y_i\})}{Z_0}\exp(-\beta V_0-\varepsilon)\equiv C\exp(-\varepsilon) \ . \tag{4.5}$$

Z_0 is the canonical quantum partition function for the particles in the well. It is evaluated by replacing the Hamiltonian of the system, Eq. (2.3), by a harmonic one near the bottom of the well. Diagonalization of the harmonic Hamiltonian leads to a familiar expression for the partition function:

$$Z_0=[2\sinh(\beta\hbar\Lambda_0/2)]^{-1}\prod_{i=1}^{N}[2\sinh(\beta\hbar\Lambda_i/2)]^{-1} \tag{4.6}$$

where Λ_i $(i=0,1,2,\ldots,N)$ are the normal mode frequencies near the bottom of the well. The partition function $Z^*(\{y_i\})$ is associated with the stable barrier modes and is given by an analogous expression

$$Z^*(\{y_i\})=\prod_{i=1}^{N}[2\sinh(\beta\hbar\lambda_i/2)]^{-1} \ . \tag{4.7}$$

The normalization constant C defined in Eq. (4.5) can be recast in the form

$$C=\frac{2\Omega}{\omega_b}\sin(\beta\hbar\lambda_0/2)\Xi\exp(-\beta V_0) \tag{4.8}$$

where Ξ is defined as

$$\Xi\equiv\frac{\omega_b}{\Omega}\frac{\sinh(\beta\hbar\Lambda_0/2)}{\sin(\beta\hbar\lambda_0/2)}\prod_{i=1}^{N}\frac{\sinh(\beta\hbar\Lambda_i/2)}{\sinh(\beta\hbar\lambda_i/2)} \ . \tag{4.9}$$

This factor can be rewritten in an equivalent form[19,20(b)] in terms of the friction function $\hat{\gamma}(p)$:

$$\Xi=\prod_{n=1}^{\infty}\frac{[\Omega^2+\tilde{\omega}_n^2+\tilde{\omega}_n\hat{\gamma}(\tilde{\omega}_n)]}{[-\omega_b^2+\tilde{\omega}_n^2+\tilde{\omega}_n\hat{\gamma}(\tilde{\omega}_n)]} \tag{4.10}$$

with $\tilde{\omega}_n\equiv 2\pi n/\beta\hbar$ being the Matsubara frequencies.

The integral equation, Eq. (4.3), has been solved by Mel'nikov[27] for the particular case of the Gaussian kernel and by Larkin and Ovchinnikov[30] in the general case via application of the Wiener-Hopf method. An alternative solution is given in the appendix. As already emphasized, an important point in the solution[27] is that the transition probability kernel $P(\varepsilon;\varepsilon')$ is a function of the energy difference $(\varepsilon-\varepsilon')$ only, so that Eq. (4.3) is a convolution-type integral equation. The final result for the quantum-mechanical escape rate Γ has the form

$$\Gamma=\frac{C\lambda_0}{4\pi\sin(\beta\hbar\lambda_0/2)}\times\exp\left[\frac{\beta\hbar\lambda_0\sin(\beta\hbar\lambda_0/2)}{2\pi}\times\int_{-\infty}^{\infty}d\tau\frac{\ln[1-\tilde{P}(\tau-i/2)]}{\cosh(\tau\beta\hbar\lambda_0)-\cos(\beta\hbar\lambda_0/2)}\right] . \tag{4.11}$$

A similar result employing the classical Gaussian transition probability kernel, (3.24), has been derived independently by Hänggi, Talkner, and Borkovec.[46] The rate expression can be recast in the more illuminating form

$$\Gamma=\frac{\Omega}{2\pi}\frac{\lambda_0}{\omega_b}\Xi\Upsilon\exp(-\beta V_0) \tag{4.12}$$

where Υ is the quantum-mechanical depopulation factor:[30]

$$\Upsilon \equiv \exp\left[\frac{\beta\hbar\lambda_0 \sin(\beta\hbar\lambda_0/2)}{2\pi} \times \int_{-\infty}^{\infty} d\tau \frac{\ln[1-\tilde{P}(\tau-i/2)]}{\cosh(\tau\beta\hbar\lambda_0)-\cos(\beta\hbar\lambda_0/2)}\right] . \quad (4.13)$$

The quantum-mechanical rate expression, Eq. (4.12), is the central result of this section. It is valid in what may be called the activated tunneling regime, which corresponds to temperatures above the crossover temperature T_0.[16,17] In this regime the escape is dominated by the energy region close to the top of the potential barrier. This accounts for the appearance of the Arrhenius factor in Eq. (4.12).

The final expression for the quantum escape rate factorizes into a product of a few terms. The first term is the classical TST result in the limit of vanishing friction. The second term is the Grote-Hynes[41] factor, which is a generalization of the intermediate friction Kramers result[1] to the case of non-Markovian dissipation. The third term is the equilibrium quantum correction and accounts for the quantum-mechanical transparency of the barrier. It has been derived by Wolynes[15] and others.[16–19,20(b)] Finally, the fourth term in the expression for the rate accounts for the depopulation due to the weak interaction of the unstable mode with the stable ones and leads to a substantial reduction of the rate in this limit. This term approaches unity exponentially fast with increasing interaction between the unstable mode and the stable normal modes.

Summarizing, the rate expression, Eq. (4.12), reduces to the known results in appropriate limits and contains them as particular cases. In this sense it constitutes a solution of the quantum Kramers turnover problem for temperatures $T > T_0$. Extension to temperatures below the crossover is possible and is outlined in Sec. VII. In Sec. V we shall use the explicit expression for the escape rate to determine the leading quantum correction to the classical result.

V. QUANTUM-MECHANICAL CORRECTION TO THE DEPOPULATION FACTOR

In this section we determine the first-order quantum-mechanical correction to the depopulation factor Υ. Qualitatively quantum mechanics affects the depopulation factor by modifying both the well and the barrier dynamics.

(i) Modification of the well dynamics expresses itself in the broadening and skewing of the transition probability kernel, $P(\varepsilon;\varepsilon')$, compared with the classical one.

(ii) The barrier dynamics is modified by the quantum tunneling through the barrier. In the parabolic barrier approximation it is taken into account via the transmission (reflection) coefficients for the unstable mode barrier.

It should be pointed out that quantum corrections to the rate enter also via the *equilibrium* factor Ξ and have been extensively studied by Wolynes[15] and others.[16–19] We shall focus our attention on quantum corrections to the depopulation factor, which reflects the *nonequilibrium* well dynamics. The quantum effects will be analyzed by employing the high-temperature expansion for the depopulation factor Υ in terms of the dimensionless parameter $\theta_0 \equiv \beta\hbar\lambda_0$. The general expression for Υ, Eq. (4.13), can be written as

$$\Upsilon = \exp\left[\int_{-\infty}^{\infty} d\tau f_1(\theta_0;\tau) f_2(\theta_0;\tau)\right] \equiv \exp[Q(\theta_0)] \quad (5.1)$$

where

$$f_1(\theta_0;\tau) = \ln[1-\tilde{P}(\tau-i/2)] , \quad (5.2a)$$

$$f_2(\theta_0;\tau) = \frac{1}{2\pi} \frac{\theta_0 \sin(\theta_0/2)}{\cosh(\tau\theta_0)-\cos(\theta_0/2)} . \quad (5.2b)$$

The classical limit corresponds to $\theta_0 \to 0$. The leading quantum correction is obtained by keeping terms up to order θ_0^2 in the expansion. Straightforward manipulations give

$$\Upsilon \simeq \Upsilon_{cl}\{1+\tfrac{1}{2}[Q_1''(0)+Q_2''(0)]\theta_0^2 + O(\theta_0^4)\} . \quad (5.3)$$

Here Υ_{cl} is the classical depopulation factor, Eq. (2.24), while

$$Q_1''(0) = -\frac{1}{12\pi}\int_{-\infty}^{\infty} d\tau \ln[1-\tilde{P}_{cl}(\tau-i/2)] \quad (5.4a)$$

and

$$Q_2''(0) = -\frac{G}{12\pi}\int_{-\infty}^{\infty} d\tau (\tau^2+\tfrac{1}{4}) \frac{\tilde{P}_{cl}(\tau-i/2)}{1-\tilde{P}_{cl}(\tau-i/2)} . \quad (5.4b)$$

$\tilde{P}_{cl}(\tau-i/2)$ is the Fourier transform of the classical transition probability, Eq. (3.24), and the parameter G is defined as

$$G \equiv \frac{\langle \delta^3 \varepsilon \rangle}{\theta_0^2} = \frac{\beta}{2\pi}\int_{-\infty}^{\infty} d\lambda (\lambda/\lambda_0)^2 \mathrm{Re}[\hat{K}(i\lambda)] f(\lambda) . \quad (5.5)$$

The first term $Q_1''(0)$ gives the tunneling correction to the depopulation factor and is *positive*. The second term $Q_2''(0)$ is associated with the quantum correction to the transition probability kernel and is *negative*. It follows that the transparency of the barrier and the quantum modification of the well dynamics (broadening and skewedness of the transition probability kernel) have opposite effects on the escape rate. The former leads to an increase of the rate with respect to the classical value, while the latter leads to a reduction.

To evaluate the leading quantum correction explicitly we substitute the Fourier transform of the classical dissipation kernel, Eq. (3.24) into Eqs. (5.4a) and (5.4b) leading to

$$Q_1''(0) = \frac{1}{12(\pi\delta)^{1/2}} e^{-\delta/4} \Phi(e^{-\delta/4}, \tfrac{3}{2}, 1) \quad (5.6a)$$

and

$$Q_2''(0) = \frac{G}{48(\pi\delta)^{1/2}} e^{-\delta/4} \times \left[\Phi(e^{-\delta/4}, \tfrac{1}{2}, 1) + \frac{2}{\delta}\Phi(e^{-\delta/4}, \tfrac{3}{2}, 1)\right] \quad (5.6b)$$

The function $\Phi(z,s,v)$ is defined as (Ref. 47, Sec. 1.11)

$$\Phi(z,s,v)\equiv\sum_{n=0}^{\infty}(v+n)^{-s}z^n$$

The limit of vanishingly small dissipation energy ($\delta\ll 1$) can be studied using the expansion [Ref. 47, Form. 1.11(8)]

$$e^{-y}\Phi(e^{-y},s,1)=\Gamma(1-s)y^{s-1}+\sum_{n=0}^{\infty}\zeta(s-n)\frac{y^n}{n!}$$

with $\Gamma(s)$ and $\zeta(s)$ being the gamma function and the Riemann's zeta function, respectively. We find

$$Q''(0)\equiv Q_1''(0)+Q_2''(0)\simeq\frac{\zeta(\frac{3}{2})}{12\pi^{1/2}}(1-G/2\delta)\delta^{-1/2}-\tfrac{1}{12}+O(\delta^{1/2})\ . \qquad (5.7)$$

This result leads to two conclusions.

(i) The sign of the quantum correction in the weak dissipation limit $\delta<1$ depends upon the ratio $G/2\delta\equiv\langle\delta^3\varepsilon\rangle/2\delta\theta_0^2$. Using Eqs. (3.20a) and (3.21) we find that

$$\frac{G}{2\delta}=\frac{\int_{-\infty}^{\infty}d\lambda(\lambda/\lambda_0)^2\mathrm{Re}[\hat{K}(i\lambda)]f(\lambda)}{2\int_{-\infty}^{\infty}d\lambda\,\mathrm{Re}[\hat{K}(i\lambda)]f(\lambda)}\ . \qquad (5.8)$$

If the product $\mathrm{Re}[\hat{K}(i\lambda)]f(\lambda)$ decays sufficiently fast then the ratio is less than unity and the quantum correction leads to an increase of the depopulation factor. Note though that, as pointed out by Mel'nikov and Sütö[31] and by Griff *et al.*,[29] the first quantum correction to the depopulation factor can be a nonmonotonic function of the dissipation strength.

(ii) It follows from Eq. (5.7) that the quantum correction has a singular behavior in the limit of vanishing dissipation ($\delta\to 0$). This nonanalyticity shows that the simple perturbation expansion (5.3) is incorrect in this limit. The leading quantum correction to the depopulation factor in this limit is obtained by using the result of Sec. III for the Fourier transform of the transition probability kernel, Eqs. (3.23) and (3.24). In the limit of vanishing dissipation ($\delta\to 0$)

$$\ln[1-\tilde{P}(\tau-i/2)]\simeq\ln(\delta)+h(\delta;\tau)\ , \qquad (5.9)$$

where $h(\delta;\tau)$ is analytic for $\delta\to 0$. Using the tabular integral

$$\frac{\theta_0\sin(\theta_0/2)}{2\pi}\int_{-\infty}^{\infty}d\tau[\cosh(\tau\theta_0)-\cos(\theta_0/2)]^{-1}=1-\frac{\theta_0}{2\pi}$$

one finds for the leading term in the depopulation factor a result obtained previously in Ref. 48.

$$\Upsilon\simeq\delta^{1-\theta_0/2\pi}\ . \qquad (5.10)$$

As noted by Mel'nikov[27] and others[29,30,46] in this limit the leading quantum correction is of the order $\hbar$ rather than $\sim\hbar^2$. It follows from Eq. (5.10) that the overall effect is an increase of the depopulation factor relative to its classical value.

It is interesting to compare this result with previous treatments of the quantum Kramers problem in the extremely underdamped limit[27–30]

$$\Upsilon_M\simeq\delta^{1-\beta\hbar\omega_b/2\pi} \qquad (5.11)$$

The difference between this result and Eq. (5.10) is that in the latter the *renormalized* frequency of the barrier replaces the bare one. Since $\lambda_0\le\omega_b$ it implies that the value of the depopulation factor is *smaller* than predicted by the previous theories. This is not surprising. As already mentioned in Sec. IV the unstable mode barrier is thicker than that for the system coordinate leading to a more classical behavior. It is also important to stress that Eq. (5.10) is valid in the limit $\delta\ll 1$. For the non-Markovian dissipation this limit may be realized also when the damping strength is very large.[35,38] In this case the prediction of Eqs. (5.10) and (5.11) will differ considerably.

VI. THE CUBIC POTENTIAL WITH OHMIC DISSIPATION

A. Preliminaries

As a practical application we consider in this section the case of the cubic potential with Ohmic (Markovian) dissipation. This choice is motivated by its extensive use as an approximation to the tilted cosine (washboard) potential for the RSJ with a biasing current close to the critical value.[7,8] It also has a few simplifying technical features, e.g., analyticity of the potential and the fact that a closed expression is known for the asymptotic trajectory.

We limit ourselves to the case of the Ohmic (Markovian) dissipation $\gamma(t)=2\gamma\delta(t)$ since it suffices for illustration of the main physical effects while avoiding unnecessary technical complications. This choice also allows us to make a direct comparison with the theory of Mel'nikov and Meshkov[34] in the classical limit, and Larkin and Ovchinnikov[30] in the quantum case.

The cubic potential (cf. Fig. 1) has the form

$$V(q)=-\tfrac{1}{2}M\omega_b^2q^2(1+q/q_0)$$
$$=-\tfrac{27}{4}V_0(q/q_0)^2(1+q/q_0) \qquad (6.1)$$

where $V_0=\frac{2}{27}M\omega_b^2q_0^2$ is the barrier height, and the origin of the coordinates is at the top of the barrier. The bare barrier frequency is ω_b and $-q_0$ is the coordinate of the second turning point. The asymptotic trajectory that starts at $t=-\infty$ at the barrier top, reaches the turning point $-q_0$ at $t=0$, and returns to the barrier top at $t=\infty$ is well known:

$$q_{\mathrm{as}}(t)\equiv q(t)=-q_0\mathrm{sech}^2(\omega_bt/2)\ . \qquad (6.2)$$

From the general discussion in Sec. II it follows that the zeroth-order equation of motion for the unstable mode can be derived from a Hamiltonian with the effective potential $V_{\mathrm{eff}}(\rho)$,

$$V_{\mathrm{eff}}(\rho)=-\tfrac{1}{2}\lambda_0^2\rho^2(1+\rho/\rho_0)\ . \qquad (6.3)$$

Here

$$\rho_0=\frac{\lambda_0^2}{\omega_b^2 u_{oo}^3}M^{1/2}q_0 \tag{6.4}$$

and u_{oo} is the projection of the system coordinate on the unstable normal mode (cf. Sec. II). The time-dependent zero-order force $F(t)$ acting on the unstable mode is given by [cf. Eqs. (2.18) and (6.3)]

$$F(t)=\tfrac{3}{2}\lambda_0^2[\rho^2(t)/\rho_0] \tag{6.5}$$

with $\rho(t)$ being the asymptotic trajectory for the effective potential. Since $V_{\rm eff}(\rho)$ is also a cubic potential the asymptotic trajectory for the unstable mode is given by

$$\rho_{\rm as}(t)\equiv\rho(t)=-\rho_0 {\rm sech}^2(\lambda_0 t/2) . \tag{6.6}$$

A closed analytic expression for the Fourier transform of the force can be derived leading to

$$f(\lambda)=216\pi^2\left[\frac{\lambda_0}{u_{oo}\omega_b}\right]^6\left[\frac{y(y^2+1)}{\sinh(\pi y)}\right]^2 V_0 \tag{6.7}$$

where $y\equiv\lambda/\lambda_0$.

We shall first consider the classical limit. The transition probability kernel $P_{\rm cl}(\varepsilon;\varepsilon')$ in this case is Gaussian and the only physical quantity that has to be evaluated is the average dissipated energy δ [cf. Eq. (2.20)]. It should be emphasized that the classical dissipation kernel $K(t)$ is nonlocal in time even in the case of Ohmic dissipation. Furthermore, $K(t)$ has a cusp at $t=0$. The Ohmic dissipation kernel is given by

$$K_0(t)=\frac{1}{2}\left[\frac{\lambda_1}{\lambda_0}e^{-\lambda_1|t|}-e^{-\lambda_0|t|}\right] . \tag{6.8}$$

The barrier frequency for the unstable mode λ_0 is related to the damping parameter γ by

$$\lambda_0=\left[\frac{\gamma^2}{4}+\omega_b^2\right]^{1/2}-\frac{\gamma}{2} \tag{6.9a}$$

and

$$\lambda_1=\left[\frac{\gamma^2}{4}+\omega_b^2\right]^{1/2}+\frac{\gamma}{2} . \tag{6.9b}$$

For future use we write an explicit expression for the ratio $\mu\equiv\lambda_1/\lambda_0$:

$$\mu\equiv\lambda_1/\lambda_0=1+\frac{\gamma^2}{2\omega_b^2}[1+(1+4\omega_b^2/\gamma^2)^{1/2}] . \tag{6.10}$$

In the extremely underdamped and the overdamped limits this expression reduces to

$$\mu\simeq 1+\gamma/\omega_b \quad (\gamma\ll\omega_b) \tag{6.10a}$$

and

$$\mu\simeq\frac{\gamma^2}{\omega_b^2}\{1+2\omega_b^2/\gamma^2\} \quad (\gamma\gg\omega_b) . \tag{6.10b}$$

The spectral function for the bath of stable modes $I(\lambda)$ is given in this case by

$$I(\lambda)=\frac{1}{\lambda}{\rm Re}[\hat{K}_0(i\lambda)]=\frac{\lambda}{2\lambda_0}\left[\frac{1}{\lambda^2+\lambda_0^2}-\frac{1}{\lambda^2+\lambda_1^2}\right] . \tag{6.11}$$

The projection of the system coordinate on the unstable normal mode u_{oo} is

$$u_{oo}^2=(1+\sigma)^{-1}=(1+\gamma/2\lambda_0)^{-1} . \tag{6.12}$$

Finally, the condition for the applicability of the perturbative approach[37] is $\sigma=\gamma/2\lambda_0<1$.

B. Cumulants of the energy loss

Substitution of Eqs. (6.7) and (6.11) into Eq. (3.20a) leads to the following result for the average energy loss of the unstable mode

$$\delta\equiv\delta(\mu)=\frac{27\pi}{4}(1+1/\mu)^3(\mu^2-1)M_4(\mu)\beta V_0$$
$$\equiv\Delta_1(\mu)\beta V_0 . \tag{6.13}$$

The integral $M_{2n}(\mu)$ is defined as

$$M_{2n}(\mu)\equiv\int_{-\infty}^{\infty}\frac{dy\, y^{2n}(y^2+1)}{(y^2+\mu^2)\sinh^2(\pi y)} \tag{6.14}$$

and satisfies the recursion relation

$$M_{2n+2}(\mu)=\frac{2}{\pi}(B_{2n+2}+B_{2n})-\mu^2 M_{2n}(\mu) \tag{6.15}$$

where B_{2n} is the Bernoulli number. This recursion relation allows us to evaluate $M_{2n}(\mu)$ easily provided that (say) $M_4(\mu)$ is known. Calculation of the integral $M_4(\mu)$ is an exercise in contour integration. The result is

$$M_4(\mu)=\frac{2}{\pi}\left[\frac{1}{5}-\frac{\mu^2}{6}+\mu^2(\mu^2-1)\left[\mu\psi'(\mu)-1-\frac{1}{2\mu}\right]\right] \tag{6.16}$$

where

$$\psi'(\mu)\equiv\frac{\partial^2\ln\Gamma(\mu)}{\partial\mu^2}=\sum_{n=0}^{\infty}\frac{1}{(\mu+n)^2}$$

is the so-called trigamma function (Ref. 49, Sec. 6.4). Analogously, the third-order cumulant of the energy loss of the unstable mode is expressed in terms of $M_6(\mu)$

$$\langle\delta^3\varepsilon\rangle=\frac{27\pi}{4}(1+1/\mu)^3(\mu^2-1)M_6(\mu)(\beta\hbar\lambda_0)^2\beta V_0$$
$$\equiv\Delta_3(\mu)(\beta\hbar\lambda_0)^2\beta V_0 \tag{6.17}$$

with

$$M_6(\mu)=\frac{4}{35\pi}-\mu^2 M_4(\mu) .$$

It is now possible to study the behavior of the average energy loss and of the third cumulant in the overdamped ($\mu\gg1$) and the extremely underdamped ($\mu\to1^+$) limits. Using the asymptotic expansion for the trigamma function $\psi'(\mu)$ [Ref. 49, Form 6.4(12)] we get

$$\Delta_1(\mu)\simeq\frac{27}{35}\left[1+\frac{3}{\mu}+\frac{1}{\mu^2}+\frac{1}{\mu^3}+O(1/\mu^4)\right], \quad (6.18a)$$

$$\Delta_3(\mu)\simeq\frac{27}{35}\left[1+\frac{3}{\mu}+O(1/\mu^2)\right]. \quad (6.18b)$$

In the extremely underdamped limit one has to employ the series expansion of $\psi'(\mu)$ around $\mu=1$ [Ref. 49, Form. 6.4(9)] leading to the results

$$\Delta_1(\mu)\simeq\frac{36(\mu-1)}{5}\{1+[60\zeta(2)-101](\mu-1)\} +O((\mu-1)^3), \quad (6.19a)$$

$$\Delta_3(\mu)\simeq\frac{36(\mu-1)}{7}\left[1-\left[42\zeta(2)-\frac{681}{5}\right](\mu-1)\right] +O((\mu-1)^3). \quad (6.19b)$$

It follows that the ratio $\langle\delta^3\varepsilon\rangle/\delta\theta_0^2$ is a slowly varying function of the damping increasing monotonically from $\frac{5}{7}$ to unity as the damping varies in the interval $0<\gamma<\infty$. From Eq. (5.7) we thus find that in the limit of weak dissipation the leading quantum correction is positive.

C. The classical limit

As already pointed out in the classical limit the present theory reduces to that of PGH.[37,38] The classical depopulation factor, Eq. (2.24) is only a function of the average energy loss δ. Results of the preceding subsection allow us as a byproduct to compare the results for the classical escape rate predicted by the theories of Grabert and co-workers[37,38] and Mel'nikov and Meshkov[34] (MM). In the particular case of Ohmic dissipation the rate expressions formally differ by the prescription for the evaluation of the average dissipated energy. The expression for the average energy loss used by MM is[27,50]

$$\delta_{\rm MM}\equiv\beta\langle\Delta E_{\rm MM}\rangle=\beta M\gamma\int_{-\infty}^{\infty}dt\,|\dot q(t)|^2=\frac{36}{5}\frac{(\mu-1)}{\mu^{1/2}}\beta V_0 \quad (6.20)$$

where $\dot q(t)$ is the asymptotic trajectory velocity for the system coordinate in the absence of dissipation. The ratio of the average energy loss of the unstable mode to the energy loss of the system coordinate $\delta_{\rm MM}$ is thus given by

$$\frac{\delta}{\delta_{\rm MM}}=\frac{15\pi(\mu+1)^4M_4(\mu)}{16\mu^{5/2}}. \quad (6.21)$$

In the extremely underdamped limit ($\mu\rightarrow 1$)

$$\frac{\delta}{\delta_{\rm MM}}\rightarrow 15\pi M_4(1)=1 \quad (6.21a)$$

and the two results coincide. In the overdamped limit ($\mu\gg 1$)

$$\frac{\delta}{\delta_{\rm MM}}\simeq\frac{15\pi}{16}\mu^{3/2}M_4(\mu)\simeq\frac{3}{28\mu^{1/2}}\rightarrow 0. \quad (6.21b)$$

This ratio of the average energy losses is exhibited in Fig. 2 for the whole range of values of the μ parameter. Inspection of the figure shows that the energy loss δ of the unstable mode is systematically smaller than that for the system coordinate. This result together with the limiting expressions for the classical depopulation factor,[27]

$$\Upsilon_{\rm cl}\simeq\delta[1+\zeta(1/2)(\delta/\pi)^{1/2}] \quad (\delta\ll 1),$$

$$\Upsilon_{\rm cl}\simeq 1-(4/\pi\delta)^{1/2}\exp(-\delta/4) \quad (\delta\geq 1),$$

enables a comparison of the classical escape rates predicted by the two methods. In the extremely underdamped limit ($\delta\ll 1$)

$$\frac{\Gamma_{\rm cl}}{\Gamma_{\rm MM}}=\frac{\Upsilon_{\rm cl}}{\Upsilon_{\rm cl}({\rm MM})}\simeq\frac{\delta}{\delta_{\rm MM}}\leq 1$$

so that the rate predicted by the PGH theory is smaller than the MM result. Furthermore, the turnover in the PGH theory is predicted at *larger* values of the damping.

D. The quantum limit

We now turn to the analysis of the quantum expression for the depopulation factor, Eq. (4.13). Combining the general expression for the Fourier transform of the dissipation kernel, Eqs. (3.11) and (3.19) with Eqs. (6.7), (6.11), and (6.13) we can recast it in the form

$$\tilde P(\tau-i/2)=\exp[-R(\tau)] \quad (6.22)$$

where

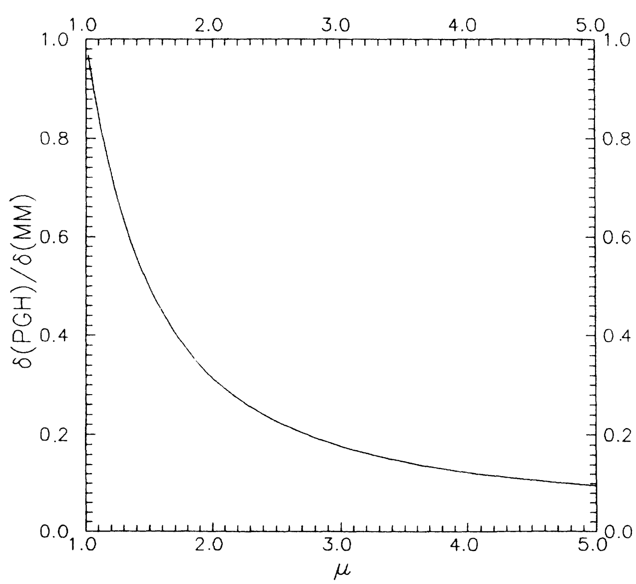

FIG. 2. The ratio of the average energy losses for the unstable mode δ and for the system coordinate $\delta_{\rm MM}$ as a function of damping strength. The results are for a model of a cubic potential with Ohmic (Markovian) dissipation.

$$R(\tau) \equiv R(\tau,\mu;\theta_0)$$
$$= \frac{27\pi}{4}\beta V_0(1+1/\mu)^3(\mu^2-1)I(\tau,\mu;\theta_0)$$
$$= \delta(\mu)\frac{I(\tau,\mu;\theta_0)}{M_4(\mu)} \quad (6.22a)$$

and the integral $I(\tau,\mu;\theta_0)$ is defined as

$$I(\tau;\theta_0,\mu) \equiv \int_0^\infty dy \frac{y^4(y^2+1)}{(y^2+\mu^2)\sinh^2(\pi y)} \times \frac{\cosh(\theta_0 y/2)-\cos(\tau\theta_0 y)}{(\theta_0 y/2)\sinh(\theta_0 y/2)} . \quad (6.22b)$$

As a result the expression for the quantum depopulation factor is given in the case of cubic potential with Ohmic dissipation by

$$\Upsilon_0 \equiv \Upsilon_0(\mu) = \exp\left[\frac{\theta_0\sin(\theta_0/2)}{2\pi}\int_{-\infty}^{\infty} d\tau \frac{\ln\{1-\exp[-\delta(\mu)I(\tau,\mu;\theta_0)/M_4(\mu)]\}}{\cosh(\tau\theta_0)-\cos(\theta_0/2)}\right] . \quad (6.23)$$

Its evaluation involves a double integration, which can be performed numerically using standard integration procedures [the integrands in Eqs. (6.22b) and (6.23) do not contain singularities].

In Fig. 3 we compare the quantum depopulation factor (solid line) with the classical one, Eq. (2.24) (dashed line), for a model potential with $\beta V_0=10$ and $\beta\hbar\omega_b=2.5$. The following observations are noteworthy.

(i) The quantum depopulation factor is substantially larger than the classical one.

(ii) Both curves are linear in the low damping domain. The classical slope is unity (as is well known from Kramers's original work[1]), while the quantum slope is substantially smaller and in close agreement with the theoretical prediction ~ 0.6 [cf. Eq. (5.10)].

(iii) The quantum and the classical factors converge at $\gamma/\omega_b \sim 0.1$. This demonstrates that a relatively weak dissipation is sufficient to suppress the quantum effects on the depopulation.

In addition to comparing the depopulation factor it is of interest to compare the effect of the quantum well dynamics as reflected in the transition probability kernel. The dash-dotted line in Fig. 3 is obtained by replacing the quantum transition probability kernel, Eq. (6.22), with the classical one $\tilde{P}_{\rm cl}(\tau-i/2)$ [cf. Eq. (3.24)] in the expression for the quantum depopulation factor, Eq. (6.23).[46] We find that this "quasiclassical" approximation is quite reasonable. It is notable though that the quantum result is somewhat lower, reflecting he skewedness of the kernel which dominates over the quantum broadening, leading to a net reduction.

The quantum Kramers turnover for the cubic potential with Ohmic dissipation is presented in Fig. 4, on a logarithmic scale. The dashed line is the classical result, derived from PGH theory. The solid lines denoted (a) and (b) are for the values of $\beta\hbar\omega_b=2.5$ and 1.0, respectively. The dimensionless barrier height is $\beta V_0=10$. The quantum enhancement of the rate is evident, although it doesn't take too high a temperature to substantially suppress the quantum effects. The turnover occurs at nearly the same values of the damping in all cases. A blown up view of the turnover region is shown in Fig. 5. We find that the quantum turnover is at values of the damping parameter which are only slightly lower than the classical.

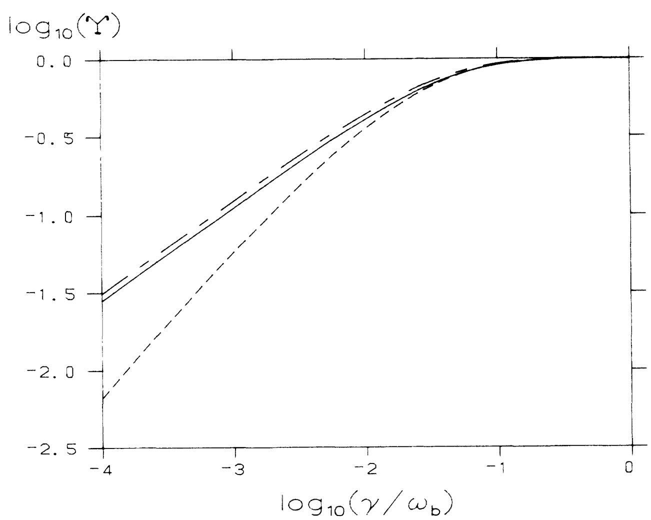

FIG. 3. The depopulation factor Υ as a function of the damping parameter for the same model with $\beta V_0=10$. The solid line represents the quantum result for $\beta\hbar\omega_b=2.5$. The dashed line represents the classical result. The dash-dotted line corresponds to using the classical transition probability kernel in the quantum expression for the depopulation factor.

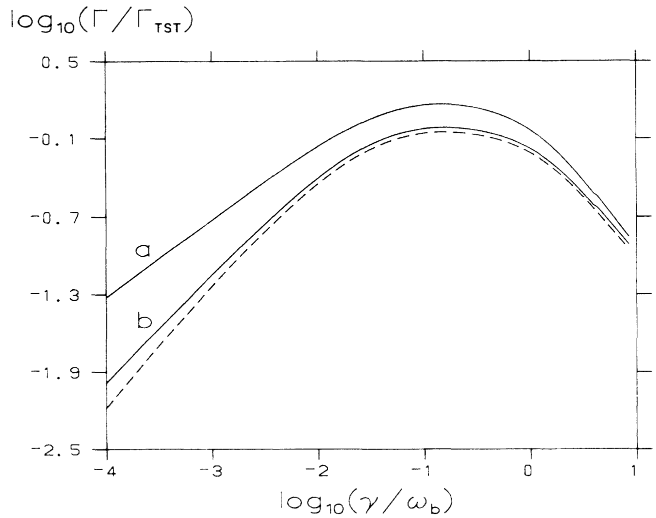

FIG. 4. The Kramers turnover on a log-log scale. The rates are normalized by the classical TST value in the limit of vanishing friction: $\Gamma_{\rm TST} \equiv (\Omega/2\pi)\exp(-\beta V_0)$. The dashed line represents the classical escape rate. Curve a is the quantum escape rate for $\beta\hbar\omega_b=2.5$; curve b is the same for $\beta\hbar\omega_b=1.0$. The (dimensionless) barrier height for all curves is $\beta V_0=10$.

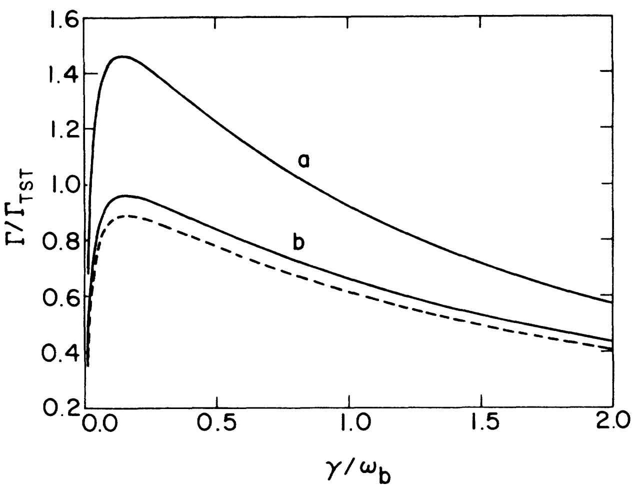

FIG. 5. A magnified plot of the turnover region on a linear scale. The input data are the same as for Fig. 4.

VII. DISCUSSION

A new expression has been derived for the quantum-mechanical decay rate of a metastable state. The result is general, applicable for the whole range of damping strengths and to non-Markovian dissipation. The derivation is based on the following new results and techniques.

1. The well dynamics leading to the redistribution of energy in the unstable normal mode have been treated systematically. The transition probability kernel was derived from the quantum forced oscillator solution for the first-order perturbation theory equations of motion for the stable modes.

2. The quantum transition probability kernel was shown to be broadened and skewed compared to the classical one. Cumulants of the energy loss for the unstable normal mode have been explicitly evaluated. The classical and the quantum average energy loss are identical. The quantum dispersion is larger than the classical. The third-order cumulant is negative.

3. The continuum limit of the kernel was obtained by defining a new spectral function for the bath of stable normal modes. This function differs qualitatively from the standard spectral density function $J(\omega)$ since it depends also on the specific form of the potential of the system.

4. The quantum rate expression was shown to factorize into a product of four terms. The nonequilibrium well and barrier dynamics is reflected by the quantum depopulation factor. The other three factors have been derived previously. The rate expression reduces to the known results for the escape rate, in the appropriate limits.

5. The depopulation factor is dependent on the barrier dynamics as well. In the presence of dissipation, the unstable mode barrier is broader (more classical) than the system coordinate barrier. As a result tunneling is hampered and the depopulation factor is smaller[48] than predicted by previous theories,[27,28,30] in which the influence of dissipation on the tunneling was neglected.

6. The first-order quantum correction to the depopulation factor was shown to have two main contributions. Tunneling as well as the quantum broadening of the transition probability kernel lead to an increase. On the other hand, the skewedness leads to a preference for transitions to lower energies, which causes a reduction of the depopulation factor.

7. An application of the theory has been illustrated on the cubic potential with Ohmic dissipation. In this case, the quantum depopulation factor is larger than the classical one. The quantum turnover occurs at lower values of the damping.

The theory presented in this paper is based on a number of assumptions and approximations which we list below.

A. The metastability assumption $\beta V_0 \gg 1$. This requirement is a standard one and assures exponential decay kinetics (so that the escape rate is well defined) and thermal equilibrium at unstable mode energies sufficiently lower than the barrier energy.

B. Classical treatment of the unstable mode. Unlike the stable normal modes, which are handled quantum mechanically, this classical approximation ignores energy quantization in the unstable mode. Justification of this important simplification is provided in Secs. III and IV.

C. Perturbative treatment of the nonlinear part of the potential. Here we follow the procedure originally suggested by Grabert.[37] The small parameter is the deviation of the unstable mode from the system coordinate. Discussion of this approximation may be found in Ref. 38.

D. Description of the well dynamics in terms of the master equation. The only physical assumption which underlies the derivation of this equation from the *exact* generalized master equation is the complete loss of coherence (phase) during the particle excursion within the well. Since the period of (conservative) motion diverges for the energy at the barrier top weak coupling to the bath of stable modes will be sufficient to destroy (randomize) the phase completely. It is more general than the diffusion equation,[1,3,25,28] which is derived from the master equation by keeping the first two terms in the Kramers-Moyal expansion.[29]

E. Parabolic barrier approximation for the tunneling probability. Derivation of the quantum escape rate expression in Sec. IV is based on the assumption that the escape is dominated by tunneling close to the top of the barrier. This assumption justifies the use of the parabolic barrier approximation and is valid for temperatures greater than T_0. The rate expression is therefore valid above the crossover temperature.

F. Extrapolation of the lower integration limit in evaluation of the flux to $-\infty$. The usual justification[27] of this procedure is based on the metastability condition $\beta V_0 \gg 1$. For the transmission coefficient of parabolic barrier this leads to the divergence of the rate expression at T_0. The divergence is therefore not a physical one.

A number of possible extensions and applications of the theory seem possible.

(i) The restriction to temperatures above T_0 may be removed. For temperatures slightly below T_0 one can employ the Affleck method.[22] Expansion of the action in the transmission coefficient to the second order in energy removes, as is well known,[16,17,23] the divergence in the Wolynes equilibrium factor Ξ. It will also modify the steady-state energy distribution function as derived from the integral equation. At very low temperatures the well dynamics is irrelevant, since the escape occurs in an energy region deep within the well, where thermal equilibrium prevails. The depopulation factor is consequently equal to unity, and one remains with the low-temperature equilibrium results for the rate.[16,17]

(ii) The example treated explicitly was limited to Ohmic dissipation. The theory has yet to be applied to a non-Markovian bath. Investigation of the validity domain of the multidimensional TST limit should prove interesting, especially for the strong damping limit where the perturbation expansion is still justified.

(iii) The theory is readily extended to a double well potential following a procedure suggested by Mel'nikov.[27]

(iv) The results of this paper are based on the lowest-order perturbation theory equations of motion for the normal modes. Systematic improvement is possible in principle, by keeping higher-order terms in the expansion of the nonlinear part of the potential. As an example, one can use the first-order equation of motion for the unstable mode, and the second-order equation of motion for the stable ones, retaining however the assumption of decoupled stable modes (keeping only the diagonal terms in the expansion).

(v) Present experimental techniques allow control of the damping in measurement of the decay rate of the zero voltage state in current biased RSJ.[10] The Kramers turnover has been reportedly observed recently.[10(a)] We believe that the present theory will be of use in analysis and interpretation of these results.

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APPENDIX: SOLUTION OF THE INTEGRAL EQUATION

In this appendix we present a solution of the integral equation, Eq. (4.3). Defining the function $N(\varepsilon)\equiv R(\varepsilon)n(\varepsilon)$ the integral equation can be written in the form

$$[1+\exp(\alpha\varepsilon)]N(\varepsilon)=\int_{-\infty}^{\infty} d\varepsilon' P(\varepsilon-\varepsilon')N(\varepsilon') . \quad \text{(A1)}$$

The two-sided Laplace transformation (Fourier transformation with imaginary argument)

$$\tilde{N}(is)=\int_{-\infty}^{\infty} d\varepsilon\, N(\varepsilon)\exp(-s\varepsilon) , \quad \text{(A2a)}$$

$$N(\varepsilon)=\frac{1}{2\pi i}\int_{z-i\infty}^{z+i\infty} ds\, \tilde{N}(is)\exp(s\varepsilon) \quad \text{(A2b)}$$

allows us to recast Eq. (A1) as

$$\tilde{N}[i(s-\alpha)]=-[1-\tilde{P}(is)]\tilde{N}(is) . \quad \text{(A3)}$$

The boundary condition, Eq. (4.5),

$$n(\varepsilon)\simeq C\exp(-\varepsilon) \quad (\varepsilon\to-\infty)$$

implies that the function $\tilde{N}(is)$ has a simple pole at $s=-1$:

$$\tilde{N}(is)=-\frac{C}{s+1} \quad (s\to-1) . \quad \text{(A4)}$$

We look for a solution of Eq. (A3) in the form of a product: $\tilde{N}(is)=\tilde{N}_1(is)\tilde{N}_2(is)$ with

$$\begin{aligned}\tilde{N}_1[i(s-\alpha)]&=[1-\tilde{P}(is)]\tilde{N}_1(is) ,\\ \tilde{N}_2[i(s-\alpha)]&=-\tilde{N}_2(is) .\end{aligned} \quad \text{(A5)}$$

Without loss of generality one can also assume that

$$\begin{aligned}\tilde{N}_1(is)&=1 \quad (s\to-1)\\ \tilde{N}_2(is)&=-\frac{C}{s+1} \quad (s\to-1) .\end{aligned} \quad \text{(A6)}$$

The function $\tilde{N}_2(is)$ is determined from the conditions (A5) and (A6)

$$\tilde{N}_2(is)=-\frac{\pi C/\alpha}{\sin[\pi(s+1)/\alpha]} . \quad \text{(A7)}$$

Introducing $\tilde{g}(is)\equiv\ln\tilde{N}_1(is)$ we get from Eq. (A5)

$$\tilde{g}[i(s-\alpha)]-\tilde{g}(is)=\ln[1-\tilde{P}(is)]\equiv\tilde{h}(is) \quad \text{(A8)}$$

which is equivalent to

$$g(x)=\frac{h(x)}{\exp(\alpha x)-1} . \quad \text{(A9)}$$

It follows that

$$\tilde{g}(is)=\int_{-\infty}^{\infty} dx\frac{(e^{-sx}-e^{x})}{e^{\alpha x}-1}\frac{1}{2\pi i}\int_{z-i\infty}^{z+i\infty} dy\, e^{yx}\tilde{h}(iy) . \quad \text{(A10)}$$

The term e^{x} has been introduced to satisfy the condition $\tilde{g}(-i)=0$ which follows from Eq. (A6). Interchanging the order of integration in Eq. (A10) and using the tabular integral

$$\int_{-\infty}^{\infty} dx\frac{\exp(-\sigma x)}{1-\exp(-x)}=\pi\cot(\pi\sigma) \quad (0<\sigma<1)$$

leads to

$$\begin{aligned}\tilde{g}(is)=\frac{1}{2i\alpha}\int_{z-i\infty}^{z+i\infty} dy\, \ln[1-\tilde{P}(iy)]\\ \times\{\cot[\pi(s-y)/\alpha]\\ +\cot[\pi(y+1)/\alpha]\} .\end{aligned} \quad \text{(A11)}$$

Combining Eqs. (A7) and (A11) we obtain

$$\tilde{N}(is)=-\frac{(\pi C/\alpha)}{\sin[\pi(s+1)/\alpha]}\exp\left[\frac{1}{2i\alpha}\int_{z-i\infty}^{z+i\infty}dy\,\ln[1-\tilde{P}(iy)]\{\cot[\pi(s-y)/\alpha]+\cot[\pi(y+1)/\alpha]\}\right] . \tag{A12}$$

The Fourier transform of the stationary energy distribution function $\tilde{n}(is)$ can now be found from

$$\tilde{n}(is)=\tilde{P}(is)\tilde{N}(is) .$$

Furthermore, the quantum-mechanical escape rate Γ of the particle is given by

$$\Gamma=\frac{1}{2\pi\beta\hbar}\int_{-\infty}^{\infty}d\varepsilon\,T(\varepsilon)n(\varepsilon)=\frac{1}{2\pi\beta\hbar}\tilde{N}(-i\alpha) \tag{A13}$$

which together with Eq. (A12) leads to

$$\Gamma=\frac{C}{2\beta\hbar\alpha\sin(\pi/\alpha)}\exp\left[\frac{i}{2\alpha}\int_{z-i\infty}^{z+i\infty}dy\ln[1-\tilde{P}(iy)]\{\cot(\pi y/\alpha)-\cot[\pi(y+1)/\alpha]\}\right] . \tag{A14}$$

The integration contour can be chosen from $-\frac{1}{2}-i\infty$ to $-\frac{1}{2}+i\infty$. Shifting the contour to the real axis by an appropriate variable change results in

$$\Gamma=\frac{C\lambda_0}{4\pi\sin(\beta\hbar\lambda_0/2)}\exp\left\{\frac{1}{2\alpha}\int_{-\infty}^{\infty}d\tau\ln[1-\tilde{P}(\tau-i/2)]\left[\cot\left[\frac{\pi}{\alpha}(\tfrac{1}{2}+i\tau)\right]-\cot\left[\frac{\pi}{\alpha}(-\tfrac{1}{2}+i\tau)\right]\right]\right\}$$

which can be further rewritten in an equivalent form used in the main text:

$$\Gamma=\frac{C\lambda_0}{4\pi\sin(\beta\hbar\lambda_0/2)}\exp\left[\frac{\beta\hbar\lambda_0\sin(\beta\hbar\lambda_0/2)}{2\pi}\int_{-\infty}^{\infty}d\tau\frac{\ln[1-\tilde{P}(\tau-i/2)]}{\cosh(\tau\beta\hbar\lambda_0)-\cos(\beta\hbar\lambda_0/2)}\right] . \tag{A15}$$

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