First Experiment on Fission Transients in Highly Fissile Spherical Nuclei Produced by Fragmentation of Radioactive Beams

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We report on a novel experimental approach for studying the dissipative spreading of collective motion in a metastable nuclear system, using, for the first time, *highly fissile nuclei with spherical shape*. This was achieved by fragmentation of 45 radioactive heavy-ion beams at GSI, Darmstadt. The use of inverse kinematics and a dedicated experimental setup allowed for the identification in atomic number of both fission fragments. From the width of their nuclear-charge distributions, a transient time of $(3.3 \pm 0.7) \times 10^{-21}$ s is deduced for initially spherical nuclei.

The problem of escape from a metastable state appears in many fields as various as fluid mechanics, chemistry, or nuclear physics [1]. An excellent test case for such a process is nuclear fission. Once an excited system has been produced, the collective coordinates start adjusting to the potential-energy landscape, and the distribution of shapes inside the fission saddle develops towards thermodynamical quasiequilibrium. After some time delay, all states in the quasibound region, including those above the fission saddle, are populated according to the available phase space. Once the system leaves this region, it is driven by the dominating Coulomb force to more elongated shapes and finally separates into two fragments.

The ideal scenario for studying the dynamics of fission is described in the pioneering theoretical work of Grangé, Weidenmüller, and collaborators [2,3]: They have chosen an initial excited system characterized by a spherical shape. In this case, the probability distribution starts from a configuration where the level density has a local maximum. Under this specific initial condition, the maximum of the probability distribution does not move with time, and the distribution only spreads out under the influence of dissipation due to the *fluctuating forces*. In contrast, the evolution from saddle to scission is characterized by a strong driving force, and it is the *friction force* which dominates the dissipation mechanism by slowing down the directed motion towards scission. Thus, in this ideal scenario starting from the bottom of a spherical potential well, the two stages of the fission process probe selectively the two dissipative phenomena, diffusion and friction, in nuclei [4]. The present work is dedicated to study the time scale of nuclear diffusion.

Attempts to selectively investigate the purely dissipative relaxation process inside the quasibound region were hampered by the unavailability of spherical highly fissile nuclei. In this Letter, we report on a novel experimental approach, which comes very close to the aforementioned ideal scenario, by investing a considerable instrumental effort utilizing the installations of GSI, Darmstadt. This approach is based on the two-step reaction mechanism illustrated in the upper part of Fig. 1. Fragmentation of a primary ^{238}U beam at 1 A GeV in a beryllium target produces a large variety of nuclei. Most of these products adapt to their ground-state configuration well before reaching the second target. In the present experiment, 45 spherical radioactive isotopes of elements between astatine and thorium, with neutron numbers around the $N = 126$ shell, were chosen as secondary projectiles. They further impinge on a lead target, where fissile nuclei are produced in peripheral nuclear collisions at relativistic energy. These, so-called prefragments, reach up to high excitation energies [5–7] and are characterized by small angular momenta [8]. According to the fast cutoff picture [5,9], the initial quadrupole deformation parameter β_2 induced by the abrasion process in the prefragment remains small and oblate with $|\beta_2| < 0.15$. This value should be compared to the corresponding saddle-point deformations of around $\beta_2 \approx 0.6$–0.8. The prefragment finally decays by a competition between fission and particle evaporation.

A schematic drawing of the experimental setup is given in the lower part of Fig. 1. In the first stage, the secondary beams produced in the primary target were identified and selected by the fragment separator (FRS). At the exit of the FRS, they have an energy of about 530 A MeV. In the second stage, the fission fragments produced in the interactions of the secondary projectiles with the lead target are detected. The inverse kinematics of the reaction allows an accurate determination of the atomic numbers $Z_{1,2}$ of both fission fragments *simultaneously* in a double ionization chamber. The excellent resolution of 0.4 charge units (FWHM) is clearly visible in the (Z_1, Z_2) correlation spectrum presented in Fig. 2; each spot corresponds to one pair of fragments. Additional detectors served as fission trigger and for background suppression. We refer to [10] for a detailed description of the setup.
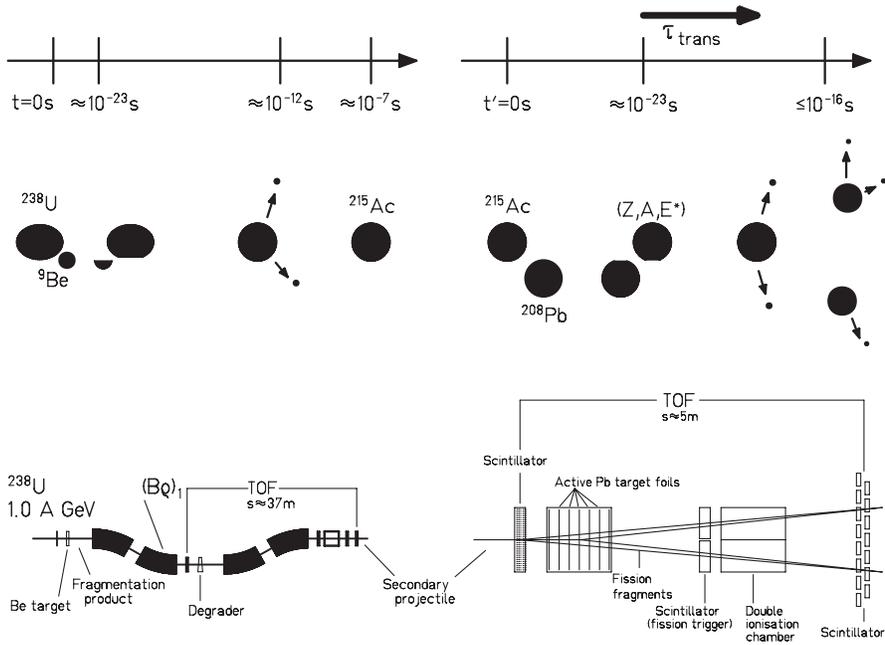
FIG. 1. Top: Illustrative drawing of the radioactive-beam production (left) and of the subsequent fragmentation-fission reaction (right). Typical time scales (t and t' refer to the primary and secondary reaction, respectively) are indicated. Bottom: Experimental setup involved along the outlined reaction scenario, consisting of the FRS (left) and the fission setup (right).

Fragmentation of the secondary projectiles leads to a large variety of prefragments. We make use of the measurement of the sum $Z_1 + Z_2$ of the nuclear charges of the two fission fragments to classify them. Full-acceptance experiments [11] have shown that in peripheral collisions at relativistic energies the impact parameter and the nuclear charge of the heaviest fragment measured in a given event are strongly correlated. This also implies a close relation between the nuclear charge of the fissioning nucleus Z_{fiss} and the impact parameter. Because the probability for the evaporation of light charged-particles is small for the systems considered in this work, the $Z_1 + Z_2$ sum is equal to the charge Z_{fiss} of the fissioning nucleus in most

cases. Hence, $Z_1 + Z_2$ scales with the fissility of the system. In addition, it can be used as a measure of the excitation energy induced in the reaction—a lower $Z_1 + Z_2$ value corresponds to a smaller impact parameter and, thus, to a higher initial excitation energy. As an example, Fig. 3 presents the calculated initial excitation energy as a function of the $Z_1 + Z_2$ sum for fragmentation-fission of a ^{215}Ra beam. For the most central collisions, multifragmentation, which proceeds on a much shorter time scale than fission, sets in. Therefore, when $Z_1 + Z_2$ falls below 70, the initial excitation energy of the prefragment relevant for the fission process decreases, corresponding to a freeze-out temperature of 5.5 MeV [7].
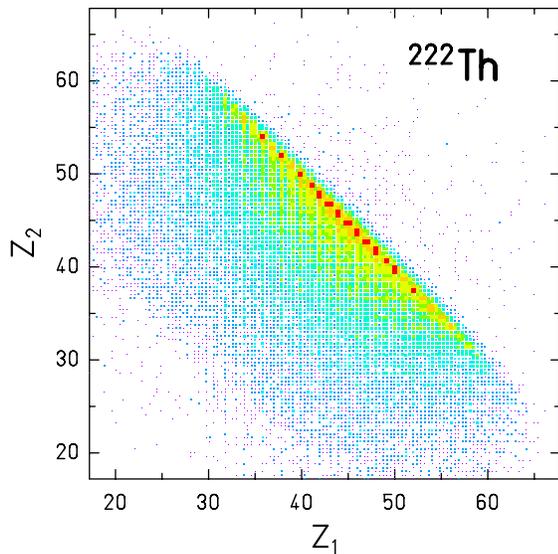

FIG. 2 (color online). Experimental correlation between the nuclear charges (Z_1, Z_2) of the fission fragments for a ^{222}Th beam. The color scale is logarithmic.
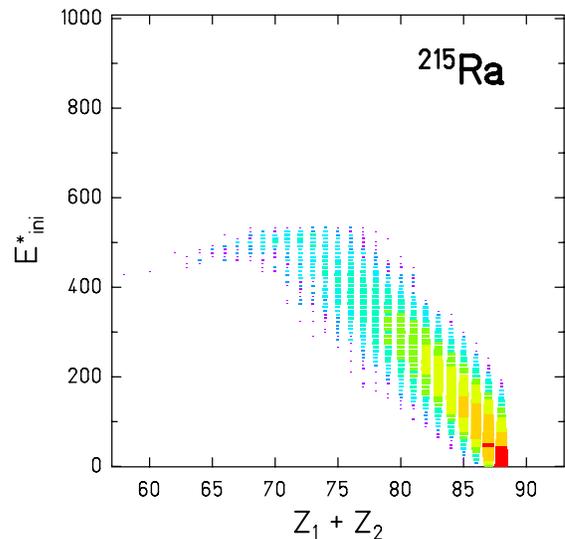

FIG. 3 (color online). Calculated correlation between the initial excitation energy of the system and the $Z_1 + Z_2$ sum for the ^{215}Ra beam. The color scale is logarithmic.

Following Ref. [12], we use the width of the fission-fragment nuclear-charge distribution σ_Z for establishing a chronometer up to the fission saddle. From a comprehensive compilation of data, the authors of [13] have shown that the width σ_A of the fission-fragment mass distribution, and accordingly σ_Z, scales with the temperature T_{sad} at saddle as $\sigma_Z^2 = T_{\text{sad}}/C_Z$, where C_Z is related to the stiffness of the potential-energy landscape at the top of the barrier. The temperature at saddle is governed by the number of particles deexciting the system before it reaches the saddle point, and, consequently, by the time spent in the potential well. Extrapolating the systematic of [13] to higher energies, we demonstrated [12] that σ_Z is closely linked to the transient delay τ_{trans} [2,3] which is the time required by the fission-decay width for reaching 90% of its quasistationary value. Hence, σ_Z can be used as a clock for the time the system needs to cross the fission barrier. The experimental σ_Z value is shown in Fig. 4 as function of $Z_1 + Z_2$ for some of the systems investigated [14].

For a quantitative analysis of the nuclear-charge width, we compare the data with results from the Monte Carlo code ABRABLA [5–7,15,16]. This code, based on the abrasion-ablation model, simulates the complex reaction scenario realized in the experiment. Its validity has been assessed by investigating various systems and observables [17]. In ABRABLA, fission is treated as a dynamical process using a *time-dependent* fission-decay width $\Gamma_f(t)$ that is based on a realistic analytical approximation [18] to the numerical solution of the one-dimensional Fokker-Planck equation for a system initially localized in the minimum of a spherical potential well. Moreover, the ratio a_f/a_n of the level-density parameter at the saddle point to that at the ground state is realistically calculated taking the influence of deformation into account [19,20]. For all 45 spherical projectiles, the experimental dependence of σ_Z on $Z_1 + Z_2$ has been compared with calculations performed varying the dissipation strength β from 1×10^{21} s^{-1} up to 7×10^{21} s^{-1} [21]. As observed in Fig. 4, the whole data set is well described with $\beta = (4.5 \pm 0.5) \times 10^{21}$ s^{-1} independent of $Z_1 + Z_2$, i.e., independent of excitation energy and fissility. Also shown in the figure are predictions based on the Bohr-Wheeler transition-state model [22] and Kramers' diffusion scenario [23]. While the former com-

pletely neglects dissipation, the latter accounts for its influence on the quasistationary value of $\Gamma_f(t)$ but omits the presaddle delay. The corresponding fission-decay widths are thus *time independent* [18]. Both models predict too steep a rise of σ_Z with decreasing $Z_1 + Z_2$. As soon as $Z_1 + Z_2$ differs by more than 3–4 charge units from the nuclear charge of the secondary projectile, calculations neglecting transients substantially deviate from the data. This shows that σ_Z becomes sensitive to the initial suppression of fission during the transient time for initial temperatures above about 3 MeV. Even more, as σ_Z is a quantitative measure of the cooling of the system by evaporation before fission, it characterizes the transient delay in a rather model-independent way [12]. The effect is sizeable: While the initial temperature reaches up to 5.5 MeV, T_{sad} does not exceed 3 MeV. The analysis of the whole set of data collected in the present work suggests a nearly constant transient time of $\langle \tau_{\text{trans}} \rangle = (3.3 \pm 0.7) \times 10^{-21}$ s.

In our previous work [12], we investigated dissipative phenomena in fission of ^{238}U projectile fragments and extracted a transient time of $(1.7 \pm 0.4) \times 10^{-21}$ s, which is appreciably shorter than the present result. We attribute this difference to the fact that the fissioning nuclei studied in [12] are deformed, since stemming from fragmentation of the primary ^{238}U projectile, which has a prolate shape characterized by $\beta_2 \approx 0.23$ [24]. To our knowledge, this constitutes the first *experimental* indication on the influence of the initial conditions on the time the system needs to reach the barrier. Our observation is consistent with one-dimensional dynamical calculations [25], which predict that the initial deformation of the system as given by the entrance channel has a strong influence on τ_{trans}. This was recently confirmed by calculations in a three-dimensional space [9]. The influence of initial deformation is customarily neglected in data analysis. The experimental confirmation of its sizeable importance suggests revisiting previous works [25,26], and, namely, those dealing with heavy-ion fusion reactions, where the capture configuration differs significantly from a compact compound nucleus. The value extracted for β might be biased when disregarding this effect.

Spherical initial shapes have also been achieved in spallation experiments with lighter systems, see, e.g., [27–29]
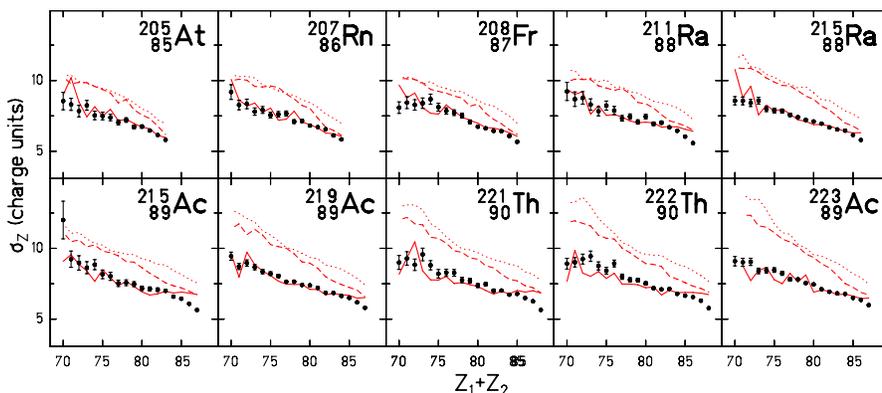

FIG. 4 (color online). Width σ_Z as a function of $Z_1 + Z_2$ for a sample of spherical secondary beams as indicated. The data (dots) are compared with calculations using Bohr-Wheeler [22] (dotted lines) and Kramers [23] (dashed lines) fission-decay width as well as the time-dependent approximation (full lines) from [18]. In the two latter cases, β is set to 4.5×10^{21} s^{-1}. Staggering in the calculations is due to statistical fluctuations.

and references therein. The present work goes a step further by utilizing for the first time *highly fissile* spherical nuclei for which the fission properties can be studied over a much wider range of excitation energy.

Our result for β suggests an overdamped motion at small deformation and high excitation energy, amounting to about 25% of what is predicted by the one-body wall theory [30]. In an extended study of a large body of observables, which in most part were sensitive to the whole path up to scission, dynamical calculations [31] led to β values comparable with the present one. This finding agrees with theoretical studies on the surface motion of a cavity wall (see [32] and references therein), which suggest that the original one-body wall damping picture [30] over-estimates the dissipation rate in nuclear matter.

The numerical value $\beta = (4.5 \pm 0.5) \times 10^{21}$ s^{-1} extracted in this work might be considered as preliminary since it is based on the Fokker-Planck equation involving a single collective coordinate. Realistic simulations [33] require at least a three-dimensional deformation space. However, the transient time, which is rather directly reflected in our experimental signature, should not be affected.

The present experiment has provided dedicated data on diffusion in the quasibound region. The consistent description of these data together with previous conclusions on scission times [31] indicates that diffusion and friction inside and outside the quasibound region are consistently described by conventional Fokker-Planck or Langevin dynamics with a rather universal dissipation strength. Hence, our data do not evidence any departure from normal diffusion dynamics in nuclear matter. According to Einstein's relation, the nearly constant dissipation strength suggests that diffusion scales with temperature.

In summary, we report on an experiment realized under very favorable conditions for studying transient effects in nuclear fission. For the first time, we succeeded in probing *diffusion in spherical highly fissile nuclei*. We benefit from a considerable instrumental effort using projectile-fragmentation in a two-step experiment at GSI, Darmstadt. The analysis of the fission-fragment nuclear-charge distributions reveals a clear manifestation of pre-saddle transient effects at high excitation energies, which delays the quasistationary flow across the fission barrier. After the essential achievement of the present work in preparing a fissile system with spherical shape, we expect further progress in characterizing the system at the future FAIR facility [34], where the coincident light particles will be recorded and the fragments will be identified in charge *and* mass.

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