

# Tunnelling in Reaction Theory: The Effect of Memory Friction

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## Abstract

The role of memory friction on dissipative quantum reaction rates is emphasized. The domain of low temperatures, bearing a variety of important tunnelling phenomena, is difficult to handle analytically. In order to fill this gap, we present new scaling relations for the bounce length  $\tau_b$  as a function of the dissipation renormalized barrier frequency  $\mu$ . In addition, we propose a simple estimate for  $\tau_b$ , which in turn yields directly the thermal low temperature enhancement of the quantum decay rate for Ohmic friction, as well as for memory friction. Finally the range of validity of the theory is discussed and pictorially represented in a rate phase diagram.

## 1. Introduction

Processes that are inhibited by the presence of a potential barrier are ubiquitous in many areas of natural sciences, ranging from nuclear physics over low-temperature solid-state physics and chemical reactions up to biological processes. It is well known and a matter of daily experience, that reactive processes proceed easier by raising the temperature. In addition, the system under consideration is usually not isolated, but coupled to a huge number of environmental degrees of freedom, which thereby act as a thermal reservoir. Thus, not only the temperature will modify the reactive process, but also the coupling to the environment is expected to influence strongly the behaviour of the system. The last decade has revealed increasing interest in such problems from a theoretical [1–7, 11–20] as well as experimental [8, and references therein] point of view. It was shown by Caldeira and Leggett [1], that at zero temperature the tunnelling rate of a metastable system coupled to a bath of harmonic oscillators is diminished, as one would expect intuitively. The next step has been the investigation of the effect of finite temperatures on dissipative tunnelling [2]. It is found, that at very low temperatures the decay rate of a system subject to Ohmic-like friction is enhanced following a universal exponential  $T^2$ -law, irrespective of the detailed dissipative mechanism [2]. Finally the next intriguing question was the behaviour at the linkage between low temperatures and higher temperatures [3, 4], where the activation rate obeys the Arrhenius Law.

## 2. Quantum reaction rate theory: A survey

First consider a particle in a one-dimensional metastable well. At sufficiently high temperatures  $T$  the decay proceeds via *thermal activation* and the rate of decay obeys the well known Arrhenius Law

$$\Gamma = \frac{\omega_0}{2\pi} \exp\left(-\frac{V_b}{kT}\right) \quad (1)$$

where  $V_b$  is the barrier height and  $\omega_0$  is the (attempt-)frequency of small oscillations in the well. Lowering the temperature would therefore result in a decrease of the decay rate, reaching  $\Gamma = 0$  at  $T = 0$ . Obviously this is not the full truth, since

one knows from quantum mechanics that the particle may leave its metastable state by *tunnelling*, leading to a small but finite decay rate even at  $T = 0$ . Intuitively it is clear, that a crossover temperature  $T_0$  must exist at which one decay mechanism becomes predominant over the other in a smooth manner. The magnitude of the crossover temperature  $T_0$  may be deduced by using a harmonic approximation for the barrier, and comparing the familiar WKB result for tunnelling through an inverted parabola of effective height  $V_b$  with the Arrhenius factor

$$\exp\left(-\frac{2\pi V_b}{\hbar\omega_b}\right) \leftrightarrow \exp\left(-\frac{V_b}{kT}\right) \quad (2)$$

Then the undamped crossover temperature is readily found to be given by

$$T_0 \equiv \frac{\hbar\omega_b}{2\pi k} \quad (3)$$

where  $\omega_b > 0$  is the angular frequency at the barrier. When the particle is subject to friction, i.e. it is coupled to a heat bath consisting of a set of harmonic oscillators, the relation for  $T_0$  remains valid, provided  $\omega_b$  is replaced by its dissipation renormalized value  $\mu$  given as the largest positive solution of [5]

$$\mu^2 + \mu\hat{\gamma}(\mu) = \omega_b^2. \quad (4a)$$

Equation (4a) can be recast into the form

$$\mu = \left(\omega_b^2 + \frac{\hat{\gamma}^2(\mu)}{4}\right)^{1/2} - \frac{\hat{\gamma}(\mu)}{2}, \quad (4b)$$

where the hat denotes the Laplace Transform of the classical memory-friction  $\gamma(t)$ , see (5) below. One may look upon eq. (4a) as being the definition of the normal mode barrier frequency induced by the coupling to the environmental bath oscillators giving rise to the damping  $\gamma(t)$  [6].

### 2.1. The decay rate from $T = 0$ up to finite temperatures

In the following we will be interested in the decay dynamics of a system, that classically obeys the following equation of motion

$$M\ddot{q} + M \int_0^t \gamma(t-s)\dot{q}(s) ds + \frac{dV}{dq} = 0. \quad (5)$$

Here  $\gamma(t)$  is a damping kernel describing memory friction exerted by the environment on the particle and  $V(q)$  is a metastable potential (see Fig. 1). It has been shown, that a dynamics as described by eq. (5) can indeed be modelled by coupling the system to an infinite set of harmonic oscillators [1].

The complete macroscopic statistical information about system plus environment in thermal equilibrium is contained

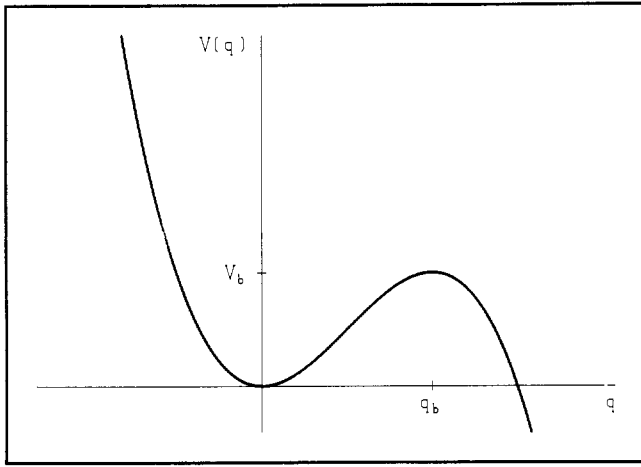


Fig. 1. A metastable potential as considered in the text.  $q_b$  locates the maximum  $V_b$  of the potential barrier.

in the partition function  $Z$ . In terms of functional integrals the quantity  $Z$  may be written as

$$Z = \int D[q(\tau)] \exp\left(-\frac{1}{\hbar} S[q(\tau), \hat{\gamma}]\right) \quad (6)$$

with the effective Euklidean action ( $\beta \equiv 1/kT$ )

$$S = \int_{-\hbar\beta/2}^{\hbar\beta/2} d\tau \left( \frac{M}{2} \dot{q}^2 + V(q) + \frac{1}{2} \int_{-\hbar\beta/2}^{\hbar\beta/2} d\tau' K(\tau - \tau') q(\tau) q(\tau') \right) \quad (7)$$

The integral runs over all periodic paths  $q(\tau)$  with period  $\hbar\beta$ . The trace over the environmental degrees of freedom has already been carried out, resulting in the last term of  $S$ . The kernel  $K(\tau)$  describes the influence of the environment on the system and is related to the classical damping  $\gamma(t)$  by [2, 3]

$$K(\tau) = \frac{M}{\hbar\beta} \sum_{n=-\infty}^{\infty} |v_n| \hat{\gamma}(v_n) e^{iv_n\tau} \quad (8)$$

where

$$v_n = \frac{2\pi n}{\hbar\beta}$$

and  $\hat{\gamma}$  again denotes the Laplace Transform of the memory damping  $\gamma(t)$ . The functional integral (eq. (6)) is dominated by such periodic paths which extremalize the effective action  $S$ , and the partition function  $Z$  is consequently given by the sum of the individual contributions  $Z = \sum_i Z_i$ .

The effective action  $S$  may be viewed as describing the motion of a particle in the inverted potential  $-V(q)$  (see Fig. 1), under the influence of the kernel  $K(\tau)$ . Obviously there are two trivial solutions, namely  $q(\tau) = 0$  and  $q(\tau) = q_b$ , where the particle sits at the top, and in the well of the potential  $-V(q)$ , respectively. The only non-trivial solution is an oscillatory motion of the particle in the well of  $-V(q)$  around  $q_b$ , the so called bounce  $q_B(\tau)$ . The bounce exists only below the crossover temperature, as will become clear later, whereas the trivial solutions are always present.

A detailed analysis shows however, that  $q(\tau) = q_b$  and  $q(\tau) = q_B(\tau)$  are not minima but saddlepoints of the effective action  $S$ . This means, that there are fluctuation modes in function space around  $q_b$  and  $q_B(\tau)$  respectively, associated with a negative eigenvalue, which would lead to a divergent

integral in the corresponding contribution to the partition function.

As was explained by Langer [9], the partition function of a metastable system is not defined as it stands, instead one has to resort to an analytic continuation, which leads from the stable to the unstable situation and thereby induces an imaginary part for the partition function  $Z$ . Finally it turns out that the imaginary part of the free energy  $F$  is given by

$$\text{Im } F = -kT \frac{\text{Im } Z_S}{Z_0} \quad (9)$$

where  $Z_0$  and  $Z_S$  are the contributions to the partition function  $Z$  of  $q(\tau) = 0$  and  $q(\tau) = q_B(\tau)$  and/or  $q(\tau) = q_b$ , respectively. Just like in field theory, the imaginary part of  $F$  is related to the decay rate of the metastable state via  $\Gamma \propto \text{Im } F$ .

## 2.2. $T > T_0$ : thermal activation

First one needs the explicit relation between the rate of decay and the imaginary part of the Free Energy  $F$ . For temperatures above  $T_0$  this is

$$\Gamma = -\frac{2}{\hbar} \frac{T_0}{T} \text{Im } F \quad (10)$$

as was shown first by Affleck [10] for an undamped system, i.e.,  $\gamma(t) = 0$ .

In the preceding section we have already found the solutions which may contribute to the statistical sum  $Z$ . The bounce however cannot exist for  $T > T_0$ , since the required period  $\hbar\beta = \hbar/kT < \hbar/kT_0 = 2\pi/\mu$  is shorter than the period of small oscillations  $2\pi/\mu$  in the dissipation renormalized well around  $q_b$ . Therefore the relevant paths are  $q(\tau) = 0$  and  $q(\tau) = q_b$ .

Taking into account fluctuations one finds with the help of eqs. (9) and (10) and the relation for the crossover temperature  $T_0$  (eq. (3)) [3-5]

$$\Gamma = \frac{\omega_0}{2\pi} \frac{\mu}{\omega_b} Q e^{-V_b/kT}. \quad (11)$$

Here,  $Q$  denotes the quantum enhancement factor

$$Q = \prod_{n=1}^{\infty} \frac{n^2 v^2 + \omega_0^2 + nv\hat{\gamma}(nv)}{n^2 v^2 - \omega_b^2 + nv\hat{\gamma}(nv)}$$

with  $v = 2\pi/\hbar\beta$  and  $V_b = V(q_b)$ .

Before discussing this result, we observe that the denominator of the infinite product may eventually vanish, indicating the breakdown of validity of formula (11). The most probable candidate for this event is the factor with  $n = 1$  and the breakdown condition reads  $v^2 - \omega_b^2 + v\hat{\gamma}(v) = 0$ ; but this is precisely the definition of the crossover temperature  $T_0$  [5] (see eq. (3)). The crossover is therefore characterized by the appearance of a new classical solution, the bounce.

Let us now inspect eq. 11 more closely. The factor  $Q$  is a quantum correction to the classical result found by Grote and Hynes [11], and by Hänggi and Mojtabai [12] using different lines of reasoning.  $Q$  approaches unity for  $T \gg T_0$ , but can be quite large even at temperatures of a few  $T_0$ , thus yielding important quantum corrections to the classical hopping rate. A simple but useful approximation of the quantum enhancement factor  $Q$  is given by [5]

$$Q \cong \exp\left[\frac{\hbar^2}{24} \cdot \frac{(\omega_0^2 + \omega_b^2)}{(kT)^2} + O(T^{-4})\right]. \quad (12)$$

Note that the approximation of order  $T^{-2}$  does not depend on the detailed dissipation mechanism which affects only the  $T^{-4}$  correction. From eqs. (11) and (12) it is seen, that the effect of the quantum correction factor is to lower the barrier height  $V_b$

$$V_b \rightarrow V_b - \frac{\hbar^2 (\omega_0^2 + \omega_b^2)}{24 kT}.$$

### 2.3. $T \cong T_0$ , the crossover region

In the proximity of the crossover temperature  $T_0$  the evaluation of the rate becomes rather difficult due to the appearance of (quasi) zero modes, which must be treated by considering the variation of  $S$  up to third and fourth order [3, 4, 13].

Slightly above  $T_0$ , the classical solution  $q(\tau) = q_b$  gives rise to two quasi zero fluctuation modes. Just below  $T_0$  we have apart from  $q(\tau) = q_b$  the bounce solution  $q(\tau) = q_B(\tau)$ . This means that every linear combination of these two solutions, approaching  $q_b$  for  $T \uparrow T_0$ , is an almost stationary point of the effective action  $S$ . Here one has one exact zero mode and one quasi zero mode.

Fortunately the crossover region is very narrow on the scale of  $T_0$ , provided the barrier is high and the potential is rather smooth. So in general there will be no need to investigate this region, except if one is interested in the detailed behaviour of the rate around the crossover temperature.

### 2.4. $T < T_0$ , quantum tunnelling

Below the crossover temperature we must take into account all of the solutions extremalizing  $S$ : the two trivial solutions  $q(\tau) = 0$ ,  $q(\tau) = q_b$  and the bounce. However it can be shown, that the action of the bounce is smaller than the action of  $q(\tau) = q_b$ , such that the contribution of the trivial saddle point may be disregarded against the bounce contribution. The rate of decay is then given by

$$\Gamma = A \frac{\{\text{Det} [\delta^2 S / \delta q^2]_{q(\tau)=0}\}^{1/2}}{\{\text{Det}' [\delta^2 S / \delta q^2]_{q(\tau)=q_B(\tau)}\}^{1/2}} \cdot \exp \{-S_B(\hbar\beta, \hat{\gamma})/\hbar\},$$

$$T < T_0 \quad (13)$$

with

$$A = \left\{ \frac{M}{2\pi\hbar} \int_{-\hbar\beta/2}^{\hbar\beta/2} \dot{q}_B^2(\tau) d\tau \right\}^{1/2}.$$

Here  $S_B(\hbar\beta, \hat{\gamma})$  is the bounce action  $S(q_B(\tau))$ , and the prime indicates that the zero eigenvalue, which is accounted for by the factor  $A$ , has been omitted in the determinant. At the crossover temperature  $T_0$  the bounce action  $S_B/\hbar$  matches smoothly with the Arrhenius factor  $V_b/kT$  [2]. An analytic evaluation of eq. (13) is possible only for a cubic potential at zero friction [1, 10, 13] and at very strong Ohmic damping [1, 4, 13], and at one particular moderate friction value [13]. In practice one has to resort to a numerical evaluation (see Ref. [14] for  $T = 0$  and Refs. [15] and [16] for  $0 < T < T_0$ ).

Just as in the case of  $T > T_0$ , where the prefactor  $Q$  decreases with increasing temperature, one finds that the prefactor of the decay rate for  $T < T_0$  (eq. (13)) becomes smaller with increasing temperature [17]. This very small temperature dependence of the prefactor, however does not offset the leading temperature enhancement given by the exponential part. Grabert, Weiss and Hänggi [2] have

shown that this temperature enhancement at low temperatures follows a universal power law

$$S(T, \hat{\gamma}) = S(T = 0, \hat{\gamma}) - \frac{\pi}{3} \alpha_0 M \omega_b \Delta q^2 \left( \frac{kT\tau_B}{\hbar} \right)^2,$$

$$\alpha_0 \equiv \frac{\gamma_0}{2\omega_b} \quad (14)$$

where in terms of the undamped tunnelling distance  $\Delta q$ , with  $V(0) = V(\Delta q) = 0$  (see Fig. 1), the bounce length  $\tau_B$  is defined as

$$\Delta q \tau_B = \int_{-\infty}^{\infty} q_B(T = 0, \hat{\gamma}; \tau) d\tau \quad (15)$$

The  $T^2$  enhancement law holds for all systems with  $\hat{\gamma}(\omega = 0) = \gamma_0 > 0$ , i.e., Ohmic like damping.

## 3. Quantum reaction theory: the effect of memory friction

In most cases the assumption of frequency-independent damping is a very poor approximation, since barrier frequencies  $\omega_b$  are often of the order  $10^{11}$ – $10^{14}$  s $^{-1}$ , and forces exerted by the environment generally will be correlated on this same time scale. A more realistic model for dissipative mechanism is therefore memory damping, and in the sequel we will focus on the archetype of memory dissipation, the Drude form

$$\gamma(t) = \gamma_0 \frac{\exp(-|t|/\tau_c)}{\tau_c}. \quad (16)$$

The corresponding Laplace transform reads

$$\hat{\gamma}(\mu) = \frac{\gamma_0}{\mu\tau_c + 1}. \quad (17)$$

Inserting eq. (17) into the definition for the dissipation renormalized barrier frequency  $\mu$  (eq. (4a)) yields a cubic equation

$$\left( \frac{\mu}{\omega_b} \right)^3 \omega_b \tau_c + \left( \frac{\mu}{\omega_b} \right)^2 + \left( \frac{\mu}{\omega_b} \right) \left\{ \frac{\gamma_0}{\omega_b} - \omega_b \tau_c \right\} - 1 = 0. \quad (18)$$

The largest real positive root of this equation immediately yields the crossover temperature  $T_0$  via eq. (3). As one would expect,  $\mu$  approaches  $\omega_b$  from below when  $\tau_c \rightarrow \infty$  (zero friction case), and approaches  $\mu = (\omega_b^2 + \gamma_0^2/4)^{1/2} - \gamma_0/2$  for  $\tau_c \rightarrow 0$  (Ohmic damping); see also eq. (4b).

### 3.1. Above the crossover temperature

For temperatures above  $T_0$  the decay rate has been found to be given by eq. (11). For the Drude damping (eq. (16)), the quantum correction factor  $Q$  (see eq. (11)) may be approximated as

$$Q = \exp \left\{ \frac{\pi^2}{6} (\omega_0^2 + \omega_b^2) \left( \frac{\hbar}{2\pi kT} \right)^2 \right. \\ \left. + \frac{\pi^4}{90} \left[ \frac{1}{2} (\omega_b^4 - \omega_0^4) - \frac{\gamma_0}{\tau_c} (\omega_0^2 + \omega_b^2) \right] \left( \frac{\hbar}{2\pi kT} \right)^4 \right. \\ \left. + \zeta(5) \frac{\gamma_0}{\tau_c^2} (\omega_0^2 + \omega_b^2) \left( \frac{\hbar}{2\pi kT} \right)^5 + O(T^{-6}) \right\} \quad (19a)$$

where  $\zeta$  is the Riemann Zeta Function.

One notes that damping affects the quantum correction

factor only at the order  $O(T^{-4})$ . Thus, memory damping influences the rate mainly via the dissipation renormalized diffusive transmission  $\kappa = \mu/\omega_b$ . Based on explicit calculations of the inclusion the fourth- and fifth-order correction in eq. (19a) does not yield a better approximation to the exact product (eq. (11)) over the simple estimate (eq. (12)), consisting only of the first term in eq. (19a), i.e., the dissipation independent contribution of order  $O(T^{-2})$ . Therefore, for most applications, the estimate

$$Q \cong \exp \left\{ \frac{(\omega_b^2 + \omega_b^2)}{24} \left( \frac{\hbar}{kT} \right)^2 \right\} \quad (19b)$$

provides a simple working expression of sufficient accuracy to describe the quantum rate enhancement at  $T > T_0$ .

### 3.2. Below $T_0$

As was already mentioned in Section 3.3, it is very difficult to obtain the decay rate explicitly in the temperature range from  $T = 0$  up to  $T_0$ . On the other hand it is just this low temperature domain, where important tunnelling phenomena, particularly in solid state physics, occur. Therefore it would be very desirable to have a simple, yet sufficiently accurate estimate for the rate of decay in this very low-temperature regime. Recently such a method, termed Sudden-Transition-State-Theory, has been proposed [6] to attack this problem. It does not resort to the difficult problem of finding the bounce solution, but instead starts from the well known WKB formula for an inverted parabola. The recipe at  $T = 0$  is very simple [6, 18]:

(1) Use the WKB formula for an inverted parabola (see eq. (2)) with frequency  $\mu$  ( $\mu$  being the dissipation renormalized frequency of the barrier under consideration).

(2) To obtain the rate of decay of the metastable potential, multiply the WKB exponent by the factor  $\Delta q^2/\Delta q_h^2$ , where  $\Delta q_h$  is the harmonic tunnelling length defined by

$$V_b \equiv \frac{M}{2} \omega_b^2 \left( \frac{\Delta q_h}{2} \right)^2 \quad (20)$$

i.e., the exponent of the decay rate scales with the square of the undamped tunnelling distance.

At zero temperature the agreement between instanton method and sudden-transition-state-theory is remarkably good for Ohmic as well as for memory damping at weak dissipation strength  $\gamma_0$  [18]. The  $T = 0$  estimate reads

$$\frac{S(\hat{\gamma}) - S(\hat{\gamma} = 0)}{M\omega_b\Delta q^2} \cong \frac{\pi}{4} \left( \frac{\omega_b}{\mu} - 1 \right), \quad T = 0 \quad (21)$$

where, as before,  $\Delta q$  is the undamped tunnelling distance under the metastable potential, see in Fig. 1.

The decay rate at low temperatures is obtained by considering the rate from the ground bath state and the first excited state, where one bath mode is excited, while the others are in their ground state.

Starting from the Sudden-TST approach of Ref. [6] we find for Ohmic damping the explicit temperature dependence of the thermal enhancement factor, defined by

$$\Delta B \equiv - \frac{S(T) - S(T = 0)}{M\omega_b\Delta q^2} \cong \frac{3\alpha_0}{2\pi} \frac{T^2}{T_0^2}, \quad \alpha_0 \equiv \frac{\gamma_0}{2\omega_b}. \quad (22)$$

The  $T$  dependence of the thermal enhancement agrees with

the general result of Ref. [2] (see eq. (14))

$$\Delta B = \frac{\pi\alpha_0}{3} \left( \frac{kT\tau_B}{\hbar} \right)^2, \quad (23)$$

where  $\tau_B$  is the zero temperature bounce length defined in eq. (15). Clearly the dissipation mechanism affects  $\tau_B$ , that means  $\tau_B = \tau_B(\hat{\gamma})$ , such that the effect of memory is entirely contained in  $\tau_B$ .

Next we define the harmonic bounce time  $\tau_B^h$ , to be the time required for a particle to perform a periodic orbit in a harmonic potential with curvature  $\mu$ , evidently this is

$$\tau_B^h \equiv \frac{2\pi}{\mu}. \quad (24)$$

What we are looking for is a simple estimate for the bounce length  $\tau_B(\hat{\gamma})$  in terms of the harmonic bounce time  $\tau_B^h$ , that is in terms of the dissipation renormalized barrier frequency  $\mu$ . With respect to their definitions, we expect these quantities to be proportional, i.e.,

$$\tau_B(\hat{\gamma}) \propto \tau_B^h; \quad \tau_B(\hat{\gamma}(\omega) = \gamma_0) = \tau_B(\gamma_0). \quad (25)$$

Put differently, we suppose eq. (22) to be valid approximately also for memory damping, in spite of the fact that eq. (22) has been derived for the Ohmic case only, i.e.,  $\hat{\gamma}(\omega) = \gamma_0$ . To this end we observe that  $T_0$  is directly proportional to  $\mu$  (see eqs. (3) and (4)), i.e.,  $T_0$  is *inversely* proportional to the harmonic bounce time  $\tau_B^h$ .

From eqs. (24) and (25) we obtain the following scaling relations, comparing Ohmic to memory friction

$$\frac{\tau_B(\hat{\gamma})}{\tau_B(\gamma_0)} \cong \frac{\mu(\gamma_0)}{\mu(\hat{\gamma})}. \quad (26)$$

The ratio of the damped to the undamped ( $\hat{\gamma} = 0$ ) bounce length is then given by

$$\frac{\tau_B(\hat{\gamma})}{\tau_B(\hat{\gamma} = 0)} \cong \frac{\omega_b}{\mu} \xrightarrow{\hat{\gamma}(\mu) \gg \omega_b} \frac{\gamma_0}{\omega_b} \quad (27)$$

and approaches  $\gamma_0/\omega_b$  for strong damping.

Substituting  $T_0$  in eq. (22), with  $\mu$  given by eq. (4), and comparing to the general result eq. (23) we obtain an estimate for  $\tau_B(\hat{\gamma})$ , i.e.,

$$\tau_B(\hat{\gamma}) = \frac{\sqrt{18}}{\mu} \cong \frac{4}{\mu}. \quad (28a)$$

Using eq. (4b), this is recast as

$$\tau_B(\hat{\gamma}) \cong \frac{4}{\omega_b[(\hat{\alpha}^2(\mu) + 1)^{1/2} - \hat{\alpha}(\mu)]}; \quad \hat{\alpha}(\mu) \equiv \frac{\hat{\gamma}(\mu)}{2\omega_b}. \quad (28b)$$

For a cubic potential (i.e.,  $\omega_0 = \omega_b$ ) and Ohmic damping ( $\hat{\alpha}(\mu) = \alpha_0$ ) the bounce time  $\tau_B(\hat{\gamma})$  has been approximated in Ref. 18 to read

$$\tau_B(\gamma_0) = \frac{12}{\pi\omega_b} \frac{1}{(\alpha_0^2 + (3/\pi)^2)^{1/2} - \alpha_0}. \quad (29)$$

In this case eq. (28b) overestimates the correct value by about 5%, inherent in the difference of factors  $12/\pi < 4$ .

## 4. The rate phase diagram

Finally let us review the range of validity of the treatment on the decay problem given above.

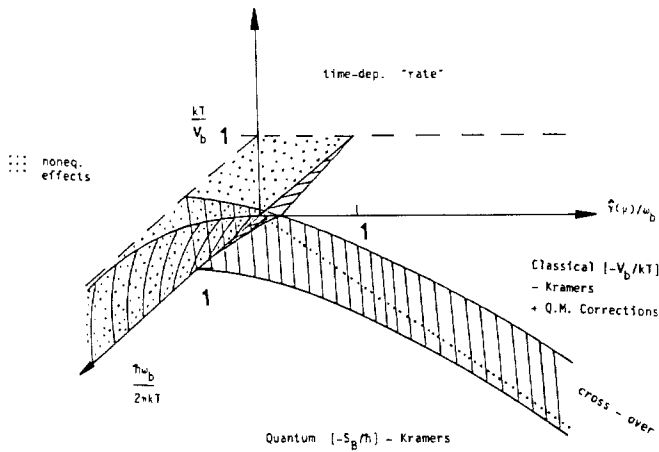


Fig. 2. The rate phase diagram. The crossover cylinder (hatched vertically) separates the classical thermal activation regime (including quantum corrections) from the tunnelling dominated regime in which the role of the Arrhenius factor is taken over by the dissipative bounce action  $S_B(T; \hat{\gamma})$  (eq. (13)). The volume (dotted) at the left side corner indicates the regime as a function of damping, barrier height and temperature in which weak damping induces nonequilibrium effects that are not accounted for by the thermodynamic rate formula  $\Gamma \propto \text{Im} F$  (imaginary free energy method). For  $kT/V_b > 1$ , the rate is generally time dependent, i.e.,  $\Gamma = \Gamma(t)$ .

The starting point was the equilibrium partition function. The theory therefore clearly can neither account for time dependent rates arising when  $E > kT$ , nor for nonequilibrium effects, which occur for very weak damping  $\hat{\gamma}(\mu) < \omega_b$ , since in this case the thermal equilibration inside the well would take a time much longer than the lifetime of the metastable state. However for high barriers, i.e.,  $kT/V_b \ll 1$  the range of validity is enlarged to lower friction values, determined by  $\hat{\gamma}(\mu) > kT\omega_b/V_b$ , since a high barrier implies an extreme long time scale for escape, allowing for the thermalization in the well. For temperatures below crossover, i.e.,  $T < T_0$ , the lifetime of the metastable state is long enough to allow for thermalization at practically all damping values.

The physics of decay problems may be summarized in a rate phase diagram ("Thomas diagram") [20] shown in Fig. 2. The dotted volume marks the region of nonequilibrium effects, and  $kT/V_b = 1$  defines the surface above which the rate generally becomes time dependent. The vertically hatched cylindrical surface separates the tunnelling region from the activation dominated domain and approaches asymptotically for strong damping the dotted line defined by

$$\frac{\hbar\omega_b}{2\pi kT_0(\hat{\gamma})} = \frac{\omega_b}{\mu} \xrightarrow{\hat{\gamma}(\mu) \gg \omega_b} \frac{\hat{\gamma}(\mu)}{\omega_b}.$$

## 5. Conclusion

In this article on decay rates we particularly emphasized the effect of memory friction. Especially in the analytically difficult and hardly accessible region below the crossover temperature we found it desirable to give scaling relations between the bounce time  $\tau_B(\hat{\gamma})$  and the dissipation renormalized barrier frequency  $\mu$  (see eqs. (26) and (27)). Moreover, we proposed an estimate for the bounce length  $\tau_B(\hat{\gamma})$  in the presence of memory friction (see eqs. (28a) and (28b)) in order to cover the thermal enhancement of the tunnelling rate (see eq. (23)) not only at Ohmic damping, but also at memory friction. We hope that the scaling relations and the estimate for  $\tau_B(\hat{\gamma})$  will prove to be useful for future experiments.

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