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BIMODAL FISSION

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ABSTRACT

In recent years, we have measured the mass and kinetic-energy distributions from the spontaneous fission of ^{258}Fm, ^{259}Md, ^{260}Md, ^{258}No, ^{262}No, and ^{260}[104]. All are observed to fission with a symmetrical division of mass, whereas the total-kinetic-energy (TKE) distributions strongly deviated from the Gaussian shape characteristically found in the fission of all other actinides. When the TKE distributions are resolved into two Gaussians, the constituent peaks lie near 200 and near 233 MeV. We conclude two modes or bimodal fission is occurring in five of the six nuclides studied. Both modes are possible in the same nuclide, but one generally predominates. We also conclude the low-energy but mass-symmetrical mode is likely to extend to far heavier nuclei; while the high-energy mode will be restricted to a smaller region, a region of nuclei defined by the proximity of the fragments to the strong neutron and proton shells in ^{132}Sn.

INTRODUCTION

Some years ago we and others had found a sudden transition to sharply symmetrical mass distributions from neutron-induced and spontaneous fission (SF) of the heaviest fermium isotopes.[1,2,3] Furthermore, the spontaneous fission of ^{258}Fm and ^{259}Fm was characterized by total kinetic energies (TKE) approaching 240 MeV or nearly the Q value of the fission reaction. Elsewhere on the nuclide chart, only the isotopes of the elements Tl through Ac fission with symmetrical mass distributions.[4] On-the-other-hand, asymmetrical (two-humped) mass distributions are a common feature in low-energy-induced fission and spontaneous fission of the actinides until ^{258}Fm is reached. The SF properties of ^{258}Fm and ^{259}Fm are remarkable because of the sudden onset of mass symmetry and the high fragment energies. To determine the range of this behavior, to provide critical tests of theory, and to improve our predictions for heavier and more distant nuclei, it was necessary to extend these fission studies to nuclides with greater atomic and neutron numbers.

Using a variety of experimental techniques that are described elsewhere,[5] we have investigated the mass and kinetic-energy distributions from the SF of ^{258}Fm, ^{259}Md, ^{260}Md, ^{258}No, ^{262}No, and ^{260}[104].[5,6,7] The total number of fission events accumulated for each nuclide ranged from 300 to ~2000. It was not possible to obtain larger numbers because of the necessity to produce these nuclides by heavy-ion reactions with actinide targets. For such reactions, the formation cross sections were often as low as 10 to 20 nb. Another unfavorable factor was the extremely short SF half-lives of three of the isotopes, each of these being 20 ms or less. To measure their fragment energies, instruments were developed especially for this purpose, however, their overall detection efficiencies were low, lying near 5 to 6%. Nevertheless, we were able to obtain accurate fragment energies without large interferences from other nuclides coproduced in the nuclear reaction and that also decayed by SF. From the energies of the coincident fragments, we calculated the sum of both fragment energies (TKE) and the provisional fragment masses on the basis of conserving mass and momentum. In this report, we shall be concerned only with the results and conclusions and refer the reader to the references cited above for additional details.

RESULTS

In Figs. 1 and 2, we show the mass and total-kinetic-energy (TKE) distributions for five of the six nuclides studied. We show in Figs. 3 and 4 the same kinds of distributions for 5-ms ^{262}No, which we recently discovered as the electron-capture daughter of ~4-h ^{262}Lr. A small ^{256}Fm contribution, amounting to at most 13%, was subtracted from the distributions for ^{258}Fm, ^{259}Md, ^{258}No, and ^{260}[104]. The nuclides ^{260}Md and ^{262}No were produced free of ^{256}Fm. Because of the pains we had taken to keep the ^{256}Fm contribution small, there was only a slight impact on any distribution from this background subtraction.

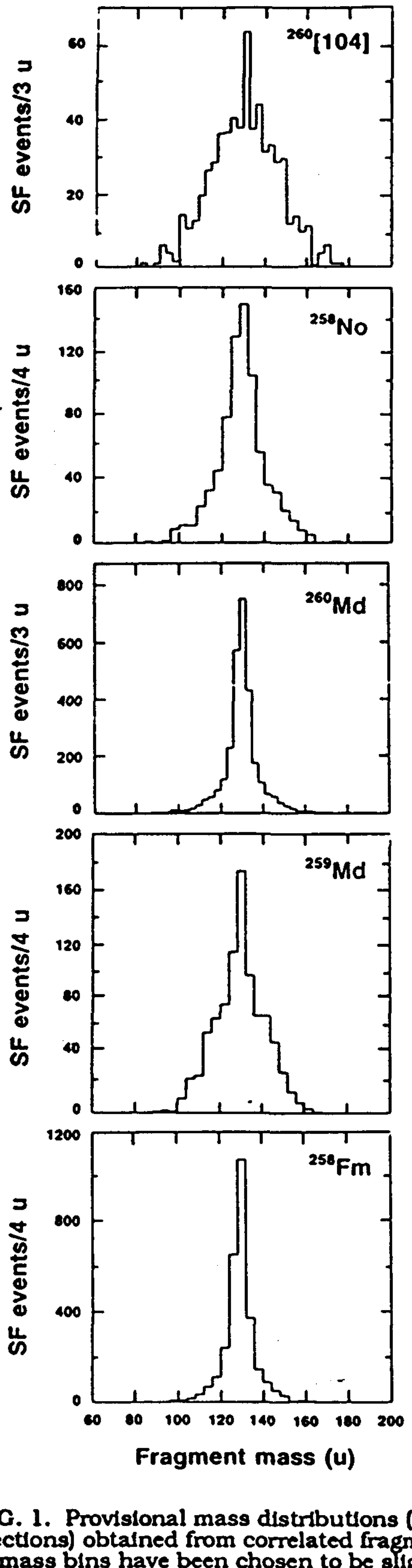

FIG. 1. Provisional mass distributions (no neutron corrections) obtained from correlated fragment energies. The mass bins have been chosen to be slightly different for each nuclide. The distributions are net after subtracting a small ^{256}Fm component.

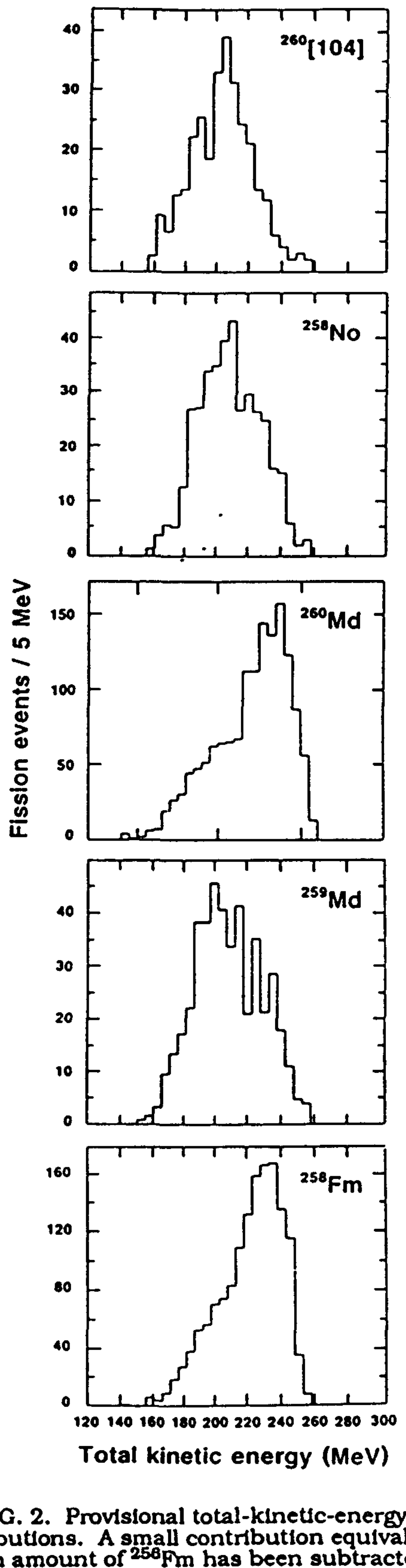

FIG. 2. Provisional total-kinetic-energy (TKE) distributions. A small contribution equivalent to the known amount of ^{256}Fm has been subtracted from all but the the ^{260}Md distribution.

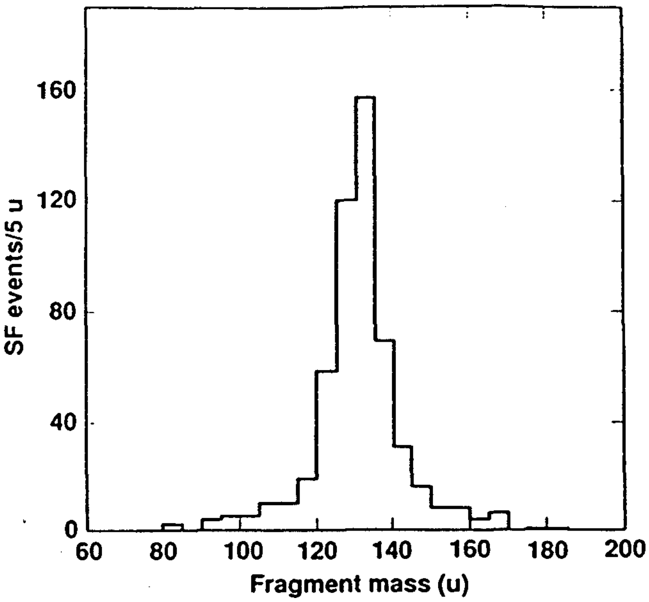

FIG. 3. Provisional mass distribution obtained for 5-ms ^{262}No.

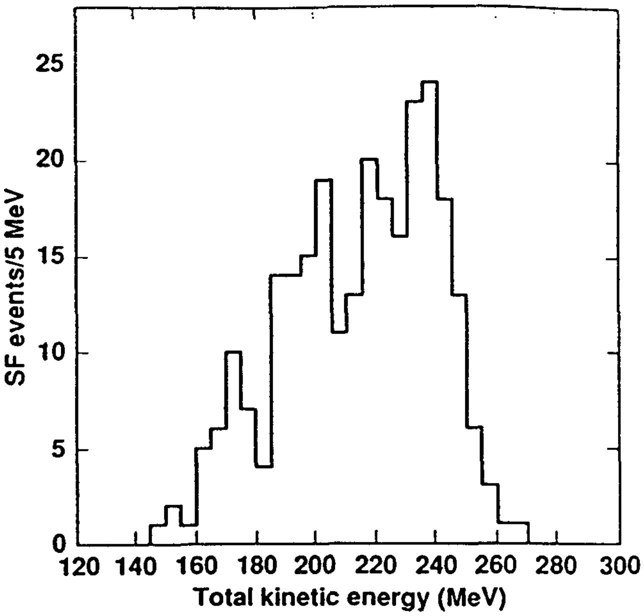

FIG. 4. Total-kinetic-energy distribution from the spontaneous fission of ^{262}No.

Aside from the highly symmetrical mass distributions, we would like to point out a unique feature in the TKE distributions that led us to suggest bimodal fission is occurring in these nuclides. If one is familiar with the TKE distributions measured for other actinide nuclei, it is apparent that five of the six TKE distributions shown here strongly deviate from the Gaussian shape found for lighter actinides. A decided asymmetry is imparted by conspicuous tailing in either energy direction from the central peak. If one inspects these distributions further, it will found that the peak of each distribution is not randomly located along the energy axis, but is positioned very near either 200 or 233 MeV. Although the major peak is at one of these locations in each case, asymmetric tailing from the peaks distributes an appreciable number of events into one or the other these two main energy locations.

When we considered these unusual features, it seemed rather clear that each of the TKE curves, except for ^{260}[104], was a composite of two energy distributions, with each most likely being Gaussian. We then attempted to test this hypothesis by decomposing each of the composite distributions into two Gaussian curves by least-mean-squares fitting. Because the TKE peak for ^{260}[104] appeared to be a single peak and, therefore, a model for the low-energy peak near 200 MeV, we used its full-width-at-half-maximum as a fixed parameter in the Gaussian fitting for the other five nuclides. We resolved each of the gross TKE distributions for ^{258}Fm, ^{258}No, ^{259}Md, ^{260}Md, and ^{262}No into two Gaussian distribution as shown in Figs. 5 and 6. To gauge the accuracy of this procedure, we calculated reduced χ^2 values, which are given in Table I together with the peak centroids and abundances. Because the reduced chi-squares

TABLE I. Parameters obtained from least-mean-squares fitting of two Gaussians to the TKE curves. Reduced χ^2 is a measure of the quality of fit, where values from 0.5 to 1.5 indicate a reasonable probability of a good match.

Nuclide	Low-energy Peak (MeV)	Abundance (%)	High-Energy Peak (MeV)	Abundance (%)	Reduced χ^2
^{258}Fm	205	50	230	50	1.32
^{258}No	204	96	232	4	0.55
^{259}Md	202	88	234	12	0.81
^{260}Md	200	42	235	58	1.10
^{260}[104]	200	100	----	----	0.63
^{262}No	199	53	235	47	0.94

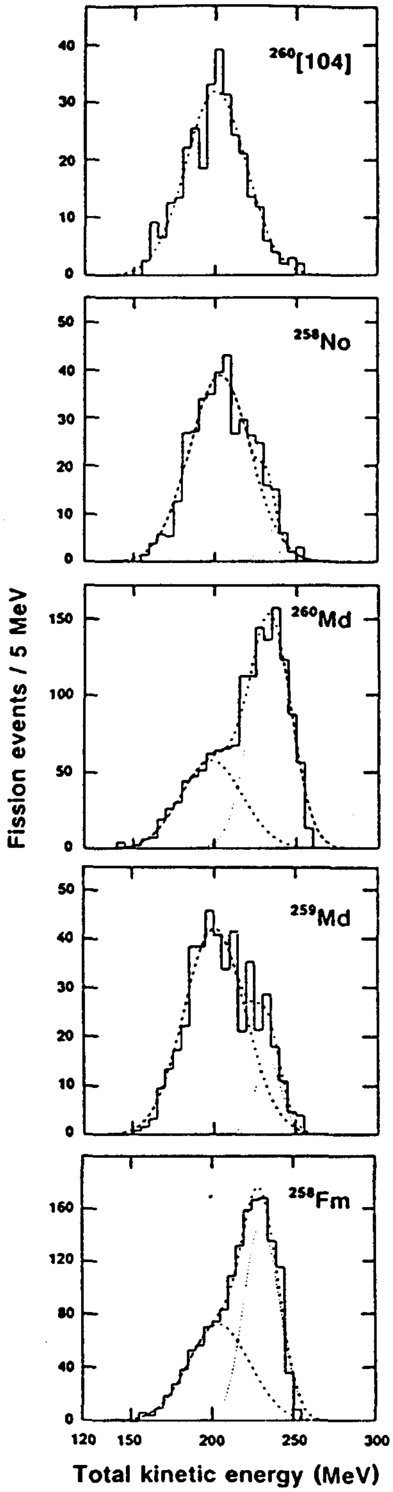

FIG. 5. Unfolding of the asymmetric TKE distributions of Fig. 2 into two Gaussian's by least-mean-squares fitting.

are near unity, the fitting of two Gaussian functions appears to be a good approximation to the parent distribution. Although the fits to two Gaussians appear to be excellent, this analytical approach should be taken only as suggestive and not as proof.

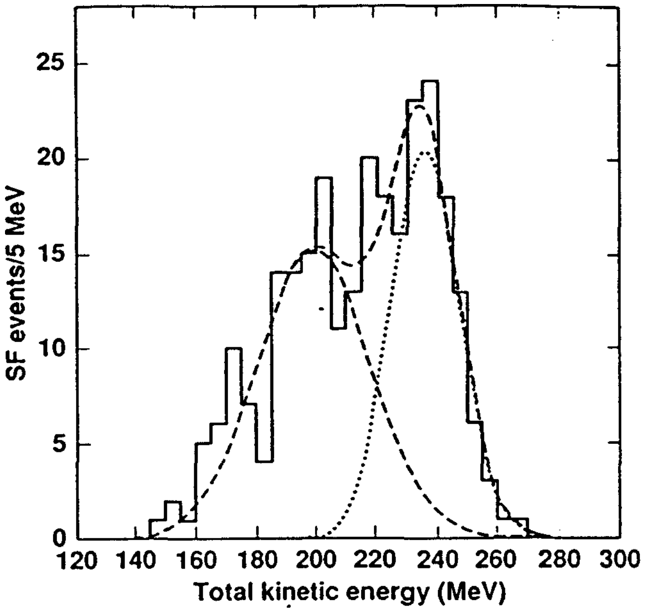

FIG. 6. Same as for Fig. 5 but for ^{262}No.

Returning to the mass distributions for these nuclides, we find that all are symmetrical, unlike those from other actinides. However, the full widths of the mass peaks vary over a range from 7.9 u to 36 u at half-maximum. That for ^{260}[104] is the broadest and is associated with a broad, low-energy TKE distribution. The remainder have a narrow and sharply symmetrical distributions around mass symmetry. The nuclides with the narrowest mass distributions also show a preponderance of high-TKE events. We can demonstrate this obvious feature by sorting events into two bins representing events with TKEs either above or below 220 MeV. This energy was arbitrarily chosen as the dividing line between the low- and high-energy TKE distributions pictured in Figs. 5 and 6. In Fig. 7, we show the mass distributions obtained for five of the nuclides from the events within each of these energy bins. The fission events with high kinetic energies produce fragments lying very close to mass symmetry while the low-energy events produce much broader but still symmetrical distributions. When we choose events with TKEs less than 200 MeV, the mass distributions become even broader and are nearly flat but remain symmetrical, with the exception of ^{258}Fm and ^{259}Md, which revert to asymmetrical distributions. We conclude from these observations that the mass distributions offer an additional feature that is as distinctive as the asymmetrical TKE distributions in pointing to two modes of fission.

CONCLUSIONS

From our observation of distinguishing features in the TKE and mass distributions from five of the six nuclides studied, we suggest there are two different fission processes that produce two modes of fission. In one mode, which we identify as the high-energy mode, the TKE approaches the Q value of the fission reaction and the fragments are closely clustered around mass 129 to 130. Because so much of the available energy has been expended as fragment kinetic energies, this mode has been described as "cold fission" for the lighter actinides for the reason that very little energy is left for internal excitation of the fragments.[8] Our other or low-energy mode is characterized by very broad symmetrical mass and TKE distributions with an average TKE around 200 MeV. The SF properties of $^{260}[104]$ would appear to typify the low-energy mode, whereas, in the remaining five nuclides, the low-energy mode is a component admixed in varying portions with the high-energy mode. It is these five nuclides that undergo bimodal fission.

From Coulomb repulsion arguments, the configuration for the high-energy mode is required to be compact at the scission point. Additionally, the two fragments must be born with very little deformation energy, again because most of the available energy is removed in the form of the kinetic energy of the fragments. Thus, the fragments are nearly spherical with masses very close to A=130. These properties are the ones known for nuclei near the doubly-closed-shell ^{132}Sn. Indeed, there are theoretical reasons for expecting the mass division to be channeled in the direction of the doubly magic Sn isotopes. There is an emergence of fragment shells between the saddle and scission point that lower the potential-energy path and, thereby, favor the mass division into spherical Sn isotopes near the 82-neutron closed shell.[9,10,11,12] We can then account for the high-energy mode on the basis of the special properties of fragments coming from mass-symmetrical division of nuclei with $Z \geq 100$ and $N \geq 156$.

An equally plausible explanation for the low-energy mode is not so obvious. We believe it is associated with the dropping of the second fission barrier below the ground state. Both spontaneous fission half-lives and theory indicates the outer fission barrier is disappearing in the region of nuclei we have studied.[13,14,15] Our reason for believing that the disappearance of this outer barrier will cause a reversion to mass symmetry is because theorists have determined that the second or outer fission barrier for mid-actinides is lowered by 0.5 to 2 MeV when shapes from asymmetrical deformations are included in their calculations of potential-energy surfaces (PES).[16] The outer barrier is large and, in some cases, the dominant barrier for many of the lighter actinides. That it favors mass-asymmetrical shapes may well be the explanation for their asymmetrical mass distributions. When this barrier drops below the ground state, passage through the remaining inner barrier would tend to promote mass-symmetrical division. This is because the PES at the first barrier lies lower for reflection- and mass-symmetrical shapes, being stiff toward any asymmetrical deformations.[15]

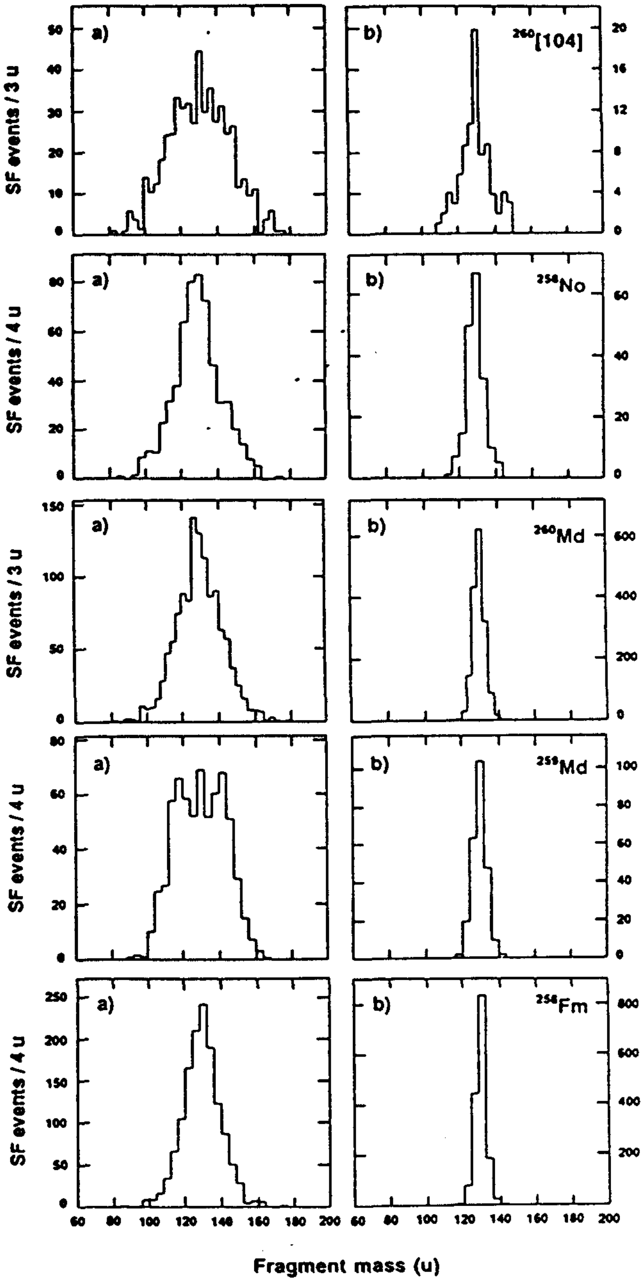

FIG. 7. Mass distributions obtained by sorting fission events according to their total-kinetic energies: (a) for events with TKEs <220 MeV and (b) those with TKEs ≥220 MeV.

If our assumption of the cause of the low-energy mode is correct, we would expect this mass-symmetrical mode to extend to far heavier nuclei, including those still undiscovered. This prediction is based on the fact that all PES calculations show the second barrier not only dropping below the ground state but completely vanishing for all nuclei with Z ≥ 106. This provides the condition for what we believe is necessary for low-energy, mass-symmetrical fission to prevail. We would expect the fission properties we observe for ^{260}[104] to be typical of nuclides with equal or greater atomic numbers. However, it would not be surprising to find an intrusion of the high-energy mode as the neutron number approaches 164, with the consequent opportunity to divide into fragments with closed 82-neutron shells. We have seen such a propensity in the SF properties of ^{262}No, but it is unknown as to the upward extent in Z that fragment-shell effects will continue to influence the fission process. Nevertheless, we would suggest that the high-energy mode will disappear as rapidly as it appeared, which is when strong fragment shells are no longer available.

The explanations we offer for each mode of bimodal fission are based upon very general features previously established by PES calculations for static deformation. Each mode is derived from the effects of shell structure: one in the parent fissioning nucleus and the other from single-particle couplings in the fragments. For essentially a dynamic process, a surprising aspect is the high degree of qualitative agreement between ours and others experimental findings with the fission properties estimated from static PES. We can only conclude that the dynamic process is adiabatic and that the collective motions are strongly coupled with the intrinsic internal structure of the nucleus during deformation. Without this hypothesis, we would be unable to explain either of the fission modes or the sharp changes in fission properties from the addition of a single nucleon such as from ^{258}Fm to ^{259}Md or ^{259}Md to ^{260}Md. All of these effects appear to approximately track variations in the ground-state Nilsson structure of the individual nuclides, which would, therefore, need to be preserved during dynamic deformation. Static PES derived from the Strutinsky/Nilsson formulation include microscopic shell energies coming from internal structure; hence, the calculated fission barriers reflect much, but not all, of the variations in shell structure from nuclide to nuclide. The ability to include microscopic features in dynamical calculations does not exist yet.

In summary, the fission properties we find for these very heavy isotopes strongly suggest two discrete modes or bimodal nuclear fission. One, the low-energy mode appears to be related to the disappearance of the second fission barrier while the high-energy mode is driven by the opportunity to divide into spherical fragments near doubly-magic ^{132}Sn. Simultaneous occurrence of both modes is likely due to a coincidental alignment of favorable shell structures in the fragments and the fissioning species within the same select group of nuclei.

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REFERENCES

1. W. JOHN, E. K. HULET, R. W. LOUGHEED, and J. J. WESOLOWSKI, Phys. Rev. Lett. 27, 45 (1971).
2. D. C. HOFFMAN, J. B. WILHELMY, J. WEBER, W. R. DANIELS, E. K. HULET, R. W. LOUGHEED, J. H. LANDRUM, J. F. WILD, and R. J. DUPZYK, Phys. Rev. C **21**, 972 (1980).
3. E. K. HULET, R. W. LOUGHEED, J. H. LANDRUM, J. F. WILD, D. C. HOFFMAN, J. WEBER, and J. B. WILHELMY, Phys. Rev. C **21**, 966 (1980).
4. *e.g.* M. G. ITKIS, V. N. OKOLOVICH, A. YA. RUSANOV and G. N. SMIRENKIN, Z. Phys. A **320**, 433 (1985).
5. E. K. HULET, J. F. WILD, R. J. DOUGAN, R. W. LOUGHEED, J. H. LANDRUM. A. D. DOUGAN, P. A. BAISDEN, C. M. HENDERSON, R. J. DUPZYK, R. L. HAHN, M. SCHÄDEL, K SÜMMERER, and G. R. BETHUNE, submitted to Phys. Rev. C (1988).
6. E. K. HULET, J. F. WILD, R. J. DOUGAN, R. W. LOUGHEED, J. H. LANDRUM. A. D. DOUGAN, M. SCHÄDEL, R. L. HAHN, P. A. BAISDEN, C. M. HENDERSON, R. J. DUPZYK, K SÜMMERER, and G. R. BETHUNE, Phys. Rev. Lett. **56**, 313 (1986).
7. R. W. LOUGHEED, E. K. HULET, J. F. WILD, K. J. MOODY, R. J. DOUGAN, C. M. GANNETT, R. A. HENDERSON, D. C. HOFFMAN, and D. M. LEE, Nucl.Chemistry Division Ann. Rept., Lawrence Livermore National Laboratory, Report No. UCAR 10062/88, 1988 (unpublished), p. 135.
8. C. SIGNARBIEUX, M. MONTOYA, M. RIBRAG, C. MAZUR, C. GUET, P. PERRIN, and M. MAUREL, J. Phys. Lettres (Paris) **42**,

L-437 (1981); C. SIGNARBIEUX, G. SIMON, J. TROCHON, and F. BRISARD, J. Phys. Lettres (Paris) **46**, L-1095 (1985).

9. J. R. NIX, Annu. Rev. Nucl. Sci. **22**, 341 (1972).
10. U. MOSEL, J. MARUHN, and W. GREINER, Phys. Lett. **34B**, 587 (1971).
11. U. MOSEL and H. W. SCHMITT, Phys. Rev. C **4**, 2185 (1971).
12. M. G. MUSTAFA, U. MOSEL, and H. W. SCHMITT, Phys. Rev. Lett. **28**, 1536 (1972); M. G. MUSTAFA, U. MOSEL, and H. W. SCHMITT, Phys. Rev. C **7**, 1519 (1973).
13. J. RANDRUP, S. E. LARSSON, P. MÖLLER, S. G. NILSSON, K. POMORSKI, and A. SOBICZEWSKI, Phys. Rev. C **13**, 229 (1976).
14. A. BARAN, K. POMORSKI, A. LUKASIAK, and A. SOBICZEWSKI, Nucl Phys. **A361**, 83 (1981); H. C. PAULI and T. LEDERGERBER, in Proceedings of the Symposium on the Physics and Chemistry of Fission, Rochester, New York, 1973 (International Atomic Energy Agency, Vienna, Austria, 1974), Vol. I, p. 463.
15. M. G. MUSTAFA and R. L. FERGUSON, Phys. Rev. C **18**, 301 (1978).
16. P. MÖLLER and S. G. NILSSON, Phys. Lett. **31B**, 283 (1970); H. C. PAULI, T. LEDERGERBER, and M. BRACK, Phys. Lett. **34B**, 264 (1971); G. GUSTAFSSON, P MÖLLER, and S. G. NILSSON, Phys. Lett. **34B**, 349 (1971).