Symmetric fission in the neutron-induced fission of ²⁵⁵Fm[†]

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The kinetic energy distributions of coincident fission fragments from the thermal-neutroninduced fission of 255 Fm and 251 Cf have been measured with phosphorus-diffused silicon detectors. The most probable values for the postneutron total kinetic energy are 192.5 ± 2.9 MeV for 255 Fm and 182.1 ± 2.7 MeV for 251 Cf. Fragment mass distributions were calculated without applying neutron emission corrections. The resultant mass and kinetic energy distributions for 255 Fm indicate a predominantly asymmetric mass division combined with appreciable symmetric fission. Fragment pairs near mass symmetry were found to be unusually energetic, which is a characteristic shared with symmetric fission in 257 Fm and 258 Fm. These results are well described by the two-center model of fission. Thermalneutron-induced fission cross sections were measured as 3400 ± 170 b for 255 Fm and 4800 ± 250 b for 251 Cf.

NUCLEAR REACTIONS, FISSION 255 Fm(n, f), 251 Cf(n, f), E = 0.025 eV; measured σ , fragment E; deduced fragment masses.

I. INTRODUCTION

Recent experiments measuring the kinetic energies of fragments produced by the spontaneous fission^{1,2} (SF) and the thermal-neutron-induced fission¹ (n, f) of ²⁵⁷Fm have shown that in the transition from ²⁵⁷Fm to ²⁵⁸Fm the primarily asymmetric mass distribution becomes predominantly symmetric. However, a recent radiochemical study³ of ²⁵⁶Fm(SF) has shown its mass distribution to be largely asymmetric, with a peak-to-valley ratio of approximately 12. We have studied prompt fission following neutron absorption by ²⁵⁵Fm in order to evaluate the effect of excitation energy in this region, where there is a rapid transition from asymmetric to symmetric mass yields, and to compare our results with theoretical predictions.

Fission theory has provided several alternative explanations for the asymmetric mass distribution in fission. In one qualitative explanation, asymmetry is explained by strong shell effect within the residual fission product nuclei.⁴ Other explanations, offering much more detail, have resulted from mapping the potential energy surface with the degree and the symmetry of nuclear deformation. Calculations with different static models have been carried out to deformations either just beyond the second or outer bar $rier^{5-12}$ using a single potential, or, by using two-center potentials, 13-20 to the point of scission. Either treatment leads to the same qualitative conclusion-that asymmetric fission is fairly well localized in the actinide region and that there are

transitions to symmetric fission just below ²²⁸Ra and just above ²⁵⁸Fm. Although the same conclusion has been reached from both the single- and the two-center potential models, the physical reasons for reaching this conclusion are quite different. Because of this, a choice of theoretical models is opened to experimental test by studying the mass and kinetic energy distributions from the prompt fission of ²⁵⁶Fm*.

Calculations made by Möller and Nilsson^{5,9} using a liquid-drop model corrected for single-particle effects show that the minimum energy path for fission is always through a symmetric inner barrier and through an outer barrier that is asymmetric or symmetric, depending on the nuclide (Z, A). Saddle points for both asymmetric and symmetric distortions are available at the outer barrier, and the one lying lower in energy would presumably govern the mass split at the scission point. For the heavier actinides the outer barrier is only a few MeV above the ground state, and asymmetric distortions are favored over symmetric ones by only a few tenths of an MeV. Thus, at excitation energies well above the level of the outer barrier, the effects of this barrier on the mass split should not be seen; a predominance of symmetric fission, which is prescribed by the inner barrier, would be seen. Basing their predictions on their calculations of the outer barrier heights, Tsang and Wilhelmy²¹ suggested that ²⁵⁶Fm* should fission symmetrically.

The two-center model developed by Mosel and co-workers¹³⁻¹⁸ allows calculation of the potential energy surfaces to much greater deformations than

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the single-potential model of Strutinski and Nilsson. As a result, it is possible to describe the energy surface from the ground state of the fissioning nucleus to the configuration of two completely separated fragment nuclei. From such calculations, it was concluded that shell structure in nascent fragments is important in the very early stages of the fission process, particularly in the descent from the outer saddle to the scission point.¹⁴ The effect of fragment shells becomes especially important in determining the mass split when both fragments are doubly magic ¹³²Sn nuclei, such as might occur in the fission of ²⁶⁴Fm. Thus, the transition from asymmetric fission in the light fermium isotopes to symmetric fission in the heavier ones is due to the fragments approaching closed proton and neutron shells (Z=50, N=82).¹⁷ If this explanation should be correct, then excitation energy from binding a neutron should not be the primary cause of symmetric fission in the heavy fermium isotopes. Based on this model, we would not expect a dramatic increase of symmetric fission for ²⁵⁵Fm-(n, f) compared to ²⁵⁶Fm (SF). Rather, some broadening of the mass distribution might be expected due to slight washing out of shell structure at ~6.5 MeV excitation energy in ²⁵⁶Fm*.

II. EXPERIMENTAL

Isotopically pure samples of 20.3-h ²⁵⁵Fm were prepared by chemically separating it from its parent, 40.6-day ²⁵⁵Es. The ²⁵⁵Es was produced in 0.035% abundance, relative to ²⁵³Es, by neutron irradiation of lighter actinides in the high flux isotope reactor at the Oak Ridge National Laboratory. A few hours before each fission experiment the ²⁵⁵Fm, in equilibrium with ²⁵⁵Es, was separated from ~2.1 μ g of Es (mainly ²⁵³Es) by elution from a cation exchange column with α -hydroxyisobutyric acid. Einsteinium and californium were reduced to undetectable amounts by repeating this ion-exhange separation once. Common elements that would contribute to the mass of the final sample were separated in a small quartz column contining Dowex 50×12 colloidal resin. After adsorption from 0.5 M HCl, the fermium was carefully eluted with 2 and 6 M ultrapure HCl. The final target for fission counting was then prepared by electroplating the 255 Fm from 0.001 *M* HNO₃ to form a 2-mm-diam spot on a ~175- μ g/cm² gold foil. We required approximately 4 h to separate and prepare the ²⁵⁵Fm target chemically, place it in the counter, and then bring the reactor to full power before starting the first fission count.

Three ²⁵⁵Fm samples were prepared and fission counted, but only the data collected from the third

and final run are reported here. This target contained $(3.37 \pm 0.10) \times 10^{10}$ atoms of ²⁵⁵Fm as determined by α -counting and α -pulse analyses. No activities other than ²⁵¹Cf (α daughter of ²⁵⁵Fm) were detected in the sample after complete decay of the ²⁵⁵Fm. As a gauge of source thickness, we measured the resolution of ²⁵¹Cf α particles, and these were found to be about 35 keV full width at half maximum (FWHM).

In addition to the ²⁵⁵Fm target, a spontaneousfission source of ²⁵²Cf and a neutron flux monitor of ²³³U were prepared on 180- and 230- μ g/cm² gold foils, respectively. The isotopically pure ²³³U (separated from ²³⁷Np) was used as a reference standard for measuring the thermal-neutronfission cross sections of ²⁵⁵Fm and its α daughter, ²⁵¹Cf. This standard contained 1.334±0.007 ng of ²³³U, a value determined by α counting.

The ²⁵⁵Fm target was mounted onto a four-position target wheel located between two phosphorus-diffused silicon detectors covered with aluminum collimators to restrict the entry angle to 50°. The other targets on the wheel consisted of the electroplated ²⁵²Cf source for energy calibration, the ²³³U source, and a blank ~175- μ g/cm² gold foil for background runs. All sources were sandwiched between the gold backing and the ~175- μ g/cm² gold cover foils in order to prevent detector contamination. The source-to-detector distance was approximately 2 mm. The assembly, along with an aluminum-covered noise detector, was mounted inside an evacuated aluminum chamber and inserted into the Livermore pool-type reactor thermal column in a thermal flux of $2 \times 10^{11} n/cm^2$ sec with a cadmium ratio for gold of 600. The detectors were cooled to -25 °C by circulating refrigerated alcohol through cooling lines attached to the detector mounting plates. The target turning shaft, cooling lines, and detector leads were carried outside the thermal column through a 2-m-long evacuated tube. Fast linear electronics were used to process the coincident fragment pulses for pulse-height analysis. Pulse pile-up rejection was employed, and noise pickup was eliminated by the use of the noise detector in an anticoincidence mode. Upon insertion into the $2 \times 10^{11} n/cm^2 \sec flux$, the leakage currents of the cooled detectors immediately rose from less than 1 μ A to 12 μ A at 200-V reverse bias and remained there until the end of the experiment.

The amplified pulses were fed to a two-parameter pulse-height analyzer operating in a 512-event buffer mode. The pulse heights of the coincident fragments were buffered onto magnetic tape for subsequent computer sorting and analysis. Details of the experimental procedure have been published previously.22

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The coincident fission-fragment counting rate from the ²⁵⁵Fm-²⁵¹Cf target increased from 500 counts/min at the beginning of the experiment to 660 counts/min at the end. Fission counts from ²⁵⁶Fm(SF) (produced by n, γ reactions) were less than 1% of the prompt fissions from ²⁵⁵Fm(n, f), since the neutron-capture cross section of ²⁵⁵Fm is only 30 b.²³

The experiment was carried out for seven days in order to follow the decay of 255 Fm to 251 Cf. Approximately 15 fission spectra were taken over the course of the experiment. For calibration and flux monitoring purposes, the fission spectra of 255 Fm and 251 Cf were interspersed with fission spectra from the 252 Cf and 233 U targets. At the midpoint of the first fission spectrum taken, the source was 80.0% 255 Fm and 20.0% 251 Cf. At the midpoint of the last spectrum taken, the target was 99.7% 251 Cf.

The fission cross section of ²⁵¹Cf was determined from the fission rate during the last few spectra, when the sample was essentially pure 251 Cf. A cross section of 4800 ± 250 b was calculated for ²⁵¹Cf from the ratio of the fission rate of ²⁵¹Cf to that of the ²³³U standard, the measured atom ratios in each, and the known thermal-neutronfission cross section²⁴ of 531 b for ²³³U. A thermal-neutron-fission cross section of 3400±170 b was calculated for ²⁵⁵Fm in a similar way, but the fission rate was determined by least-meansquares fitting of the growth-decay curve of the gross fission rates. In the fitting, which included 14 decay points taken over 7 days, gross fission rates were normalized to a constant neutron flux by using the fission rate of the ²³³U standard. The half-lives of 251 Cf (898 yr) and 255 Fm (20.3 h) were held as fixed parameters. A standard deviation of 3.7% was obtained in this least-mean-squares fit.

Energy calibrations were based on an average post neutron emission energy of 183.9 MeV for ²⁵²Cf, which is an average of the results of Schmitt, Neiler, and Walter²⁵ and Whetstone.²⁶ A correction was made for the pulse-height defect according to the method of Schmitt, Kiker, and Williams.²⁷ Care was taken to operate the detectors in the saturation region to ensure the validity of the calibration procedure. In order to include the effects of energy losses in the gold foil and the detector window, the ²⁵²Cf fragments were analyzed under the identical conditions as were the ²⁵⁵Fm and ²⁵¹Cf fragments. The calibration constants thus obtained included the corrections for energy losses. These calibration constants were then used to analyze the energy spectrum of the ²⁵⁵Fm and ²⁵¹Cf fragments.



FIG. 1. Contour diagrams of counts vs provisional fragment mass and postneutron total kinetic energy: (a) 255 Fm (n, f) for 172 881 events are grouped into 5.0-MeV and 3.4-amu bins; (b) 251 Cf $(n_x f)$ for 367 114 events are grouped into 2.5-MeV and 1.7-amu bins; 252 Cf(SF) for 332 434 events are grouped into 2.5-MeV and 1.7-amu bins.



FIG. 2. Postneutron emission total kinetic energy distribution for the thermal-neutron-induced fission of 255 Fm. The most probable kinetic energy is 192.5 ± 2.9 MeV. The FWHM is 42.5 MeV.

No corrections were made for neutron emission since Balagna *et al.*² had shown that the mass distribution is extremely sensitive to $\nu(M)$. Their use of a $\nu(M)$ correction corresponding to the ²⁵²Cf $\nu(M)$ created a double-humped preneutron emission massyield curve for ²⁵¹Fm(SF) with a peak-to-valley ratio of 1.5, whereas a constant $\nu(M)$ value of 2 created a flat-topped curve with no valley.² Therefore, in order not to distort the data in any arbitrary way, we present the mass data which correspond to the provisional masses of Schmitt, Neiler, and Walter.²⁵ No corrections were made for instrumental resolution.

III. RESULTS

Figure 1(a) shows the results for thermal-neutron fission ²⁵⁵Fm represented as a contour diagram of counts vs fragment mass and total kinetic energy. This plot was obtained by subtracting out the events from the thermal-neutron fission of ²⁵¹Cf, which was done by weighting the ²⁵¹Cf fission spectra according to the amount of ²⁵¹Cf in the target and using the fission cross sections for 255 Fm and 251 Cf derived from fission rates as a function of time. This procedure amounted to a subtraction of 141 045 ²⁵¹Cf fission events out of a total of 313 926 gross fission events, a 45% correction. The highest points (2787 events) occur at the asymmetric fission masses of 114 and 142 for a postneutron emission energy of 194 MeV. The most energetic contour on the plot occurs at mass 128 and equals 254 MeV (473 events out of a total of 172 881 events). Figure 2 shows a plot of the kinetic energy for 255 Fm(n, f) obtained by summing over all fragment masses. The average postneutron kinetic energy is computed $\langle E \rangle$



FIG. 3. Provisional mass distributions for the thermalneutron-induced fission of 255 Fm and of 257 Fm. The 257 Fm distribution is taken from Ref. 1 and represents 15900 fissions.

 $= \sum_{i} N_{i} (E_{i}) E_{i} / N$ to be 192.5±2.9 MeV.

In Fig. 3, we show the mass distribution from 255 Fm(n, f) without neutron correction, a relatively flat distribution. A neutron correction $\nu(M)$ similar to that for 252 Cf(SF), in which there is a sharp drop in $\nu(M)$ at symmetric fission, would produce a dip at mass symmetry in the calculated preneutron-emission mass yields. Since a detailed behavior of $\nu(M, E)$ is not known for the fermium isotopes, it was felt that the provisional mass distribution for 255 Fm(n, f) would be the most valid method of presenting the data.

Figure 1(b) represents the counts vs mass and energy for 251 Cf. This contour diagram is very similar to the analogous plot for 252 Cf (SF) shown



FIG. 4. Provisional mass distributions for the thermalneutron-induced fission of 251 Cf and for the spontaneous fission of 252 Cf.

in Fig. 1(c), except that there are more events in the region near a symmetric mass split. Figure 4 shows the mass distribution for $^{251}Cf(n, f)$, which again resembles the $^{252}Cf(SF)$ mass distribution. The peak-to-valley ratios are 3.1 for $^{251}Cf(n, f)$ and 5.3 for $^{252}Cf(SF)$. The FWHM for the ^{251}Cf -(n, f) kinetic curve was 36.5 MeV, compared to 31.7 MeV for the $^{252}Cf(SF)$ energy curve. The exact difference in target thicknesses is unknown, and therefore the effect of this difference on the mass distribution is not known. The average postneutron total kinetic energy for ^{251}Cf thermal-neutron-induced fission was computed to be 182.1 ± 2.7 MeV.

Table I compares our kinetic energy measurements to those for other fermium and californium isotopes. Our reported errors of ± 2.9 and ± 2.7 MeV for ²⁵⁵Fm and ²⁵¹Cf, respectively, include a 2σ value (1.0 MeV) for the variation of the different runs about the mean energies reported. The errors also include an estimate to account for any differences in thickness between the combined ²⁵⁵Fm-²⁵¹Cf target and the ²⁵²Cf target. In general, the results seem quite reasonable for the californium isotopes, considering the errors quoted for the measurements. The $\frac{255}{5}$ Fm(n, f) and $\frac{257}{5}$ Fm(SF)data agree quite well; however, the 257 Fm(n, f) $\langle E \rangle$ value appears too low. Balagna *et al.*² have measured the average energy for 257 Fm(SF) to be 195.1 ± 2.9 MeV, whereas John *et al.*¹ measured the average postneutron energy for 257 Fm(n, f) to equal 180 MeV. This may have been the result of inadequate statistics. The approximate total preneutron kinetic energy of 195 MeV for 255 Fm (n, f)is higher than the value of 191 MeV calculated from Viola's³⁰ empirical relationship $\langle E \rangle$ = 0.1071 $Z^2/A^{1/3}$ + 22.2, based on the liquid-drop model. This disagreement is understandable considering that 12% of the events have energies above 220 MeV. Since the predicted energies decrease as A increases, the preneutron kinetic energy (198 MeV) reported² for ²⁵⁷Fm(SF) is in greater disagreement with the empirical curve

TABLE I. Average postneutron total kinetic energies.

	$\langle E \rangle$ (MeV)	FWHM (MeV)	Ref.
²⁵⁴ Fm (SF)	186 ± 2	27.6	28
255 Fm (n, f)	192.5 ± 2.9	42.5	This work
²⁵⁷ Fm (SF)	195.1 ± 2.9	36.4	2
257 Fm(n, f)	180		1
²⁵⁰ Cf (SF)	181.8 ± 2.7	30.2	29
${}^{251}Cf(n, f)$	182.1 ± 2.7	36.5	This work
²⁵² Cf (SF)	184.1 ± 2.8	30.8	29
²⁵⁴ Cf (SF)	181.8 ± 2.7	33.8	29

(190 MeV) than the 255 Fm(n, f) energy. It should be pointed out that Viola's relationship was intended to predict the trend for asymmetric fission and did not anticipate the symmetric fission which occurs in 255 Fm(n, f) and 257 Fm(SF).

Since fragments coming from symmetric fission have higher kinetic energies than those associated with asymmetric fission, we can differentiate symmetric fission by the unusually high total energies of these events. Figure 5 illustrates the changes in mass distribution as the total kinetic energy of fission increases. The double-humped distribution gradually shifts over to a singlehumped curve which becomes narrower as the kinetic energy rises. A single-humped distribution first occurs at 220–225 MeV, and we have assumed that all events at or above this energy



FIG. 5. Provisional mass distributions for 256 Fm* fission grouped as a function of their total kinetic energies. Each curve shows the mass distribution at a fixedtotal kinetic energy (within a 5-MeV band). They are constant energy cuts across the mass-energy contours of Fig. 1(a).

originated from symmetric fissions. Inasmuch as we have no way of distinguishing symmetric events below this energy, our cutoff at 220 MeV is merely qualitative and has been used arbitrarily in estimating the percentage of symmetric fission. Approximately 12% of the events have kinetic energies greater than 220 MeV. From the relative portion of such events, we describe the mass distribution from ²⁵⁶Fm* fission as primarily asymmetric with an appreciable symmetric component. We believe this method of estimating symmetry offers a higher sensitivity than total peak-tovalley ratios obtained from radiochemical measurements, since for the heavier fermium isotopes. a valley is barely discernible, if it appears at all. Until the valley is fully filled, the conclusions from radiochemical peak-to-valley ratios will likely be quite different from the conclusions reached from coincident-fragment energy measurements. Keeping these limitations in mind, we compare only those features in the fission of ²⁵⁶Fm* that are common to both types of measurements, namely the mass-yield curves.

Using radiochemical techniques, Flynn and coworkers³ have shown the mass-yield curve from 256 Fm(SF) to have a peak-to-valley ratio of 12. We observe no valley in our provisional mass distribution (Fig.3); therefore, it seems evident that there is more symmetric mass division from the fission of ²⁵⁶Fm* than from the spontaneous fission of $^{\rm 256}{\rm Fm}.$ This increase in near-symmetric mass division with an increase in excitation energy is consistent with the behavior of the lighter actinides. Flynn et al.³¹ used radiochemical methods to determine a peak-to-valley ratio of 2.5 for ²⁵⁵Fm-(n, f). Our data are consistent with their radiochem ical results after taking into account the broadening due to the resolution of the silicon detectors and the broadening from neutron emission.

IV. DISCUSSION

The most notable feature of our ²⁵⁶Fm* fission results is the unusually high fragment energies associated with masses near symmetric division This is seen by comparing Fig. 1(a) with Figs. 1(b) and 1(c). This same high-energy component is also characteristic of symmetric fission in ²⁵⁷Fm and ²⁵⁸Fm*. These distinctive symmetric events predominate in the fission of ²⁵⁸Fm*. Since the mass-energy contours shown in Fig. 1(c) are remarkably similar to those for ²⁵⁷Fm (SF), we have concluded that the neutron-induced fission of ²⁵⁵Fm gives mass and energy distributions equivalent to the spontaneous fission of ²⁵⁷Fm.

High kinetic energies from fragments near mass symmetry have now been observed in the neutron-

induced fission of ²⁵⁵Fm and ²⁵⁷Fm and the spontaneous fission of ²⁵⁷Fm, but not in the fission of lighter actinides. As noted in earlier reports,^{1,2} this is obviously related to the low internal excitation energy of the fragments caused by their spherical rigidity upon approaching the magic nucleon numbers Z = 50, N = 82. The total kinetic energy of fragments from some events approximates the Q value of the reaction, indicating nearly zero internal excitation energy. These properties of symmetric fission in the fermium isotopes are the inverse of those found in symmetric fission in ²³³U. ²³⁵U. and ²³⁹Pu. For the latter nuclides, there is a pronounced dip in the kinetic energy released for near-symmetric mass division and a corresponding increase in fragment excitation energy (an increase in ν).³² Fission products from nearsymmetric division of these nuclei lie in a region that is softer toward deformation than products from the near-symmetric fission of the fermium isotopes. Thus, the contrasting behavior in the division of kinetic and excitation energy by the lighter and heavier actinides is apparently related to the softness of the fragments toward deformation. We infer from such correlations that the partitioning of energy and mass in the fission process is governed by fragment shell structures.

We discern two modes of fission (asymmetric and symmetric) from Fig. 1(a), since symmetric fission -as a separate, observable mode-is distinguishable by having unusually high fragment energies. From this contour plot and also from the ones for 257 Fm(SF) and 257 Fm(n, f), ¹ a new type of symmetric division appears superimposed upon a normal asymmetric one. This suggests a genuine form of symmetric fission arising only from symmetric deformations. The mass and energy distributions from the asymmetric mode of fissioning in these isotopes seem about as expected for nuclei in this mass range. In this mode, near-symmetric masses come from symmetric scission of asymmetrically deformed nuclei. In this respect a distinction is made between a symmetric mass division and genuine symmetric fission

Our conclusion regarding the appearance of two distinct fission modes suggests a relationship to the two-mode fission hypothesis first proposed by Turkevitch and Niday.³³ This hypothesis has been used to interpret mass distributions from fission of excited nuclei in the Ra-Ac region where the relative probabilities for symmetric and asymmetric fission are about equal at excitation energies near 20 MeV. A three-peaked mass distribution representing symmetric and asymmetric fission modes has also been observed in the heliumion-induced fission of ²³³U and the fission of ²³²Th

caused by reactor spectrum neutrons.³⁴ Most importantly, there was an increasing yield of the symmetric mode with increasing excitation energy. Therefore, a possibility exists that the symmetric component that we find in the neutron fission of 255 Fm and 257 Fm is a result of excitation energy. An enhancement of symmetric division caused by excitation from the neutron separation energy is very evident from the two radiochemical studies of 256 Fm fission, but this increase of valley-to-peak ratios with excitation energy seems similar to the general trend established for the lighter actinides and for the prompt fission of ²⁵²Cf* reported here. Thus, this customary increase of the symmetric mode with rising excitation energy appears unrelated to the onset of the new type of symmetric fission in the heavy fermium isotopes. Our conclusion here is also based on the large increase in symmetric fission for ²⁵⁷Fm-(n, f) compared to ²⁵⁵Fm(n, f) in which the excitation energies of each of these nuclides before fissioning are comparable. At the moment, we view this symmetric mode as a direct consequence of strong shell effects in fragment nuclei near doubly closed-shell ¹³²Sn.

The mass and kinetic energy distributions from the fission of ²⁵⁶Fm* are in good agreement with the qualitative predictions of the two-center model of fission. Results from calculations using this model correctly anticipated a transition from asymmetric fission in the light fermium isotopes to symmetric fission in the heavier $ones^{17}$ and an increase in the total kinetic energy of fragments near mass symmetry, together with a decrease in internal excitation energy.¹⁶ A quantitative comparison is available from the total kinetic energy of the fragments. Assuming a symmetric division of charge and mass from the fission of ²⁵⁶Fm, Schmitt and Mosel¹⁶ calculate a total kinetic energy of ~210 MeV, while we obtained 212 MeV for \overline{E}_{k} (A = 128).

On the other hand, our results disagree somewhat with the prediction of Tsang and Wilhelmy²¹ that ²⁵⁶Fm* should fission primarily symmetrically.³⁵ Their prediction was based largely upon the relation of the excitation energy to the height of the second or outer barrier. In the case of ²⁵⁶Fm*, asymmetric deformations at this barrier are favored over symmetric ones by a saddle ~ 0.2 MeV lower in energy. However, Tsang and Wilhelmy expected the excitation energy after neutron capture to be ~3.2 MeV greater than the outer barrier, leading them to propose that symmetric distortions should be preferred equally to asymmetric ones. This theory excludes completely the effects of fragment shells upon the mass division and depends instead on potential energy

surfaces originating from shell structure of the parent fissioning nucleus. Since the mass division of ²⁵⁶Fm* fission is primarily asymmetric and, therefore, since their model failed this test, we feel the influence of fragment shells needs to be included in a suitable theory.

Wilkins and Steinberg,³⁶ in extending earlier approaches,³⁷⁻⁴⁰ have incorporated the effect of fragment shells on the total potential energy of the system when at large deformations. In this model, potential energies for strongly deformed fragments were calculated for the case where the fragments were still joined (somewhere between the outer saddle and scission points). Fragment pairs yielding the lowest total potential energy were assumed to determine the most probable mass and charge distributions. With this simplified model, the mass distributions of many nuclides were surprisingly well fitted, particularly by slightly adjusting the deformation parameter β of the fissioning nucleus where the mass division is decided.

An asymmetric mass distribution for the thermal-neutron-induced fission of ²⁵⁵Fm was anticipated by Wilkins and Steinberg.³⁶ However, contrary to our results, they estimated that the total kinetic energy released in symmetric division would be a nominal 200 MeV rather than the unusually high energies observed. To account for this discrepancy, it would have been necessary for them to let some portion of the mass divisions be determined at a much smaller β , such as that β associated with the inner symmetric barrier. Further refinements of this model may allow this to be done, but at this time it appears to have some arbitrary features.

All of the theoretical approaches that we have compared with our experimental data are limited to the extent that they are unable to fully reproduce the details given in the contour plot of Fig. 1(a). We note that these approaches are rooted in a common base, namely, the calculating of quasistatic potential energy surfaces for nuclei at very large deformations. Since static calculations are inherently limited in a dynamic process, further advancements in providing a detailed explanation of fission will eventually require a full dynamic treatment for deformations from the ground state to the scission point.

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- ¹W. John, E. K. Hulet, R. W. Lougheed, and J. J. Wesolowski, Phys. Rev. Lett. <u>27</u>, 45 (1971).
- ²J. P. Balagna, G. P. Ford, D. C. Hoffman, and J. D. Knight, Phys. Rev. Lett. <u>26</u>, 145 (1971).
- ³K. F. Flynn, E. P. Horwitz, C. A. A. Bloomquist, R. F. Barnes, R. K. Sjoblom, P. R. Fields, and L. E. Glendenin, Phys. Rev. C 5, 1725 (1972).
- ⁴P. Fong, Phys. Rev. <u>102</u>, 434 (1956).
- ⁵P. Möller and S. G. Nilsson, Phys. Lett. <u>31B</u>, 283 (1970).
- ⁶H. C. Pauli, T. Ledergerber, and M. Brack, Phys. Lett. 34B, 264 (1971).
- ⁷C. Gustafsson, P. Möller, and S. G. Nilsson, Phys. Lett. <u>34B</u>, 349 (1971).
- ⁸H. Schultheis and R. Schulteis, Phys. Lett. <u>34B</u>, 245 (1971).
- ⁹P. Möller, Nucl. Phys. A192, 529 (1972).
- ¹⁰V. V. Pashkevich, Nucl. Phys. <u>A169</u>, 275 (1971).
- ¹¹J. R. Nix, Annu. Rev. Nucl. Sci. <u>22</u>, 341 (1972).
- ¹²M. Bolsterli, E. O. Fiset, J. R. Nix, and J. L. Norton, Phys. Rev. C 5, 1050 (1972).
- ¹³D. Scharnweber, W. Greiner, and U. Mosel, Nucl. Phys. A164, 257 (1971).
- ¹⁴U. Mosel, J. Maruhn, and W. Greiner, Phys. Lett. <u>34B</u>, 587 (1971).
- ¹⁵U. Mosel and H. W. Schmitt, Phys. Rev. C <u>4</u>, 2185 (1971).
- ¹⁶H. W. Schmitt and U. Mosel, Nucl. Phys. <u>A186</u>, 1 (1972).
- ¹⁷M. G. Mustafa, U. Mosel, and H. W. Schmitt, Phys. Rev. Lett. <u>28</u>, 1536 (1972).
- ¹⁸M. G. Mustafa, U. Mosel, and H. W. Schmitt, Phys. Rev. C 7, 1519 (1973).
- ¹⁹B. Slavov, J. E. Galonska, and A. Faessler, Phys. Lett. 37B, 483 (1971).
- ²⁰G. D. Adeev, P. A. Cherdantsev, and I. A. Gamalya, Phys. Lett. <u>35B</u>, 125 (1971).
- ²¹C. F. Tsang and J. B. Wilhelmy, Nucl. Phys. <u>A184</u>, 417 (1972).
- ²²J. J. Wesolowski, W. John, and J. Held, Nucl. Instrum. Methods <u>83</u>, 208 (1970).
- ²³R. W. Hoff, J. E. Evans, E. K. Hulet, R. J. Dupzyk,

and B. J. Qualheim, Nucl. Phys. <u>A115</u>, 225 (1968). ²⁴Nucl. Data <u>B6</u>(No. 3), 275 (1971).

- ²⁵H. W. Schmitt, J. H. Neiler, and F. J. Walter, Phys. Rev. <u>141</u>, 1146 (1966).
- ²⁶S. L. Whetstone, Jr., Phys. Rev. <u>131</u>, 1232 (1963).
- ²⁷H. W. Schmitt, W. E. Kiker, and C. W. Williams, Phys. Rev. <u>137</u>, B837 (1965).
- ²⁸R. Brandt, S. G. Thompson, R. C. Gatti, and L. Phillips, Phys. Rev. 131, 2617 (1963).
- ²⁹D. C. Hoffman, G. P. Ford, and J. P. Balagna, Phys. Rev. C 7, 276 (1973).
- ³⁰V. E. Viola, Jr., Nucl. Data <u>A1</u>, 391 (1966).
- ³¹K. F. Flynn, J. E. Gindler, R. K. Sjoblom, and L. E. Glendenin, in *Third International Symposium on the Physics and Chemistry of Fission, Rochester, N. Y., 13-17 August 1973* (International Atomic Energy Agency, Vienna, 1973), Paper 209 reported by J. P. Unik.
- ³²E. K. Hyde, The Nuclear Properties of the Heavy Elements (Prentice Hall, Englewood Cliffs, N. J., 1964), Vol. III, p. 187.
- ³³A. Turkevich and J. B. Niday, Phys. Rev. <u>84</u>, 52 (1951).
 ³⁴See Ref. 32, p. 330.
- ³⁵C. F. Tsang and J. B. Wilhelmy's definition of "symmetric fission" is unclear. Although they have stated that ²⁵⁶Fm* should fission symmetrically, they have also equated this term to a mass distribution having a peakto-valley ratio near 1. They also indicated that the mass distributions from the neutron-induced fission of ²⁵⁵Fm and ²⁵⁷Fm should be much alike, which they are not.
- ³⁶B. D. Wilkins and E. P. Steinberg, Phys. Lett. <u>42B</u>, 141 (1972).
- ³⁷F. Dickmann and K. Dietrich, Nucl. Phys. <u>A129</u>, 241 (1969).
- ³⁸H. W. Schmitt, in Proceedings of the Second International Symposium on Physics and Chemistry of Fission, Vienna, Austria, 1969 (International Atomic Energy Agency, Vienna, 1969), p. 67.
- ³⁹V. S. Ramamurthy and R. Ramanna, in Proceedings of the Second International Symposium on Physics and Chemistry of Fission, Vienna, Austria, 1969 (see Ref. 38), p. 50.
- ⁴⁰A. V. Ignatyuk, Yad. Fiz. <u>7</u>, 1043 (1968) [transl.: Sov. J. Nucl. Phys. <u>7</u>, 626 (1968)].

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