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S. A. Karamian

FISSION-TO-SPALLATION RATIO AND FISSION DYNAMICS MANIFESTATION

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Карамян С. А. Е15-2008-95
Отношение сечений деление–скалывание
и проявление динамики деления

Для тяжелых ядер с $Z \geqslant 90$ вязкий характер движения системы в координатах деформации влияет на механизм деления высоковозбужденных ядер. В реакциях с тяжелыми ионами доказана немалая длительность временной шкалы деления таких систем. При диабатическом сверхзаторможенном движении эти эффекты могут влиять также на деление ядра-остатка реакции скалывания под действием протонов промежуточной энергии. Анализируются экспериментальные результаты по отношениям сечений деление–скалывание и найдены доказательства, что длительная шкала времени деления проявляется в функции возбуждения деления среднетяжелых мишеней с $Z = 70{-}75$.

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Karamian S. A. E15-2008-95
Fission-to-Spallation Ratio and Fission Dynamics Manifestation

Fission of highly excited nuclei is affected by the viscous character of the system motion in deformation coordinates for very heavy nuclei with $Z \geqslant 90$. The long-time scale fission was proved for such systems formed in heavy-ion induced reactions. The overdamped diabatic motion may influence also fission of the spallation-residue products in reactions with protons at intermediate energy. The experimental results on fission-to-spallation ratio are analyzed and the evidences for the long-time scale fission are found in the fission excitation functions for medium-mass targets with $Z = 70{-}75$.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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INTRODUCTION

Traditionally, the «intermediate» energy of protons is defined as corresponding to the range of $100 < E_p < 1000$ MeV, where the mechanisms of pre- and non-equilibrium emission of nucleons are switched-on and become competitive to the statistical evaporation mechanism. Near 100–200 MeV, it would not be easy to isolate clearly the dominance of one of the mentioned mechanisms. We may assume that at 100 MeV, the equilibrium and non-equilibrium mechanisms make comparable contributions, in abundance. Then, this energy can be taken as a start point for the analysis of the fissile nucleus properties changing at higher energies, $E_p > 100$ MeV. The characteristic neutron-evaporation time for nuclei near ^{181}Ta at the excitation energy of 100 MeV is as short as of about 10^{-20} s, according to the standard statistical model estimates with the Fermi-gas level density. This is still by two orders of magnitude longer than the time of nucleon passage across the nucleus. Therefore, fast intranuclear cascade may happen prior the evaporation being isolated in time scale. For simplicity, we will join pre- and non-equilibrium processes under common term of the fast reaction mechanism.

In the series of experiments carried out [1–3] at the Dubna synchrocyclotron, the yields of products have been systematically measured in reactions of protons with the Hf, Ta, W and Re targets at $E_p = 100-650$ MeV. Main objective was to study the production of high-spin Lu and Hf isomers in the spallation reactions. Along the course of these works, the fission products have also been detected, and fission-to-spallation ratio is finally deduced for many targets at different proton energies. Despite the data has appeared as byproduct, it should be analyzed because the fission mechanisms are still under discussion in modern publications.

Unexpected manifestation of the long time-scale fission for strongly excited nuclei has been established [4–6] in experiments applying the crystal-blocking and atomic-clock methods. Prefission particle emission experiments [7, 8] had stimulated the explanations of long time as a consequence of the reduced excitation energy past emission of many neutrons and other particles. Statistical model calculations were attracted to quantify the «neutron-clock» approach and to deduce the characteristic fission time. However, recent blocking experiments [4, 5] bring evidences for inherent origin of the fission time-scale, while particle emission does accompany the evolution of a system to fission being not a main reason for time delay. This conclusion follows from the experiment [4] on fission of the W-crystal target induced with ^{32}S, ^{48}Ti and ^{58}Ni ions, and from measurements [5] of the fission time for superheavy composite systems formed in reactions of ^{238}U with the Si, Ni and Ge crystal targets. At the latter cases, long times could not arise due to emission of many neutrons because this contradicts to extremely low cross sections for the evaporation residues detected in experiments on a new element production. The overdamped viscous deformation was attracted for explanation of the long-time evolution of the fissile system. It would be interesting to find this effect manifestation not only in heavy-ion induced fission but also at other reactions, in particular at the case of fission of the spallation residue.

1. FISSION OF THE SPALLATION RESIDUE

Nuclear reactions induced by proton at intermediate energy are schematically described as a consequence of fast and slow stages. Each of them may be a multistep process, as well. The fast intranuclear cascade releases nucleons due to the direct proton impact when it crosses the nucleus, and then, the pre-equilibrium emission takes place. Fast stage remains nucleus excited, and the residue undergoes to the slow statistically equilibrated evolution with evaporation of particles, mostly neutrons. Final steps are similar to the well-known scenario of the compound nucleus deexcitation. Fission may happen on the stage of the residual decay, and its entrance parameters make significance for the fission probability estimates. Within traditional schemes, the major problem arises in simulation of Z, A, and excitation energy E^* distributions for the residual nucleus past intranuclear cascade. The sophisticated computer codes are created for that and applied up to the GeV proton energies. However, at E_p lower 200 MeV, the compound nucleus decay codes were used in some cases, for instance, the GNASH code [9] was specially developed to follow up the angular momentum evolution in the statistical decay of excited nucleus. At the same time, some encounter coordinates are achievable basing on the measured mass distributions of the spallation products.

In experiments, Z and A numbers of the detected products are reached after ending of both stages of the reaction. Mass distributions measured in [1–3] show that a mean number of emitted nucleons does not exceed 8 to 15 particles at $E_p = 100-200$ MeV, respectively. During the fast stage, neutrons and protons are emerged with almost equal probability without specific selectivity, unlike to the statistical evaporation of mostly neutrons. Thus, the residual product remains near the β-stability line if the β-stable nucleus is used as a target. Mean mass number of a residue can be estimated assuming that only half of the total number of nucleons is released at the fast reaction. Each nucleon struck out from nucleus at the fast stage indeed remains the nucleus excited to E^* that may be enough for evaporation of one neutron more at the slow stage. Support for the assumption can be found, for instance, in the widely used percolation mechanism of the proton–nucleus interaction. «Percolation» means the creation of a vacancy in the deep nuclear orbital. Then, after relaxation, the excitation energy comparable to the neutron binding energy may arise. Residual nucleus parameters are estimated using the assumptions above, and then the statistical calculations might be productive for the fission probability prediction. Certainly, the described approach looks schematic, but in restricted range of the proton energies from 100 to 200 MeV, the accuracy of estimates must be acceptable.

The experimental results [1–3] on the fission-to-spallation ratio dependent on the proton energy for Hf to Re targets are presented in Fig. 1. Relatively flat function is manifested contradicting to the predictions by the advanced computer code LAHET [10], as is clear from Fig. 1. Thus, some model should be attracted for understanding of the unusual fission probability function. In addition to the LAHET simulations, the simplified scheme is developed here mostly to verify that physical essence of the reaction mechanism is not missed, either screened by the mathematical and program complications. The conclusions made from

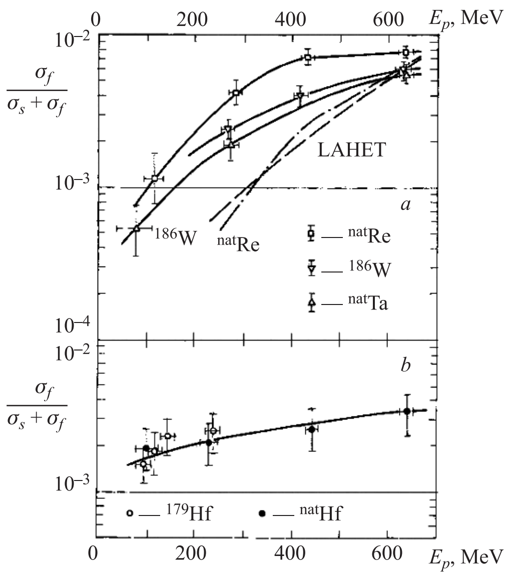

Fig. 1. Fission-to-spallation ratio displayed as a function of the proton energy: *a*) for targets made of Ta to Re according to measurements [1, 2], and *b*) for ^{179}Hf and ^{nat}Hf targets [3]. Solid lines are the guide-lines and dash-dotted lines show the results of calculations using the LAHET code

the experiment-to-theory comparison would be more reliable if they are similar in two different schemes used for the calculations. Quantitative details of the present calculations are given below.

The fission probability is calculated for the ^{174}Yb nucleus which can serve as a typical example of the spallation residue in interaction of protons with Hf and Ta targets at energies $E_p \leqslant 200$ MeV. In statistical model, the probability depends on E^* and is expressed as a ratio of decay widths: $w_f(E^*) = \Gamma_f(E^*)/\Gamma_t(E^*)$, where Γ_t is a total width contained all partial widths of the opened decay channels with emission of neutrons, protons, gammas, etc.: $\Gamma_t = \Gamma_f + \Gamma_n + \Gamma_p + \Gamma_\gamma + \ldots$

At moderate E* values, main contribution corresponds to the neutron emission, and $w_f \approx \Gamma_f/(\Gamma_n + \Gamma_f)$. In numerical calculations, the level density $\rho(E^*)$ function was taken by the Gilbert–Cameron formula [11], and the Moretto equation [12] was used for Γ_n calculations. Fission width was obtained in the Bohr–Wheeler approach multiplied by the Kramers factor:

$$\Gamma_f(E^*) = \frac{(\sqrt{1+\eta^2} - \eta)}{2\pi\rho_c(E^*)} \int\limits_0^{E^*-B_f} \rho_f(E^* - B_f - \varepsilon)d\varepsilon. \quad (1)$$

The Kramers factor takes into account a nuclear matter viscosity parameter η, and its numerical value being constant does not influence the energy dependence of Γ_f, only defines the absolute scale. For better approximation to the absolute Γ_f values, we took a moderate value $\eta = 5$ in our calculations, according to [13]. But the main idea of the present calculations would be to reproduce the energy dependence of a fission probability obtained in the spallation experiments [1–3]. Therefore, a choice of the viscosity parameter should not make strong influence to our analysis, until η is independent on energy. Definitely, there is a freedom for special assumptions on the $\eta(E^*)$ function, but the information available in literature is not enough for the reliable and unique choice.

The level density parameters were taken as in Ref. [13]: $a_n = a_c = A/10$ and $a_f = 1.1\, a_n$. The calculated fission probability $w_f(E^*)$ is a differential function, thus for the comparison to experiments one has to account the multichance fission probability after emission of neutrons. The corresponding sum is expressed as:

$$W_f(E^*) = \sum_{x=0}^{x_{\max}} \left[\frac{\Gamma_f}{\Gamma_t}(x) \prod_{k=0}^{(x-1)} \frac{\Gamma_n}{\Gamma_t}(k) \right], \quad (2)$$

where x is a number of neutrons emitted prior the successful fission, and parameters Γ correspond to the mean excitation energy at the neutron cascade steps. The W_f sum is not yet enough for realistic description of the experimental results. Measured fission-to-spallation ratio characterizes a total fission yield for the whole set of the spallation residues arising with different excitation energy at fixed proton energy. Therefore, measured fission-to-spallation ratio should be compared to the integral:

$$P_f(E_p) = \int\limits_{B_f}^{(E_p+B_p)} W_f(E^*)S(E^*, E_p)dE^*, \quad (3)$$

where $S(E^*, E_p)$ is the normalized to unity spectral distribution of the residual excitation energy at definite E_p; and B_p is a binding energy of the proton with the target nucleus.

In Fig. 2 the spectral functions $S(E^*, E_p)$ at $E_p = 100$, 150 and 200 MeV are shown according to estimates based on the discussed above reaction properties. Possible E^* values may appear within whole range from 0 to maximum energy $(E_p + B_p)$. Population of the low-energy domain has been proved by the reactions of exclusive emission of only a few nucleons, from 1 to 3, as is detected in [3]. Maximum E^* values are also visible in a form of the spallation products corresponding to the emission of many nucleons: as many as is allowed by the energy available. In general, the spallation peaks in mass distributions [1–3] satisfy the requirements both of energy balance and of the integral cross section close to the geometrical one, $\sigma_t \approx 1.5$ b. At 100 MeV, the equilibrium and pre-equilibrium processes are equally abundant and S distribution shows-up a peak

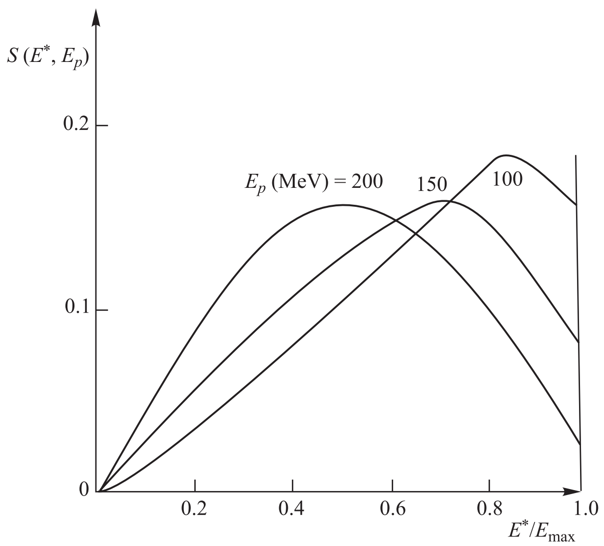

Fig. 2. Spectral function $S(E^*, E_p)$ or excitation energy distribution for the residual nucleus in the spallation reactions

near the maximum energy. But at 200 MeV, fast non-equilibrium interaction is responsible for the direct knock-out of several nucleons, and the residual energy is decreased shifting the peak of S function to lower energies.

The differential fission probability $w_f(E^*)$ was calculated using the statistical model procedure commented above for the nucleus with fission barrier of $B_f = 25$ MeV that is trustable for the ^{174}Yb residual nucleus, according to [14]. The $w_f(E^*)$ and $W_f(E^*)$ functions are given in Fig. 3. Finally, one can calculate using Eqs. (2), (3) the integral fission-to-spallation ratio $P_f(E_p)$ and the resulted function is displayed in Fig. 3 as well. Calculated $P_f(E_p)$ dependence clearly deviates from the experimental results shown in Fig. 1. We did not try to reproduce the absolute values, but the slope of $P_f(E_p)$ function looks more or less similar to the LAHET code predictions. Thus, two different schemes of calculations predict drastic energy dependence of fission probability, but the experiment demonstrates a flat behavior, in contradiction. Some modification of the basic assumptions is necessary for theoretical description of the results shown in Fig. 1. The w_f values cannot be significantly changed by the angular momentum account because of moderate spin value of the residual nucleus, as follows from

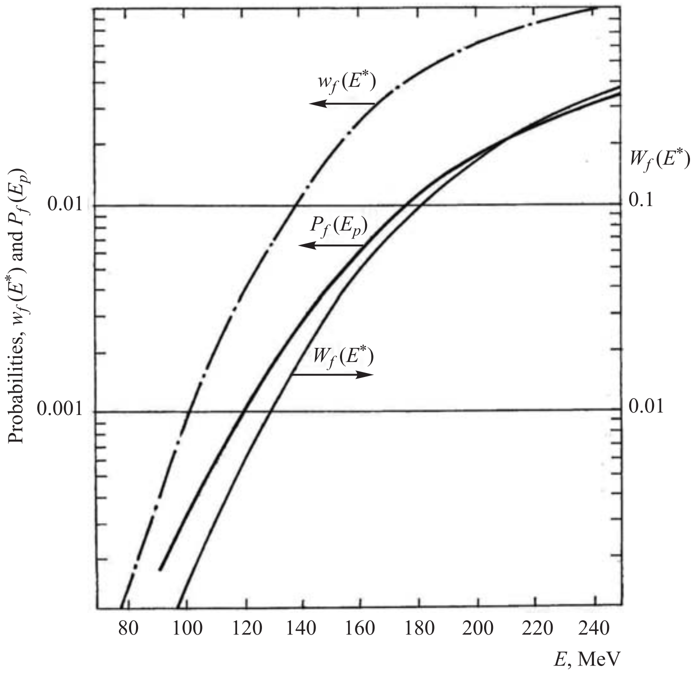

Fig. 3. Calculated differential $w_f(E^*)$ and total $W_f(E^*)$ probabilities of fission together with the overall probability $P_f(E_p)$ representing the fission-to-spallation ratio

the analysis of [15]. The angular momentum lower 10 units practically conserves fission barrier without changes [16].

One may try another approach with deep modification of the Γ_f calculations, assuming the viscosity parameter η strongly-dependent on excitation energy. But in the popular one-body dissipation mechanism [17], the viscosity should be almost constant. In addition, the idea to replace a fission probability values with the $\eta(E^*)$ function varied for the best fit does not look correct in general philosophy. This is just a trivial translation from one parameter to another, and it does not bring a new knowledge. Possible interpretation that $P_f(E_p)$ dependence reflects weak variation of the mean residual excitation energy E^* with E_p growth, looks also inadequate. In addition to the model arguments, such an assumption would be in conflict with the observed more flat $P_f(E_p)$ behavior for Hf than for heavier W and Re targets, see Fig. 1. For lower Z nuclei, the fission barrier growth reduces P_f in magnitude and makes it more drastic function of the excitation energy. The experiment shows however another trend. A necessity of some new interpretation becomes evident for explanation of the spallation-residue fission probability — $P_f(E_p)$ function.

2. DEFORMATION TIME SUPPRESSES FISSION YIELD

Unexpected manifestations have been observed earlier for fission of excited nuclei in heavy-ion induced reactions. In experiments [4–6], there is found that fission time-scale typically covers a range near 1 as $= 10^{-18}$ s, and this time is weakly-dependent on Z and excitation energy of the fissile nuclear system. For superheavy nuclei, similar properties [5] should be combined with extremely low cross sections for the evaporation residue formation as is observed in the experiments on the new elements synthesis [18]. Such a combination cannot be explained within standard statistical model calculations. More specifically, low fission barrier for heavy nuclei with $Z^2/A \geqslant 38$ provides in statistical approach a dominance of fission expressed in almost 100% probability for fission. Thus, low cross section for the evaporation product may be reproduced, but not a long time-scale of fission. The only idea for understanding could be found in an assumption that the fission-evaporation competition is causally decoupled from the fission evolution time.

In Fig. 4, the potential energy of a fissile system is schematically illustrated for two values of the parameter $\mathrm{Z}^2/A \approx 40$ and ≈ 32. In our discussion, the quantitative details and inaccuracies practically make no significance, because only a right scheme we are looking for the interpretation of experiments. Therefore, the fission barrier profile in Fig. 4 is taken according to the classical liquid-drop model potential [19]. For the case of a heavy nucleus, the saddle point is located at relatively low deformation, and a long path corresponds to the motion from

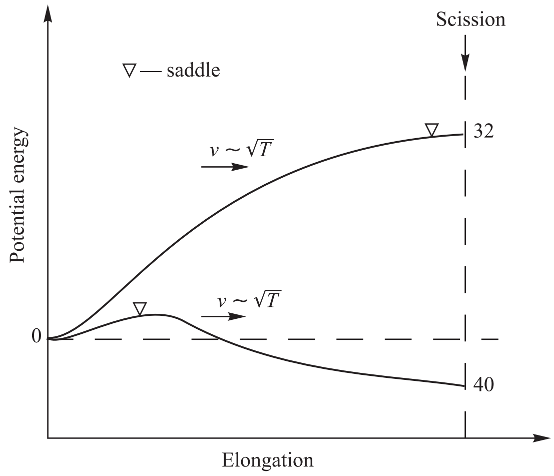

Fig. 4. Nuclear potential energy dependent on generalized deformation parameter as is schematically reproduced by the liquid-drop model calculations [19] for very heavy system with $Z^2/A = 40$ and for relatively light, $Z^2/A = 32$, fissile nucleus

saddle to scission. Obviously, the latter stage of a system evolution is responsible for the observed long time-scale of fission. A survival of the compound nucleus is decoupled from this stage and is defined by the earlier steps. The dynamical deformation of a composite nuclear system has been theoretically studied in many works, see, for instance, Refs. [20, 21] and references therein. There was shown that evolution to scission may correspond to long time at the case of overdamped diabatic motion. The puzzling situation for heavy system can be resolved within the discussed scheme.

For relatively light fissile nuclei with $Z^2/A \sim 32$, fission barrier is as high as $B_f \approx 25$ MeV, and the corresponding saddle point is located near the scission deformation. Thus, long path corresponds to the motion from initial quasi-spherical configuration to the saddle (scission) elongation. The time parameter of a latter dynamical motion will define not only the fission time-scale but also will make influence to the integral fission probability. Long deformation time may allow the evaporation of many neutrons with the reduction of excitation energy down to $E^* \leqslant B_f$. A conclusion follows that fission probability studies for relatively light systems may throw some light onto the dynamical deformation time in addition to the direct time-scale measurements. Our fission-to-spallation probability functions shown in Fig. 1 and commented above meet the requirement of such an advanced analysis.

A question arises: is this really a new approach as compared to the known «neutron-clock» method [7]? At the latter case, a whole model operates within the statistical decay scheme. The fission-evaporation competition and partial neutron and fission rates are defined by the statistical width values. Time-scale and probability of fission originate from the same stuff. In the present consideration, we suppose that fission time is defined inherently by the overdamped dynamics, and neutron evaporation just accompanies a system evolution. The causal relation between two processes is now reversed. Due to that, whole logics of a model should be revisited: not only for the fission time results, but also for the fission probability functions.

Within the proposed new approach, a fission evolution is defined by the deformation time τ_d from the compact shape to the strongly deformed saddle point. The τ_d parameter is connected mostly with the path length unlike to the statistical fission time, $\tau_f = \hbar/\Gamma_f$, that is governed mostly by the potential barrier B_f, see Eq. (3). The deformation time of the dissipative inertial process should depend on the path length. Even at high temperature, a system needs time for the successful elongation to scission. Thus, relatively weak variation of the fission time-scale with E^* revealed in Refs. [4–6] becomes understood in this approximation.

Despite another physical content, one may assume that in the new model, a fission probability is still defined by the rate linked to the characteristic time parameter, thus, a probability is reversely proportional to the deformation time: $W_f \sim \tau_d^{-1}$. The assumption looks natural that a process rate is bound to the characteristic time parameter τ_d, even despite another origin of this time.

The collective velocity should be proportional to the square-root of a temperature $\upsilon_d \sim T^{1/2}$, therefore:

$$W_f \sim \tau_d^{-1} \sim \left[(\upsilon_d)^{-1}\right]^{-1} \sim T^{1/2}. \tag{4}$$

In these schematic considerations, the standard thermodynamic relations are involved in the estimates of the partial energy pumped into the collective degree of freedom of the equilibrated system at definite temperature. For quantitative description of the fission features, the calculations by Eq. (4) are not enough, and more developed theory should be created, in particular for the τ_d parameter evaluation. But the presently introduced approach may play a key role for understanding of the fission probability and time-scale in the unified scheme without contradiction to the experimental results. The observed flat energy dependence of the fission-to-spallation ratio shown in Fig. 1 may confirm and justify the physical content of Eq. (4), in addition to the above explanations.

For better understanding, let us continue with additional comments on the comparison to standard statistical model scheme. The neutron width Γ_n at the case of light fissile nuclei is typically much higher than the fission width Γ_f calculated by Eq. (3). The Γ_f value influences mainly the fission probability $P_f = \Gamma_f/(\Gamma_n + \Gamma_f)$, while real time scale is defined by the compound nucleus lifetime $\tau_c = \hbar/(\Gamma_n + \Gamma_f)$, i.e., practically, by the neutron time $\hbar/\Gamma_n$. The logic is absolutely identical to the well-known case of radioactive decay, when all branches together are characterized by the common lifetime, and partial half-lives define only the branching ratios.

In the discussed here new scheme, fission is a slow process that is not completed within short time of the neutron-evaporation step. Thus, the system evolution to scission is accompanied by the neutrons emission, but they cannot influence the inherent fission time, neither stop the successful fission until the excitation energy is enough for the fission-barrier transmission. The total time $\Sigma\tau_n$ of the neutron cascade reducing E^* down to B_f value should be compared to the dynamical deformation time τ_d, and this ratio will define the probability of successful fission:

$$W_f = \frac{\sum \tau_n}{\tau_d}. \tag{5}$$

For some definite nucleus, the nominator would not be strongly-dependent on the initial excitation energy, because $\Sigma\tau_n$ is in major defined by the last step of the cascade. Therefore $W_f \sim \tau_d^{-1}$, as we have assumed above.

More complicated scenarios may also take place, and the simplified version is discussed here just for illustration of the scheme capability for the self-consistent simulations of the fission probability and time-scale. In particular, the τ_d magnitude should obviously decrease with the growth of Z^2/A parameter because of shortening of the path-length for the system evolution from a compact shape to the saddle point. A model relevant for quantitative calculations should yet be created elsewhere.

The statistical model calculations are attracted for the simulation of fission properties in many publications. But recent experiments bring the evidence that this model is incapable to reproduce the fission time-scale for very heavy systems, neither fission probability for relatively light nuclei. Probably, the statistical model can be applied within some restricted range of the fissility parameter, near the moderate value of $Z^2/A \approx 35$, and many works are successful just because they operate within such a range. It is probable that for medium Z nuclei, the time parameters of both neutron cascade $\Sigma\tau_n$ and fission-deformation path τ_d are comparable in magnitude. Therefore, a scheme of coupling between both processes could be applied. However, at much higher and much lower Z^2/A values, the traditional scheme obviously fails.

SUMMARY

The present paper is written mostly to stress a manifestation of the dynamical deformation time in the fission probability P_f of relatively light fissile nuclei formed as a spallation residue of the proton-induced reactions. Fission probability was determined in experiments [1–3] after measurements of the fission-to-spallation ratio dependent on the proton energy in irradiations of the Hf to Re targets. Relatively flat energy dependence $P_f(E_p)$ contradicts the statistical model predictions and the results of the advanced computer simulation codes. Therefore, it is now analyzed within the alternative model. A traveling time of the fissile system in deformation coordinates is affected by the inertial dissipative dynamics of the motion, and that finally influences the experimentally measured fission probability and time-scale parameters. This new approach allows one to understand without internal contradictions the behavior of the above-mentioned parameters.

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141980, г. Дубна, Московская обл., ул. Жолио-Кюри, 6.
E-mail: publish@jinr.ru
www.jinr.ru/publish/