Fission of Ra^{226} by 11-Mev Protons*

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The fission of Ra^{226} by 11-Mev protons has been studied by using radiochemical techniques. The mass-yield curve of fission products shows three humps: a narrow hump centered about mass 112, corresponding to a symmetric fission mode, with two humps on either side, corresponding to an asymmetric fission mode. The measured fission yields of fission fragments at the peaks of all three humps are about the same. The heavy-fragment "wing" of the asymmetric portion of the mass-yield curve is very similar in position and shape to the corresponding member of other asymmetric mass-yield curves. The light-fragment "wing" has accordingly shifted to lower mass numbers, continuing the trend observed in the asymmetric fission-yield curves of elements heavier than radium. The central, symmetric-fission hump is of the same width as the narrow, symmetric-fission curve observed for fission of bismuth by 20-Mev deuterons. The cross section for fission of Ra^{226} by 11-Mev protons is measured to be $2\pm1\times10^{-27}$ cm^2, and drops by a factor of about 60 when the proton energy is degraded to 9 Mev.

I. INTRODUCTION

TWO distinctly different types of mass division have been observed to occur in nuclear fission: asymmetric fission, where the fragments have a mass ratio of roughly two to three; and symmetric fission where the fragments have nearly the same mass. Asymmetric fission is characteristic of spontaneous and very low-energy fission of the heaviest elements (thorium and above),[1–9] while symmetric fission is observed to occur in bismuth[10] and lead[11] when these elements are bombarded with moderate energy particles (15 to 40 Mev).

When the heaviest elements are bombarded with moderate energy particles or photons the mass division becomes more complicated.[12–21] Up to about 40 Mev, asymmetric fission is the most probable fission mode. However, the probability of symmetric fission increases strongly with energy until it becomes the most probable fission mode at energies above 40 Mev. Hence at moderate energies both types of mass division are observed to occur in the heaviest elements, whereas in the lighter elements of bismuth and lead only symmetric fission is observed.

In view of the distinctly different mass distributions observed for thorium at moderate energies on the one hand and for bismuth and lead at moderate energies on the other, it was thought to be of interest to examine the mass distributions of elements having atomic numbers between bismuth and thorium. Unfortunately, because these elements are all radioactive with short half-lives for the most part, the only element in this region which could be obtained in sufficient quantity for such studies is radium. The isotope Ra^{226} is the only one commerically available in macroquantities. But having an atomic number of 2 units less and mass number of 6 units less than thorium it lies rather closer to the heaviest elements than to elements in the region of bismuth, and it would therefore be expected to have a fission-fragment mass distribution similar to thorium. Nevertheless it was decided to study its fission behavior on bombardment with 11-Mev protons.

Among the projectiles available from a cyclotron, 11-Mev protons make the best choice for several reasons. The proton flux available from the University of Washington 60-inch cyclotron is high (10^{14} protons/cm^2 sec), making it possible to use rather small amounts of somewhat hazardous and expensive material. Except for neutrons, which are unobtainable in high flux and

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[1] C. D. Coryell and N. Sugarman, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951); National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.

[2] W. E. Grummitt and G. Wilkenson, Nature **161**, 520 (1948).

[3] A. Turkevich and J. B. Niday, Phys. Rev. **84**, 52 (1951).

[4] Keller, Steinberg, and Glendenin, Phys. Rev. **94**, 969 (1954).

[5] E. P. Steinberg and L. E. Glendenin, Phys. Rev. **95**, 431 (1954).

[6] Harold G. Richter and Charles D. Coryell, Phys. Rev. **95**, 1550 (1954).

[7] E. P. Steinberg and L. E. Glendenin, J. Inorg. & Nuclear Chem. **1**, 45 (1955).

[8] J. G. Cuninghame, J. Inorg. Nuc. Chem. **4**, 1 (1957).

[9] L. E. Glendenin and E. P. Steinberg, J. Inorg. & Nuclear Chem. **1**, 45 (1955).

[10] A. W. Fairhall, Phys. Rev. **102**, 1335 (1956).

[11] E. F. Nuezil and A. W. Fairhall (to be published).

[12] A review of work prior to 1952 is given by R. W. Spence and G. P. Ford [*Annual Review of Nuclear Science* (Annual Reviews, Inc., Stanford, 1953), Vol. 2, p. 399].

[13] Howard A. Tews and Ralph A. James, Phys. Rev. **88**, 860 (1952).

[14] Turkevich, Niday, and Tompkins, Phys. Rev. **89**, 552 (1953).

[15] J. S. Wahl, Phys. Rev. **95**, 126 (1954).

[16] Roman A. Schmitt and Nathan Sugarman, Phys. Rev. **95**, 1260 (1954).

[17] Jones, Timmick, Paehler, and Handley, Phys. Rev. **99**, 184 (1954).

[18] Glass, Carr, Cobble, and Seaborg, Phys. Rev. **104**, 434 (1956).

[19] W. M. Gibson, University of California Radiation Laboratory Report UCRL-3493, November, 1956 (unpublished).

[20] Katz, Kavanagh, Cameron, Bailey, and Spinks, Phys. Rev. **99**, 98 (1955).

[21] Sugihara, Drevinsky, Troianello, and Alexander (to be published).

of homogeneous energy from a cyclotron, protons have the lowest Z, thus keeping the atomic number of the compound nucleus as small as possible. Unlike deuterons which are likely to be stripped, giving rise to a large number of different compound-nuclear states, protons have the advantage that one may more safely infer the formation of a single compound nucleus. Furthermore, at the relatively low bombarding energy of 11 Mev only two fissioning species would be expected: Ac^{227} excited to 16 Mev from the capture of an 11-Mev proton in Ra^{226}, and Ac^{226} excited to less than 9 Mev from the $Ra^{226}(p,n)$ reaction. Finally, unlike deuterons, 11-Mev protons give rise to relatively few secondary neutrons. Hence neutron-induced reactions will contribute negligibly to the observed fission fragments.

The fission of radium by helium ions of 20 and 42 Mev and by 9- to 22-Mev deuterons is also being investigated. The results of these experiments will be published at a later date.

II. EXPERIMENTAL PROCEDURE

Radium was obtained commercially in ampoules containing approximately 1 mg of radium as the bromide salt. Because of the extensive physical and chemical processing used in its recovery, the radium was assumed to be free of thorium and uranium. No attempt was made to purify it further.

This assumption was checked experimentally by spiking 20 g of barium bromide with 3 g each of thorium nitrate and uranium nitrate. After only 12 fractional crystallizations of barium bromide an aliquot of the purified material was bombarded with 11-Mev protons and a search was made for silver, molybdenum, and strontium fission products. No fission-product activity was found in the sample, indicating that thorium or uranium impurity in the radium target would be very unlikely.

To lessen the hazard of using radium, and to considerably lessen the cost of extensive experimentation, the radium, prepared in the form of radium carbonate, was sandwiched between two thin gold foils. This radium sandwich was bombarded between thin aluminum foils which served to catch fission fragments recoiling out of the gold-radium targets. By processing only the catcher foils, chemical separation could be performed that was relatively free from complications that would arise if radium were present. Furthermore, the radium sandwich could be used over again.

The radium sandwich was prepared by precipitating about 1 mg of radium as the carbonate, and washing the precipitate with water to remove unwanted salts. A slurry of the precipitate in alcohol was then allowed to settle uniformly by sedimentation onto a thin gold foil of thickness 2.7 mg/cm^2. The precipitate was confined to a definite area by means of a rectangular hole in an aluminum block which was clamped tightly against the gold foil. After the alcohol had evaporated the block was removed and another sheet of cohesive gold foil was laid over the radium. The foils were then placed between two smooth blocks of aluminum and the whole assembly compressed in a hydraulic press to a pressure of 1000 psi. In this way the two gold foils, with the radium carbonate between, were compacted into a single foil.

Because of the thickness of the radium deposit (about 0.5 mg/cm^2) and the overlapping gold, the fission fragments emitted at oblique angles to the foil normal could not escape from the target. Since they have shorter ranges, fewer heavy fragments would be able to escape compared with complementary light fragments. Assuming the fission fragments are emitted isotropically, and using the measured values for the ranges in gold[22] and aluminum[23] of fission fragments from thermal-neutron-induced fission of uranium, it was computed that for the most extreme asymmetric pair of fragments about 20% fewer heavy fragments would be collected than complementary light fragments. Correction factors to compensate for the effect of source thickness were computed and applied to the measured yields. Perturbations in these correction factors due to nonisotropic fission are expected to be completely negligible in view of the very small angular anisotropies observed for fission products in 11-Mev proton-induced fission of thorium and uranium.[24]

For bombardment the radium sandwich was placed between two hyperpure aluminum catcher foils[25] and fastened tightly to a water-cooled pure aluminum target plate. The target was then bombarded for either one or two hours with a proton flux of 10^{14} particles/cm^2 sec.

After bombardment the catcher foil was dissolved in an acid or basic solution containing carrier amounts, usually 10 mg, of the fission product elements to be separated. The type of solvent used depended on the elements to be separated. The strong reducing properties of the aluminum catcher foil made carrier exchange difficult and extreme oxidizing conditions were required. When solutions of sodium hydroxide or sulfuric acid were used to dissolve the catcher foils the results were not reproducible, presumably owing to difficulties of carrier-tracer exchange of the silver monitoring species. The observation that the ratio of the yields of Ag^{111} to Ag^{113} was not reproducible in those cases is evidence that this explanation is the correct one. A strong oxidizing solution, such as HCl and HNO_3, NaOH and NaClO, or H_2SO_4 and $HClO_4$, and a half-hour reflux period was found to give reproducible results.

Ag^{111}, which was separated in every experiment,

[22] E. Segrè and C. Wiegand, Phys. Rev. **70**, 808 (1946).

[23] Finkle, Hoagland, Katcoff, and Sugarman, *Radiochemical Studies: The Fission Products*, (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 46, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.

[24] C. T. Coffin, Department of Physics, University of Washington, Ph.D. thesis, 1956 (to be published).

[25] We are very grateful to Mr. J. A. Nock, Jr., of the Aluminum Research Laboratories, Aluminum Company of America, for supplying the very high purity (99.99%) aluminum used.

served as an internal standard to which the fission yields of the other species were normalized. The separation and purification of each element followed well known chemical procedures,[26–29] modified slightly to allow for the presence of aluminum, which was sometimes annoying and generally lowered the chemical yields.

Each purified element was precipitated in some suitable form, filtered onto a small filter paper, dried, and weighed to determine chemical yield. The sample was mounted on Scotch tape covering a one-inch hole in a sample mounting card, and covered with thin (2 mg/cm^2) polystyrene foil. Each sample was counted in one of a set of four end-window G-M counters with nearly identical shelf assemblies. The detection efficiencies between counters were cross-calibrated with a Pb^{210} standard. The decay curves were plotted and analyzed in the usual way, and saturation counting rates were determined for each radioactive species. The half-lives of the various components of the decay curves always agreed closely with the values for known fission product species of the elements being counted.[30] Corrections for absorption of the radiations by air, polystyrene, and counter window; and for scattering by Scotch tape, filter paper, polystyrene, air, and counter housing were made from data of Pappas.[31] Sample absorption and scattering corrections were made according to the curves of Nervik and Stevenson.[32] Since no absorption curves were measured, tabulated energy values for the radioactive species[30] were used in making counting corrections.

For the purpose of computing a cross section for fission, the geometry factors of the several shelves were determined using a sample of known disintegration rate. This standard was obtained by chemically separating in a quantitative manner the Th^{234} daughter of an aged uranium solution of known concentration.

III. EXPERIMENTAL RESULTS

The fission yields measured for the 20 nuclides which have been studied so far are given in Table I. The number of measurements of each yield is given in column 1. Most of the measurements have been made on those elements which represent symmetric fission. The tabulated errors are the standard deviations of separate measurements. The values for Y^{93} and Ba^{139}, which have been measured only once, are probably reliable within 20%. Pd^{112} was determined both by milking the Ag^{112} daughter and by analyzing the total decay curve for Pd^{109} and Pd^{112}. Because of the uncertain correction factors for the low-energy Pd^{112}, the milking procedure is probably the more accurate.

All yields are relative to Ag^{111}, which was measured in each experiment. As is customary with fission yield measurements, the yields have been normalized to give a total mass yield of 200%. The tabulated values represent cumulative yields of each mass chain up to the species listed. Species independently formed closer to stability are not included. Calculations of independent fission yields on the basis of the equal-chain-length hypothesis,[33] the unchanged charge distribution hypothesis, and the hypothesis of maximum energy release[34] indicate that the yields of the more asymmetric fission products which were measured might be somewhat low. However, none of these postulated charge distributions indicate significant changes in the measured yields for fragments having mass ratios up to a value of about 1.3.

Any fission of the gold foil or activation of impurities in the aluminum catcher foil and gold sandwich might affect the results. Of the species tabulated in Table I, only Br^{83} could be expected to be formed by proton-activation of an impurity (selenium). The aluminum foil used as the fission fragment catcher had previously been shown[10] to be of extremely high purity, so that only the gold could be expected to be the source of

TABLE I. Relative yields of fission fragments of radium irradiated with 11-Mev protons.

Number of determinations	Nuclide	Fission yield (%)
2	Br^{83}	3.1±0.2
2	Br^{84}	2.9±0.2
6	Sr^{91}	3.5±0.4
6	Sr^{92}	3.0±0.4
1	Y^{93}	3.4
4	Zr^{97}	1.9±0.3
15	Mo^{99}	1.9±0.2
2	Ru^{105}	2.9±0.4
4	Pd^{109}	3.6±0.4
25	Ag^{111}	4.0[a]
4	Pd^{112}	3.3±0.4[b]
4	Pd^{112}	3.8±0.3[c]
25	Ag^{113}	3.1±0.1
3	Cd^{115}	3.9±0.6
3	Sn^{121}	2.0±0.2
3	Sn^{125}	0.9±0.1
4	Te^{132}	2.8±0.5[d]
2	I^{134}	4.7±0.4
1	Ba^{139}	4.0
2	Ba^{140}	2.8±0.2[e]
2	Ce^{133}	1.9±0.2

[a] The fission yields of the other fission products are measured relative to this nuclide. The fission yields of all mass numbers integrate to 200%.
[b] By analysis of the total palladium decay curve for Pd^{109} and Pb^{112}.
[c] By milking Ag^{112} daughter from Pd^{112}.
[d] By milking I^{132} daughter from Te^{132}.
[e] By milking La^{140} daughter from Ba^{140}.

[26] See reference 1, part VI.
[27] Jacob Kleinberg, Los Alamos Scientific Laboratory of the University of California Report LA-1566, 1953 (unpublished).
[28] W. Wayne Meinke, Atomic Energy Commission Report AEC-2738 (UCRL-432), 1949 (unpublished).
[29] Hillebrand, Lundell, Bright, and Hoffman, *Applied Inorganic Analysis* (John Wiley and Sons, Inc., New York, 1953).
[30] *Nuclear Data Cards* (Nuclear Data Group, National Research Council, Washington 25, D. C.).
[31] A. C. Pappas, Massachusetts Institute of Technology Technical Report No. 63, 1953 (unpublished).
[32] W. Nervik and P. Stevenson, Nucleonics **10**, No. 3, 18, (1952).
[33] Glendenin, Coryell, and Edwards, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 52, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.
[34] K. Way and E. P. Wigner, Phys. Rev. **73**, 1318 (1948).

contamination of the fission product species. The most likely impurities in the gold would be copper and zinc, with silver and the platinum metals being possible impurities. In an exploratory run to test for impurities a stack of gold and aluminum foils was bombarded with 11-Mev protons, and silver, zinc, gallium, and palladium were separated radiochemically. No silver or palladium activities were found, but appreciable zinc and gallium activities were observed that would mask the presence of any fission activities of these two elements. These activities are probably due to (p,n) reactions on copper and zinc impurities in the gold foil.

The data of Table I are plotted in Fig. 1. In Fig. 2 they are plotted along with complementary fragments, assuming a total mass of 227 and the loss of either 3 or 5 neutrons during the course of the reaction. With the exception of 3 points the data show a smooth pattern, and a value of between 3 and 5 would seem to fit the data fairly well.

The low yield of Sn^{125} relative to neighboring mass numbers is attributed to the independent formation of the 9.5-minute isomer, which is not measured. The yield of Br^{84} appears to be too low by a factor of about 20%. This may be due to the short half-life of Br^{84}, which results in rather poor resolution of this species from the decay curve. The yield of Ag^{113} was consistently lower than the yields of neighboring species. It is doubtful that experimental error could account for this low yield. A short-lived isomer of this nuclide is known to appear in thermal-neutron fission of uranium.[35] This species would not have been detected in these experiments and presumably accounts for the difference.

Radium was observed to contaminate the barium fraction, and the measured value of Ba^{139} is therefore somewhat uncertain. Iodine was separated before complete decay of the 44-minute Te^{134} precursor. Corrections were made for this assuming that all the I^{134} came from Te^{134}. This may not be the case, so that the tabulated yield may be high.

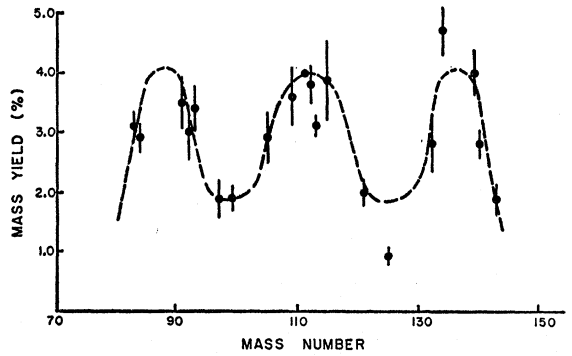

FIG. 1. Fission yields of 20 radionuclides in the fission of Ra^{226} induced by 11-Mev protons.

[35] U. Schindewolf and C. D. Coryell, Massachusetts Institute of Technology Laboratory of Nuclear Science Annual Progress Report, June, 1955 to May, 1956 (unpublished) p. 35.

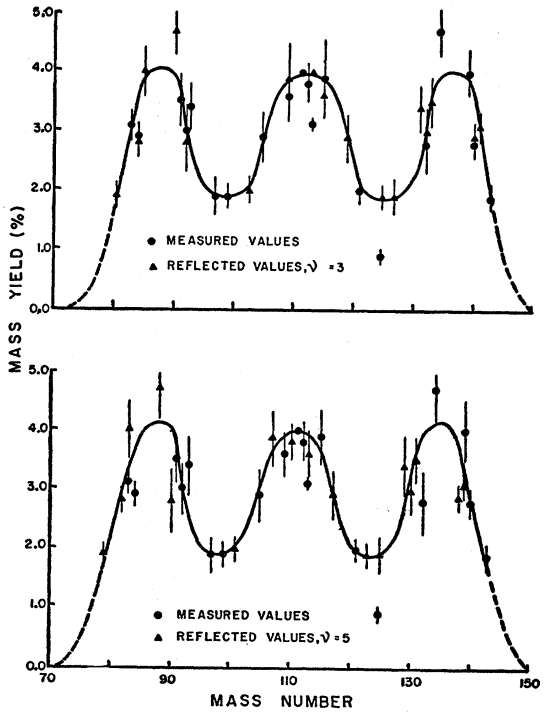

FIG. 2. Fission yield curves for 11-Mev proton-induced fission of Ra^{226}. Data of Table I plotted with reflected points on the basis of $\nu=3$ and $\nu=5$.

IV. CROSS SECTION FOR FISSION

The cross section for fission with 11-Mev protons is calculated from the data to be 2 ± 1 millibarns. When the protons were degraded to 9 Mev it was found that the cross section dropped by a factor of 60. This shows, among other things, that contamination of the proton beam by 22-Mev deuterons or 44-Mev helium ions is less than 1%. It also shows that the fission product species reported here cannot arise from fission of thorium or uranium impurities present in the radium target. The cross section for the fission of thorium by 11-Mev protons is 50 millibarns, and drops by a factor of about 2 for fission by 9-Mev protons. The cross section for proton fission of uranium would be expected to change with energy in a similar manner. The rapid decrease in the cross section observed for radium indicates that less than a few percent of the asymmetric fission products observed for radium with 11-Mev protons could be due to thorium or uranium impurities.

V. DISCUSSION

The most striking feature of the results displayed in Figs. 1 and 2 is the triple-humped nature of the mass-yield curve. The fission of radium with 11-Mev protons exhibits two distinctly different fission modes: typically asymmetric fission, and symmetric fission with a narrow range of mass values. It was suggested[3] some years ago that it was meaningful to discuss the mass distribution in terms of two modes, "symmetric" and

"asymmetric." This division was based largely on the observation that symmetric fission becomes increasingly probable in the heavy elements as the energy of the fission-inducing particle is raised. The present results exhibit the two modes separately for the first time.

The present results differ from the earlier ones in at least two respects. First of all, the symmetric fission mode of radium stands out clearly by itself, whereas for the heavier elements asymmetric fission is still the predominant fission mode. Only recently has evidence been obtained that a small peak occurs at the symmetric point in the fission of U^{238} by 31-Mev bremsstrahlung.[36]

Secondly, there is considerably less asymmetric fission relative to symmetric fission in radium than would be expected by extrapolation of the systematic behavior observed for elements heavier than radium. On the basis of the correlations pointed out by Fowler, Jones, and Paehler,[37] the fission yield of Ba^{140} would be expected to be at least five times as large as Ag^{111}, whereas they have nearly the same fission yield.

It is of interest to compare the two fission modes observed for radium with the symmetric mode observed for bismuth at moderate bombarding energies, and the asymmetric mode observed for low-energy fission of heavier elements. The central peak of the radium mass yield curve is quantitatively similar to the narrow peak observed for bismuth, up to the point of its merger with the asymmetric part of the curve. The width of the symmetric fission curve at half the maximum yield is only 17 mass units for both radium and bismuth.

The heavy "wing" of the asymmetric fission mode observed for radium fission is very similar to those observed in the asymmetric fission-yield curves of other elements. Within experimental error it is of the same width at half the maximum yield as the curve for thermal-neutron-induced fission of uranium; and it is of interest that the position of the peak occurs in the same mass region as it is observed to occur for the other elements which undergo asymmetric fission. Thus, the observation that the location of the heavy-fragment "wing" stays fixed and the light-fragment "wing" shifts compensatingly as one goes to lighter fissioning elements also applies to the asymmetric fission of radium. It is probably this fact that has permitted the observation of the central hump: the "wings" have moved sufficiently far apart to allow dips to appear between the central hump and the insides of the asymmetric "wings." The correlation of the degree of separation of the asymmetric peaks with Z^2/A of the compound nucleus is in agreement with the empirical equation of Swiatecki.[38]

The picture presented by the fission product mass distribution of all of the elements heavier than lead is as follows: symmetric fission is common to all of them when excitation energies are moderate. So far only the heaviest elements, radium and above, are observed to exhibit asymmetric fission at moderate energies; and for these elements the ease with which they undergo fission at low energy decreases as one goes to species of lower atomic number.

The probability of fission taking place at low excitation energies is presumably related in some way to the fissionability parameter Z^2/A, to nuclear excitation energy, and to neutron binding energies. Where Z^2/A is unfavorably small and neutron binding energy low, low-energy fission would not be expected to compete favorably with neutron emission. The rapid increase in fissionability with the energy deposited in the compound nucleus which was observed for bismuth, along with its symmetric mass distribution suggests as one possibility that this mode of fission may be associated with prompt fission at higher excitation energy before the loss of many neutrons cools off the nucleus. The central peak in the radium mass-yield curve could on this basis be ascribed to the fission of Ac^{227} excited to 16 Mev formed by capture of an 11-Mev proton in Ra^{226}. The asymmetric fission of radium would be ascribed to fission taking place in Ac^{227} excited to 16 Mev in competition with symmetric fission, and in addition, to the fission of Ac^{226} excited to 7 or 8 Mev following the evaporation of a neutron from Ac^{227}. This latter species would be expected to show only the asymmetric fission behavior common to the heaviest elements at comparable excitation energies.

VI. SUMMARY

Moderate-energy fission of radium exhibits two distinctly different mass divisions corresponding to separate asymmetric and symmetric fission modes. The occurrence together of two such different fission modes has been inferred to be the case; the present results confirm this hypothesis. The symmetric-fission mode shows the same type of mass distribution as is observed for bismuth at comparable excitation energies. The asymmetric-fission mode continues a trend already observed for elements heavier than radium: the mass distribution of the heavy fragments is more or less the same as is observed for all the heavy elements, and the complementary light-fragment distribution has shifts to smaller mass numbers.

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[36] A. C. Pappas (private communication).
[37] Fowler, Jones, and Paehler, Phys. Rev. **88**, 71 (1952).
[38] W. J. Swiatecki, Phys. Rev. **100**, 936 (1955).