The nuclear fission process as Brownian motion: modifying the Kramers fission rates

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Abstract. In 1940 Kramers demonstrated theoretically the influence of dissipation on the rate of the thermal escape of a Brownian particle from metastable state. He pointed out the nuclear fission process as a possible application of his results. In his derivation only the canonical ensemble and harmonical shapes of the potential were considered. We generalize the Kramers results for the case of the microcanonical ensemble which is more relevant for the fission process and derive the corrections to the original Kramers formulas accounting for the anharmonic character of the collective potential near its quasistationary and barrier points. The finite "distance" between the barrier and scission points is accounted for as well. We perform quantitative study of the agreement between the generalized and corrected Kramers fission rates and the exact dynamical quasistationary rates in the case of typical fission potentials.

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1. Introduction

Two remarkable papers had been published in the very same year of 1909. First, Hans Geiger and Ernest Marsden had performed the decisive experiments [1] that had lead Ernest Rutherford to the discovery of atomic nucleus [2]. Second, Jean Perrin reported his experiments [3] confirming the theory of Brownian motion developed by Albert Einstein [4]. At that time these two physical systems, atomic nucleus and Brownian Particle (BP), seemed to have nothing in common. Indeed their sizes are 9 orders of magnitude different, the BP moves under the constant influence of the surrounding media whereas the excited atomic nucleus is isolated at least between the rare acts of emission of neutrons etc.

The revolutionary idea to apply the laws of the Brownian motion to the nuclear fission process was put forward by Kramers 30 years later [5]. In 1940 he obtained the formulas for the quasistationary thermal decay rate of a metastable state. The idea is that the BP, due to thermal fluctuations of its linear momentum, escapes from a shallow potential well to a deeper one. The intensity of these fluctuations is proportional to the friction parameter according to the Einstein relation. Evidently Ref. [5] was the first work in which the BP has been regarded not only as the real object, but as the physical model as well. The model is described mathematically by either the stochastic Langevin equations or by the partial differential equations (the Fokker-Plank equation or the Smoluchowski equation (SE)).

One visualizes the classical Brownian motion as the random changes of the Descartes coordinates and conjugate momenta of a comparatively large ball experiencing numerous outside strikes of the much smaller balls. The latter provide the thermostat whose temperature does not change as the BP moves. This corresponds to the Canonical Ensemble (CE).

Similarly, one can imagine the nucleus as the elastic shell inside which many small particles are moving and impinging the shell [6]. In this case the deformation parameters of the nucleus (e.g. its length from pole to pole) and the conjugate momenta play part of the dynamical stochastic variables. The nucleonic degrees of freedom form a heat bath (intrinsic subsystem) which is characterized by a thermodynamical potential and the

temperature. The total excitation energy of the system, E, is shared between the intrinsic and collective subsystems: $E = E_{\text{int}} + E_{coll}$. It is E_{int} that defines the temperature T and thus the intensity of the fluctuations. For the excited nucleus in course of fluctuations of its shape the ratio $E_{coll} / E_{\text{int}}$ can easily reach several percents. In our work we restrict ourselves with the case of strong dissipation. Thus E_{coll} is dominated by the deformation (coordinate) dependent potential energy $U(q)$ and for the intrinsic excitation energy E_{int} equation $E_{\text{int}}(q) = E - U(q)$ seems to be good approximation. In this case the temperature becomes of course the deformation-dependent quantity. This corresponds to the MicroCanonical Ensemble (MCE) approximation.

The fission rate, although not an observable, is the principal characteristic of the fission process (see [7] for the relation between the fission rate and the observables). In [5] Kramers derived several formulas for the decay rate and indicated the nuclear fission process as one of the potential application of his results. One of the Kramers formulas is widely used in modern nuclear physics (see e.g. Eq.(1) of [8], Eq.(20) of [9], Eq.(11) of [10]). Alternative way is to calculate the quasistationary fission rate (QSFR) by means of solving on grids the SE. The latter approach is more accurate but very time consuming. The difference between the Kramers fission rates and the QSFR was shown to reach approximately 20% [11 – 13]. This discrepancy was addressed in [14, 15]. The 20% inaccuracy is comparable to the quantum [16], non-Markovian [17, 18] and multidimensional effects [19] discussed in literature.

The aim of the present study is to find a way for diminishing the discrepancy between the analytical and dynamical rates down to about 2%. This value is comparable with the statistical errors of the QSFR achievable during a reasonable time of computer modeling.

This is an ambitious program, therefore only the one-dimensional overdamped motion corresponding to the symmetric fission at zero angular momentum is considered. The deformation dependence of the friction η and inertia m is ignored whereas for the temperature T (see also [15]) this effect is accounted for.

2. Analytical formulas for the fission rate

The nucleus shape is characterized by the elongation over the diameter of the spherical nucleus, q. Initially all the nuclei are assumed to be concentrated near the quasistationary point $q_c = 1.00$. Three typical nuclei are considered possessing significantly different fission barrier height B_f and barrier point locations q_b: ${}^{216}_{90}Th$ ($B_f = 5.46$ MeV, $q_b = 1.60$), ${}^{204}_{84}Po$ ($B_f = 11.07$ MeV, $q_b = 1.78$) and ${}^{196}_{80}Hg$ ($B_f = 16.23$ MeV, $q_b = 1.90$).The scission point coordinate is $q_a = 2.14$.

The SE with the coordinate-dependent diffusion coefficient reads

$$\frac{\partial g}{\partial t} = -\frac{\partial}{\partial q}\left(D_1 g\right) + \frac{\partial^2}{\partial q^2}\left(\frac{gT}{\eta}\right). \qquad (1)$$

Here $g(q,t)$ is the probability density. The drift coefficient D_1 reads for the cases of the CE and the MCE respectively:

$$D_1 = -\frac{1}{\eta}\cdot\frac{dU}{dq} \quad \text{(CE)}, \qquad D_1 = \frac{1}{\eta}\left[T(q)\frac{dS}{dq} + \frac{dT}{dq}\right] \quad \text{(MCE)}. \tag{2}$$

In the case of the CE the temperature T is calculated using the Fermi-gas relation at the quasistationary point, $T = T_c = \sqrt{(E - U_c)/a}$ and is supposed to be deformation-independent. In the case of the MCE the temperature depends upon the coordinate: $T(q) = \sqrt{\left[E - U(q)\right]/a}$.

Following Kramers we used the flux over population method in order to calculate the fission rate. In the case of the CE it results in

$$R_{I\,T} = \left\{\frac{\eta}{T}\int_{-\infty}^{q_b}\exp\left[-\frac{U(y)}{T}\right]dy\int_{q_c}^{q_a}\exp\left[\frac{U(x)}{T}\right]dx\right\}^{-1}. \tag{3}$$

This relation is implicit in [5] and we call it the Integral Kramers Formula (IKF). The IKF for the case of the microcanonical ensemble includes the entropy $S(q)$ and reads

$$R_{I\,E} = \left\{\eta\int_{q_c}^{q_a}\frac{\exp\left[-S(y)\right]}{T(y)}dy\int_{-\infty}^{q_b}\exp\left[S(x)\right]dx\right\}^{-1}. \tag{4}$$

Geometry used in the derivation of (3) and (4) is illustrated by Fig. 1. These equations are not used in practical modeling of fission at high excitation energy. Instead the following approximate formulas are applied

$$R_{O\,T} = \frac{1}{2\pi\eta}\sqrt{\left|\left(\frac{d^2U}{dq^2}\right)_c\left(\frac{d^2U}{dq^2}\right)_b\right|}\exp\left(-\frac{U_b - U_c}{T_c}\right), \tag{5}$$

$$R_{O\,E} = \frac{T_b}{2\pi\eta}\sqrt{\left|\left(\frac{d^2S}{dq^2}\right)_c\left(\frac{d^2S}{dq^2}\right)_b\right|}\exp\left(S_b - S_c\right). \tag{6}$$

These zero-order formulas are obtained by extending the limits of integration q_c, q_b and q_a to minus/plus infinity, and expanding the potential (entropy) in the integrands up to the quadratic terms in $y - q_c$ and $x - q_b$.

Extending the potential further up to the 4-th order terms and accounting for the finite value of q_a results in the first-order formula ($U^i \equiv \frac{d^iU}{dq^i}$)

$$R_{1\,T} = R_{O\,T}\left[1 - \frac{1}{8}\frac{U_c^{IV}}{U_c^{II}}b_c + \frac{5}{24}\left(\frac{U_c^{III}}{U_c^{II}}\right)^2 b_c\right]^{-1}\left[F + P\right]^{-1}. \tag{7}$$

$$F=\frac{1}{2}\left\{1+erf\left(\Delta q_{ba}/\sqrt{2b_b}\right)\right\}\left[1-\frac{1}{8}\frac{U_b^{IV}}{U_b^{II}}b_b+\frac{5}{24}\left(\frac{U_b^{III}}{U_b^{II}}\right)^2 b_b\right], \tag{8}$$

$$P=\frac{1}{12U_b^{II}}\sqrt{\frac{2}{\pi b_b}}\cdot\exp\left[-\left(\Delta q_{ba}\right)^2/\left(2b_b\right)\right]\cdot\left\{U_b^{III}\left(\left(\Delta q_{ba}\right)^2+2b_b\right)+\right.$$
$$\left.\frac{1}{4}U_b^{IV}\left(\left(\Delta q_{ba}\right)^2+3b_b\right)\Delta q_{ba}+\frac{1}{12b_b}\frac{\left(U_b^{III}\right)^2}{U_b^{II}}\left[\left(\Delta q_{ba}\right)^5+5b_b\Delta q_{ba}\left(\left(\Delta q_{ba}\right)^2+3b_b\right)\right]\right\}. \tag{9}$$

Here $b_c=T\left(U_c^{II}\right)^{-1}$ and $b_b=T\left|U_b^{II}\right|^{-1}$ are the small parameters of our approach. The second-order formula $R_{2\,T}$, also obtained by us; is rather cumbersome and is not presented here. The first- and second-order formulas for the MCE are in the process of derivation.

3. Dynamical modeling and representation of results

All Kramers formulas are approximate. The exact fission rate, been based on the solution of Eq.(010) on grids, reads

$$R_{f\,a}(t)=\left\{-D_1(q_a)g\left(q_a,t\right)+\frac{\partial}{\partial q}\left[D_a\left(q_i\right)g(q_a,t)\right]\right\}\left\{\int_{-\infty}^{a}g(x,t)dx\right\}^{-1}. \tag{10}$$

The initial probability density is chosen to have a Gaussian shape with the variance σ_0^2 centered at q_c. According to the very concept of the quasistationary decay, $R_{f\,a}$ must reach a time-independent value R_D, which should be independent of the initial conditions. Fig. 2 proves that this is really the case in our modeling.

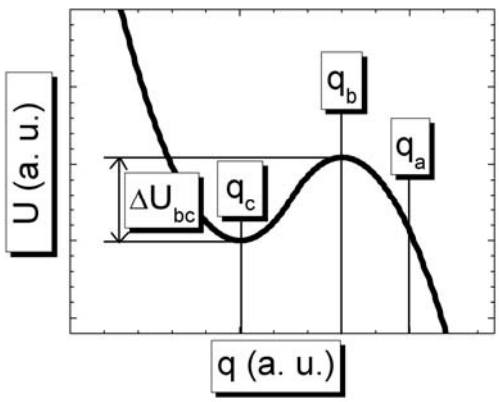

Figure 1. Schematic deformation dependence of the potential (arbitrary units). The coordinates corresponding to the absorptive border (q_a), the saddle point (q_b) and the metastable state (q_c) are shown by thin vertical lines. The height of the potential barrier, ΔU_{bc}, is indicated by two thin horizontal lines.

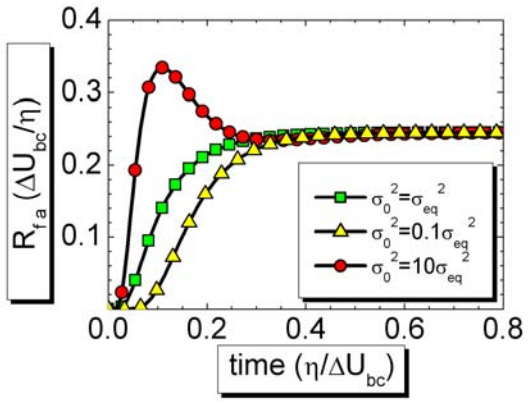

Figure 2. The time dependence of the fission rates calculated according to Eq.(10) at three values of σ_0^2. The rates reach the same quasistationary value R_D irrespectively of the width of initial coordinate distribution. $\sigma_{eq}^2=T_c/U_c^{II}$.

The approximate rates of Eqs.(3 – 7) must be compared with the exact quasistationary rate R_D. As the excitation energy E changes, the rates cover several orders of magnitude, whereas the difference between them is expected to be about 10%. Therefore it is convenient to characterize the deviation of an approximate fission rate R_i from R_D by means of the fractional difference $\xi_i = \left(R_i - R_D\right)/R_D$.

4. Numerical fission rates versus the analytical ones

In Fig. 3 six typical examples of the dependence of the fractional difference upon the controlling parameter are presented. These correspond to the three nuclei with the realistic barrier heights and the saddle and scission point locations. Note that in the case of the canonical ensemble (panels a-c) $\varepsilon = \Delta U_{bc}/T_c$ whereas for the microcanonical ensemble (panels d-f) $\varepsilon = S_c - S_b$. In all cases the absolute values of the fractional deviations become smaller at larger values of ε as it should be expected.

Let us first focus on the CE. For all three cases, $R_{0\,T}$ exceeds R_D by more than 2% in the wide range of ε ($3 < \varepsilon < 10$) where good agreement might be expected. In particular, at $\varepsilon \approx 3$ the fractional difference $\xi_{0\,T}$ increases up to 10% (see panels a) and b)). It is interesting that for lighter nuclei (when $\Delta q_{ba} = q_a - q_b$ decreases) the agreement between the $R_{0\,T}$ and R_D improves. We attribute this effect due to mutual compensation of two errors in $R_{0\,T}$ caused by ignoring Δq_{ba} and the higher derivatives (see details in [20]). In panels a) and b) $\xi_{I\,T}$, $\xi_{1\,T}$ and $\xi_{2\,T}$ are rather close to each other providing acceptable accuracy at $\varepsilon > 3$. This is due to large value of Δq_{ba}. As Δq_{ba} becomes comparatively small (panel c)), $R_{2\,T}$ agrees with R_D worse than $R_{I\,T}$ and $R_{1\,T}$ do.

For a given nucleus behavior of ξ_O and ξ_I for the MCE and CE is very similar. However the fission rates calculated using the constant temperature (which is not the case in the nuclear fission problem) and the constant excitation energy (which is true for the excited nuclei) are very different (see Figs. 5, 7, 8 of Ref.[15] for details).

5. Conclusions

The accuracy of the Kramers formulas for fission rate of heated nuclei has been systematically studied earlier in Ref. [11, 14, 15]. It was found that the integral Kramers formulas (3, 4) agree with the corresponding long time limits of the dynamical modeling significantly better than the zero-order approximation formulas (5, 6). In the present work we have gone beyond the zero-order approximation for the case of the canonical ensemble and have derived the formulas accounting for the first- and second-order corrections.

The accuracy of the analytical formulas was studied for the case of overdamping by comparing with the dynamical decay rate (QDR, R_D). The latter was obtained by solving numerically the Smoluchowski equation (1). The maximum error of the QDR is 2%.

Our results indicate that taking into account the higher derivatives of the potential and the finite value of the distance between the barrier and scission points, Δq_{ba}, in $R_{1\,T}$ results to significantly better agreement with R_D comparing to the conventionally used $R_{O\,T}$: the differences between $R_{1\,T}$ and R_D is within the accuracy of the dynamical modelling (2%) when the fission barrier height becomes approximately thrice as large as the temperature at the quasistationary point. Further expanding of the potential resulting in the $R_{2\,T}$ does not improve the agreement.

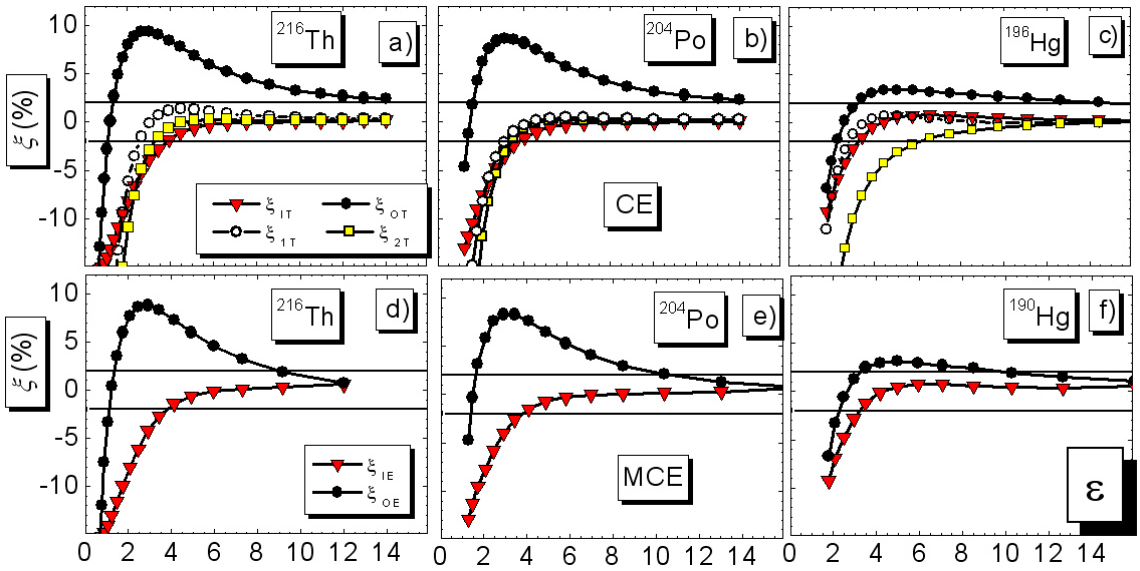

Figure 3. Fractional deviations of the approximate rates from the exact ones versus controlling parameter ε. Upper row of panels: canonical ensemble ($\varepsilon = \Delta U_{bc} / T_c$), lower row – microcanonical ensemble ($\varepsilon = S_c - S_b$). $\eta = 1000$ MeV$\cdot$zs.

The zero-order Kramers formulas $R_{O\,T}$ (5) and $R_{O\,E}$ (6) are not acceptable unless one agrees with the 10% error. In order to reduce the deviation from the dynamical QDR down to 2%, one should apply either the integral formulas (3) and (4) or the first-order formula (7). However, the latter is represented by a simple analytical formula in contrast to the former, which require calculating the double integrals numerically.

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