

## Fission of $^{240}\text{Pu}$ with 14.8-MeV neutrons\*

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We have measured the mass-yield distribution from the fission of  $^{240}\text{Pu}$  induced by 14.8-MeV neutrons. Seventy-five fission products from  $^{66}\text{Ni}$  to  $^{169}\text{Er}$  were detected. From these measurements we obtained the total chain yields for 49 mass numbers and constructed the mass-yield curve. Absolute fission yields were obtained by using the  $^{238}\text{U}(n,f)$  monitor reaction. The total yield determined from each half of the mass-yield curve is 0.984. From the agreement in these two methods we conclude that our yield determination is accurate to approximately 4%. The peak-to-valley ratio is 5.5, a significantly larger value than the ratio of 4.1 found for 14.8-MeV neutron fission of  $^{239}\text{Pu}$ . This difference agrees with the trend noted previously for 14.8-MeV neutron fission of uranium isotopes. A number of independent fission yields were measured. Values of  $Z_p$  inferred from these yields agree with those estimated from independent-yield systematics, but are approximately 0.1 charge unit greater than predicted. Several partial isomer yields and isomer ratios were measured. These results provide additional information about the fractional production of isomeric states either directly from fission or from  $\beta$  decay. The absolute photon intensity for the 342-keV transition in the decay of  $^{111}\text{Ag}^g$  was measured to be  $0.0668(\pm 5\%)$ .

[ NUCLEAR REACTIONS, FISSION  $^{240}\text{Pu}(n,f)$ ,  $E = 14.8$  MeV; measured fission yields, deduced fission mass distribution. Measured independent fission yields and partial isomeric yields.  $^{111}\text{Ag}^g$  measured  $I_\gamma$ . ]

### INTRODUCTION

Many measurements of  $^{239}\text{Pu}$  fission-product yields are made on targets of  $^{239}\text{Pu}$  containing a significant fraction of  $^{240}\text{Pu}$ . The  $^{240}\text{Pu}$  may exist as an isotopic impurity, or arise from the formation of  $^{240}\text{Pu}$  by the  $(n,\gamma)$  reaction during high-fluence neutron irradiation. Because earlier results for the fission of different uranium isotopes have shown there can be significant differences in the yield distribution from adjacent isotopes,<sup>1</sup> it is important to know the mass-yield distribution from neutron-induced fission of  $^{240}\text{Pu}$ . Until now, no measurements have been reported, and it has not been possible to correct the results from  $^{239}\text{Pu}$  fission experiments for the effects of  $^{240}\text{Pu}$  fission.

The mass-yield distribution from fission of  $^{240}\text{Pu}$  induced by 14.8-MeV neutrons was determined by measurements on the  $\text{PuO}_2$  targets, on aliquots of the dissolved  $\text{PuO}_2$  targets, and on chemically separated samples. We have results for 75 nuclides from the mass range 66 through 169. Absolute fission yields were obtained by using the  $^{238}\text{U}(n,f)$  monitor reaction.

### EXPERIMENTAL PROCEDURE

Enriched  $^{240}\text{Pu}$ , as  $\text{PuO}_2$ , was obtained from Oak Ridge National Laboratory. It had the isotopic

composition  $^{238}\text{Pu}$ , 0.017%;  $^{239}\text{Pu}$ , 0.67%;  $^{240}\text{Pu}$ , 98.39%;  $^{241}\text{Pu}$ , 0.55%; and  $^{242}\text{Pu}$ , 0.37% at the time of irradiation. Because of the high level of  $\alpha$  radioactivity associated with the  $^{240}\text{PuO}_2$ , all target preparation and most of the postirradiation chemistry was done in a glove box. Before each irradiation, the Pu was purified to remove decay products of  $^{241}\text{Pu}$  ( $^{241}\text{Am}$ ,  $^{237}\text{Np}$ , and  $^{237}\text{U}$ ), spontaneous fission products, and inert contaminants. Up to 300 mg of  $^{240}\text{PuO}_2$  was dissolved in 6 M HCl, a few drops of  $\text{HBF}_4$ , and a few drops of  $\text{HNO}_3$  to maintain Pu as Pu(IV). The solution was boiled down, mixed with 12 M HCl and loaded onto a Dowex 1 anion exchange column which had been preconditioned with 12 M HCl containing a few drops of  $\text{HNO}_3$ . The adsorbed Pu(IV) was washed with 12 M HCl. The Pu(IV) was reduced to Pu(III) and eluted with a mixture of 12 M HCl and 0.5 M HI. This reduction-elution step is rather specific for Pu. Following removal of I from the eluate by boiling with aqua regia, Pu was precipitated as the oxalate, washed, and converted to the oxide.

Individual  $^{240}\text{PuO}_2$  targets, ranging from 10 to 200 mg, were doubly encapsulated, first in either 0.012-mm Mylar or 0.025-mm aluminum foil to prevent loss of fission fragments and then in 0.2-mm polyvinyl chloride to prevent contamination by  $\alpha$  emitters. Monitor foils of enriched  $^{238}\text{U}$  (99.8%  $^{238}\text{U}$ , 0.2%  $^{235}\text{U}$ ) were similarly encapsu-

lated. Each target assembly consisted of a  $^{240}\text{PuO}_2$  target, either alone or sandwiched between two monitor foils. The dimensions of the assembly were approximately 1.0 cm square by less than 0.5 cm between the front and back monitor foils. Each assembly was encapsulated in 0.5-mm-thick aluminum and contained in a Cd target holder to minimize the possibility of extraneous low-energy neutrons causing fission of the isotopic impurities  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$ . The Cd target holder had no line-of-sight joints. Its minimum thickness (the side facing the neutron source) was 0.5 mm.

The 14.8-MeV neutron irradiations were made at the LLL insulated-core-transformer (ICT) accelerator, where neutrons are produced by the reaction of a beam of 400-keV deuterons on a rotating titanium tritide target.<sup>2</sup> The maximum deuteron beam current was 20  $\mu\text{A}$ , producing a 14.8-MeV source of up to  $5 \times 10^{12}$  n/s. The neutron flux density was monitored with a proton-recoil counter so corrections could be made for the small changes in beam intensity. We had five irradiations, varying in duration from 1.5 to 15 h and producing up to  $4 \times 10^{12}$  fissions. The target holder was placed at  $0^\circ$  to the deuteron beam and either as close as possible to the neutron source or 10 cm away. The latter position ensured similar irradiation conditions for front and back monitor foils during irradiations using  $^{238}\text{U}$  foils as fission monitors. The mean neutron energy in the  $\text{PuO}_2$  and U targets was  $14.8 \pm 0.3$  MeV.

After irradiation, the polyvinyl chloride package containing the  $^{240}\text{PuO}_2$  was either sealed in a new aluminum container for  $\gamma$ -ray spectrometric analysis or transferred to a glove box for radiochemical separations. The target, Mylar, and polyvinyl chloride were dissolved in a mixture of  $\text{HNO}_3$  and  $\text{HClO}_4$  containing microgram quantities of Zr, Mo, Pd, and Ag. The solution was diluted to known volume. The actual  $^{240}\text{Pu}$  content of the target was determined from an aliquot of this solution by thermal ionization mass spectrometry at McClellan Central Laboratory (MCL) using the isotope dilution technique with  $^{242}\text{Pu}$  as tracer. Other aliquots were taken for radiochemical separations and direct  $\gamma$ -ray spectrometric measurements. Standard radiochemical procedures<sup>3-6</sup> were modified to allow the removal of Pu and  $^{241}\text{Am}$  and the sequential separation of up to 15 elements from a single aliquot. Rare earth elements were separated on a Dowex 50 cation exchange column. The eluent was a solution of  $\alpha$ -hydroxyisobutyric acid at pH 4.2, the concentration of which was increased exponentially from 0.2M to 0.45 M. Those rare earth elements having very low fission yield (Er and Tb) were further purified by a repeated separation on a similar ion exchange column. In one

irradiation we measured rare earth fission products having half-lives shorter than 6 h. The more rapid separation of rare earth elements required for this irradiation was achieved by operating the ion exchange column at elevated pressure.<sup>7</sup>

The disintegration rates of individual radioactive fission products were determined by  $\gamma$ -ray spectrometry at LLL or MCL, by  $\beta$ -particle assay at MCL, or by both measurement techniques. The characteristic radiation detected for each nuclide is presented in Table I. Four types of samples were analyzed by  $\gamma$ -ray spectrometry: undissolved  $^{240}\text{PuO}_2$  targets and  $^{238}\text{U}$  monitor foils (at LLL), aliquots of dissolved targets (LLL), group-separated rare earth samples (LLL, MCL), and radiochemically purified samples (LLL, MCL). We used 10 coaxial Ge(Li) detectors ranging in active volume from 45 to 85  $\text{cm}^3$  with resolutions from 2.39 to 1.86 keV full width at half maximum at 1332 keV. Spectra were interpreted with the GAMANAL code.<sup>46</sup> The absolute efficiency curve for each detector was determined with standardized sources from Amersham Searle Corp; Centre d'Etalonnage des Rayonnements Ionisants, France; International Atomic Energy Agency; Lawrence Livermore Laboratory; Los Alamos Scientific Laboratory; McClellan Central Laboratory; National Office of Measures, Hungary; and U. S. National Bureau of Standards.

Radiochemically purified samples were subjected to  $\beta$ -particle assay at MCL where we used 12 gas-flow proportional counters. Data for count rate as a function of time were resolved by multi-component weighted least-squares analysis. The absolute efficiencies for detected fission products were determined as a function of precipitate thickness using standardized sources from Amersham Searle Corp., Lawrence Livermore Laboratory, Los Alamos Scientific Laboratory, McClellan Central Laboratory, and U. S. National Bureau of Standards. The efficiencies of nuclides unavailable as standardized sources (for instance,  $^{66}\text{Ni}$  and  $^{157}\text{Eu}$ ) were estimated from a general curve relating efficiency to mean  $\beta$ -particle energy.

Aliquots free of Cs carrier were analyzed by thermal ionization mass spectrometry for atom ratios of fission product cesium isotopes. Targets for these measurements were dissolved several weeks after irradiation. Absolute atom yields were obtained by relating the atom ratios  $^{134}\text{Cs}/^{137}\text{Cs}$  and  $^{135}\text{Cs}/^{137}\text{Cs}$  to the atom yield of  $^{137}\text{Cs}$  determined by  $\gamma$ -ray spectrometry.

## RESULTS AND DISCUSSION

### Fission yield measurements

The measured amount of each fission product was corrected for decay back to the end of irradiation.

TABLE I. Nuclear properties of fission products detected in  $^{240}\text{Pu}$  fission.

Nuclide	Radiation detected ( $\gamma$ energy in keV)	Absolute $\gamma$ intensity ( $\gamma$ /decay)	Half-life	References
$^{66}\text{Ni}^a$	$\beta$		54.6 h	8
$^{67}\text{Cu}$	91.23	0.0683	2.575 day	9
	93.29	0.161		
	184.56	0.487		
$^{72}\text{Zn}$	144.7	0.83	46.5 h	10
$^{73}\text{Ga}$	295	0.77	4.91 h	11
	328	0.135		
	402.47	0.473		
$^{87}\text{Kr}$	2554.92	0.0847	1.267 h	12
	2195.90	0.141		
$^{88}\text{Kr}$	2392.10	0.379	2.803 h	12
	$\beta$			
$^{89}\text{Sr}$	$\beta$		50.55 day	13
$^{90}\text{Sr}^b$	$\beta$		28.6 yr	14
$^{91}\text{Sr}$	555.63 <sup>c</sup>	0.58	9.48 h	15
	749.77	0.24		
	1024.25	0.33		
$^{91}\text{Y}$	$\beta$		58.51 day	16
$^{92}\text{Sr}$	1383.94	0.90	2.71 h	15
$^{92}\text{Y}$	934.52	0.137	3.53 h	17
$^{93}\text{Y}$	266.87	0.068	10.24 h	12, 18
	947.1	0.0194		
	1917.8	0.014		
$^{95}\text{Zr}$	$\beta$		65.0 day	15
	756.72	0.546		
	765.80 <sup>c</sup>	0.990		
$^{96}\text{Nb}$	568.86	0.558	23.4 h	19
	778.22	0.968		
	1091.31	0.495		
$^{97}\text{Zr}$	$\beta$		16.82 h	15
	658.18 <sup>c</sup>	0.990		
	743.37	0.940		
$^{99}\text{Mo}$	$\beta$		2.752 day	9, 12, 15
	140.514 <sup>c</sup>	0.805		
	181.092	0.0604		
	739.481	0.120		
	777.900	0.0424		
$^{103}\text{Ru}$	497.09	0.900	39.6 day	12, 15
$^{105}\text{Ru}$	469.38	0.175	4.44 h	15, 20
	676.40	0.167		
	319.24	0.196		
$^{105}\text{Rh}$	319.24	0.196	1.476 day	15, 20
$^{106}\text{Ru}$	622.10 <sup>c</sup>	0.098	369 day	15
$^{109}\text{Pd}$	88.04	0.0379	13.47 h	12, 21
$^{110}\text{Ag}^m$	937.3	0.336	260 day	15
	1384.3	0.24		
$^{111}\text{Pd}^m$	172.2	0.324	5.5 h	22
$^{111}\text{Ag}^f$	$\beta$		7.431 day	9
	342.14	0.0668 <sup>d</sup>		
$^{112}\text{Pd}$	617.40 <sup>c</sup>	0.435	21.12 h	15, 23
$^{113}\text{Ag}^f$	$\beta$		5.371 h	24
	298.4	0.093		
$^{115}\text{Cd}^m$	$\beta$		44.6 day	25
$^{115}\text{Cd}^f$	$\beta$		2.208 day	9, 12
	336.25 <sup>c</sup>	0.465		
	492.29	0.0826		
	527.86	0.280		
$^{117}\text{Cd}^m$	564.4	0.153	3.35 h	26, 27
	1066.0	0.231		
	1997.4	0.254		

TABLE I. (Continued)

Nuclide	Radiation detected ( $\gamma$ energy in keV)	Absolute $\gamma$ intensity ( $\gamma$ /decay)	Half-life	References
$^{117}\text{Cd}^{\text{e}}$	273.31	0.284	2.56 h	26, 27
	344.51	0.173		
	1303.4	0.176		
$^{118}\text{Cd}$	1229.5 <sup>c</sup>	0.15	0.838 h	28
$^{125}\text{Sn}^{\text{e}}$	822.6	0.0387	9.65 day	17
	915.5	0.0376		
	1066.6	0.0887		
$^{125}\text{Sb}$	427.88	0.304	2.77 yr	12, 15
$^{126}\text{Sb}^{\text{e}}$	414.7	0.810	12.5 day	15
	666.2	1.00		
	695.1	1.00		
$^{127}\text{Sn}^{\text{e}}$	1095.6	0.195	2.12 h	29
$^{127}\text{Sb}$	473.20	0.248	3.87 day	9
	684.90	0.368		
$^{128}\text{Sn}$	482.0	0.66	1.00 h	17
$^{128}\text{Sb}^{\text{e}}$	636.2	0.360	9.01 h	17
$^{129}\text{Sb}$	544.7	0.181	4.32 h	30
	914.6	0.202		
	1030.1	0.127		
$^{130}\text{Sb}^{\text{e}}$	330.9	0.78	0.667 h	17
	839.4	1.00		
$^{130}\text{I}^{\text{e}}$	536.09	0.99	12.36 h	31
$^{131}\text{Te}^{\text{m}}$	793.80	0.159	1.25 day	15
	852.30	0.256		
	1125.50	0.148		
	1206.6	0.118		
$^{131}\text{I}$	364.46	0.79	8.022 day	12
$^{132}\text{Te}$	228.2	0.88	3.24 day	15
	522.60 <sup>c</sup>	0.156		
	630.20 <sup>c</sup>	0.135		
$^{132}\text{Cs}$	667.5	0.974	6.475 day	32
$^{133}\text{I}$	529.91	0.83	21.0 h	9
$^{134}\text{Cs}^{\text{m}}$	127.42	0.143	2.90 h	33
$^{134}\text{Cs}^{\text{e}}$	604.70	0.98	767 day	15
$^{135}\text{I}$	1260.41	0.284	6.610 h	9
	1457.56	0.0858		
	1678.03	0.0947		
$^{135}\text{Xe}$	249.65	0.92	9.106 h	12, 15
$^{135}\text{Cs}^{\text{m}}$	786.34	0.997	0.883 h	12, 34
	845.05	0.959		
$^{135}\text{Cs}^{\text{e}}$	e		$2.3 \times 10^6$ yr	34
$^{136}\text{Cs}$	818.48	1.000	13.0 day	15
	1048.10	0.805		
	1235.41	0.197		
$^{137}\text{Cs}$	661.62 <sup>c</sup>	0.850	30.01 yr	15
$^{138}\text{Cs}$	462.7	0.270	0.557 h	17
	1009.7	0.285		
	1435.7	0.750		
$^{139}\text{Ba}$	165.85	0.220	1.385 h	15
$^{140}\text{Ba}$	$\beta$		12.80 day	12
	537.261	0.2446		
$^{141}\text{Ce}$	1596.20 <sup>c</sup>	0.9552	32.38 day	12
	$\beta$			
$^{142}\text{La}$	145.44	0.493	1.545 h	15, 35
	641.21	0.465		
	2398.0	0.117		
$^{143}\text{Ce}$	2542.9	0.088	1.379 day	12
	293.20	0.435		

TABLE I. (Continued)

Nuclide	Radiation detected ( $\gamma$ energy in keV)	Absolute $\gamma$ intensity ( $\gamma$ /decay)	Half-life	References
$^{144}\text{Ce}$	$\beta$ 133.50	0.110	284.6 day	15
$^{145}\text{Pr}$	674.0 748.0	0.0043 0.0043	5.98 h	36
$^{147}\text{Nd}$	$\beta$ 531.00	0.1295	11.04 day	12
$^{149}\text{Nd}$	114.321 211.307	0.161 0.234	1.721 h	12
$^{149}\text{Pm}$	285.90	0.031	2.208 day	15, 37
$^{151}\text{Pm}$	167.73 + 168.38 240.08 340.08	0.092 0.036 0.224	1.183 day	15, 38
$^{153}\text{Sm}$	$\beta$ 103.17	0.284	1.928 day	15, 39
$^{155}\text{Eu}$	86.55 105.32	0.335 0.224	4.68 yr	15, 40
$^{156}\text{Eu}$	$\beta$		15.19 day	15
$^{157}\text{Eu}$	$\beta$ 413	0.186	15.15 h	41
$^{159}\text{Gd}$	$\beta$ 363.56	0.103	18.56 h	42
$^{160}\text{Tb}$	$\beta$		72.1 day	15
$^{161}\text{Tb}$	$\beta$		6.91 day	43
$^{169}\text{Er}$	$\beta$		9.40 day	44

<sup>a</sup> Confirmed by observation of the 1039-keV  $\gamma$  ray from the  $^{66}\text{Cu}$  daughter in equilibrium.

<sup>b</sup> Determined by milking of the 2.66-day  $^{90}\text{Y}$  daughter.

<sup>c</sup>  $\gamma$  ray emitted by the daughter in equilibrium.

<sup>d</sup> A value of 0.0668( $\pm 5\%$ ) for the absolute photon intensity ( $I_\gamma$ ) of the 342-keV  $\gamma$  ray was determined by the  $4\pi\beta\gamma$  coincidence technique. This result is in agreement with the recommended value of Thierens *et al.* (0.068  $\pm$  0.006) (Ref. 45).

<sup>e</sup> Determined from 135/137 atom ratio and  $^{137}\text{Cs}$  disintegration rate. See text.

tion. A correction was also made for decay during irradiation, based on the time history of neutron flux density as measured by a proton-recoil counter. In cases where both the parent and daughter nuclides have half-lives within a factor of 100 of each other (such as the  $^{91}\text{Sr}$ -Y,  $^{92}\text{Sr}$ -Y,  $^{105}\text{Ru}$ -Rh,  $^{135}\text{I}$ -Xe pairs), it was necessary to consider both the parent and daughter decay in making this correction.

Results of the fission yield measurements are summarized in Table II. Relative yields were first calculated as the ratio of a particular yield to that of  $^{99}\text{Mo}$ , and the ratios for each product were averaged over all the experiments.  $^{238}\text{U}$  monitor foils were used with four  $^{240}\text{PuO}_2$  targets in two of the irradiations. The number of fissions occurring in these  $^{240}\text{PuO}_2$  targets was calculated from the  $^{240}\text{Pu}$  and  $^{238}\text{U}$  masses, the 14.8-MeV neutron fission cross section ratio,  $\sigma_f(^{240}\text{Pu})/\sigma_f(^{238}\text{U}) = 1.98 \pm 0.06$ ,<sup>47, 48</sup> and the average number of  $^{238}\text{U}$  fissions. The latter quantity was determined

from numerous (10 to 15) prominent fission products for each pair of foils using yields given in Ref. 49. The average front-to-back ratio was 1.03. From these four targets used with  $^{238}\text{U}$  the absolute fission yield of  $^{99}\text{Mo}$  was determined to be 0.0485 atoms per fission with an estimated uncertainty of 5%, based on the uncertainties in the  $^{238}\text{U}$  and  $^{240}\text{Pu}$  fission cross sections and the  $^{238}\text{U}$  fission yields. The absolute fission yields of the other products from  $^{240}\text{Pu}$  fission were calculated from their relative yields and the  $^{99}\text{Mo}$  absolute yield. Since the fission cross sections of  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ , and  $^{242}\text{Pu}$  are all about the same at 14.8 MeV, the corrections for fission of the isotopic impurities  $^{239}\text{Pu}$ ,  $^{241}\text{Pu}$ , and  $^{242}\text{Pu}$  are each less than 1%, and tend to be self-cancelling.

In column 4 of Table II we have listed the estimated fraction of the total chain yield for each product nuclide. These estimates are based on tabulated  $Z_p$  values<sup>50</sup> and a Gaussian charge dispersion curve with  $\sigma = 0.56$ . The  $Z_p$  values were

TABLE II. Yields of products from fission of  $^{240}\text{Pu}$  with 14.8-MeV neutrons.

Product nuclide	Measured fission yield (atoms/fission)	Percent error	Est. fraction of total chain yield	Total chain yield <sup>a</sup> (atoms/fission)
$^{66}\text{Ni}$	$6.04 \times 10^{-7}$	44	$0.999 \pm 0.001$	$(6.1 \pm 2.7) \times 10^{-7}$
$^{67}\text{Cu}$	$1.31 \times 10^{-6}$	35	1	$(1.3 \pm 0.5) \times 10^{-6}$
$^{72}\text{Zn}$	$4.47 \times 10^{-5}$	10	$0.993 \pm 0.005$	$(4.5 \pm 0.5) \times 10^{-5}$
$^{73}\text{Ga}$	$8.34 \times 10^{-5}$	5.8	1	$(8.3 \pm 0.5) \times 10^{-5}$
$^{87}\text{Kr}$	0.0122	8.2	$0.990 \pm 0.004$	$0.0123 \pm 0.0010$
$^{88}\text{Kr}$	0.0128	4.9	$0.943 \pm 0.016$	$0.0136 \pm 0.0007$
$^{89}\text{Sr}$	0.0174	3.0	1	$0.0174 \pm 0.0005$
$^{90}\text{Sr}$	0.0194	3.0	1	$0.0194 \pm 0.0006$
$^{91}\text{Sr}$	0.0239	3.0	$0.999 \pm 0.001$	0.0239
$^{91}\text{Y}$	0.0249	3.0	1	0.0249
Mass 91				$0.0244 \pm 0.0005$
$^{92}\text{Sr}$	0.0277	3.0	$0.989 \pm 0.004$	0.0281
$^{92}\text{Y}$	0.0293	4.0	1	0.0293
Mass 92				$0.0284 \pm 0.0006$
$^{93}\text{Y}$	0.0288	3.0	1	$0.0288 \pm 0.0009$
$^{95}\text{Zr}$	0.0368	3.0	1	$0.0368 \pm 0.0007$
$^{96}\text{Nb}$	$8.34 \times 10^{-5}$	3.0	b	
$^{97}\text{Zr}$	0.0418	3.0	$0.990 \pm 0.004$	$0.0422 \pm 0.0013$
$^{99}\text{Mo}$	0.0485	3.0	1	$0.0485 \pm 0.0015$
$^{103}\text{Ru}$	0.0527	3.0	1	$0.0527 \pm 0.0016$
$^{105}\text{Ru}$	0.0525	3.0	1	0.0525
$^{105}\text{Rh}$	0.0432	3.0	1	0.0432
Mass 105				$0.0432 \pm 0.0013$ <sup>c</sup>
$^{106}\text{Ru}$	0.0400	3.0	$0.999 \pm 0.001$	$0.0400 \pm 0.0012$
$^{109}\text{Pd}$	0.0229	3.0	1	$0.0229 \pm 0.0007$
$^{110}\text{Ag}^m$	$\leq 4.9 \times 10^{-6}$		b	
$^{111}\text{Pd}^m$	$9.90 \times 10^{-3}$	8.6	$0.997 \pm 0.003$	$(9.9 \pm 0.8) \times 10^{-3}$ <sup>d</sup>
$^{111}\text{Ag}$	0.0166	3.0	1	$0.0166 \pm 0.0005$
$^{112}\text{Pd}$	0.0156	3.0	$0.987 \pm 0.016$	$0.0158 \pm 0.0005$
$^{113}\text{Ag}^f$	0.0153	5.2	1	$0.0153 \pm 0.0008$
$^{115}\text{Cd}^f$	0.0109	3.0	1	$0.0109 \pm 0.0003$
$^{115}\text{Cd}^m$	$7.18 \times 10^{-4}$	3.0	1	$(7.18 \pm 0.22) \times 10^{-4}$
Mass 115				$0.0116 \pm 0.0003$
$^{117}\text{Cd}^f$	$6.01 \times 10^{-3}$	3.0 <sup>e</sup>	$0.996 \pm 0.004$	$(6.0 \pm 1.2) \times 10^{-3}$
$^{117}\text{Cd}^m$	$2.57 \times 10^{-3}$	3.0 <sup>e</sup>	$0.996 \pm 0.004$	$(2.6 \pm 0.5) \times 10^{-3}$
Mass 117				$(8.6 \pm 1.3) \times 10^{-3}$
$^{118}\text{Cd}$	$4.95 \times 10^{-3}$	6.0	$0.979 \pm 0.018$	$(5.1 \pm 1.0) \times 10^{-3}$ <sup>f</sup>
$^{125}\text{Sn}^f$	$8.44 \times 10^{-3}$	3.0	$0.959 \pm 0.022$	$(8.80 \pm 0.33) \times 10^{-3}$ <sup>d</sup>
$^{125}\text{Sb}$	0.0157	3.0	1	$0.0157 \pm 0.0005$
$^{126}\text{Sb}^f$	$3.60 \times 10^{-3}$	3.2	b	
$^{127}\text{Sn}^f$	0.0143	4.0 <sup>g</sup>	$0.732 \pm 0.062$	$0.0195 \pm 0.0042$ <sup>d</sup>
$^{127}\text{Sb}$	0.0195	3.0	$0.992 \pm 0.004$	$0.0197 \pm 0.0006$
$^{128}\text{Sn}$	$9.94 \times 10^{-3}$	3.0	b	
$^{128}\text{Sb}^f$	0.0101	3.0	$0.963 \pm 0.014$	$0.0105 \pm 0.0003$ <sup>d</sup>
$^{129}\text{Sb}$	0.0191	4.2	$0.875 \pm 0.030$	$0.0218 \pm 0.0012$
$^{130}\text{Sb}^f$	0.0171	5.0	$0.686 \pm 0.049$	$0.0250 \pm 0.0022$ <sup>d</sup>
$^{130}\text{I}^f$	$2.09 \times 10^{-3}$	4.5	b	
$^{131}\text{Te}^m$	0.0154	3.0	$0.940 \pm 0.016$	$0.0164 \pm 0.0006$ <sup>d</sup>
$^{131}\text{I}$	0.0445	3.2	1	$0.0445 \pm 0.0014$
$^{132}\text{Te}$	0.0355	3.0	$0.796 \pm 0.037$	$0.0445 \pm 0.0025$
$^{132}\text{Cs}$	$1.7 \times 10^{-5}$	60	b	
$^{133}\text{I}$	0.0538	3.4	$0.968 \pm 0.010$	$0.0556 \pm 0.0020$
$^{134}\text{Cs}^f$	$4.22 \times 10^{-4}$	3.6	b	
$^{134}\text{Cs}^m$	$2.23 \times 10^{-4}$	3.8	b	
$^{135}\text{I}$	0.0386	3.2	$0.629 \pm 0.049$	$0.0614 \pm 0.0052$

TABLE II. (Continued)

Product nuclide	Measured fission yield (atoms/fission)	Percent error	Est. fraction of total chain yield	Total chain yield <sup>a</sup> (atoms/fission)
$^{135}\text{Xe}$	0.0579	6.2	$0.983 \pm 0.006$	$0.0589 \pm 0.0037$
$^{135}\text{Cs}^f$	0.0497	4.1	1	$0.0497 \pm 0.0020$
Mass 135				$0.0520 \pm 0.0033$
$^{135}\text{Cs}^m$	$6.55 \times 10^{-4}$	3.3	b	
$^{136}\text{Cs}$	$5.03 \times 10^{-3}$	3.0	b	
$^{137}\text{Cs}$	0.0448	4.0	$0.996 \pm 0.002$	$0.0450 \pm 0.0018$
$^{138}\text{Cs}$	0.0519	10	$0.977 \pm 0.007$	$0.053 \pm 0.005$
$^{139}\text{Ba}$	0.0456	3.0	$0.999 \pm 0.001$	$0.0457 \pm 0.0014$
$^{140}\text{Ba}$	0.0377	3.0	$0.993 \pm 0.002$	$0.0380 \pm 0.0011$
$^{141}\text{Ce}$	0.0357	3.0	1	$0.0357 \pm 0.0011$
$^{142}\text{La}$	0.0388	8.0	$0.998 \pm 0.001$	$0.0389 \pm 0.0031$
$^{143}\text{Ce}$	0.0309	3.0	1	$0.0309 \pm 0.0009$
$^{144}\text{Ce}$	0.0265	3.0	1	$0.0265 \pm 0.0008$
$^{145}\text{Pr}$	0.0269	10	1	$0.0269 \pm 0.0027$
$^{147}\text{Nd}$	0.0178	3.0	1	$0.0178 \pm 0.0005$
$^{148}\text{Nd}$	0.0145	3.2	$0.999 \pm 0.001$	$0.0145 \pm 0.0004$
$^{149}\text{Pm}$	0.0129	3.0	1	$0.0129 \pm 0.0004$
Mass 149				$0.0136 \pm 0.0008$
$^{151}\text{Pm}$	$8.05 \times 10^{-3}$	6.2	1	$(8.1 \pm 0.5) \times 10^{-3}$
$^{152}\text{Sm}$	$6.55 \times 10^{-3}$	3.0	1	$(6.55 \pm 0.20) \times 10^{-3}$
$^{155}\text{Eu}$	$2.78 \times 10^{-3}$	3.0	1	$(2.78 \pm 0.08) \times 10^{-3}$
$^{156}\text{Eu}$	$2.25 \times 10^{-3}$	3.0	1	$(2.25 \pm 0.07) \times 10^{-3}$
$^{157}\text{Eu}$	$1.58 \times 10^{-3}$	3.0	$0.999 \pm 0.001$	$(1.58 \pm 0.05) \times 10^{-3}$
$^{159}\text{Gd}$	$6.8 \times 10^{-4}$	15	1	$(6.8 \pm 1.0) \times 10^{-4}$
$^{160}\text{Tb}$	$\leq 1.5 \times 10^{-6}$		b	
$^{161}\text{Tb}$	$3.46 \times 10^{-4}$	3.0	1	$3.46 \pm 0.10 \times 10^{-4}$
$^{169}\text{Er}$	$1.15 \times 10^{-5}$	4.0	1	$(1.15 \pm 0.05) \times 10^{-5}$

<sup>a</sup>The experimental standard deviations given here do not include the systematic uncertainty in the yield determination. This additional uncertainty is estimated to be about 4%.

<sup>b</sup>Independent fission yield—see Table IV.

<sup>c</sup>The  $^{105}\text{Rh}$  yield is preferred. The  $^{105}\text{Ru}$  yield is not consistent with the mass-yield curve. The photon abundances of  $^{105}\text{Ru}$  and  $^{105}\text{Rh}$  are well known, and replicate analyses agreed in both cases. The large discrepancy in the fission yields could not be resolved.

<sup>d</sup>Partial isomeric yield only.

<sup>e</sup>The disagreement in the photon abundances found in the literature for  $^{117}\text{Cd}^{m,*f}$  is such that we have arbitrarily increased the total uncertainty in the yields to 20%.

<sup>f</sup>The yield of  $^{118}\text{Cd}$  is in disagreement with the expected shape of the mass-yield curve, and has not been used. We believe that the absolute photon abundance is in error. We have also measured the  $^{118}\text{Cd}$  yield from thermal fission of  $^{235}\text{U}$  and found similar disagreement.

<sup>g</sup>The uncertainty in the yield of  $^{127}\text{Sn}^f$  has been increased to 20% to allow for the extra uncertainty in the photon abundance.

derived from systematics inferred from an analysis of measured independent and cumulative fractional chain yields.<sup>51</sup>

Of the 75 individual measurements given in Table II, we have calculated the total chain yield for 49 mass numbers. The remaining results are independent yields, partial isomer yields, or multiple measurements for the same mass number. We show the total chain yields greater than  $10^{-4}$  as a mass-yield curve in Fig. 1. Because we found no distinct structure, we drew a smooth curve

through the data points using their reflected values as an aid. We assumed the average mass of the fissioning nucleus was 236.0, corresponding to an emission of 5.0 neutrons.<sup>52</sup> The total yield in each half of the mass-yield curve is 0.984. The closeness of this value to unity confirms the reliability of the yields based on the  $^{238}\text{U}$  fission monitors. Since both methods have uncertainties of about 5%, we estimate then that the overall uncertainty in the yield determination is approximately 4%.

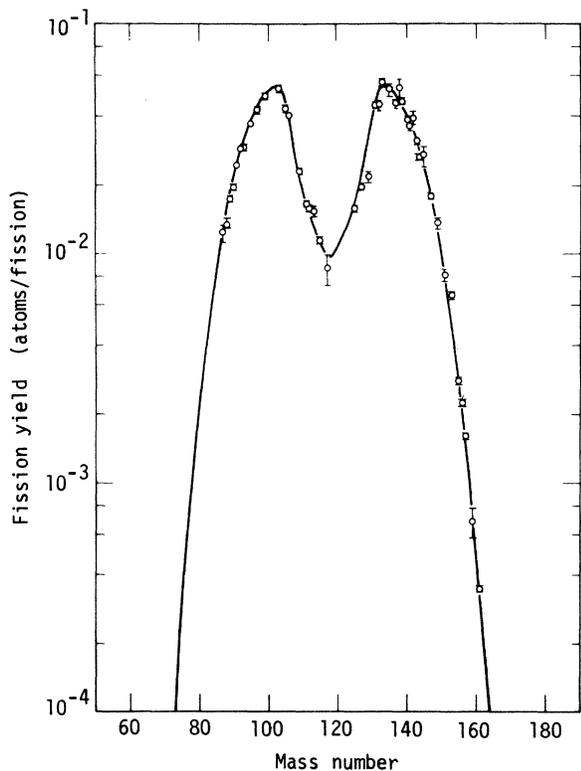


FIG. 1. Mass-yield curve for fission of  $^{240}\text{Pu}$  with 14.8-MeV neutrons. Measured yields are indicated by open circles with error bars. The curve was drawn with aid of mirror points reflected about mass 118.0.

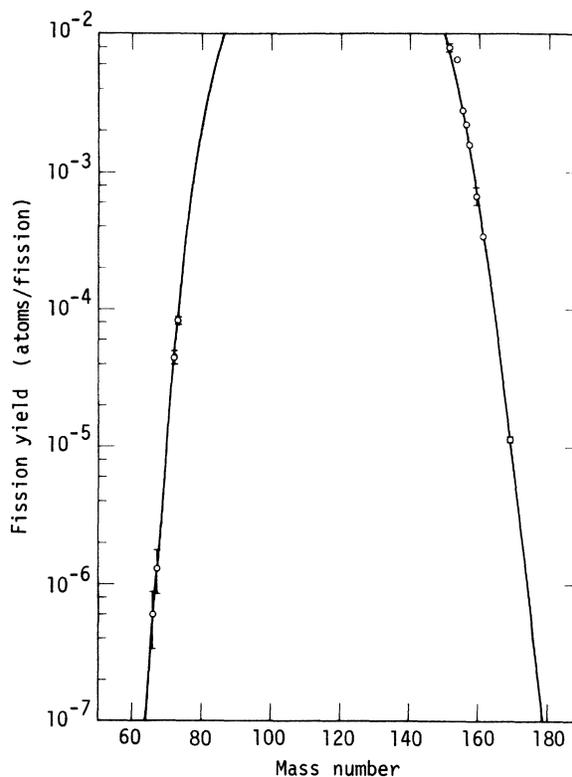


FIG. 2. Low-yield products from fission of  $^{240}\text{Pu}$  with 14.8-MeV neutrons. Measured yields are indicated by open circles with error bars.

The peak-to-valley ratio in the mass-yield distribution is 5.5 compared with the value of 4.1 for 14.8-MeV neutron fission of  $^{239}\text{Pu}$ .<sup>49</sup> This agrees with the trend found for 14.8-MeV fission of the isotopes of uranium,<sup>1</sup> i.e., the peak-to-valley ratio increases with increasing target mass number from 3.2 (for  $^{233}\text{U}$ ) to 6.8 (for  $^{238}\text{U}$ ).

The total chain yields of products on the lower portions of the mass-yield curve are shown in Fig. 2 and appear to be skewed with no common central mass number. Two experimental factors may account for this effect. The first is the imprecise low yields determined for  $^{66}\text{Ni}$  and  $^{67}\text{Cu}$ . The reported yield of  $^{67}\text{Cu}$  represents only 26% of the measured yield. This is caused by its production from zinc impurity in the  $^{240}\text{Pu}$  target material. The correction is based on an observed apparent yield of  $^{65}\text{Zn}$  and the results of a separate experiment in which the relative yields of  $^{65}\text{Zn}$ ,  $^{66}\text{Ni}$ , and  $^{67}\text{Cu}$  were measured in a zinc target irradiated with 14.8-MeV neutrons. The correction to the  $^{66}\text{Ni}$  fission yield is negligible. Second, the skewness and lack of common central mass number may be caused by the result for  $^{169}\text{Er}$ . It is

possible that the majority of the observed  $^{169}\text{Er}$  yield was produced from reactions on Er impurity. The observed  $^{169}\text{Er}$  could have been produced from an erbium impurity of approximately 70 ppm. The actual erbium content is not known, but, because of the rather specific purification of our target material, we would not expect it to be that high. We have not made any adjustment on the observed  $^{169}\text{Er}$  to account for this, but we note that it may have been significant.

We should also note that the yields of the products on the lower part of the mass-yield curve in Fig. 2 might not have the same central mass number as the high-yield products. For example, in the fission of five uranium isotopes the central mass number of the low-yield products was about one mass number higher than for the peak-yield products.<sup>1</sup> This effect may be caused by an increased energy requirement for reactions leading to the low-yield products and which are accompanied by less neutron emission.

A listing of the recommended total chain yields for mass numbers from 66 to 170 is in Table III. The yields are taken from the smooth curves shown in Figs. 1 and 2.

TABLE III. The recommended total chain yields for mass numbers 66 to 170. These yields were taken from the smooth curves drawn through the data points in Figs. 1 and 2. The yields are given as atoms/fission.

Mass number	Total chain yield	Mass number	Total chain yield
66	$6.1 \times 10^{-7}$	118	0.0097
67	$1.3 \times 10^{-6}$	119	0.0099
68	$2.4 \times 10^{-6}$	120	0.0105
69	$4.9 \times 10^{-6}$	121	0.0115
70	$9.8 \times 10^{-6}$	122	0.0123
71	$2.1 \times 10^{-5}$	123	0.0135
72	$4.5 \times 10^{-5}$	124	0.0150
73	$9.0 \times 10^{-5}$	125	0.0167
74	$1.6 \times 10^{-4}$	126	0.0192
75	$3.2 \times 10^{-4}$	127	0.0224
76	$5.1 \times 10^{-4}$	128	0.0261
77	$7.5 \times 10^{-4}$	129	0.0310
78	0.00108	130	0.0375
79	0.00156	131	0.0450
80	0.00217	132	0.0516
81	0.00293	133	0.0530
82	0.0039	134	0.0538
83	0.0050	135	0.0534
84	0.0064	136	0.0515
85	0.0082	137	0.0494
86	0.0102	138	0.0470
87	0.0124	139	0.0436
88	0.0149	140	0.0408
89	0.0176	141	0.0375
90	0.0210	142	0.0345
91	0.0244	143	0.0311
92	0.0281	144	0.0281
93	0.0311	145	0.0244
94	0.0345	146	0.0210
95	0.0375	147	0.0176
96	0.0408	148	0.0149
97	0.0436	149	0.0124
98	0.0470	150	0.0102
99	0.0494	151	0.0082
100	0.0515	152	0.0064
101	0.0534	153	0.0050
102	0.0538	154	0.0039
103	0.0530	155	0.00293
104	0.0516	156	0.00217
105	0.0450	157	0.00156
106	0.0375	158	0.00108
107	0.0310	159	$7.5 \times 10^{-4}$
108	0.0261	160	$5.1 \times 10^{-4}$
109	0.0224	161	$3.5 \times 10^{-4}$
110	0.0192	162	$2.3 \times 10^{-4}$
111	0.0167	163	$1.6 \times 10^{-4}$
112	0.0150	164	$1.0 \times 10^{-4}$
113	0.0135	165	$6.8 \times 10^{-5}$
114	0.0123	166	$4.4 \times 10^{-5}$
115	0.0115	167	$2.8 \times 10^{-5}$
116	0.0105	168	$1.8 \times 10^{-5}$
117	0.0099	169	$1.1 \times 10^{-5}$
		170	$7.2 \times 10^{-6}$

#### Independent yields

Independent fission yields were measured for several nuclides. These results are summarized in Table IV, along with the total chain yields (see Table III), the independent fractional chain yields, and the values of  $Z_p$  inferred from the measurements using a Gaussian charge dispersion curve with  $\sigma=0.56$ .<sup>53</sup> Measured values of  $Z_p$  are compared with predicted values on the basis of independent-yield systematics.<sup>50,51</sup>

Measured and predicted  $Z_p$  values generally agree, although the predicted values are approximately 0.1 charge unit smaller. The measured yield of  $^{132}\text{Cs}$ , about 60 times higher than expected, may be partly caused by the  $(n, 2n)$  reaction on Cs impurity in the  $^{240}\text{Pu}$ . The amount of  $^{132}\text{Cs}$  observed could be formed from 14 ppm Cs, but the actual level was not measured. The high yield of  $^{134}\text{Cs}$  could also be partly caused by the  $(n, \gamma)$  reaction.

#### Isomer ratios

Several partial isomer yields and isomer ratios were measured and are summarized in Table V. The estimated fractional production of each isomer pair from direct fission and from  $\beta$  decay is given.

The measured isomer ratio for  $^{115}\text{Cd}$  (0.0658  $\pm$  0.0020), primarily formed by  $\beta$  decay, is in agreement with the average value of approximately 0.071 found for the 14.8-MeV fission of five uranium isotopes.<sup>1</sup>

When the yield is primarily from fission ( $^{127}\text{Sn}$ ,  $^{130}\text{Sb}$ ,  $^{134}\text{Cs}$ ), the measured isomer ratio can be compared with recent theoretical calculations for the distribution of independent fission-product yields to isomeric states.<sup>54</sup> The calculated isomer ratios are 0.22 ( $^{127}\text{Sn}$ ), 0.75 ( $^{130}\text{Sb}$ ), and 1.59 ( $^{134}\text{Cs}$ ). The  $^{127}\text{Sn}$  and  $^{130}\text{Sb}$  calculated yields are in fair agreement with the experimental values; the disagreement in the case of  $^{134}\text{Cs}$  may be caused by preferential formation of  $^{134}\text{Cs}^f$  by the  $(n, \gamma)$  reaction on cesium impurity in the target (the measured independent yield of  $^{134}\text{Cs}$  is higher than expected).

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TABLE IV. Measured independent fractional chain yields (FCY) and a comparison of measured and predicted values of  $Z_p$ , the most probable charge for a given mass number.

Nuclide	Measured fission yield <sup>a</sup>	Total chain yield <sup>b</sup>	Independent FCY	Value of $Z_p$	
				Measured <sup>c</sup>	Predicted
<sup>96</sup> Nb	$(8.34 \pm 0.25) \times 10^{-5}$	0.0408	$(2.04 \pm 0.06) \times 10^{-3}$	$38.89 \pm 0.17$	$38.80 \pm 0.08$
<sup>110</sup> Ag <sup>m</sup>	$\leq 4.9 \times 10^{-6}$	0.0192	$\leq 2.6 \times 10^{-4}$	$\leq 44.56 \pm 0.21$	$44.58 \pm 0.16$
<sup>126</sup> Sb <sup>g</sup>	$(3.60 \pm 0.12) \times 10^{-3}$	0.0192	$0.188 \pm 0.006$	$\geq 50.01 \pm 0.05$ <sup>d</sup>	$49.84 \pm 0.12$
<sup>128</sup> Sn	$(9.94 \pm 0.31) \times 10^{-3}$	0.0261	$0.381 \pm 0.012$ <sup>e</sup>	$50.67 \pm 0.02$	$50.50 \pm 0.09$
<sup>130</sup> I <sup>g</sup>	$(2.09 \pm 0.09) \times 10^{-3}$	0.0375	$0.0557 \pm 0.0025$	$\geq 51.61 \pm 0.09$ <sup>d</sup>	$51.23 \pm 0.08$
<sup>132</sup> Cs	$(1.7 \pm 1.0) \times 10^{-5}$	0.0516	$(3.3 \pm 2.0) \times 10^{-4}$	$52.59 \pm 0.31$	$52.04 \pm 0.07$
<sup>134</sup> Cs	$(6.45 \pm 0.17) \times 10^{-4}$	0.0538	$0.0120 \pm 0.0003$	$53.24 \pm 0.14$	$52.91 \pm 0.07$
<sup>135</sup> Cs <sup>m</sup>	$(6.55 \pm 0.22) \times 10^{-4}$	0.0534	$0.0123 \pm 0.0004$	$\geq 53.24 \pm 0.14$ <sup>d</sup>	$53.32 \pm 0.07$
<sup>136</sup> Cs	$(5.03 \pm 0.15) \times 10^{-3}$	0.0515	$0.0977 \pm 0.0029$	$53.78 \pm 0.08$	$53.69 \pm 0.07$
<sup>160</sup> Tb	$\leq 1.5 \times 10^{-6}$	$5.10 \times 10^{-4}$	$\leq 2.9 \times 10^{-3}$	$\leq 62.96 \pm 0.16$	$62.89 \pm 0.13$

<sup>a</sup>Measured fission yield from Table II.<sup>b</sup>Total chain yield from Table III.<sup>c</sup>The "measured" value of  $Z_p$  is inferred from the measured independent FCY. The uncertainty given for the measured  $Z_p$  includes the uncertainty in the width of the Gaussian charge dispersion curve,  $\sigma = 0.56 \pm 0.06$ . This is by far the major part of the total uncertainty.<sup>d</sup>The result is a lower limit to  $Z_p$  since only the partial isomeric yield was measured.<sup>e</sup>This yield is a cumulative fractional chain yield.TABLE V. Measured isomer yields from fission of <sup>240</sup>Pu with 14.8-MeV neutrons.

Nuclide	Spin-parity	Fission yield	Isomer ratio ( <i>m/g</i> )	Fractional production <sup>a</sup>	
				Fission	$\beta$ decay
<sup>111</sup> Pd <sup>m</sup>	$\frac{11}{2}^-$	$9.9 \times 10^{-3}$ <sup>b</sup>	1.49 $\pm$ 0.27	0.15	0.85
<sup>111</sup> Pd <sup>g</sup>	$\frac{5}{2}^+$	$6.77 \times 10^{-3}$ <sup>c</sup>			
<sup>115</sup> Cd <sup>m</sup>	$\frac{11}{2}^-$	$7.18 \times 10^{-4}$ <sup>b</sup>	$0.0658 \pm 0.0020$	0.01	0.99
<sup>115</sup> Cd <sup>g</sup>	$\frac{1}{2}^+$	$0.0109$ <sup>b</sup>			
<sup>117</sup> Cd <sup>m</sup>	$\frac{11}{2}^-$	$2.57 \times 10^{-3}$ <sup>b</sup>	0.43 $\pm$ 0.12	0.18	0.82
<sup>117</sup> Cd <sup>g</sup>	$\frac{1}{2}^+$	$6.01 \times 10^{-3}$ <sup>b</sup>			
<sup>125</sup> Sn <sup>m</sup>	$\frac{3}{2}^+$	$7.6 \times 10^{-3}$ <sup>c</sup>	0.90 $\pm$ 0.09	0.48	0.52
<sup>125</sup> Sn <sup>g</sup>	$\frac{11}{2}^-$	$8.44 \times 10^{-3}$ <sup>b</sup>			
<sup>127</sup> Sn <sup>m</sup>	$\frac{3}{2}^+$	$2.1 \times 10^{-3}$ <sup>c</sup>	0.15 $\pm$ 0.23	0.83	0.17
<sup>127</sup> Sn <sup>g</sup>	$\frac{11}{2}^-$	$0.0143$ <sup>b</sup>			
<sup>128</sup> Sb <sup>m</sup>		$0.0150$ <sup>c</sup>	1.49 $\pm$ 0.12	0.48	0.52
<sup>128</sup> Sb <sup>g</sup>	$8^-$	$0.0101$ <sup>b</sup>			
<sup>130</sup> Sb <sup>m</sup>		$8.6 \times 10^{-3}$ <sup>c</sup>	0.50 $\pm$ 0.14	0.86	0.14
<sup>130</sup> Sb <sup>g</sup>		$0.0171$ <sup>b</sup>			
<sup>131</sup> Te <sup>m</sup>	$\frac{11}{2}^-$	$0.0154$ <sup>b</sup>	0.57 $\pm$ 0.04	0.57	0.43
<sup>131</sup> Te <sup>g</sup>	$\frac{3}{2}^+$	$0.0269$ <sup>c</sup>			
<sup>134</sup> Cs <sup>m</sup>	$8^-$	$2.23 \times 10^{-4}$ <sup>b</sup>	0.53 $\pm$ 0.03	1.00	0
<sup>134</sup> Cs <sup>g</sup>	$4^+$	$4.22 \times 10^{-4}$ <sup>b</sup>			

<sup>a</sup>The fractional production was estimated from the cumulative fractional chain yields of the isomer pair and its  $\beta$ -decay precursor.<sup>b</sup>Measured yield from Table II.<sup>c</sup>Yield estimated from the total chain yield (Table III), the cumulative fractional chain yield, and the measured yield of the other isomer.

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