Dissipative quantum systems with a potential barrier. III. Steady state nonequilibrium flux and reaction rate

Joachim Ankerhold* and Hermann Grabert
Fakultät für Physik der Albert-Ludwigs-Universität, Hermann-Herder-Strasse 3, D-79104 Freiburg, Germany
(Received 4 September 1996)

We study the real time dynamics of a dissipative quantum system in a metastable state which may decay by crossing a potential barrier. Starting from an initial state where the system is in thermal equilibrium on one side of the barrier, the time evolution of the density matrix is evaluated analytically in the semiclassical approximation for coordinates near the barrier top. In a region about a critical temperature T_c large quantum fluctuations render the harmonic approximation of the potential insufficient and anharmonicities become essential. Accounting for non-Gaussian fluctuation modes, we show that the density matrix approaches a quasistationary state with a constant flux across the barrier. This extends our earlier results [Phys. Rev. E **51**, 4267 (1995)] on the quantum generalization of the Kramers flux state to the region about T_c. By matching the flux state onto the equilibrium state on one side of the barrier, we determine the decay rate out of the metastable state. The rate constant shows a changeover from thermally activated decay to quantum tunneling for temperatures below T_c. [S1063-651X(97)01002-7]

PACS number(s): 05.40.+j, 03.65.Sq, 82.20.Db

I. INTRODUCTION

Quantum mechanical barrier penetration phenomena can be found in various areas in physics and chemistry [1]. While usually the systems in question exhibit a large number of degrees of freedom, in most cases only one variable, the so-called reaction coordinate, governs the escape process. However, the interaction of the reaction coordinate with the remaining degrees of freedom is essential and cannot be neglected. As a consequence, theory must incorporate effects of a heat-bath environment. In the classical region of thermally activated decay, generalized Langevin equations and related methods are adequate. This way, based on the seminal work by Kramers [2], escape rates can be determined from the stationary nonequilibrium flux across the barrier.

In the past decade various theories were formulated for rate calculations in the quantum mechanical regime [3–9] which are based on thermodynamic methods and ultimately *ad hoc* rate formulas. The most famous of these rate expressions proposed by Langer [10] relates the decay rate with the imaginary part of an analytically continued free energy. Foundations for a dynamical quantum rate theory, starting from first principles, have been layed only very recently in a sequence of two articles [11,12], which are referred to as I and II henceforth. The approach is based upon the path integral description of dissipative quantum systems [13] pioneered by Feynman and Vernon [14]. This method was used by Caldeira and Leggett [15] to describe metastable systems and extended by Grabert, Schramm, and Ingold [16] to a wider class of useful initial conditions. In this article the theory is applied to a dissipative quantum system with a metastable state which may decay by crossing a high potential barrier. Then, a semiclassical evaluation of the time dependent density matrix is adequate. Previously, in article I [11] we have considered the region of moderately high temperatures where quantum effects are important but the harmonic approximation of the barrier potential is still sufficient. We have shown that the nonequilibrium state becomes stationary for a large plateau of intermediate times. The corresponding quasistationary flux state is the quantum generalization of Kramers' flux solution of the Fokker-Planck equation [2] and can be used to determine the escape rate. For lower temperatures, near a critical temperature T_c, the simple Gaussian semiclassical approximation breaks down. The instability arises since near T_c new classical paths emerge in the inverted barrier potential [17]. In article II [12] we have extended the calculation of the semiclassical time dependent density matrix to the region about T_c. For a high barrier analytical results for the density matrix are available also in the critical region where large quantum fluctuations explore the anharmonic range of the barrier potential. However, one has to go beyond the Gaussian approximation in the semiclassical evaluation of the path integrals.

In the present article we use the results of I and II to determine the stationary flux state and the related escape rate in the temperature region about T_c. We derive results that are valid from high temperature down to temperatures slightly below T_c. At even lower temperatures a simple semiclassical approximation of the path integrals is again possible but analytical results are usually not available.

The article is organized as follows. In Sec. II we briefly introduce the real time formulation of the problem and our basic notation. In Secs. III and IV the semiclassical time dependent density matrix is evaluated by determining the minimal action paths, the corresponding minimal action, and the contribution of the quantum fluctuations. This result is used in Sec. V to derive the stationary nonequilibrium flux state. The matching of the flux solution onto the thermal equilibrium state in the well investigated in Sec. VI leads to a condition on the minimal damping strength required for our

*Present address: Department of Chemistry, Columbia University, New York, NY 10027.

results to be valid. We also calculate the escape rate, which is shown to be identical with the rate for thermally activated decay for temperatures above T_c but deviates for lower temperatures. Parts of these results are contained in a thesis by one of us [18].

II. TIME DEPENDENT DENSITY MATRIX

In this section we briefly introduce the real time formulation for the dynamics of dissipative quantum systems and specify the barrier potential. Afterwards the initial preparation is considered.

A. Time evolution of the reduced density matrix and barrier potential

As shown in detail in [16] the position representation of the time dependent reduced density matrix can be written as

$$\rho(x_f,r_f,t)=\int dx_i dr_i d\overline{x}d\overline{r}\, J(x_f,r_f,t,x_i,r_i,\overline{x},\overline{r})\times\lambda(x_i,r_i,\overline{x},\overline{r}). \tag{1}$$

Here, $J(x_f,r_f,t,x_i,r_i,\overline{x},\overline{r})$ denotes the propagating function given as a threefold path integral where two path integrals are in real time and one is in imaginary time. The preparation function $\lambda(x_i,r_i,\overline{x},\overline{r})$ describes the deviation from thermal equilibrium in the initial state

$$\rho(x_f,r_f,0)=\int d\overline{x}d\overline{r}\,\lambda(x_f,r_f,\overline{x},\overline{r}')\rho_\beta(\overline{x},\overline{r}), \tag{2}$$

where $\rho_\beta=\mathrm{tr}_R(W_\beta)$ in which W_β is the equilibrium density matrix of the entire system. A brief summary of the underlying theory is given in I.

In the following we consider a system in a metastable state which may decay by crossing a potential barrier. Assuming that the barrier top is at $q=0$ and $V(0)=0$, the general form of a symmetric barrier potential reads

$$V(q)=-\frac{1}{2}M\omega_0^2q^2\left[1-\sum_{k=2}^{\infty}\frac{c_{2k}}{k}\left(\frac{q}{q_a}\right)^{2k-2}q^{2k-2}\right]. \tag{3}$$

Here, the c_{2k} are dimensionless coefficients. We assume $c_4>0$ so that the barrier potential becomes broader than its harmonic approximation at lower energies. q_a is a characteristic length indicating a typical distance from the barrier top at which anharmonic terms of the potential become essential. For small $|q|\ll q_a$ the barrier potential is harmonic with frequency ω_0. Now, we imagine that the system starts out from a potential well to the left of the barrier. Metastability then means that the barrier height V_b is much larger than other relevant energy scales of the system such as k_BT and $\hbar\omega_0$, where $\hbar\omega_0$ is the excitation energy in the well of the inverted potential. In I we have introduced $q_0=\sqrt{\hbar/2M\omega_0}$ as a typical quantum mechanical length scale which is the variance of the coordinate in the ground state of a harmonic oscillator with oscillation frequency ω_0. Since V_b is related with q_a, the condition $V_b\gg\hbar\omega_0$ implies that

$$\epsilon=q_0/q_a \tag{4}$$

is a small dimensionless parameter.

Clearly, for an anharmonic potential field the threefold path integral defining the propagating function in Eq. (1) cannot be solved exactly. However, since $\epsilon\ll1$, the dynamics of the escape process may be determined via a semiclassical approximation of the propagating function for coordinates near the barrier top with the small parameter ϵ serving as an expansion parameter. This calculation becomes more transparent if we introduce a *dimensionless formulation*. In the following all coordinates are measured in units of q_0, all frequencies in units of ω_0, and all times in units of $1/\omega_0$.

For sufficiently high temperatures the smallest eigenvalue for fluctuations about the barrier top

$$\Lambda(\theta)=-\frac{1}{\theta}+\frac{2}{\theta}\sum_{n=1}^{\infty}\frac{1}{\nu_n^2-1+\nu_n\hat{\gamma}(\nu_n)} \tag{5}$$

is finite and of order 1. Here, $\nu_n=2\pi n/\theta$ are the dimensionless Matsubara frequencies where $\theta=\omega_0\hbar\beta$ denotes the dimensionless inverse temperature, and $\hat{\gamma}(z)$ is the Laplace transform of the macroscopic damping kernel $\gamma(s)$ describing the influence of the heat-bath environment. As a consequence, in this temperature range and for moderate to strong damping the barrier dynamics is not affected by anharmonic terms of the potential over a wide range of times (see I). For lower temperatures, however, $|\Lambda(\theta)|$ decreases and vanishes at a critical temperature T_c determined by $\Lambda(\theta_c)=0$. Then, the propagating function diverges within the harmonic approximation and one has to go beyond a simple semiclassical approximation (see II). Hence, near T_c large quantum fluctuations render the harmonic approximation insufficient and anharmonicities are essential even for coordinates close to the barrier top and for all times.

B. Initial preparation

The initial nonequilibrium state at time $t=0$ is described by the preparation function

$$\lambda(x_i,r_i,\overline{x},\overline{r})=\delta(x_i-\overline{x})\delta(r_i-\overline{r})\Theta(-r_i) \tag{6}$$

so that the initial state (2) is a thermal equilibrium state restricted to the left side of the barrier only,

$$\rho(x_f,r_f,0)=\rho_\theta(x_f,r_f)\Theta(-r_f). \tag{7}$$

Here, $\rho_\theta(x,r)$ is the position representation of the equilibrium density matrix. Then, according to Eq. (1), the time dependent density matrix is given by

$$\rho(x_f,r_f,t)=\int dx_i dr_i J(x_f,r_f,t,x_i,r_i,x_i,r_i)\Theta(-r_i). \tag{8}$$

As in I, it is convenient to write this equation as

$$\rho(x_f,r_f,t)=\rho_\theta(x_f,r_f)g(x_f,r_f,t), \tag{9}$$

where $g(x_f,r_f,t)$ is a form factor describing deviations from thermal equilibrium. From Eqs. (8) and (I26) [19] one has

$$g(x_f,r_f,t)=\int dx_i dr_i \widetilde{J}(x_f,r_f,t,x_i,r_i)\Theta(-r_i)/\rho_\theta(x_f,r_f), \tag{10}$$

where

$$\widetilde{J}(x_f,r_f,t,x_i,r_i)\equiv J(x_f,r_f,t,x_i,r_i,x_i,r_i) = \frac{1}{Z}\int \mathcal{D}x\mathcal{D}r\mathcal{D}\overline{q}\exp\left[\frac{i}{2}\Sigma[x,r,\overline{q}]\right]. \tag{11}$$

The above path integral is over all paths $x(s),r(s)$, $0\leqslant s\leqslant t$ in real time with

$$x(0)=x_i,\quad r(0)=r_i,\quad x(t)=x_f,\quad r(t)=r_f,$$

and over all paths $\overline{q(\sigma)}$, $0\leqslant\sigma\leqslant\theta$ in imaginary time with $\overline{q(0)}=r_i-x_i/2$, $\overline{q(\theta)}=r_i+x_i/2$. We note that x_i,x_f and r_i,r_f are difference and sum coordinates defined in Eqs. (I22) and (I23). Z is an appropriate normalization factor and the effective action $\Sigma[x,r,\overline{q}]$ is given by Eq. (I28),

$$\begin{aligned}\Sigma[x,r,\overline{q}]=&\, i\int_0^\theta d\sigma\left[\frac{1}{2}\dot{\overline{q}}^2+V(\overline{q})+\frac{1}{2}\int_0^\theta d\sigma' k(\sigma-\sigma')\overline{q(\sigma)}\,\overline{q(\sigma')}\right]+\int_0^\theta d\sigma\int_0^t ds K^*(s-i\sigma)\overline{q(\sigma)}x(s)\\ &+\int_0^t ds[\dot{x}\dot{r}-V(r+x/2)+V(r-x/2)-r_i\gamma(s)x(s)]\\ &-\int_0^t ds\left[\int_0^s ds'\gamma(s-s')x(s)\dot{r}(s')-\frac{i}{2}\int_0^t ds' K'(s-s')x(s)x(s')\right].\end{aligned} \tag{12}$$

Here, the asterisk denotes complex conjugation and the kernel $K(s-i\sigma)$ is specified in Eq. (I29). The dimensionless potential field reads

$$V(q)=-\frac{1}{2}q^2\left(1-\sum_{k=2}^{\infty}\frac{c_{2k}}{k}\epsilon^{2k-2}q^{2k-2}\right). \tag{13}$$

C. Coordinate transformation

In view of Eq. (9), the dynamics of the escape process is determined by the time dependent form factor (10). In particular, we are interested in the stationary flux over the barrier in a region of time (plateau region) where the form factor becomes nearly independent of time. For sufficiently strong damping the nonequilibrium region of the flux state is localized in coordinate space, so that the form factor reaches 1 on the left side of the barrier and 0 on the right side within a small region around the barrier top. As shown in I, the dimensionless width of this nonequilibrium region is of order 1 for high temperatures where $|\Lambda|$ is of order 1 and decreases with decreasing temperature. Hence, to extend the investigations of I to the temperature region around T_c, we calculate the density matrix $\rho(x_f,r_f,t)$ for coordinates x_f and r_f smaller than order 1, i.e., for dimensional coordinates q and q' smaller than q_0.

From I and II we also know that for lower temperatures the range of initial coordinates x_i and r_i which are relevant for the density matrix (8) in the barrier region increases. In particular, near T_c these coordinates can be restricted to be of order 1 only for small times. Furthermore, we have shown in II that near T_c even for endpoints near the barrier top the amplitude of the imaginary time path $\overline{q(\sigma)}$ becomes very large. To determine the relevant range of initial coordinates, we recall that in I we have calculated the stationary form factor within the harmonic approximation by performing a transformation $(x_i,r_i)\rightarrow(x_i',r_i)$ defined by $x_i'=x_i-ir_i/a$, where

$$a=\lim_{\omega_R t\gg 1}\frac{S(t)}{2A(t)}=\frac{1}{2}\cot\left(\frac{\omega_R\theta}{2}\right). \tag{14}$$

Here, ω_R is the Grote-Hynes frequency [20] given by the positive solution of $\omega_R^2+\omega_R\hat{\gamma}(\omega_R)=1$. The functions $A(t)$ and $S(t)$ describe the unbounded motion at the parabolic barrier and are specified in Eqs. (I72) and (I73). Evaluating these functions for times larger than $1/\omega_R$ one gets apart from corrections decaying exponentially in time [Eqs. (I83, I84)]

$$A(t)=-\frac{1}{2}\frac{1}{2\omega_R+\hat{\gamma}(\omega_R)+\omega_R\hat{\gamma}'(\omega_R)}\exp(\omega_R t) \tag{15}$$

and

$$S(t)=-\frac{a}{2\omega_R+\hat{\gamma}(\omega_R)+\omega_R\hat{\gamma}'(\omega_R)}\exp(\omega_R t), \tag{16}$$

where $\hat{\gamma}'(\omega)$ is the derivative of $\hat{\gamma}$ with respect to ω. The evaluation of the flux solution for higher temperatures carried out in I shows that in the new coordinates x_i' and r_i, the integrals in Eq. (10) decouple. The relevant values of x_i' are of order $\sqrt{|\Lambda|}$ or smaller and those of r_i at most of order $S(t)/\sqrt{|\Lambda|}$. Consequently, the relevant range of the x_i' coordinate decreases with decreasing temperature while the relevant range of r_i increases. This suggests the use of a transformation similiar to that for high temperatures also near T_c, i.e.,

$$x_i' = x_i - i r_i / a + \Delta(r_i), \tag{17}$$

where $\Delta(r_i)$ is a function of r_i only with $\Delta(0)=0$ which takes into account nonlinear corrections to the linear transformation. In particular, with the help of the transformation (17) one can demonstrate that the relevant values of the amplitude of the imaginary time path $\overline{q(\sigma)}$ remain of the same order of magnitude as the endpoint r_i which simplifies the semiclassical approximation enormously.

Now, to determine the stationary density matrix near T_c we make use of Eq. (17) and the following assumptions which will be confirmed self-consistently. First, we assume that anharmonicities of the potential become important for temperatures where $|\Lambda|$ is of order ϵ or smaller. Second, we evaluate the flux solution only for large times $\omega_R t \gg 1$ where $A(t)$ and $S(t)$ are of the form (15) and (16). Hence, we consider times where $A(t)$ and $S(t)$ are large. To keep track of the relevant orders of magnitude, $A(t)$ and $S(t)$ are estimated by $\epsilon^{-\alpha(t)}$ with an appropriate exponent $\alpha>0$. Third, following the above discussion, x_i' is assumed to be at most of order $\epsilon^{1/2-\alpha}$ and r_i to be at most of order $\epsilon^{-1/2-\alpha}$. Finally, we assume that the function $\Delta(r_i)$ is at most order $\epsilon^{1/2-3\alpha}$. In practice, it is sufficient to consider the limit $\alpha \to 0$ since the plateau region is reached as soon as $A(t)$ and $S(t)$ are given by the asymptotic formulas (15) and (16).

With these assumptions we evaluate the semiclassical propagating function in Secs. III and IV. Thereby, the propagating function is calculated for real time paths $x(s)$ and $r(s)$ and imaginary time paths $\overline{q(\sigma)}$ that are at most of order $\epsilon^{-1/2-\alpha}$. We first determine the minimal action paths and the corresponding minimal action. Afterwards, the contribution of the quantum fluctuations is calculated. Most of these steps have already been done in II so that we omit details here. By use of the semiclassical propagating function the form factor is then evaluated in Sec. V.

III. MINIMAL ACTION PATHS

Here, we briefly outline the determination of the minimal action paths. For a general initial state a corresponding calculation is performed in II to which we refer for further details. However, some differences should be noted. First, in II we have distinguished between a classical small parameter ξ characterizing the influence of potential anharmonicities and the quantum mechanical small parameter ϵ given in Eq. (4). The parameter ξ serves as an expansion parameter for the classical motion while ϵ governs the semiclassical approximation. Since in the present case the density matrix is calculated for dimensional coordinates within a quantum mechanical range (smaller than q_0) around the barrier top, the distinction between ξ and ϵ is not necessary and we set $\xi=\epsilon$. Second, due to the particular initial state (6) considered here, the transformation (17) is used advantageously already for the solution of the equations of motion. The amplitude of the imaginary time path $\overline{q(\sigma)}$ is then readily seen to be at most of the same order of magnitude than the initial coordinates x_i and r_i. In the case of a general initial state considered in II, the amplitude of $\overline{q(\sigma)}$ may be much larger then x_i and r_i. On the other hand, the assumption made in II that the initial coordinates x_i and r_i are at most of order $\epsilon^{-1/4}$ is too restrictive for the present case, since the relevant values of r_i become of order $\epsilon^{-1/2-\alpha}$ as discussed above. Hence, we have to extend the perturbative expansion in II and retain terms neglected there. However, we perform this expansion only for the range of coordinates required for the particular initial state (6) which simplifies the expansion substantially. We remark that in this section the explicit dependence of the function $x_i = x_i(x_i', r_i)$ according to Eq. (17) is suppressed to keep the formulas more transparent.

A. Extremal imaginary time path

From Eq. (I43) the minimal action path in imaginary time obeys the equation of motion

$$\ddot{\overline{q}} - \int_0^\theta d\sigma' k(\sigma-\sigma')\overline{q(\sigma')} - \frac{dV(\overline{q})}{d\overline{q}}$$

$$= -i\int_0^t ds K^*(s-i\sigma)x(s), \tag{18}$$

where $\overline{q(0)} = r_i - x_i/2$ and $\overline{q(\theta)} = r_i + x_i/2$. The inhomogeneity on the right hand side couples $\overline{q(\sigma)}$ to the real time motion.

As already addressed, the amplitude of $\overline{q(\sigma)}$ grows near T_c and the harmonic approximation breaks down. A detailed analysis shows (see II) that the imaginary time motion becomes marginally stable only in one direction in function space. Hence, to determine the imaginary time path for an anharmonic potential near T_c we follow II and make the ansatz

$$\overline{q(\sigma)} = \frac{1}{\theta}\sum_{l=1}^{\infty} Q_{2l}\sin(\nu_l\sigma) + \hat{Q}\phi(\sigma) + \hat{q}(\sigma). \tag{19}$$

Here, $\phi(\sigma)$ denotes the marginal direction in function space [Eq. (II23)] and $\hat{Q}$ the corresponding amplitude, while the function $\hat{q}(\sigma)$ contains boundary terms and is specified in Eq. (II26). Inserting Eq. (19) into Eq. (18) one gets equations for the amplitudes Q_{2l} and for $\hat{Q}$ (see II). The equations for the amplitudes Q_{2l} can be solved perturbatively by expanding about the harmonic path. We set

$$Q_{2l} = Q_{2l,0} + \epsilon Q_{2l,1} + O(\epsilon^{3/2-5\alpha}), \tag{20}$$

where the $Q_{2l,0}$ are of order $\epsilon^{-1/2-\alpha}$ and describe the solution in the harmonic approximation. The $Q_{2l,1}$ are of order $\epsilon^{-1/2-3\alpha}$ and take into account corrections due to anharmonic terms. This way we obtain from Eq. (18)

$$Q_{2l,0} = -u_l\{2\nu_l x_i + 2 f_l[x(s)]\} \tag{21}$$

and

$$Q_{2l,1} = -2c_4\epsilon u_l\int_0^\theta d\sigma \sin(\nu_l\sigma)\left[\frac{1}{\theta}\sum_{l=1}^{\infty} Q_{2l,0}\sin(\nu_l\sigma) + \hat{Q}\phi(\sigma) + \hat{q}(\sigma)\right]^3. \tag{22}$$

Here, we used the abbreviation

$$u_l=[\nu_l^2-1+|\nu_l|\hat{\gamma}(|\nu_l|)]^{-1}. \quad (23)$$

Furthermore, the functionals $f_n[x(s)]$ and also $g_n[x(s)]$ that appear in the equation for $\hat{Q}$ (see below) describe the coupling to the real time motion and were already given in Eqs. (II27) and (II28).

For the marginal mode amplitude one has from Eq. (18) apart from corrections of order $\epsilon^{3/2-5\alpha}$

$$\frac{\Lambda}{\epsilon^{1/2}}Q-c_4\epsilon^2\int_0^\theta d\sigma\phi(\sigma)^3\left[\frac{1}{\theta}\sum_{l=1}^{\infty}Q_{2l,0}\sin(\nu_l\sigma)+\frac{2\theta}{\epsilon^{1/2}}Q\phi(\sigma)+\hat{q}(\sigma)\right]^3=\overline{b} \quad (24)$$

with the inhomogeneity

$$\overline{b}=\frac{1}{2\theta}\left\{r_i-\frac{i}{\theta}\sum_{n=-\infty}^{\infty}u_n g_n[x(s)]\right\}. \quad (25)$$

Here, as in II, we have introduced

$$Q\equiv\frac{1}{2\theta}\epsilon^{1/2}\hat{Q} \quad (26)$$

to make the ϵ dependence more explicit. A cubic equation similiar to Eq. (24) has been derived previously in Eq. (II41). However, due to the differences discussed at the beginning of this section, (II41) contains at most terms linear in r_i, while in Eq. (24) powers of r_i up to r_i^3 have been retained. Yet, the basic properties of the cubic equation (24) are the same as those discussed in II, to where we refer for further details. We only note that for endpoints of order 1 and for high temperatures where $|\Lambda|$ is larger than order $\epsilon^{2/3}$ we recover from Eq. (24) the harmonic solution.

B. Extremal real time paths

Let us start by considering the equation of motion for the path $r(s)$ which reads according to Eq. (I42)

$$\ddot{r}+\frac{d}{ds}\int_0^s ds'\gamma(s-s')r(s')+\frac{1}{2}\frac{d}{dr}[V(r+x/2)+V(r-x/2)]=i\int_0^t ds'K'(s-s')x(s')+\int_0^\theta d\sigma K^*(s-i\sigma)\overline{q(\sigma)}. \quad (27)$$

For endpoints in the range assumed above, anharmonic terms in the potential become larger than order $\epsilon^{1/2}$. Hence, these terms cannot be neglected, since they lead to terms at least of order 1 in the minimal effective action. We have from Eq. (13)

$$\frac{1}{2}\frac{d}{dr}[V(r+x/2)+V(r-x/2)]=-r+c_4\epsilon^2r^3+\frac{3}{4}c_4\epsilon^2rx^2+O(\epsilon^{3/2-5\alpha}). \quad (28)$$

The equation of motion (27) can then be solved perturbatively using the ansatz

$$r(s)=r_0(s)+\epsilon r_1(s)+O(\epsilon^{3/2-5\alpha}) \quad (29)$$

and also

$$x(s)=x_0(s)+\epsilon x_1(s)+O(\epsilon^{3/2-5\alpha}), \quad (30)$$

where $r_0(s)$ and $x_0(s)$ may be of order $\epsilon^{-1/2-\alpha}$ with $r_0(0)=r_i$, $r_0(t)=r_f$ and $x(0)=x_i$, $x(t)=x_f$. The corrections $r_1(s)$ and $x_1(s)$ are of order $\epsilon^{-1/2-3\alpha}$ and obey $r_1(0)=r_1(t)=0$ and $x_1(0)=x_1(t)=0$.

Following the steps outlined in detail in II, we then obtain in leading order

$$r_0(s)=r_h(s)-\frac{G_+(s)}{\Lambda}[C_1^+(s)-C_1^+(t)][r_i-ix_iC_1^+(t)]+r_0^a(s)+G_+(s)[\gamma_i(s)-\gamma_i(t)]\left(\overline{b}-\frac{2\theta\Lambda Q}{\epsilon^{1/2}}\right). \quad (31)$$

The real time path in the harmonic potential $r_h(s)$ is given in Eq. (I66). Further,

$$r_0^a(s)=2\theta\frac{Q}{\epsilon^{1/2}}G_+(s)[C_1^+(s)-C_1^+(t)]. \quad (32)$$

Here, we have introduced the propagator $G_+(s)$ of the homogeneous equation (27) with the initial conditions $G_+(0)=0$, $\dot{G}_+(0)=1$, which has the Laplace transform

$$\hat{G}_+(z)=[z^2+z\hat{\gamma}(z)-1]^{-1}. \quad (33)$$

The time dependent functions $C_n^+(t)$ are specified in terms of $A(t)$ and $S(t)$ in Eq. (II102). The function $\gamma_i(s)$ is given in Eq. (II83) and $\overline{b}$ in Eq. (25). Clearly, the last three terms in Eq. (31), which depend through the amplitude Q on the anharmonicities of the potential, obey $(r_0-r_h)(0)=(r_0-r_h)(t)=0$. The first part $r_h(s)$ in Eq. (31) guarantees that $r(s)$ fulfills the boundary conditions $r(0)=r_i$ and $r(t)=r_f$. This harmonic path diverges for $\Lambda\to 0$ while $r_0(s)$ remains finite. In first order we gain

$$r_1(s)=\int_0^s ds'G_+(s-s')R_1(s')-\frac{G_+(s)}{G_+(t)}\int_0^t ds'G_+(t-s')R_1(s') \quad (34)$$

with

$$R_1(s)=\int_0^\theta d\sigma K^*(s-i\sigma)\overline{q_1}(\sigma)+i\int_0^t ds'K'(s-s')x_1(s')-c_4\epsilon\left[r_0(s)^3+\frac{3}{4}r_0(s)x_0(s)^2\right]. \quad (35)$$

Here, we have used the decomposition

$$\overline{q(\sigma)}=\overline{q_0}(\sigma)+\epsilon\overline{q_1}(\sigma) \quad (36)$$

of the imaginary time path (19) where $\overline{q_0}(\sigma)$ is of order $\epsilon^{-1/2-\alpha}$ with $\overline{q_0}(0)=r_i-x_i/2$ and $\overline{q_0}(\theta)=r_i+x_i/2$. The correction $\epsilon\overline{q_1}(\sigma)$ collects terms in $\overline{q(\sigma)}-\hat{Q}\phi(\sigma)$ which are at most of order $\epsilon^{1/2-3\alpha}$.

Finally, let us investigate the equation of motion for the real time path $x(s)$. According to Eq. (I43) we have

$$\ddot{x}-\frac{d}{ds}\int_s^t ds'\,\gamma(s'-s)x(s')+2\frac{d}{dx}[V(r+x/2)+V(r-x/2)]=0. \tag{37}$$

Further, from Eq. (13) one obtains

$$2\frac{d}{dx}[V(r+x/2)+V(r-x/2)]=-x+3c_4\epsilon^2xr^2+\frac{c_4}{4}\epsilon^2x^3+O(\epsilon^{3/2-5\alpha}). \tag{38}$$

To solve Eq. (37) perturbatively we use the ansatz (30). The equation of motion for $x_0(s)$ is homogeneous and the backward equation of motion for $r_0(s)$ for vanishing inhomogeneity [16]. Hence, we gain

$$x_0(s)=x_i\frac{G_+(t-s)}{G_+(t)}+x_f\left[\dot{G}_+(t-s)-\frac{G_+(t-s)}{G_+(t)}\dot{G}_+(t)\right]. \tag{39}$$

Further, to next order in ϵ one obtains (see also II) the solution for $x_1(s)$ as

$$x_1(s)=-c_4\epsilon\Bigg\{\int_s^t ds'\,G_+(s'-s)\left[3r_0(s')^2x_0(s')+\frac{1}{4}x_0(s')^3\right]-\frac{G_+(t-s)}{G_+(t)}\int_0^t ds'\,G_+(s')\times\left[3r_0(s')^2x_0(s')+\frac{1}{4}x_0(s')^3\right]\Bigg\}. \tag{40}$$

With Eqs. (31) and (34) [(39) and (40)] the solution for the real time path $r(s)$ [$x(s)$] has been found for end points x_f and r_f smaller than order 1 but possibly large x_i and r_i at most of order $\epsilon^{-1/2-\alpha}$. In particular, the above analysis shows that both real time paths are also at most of order $\epsilon^{-1/2-\alpha}$. Hence, these solutions correspond to trajectories with a turning point near the barrier top which remain therefore in the barrier region for all times. In principle, there exist also trajectories going forth and back through the well region of the metastable potential. However, as we have discussed in detail in II, these trajectories give relevant contributions to the density matrix only in a parameter region which is not relevant here, namely, for extremely long times and small damping.

IV. SEMICLASSICAL DENSITY MATRIX

In the preceding section we have evaluated the minimal action paths. Employing the semiclassical approximation we now expand the functional integrals about these paths. In a first step one determines the corresponding minimal effective action (Sec. IV A) and in a second step the quantum fluctuations are considered (Sec. IV C) leading to the semiclassical propagating function.

A. Minimal effective action

According to the investigations in II, the minimal effective action in an anharmonic potential field can be written in the form

$$\Sigma(x_f,r_f,t,x_i,r_i)=\Sigma^h(x_f,r_f,t,x_i,r_i)+\Sigma^a(x_f,r_f,t,x_i,r_i), \tag{41}$$

where $\Sigma^h(x_f,r_f,t,x_i,r_i)$ denotes the harmonic result (I68) and $\Sigma^a(x_f,r_f,t,x_i,r_i)$ is formally specified in Eq. (II78). Now, we insert the imaginary time path $\overline{q(\sigma)}$ and the two real time paths $r(s)$ and $x(s)$ determined in the previous section into Σ^a. Using the equations of motion, we then get after some algebra up to terms of order 1

$$\Sigma^a(x_f,r_f,t,x_i,r_i)=\Sigma_0^a(x_f,r_f,t,x_i,r_i)-c_4\epsilon^2\Sigma_1^a(x_f,r_f,t,x_i,r_i)+O(\epsilon^{1-6\alpha}) \tag{42}$$

where

$$\Sigma_0^a(x_f,r_f,t,x_i,r_i)=\frac{i}{2}\left\{\left(r_i+\frac{x_i}{2}\right)[\overline{q_0}-\overline{q_h}](\theta)-\left(r_i-\frac{x_i}{2}\right)[\overline{q_0}-\overline{q_h}](0)\right\}+x_f[\dot{r}_0-\dot{r}_h](t)-x_i[\dot{r}_0-\dot{r}_h](0)-\frac{1}{2}\int_0^t ds\int_0^\theta d\sigma K^*(s-i\sigma)x_0(s)[\overline{q_0}-\overline{q_h}](\sigma), \tag{43}$$

with the harmonic paths $\overline{q_h}(\sigma)$ and $r_h(s)$ given in Eqs. (I44) and (I66), respectively. Furthermore, one gains for the second term in Eq. (42) the compact result

$$\Sigma_1^a(x_f,r_f,t,x_i,r_i)=\frac{i}{4}\int_0^\theta d\sigma\overline{q_0}(\sigma)^4-\frac{i}{2}\int_0^\theta d\sigma\overline{q_0}(\sigma)^3\left[\frac{1}{\theta}\sum_{l=1}^\infty Q_{2l,0}\sin(\nu_l\sigma)\right]+\int_0^t ds\,x_1(s)C_1(s)\left[\frac{\theta Q}{c_4\epsilon^{3/2}}+\frac{i}{2\epsilon c_4}\int_0^t ds'\,x_0(s')\gamma(s')\right]+\frac{1}{4}\int_0^t ds[r_0(s)x_0(s)^3+4r_0(s)^3x_0(s)]. \tag{44}$$

The two expressions (43) and (44) are now used to evaluate the minimal effective action explicitly as a function of the coordinates for times where $\omega_Rt\gg1$. It is obvious that the function Σ^a contains anharmonic x_i and r_i dependent terms, so that an analytical evaluation of the form factor seems impossible. However, transforming to the coordinate x_i' according to Eq.

(17), one finds that the leading order $x_i' r_i$ coupling terms vanish. Also, the amplitude Q studied in the following section is seen to become at most of order $\epsilon^{-\alpha}$. With the transformation (17) one gains after a tedious but straightforward calculation

$$\Sigma_0(x_f,r_f,t,x_i',r_i)\equiv\Sigma_h(x_f,r_f,t,x_i(x_i',r_i),r_i)+\Sigma_0^a(x_f,r_f,t,x_i(x_i',r_i),r_i)=-\frac{\theta Q}{\epsilon^{1/2}}ax_i'+\frac{\theta Q}{\epsilon^{1/2}}a\Delta-\frac{i\theta\Lambda Q}{a\epsilon^{1/2}}\omega_R r_i$$
$$-\frac{i}{2a^2}\Lambda r_i^2\left(\frac{1}{4A(t)^2}+\omega_R^2\right)-x_i' r_i\omega_R+\omega_R r_i\Delta+\frac{i}{2aA(t)}r_f r_i+\omega_R x_f r_f+\frac{i}{2}\Omega x_f^2, \tag{45}$$

where we have neglected corrections which are exponential small for large times but have kept the leading order x_f and r_f dependent terms. Evaluating Σ_1^a as a function of the transformed coordinates x_i', r_i, one finds that apart from corrections that are smaller than order 1 this part of the action is independent of the coordinates x_f and r_f. We obtain

$$\Sigma_1(x_i',r_i,t)\equiv\Sigma_1^a(x_f,r_f,t,x_i(x_i',r_i),r_i)=i2\theta^5 D_4\frac{Q^4}{\epsilon^2}-i\frac{4\theta^3Q^3}{a\epsilon^{3/2}}r_i\left[F_{3,1}(t)-8A(t)^3C_{3,1}(t)-\frac{\theta}{2}\gamma_i(t)D_4\right]$$
$$-i\frac{3\theta Q^2}{a^2\epsilon}r_i^2[F_{2,2}(t)+\theta F_{3,1}(t)\gamma_i(t)]-i\frac{Q}{a^3\theta\epsilon^{1/2}}r_i^3\left\{F_{1,3}(t)+\theta^2A(t)J_1(t)-\frac{3}{2}\gamma_i(t)\theta[F_{2,2}(t)-4\overline{F_{2,2}}(t)]\right\}$$
$$+\frac{i}{8\theta^3a^4}r_i^4[F_4(t)-16\overline{F_{0,4}}]-\frac{i}{a^2}r_i^4\left[\frac{1}{2a^2}A(t)\psi_{1,3}(t)-\frac{8}{a^2}A(t)^3\psi_{3,1}(t)-a\Gamma_3(t)+\frac{1}{4a}\Gamma_1(t)\right.$$
$$\left.-\frac{12}{a}A(t)^2\psi_{2,1}(t)-6A(t)\psi_{1,1}(t)+\frac{\gamma_i(t)}{4a^2\theta^2}F_{1,3}(t)\right]. \tag{46}$$

Here, according to Eq. (II37), one has

$$D_n=\frac{2}{\theta}\int_0^\theta d\sigma\phi(\sigma)^n. \tag{47}$$

Moreover, we have introduced a set of auxiliary time dependent functions, which were in part already used in II, to describe the influence of the potential anharmonicities on the real time motion. These functions are given explicitly in Appendix A.

B. Amplitude of the marginal mode

Before we evaluate the contribution of the quantum fluctuations, let us write Eq. (24) for the amplitude Q in explicit form. First, by use of the solution (39) and (40) of the real time path $x(s)$, one obtains for the inhomogeneity on the right hand side of the cubic equation

$$\overline{b}\equiv\frac{1}{2\theta}\left\{r_i-\frac{i}{\theta}\sum_{n=-\infty}^{\infty}u_ng_n[x(s)]\right\}$$
$$=\frac{1}{2\theta}[r_i-ix_iC_1^+(t)-ix_f2A(t)\dot{C}_1^+(t)]$$
$$-\frac{i\epsilon}{2\theta}\int_0^t ds x_1(s)C_1(s), \tag{48}$$

where $C_1^+(t)$ is given in Eq. (II102). Note that the last term on the right hand side is of order $\epsilon^{1/2}$. For times $\omega_R t\gg1$ the time dependent term $A(t)\dot{C}_1^+(t)$ becomes exponentially small (see I) and can therefore be neglected. We remark that a naive estimate of the magnitude of the amplitude Q from the cubic equation (24) and (48) for untransformed coordinates x_i,r_i of order $\epsilon^{-1/2}$ leads near T_c to values of Q of order $\epsilon^{-1/3}$, i.e., $\hat{Q}$ of order $\epsilon^{-5/6}$. Such large values of $\hat{Q}$ would require perturbation theory for the minimal action paths at quite large orders. However, using the transformation (17) we find from Eq. (48) for large times

$$\overline{b}=i\frac{a}{2\theta}x_i'-\frac{\Lambda\omega_R}{2\theta a}r_i-i\frac{a}{2\theta}\Delta(r_i)-\frac{i\epsilon}{2\theta}\int_0^t ds x_1(s)C_1(s). \tag{49}$$

For x_i' smaller than order 1 and Δ of order $\epsilon^{1/2-3\alpha}$, it is now seen from Eq. (24) that near T_c the relevant values of the amplitude Q remain of order $\epsilon^{-\alpha}$ also for large r_i, i.e., $\hat{Q}$ is then of order $\epsilon^{-1/2-\alpha}$. Hence, the imaginary time path $\overline{q(\sigma)}$ is of same order of magnitude as r_i. This and the decoupling of x_i' and r_i in the minimal effective action exemplify the advantage in using the coordinate transformation (17) to determine the stationary flux solution.

Now, Eq. (49) combines with the right hand side of Eq. (24) to yield for the cubic equation for the amplitude Q the explicit form

$$\frac{\Lambda}{\epsilon^{1/2}}Q-c_4\epsilon^{1/2}2\theta^3D_4Q^3+c_4\epsilon\frac{3\theta}{a}[F_{3,1}(t)$$
$$-16A(t)^3C_{3,1}(t)]Q^2r_i-c_4\epsilon^{3/2}\frac{3}{a^2\theta}$$

$$\times\left[\frac{1}{2}F_{2,2}(t)-2\overline{F_{2,2}}(t)+8\,\theta A(t)^2J_2(t)\right]Qr_i^2$$

$$+c_4\epsilon^2\frac{1}{4a^3\theta^3}[F_{1,3}(t)+4\,\theta^3A(t)J_1(t)]r_i^3$$

$$=i\,\frac{a}{2\,\theta}x_i'-\frac{\Lambda\,\omega_R}{2\,\theta a}r_i-i\,\frac{a}{2\,\theta}\Delta(r_i). \qquad (50)$$

The time dependent functions $F_{n,m}(t)$ and $\overline{F_{n,m}}(t)$ as well as $C_{n,m}(t)$ and $J_n(t)$ are given in Appendix A. The behavior of the amplitude Q as a function of temperature and coordinates following from a cubic equation of the above form has been discussed in detail in II and in [17] to where we refer for further details.

Since the x_i' coordinate remains small while r_i becomes large, it is convenient to set

$$Q=Q_x+Q_r, \qquad (51)$$

where Q_x and Q_r vanish for $x_i'=0$ and $r_i=0$, respectively. From Eq. (50) the amplitudes Q_x and Q_r are found to be determined by the coupled equations

$$\frac{\tilde{\Lambda}_1(Q_r,r_i)}{\epsilon^{1/2}}Q_x+2c_4\epsilon^{1/2}\theta^3D_4Q_x^3-\epsilon\frac{3\,\theta c_4}{a}[F_{3,1}(t)$$

$$-16A(t)^3C_{3,1}(t)]Q_x^2r_i=-i\,\frac{a}{2\,\theta}x_i' \qquad (52)$$

and

$$\frac{\Lambda_1(Q_x,0)}{\epsilon^{1/2}}Q_r+c_4\epsilon^{1/2}2\,\theta^3D_4Q_r^3-\epsilon\frac{3\,\theta c_4}{a}[F_{3,1}(t)-16A(t)^3C_{3,1}(t)]Q_r^2r_i+\epsilon^{3/2}\frac{3c_4}{2a^2\theta}[F_{2,2}(t)-4\overline{F_{2,2}}(t)$$

$$+16\theta A(t)^2J_2(t)]Q_rr_i^2-\epsilon^2\frac{c_4}{4a^3\theta^3}[F_{1,3}(t)+4\,\theta^3A(t)J_1(t)]r_i^3=\frac{\Lambda\,\omega_R}{2\,\theta a}r_i+i\,\frac{a}{2\,\theta}\Delta(r_i). \qquad (53)$$

In the above equations we have introduced

$$\tilde{\Lambda}_1(Q,r)=\Lambda_1(Q,r)+c_4\epsilon^{3/2}\frac{24A(t)^2}{a\,\theta}$$

$$\times\left[4\,\theta^2QrA(t)C_{3,1}(t)+\epsilon^{1/2}\frac{J_2(t)}{a}r^2\right], \qquad (54)$$

where

$$\Lambda_1(Q,r)=-\Lambda+6c_4\epsilon D_4\theta^3Q^2-\epsilon^{3/2}\frac{6\,\theta c_4}{a}F_{3,1}(t)Qr$$

$$+\epsilon^2\frac{3c_4}{2a^2\theta}[F_{2,2}(t)-4\overline{F_{2,2}}(t)]r^2. \qquad (55)$$

As mentioned previously, for high temperatures the coordinate r_i is of order $1/\sqrt{|\Lambda|}$ in the limit $\alpha\to 0$ and Eq. (50) becomes linear. Then, the part Σ_0 of the minimal effective action given in Eq. (45) leads to the harmonic result (I69) and (I74) while the second part Σ_1 in Eq. (46) becomes smaller than order 1 and can be neglected. This is no longer the case for temperatures $T\geqslant T_c$, where $|\Lambda|$ is of order ϵ or smaller and for all temperatures $T<T_c$.

C. Quantum fluctuations and semiclassical propagating function

With the minimal action (45) and (46) we have found the leading order term of the path integral for the propagating function (11). The path integral now reduces to integrals over periodic paths $\eta(0)=\eta(t)=0$ and $\eta'(0)=\eta'(t)=0$ in real time and $y(0)=y(\theta)=0$ in imaginary time. Thereby the relevant fluctuations give a contribution of order 1 to the full effective action. We have shown in I and II that the contribution of the real time fluctuations is given by the harmonic result (I75) also for temperatures near T_c, while a corresponding Gaussian approximation for the fluctuations about the imaginary time path is only valid for high temperatures. The simple semiclassical approximation diverges for temperatures near T_c where $\Lambda\to 0$. As discussed in II, this comes from the fact that the ϕ direction in function space becomes unstable. As a consequence, higher order contributions in the expansion of the imaginary time path about the minimal action path have to be taken into account. The detailed analysis is given in II. One finds for the contribution of the quantum fluctuations for temperatures near T_c

$$\int\mathcal{D}[\eta]\mathcal{D}[\eta']\mathcal{D}[y]\exp\left\{\frac{i}{2}(\Sigma[q,q',\overline{q}]\right.$$

$$\left.-\Sigma[q_{ma},q_{ma}',\overline{q_{ma}}])\right\}$$

$$=\frac{1}{8\pi|A(t)|}\frac{1}{\sqrt{4\pi\theta^2}}\left(\prod_{n=1}^{\infty}\nu_n^2u_n\right)K(Q). \qquad (56)$$

Here, we have decomposed an arbitrary real time path into $q(s)=q_{ma}(s)+\eta(s)$ where $q_{ma}(s)$ denotes the minimal action path in real time. Thereby, the paths $q(s)$ and $q(s')$ are related to the sum and difference paths considered in the previous sections by $r(s)=[q(s)+q'(s)]/2$ and $x(s)=q(s)-q'(s)$. Correspondingly, one has for the imaginary time path $\overline{q(\sigma)}=\overline{q_{ma}}(\sigma)+y(\sigma)$. The contribution of fluctuations in the marginal direction in function space is given by

$$K(Q)=\frac{1}{\sqrt{4\pi}}\int_{-\infty}^{\infty}dY\exp[-V(Q,Y)], \qquad (57)$$

which remains finite for $\theta=\theta_c$. The fluctuation potential for the marginal mode amplitude is obtained as

$$V(Q,Y)=\frac{1}{4}\left[\Lambda_1(Q,r_i)Y^2+2\theta^2 c_4 D_4 \epsilon^{3/2} Q Y^3 + \frac{\theta}{4} c_4 D_4 \epsilon^2 Y^4\right]+o(1), \tag{58}$$

where the eigenvalue $\Lambda_1(Q,r)$ is given in Eq. (55). Here, Q, which depends on x_i' and r_i, is determined by the cubic equation (50) or, equivalently, by Eqs. (52) and (53). For high temperatures Eq. (56) reduces to the harmonic result (I77). The features of the fluctuation potential $V(Q,Y)$ and various limits of the fluctuation integral $K(Q)$ as a function of coordinates and temperature are discussed in II.

Now, with the minimal action and the contribution of the quantum fluctuations evaluated for small ϵ we obtain the semiclassical propagating function (11) as

$$\tilde{J}(x_f,r_f,t,x_i',r_i)=\frac{N}{8\pi|A(t)|}K(Q) \times \exp\left\{\frac{i}{2}[\Sigma_0(x_f,r_f,t,x_i',r_i) - c_4\epsilon^2\Sigma_1(x_i',r_i,t)]\right\}. \tag{59}$$

Here,

$$N=\frac{1}{Z}\frac{1}{\sqrt{4\pi\theta^2}}\left(\prod_{n=1}^{\infty}\nu_n^2 u_n\right), \tag{60}$$

where Z is a normalization constant which cannot be calculated from the equilibrium density matrix near the barrier top but depends on the equilibrium distribution in the well region. The function Σ_0 is given in Eq. (45) and the function Σ_1 in Eq. (46). The fluctuation integral $K(Q)$ is defined in Eq. 57) and the marginal mode amplitude Q is determined by Eq. (50). While related to Eq. (II115), the result (59) gives only the reduced propagating function introduced in Eq. (11) but allows for larger values of r_i.

V. FORM FACTOR

Having evaluated the semiclassical propagating function, we are now able to determine the time dependent density matrix for large times. According to Eq. (9), the nonequilibrium state near the barrier top is given by the equilibrium density matrix $\rho_\theta(x_f,r_f)$ and the time dependent form factor $g(x_f,r_f,t)$. The equilibrium density matrix for coordinates near the barrier top was already evaluated in Eq. (II123). For the range of end coordinates considered here, namely, x_f and r_f smaller than order 1, this previous result reduces to

$$\rho_\theta(x_f,r_f)=NK(\bar{Q})\exp\left[\frac{i}{2}S_\theta(x_f,r_f)\right]. \tag{61}$$

The amplitude $\bar{Q}$ is determined by the cubic equation

$$\frac{\Lambda}{\epsilon^{1/2}}\bar{Q}-2\theta^3 c_4 D_4 \epsilon^{1/2}\overline{Q^3}=\frac{r_f}{2\theta} \tag{62}$$

apart from corrections smaller than order $\epsilon^{2/3}$. Furthermore, the fluctuation integral $K(\bar{Q})$ is given by Eq. (57) with a fluctuation potential $V(\bar{Q},Y)$ of the form (58), however, the eigenvalue $\Lambda_1(Q,r)$ replaced by $\Lambda_1(\bar{Q},0)$. According to Eq. (II125), the minimal Euclidean action reads

$$S_\theta(x_f,r_f)=i\left(\theta\frac{\overline{Q}r_f}{\epsilon^{1/2}}+\frac{\Omega}{2}x_f^2-c_4 2\theta^5 D_4\overline{Q^4}\right)+o(1), \tag{63}$$

where we have kept also the leading order x_f dependent term. Note that this action becomes of order 1 for coordinates of order $\epsilon^{1/2}$.

According to Eqs. (10), (59), and (61) the form factor takes the form

$$g(x_f,r_f,t)=\int dr_i U(x_f,r_f,t,r_i), \tag{64}$$

where

$$U(x_f,r_f,t,r_i)=\frac{1}{8\pi|A(t)|}\int dx_i'\frac{K(Q)}{K(\bar{Q})} \times\exp\left\{\frac{i}{2}[\Sigma_0(x_f,r_f,t,x_i',r_i) - c_4\epsilon^2\Sigma_1(x_i',r_i,t)-S_\theta(x_f,r_f)]\right\}\Theta(-r_i). \tag{65}$$

In the semiclassical limit the relevant contributions to the integrals come from those coordinates x_i' and r_i for which the exponent in Eq. (65) is of order 1 or smaller. Since the action $S_\theta(x_f,r_f)$ is of order 1 or smaller only for coordinates x_f,r_f of order $\epsilon^{1/2}$ or smaller, we may restrict ourselves to this range of end coordinates. Furthermore, we assume $|\Lambda|$ to be smaller than order ϵ and show later that the final result can be extended to larger values of $|\Lambda|$. We start by evaluating the x_i' integral (65) in the following way. First, we consider the integrand for fixed r_i with r_i smaller than order $\epsilon^{-1/2}$ (region 1). Second, values of r_i larger than order $\epsilon^{-1/2}$ (region 2) are investigated. Both results are then combined to determine $U(x_f,r_f,t,r_i)$. It is advantageous to use the decomposition $Q=Q_x+Q_r$ introduced in Eq. (51) where Q_x is determined by Eq. (52) and Q_r by Eq. (53). Also, from Eqs. (45) and (46), we write the minimal effective action in the form

$$\begin{aligned}\Sigma(x_f,r_f,t,x_i',r_i)&=\Sigma_0(x_f,r_f,t,x_i',r_i)-c_4\epsilon^2\Sigma_1(x_i',r_i,t)\\&=\Sigma_x(x_i',t)+\Sigma_r(x_f,r_f,t,r_i)+\Sigma_{xr}(x_i',r_i,t)\end{aligned} \tag{66}$$

with a x_i' dependent part $\Sigma_x(x_i',t)=\Sigma(0,0,t,x_i',0)$ governing the convergence of the x_i' integral, a x_i' independent part

$\Sigma_r(x_f,r_f,t,r_i)=\Sigma(x_f,r_f,t,0,r_i)$, and a term $\Sigma_{xr}(x_i',r_i)$ containing the remaining $x_i'r_i$ coupling terms.

A. Region 1: Small r_i

First let us consider the case $r_i=0$. Then, from Eq. (53) we see that $Q_r=0$ so that Q_x in Eq. (52) is a time independent function of x_i' only. Accordingly, we obtain from Eqs. (45) and (46)

$$\Sigma_x(x_i',t)=\Sigma_x(x_i')=-\frac{\theta Q_x}{\epsilon^{1/2}}ax_i'-2ic_4D_4\theta^5Q_x^4, \quad (67)$$

which is also independent of time and coincides after the formal replacement $iax_i'\to r_f$ with the diagonal part of the action (63) in the exponent of the equilibrium density matrix (61). In the same way, the fluctuation integral $K(Q)$ from Eq. (57) reduces to $K(\bar{Q})$ in the prefactor of the equilibrium density matrix (61) with $\bar{Q}$ substituted by Q_x. Further, $\Sigma_{xr}(x_i',0)=0$ and the remaining term in Eq. (66) is independent of x_i' and given by

$$\Sigma_r(x_f,r_f,t,0)=x_fr_f\omega_R+\frac{i}{2}\Omega x_f^2. \quad (68)$$

This is of order ϵ for coodinates of order $\epsilon^{1/2}$. Hence, one obtains for the x_i' integration in Eq. (65) for $r_i=0$ an integral of the form

$$\Upsilon\equiv\frac{a}{\sqrt{4\pi}}\int dx_i'K(Q_x)\exp\left[\frac{i}{2}\Sigma_x(x_i')\right]. \quad (69)$$

Here, a is the coefficient (14) in the transformation (17), and the prefactor is chosen for convenience. Since $K(Q_x)$ is at most of order $\epsilon^{-1/2}$, it is readily seen that the relevant contribution to the integral comes from those x_i' values that are of order $\epsilon^{1/2}$ or smaller. Correspondingly, Q_x is then at most of order 1. For high temperatures where $|\Lambda|$ is larger than order ϵ, the fluctuation integral reduces to $K(Q_x)=1/\sqrt{|\Lambda|}$ and the action to $\Sigma_x=-ia^2x_i'^2/2\Lambda$. Thus, we regain the harmonic result $\Upsilon=1$. For temperatures near T_c the integral Υ is of order 1 but the precise value must be calculated numerically. In terms of the integral Υ the function $U(x_f,r_f,t,r_i)$ defined in Eq. (65) reads for $r_i\to0$

$$U(x_f,r_f,t,r_i)=\frac{1}{\sqrt{4\pi}|S(t)|}\frac{\Upsilon}{K(\bar{Q})}\exp\left\{-\frac{i}{2}[\Sigma_r(x_f,r_f,t,0)-S_\theta(x_f,r_f)]\right\}\Theta(-r_i), \quad (70)$$

where we have kept $\Sigma_r(x_f,r_f,t,0)$, which is at most of order ϵ but contains the leading order x_fr_f dependent terms. Now, with increasing r_i the amplitude Q_r also increases and $x_i'r_i$ coupling terms might become relevant, i.e., of order 1 or larger. Inserting $Q=Q_x+Q_r$ into Eqs. (45) and (46), one finds that for x_i' of order $\epsilon^{1/2}$ and small r_i the leading order coupling terms in Σ_{xr} are removed by a function $\Delta\propto\epsilon^{1/2}Q_r$. From Eq. (53) we then conclude that Q_r is smaller than order 1 for coordinates r_i that are smaller than order $\epsilon^{-1/2}$. Consequently, the amplitude Q_x is independent of r_i apart from corrections smaller than order 1 and it can be determined from Eq. (52) for $Q_r=r_i=0$. Furthermore, Σ_{xr} and Σ_r are both smaller than order 1 so that the above analysis for $r_i\to0$ extends in the semiclassical limit to finite r_i values smaller than order $\epsilon^{-1/2}$.

We are interested in the leading order x_f dependence of the form factor which determines the flux (I125) across the barrier. Now, x_f dependent terms in Σ_r are at most of order ϵ and are therefore relevant for the r_i integration (64) only for small values of r_i. From Eqs. (45) and (46) we get for $|r_i|\ll1$

$$\Sigma_r(x_f,r_f,t,r_i)=\left[x_fr_f\omega_R+\frac{i}{2}\Omega x_f^2+\frac{i}{S(t)}r_fr_i\right][1+o(1)]. \quad (71)$$

Here, the second term proportional to x_f^2 is removed in the exponent of Eq. (65) by the x_f dependence of the action $S_\theta(x_f,r_f)$. Further, the Θ function restricts the r_i integration (64) to the halfplane $r_i\leqslant0$. Now, as in Eq. (I90) we perform a shift smaller than order 1,

$$r_i=r_i'+i\dot{S}(t)x_f. \quad (72)$$

In view of $\dot{S}(t)/S(t)=\omega_R$ following from Eq. (16), the exponent in Eq. (65) then becomes independent of x_f. Since this shift is smaller than order 1 it causes only additional x_f dependent terms in Σ_{xr} which are smaller than order $\epsilon^{3/2}$ and can therefore be neglected. Thus, one has for the r_i' dependent exponent in Eq. (65) in the region of small values of r_i'

$$\Sigma_r(x_f,r_f,t,r_i')+\Sigma_{xr}(x_i',r_i')-S_\theta(x_f,0)=\frac{i}{S(t)}r_fr_i'+O(\epsilon^{3/2}), \quad (73)$$

while the upper bound of the r_i' integration is given by $i\dot{S}(t)x_f$.

The above analysis gives the integral in Eq. (64) for all values of r_i' smaller than order $\epsilon^{-1/2}$ apart from corrections which are vanishing in the limit $\epsilon\to0$. We find

$$U(x_f,r_f,t,r_i)=\frac{1}{\sqrt{4\pi}|S(t)|}\frac{\Upsilon}{K(\bar{Q})}\exp\left[-\frac{i}{2}S_\theta(0,r_f)\right]\times\Theta[-r_i'-i\dot{S}(t)x_f]. \quad (74)$$

Due to the shift (72) the integrand in Eq. (64) is over r_i' values on a line parallel to the real axis where the argument of the Θ function is real.

B. Region 2: Large r_i

As a next step we consider the integrand in Eq. (65) for values of r_i which are larger than order $\epsilon^{-1/2}$ but at most of order $\epsilon^{-1/2-\alpha}$. In this region $x_i'r_i$ coupling terms in Σ_{xr} become of order 1 or larger and are therefore essential. Furthermore, for $\Delta(r_i)$ of order $\epsilon^{1/2-3\alpha}$ as assumed above, one readily sees from Eq. (53) that Q_r is then of order $\epsilon^{-\alpha}$.

Accordingly, r_i dependent terms in the cubic equation (52) for Q_x can no longer be neglected. It should be noted that in region 2 the coordinate r_i and the shifted coordinate r_i' coincide apart from terms that are negligible in the semiclassical limit. We now proceed in the following way. First, the amplitudes Q_x and Q_r are calculated in the approximation needed, and the as yet unknown function Δ is determined by the condition that leading order $x_i' r_i$-coupling terms in Σ_{xr} are removed. Afterwards, the x_i' integral is evaluated leading to $U(x_f,r_f,t,r_i)$ in region 2.

For x_i' of order $\epsilon^{1/2-\alpha}$ and Q_r of order $\epsilon^{-\alpha}$ Eq. (52) can be solved perturbatively using the ansatz

$$Q_x = Q_{x,0} + \epsilon^{2\alpha} Q_{x,1} + O(\epsilon^{5\alpha}), \tag{75}$$

where $Q_{x,0}$ and $Q_{x,1}$ are both of order ϵ^{α}. Inserting this ansatz into Eq. (52) we find

$$Q_{x,0} = -\frac{ia}{2\theta}\frac{\epsilon^{1/2} x_i'}{\tilde{\Lambda}_1(Q_{r,0},r)} \tag{76}$$

and

$$\epsilon^{2\alpha} Q_{x,1} = 3\theta c_4 \epsilon^{3/2} \frac{F_{3,1}(t) - 16A(t)^3 C_{3,1}(t)}{a\tilde{\Lambda}_1(Q_{r,0},r_i)} r_i Q_{x,0}^2 . \tag{77}$$

The leading order terms of the amplitude Q_r, that are relevant for the exponent in Eq. (65), can be evaluated in a similiar way using

$$Q_r = Q_{r,0} + \epsilon^{4\alpha} Q_{r,1} + O(\epsilon^{5\alpha}), \tag{78}$$

where $Q_{r,0}$ and $Q_{r,1}$ are both of order $\epsilon^{-\alpha}$ (for $\Lambda \to 0$). In leading order the amplitude $Q_{r,0}$ is independent of x_i' and given by the cubic equation (53) with $\Lambda_1(Q_x,0)=0$. For the next order term we simply obtain

$$\epsilon^{4\alpha} Q_{r,1} = -\frac{\Lambda_1(Q_{x,0},0)}{\tilde{\Lambda}_1(Q_{r,0},r_i)} Q_{r,0} . \tag{79}$$

Now, inserting Q_x and Q_r into Eqs. (45) and (46), we find for the dominant coupling terms of order $\epsilon^{-2\alpha}$ the result

$$\Sigma_{xr}(x_i',r_i,t) = \Sigma_{xr}^0(x_i',r_i,t)[1 + O(\epsilon^{2\alpha})] \tag{80}$$

with

$$\begin{aligned} \Sigma_{xr}^0(x_i',r_i,t) = {} & -\frac{\theta Q_{r,0}}{\epsilon^{1/2}} a x_i' + \frac{\theta Q_{x,0}}{\epsilon^{1/2}} a\Delta - x_i' r_i \omega_R \\ & - 8ic_4\theta^5 D_4 Q_{r,0}^3 Q_{x,0} + ic_4\epsilon^{1/2}\frac{12\theta^3}{a} Q_{r,0}^2 Q_{x,0} r_i \\ & \times\left[F_{3,1}(t) - 8A(t)^3 C_{3,1}(t) - \frac{\theta}{2}\gamma_i(t) D_4\right] \\ & + ic_4\epsilon\frac{6\theta}{a^2} Q_{r,0} Q_{x,0} r_i^2 [F_{2,2}(t) + \theta F_{3,1}(t)] \\ & + ic_4\epsilon^{3/2}\frac{Q_{x,0}}{a^3\theta} r_i^3 \Big\{ F_{1,3}(t) + \theta^2 A(t) J_1(t) \\ & - \frac{3}{2}\gamma_i(t)\theta[F_{2,2} - 4\overline{F}_{2,2}]\Big\} . \end{aligned} \tag{81}$$

Inserting $Q_{x,0}$ from Eq. (76), these terms are removed by choosing a function $\Delta \propto \epsilon^{1/2} Q_{r,0}^3$. In view of Eq. (53), this implies that $Q_{r,0} \propto \epsilon^{1/2} r_i$. To obtain explicit results we set

$$Q_{r,0} = \epsilon^{1/2} r_i q/2a \tag{82}$$

and

$$\Delta(r_i) = ic_4 \frac{\delta(q)}{a^4\theta}\epsilon^2 r_i^3 . \tag{83}$$

Inserting these expressions into Eq. (81), one obtains from the condition $\Sigma_{xr}^0 = 0$ that

$$\begin{aligned} \delta(q) = {} & \frac{5}{2}\theta^5 D_4 q^3 - 6\theta^3 q^2 \Big[F_{3,1}(t) - 12A(t)^3 C_{3,1}(t) \\ & - \frac{\theta}{4}\gamma_i(t) D_4\Big] - \frac{3}{2}\theta q[F_{2,2}(t) + 4\overline{F}_{2,2}(t) + 4\theta F_{3,1}(t) \\ & - 16\theta A(t)^2 J_2(t)] + 2\theta\omega_R \tilde{\lambda}_1(q) \\ & + \frac{3}{2}\gamma_i(t)[F_{2,2}(t) - 4\overline{F}_{2,2}(t)] - \theta A(t) J_1(t) \\ & - \frac{1}{\theta} F_{1,3}(t). \end{aligned} \tag{84}$$

Here, we have introduced the abbreviation

$$\tilde{\lambda}_1(q) = \tilde{\Lambda}_1(\epsilon^{1/2} q/2,a)/c_4\epsilon^2|_{\theta=\theta_c}. \tag{85}$$

This result is now inserted into the cubic equation (53) for $Q_{r,0}$, leading to a cubic equation for q. We obtain

$$\begin{aligned} & 3\theta^3 D_4 q^3 - 3\theta q^2 \Big\{\frac{5}{2} F_{3,1}(t) - \frac{\theta}{2} D_4[2\omega_R + \gamma_i(t)] \\ & \quad - 32A(t)^3 C_{3,1}(t)\Big\} - 3q\Big\{F_{3,1}(t)[2\omega_R + \gamma_i(t)] \\ & \quad - 32\omega_R A(t)^3 C_{3,1}(t) - 16A(t)^2 \frac{J_2(t)}{\theta} + \frac{4}{\theta}\overline{F}_{2,2}\Big\} \\ & = \frac{F_{1,3}(t)}{2\theta^3} + A(t)\frac{J_1(t)}{\theta} - \frac{\delta(0)}{\theta^2}. \end{aligned} \tag{86}$$

In particular, the above results confirm the assumptions made previously that Δ is at most of order $\epsilon^{1/2-3\alpha}$ while Q_r is of order $\epsilon^{-\alpha}$ or smaller for values of r_i that are at most of order $\epsilon^{-1/2-\alpha}$.

The remaining coupling terms in Σ_{xr} are of order 1. These terms are simply removed by scaling the x_i' coordinate in the following way. For Q_x of order ϵ^{α} and Q_r of order $\epsilon^{-\alpha}$ the fluctuation integral $K(Q)$ in Eq. (65) reduces to $K(Q)=K(Q_r)=1/\sqrt{\Lambda_1(Q_{r,0},r_i)}$ apart from corrections smaller than order 1. Hence, scaling the x_i' coordinate according to $x_i''=x_i'/\sqrt{\Lambda_1(Q_{r,0},r_i)}$ and inserting Q_x and Q_r as well as Δ from Eq. (83) into the action, one can show that $\Sigma_{xr}(x_i''\sqrt{\Lambda_1(Q_{r,0},r_i)},r_i,t)$ is independent of r_i. This combines with the contribution from Σ_x to yield the exponent for the x_i'' integration in Eq. (65)

$$\Sigma_x(x_i''\sqrt{\Lambda_1(Q_{r,0},r_i)},t)+\Sigma_{xr}(x_i''\sqrt{\Lambda_1(Q_{r,0},r_i)},r_i,t)$$
$$=\frac{i}{2}\zeta(t)a^2x_i''^2+o(1). \tag{87}$$

Here, the function $\zeta(t)$, which is positive and of order 1, is given in Appendix B. In view of the above result the x_i' integral in Eq. (65) becomes Gaussian with relevant contributions from the domain where x_i' is of order $\sqrt{\Lambda_1(Q_{r,0},r_i)}$ or smaller, i.e., x_i' is at most of order $\epsilon^{1/2-\alpha}$ as assumed previously.

Accordingly, we get in the semiclassical limit for values of r_i larger than $\epsilon^{-1/2}$ the result

$$U(x_f,r_f,t,r_i)=\frac{1}{\sqrt{4\pi\zeta(t)}|S(t)|K(\overline{Q})}\exp\left\{\frac{i}{2}[\Sigma_r(0,r_f,t,r_i')-S_\theta(0,r_f)]\right\}\Theta[-r_i'-i\dot{S}(t)x_f]. \tag{88}$$

Here, from Eqs. (45) and (46), the exponent Σ_r reads

$$\Sigma_r(0,r_f,t,r_i')=-ir_f\frac{r_i'}{|S(t)|}+ic_4\epsilon^2\frac{\sigma_r(t)}{a^4}r_i'^4, \tag{89}$$

where $\sigma_r(t)$ is given in Appendix B.

C. Stationary flux solution near T_c

To gain the form factor we now combine the results (74) and (88) for $U(x_f,r_f,t,r_i,x_i)$ in regions 1 and 2. It is useful to scale the r_i' coordinate according to $r_i''=r_i'\Upsilon$ in region 1 and $r_i''=r_i'/\sqrt{\zeta(t)}$ in region 2. Since both scaling factors are of order 1, these transformations do not modify the regions 1 and 2. Now, for r_f of order $\epsilon^{1/2}$ the action Σ_r in Eq. (89) is of order 1 if r_i'' is of order $\epsilon^{-1/2-\alpha}$, but Σ_r becomes smaller than order 1 for smaller values of r_i'' within region 2. Hence, the exponents in Eqs. (74) and (88) coincide near the boundary of regions 1 and 2. Furthermore, the Θ function in Eq. (74) may be approximated near the boundary by $\Theta[-r_i''-i\Upsilon\dot{S}(t)x_f]=\Theta[-r_i'']$, while in region 2 we have $\Theta[-r_i''-i\dot{S}(t)x_f/\zeta(t)^{1/2}]=\Theta[-r_i'']$ apart from negligible corrections. The results in region 1 and 2 can thus be matched to yield the form factor near T_c

$$g(x_f,r_f,t)=\frac{1}{\sqrt{4\pi}|S(t)|K(\overline{Q})}\int dr_i''\times\exp\left\{\frac{i}{2}[\Sigma_r(0,r_f,t,r_i''\zeta(t)^{1/2})-S_\theta(0,r_f)]\right\}\times\Theta[-r_i''-i\Upsilon\dot{S}(t)x_f]. \tag{90}$$

For small values of r_i'' in region 1 the x_f dependence of the argument of the Θ function is essential while Σ_r is smaller than order 1. Correspondingly, the above integrand reduces to Eq. (74). On the other hand, for r_i'' in region 2 the x_f dependence of the Θ function can be neglected and the integrand in Eq. (88) is regained for $\epsilon\to 0$.

The result (90) can be brought into a more convenient form. To this purpose we put

$$z=(r_i''-r_i^0)/|S(t)|, \tag{91}$$

where the shift r_i^0 is defined implicitly by

$$\Sigma_r(0,r_f,t,r_i^0\zeta(t)^{1/2})-S_\theta(0,r_f)=0. \tag{92}$$

For temperatures close to T_c where Λ is smaller than order ϵ, the r_f dependence of r_i^0 can be determined analytically. Then, the linear term in Eq. (62) can be neglected against the cubic one with the result

$$\overline{Q}=-\left(\frac{r_f}{4\theta^4c_4D_4\epsilon^{1/2}}\right)^{1/3}. \tag{93}$$

Inserting this solution into Eq. (63), one obtains from Eqs. (89) and (92)

$$r_i^0=\eta(t)S(t)\frac{2\theta}{\epsilon^{1/2}}\overline{Q}, \tag{94}$$

where $\eta(t)$ is determined by the quartic equation

$$\eta(t)^4\frac{2D_r(t)}{\theta D_4}-\eta(t)\zeta(t)^{1/2}+\frac{3}{4}=0 \tag{95}$$

with

$$D_r(t)=S(t)^4\frac{\sigma_r(t)}{a^4}\zeta(t)^2=16A(t)^4\sigma_r(t)\zeta(t)^2. \tag{96}$$

This way the exponent in Eq. (90) takes the form

$$\begin{aligned}\tilde{\Sigma}(r_f,z,t)&\equiv\Sigma_r(0,r_f,t,z|S(t)|\zeta(t)^{1/2}+r_i^0\zeta(t)^{1/2})-S_\theta(0,r_f)\\&=iD_r(t)\epsilon^2z^4-i\eta(t)D_r(t)8\theta c_4\epsilon^{3/2}\overline{Q}z^3\\&\quad+i\eta(t)^2D_r(t)24\theta^2c_4\epsilon\overline{Q}^2z^2\\&\quad+ir_fz\left(\frac{8D_r(t)}{\theta D_4}\eta^3(t)-\zeta(t)^{1/2}\right).\end{aligned} \tag{97}$$

We note that in deriving this equation one has to take into account that $S(t)<0$.

Now, with Eq. (97) we have transformed the exponent in Eq. (90) into a quartic polynomial in z with $\overline{Q}$-dependent coefficients. Clearly, for an initial equilibrium distribution, that is formally for $\Theta[\cdot]$ replaced by 1, the form factor must reduce to $g(x_f,r_f,t)=1$. This suggests to compare the exponent $\tilde{\Sigma}$ with the fluctuation potential $V(\overline{Q},z)$ in Eq. (58) which is also a quartic polynomial in z with the same $\overline{Q}$ dependence of the coefficients as in Eq. (97). We recall that the above results are valid provided the asymptotic formulas (15) and (16) for the functions $A(t)$ and $S(t)$ can be used. Then, a detailed analysis given in Appendix B shows that up to corrections which are negligible in the semiclassical limit, the functions $\zeta(t)$ and $D_r(t)$ are independent of time and given by

$$\zeta(t)=\zeta=1,\quad D_r(t)=D_r=\frac{\theta}{8}D_4. \tag{98}$$

As a consequence, one obtains from Eq. (95) the real and time independent solution $\eta(t)=\eta=1$, so that from Eq. (94) the shift $r_i^0=S(t)2\theta\overline{Q}/\epsilon^{1/2}$. We note that $2\theta\overline{Q}/\epsilon^{1/2}$ is the unscaled marginal mode amplitude $\hat{\overline{Q}}$. Now, we find that the exponent in Eq. (97) is also independent of time with

$$i\tilde{\Sigma}(r_f,z,t)/2=i\tilde{\Sigma}(r_f,z)/2=-V(-\overline{Q},z), \tag{99}$$

and the form factor may be written as

$$g(x_f,r_f,t)=\frac{1}{\sqrt{4\pi K(\overline{Q})}}\int dz\exp[-V(-\overline{Q},z)]\times\Theta[-z|S(t)|-r_i^0-i\Upsilon\dot{S}(t)x_f]. \tag{100}$$

The two relations in Eq. (99) are valid only in an intermediate region of time (plateau region). A lower bound comes from employing the asymptotic formulas (16) and (17) that are only valid for $1\ll\exp(\omega_R t)$. There is also an upper bound of time since corrections to the minimal effective action (41) must be smaller than order 1. Accordingly, we obtain from Eq. (42) the relation $\alpha<1/6$. In particular, anharmonic terms in the barrier potential (13) of the form $c_{2k}\epsilon^{2k-2}q^{2k}$ with $k>2$ are then smaller than order 1. Hence, in the limit of small ϵ the plateau region can be estimated, as far as orders of magnitude are concerned, by $\omega_R^{-1}\ll t\ll|\ln(\epsilon)|$. Furthermore, from the fluctuation potential (58), the relevant values of z in Eq. (100) are at most of order $\epsilon^{-1/2}$. Since the order of magnitude of r_i is given by $S(t)z$, this confirms the basic assumption that r_i is at most of order $\epsilon^{-1/2-\alpha}$.

Now, after the transformation $z'=z-i\Upsilon\omega_R x_f$ the integral (100) gives only contributions if $z'<r_i^0/S(t)$ where $S(t)<0$. Hence, the stationary form factor for temperatures near T_c may be written as

$$g_{fl}(x_f,r_f)=\frac{1}{\sqrt{4\pi K(\overline{Q})}}\int_{-\infty}^{u(x_f,r_f)}dz\exp[-V(-\overline{Q},z)], \tag{101}$$

where

$$u(x,r)=\frac{2\theta}{\epsilon^{1/2}}\overline{Q}+i\Upsilon\omega_R x. \tag{102}$$

Here, the fluctuation integral as well as the corresponding fluctuation potential are defined in Eqs. (57) and (58), respectively, with $\Lambda_1(Q,r)$ replaced by $\Lambda_1(Q,0)$. Further, the integral Υ is given in Eq. (69). Since $K(\overline{Q})$ is of order $\epsilon^{-1/2}$, it is readily seen that the width of the diagonal part $g_{fl}(0,r_f)$ is of order $\epsilon^{1/2}$. For larger positive coordinates r_f the function $u(0,r_f)$ is negative and larger than order 1 so that $g_{fl}(x_f,r_f)\rightarrow 0$. On the other hand, for negative r_f and $|r_f|$ larger than order $\epsilon^{1/2}$ the function $u(0,r_f)$ is positive and larger than order 1 so that $g_{fl}(x_f,r_f)\rightarrow 1$. In particular, $g_{fl}(0,0)=1/2$. We note that a formal continuation of the high temperature result (I102) would lead to a vanishing width at T_c showing again the breakdown of the harmonic approximation. Further, evaluating Eq. (97) in the region of time where Eq. (99) is valid and for $z=r_i^0/S(t)$, we find with $\Sigma_r(0,r_f,t,0)=0$ that

$$\frac{i}{2}S_\theta(0,r_f)=V(-\overline{Q},u(0,r_f)). \tag{103}$$

This identity holds also for high temperatures and ensures that the form factor describes a nonequilibrium state with a stationary flux across the barrier which is independent of position (i.e., of r_f) as will be seen below.

Although the calculation presented above was carried out for temperatures where $|\Lambda|$ is smaller than order ϵ, the fluctuation potential can now be used to extend Eq. (101) both to higher and lower temperatures. This way we gain the central result of this article, namely, the expression

$$\rho_{fl}(x_f,r_f)=\rho_\theta(x_f,r_f)\frac{1}{\sqrt{4\pi K(\overline{Q})}}\int_{-\infty}^{u(x_f,r_f)}dz\times\exp[-V(-\overline{Q},z)] \tag{104}$$

for the stationary flux solution valid from high temperatures down to temperatures slightly below T_c. Here, the equilibrium density matrix near the barrier top $\rho_\theta(x_f,r_f)$ is given in Eq. (61) and $u(x_f,r_f)$ in Eq. (102). For temperatures above T_c where $|\Lambda|$ is larger than order ϵ the solution of the cubic equation (62) reads $\overline{Q}=\epsilon^{1/2}r_f/2\theta\Lambda$. Then, the fluctuation potential reduces to $V(\overline{Q},z)=-\Lambda z^2/4$ and therefore $K(\overline{Q})=1/\sqrt{\Lambda}$. Hence, the high temperature result (I102) is regained. For temperatures below T_c the amplitude $\overline{Q}$ grows and terms such as $c_6\epsilon\overline{Q}^6$ neglected in the action (63) become of order 1. Accordingly, the above flux solution can be used only down to temperatures $T<T_c$ where Λ is smaller than order $\epsilon^{2/3}$.

D. Relation to equilibrium quantities

Before we proceed, let us collect the main formulas needed to get explicit values for $\rho_{fl}(x_f,r_f)$. For given damping mechanism $\hat{\gamma}(z)$ and inverse temperature θ the marginal mode amplitude $\overline{Q}$ is determined from the cubic equation

(62) as a function of r_f. This leads to the action $S_\theta(x_f,r_f)$ in Eq. (63) and the fluctuation potential

$$V(\overline{Q},z)=\frac{1}{4}\left[(-\Lambda+6c_4\epsilon D_4\theta^3\overline{Q^2})z^2+2\,\theta^2c_4D_4\epsilon^{3/2}\overline{Q}z^3+\frac{\theta}{4}c_4D_4\epsilon^2z^4\right] \tag{105}$$

from which we can gain the fluctuation integral $K(\overline{Q})$ introduced in Eq. (57). On the one hand, these quantities give the equilibrium density matrix

$$\rho_\theta(x_f,r_f)=NK(\overline{Q})\exp\left[\frac{i}{2}S_\theta(x_f,r_f)\right]. \tag{106}$$

On the other hand, they also determine the form factor of the stationary flux solution (104).

In particular, we may relate the function Υ defined in Eq. (69), which appears in formula (102) for $u(x_f,r_f)$, with equilibrium quantities. Scaling the integration variable x_i' according to $q=ax_i'$ and comparing the cubic equations for Q_x in Eq. (52) and Q in Eq. (62) as well as the exponent $\Sigma_x(x_i')$ in Eq. (67) with the action $S_\theta(0,r_f)$ in Eq. (63), we see that $Q_x(q)=Q(iq)$ and $\Sigma_x(q)=S_\theta(0,iq)$. Hence, Υ can be expressed in terms of the analytically continued equilibrium density matrix

$$\Upsilon=\frac{1}{\sqrt{4\pi}N}\int dq\rho_\theta(0,iq)=\frac{1}{\sqrt{4\pi}}\int dqK(\overline{Q(iq)})\exp\left[\frac{i}{2}S_\theta(0,iq)\right]. \tag{107}$$

Here, the amplitude $\overline{Q}$ must be evaluated from Eq. (62) with r_f replaced by iq. The analytic continuation of $\rho_\theta(0,q)$ leads to a convergent integral for coordinates within the barrier region. Moreover, the quantity Υ can be shown to be only a function of the scaled bifurcation parameter

$$\overline{\Lambda}=\Lambda/\sqrt{\epsilon^2 2c_4\theta D_4}. \tag{108}$$

From Eqs. (105) and (107) we find that Υ may be written as

$$\Upsilon(\overline{\Lambda})=\frac{1}{4\pi}\int d\overline{q}\,d\overline{y}\exp\left[-\overline{\Lambda}\widetilde{Q}^2+\frac{3}{2}\widetilde{Q}^4\right]\exp[-\overline{V}(\widetilde{Q},\overline{y})]. \tag{109}$$

Here, $\widetilde{Q}=\overline{Q}(2\theta^5c_4D_4)^{1/4}$ is determined from $\overline{\Lambda}\widetilde{Q}-\widetilde{Q}^3=i\overline{q}$ with $\overline{q}=q/(2\theta\epsilon^2c_4D_4)^{1/4}$, and the scaled fluctuation potential reads

$$\overline{V}(\widetilde{Q},\overline{y})=\frac{1}{4}\left[(-\overline{\Lambda}+3\widetilde{Q}^2)\overline{y}^2+\widetilde{Q}\overline{y}^3+\frac{\overline{y}^4}{8}\right]. \tag{110}$$

As a consequence, we obtain the important result that the stationary flux solution is completely determined by properties of the equilibrium density matrix near the barrier top evaluated already in II.

E. Form factor slightly below T_c

As we have already discussed in II and [17], for temperatures below T_c where Λ is larger than order ϵ, the fluctuation integral matches again onto a simple semiclassical approximation. Here, we consider the corresponding limit for the flux solution. First, we investigate the equilibrium density matrix (61) and afterwards the form factor (101).

For coordinates near the barrier top the cubic equation can be solved perturbatively for temperatures where Λ is larger than order ϵ. Using $\epsilon/\Lambda^{3/2}$ as a small parameter we obtain for the stable branches the approximate result

$$\overline{Q}_{\substack{se\\sn}}=\text{sgn}(-r_f)\left(\pm\overline{Q}_0+\frac{\epsilon^{1/2}}{4\theta\Lambda}|r_f|\mp\frac{\epsilon^{3/2}}{\Lambda^{5/2}}\frac{3r_f^2}{16\theta^2(2c_4\theta^3D_4)^{1/2}}\right)+O(\epsilon^{5/2}/\Lambda^4), \tag{111}$$

which is valid for end points r_f smaller than order $\Lambda^{3/2}/\epsilon$. Here, Q_{se} denotes the branch which extends from the high temperature region to lower temperatures while Q_{sn} is the branch which newly emerges near T_c. The amplitude $\overline{Q}_0$ of the positive stable branch for $r_f=0$ is given by

$$\overline{Q}_0=\left(\frac{\Lambda}{2c_4\epsilon\theta^3D_4}\right)^{1/2}. \tag{112}$$

Now, inserting Eq. (111) into the action (63) we have

$$S_{\substack{\theta;se\\sn}}(x_f,r_f)=-i\left(\frac{\Lambda}{\epsilon}\right)^2\frac{1}{2c_4\theta D_4}\mp i\left(\frac{2\Lambda}{\epsilon^2c_4\theta D_4}\right)^{1/2}|r_f|-i\frac{r_f^2}{4\Lambda}+\frac{i}{2}\Omega x_f^2+O(\epsilon/\Lambda^{5/2}). \tag{113}$$

Since the equilibrium density matrix near T_c and for coordinates near the barrier top is independent of the particular root of the cubic equation (62), it is convenient to evaluate the fluctuation potential at the stable branch $\overline{Q}_{se}$. Then, $V(\overline{Q}_{se},Y)$ exhibits two minima and one local maximum. The first minimum at $Y=0$ corresponds to Q_{se} while the second one at $Y_+=(Q_{sn}-Q_{se})2\theta/\epsilon^{1/2}$ is associated with the stable branch Q_{sn}. These minima are well separated for temperatures where Λ is larger than order ϵ by a local maximum the height of which is larger then order 1. Accordingly, in this temperature range the fluctuation potential may be written in the form [17]

$$V(\overline{Q}_{se},Y)=\frac{\Lambda}{2}Y^2 \tag{114}$$

for fluctuation amplitudes Y around $Y=0$ and

$$V(\overline{Q}_{se},Y)=\Delta S(r_f)+\frac{\Lambda}{2}(Y-Y_+)^2 \tag{115}$$

near $Y=Y_+$ with

$$\Delta S(r_f)\equiv\frac{i}{2}[S_{\theta;se}(0,r_f)-S_{\theta;sn}(0,r_f)]=\left(\frac{2\Lambda}{\epsilon^2 c_4\theta D_4}\right)^{1/2}|r_f|. \tag{116}$$

Hence, the fluctuation integral (57) reduces to

$$K(\overline{Q_{se}})=\frac{1}{\sqrt{2\Lambda}}\{1+\exp[-\Delta S(r_f)]\}. \tag{117}$$

This combines with Eq. (61) to yield the equilibrium density matrix for temperatures below T_c. For coordinates of order $\epsilon/\sqrt{\Lambda}$ or smaller we then have

$$\rho_\theta(x_f,r_f)=\frac{2N}{\sqrt{2\Lambda}}\exp\left[\frac{i}{2}S_{\theta;se}(x_f,0)\right]\cosh\left[\left(\frac{\Lambda}{2\epsilon^2 c_4\theta D_4}\right)^{1/2}r_f\right], \tag{118}$$

where N is given by Eq. (60). The typical width of the minimum is of order $\epsilon/\sqrt{\Lambda}$. For end points r_f within this region both contributions of the stable branches must be taken into account, while for larger end points the consistent semiclassical approximation is determined by the branch $\overline{Q_{se}}$ only. Accordingly, for r_f of order $\epsilon/\sqrt{\Lambda}$ or larger but smaller than order $\Lambda^{3/2}/\epsilon$ we gain

$$\rho_\theta(x_f,r_f)=\frac{N}{\sqrt{2\Lambda}}\exp\left[\frac{i}{2}S_{\theta;se}(x_f,0)\right]\times\exp\left[\left(\frac{\Lambda}{2\epsilon^2 c_4\theta D_4}\right)^{1/2}|r_f|+\frac{r_f^2}{8\Lambda}\right]. \tag{119}$$

Clearly, for end points of order $\epsilon/\sqrt{\Lambda}$ the result (118) matches onto Eq. (119).

Let us now turn to the form factor (101). Since $-\overline{Q_{se}}(q)=\overline{Q_{se}}(-q)$, the fluctuation potential $V(-\overline{Q_{se}},z)$ can be written in the form (114) and (115) with Y_+ replaced by $-Y_+$. From Eq. (103) we then find for $Y=u_{se}(0,r)$ and $Y+Y_+=u_{sn}(0,r)$, respectively,

$$\frac{\Lambda}{2}[u_{\substack{se\\sn}}(0,r)]^2=\frac{i}{2}S_{\substack{\theta;se\\sn}}(0,r). \tag{120}$$

A simple calculation using Eq. (113) leads in leading order to

$$u_{\substack{se\\sn}}(x,r)=\mathrm{sgn}(-r)\left(\pm\frac{\theta\overline{Q_0}}{\epsilon^{1/2}}+\frac{|r|}{\Lambda}\right)+i\Upsilon\omega_R x. \tag{121}$$

The integral Υ is expressed in Eq. (109) in terms of the analytical continued equilibrium density matrix $\rho_\theta(0,iq)$. Now, the results (118) and (119) can be used to evaluate Υ in the semiclassical limit. For convenience, we choose the matching point of both results at $r_f=\pi(\epsilon^2 c_4\theta D_4/2\Lambda)^{1/2}$ With $\cosh(ix)=\cos(x)$, where $x=q(\Lambda/2\epsilon^2 c_4\theta D_4)^{1/2}$, we then obtain from Eq. (109)

$$\Upsilon=\frac{1}{\sqrt{2\pi\Lambda^2}}\exp\left[\frac{i}{2}S_{\theta;se}(0,0)\right]\left[\int_0^{\pi/2}dx\cos(x)+\int_0^\infty dx\cos(x)\exp\left(-\frac{x^2}{8\Lambda^2}\right)+i\int_{\pi/2}^\infty dx\sin(x)\exp\left(-\frac{x^2}{8\Lambda^2}\right)\right]. \tag{122}$$

The second integral is exponentially small [of order $\exp(-\Lambda^2/\epsilon^2)$], while the third one can be estimated to be of order ϵ^2/Λ^2 which formally is of order $\hbar$. Hence, in the semiclassical limit, the first integral which is of order 1 leads to

$$\Upsilon=\frac{1}{\sqrt{2\pi\Lambda^2}}\exp\left(\frac{\overline{\Lambda}^2}{2}\right), \tag{123}$$

where $\overline{\Lambda}$ is given in Eq. (108).

Now, from Eq. (104) one gains with Eqs. (114)–(117) and (121) the form factor for temperatures slightly below T_c where a simple semiclassical approximation is again valid as

$$g_{fl}(x_f,r_f)=\frac{1}{2}\{1+\exp[-\Delta S(r_f)]\}^{-1}\times\left\{\mathrm{erfc}\left[-\left(\frac{\Lambda}{2}\right)^{1/2}u_{se}(x_f,r_f)\right]+\exp[-\Delta S(r_f)]\mathrm{erfc}\left[-\left(\frac{\Lambda}{2}\right)^{1/2}u_{sn}(x_f,r_f)\right]\right\}. \tag{124}$$

Here, $\mathrm{erfc}(x)=(2/\sqrt{\pi})\int_x^\infty dz\exp(-z^2)$. For $r_f<(>)0$ the function $u_{se}(0,r)$ is positive (negative) while $u_{sn}(0,r)$ is negative (positive) and the arguments of the erfc functions are both larger than order 1. Furthermore, for values of $|r_f|$ larger then order $\epsilon/\sqrt{\Lambda}$ the function $\exp[-\Delta S(r_f)]$ becomes exponentially small. Accordingly, we then have $g_{fl}(x_f,r_f)\approx\frac{1}{2}[1-\mathrm{sgn}(r_f)]$ as expected. In particular, we gain $g_{fl}(0,0)=1/2$ with $\Delta S(0)=0$. The width of the diagonal part $g_{fl}(0,r_f)$ of the nonequilibrium state is therefore of order $\epsilon/\sqrt{\Lambda}$ which is again of the same order as the width of the minimum of $\rho_\theta(0,r_f)$. Since the amplitude $\overline{Q_0}$ increases with increasing inverse temperature the above analytical results are restricted to temperatures below T_c where Λ is smaller than order $\epsilon^{2/3}$.

To illustrate these results we have depicted in Fig. 1 the diagonal part $g_{fl}(0,q)$ of the form factor for various temperatures and Ohmic damping with $\gamma=3$ and $\epsilon=0.05$. For these parameters $\theta_c=5.079\ldots.$ For $T/T_c=1.15$ above and $T/T_c=0.85$ below T_c the function $|\Lambda|$ is slightly larger than ϵ. As one sees, the width of $g_{fl}(0,q)$ decreases with decreasing temperature since thermal fluctuations also decrease. Near T_c the width is of order $\epsilon^{1/2}$ while below T_c the width saturates at a value of order ϵ.

Figure 2 shows the quantity Υ as a function of $\overline{\Lambda}$. Thereby, the integrals in Eq. (109) are evaluated numerically by inserting the branch $\overline{Q_{se}}$. For $\Lambda>0$ and coordinates q

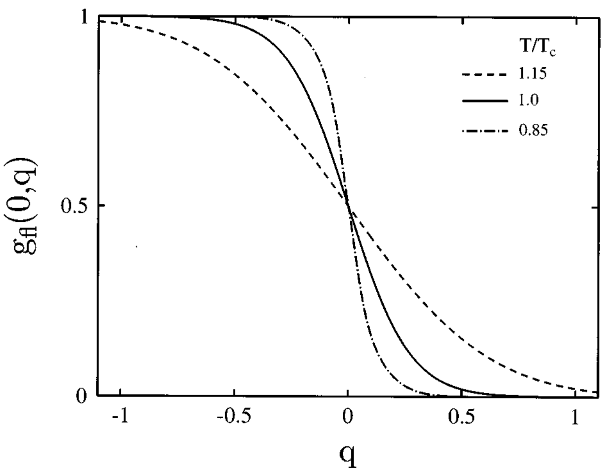

FIG. 1. Diagonal part $g_{fl}(0,q)$ of the form factor of the stationary flux solution (104) as a function of the scaled coordinate q for Ohmic damping with $\hat{\gamma}=3$ and various values of the temperature ratio T/T_c.

where $\exp[-\Delta S(q)]\ll 1$, the contribution of the branch $\overline{Q_{sn}}$, which corresponds to a second minimum in $V(\overline{Q_{se}},Y)$, is neglected according to Eq. (117). Note that in the semiclassical limit Υ is real. One sees that $\Upsilon=1$ up to negligible corrections down to temperatures near T_c. For temperatures below T_c, however, Υ increases exponentially according to Eq. (123). This reflects the fact that below T_c the equilibrium density matrix (118) depends strongly on the classical action $S_\theta(0,0)$ of nonlocal paths in the inverted potential and is no longer of a Boltzmann-like form. This means that below T_c quantum tunneling strongly enhances the probability to find states near the barrier top.

VI. MATCHING TO EQUILIBRIUM STATE IN THE WELL AND DECAY RATE

In the previous section we have found an analytical expression for the stationary flux state of a metastable system

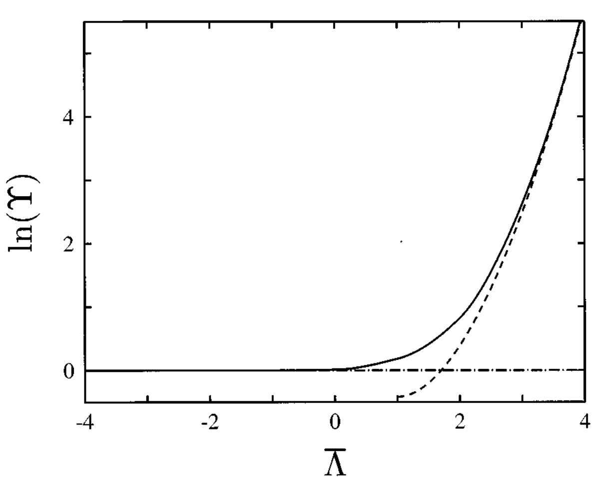

FIG. 2. The function Υ given in Eq. (69) as a function of the scaled bifurcation parameter $\overline{\Lambda}$ (solid line). The dashed line represents the approximate result (123) valid for temperatures slightly below T_c. See text for details.

valid from high temperatures down to temperatures slightly below T_c. As we have shown, the density matrix $\rho_{fl}(x_f,r_f)$ depends on local properties of the barrier potential only. Since, the metastable system is in thermal equilibrium in the well region, the flux state must reduce to a thermal equilibrium state for coordinates on the left side of the barrier at a distance from the barrier top smaller than the typical distance $1/\epsilon$ between the barrier top to the well bottom. In I we have shown that this implies a condition on the minimal damping strength. Here, we first give a corresponding condition for temperatures near T_c. Afterwards, this result is used to derive the decay rate out of the metastable state from the flux solution.

A. Matching to equilibrium state in the well

For coordinates q_f,q_f' on the left side of the barrier the form factor (101) must approach 1 as one moves away from the barrier top. In $x_f r_f$ coordinates this means that

$$|1-g_{fl}(x_f,r_f)|\ll 1 \tag{125}$$

for values of x_f,r_f away from the barrier top. One can estimate the region in the half-plane $r_f<0$ where Eq. (125) is valid following the lines of reasoning in I. We find

$$|x_f|<\frac{1}{2\Upsilon\,\omega_R\sqrt{\theta}}\left(\frac{2|r_f|}{\epsilon^2\theta c_4 D_4}\right)^{1/3}. \tag{126}$$

Further, from Eq. (61) the equilibrium density matrix is nonvanishing essentially only for

$$|x_f|<\frac{r_f^{2/3}}{\epsilon^{1/3}}\left(\frac{3}{\Omega}\right)^{1/2}\frac{1}{(4\theta c_4 D_4)^{1/6}}. \tag{127}$$

On the one hand, there should be values $r_f<0$ with $|r_f|\ll 1/\epsilon$ where the two conditions (126) and (127) hold simultaneously. On the other hand, we have to ensure that in Eqs. (126) and (127) the coordinate $|x_f|$ is also much smaller than $1/\epsilon$. To estimate this latter condition we consider values of r_f within the typical width of the diagonal part of the nonequilibrium state. Then, only relation (127) is relevant and yields

$$\Omega\gg\epsilon^2. \tag{128}$$

Now, a detailed analysis shows that Eq. (128) gives the most stringent condition on the minimal damping strength.

Up to this point we have used the dimensionless formulation introduced in Sec. II. Now, in order to facilitate a comparison with earlier results we shall return to *dimensional units* for the remainder of this section. Then, condition (128) reads

$$\Omega\gg\frac{\hbar\omega_0^2}{V_b}, \tag{129}$$

where ω_0 denotes the oscillation frequency at the barrier top, V_b the barrier height with respect to the well bottom, and Ω is given in Eq. (I111). Following the discussion in I, we make Eq. (129) more explicit by considering a Drude model with $\gamma(t)=\gamma\omega_D\exp(-\omega_D t)$. Since the condition (129) is relevant for small damping only, the function $\Omega(\beta,\gamma)$ can be expanded according to Eq. (I118). In particular, for vanishing damping one has $\theta_c \equiv \omega_0\hbar\beta_c = \pi$ which leads to $\Omega(\pi/\omega_0\hbar,0)=0$. To determine the critical inverse temperature for small damping from $\Lambda(\beta_c,\gamma)=0$ we expand Λ according to Eq. (I116). Then, we put $\omega_0\hbar\beta_c=\pi+\xi$ with $\xi\ll 1$ and obtain in leading order

$$\xi = -4\gamma\Lambda'(\pi/\omega_0\hbar), \tag{130}$$

where $\Lambda'(\beta)=\partial\Lambda(\beta,\gamma)/\partial\gamma|_{\gamma=0}$ is given in Eq. (I117). Note that the correction ξ is positive since $\Lambda'(\beta)<0$. With this value for β_c the condition (129) simply reads

$$\gamma \gg \frac{\hbar\omega_0}{V_b\kappa}, \tag{131}$$

where κ is given in Eq. (I121).

In the limit $\omega_D \gg \omega_0,\gamma$ the Drude model behaves like an Ohmic model except for very short times of order $1/\omega_D$. Then, near β_c one obtains $\kappa=\ln(\omega_D\hbar\beta)/\pi$ [see Eq. (I122)]. Accordingly, Eq. (131) reads

$$\gamma \gg \frac{\hbar\omega_0\pi}{V_b\ln(\omega_D\hbar\beta)}. \tag{132}$$

Comparing this with the high temperature result we see that both conditions coincide for temperatures slightly above T_c where Eq. (I123) is still valid. However, in contrast to the high temperature case, the region of damping where the flux state derived here becomes invalid is very narrow for temperatures near and below T_c.

B. Decay rate

If condition (131) is satisfied the flux solution can be used to determine expectation values, in particular, the decay rate Γ out of the metastable state. From Eq. (I124) one has in coordinate representation

$$\Gamma = \frac{J_{fl}}{Z} = \frac{\hbar}{iM}\left(\frac{\partial}{\partial x_f}\rho_{fl}(x_f,0)\right)\Bigg|_{x_f=0}. \tag{133}$$

Here, J_{fl} denotes the unnormalized flux at the barrier top. It is worthwhile to note that $r_f=0$ is chosen in Eq. (133) for simplicitly only since the flux is indeed independent of the particular value of r_f as can readily be verified with Eq. (103). The normalization constant Z is approximated as in I by the partition function of a damped harmonic oscillator with frequency ω_w

$$Z = \frac{1}{\omega_w\hbar\beta}\left(\prod_{n=1}^{\infty}\frac{\nu_n^2}{\nu_n^2+\nu_n\hat{\gamma}(\nu_n)+\omega_w^2}\right)\exp(\beta V_b). \tag{134}$$

Here, V_b denotes the dimensional barrier height with respect to the well bottom and ω_w is the well frequency. Inserting Eq. (104), which is valid for high temperatures as well as for temperatures slightly below T_c, into Eq. (133) we obtain in dimensional units

$$\Gamma = \frac{\omega_w}{2\pi}\frac{\omega_R\Upsilon}{\omega_0}\left(\prod_{n=1}^{\infty}\frac{\nu_n^2+\nu_n\hat{\gamma}(\nu_n)+\omega_w^2}{\nu_n^2+\nu_n\hat{\gamma}(\nu_n)-\omega_0^2}\right)\exp(-\beta V_b). \tag{135}$$

Here, the Grote-Hynes frequency ω_R [20] is given by the positive solution of $\omega_R^2+\omega_R\hat{\gamma}(\omega_R)=\omega_0^2$. When this is compared with the result (I127) we see that the anharmonicities of the barrier potential lead to an additional factor Υ in the vicinity of T_c. The function Υ is expressed in Eq. (107) in terms of the analytically continued equilibrium density matrix $\rho_\theta(0,iq)$ near the barrier top and depicted in Fig. 2. As already discussed in Sec. V E one has $\Upsilon=1$ from high temperatures down to temperatures near T_c while below T_c quantum tunneling causes an exponential increase of Υ. This behavior depends essentially on $S_\theta(0,0)$ that is the action of classical paths in the inverted barrier potential with $q(0)=-q(\theta)$ and $\dot{q}(0)=-\dot{q}(\theta)$. For $T>T_c$ the only possible path is then the trivial one $q(\sigma)=0$ with $S_\theta(0,0)=0$ while for $T<T_c$ oscillating solutions with different $q(0)$ and $\dot{q}(0)$ but the same nonvanishing action emerge.

Hence, for temperatures above T_c we recover from Eq. (135) the well-known result for thermally activated decay including quantum corrections [21] while for lower temperatures quantum tunneling leads to an enhancement of the rate. This behavior of the escape rate differs from predictions based on purely thermodynamic methods [8,9]. A semiclassical approximation of the functional integral for the partition function together with an analytic continuation according to Langer [10] yields for the fluctuation modes the eigenvalues $\lambda_n^b \equiv \nu_n^2+\nu_n\hat{\gamma}(\nu_n)-\omega_0^2$. The first eigenvalue λ_1^b vanishes at a temperature $T_0<T_c$. In the undamped case one has $T_0=T_c/2$. The eigenvalues λ_n^b are characteristic for the imaginary time motion in a harmonic barrier potential and the thermodynamic rate theory does not lead to an instability near T_c.

In the rate formula (135) the λ_n^b appear since around T_c only the marginal mode amplitude Q is affected essentially by anharmonicities of the potential. However, for lower temperatures the magnitude of the imaginary time path increases and anharmonicities are important for all eigenvalues of the second order variational operator in the fluctuation path integral. As a consequence, the rate formula (135) is then no longer valid and its corresponding extension to low temperatures exhibits no singularities for temperatures near and below T_0.

ACKNOWLEDGMENTS

The authors would like to thank G.-L. Ingold and E. Pollak for helpful discussions. This work was supported by the Deutsche Forschungsgemeinschaft (Bonn).

APPENDIX A: AUXILIARY FUNCTIONS

In this appendix we collect functions needed in Sec. IV. These functions can be evaluated for given inverse temperature θ and macroscopic damping kernel $\gamma(s)$ or Laplace transform $\hat{\gamma}(z)$.

The $F_{n,m}(t)$ and $\overline{F_{n,m}}(t)$ describing the influence of anharmonicities on the imaginary time motion are defined by

$$F_{n,m}(t)=\frac{2}{\theta}\int_0^\theta d\sigma\,\phi(\sigma)^n \times\left[\sum_{n=-\infty}^{\infty} u_n\cos(\nu_n\sigma)\int_0^t ds\,g_n(s)G_i(t,s)\right]^m \tag{A1}$$

and

$$\overline{F}_{n,m}(t)=\frac{2}{\theta}\int_0^\theta d\sigma\,\phi(\sigma)^n\left\{\frac{1}{4}(2\sigma-\theta)+\sum_{l=1}^{\infty} u_l\left[\frac{1-\nu_l\hat{\gamma}(\nu_l)}{\nu_l} + \int_0^t ds\,f_l(s)G_i(t,s)\right]\sin(\nu_l\sigma)\right\}^m. \tag{A2}$$

Here, the auxiliary functions $g_n(s)$ and $f_l(s)$ are defined in Eqs. (II29) and (II30), respectively, and $G_i(t,s)=G_+(t-s)/G_+(t)$.

To describe the anharmonic real time motion we need five classes of functions. The functions

$$\Gamma_m(t)=\int_0^t ds\,G_i(t,s)^m G_f(t,t-s)^{4-m}, \tag{A3}$$

where $G_f(t,s)=G_+(t)\partial G_i(t,s)/\partial t$ remain finite for vanishing damping. The other functions describe damping induced couplings between real time and imaginary time paths. The function $\gamma_i(t)$ was already introduced in Eq. (II83). Further,

$$C_{n,m}(t)=\int_0^t G_i(t,t-s)^n G_i(t,s)^m G_f(t,t-s)^{4-n-m} \times[C_1^+(s)-C_1^+(t)]^n \tag{A4}$$

and

$$\psi_{n,m}(t)=\int_0^t ds\,G_i(t,t-s)^n G_i(t,s)^m \times G_f(t,t-s)^{4-n-m}[\hat{R}(t,s)-\hat{R}(t,t)]^n. \tag{A5}$$

Here, the function $C_1^+(t)$ defined in Eq. (II102) reduces to $C_1^+(t)=-a$ for times where the asymptotic formulas (15) and (16) are valid. In this region of time one has

$$\hat{R}(t,s)=-2\omega_R a+\omega_R\frac{S(s)}{2A(s)}+a\frac{\dot{A}(s)}{A(s)} +\frac{1}{4\theta A(t)A(s)}\sum_{n=-\infty}^{\infty}\left\{u_n\cosh[\nu_n(t-s)] +\int_s^t ds'[A(s'-s)-A(s-s')]\cosh[\nu_n(t-s')]\right\}. \tag{A6}$$

Finally, the functions

$$I_{n,m}(t)=\int_0^t ds\,G_i(t,s)G_i(t,t-s)^{n+l}G_f(t,t-s)^{3-n-l} \times[C_1^+(s)-C_1^+(t)]^l[\hat{R}(t,s)-\hat{R}(t,t)]^n \tag{A7}$$

contain both $C_1^+(s)$ and $\hat{R}(t,s)$. These latter functions do not appear explicitly in the main text since for convenience we have introduced the linear combinations

$$J_1(t)=\frac{1}{2}C_{1,3}(t)-6a^2C_{1,1}(t)-24A(t)^2I_{2,1}(t) +24aA(t)I_{1,1}(t) \tag{A8}$$

and

$$J_2(t)=2A(t)I_{1,2}(t)+aC_{2,1}(t). \tag{A9}$$

The functions $\Gamma_n(t)$, $C_{n,m}(t)$, $\psi_{n,m}(t)$, and $I_{n,m}(t)$ are given as integrals over times $0\leqslant s\leqslant t$ with integrands which vanish at $s=0$ and $s=t$. The integrands become exponentially small for times s where the asymptotic formulas (15) and (16) are valid.

In general, these functions can be evaluated explicitly only numerically. However, in the limits of vanishing and very strong damping analytical results are available. Let us first consider the case of vanishing damping. Then, only the functions D_n defined in Eq. (47), and $\overline{F}_{n,m}(t)$ as well as $\Gamma_m(t)$ are finite while the other ones vanish. Since the auxiliary functions $g_n(s)=f_n(s)=0$, the functions $\overline{F}_{n,m}(t)$ are independent of time. Then, with $\phi=-\sin(\sigma)/2$ the integrals defining the relevant functions can be done analytically. We obtain

$$D_4=\frac{3}{64},\quad \overline{F}_{2,2}=\frac{\pi^2}{256}. \tag{A10}$$

The functions $\Gamma_m(t)$ are not relevant since the corresponding prefactors in the minimal effective action vanish in the undamped case.

For very strong damping the equation $z^2+z\hat{\gamma}(z)=1$ has a very small positive solution $\omega_R\approx 1/\gamma$ with $\gamma=\hat{\gamma}(0)$, while the negative solutions are of order γ or larger. As a consequence, the function $A(t)$ contains transient terms for times of order $1/\gamma$ or smaller only. Then, from the asymptotic form (15) we see that for times $1/\gamma\ll t\ll\gamma$ one has $A(t)\approx -1/2\gamma$. Hence, we gain in leading order in this range of time $\gamma_i(t)=\gamma$. Furthermore, with increasing damping strength also θ_c increases so that ν_1 becomes very small. Thus, the leading order terms of the functions defined above are determined by the static friction γ. As a consequence, for times $1/\gamma\ll t\ll\gamma$ these functions become in leading order independent of time with

$$D_4=\frac{2}{\theta^4},\quad F_{n,m}=\frac{2\gamma^m}{\theta^n},\quad \overline{F}_{2,2}=\frac{1}{24},\quad \overline{F}_{0,4}=\frac{\theta^4}{640}. \tag{A11}$$

The functions $\Gamma_n(t)$, $C_{n,m}(t)$, $\psi_{n,m}(t)$, and $I_{n,m}(t)$ are at most of order $1/\gamma$ since the integrands in these time integrals give essential contributions only within a time interval of order $1/\gamma$. We have evaluated the above functions also numerically for Ohmic damping with $\gamma(t)=2\gamma\delta(t)$. This shows that the results for strong damping can be used for $\gamma\gtrsim 4$.

APPENDIX B: $\zeta(t)$ AND $D_r(t)$

In Sec. V we have introduced two auxiliary functions $\zeta(t)$ and $D_r(t)$ given by

$$\zeta(t)=\frac{3}{\tilde{\lambda}_1(q)}\left\{D_4\theta^3q^2-\theta q\left(\frac{7}{4}F_{3,1}(t)-24A(t)^3C_{3,1}(t)-\frac{\theta}{4}[\gamma_i(t)+2\omega_R]D_4\right)+\frac{1}{4}F_{3,1}(t)\times[\gamma_i(t)+2\omega_R]\right.$$
$$\left.+\frac{1}{4\theta}[F_{2,2}(t)-8\overline{F_{2,2}}(t)]+\frac{8}{\theta}A(t)^2J_2(t)+8\omega_RA(t)^3C_{3,1}(t)\right\} \tag{B1}$$

and

$$D_r(t)=16A(t)^4\zeta(t)^2\sigma_r(t) \tag{B2}$$

with

$$\sigma_r(t)=\frac{9}{8}\theta^5D_4q^4+\theta^3q^3\left\{\frac{\theta}{2}D_4[8\omega_R+\gamma_i(t)]+32A(t)^3C_{3,1}(t)-\frac{5}{2}F_{3,1}(t)\right\}-12\theta^2q^2\left\{\frac{\overline{F_{2,2}}(t)}{4\theta}-\frac{\theta}{8}\omega_RD_4[2\omega_R+\gamma_i(t)]\right.$$
$$\left.+\frac{\theta}{16}F_{3,1}(t)[12\omega_R+\gamma_i(t)]-A(t)^2\frac{J_2(t)}{\theta}-10\omega_RA(t)^3C_{3,1}(t)\right\}-6\omega_Rq\left\{2\overline{F_{2,2}}(t)+\frac{\theta}{2}F_{3,1}(t)[2\omega_R+\gamma_i(t)]\right.$$
$$\left.-16\theta\omega_RA(t)^3C_{3,1}(t)-8A(t)^2J_2(t)\right\}+\frac{1}{8\theta^3}[16\overline{F_{0,4}}(t)-F_{0,4}(t)]+\frac{3}{2\theta}[F_{2,2}(t)-4\overline{F_{2,2}}(t)][2\omega_R+\gamma_i(t)]-\frac{1}{4\theta^2}F_{1,3}(t)$$
$$\times[4\omega_R-\gamma_i(t)]-\omega_RA(t)J_1(t)+\frac{48}{\theta}\omega_R^2J_2(t)+\frac{1}{2}A(t)\psi_{1,3}(t)-8A(t)^3\psi_{3,1}(t)-a^3\Gamma_3(t)+\frac{a}{4}\Gamma_1(t)-12aA(t)^2\psi_{2,1}(t)$$
$$-6A(t)a^2\psi_{1,1}(t). \tag{B3}$$

Obviously, the functions $\zeta(t)$ and $D_r(t)$ depend explicitly on $A(t)$ but not on $S(t)$. After determining the solution q of the cubic equation (86) they can be calculated numerically with the functions defined in Appendix A.

In the sequel we study ζ and D_r for vanishing and strong damping which allows for analytical results. First, for vanishing damping one has $A(t)=-\sinh(t)/2$ and $S(t)=aA(t)$ so that the transient term $\exp(-t)$ in $A(t)$ and $S(t)$ decreases on the same time scale on which the asymptotic term increases in time. As a consequence, $\dot{A}(t)/A(t)$ contains exponentially small terms for times $t\gg 1$ which are, however, not negligible, and one must resubstitute ω_R by $\dot{A}(t)/A(t)$ in the above formulas. This way using Eq. (A10) the cubic equation (86) reduces to

$$\pi q^3+q^2\frac{\dot{A}(t)}{A(t)}-\frac{q}{3\pi}=\frac{\dot{A}(t)}{3\pi^2A(t)}. \tag{B4}$$

This equation has three real solutions

$$q_1=-\frac{\dot{A}(t)}{\pi A(t)},\quad q_{2,3}=\pm\frac{1}{\sqrt{3}\pi}. \tag{B5}$$

From Eq. (55) one readily sees that only the solution q_1 is stable while the other ones are unstable. Inserting q_1 into Eq. (85) one obtains $\tilde{\lambda}_1(q_1)=3\pi/64$ up to exponentially small corrections. Further, evaluating then ζ from Eq. (B1) one finds $\zeta(t)=\zeta=1$. To determine D_r we have to take into account that for vanishing damping $\theta_c=\pi$ so that $a=\cot(\theta_c/2)/2=S(t)=0$. Since $S(t)/a$ remains finite for large times, the function $D_r(t)$ is nonvanishing also for vanishing damping. Now, due to $a=0$ the terms containing the functions $\Gamma_n(t)$ vanish. Then, inserting q_1 into Eq. (B3) and collecting the remaining terms one obtains

$$\sigma_r(t)=\frac{3\pi}{512}\left(\frac{\dot{A}(t)^2}{A(t)^2}-1\right)^2=\frac{3\pi}{512}\frac{1}{\sinh(t)^4}. \tag{B6}$$

This leads with Eq. (B2) to $D_r(t)=D_r=3\pi/512$. On the other hand Eq. (A10) leads to $\theta D_4/8=3\pi/512$ and thus to Eq. (98) in this limit.

For very strong damping and times $1/\gamma\ll t\ll\gamma$ one can set $\dot{A}(t)/A(t)=1/\gamma$. Accordingly, we gain in leading order from Eq. (86) in this range of time

$$q^3-2\frac{\gamma}{\theta}q^2-\frac{\gamma^2}{\theta^2}q=0. \tag{B7}$$

The only solution which is stable and extends continuously to the high temperature solution $q=-1/\gamma\theta$ is given by

$q_1=0$. A more detailed analysis taking into account also next order terms shows that $q_1 \propto O(1/\gamma)$. Then, we obtain from Eq. (B1)

$$\zeta(t)=\zeta=\frac{3}{\tilde{\lambda}_1(0)}\frac{\gamma^2}{\theta^3}[1+O(1/\gamma)]=1+O(1/\gamma). \quad \text{(B8)}$$

Furthermore, one has from Eq. (B3)

$$\sigma_r(t)=\sigma_r=\frac{\gamma^4}{4\theta^3}[1+O(1/\gamma)], \quad \text{(B9)}$$

which leads to $D_r=1/4\theta^3$. On the other hand, Eq. (A10) gives $\theta D_4/8=1/4\theta^3$.

This way we have shown analytically for vanishing and very strong damping in the region of time where the asymptotic formulas (15) and (16) are valid that $\zeta=1$ and $D_r=\theta D_4/8$ up to corrections negligible in the semiclassical approximation. Since we were not able to verify these relations in general, we have performed numerical calculations for Ohmic damping with $\gamma(t)=2\gamma\delta(t)$ confirming the validity of Eq. (98).

[1] P. Hänggi, P. Talkner, and M. Borkovec, Rev. Mod. Phys. **62**, 251 (1990).

[2] H. A. Kramers, Physica **7**, 284 (1948).

[3] W. H. Miller, J. Chem. Phys. **62**, 1899 (1975); W. H. Miller, Adv. Chem. Phys. **25**, 69 (1974).

[4] A. O. Caldeira and A. J. Leggett, Phys. Rev. Lett. **46**, 211 (1981); A. O. Caldeira and A. J. Leggett, Ann. Phys. (NY) **149**, 374 (1983); **153**, 445(E) (1984).

[5] A. I. Larkin and Yu. N. Ovchinnikov, Pis'ma Zh. Éksp. Teor. Fiz. **37**, 322 (1983) [JETP Lett. **37**, 382 (1983)]; A. I. Larkin and Yu. N. Ovchinnikov, Zh. Éksp. Teor. Fiz. **86**, 719 (1984) [Sov. Phys. JETP **59**, 420 (1984)].

[6] H. Grabert, U. Weiss, and P. Hänggi, Phys. Rev. Lett. **52**, 2193 (1984); H. Grabert and U. Weiss, *ibid.* **53**, 1787 (1984).

[7] E. Pollak, Phys. Rev. A **33**, 4244 (1986); E. Pollak, Chem. Phys. Lett. **177**, 178 (1986).

[8] H. Grabert, P. Olschowski, and U. Weiss, Phys. Rev. B **36**, 1931 (1987).

[9] P. Hänggi and W. Hontscha, J. Chem. Phys. **88**, 4094 (1988); Ber. Bunsenges. Phys. Chem. **95**, 379 (1991).

[10] J. S. Langer, Ann. Phys. (N.Y.) **41**, 108 (1967); M. Stone, Phys. Lett. **67B**, 186 (1977); C. G. Callan and S. Coleman, Phys. Rev. D **16**, 1762 (1977); S. Coleman, in *The Whys of Subnuclear Physics*, edited by A. Zichichi (Plenum, New York, 1979).

[11] J. Ankerhold, H. Grabert, and G. -L. Ingold, Phys. Rev. E **51**, 4267 (1995), referred to as I in the following.

[12] J. Ankerhold and H. Grabert, Phys. Rev. E **52**, 4704 (1995), referred to as II in the following.

[13] U. Weiss, *Quantum Dissipative Systems* (World Scientific, Singapore, 1993).

[14] R. P. Feynman and F. L. Vernon, Ann. Phys. (N.Y.) **243**, 118 (1963).

[15] A. O. Caldeira and A. J. Leggett, Physica **121A**, 587 (1983).

[16] H. Grabert, P. Schramm, and G.-L. Ingold, Phys. Rep. **168**, 115 (1988).

[17] J. Ankerhold and H. Grabert, Physica **A188**, 568 (1992).

[18] J. Ankerhold, Ph.D. thesis, University of Essen, 1994 (unpublished).

[19] We refer to Eq. (x) in I (Ref. [7]) and II (Ref. [8]) as (Ix) and (IIx), respectively.

[20] R. F. Grote and J. T. Hynes, J. Chem. Phys. **73**, 2715 (1980); P. Hänggi and F. Mojtabai, Phys. Rev. A **26**, 1168 (1982).

[21] P. G. Wolynes, Phys. Rev. Lett. **47**, 968 (1981); P. Hänggi, H. Grabert, G. -L. Ingold, and U. Weiss, *ibid.* **55**, 761 (1985).