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Charge distribution for the photofission of ^{238}U with 20-MeV bremsstrahlung

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Fractional independent or cumulative chain yields of 35 fission products were determined for the photofission of ^{238}U with 20-MeV bremsstrahlung, using direct γ spectrometry of irradiated uranium samples, γ-ray spectrometry of fission product catcher foils, and chemical separation techniques. For the mass chains 131–136 the width parameter c of the charge distribution was obtained. An average value of 0.93 ± 0.06 was deduced for c. A comparison of this c value with the results for other low-energy fissioning systems indicates that the width of the charge distribution is practically independent of the excitation energy of the compound nucleus. Using the average c value, the most probable charges Z_p(20 MeV) were calculated for 8 light and 12 heavy mass chains. A comparison of the deduced Z_p(20 MeV) behavior with the expectations of the unchanged charge density hypothesis shows the higher charge-to-mass ratio of the light fragments compared to the heavy ones and the influence of the closed 50-proton shell on the charge distribution. The Z_p(20 MeV) values determined are very well reproduced by the empirical relation of Nethaway except in the vicinity of the $Z = 50$ line.

[NUCLEAR REACTIONS, FISSION $^{238}U(\gamma,F)$, $E_{\gamma max} = 20$ MeV; measured: fragment γ-ray spectra; deduced: charge distributions, width and most probable charges.]

I. INTRODUCTION

The nuclear charge distribution of the fragments is one of the most interesting observables in fission, as its parameters can be related to the dynamics of the fission process.[1]

For low-energy fissioning systems the independent yields of many fission products were determined using chemical separation techniques[2] and more recently by direct physical methods.[3] Wahl *et al.*[2] proposed a Gaussian shape for the charge distribution of $^{235}U(n_{th},f)$. Amiel and Feldstein[4] found that a proton pairing effect of 25% which decreases strongly with increasing excitation energy and Z of the fissioning system[5] was superimposed on this Gaussian distribution.

In medium energy fission very little information concerning the shape of the charge distribution is available in the literature. McHugh and Michel[6] determined the independent yields of ^{135}I, ^{135}Xe, and ^{135}Cs for the fissioning nucleus ^{236}U, produced at an excitation energy between 15 and 39 MeV by the $^{232}Th(\alpha,f)$ reaction. These data for mass 135 were consistent with a Gaussian charge distribution curve with a constant width $c = 0.95 \pm 0.05$, independent of the excitation energy. Comparing this c value to the value of 0.80 ± 0.14 for $^{235}U(n_{th},f)$ (Ref. 2) one can conclude that the width of the charge distribution is nearly independent of the excitation energy of the compound nucleus up to 39 MeV. Yaffe,[7] however, found an increase of c with increasing excitation energy for the fission of several actinides induced by protons in the energy range from 20 to 85 MeV. These results were not based on direct measurements of the charge distribution but deduced from excitation functions for various Cs isotopes.

Concerning the Z_p behavior in medium energy fission, more information is available. A survey of the existing data is given by Umezawa *et al.*[8] and by Nethaway.[9] They calculated Z_p values from measured independent yields by assuming a pure Gaussian charge distribution with $c = 0.95$ (Umezawa *et al.*[8]) or $c = 0.80$ (Nethaway[9]).

Recently, by using γ spectrometry of fission product catcher foils and by performing chemical separations of the cesium fraction, we deduced the width of the charge distribution for mass chain 134 for the photofission of ^{238}U with 20- and 30-MeV bremsstrahlung (Ref. 10). The values 0.86 $\pm$ 0.09 and 0.84 $\pm$ 0.09, respectively, were obtained. Apart from these results, the data on independent yields for photofission available in the literature are very scarce (Refs. 11–14).

To extend our knowledge on the charge distribution in medium energy fission, especially in photofission, we determined the yield of a number of short-lived (5 s $< T_{1/2} <$ 7 min) fission products for the photofission of ^{238}U with 20-MeV bremsstrahlung by direct γ spectrometry of irradiated uranium samples. In addition, the independent yields of longer-lived fission products were determined by γ spectrometry of fission product catcher foils and chemical separation procedures. The charge distribution for the mass chains 131, 132, 133, 135, and 136, for which the yield of at least three members of the chain could be determined, was investigated. Furthermore, we deduced from our measurements the most probable charges Z_p(20 MeV) for 7 other mass chains in the heavy wing and for 8 mass chains in the light mass wing of the mass distribution, adopting for the width of the charge distribution the average of the values deduced from the charge distribution of the masses 131–136. The experimentally determined Z_p(20 MeV) values are compared with the most probable charge of the fragments, according to the unchanged charge density (UCD) hypothesis and to the Z_p values expected from the empirical relation of Nethaway.[9] The influence of the closed 50-proton shell is discussed.

II. EXPERIMENTAL PROCEDURE

The experimental setup and procedure used for the catcher foil experiments are the same as described in a previous paper.[10] For the determination of the independent yields of ^{96}Nb, $^{132}I^g$, $^{132}I^m$, ^{133}I, $^{134}I^g$, $^{134}I^m$, ^{134}Cs, and ^{136}Cs, 1 g $UO_2(NO_3)_2 \cdot 6H_2O$ (natural uranium) was irradiated with 20-MeV thin-target bremsstrahlung for appropriate times. The niobium, iodine, and cesium fractions were separated from the irradiated sample using the methods of Morris *et al.*,[15] Wahl,[16] and Cuninghame *et al.*,[17] respectively. Successive γ spectra of the separated samples were taken with the same detector-amplifier system used for the catcher foil experiments. To increase the reliability of the results, several experiments with different irradiation times, various cooling times before the chemical separation, and appropriate measuring cycles were performed.

The fractional independent or cumulative chain yields of short-lived (5 s$< T_{1/2} <$ 7 min) fission products were, as already mentioned in the Introduction, determined by direct γ spectrometry of irradiated uranium samples. These samples, consisting of 0.1 mm-thick natural uranium disks, prepared at the Central Bureau for Nuclear Measurements (Euratom, Geel), were enclosed in very pure (99.99%) nickel capsules and irradiated with 20-MeV bremsstrahlung for 15 or 30 s. After the irradiations, the capsules were transported from the irradiation site to the Ge(Li) detector with a pneumatic transport system.

After a cooling time of 5 or 10 s, according to the irradiation time, 60 successive γ spectra of 4096 channels each were taken with a detector-amplifier system consisting of a 19 cm^3 Ortec Ge(Li) detector followed by an Ortec 120-4 preamplifier, a Tennelec TC 205A linear amplifier, a Northern Scientific NS624 analog-to-digital converter, and a PDP 11/10 system with a CA11C Camac interface. A pulser was added for deadtime correction. The measuring time of one single spectrum was 0.5 or 3 s in the experiments with irradiation times of 15 or 30 s, respectively. The time interval between two spectra was about 160 μs. These measurements were possible only by the registration of the 60 spectra as one single file on an RK05 disk of the PDP 11/10 system. This procedure enabled us to add a number of successive spectra chosen in accordance to the half-life of the fission products and to perform an accurate deadtime correction, which is necessary in view of the large variation of the deadtime (35–10%) during the measurements. The resolution of the system in the measuring conditions was 2.3 keV at 1333 keV.

The identification of the γ rays was mainly based on the γ-ray catalogs of Blachot and Fiche[18] and Reus *et al.*[19] A list of the spectroscopic data used for the short-lived fission products is given in Table I. In this table are summarized the

TABLE I. Nuclear data for the fission products.

Isotope	E_γ (keV)	I_γ (%)	$T_{1/2}$	$T_{1/2p}$	Ref.
^{88}Br	775.2	63	15.9 s	1.5 s	20
^{90}Kr	1118.7	53	32.3 s	1.96 s	21
^{91}Kr	506.6	19	8.57 s	0.54 s	22
^{93}Rb	432.4	18.7	5.89 s	1.2 s	18
^{94}Sr	1428.3	95.4	74.1 s	2.69 s	23
^{95}Sr	685.9	24.0	28.0 s	0.36 s	18
	826.9	3.04			
^{100}Nb	535.2	50.0	1.5 s	7.1 s	18
$^{131}Sn^g$	798.4	100	39 s	0.3 s	24
$^{131}Sn^m$	1226.2	90	50 s	0.3 s	24
^{132}Sn	247	41.7	40 s	0.12 s	25
$^{132}Sb^g$	696	100	4.2 min		25
	974	100			
$^{132}Sb^m$	635.6	9.9	2.8 min	40 s	26
	989.6	14.8			
^{133}Sb	1096	32	2.34 min	1.7 s	25
$^{133}Te^g$	312	70	12.45 min	2.34 min	25
$^{133}Te^m$	912	62	55.4 min	2.34 min	25
$^{134}I^m$	272	79	3.8 min		25
^{135}Te	603	25.4	18 s	1.7 s	25
^{136}Te	332	36	17.5 s	0.82 s	25
$^{136}I^g$	1313	67	83 s	17.5 s	25
	1321	25			
$^{136}I^m$	381	100	46 s		25
^{137}Xe	455	31	3.83 min	24.75 s	25
^{138}I	588.9	97.7	6.46 s	1.6 s	18
^{139}Xe	296.5	22.11	39.7 s	2.61 s	18
^{139}Cs	1283.3	7.4	9.4 min	39.7 s	27
^{140}Xe	805.5	21	13.6 s	0.59 s	19
^{140}Cs	602.3	46	63.7 s	13.6 s	28, 29
	908.4	7.3			
^{144}La	397.3	90.3	42.4 s	10.7 s	18

fission product nuclide, the energy (E_γ) and absolute intensity (I_γ) of the observed γ transition, the half-lives of the nuclide ($T_{1/2}$) and its precursor ($T_{1/2p}$), and the reference (Ref.) from which these data are taken. In Table I the decay data of some longer-lived fission products, which were not already tabulated in our previous work,[10,30-31] are also included. From the peak areas in the spectra, fractional independent and cumulative chain yields were deduced in the same way as described in Ref. 30. For the correction for the γ attenuation in the uranium target and the nickel capsules, we used the attenuation coefficients of Storm and Israel.[32]

An estimate 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. A decrease in the fission yield of more than a factor 200 was found, indicating that the contribution of neutron-induced fission was less than 0.5% in the experiments performed with 20-MeV bremsstrahlung.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Our results on the fractional independent and cumulative chain yields for the photofission of ^{238}U with 20-MeV bremsstrahlung are summarized in Table II. The charge distribution in our bremsstrahlung experiments has the following shape:

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

with E_e the end-point energy of the bremsstrahlung, k the photon energy, $\sigma_{\gamma,F}(k)$ the photofission cross section for ^{238}U (Ref. 33, 34), and $\phi(E_e,k)$ the Schiff thin-target bremsstrahlung spectrum (Ref. 35).

This expression is based on the assumption of a Gaussian shape without odd-even effects for the charge distribution with monoenergetic photons with energy k. The width parameter c of the charge distribution was assumed to be energy independent in the excitation energy range of our experiments. A support for this assumption was given by the work of McHugh and Michel,[6] mentioned earlier. The dependence of the maximum of the charge distribution $Z_p(k)$ on the photon energy k was taken from the work of Nethaway.[9] The existence of an odd-even effect in the charge distribution in our experiments was neglected on the basis of the strong decrease of

TABLE II. Fractional independent and cumulative chain yields and deduced Z_p (20 MeV) values for the photofission of ^{238}U.

M_{post}	Product	Fractional chain yield	Z_p(20 MeV)
88	^{88}Br	0.740 ± 0.080(c)	35.10 ± 0.16
90	^{90}Kr	0.838 ± 0.093(c)	$35.90^{+0.22}_{-0.36}$
91	^{91}Kr	0.665 ± 0.087(c)	36.17 ± 0.20
93	^{93}Rb	0.736 ± 0.129(c)	$37.11^{+0.24}_{-0.31}$
94	^{94}Sr	0.884 ± 0.099(c)	$37.76^{+0.29}_{-0.64}$
95	^{95}Sr	0.848 ± 0.071(c)	$37.87^{+0.19}_{-0.28}$
96	^{96}Nb	$(1.1\pm0.5)\times10^{-4}$	$38.17^{+0.16}_{-0.20}$
100	^{100}Zr	0.870 ± 0.108(c)	$39.81^{+0.27}_{-0.62}$
128	^{128}Sn	0.700 ± 0.090(c)	50.17 ± 0.20
131	^{131}Sn^g	0.096 ± 0.013	50.86 ± 0.09
	^{131}Sn^m	0.159 ± 0.024	
	^{131}Sb	0.573 ± 0.057	
	^{131}Te^g	$(8.5\pm4.5)\times10^{-2}$	
	^{131}Te^m	$(7.7\pm1.1)\times10^{-2}$	
132	^{132}Sn	0.181 ± 0.019(c)	$51.10^{+0.05}_{-0.04}$
	^{132}Sb^g	0.169 ± 0.037	
	^{132}Sb^m	0.339 ± 0.051	
	^{132}Te	0.299 ± 0.096	
	^{132}I^g	$(7.4\pm1.5)\times10^{-3}$	
	^{132}I^m	$(5.7\pm0.9)\times10^{-3}$	
133	^{133}Sb	0.450 ± 0.046(c)	$51.62^{+0.06}_{-0.09}$
	^{133}Te^g	0.147 ± 0.040	
	^{133}Te^m	0.348 ± 0.034	
	^{133}I	$(9.5\pm2.6)\times10^{-2}$	
134	^{134}I^g	0.118 ± 0.018	52.07 ± 0.08
	^{134}I^m	0.109 ± 0.010	
	^{134}Cs	$(4.0\pm1.5)\times10^{-5}$	
135	^{135}Te	0.567 ± 0.062(c)	$52.40^{+0.10}_{-0.12}$
	^{135}I	0.393 ± 0.078	
	^{135}Xe	0.051 ± 0.012	
136	^{136}Te	0.230 ± 0.028(c)	$52.87^{+0.02}_{-0.04}$
	^{136}I^g	0.109 ± 0.017	
	^{136}I^m	0.304 ± 0.039	
	^{136}Cs	$(1.86\pm0.16)\times10^{-3}$	
137	^{137}I	0.679 ± 0.080(c)	$53.15^{+0.10}_{-0.12}$
	^{137}Xe	0.266 ± 0.056	
138	^{138}I	0.300 ± 0.036(c)	$53.87^{+0.10}_{-0.08}$
139	^{139}Xe	0.812 ± 0.084(c)	$53.95^{+0.22}_{-0.26}$

TABLE II. *(Continued.)*

$M_{\rm post}$	Product	Fractional chain yield	Z_p(20 MeV)
140	^{140}Xe	0.594 ± 0.061(c)	54.40 ± 0.07
	^{140}Cs	0.416 ± 0.053	
144	^{144}Ba	0.788 ± 0.084(c)	$56.07^{+0.11}_{-0.18}$
	^{144}La	0.237 ± 0.078	

(c) = Fractional cumulative chain yield.

this effect found in neutron-induced fission with increasing excitation energy. Following Nifenecker *et al.*[1] the odd-even effect is already reduced by a factor of 3 to 5 by an increase of the excitation energy by 2 to 3 MeV above the barrier.

By a least-squares fit of the data of Table II to expression (1), the width parameter c and the maximum of the charge distribution for the photofission of ^{238}U with 20-MeV bremsstrahlung, Z_p(20 MeV) could be calculated for the mass chains 131 to 135, for which at least two fractional independent or cumulative chain yields were determined. The c values are given in Table III. Figure 1 shows the charge distribution curves, calculated with expression (1), fitting the fractional yield data of the masses 131–133, 135, and 136. For these mass chains, at least three fractional yields were determined. From this figure it is clear that, with the exception of mass 136, the expression (1) describes very well the charge distribution for the photofission of ^{238}U with 20-MeV bremsstrahlung without any need to consider proton pairing effects. For mass chain 136 it was impossible to draw within the error bars a curve

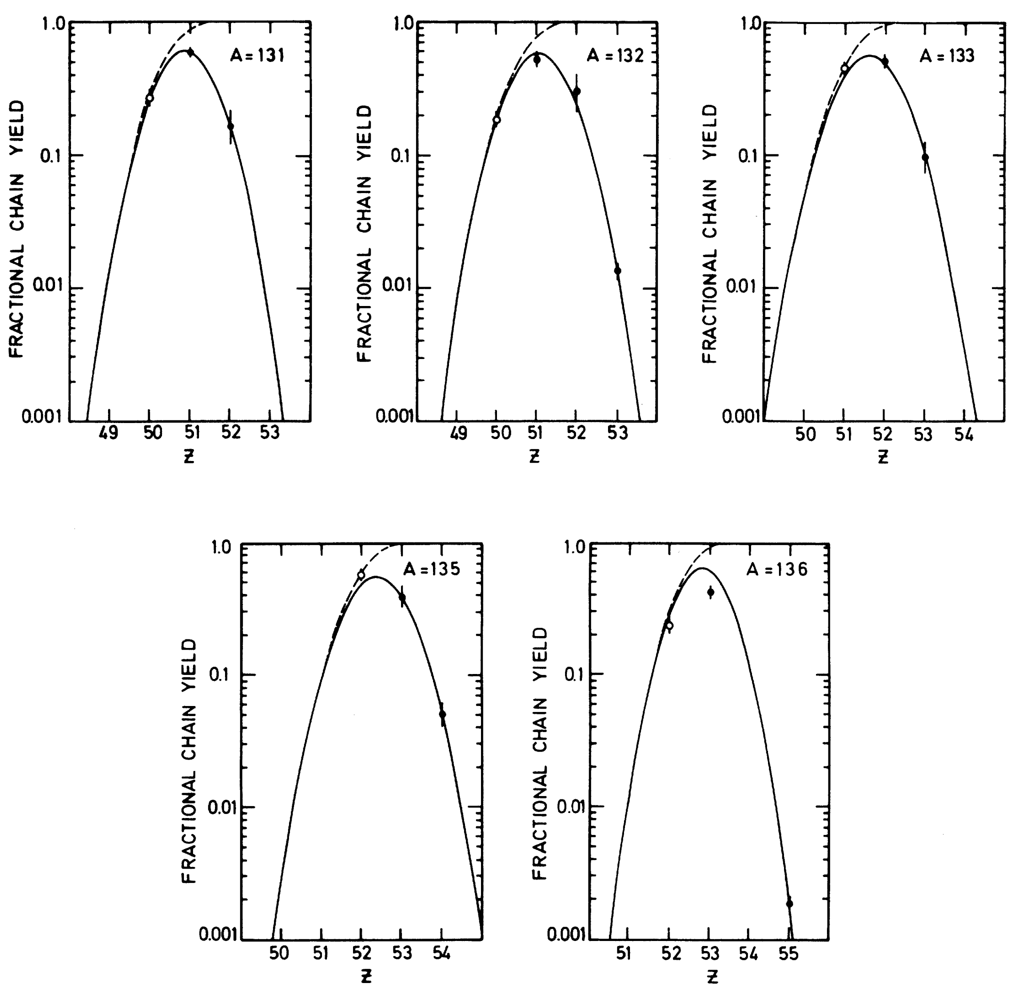

FIG. 1. Charge distribution for the mass chains 131, 132, 133, 135, and 136 for the photofission of ^{238}U with 20-MeV bremsstrahlung. ○ is the experimentally determined fractional cumulative chain yield (FCY) and ● is the experimentally determined fractional independent chain yield (FIY). A least-squares fit to the data based on expression (2) is represented by the full line (FIY) or by the dotted line (FCY).

of the shape (1) through the measured independent yields, due to an abnormally low independent yield of ^{136}I. Denschlag *et al.*[25] found in the thermal neutron induced fission of ^{235}U an unusually high odd-even effect (varying from 40 to 60% depending on the kinetic energy of the fragments) for mass 136, which can also be attributed to the low yield of ^{136}I. Furthermore, also in 3-MeV neutron-induced fission of ^{235}U, a Gaussian cannot fit the yield data for mass 136 (see Nifenecker *et al.*[1]), where for the other masses no important deviations are observable. Possible explanations for the observed abnormally low yield of ^{136}I are the influence of the closed 82-neutron shell configuration or the use of unreliable spectroscopic data for mass chain 136.

From the results given in Table III, an average value of $c = 0.93 \pm 0.06$ can be deduced for the photofission of ^{238}U with 20-MeV bremsstrahlung (corresponding average excitation energy of the compound nucleus 13.4 MeV—see Ref. 10). This value is somewhat higher, although within the experimental errors, than the value 0.80 ± 0.14 given by Wahl *et al.*[2] for $^{235}U(n_{th},f)$ and which was found to be generally valid for low-energy fission (Ref. 5). It is in excellent agreement with the value 0.95 ± 0.05, obtained by McHugh and Michel[6] for mass chain 135 in the medium energy fission of the compound nucleus ^{236}U and confirms that the width of the charge distribution is almost independent of the excitation energy of the fissioning nucleus.

The observed quasi-independence of c of the excitation energy can be qualitatively understood in the framework of the scission-point model of Wilkins *et al.*[36] in which a statistical equilibrium among the collective degrees of freedom at the scission point is assumed. As the collective temperature is proportional to the width parameter of the charge distribution, the observed constancy of c with excitation energy indicates that most of the additional energy above the barrier should contribute to intrinsic excitations.

Another possible qualitative explanation for the constancy of c is given by the thermodynamical equilibrium model if the quantum mechanical zero-point oscillation dominates over the temperature effects.[37]

Clerc *et al.*[3] also observed a constancy of c with changing fragment excitation energy for $^{235}U(n_{th},f)$. Although the observed independence of c on the fragment excitation energies in $^{235}U(n_{th}f)$ as well as on the compound nucleus excitation energy can be understood in the framework of the two mentioned models, Clerc *et al.*[3] rejected both theoretical interpretations, owing to the fact that they fail to reproduce quantitatively the measured c value.

TABLE III. Values of the width parameter c determined for the photofission of ^{238}U with 20-MeV bremsstrahlung.

M_{post}	c
131	$0.90^{+0.25}_{-0.18}$
132	$0.95^{+0.08}_{-0.07}$
133	$1.05^{+0.22}_{-0.23}$
134	0.86 ± 0.09
135	$1.06^{+0.34}_{-0.26}$

As already mentioned, for the mass chains for which the width parameter c could be deduced, the maximum of the charge distribution Z_p(20 MeV) was determined simultaneously. For the isobaric chains, for which the fractional independent or cumulative yield of only one member was determined, Z_p(20 MeV) values were calculated using expression (1) and adopting for c the average value 0.93 ± 0.06. A summary of the Z_p(20 MeV) results obtained is also given in Table II.

As is usually done in charge distribution studies, the deviations of the Z_p(20 MeV) values from the charge of the fragments calculated assuming unchanged charge density of compound nucleus and fragments (Z_{UCD}) are plotted in Fig. 2 versus fragment mass number for the light (crosses) and heavy (circles) fragments. For the necessary conversion of the postneutron masses into preneutron masses, we used the neutron emission curve for the photofission of ^{238}U with 25-MeV bremsstrahlung,[38] multiplied with an appropriate factor to reproduce the measured $\langle\nu\rangle$ value for 20-MeV bremsstrahlung induced fission.[10] The lines corresponding to the closed 50-proton and 82-neutron shells are also indicated in Fig. 2. From this figure it is apparent that the Z_p function has a smooth behavior with the exception of the Z_p(20 MeV) value deduced from the fractional independent yield of ^{138}I. An explanation for this deviation cannot be given at the moment. Also in the thermal neutron induced fission of ^{235}U (Ref. 4) the fractional independent yield of ^{138}I has an abnormally low value.

The behavior of the Z_p function in the vicinity of the $Z = 50$ line shows the strong influence of the closed 50-proton shell on the charge distribution for the photofission of ^{238}U with 20-MeV bremsstrahlung, as was also observed in the thermal neutron-induced fission of ^{235}U (Ref. 39). This indicates that at an average excitation energy of 13.4 MeV shell effects are still important in the determination of the fission characteristics.

Figure 2 shows that, excluding the mass region

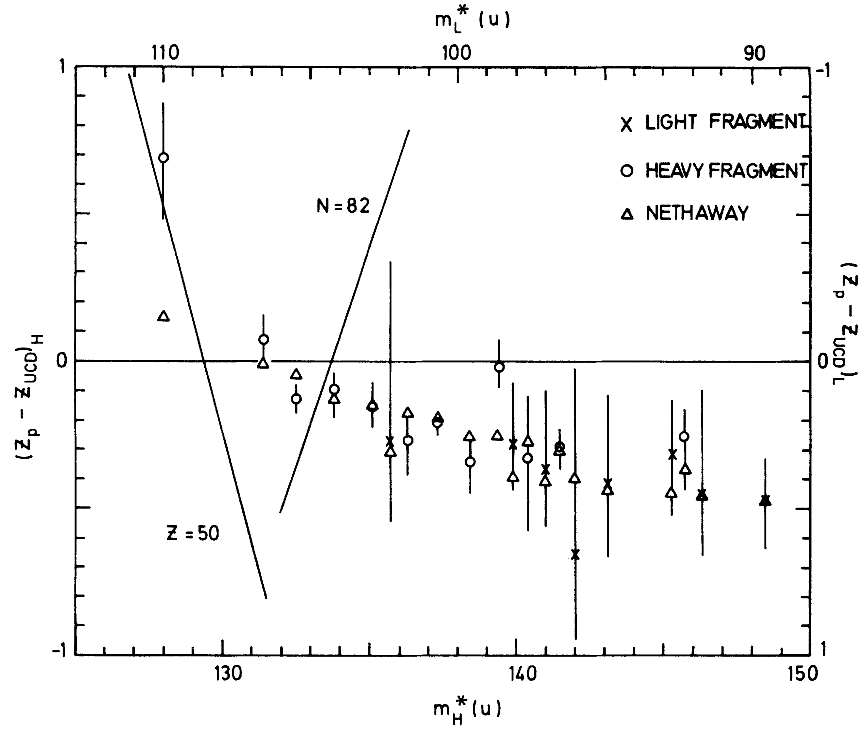

FIG. 2. Z_p-Z_{UCD} versus fragment mass number for the photofission of ^{238}U with 20-MeV bremsstrahlung. The Z_p values calculated following Nethaway (Ref. 9) are indicated by the triangles.

around $Z = 50$, there is a charge polarization resulting in a higher charge-to-mass ratio for the light fragment. This was also observed in the thermal neutron-induced fission of ^{235}U. The deviation of Z_p from Z_{UCD} for $^{235}U(n_{th},f)$ fluctuates between the values 0.40 and 0.60 (Ref. 40). As can be seen in Fig. 2, $(Z_p - Z_{UCD})$ is not so large in our photofission experiments. By an appropriate choice of the distance between the charge centers of the two fragments at the scission point, the scission-point model of Wilkins *et al.*[36] provides $(Z_p$-$Z_{UCD})$ values in reasonable agreement with experimental information obtained for $^{235}U(n_{th},f)$. The strong influence of the closed 50-proton shell on the charge distribution, however, observed in $^{235}U(n_{th},f)$ and in our photofission experiments on ^{238}U, is not predicted by the model.

The triangles in Fig. 2 represent the Z_p values, calculated following the empirical relation of Nethaway[9] for the fission of the compound system ^{238}U at an excitation energy of 13.4 MeV. The empirical relation of Nethaway gives for a mass chain the difference between the Z_p values for the particular fissioning system and the reference system $^{235}U(n_{th},f)$. For the reference Z_p values we used the values of Wahl *et al.*[2]

As can be seen in Fig. 2, there is in general a very good agreement between the experimentally determined Z_p values and the Z_p behavior calculated following Nethaway,[9] as well for the light as for the heavy fragments. A discrepancy of about 0.5 charge units exists for the mass chain 128, due to the influence of the closed 50-proton shell on the charge distribution. As pointed out in Ref. 41, this influence is not taken into account in the method of Nethaway,[9] so that in the mass region where the Z_p function approaches the closed 50-proton shell, this approximation cannot yield reasonable estimates. The close agreement, except for the $Z = 50$ mass region, between the calculated and experimentally determined Z_p values proves the usefulness of Nethaway's expression for the prediction of the most probable charges in photofission studies.

IV. CONCLUSIONS

A comparison of the results of our study of the charge distribution for the photofission of ^{238}U with 20-MeV bremsstrahlung (corresponding to an average excitation energy of the compound nucleus ^{238}U of 13.4 MeV) with the results for thermal neutron-induced fission shows that the width of the charge distribution is almost independent of the compound nucleus excitation energy as previously reported by McHugh and Michel.[6] Although this independence can be qualitatively understood in the framework of the scission-point model of Wilkins *et al.*,[36] or in terms of a quantum mechanical zero-point oscillation,[37] serious remarks can be made against these models in view of the recent work of Clerc *et al.*[3]

From our photofission studies we can also conclude that as observed in ^{235}U(n_{th},f), there is a charge polarization in the fissioning nucleus resulting in a higher charge-to-mass ratio of the light fragment. In addition, the behavior of the Z_p function shows the influence of the closed 50-proton shell on the charge distribution, as also observed in ^{235}U(n_{th},f). Finally, a good agreement between the experimentally determined Z_p values and those calculated following Nethaway[9] is obtained in our photofission studies, except for the mass region where the Z_p function reaches the $Z = 50$ line.

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