# Bimodal Symmetric Fission Observed in the Heaviest Elements

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(Received 26 August 1985; revised manuscript received 23 December 1985)

We measured the mass and kinetic-energy partitioning in the spontaneous fission of $^{258}$Fm, $^{259}$Md, $^{260}$Md, $^{258}$No, and $^{260}$[104]. All fissioned with mass distributions that were symmetric. Total-kinetic-energy distributions peaked near either 200 or 235 MeV. Surprisingly, these energy distributions were skewed upward or downward from the peak in each case, except for $^{260}$[104], indicating a composite of two energy distributions. We interpret this as a mixture of liquid-drop-like and fragment-shell–directed symmetric fission, although theory had not anticipated this phenomenon.

PACS numbers: 25.85.Ca, 27.90.+b

A central feature of low-energy and spontaneous fission of the heavy elements is the division into fragments of unequal mass. Mass-symmetric fission is a rarer mode, occurring only in two restricted regions of the chart of nuclides: the Tl-to-Ac region preceding the actinides and the region near the termination of the actinide series of elements. The causes advanced for mass symmetry and asymmetry differ markedly for each region of nuclides. The liquid-drop model describes the broadly symmetric mass distributions and moderate kinetic energies found for the low-energy–induced fission of nuclei between Tl and Ac.[1] In contrast, the highly symmetric mass division found in the heaviest Fm isotopes has been ascribed to the strong shell effects emerging near scission from fragments approaching the magic $^{132}$Sn nucleus.[2,3] Because these fission-product nuclei are spherical and stiff toward deformation, the compact configuration at scission results in a high Coulomb energy which is translated into unusually large kinetic energies for the fragments. This we call "fragment-shell–directed" symmetric fission. These two causes for mass symmetry were thought to be mutually exclusive, because they depended on the very different balance between macroscopic forces and single-particle couplings in separate regions of nuclides.

We have gathered evidence from fragment energy measurements in the spontaneous fission (sf) of the heaviest nuclides that strongly suggests that symmetric mass division in this region may arise as much from the liquid-drop process as from the influence of emerging fragment shells. We found that the total-kinetic-energy (TKE) distributions of the fragments strongly deviated from the Gaussian distributions which are observed in the fission of lighter actinides. In four of the five nuclides studied, the anomalous TKE distribution was skewed sufficiently that it could be decomposed into two Gaussian distributions. From the multiple TKE distributions and the shapes of the mass distributions, we conclude that there is a lower-energy fission component with liquid-drop characteristics which is admixed with a much higher-energy component due to fragment shells. These observations provide new insights into the fission process which we hope will inspire major theoretical advances.

We are reporting the results of coincident-fragment energy measurements on the sf of five nuclides with $Z \geqslant 100$ and $N \geqslant 156$. The very-short-lived nuclides, 1.2-ms $^{258}$No (Nurmia *et al.*[4]) and 20-ms $^{260}$[104] (Nitschke *et al.*[5] and Sommerville *et al.*[5]), were produced in fusion reactions of 68-MeV $^{13}$C ions with $^{248}$Cm and of 81-MeV $^{15}$N ions with $^{249}$Bk, respectively. Data for these were collected with a new instrument that provides a continuous on-line method for producing short-lived isotopes and measuring the energy of the fragments emitted in sf. Recoil products emerging from the target were stopped in a band of thin Al foils mounted on the rim of a 30-cm-diam wheel that spins up to 5000 rpm. These foils were rotated past opposing banks of trapezoidal-shaped surface-barrier detectors, which measured the energies deposited by coincident fragments. With event rates averaging only 5–8 coincident fissions per hour, we obtained 382 $^{258}$No sf events together with 59 $^{256}$Fm sf events. For $^{260}$[104], only 300 events were recorded along with 41 $^{256}$Fm events. A full paper will follow this brief Letter, although some information concerning these particular experiments was reported earlier.[6,7]

The half-lives of $^{258m}$Md, $^{259}$Md, and $^{260}$Md are sufficiently long to allow time for off-line mass separation for the preparation of isotopically pure sources. These Md isotopes were produced by transfer reactions in bombardments of an $^{254}$Es target with beams of 105-MeV $^{18}$O and 126-MeV $^{22}$Ne from the 88-in. cyclotron at the Lawrence Berkeley Laboratory. Recoil products from the bombardments were trapped in Ta foils, which were then flown by helicopter from the cyclotron to the Lawrence Livermore National Laboratory for mass separation. Because $^{256}$Fm (157-min sf) is one of the most abundant products of these synthesizing reactions, mass separation was a key technique in freeing $^{258-260}$Md from the massive interference caused by the sf of $^{256}$Fm. The background correction due to this isotope ranged from 8.6% for $^{259}$Md to zero for $^{260}$Md.

The desired mass fraction was collected on an Al foil (50 $\mu$g cm$^{-2}$), which was subsequently placed between two surface-barrier detectors for the measurement of the correlated fission-fragment energies. The elapsed time between the end of bombardment and the start of counting was 1 h. This short interval was an important factor which allowed us to study the sf of $^{258}$Fm and $^{259}$Md. Fragment energies were determined from calibrations of the detectors with $^{252}$Cf for which we used 181.25 MeV for the average TKE.[8] Fragments that passed through the Al supporting foil were corrected for energy losses amounting to an average of 3.2 MeV. For all of our results, fragment masses were calculated on the basis of the conservation of mass and linear momentum in the fission process.

The sf properties of 380-$\mu$s $^{258}$Fm (Hulet *et al.*[9]) produced by the electron-capture decay of 60-min $^{258m}$Md, had been roughly determined in an earlier study.[3] We are reporting new measurements based on counting of the $A=258$ mass fraction. Previous fission studies had also been made on 95-min $^{259}$Md which decays directly by sf.[10] The observation of this same activity in the $A=259$ sample confirms our earlier isotopic assignment. In comparison with our older work,[3,10] these remeasurements on mass-separated samples of $^{258}$Fm and $^{259}$Md resulted in more events, much lower contributions from the sf of $^{256}$Fm, and better energy resolution. Indeed, skewing of the TKE distributions for these nuclides became visible for the first time.

The longest-lived nuclide that we report is a hitherto unknown sf emitter with a half-life of 32 d that has been discovered in the $A=260$ mass fraction after off-line mass separation.[11] Chemical methods have identified a Md isotope as the source of this activity; thus the $Z$ and $A$ are certain. Although we attribute the decay period to $^{260}$Md, this sf activity could, in principle, result from the decay of its possible daughters, $^{260}$No or $^{260}$Fm. These nuclides are expected to have subsecond sf half-lives, and therefore, possibly be in secular equilibrium with the parent $^{260}$Md. We have eliminated $\beta$ decay to $^{260}$No as a possible source of the observed fissions by measuring the time correlations between 0.04–1-MeV $\beta$'s and subsequent fissions. We found that the time distribution was random and the same as between any two successive $\beta$ particles. Similarly, preliminary measurements of the time correlation between Fm $K$ x-rays and fission events showed that $K$-electron capture to $^{260}$Fm was less than 10%, provided that the mean lifetime of $^{260}$Fm is less than 100 ms. $L$-electron capture, due to insufficient decay energy for $K$ capture, is a marginal possibility and remains to be investigated. For now, we ascribe our sf results for the $A=260$ fraction to the direct sf of $^{260}$Md.

Figure 1 shows the mass distributions that we obtained for the five nuclides. All are symmetric but some more sharply than others. The narrowest, from $^{258}$Fm and $^{260}$Md, have full widths at half maximum of 7.5 u, while the broadest, that for $^{260}$[104], is 36 u. In a majority of these nuclides, there are wings extending far outward in mass from the central peak, a feature not so clearly obvious from earlier studies. The fraction of events in these wings is the lowest in $^{258}$Fm and $^{260}$Md and increases with $Z$. A finding common to all these five and to other very heavy nuclides[12] is that events with masses residing in these wings are associated with low TKE values while events with TKE values $>220$ MeV give an exceedingly sharp mass distribution around symmetry. When sf events with TKE values less than 200 MeV are chosen, the resulting mass distributions are very broad and flat. Some might even be characterized as asymmetric if our statistical samples were larger. Thus we find a low-energy form of fission with very broad mass distributions and a high-energy form associated with sharply symmetric mass division.

In four of the nuclides, the TKE distributions deviated substantially from Gaussian distributions. This is a phenomenon not previously observed in the sf of actinide nuclei.[13] After first observing this anomaly last year for $^{258}$No,[7] we have since found skewed TKE distributions in three additional isotopes, as shown in Fig. 2. With this added confirmation, it appears to be more of a general trait rather than a singular oddity. Figure 2 makes it clear that asymmetric tailing from the peak energy can occur toward either higher or lower energies. Furthermore, we note that the peak in each of the TKE curves falls in one of two distinct positions, either near 200 or 235 MeV. Skewing of the TKE curves results in the distribution of an appreciable portion of the events into each of these two main energy locations. Least-mean-squares fitting of two Gaussian distributions to the TKE curves gave the following centroids: $^{258}$Fm, 205 and 232 MeV; $^{259}$Md, 201 and 235 MeV; $^{258}$No, 203 and 235 MeV; $^{260}$Md, 195 and 234 MeV. These values were calculated by our setting the width of the lower-energy Gaussian to the value obtained for $^{260}$[104].

The sf of $^{260}$[104], in which the low-energy mode appears to dominate, provides an exception to the skewing behavior noted above for the other nuclides. Because of the low average TKE and the broad TKE and mass distributions obtained for this nuclide, we have interpreted these properties as being characteristic of a liquid-drop mode of fission.[6] The outer asymmetric fission barrier, which may be responsible for asymmetric mass distributions in the fission of all but

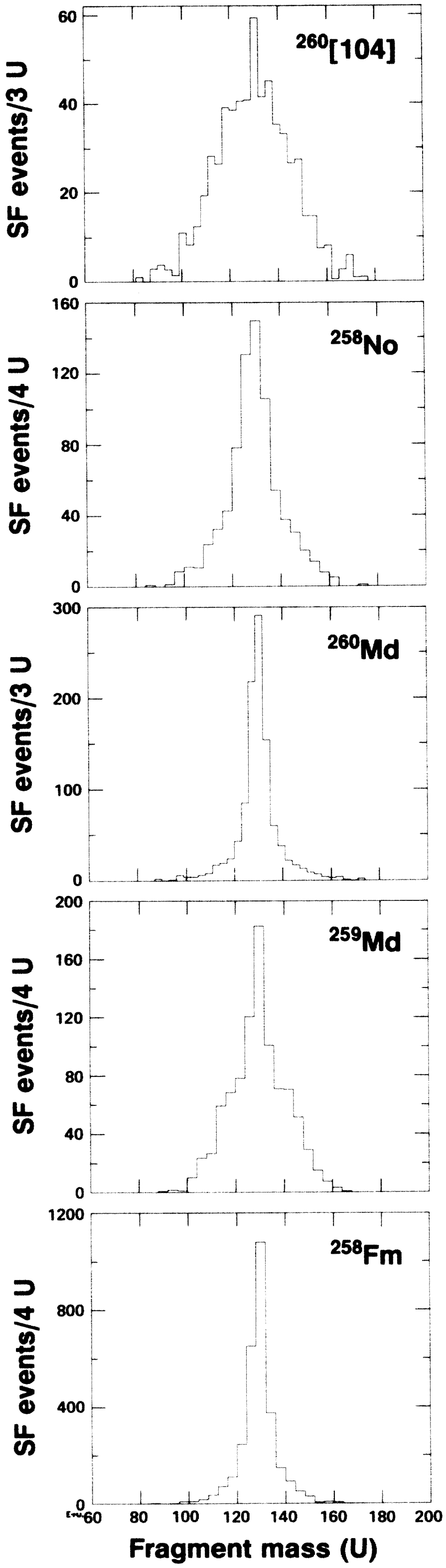


FIG. 1. Provisional mass distributions obtained from correlated fragment energies. A small contribution from $^{256}$Fm has been subtracted from all but the $^{260}$Md distribution. The mass bins have been chosen to be slightly different for each nuclide.

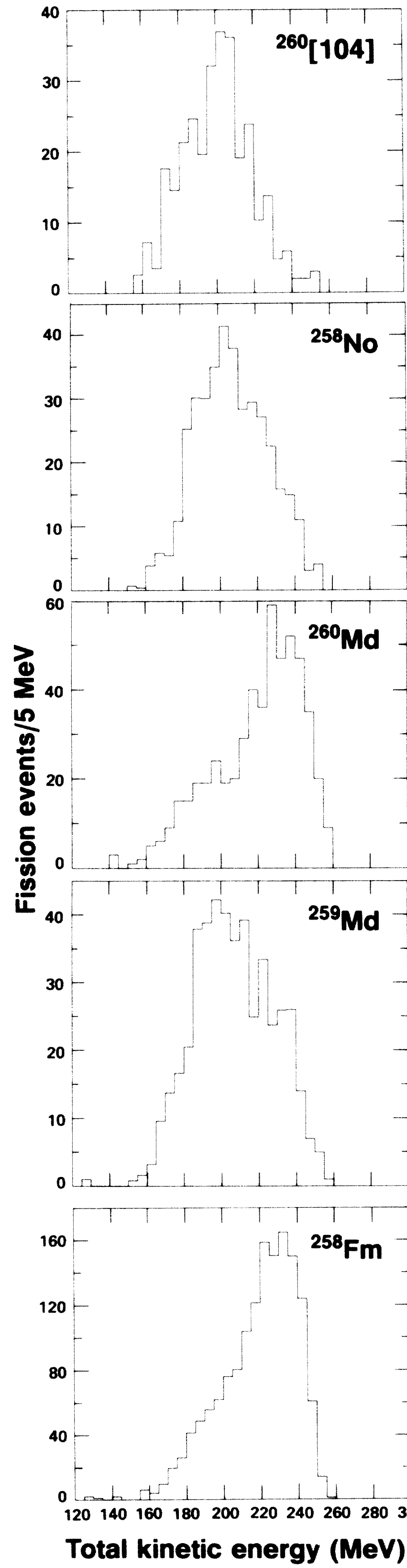


FIG. 2. Provisional total-kinetic-energy distributions. A small contribution equivalent to the known amount of $^{256}$Fm has been subtracted from all but the $^{260}$Md distribution.

the heaviest actinides,[14] is predicted to have disappeared below the ground state in $^{260}[104]$.[15] Therefore, passage through the remaining inner barrier, which contains liquid-drop and shell components, should yield symmetric mass distributions and average TKE values that conform to the estimates of Viola, Kwiatkowski, and Walker for liquid-drop fission.[16] Mass symmetry is expected because only reflection-symmetric shapes are allowed by the underlying liquid-drop fission barrier.

The best explanation for the high-TKE mode of sf depends on the influence of fragment shells which are emerging between the saddle and scission points.[2,3] In this process, fragment shells near the doubly magic $^{132}$Sn lower the potential-energy path and, thus, guide the mass division toward Sn isotopes near the 82-neutron closed shell. As $N$ decreases below 158 neutrons and $Z$ of the fissioning species increases beyond 100, the opportunity to divide into two Sn fragments diminishes. Hence we observe a trend away from sf characterized by unusually high TKE values and sharply symmetric mass splits and toward the liquid-drop mode represented by $^{260}[104]$.

Because there are unmistakable high- and low-energy fission distributions occurring concurrently in the same nuclide, we conclude that these nuclides sustain two strikingly different modes of fission. We suggest that the liquid-drop and the "fragment-shell" processes are separately responsible for the distinct portions of the TKE distributions. We arrived at this interpretation on the basis that only these two fission models can account for the large ($\sim 35$ MeV) difference between the low- and high-energy modes. In addition, the symmetric mass distributions observed, with their disparate widths, are consistent with these two models. Our current view is that passage through a single barrier, where only symmetric oscillations prevail, would, presumably, lead only to a broadly symmetric mass division and liquid-drop TKE's. However, the presence of strong shells in the fragments from fission of the very heavy Fm and Md isotopes produces an alternate potential-energy valley which is competititve with the liquid-drop valley. A ridge in the potential-energy surface between these two valleys would seem necessary to prevent further equilibration after the saddle point. In support of this picture, we note that a potential-energy surface containing two separated valleys has been calculated by the Hartree-Fock approach for the asymmetric fission of $^{240}$Pu.[17]

We wish to thank M. G. Mustafa, P. Möller, W. J. Swiatecki, and W. D. Myers for helpful discussions of the theoretical problems presented by this work. We also thank the staff and operating crew of the 88-in. cyclotron for the irradiations. This work was supported by the U.S. Department of Energy under Contract No. W-7405-Eng-48.

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