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Phys. Rev. Lett. **106**, 132503 — Published 30 March 2011

DOI: 10.1103/PhysRevLett.106.132503

Brownian shape motion on five-dimensional potential-energy surfaces: Nuclear fission-fragment mass distributions

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Although nuclear fission can be understood qualitatively as an evolution of the nuclear shape, a quantitative description has proven to be very elusive. In particular, until now, there exists no model with demonstrated predictive power for the fission fragment mass yields. Exploiting the expected strongly damped character of nuclear dynamics, we treat the nuclear shape evolution in analogy with Brownian motion and perform random walks on five-dimensional fission potential-energy surfaces which were calculated previously and are the most comprehensive available. Test applications give good reproduction of highly variable experimental mass yields. This novel general approach requires only a single new global parameter, namely the critical neck size at which the mass split is frozen in, and the results are remarkably insensitive to its specific value.

PACS numbers: 25.85.-w, 24.10.-i, 24.60.Ky, 24.10.Lx

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Since nuclear fission was discovered in 1938 [1], its theoretical modeling has presented significant challenges. As discussed already in the pioneering papers by Meitner and Frisch [2] and Bohr and Wheeler [3, 4] in 1939, nuclear fission can be viewed qualitatively as an evolution of the nuclear shape from that of a single compound nucleus to two receding fragments. But the character of the shape dynamics is still not well established. Nor is it yet understood in detail how the original compound nucleus is transformed into a variety of different fragmentations. Therefore models for the resulting distribution of mass splits have, until now, had limited predictive power.

The currently used methods for calculating fragment yields include a variety of phenomenological approaches. These invariably introduce a number of parameters whose values are determined from adjustments to measured mass yields and other observables [5–7]. Such approaches typically reproduce experimental data in the regions where their parameters were determined, but they do not advance our understanding of fission and they also tend to fail when applied to other regions.

Scission models consider the relative statistical weights of various fragmentations at the time of scission [8, 9]. Such calculations are entirely static in nature so they cannot take account of any dynamical effects. While scission models often yield reasonable agreement with the observed mass distributions, they are not universally successful and their failures suggests that the resulting divisions are sensitive to the pre-scission dynamics.

A number of dynamical models of fission have also been developed. Most of these concentrate on the average evolution and they are often macroscopic. Langevin treatments have been developed and applied for excitations sufficiently high to render the dynamics macroscopic [10–12]. The presently most refined microscopic approach uses the time-dependent generator-coordinate method with Hartree-Fock-Bogoliubov states based on an effective interaction [13, 14]. This treatment is rather computer intensive and, consequently, only a few systems have been studied so far. In particular, fragment mass distributions have been calculated only for fission of ^{238}U [13] (though results for ^{240}Pu are underway [15]).

We introduce here a novel method for calculating fission fragment mass distributions. It invokes the expected dissipative character of the coupling between the nuclear surface and the internal degrees of freedom [16] which, in the Smoluchowski limit of strong coupling [17], gives the shape dynamics the character of Brownian motion. Consequently, once the potential energy is known as a function of deformation for a sufficiently rich family of fission shapes, the required calculation is relatively straightforward, amounting effectively to a random walk on the corresponding potential-energy surface. Because suitable deformation-energy surfaces are available for essentially all nuclei of potential interest [18], this treatment of the fission dynamics provides a powerful predictive tool. We describe here the key features of the approach and present several test applications.

As mentioned above, our method exploits the limit of strong dissipative coupling between the nuclear surface motion and the internal degrees of freedom, as is characteristic of systems dominated by one-body dissipation [16]. The basic mechanism is the reflection of individual nucleons off the moving surface which generates a dissipative force that is rather strong due to the nucleonic Fermi motion. The average nuclear shape evolution is then determined by the balance of the associated dissipative force on a surface element and the conservative force due to the deformation energy. The resulting equations of motion for the shape evolution contain no adjustable parameters and dynamical fission calculations yield remarkably good agreement with data for the most probable fragment-kinetic energies [16].

Because the individual nucleons reach the moving surface at random times, the associated force is stochastic, in accordance with the fluctuation-dissipation theorem [19].

The present treatment is the first implementation of the stochastic part of the one-body mechanism for mononuclear dynamics in the Smoluchowski limit (it was implemented early on for nucleon exchange in the dinucleus [20] where it proved to be essential for understanding the dependence of the mass distribution on energy loss in damped nuclear reactions [21]).

In this physical picture, the evolution of the nuclear shape is akin to Brownian motion. For the fissioning nucleus, the shape plays the role of the Brownian body, while the environment consists of the microscopic degrees of freedom associated with a nucleon gas. For a given shape, the nuclear surface is being continually assaulted by those nucleons and the average of these impulses constitues the associated friction force and the residual fluctuations give the shape evolution a diffusive character.

While it is generally somewhat complicated to treat this dynamical problem, considerable simplicity emerges in the strongly damped limit which is expected to be a reasonable starting point for the description of nuclear dynamics [22]. Indeed, the strongly damped (Smoluchowski) limit of standard Brownian motion can be treated as a random walk in configuration space and the magnitudes of the inertial mass and the friction affect only the overall time scale of the evolution but not the resulting ensemble of random walks in configuration space.

We therefore simulate the shape evolution as a random walk of the nuclear shape. More precisely, we consider a parametrized multi-dimensional family of shapes suitable for the fission process and let $\boldsymbol{\chi}=(\chi_1,\chi_2,\dots)$ denote the associated shape parameter. Let, at some point in the evolution, the nuclear shape be that defined by a given value of $\boldsymbol{\chi}$. Then, in the course of a brief time Δt, the accumulated effect of the nucleons impinging on the surface is a stochastic change in the shape, $\Delta\boldsymbol{\chi}$, which can be sampled from the appropriate distribution $P(\Delta\boldsymbol{\chi};\boldsymbol{\chi})$.

Because the potential energy $V(\boldsymbol{\chi})$ is given on a fixed lattice $\{\boldsymbol{\chi}_i\}$, it is covenient to recast the process as a random walk on that lattice. This can be accomplished by standard methods. Due to detailed balance, the ratio between the resulting transition probabilities for reverse processes equals the Boltzmann factor, $P(i\to i'):P(i'\to i)=\exp(-\Delta V/T)$, where $\Delta V\equiv V(\boldsymbol{\chi}_{i'})-V(\boldsymbol{\chi}_i)$ is the change in the potential energy associated with the shape change from $\boldsymbol{\chi}_i$ to $\boldsymbol{\chi}_{i'}$ and T is the (local) nuclear temperature. Such a random walk can readily be simulated by means of the familiar Metropolis procedure [23].

The above described procedure is merely preliminary and serves to illustrate the utility of this type of approach. There is obviously a need to take account of the metric in the shape space, which is related to the lattice spacings in the employed table of deformation energies. These were chosen in order to achieve typical changes of $|\Delta V|\approx 1$ MeV and our preliminary studies (involving changing the spacings or introducing a specific metric based on shape overlaps) suggest that this guiding principle was quite reasonable, since only relatively extreme modifications have a significant influence on the results. A more formal treatment is being developed [26].

While the Smoluchowski shape trajectory is independent of the overall friction strength, the present simplified treatment assumes that the friction tensor does not introduce significant misalignments between the dissipative force and the resulting shape change. Our results are reassuring in this regard and a thorough study of this central issue is underway [24].

The five shape parameters used in Ref. [18] are approximately orthogonal, especially near scission and, furthermore, the mass-asymmetry lattice is equidistant in the fragment mass number $A_{\rm f}$. Therefore there should be no significant Jacobian distortion involved in extracting the mass distribution and, together with the insensitivity to both mass and friction, this in turn should render the extracted $P(A_{\rm f})$ rather robust, reflecting primarily the features of the potential-energy surface, $V(\boldsymbol{\chi})$.

For the potential energy, $V(\boldsymbol{\chi})$, we employ tabulated values calculated for the three-quadratic-surface shape family [25] with the macroscopic-microscopic finite-range liquid-drop model [18]. For more than five thousand nuclei, these tables provide the potential energy of over five million shapes in terms of five convenient shape parameters. They are the most comprehensive available and have proven to form a good framework for understanding many important features of fission [26].

The temperature is obtained by the Fermi-gas formula, $T^2=[E^*-V]/a_A$, where E^* is the total excitation energy of the nucleus and $a_A=A/(8\text{ MeV})$ is the level-density parameter. While appropriate for the present explorative study, this simple approximation may need future refinement, such as inclusion of pairing and shell effects.

In order to illustrate the quantitative utility of the dynamical treatment described above, we have used it to calculate the fission fragment mass distribution for a number of cases of practical interest. (The calculated mass yields have been reexpressed as charge yields by means of a simple scaling, $P(Z_f)=P(A_f)A_0/Z_0$.)

Considering the fission process as a temporal evolution of the nuclear shape, we combine the original fission theory concepts introduced by Bohr [3] with the recognition that the associated statistical distribution of nuclear shapes is not established instantly when the nucleus is agitated (by the absorption of a neutron or a photon) but builds up over time. Our key assumption is that this equilibration process will terminate at scission, *i.e.* when the system finds itself with a shape for which the neck radius $c_{\rm neck}$ is so small that the nucleus will irreversibly proceed to separate into two distinct fragments without any further change in the mass asymmetry. We define scission to occur when $c_{\rm neck}$ has decreased to a specified value c_0. We employ the value $c_0=2.5$ fm but our results tend to be rather insensitive to the precise value.

By discouraging but not prohibiting uphill steps, the

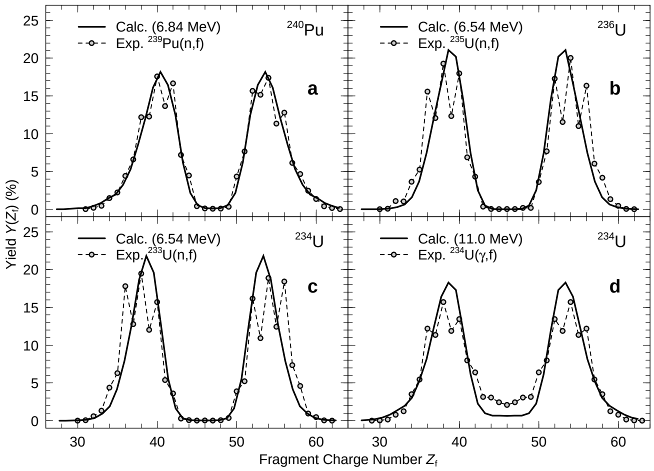

FIG. 1: Calculated and measured charge yields for fission of ^{240}Pu and ^{236,234}U. The data in (a–c) are for (n_{th},f) reactions leading to $E^* \approx 6.5$ MeV [27], while the data in (d) is for (γ,f) reactions leading to $E^* \approx 8-14$ MeV; they include contamination from fission of ^{233}U ($\approx$15%) and ^{232}U ($\approx$5%) [28]; the corresponding calculation was made for $E^* = 11$ MeV.

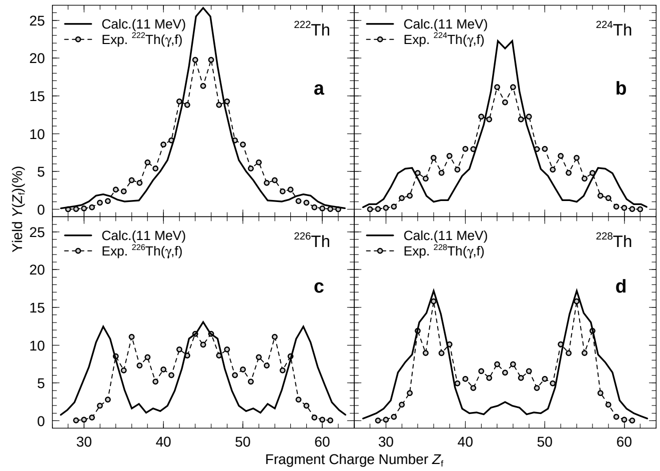

FIG. 2: Calculated charge yields for four even-even thorium isotopes compared to experimental data [28].

Metropolis sampling method ensures that a sufficiently long walk will visit each shape in accordance with its appropriate statistical weight. Initially the nuclear shape is close to that of the ground state, where it resides before being agitated. The shape will then typically make excursions around this favored shape. However, every now and then, according to the statistical probability, the path in deformation space may lead over the fission barrier and the shape is then more likely to continue towards division than to revert to a compact shape.

It is thus evident that the random walk will tend to wander around inside the barrier for many steps before eventually surmounting it. To speed up the calculation without unduly influencing the final outcome, we have augmented the potential energy by a bias term, $V_{\rm bias} = V_0\, Q_0^2/Q^2$, where Q is the quadrupole moment of the deformed nucleus and Q_0 represents the average ground-state quadrupole moment of deformed actinide nuclei. Thus, in the region of compact shapes, where Q is small, $V_{\rm bias}$ will encourage increases of Q, while it will have relatively little effect for highly deformed shapes where the mass division is decided. The resulting mass yields are not sensitive to variations in the bias strength V_0, as long as it remains small (we use $V_0 = 15$ MeV).

Figure 1 shows the calculated charge distributions for ^{239}Pu, ^{235,233}U($n_{\rm th}$, f), and ^{234}U(γ, f) together with the corresponding experimental data. (We focus on charge distributions to avoid issues related to neutron evaporation.) The results agree quite well with the data which is remarkable because no parameter was adjusted. The features of the calculated yields are thus determined essentially only by the structure of the potential-energy surfaces. (The odd-even staggering seen in the data is due to pairing and this effect is not present in the potential-energy surfaces because existing pairing models treat the fissioning nucleus as a single system, even near scission.)

The most noticeable discrepancy is an underestimate of the symmetric yield for the (γ,f) data (Fig. 1d). These were obtained with photons having energies of 8–14 MeV [28], while the calculations were made for $E^* = 11$ MeV. Furthermore, the experimental data contain contaminations from multi-chance fission.

We also compare with a sequence of thorium isotopes which constitute a more challenging test because their yield curves change qualitatively from the light to the heavy isotopes. Figure 2 shows calculated and measured charge yields for ^{222,224,226,228}Th. As for ^{234}U(γ, f) discussed above, these data [28] were obtained with photons having a wide energy range and they contain multi-chance contaminations. Although the differences between calculation and experiment are larger than seen in Fig. 1, the onset of asymmetric fission towards the heavier thorium isotopes is quite well reproduced. In fact, the deviations of the model results from the corresponding data are smaller than the differences between neighboring experimental results, which in turn differ by just one occupied neutron orbital.

The generality of the approach makes it possible to gain new insight about the fission process from the remaining differences between calculated results and experimental data. For example, the energy dependence of the symmetric-valley yield in ^{234}U (Fig. 1c-d) can be improved by refining the shape dependence of the temperature and an alternate shape dependence of the Wigner term in the potential energy [29] moves the asymmetric peaks in Fig. 2c into agreement with experiment.

The case of ^{222}Th (Fig. 2a) is particularly instructive, because the calculation yields a symmetric mass distribution even though the nuclear shape at the saddle point of the potential energy surface is reflection asymmetric with a mass ratio of 129:93. This remarkable finding implies that either the path from the isomeric minimum (which is located at a symmetric shape) to scission does not cross the potential barrier in the most favorable region (*i.e.* near the saddle point) or the shape distribution reverts from asymmetric to symmetric during the descent from saddle to scission. In either case, the result invalidates the hypothesis (see *e.g.* Refs. [7, 30–32]) that the character of the mass distribution, whether symmetric or asymmetric, is determined by the saddle shape. Rather, our result suggests that the fragment mass distribution is determined by the relatively complicated structure of the potential-energy landscape between the isomeric minimum and scission. Therefore any plausible model of the mass yields must take this into account.

It is interesting to compare the calculated mass yields with the result of statistical scission models [8, 9]. We have found that while a statistical sampling of our scission shapes often (but not always) lead to reasonable agreement with the data, it is never as good as the results of the random walk, suggesting that the shape evolution in the pre-scission landscape generally plays an important role for the resulting mass distribution [24].

In summary, we have presented a novel treatment of the shape dynamics in moderately excited nuclei and we have illustrated its practical and quantitative utility by using it to calculate fission fragment yields for several cases that have been studied experimentally, including some particularly challenging ones. Relative to previously employed methods, the present approach represents a significant advance with regard to predictive power. (We have here concentrated on nuclei with 5-20 MeV excitation but we plan to explore extensions to both higher and lower energies, including spontaneous fission.)

Taking explicit account of the equilibration process, our treatment extends in a natural way the compound nucleus concept invoked in 1939 to describe many aspects of the newly discovered fission phenomenon. It builds directly on the general picture of low-energy nuclear dynamics as being dominated by the dissipative interaction between the evolving surface and individual nucleons. This mechanism causes the nuclear shape dynamics to resemble Brownian motion and the present dynamical treatment is the first to treat the resulting stochastic shape evolution in the Smoluchowski limit where the inertial mass is immaterial. A particularly attractive feature of the approach is its generality: once the potential energy has been calculated as a function of deformation, for a sufficiently rich class of mononuclear shapes, the dynamics can readily be studied. Studies are well underway to clarify the importance of the shape metric, the friction tensor, and pairing and shell effects in the entropy [24].

We have here concentrated on applications of this treatment to the calculation of fission fragment mass distributions for which a variety of data is available. Importantly, only a single new parameter is required for this purpose, namely the critical neck radius characterizing a scission shape, and the mass yields are rather insensitive to its specific value. This degree of robustness gives the method unprecedented predictive power with regard to fission-fragment mass distributions. In particular, it can be readily employed in regions of the nuclear chart that are of special astrophysical interest and it may, for example, help to clarify the importance of fission recycling for the r-process [33, 34].

Acknowledgments

We are grateful to K.-H. Schmidt for providing computer-readable files of the data in Ref. [28] and to L. Bonneau, H. Goutte, D.C. Hoffman, A.J. Sierk, and R. Vogt for helpful discussions; T. Watanabe kindly extracted the (n,f) data from the ENDF/B-VII.0 data base.

This work was supported by the Director, Office of Energy Research, Office of High Energy and Nuclear Physics, Nuclear Physics Division of the DOE under Contract No. DE-AC02-05CH11231 (JR) and by the National Nuclear Security Administration of the U.S. Department of Energy at Los Alamos National Laboratory under Contract No. DE-AC52-06NA25396 (PM).

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