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Fission decay rates from a quantal transport equation

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The decay of a metastable system is described by extending Kramers' method to the quantal regime. For temperatures above twice the crossover value we recover the result known from applying Euclidean path integrals to solvable models. Our derivation is not restricted to a linearly coupled heat bath of oscillators, and thus applicable to nuclear systems.

In his famous paper Kramers [1] derived a formula which took into account the implications of dynamics on the decay rate of a metastable system. Applied to the situation portrayed in fig. 1 it can be written as

$$R_{\mathrm{K}} = \frac{\varpi_{\mathrm{a}}}{2\pi}\left(\sqrt{1+\eta_{\mathrm{b}}^2}-\eta_{\mathrm{b}}\right)\exp(-\beta B)\,. \qquad (1)$$

Here, $\beta=(kT)^{-1}$, B is the barrier height and ϖ_{a} the frequency of the motion around the potential minimum (fulfilling the relation $M_{\mathrm{a}}\varpi_{\mathrm{a}}^2=C_{\mathrm{a}}=\partial^2V(Q)/\partial Q^2|_{\mathrm{a}}$ with M_{a} being the inertia). $\eta_{\mathrm{b}}=\gamma_{\mathrm{b}}/2\sqrt{M_{\mathrm{b}}|C_{\mathrm{b}}|}$ measures the effective damping rate, calculated at the barrier from the friction coefficient γ_{b}, the inertia M_{b} and the stiffness coefficient $C_{\mathrm{b}}=\partial^2V(Q)/\partial Q^2|_{\mathrm{b}}$.

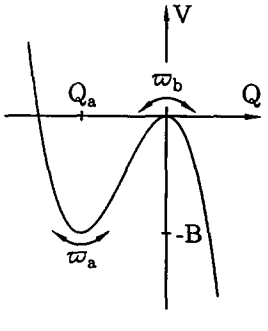

Fig. 1. A schematic plot of the effective potential.

Expression (1) is valid for damping rates η_{b} not smaller than about 0.2. Furthermore, one has to assume the barrier high enough to keep the decay rate R sufficiently small. Under such conditions the process can be viewed as quasi-stationary with the outward flux j_{b} at Q_{b} being constant for macroscopically large times. The decay rate R is then simply given by the ratio of the current divided by the number of particles N_{a} caught in the well near Q_{a}, $R=j_{\mathrm{b}}/N_{\mathrm{a}}$.

It has always been a challenge to extend Kramers' result to the quantal regime. However, it was only after one had learned to apply the instanton trick to this problem that a decent solution could be found. Among the vast literature on this subject of "dissipative tunneling" we would only like to refer to the classic paper [2], to ref. [3] as a general recent re-

view and to ref. [4] as a reference to formal details. Common to all these approaches is the application of the technique of path integrals for imaginary time propagation. This is feasible if for the basic Hamiltonian one assumes a form like

$$\hat{\mathscr{H}}(x_i, p_i, Q, P) = \hat{H}_B(x_i, p_i) + \hat{H}_{SB}(x_i, p_i, Q, P) + \hat{H}_S(Q, P), \quad (2)$$

in which two restrictions are imposed. First, the coupling is assumed to be of the form $H_{SB}=QF$ with $F=\sum_i c_i x_i$. Secondly, the heat bath must be represented by a set of oscillators. It is needless to stress that some measures have to be taken in order to render the final equations of motion irreversible.

A first major step to unify these results with the usual concepts of transport theory has been undertaken in refs. [5,6]. Still using path integrals, the author has been able to obtain from real time propagation a quantal version of Kramers' stationary solution for the inverted oscillator, which he could then use to rederive the form of the known quantal correction factor. This derivation is valid above a critical temperature $T_c=2T_0$ with T_0 being the so called crossover value [3,4]. We will come back to the physical significance of this restriction.

A full-fledged transport theory has been formulated in ref. [7] within a quasi-classical approach based on a quantal Langevin equation. Unfortunately, the authors have not been able to establish a direct connection to the results mentioned above. This will be the main goal of the discussion to come. We will proceed by exhibiting first the formal details, and then discuss later both the physical conditions of the derivation as well as consequences for possible applications.

Let us begin by formulating the decay rate in terms of Wigner functions $k^W(\beta; Q, P)$. Borrowing from the ideas of Kramers we may write

$$R \equiv \frac{j_b}{N_a} = \frac{\int_{-\infty}^{\infty} dP (P/M) k^W_{glob}(\beta; Q \sim Q_b = 0, P)}{\int_{Q_a - \Delta}^{Q_a + \Delta} dQ \int_{-\infty}^{\infty} dP\, k^W_{glob}(\beta; Q, P)}. \quad (3)$$

Here $k^W_{glob}(Q, P)$ is meant to represent the full global solution to our problem. Under the conditions mentioned already above, and for not too small temperatures (see below), this global function can be approximated by local ones valid at the barrier or at the potential minimum, respectively. They are obtained by appropriate linearizations. Trivially an overall normalization factor drops out of eq. (3) and it is almost obvious that the local solutions should not be normalized in themselves. In this context it is useful, for a moment, to think of a stationary situation. Then $k^W_{glob}(Q, P)$ would simply be the Wigner transform of $\hat{\kappa}_{glob} = \exp(-\beta \hat{\mathscr{H}})$. As a consequence of this transformation one gets a prefactor (to the exponential factor depending on the coordinates and momenta), which in the end may be found from the partition function $\mathscr{Z}_{glob}$. The linearization procedure we spoke of will in such a case have to be performed on $\hat{\mathscr{H}}$. In this way it will lead to both a local partition function as well as to a local prefactor ζ. Both will depend on the local frequency rendering obvious that the ζ will have to be different at the well and at the barrier. Our situation differs from the one just described only in the sense that we have to deal with a dynamical situation, albeit in quasi stationary approximation, whose solutions must be found from transport equations.

In ref. [8] a Hamiltonian of type (2) has been used to derive a transport equation for local collective motion. This Hamiltonian can be obtained self-consistently by applying an appropriate linearization procedure with respect to the macroscopic degrees of freedom (see ref. [9]). No restrictions need to be imposed on the "bath" Hamiltonian $\hat{H}_B$ and the F appearing as a factor in $\hat{H}_{SB}$ (see text below eq. (2)). As we shall see, for the present purpose it suffices to expand around values Q_0 which correspond to extremal points of the effective potential (for the general case see eg. ref. [8] or ref. [10]). Then the result can be written as

$$\frac{\partial}{\partial t} f(q, P, t) = \left(-\frac{\partial}{\partial q}\frac{P}{M} + \frac{\partial}{\partial P} C q + \frac{\partial}{\partial P}\frac{P}{M}\gamma + D_{qp}\frac{\partial^2}{\partial q\, \partial P} + D_{pp}\frac{\partial^2}{\partial P\, \partial P} \right) f(q, P, t) \quad (4)$$

with $q=Q-Q_0$. (With respect to P we assume from the start that our Hamiltonian is at most quadratic). Besides the transport coefficients for average (local) motion, there appear the two diffusion coefficients

$$D_{pp} = \frac{\gamma}{M}\Sigma_{pp}, \quad D_{qp} = C\Sigma_{qq} - \frac{1}{M}\Sigma_{pp},$$

with the equilibrium fluctuation Σ_{qq} given by the fluctuation dissipation theorem (FDT)

$$\Sigma_{qq} = \int \frac{d\omega}{2\pi} \coth\left(\frac{\omega}{2T}\right) \chi''_{qq}(\omega) \,. \tag{5}$$

Similar expressions hold for Σ_{pp} and Σ_{qp} if for each appearance of the kinetic momentum P a factor $M\omega$ is multiplied to the integrand. As a consequence, Σ_{qp} vanishes because of a simple symmetry, which is the reason for $D_{qq}=0$.

$\chi''_{qq}(\omega)$ is the dissipative part of the susceptibility $\chi_{qq}(\omega)$ which measures the response of $\langle q\rangle_\omega$ to an external "field" $q_{\rm ext}(\omega)$ in linear order. These functions contain information about all possible modes of our system, as given by the Hamiltonian (2). Clearly, we are interested only in the collective ones, namely those whose average behaviour we have parametrized in terms of the transport coefficients M, γ and C, defined for local motion. To render the dynamics of the fluctuations consistent, we have to calculate the diffusion coefficients from the corresponding approximations. That means to replace in (5) the locally valid $\chi''_{qq}(\omega)$ by the dissipative part of the oscillator response function being defined by

$$\chi^{-1}_{\rm osc}(\omega) q(\omega) \equiv (-\omega^2 M - i\omega\gamma + C) q(\omega) = -q_{\rm ext}(\omega) \,.$$

We are aware that this restriction will finally cause the expected problem of a diverging Σ_{pp}. To regularize the integral appearing there one has to introduce one other constant. For example, this could be done by way of a cut off [8] or through a Drude regularization (cf. refs. [11,12]). For our present purpose this problem is not really relevant.

We are ready now to evaluate the decay rate. Let us look first at the denominator of (3). The linearization we spoke of before means to put $k^{\rm W}_{\rm glob}(Q\sim Q_{\rm a}, P)\sim k^{\rm W}_{\rm osc}(q,P)$ and to use the following stationary solution of (4):

$$k^{\rm W}_{\rm osc}(q,P) = \zeta_{\rm a} \exp(\beta B) \times \exp\left(-\frac{P^2}{2\Sigma^{\rm a}_{pp}} - \frac{(q-q_{\rm a})^2}{2\Sigma^{\rm a}_{qq}}\right), \tag{6}$$

with the prefactor given as

$$2\pi\zeta_{\rm a} = \frac{\mathscr{Z}_{\rm a}}{\sqrt{\Sigma^{\rm a}_{qq}\Sigma^{\rm a}_{pp}}} \,,$$

which normalizes (6) to $Z_{\rm a}$ as discussed above. (Here and in the following the $\mathscr{Z}$'s always refer to the effective macroscopic part of the partition function). In order to calculate the number of particles $N_{\rm a}$ we may replace Δ by ∞, provided we have $|Q_{\rm a}| \gg \sqrt{\Sigma^{\rm a}_{qq}}$. Next we turn to the barrier region. Here the stiffness is negative and the basic expressions for the diffusion coefficients become somewhat delicate, as the "equilibrium fluctuations" loose their immediate physical meaning. Nevertheless, all formulas can be continued analytically by replacing

$$\varpi_{\rm a} = \sqrt{\frac{C_{\rm a}}{M_{\rm a}}} \Rightarrow i\varpi_{\rm b} = \sqrt{\frac{-|C_{\rm b}|}{M_{\rm b}}} \tag{7}$$

everywhere. In this way the Σ_{qq} becomes negative, as already known from Kramers' case of high temperatures. For the prefactor at $Q_{\rm b}$ this implies to write

$$2\pi\zeta_{\rm b} = \frac{\mathscr{Z}_{\rm b}}{\sqrt{\Sigma^{\rm b}_{qq}\Sigma^{\rm b}_{pp}}} = \frac{|\mathscr{Z}_{\rm b}|}{\sqrt{-\Sigma^{\rm b}_{qq}\Sigma^{\rm b}_{pp}}} \,.$$

As for the functional dependence on Q and P, a form like the one obtained from (6) still solves our equation of motion (4), now written for the local motion around $Q_{\rm b}=0$. However, this would not be a decent approximation to $k^{\rm W}_{\rm glob}(Q,P)$. Indeed, since the barrier is unbound to the right we expect a solution with a finite current. Such a solution has been displayed in ref. [13] and applied to computations of interest in nuclear fission, but not to the decay rate. Its form had been found before in ref. [5] within a path integral description. In our present notation it reads

$$k^{\rm W}_{\rm glob}(Q\sim Q_{\rm b}, P) \sim k^{\rm W}_{\rm I}(q,P) = \zeta_{\rm b} \exp\left(-\frac{P^2}{2\Sigma^{\rm b}_{pp}} - \frac{q^2}{2\Sigma^{\rm b}_{qq}}\right) \times \int_{-\infty}^{P-Aq} \frac{du}{\sqrt{2\pi\sigma}} \exp\left(-\frac{u^2}{2\sigma}\right), \tag{8}$$

with

$$\sigma = -\Sigma^{\rm b}_{qq} A^2 - \Sigma^{\rm b}_{pp} \,,$$

$$M_{\rm b}\varpi_{\rm b}(\sqrt{1+\eta_{\rm b}^2} - \eta_{\rm b}) A = \frac{\Sigma^{\rm b}_{pp}}{-\Sigma^{\rm b}_{qq}} \,.$$

The final expression for the decay rate is easily obtained. After putting into (3) both (6) and (8) the integrals can be calculated by elementary methods to give

$$R=\frac{|\mathscr{Z}_b|}{\mathscr{Z}_a}\frac{\varpi_b}{2\pi}\left(\sqrt{1+\eta_b^2}-\eta_b\right)\exp(-\beta B)$$

$$\equiv f_Q R_K\,. \tag{9}$$

As observed already in ref. [5], the quantum correction factor f_Q can be expressed by the absolute value of the ratio of the local partition functions at a and b. In the remaining part of our letter we will discuss the physical relevance of our result together with a critique of the present derivation.

(1) *Comparison with previous derivations.* The partition functions appearing in (9) are readily calculated for the model of the bilinear coupling to a set of oscillators, for example with path integrals (cf. e.g. ref. [4] and our discussion below eq. (2)). Again take first the case of the potential minimum where one gets

$$\mathscr{Z}_a=\mathscr{N}\left(\prod_{n=-\infty}^{+\infty}\frac{\lambda_n^a}{M_a}\right)^{-1/2} \tag{10}$$

with the eigenvalues

$$\lambda_n^a=M_a\nu_n^2+|\nu_n|\gamma_a(|\nu_n|)+M_a\varpi_a^2\,.$$

Here, $\nu_n=2n\pi/\hbar\beta$ and $\mathscr{N}$ is the usual reference factor being independent of ϖ_a, M_a and γ_a. (We follow the notation of ref. [4] allowing frequency dependent friction coefficients $\gamma(\omega)$ for an eventual Drude regularization). $\mathscr{Z}_b$ is obtained from the transformation (7). We observe that the mode $n=0$ has a negative eigenvalue, which for this model proves the conjecture about $\mathscr{Z}_b$ stated above, namely that $\mathscr{Z}_b$ becomes purely imaginary. For temperatures above T_0, with the corresponding β_0 being a solution of $\lambda_1^b=\lambda_{-1}^b=0$, all other eigenvalues are positive. For the quantum correction factor one thus obtains

$$f_Q=\prod_{n=1}^{\infty}\frac{\nu_n^2+\nu_n\Gamma_a(\nu_n)+\varpi_a^2}{\nu_n^2+\nu_n\Gamma_b(\nu_n)-\varpi_b^2} \tag{11}$$

with $\Gamma=\gamma/M$. This expression has been derived before within the formulation of "dissipative tunneling", but for the case $M_a=M_b$ and $\gamma_a=\gamma_b$ [#1]. We present in fig. 2 for a whole range of effective damping factors η_b. At the crossover temperature T_0 this formula diverges, but it can be regularized after considering deviations of third order from the saddle points [4]. As we shall see now our derivation is restricted to temperatures of approximately twice that value, $T>2T_0$.

(2) *The limits of our approach.* In our discussion above we have been very sloppy about any convergence problem appearing in the integrals. Looking back at formula (3) we realize that problems may come from the numerator of this expression. We certainly need a positive Σ_{pp}^b. However, the more crucial quantity turns out to be the Σ_{qq}^b. In Kramers' case it is known to be negative such that the Gaussian factor in (8) increases with increasing q. To study these expressions in general and for sizeable friction is somewhat elaborate, at least for the regime where quantal effects are important. For a first orientation, let us therefore look at weakly damped motion, to postpone a discussion of the general case to future publications (see e.g. ref. [10]). Evaluating the $\Sigma_{\mu\nu}$ from (5) to zero order in γ one gets

$$\langle C_b q^2/2\rangle_b=\langle P^2/2M_b\rangle_b=\tfrac{1}{2}T^*(i\varpi_b)$$

$$=\frac{1}{2}\frac{\hbar\varpi_b}{2}\cot\left(\frac{\hbar\varpi_b}{2T}\right), \tag{12}$$

which is nothing else but the quantal virial theorem continued analytically to the imaginary frequency of the barrier, making apparent the fact that the "equilibrium fluctuation" in q is negative at the barrier.

[#1] The case of variable inertia and friction has been treated in ref. [14] within a phenomenological ansatz; see our discussion to come in point (4).

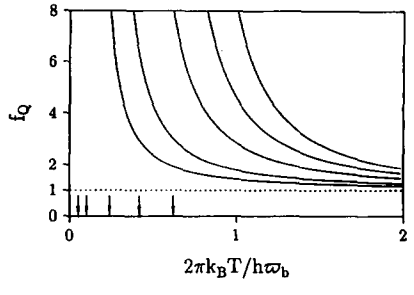

Fig. 2. The quantum correction factor f_Q for the case of both constant inertia $M_a=M_b$ and friction $\gamma_a=\gamma_b$, and the following values of the effective damping rate $\eta_b=10, 5, 2, 1, 0.5$, from left to right, respectively. The arrows show the corresponding crossover temperatures T_0.

(To get Kramers' case of high temperature we just have to replace T^* by T). For a situation like the one behind (12) the condition we have to pose clearly is $T > T_c = 2T_0 = \hbar\varpi_b/\pi$. It guarantees that Σ_{pp} is positive and Σ_{qq} negative. Notice, that for this situation it is easy to observe that also the σ of eq. (8) is positive. At $T = T_c = 2T_0$ the Gaussian factor appearing in (8) becomes indifferent as function of q and regularization procedures would have to be employed [15,6].

(3) *On the quantum nature of this approach.* Our result clearly demonstrates that the transport equation (4) contains quantum effects. It is clear that the latter are there on a semiclassical level only, as global motion is treated in a locally harmonic fashion. Nevertheless it is interesting to see the effects contained in a differential transport equation.

Without any doubt this equation causes problems for the basic uncertainty relations *at short times*. This is no surprise as we have left out memory effects as well as the implications of initial correlations. That both these effects must be there can be studied in the oscillator model again. One may look, for instance, at the results of ref. [16] for the time evolution of the second moments, say for the case of a positive stiffness. Compared with the evolution obtained from (4), one sees that both agree for large times. This is to be expected, of course, from the very construction of the transport equation [8]: The diffusion coefficients are chosen such as to guarantee the correct equilibrium, as defined by the FDT.

The lesson we learn from our present results is that these features prevail also for the case of the inverted oscillator. Again, this does not come unexpected. One knows both from ref. [17] and ref. [5] that for the inverted oscillator time dependent solutions "relax" to stationary ones. Apparently, in the regime of temperatures we look upon here, it is the long time behaviour of the solution which matters – and this behaviour contains the adequate quantum feature. Further details will be discussed elsewhere, see e.g. ref. [10].

(4) *Advantages of our approach.* Although our method does not apply at very low temperatures, we hope to contribute to the understanding of quantum effects in dissipative dynamics. We see at least the following three favorable circumstances.

(i) It should be of some theoretical interest to see effects of "dissipative tunneling" come out of a transport equation proper.

(ii) Our derivation is not based on the assumption of a *linear coupling* to a *heat bath* which does *not change with Q*. It is commonly understood that such a model would be quite unrealistic for nuclear fission, a case discussed already by Kramers. In a Hamiltonian like (2) the x_i, p_i represent the dynamics of the nucleons. For a first approximation there is no way around a mean field approach, which poses important constraints both on the Hamiltonian itself as well as on the treatment of the coupling (see ref. [9] and further literature cited there). First of all, $\hat{H}_{SB}$ *must* have a term involving the collective momentum P. Secondly, the dependence of $\hat{H}_B$ and $\hat{H}_{SB}$ on the x_i cannot just be of first and second order. This feature becomes already evident by looking at the simple but well examined case of multipole vibrations around a potential minimum. It is only for dipole excitations that the $\hat{H}_{SB}$ can be taken linear in the x_i. Thirdly, for large scale motion both $\hat{H}_B$ as well as $\hat{H}_{SB}$ must vary along the collective path. Within the locally harmonic approach one is able to incorporate these effects both for the construction of the Hamiltonian $\hat{\mathscr{H}}(x_i, p_i, Q, P)$ itself [9] as well as for the derivation of the equations of motion [8].

We have mentioned already ref. [14]. There the effects of variable inertia and friction on the decay rate have been taken into account by the following procedure: It was assumed that a Hamiltonian like (2) exists for *global* motion, with a factorized form for the $\hat{H}_{SB}$; the latter was supposed just to depend on the coordinates, thus neglecting self-consistency; some phenomenological ansatz was made both for the form factor of $\hat{H}_{SB}$ as well as for the (unperturbed) inertia in $\hat{H}_S$, neglecting the renormalization of the conservative forces through the coupling; the collective variables were linearly coupled to a heat bath of oscillators with fixed frequencies.

(iii) Our approach may allow for interesting practical applications, even beyond the mere calculation of decay rates. For instance, one is not bound to look at cases where the temperature stays constant. Moreover, as we look at real time propagation our transport equation can easily be used for Monte Carlo simulations. Notice, that our locally harmonic approximation concurs ideally with the small time steps employed in these calculations. Such simulations have

proven successfully in many fields of physics. Let us only allude to the following examples where instabilities play a dominant role: once more nuclear fission, chemical and surface reactions, nucleation and spinodal decompositions. The lesson we learn from the present study is how simple it can be to incorporate quantum effects: Suppose one wants to treat time evolution of the average values on the basis of classical physics. The use of proper diffusion coefficients restores quantum features on the level of the second moments, a procedure which is quite easy to perform within the locally harmonic approximation. In the case discussed here average dynamics is represented by our collective variable $Q(t)$, but an extension to time evolution of the general mean field is straightforward. For the latter the appropriate classical means is the Landau–Vlasov equation. A linearization allows to deduce response functions which in turn determine quantum statistical fluctuations and hence generalized diffusion coefficients (cf. ref. [18]).

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