

We have searched diligently for alpha radiation from the isotope $^{258}\text{102}$ without success. From the data for all of the nuclides in this region we would predict an alpha-decay half-life of about a minute and a production cross section via the reaction $^{248}\text{Cm}(^{13}\text{C}, 3n)$ of the order of 10^{-31} cm^2 . We should have readily observed its presence either directly or via its daughter ^{254}Fm in alpha-recoil milking experiments and consequently we feel that its most likely mode of decay is by spontaneous fission. Preliminary experiments set a half-life limit of much less than a second for spontaneous fission.

This work was aided greatly by the efforts of many people but we would particularly like to acknowledge the assistance of the following: T. Bowman for preparation of the targets, R. Latimer for target material purification, P. Fields and J. Lerner of the Argonne National Laboratory for the isotopically separated ^{246}Cm , A. Larsh for electronics support, C. Corum for mechanical design, and F. Grobelch and the HILAC crew for the superb performance

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TERNARY FISSION OF HEAVY NUCLEI*

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A heightened interest has lately centered on that fission process by which three large fragment masses are formed. We wish to present preliminary results from the study of ternary fission of a variety of heavy nuclei. Although the energy and mass distributions (and implications therefrom) are in themselves of considerable interest, the main purpose of this communication is to point out how these data are inconsistent with an explanation based on scattering phenomena, a view which is currently popular in some circles.

The experimental arrangement has been previously described.¹ Briefly, it consists of three solid-state detectors positioned 120° apart in a plane about a fission source. The physical properties of the sources were essentially identical (excepting the Cf^{252} source¹) and consisted of a tetrafluoride deposit on $20\text{-}\mu\text{g}/\text{cm}^2$ VYNS film support; areal densities varied around $0.100 \mu\text{g}/\text{cm}^2$. Calibration against the binary-fission fragment spectrum was made for each

foil before and after each experiment. The output pulse from each detector was digitally analyzed and recorded event by event on punched paper tape whenever a parallel triple-coincidence circuit was satisfied. Energy calibration was achieved by comparing the binary-fission fragment spectrum with that from time-of-flight data² in order to locate the energy position of the average light and heavy masses. A straight-line calibration was then applied; alternatively, a mass-dependent approach as proposed by Schmitt *et al.*³ was used.

The following fissioning systems were investigated:

$$\text{Cf}^{252} \rightarrow (\text{spontaneous})\text{TF}, \quad (1)$$

$$\text{Pu}^{241} + n_{\text{th}} \rightarrow \text{TF}, \quad (2)$$

$$\text{Pu}^{239} + n_{\text{th}} \rightarrow \text{TF}, \quad (3)$$

$$\text{U}^{235} + n_{\text{th}} \rightarrow \text{TF}, \quad (4)$$

and

$$U^{233} + n_{th} \rightarrow TF, \quad (5)$$

where TF stands for ternary fission.

For the latter four systems, irradiation of the experimental assembly was made in the thermal column of the University of Florida Training Reactor (UFTR).

The frequency of occurrence of ternary fission was found to be greater than about 1 per 10^6 binary fissions for all systems studied.

The energy distributions of the individual fragments are shown in Fig. 1. If we compare the two fissioning systems U^{234*} and U^{236*} , a distinct difference is evident in the fragment kinetic energy spectrum in the range (60–80 MeV) corresponding to the energy of the heavy fragment from binary fission.

It is well known⁴ that the features of (elastic) Coulomb scattering, as applied to fission fragments colliding with heavy nuclei, depend

only upon the following properties: (a) mass, energy, and charge of scattered fragment, and (b) mass and charge of the recoil nucleus (initially at rest in the laboratory system). If one assumes that the data result from binary-fission fragment scattering with uranium nuclei, then one would expect virtually identical fragment kinetic-energy spectra because of the following qualitative reasons. The energy, mass, and charge spectra of the binary-fission fragments would be essentially equivalent² for the fissioning systems U^{234*} and U^{236*} , and the recoil nuclei ($_{92}U^{233}$ vs $_{92}U^{235}$) would be different by only two neutron numbers.

Thus, the similarity of properties bearing upon the characteristics of (fission-fragment-uranium) Coulomb scattering indicates that similarities in kinetic-energy spectra would result if scattering phenomena were responsible, an expectation clearly at odds with the observed data.

Another way of stating this inconsistency is as follows [referring to Figs. 1(a) and 1(b)]: If scattering phenomena are indeed responsible, for some unexplained reason the scattered fragment is more often the heavy fragment (lower energy) for the U^{234*} system, whereas for the U^{236*} system, scattering by the light and heavy fragments are about equally probable.

Without lengthening this paper unnecessarily we state simply that a distinct dissimilarity (even though based on fewer events) is also apparent in the two comparable fissioning systems Pu^{240*} and Pu^{242*} [Figs. 1(c) and 1(d)]. The ternary-fissioning system of Cf^{252} is shown for collation [Fig. 1(e)]. It is difficult, if not impossible, to assign these data to scattering phenomena without postulating a most unusual scattering mechanism. We interpret the marked difference in the kinetic-energy spectra associated with a two-neutron difference in fissioning systems as reflecting the commanding influence of shell effects on the scission configuration leading to tripartite division.

In this comparison of fragment kinetic energies (Fig. 1), we have used a calibration that is independent of the origin of the events. Using a mass-dependent calibration³ (and accepting the events as originating from a ternary-fission process) the kinetic-energy spectra for the various fissioning nuclei appear as in Fig. 2. A generally lower value for the fragment kinetic energies is characteristic of the

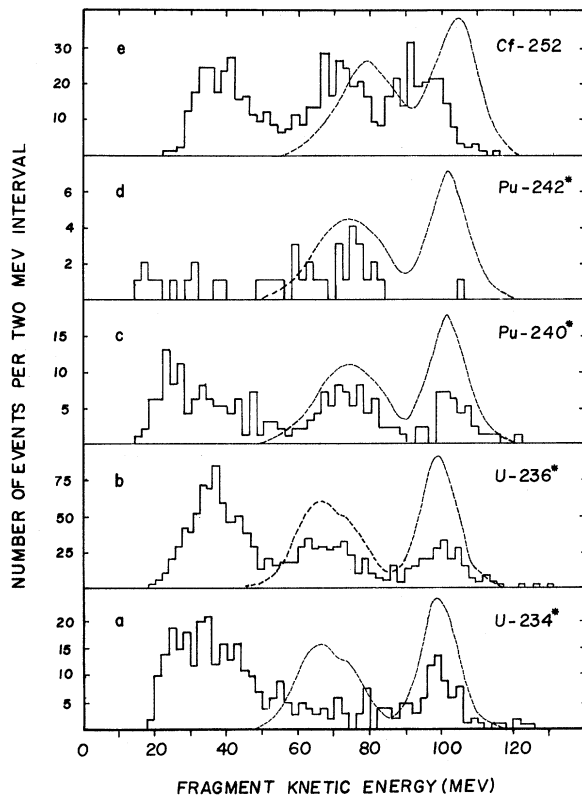


FIG. 1. Fragment kinetic-energy distribution using a straight-line calibration. The broken curve here and in Fig. 2 represents the kinetic-energy distribution from binary fission as reported in Ref. 2 and as reproduced in these experiments.

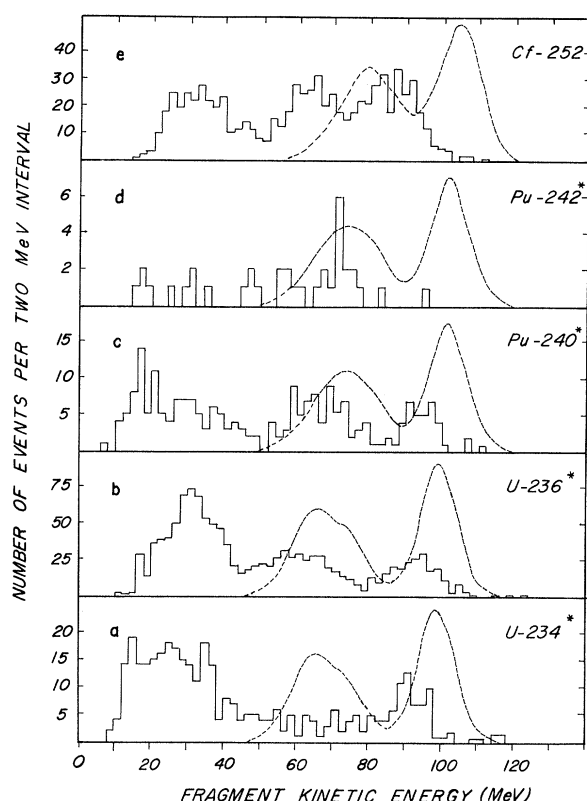


FIG. 2. Fragment kinetic-energy distribution using a mass-dependent calibration.

mass-dependent calibration.

The total fragment kinetic-energy release is slightly less than that for binary fission; average values are listed in Table I. These values are lower than those reported in Ref. 1 because of the use of a different calibration (mass-dependent) scheme. The lower kinetic-energy release coupled with the higher total mass-energy release (as estimated from semi-empirical mass formulas⁵) for ternary fission indicates a rather large amount of excitation energy for the final products and a generally greater distortion for the scission configuration as compared to binary fission.

The distinctive features of each fissioning system are shown in more striking form in Fig. 3. Here the spectra of the lightest-mass fragment are shown for comparison; a prominent feature is the appearance of two peaks, a lower one in the mass range 30-40 and a higher one centered near mass 50.

As suggested by earlier work,¹ we distinguish events as to type I or II based on the mass value of the lightest fragment. Using the mass

Table I. Total kinetic-energy release.

Fissioning system	\bar{E}_{total} (MeV)	
	Ternary fission ^a	Binary fission
Cf ²⁵²	185 ± 3	185
Pu ^{242*}	167 ± 2	174
Pu ^{240*}	153 ± 5	174
U ^{236*}	155 ± 5	168
U ^{234*}	144 ± 4	167

^aBased on mass-dependent calibration. Errors shown are average errors based on differences in average total kinetic energies from different data sets.

39-40 cut to separate the two peaks of Fig. 3 (type I corresponds to the higher peak, and type-II events to the lower), the ratio of type I to type II (I/II) is taken as a measure of the effect of underlying shell structure in determining the choice of division. This ratio is listed in Table II as a function of the fissioning system and other popular parameters.

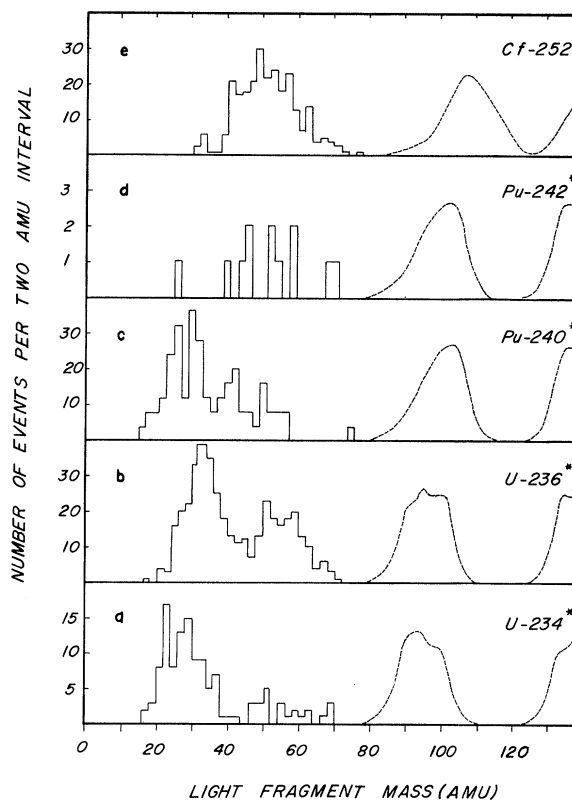


FIG. 3. Light-mass fragment distribution from ternary fission using mass-dependent calibration. The broken curve represents the mass distribution from binary fission.

Table II. Ratio of type-I to type-II events as function of fissioning system and other popular parameters.

Fissioning system	Type I ^a Type II	Fission parameter Z^2/A	Neutron excess $N-Z$
Cf ²⁵²	15 ± 4	38.1111	56
Pu ^{242*}	11	36.6639	54
Pu ^{240*}	0.5 ± 0.1	36.9707	52
U ^{236*}	0.9 ± 0.1	36.0170	52
U ^{234*}	0.3 ± 0.1	36.3262	50

^aErrors shown are statistical errors of counting. No error shown for Pu^{242*} system since only one type-II event was observed.

Quantitatively, the difference in the ratio (I/II) is quite distinct within each of the two pairs (Pu and U) of fissioning nuclei.

With the exception of the data of plots *d* of Figs. 1-3, the data are composites from multiple series of identical experiments for which, typically, 10-30 triple events were recorded per experiment. The 12 events obtained from the Pu^{242*} fissioning system (plots *d* of accompanying figures) were obtained from two separate runs (7 and 5 events, respectively) performed with two different experimental chambers, two different Pu²⁴¹ fission source foils, and two different sets of detectors. In all cases, for a given set of experimental conditions the results from separate runs were quite consistent and were reproducible within statistical limits.

Next, we wish to correct a slight, yet significant, misinterpretation of earlier work reported concerning the mass distribution from ternary fission. Whereas this distribution (in histogram form) appears in Fig. 3, it was carefully pointed out^{8,7} that this mass dispersion could be attributed to the finite angle subtended by the detectors; further, the possibility of very sharp mass peaks could not be excluded. This point seems to have been overlooked by some authors^{8,9} in evaluating the radiochemical work^{8,10,11} in the low-mass region (below 60 amu).

There is no reason, in fact, for precluding the possibility of forming one particular nuclide (perhaps stable) in the ternary-fission process. The very infrequency of the act suggests that stringent conditions must be satisfied as the nucleus glides over the barrier; these requirements must involve selective pathways leading

to at least one unique mass product. Indeed, the decided effect of shell structure (as shown in this work) in influencing the division appears to support this idea in a beautiful and striking manner. Hence, a broad interpolation covering all masses in the low-mass range (below 60 amu), and based on a few reported radiochemical mass yields, is premature.

In connection with the recent report⁹ on ternary fission at higher excitation energies, we wish to point out that radiochemical detection of a light-mass fragment alone does not in itself distinguish between the ternary- and binary-fission processes. To prove ternary fission (as opposed to binary) one must in addition show the absence of heavy fragments complementary to the light fragment in question. To assume (no matter what the basis) that such very asymmetric binary fission does not occur is equivalent to assuming that the alternative process, ternary fission, does occur, i.e., assuming what is to be proved. The interpretation of these groups is less analytic than their very fine experimental work. We concur, nonetheless (indeed it would be embarrassing if it were not so), that ternary fission very likely exists at these higher excitation energies.

In summary we emphasize the following themes:

(1) The fragment kinetic-energy spectra from selected pairs of fissioning systems (U^{234*,236*} and Pu^{240*,242*}) are quite inconsistent with an explanation based on scattering phenomena. Although some of the data may result from scattering, we interpret the major portion as arising from a ternary-fission process.

(2) The general trend and marked change in the ratio I/II as the fissioning system varies indicates the overriding influence of shell effects, which in turn suggests the possibility of unique mass formation. Because of this indication it is incorrect to state (as others have) that radiochemical evidence is in disagreement with instrumental results of ternary-fission studies.

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EXCITATION OF SIMPLE CONFIGURATIONS IN THE OUTGOING REACTION CHANNEL

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The existence of states due to simple particle-hole (p-h) configurations in proton closed shell nuclei has been the object of several recent letters.^{1,2} In particular, states in Ni and Sn isotopes observed by the $(p, p'\gamma)_{g.s.}$ ³ and $(^3\text{He}, d)$ processes, respectively, have been interpreted as intrinsic "core" states due to simple p-h configurations in the proton closed shell. Bloch⁴ has suggested that these reactions could be used to investigate gross structure in the outgoing reaction channel by exciting simple configurations in the residual nucleus.

Some preliminary results on ^{60}Ni have been presented at the Heidelberg conference.⁴ A comparison of the free-proton spectrum from the reaction $^{60}\text{Ni}(p, p')^{60}\text{Ni}^*$ with the coincidence-proton spectrum from the reaction $^{60}\text{Ni}(p, p'\gamma)^{60}\text{Ni}^*$ gated with corresponding ground state (g.s.) gamma transitions showed that several high-lying levels or level groups in ^{60}Ni were relatively strongly excited when gated with the corresponding ground-state transitions.¹ The above levels were associated with simple p-h configurations in the $Z=28$ (proton) core and the particular way of discrimination favored their selection.

We present here the results of an investigation of the reaction $^{40}\text{Ca}(p, p'\gamma)^{40}\text{Ca}$ using the

13-MeV proton beam of the Saclay tandem accelerator. The experimental setup is described in detail elsewhere.^{5,6} The signals from the proton and gamma detectors were led through a fast-slow coincidence apparatus⁶ to a multi-parametric system for information storage.⁷ The information was read into a CAE 510 computer which delivered two-dimensional p - γ coincidence contour plots and permitted the summing over the range corresponding to g.s. transitions.⁸

A comparison of proton coincidence spectra from the reaction $^{40}\text{Ca}(p, p'\gamma)^{40}\text{Ca}^*$ gated, respectively, with all gamma transitions and with corresponding g.s. transitions only is presented in Fig. 1.

Peaks at $E_x = 5.27$ -, 5.62 -, 5.90 -, 6.95 -, 7.95 -, 8.15 -, and 9.0 -MeV excitation show clearly when gated with g.s. gamma transitions. The latter four (6.95 , 7.95 , 8.15 , and 9.0 MeV) are practically washed out when gated with all gammas.

States observed in (p, p') ,⁹ (α, α') ,¹⁰ $(^3\text{He}, d)$,¹¹ and $(p, p'\gamma)_{g.s.}$ reactions leading to ^{40}Ca are compared in Fig. 2. The negative-parity states predicted by Gillet and Sanderson¹² are shown, too. Most of the experimental spin and parity attributions¹⁰ are tentative. It is evident from