Product yields for the photofission of ^{235}U and ^{238}U with 25-MeV bremsstrahlung

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The photofission of ^{235}U and ^{238}U with 25-MeV bremsstrahlung was studied. For both nuclei cumulative yields for about 40 mass chains were measured. For ^{235}U fractional independent chain yields of $^{126}Sb^g$, $^{126}Sb^m$, ^{128}Sn, ^{128}Sb(9 h), ^{128}Sb(10 min), ^{131}Sb, $^{131}Te^g$, $^{131}Te^m$, ^{132}I, ^{134}I, ^{135}Xe, ^{136}Cs, and ^{140}La were determined and for ^{238}U those of ^{128}Sn, ^{128}Sb(9 h), ^{131}Sb, $^{131}Te^g$, $^{131}Te^m$, ^{132}I, ^{134}I, ^{135}Xe, and ^{136}Cs. Charge distributions and most probable charges are deduced and compared to the unchanged charge distribution hypothesis. Fragment shell effects are discussed. In addition the isomeric ratios $^{126}Sb^g$-$^{126}Sb^m$, ^{128}Sb(9 h)-^{128}Sb(10 min), and $^{131}Te^g$-$^{131}Te^m$ for the photofission of ^{235}U and ^{128}Sb(9 h)-^{128}Sb(10 min) and $^{131}Te^g$-$^{131}Te^m$ for the photofission of ^{238}U are determined and discussed.

[NUCLEAR REACTIONS, FISSION $^{235,238}U(\gamma,F)$, $E_{\gamma\,max}=25$ MeV; measured: fragment γ-ray spectra; deduced: mass distributions, most probable charges, isomeric ratios.]

I. INTRODUCTION

The postneutron fragment yields for the photofission of ^{238}U have been measured by several investigators.[1-9] The irradiations in all these experiments were performed with bremsstrahlung with end-point energies between 10 and 50 MeV, except for the experiment of Meason and Kuroda,[5] who used the $^7_3Li(p,\gamma)^4_2He$ reaction to produce 17.5-MeV γ rays. Almost all the information on the fission fragment yields was obtained after chemical separation of some fragments followed by counting the β and/or γ activity.[1-7] In recent years the extremely good resolution of Ge(Li) detectors has been exploited to obtain yields from direct γ-ray spectra of the gross fission products. This procedure, together with chemical separation of some isotopes, was used by Chattopadhyay *et al.*,[8] who measured the γ-ray spectrum of the irradiated uranium target. This method is limited by the presence of γ rays emitted by the uranium isotopes produced by (γ, n) reactions in the target. All the authors give cumulative chain yields and mass distributions, but only Cuninghame *et al.*[6] (for ^{82}Br, ^{96}Nb, ^{136}Cs, ^{140}La) and Chattopadyay *et al.*[8] [for ^{126}Sb(12.5 day), ^{128}Sb(9 h), ^{132}I, ^{133}I, ^{134}I, ^{135}Xe, ^{136}Cs] give fractional independent yields.

The general aspect of the asymmetric mass distribution curve agrees very well for all the authors. Concerning the presence of fine structure in the mass region 130–140, many discrepancies exist. Chattopadhyay *et al.*[8] mention a large "negative" fine structure at mass 135. Other authors observe a fine-structure peak around the masses 132, 133, or 134. To clarify this situation Petrzhak *et al.*[9] measured the yields of ^{131}Xe, ^{132}Xe, ^{134}Xe, and ^{136}Xe, and concluded that there is a peak at $A=134$.

Less information is available concerning the postneutron fragment yields for the photofission of ^{235}U. Only Kondrat'ko *et al.*[10,11] report photofission studies on ^{235}U. These authors observed an asymmetric mass distribution without fine structure in the mass region $A=130-140$, but with an excess of the yields of the most symmetric fission ($A=115$ and 117) over the yields of neighboring fragments in the symmetric region ($A=111$, 112, and 113). No independent yields were determined.

We studied the photofission of ^{235}U and ^{238}U with 25-MeV bremsstrahlung, using the technique of γ-ray spectrometry of fission product catcher foils. Cumulative yields for about 40 mass chains were measured for both nuclei. Fractional independent yields for $^{126}Sb^g$, $^{126}Sb^m$, ^{128}Sn, ^{128}Sb(9 h), ^{128}Sb(10 min), ^{131}Sb, $^{131}Te^g$, $^{131}Te^m$, ^{132}I, ^{134}I, ^{135}Xe, ^{136}Cs, and ^{140}La in the case of $^{235}U(\gamma,F)$ and for ^{128}Sn, ^{128}Sb(9 h), ^{131}Sb, $^{131}Te^g$, $^{131}Te^m$, ^{132}I, ^{134}I, ^{135}Xe, and ^{136}Cs in the case of $^{238}U(\gamma,F)$ were determined. Charge distributions and most probable charges (Z_p) are deduced and compared to the unchanged charge distribution (UCD) hypothesis. Fragment shell effects are discussed. In addition, the isomeric ratios $^{126}Sb^g$-$^{126}Sb^m$, ^{128}Sb(9 h)-^{128}Sb(10 min) and $^{131}Te^g$-$^{131}Te^m$ for the photofission of ^{235}U, and ^{128}Sb(9 h)-^{128}Sb(10 min) and $^{131}Te^g$-$^{131}Te^m$ for the photofission of ^{238}U, are determined and discussed.

II. EXPERIMENTAL PROCEDURE

A 15-mg/cm^2 U_3O_8 layer, enriched to 97% ^{235}U, on a 1-mm thick aluminium disk or a 18-mg/cm^2 U_3O_8 layer, enriched to 99.6% ^{238}U, on a 5-mm

thick graphite disk, followed by an aluminium catcher foil, were irradiated with 25-MeV endpoint bremsstrahlung at the linac of the Nuclear Physics Laboratory. The targets were prepared at the Central Bureau of Nuclear Measurements (CBNM), Euratom Geel. The diameter of the active layer was 30 mm.

The bremsstrahlung was produced by 25-MeV electrons of an analyzed beam of the linac, in a 0.5-mm-thick tungsten bremsstrahlung target, followed by 10-cm graphite to stop the electrons. The uranium targets were placed directly behind the graphite block, without any collimation of the bremsstrahlung beam. After appropriate irradiation and cooling times, the γ-ray spectra of the catcher foils were measured, using a 19-cm^3 Ge (Li) detector followed by an Ortec 120-4 preamplifier, a Tennelec TC 205A linear amplifier, a Northern Scientific NS 624 analog-to-digital converter, and a PDP-15 system. Cumulative yields or fractional independent yields are determined. The resolution of the system was 2.2 keV at 1333 keV in the measuring conditions. More details concerning the experimental setup and the data handling can be found in a previous paper.[12] In addition to the data of Table I of Ref. 12, the decay data given in Table I of this paper are used. In this table we give the isotope used for the determination of the independent or cumulative chain yield, its half-life ($T_{1/2}$), the energy (E_γ), and absolute intensity (I_γ) of the used γ transition, the half-life of the precursor ($T_{1/2P}$), and the reference for these data (Ref.).

The average electron current at the W-bremsstrahlung target was 10 μA, with a stability better than 5%. This enables us to use the formulas given in our previous paper[12] for the calculation of the fragment yields.

The ^{24}Na produced by the ^{27}Al$(n,\alpha)^{24}$Na reaction on the aluminium catcher foil, enables an estimation of the contribution of the fast neutron induced fission in our targets, using the known cross sections for the ^{235,238}U(n,F),[18] ^{235,238}U(γ,F),[19,20] and ^{27}Al(n,α)[21] reactions. The contribution of fast neutron induced fission was in both cases less than 0.1%. By introducing Sc and In samples in the bremsstrahlung beam at the position of the uranium target, and using the cross sections for the reactions ^{45}Sc$(\gamma,n)^{44}$Sc,[22] ^{115}In$(n,\gamma)^{116}$In^m,[23] and ^{235,238}U(n,F), we found that the contribution of slow neutron induced fission was less than 2% for our photofission studies on ^{235}U.

TABLE I. Additional nuclear data for studied fission products.

Isotope	E_γ (keV)	I_γ (%)	$T_{1/2}$	$T_{1/2P}$	Ref.
^{78}As	613.7	35.0	91 min	88 min	13
^{84}Br	802.3	6.3	31.8 min	3.3 min	14
	1015.9	6.4			
^{125}Sb	427.9	29.8	2.77 yr	9.64 day	15
^{126}Sb^m	414.8	81.9	19 min	10^5 yr	16
^{126}Sb	414.8	81.9	12.4 day	10^5 yr	16
^{131}Te^m	200.7	7.5	30 h	23 min	17
	793.7	13.7			
	852.2	21.2			
	1206.6	9.7			
^{131}Te	149.7	68.1	25 min	30 h	17
^{136}Cs	1048.1	80.5	13.7 day	...	13

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Mass distribution

The cumulative yields for 41 and 43 mass chains have been measured for the photofission with 25-MeV bremsstrahlung of ^{235}U and ^{238}U, respectively. The results are given in Tables II and III. Our mass distributions are normalized to a total yield of 200%. In addition to the uncertainties mentioned in our previous paper,[12] we introduced a supplementary error of 5% on the branching ratios of the used γ rays in calculating the experimental error of our results.

As mentioned in the introduction only Kondrat'ko *et al.*[10,11] have studied the postneutron mass distribution for the photofission of ^{235}U with bremsstrahlung in a comparable energy region. Their target was enriched to 90% ^{235}U. As they concentrated their attention on changes of fission product yields in the symmetric region with increasing maximum bremsstrahlung energy, they give only six mass yields for 25-MeV bremsstrahlung. These are given together with our results in Table II. For 14-MeV bremsstrahlung they determined the yields of 18 mass chains. The excess of the yield of the most symmetric fission fragments ($A = 115, 117$) over the yield of the neighboring fragments in the symmetric region as mentioned by Kondrat'ko *et al.*[10,11] was not observed in our experiment.

In Table III we compare our results for the photofission of ^{238}U with the results of Chattopadhyay *et al.*[8] and Swindle *et al.*[7] Chattopadhyay *et al.* measured the cumulative yield for 26 mass chains by γ-ray spectroscopy of the uranium target, combined with chemical separation for some isotopes. No individual error for each mass yield is given, but an over-all error of 8% is adopted. Using chemical separation techniques, Swindle *et al.*[7] determined the cumulative yield of 22 masses for the photofission of ^{238}U with 24- and 26-MeV bremsstrahlung. In the available literature, no values for the cumulative yields of masses 85, 87,

88, 94, 101, 104, 107, 146, and 155 for the photofission of ^{238}U with bremsstrahlung in an energy region from 10 to 50 MeV, were found.

Figure 1 gives our mass distributions for the photofission of ^{235}U and ^{238}U. Except for the mass region 133–134 in the case of ^{238}U, no fine structure is observed. A proof that the observed fine structure is a real effect can be found in our previous studies,[12] with the same spectroscopic data, of the well-known mass distributions for the spontaneous fission of ^{252}Cf, where no fine-structure effect is present, and the thermal neutron induced fission of ^{235}U, where the fine-structure peak at mass 134 is reproduced. The fine-structure peak in the mass region 133–134 for the photofission of ^{238}U was also reported by Petrzhak *et al.*,[9] Chattopadhyay *et al.*,[8] Richter and Coryell,[2] and Schmitt and Sugarman.[1] A fine structure at mass 133–134 as observed in the photofission of ^{238}U also exists in several other fissioning systems, e.g., ^{235}U (n_{th},f),[24] ^{239}Pu(n_{th},f),[24] ^{241}Pu(n_{th},f),[24] and ^{238}U $(n_{14.8\text{MeV}},f)$.[8] Neither the mass yield excess for mass 132, as observed by Meason and Kuroda[5] in the photofission of ^{238}U with 17.5-MeV monochromatic γ-rays, nor the "negative fine structure" at mass 135, reported by Meason and Kuroda[5] and Chattopadhyay *et al.*,[8] is present in our measured mass distribution for the bremsstrahlung induced fission of ^{238}U. This "negative fine structure" was never observed in any other fissioning system.

TABLE II. Cumulative chain yields for the photofission of ^{235}U.

Mass chain	Yield This experiment	Kondrat'ko
78	0.136 ± 0.018	•••
84	1.47 ± 0.15	•••
85	2.01 ± 0.15	•••
87	2.97 ± 0.21	•••
88	3.61 ± 0.14	•••
89	4.21 ± 0.23	•••
91	5.57 ± 0.23	•••
92	5.69 ± 0.31	•••
93	5.98 ± 0.33	•••
94	5.78 ± 0.42	•••
95	6.06 ± 0.25	•••
97	5.47 ± 0.27	•••
99	5.39 ± 0.24	•••
101	3.84 ± 0.25	•••
103	2.60 ± 0.18	•••
104	1.84 ± 0.14	•••
105	1.29 ± 0.05	•••
106	0.854 ± 0.080	•••
111	0.511 ± 0.061	0.561 ± 0.040
112	0.542 ± 0.039	•••
113	0.466 ± 0.041	0.454 ± 0.050
115	0.486 ± 0.036	0.528 ± 0.040
125	0.726 ± 0.073	•••
127	1.20 ± 0.06	•••
128	1.72 ± 0.14	•••
131	4.00 ± 0.29	•••
132	4.66 ± 0.18	•••
133	5.45 ± 0.29	•••
134	5.82 ± 0.17	•••
135	6.08 ± 0.22	•••
137	6.34 ± 0.48	•••
139	•••	5.90 ± 0.70
140	5.46 ± 0.20	5.39 ± 0.30
141	5.83 ± 0.41	•••
142	4.80 ± 0.36	•••
143	4.23 ± 0.22	•••
144	3.62 ± 0.26	•••
146	2.25 ± 0.12	•••
147	1.71 ± 0.10	•••
149	0.899 ± 0.058	•••
151	0.509 ± 0.041	•••
153	0.217 ± 0.031	•••

Our peak-to-valley ratio value of 13 ± 1 for the photofission of ^{235}U agrees with the result of Kondrat'ko *et al.*,[10] who found a value of 13 ± 2. For the photofission of ^{238}U we found the value 19 ± 2 for the peak-to-valley ratio. A significant discrepancy exists between the different values given in the literature; e.g., Swindle *et al.*[7] and Chattopadhyay *et al.*[8] give, respectively, 30 ± 3 and 12.9 ± 2. The higher value for the peak-to-valley ratio for the photofission of ^{238}U, compared to the value for the photofission of ^{235}U, is consistent with a larger fissility parameter (Z^2/A) exhibiting larger yields at symmetry[25] and with the fact that, compared to even-odd compound nuclei, even-even fissioning nuclei have a greater peak-to-valley ratio for the same excitation energy.[26] The assumption of equal excitation energy for ^{235}U and ^{238}U seems very reasonable as our experimental setup was the same in both cases, while the cross sections for the photofission of ^{235}U and ^{238}U have practically the same shape in the energy region of interest for our experiments.[19,20]

Some characteristics of the mass distributions are summarized in Table IV. The peak-to-valley ratio is given (P/V). MLM and MHM stand for the median mass number at half-maximum height for the light and heavy fragment peaks, FWHM for the full width at half-maximum of the light and heavy fragment peaks (the mass distribution is supposed to be symmetric), and $\langle\nu\rangle$ for the average number of emitted neutrons. As generally observed, the MHM remains practically constant, while the MLM varies almost linearly with the mass of the fissioning nucleus. Following Vandenbosch and Huizenga,[27] a change of 2.6 amu is expected, for the MLM, in going from ^{235}U to ^{238}U. We observed 2.5 ± 0.4 amu.

Within the error, our experiment gives the same value of about 15.5 amu for the FWHM for the pho-

tofission of ^{235}U and ^{238}U. Swindle *et al.*[7] found a value of 16.0 amu for the photofission of ^{238}U with 24- and 26-MeV bremsstrahlung. For the thermal neutron and fission neutron induced fission, Vandenbosch and Huizenga[27] adopted a value of 15 amu, and for the fission neutron induced fission on ^{238}U and the 14-MeV neutron induced fission on ^{235}U and ^{238}U, a value of 16 amu.

For $\langle\nu\rangle$, obtained as the difference between the compound nucleus mass number and the sum of the

TABLE III. Cumulative chain yields for the photofission of ^{238}U.

Mass chain	Yield Present work	A. Chattopadhyay *et al.*[a] (Ref. 8)	Swindle *et al.* (Ref. 7)
77	•••	0.0673	•••
84	0.67 ±0.12	•••	•••
85	1.10 ±0.11	•••	•••
87	2.20 ±0.16	•••	•••
88	2.56 ±0.10	•••	•••
89	3.06 ±0.16	•••	4.22 ±0.29
91	4.09 ±0.20	4.53	5.35 ±0.37
92	4.63 ±0.27	4.33	•••
93	5.13 ±0.30	4.50	5.84 ±0.41
94	5.21 ±0.36	•••	•••
95	5.55 ±0.30	5.40	•••
97	5.73 ±0.29	5.64	5.75 ±0.40
99	6.48 ±0.28	5.98	•••
101	5.57 ±0.29	•••	•••
103	4.79 ±0.36	4.43	•••
104	3.69 ±0.26	•••	•••
105	2.81 ±0.16	2.61	2.10 ±0.15
106	1.84 ±0.16	2.39	•••
107	1.17 ±0.11	•••	•••
109	•••	•••	0.48 ±0.05
111	0.392±0.048	•••	•••
112	0.408±0.031	0.436	0.21 ±0.02
113	0.324±0.039	•••	•••
115	0.334±0.032	0.475	0.22 ±0.02
125	0.454±0.034	•••	•••
127	0.749±0.035	0.972	1.13 ±0.11
129	1.50 ±0.10	1.81	•••
131	3.74 ±0.27	3.31	2.34 ±0.16
132	4.62 ±0.20	5.07	3.40 ±0.24
133	6.31 ±0.32	6.60	5.61 ±0.39
134	6.59 ±0.33	•••	•••
135	6.26 ±0.20	3.77	4.92 ±0.34
137	6.06 ±0.52	•••	•••
138	5.41 ±0.30	•••	•••
140	5.39 ±0.22	5.53	5.00
141	5.55 ±0.40	6.03	•••
142	4.76 ±0.36	5.40	•••
143	4.51 ±0.33	4.51	5.67 ±0.40
144	4.24 ±0.31	3.18	•••
146	3.05 ±0.17	•••	•••
147	2.46 ±0.18	1.78	2.76 ±0.19
148	•••	0.77	•••
149	1.51 ±0.09	•••	2.09 ±0.15
151	0.758±0.061	•••	1.03 ±0.07
153	0.287±0.061	•••	0.49 ±0.03
155	0.085±0.018	•••	•••
156	•••	0.029	0.095±0.007
157	•••	•••	0.080±0.006

[a]Overall error 8%.

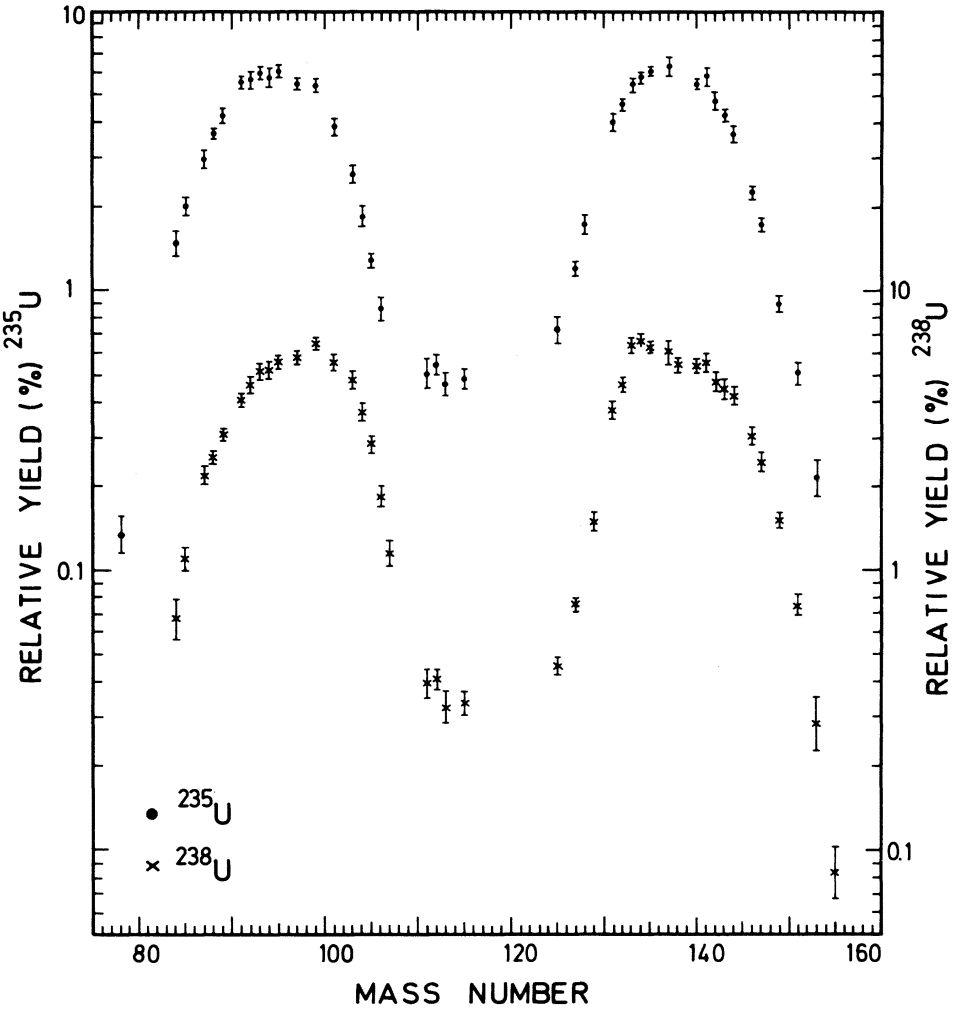

FIG. 1. Postneutron mass distributions for the photofission of ^{235}U and ^{238}U with 25-MeV bremsstrahlung.

average mass of light and heavy fragment group, we found 3.0 ± 0.4. This is in agreement with the value 3 of Swindle *et al.*[7] Based on the relation between $\langle\nu\rangle$ and the average excitation energy of the compound nucleus, given by Veyssière *et al.*[20] for the photofission of ^{238}U and by Bowman, Auchampaugh, and Fultz[19] for the photofission of ^{235}U, we find for the average excitation energy of the ^{235}U and ^{238}U nuclei in our experiment, respectively, the values 10.5 ± 2.5 MeV and 10.0 ± 2.5 MeV. The given errors take into account only our experimental error on $\langle\nu\rangle$. No uncertainty on the relation or on the coefficients of the relation given by Bowman *et al.*[19] or Veyssière *et al.*[20] was included.

TABLE IV. Parameters of the mass distributions.

	^{235}U	^{238}U
P/V	13 ± 1	19 ± 2
MLM (amu)	94.6 ± 0.3	97.1 ± 0.3
MHM (amu)	137.4 ± 0.3	138.0 ± 0.3
FWHM(amu)	15.3 ± 0.4	15.6 ± 0.4
$\langle\nu\rangle$	3.0 ± 0.4	3.0 ± 0.4

B. Independent yields

In Table V we give our results concerning the independent chain yields of a number of fission products for the photofission of ^{235}U and ^{238}U with 25-MeV bremsstrahlung together with the related data available in the literature. For ^{235}U no independent yield data are available, while for ^{238}U, Chattopadhyay *et al.*[8] determined a few independent yields. Generally our results agree with the data of the literature except for ^{136}Cs, where there also

TABLE V. Fractional independent chain yields.

Isotope	$t_{1/2}$	^{235}U This work	^{238}U This work	^{238}U Chattopadhyay *et al.* (Ref. 8)	^{238}U Cunninghame *et al.* (Ref. 6)
^{126}Sb^{g}	12.4 day	0.089 ± 0.010	•••	0.12 ± 0.05	•••
^{126}Sb^{m}	19 min	0.100 ± 0.016	•••	•••	•••
^{128}Sn[a]		0.576 ± 0.060	0.741 ± 0.083	•••	•••
^{128}Sb	9 h	0.216 ± 0.024	0.095 ± 0.020	0.10 ± 0.02	•••
^{128}Sb	10 min	0.173 ± 0.060	•••	•••	•••
^{131}Sb[a]		0.492 ± 0.049	0.802 ± 0.079	•••	•••
^{131}Te^{g}	25 min	0.187 ± 0.035	0.107 ± 0.036	•••	•••
^{131}Te^{m}	30 h	0.265 ± 0.030	0.086 ± 0.013	•••	•••
^{132}I		0.091 ± 0.010	0.019 ± 0.005	0.02 ± 0.005	•••
^{133}I		•••	•••	0.08 ± 0.015	•••
^{134}I		0.485 ± 0.031	0.25 ± 0.03	0.19 ± 0.02	•••
^{135}Xe		0.257 ± 0.010	0.066 ± 0.010	0.09 ± 0.02	•••
^{136}Cs		0.034 ± 0.004	0.004 ± 0.001	0.008 ± 0.0015	0.0018
^{140}La		0.0074 ± 0.0016	•••	•••	0.0011

[a] Fractional cumulative chain yield.

exists a considerable discrepancy among the results of Chattopadhyay *et al.*[8] and those of Cuninghame *et al.*[6]

In the thermal neutron induced fission of ^{235}U, Wahl *et al.*[28] found that within a given mass chain the charge of the fission fragments has a Gaussian distribution

$$P(Z)=\frac{1}{\sqrt{\pi c}}\exp\left[\frac{-(Z-Z_p)^2}{c}\right]. \quad (1)$$

For the width parameter c of this distribution he found the value 0.80 ± 0.14, constant over the whole range of fission products. From a review of thermal neutron induced fission studies Amiel and Feldstein[29] concluded that superimposed on the Gaussian distribution proposed by Wahl *et al.*,[28] they had to include a proton pairing effect with an average value of + 25% for even-Z nuclei and −25% for odd-Z nuclei. In their studies of the α particle induced fission of ^{232}Th and ^{235}U from fractional independent yield data for mass 135, McHugh and Michel[30] observed for the compound nucleus ^{236}U that the c parameter has the constant value 0.95 ± 0.05 over the excitation energy range from 15–18 MeV to 39 MeV. No noteworthy deviation of the charge of the fragments in a given mass chain from a Gaussian distribution could be observed, so that in the experiments of McHugh and Michel[30] the importance of shell structure or proton pairing effects was very small.

As for photofission no experimental data are available concerning the width parameter or proton pairing effects, we calculated from the independent chain yields for the photofission of ^{235}U and ^{238}U the value of Z_p, both by using $c = 0.80$ including 25% odd-even proton pairing effect, and by using $c = 0.95$ without proton pairing effect. The results are given in Table VI. One might expect that the

TABLE VI. Z_p and $\Delta Z_p/\Delta A$ values.

		$c = 0.80$ 25% odd-even effect			$c = 0.95$ No odd-even effect		
M_{post}	M_{pre}	$Z_p(^{235}U)$	$Z_p(^{238}U)$	$-\frac{\Delta Z_p}{\Delta A}$	$Z_p(^{235}U)$	$Z_p(^{238}U)$	$-\frac{\Delta Z_p}{\Delta A}$
126	126.00	50.14 ± 0.05	•••	•••	49.97 ± 0.05	•••	•••
128	128.22	50.56 ± 0.11	50.29 ± 0.17	0.09 ± 0.07	50.38 ± 0.12	50.10 ± 0.18	0.09 ± 0.07
131	131.56	51.34 ± 0.07	50.91 ± 0.08	0.14 ± 0.04	51.51 ± 0.09	50.97 ± 0.11	0.18 ± 0.05
132	132.67	51.85 ± 0.04	51.40 ± 0.08	0.15 ± 0.03	51.67 ± 0.04	51.20 ± 0.08	0.16 ± 0.03
134	134.89	52.75 ± 0.05	52.32 ± 0.06	0.14 ± 0.03	52.59 ± 0.06	52.12 ± 0.06	0.16 ± 0.03
135	136.00	53.03 ± 0.02	52.59 ± 0.04	0.15 ± 0.02	53.12 ± 0.02	52.56 ± 0.05	0.19 ± 0.02
136	137.11	53.55 ± 0.04	53.04 ± 0.05	0.17 ± 0.02	53.36 ± 0.04	52.83 ± 0.07	0.18 ± 0.03
140	141.56	55.17 ± 0.05	•••	•••	54.96 ± 0.05	•••	•••

actual Z_p values for the photofission of ^{235}U and ^{238}U should lie between the two calculated values.

In Fig. 2 the calculated Z_p values are compared with the most probable charges obtained with the UCD hypothesis (Z_{UCD}). As the neutron emission curves $\nu(M)$ for photofission are not determined, the postneutron emission masses were converted into preneutron emission masses using the empirical neutron emission saw tooth curve of Terrell[31] obtained from a study of the spontaneous fission of ^{252}Cf and the thermal neutron induced fission of ^{233}U, ^{235}U, and ^{239}Pu.

No definite conclusions concerning the existence of odd-even effects could be drawn. The lines corresponding to $Z = 50$ and $N = 82$ are also indicated in Fig. 2. For both ^{235}U and ^{238}U the Z_p values tend to remain close to the $Z = 50$ line. On the other hand, no pronounced $N = 82$ effect was observed. These results are in agreement with the observations in thermal neutron induced fission of ^{235}U.[32]

Based on the available data for thermal neutron induced fission and spontaneous fission, Nethaway[33] investigated the variation of Z_p with the number of protons (Z_c), the number of neutrons (N_c), and the excitation energy (E_c^*) of the compound nucleus.

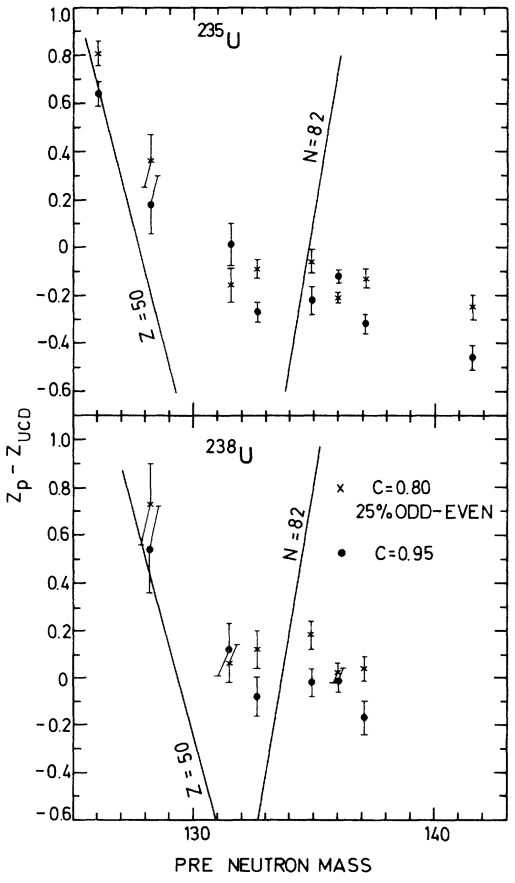

FIG. 2. Z_p-Z_{UCD} as a function of the preneutron fragment mass for the photofission of ^{235}U and ^{238}U with 25-MeV bremsstrahlung.

For the difference $\Delta' Z_p$ between the Z_p of a given mass chain in the fission of a compound nucleus (N_c, Z_c) with an excitation energy E_c^*, and the corresponding Z_p value in the thermal neutron induced fission of ^{235}U Nethaway found

$$\Delta' Z_p = a(Z_c - 92) + b(A_c - 236) + c'(E_c^* - 6.52) \ . \tag{2}$$

For the heavy fragment mass region the parameters a and b have the constant values 0.547 ± 0.010 and -0.188 ± 0.004, respectively, while the parameter c' depends strongly on the fragment mass. An expression of the form of Eq. (2) was first proposed by Coryell, Kaplan, and Fink[34] to generalize the equal charge displacement (ECD) hypothesis, where a and b have the values 0.5 and -0.21.

Using the value of 10.5 MeV for the average excitation energy (E_c^*) of the compound nucleus, as is obtained from the average number of emitted neutrons, we calculated the expected Z_p values from relation (2) and the known Z_p values for the thermal neutron induced fission of ^{235}U. We found Z_p values too small as compared to the experimentally determined ones. By using 13.0 MeV, the upper limit of E_c^* obtained from the average number of emitted neutrons, agreement between the calculated and measured Z_p values can be obtained. Only for the mass chains 126 and 128 are relatively important deviations (about 0.30 to 0.40 charge units) between the measured and calculated Z_p values observed. However, the tendency of the Z_p function to follow the $Z = 50$ line was not taken into account in relation (2).

The parameter $b = \Delta Z_p / \Delta A = [Z_p(^{238}\mathrm{U}) - Z_p(^{235}\mathrm{U})] / [A(^{238}\mathrm{U}) - A(^{235}\mathrm{U})]$, as obtained from our experiments (see Table VI), has a constant value in the mass region 130–140. For $c = 0.80$ [relation (1)] and including 25% proton pairing effect we find an average value of $b = -0.154 \pm 0.012$, and for $c = 0.95$ without odd-even effect, an average value of $b = -0.177 \pm 0.013$. The difference between the two values of b is mainly due to the difference in the c parameter, and is scarcely influenced by introducing proton pairing effects. Due to the vicinity of the closed $Z = 50$ shell a lower value for b is found for mass chain 128. The value $b = -0.177 \pm 0.013$ obtained for $c = 0.95$ without including proton pairing effects is in agreement with the value -0.188 ± 0.004 of Nethaway.[33] The determined values of b are significantly smaller than those expected from the ECD hypothesis, giving $b = -0.21$, or from the UCD hypothesis, giving a value of b

from −0.21 to −0.23 for preneutron masses in the region 128–137.

C. Isomeric ratios

The fractional independent chain yields of the isomeric pairs $^{126}Sb^g$-$^{126}Sb^m$, ^{128}Sb(10 min)-^{128}Sb (9 h) and $^{131}Te^g$-$^{131}Te^m$ were measured for the photofission of ^{235}U. For the photofission of ^{238}U the fractional independent chain yields of the isomeric pair $^{131}Te^g$-$^{131}Te^m$ and of the ^{128}Sb(9 h) isomer were measured, while the yield of ^{128}Sb(10 min) was determined from the Z_p value for mass chain 128 deduced from the fractional independent cumulative chain yield of ^{128}Sn. The value 0.80 for the c parameter of the charge distribution [see relation (1)], including 25% odd-even proton effect, gives the same result for the isomeric ratio ^{128}Sb(10 min)-^{128}Sb(9 h) in the photofission of ^{238}U as the value 0.95 without odd-even proton effects.

The following isomeric transition intensities were used: $^{126}Sb^m \rightarrow {}^{126}Sb^g$: 14%[16]; no isomeric transition between ^{128}Sb(10 min) and ^{128}Sb(9 h)[35]; $^{131}Te^m \rightarrow {}^{131}Te^g$: 22.2%.[17] We assumed that ^{128}Sn decays entirely to ^{128}Sb(10 min),[35] and that the $^{131}Te^m$ branch from ^{131}Sb is 6.8%.[36]

Our isomeric ratios $\sigma_m/(\sigma_m+\sigma_g)$, calculated from the independent yields of the isomeric pairs, are given in Table VII together with similar results of other authors. The spins of the ^{126}Sb, ^{128}Sb, and ^{131}Te isomers and ground states were taken from Orth, Dropesky, and Freeman,[16] Auble,[35] and Jackson,[17] respectively. Only Warhanek and Vandenbosch[37] have measured isomeric ratios for photofission; they studied the $^{134}Cs^m$-$^{134}Cs^g$ pair for the fission of ^{233}U with 16-MeV bremsstrahlung.

If we assume a functional form of the spin distribution of the primary fission fragments, we can determine approximately the average primary fragment spin corresponding to the observed isomeric ratio.[36,37] As pointed out by Sarantites, Gordon, and Coryell[36] and Warhanek and Vandenbosch,[37] the statistical theory gives for the primary fragment spin the following distribution:

$$P(J_i) = (2J_i+1)\exp\left[\frac{-J_i(J_i+1)}{B^2}\right] . \quad (3)$$

$P(J_i)$ is the probability of a primary fragment with spin J_i, and B is a kind of spin-cutoff parameter which is to be determined. The disturbance of the primary spin distribution by the emission of neutrons and γ rays is calculated using the statistical treatment of neutron and γ emission, introducing the spin cutoff factors σ_n and σ_γ.[38,39]

In a careful investigation of the dependence of the isomeric ratios for ^{131}Te and ^{133}Te on variations of different parameters in the thermal neutron induced fission of ^{235}U, Sarantites *et al.*[36] showed that the average spin $\overline{J_i}$ is not strongly dependent on the assumption concerning the emission of neutrons and γ rays. Therefore, in calculating the average value of the initial spin for ^{131}Te, we used for photon induced fission the relation between B and the isomeric ratio of Sarantites *et al.*[36] for the thermal neutron induced fission of ^{235}U. They proposed for the uncertainty on the average initial spin, inherent in the method, a value of 1.2 $\hbar$.

In the mass region 126–128 practically no neutrons are emitted according to the neutron emission curve of Terrell.[31] Therefore, for the isomeric ratios in ^{126}Sb and ^{128}Sb, we calculated only the change of angular momentum due to the emis-

TABLE VII. Independent isomeric yield ratios in fission.

Target	Projectile	Projectile energy (MeV)	Isomeric pair	Spin	$\frac{\sigma_m}{\sigma_m+\sigma_g}$	Ref.
^{233}U	γ (bremsstrahlung)	16	$^{134}Cs^m$-$^{134}Cs^g$	8-4	0.43	37
	n	Thermal	$^{131}Te^m$-$^{131}Te^g$	$\frac{11}{2}-\frac{3}{2}$	0.66 ± 0.03	36
^{235}U	γ (bremsstrahlung)	25	$^{126}Sb^m$-$^{126}Sb^g$	5-8	0.53 ± 0.05	This work
			^{128}Sb(10 min)-^{128}Sb(9 h)	5, 6, 7-8	0.45 ± 0.09	This work
			$^{131}Te^m$-$^{131}Te^g$	$\frac{11}{2}-\frac{3}{2}$	0.58 ± 0.06	This work
	n	Thermal	$^{131}Te^m$-$^{131}Te^g$	$\frac{11}{2}-\frac{3}{2}$	0.64 ± 0.05	36
			$^{133}Te^m$-$^{133}Te^g$	$\frac{11}{2}-\frac{3}{2}$	0.61 ± 0.010	36
^{238}U	γ (bremsstrahlung)	25	^{128}Sb(10 min)-^{128}Sb(9 h)	5, 6, 7-8	0.62 ± 0.22	This work
			$^{131}Te^m$-$^{131}Te^g$	$\frac{11}{2}-\frac{3}{2}$	0.45 ± 0.09	This work
^{239}Pu	n	Thermal	$^{131}Te^m$-$^{131}Te^g$	$\frac{11}{2}-\frac{3}{2}$	0.77 ± 0.04	36

sion of γ rays. The emission of one neutron would increase our $\overline{J_i}$ value by 0.5 to 1 $\hbar$, depending on the B and σ_n values. Because of the lack of data concerning γ-ray emission in photofission, we made the assumption that three $E1$ γ rays are emitted per fission by each of the fragments. A value of 0 to 1 given in this mass region by Pleasonton, Ferguson, and Schmitt[40] for $^{235}U(n_{th},f)$ seems to us to be too low for our photofission experiments, taken into account the excitation energy of the compound nucleus, so that rather the average number of emitted γ rays was taken. In addition, we may remark that for a given B value the number of emitted γ rays, including also eventually the emission of $E2$ γ rays, does not much influence the observed isomeric ratio. In the region of the experimental values of the isomeric ratios, the variation of $\overline{J_i}$ by emission of 0 to 3 γ rays amounts to 0.7 $\hbar$. In Fig. 3 we show the isomeric ratio, for a ground-state spin value 8 and a spin 5 for the isomeric state (e.g., case of ^{126}Sb), as a function of B, supposing no emission of γ rays (curve A) and supposing the emission of 3 γ rays, once taking the spin cutoff factor $\sigma_\gamma = 4$ (curve B), and once taking $\sigma_\gamma = 3$ (curve C). Following Sarantites *et al.*[36] we have taken for the isomeric ratio corresponding to a given B value, the average value of the isomeric ratios for $\sigma_\gamma = 3$ and $\sigma_\gamma = 4$.

In Table VIII we give the average initial spin values, $\overline{J_i}$ calculated from our observed isomeric ratios. In the experimental error a contribution of 1.2 $\hbar$, inherent in the method, was taken into account. For ^{128}Sb the $\overline{J_i}$ values, corresponding to the three proposed spin values of ^{128}Sb(10 min) are calculated. Our results are in agreement with those given in the literature for spontaneous and low energy fission.[36,37,41,42] For the $^{134}Cs^m$-$^{134}Cs^g$ pair in the photofission of ^{233}U with 16-MeV bremsstrahlung, Warhanek and Vandenbosch[37] found a $\overline{J_i}$ value of 6.5 $\hbar$.

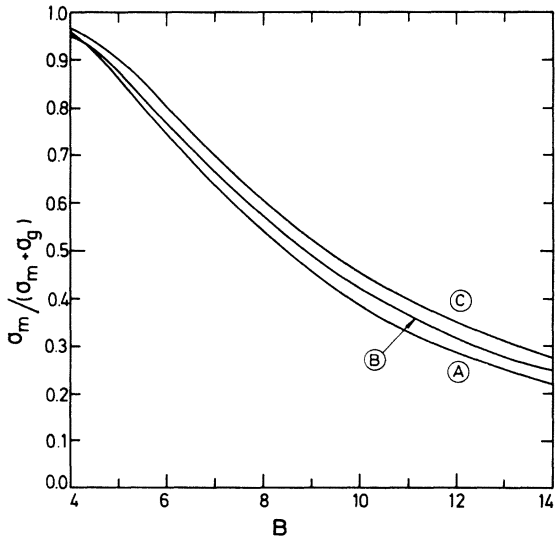

FIG. 3. Isomeric ratio $\sigma_m/(\sigma_m+\sigma_g)$ as a function of B for a ground-state spin 8 and isomer spin 5. Curve A: no emission of γ rays; Curve B: emission of 3 $E1$ γ rays, $\sigma_\gamma = 4$; Curve C: emission of 3 $E1$ γ rays, $\sigma_\gamma = 3$.

Since the photon absorption in our energy region is predominantly $E1$ absorption, the spin and parity of the compound nucleus ^{235}U is $\frac{5}{2}^+$, $\frac{7}{2}^+$, or $\frac{9}{2}^+$, and the spin and parity of the compound nucleus ^{238}U is 1^-. The discrepancy between the spin values of the compound nucleus and the primary fragments (see Table VIII), was also observed in other fissioning systems.[36,37,41,42] It was explained as a generation of angular momentum in the fission fragments. A compilation of the theory of different mechanisms leading to production of angular momentum is given by Vandenbosch and Huizenga.[27]

Where a comparison is possible, the obtained primary fragment spin ($\overline{J_i}$) values are systematically higher for the photofission of ^{235}U than for the photofission of ^{238}U, although the differences remain within the experimental error. This is in agreement with the results of Warhanek and Vandenbosch,[37] who observed that the isomeric ratio of the $^{134}Cs^m$-$^{134}Cs^g$ pair is significantly larger for particle induced fission than for photofission. This implies that the spin of the compound nucleus is transferred to a considerable extent into intrinsic angular momentum of the fragments. This seems to be in contradiction with results of Loveland and Shun[43] in medium energy fission, where no correlation between the angular momentum of the compound nucleus and the primary fragments was observed, although the primary fragment angular momenta are found to be higher than those in low energy fission.

The $\overline{J_i}$ value determined from the isomeric ratio of $^{131}Te^m$-$^{131}Te^g$ is relatively low compared to the results for ^{126}Sb and ^{128}Sb. Also, in the thermal neutron induced fission of ^{235}U, where predominantly 3^- and 4^- states in the compound nucleus ^{236}U are produced, the $\overline{J_i}$ value of 5 ± 1.5 $\hbar$ ob-

TABLE VIII. $\overline{J_i}$ values.

Isomeric pair	$\overline{J_i}$ ($\hbar$) $^{235}U(\gamma, F)$	$^{238}U(\gamma, F)$
$^{126}Sb^m$-$^{126}Sb^g$	7.2 ± 1.4	•••
^{128}Sb(10 min)-^{128}Sb(9 h)	$8.1 \pm 1.7(5^+)$	$6.3 \pm 2.8(5^+)$
	$8.8 \pm 1.7(6^+)$	$6.8 \pm 2.8(6^+)$
	$9.5 \pm 1.7(7^+)$	$7.3 \pm 2.8(7^+)$
$^{131}Te^m$-$^{131}Te^g$	4.4 ± 1.4	3.3 ± 1.6

tained for ^{131}Te [37] is lower than the average value of $8 \pm 2\ \hbar$ [41] found for gross fission products.

IV. CONCLUSION

The observation of a fine-structure peak at mass 133–134 in the mass distribution for ^{238}U, and the behavior of the Z_p value in the vicinity of $Z = 50$ in our photofission studies of ^{235}U and ^{238}U with 25-MeV bremsstrahlung, provide evidence that at an average excitation energy of the order of 11–12 MeV the influence of fragment shell effects is still observed.

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