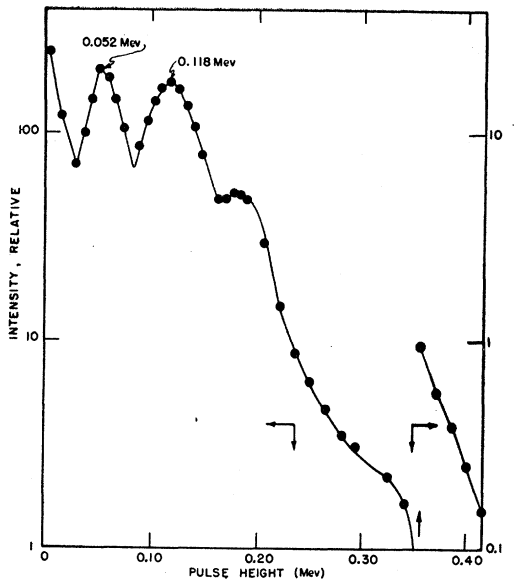

FIG. 3. Gamma-ray spectrum for Yb^{167}.

the gross decay curve obtained before separation. The products were examined specifically for such a half-life by using different counting techniques and by following the decay of various points on the spectrum. If the activity does have a mass of 167, it should have been produced in this bombardment.

For further proof of the mass assignment of the 18.5-minute Yb^{167}, an excitation function, Fig. 2, was determined for production of the activity. This shows a threshold of 19.0±0.5 Mev, which indicates a $(p,3n)$ reaction and, of course, assigns a mass of 167 to the activity.

The gamma-ray spectrum, as measured with a NaI(Tl) scintillation spectrometer, is shown in Fig. 3. A gamma ray of 0.118-Mev energy is apparent, also possible low intensity peaks near 0.18 and 0.33 Mev. The 0.052-Mev peak is the x-ray peak. The 0.51-Mev annihilation peak was not observed; if positrons are present, they constitute a very small fraction of the total radiation.

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Yields of Fission Products from U^{238} Irradiated with Fission Spectrum Neutrons

R. N. KELLER,* E. P. STEINBERG, AND L. E. GLENDENIN
Chemistry Division, Argonne National Laboratory, Lemont, Illinois
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A radiochemical investigation of the fission yields of 15 products in the fission of U^{238} is described. The average energy of the neutrons effective in inducing fission is estimated as 2.8 Mev. The familiar twin-peaked yield-mass distribution is observed with a peak-to-trough ratio of 200 to 1. A comparison of the fission yield-mass curve for U^{238} with those for other fissile nuclei is made.

I. INTRODUCTION

RADIOCHEMICAL determinations of the yields of fission products from the fission of various fissile nuclei have indicated that, in general, asymmetric fission results at low energies[1,2] with the contribution from symmetrical modes ("trough"), and perhaps, also very asymmetric modes ("wings"), increasing as the energy content of the compound nucleus is increased.[3–9] With increasing mass of the fissioning nucleus, the light group distribution shifts toward higher masses, while the heavy group remains relatively constant, with perhaps a slight shift in the direction of smaller masses.[3] A tendency toward wider mass distribution with increasing mass of the fissile nucleus has also been noted.[3,10]

Studies have been made of the fission of Th^{232} [10] and U^{238} [11] with fission spectrum (fast) neutrons to extend the knowledge of the characteristics of fission mentioned above. In the earlier U^{238} investigation[11] samples of of U_3O_8 powder, depleted seventeenfold in U^{235}, and normal uranium metal disks were irradiated in a cadmium can placed inside a hollow uranium cylinder. The U_3O_8 powder became contaminated with Cd^{115} in the process of opening the activated cadmium container making it impossible to determine directly in the depleted uranium the fission yield of Cd^{115} (which essentially establishes the trough yield). An attempt was therefore made to calculate this yield from the

* On leave from the University of Michigan. Present address: Department of Chemistry, University of Colorado, Boulder, Colorado.

[1] *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. Grummit and G. Wilkinson, Nature **161**, 520 (1948).

[3] E. P. Steinberg and M. S. Freedman, reference 1, Paper No. 219.

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[5] A. S. Newton, Phys. Rev. **75**, 17 (1949).

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[7] Fowler, Jones, and Paehler, Phys. Rev. **88**, 71 (1952).

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

[9] H. A. Tewes and R. A. James, Phys. Rev. **88**, 860 (1952).

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

[11] Engelkemeir, Seiler, Steinberg, and Winsberg, reference 1, Paper No. 218.

observed Cd^{115} yield in the normal uranium. This calculation involved a correction for the Cd^{115} contributed by the U^{235} in normal uranium based on the known fission yield of Cd^{115} in thermal neutron fission of U^{235}. Since the Cd^{115} yield in fast neutron fission of U^{235} was not known, the correction was possibly in error. Moreover, the heavy mass peak was not well defined.

It seemed advisable to reexamine the fast neutron fission of U^{238} radiochemically with particular attention to the yield of Cd^{115} and yields in the region of the heavy mass peak. The accuracy of the fission yield determinations was also improved by the application of somewhat better corrections to the observed beta counting rates for absorption and scattering in the sources.

II. EXPERIMENTAL

A. General Procedure

Samples of U_3O_8 depleted in U^{235} (about 0.02 percent U^{235}) were sealed in cadmium-lined aluminum capsules and irradiated in a hollow uranium rod in the Oak Ridge pile. This arrangement enables irradiation with nearly unmoderated fission spectrum neutrons. The average neutron energy effecting fission in U^{238}, which has a fission threshold of about 1 Mev, is estimated to be approximately 2.8 Mev. Efficient filtering of neutrons of thermal energy was accomplished by the cadmium, minimizing the contribution of U^{235} to the total number of fissions. This contribution was shown to be less than 1.5 percent by comparison with normal uranium samples (0.7 percent U^{235}) irradiated under the same conditions. Several irradiations were carried out varying in duration to suit the half-life ranges of the nuclides under investigation. After irradiation and removal from the pile, the U_3O_8 was dissolved in a minimum quantity of $6N$ HNO_3 and the resulting solution diluted to known volume. This solution was then used for all subsequent aliquots and dilutions.

The fission products were isolated from the active solutions by adding a known amount of isotopic carrier, subjecting it to chemical separations which are specific for the element, and recovering the carrier in a chemically pure and weighable form suitable for counting. Care was exercised that interchange between the carrier and the fission-produced species was effected. If this condition is realized, the chemical yield of the recovered carrier, which need not be quantitative, also can be taken to be equal to the radiochemical yield of the fission product. The aliquot used and the amount of carrier added were adjusted to the chemical yield of the particular procedure and to the approximate fission yield of the isotope so that the recovered carrier in the form of a compound suitable for mounting weighed approximately 20 mg and had a counting rate of approximately 5000 counts per minute. In general, the radiochemical procedures used for the isolation of the fission-produced elements are those given in reference 1, Part VI, with minor modifications. The final precipitate in each case was collected on a 1.75-cm^2 circle of filter paper, mounted at the center of a 2.5-in. $\times$3.25-in. cardboard support, covered with thin Cellophane, and its counting rate measured with an end-window Geiger counter. A standard beta source was used to monitor any changes in the characteristics of the counter. The radiochemical purity of each fission product was determined by its absorption and decay characteristics.

The observed counting rates were corrected for aliquot of fission solution used, chemical yield, decay from the end of the irradiation to the time of counting, lack of saturation during irradiation, the presence of other activities due to genetically related or isotopic species, and resolution losses in the counting apparatus. Empirical self-absorption and self-scattering factors[12] were used to correct for the effects of sample weight, and an exponential absorption correction was made for absorption in the Cellophane covering over the sample, in the air layer between the source and the counter, and in the window of the counting tube. Inasmuch as all samples, including the beta standard, were counted with identical geometry, no correction for counting geometry was required.

B. Individual Determinations

Arsenic (40-hr As^{77})

The radiochemical procedure of Winsberg[13] was modified to include a preliminary reduction of As(V) to As(III) with iodide ion and subsequent oxidation back to As(V) with concentrated HNO_3 to insure interchange between the carrier and the fission-produced species.

According to the genetic relations of the chain of mass 77 the total fission yield of arsenic includes contributions from two Ge^{77} precursors, a 59-second isomer and a 12-hour isomer. In the calculations of the fission yield the assumption was made that the fraction of the total As^{77} that is formed by decay of the 59-second isomer of Ge^{77} is 55 percent and the fraction formed by decay of the 12-hour isomer is 45 percent as has been observed in the fission of U^{233}, U^{235}, and Th^{232}. The aluminum absorption curve for the arsenic activity obtained in this work showed a hard component, apparently due to 26.8-hr As^{76} formed by an (n,γ) process on minute traces of arsenic imputities in the U_3O_8. A correction was made for the contribution of this contaminant to the total observed activity.

Strontium (53-day Sr^{89})

The procedure used for isolating strontium was essentially that of Glendenin.[14]

The determination of Sr^{89} was straightforward since

[12] Engelkemeir, Seiler, Steinberg, and Winsberg, reference 1, Paper No. 4.
[13] Reference 1, Paper No. 228.
[14] Reference 1, Paper No. 236; also E. Hoagland, Paper No. 237.

complete decay of the 9.7-hr Sr^{91} had taken place before the radiochemical separations were made and the activity due to 20-yr Sr^{90} was negligible.

Zirconium (65-day Zr^{95})

The radiochemical procedure of Hume[15] was followed.

Determinations of Zr^{95} were made after allowing sufficient time for the complete decay of 17-hr Zr^{97}, and counting of samples was done before appreciable growth of the 90-hr Nb^{95m} and 35-day Nb^{95} daughters could occur.

Molybdenum (65-hr Mo^{99})

The radiochemical procedure of Ballou[16] was followed, with the exception that molybdenum was finally precipitated as $PbMoO_4$ rather than Ag_2MoO_4.

The 67-hr Mo^{99} decays by beta emission to 5.9-hr Tc^{99m}, which in turn decays by isomeric transition to the ground state, 2×10^5-yr Tc^{99}. Samples of 67-hr Mo^{99} were consequently counted through 15 mg/cm^2 of aluminum to filter out the low-energy conversion electrons from 5.9-hr Tc^{99m}. Allowance for this additional absorber was then made in the absorption correction.

Ruthenium (42-day Ru^{103} and 1.0-yr Ru^{106})

The radiochemical procedure of Glendenin[17] was used.

The isolated ruthenium contained activities from 40-day Ru^{103}, 1.0-yr Ru^{106}, and 30-sec Rh^{106}. The relative contributions from the two ruthenium isotopes were evaluated by analysis of an absorption curve.

Silver (7.6-day Ag^{111})

Ag^{111} activity was isolated by the procedure of Glendenin.[18] No other long-lived silver activities are produced in fission and no attempt was made to determine the fission yields of shorter-lived isotopes.

Cadmium (2.33-day Cd^{115} and 43-day Cd^{115})

The procedure of Metcalf[19] was used except that the cadmium was mounted for counting as CdS rather than $CdNH_4PO_4\cdot H_2O$.

The cadmium activity obtained was composed of 2.33-day Cd^{115} and 43-day Cd^{115}. The fraction due to the longer-lived isomer, which decays by beta emission to stable In^{115}, was evaluated by an analysis of the decay curves. The 2.33-day Cd^{115} decays to 4.53-hr In^{115} which in turn decays partly to stable In^{115} by isomeric transition and partly by beta emission to stable Sn^{115}. Counting of the samples was delayed until transient equilibrium was reached between 2.33-day Cd^{115} and 4.53-hr In^{115}. The contribution of the 2.33-day Cd^{115} to the total activity was determined from the equations of radioactive growth and decay and the known decay schemes.[20]

Antimony (93-hr Sb^{127})

The procedure used for isolating antimony activity was that of Stanley and Glendenin[21] with final precipitation as pyrogallate after Boldridge and Hume.[22]

The 93-hr Sb^{127} decays to two isomers of Te^{127}, a 90-day upper state and a 9.3-hr ground state with branching of 16 percent and 84 percent, respectively.[23] The 90-day isomer decays by isomeric transition to the 9.3-hr isomer, which in turn decays by beta emission to stable I^{127}. The Sb^{127} was determined after decay of the shorter-lived antimony isotopes by counting the transient equilibrium mixture of 93-hr Sb^{127} and 9.3-hr Te^{127}. The contribution of 93-hr Sb^{127} to the observed activity could then be calculated from the growth and decay relations of parent and daughter. Longer-lived antimony activities were present in negligible intensities.

Tellurium (77-hr Te^{132})

The radiochemical procedure of Glendenin[24] was followed. The method for determining 77-hr Te^{132} activity in the presence of the other tellurium activities from fission involved isolating samples of elementary tellurium of known weight from the fission solution, dissolving these and diluting to known volume, allowing sufficient time for transient equilibrium to be reached between 77-hr Te^{132} and its daughter, 2.4-hr I^{132}, and then analyzing for 2.4-hr I^{132} in aliquots of these solutions.[25] Once the 2.4-hr I^{132} activity in equilibrium with 77-hr Te^{132} is determined, the parent-daughter relations allow calculation of the activity due to 77-hr Te^{132}.

Cesium (33-yr Cs^{137})

The procedure for isolating this activity was essentially that of Glendenin and Nelson.[26]

The 33-yr Cs^{137} decays to stable Ba^{137}, partly by the direct route and partly through 2.6-min Ba^{137m}. The branching ratio for these two paths was taken as 8 percent and 92 percent, respectively, and the conversion in Ba^{137m} was taken as 10 percent.[20]

Barium (12.8-day Ba^{140})

The method for isolating this activity combined the procedures outlined by Glendenin for separating

[15] Reference 1, Paper No. 245.
[16] Reference 1, Paper No. 257.
[17] Reference 1, Paper No. 260.
[18] Reference 1, Paper No. 267.
[19] Reference 1, Paper No. 268.
[20] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).
[21] Reference 1, Paper 271.
[22] Reference 1, Paper 272.
[23] J. Beydon, Compt. rend. **227**, 1159 (1948).
[24] Reference 1, Paper 274.
[25] Reference 1, Paper 278.
[26] Reference 1, Paper 283.

strontium and barium activities together[14] and that for separating barium alone.[27]

The 12.8-day Ba^{140} decays by beta emission to 40-hr La^{140} which in turn decays to stable Ce^{140}. For several hours after the removal of 40-hr La^{140} from 12.8-day Ba^{140} the growth of activity due to the former is essentially linear with time. The measurements of the beta activity of the barium samples were made as soon as possible after the last precipitation, and the observed beta activity was corrected to zero time for the 40-hr La^{140} growth by the relation $A_0=A_t/(1+0.015t)$, where A_0 is the original activity and A_t is the activity after t hours.

Cerium (282-day Ce^{144})

The procedure used was essentially that of Boldridge and Hume.[28]

The isolated cerium contained activities from 30-day Ce^{141}, 282-day Ce^{144}, and 17.5-min Pr^{144}. The contribution of Ce^{144} to the total activity was determined by analysis of an absorption curve after the 17.5-min Pr^{144} had reached secular equilibrium.

Europium (15.4-day Eu^{156})

A modification of the procedure by Winsberg[29] was used for separating europium activity. Barium and strontium sulfate precipitations were carried out in the presence of europium carrier prior to the initial EuF_3 precipitation of Winsberg's method and the europium was finally mounted as oxalate. The 15.4-day Eu^{156} was determined in the presence of 1.7-yr Eu^{155} (in low intensity) by analysis of absorption curves after the shorter-lived isotopes had decayed.

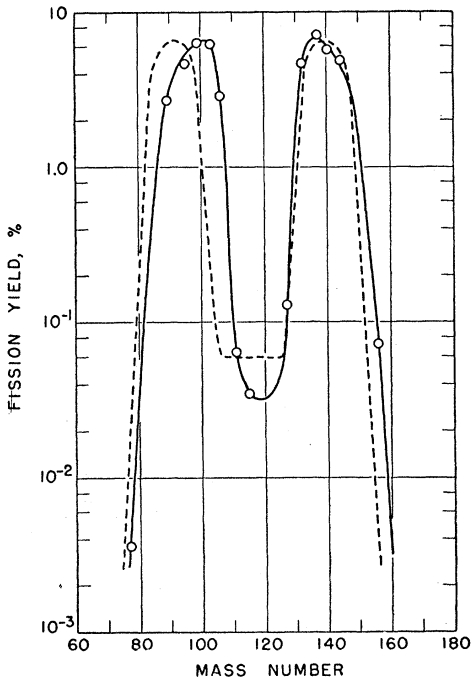

FIG. 1. Yield-mass curve for fast-neutron-induced fission of U^{238} (solid line), compared with that for Th^{232} (dotted line).

[27] Reference 1, Paper 288.
[28] Reference 1, Paper 294.
[29] Reference 1, Paper 303.

III. RESULTS

The fission yield of a nuclear species may be defined as the percentage of fissions which eventually result in the formation of the particular species. Since two fragments are formed per fission, the sum of the fission yields for all mass numbers is 200 percent. If the absolute fission yield for one nuclide has been determined, this can be used as a fission yield reference standard and the yields of other nuclides determined relative to it. Alternatively, an arbitrary value for the fission yield of one nuclide may be selected, the fission yields of other nuclides determined relative to this, and finally the resulting yield-mass curve normalized to 200 percent. The latter method has been employed in the present case. The reference nuclide used was 12.8-day Ba^{140}. The fission yield Y_A of any species A can be determined relative to the arbitrarily chosen value, $Y_{Ba^{140}}$, for 12.8-day Ba^{140} by the relation

$$Y_A=\frac{S_A}{S_{Ba^{140}}}Y_{Ba^{140}},$$

where S_A and $S_{Ba^{140}}$ are the saturation activities of A and Ba^{140} in a given irradiation.

The relative yields of fifteen nuclides obtained by the normalization method indicated above are given with estimates of reliability in Table I, column 3. The data from the earlier work of Engelkemeir *et al.*,[11] normalized to a value of 5.7 percent for Ba^{140}, are given in column 4 for comparison. The fission yields from column 3 are plotted against mass number in Fig. 1 and a smooth yield-mass curve (solid line) has been drawn through

TABLE I. U^{238} fission yields.

Fission product	Mass number	Fission yield (%) Present investigation	Reference 11
40-hr As	77	0.0036±0.001	0.0040
53-day Sr	89	2.7 ±0.3	3.1
65-day Zr	95	4.7 ±0.7	6.8
67-hr Mo	99	6.4 ±0.7	5.5
42-day Ru	103	6.3 ±1.0	6.9
1.0-yr Ru	106	2.9 ±0.3	2.5
7.6-day Ag	111	0.064 ±0.006	0.067
2.33-day Cd	115	0.032 ±0.006	0.053
43-day Cd	115	0.0025±0.0003	0.006
Total chain	115	0.035 ±0.007	0.059
93-hr Sb	127	0.13 ±0.03	0.11
77-hr Te	132	4.7 ±0.7	...
33-yr Cs	137	7.1 ±0.7	...
12.8-day Ba	140	5.7[a]	5.7[a]
282-day Ce	144	4.9 ±0.5	...
15.4-day Eu	156	0.073 ±0.01	0.059

[a] Ba^{140} was used as a fission yield standard for each irradiation.

them symmetrically about mass 118.5 (i.e., assuming binary fission with the emission of two neutrons per fission). The yield summation under this curve is 200 percent, and since the nuclides chosen for analysis are close to the stable ends of the fission chains, the curve should represent total chain yields.[30]

IV. DISCUSSION

The yield-mass curve for the fission of U^{238} with fission spectrum neutrons exhibits the familiar double-maxima and is in essential agreement with that observed previously[11] except for the yield of the trough (i.e., Cd^{115}). As mentioned above (Sec. I), the yield of Cd^{115} in U^{238} fission in the previous work was calculated from data on normal uranium assuming the yield of Cd^{115} in thermal neutron fission of U^{235} (0.011 percent) in the correction for the U^{235} contribution. It is now known that in the fission of U^{235} with fission spectrum neutrons the yield of Cd^{115} rises to a value of about 0.04 percent.[31] If this value is used to correct the old data, a yield of 0.04 percent is calculated for U^{238} fission, in good agreement with the result obtained in the present work using uranium depleted in U^{235}.

A comparison of the yield-mass curves of U^{238} and Th^{232} [10] is given in Fig. 1 to illustrate the effect of increasing the mass of the fissioning nucleus. A shift in mass distribution toward higher mass numbers is seen to take place almost exclusively in the light group. Also, the tendency toward a wider mass distribution is apparent in the heavy group.

The yield of symmetrical fission modes is definitely lower in U^{238} than in Th^{232}. Although the exact shape of the trough in U^{238} is not well defined, the condition of symmetry imposed in drawing a smooth curve through the yield of mass number 115 and its complement, mass number 122, leaves little latitude in the position of the minimum. A trough yield of about one-half that in Th^{232} is indicated. The average energy of the fission spectrum neutrons effective in inducing fission in both U^{238} and Th^{232} is about 2.8 Mev, as deduced from the known neutron energy spectrum[32] and the fission cross sections.[33] Since the neutron binding energies in Th^{233} and U^{239} are 5.1 Mev and 4.9 Mev, respectively,[34] the compound nucleus is slightly more excited in the case of Th^{232} fission and a somewhat higher trough yield might therefore be expected. It has also been suggested by Turkevich[10] that the peak-to-trough ratios in the fission of all heavy nuclei may be a function of the rate of break-up of the compound nucleus. Thus, smaller peak-to-trough ratios would result at excitations where the rate of break-up is faster. It is, of course, difficult to interpret the effect of energy on the peak-to-trough ratios when fission is induced by a spectrum of neutrons. Obviously, more data are needed utilizing various fissile nuclei and monoenergetic neutrons.

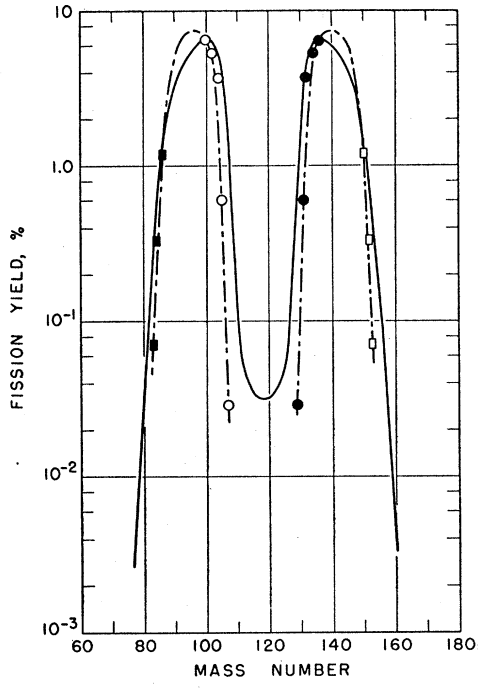

FIG. 2. Yield-mass curves for fast-neutron-induced fission of U^{238} (solid line) and spontaneous fission of U^{238} (dotted line). Solid squares and circles represent data for Kr and Xe isotopes, respectively, from reference 35; open squares and circles represent mirror points.

A comparison of the yield-mass curves for fast neutron-induced fission of U^{238} and spontaneous fission of U^{238} is given in Fig. 2. The data for spontaneous fission are the relative yields of the stable krypton and xenon isotopes produced by spontaneous fission of U^{238} in uranium ores.[35] A dotted curve has been drawn through the data assuming symmetrical reflection about mass 118 (i.e., two neutrons emitted per fission) and normalized to 200 percent yield summation. Narrower peaks and lower yields of symmetrical fission modes in spontaneous fission are apparent.[36] These results are consistent with the trends noted previously regarding the effect of excitation energy on the yield-mass curve.

[30] Glendenin, Coryell, and Edwards, reference 1, Paper No. 52.
[31] R. W. Spence (private communication, September, 1952).
[32] Bonner, Ferrell, and Rinehart, Phys. Rev. **87**, 1032 (1952); D. L. Hill, Phys. Rev. **87**, 1034 (1952); B. E. Watt, Phys. Rev. **87**, 1037 (1952); D. B. Nicodemus and H. H. Staub, Phys. Rev. **89**, 1288 (1953).
[33] Neutron Cross Sections, U. S. Atomic Energy Commission Report AECU-2040 (Technical Information Service, Department of Commerce, Washington, D. C., 1952).
[34] J. R. Huizenga and L. B. Magnusson, Argonne National Laboratory Report ANL-5158, November, 1953 (unpublished).
[35] W. H. Fleming and H. G. Thode, Phys. Rev. **92**, 378 (1953).
[36] More recent mass spectrometric work of G. W. Wetherill [Phys. Rev. **92**, 907 (1953)] on the mass distribution of xenon and krypton from spontaneous fission of uranium and thorium indicates even narrower peaks and lower yields for modes near symmetrical fission.