

## Product yields for the photofission of $^{235}\text{U}$ with 12-, 15-, 20-, 30-, and 70-MeV bremsstrahlung

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The mass distributions for the photofission of  $^{235}\text{U}$  using bremsstrahlung with end-point energy of 12, 15, 20, 30, and 70 MeV were determined by  $\gamma$  spectrometry of fission product catcher foils. Smoothly varying curves without fine structure were obtained. A third hump in the symmetric region is not present. Several fractional independent chain yields in the mass region 126–140 were measured and the corresponding most probable charges  $Z_p(E_e)$  were deduced. A comparison with the charge expected from the unchanged charge density hypothesis is made and an influence of the 50-proton shell on the behavior of the  $Z_p$  function is observed. Except in this mass region the determined  $Z_p(E_e)$  values are very well described by the empirical relation of Nethaway. From the isomeric ratios of  $^{126}\text{Sb}^g$ - $^{126}\text{Sb}^m$ ,  $^{128}\text{Sb}^g$ - $^{128}\text{Sb}^m$ , and  $^{131}\text{Te}^g$ - $^{131}\text{Te}^m$  average initial fragment spins are calculated using a statistical model analysis. The values increase with increasing end-point energy of the bremsstrahlung.

NUCLEAR REACTIONS, FISSION  $^{235}\text{U}(\gamma, F)$ ,  $E_{\gamma \text{ max}} = 12, 15, 20, 30, 70$  MeV; measured: fragment  $\gamma$ -ray spectra; deduced: mass distributions, most probable charges, isomeric ratios, average initial fragment spins.

### I. INTRODUCTION

Very little information concerning the product yields for the photofission of  $^{235}\text{U}$  is available in the literature. Kondrat'ko *et al.*<sup>1,2</sup> investigated the mass distribution for the photofission of  $^{235}\text{U}$  with 14-MeV bremsstrahlung by radiochemical analysis of the target. They measured the cumulative yields for 18 mass chains and obtained an asymmetric mass distribution with an additional third peak in the symmetric region. By measuring the yields of  $^{111}\text{Ag}$ ,  $^{113}\text{Ag}$ ,  $^{115}\text{Cd}$ ,  $^{117}\text{Cd}^m$ ,  $^{139}\text{Ba}$ , and  $^{140}\text{Ba}$  for fission induced by bremsstrahlung with 10-, 12-, 14-, 16-, 18-, 20-, 22-, and 25.5-MeV end-point energy, they concluded that the mass distribution for the photofission of  $^{235}\text{U}$  with 10- to 25-MeV bremsstrahlung has a maximum for mass chain 116 and minima for the complementary masses 112 and 120. In a more recent paper Petrzhak *et al.*<sup>3</sup> mentioned a fine structure peak at mass 134 in the mass distributions for the photofission of  $^{237}\text{Np}$  and  $^{235}\text{U}$  with 20-MeV bremsstrahlung. These results were obtained by the determination of the relative yields of xenon isotopes by mass spectrometry.

In a previous paper<sup>4</sup> we studied the mass distribution and the independent yields of a number of fission products for the photofission of  $^{235}\text{U}$  with 25-MeV bremsstrahlung. A yield excess of mass chain 115 over the yield of the neighboring masses in the symmetric region as mentioned by Kondrat'ko *et al.*<sup>1,2</sup> was not observed. Up to now no other data concerning the photofission of  $^{235}\text{U}$  have

been reported in the literature. A systematic investigation of the changes of the mass distribution and the independent yields of fission products with increasing bremsstrahlung end-point energy for the photofission of  $^{235}\text{U}$  has never been done.

In the present work cumulative yields for about 40 mass chains and fractional independent chain yields for  $^{126}\text{Sb}^g$ ,  $^{126}\text{Sb}^m$ ,  $^{128}\text{Sn}$ ,  $^{128}\text{Sb}^g$ ,  $^{128}\text{Sb}^m$ ,  $^{131}\text{Sb}$ ,  $^{131}\text{Te}^g$ ,  $^{131}\text{Te}^m$ ,  $^{134}\text{I}$ ,  $^{135}\text{Xe}$ ,  $^{136}\text{Cs}$ , and  $^{140}\text{La}$  were measured for the photofission of  $^{235}\text{U}$  with thin target bremsstrahlung with 12-, 15-, 20-, 30-, and 70-MeV end-point energy, using the technique of  $\gamma$ -ray spectrometry of fission product catcher foils. For the determination of the cumulative yield of mass 117 and the independent yields of  $^{130}\text{I}$  and  $^{132}\text{I}$ , the cadmium and iodine fractions were separated radiochemically from the catcher foils.

For each bremsstrahlung end-point energy (except for 70 MeV) the average excitation energy of the  $^{235}\text{U}$  nucleus is calculated. The changes of the characteristics of the mass distribution with the bremsstrahlung end-point energy are studied, and a comparison with other  $^{238}\text{U}$  photofission results<sup>5</sup> is made.

Using the value 0.85 for the width parameter  $c$  of the charge distribution, obtained in our previous work,<sup>5</sup> the most probable charges  $[Z_p(E_e)]$ , corresponding to the measured independent yields are calculated ( $E_e$  being the energy of the electrons producing the bremsstrahlung). The determined  $Z_p(E_e)$  values are compared to the  $Z_p$  values, following the unchanged charge density (UCD) hypothesis and to those expected from the empirical

relation of Nethaway.<sup>6</sup> The coefficient  $b$  in the relation of Nethaway, giving the change of  $Z_p$  with increasing mass of the compound nucleus, is deduced from a comparison between our  $^{235}\text{U}$  and  $^{238}\text{U}$  results.

From the isomeric ratios of  $^{126}\text{Sb}^{\epsilon}$ - $^{126}\text{Sb}^m$ ,  $^{128}\text{Sb}^{\epsilon}$ - $^{128}\text{Sb}^m$ , and  $^{131}\text{Te}^{\epsilon}$ - $^{131}\text{Te}^m$ , the average initial spin of the corresponding fragments are calculated for each bremsstrahlung end-point energy, using a statistical model analysis of the neutron and  $\gamma$ -ray emission of the fragments.

## II. EXPERIMENTAL PROCEDURE

The experimental setup for the irradiations, the measuring chain, and the data handling system for the catcher foil experiments on  $^{235}\text{U}$  were the same as in our  $^{238}\text{U}$  photofission studies and are described in our previous paper.<sup>5</sup> The target for the  $^{235}\text{U}$  experiments, prepared at the Central Bureau for Nuclear Measurements (CBNM) Euratom Geel, consisted of a  $15\text{ mg/cm}^2$   $\text{U}_3\text{O}_8$  layer, enriched up to 97%  $^{235}\text{U}$ , on a 1 mm thick aluminum disk. The diameter of the active layer was 40 mm. At a distance of 1 mm the target was followed by a 0.1 mm thick aluminum catcher foil with a purity of 99.999%.

For the determination of the cumulative yield of  $^{117}\text{Cd}^{\epsilon+m}$  and the independent yields of  $^{130}\text{I}^{\epsilon+m}$  and  $^{132}\text{I}^{\epsilon+m}$ , the cadmium and iodine fractions were separated from the catcher foils, following radiochemical procedures close to those of Gleit and Coryell<sup>7</sup> for cadmium and of Wahl<sup>8</sup> for iodine. Owing to the low activity of the irradiated catcher foils it was not possible to determine the independent yield of the ground and isomeric state of  $^{130}\text{I}$  and  $^{132}\text{I}$  separately. For the determination of the independent yield of  $^{130}\text{I}^{\epsilon+m}$  we used the 536.5 keV  $\gamma$ -ray with an absolute intensity of 99.8% in the decay of the ground state of  $^{130}\text{I}$  (Ref. 9). The small correction for the fraction of  $^{130}\text{I}^m$  that does not decay to  $^{130}\text{I}^{\epsilon}$  (17% following Ref. 9) was made by assuming the value 0.81 found in proton induced fission<sup>10</sup> for the isomeric ratio  $\sigma_m/\sigma_g + \sigma_m$ . The uncertainty on the independent yield of  $^{130}\text{I}^{\epsilon+m}$  contains a contribution, owing to this correction. For the determination of the independent yield of  $^{132}\text{I}^{\epsilon+m}$  the 667.7 keV  $\gamma$  ray in the decay of the ground state was used. By an appropriate choice of the irradiation time and of the cooling and measuring times, the sum of the independent yields of the ground and isomeric state of  $^{132}\text{I}$  can be obtained.

For an estimation of the contribution of slow neutron induced fission, the  $^{235}\text{U}$  target was replaced by scandium and indium samples in the experimental setup. Based on the cross sections  $^{45}\text{Sc}(\gamma, n)^{44}\text{Sc}$  (Ref. 11),  $^{115}\text{In}(n, \gamma)^{116}\text{In}^m$  (Ref. 12),

and  $^{235,238}\text{U}(n, F)$  (Ref. 13), an upper limit of 2 and 3% for this contribution in our  $^{235}\text{U}$  photofission experiments with 30- and 70-MeV bremsstrahlung was obtained.

## III. EXPERIMENTAL RESULTS AND DISCUSSION

### A. Average excitation energy

The average excitation energy of the compound nucleus  $^{235}\text{U}$  in our photofission studies was calculated in the same way as outlined in a previous paper.<sup>5</sup> For the behavior of the  $^{235}\text{U}$  photofission cross section  $\sigma(\gamma, F)$  up to 18 MeV the experimental data of Caldwell *et al.*<sup>14</sup> and Bowman, Auchampaugh, and Fultz<sup>15</sup> were used. Above this energy no experimental information on  $\sigma(\gamma, F)$  is available in the literature. To estimate the  $^{235}\text{U}$  photofission cross section for photon energies between 18 and 30 MeV, a similar behavior as given by Shotter *et al.*<sup>16</sup> for the  $^{238}\text{U}$  photofission cross section was assumed. This is based on the similarity of the experimentally determined cross sections below 18 MeV. The average excitation energies of the  $^{235}\text{U}$  nucleus after irradiation with 12-, 15-, 20-, and 30-MeV bremsstrahlung, calculated in this way, are 9.7, 11.6, 13.1, and 14.1 MeV, respectively. The corresponding values of the average excitation energy of the  $^{238}\text{U}$  nucleus, given in our previous paper<sup>5</sup> are 9.7, 11.6, 13.4, and 14.7 MeV. A comparison between our  $^{235}\text{U}$  and our earlier  $^{238}\text{U}$  results is thus meaningful.

### B. Mass distribution

Our results on the mass distribution for the photofission of  $^{235}\text{U}$  are presented in Table I. Cumulative yields of 34, 38, 39, 39, and 38 mass chains were obtained for fission induced with respectively 12-, 15-, 20-, 30- and 70-MeV bremsstrahlung. The mass distributions are normalized to a total yield of 200% as usual. The not-measured mass yields needed for these normalizations and the uncertainties given in Table I are obtained as described in our previous paper.<sup>5</sup> As already mentioned in the Introduction, up to now a systematic study of the mass distribution for the photofission of  $^{235}\text{U}$  with changing end-point energy of the bremsstrahlung was not performed. Kondrat'ko and Petrzhak<sup>1</sup> determined the cumulative yield of 18 mass chains for 14-MeV bremsstrahlung and investigated the behavior of four product yields in the symmetric region for an increase of the bremsstrahlung end-point energy from 10 to 25 MeV. Petrzhak *et al.*<sup>3</sup> measured the relative yields of  $^{131}\text{Xe}$ ,  $^{132}\text{Xe}$ ,  $^{134}\text{Xe}$ , and  $^{136}\text{Xe}$  for 20-MeV bremsstrahlung.

The determined mass yields from our measurements are presented graphically in Fig. 1. For

TABLE I. Cumulative yields for the photofission of  $^{235}\text{U}$ .

$M_{\text{post}} \setminus E_e$ (MeV)	12	15	20	30	70
78	0.095 ± 0.047	0.096 ± 0.033	0.133 ± 0.025	0.135 ± 0.027	0.155 ± 0.019
84	1.18 ± 0.13	1.44 ± 0.19	1.33 ± 0.16	1.35 ± 0.19	1.29 ± 0.14
85	1.74 ± 0.12	1.90 ± 0.14	1.91 ± 0.14	1.95 ± 0.14	1.81 ± 0.13
87	2.94 ± 0.25	3.21 ± 0.24	2.79 ± 0.20	2.84 ± 0.21	2.81 ± 0.21
88	3.88 ± 0.21	3.88 ± 0.16	3.67 ± 0.15	3.63 ± 0.15	3.55 ± 0.15
89	4.55 ± 0.24	4.74 ± 0.25	4.32 ± 0.22	4.39 ± 0.22	4.24 ± 0.22
91	5.91 ± 0.25	5.73 ± 0.25	5.49 ± 0.26	5.64 ± 0.24	5.36 ± 0.23
92	6.02 ± 0.37	5.76 ± 0.30	5.70 ± 0.29	5.80 ± 0.32	5.72 ± 0.33
93	6.17 ± 0.50	6.14 ± 0.50	6.03 ± 0.40	5.91 ± 0.44	5.55 ± 0.40
94	6.73 ± 0.48	6.44 ± 0.48	6.01 ± 0.42	5.98 ± 0.43	5.95 ± 0.43
95	6.41 ± 0.36	6.33 ± 0.28	6.51 ± 0.32	6.18 ± 0.27	6.01 ± 0.26
97	6.02 ± 0.31	5.71 ± 0.30	5.70 ± 0.28	5.55 ± 0.29	5.27 ± 0.27
99	5.58 ± 0.26	5.29 ± 0.22	5.38 ± 0.23	5.23 ± 0.24	5.13 ± 0.23
101	4.10 ± 0.25	4.18 ± 0.24	3.80 ± 0.18	3.99 ± 0.20	3.98 ± 0.22
103	2.74 ± 0.23	2.67 ± 0.19	2.74 ± 0.20	2.73 ± 0.20	2.76 ± 0.20
104	1.66 ± 0.13	1.77 ± 0.15	1.79 ± 0.13	1.77 ± 0.15	1.84 ± 0.14
105	1.03 ± 0.07	1.24 ± 0.07	1.27 ± 0.14	1.31 ± 0.11	1.35 ± 0.14
106			0.745 ± 0.106	0.953 ± 0.143	1.11 ± 0.24
112	0.210 ± 0.043	0.310 ± 0.032	0.476 ± 0.041	0.568 ± 0.044	0.774 ± 0.060
113		0.307 ± 0.049	0.419 ± 0.063	0.512 ± 0.071	0.808 ± 0.081
115	0.180 ± 0.024	0.295 ± 0.029	0.415 ± 0.043	0.522 ± 0.040	0.711 ± 0.051
117			0.401 ± 0.076	0.534 ± 0.071	0.715 ± 0.078
123	0.218 ± 0.040	0.308 ± 0.037	0.441 ± 0.073	0.517 ± 0.051	
125		0.454 ± 0.091	0.576 ± 0.068	0.716 ± 0.077	0.847 ± 0.073
127	0.849 ± 0.063	0.916 ± 0.055	1.15 ± 0.06	1.17 ± 0.06	1.21 ± 0.06
128		1.38 ± 0.13	1.40 ± 0.13	1.48 ± 0.14	1.48 ± 0.12
131	4.02 ± 0.33	4.16 ± 0.25	4.30 ± 0.36	4.06 ± 0.23	3.93 ± 0.21
132	4.73 ± 0.19	4.79 ± 0.18	4.93 ± 0.16	4.63 ± 0.21	4.46 ± 0.21
133	5.83 ± 0.30	5.55 ± 0.28	5.64 ± 0.29	5.46 ± 0.28	5.30 ± 0.28
134	5.96 ± 0.24	5.88 ± 0.20	5.67 ± 0.21	5.59 ± 0.27	5.67 ± 0.37
135	6.59 ± 0.28	6.19 ± 0.26	6.32 ± 0.26	6.03 ± 0.25	5.80 ± 0.26
137			6.43 ± 0.62	6.21 ± 0.57	6.19 ± 0.53
140	5.90 ± 0.20	5.62 ± 0.21	5.79 ± 0.20	5.73 ± 0.22	5.34 ± 0.23
141	5.73 ± 0.32	5.45 ± 0.37	5.36 ± 0.28	5.44 ± 0.29	5.18 ± 0.37
142	5.03 ± 0.37	5.22 ± 0.39	4.76 ± 0.35	4.78 ± 0.35	4.48 ± 0.34
143	4.72 ± 0.38	4.40 ± 0.31	4.43 ± 0.31	4.25 ± 0.30	4.18 ± 0.30
144		3.50 ± 0.52	3.65 ± 0.31	3.60 ± 0.30	3.62 ± 0.28
146	2.20 ± 0.16	2.24 ± 0.14	2.17 ± 0.12	2.24 ± 0.13	2.23 ± 0.13
147	1.86 ± 0.13	1.64 ± 0.12	1.60 ± 0.11	1.67 ± 0.11	1.60 ± 0.09
149	0.860 ± 0.080	0.811 ± 0.099	0.842 ± 0.067	0.847 ± 0.069	0.818 ± 0.060
151	0.485 ± 0.050	0.473 ± 0.041	0.470 ± 0.056	0.408 ± 0.036	0.463 ± 0.040
153		0.207 ± 0.028	0.189 ± 0.032	0.201 ± 0.034	0.215 ± 0.037

each end-point energy a smooth mass distribution without fine structure is obtained. As already observed in our photofission studies on  $^{238}\text{U}$  (Ref. 5), there is a strong increase of the mass yields for symmetric fission with increasing end-point energy of the bremsstrahlung and a nearly independence of the asymmetric yields. The valley region is flat: A third peak with a maximum for mass chain 116 and minima for the complementary masses 112 and 120 as observed by Kondrat'ko and Petrzhak<sup>1</sup> is not present in our results. We also do not observe a yield excess for mass 134 compared to the neighboring masses, in contradiction to the results of Petrzhak *et al.*<sup>3</sup>

Some characteristics of the mass distributions are summarized in Table II. The peak-to-valley ratio ( $P/V$ ) decreases from  $37 \pm 5$  to  $8.4 \pm 0.6$ , by increasing the maximum bremsstrahlung energy from 12 to 70 MeV. From the fragment yields reported by Kondrat'ko and Petrzhak<sup>1</sup> values of  $45 \pm 7$ ,  $32 \pm 3$ ,  $21 \pm 2$ , and  $16 \pm 2$  for the peak to valley ratio for the photofission with 12-, 14-, 16-, and 20-MeV bremsstrahlung can be deduced. For the determination of the symmetric fission yield, the average of the available mass yields in this region was taken. Although in the experiments of Kondrat'ko and Petrzhak<sup>1</sup> the bremsstrahlung was produced in the uranium target itself, there is a

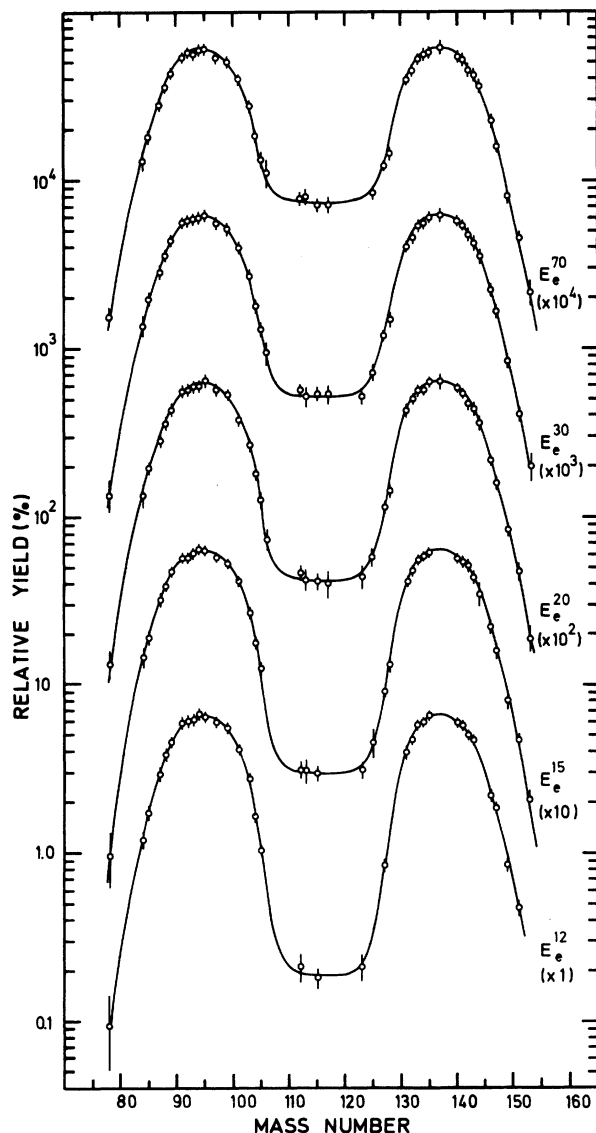


FIG. 1. Postneutron mass distributions for the photofission of  $^{235}\text{U}$  with 12-, 15-, 20-, 30-, and 70-MeV bremsstrahlung. The yields have been multiplied by the scale factor given in parentheses.

general agreement between the  $P/V$  ratio deduced from their and our work.

The mean masses of the light and heavy fragment peaks, MLM and MHM, calculated in the same way as in Ref. 5, are practically independent of the end-point energy of the bremsstrahlung. Comparing  $^{235}\text{U}$  with  $^{238}\text{U}$ , one can conclude that also in photofission an increase of the mass of the fissioning nucleus results mainly in a shift of the MLM towards heavier masses, while the MHM remains practically constant:  $\Delta\text{MLM}/\Delta A$  varies from  $0.80 \pm 0.04$  to  $0.71 \pm 0.04$  and  $\Delta\text{MHM}/\Delta A$  from  $0.20 \pm 0.03$  to  $0.16 \pm 0.03$ , increasing the maximum bremsstrahlung energy from 12 to 30 MeV. ( $A$  represents the mass of the fissioning nucleus.)

For the full width at half maximum height, FWHM, a small increase with increasing end-point energy of the bremsstrahlung is observed. Although the differences with our  $^{238}\text{U}$  photofission values are small, the FWHM for the photofission of  $^{238}\text{U}$  at the same bremsstrahlung energy is systematically higher. Also for neutron induced fission the width of the mass peaks is found to increase for increasing mass of the fissioning nucleus (Ref. 17).

As could be expected, the average number of emitted neutrons ( $\nu$ ), calculated from the mass distribution, increases with the bremsstrahlung end-point energy. From their systematic study of photofission neutron multiplicities for different uranium isotopes using monoenergetic photons in the energy range 6–18 MeV, Caldwell *et al.*<sup>18</sup> found for the photofission of  $^{235}\text{U}$  the linear dependence of the average number of emitted neutrons ( $\nu$ ) on the photon energy  $E$ :

$$\langle \nu \rangle = 1.610 + 0.133E.$$

Following this expression the  $\langle \nu \rangle$  values, to be expected for the photofission of  $^{235}\text{U}$  with 12-, 15-, 20-, and 30-MeV bremsstrahlung, obtained by substituting our values for the average excitation energy for  $E$ , are respectively 2.90, 3.15, 3.35, and 3.48. As for our photofission studies on  $^{238}\text{U}$  (Ref. 5) also for  $^{235}\text{U}$ , there is a good agreement

TABLE II. Mass distribution characteristics.

$E_e$ (MeV)	12	15	20	30	70
$P/V$	37 $\pm$ 5	22 $\pm$ 2	15.5 $\pm$ 1.4	11.7 $\pm$ 0.8	8.4 $\pm$ 0.6
MLM <sup>a</sup>	94.85 $\pm$ 0.07	94.70 $\pm$ 0.07	94.80 $\pm$ 0.07	94.68 $\pm$ 0.07	94.77 $\pm$ 0.07
MHM <sup>a</sup>	137.28 $\pm$ 0.07	137.24 $\pm$ 0.07	137.09 $\pm$ 0.07	137.15 $\pm$ 0.07	137.14 $\pm$ 0.07
FWHM	14.7 $\pm$ 0.4	15.0 $\pm$ 0.4	15.0 $\pm$ 0.4	15.0 $\pm$ 0.4	15.2 $\pm$ 0.4
$\langle \mu \rangle^a$	2.78 $\pm$ 0.12	3.06 $\pm$ 0.12	3.15 $\pm$ 0.11	3.25 $\pm$ 0.11	3.26 $\pm$ 0.11

<sup>a</sup>Uncertainties on MLM, MHM, and  $\langle \mu \rangle$  were obtained by a statistical treatment of the uncertainties of the yields.



TABLE IV.  $Z_p(E_e)$  values for the photofission of  $^{235}\text{U}$ .

$M_{\text{post}} \setminus E_e$ (MeV)	12	15	20	30
126		$49.82 \pm 0.09$	$49.89 \pm 0.06$	$49.97 \pm 0.06$
128		$50.21 \pm 0.12$	$50.24 \pm 0.11$	$50.32 \pm 0.11$
130		$50.89^{+0.09}_{-0.13}$	$50.97 \pm 0.05$	$51.04 \pm 0.05$
131	$51.24 \pm 0.11$	$51.34 \pm 0.07$	$51.45 \pm 0.08$	$51.47 \pm 0.08$
132	$51.51^{+0.10}_{-0.15}$	$51.60 \pm 0.03$	$51.65 \pm 0.03$	$51.68 \pm 0.04$
134	$52.41 \pm 0.10$	$52.50 \pm 0.04$	$52.55 \pm 0.04$	$52.60 \pm 0.04$
135	$52.93 \pm 0.06$	$53.04 \pm 0.04$	$53.08 \pm 0.03$	$53.10 \pm 0.02$
136		$53.28 \pm 0.06$	$53.37 \pm 0.05$	$53.40 \pm 0.04$
140		$55.01 \pm 0.07$	$55.02 \pm 0.06$	$55.09 \pm 0.04$

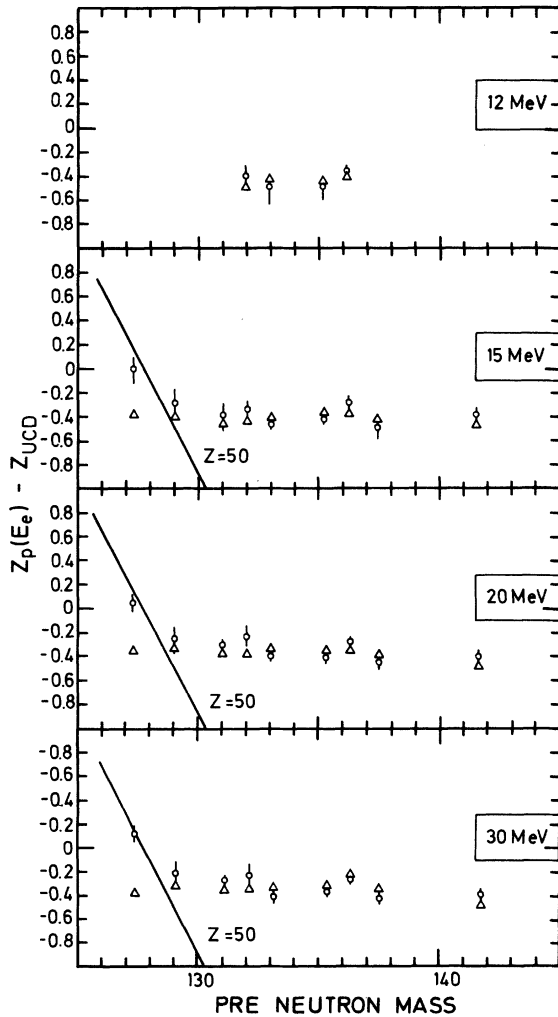


FIG. 2.  $Z_p(E_e) - Z_{\text{UCD}}$  versus fragment mass number for the photofission of  $^{235}\text{U}$  with 12-, 15-, 20-, and 30-MeV bremsstrahlung. The open circles represent the  $Z_p(E_e)$  values deduced from the measured independent yields, the triangles the  $Z_p$  values calculated following Nethaway (Ref. 6).

of the charge to mass ratio of the fragments from that of the fissioning nucleus, but not as large as observed in thermal neutron induced fission of  $^{235}\text{U}$  (Ref. 21). This deviation was explained as the result of a polarization effect during deformation or a two-neutron transfer process through the neck.

The  $Z_p$  values, calculated from the empirical relation of Nethaway<sup>6</sup> using for the excitation energy of the compound nucleus the calculated average excitation energy and using the reference  $Z_p$  function of Wahl *et al.*<sup>22</sup> for the thermal neutron fission of  $^{235}\text{U}$ , are indicated in Fig. 2 by the triangles. A good agreement between the calculated and experimentally determined  $Z_p(E_e)$  values is obtained at each bremsstrahlung energy, except for the  $Z_p(E_e)$  values, deduced from the independent yield of  $^{126}\text{Sb}$ .

As already mentioned in our previous work<sup>5</sup> on the photofission of  $^{238}\text{U}$ , the influence of the closed 50-proton shell on the  $Z_p$  function, clearly demonstrated in Ref. 20, was not taken into account in the used reference  $Z_p$  behavior. The presence of this tendency caused a discrepancy of about 0.5 charge units between the experimentally determined  $Z_p(E_e)$  values and the  $Z_p$  values calculated using the  $Z_p$  reference value of Wahl *et al.*<sup>22</sup> for mass chain 128 in the photofission of  $^{238}\text{U}$ . Owing to the shift of the  $Z=50$  line towards lower masses (about 1.7) for the photofission of  $^{235}\text{U}$  compared to  $^{238}\text{U}$ , the influence of the closed  $Z=50$  shell is not observed around mass 128 but around mass 126 for the photofission of  $^{235}\text{U}$ . As a consequence, in this case a good agreement between the experimental  $Z_p(E_e)$  values and the  $Z_p$  values calculated using the  $Z_p$  reference values of Wahl *et al.*<sup>22</sup> is obtained for mass 128 and a discrepancy of about 0.4 charge units is observed for mass 126. From our photofission studies on  $^{235}\text{U}$  and  $^{238}\text{U}$ , we can conclude that the method of Nethaway<sup>6</sup> combined

with the reference  $Z_p$  function of Wahl *et al.*<sup>22</sup> is very useful for the calculation of  $Z_p$  values in photofission, except in the mass region where the 50-proton shell has an influence on the  $Z_p$  behavior

From a comparison between the  $Z_p(E_e)$  values, determined for the photofission of  $^{235}\text{U}$  and  $^{238}\text{U}$ , the shift of  $Z_p$  with increasing mass of the compound nucleus  $A$  (the coefficient  $b = \Delta Z_p / \Delta A$  of the relation of Nethaway<sup>6</sup>) can be deduced. Excluding the mass region, where the closed 50-proton shell is important, average values  $b = -0.165 \pm 0.023$ ,  $-0.180 \pm 0.022$ ,  $-0.186 \pm 0.020$ , and  $-0.174 \pm 0.020$  are obtained for 12-, 15-, 20-, and 30-MeV bremsstrahlung induced fission, respectively. The coefficient  $b$  is found to be rather independent of the excitation energy in the energy range of our experiments. Averaging the obtained  $b$  values, a value  $\langle b \rangle = -0.177 \pm 0.011$  is found, which is close to the value  $-0.188 \pm 0.004$  deduced by Nethaway<sup>6</sup> from neutron induced fission data for the heavy fragment mass region.

#### D. Isomeric ratios

From the measured independent yields of the isomeric pairs  $^{126}\text{Sb}^m(8^-)$ - $^{126}\text{Sb}^m(5^+)$ ,  $^{128}\text{Sb}^m(8^-)$ - $^{128}\text{Sb}^m(5^+)$ , and  $^{131}\text{Te}^m(\frac{3}{2}^-)$ - $^{131}\text{Te}^m(\frac{1}{2}^-)$  for the photofission of  $^{235}\text{U}$  (see Table III), the corresponding isomeric ratios  $\sigma_m / (\sigma_g + \sigma_m)$  are deduced directly. They are given in Table V. A systematic decrease of  $\sigma_m / (\sigma_g + \sigma_m)$  for  $^{126}\text{Sb}$  and  $^{128}\text{Sb}$  with increasing bremsstrahlung energy is observed, indicating that an increase of the bremsstrahlung end-point energy favors the production of the high spin isomer. This tendency is not so pronounced for  $^{131}\text{Te}$ , for which the relative yield of the high spin isomer increases only very slightly with the bremsstrahlung end-point energy. Also in the study of the production of the iodine and tellurium isomers in the fission of  $^{238}\text{U}$ , induced by protons with energy between 30 and 70 MeV,<sup>10</sup> the isomeric ratio of  $^{131}\text{Te}$  was found to be almost constant with increasing bombarding energy, while in the case of  $^{130}\text{I}$ ,  $^{132}\text{I}$ , and  $^{133}\text{Te}$  the high spin isomer was systematically favored by an increase of the proton energy.

Although the differences remain within the experimental errors the measured isomeric ratios for  $^{131}\text{Te}$  are systematically higher for the photo-

fission of  $^{235}\text{U}$  compared to  $^{238}\text{U}$ . A comparison with photofission data of other authors on the isomeric ratios of the studied fission products is not possible. Also concerning the isomeric ratio of  $^{126}\text{Sb}$  in thermal neutron induced fission no information was available in the literature. Imanishi, Fujiwara, and Nishi<sup>23</sup> give an isomeric ratio of  $0.47 \pm 0.04$  and  $0.48 \pm 0.04$  for  $^{126}\text{Sb}$  in the thermal neutron induced fission of  $^{235}\text{U}$  and  $^{239}\text{Pu}$ , respectively. In  $^{235}\text{U}(n_{\text{th}}, f)$  an upper limit of 0.587 for this isomeric ratio was found (Ref. 24). For the isomeric ratio of  $^{131}\text{Te}$  values  $0.65 \pm 0.03$ ,  $0.66 \pm 0.05$ , and  $0.68 \pm 0.03$  for  $^{233}\text{U}(n_{\text{th}}, f)$ ,  $^{235}\text{U}(n_{\text{th}}, f)$ , and  $^{239}\text{Pu}(n_{\text{th}}, f)$ , respectively were given in Ref. 23. It appears that in our photofission experiments approximately the same isomeric ratio values are obtained as in thermal neutron induced fission. For  $^{131}\text{Te}$  a still better agreement is obtained by using the value 0.72 of Imanishi, Fujiwara, and Nishi<sup>23</sup> for the absolute intensity of the 149 keV  $\gamma$  ray in the decay of  $^{131}\text{Te}^g$  instead of 0.681 (Ref. 25). This leads to an increase of about 0.05 of the isomeric ratios of  $^{131}\text{Te}$  in our photofission studies on  $^{235}\text{U}$ . Also the difference between the  $^{131}\text{Te}$  isomeric ratios for photofission of  $^{235}\text{U}$  and  $^{238}\text{U}$  can partly be eliminated by adopting the value 0.72 for the absolute intensity of the 149 keV  $\gamma$  ray.

Performing a statistical model analysis in the same way as in our photofission studies on  $^{238}\text{U}$  (Ref. 5), the average initial spin of the fragments, leading to the measured isomeric pairs by the emission of prompt neutrons and  $\gamma$  rays, was derived. The different quantities, necessary in this calculation: the number, energy, and transmission coefficients of the emitted neutrons, the number and multipolarity of the emitted  $\gamma$  rays, and the spin cutoff parameters  $\sigma_n$  and  $\sigma_\gamma$  are determined as outlined in our previous work.<sup>5</sup> The assumptions, made in Sec. III C of this paper for the determination of the average number of neutrons emitted by the fragments, leading to the considered fission products, do not much influence the results, as the emitted neutrons are predominantly s wave.

The average initial spin values,  $\bar{J}_i$ , deduced from the measured isomeric ratios are given in

TABLE V. Isomeric ratios  $\sigma_m / (\sigma_g + \sigma_m)$ .

Isomeric pair \ $E_e$ (MeV)	12	15	20	30	70
$^{126}\text{Sb}^m(5^+)$ - $^{126}\text{Sb}^g(8^-)$		$0.52 \pm 0.12$	$0.45 \pm 0.08$	$0.42 \pm 0.07$	$0.34 \pm 0.05$
$^{128}\text{Sb}^m(5^+)$ - $^{128}\text{Sb}^g(8^-)$		$0.53 \pm 0.10$	$0.49 \pm 0.10$	$0.45 \pm 0.11$	$0.39 \pm 0.08$
$^{131}\text{Te}^m(\frac{11}{2}^-)$ - $^{131}\text{Te}^g(\frac{3}{2}^+)$	$0.59 \pm 0.11$	$0.59 \pm 0.06$	$0.61 \pm 0.05$	$0.62 \pm 0.07$	$0.61 \pm 0.05$

TABLE VI.  $\bar{J}_i$  values ( $\hbar$ ).

Isomeric pair \ $E_e$ (MeV)	12	15	20	30	70
$^{126}\text{Sb}^m(5^+) - ^{126}\text{Sb}^g(8^-)$		$7.5 \pm 1.6$	$8.4 \pm 1.7$	$9.0 \pm 1.6$	$10.4 \pm 1.6$
$^{128}\text{Sb}^m(5^+) - ^{128}\text{Sb}^g(8^-)$		$7.0 \pm 1.6$	$7.9 \pm 1.7$	$8.4 \pm 1.8$	$9.4 \pm 1.8$
$^{131}\text{Te}^m(\frac{11}{2}^-) - ^{131}\text{Te}^g(\frac{3}{2}^+)$	$5.6 \pm 1.8$	$5.6 \pm 1.4$	$5.9 \pm 1.4$	$6.0 \pm 1.4$	$5.9 \pm 1.4$

Table VI. As in our  $^{238}\text{U}$  photofission work a systematic contribution of  $1.2\hbar$ , inherent in the method, was included in the given uncertainties. As observed in different other fissioning systems<sup>23,26-30</sup> and in our  $^{238}\text{U}$  photofission study,<sup>5</sup> the deduced  $\bar{J}_i$  values of the fragments are systematically higher than the spin of the compound nucleus  $^{235}\text{U}$  (as the photon absorption in our experiments is predominantly  $E1$ , only  $\frac{5}{2}^+$ ,  $\frac{7}{2}^+$ , and  $\frac{9}{2}^+$  states are excited).

As could be expected from the changes of the measured isomeric ratios in our  $^{235}\text{U}$  photofission experiments, there is a systematic increase with increasing end-point energy of the bremsstrahlung of the average initial spin values, determined from the isomeric ratio of  $^{126}\text{Sb}$  and  $^{128}\text{Sb}$ . For  $^{131}\text{Te}$  this tendency is not so pronounced. Also in the proton induced fission of  $^{238}\text{U}$  (Ref. 10) an increase of the calculated fragment angular momentum with increasing bombarding energy has been found, except in the case of  $^{131}\text{Te}$ , in which the deduced average initial spin remained practically constant.

The  $\bar{J}_i$  values deduced from the isomeric ratio of  $^{128}\text{Sb}$  and  $^{131}\text{Te}$  in our photofission experiments on  $^{235}\text{U}$  are almost the same as those obtained in thermal neutron induced fission. Imanishi, Fujiwara, and Nishi<sup>23</sup> deduced from the isomeric ratio of  $^{128}\text{Sb}$  for  $^{233}\text{U}(n_{\text{th}}, f)$  and  $^{239}\text{Pu}(n_{\text{th}}, f)$  the  $\bar{J}_i$  values  $9.5 \pm 0.7\hbar$  and  $9.3 \pm 0.7\hbar$ , respectively. For  $^{131}\text{Te}$  in  $^{233}\text{U}(n_{\text{th}}, f)$ ,  $^{235}\text{U}(n_{\text{th}}, f)$ , and  $^{239}\text{Pu}(n_{\text{th}}, f)$ , they obtained the values  $6.0 \pm 0.4\hbar$ ,  $6.1 \pm 0.6\hbar$ , and  $6.2 \pm 0.4\hbar$ . The uncertainties on the  $\bar{J}_i$  values, given by these authors, include only the statistical errors and do not include the uncertainty, introduced by the assumptions made during the calculation. As far as the small differences between these  $\bar{J}_i$  values and our photofission results on  $^{235}\text{U}$  are not due to differences in the observed isomeric ratio values, they are caused by a different choice of the parameters in the statistical treatment of the de-excitation process of the fragments.

Aumann *et al.*<sup>30</sup> studied the isomeric ratio of the  $^{148}\text{Pm}$  isomeric pair for the thermal neutron induced fission of  $^{233}\text{U}$  and  $^{235}\text{U}$  and for fission of  $^{232}\text{Th}$  induced by  $^4\text{He}$  ions with energy in the range

25.8–41.4 MeV. They also found an increase of the fragment angular momentum, derived from the  $^{148}\text{Pm}$  isomeric ratio, with the excitation energy and angular momentum of the fissioning nucleus. As the average angular momentum of the compound nucleus  $^{235}\text{U}$  in our photofission experiments remains practically constant with changing end-point energy of the bremsstrahlung, the observed increase of the determined  $\bar{J}_i$  values of the fragments in our experiments must be attributed to an increase of the excitation energy of the compound nucleus.

#### IV. CONCLUSIONS

The mass distribution for the photofission of  $^{235}\text{U}$  with bremsstrahlung with end-point energy in the range 12–70 MeV shows a doubly peaked shape without fine structure. As in our  $^{238}\text{U}$  photofission studies (Ref. 5) a strong increase of the symmetric mass yield with increasing bremsstrahlung energy and a near independence on the excitation energy of the mass yields corresponding to asymmetric fission are observed.

From the measured independent yields  $Z_p(E_e)$  values are deduced in the mass region 126–140. A tendency of the  $Z_p$  function to remain close to the  $Z=50$  line is present, showing the influence of the closed 50-proton shell on the charge distribution in the excitation energy range of our experiments. Outside this region a nearly constant deviation of  $Z_p(E_e)$  from  $Z_{\text{UCD}}$  is observed. Except in the mass region where the  $Z_p$  function reaches the closed  $Z=50$  shell, a good agreement between the experimentally determined  $Z_p(E_e)$  values and those calculated following the method of Nethaway<sup>6</sup> is obtained.

The behavior of the isomeric ratios  $\sigma_m/(\sigma_g + \sigma_m)$  of the isomeric pairs  $^{126}\text{Sb}^g - ^{126}\text{Sb}^m$ ,  $^{128}\text{Sb}^g - ^{128}\text{Sb}^m$ , and  $^{131}\text{Te}^g - ^{131}\text{Te}^m$  with increasing bremsstrahlung energy indicate that the production of the high spin isomer is favored at higher excitation energy of the compound nucleus. The average initial fragment spin values, calculated from these isomeric ratios are approximately the same as those deduced in thermal neutron induced fission.



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- <sup>1</sup>M. Ya. Kondrat'ko and K. A. Petrzhak, *At. Énerg.* **23**, 559 (1967).
- <sup>2</sup>M. Ya. Kondrat'ko, U. N. Korinetz, and K. A. Petrzhak, *At. Énerg.* **35**, 214 (1973).
- <sup>3</sup>K. A. Petrzhak, E. V. Platygina, Yu. A. Solov'ev, and V. F. Teplykh, *At. Énerg.* **41**, 44 (1976).
- <sup>4</sup>H. Thierens, D. De Frenne, E. Jacobs, A. De Clercq, P. D'hondt, and A. J. Deruytter, *Phys. Rev. C* **14**, 1058 (1976).
- <sup>5</sup>E. Jacobs, H. Thierens, D. De Frenne, A. De Clercq, P. D'hondt, P. De Gelder, and A. J. Deruytter, *Phys. Rev. C* **19**, 422 (1979).
- <sup>6</sup>D. R. Nethaway, Report No. UCRL-51538 (unpublished).
- <sup>7</sup>C. E. Gleit and C. D. Coryell, *Phys. Rev.* **122**, 229 (1961).
- <sup>8</sup>A. C. Wahl, *Phys. Rev.* **99**, 730 (1955).
- <sup>9</sup>H. R. Hiddleston and C. P. Browne, *Nucl. Data Sheets* **13**, 133 (1974).
- <sup>10</sup>M. Diksic and L. Yaffe, *Can. J. Chem.* **53**, 3116 (1975).
- <sup>11</sup>R. M. Sambell and B. M. Spicer, *Nucl. Phys.* **A205**, 139 (1973).
- <sup>12</sup>W. E. Alley and R. M. Lessler, *Nucl. Data* **A11**, 734 (1973).
- <sup>13</sup>Neutron Cross Sections, compiled by J. R. Stehn, M. D. Goldberg, R. Wiener-Chasman, S. F. Mughabghab, B. A. Magurno, and V. M. May, Brookhaven National Laboratory Report No. BNL-325 (National Technical Information Service, Virginia, 1965), 2nd ed., 2nd suppl., Vol. III,  $z = 88$  to 98.
- <sup>14</sup>J. T. Caldwell, E. J. Dowdy, B. Berman, R. Alvarez, and P. Meyer, Report No. LA-UR76-1615 (unpublished).
- <sup>15</sup>C. D. Bowman, C. F. Auchampaugh, and S. C. Fultz, *Phys. Rev.* **133**, B676 (1964).
- <sup>16</sup>A. C. Shotton, D. Branford, J. C. McGeorge, and J. M. Reid, *Nucl. Phys.* **A210**, 55 (1977).
- <sup>17</sup>K. F. Flynn, E. P. Horwitz, C. A. A. Bloomquist, R. F. Barnes, R. K. Sjolbom, P. R. Fields, and L. E. Glendenin, *Phys. Rev. C* **5**, 1725 (1972).
- <sup>18</sup>J. T. Caldwell, E. J. Dowdy, R. A. Alvarez, B. L. Berman, and P. Meyer, private communication and *Nucl. Sci. Eng.* (to be published).
- <sup>19</sup>E. Jacobs, H. Thierens, A. De Clercq, D. De Frenne, P. D'hondt, and A. J. Deruytter, *Phys. Rev. C* **14**, 1874 (1976).
- <sup>20</sup>J. Kratz and G. Hermann, in *Proceedings of the Third International Symposium on the Physics and Chemistry of Fission, Rochester, 1973* (International Atomic Energy Agency, Vienna, 1974), Vol. II, p. 95; H. G. Clercq, W. Lang, J. Wohlforth, K. H. Schmidt, H. Schrader, K. E. Pferdekämper, and J. Jungmann, *Z. Phys.* **A274**, 203 (1975); R. B. Strittmatter, Ph.D. thesis, University of Illinois, 1979 (unpublished); A. C. Wahl, Report No. IAEA INDC(NDS) -87, 215, 1979 (unpublished).
- <sup>21</sup>S. Amiel and H. Feldstein, in *Proceedings of the Third International Symposium on the Physics and Chemistry of Fission, Rochester, 1973* (International Atomic Energy Agency, Vienna, 1974), Vol. II, p. 65.
- <sup>22</sup>A. C. Wahl, A. E. Norris, R. A. Rouse, and J. C. Williams, in *Proceedings of the Second International Symposium on the Physics and Chemistry of Fission, Vienna, Austria, 1969* (International Atomic Energy Agency, Vienna, 1969), p. 813.
- <sup>23</sup>N. Imanishi, I. Fujiwara, and T. Nishi, *Nucl. Phys.* **A263**, 141 (1976).
- <sup>24</sup>M. M. Fowler and A. C. Wahl, *J. Inorg. Nucl. Chem.* **36**, 1201 (1974).
- <sup>25</sup>S. V. Jackson, Ph.D. thesis, University of California Report No. UCRL-51846, 1975 (unpublished).
- <sup>26</sup>H. Warhanek and R. Vandenbosch, *J. Inorg. Nucl. Chem.* **26**, 669 (1964).
- <sup>27</sup>M. M. Hoffman, *Phys. Rev.* **133**, B714 (1964).
- <sup>28</sup>J. B. Wilhelmy, E. Cheifetz, R. C. Jared, S. G. Thompson, and H. R. Bowman, *Phys. Rev. C* **5**, 2041 (1972).
- <sup>29</sup>W. D. Loveland and Y. S. Shun, *Phys. Rev. C* **4**, 2282 (1971).
- <sup>30</sup>D. C. Aumann, W. Gückel, E. Nirschl, and H. Zeising, *Phys. Rev. C* **16**, 254 (1977).