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Product yields for the photofission of ^{238}U with 12-, 15-, 20-, 30-, and 70-MeV bremsstrahlung

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Cumulative yields for about 40 mass chains and fractional independent yields of ^{128}Sn, ^{128}Sb^{g}, ^{131}Te^{g}, ^{131}Te^{m}, ^{132}I^{g}, ^{132}I^{m}, ^{134}I, ^{134}Cs, ^{135}Xe, and ^{136}Cs were determined for the photofission of ^{238}U with 12-, 15-, 20-, 30-, and 70-MeV bremsstrahlung. Changes in the characteristics of the mass distribution are studied. An enhanced yield in the mass region 133-134, decreasing with increasing end-point energy of the bremsstrahlung (E_e), is observed. From the determined independent yields the most probable charges $Z_p(E_e)$ are calculated using the value 0.85 for the width parameter c of the charge distribution as deduced from our results for mass chain 134. The determined $Z_p(E_e)$ values are very well described by the empirical relation of Nethaway except for the mass chain 136. This discrepancy for mass chain 136, which decreases with increasing end-point energy of the bremsstrahlung, is attributed to an abnormal low yield of ^{136}Cs. Fragment shell effects are discussed. From the isomeric ratios for ^{131}Te^{g}-^{131}Te^{m} and ^{132}I^{g}-^{132}I^{m} average initial fragment spins are calculated using a statistical model analysis.

[NUCLEAR REACTIONS, FISSION ^{238}U(γ,F), $E_{\gamma\,\max}=12$, 15, 20, 30, 70 MeV; measured: fragment γ-ray spectra; deduced: mass distributions, most probable charges, isomeric ratios, average initial fragment spins.]

I. INTRODUCTION

Photofission mass distributions of ^{238}U have been reported already by different authors.[1-10] However, for the photofission of ^{238}U, changes in the characteristics of the mass distribution with the end-point energy of the bremsstrahlung were only studied by Schmitt and Sugarman,[1] Katz *et al.*,[3] Duffield *et al.*,[4] and Chattopadhyay *et al.*[8] By comparing the photofission yields with the yields of the corresponding mass chains in the thermal neutron induced fission of ^{235}U Schmitt and Sugarman[1] determined the cumulative yield of respectively 14, 26, 18, and 16 mass chains for the photofission of ^{238}U with 21-, 48-, 100-, and 300 MeV-bremsstrahlung. In addition they were able to deduce the asymmetric-to-symmetric fission ratio for 7-, 10-, and 16-MeV bremsstrahlung. Katz *et al.*[3] investigated in more detail the variation of the peak-to-valley ratio with the end-point energy for bremsstrahlung in the energy range from 12 to 24 MeV. Duffield *et al.*[4] measured photofission yields for 5.5-MeV to 8-MeV thick-target bremsstrahlung. Mass distributions for the photofission of ^{238}U with thick-target bremsstrahlung with end-point energy between 25 and 40 MeV have been determined by Chattopadhyay *et al.*[8] They measured yields of 23 nuclides by direct γ-ray spectrometry of the target. The results of the other investigators mentioned above were obtained by chemical separation methods. Large differences exist between the photofission yields, reported in the literature for the same bremsstrahlung end-point energy. The use of either thin- or thick-target bremsstrahlung by different authors is probably one of the reasons for these large discrepancies. A systematic investigation of the changes of all the characteristics of the mass distribution for the photofission of ^{238}U as a function of increasing end-point energy of the bremsstrahlung (especially in the giant resonance region) using thin-target bremsstrahlung has not been reported in the literature.

Less information is available concerning the changes of the independent yields of fission products with the end-point energy of the bremsstrahlung for the photofission of ^{238}U. Only Chattopadhyay *et al.*[8] determined the independent yields of five fission products for 25- and 30-MeV end-point bremsstrahlung.

We studied the mass distribution and the independent yields of a number of fission products for the photofission of ^{238}U with thin-target bremsstrahlung with 12-, 15-, 20-, 30-, and 70-MeV end-point energy, using the technique of γ-ray spectrometry of fission product catcherfoils. In addition chemical separation of Cd, I, and Cs were carried out. The average excitation energy of the ^{238}U nucleus was calculated for each end-point energy of the bremsstrahlung. Cumulative yields for about 40 mass chains as well as the fractional independent yields for ^{128}Sn, ^{128}Sb^{g}, ^{131}Sb, ^{131}Te^{g}, ^{131}Te^{m}, ^{132}I^{g}, ^{132}I^{m}, ^{134}I, ^{134}Cs, ^{135}Xe, and ^{136}Cs were determined. From our results the width parameter c of the charge distribution of mass chain 134 could be deduced for 20-, and 30-MeV bremsstrahlung. The most probable charges are calculated and compared with the values predicted by the unchanged charge distribution (UCD) hypothesis and with the values expected from the empirical relation of Nethaway.[11] Fragment shell effects are discussed. For the photofission of ^{238}U with 15-, 20-, 30-, and 70-MeV bremsstrahlung the isomeric ratios for ^{131}Te^{g} – ^{131}Te^{m} and ^{132}I^{g} – ^{132}I^{m} are determined and the corresponding average initial fragment spins are calculated.

II. EXPERIMENTAL PROCEDURE

For our catcherfoil experiments with 15-, 20-, 30-, and 70-MeV bremsstrahlung, the ^{238}U target consisted of an 18 mg/cm^2 U_3O_8 layer, enriched up to 99.6% ^{238}U, on a 5 mm thick graphite disk. The active layer with a diameter of 30 mm was followed at a distance of 1 mm by a 0.1 mm thick very pure aluminum catcherfoil. For our experiments with 12-MeV bremsstrahlung the U_3O_8 target was replaced by a foil stack, consisting of 5 natural uranium plates, separated by 0.025 mm thick aluminum catcherfoils. The natural uranium targets were 0.1 mm thick and had a diameter of 35 mm. The targets were prepared at the Central Bureau for Nuclear Measurements Euratom Geel.

The bremsstrahlung was produced by an analyzed electron beam of the linac of the Nuclear Physics Laboratory in a 0.1 mm thick gold target. The photon beam was cleared of electrons by a cleaning magnet and the deflected electrons were caught in a watertank. Irradiation times varied from 15 min up to 36 h. After appropriate cooling times γ-ray spectra of the catcherfoils were measured using a 19 cm^3 or a 50 cm^3 Ortec 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 the data handling system described in Ref. 12. A precision pulser was added for deadtime correction. In the measuring conditions the resolution of the 19 cm^3 and 50 cm^3 Ge(Li) detector systems for the 1.33 MeV ^{60}Co γ ray was 2.2 and 2.4 keV, respectively. From the recorded spectra the cumulative or the independent yields are determined as described in our previous papers.[10,13]

In order to measure the cumulative yield of ^{117}Cd^{g+m} and the independent yields of ^{132}I^{g}, ^{132}I^{m}, ^{134}Cs, and ^{136}Cs, we separated the cadmium, iodine, and cesium fractions from the fission products following radiochemical procedures based on the method of Gleit and Coryell[14] for cadmium, Wahl[15] for iodine, and Cuninghame *et al.*[16] for cesium. For these chemical separations samples of 1 g $UO_2(NO_3)_2 \cdot 6H_2O$ (natural uranium) were irradiated. In addition to the necessary nuclear data of the fission products already given in our previous work,[10,13] we summarize in Table I the adopted values of the absolute intensity (I_γ) of the used γ transitions in the decay of ^{117}Cd^{m}, ^{132}I^{m}, and ^{134}Cs, and the half-lives of these isotopes ($T1/2$). As estimation of the upper limit of the contribution of neutron induced fission in our experiments is obtained by inserting a 13 cm thick lead filter in the photon beam. The decrease in the fission yield compared to the yield without lead shield indicates that even in our experiments with 70-MeV bremsstrahlung the contribution of neutron induced fission was less than 1%.

TABLE I. Additional nuclear data for studied fission products.

Isotope	E_γ (keV)	I_γ (%)	$T\frac{1}{2}$	Ref.
^{117}Cd^{m}	564.4	12.4	3.5 *h*	17
^{132}I^{m}	175.0	8.3	83.6 min	18
^{134}Cs	604.7	97.6	2.062 yr	19
	795.8	85.4		

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Average excitation energy

The average excitation energy of the ^{238}U compound nucleus corresponding to the different end-point energies of the bremsstrahlung $\langle E_{\text{exc}}(E_e)\rangle$ is given by

$$\langle E_{\text{exc}}(E_e)\rangle = \frac{\int_0^{E_e} k\sigma_{\gamma,F}(k)\phi(E_e,k)dk}{\int_0^{E_e}\sigma_{\gamma,F}(k)\phi(E_e,k)dk}\,;$$

E_e and k are respectively the electron and photon energy, $\sigma_{\gamma,F}(k)$ is the cross section for photofission, and $\phi(E_e,k)$ represents the bremsstrahlung spectrum. In our calculations we used for $\phi(E_e,k)$ the Schiff[20] form for thin-target bremsstrahlung. Direct measurements of the cross section for photofission of ^{238}U exist only in the energy range from 5 to 18 MeV.[21,22] Above this energy we used the extrapolation of the cross section deduced by Shotter *et al.*[23] This extrapolation is based on the cross sections for symmetric and asymmetric fission derived by Katz *et al.*[3] The calculated average excitation energies of the ^{238}U compound nucleus after irradiation with bremsstrahlung with an end-point energy of 12, 15, 20, 30, and 70 MeV are 9.7, 11.6, 13.4, 14.7, and 19.9 MeV respectively. Owing to the important contribution of the not directly measured tail of the cross section above the giant resonance, in the calculation of the average excitation energy for 70-MeV bremsstrahlung the value of 19.9 MeV is rather unreliable.

B. Mass distribution

The cumulative yields of 39, 40, 40, 41, and 43 mass chains respectively were determined for the photofission of ^{238}U with 12-, 15-, 20-, 30-, and 70-MeV bremsstrahlung. The results are given in Table II. The mass distributions are normalized to a total yield of 200% as usual. For these normalizations, the unmeasured mass yields were determined by extra- or interpolation as described in Ref. 24. The uncertainties given in Table II were obtained in the same way as lined out in our previous paper.[10] As it was only possible to measure the cumulative yield of $^{115}Cd^g$ and not that of $^{115}Cd^m$, using γ-ray spectrometric methods, we have calculated the contribution of $^{115}Cd^m$ in the yield of mass 115, using the value 14 (see Ref. 1) for the ratio of the yield of $^{115}Cd^g$ to $^{115}Cd^m$. This ratio was found to be constant for the photofission with bremsstrahlung with an end-point energy ranging from 16 to 100 MeV.[1] Up till now the cumulative yields of the masses 85, 87, 88, 94, 101, 104, 107, 146, and 155 were never published by other authors for the photofission of ^{238}U with bremsstrahlung in the energy range of our experiments.

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

M_{post} \ E_e (MeV)	12	15	20	30	70
84	0.76 ±0.14	0.90 ±0.09	0.84 ±0.13	0.90 ±0.10	0.93 ±0.12
85	0.930±0.069	1.09 ±0.09	1.21 ±0.09	1.20 ±0.09	1.17 ±0.09
87	1.87 ±0.14	2.12 ±0.16	2.06 ±0.15	1.99 ±0.14	1.99 ±0.14
88	2.27 ±0.16	2.45 ±0.12	2.60 ±0.11	2.62 ±0.14	2.65 ±0.12
89	3.00 ±0.18	3.10 ±0.17	3.01 ±0.16	3.04 ±0.16	2.95 ±0.16
91	4.41 ±0.19	4.39 ±0.19	4.30 ±0.19	4.20 ±0.19	3.99 ±0.17
92	4.56 ±0.29	4.78 ±0.25	4.80 ±0.27	4.76 ±0.25	4.69 ±0.25
93	4.93 ±0.43	5.15 ±0.39	5.02 ±0.36	4.93 ±0.35	4.82 ±0.35
94	5.52 ±0.40	5.57 ±0.40	5.33 ±0.38	5.32 ±0.38	5.13 ±0.37
95	5.92 ±0.25	6.10 ±0.34	5.70 ±0.25	5.75 ±0.25	5.49 ±0.25
97	6.03 ±0.30	5.82 ±0.29	5.87 ±0.29	5.69 ±0.29	5.42 ±0.27
99	6.76 ±0.28	6.13 ±0.26	6.17 ±0.26	6.09 ±0.25	5.90 ±0.25
101	5.82 ±0.33	5.71 ±0.31	5.59 ±0.30	5.51 ±0.28	5.51 ±0.31
103	5.61 ±0.40	5.14 ±0.37	4.97 ±0.37	4.82 ±0.35	4.62 ±0.33
104	3.69 ±0.27	3.51 ±0.26	3.56 ±0.26	3.37 ±0.26	3.32 ±0.24
105	2.71 ±0.28	2.67 ±0.18	2.68 ±0.11	2.57 ±0.24	2.66 ±0.11
106	1.74 ±0.17	1.67 ±0.23	1.64 ±0.24	1.75 ±0.18	1.95 ±0.17
111	• • •	• • •	• • •	• • •	0.718±0.089
112	0.115±0.022	0.184±0.027	0.311±0.048	0.491±0.049	0.744±0.060
113	• • •	0.195±0.040	0.290±0.039	0.441±0.052	0.745±0.074
115	0.075±0.007	0.172±0.021	0.270±0.025	0.444±0.036	0.695±0.052
117	0.087±0.011	0.171±0.024	0.281±0.031	0.446±0.045	0.737±0.064
123	0.083±0.015	0.176±0.022	0.277±0.034	0.450±0.059	0.683±0.064
125	• • •	• • •	• • •	0.553±0.073	0.746±0.075
127	0.307±0.018	0.533±0.036	0.667±0.042	0.894±0.053	1.045±0.058
129	1.07 ±0.13	1.38 ±0.11	1.403±0.092	1.516±0.095	1.81 ±0.17
131	3.73 ±0.24	4.02 ±0.26	3.90 ±0.24	3.95 ±0.24	3.85 ±0.23
132	4.95 ±0.16	4.68 ±0.31	4.74 ±0.39	4.63 ±0.15	4.34 ±0.24
133	6.80 ±0.34	6.34 ±0.37	6.30 ±0.35	6.10 ±0.31	5.84 ±0.30
134	6.88 ±0.23	6.87 ±0.25	6.84 ±0.22	6.43 ±0.21	6.12 ±0.27
135	6.73 ±0.28	6.58 ±0.29	6.65 ±0.28	6.27 ±0.26	5.72 ±0.25
137	6.20 ±0.48	6.13 ±0.55	6.11 ±0.55	5.97 ±0.52	5.75 ±0.48
140	6.10 ±0.20	5.91 ±0.20	5.59 ±0.20	5.62 ±0.18	5.33 ±0.17
141	5.40 ±0.38	5.38 ±0.29	5.05 ±0.37	5.34 ±0.29	5.21 ±0.27
142	5.07 ±0.47	5.02 ±0.44	4.88 ±0.37	4.60 ±0.38	4.60 ±0.35
143	4.80 ±0.34	4.72 ±0.34	4.53 ±0.32	4.39 ±0.31	4.20 ±0.32
144	4.60 ±0.32	4.24 ±0.46	3.97 ±0.34	3.82 ±0.32	3.65 ±0.26
146	3.05 ±0.22	3.15 ±0.21	2.90 ±0.17	2.82 ±0.18	2.70 ±0.15
147	2.40 ±0.17	2.22 ±0.15	2.16 ±0.12	2.22 ±0.13	2.13 ±0.16
149	1.36 ±0.11	1.22 ±0.15	1.38 ±0.11	1.222±0.089	1.27 ±0.10
151	0.792±0.062	0.742±0.067	0.757±0.070	0.694±0.068	0.720±0.065
153	0.300±0.047	0.324±0.053	0.316±0.039	0.316±0.038	0.326±0.050
155	• • •	• • •	• • •	• • •	0.110±0.034

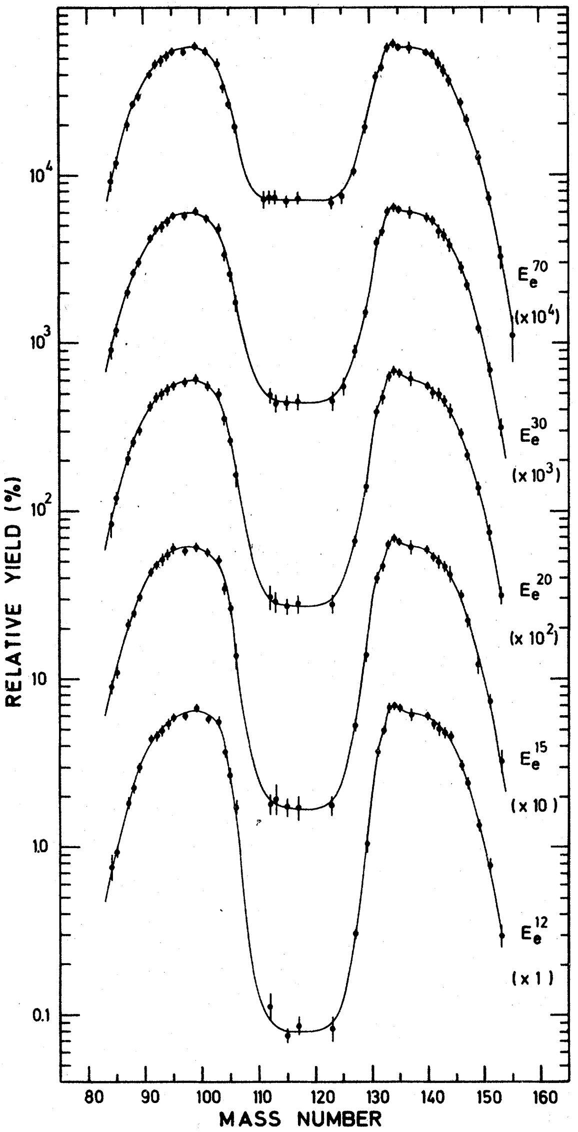

FIG. 1. Post neutron mass distributions for the photofission of ^{238}U with 12-, 15-, 20-, 30-, and 70-MeV bremsstrahlung. The yields have been multiplied by the scale factor given in parentheses.

The obtained mass distributions are plotted in Fig. 1. Striking features of the mass distributions, directly observable from this figure, are a strong dependence of the mass yields for symmetric fission on the end-point energy of the bremsstrahlung, a near independence of the asymmetric yields, and the presence of a fine-structure peak in the mass region 133–135 persisting even for 70-MeV bremsstrahlung induced photofission.

This fine-structure peak is also observed in other fissioning systems e.g. ^{235}U($n_{\rm th}$, f), ^{239}Pu($n_{\rm th}$, f), and ^{241}Pu($n_{\rm th}$, f) (Ref. 25). Nagy *et al.*[26] studied the variation of the mass distribution for the neutron induced fission of ^{238}U in a neutron energy range $E_n = 1.5$ to 7.7 MeV and compared their results with 14.8-MeV neutron data. They also observed an enhanced yield around mass 134 persisting at 14.8-MeV neutron energy. In order to compare quantitatively the fine-structure peaks at different end-point energies, we fitted a quadratic function to the measured complementary points of the fine-structure mass region ($A = 133$–135) and subtracted the yields obtained by interpolation of the complementary masses from the measured yields for masses around $A = 133$–135. The intensity of the fine-structure peak calculated in this way remains practically constant, 2% for 12 to 20-MeV bremsstrahlung, decreases to 1.5% for 30-MeV and to 1% for 70-MeV bremsstrahlung, the total fission yield being normalized to 200% as mentioned above. As Z and N odd-even effects are expected to be small (see also Sec. III C), if not completely negligible at an excitation energy above 10 MeV, the yield excess observed in our photofission studies must be mainly due to neutron emission. Some characteristics of the mass distributions are summarized in Table III. The peak-to-valley ratio (P/V) decreases from 78± 7 to 8.2± 0.7 for bremsstrahlung end-point energies increasing from 12 to 70 MeV. The known bremsstrahlung peak-to-valley ratio results for ^{238}U(γ, F) are summarized in Fig. 2. Our values lie systematically above the ratios determined by Katz *et al.*[3] and are in rather good

TABLE III. Mass distribution characteristics.

E_e (MeV)	12	15	20	30	70
P/V	78 ±7	35 ±4	22 ±2	13.5 ±0.9	8.2 ±0.7
MLM u[a]	97.25±0.08	97.01±0.07	96.99±0.07	96.80±0.08	96.89±0.07
MHM u[a]	137.87±0.07	137.80±0.07	137.61±0.07	137.64±0.07	137.55±0.07
FWHM u	15.3 ±0.4	15.7 ±0.4	15.7 ±0.4	15.7 ±0.4	15.7 ±0.4
$\langle\nu\rangle$	2.85±0.11	3.39±0.12	3.44±0.11	3.57±0.11	3.66±0.11

[a] Uncertainties on MLM, MHM, and $\langle\nu\rangle$ are obtained by a statistical treatment of the uncertainties of the yields.

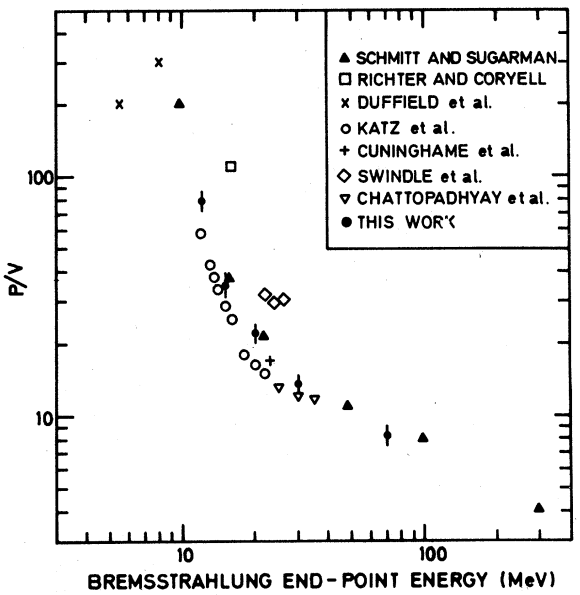

FIG. 2. Summary of the available data on the peak-to-valley ratio for the photofission of ^{238}U.

agreement with the older results of Schmitt and Sugarman.[1]

Other tabulated characteristics of the mass distributions are the mean mass of the light- and heavy-fragment peaks, MLM and MHM (taken into account only those masses having a yield higher than half the maximum yield, to eliminate the contribution of the symmetric fission component), the full width at half maximum of the light and heavy fragment peaks FWHM (the mass distribution is supposed to be symmetric), and the average number of emitted neutrons $\langle\nu\rangle$. Both MLM and MHM show a slow decrease with increasing excitation energy of the compound nucleus due to increasing neutron emission. The FWHM remains practically constant 15.7 ± 0.4 u. The value of $\langle\nu\rangle$ is obtained as the difference between the mass of the fissioning system and the sum of the mean masses of the light and heavy fission product groups, this time taking into account all the fission products including those corresponding to symmetric mass splits. The value of $\langle\nu\rangle$ increases from 2.85 for 12-MeV bremsstrahlung to 3.63 for 70-MeV bremsstrahlung. Caldwell *et al.*[21] have studied the variation of $\langle\nu\rangle$ with the photon energy k for the photofission of ^{238}U with monochromatic γ rays in an energy range from 6 to 17 MeV. They found a good description of the data by the relation

$$\langle\nu\rangle = 1.862 + 0.1234 \times k\,.$$

Substituting in this expression our values of the average excitation energy for k we obtain as the number of emitted neutrons to be expected for the photofission of ^{238}U with 12-, 15-, 20-, and 30-MeV bremsstrahlung respectively 3.06, 3.29, 3.51, and 3.68. These values are in reasonable agreement with our experimental results. In the work of Caldwell *et al.*[21] too the deviations of the experimental points from the given straight line amount to about some 0.2 neutrons. Nagy *et al.*[26] found for the neutron induced fission of ^{238}U good agreement between the directly measured $\langle\nu\rangle$ values and those determined from the mass distribution. Compared to the results of Caldwell *et al.*[21] our measured $\langle\nu\rangle$ value for 70-MeV bremsstrahlung seems to be too low, indicating that the calculated average excitation energy for this bremsstrahlung end-point energy could be too high. One can note, however, that the extrapolation of the cross section for the photofission of ^{238}U by Shotter *et al.*[23] was based on the results of Katz *et al.*[3] The systematic discrepancy between our observed peak-to-valley ratios and those given by Katz *et al.*[3] introduces a large uncertainty on this extrapolated cross section and on the deduced average excitation energy for 70-MeV bremsstrahlung.

C. Independent yields and charge distributions

Our results concerning the fractional independent yields of a number of fission products and the fractional cumulative yields of ^{128}Sn and ^{131}Sb for the photofission of ^{238}U are shown in Table IV. A comparison with data from the literature was not possible as hitherto no systematic study of changes of the independent yields with the end-point energy of the bremsstrahlung for the photofission of ^{238}U was made. A first general trend, directly observable from our data, is the increase of the measured fractional independent chain yields with the average excitation energy.

From an accurate examination of the independent yields in neutron induced fissioning systems Amiel *et al.*[27] concluded that to describe the charge distribution an odd-even effect has to be included, superimposed on a Gaussian distribution with a constant width parameter $c = 0.80 \pm 0.14$. This odd-even effect decreases with increasing excitation energy and proton number of the fissioning nucleus. For the thermal neutron induced fission of ^{235}U (excitation energy of the compound nucleus ^{236}U: 6.52 MeV) the enhancement for even Z elements and the suppression for odd Z elements has the value of $(22 \pm 7)\%$ while for fast neutron induced fission of ^{235}U (average excitation energy of the compound nucleus 8.5 MeV) it is reduced to $(10 \pm 10)\%$, due to the increase of the number of particle-hole excitations during the fission pro-

TABLE IV. Fractional independent yields.

Isotope \ E_e (MeV)	12	15	20	30	70
^{128}Sn [a]	0.77 ± 0.12	0.67 ± 0.11	0.700 ± 0.090	0.702 ± 0.090	0.615 ± 0.076
^{128}Sb^{g}	∘ ∘ ∘	$(4.9 \pm 1.9) \times 10^{-2}$	$(6.2 \pm 2.3) \times 10^{-2}$	$(8.5 \pm 2.4) \times 10^{-2}$	0.188 ± 0.030
^{131}Sb [a]	0.915 ± 0.071	0.855 ± 0.050	0.828 ± 0.046	0.798 ± 0.049	0.761 ± 0.061
^{131}Te^{g}	∘ ∘ ∘	$(7.8 \pm 4.8) \times 10^{-2}$	$(8.5 \pm 4.5) \times 10^{-2}$	$(9.3 \pm 4.3) \times 10^{-2}$	0.109 ± 0.041
^{131}Te^{m}	$(2.9 \pm 0.7) \times 10^{-2}$	$(5.7 \pm 1.1) \times 10^{-2}$	$(7.7 \pm 1.1) \times 10^{-2}$	0.100 ± 0.013	0.110 ± 0.013
^{132}I^{g}	∘ ∘ ∘	$(5.7 \pm 1.4) \times 10^{-3}$	$(7.4 \pm 1.5) \times 10^{-3}$	$(1.17 \pm 0.21) \times 10^{-2}$	$(2.82 \pm 0.51) \times 10^{-2}$
^{132}I^{m}	∘ ∘ ∘	$(3.9 \pm 1.2) \times 10^{-3}$	$(5.7 \pm 0.9) \times 10^{-3}$	$(1.26 \pm 0.17) \times 10^{-2}$	$(2.13 \pm 0.39) \times 10^{-2}$
^{134}I	0.169 ± 0.041	0.213 ± 0.024	0.227 ± 0.028	0.255 ± 0.023	0.275 ± 0.025
^{134}Cs	∘ ∘ ∘	∘ ∘ ∘	$(4.0 \pm 1.5) \times 10^{-5}$	$(8.6 \pm 3.4) \times 10^{-5}$	$(1.33 \pm 0.14) \times 10^{-3}$
^{135}Xe	$(3.5 \pm 1.5) \times 10^{-2}$	$(4.4 \pm 1.7) \times 10^{-2}$	$(5.1 \pm 1.2) \times 10^{-2}$	$(6.0 \pm 1.2) \times 10^{-2}$	$(7.7 \pm 1.3) \times 10^{-2}$
^{136}Cs	$(4.6 \pm 1.1) \times 10^{-4}$	$(1.06 \pm 0.11) \times 10^{-3}$	$(1.86 \pm 0.16) \times 10^{-3}$	$(3.62 \pm 0.32) \times 10^{-3}$	$(1.15 \pm 0.12) \times 10^{-2}$

[a] Fractional cumulative yield.

cess. Following the assumptions of Amiel *et al.*[27] for a rough calculation of the number of particle-hole excitations and using the value 5.8 ± 0.2 MeV for the ^{238}U fission barrier height we expect a 3% odd-even effect in our photofission experiments with 12-MeV bremsstrahlung. For the 14.8-MeV neutron induced fission of ^{235}U too the odd-even fluctuation is found to be absent.[11] Consequently we do not have to take into account an even Z enhancement in our photofission studies.

The charge distribution for photofission with bremsstrahlung is described by the following expression:

$$P(Z)=\frac{\int_0^{E_e} 1/\sqrt{\pi c}\,\exp\{-[Z-Z_p(k)]^2/c\}\,\sigma_{\gamma,F}(k)\,\phi(E_e,k)\,dk}{\int_0^{E_e}\sigma_{\gamma,F}(k)\,\phi(E_e,k)\,dk}. \tag{1}$$

As already mentioned (in Sec. III A) E_e and k are the electron or bremsstrahlung end-point and the photon energy, $\sigma_{\gamma,F}(k)$ and $\phi(E_e,k)$ represent the photofission cross section and the bremsstrahlung spectrum. The charge distribution at a photon energy k was assumed to be Gaussian in shape with a most probable charge $Z_p(k)$ and a width parameter c independent of k. The latter assumption is supported by the work of McHugh and Michel,[28] who found a constant value of 0.95 ± 0.05 for the c parameter in an excitation energy range from 15–18 MeV to 39 MeV of the compound nucleus ^{236}U (α induced fission of ^{232}Th) differing only to a small extent from the value 0.80 ± 0.14 determined for low energy fission. The dependence of $Z_p(k)$ on k is taken from the work of Nethaway,[11] who investigated the variation of the most probable charge with changes in excitation energy and with proton and mass number of the compound nucleus for neutron induced and spontaneously fissioning systems.

From the determined independent yields of ^{134}I and ^{134}Cs, we deduced for the isobaric chain 134 using the expression (1) the value of $c = 0.86 \pm 0.09$ and $c = 0.84 \pm 0.09$ for the photofission of ^{238}U with 20- and 30-MeV bremsstrahlung respectively. These values lie as previously mentioned very close to the values 0.80 ± 0.14 and 0.95 ± 0.05 found in other fissioning systems.

Owing to the inaccuracy of the extrapolated cross section for ^{238}U(γ, F), it is not possible to get information from our 70-MeV bremsstrahlung results.

Using the average of our c values, $c = 0.85$ in expression (1), the $Z_p(E_e)$ values (the top of the charge distribution) of the isobaric chains corresponding to the measured independent yields are calculated for each bremsstrahlung end-point energy (E_e). These results are given in Table V. In the given uncertainties no error on the c value is taken into account, except in the cases where the width parameter c could be determined to-

TABLE V. $Z_p(E_e)$ values for the photofission of ^{238}U.

M_{post} \ E_e(MeV)	12	15	20	30
128	$50.08^{+0.21}_{-0.29}$	$50.24^{+0.17}_{-0.19}$	$50.19^{+0.15}_{-0.17}$	$50.17^{+0.15}_{-0.17}$
131	$50.68^{+0.22}_{-0.50}$	$50.86^{+0.13}_{-0.16}$	$50.92^{+0.11}_{-0.14}$	$50.98^{+0.11}_{-0.13}$
132	∘ ∘ ∘	51.10 ± 0.04	51.16 ± 0.03	51.26 ± 0.03
134	51.95 ± 0.11	52.05 ± 0.06	52.07 ± 0.08	52.12 ± 0.07
135	$52.43^{+0.10}_{-0.15}$	$52.50^{+0.10}_{-0.13}$	$52.52^{+0.06}_{-0.08}$	$52.55^{+0.06}_{-0.08}$
136	52.51 ± 0.05	52.66 ± 0.02	52.75 ± 0.02	52.83 ± 0.02

gether with the top of the charge distribution.

It can be noted that the charge distribution described by expression (1) has practically a Gaussian shape. Assuming a pure Gaussian charge distribution for the photofission of ^{238}U with bremsstrahlung we obtain from our measured independent yields the values 0.90 ± 0.10 and 0.94 ± 0.10 for the width parameter c for the isobaric chain 134 for 20- and 30-MeV bremsstrahlung respectively. The $Z_p(E_e)$ values, calculated from our measured independent yields using a pure Gaussian with $c = 0.90$ for 12-, 15-, and 20-MeV end-point bremsstrahlung and $c = 0.94$ for 30-MeV bremsstrahlung, deviate only to a small extent (< 0.05) from those calculated with expression (1). It can thus be concluded that our independent yield results for photofission of ^{238}U with bremsstrahlung with an end-point energy up to 30 MeV are very well described by assuming a Gaussian shape for the charge distribution.

The $Z_p(E_e)$ values are compared with those expected from the unchanged charge density (UCD) hypothesis (Z_{UCD}) in Fig. 3. As the neutron emission curves for photofission with thin-target bremsstrahlung were not determined, the post-neutron masses are converted into preneutron masses using the neutron emission curve for the photofission of ^{238}U with thick-target bremsstrahlung with an end-point energy of 25 MeV.[24] To obtain neutron emission curves, giving the determined average number of emitted neutrons (see Table III) at the different end-point energies of the bremsstrahlung, the curve of Ref. 24 was multiplied with the appropriate ratio.

The $Z_p(E_e)$ values, obtained from the fractional cumulative chain yields of ^{128}Sn, indicate (see Fig. 3) that the Z_p function tends to remain close to the $Z = 50$ line as observed in the thermal neutron induced fission of ^{235}U (Ref. 29). Thus also in the photofission of ^{238}U with bremsstrahlung the 50-proton shell has a strong influence on the charge distribution.

Following Nethaway[11] the difference, ΔZ_p, between the Z_p value of a given mass chain for a

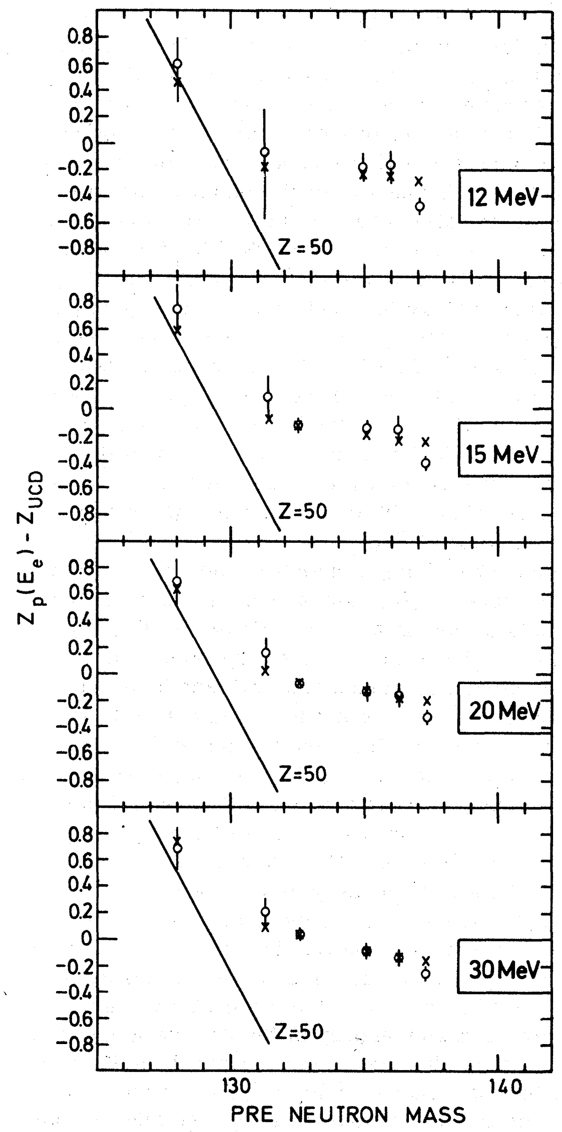

FIG. 3. Z_p-Z_{UCD} versus fragment mass number for the photofission of ^{238}U with 12-, 15-, 20-, and 30-MeV bremsstrahlung. The Z_p values deduced from the measured independent yields are represented by open circles. The Z_p values calculated following Nethaway (Ref. 11) are indicated by the crosses.

fissioning system with mass A_C, proton number Z_C, and excitation energy E_C^*, and the Z_p value for the thermal neutron induced fission of ^{235}U is given by the relation

$$\Delta Z_p = a(Z_C - 92) + b(A_C - 236) + c'(E_C^* - 6.52);$$

a, b, c' are parameters tabulated by Nethaway.

We have calculated the Z_p values for our photofission experiments using this expression. For the value of E_c^* the calculated average excitation energies (see A) were taken. The Z_p values for $^{235}\mathrm{U}(n_{\mathrm{th}}, f)$ were taken from Wahl *et al.*[30] However, the Z_p value 49.66 given by these authors for mass chain 128 was found by extrapolation of Z_p without taking into account the tendency of the Z_p function to remain close to the $Z = 50$ line and without taking into account the measured value $(4 \pm 1) \times 10^{-5}$ (see Ref. 30) of the independent yield of ^{128}I. The corresponding calculated independent yield of ^{128}I is 0.02×10^{-5}. Blachot *et al.*[31] measured the fractional cumulative chain yield of ^{128}Sn and the independent yields of $^{128}\mathrm{Sb}^g$ and $^{128}\mathrm{Sb}^m$ for the thermal neutron induced fission of ^{235}U. They derived a Z_p value of 50.16 for mass chain 128. Using this value one obtains as an estimation of the independent yield of ^{128}I 2×10^{-5}, which is closer to the measured yield. From the behavior of Z_p given in the paper of Kratz and Herrmann[29] too, a Z_p value of 50.16 can be deduced. In view of these results, we have adopted a Z_p value of 50.16 for the mass chain 128 for the thermal neutron induced fission of ^{235}U.

For each end-point energy of the bremsstrahlung the expected $Z_p(E_e)$ values, calculated using the formula of Nethaway, are indicated in Fig. 3 by crosses. There is very good agreement between these calculated values and our experimentally determined ones, except for the most probable charges, deduced from the independent yields of ^{136}Cs. Our experimental $Z_p(E_e)$ values are too low compared to the estimation following Nethaway. This is probably due to an "abnormally" low fission yield of ^{136}Cs. This discrepancy between the experimentally determined and calculated Z_p values for the (postneutron) mass chain 136 decreases with increasing end-point energy of the bremsstrahlung, indicating an independent yield of ^{136}Cs closer to the estimated value at higher excitation energy of the compound nucleus. Also, in the thermal neutron induced fission of ^{235}U the independent yield of ^{136}Cs is found to be too small: The measured fractional independent yield is $(9.3 \pm 0.5) \times 10^{-4}$ (Ref. 30) while the estimated yield including an odd-even effect of 22% amounts to 3.45×10^{-3}. At higher excitation energies of the compound nucleus ^{236}U the discrepancy decreases: The measured yield for fast neutron induced fission of ^{235}U is $(2.33 \pm 0.28) \times 10^{-3}$, the estimated value including 10% odd-even effect is 4.98×10^{-3} (Ref. 11) for 14.8-MeV neutron induced fission of ^{235}U the measured and expected values are 4.81×10^{-2} and 5.35×10^{-2} respectively (Ref. 11). This behavior is in accordance with our photofission results. The "abnormally" low yield of ^{136}Cs can be attributed to the influence of nuclear structure (see Ref. 30).

From the preceding discussion it is clear that generally the formula of Nethaway is also very useful in photofission studies to estimate unmeasured independent yields.

It is interesting to note that for the photofission of ^{238}U with bremsstrahlung with an end-point energy between 20 and 30 MeV, the Z_p behavior in the product mass region of our experiments (128–136) is practically the same as for the thermal neutron induced fission of ^{235}U. Because of the enhancement for $^{235}\mathrm{U}(n_{\mathrm{th}}, f)$ of the fine-structure peak in the mass region 133–134, by the neutron emission, the yield excess in the same mass region in the postneutron mass distribution for the photofission of ^{238}U with bremsstrahlung with an end-point energy in the energy range of our experiments is at least to a large amount due to neutron emission. The presence of this fine-structure peak indicates that shell effects in the fission fragments still are observed in photofission at excitation energies in the region of the giant resonance.

D. Isomeric ratios

In our study the fractional independent yields of the isomeric pairs $^{131}\mathrm{Te}(\frac{3}{2}^+, \frac{11}{2}^-)$ and $^{132}\mathrm{I}(4^+, 8^-)$ were measured for the photofission of ^{238}U with 15-, 20-, 30-, and 70-MeV bremsstrahlung (for 12-MeV bremsstrahlung the obtained activities are too low to yield a meaningful result). The isomeric ratios $\sigma_m/(\sigma_g + \sigma_m)$, calculated from the measured independent yields, are given in Table VI. Again a comparison with other photofission data is impossible.

The measured isomeric ratios for the isomeric pair $^{131}\mathrm{Te}^g - {}^{131}\mathrm{Te}^m$ are lower than the values obtained in the thermal neutron induced fission of ^{235}U, namely 0.66 ± 0.05 (Ref. 32). This can partly be explained by the difference in the adopted values for the absolute intensity of the 149-keV γ ray in the decay of $^{131}\mathrm{Te}^g$: We used 0.681 (Ref. 33). Imanishi *et al.*[32] used 0.72. An increase of the ^{131}Te isomeric ratios of about 0.1 is obtained by using the value 0.72 instead of 0.681.

Although in our photofission studies the changes of the measured isomeric ratios remain between the experimental errors, the results show an

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

Isomeric pair \ E_e (MeV)	15	20	30	70
^{131}Te^m–^{131}Te^g	0.43 ± 0.15	0.48 ± 0.14	0.52 ± 0.12	0.50 ± 0.10
^{132}I^m–^{132}I^g	0.41 ± 0.09	0.44 ± 0.06	0.51 ± 0.06	0.44 ± 0.05

increase of the isomeric ratios with the end-point energy of the bremsstrahlung for both studied isomeric pairs, indicating that the higher spin level is more populated at higher excitation energies. In the framework of the study of the behavior of the isomeric ratios with the excitation energy of the compound nucleus, the errors given in Table VI are certainly overestimated as they are absolute uncertainties, containing a systematic contribution of the uncertainty on the detector efficiency and the absolute intensities of the used γ rays.

From the experimentally determined isomeric ratios the average initial spin of the primary fission fragments, leading by the emission of prompt neutrons and γ rays to the studied isomeric pairs, was deduced using the statistical model analysis, developed by Vandenbosch and Huizenga.[34,35]

As in our previous paper[10] the probability distribution of the initial spin states of the fragments, $P(J_i)$, is assumed to be represented by

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

with B a kind of spin cutoff parameter, to be determined from the measured isomeric ratios.

The modification of the initial spin distribution by the emission of prompt neutrons and γ rays is calculated using the statistical treatment of neutron and γ emission.[34,35] This calculation requires the determination of the number, energy, and transmission coefficients of the emitted neutrons, the number and multipolarity of the emitted γ rays, and the spin cutoff parameters σ_n and σ_γ.

The average number of emitted neutrons of the fragments, leading to the product masses 131 and 132, were deduced from our previous measurements as described in Sec. III C. We assumed the variation of the number of emitted neutrons with the Z value of the fission fragments within a given mass A independent of Z, due to the lack of knowledge on this subject. For the determination of the neutron energy we used the relationship between the average neutron energy and the average number of emitted neutrons for photofission, deduced by Caldwell *et al.*[21] The neutron transmission coefficients were derived from a simple square well potential[36] with a depth of 50 MeV.[37] Because the emitted neutrons are predominantly s-wave neutrons and because of the expected small number of neutrons emitted by the fragments in the considered mass region, the initial spin distribution is not seriously altered by the neutron emission so that the introduced simplifications practically do not influence the calculated average initial spin.

As in our previous paper[10] we assume that after neutron evaporation the residual nucleus deexcites by the emission of three γ rays. These γ cascades are $E1$ with an $E2$ contribution. Based on the work of Aumann *et al.*[38] a quadrupole component of 10% is taken.

The isomeric ratios corresponding to different values of the parameter B were calculated using for σ_n the value 4 and for σ_γ the values 3 and 4. The average of both calculations was taken, following Sarantites *et al.*[39]

To support the choice of the value for σ_n, rough calculations of this spin cutoff parameter for the fission fragments, leading to the fission products ^{131}Te and ^{132}I, were done for the photofission of ^{238}U with 20-MeV bremsstrahlung. As has been done by Aumann *et al.*,[38] for the calculation of the excitation energy of complementary fragments, we assumed once that the excitation energy of all fragments of a given mass division is the same independent of the charge division and once that the kinetic energy release for a given mass split is independent of the charge division. Using the values of our previous work[40] for the average kinetic energy and the Q values of Ref. 41, we obtain depending on the assumption, for ^{131}Te as well as for ^{132}I σ_n, values between 4 and 5. The obtained average initial spin values $\bar{J}_i$ are given in Table VII. In the experimental uncertainties a contribution of 1.2 $\hbar$ inherent in the method was included as proposed by Sarantites *et al.*[39] In the thermal neutron induced fission of ^{235}U Sarantites *et al.*[39] found for the average initial spin, determined from the isomeric pair ^{131}Te^g – ^{131}Te^m, the value 5 ± 1 $\hbar$ and Imanishi *et al.*[32] the value 6.1 ± 0.6 $\hbar$. These values are higher than our photofission values. As already mentioned the ^{131}Te isomeric ratios, determined in our photofission studies, increase by about 0.1 by using the value of Imanishi *et al.*[32] for the absolute intensity of

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

Isomeric pair \ E_e (MeV)	15	20	30	70
$^{131}\mathrm{Te}^m$–$^{131}\mathrm{Te}^g$	3.5 ± 1.7	3.9 ± 1.6	4.3 ± 1.6	4.1 ± 1.5
$^{132}\mathrm{I}^m$–$^{132}\mathrm{I}^g$	6.5 ± 1.6	6.9 ± 1.4	7.8 ± 1.4	6.9 ± 1.4

the 149-keV γ ray of $^{131}\mathrm{Te}^g$. The corresponding increase of $\bar{J}_i$ is about 1 $\hbar$.

Even for the lowest excitation energy (15-MeV bremsstrahlung) the $\bar{J}_i$ values deduced from the ^{131}Te and ^{132}I isomeric ratios are considerably higher than the spin of the compared nucleus ^{238}U (1 $\hbar$, as only $E1$ absorption is assumed) indicating a generation of angular momentum in the fragments during the fission process.

Although the differences between the $\bar{J}_i$ values remain within the experimental uncertainties, our results show a slight increase of the average initial spin values of the fragments with the end-point energy of the bremsstrahlung. These observations indicate that the higher fragment spin states are more often formed at higher excitation energy of the compound nucleus.

IV. CONCLUSIONS

Our studies of the behavior of the product yields for the photofission of ^{238}U shows, as already reported by other authors, a strong dependence of the yields for symmetric mass splits on the excitation energy of the nucleus: The peak-to-valley ratio decreases from 78 to 8 when the bremsstrahlung end-point energy is increased from 12 to 70 MeV. On the contrary, in the region of the asymmetric mass splits the yields remain practically unchanged. The fine-structure peak in the mass region 133–135 mainly due to neutron emission is reduced, but still persists for 30- and 70-MeV end-point bremsstrahlung.

The measured independent yields in the photofission of ^{238}U follow the systematics deduced from the results obtained in neutron induced fission. This is illustrated by the determined value 0.85 for the width parameter c of the charge distribution for mass chain 134 and by the agreement between the experimentally determined Z_p values and those calculated following the method of Nethaway.

The average initial fragment spin values calculated from the isomeric ratios of the isomeric pairs $^{131}\mathrm{Te}^g$ – $^{131}\mathrm{Te}^m$ and $^{132}\mathrm{I}^g$ – $^{132}\mathrm{I}^m$ are systematically higher than the spin of the compound nucleus, as usually observed. They show a tendency to increase slightly with the end-point energy of the bremsstrahlung.

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