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Unusually low fragment energies in the symmetric fission of ^{259}Md

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The 103-min isotope ^{259}Md has been identified as the daughter of an electron-capture decay branch of ^{259}No produced via the ^{248}Cm(^{18}O,$\alpha 3n$) reaction. Chemical separations were used to confirm the identity of ^{259}Md, which decays by spontaneous fission. The kinetic energies of coincident fission fragments were measured, corresponding to a fragment mass which is highly symmetric, similar to those of ^{258}Fm and ^{259}Fm. However, the total kinetic energy distribution for ^{259}Md is considerably broader (FWHM $\sim$60 MeV) than those of ^{258}Fm and ^{259}Fm, and peaks at 201 MeV, about 35–40 MeV lower in energy. Furthermore, the maximum total kinetic energy of 215 MeV for mass-symmetric events is about 30 MeV lower than for similar events from the spontaneous fission of ^{258}Fm and ^{259}Fm. A hypothesis that this energy difference resulted from the emission of light, hydrogen-like particles at scission in a large fraction of ^{259}Md spontaneous fission decays was shown to be unfounded. From experiments to observe such particles with counter telescopes, an upper limit of 5% was determined for the fraction of fission events accompanied by light-particle emission. The total kinetic energy deficit at mass symmetry must, therefore, be distributed between internal excitation energy and fragment deformation energy at scission. Although the presence of a large amount of fragment deformation energy seems incompatible with symmetric fission into spherical Sn-like fragments, we prefer this explanation because the low total kinetic energy suggests a lowered Coulomb energy resulting from greater separation of the charge centers of deformed fragments at scission.

[RADIOACTIVITY, FISSION ^{259}Md (SF); measured $T_{1/2}$, fragment-fragment coin, deduced mass, TKE distributions. ^{259}No; measured EC decay to ^{259}Md, upper limit to SF decay.]

I. INTRODUCTION

A major transition in fission behavior appears for heavy nuclei lying between $Z = 98$ and 104. The earliest manifestations of this behavior are the extremely sharp decreases in the spontaneous fission (SF) half-lives[1a] and the appearance of symmetric fission in the heaviest Fm ($Z = 100$) isotopes.[1b–3] Another indication of abrupt changes in systematic behavior was the reversal in the trend of SF half-lives rapidly decreasing with increasing N after the 152-neutron subshell.[4] Between ^{254}No and ^{258}No, SF half-lives fall by a factor of 10^8, whereas in the element 104 isotopes they increase by a factor of about 40 between ^{254}[104] and ^{260}[104].[5] These sharp departures from the fission behavior of the lighter actinides tend to make this region of nuclei an important one for the study of the fission process from both an experimental and theoretical viewpoint.

Fission fragment kinetic energies and the mass distributions derived from them provide the most detailed information on the fission process. For elements beyond Fm these properties have been measured for only ^{252}No (Refs. 6 and 7) and ^{262}[105].[8] The mass distribution of ^{252}No SF is clearly asymmetric, but this fissioning nuclide is too light to provide a relevant test of fission theories. In the case of ^{262}[105], the measured mass and kinetic energy distributions were masked to a large extent by a high SF background from 2.6-h ^{256}Fm, rendering the conclusion that its mass distribution is asymmetric as problematical.

The discovery of ^{259}No as a 1-h α emitter with an associated SF activity[9] provided an opportunity to study the SF properties of a heavy transfermium nuclide without interference from the SF of lighter actinides. Although earlier workers had tentatively

TABLE I. Decay-energy predictions for ^{259}Md and ^{259}No.

^{259}No Q_{EC} (MeV)	^{259}Md Q_{EC} (MeV)	^{259}Md Q_α (MeV)	Reference
+1.24	+0.33	7.17	10
+0.32	−0.30	7.18	11
+0.45	−0.57	6.91	12
+1.12	−0.31	7.06	13
+1.1	−0.4	6.9	14
+0.87	−0.37	6.93	15
+0.89	−0.71	6.60	16
+1.18	+0.33	7.35	17 (Myers)
+1.01	+0.12	7.34	17 (Groote, Hilf, and Takahashi)
+0.9	+0.2	6.8	17 (Seeger and Howard)
+0.51	−0.27	7.17	17 (Liran and Zeldes)

assigned this SF activity to ^{259}No with an abundance of about 20%, we felt that there was a possibility that the fissions arose from the decay of ^{259}Md produced by electron-capture (EC) decay of ^{259}No. Calculations of Q_{EC} for ^{259}No (Refs. 10−17) (see Table I) indicated a possible EC-decay branch of a few to over 40% for an allowed ground-to-ground state transition with $\log ft = 6.2$, an average value for this mass region.[18] More importantly the fission properties of either ^{259}Md, a nuclide with 158 neutrons, or ^{259}No would be of considerable interest in testing theories concerning fission-fragment mass-energy distributions and potential energy surfaces.

Once we had identified the source of the SF activity as ^{259}Md, we aimed our investigation toward obtaining the fission mass distribution to determine if a trend toward asymmetry would appear as the proton number is increased beyond that of Fm. It should be noted that ^{259}Md is only one proton added to ^{258}Fm, which undergoes SF in a highly mass-symmetric mode.[2] In addition, the total kinetic energy (TKE) released in the SF of ^{258}Fm is 238 MeV, a very high value and one close to the Q value for the fission process.

In recent years, fragment shell structure has appeared to be the more likely factor in determining the mass and TKE distributions from the fission process, as evidenced by the appearance of symmetric fission in the heaviest Fm isotopes. The magnitude of the TKE is related to the ability of the fragments to resist deformation. In the case of the SF of ^{258}Fm and ^{259}Fm, the mass division is highly symmetric due to the energetically-favored formation of two very stable, Sn-like fragments with $Z = 50$ and $N \sim 79$. These fragments possess high spherical rigidity and, therefore, very little of the total energy available from fission is lost to deformation, resulting in high values of TKE. According to Mustafa and Ferguson,[19] increasing Z above 100 while maintaining N essentially constant results in a transition back to asymmetry. Fission then remains asymmetric until N approaches 164, when the effect of the emergence of the $N = 82$ shell during fragment formation becomes dominant. Thus, for ^{259}Md, we expected the first indications of this transition from symmetry to asymmetry in the mass distribution.

Our measurements of the kinetic energy and mass distributions for the SF of ^{259}Md reported in this paper did indeed show a slightly larger asymmetric fission component than does the mass distribution obtained from the SF of ^{258}Fm. However, several surprising dissimilarities were observed that prompted further measurements of these distributions and a search for a very large enhancement in the emission of hydrogen-like particles accompanying fission. The main difference between the SF of ^{259}Md and ^{258}Fm is the broader and lower (by 37 MeV) TKE distribution measured for ^{259}Md. The question posed by this unexpected reduction in kinetic energy is the form in which the excess energy exists at scission. Our measurement of light-particle emission in ^{259}Md was directed at examining an attractive possibility.

II. DISCOVERY EXPERIMENTS

We performed experiments to identify the source of the SF activity associated with ^{259}No and, subsequently, to determine fragment mass and kinetic energy distributions and other decay characteristics of this activity. In all of our experiments, we initially prepared a radiochemically-pure sample of ^{259}No, produced via the (^{18}O,$\alpha 3n$) reaction with ^{248}Cm at the 88-inch Cyclotron at the Lawrence Berkeley Laboratory. The ^{248}Cm targets, typically about 0.9 mg/cm^2 thick, were prepared by vacuum sublimation[20] of ^{248}CmF$_3$ in a 6-mm-diameter spot onto a 0.013-mm-thick Be foil. The energy of the ^{18}O ions entering the target was 97 MeV; about 2 MeV was lost upon passing through the target.

The target was clamped in a Cu block and mounted facing away from the incoming cyclotron beam. The target was cooled by a stream of N$_2$ gas impinging on the face of the Be foil opposite the Cm deposit. Reaction products recoiling from the target were collected on a thin foil of either Au or Pd mounted about 3 mm directly behind the target. At the end of a bombardment, typically 2 h long, the foil was dissolved in HCl-H$_2$O$_2$ and the Au or Pd removed by adsorption onto an anion-exchange column. The eluate from this column was evaporated to dryness, redissolved in 0.1 *N* HCl solution, and loaded onto a chromatographic-extraction column consisting of di-(2-ethylhexyl)-orthophosphoric acid (HDEHP) dissolved in *n*-heptane and supported on an inert fluorocarbon powder. Only No, among the actinides produced in the bombardment, exists commonly in the divalent oxidation state in aqueous solution rather than the trivalent state usual to the transplutonium actinides. Thus, it was not extracted by the chromatographic column from 0.1 *N* HCl solution. Other contaminating SF activities produced in the bombardment, such as ^{256}Fm, ^{254}Fm, and ^{248}Cm transferred from the target, were extracted, separating them from the No. Any transactinides produced in the bombardments are too short-lived to have survived the time required for the chemical separations. Therefore we were assured of starting with a pure source of ^{259}No from which we could observe the decay of a SF branch or the growth of ^{259}Md SF activity following EC decay. Isotopic purity was assured by measuring α energies using surface-barrier detectors. The analog signals from our detectors were routed through an analog-to-digital converter (ADC) to a small computer which recorded the energy and sequential time of occurrence of each counting event for subsequent off-line analysis. Pulses from fission events were recorded in a channel at the high-energy end of the pulse-height spectrum.

We obtained a half-life of 59±13 min for ^{259}No from the measured time history of α events in the energy range 7.4 – 7.8 MeV, corresponding to ^{259}No α decay. This value is in good agreement with the half-life of 58±5 min obtained by Silva *et al.*[9] A background correction was made for the contribution from the parent-daughter pair 7.2-h ^{211}At/0.5-s ^{211}Po; the ^{211}At resulted from the ^{18}O bombardment of the Au recoil collection foil and was not completely removed by our chemical procedure. The ^{211}Po daughter decays primarily via a 7.45-MeV α transition.

There was no long-lived SF activity in these samples which would have indicated the presence of ^{248}Cm contamination. However, the recorded time history of the fission events in these samples indicated a SF activity that could not have originated directly from ^{259}No decay, since fitting a single-component decay curve to these counting data yielded half-lives which were more than twice as long as the value determined for ^{259}No. This was evidence that we were observing fissions from the decay of ^{259}Md, which would be the EC daughter of ^{259}No.

To confirm our tentative identification, we adsorbed pure ^{259}No on a column of cation-exchange resin and eluted trivalent species at measured intervals of 20 to 30 min with a solution of 0.5 *M* ammonium α-hydroxyisobutyrate at pH 5. Samples of the eluate were evaporated on Pt disks and the α energies analyzed. The ^{259}No, being a divalent ion, is complexed considerably less by the α-hydroxyisobutyrate solution than are trivalent ions. Thus, the ^{259}No remained essentially at the top of the column while the daughter atoms ^{255}Fm and ^{259}Md, produced by the α and EC decay of ^{259}No between elutions, were removed rapidly. A decay curve derived from summing the SF activity in the eluate fractions indicated a single component decaying with a 1.5- to 2-h half-life. The number of fission events observed in this experiment is consistent with that observed after complete decay of initially pure ^{259}No samples, suggesting that essentially all of the born SF activity is associated with a trivalent daughter, which we believe to be ^{259}Md. There were no α events in the energy range associated with the α decay of ^{259}No, thus none of the observed SF events were due to ^{259}No.

We calculated a weighted-average half-life of 103±12 min for ^{259}Md, based on four measurements. One of these was the milk experiment just described, and the other three were decay measurements of SF activity in initially pure ^{259}No samples. For these latter three measurements, we fit two-component decay curves to the SF activity data using a fixed half-life of 59 min for the ^{259}No parent. Although there are too few events counted in these experiments to indicate whether a SF-decay curve consists of one or two components, the chemical separations we performed assure us that each experiment was initiated with ^{259}No free of other actinide species. We know from the milk experiment that all of the SF activity originated from ^{259}Md. Any contribution to the SF activity from 20.1-h ^{255}Fm (100% α; 2.4×10^{-5}% SF), the α-decay daughter of ^{259}No, or from 39.8-d ^{255}Es (92% β^-; 8% α; 4.1×10^{-3}% SF), the α-decay daughter of ^{259}Md, was negligible due to the long SF half-lives of these isotopes. Electron-capture decay of ^{259}Md to give 1.5-s ^{259}Fm, a known SF emitter,[3] is expected to be either energetically impossible (see the Q_{EC} predictions for ^{259}Md in Table I) or of low abundance. The most positive estimate for the Q_{EC} of ^{259}Md, 0.33 MeV, results in a half-life of 1.5 d for a transition with $\log ft=6.0$, using the $\log f$ tables of Gove and Martin.[21] This corresponds to an upper limit of 5% for ^{259}Md EC decay. Therefore, we interpret the fission-counting data obtained from direct counting of initially pure ^{259}No samples as a parent-daughter two-component decay curve, and the decay data from the Md milk experiments as a one-component curve.

We observed no α events in the energy range 6.5–7.0 MeV in the Md-milk experiment. Predictions of Q_α for ^{259}Md, presented in Table I, range from 6.6 to 7.3 MeV. Mendelevium-259 presumably has a ground-state configuration of $\frac{7}{2}-[514]\downarrow$, and would likely α decay to a similar state in ^{255}Es at an excitation energy of about 0.4 MeV. With the subtraction of another 0.15 MeV for recoil and screening corrections from the Q_α estimates, it is apparent that α particles from ^{259}Md would have energies of 6.8 MeV or less. According to the α-decay systematics proposed by Viola and Seaborg,[18] such a transition would occur in less than 0.1% of the total decays. From our observations we can set an upper limit of about 3% for ^{259}Md α decay. Because EC decay and α decay could contribute at most only a few percent to the decay of ^{259}Md, we conclude that SF is the predominant decay mode.

The branching ratio for ^{259}No EC decay was determined from these experiments to be 25 ± 4%. An estimate of the SF-decay branch for ^{259}No was also obtained, although too few SF counts in the growth and decay curves render it qualitative at best; a value of 1 ± 9% was calculated for this branching ratio.

III. INITIAL FISSION COINCIDENCE STUDIES

Following the discovery of ^{259}Md, we performed experiments to determine the kinetic energies of fragment pairs from its SF decay. We prepared two coincidence counting systems, each consisting of two 450-mm^2 surface-barrier detectors mounted facing one another inside a vacuum chamber. Samples of ^{259}No, chemically purified as described in the preceding section, were evaporated on thin polyvinyl-acetate-chloride copolymer (VYNS) films (typically 25 to 35 μg/cm^2) and placed between the detectors. Fission fragments from the decay of the daughter ^{259}Md were detected in coincidence in opposing detectors, and the kinetic energy of each fragment and the sequential time of occurrence of each fission event were measured. As before, the digitized signals were processed by a small computer which recorded the fragment energies and event times on magnetic tape. The mass of each fragment in a coincident event was deduced from kinematic considerations.

We used a source of ^{252}Cf mounted on VYNS film to calibrate the fragment-energy response of the detectors, and employed the mass-dependent calibration procedure of Schmitt, Kiker, and Williams[22] to correct the detector responses for the pulse-height defect. The ^{252}Cf SF calibration source was prepared in the same manner as our ^{259}Md SF sources, and the thickness appeared to be similar.

We derived fragment mass and kinetic-energy distributions from the detection of 397 coincident SF events obtained in 18 separate bombardments. We only note the main characteristics at this time, because we later obtained mass and kinetic energy distributions for ^{259}Md SF using thinner sources because of improved chemical separation procedures. The provisional mass distribution (no fragment neutron-emission correction) that we obtained from these initial fission studies was quite symmetric, with small wings indicative of a small asymmetric component. In this respect, ^{259}Md is quite like ^{258}Fm and ^{259}Fm in undergoing principally symmetric mass division. However, the provisional TKE distribution from ^{259}Md SF in these studies was considerably different from those of ^{258}Fm and ^{259}Fm; the most probable TKE was about 190 MeV and the distribution was quite broad, with a FWHM of about 106 MeV. These earlier results are described fully in Ref. 23 and the newer results are reported in Sec. V of this paper.

IV. LIGHT-PARTICLE EMISSION STUDIES

Unlike that of ^{258}Fm and ^{259}Fm, the low TKE measured for ^{259}Md suggested that the fragments were considerably deformed at scission. Since, how-

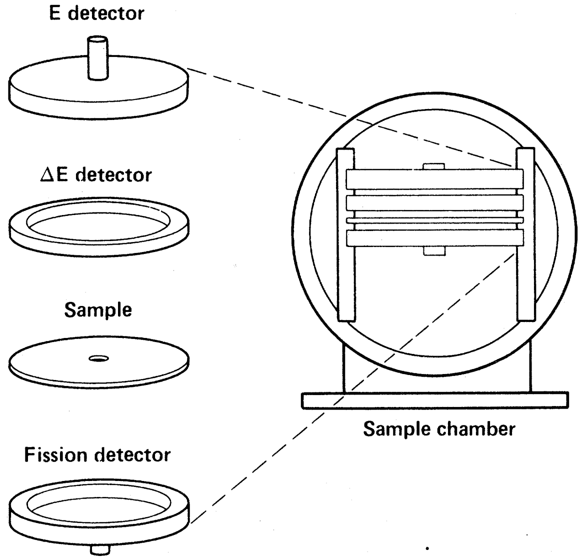

FIG. 1. An exploded, schematic diagram of the counter telescope used to search for light-particle emission by ^{259}Md.

ever, calculations based on Strutinsky shell corrections from the two-center shell model[24] do not show a stable minimum in that region of the potential energy surface favoring highly-deformed mass-symmetric fragments (see the Discussion), we decided to test the hypothesis of light-particle emission at scission. If a light, preferably hydrogen-like particle (*p*, *d*, or *t*) were emitted at scission, it could remove 25 MeV or more of the energy available (*Q* value) in the fission process. Furthermore, the removal of a $Z=1$ particle during symmetric mass division occurring in about half of the SF events could account for most or all of the 30 MeV energy deficit in the vicinity of mass symmetry.

We assembled two similar counter telescopes, each consisting of a ΔE and E detector placed above the sample, and a 450 mm^2 Si(Au) surface-barrier detector below it (see Fig. 1). A coincidence ($2\tau=470$ ns) was required between the ΔE and E detectors before the energies deposited in each detector were recorded; the ΔE detector measured energies between 0.7 and 15 MeV, while the E detector measured energies between 0.5 and 30 MeV. The energies obtained from a ΔE-E coincident event were required to fall within certain limits based on known range-energy relationships of light particles in Si. Fission-fragment kinetic energies were obtained during these experiments by analyzing coincident fission pulses from the detector below the sample and the ΔE detector above it.

We calibrated our detector systems with sources of ^{252}Cf evaporated from aqueous solution onto VYNS films as before. To determine the detection efficiency of light particles from ^{252}Cf SF for our two counter telescopes, we fit the long-range α energy distribution accumulated for each system to a Gaussian curve with the mean energy and the distribution width fixed at values determined by Cosper, Cerny, and Gatti.[25] We then corrected the area under this curve to account for the non-Gaussian α-energy distribution at low energies observed by Loveland.[26] On this basis, and assuming a value for ^{252}Cf SF of 3.30 ± 0.20 long-range α's per 10^3 SF derived from Cosper *et al.*,[25] Loveland,[26] and Raisbeck and Thomas,[27] we obtained effective geometric efficiencies of 10.6% and 14.0% for our two systems. Owing to the thicknesses of our ΔE detectors, which were more favorable for the detection of $Z=1$ particles than α's, the minimum α energies observable in the two systems were 11 and 15 MeV. Additionally, since the geometries were high for these systems, there was a significant chance of not detecting light particles because one of the fission fragments would strike the ΔE detector at the time that a light particle from the same event passed through it, thus masking the critical light-particle ΔE energy. However, since the probability was the same for ^{259}Md light particles as for ^{252}Cf light particles, it was canceled out. The number of tritons

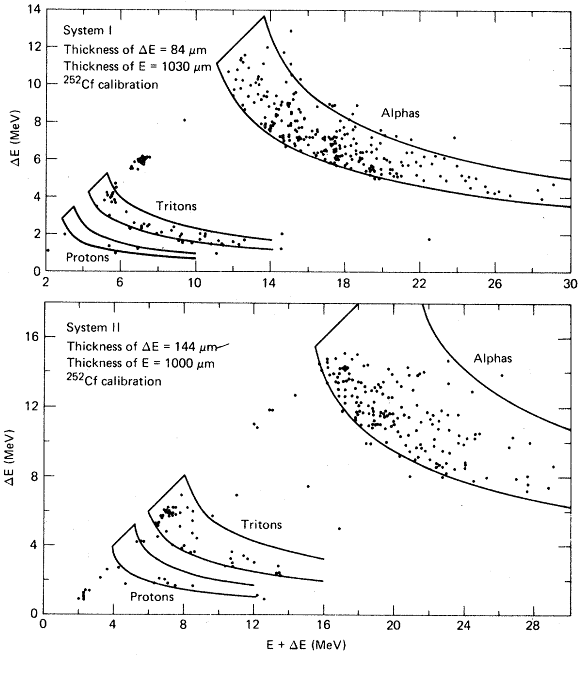

FIG. 2. Energy deposited in the ΔE detector versus incident particle energy for the ^{252}Cf calibration of the two counter telescopes used to measure light-particle emission by ^{259}Md.

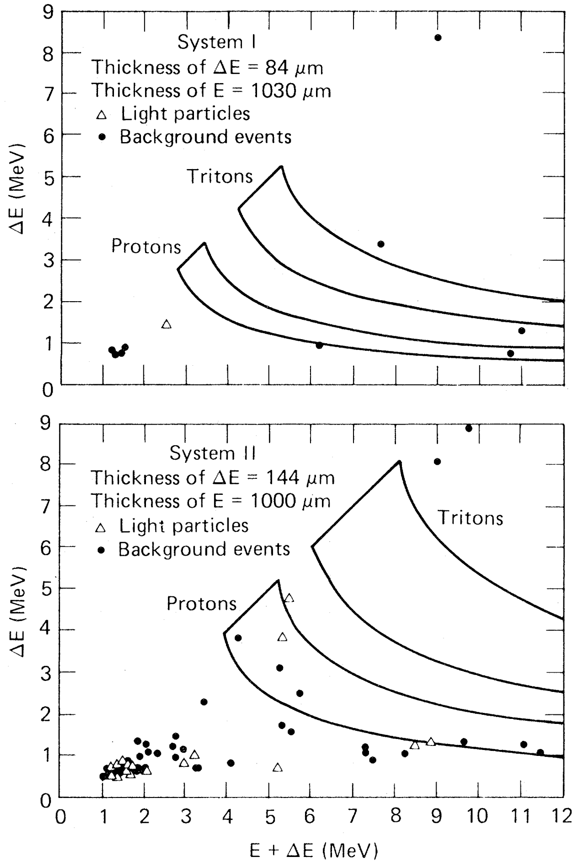

FIG. 3. Energy deposited in the ΔE detector versus incident particle energy for the light particle events detected from eleven samples of ^{259}Md and also the events obtained during over 17000 min of background counting.

observed during the ^{252}Cf calibrations agreed well with literature values for the relative abundances of α particles and tritons.[25,27] The geometric efficiency of each system for detecting singles fission events in the detector below the sample was about 65%.

The ^{252}Cf calibrations for each of the counter telescopes are shown in Fig. 2 as plots of energy deposited in the ΔE detector versus total particle energy. The envelopes shown for each type of particle represent the range of energies which would be deposited in the ΔE detector as a function of the incident particle energy and the angle at which the particle entered the detector. The ratio between the number of fission-originated α particles (long-range α's) and the number of fissions recorded by the detectors below the samples provided a calibration with which to compare our ^{259}Md results.

Samples of ^{259}Md on VYNS film from 11 bombardments were counted for a total of over 4800 min in the two counting systems; in addition, background counts were taken in each system for over 17000 min. The events detected in both the ^{259}Md light-particle counts and the background counts are shown in Fig. 3 as plots of ΔE versus total particle energy for each system. Unfortunately, we had contaminated the ΔE detector in system II with a small amount of ^{252}Cf during our calibration procedure. This contamination amounted to about one fission per 8–9 min. In order to reduce the effect of this contamination, we turned the ΔE detector over so that the contaminated side faced the E detector. In this way, no light particles from the Cf could pass through both the ΔE and E detectors simultaneously. However, coincidences were generated between the detectors by such species as degraded fission fragments, fission-product betas and gamma rays, and 6.1-MeV α particles from ^{252}Cf decay; these are the source of the relatively high sample and background counting rates seen in the bottom plot in Fig. 3.

We observed a net total of 2 ± 2 light particles during these runs, all of them protons, and a total of 443 fissions as recorded by the detectors below the samples. After corrections for geometrical efficiency based on our ^{252}Cf calibrations, these values correspond to an emission rate of 1 ± 4 light particles per 100 ^{259}Md SF decays.

V. FISSION PROPERTIES OF ^{259}Md

We also redetermined the fission properties of ^{259}Md by measuring the kinetic energies of coin-

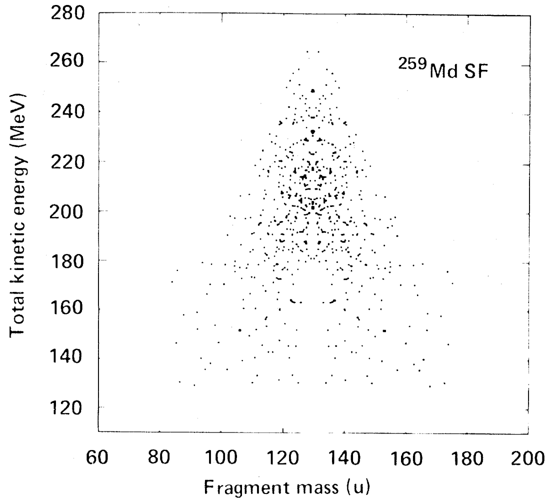

FIG. 4. Mass-TKE distribution for the SF of ^{259}Md. Each coincident fission event provides two points which are reflected in the mass plane.

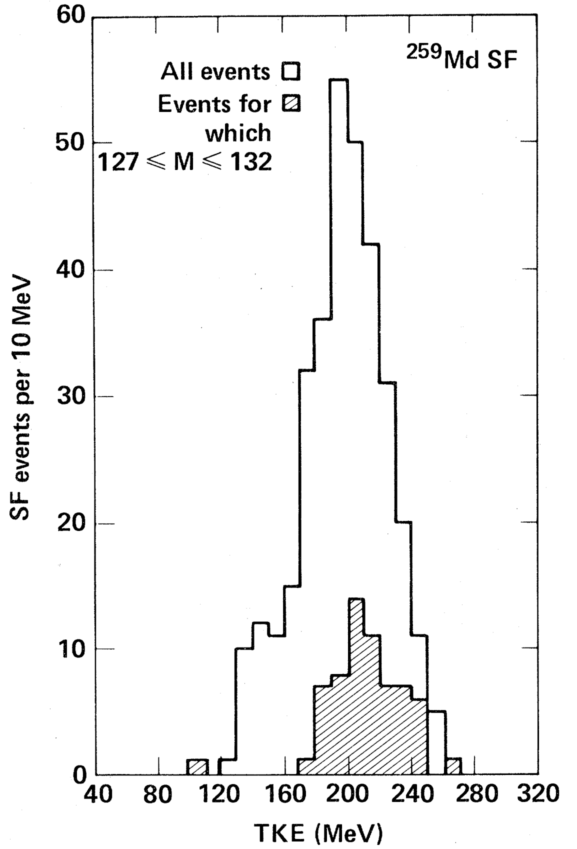

FIG. 5. Provisional TKE distribution for the SF of ^{259}Md; the hatched area is the distribution of TKE from those events with both fragments having masses between 127 and 132 u.

cident fission fragments concurrently with the acquistion of the light-particle data, as described in Sec. IV. Our initial fission-coincidence studies, described in Sec. II of this article, suffered by comparison from relatively thicker sources of ^{259}Md. This thickness resulted from mass contamination by such common cations as Ca^{++}, Mg^{++}, Na^{+}, etc., which were not separated from No^{++} on the extraction-chromatographic column we used to purify the No from other bombardment products. In order to reduce the quantity of mass contamination in our samples, we performed a gradient elution separation of the No from the contaminating monovalent and divalent ions with dilute HNO_3.

We measured fragment energies for 333 coincident fission events; these are shown in Fig. 4 as a reflected plot of TKE versus fragment mass for each event. It can be seen that there is a high concentration of events at mass symmetry, as there was in our initial studies, and a smaller contribution from asymmetric fission at lower TKE's. The provisional TKE distribution (no fragment neutron-emission correction), shown in Fig. 5, is considerably narrower than that obtained from our first studies, but the most probable TKE is essentially the same. The most probable TKE for the SF of ^{259}Md, obtained by fitting a Gaussian curve to the distribution, is 200.7±1.4 MeV, while the FWHM of the distribution is 60.6±3.4 MeV, considerably broader than the values for ^{252}Cf (40.6 MeV) and ^{256}Fm (41.4 MeV) that we measured in the same counting systems using sources evaporated from solution onto VYNS films.

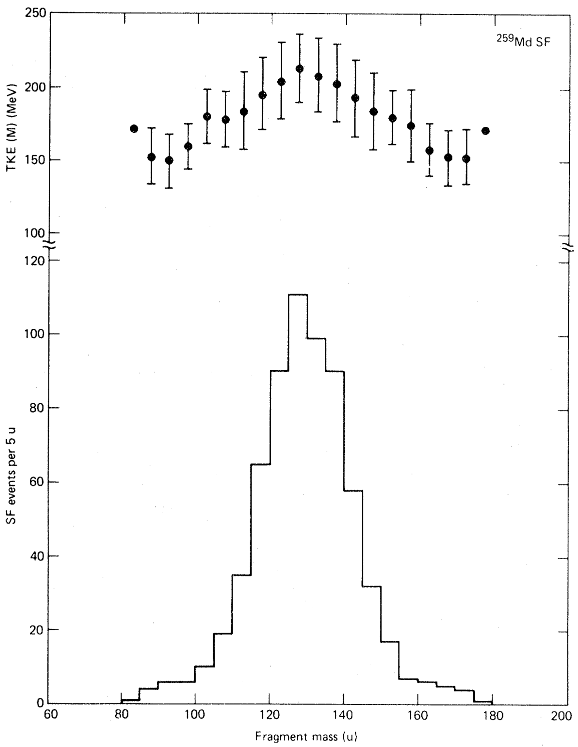

FIG. 6. Provisional mass distribution from the SF of ^{259}Md (bottom) and the average TKE (top) associated with each mass increment shown in the histogram below.

Also shown in Fig. 5 is the TKE distribution of those fission events with both fragment masses between 127 and 132 u, i.e., at mass symmetry. These events, about 19% of the total, have an average TKE of 215 MeV, still about 30 MeV below what would be expected for similar events from the SF of ^{258}Fm and ^{259}Fm. This distribution is also quite broad (FWHM ~50 MeV), with one quarter of the events having a TKE less than 200 MeV.

The provisional mass distribution is shown in Fig. 6 along with a plot of the average TKE for each 5-u mass bin. The mass distribution is decidedly symmetric, with a FWHM of 27.8±0.6 u. Although this is more than twice as broad as the mass

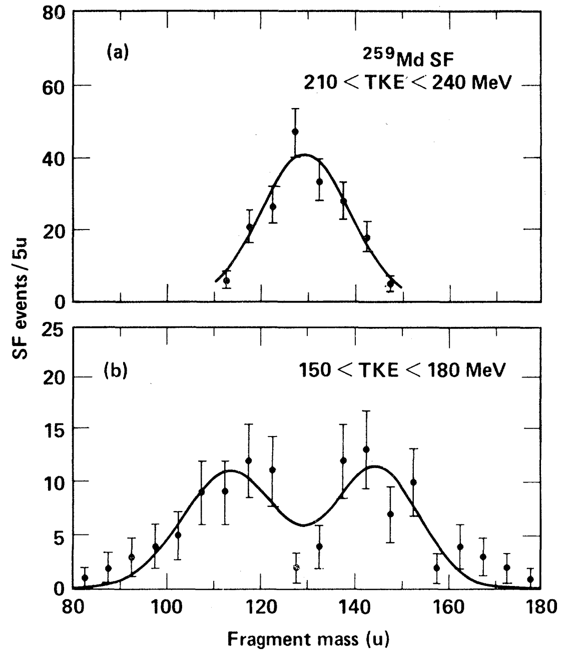

FIG. 7. Provisional mass distribution for the SF of ^{259}Md (this work); (a) events with TKE between 210 and 240 MeV; (b) events with TKE between 150 and 180 MeV.

distributions for ^{258}Fm (FWHM ~8 u) (Ref. 2) and ^{259}Fm (FWHM ~11 u),[3] it is narrower by about one-third than the provisional mass distribution obtained by Balagna *et al.*[28] for the SF of ^{257}Fm.

The SF events from ^{259}Md with lower TKE values clearly form an asymmetric mass distribution. In Fig. 7 we show plots of the mass distribution for events with TKE between 210 and 240 MeV and for events with TKE between 150 and 180 MeV. For comparison, we show similar plots in Figs. 8 and 9 for ^{257}Fm (Ref. 28) and ^{256}Fm (unpublished data). The solid curves are Gaussian fits to the mass distributions. The lower-TKE mass distribution for ^{259}Md shown in Fig. 7(b) is no longer as symmetric as it was in our initial fission studies; this was apparently due to unequal energy losses by coincident fragments caused by the thicker ^{259}Md samples.

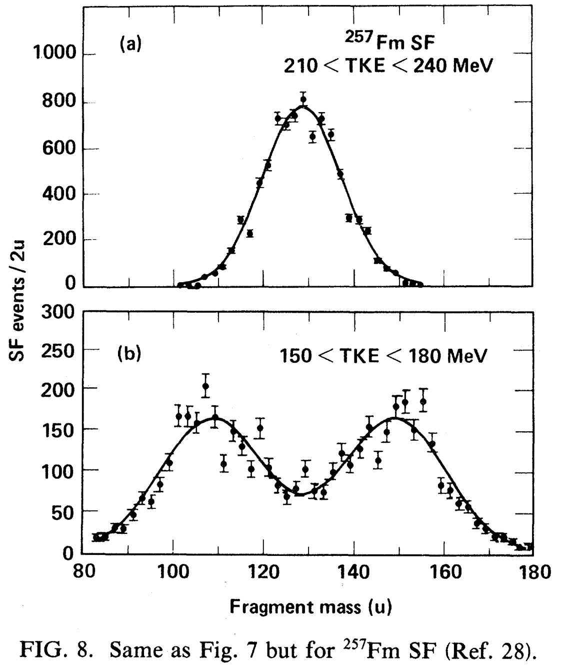

FIG. 8. Same as Fig. 7 but for ^{257}Fm SF (Ref. 28).

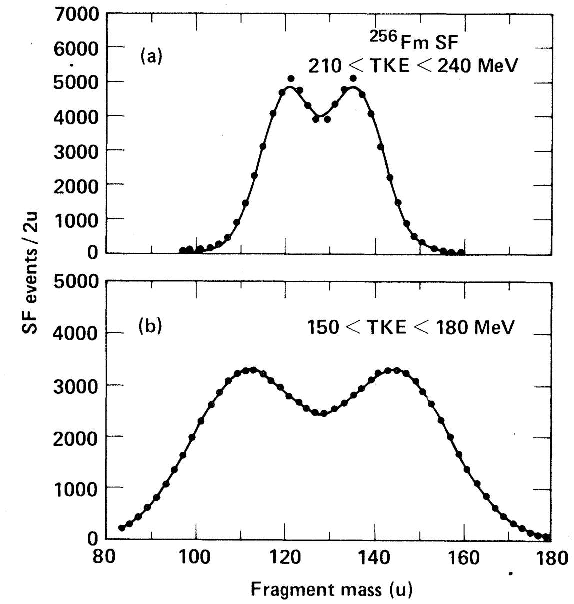

FIG. 9. Same as Fig. 7 but for ^{256}Fm SF (unpublished data).

Our study of the SF properties of ^{259}Md was not complicated by any background from the SF of ^{256}Fm, as were those of ^{258}Fm and ^{259}Fm, due to our chemical purification of the ^{259}No parent. Since there were fewer than 500 gross fission events in either of the Fm experiments, the subtraction of this sizable background (45% to 65% of the total events) resulted in considerable distortion of the TKE and mass distributions. The total number of fission events in our study is not large, either, but we are confident that they have all arisen from ^{259}Md, resulting in greater reliability in our determination of the mass and TKE distributions.

VI. DISCUSSION

The important questions posed by the experimental results we have presented here are the following:

TABLE II. Calculated fragment excitation energies for SF-emitting actinide nuclides.

Nuclide	Q (MeV)	$\overline{\text{TKE}}$ (MeV)[a]	Q-$\overline{\text{TKE}}$ (MeV)	Reference[b]
^{240}Pu	199.8	177.0	22.8	31, 41
^{242}Pu	197.9	178.5[c]	19.4	31, 42
^{242}Cm	210.8	183[d]	27.8	39, 40
^{244}Cm	210.1	183.7	26.4	43
^{246}Cm	210.4	183.9	26.5	39
^{248}Cm	209.1	182.2	26.9	39
^{250}Cm	208.7	179.8	28.9	44
^{250}Cf	220.5	187.0	33.5	39
^{252}Cf	218.9	185.9	33.0	39
^{254}Cf	220.3	186.9	33.4	39
^{254}Fm	233.9	195.1	38.8	45
^{256}Fm	235.3	197.9	37.4	39
^{257}Fm	237.2	199.0[e]	38.2	28, 33
^{258}Fm	249.3	238[f]	11.3	2
^{259}Fm	249.6	242.4[f]	7.2	3
^{259}Md	246.6	202.9[g]	43.7	this work
^{252}No	247.3	202.4	44.9	7

[a] Average preneutron TKE except where noted to be provisional.

[b] Refers to $\overline{\text{TKE}}$ values only.

[c] $\overline{\text{TKE}}$ obtained by reducing $\overline{\text{TKE}}$ for ^{241}Pu(n,f) from Ref. 42 by a factor of $\overline{\text{TKE}}$ (^{240}Pu SF)/$\overline{\text{TKE}}$ (^{239}Pu[n,f]) from Ref. 31.

[d] Average value between $\overline{\text{TKE}}$ vs $Z^2/A^{1/3}$ systematics of Unik *et al.* (Ref. 39) and Viola (Ref. 40).

[e] Data from Ref. 28 using different neutron-emission correction derived from the data in Ref. 33.

[f] Provisional values.

[g] Most probable TKE using kinetic energies corrected for neutron emission with the $\bar{\nu}(M)$ function scaled to $\bar{\nu}_T = 4.15$.

Why is there a deficit of 30 MeV in the TKE of fragments at mass symmetry in the SF of ^{259}Md, and what has happened to the remaining energy? We show in Table II calculations of the energy available from fission after the Coulomb energy, i.e., the TKE, has been subtracted, for a number of SF-emitting actinide nuclides. These energies were obtained by subtracting the measured preneutron average TKE value for each nuclide from a preneutron mass distribution-weighted fission Q value. The Q values were calculated from the Comay-Kelson ensemble-averaged mass-excess values,[29] with the relative abundance of each mass split determined from Gaussian-curve parameters used to fit the experimentally-determined mass distribution. Fragment atomic numbers were calculated from the prescription of Nethaway.[30] In the cases of ^{240}Pu SF (Ref. 31) and ^{252}Cf SF (Ref. 32), for which the total fission energy balance has been determined, the experimentally determined residual energy values are in good agreement with the values in Table II. This energy at scission is divided between internal excitations (vibrational, rotational, and single-particle motions) and deformation energy. All forms are ultimately released with the emission of prompt neutrons and gamma rays from the fragments and, to a very minor extent, light charged particles emitted at scission.

The excitation energies in Table II increase monotonically with Z, expect for the nuclides ^{258}Fm and ^{259}Fm. These nuclides have predictably low excitation energies due to the proximity of the fragments to the doubly magic ^{132}Sn configuration. We would expect ^{259}Md to exhibit a similarly low excitation energy, which is not the case. This means that ^{259}Md SF has available some 35 MeV more of excitation energy at scission to dissipate than do ^{258}Fm and ^{259}Fm. For SF events with both fragment masses between 127 and 132 u (for which the fission Q value is about 256 MeV), the available energy is 41 MeV, which must be principally in the form of internal energy and deformation energy.

The proximity of ^{259}Md to ^{258}Fm and ^{259}Fm (only one proton or one neutron and proton different)

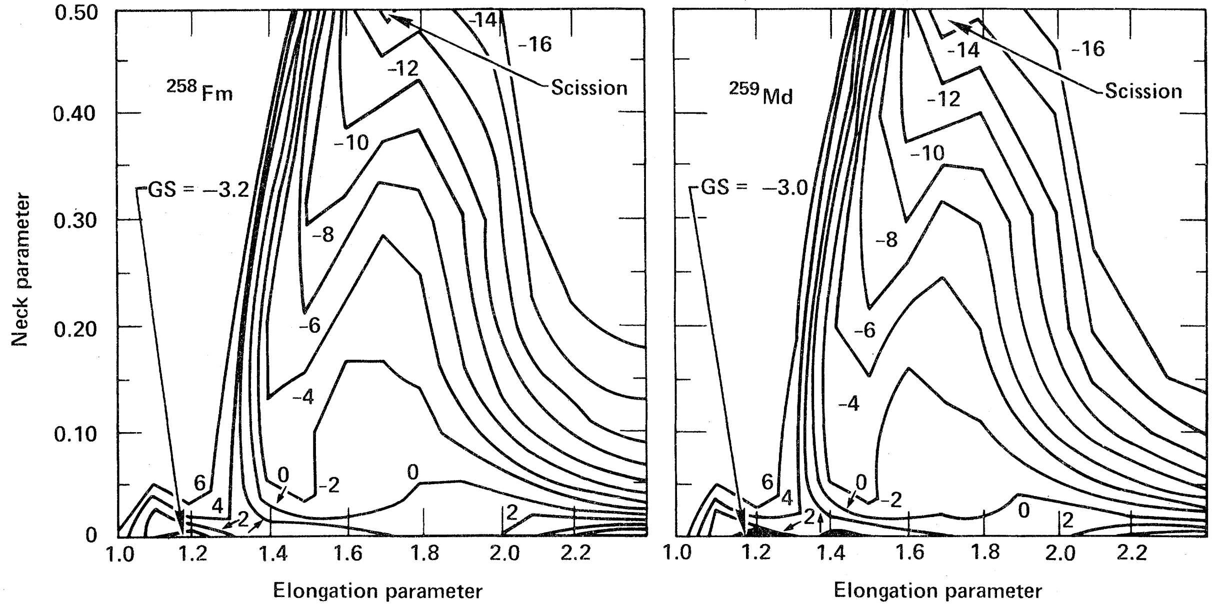

FIG. 10. Potential energy surfaces of ^{258}Fm and ^{259}Md calculated as a function of an elongation and a neck parameter (Refs. 24 and 46). The numbers labeling the contour lines are energies in MeV.

strongly suggests that all three nuclides should have similar properties. This is in accordance with the two-center shell model in which fragment shell structure is the factor determining mass and TKE distributions from fission.

Calculations of the potential-energy surfaces for ^{258}Fm and ^{259}Md have been carried out by using the two-center plus Strutinsky model including odd-particle effects. These calculations, shown in Fig. 10, do not show any measurable difference between these two nuclei, and certainly no minimum potential energy path can be found where one or both fragments are highly deformed. The fragment shell effect dominates the potential-energy surface leading to a narrow path to scission with nearly spherical, symmetric fragments. The mass distribution for ^{259}Md SF is certainly similar to that for ^{258}Fm and ^{259}Fm, being symmetric although broader by more than a factor of 2. However, this additional breadth should not be a factor in the 30 MeV TKE deficit at symmetry for ^{259}Md relative to ^{258}Fm and ^{259}Fm.

Four possible explanations for this TKE deficit are the following: (a) the mass distribution for ^{259}Md SF could become asymmetric after correcting the fragment kinetic energies for neutron emission, and therefore the fission properties of ^{259}Md are not similar to those of ^{258}Fm and ^{259}Fm; (b) there is a significant light charged-particle emission at scission, which removes Coulomb potential energy from the fissioning system without perturbing the mass distribution much; (c) there is a large amount of available energy distributed into internal excitation at scission; and (d) there is a large amount of potential energy in deformation at scission. We explored each of these possibilities as thoroughly as was practical to try to account for this TKE deficit.

We had not to this point corrected the fragment kinetic energies from ^{259}Md SF for the effects of neutron emission because there is no experimental information available for this purpose. When applied to the SF of ^{257}Fm,[28] this correction caused a noticeable valley to appear in the mass distribution at symmetry. To examine this possibility, we constructed a sawtooth $\bar{\nu}(M)$ function from the ^{257}Fm neutron multiplicity distribution of Hoffmann *et al.*[33] and normalized the function to a value for $\bar{\nu}_T$ (total average neutron emission per fission) of 4.15, which is the measured value for ^{252}No,[6] and the highest yet observed in SF. The mass distribution obtained after applying this neutron-emission correction to ^{259}Md is shown in Fig. 11. This distribution is somewhat less sharply peaked than the provisional one shown in Fig. 6, but there is no signficiant dip at mass symmetry as there is in the case of ^{257}Fm SF. The application of this neutron-emission correction to the ^{259}Md fragment kinetic energies broadened the mass distribution by about 4 u and increased the most probable TKE by 2.2 MeV. We cannot, therefore, explain the TKE deficit by saying that the true mass distribution for ^{259}Md SF is asymmetric and that its fission proper-

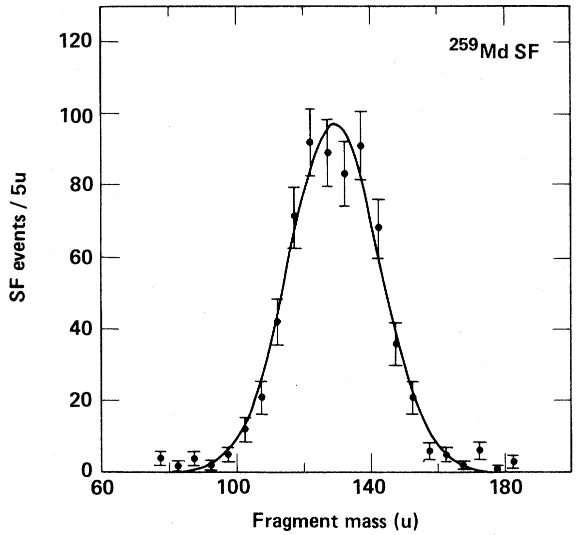

FIG. 11. Preneutron emission mass distribution for ^{259}Md SF. The fragment neutron-emission correction was assumed to be that of ^{257}Fm, but scaled to $\bar{\nu}_T = 4.15$.

ties are completely dissimilar to those of ^{258}Fm and ^{259}Fm.

Another possibility for reducing the Coulomb potential energy at scission, and thus the TKE, is the occurrence of three-body fragmentation. Three-body fragmentation in fission is well known and has been the subject of much experimental and theoretical study in the past two decades (see, for example, Refs. 34 and 35). Three-body fragmentation usually occurs at the rate of a few events per thousand binary fissions; the third particle emitted is typically much smaller than either of the other two fragments, and the process is therefore often referred to as light charged-particle (LCP) fission.

Angular distributions of these light particles with respect to the fission axis indicate that they are emitted before the heavier fragments are able to attain any significant velocity, i.e., at or before scission. Alpha particles account for about 90% of these light particles followed in abundance by tritons, ^{6}He, and protons. Nuclei as large as oxygen have been observed in LCP fission.[36] The emission of light particles appears to cost the fissioning system at least 25 MeV in potential energy,[35] composed of the loss in binding energy of the light particle, the initial kinetic energy of the light particle, and the disturbance of the Coulomb energy repelling the two major fragments caused by the light particle between them.

The SF of ^{259}Md might represent a special case of LCP fission in which the rate of emission of $Z=1$ particles is greatly enhanced over the fraction of a percent observed for the SF of other nuclides.[37] If the path from saddle to scission in the SF of ^{259}Md is sufficiently slow to permit the increasing influence of nucleon shells in the developing fragments (the adiabatic approximation), the preformation and subsequent emission of a $Z=1$ particle might result in a path of minimum potential energy by which the residual fissile nucleus, now a heavy Fm isotope, could fission symmetrically into two $Z=50$ fragments. Owing to the Coulomb potential energy removed by this charged particle, the fragment kinetic energies would be lower than those observed for the binary SF of ^{258}Fm and ^{259}Fm.

However, the LCP emission rate that we measured, 1 ± 4 light particles per 100 ^{259}Md SF decays, was significantly below the frequency of $\sim 50\%$ required to explain the magnitude of the TKE deficit in the symmetric fission of ^{259}Md. Our results provide no evidence that the LCP emission rate is significantly different from that which would be extrapolated from LCP emission rates already measured for some actinide SF emitters.[37] If the LCP emission rate for ^{259}Md SF were enhanced to 50%, we would have observed about 40 light particles.

The two final explanations, the presence of a large amount of internal excitation energy and the presence of a large amount of fragment deformation energy, can be addressed together, since these two degrees of freedom are complementary in the fission process at the point of scission.

Schultheis and Schultheis have calculated limits for energy dissipated by the SF of ^{252}Cf,[38] based on experimental measurements of energies of fission fragments, prompt neutrons and gamma rays, and calculated static fragment potential energies. These calculations show that, for symmetric mass division and under the assumption of reasonable fragment shapes, only about 20%, or 7 MeV, of the available excitation energy (33 MeV) is present as internal energy; the remainder is presumably deformation energy which is converted to internal energy as the fragments move apart from scission. Furthermore, Schultheis and Schultheis calculate maximum fragment deformations for ^{252}Cf SF, which form a sawtooth-shaped curve with fragment mass, showing the expected trend of low deformation (high sphericity) for fragments with $A = 130-132$. It is reasonable to consider these shapes valid for fragments from the SF of ^{259}Md; thus a distribution of available energy at scission for ^{259}Md weighted heavily in favor of fragment deformation energy is implausible, because a larger portion of the mass division yields fragments near the region of minimum distortion for ^{259}Md than for ^{252}Cf. This means that the "average" total deformation of the fragments from ^{259}Md SF at scission should be less than that of the fragments from ^{252}Cf SF. This argument suggests, then, that a significantly larger portion of the excitation energy at scission available from the SF of ^{259}Md (41 MeV at symmetry; 44 MeV overall) is present as internal excitation energy.

There is one important factor, however, which speaks against a low fragment deformation energy for ^{259}Md SF, and that is that the measured TKE is only about 200 MeV. That this TKE is about 40 MeV lower than those of ^{258}Fm and ^{259}Fm indicates clearly that the scission configuration for ^{259}Md cannot be as compact as those of ^{258}Fm and ^{259}Fm; the charge centers of the fragments at scission must be farther apart. This means that the fragments from ^{259}Md SF must be more deformed than those fragments from the SF of ^{258}Fm and ^{259}Fm, even though the fissioning mass and the mass division are essentially the same. There is no readily apparent reason within the framework of current fission theory why the fragments from ^{259}Md at scission should be considerably more deformed than those from ^{258}Fm and ^{259}Fm.

A resolution of the form of this excitation energy for ^{259}Md might be obtained by performing experiments to measure the neutron-emission angular distribution for ^{259}Md SF. If there is a large amount of internal excitation energy at scission, it would likely be dissipated through the emission of neutrons before the fragments have attained any significant velocity. These neutrons would be emitted isotropically in the laboratory frame of reference with respect to the fission axis. If, however, the energy were mostly deformation energy, it would not be dissipated through neutron emission until the fragments had achieved essentially their final velocities and, in the laboratory frame of reference, these neutrons would be strongly focused in the direction of the fission axis. Considering the small number of atoms that can be produced, this is an exceedingly difficult experiment.

In sum, the SF decay of ^{259}Md is principally symmetric, as is that of ^{258}Fm and ^{259}Fm, but the average TKE is about 40 MeV lower, about what one would expect from the systematics of $\overline{\text{TKE}}$ vs $Z^2/A^{1/3}$.[39,40] The deficit in TKE at mass symmetry compared with that expected based on the fission properties of ^{258}Fm and ^{259}Fm is not caused by significant light charged-particle emission at scission, and is therefore most likely in the form of unusually large deformation or excitation energy.

It is possible, then, that the SF properties of ^{259}Md represent a transition from the highly symmetric, high-TKE fission of ^{258}Fm and ^{259}Fm back to asymmetric, low-TKE fission as Z increases beyond 100. The observed SF properties of ^{259}Md are not consistent with predictions based on fragment shell effects within the Strutinsky method. Finding a consistent fission model which is able to explain the SF properties of all three nuclei, ^{258}Fm, ^{259}Fm, and ^{259}Md, is at this time an open challenge.

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[1a] E. K. Hulet, J. F. Wild, R. W. Lougheed, J. E. Evans, B. J. Qualheim, M. Nurmia, and A. Ghiorso, Phys. Rev. Lett. 26, 523 (1971).

[1b] 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] Yu. Ts. Oganessian, A. G. Demin, A. S. Iljinov, S. P. Tretyakova, A. A. Pleve, Yu. E. Penionzhkevich, M. P. Ivanov, and Yu. P. Tretyakov, Nucl. Phys. A239, 157 (1975).

[5] G. M. Ter-Akopyan, A. S. Iljinov, Yu. Ts. Oganessian, O. A. Orlova, G. S. Popeko, S. P. Tretyakova, V. I. Chepigin, B. V. Shilov, and G. N. Flerov, Nucl. Phys. A255, 509 (1975).

[6] Yu. A. Lazarev, O. K. Nefediev, Yu. Ts. Oganessian, and M. Dakowski, Phys. Lett. 52B, 321 (1974).

[7] C. E. Bemis, R. L. Ferguson, F. Plasil, R. J. Silva, F. Pleasonton, and R. L. Hahn, Phys. Rev. C 15, 705 (1977).

[8] C. E. Bemis, R. L. Ferguson, F. Plasil, R. J. Silva, G. D. O'Kelley, M. L. Kiefer, E. K. Hulet, and R. W. Lougheed, Phys. Rev. Lett. 39, 1246 (1977).

[9] R. J. Silva, P. F. Dittner, M. L. Mallory, O. L. Keller, K. Eskola, P. Eskola, M. Nurmia, and A. Ghiorso, Nucl. Phys. A216, 97 (1973).

[10] W. D. Myers and W. J. Swiatecki, Nucl. Phys. 81, 1 (1966).

[11] B. M. Foreman, Jr., and G. T. Seaborg, J. Inorg. Nucl. Chem. 7, 305 (1958).

[12] V. E. Viola and G. T. Seaborg, J. Inorg. Nucl. Chem. 28, 697 (1966).

[13] P. A. Seeger and R. C. Perisho, Los Alamos National Laboratory Report LA-3751, 1967.

[14] P. A. Seeger, *American Institute of Physics Handbook*, edited by D. E. Gray (McGraw-Hill, New York, 1972), Sec. 8 , pp. 136 and 137.

[15] V. E. Viola, J. A. Swant, and J. Graber, At. Data Nucl. Data Tables 13, 35 (1974).

[16] N. N. Kolesnikov and A. G. Demin, Joint Institute for Nuclear Research Report P6-9420, 1976.

[17] W. D. Myers, At. Data Nucl. Data Tables 17, 591 (1976); H. V. Groote, E. R. Hilf, and K. Takahashi, *ibid.* 17, 591 (1976); P. A. Seeger and W. M. Howard, *ibid.* 17, 591 (1976); S. Liran and Zeldes, *ibid.* 591 (1976).

[18] V. E. Viola and G. T. Seaborg, J. Inorg. Nucl. Chem. 28, 741 (1966).

[19] M. G. Mustafa and R. L. Ferguson, Phys. Rev. C 18, 301 (1978).

[20] R. W. Lougheed and E. K. Hulet, Nucl. Instrum. Methods 166, 329 (1979).

[21] N. B. Gove and M. J. Martin, At. Data Nucl. Data Tables 10, 262 (1971).

[22] H. W. Schmitt, W. E. Kiker, and C. W. Williams, Phys. Rev. 137, B837 (1965).

[23] E. K. Hulet, J. F. Wild, R. W. Lougheed, P. A. Baisden, J. H. Landrum, R. J. Dougan, M. Mustafa, A. Ghiorso, and J. M. Nitschke, *Proceedings of an International Symposium on the Physics and Chemistry of Fission, Jülich, 1979* (IAEA, Vienna, 1980), Vol. II, p. 299.

[24] M. G. Mustafa, Bull. Am. Phys. Soc. 24, 848 (1979).

[25] S. W. Cosper, J. Cerny, and R. C. Gatti, Phys. Rev. 154, 1193 (1967).

[26] W. Loveland, Phys. Rev. C 9, 395 (1974).

[27] G. M. Raisbeck and T. D. Thomas, Phys. Rev. 172, 1272 (1968).

[28] J. P. Balagna, G. P. Ford, D. C. Hoffman, and J. D. Knight, Phys. Rev. Lett. 26, 145 (1971).

[29] E. Comay and I. Kelson, At. Data Nucl. Data Tables 17, 463 (1976).

[30] D. R. Nethaway, University of California Radiation Laboratory Report UCRL-51640, 1974.

[31] A. J. Deruytter and G. Wegener-Penning, *Proceedings of the International Symposium on the Physics and Chemistry of Fission, Rochester, 1973* (IAEA, Vienna, 1974), Vol. II, p. 51.

[32] H. Nifenecker, C. Signarbieux, R. Babinet, and J. Poitou, *Proceedings of the International Symposium on the Physics and Chemistry of Fission, Rochester, 1973* (IAEA, Vienna, 1974), Vol. II, p. 117.

[33] D. C. Hoffman, G. P. Ford, J. P. Balagna, and L. R. Veeser, Phys. Rev. C 21, 637 (1980).

[34] I. Halpern, Annu. Rev. Nucl. Sci. 21, 245 (1971).

[35] R. Vandenbosch and J. R. Huizenga, *Nuclear Fission* (Academic, New York, 1973), pp. 374—400.

[36] J. B. Natowitz, A. Khodai-Joopari, J. M. Alexander, and T. D. Thomas, Phys. Rev. 169, 993 (1968).

[37] R. A. Nobles, Phys. Rev. 126, 1508 (1962).

[38] H. Schultheis and R. Schultheis, Phys. Rev. C 18, 317 (1978).

[39] J. P. Unik, J. E. Gindler, L. E. Glendenin, K. F. Flynn, A. Gorski, and R. K. Sjoblom, *Proceedings of the International Symposium on the Physics and Chemistry of Fission, Rochester, 1973* (IAEA, Vienna, 1974), Vol. II, p. 19.

[40] V. Viola, Nucl. Data 1, 391 (1966).

[41] R. L. Walsh, J. W. Boldeman, and M. E. Elcombe, *Proceedings of an International Symposium on the Physics and Chemistry of Fission, Jülich,* 1979 (IAEA, Vienna, 1980), Vol. II, p. 129.

[42] J. N. Neiler, F. J. Walter, and H. W. Schmitt, Phys. Rev. 149, 894 (1966).

[43] Yu. A. Barashkov, Yu. A. Vasil'ev, A. N. Maslov, E. S. Pavlovskii, M. K. Sareava, L. V. Sidorov, V. M. Surin, and P. V. Toropov, Yad. Fiz. 13, 1162 (1971) [Sov. J. Nucl. Phys. 13, 668 (1971)].

[44] D. C. Hoffman, G. P. Ford, and J. P. Balagna, Phys. Rev. C 7, 276 (1973).

[45] J. E. Gindler, K. F. Flynn, L. E. Glendenin, and R. K. Sjoblom, Phys. Rev. C 16, 1483 (1977).

[46] U. Mosel and H. W. Schmitt, Nucl. Phys. A165, 73 (1971).