# Comparison of Yields from Fission of  $^{233}$ U,  $^{234}$ U,  $^{235}$ U,  $^{236}$ U, and  $^{238}$ U with 14.8-Mev Neutrons\*

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We analyzed mass-yield distributions from fission of  $^{233}$ U,  $^{234}$ U,  $^{235}$ U,  $^{236}$ U, and  $^{238}$ U with 14.8-MeV neutrons to study the details of the fission distribution as a function of target mass number at a fairly low excitation energy. The fission yields of individual products, both cumulative and independent, vary smoothly and as expected with uranium target mass number. The over-all distribution of yields becomes significantly narrower as the uranium mass number is increased. The average neutron emission associated with the very-low-yield products is about 3.5, significantly less than the value of about 4.<sup>5</sup> for the fission fragments as a whole. This difference may be due to an increase in energy required for reactions leading to the lowyield products. The peak/valley ratio increases by a factor of more than <sup>2</sup> as the mass number is increased from 233 to 238, and decreases linearly with increasing excitation energy, with the even-mass and odd-mass uranium isotopes defining lines that are approximately parallel and separated by 1.<sup>4</sup> MeV. This difference can be explained either by an odd-even effect in the neutron-evaporation/fission competition as a function of target mass number, or by an odd-even effect in the ratio of symmetric to asymmetric fission as a function of mass number. One consequence of the rapid change in peak/valley ratio with mass number is that fission of  $^{232}$ U with 14.8-MeV neutrons would be expected to produce a flat-topped mass-yield distribution, with a peak/valley ratio of about unity, while fission of lower-mass uranium isotopes would be expected to be predominantly symmetric. The average value of  $\Delta Z_{b}/\Delta A$ was found to be  $-0.16 \pm 0.02$  for the shielded product <sup>96</sup>Nb and  $-0.20 \pm 0.01$  for <sup>136</sup>Cs. This difference may be due to the change in prefission neutron emission with uranium mass number. All of the available data on fractional chain yields for fission of <sup>236</sup>U\* at  $E^* = 6.4$  and 21.3 MeV were used to estimate  $\Delta Z_{\rho}(A)$  for  $\Delta E^*=15$  MeV. The scatter in the  $\Delta Z_{\rho}$  values is too large to allow a smooth curve to be drawn relating  $\Delta Z_b$  as a function of mass number. It was found, however, that  $\Delta Z_{\rho}$  values are larger on the average for heavy-fragment products ( $\Delta Z_{\rho} = 0.49$ )  $\pm 0.05$ ) than for light-fragment products ( $\Delta Z_{p} = 0.26 \pm 0.03$ ) over this range of excitation energy. The well-known  $Z_{\nu}(A)$  function for thermal-neutron fission of <sup>235</sup>U, together with the  $\Delta Z_{\nu}/\Delta A$ and  $\Delta Z_{\rm b}/\Delta E^*$  relationships reported here, can be used to estimate  $Z_{\rm b}$  values and fractional chain yields for fission of the various uranium isotopes with  $14.8-MeV$  neutrons.

#### INTRODUCTION

We have measured the distribution of fission products for 14.8-MeV neutron-induced fission of five isotopes of uranium, which allows us to examine the details of the mass-yield distribution as a function of uranium mass number at a fairly low excitation energy —around 20 MeV. This paper is based on recent results for fission of  $^{233}$ U (Ref. 1),  $^{234}$ U and  $^{236}$ U (Ref. 2), and on part of the extensive literature for  $^{235}$ U and  $^{238}$ U (Refs. 3-19). We also introduce some new and revised results for  $^{233}$ U fission.

Of the five long-lived isotopes of uranium,  $^{234}U$ ,  $^{236}$ U, and  $^{238}$ U have thresholds for fission with neutrons of about  $0.1-1$  MeV, while  $^{233}$ U and  $^{235}$ U are fissionable with thermal neutrons. The fission cross sections in the 8-15-MeV range are shown in Fig. 1. Smooth curves have been drawn through points taken from the tabulation of evaluated data

given by Hart.<sup>20</sup> The 14.8-MeV fission cross section drops from 2.4 b for  $^{233}$ U to 1.2 b for  $^{238}$ U.

In this paper we examine the variation in the following facets of the fission-product distribution as the uranium target is varied from  $^{233}$ U through  $^{238}$ U: the fission yields of individual products, the yields of the shielded products  $96$ Nb and  $136$ Cs, the ratio of asymmetric to symmetric fission as shown by the peak/valley ratio, and the shape of the fission distribution on the wings of the mass-yield curve. The role of neutron-emission/fission competition in determining the prefission neutron evaporation and hence the excitation energy is important in these studies.

Finally, we use charge-distribution systematics to estimate the change in  $Z_{\nu}$ , the most probable charge for a given fission-product mass number, with a change in the mass number of the target nuclide. This is then coupled with other data on the change in  $Z_{\rho}$  with excitation energy as a func-

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FIG. 1. The fission cross section for uranium isotopes in the energy range <sup>8</sup>—15 MeV.

estimating independent fractional chain yields in 14.8-MeV fission.

#### FISSION YIELD DATA

# A. <sup>233</sup>U Data

The <sup>233</sup>U fission-yield measurements in Ref. 1 were based on a measurement of the neutron fluence in the target foil, using yttrium monitor foils. A cross section for the monitor reaction  ${}^{89}Y(n, 2n)$ - $^{28}$ Y of 1.02 b was used, but the preferred value is now 1.06  $\pm$  0.05 b.<sup>21</sup> In Ref. 1 we also noted that the now  $1.06 \pm 0.05$  b. $^{21}$  In Ref. 1 we also noted that the area under each half of the mass-yield curve was about 1.08 (or  $8\%$  high), but we were unable to resolve the discrepancy. With the change in monitor cross section, the discrepancy has increased to 12%. In Ref. 2, we found a similar difference between results for  $236$ U fission based on yttrium monitor foils and on niobium monitor foils. This difference is thought to be due to the high threshold of the  $(n, 2n)$  reaction on  $^{89}Y$  (11.6 MeV), so that the shape of the excitation function in the region 12-15 MeV is very different from that for the fission reaction. The  $^{93}Nb(n, 2n)^{92}Nb$  excitation function, with a threshold of 8.9 MeV, is more

TABLE I. Corrected yields for fission of  $^{233}U$  with 14.8-MeV neutrons.

Product	Fission	Product	Fission
nuclide	yield <sup>a</sup>	nuclide	yield <sup>a</sup>
$^{66}{\rm Ni}$	$(7.1 \pm 0.7) \times 10^{-6}$	$140$ Ba	$0.0401 \pm 0.0016$
${}^{67}Cu$	$(1.64 \pm 0.19) \times 10^{-5}$	$141$ Ce	$0.042 \pm 0.003$
${}^{72}Zn$ b	$(1.35 \pm 0.06) \times 10^{-4}$	$143$ Ce	$0.0328 \pm 0.0017$
93y	$0.055 \pm 0.006$	$144$ Ce	$0.0249 \pm 0.0016$ <sup>d</sup>
95Zr	$0.052 \pm 0.002$	147 <sub>Nd</sub>	$0.0119 \pm 0.0008$
97Zr	$0.048 \pm 0.003$	$^{153}\mathrm{Sm}$	$(1.44 \pm 0.12) \times 10^{-3}$
96Nb	$(2.0 \pm 0.2) \times 10^{-4}$ c	$156$ $\rm Eu$	$(4.4 \pm 0.2) \times 10^{-4}$
$^{99}$ Mo	$0.038 \pm 0.003$	$^{159}\mathrm{Gd}$	$(1.07 \pm 0.11) \times 10^{-4}$
$105$ Rh	$0.0204 \pm 0.0019$	$160 \text{Th}$	$(3.0 \pm 0.8) \times 10^{-7}$ <sup>c</sup>
$^{112}Pd$	$0.0175 \pm 0.0011$	$^{161}$ Tb	$(4.7 \pm 0.3) \times 10^{-5}$
$^{111}$ Ag	$0.0164 \pm 0.0007$ <sup>d</sup>	$166_{\text{Dy}}$	$(2.4 \pm 0.2) \times 10^{-6}$
$115m + g$ Cd	$0.0155 \pm 0.0016$ <sup>d,e</sup>	$^{169}\mathrm{Er}$	$(8.4 \pm 0.6) \times 10^{-7}$
$132$ Te $^{136}$ Cs $^{137}$ Cs	$0.032 \pm 0.003$ $(9.2 \pm 0.4) \times 10^{-3}$ c $0.051 \pm 0.002$	172 <sub>Er</sub> $175$ Yb	$(1.81 \pm 0.15) \times 10^{-7}$ $(1.9 \pm 0.3) \times 10^{-8}$

<sup>a</sup> The experimental standard deviations given here do not include a systematic uncertainty of about  $6\%$  in the normalization of the mass-yield curve.

 $b$  The <sup>72</sup>Zn half-life was erroneously given as 2.45 days in Ref. 1. The correct value is 1.942 days.

<sup>c</sup> The independent fractional chain yields of <sup>96</sup>Nb, <sup>136</sup>Cs, and <sup>160</sup>Tb are  $(4.1\pm0.5)\times10^{-3}$ , 0.174 $\pm0.011$ , and  $(4.2\pm1.0)$  $\times 10^{-3}$ , respectively.

 $d$  The  $^{111}$ Ag,  $^{115m}$ Cd, and  $^{144}$ Ce fission yields were revised after a remeasurement of their counting efficiencies. The new counting efficiencies are the same as those used for the  $^{234}$ U and  $^{236}$ U results in Ref. 2.

<sup>e</sup> The measured  $^{115m}$  Cd/ $^{115g}$ Cd ratio is 0.075.

similar. Use of the  ${}^{89}Y(n, 2n) {}^{88}Y$  reaction to monitor the neutron fluence in the target apparently leads to low estimates of the number of fissions actually occurring, and hence to erroneously high values for the fission yields. The containers used for the  $^{233}U$ ,  $^{234}U$ , and  $^{236}U$  irradiations were more massive than those used for the  $^{235}$ U and  $^{238}$ U irramassive than those used for the countries of the necessity diations,<sup>3</sup> for example, because of the necessity of confining the intensely  $\alpha$ -active material. This may have resulted in a significant increase in fissions due to scattered neutrons.

We have chosen, then, to discard the results of the yttrium monitors given in Ref. l (as was also done for the  $^{236}U$  fission yields in Ref. 2), and to use instead the method of normalizing the area. under each half of the mass-yield curve to unity. This means that all of the  $233$ U fission yields as given in Ref. 1 should be reduced by the factor 1.08, after taking into account the contribution from fission of other isotopes of uranium in the target foils. The corrected yields are given in Table I.

Also included in Table I are new results for  $^{96}Nb$ ,  $^{136}Cs$ ,  $^{137}Cs$ , and  $^{156}Eu$ . These results are based on one additional  $14.8-MeV$  irradiation of  $^{233}U$  that was made after Ref. 1 was published. This was done primarily for the  $^{96}$ Nb and  $^{136}$ Cs independent yields so that they could be compared with similar data for the other uranium isotopes. The experimental details of the irradiation<sup>1</sup> and the radioactivity measurements<sup>1,2</sup> are the same as before and are not repeated here.

B. 
$$
^{234}
$$
U and  $^{236}$ U Data

The results given in Ref. 2 are used.

C. 
$$
^{235}
$$
U and  $^{238}$ U Data

The  $^{235}$ U and  $^{238}$ U fission yields that have been used in the following sections are summarized in Table II. The values are averages of the results given in the literature, together with some preliminary data' based on measurements with a Ge(Li) detector, both with and without chemical separation of individual products. The more recent results were generally given more weight in the averaging. We are also reporting values for the independent yields of  $^{96}$ Nb and  $^{160}$ Tb, based on one irradiation of  $^{235}$ U and  $^{238}$ U foils with 14.8-MeV neutrons. The experimental details are similar to those given before.<sup>3</sup> The number of fissions occurring in the uranium foils was calculated from the yields of the products  $147$ Nd and  $144$ Ce.

The fission yields given in Ref. 3 were based on the use of the  $^{89}Y(n, 2n)^{88}Y$  reaction to monitor the neutron fluence in the target foils. Despite the apparent difficulty with this method, the results have been used without further changes. The highyield products appear to be in good agreement with the normalized mass-yield curve and with the results of others.

TABLE II. Evaluated yields of selected products from fission of  $^{235}$ U and  $^{238}$ U with 14.8-MeV neutrons.

Product	235 <sub>11</sub>	$238$ U			
nuclide	fission <sup>a</sup>	Reference	fission <sup>a</sup>	Reference	
$^{66}{\rm Ni}$	$(2.8 \pm 0.3) \times 10^{-6}$	3, 4	$(8.5 \pm 0.9) \times 10^{-7}$	3	
${}^{67}Cu$	$(6.5 \pm 0.9) \times 10^{-6}$	3	$(1.4 \pm 0.4) \times 10^{-6}$	3	
${}^{72}Zn$	$(6.3 \pm 0.3) \times 10^{-5}$	$3 - 5$	$(3.0 \pm 0.4) \times 10^{-5}$	3	
95Zr	$0.050 \pm 0.005$	5,6	$0.049 \pm 0.002$	6, 7	
$^{97}Zr$	$0.051 \pm 0.005$	5, 6, 8	$0.051 \pm 0.002$	$6 - 10$	
96Nb b	$(3.9 \pm 0.2) \times 10^{-5}$	This work	$(1.07 \pm 0.09) \times 10^{-6}$	This work	
$^{99}$ Mo	$0.051 \pm 0.005$	$8 - 11$	$0.058 \pm 0.003$	$8 - 15$	
$^{111}$ Ag	$0.0109 \pm 0.0010$	5, 6, 8, 16	$0.0106 \pm 0.0006$	6, 8, 9, 13, 14, 16, 17	
$^{112}\mathrm{Pd}$	$(9.2 \pm 1.3) \times 10^{-3}$	5, 8, 16	$0.0114 \pm 0.0006$	7, 9, 13, 16	
$115m + g$ Cd	$0.0112 \pm 0.006$	$6-8, 16, 18$	$(8.3 \pm 0.4) \times 10^{-3}$	$6-8$ , 13, 14	
$136C_S$ b	$(2.47 \pm 0.12) \times 10^{-3}$	7	$(2.19 \pm 0.10) \times 10^{-4}$	7	
140 <sub>Ba</sub>	$0.0425 \pm 0.0017$	3, 5, 6, 8	$0.0454 \pm 0.0023$	$3, 6, 8-10, 14, 17$	
$144$ Ce	$0.031 \pm 0.003$	5,8	$0.0367 \pm 0.0018$	7, 8, 14, 19	
$^{147}\mathrm{Nd}$	$0.0164 \pm 0.0011$	3, 5	$0.0210 \pm 0.0010$	3, 7, 10, 14	
$^{153}$ Sm	$(2.2 \pm 0.2) \times 10^{-3}$	3, 5	$(4.2 \pm 0.4) \times 10^{-3}$	3, 14	
$^{156}\mathrm{Eu}$	$(5.5 \pm 0.3) \times 10^{-4}$	5, 7, 8, 16	$(1.07 \pm 0.05) \times 10^{-3}$	7, 8, 14, 16	
$^{159}\mathrm{Gd}$	$(1.27 \pm 0.13) \times 10^{-4}$	3	$(2.6 \pm 0.3) \times 10^{-4}$	3	
$160$ Th $b$	$(2.2 \pm 0.2) \times 10^{-7}$	This work	$(2.1 \pm 0.2) \times 10^{-7}$	This work	
$161$ Tb	$(5.4 \pm 0.4) \times 10^{-5}$	3, 5, 7	$(8.5 \pm 0.4) \times 10^{-5}$	3, 7	

<sup>a</sup> The uncertainties given with each value are estimates of the precision of the measurement, and (probably) do not include any systematic uncertainty in the absolute normalization of the mass-yield curve,

<sup>b</sup> The independent fractional chain yields of <sup>96</sup>Nb, <sup>136</sup>Cs, and <sup>160</sup>Tb are, respectively: for <sup>235</sup>U fission (7.4 ± 0.5) × 10<sup>-4</sup>. 0.048 ± 0.003, and  $(2.7 \pm 0.3) \times 10^{-3}$ ; for <sup>238</sup>U fission  $(2.1 \pm 0.2) \times 10^{-5}$ ,  $(3.7 \pm 0.3) \times 10^{-3}$ , and  $(1.29 \pm 0.15) \times 10^{-3}$ .

### PEAK-TO-VALLEY RATIO

The peak/valley ratios for fission of the five uranium isotopes with 14.8-MeV neutrons are plotted in Fig. 2. The peak/valley ratios were taken from smoothed plots of the mass-yield curves. These ratios are not dependent on the normalization of absolute fission yields. They are limited in accuracy only by the measurement of isotope ratios. The peak/valley ratio shows a rather large change from a value of  $3.2$  for  $233$ U fission to  $6.8$  for  $^{238}$ U fission.

The peak/valley ratio in low-energy fission is known to be strongly dependent on the excitation energy and is a function of the ratio of asymmetricto-symmetric fission. It varies, for example, from a value of about 600 for thermal-neutron fission of  $^{235}$ U to a value of 4.9 for 14.8-MeV neutron fission. In Fig. 3 we have plotted the peak/valley ratio as a function of the excitation energy in the compound nucleus. The points for the even-mass and odd-mass uranium target isotopes fall on two lines separated by 1.44 MeV. The lines are approximately parallel and show the expected decrease in peak/valley ratio with increase in excitation energy. The rate of change of the peak/ valley ratio is about  $-6.2 \text{ MeV}^{-1}$ .

At an excitation energy of about 20 MeV, the reactions  $(n, f)$ ,  $(n, nf)$ , and  $(n, 2nf)$  are expected to take place. The degree to which each reaction occurs for the various uranium isotopes is not



FIG. 2. The peak/valley ratio for fission of the different isotopes of uranium with 14.8-MeV neutrons. The line through the points was drawn as a visual aid.

known with any certainty. However, it is reasonably well established that the neutron-emission/ fission competition changes as a result of an exponential increase in the ratio of neutron evaporation width to fission width  $(\Gamma_n/\Gamma_f)$  with increasing mass number. $2^{2-24}$  This would lead, as a result, to a lowering of the average excitation energy for fission of  $^{238}U$  compared to  $^{233}U$ . For  $^{235}U$  the three reactions  $(n, f)$ ,  $(n, nf)$ , and  $(n, 2nf)$  are expected<sup>24</sup> to occur roughly in the ratio  $0.3:0.2:0.5$ , and, for  $^{238}U$ , in the ratio 0.1:0.2:0.7. In contrast, the recent analysis by Davey<sup>25</sup> of the fission excitation functions of  $^{235}$ U and  $^{238}$ U indicates that the ratios for  $^{235}U$  are (very approximately) 0.0:  $0.6:0.4$ , and, for  $238$ U,  $0.4:0.4:0.2$ .

There are two possible explanations for the striking difference between the lines for the even-mass and odd-mass isotopes in Fig. 3. The first is that there may be an odd-even effect in the neutronevaporation width as a function of target mass number, such that even-mass uranium isotopes have a higher  $\Gamma_n$  than odd-mass isotopes, because of the higher level density in the even-odd product nuclei. This would tend to cause fission of the odd-mass target isotopes to occur at a lower average excitation energy than for the even-mass target isotopes. In essence, this means that we should really plot the peak/valley ratio against the average excitation energy of the fissioning species, rather than against the excitation energy of the initial compound nucleus.



FIG. 3. The peak/valley ratio as a function of the excitation energy in the compound nucleus. The points are plotted for the target uranium isotope.

The second explanation is that there may be an odd-even effect in the ratio of symmetric-to-asymmetric fission as a function of target mass number. Asymmetric fission is presumably a lowenergy process, while symmetric fission occurs via higher-energy channels that are opened at higher excitation energies. Odd-even nuclei are known to have a higher level density than even-even nuclei, and therefore might be expected to undergo symmetric fission to a greater extent. In Fig. 3 we see that, at a given excitation energy, the evenmass target isotopes have a higher symmetric fission component than do the odd-mass isotopes. One might infer, then, that fission of the evenmass target isotopes occurs mainly from oddmass nuclei and that fission of the odd-mass target isotopes occurs mainly from even-mass nuclei. In other words, our data tend to support the  $(n, f)$ or, less likely, the  $(n, 2nf)$  reaction as being the most important.

It is also interesting to note that the fission of  $^{232}$ U with 14.8-MeV neutrons, leading to a  $^{233}$ U compound nucleus with  $E^* = 20.5$  MeV, would be expected to have a peak/valley ratio of about unity. If the trend shown in Fig. 3 continues, other isotopes of uranium with lower mass numbers should show mainly symmetric fission.

## COMPARISON OF FISSION YIELDS

We compared the fission yields of a selected list of products as a function of the target uranium



FIG. 4. Comparison of the  $^{66}$ Ni,  $^{67}$ Cu, and  $^{72}$ Zn fission yields as a function of uranium mass number.

mass number, to determine whether or not the yields vary smoothly with uranium mass. The fission yields of  $^{66}\rm{Ni},~^{67}\rm{Cu},~^{72}\rm{Z}$ n,  $^{95}\rm{Z}$ r,  $^{97}\rm{Z}$ r,  $^{99}\rm{Mo}$  $^{111}$ Ag,  $^{112}$ Pd,  $^{115}$ Cd,  $^{140}$ Ba,  $^{144}$ Ce,  $^{147}$ Nd,  $^{153}$ Sm,  $^{156}$ Eu,  $^{159}$ Gd, and  $^{161}$ Tb are plotted in Figs. 4-8. The data from this paper and from Refs. 1-3 and 7 are all on consistent atom scales. The error bars shown in the figures are those given in the original references. To be strictly accurate in this comparison, that part of the uncertainty in the data due to uncertainty in counting efficiencies should be omitted, and the uncertainty due to absolute fission-yield normalization should be added. However, these two uncertainties are about the same, and the changes would tend to cancel each other.

Two of the best measured yields are those of  $^{140}$ Ba and  $^{147}$ Nd, shown in Fig. 7. The yields of both products vary smoothly with uranium mass number. No evidence is found for any odd-even variation, and we conclude that the absolute yield normalization for each uranium target is reasonable within experimental uncertainty.

The yields of the various products shown in Figs. 4-8 exhibit the expected trends with varying ura-



FIG. 5. Comparison of the  $^{95}Zr$ ,  $^{97}Zr$ , and  $^{99}Mo$  fission yields as a function of uranium mass number.

nium mass number. Both fragments shift to hi ass numbers as the uranium mass is increased, mass numbers as the uranium mass is increased<br>with the light fragment shifting more than the h<br>fragment. This wealth is a house descense is with the light fragment shifting more than the hea<br>fragment. This results in a large decrease in the<br>siable of  $^{66}$ N'.  $^{67}$ C. N<sup>72</sup>C. regiment. This results in a rarge decrease in the<br>yields of  $^{66}Ni$ ,  $^{67}Cu$ , and  $^{72}Zn$  and a small increase in the yields of  $^{153}\mathrm{Sm},~^{156}\mathrm{Eu},~^{159}\mathrm{Gd},~$  and  $^{161}\mathrm{Tb}$  as ings of the mass-yield distribution shift to igher masses. In addition, the yields of the proc ucts on the peaks of the distribution change in a  $\mathop{\mathrm{corresponding}}\nolimits\mathop{\mathrm{fashion}}\nolimits.$  The yields of the vall $\mathop{\mathrm{d}}\nolimits$ corresponding fashion. The<br>products <sup>111</sup>Ag, <sup>112</sup>Pd, and <sup>1</sup> products<br>in the previ error in the counting efficiency  $5$ U fission may be due to in peak/valley ratio. The apparent disagreement

of all products vary smoothly as a function of the vielas e can make the generalization that the yields uranium target mass.

amum target mass.<br>A summary of the <sup>115</sup>Cd isomer ratio measure ments is given in Table III. There is no evident correlation of isomer ratio with mass numbe probably because of the multiplicity of fissioning prostasty socialist of the matchprietry of fissioning<br>nuclei of different spins and parities. The average  $5<sup>m</sup>$  Cd<sup>/115</sup><sup>s</sup>Cd ratio is 0.071.



FIG. 6. Comparison of the  $^{111}\text{Ag},\ ^{112}\text{Pc}$ ion yields as a function of uranium mass numbe is



FIG. 7. Comparison of the  $^{140}\text{Ba}$ ,  $^{144}\text{Ce}$ , and  $^{147}\text{Nd}$  fis sion yields as a function of uranium mass number.

#### WINGS OF THE MASS-YIELD CURVES

In Refs. 1–3 we made use of a Gaussian function tted to the yields of products on the wings of the ntied to the yields of products on the<br>mass-yield curve to aid in the estimat of unmeasured product

yield = 
$$
\frac{\text{area} \times \text{exp}\{-0.5[(A - A_0)/\sigma]^2\}}{\sigma(2\pi)^{0.5}}
$$
.



FIG. 8. Comparison of the  $^{153}$ Sm,  $^{156}$ Eu,  $^{159}$ Gd, and rich 3. Comparison of the ""Sm, ""Eu, ""Gd, and<br>Tb fission yields as a function of uranium mass numb

Uranium target	Measured $115mCd/115sCd$ ratio <sup>a</sup>	
233 <sub>11</sub>	$0.075 \pm 0.001$	
$234$ <sub>TT</sub>	$0.067 \pm 0.002$	
$235$ <sup>T</sup>	$0.064 \pm 0.003$	
$236$ <sub>TT</sub>	$0.071 \pm 0.002$	
238 <sub>T1</sub>	$0.075 \pm 0.002$	

TABLE III. The  $^{115m}$  Cd/ $^{115g}$ Cd isomer ratio in the fission of uranium with 14.8-MeV neutrons.

 $^a$  An uncertainty of about 10% in the counting efficiencies has not been included.

The parameters  $\sigma$ ,  $A_0$ , and area are calculated by a least-squares procedure in which the data are weighted by the reciprocal of the square of their standard deviations. Reflected data points were not used in the calculations. The final values of the three parameters, including yield revisions given in this paper, are summarized in Table IV.

The value of  $\sigma$ , the width parameter of the Gaussian curve, decreases smoothly with increasing mass number. Thus we find that, in going from fission of  $^{233}U$  to  $^{238}U$ , the over-all distribution of yields becomes significantly narrower, as well as deeper (higher peak/valley ratio).

The center-of-the-mass distribution of the verylow-yield products shifts uniformly toward higher mass numbers as the uranium target mass is increased. The position of the center is given by the equation  $A_0 = 0.506T - 2.70$ , where T is the uraniumtarget mass number. This equation was obtained by a least-squares fit to the data in Table IV. The average numbers of neutrons emitted per fission for the low-yield products can be calculated from the positions of the center mass numbers and are ' $3.4 \pm 0.2$  ( $233$ U),  $3.7 \pm 0.1$  ( $234$ U),  $3.5 \pm 0.2$  ( $235$ U),

TABLE IV. Parameters for the Gaussian function fitted to the wings of the mass-yield curves.

Target nuclide	Area	Center mass number	σ
233 <sub>TT</sub>	$53 \pm 11$	$115.29 \pm 0.11$	$9.85 \pm 0.10$
$234$ TT	$89 \pm 19$	$115.64 \pm 0.07$	$9.55 \pm 0.10$
235 <sub>TT</sub>	$86 \pm 24$	$116.24 \pm 0.11$	$9.47 \pm 0.12$
236 <sub>TT</sub>	$113 \pm 16$	$116.80 \pm 0.05$	$9.35 \pm 0.06$
238 <sub>TT</sub>	$145 \pm 25$	$117.68 \pm 0.09$	$9.21 \pm 0.07$

3.4  $\pm$  0.1 (<sup>236</sup>U), and 3.6  $\pm$  0.2 (<sup>238</sup>U). These are all about 1 less than the measured average values of about 1 less than the measured average values o<br>about 4.5 for 14.8-MeV fission.<sup>25</sup> This differenc is perhaps a result of an increased energy requirement for reactions leading to very-low-yield fragments, compared to those leading to high-yield fragments. This would result in a lower excitation energy and less neutron emission associated with such low-yield products.

# CHARGE DISTRIBUTION IN FISSION

### A. Variation of  $Z_{p}$  with A

We measured the yields of two shielded products for fission of each of the five isotopes of uranium. The independent fractional chain yields of  $^{96}$ Nb and  $136$ Cs are summarized in Table V and are plotted against the uranium mass number in Fig. 9. The fractional chain yields are essentially isotope ratios and are independent of the absolute fissionyield normalization. The  $^{96}$ Nb and  $^{136}$ Cs yields are found to be rapidly, but smoothly, varying functions of the target mass number.





The values of  $Z - Z_p$  have been calculated for each <sup>96</sup>Nb and <sup>136</sup>Cs datum by assuming a Gaussian charge-dispersion curve with  $\sigma$  =0.56. This "standard" charge-dispersion curve has been found to be in agreement with most low-energy fission be in agreement with most low-energy fission<br>data.<sup>26</sup> The values of  $Z - Z_p$  and of  $\Delta Z_p / \Delta A$  are given in the last columns of Table V. The average value of  $\Delta Z_{p}/\Delta A$  is  $-0.16\pm0.02$  for <sup>96</sup>Nb and  $-0.20 \pm 0.01$  for  $^{136}$ Cs.

A formula for estimating the change in  $Z_{\rho}$  due to a change in mass, charge, or excitation energy of the fissioning nucleus was first given by Coryell, Kaplan, and Fink<sup>27</sup>:

$$
\Delta Z_{\pmb{\rho}}(A) = 0.5(Z_c-92) - 0.21(A_c-236) + 0.19(\nu_T-2.5),
$$

where the subscript  $c$  refers to the compound nucleus, and  $\nu<sub>r</sub>$  is the total neutron emission. The intent is to use the well-known  $Z_{p}(A)$  function for thermal-neutron fission of  $^{235}U$  and calculate adjustments to the  $Z_{\rho}$  values for other fissioning systems. One difficulty with this method is that the three right-hand terms in the equation above are not strictly separable. A change in A with constant Z probably leads to a change in the excitation energy of the over-all fissioning species (and hence in  $\nu_r$ ), because of both the odd-even effect on the neutron binding energy and the change in prefission neutron emission. The value of  $\Delta Z_{\rm A}/\Delta A$ 



FIG. 9. The independent fractional chain yields of  $^{96}Nb$ and  $136$ Cs as a function of uranium mass number.

that we find for  $^{136}Cs$  (-0.20  $\pm$  0.01) agrees well with that we find for <sup>136</sup>Cs (–0.20±0.01) agrees well with<br>the value proposed by Coryell, Kaplan, and Fink.<sup>27</sup> The value for  $^{96}$ Nb (-0.16 ± 0.01) is not in agreement, however, and this difference is probably due to the difference in  $\Delta Z_{\phi}/\Delta E^*$  for the two products, as shown later in this paper. The total change in  $Z_{\rho}$ between  $^{233}$ U and  $^{238}$ U is 0.82 for  $^{96}$ Nb and 0.98 for  $^{136}$ Cs. The difference of 0.16 could be due to a further lowering of the  $Z_p$  value for mass 136 relative to that for mass 96, if the average fission event for <sup>238</sup>U occurs at a lower excitation energy than for  $233$ U.

# B. Variation of  $Z_{p}$  with Excitation Energy

It has been shown before<sup>28, 29</sup> that the rate of change of  $Z_{p}$  with excitation energy,  $dZ_{p}/dE^{*}$ , is greater for heavy-fragment nuclides on the average than for light-fragment nuclides. This implies that, as the excitation energy is increased, neutron emission increases more from the heavy fragment than from the light fragment. This has been substantiated more recently, for example, by Ume-'zawa, $^{30}$  who measured  $\Delta Z_{\rho}/\Delta E^{*}$  for several nuclides from proton-induced fission of  $^{238}$ U, and by<br>Burnett *et al*.,<sup>31</sup> who measured the change in Burnett  ${et}$   ${al.},^{31}$  who measured the change in prompt-neutron emission with energy for protoninduced fission of  $^{233}U$ .

The method used in Ref. 28 for obtaining the value of  $dZ_{p}/dE^{*}$  as a function of mass number was to use the difference in  $Z_{p}$  values calculated from measured independent and cumulative fractional chain yields for fission of  $236$  U\* at excitation energies of 6.4 and 21.0 MeV. We assume that the width of the charge-dispersion curve is constant for each mass number over this range of energies (but not that it is constant for all mass numbers). An up-to-date version of this comparison of  $Z_{\rho}$ values is given in Table VI. The  $Z_p$  values for thermal-neutron fission of <sup>235</sup>U were taken from Ref. 26. The fractional-chain-yield data at  $E^*$ =21.3 MeV from which  $Z_p$  values were calculated are listed in the table. A charge-dispersion curve with  $\sigma$  =0.56 was used. The uncertainties in the final  $\Delta Z_{\rho}$  values include an estimate of the uncertainty in the  $Z_p$  function for thermal-neutron fission of about  $\pm 0.05$  to  $\pm 0.15$ , depending on the amount of data available for each isobaric mass chain, and the uncertainty in the  $Z_{p}$  value for  $E^{*}$ =21.<sup>3</sup> MeV due to the uncertainty in the measured yield.

An examination of Table VI reveals that, indeed, the  $\Delta Z_{p}$  values for  $\Delta E^{*}$  = 15 MeV are larger for heavy-fragment nuclides on the average. We attempted to fit a smooth curve to the data points in a plot of  $\Delta Z_{\rho}$  versus mass number, but found that the data scatter too badly to permit a reasonable fit. This may be due in part to the inherent difficulty in making accurate independent yield measurements by radiochemical methods. We had hoped to be able to fit a curve to the  $\Delta Z_{\rho}$ -massnumber data, similar in shape to that found by number data, similar in shape to that found by<br>Burnett *et al*.<sup>31</sup> for the change in prompt-neutr emission with excitation energy. A peak in the value of  $\Delta Z_{b}/\Delta E$  should occur at about mass 130 because of the reduction of shell effects with increasing excitation energy that leads to a filling in of the dip in the prompt-neutron-emission curve<br>for thermal-neutron fission.<sup>32</sup> for thermal-neutron fission.

We are left then with the simple alternative of using the average values of  $\Delta Z_b$  for  $\Delta E^*$  = 15 MeV:  $0.26 \pm 0.03$  (light fragment) and  $0.49 \pm 0.05$  (heavy fragment). The corresponding values of  $dZ_{\alpha}/dE$ are  $0.0176 \text{ MeV}^{-1}$  (light) and  $0.0333 \text{ MeV}^{-1}$  (heavy) These averages provide the best method of estimating the change in  $Z_{p}$  with excitation energy for any fissioning system until more experimental data become available.

#### SUMMARY

We have collected and analyzed the mass-yield data available for fission of  $^{233}U$ ,  $^{234}U$ ,  $^{235}U$ ,  $^{236}U$ , and  $^{238}$ U with 14.8-MeV neutrons. The fission yields of individual products, both cumulative and independent, vary smoothly, and in the expected manner with uranium target mass number. The peak/valley ratio increases by over a factor of 2 in going from  $^{233}U$  fission to  $^{238}U$  fission. The peak/ valley ratio decreases linearly with increasing excitation energy in the 19-22-MeV range, with the even-mass and odd-mass uranium isotopes defining parallel lines separated by 1.4 MeV. This difference can be explained by an odd-even effect in the degree of prefission neutron emission that would lead to average excitation energies higher by 1.4 MeV for the odd-mass uranium target nuclides. An alternative explanation is that there might be a predominance of the  $(n, f)$  or  $(n, 2nf)$ reaction over the  $(n, nf)$  reaction, based on: (1) the

TABLE VI. Change in  $Z_p$  for fission of <sup>236</sup>U\* at  $E^*=6.4$  and 21.3 MeV.

	Fractional		Value of $Z_p$		
Nuclide	chain yield <sup>a</sup>	Reference	$E^* = 21.3 \text{ MeV}^b$	$E^* = 6.4$ MeV <sup>c</sup>	$\Delta Z_{p}$
${}^{82}\text{Br}$	$(2.4 \pm 0.7) \times 10^{-3}$	8, 29, d	32.92	32.75	$0.17 \pm 0.12$
$^{86}\mathrm{Rb}$	$(4.2 \pm 1.3) \times 10^{-4}$	29, e, f	34.63	34.40	$0.23 \pm 0.11$
90Y	$(2.2 \pm 0.4) \times 10^{-4}$	28	36.53	36.06	$0.47 \pm 0.06$
$^{91}\mathrm{Kr}$	$0.36^{+0.02}_{-0.01}*$	g	36.70	36,46	$0.24 \pm 0.05$
$^{92}\mathrm{Kr}$ $^{92}Y$	$0.156\substack{+0.013 \\ -0.005}$ $(7.4 \pm 1.7) \times 10^{-3}$	g 28	37,07) 37.14)	36.86	$0.23 \pm 0.06$
$^{93}$ Kr	$0.039^{+0.005}_{-0.002}$ *	g	37.49	37,27	$0.22 \pm 0.05$
$^{96}Nb$	$(7.4 \pm 0.5) \times 10^{-4}$	This work	38.72	38,47	$0.25 \pm 0.05$
97Nb	$0.069 \pm 0.015$	h	39.67	38.87	$0.80 \pm 0.08$
112Ag	$0.051 \pm 0.006$	h	45.59	44.70	$0.89 \pm 0.11$
128 <sub>I</sub>	$(1.9 \pm 0.2) \times 10^{-3}$	29	50.88	49.66	$1,22 \pm 0.10$
130 <sub>T</sub>	$0.029 \pm 0.003$	29	51.44	50.39	$1.05 \pm 0.10$
132 <sub>T</sub>	$0.16 \pm 0.01$	i	51.95	51,21	$0.74 \pm 0.05$
134 <sub>T</sub>	$0.43 \pm 0.02$	i	52.44	52.12	$0.32 \pm 0.06$
$^{135}\text{Xe}$	$0.305 \pm 0.03$	29	53,23	52.56	$0.67 \pm 0.07$
136 <sub>Cs</sub>	$0.048 \pm 0.003$	7	53,57	52.98	$0.59 \pm 0.05$
$^{138}\mathrm{Xe}$	$0.75 \pm 0.02*$	35	54,12	53.79	$0.33 \pm 0.10$
$^{139}\mathrm{Xe}$	$0.429 \pm 0.009*$	g	54.60	54,19	$0.41 \pm 0.05$
140 <sub>xe</sub>	$0.204^{+0.008}_{-0.006}$ *	g	54,96)		
$140$ La	$(7.2 \pm 0.5) \times 10^{-3}$	28	55,13	54.59	$0.46 \pm 0.09$
$^{141}$ Xe	$0.056^{+0.006}_{-0.002}$ *	g	55.39	54.99	$0.40 \pm 0.05$
160 <sub>Tb</sub>	$(2.7 \pm 0.3) \times 10^{-3}$	This work	62.94	62,47	$0.47 \pm 0.15$

<sup>a</sup> Fractional chain yield for fission of <sup>236</sup>U\* at  $E^*=21.3$  MeV. Values with an asterisk are cumulative yields; others are independent yields.

 ${}^{\text{b}}Z_p$  value obtained by using a charge-dispersion curve with  $\sigma = 0.56$ .

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assumption that the symmetric fission mode may be favored in odd-even nuclei and (2) the observation that even-even uranium target nuclides have a higher symm etric fission component than do even-odd target nuclides at the same excitation energy.

From an analysis of the shapes of the wings of the mass-yield curves, we found that the over-all distribution of yields becomes significantly narrower as the uranium target mass is increased. The variation is so smooth that it seems unlikely to be due completely to a decrease in average excitation energy, because of the large odd-even variation with neutron binding energy. It could perhaps be due also to a gradual change in the degree of mixing of the various reactions  $(n, f)$ ,  $(n, nf)$ , and  $(n, 2nf)$ , such that fission of  $^{238}U$ , for example, might occur more predominantly by a single reaction, while fission of lighter-mass isotopes might occur more from a mixture of reactions. Fission of a variety of isotopes would be expected to lead to a broader distribution than fission of a single species.

The center-of-the-mass distribution of low-yield products shifts uniformly toward higher mass numbers with increasing uranium mass. The average neutron emission associated with the very-lowyield products is about one less than that for the high-yield products. This may be associated with

- \*Work performed under the auspices of the U. S. Atomic Energy Commission.
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an increased energy requirement for the rare fission reactions leading to the low-yield products.

The value of  $\Delta Z_{p}$  per unit change in target mass number was  $-0.16 \pm 0.02$  for  $^{96}$  Nb and  $-0.20 \pm 0.01$  for  $^{136}Cs$ . This difference can be explained by a decrease in average excitation energy as the uranium target mass is increased, because of an increase in prefission neutron emission. We attempted to obtain the best correlation of  $\Delta Z_{\rho}/\Delta E^*$  with fragment mass number for the excitation energy range 6-21 MeV. Because of the scatter in the experimental data, we concluded that it is probably best to use the average values of  $\Delta Z_{\rho}$  for  $\Delta E$  $= 15$  MeV of  $0.26 \pm 0.03$  for light-fragment products and  $0.49 \pm 0.05$  for heavy-fragment products.

The estimation of  $Z_{\rho}$  values for 14.8-MeV fission of the various uranium isotopes is best done by starting with the well-known  $Z_{p}(A)$  function for thermal-neutron fission of  $^{235}U$  and using the average values of  $\Delta Z_{p}$  given above for each fragment to calculate  $Z_{p}(A)$  for 14.8-MeV fission of <sup>235</sup>U. The values of  $\Delta Z_{b}$  per unit change in target mass number, -0.<sup>16</sup> for light fragments and -0.<sup>20</sup> for heavy fragments, can be used to obtain  $Z_{\rho}(A)$  values for other isotopes of uranium. Independent fractional chain yields can then be estimated for a charge-dispersion curve with  $\sigma = 0.56 \pm 0.06$ . This method was used for estimating total chain yields for  $^{234}$ U and  $^{236}$ U fission in Ref. 2.

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#### PHYSICAL REVIEW C VOLUME 6, NUMBER 5 NOVEMBER 1972

# $^{107}$ Ag(d, p)<sup>108</sup>Ag and <sup>109</sup>Ag(d, p)<sup>110</sup>Ag Reactions<sup>\*</sup>

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The level structures of  $^{108}$ Ag and  $^{110}$ Ag have been investigated via the  $(d, p)$  stripping reaction on isotopically enriched targets of  $^{107}$ Ag and  $^{109}$ Ag at an incident deuteron energy of 10.0 MeV. The over-all experimental resolution was approximately 20 keV. Proton groups leading to 57 states in  $108\text{Ag}$  with excitation energies up to 2.78 MeV and 37 states in  $110\text{Ag}$  with excitation energies up to 1.66 MeV have been identified. Orbital angular momentum transfer values and spectroscopic factors have been extracted for 19 states in  $^{108}$ Ag and for 23 states in <sup>110</sup>Ag using the zero-range distorted-wave Born approximation including an approximation for the nonlocality of the optical potential and a finite-range correction. In  $^{108}Ag$ , six  $l = 0$ , nine  $l = 2$ , one  $l = 4$ , one admixture of  $l = 0$  and  $l = 2$ , and two admixtures of  $l = 2$  and  $l = 4$  are assigned. In <sup>110</sup>Ag, six  $l = 0$ , sixteen  $l = 2$ , and one admixture of  $l = 2$  and  $l = 0$  are assigned. It is possible that a small fraction of  $l = 2$  transitions in <sup>110</sup>Ag are masked with  $l = 4$  transitions. On this assumption, the effective numbers of neutrons outside the closed  $g_{9/2}$  orbital in  $108\text{Ag}$  and  $110\text{Ag}$  are consistent with the expected values. The summed strengths of the single-particle orbitals are consistent with those of neighboring isotones.

#### I. INTRODUCTION

The deuteron stripping reaction has been extensively used to obtain nuclear spectroscopic information. Most of these investigations, however, have been restricted to even-even target nuclei. The shapes of the angular distributions are uniquely related to the orbital angular momentum transfer to the target nucleus,<sup>1</sup> and since the ground-stat spin is  $0^+$ , the final-state spin of the residual nucleus is known within limits of  $\pm \frac{1}{2}$ . It is natural to extend these investigations to odd-A target nuclei with an even number of neutrons. The  $(d, p)$ angular distributions are still characteristic of the l-value transfer to the target nucleus, but except for the s-wave transfer, the simplicity of restricting the choice of the final-state spin to two possible values only is lost. It is, however, of interest to study the effect of the odd proton on the spectroscopic factors of the neutron singleparticle states.

In this paper we report the results of our investigation of the level structures of  $^{108}$ Ag and  $^{110}$ Ag via the  $(d, p)$  stripping reaction using <sup>107</sup>Ag and

<sup>109</sup>Ag targets ( $Z = 47$ ), respectively. A comparison of the level structures of the isotopic nuclei  $^{108}$ Ag and  $^{110}$ Ag with the structure information available on the respective isotonic nuclei  $^{107}Pd$  (Ref. 2),  $^{109}$ Pd (Ref. 2), and  $^{113}$ Sn (Refs. 3, 4) should yield information about the order and the degree of filling of both proton and neutron single-particle orbitals in the mass neighborhood under consideration.

The  $(d, p)$  reaction on the stable Ag isotopes has not been extensively investigated. Earlier experiments of Sperduto<sup>5</sup> and of Mazari<sup>5</sup> were restricted to the determination of the excitation energies of low-lying states of <sup>108</sup>Ag and <sup>110</sup>Ag, and no attempt was made to extract the l-value transfers. Recently, the  $(d, p)$  stripping reaction has been studied by Lopez<sup>6</sup> at an incident deuteron energy of 7.<sup>5</sup> MeV. Shugart, Curry, Lock, Moore, and Riley' have studied the isobaric analogs of the lowlying states of <sup>108</sup>Ag and <sup>110</sup>Ag by means of proton elastic scattering from <sup>107</sup>Ag and <sup>109</sup>Ag targets. It was found that the weak-coupling approximation gives a reasonably good description of the lowlying states of the parent nuclei. A study of the  $(d, p)$  reactions on the <sup>107</sup>Ag and <sup>109</sup>Ag isotopes in