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Fragment mass and kinetic energy distributions for the photofission of ^{235}U with 12-, 15-, 20-, 30-, and 70-MeV bremsstrahlung

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Energy correlation measurements were performed for the photofission of ^{235}U with 12-, 15-, 20-, 30-, and 70-MeV bremsstrahlung. Overall fragment mass and kinetic energy distributions are deduced. The behavior of the total fragment kinetic energy as a function of the fragment mass and excitation energy of the compound nucleus is studied. The results are interpreted in terms of the scission-point model of Wilkins *et al.*

[NUCLEAR REACTIONS, FISSION ^{235}U(γ,f), $E_{\gamma\max} = 12$, 15, 20, 30, and 70 MeV; measured fragment energies E_1, E_2; deduced $N(\mu,E_K)/\langle E_{\text{exc}}(E_e)\rangle$.]

I. INTRODUCTION

An investigation of the kinetic energy of the fragments as a function of the fragment mass and the excitation energy of the fissioning nucleus provides interesting information on the decrease of the importance of fragment shells in the fission process at higher excitation energies.[1] Up to now such a systematic study of the fragment kinetic energy was not performed for the photofission of ^{235}U. Energy correlation measurements for 25-MeV bremsstrahlung-induced fission of ^{235}U were already reported by Patrzhak and Tutin[2] and by our group.[3] Recently, Günther *et al.*[4] measured fragment mass and energy distributions for the photofission of ^{235}U with bremsstrahlung varying the end-point energies from 15 – 55 MeV. They investigated in detail the competition between the symmetric and asymmetric fission modes and observed no significant changes in the fragment kinetic energies at different bremsstrahlung end-point energies. Energy correlation measurements for the electrofission of ^{235}U were performed by Shotter *et al.*[5] and McGeorge *et al.*[6] but they did not study the behavior of the fragment kinetic energy distribution with increasing bombarding energy.

We performed energy correlation measurements for the photofission of ^{235}U with 12-, 15-, 20-, 30-, and 70-MeV bremsstrahlung, using the same experimental setup as described in our paper on the photofission of ^{238}U.[7] The target consisted of a 65 μg/cm^2 UF$_4$ layer (enriched up to 97% ^{235}U) on a 58 μg/cm^2 polyvinyl-acetate chloride-copolymer (VYNS) backing coated with 10 μg/cm^2 gold. The diameter of the active layer was 30 mm. The target was prepared by the evaporation technique in the Central Bureau for Nuclear Measurements, Euratom, Geel. The uncertainty on the different thicknesses was 10%. As the changes in the kinetic energies between different end-point energies were expected to be quite small, extreme care was taken to have the same experimental conditions throughout the measurements. Each run at a given bremsstrahlung end-point energy was preceded and followed by a calibration run with 20-MeV bremsstrahlung, while the 20-MeV measurements were calibrated with the spontaneous fission of ^{252}Cf following the Schmitt calibration procedure.[8,9] The data were sorted off line in two dimensional $N(\mu,E_K)$ arrays of 120 × 120 channels as described in our previous work.[7] Here, μ is the provisional mass of a fragment and E_K the total kinetic energy of the two fragments.

The contribution of thermal and slow neutron induced fission in our photofission experiments on ^{235}U, obtained by measuring the fission yield between the linac pulses, was found to be less than 1%. The contribution of fast neutron induced fission was measured by inserting a 13 cm thick lead filter in the photon beam. Here, an upper limit of 1% was also obtained.

By integrating the two dimensional arrays

$N(\mu,E_K)$ over the variables μ or E_K, the overall kinetic energy and provisional mass distributions can be deduced. The characteristic of these distributions, together with the total number of analyzed events (NEV) and the average excitation energy of the ^{235}U compound nucleus $\langle E_{\mathrm{exc}}(E_e)\rangle$, are given in Table I. The values of $\langle E_{\mathrm{exc}}(E_e)\rangle$ were calculated as outlined in our ^{238}U work[7] using the Schiff form for thin target bremsstrahlung.[10] For the cross section of $^{235}\mathrm{U}(\gamma,f)$ in the photon energy range of 5 – 18 MeV we adopted the measured values of Caldwell *et al.*[11] As no experimental information is available at higher photon energies, an estimation was obtained by assuming a similar shape for the photofission cross section of ^{235}U above 18 MeV as deduced for the photofission of ^{238}U.[7] No $\langle E_{\mathrm{exc}}(E_e)\rangle$ value for 70 MeV bremsstrahlung was calculated because of the large uncertainty on the extrapolated cross section at higher energies. Parameters of the overall provisional mass distributions, listed in the table, are the average mass of the light and heavy fragments $\langle \mu_L\rangle$ and $\langle \mu_H\rangle$, the corresponding standard deviations $\sigma(\mu_L)$ and $\sigma(\mu_H)$, and the peak-to-valley ratio P/V. The values of $\sigma(\mu_L)$ and $\sigma(\mu_H)$ are equal as they are obtained after symmetrization around mass $A/2$. In addition, the midpoints between the $\frac{3}{4}$ maximum points in the light and heavy fragment provisional mass peaks $\langle \mu_L\rangle_{3/4}$ and $\langle \mu_H\rangle_{3/4}$, analogous to P_L and P_H in Ref. 9, and the full width at the $\frac{3}{4}$ maximum points for the fragment peaks FW($\frac{3}{4}$) are also tabulated. The average total kinetic energy of the two fragments and the corresponding standard deviations are indicated in Table I by $\langle E_K\rangle$ and $\sigma(E_K)$. Except for the (P/V) values, the errors, given in the table, are relative uncertainties. They are the root-mean-square deviation for at least five experimental runs. The errors based on the statistical accuracy of the data are of the same order but slightly smaller. The uncertainties on $\langle \mu_L\rangle_{3/4}$, $\langle \mu_H\rangle_{3/4}$ and FW($\frac{3}{4}$) are of the same order as those on $\langle \mu_L\rangle$, $\langle \mu_H\rangle$, and $\sigma(\mu_L)=\sigma(\mu_H)$, respectively. As pointed out in our previous paper[7] the absolute uncertainty on the $\langle E_K\rangle$ values is of the order of 2 MeV and the corresponding accuracy of $\langle \mu_L\rangle$ and $\langle \mu_H\rangle$ is about 0.5 u.

Two important features of the provisional mass distribution, directly apparent from Table I, are the strong decrease of the peak-to-valley ratio with increasing excitation energy and the constancy of the average mass of the light and heavy fragment peaks. This behavior was also observed by Günther *et al.*[4] The P/V values of the provisional mass distribution, deduced in the present study, are in very good agreement with values of the post neutron mass distribution,[12] obtained using γ-spectrometric methods with perfect mass resolution. The P/V values, reported by Günther *et al.*,[4] are slightly higher than those determined in our studies. However, the bremsstrahlung in the Giessen experiments is produced in a 1 mm thick tantalum target, where we are using a 0.1 mm gold foil. As the resulting bremsstrahlung spectra are seriously different, a direct comparison of the P/V values is not meaningful. The constancy of $\langle \mu_L\rangle_{3/4}$ and $\langle \mu_H\rangle_{3/4}$ shows that the small changes in $\langle \mu_L\rangle$ and $\langle \mu_H\rangle$ can be attributed completely to the increase of the symmetric fission mode at higher excitation energies. Our results show also a slight increase of the width of the mass distribution peaks with increasing end-

TABLE I. Parameters of the overall kinetic energy and provisional mass distributions for the photofission of ^{235}U.

E_e(MeV)	12	15	20	30	70
$\langle E_{\mathrm{exc}}(E_e)\rangle$ (MeV)	9.7	11.6	13.1	14.1	-
NEV	16.10^3	99.10^3	241.10^3	113.10^3	145.10^3
$\langle \mu_L\rangle$ (u)	96.89 ± 0.35	96.96 ± 0.16	97.23 ± 0.16	97.41 ± 0.24	97.75 ± 0.15
$\langle \mu_L\rangle_{3/4}$ (u)	97.00	96.87	96.96	96.87	96.99
$\langle \mu_H\rangle$ (u)	138.11 ± 0.16	138.04 ± 0.11	137.77 ± 0.20	137.59 ± 0.31	137.25 ± 0.22
$\langle \mu_H\rangle_{3/4}$ (u)	138.00	138.13	138.04	138.13	138.01
$\sigma(\mu_L)=\sigma(\mu_H)$ (u)	6.15 ± 0.31	6.47 ± 0.26	6.79 ± 0.20	7.05 ± 0.20	7.33 ± 0.22
FW($\frac{3}{4}$) (u)	10.39	10.69	10.80	10.93	11.11
P/V	41 ± 4	24 ± 1	14.2 ± 0.3	10.7 ± 0.4	7.5 ± 0.3
$\langle E_k\rangle$ (MeV)	171.03 ± 0.65	170.50 ± 0.30	169.97	169.64 ± 0.13	169.36 ± 0.30
$\sigma(E_k)$ (MeV)	10.43 ± 0.10	10.63 ± 0.10	10.81 ± 0.10	10.94 ± 0.11	11.35 ± 0.33

point energy of the bremsstrahlung.

The average total kinetic energy of the fragments $\langle E_K \rangle$ decreases systematically from 171.03 – 169.64 MeV for an increase of the average excitation energy $\langle E_{\mathrm{exc}}(E_e) \rangle$ from 9.7 – 14.1 MeV. Günther *et al.* observed no significant dependence of $\langle E_K \rangle$ on the bremsstrahlung end-point energy. This can easily be understood, taking into account the error on $\langle E_K \rangle$, 3 MeV, quoted in this paper, and the small changes in $\langle E_K \rangle$ observed in our work. By fitting a straight line to the $\langle E_K \rangle$ values as a function of the average excitation energy of the compound nucleus $^{235}\mathrm{U}\langle E_{\mathrm{exc}}(E_e) \rangle$ using a least squares procedure, one obtains for the slope

$$\frac{d\langle E_K \rangle}{d\langle E_{\mathrm{exc}}(E_e) \rangle} = -0.32 \pm 0.14 \; .$$

This value is within the given uncertainty the same as the value of the slope deduced from the photofission data of ^{238}U: -0.25 ± 0.04.[7]

In Fig. 1 we plotted the slope of the excitation energy dependence of the total kinetic energy of the fragments as a function of the fragment mass

$$\frac{d\langle E_K(\mu) \rangle}{d\langle E_{\mathrm{exc}}(E_e) \rangle}$$

for the photofission of ^{235}U. The indicated uncertainties are based on the statistical accuracy of the $\langle E_k(\mu) \rangle$ values. Figure 1 shows clearly that for strongly asymmetric mass splits ($\mu_H > 140$) the total kinetic energy of the fragments is almost independent on the excitation energy of the compound nucleus. In the symmetric fission region the slope $d\langle E_K(\mu) \rangle / d\langle E_{\mathrm{exc}}(E_e) \rangle$ is positive or close to zero. A definite conclusion here is difficult because of the large experimental uncertainties, due to the poor statistics. By grouping 10 masses around the symmetry point a value 0.36 ± 0.50 for the slope is obtained. In the transition region between symmetric and asymmetric fission $d\langle E_K(\mu) \rangle / d\langle E_{\mathrm{exc}}(E_e) \rangle$ is strongly negative with a minimum of about -0.8 for the mass splits with the heavy mass around 130. A similar behavior of the excitation energy dependence of the fragment kinetic energy as a function of the fragment mass, including the tendency of a positive slope for symmetric mass splits, was observed in our photofission studies of ^{238}U.[7]

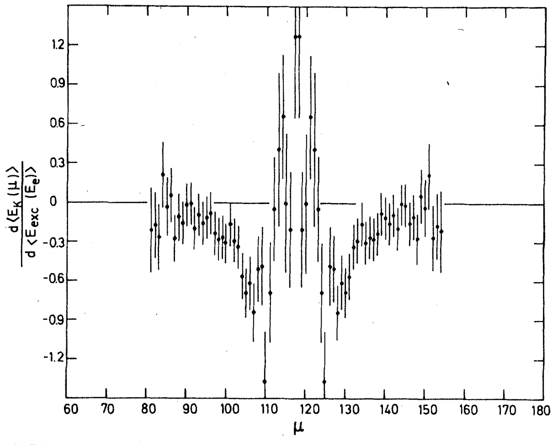

FIG. 1. Compound nucleus excitation energy dependence of the total fragment kinetic energy $d\langle E_K(\mu) \rangle / d\langle E_{\mathrm{exc}}(E_e) \rangle$ as a function of the provisional mass μ for the photofission of ^{235}U.

The variations of E_K with fragment mass and compound nucleus excitation energy can be intepreted in the framework of the scission point model based on deformed shell effects of Wilkins *et al.*[1] They showed that a large amount of the observed trends in the fission data can be understood in terms of the shell closures in the nascent fragments. In this model statistical equilibrium among the collective degrees of freedom at the scission point is assumed, resulting in a collective temperature T_{coll}. An effective intrinsic temperature τ_{int} determines the population of the single particle levels. An increase of the excitation energy of the compound nucleus above the fission barrier increases the value of τ_{int} and consequently changes the shell correction terms in the calculation of the potential energy of the system. The variations of the kinetic energy of the fragments with the excitation energy reflect the changes in the deformation of the fragments at the scission point, due to the changes in the shell corrections. Especially large effects on E_K are expected for the mass splits, where two minima in the potential energy surface, associated with configurations with different total deformation and liquid-drop potential energy are present. In this way the strong dip of $d\langle E_K \rangle / d\langle E_{\mathrm{exc}}(E_e) \rangle$ for the mass splits, with heavy mass around 130, observed in our experiments, can be explained in terms of a competition between a configuration with low total deformation, associated with the closed 50-proton and 82-neutron shells and favored at low excitation energy, and a secondary configuration with total deformation close to the liquid-drop value. At higher values of τ_{int} the shell corrections diminish and the secondary liquid-drop configuration, with larger deformation, becomes more important, causing the drop in the kinetic energy.

According to this model the kinetic energy is expected to increase with the compound nucleus excitation energy in the symmetric fission region around mass 118. Our data, although not conclusive, indeed show this trend. For strongly asymmetric mass splits ($\mu_H > 140$), where the total deformation of the scission configuration is not strongly influenced by introducing shell corrections in the potential energy surface, the kinetic energy is almost independent on the excitation energy. This shows also that the fission mode is not, or very weakly, coupled to the quasiparticle excitations.

As the (γ,f) and (γ,nf) cross sections for ^{235}U are not measured, the contribution of second chance fission in our experiments cannot be calculated directly. Based on the results of Caldwell *et al.*[13] we found for the second chance fission contribution in our experiments[7] on ^{238}U with 12-, 15-, and 20-MeV bremsstrahlung 0%, 15%, and 25%, respectively. In view of the Γ_n/Γ_f values for ^{234}U, ^{235}U, ^{237}U, and ^{238}U, determined by Caldwell *et al.*,[14] the second chance fission contribution is expected to be less in our ^{235}U photofission studies than in our experiments on ^{238}U. As discussed in our previous paper,[7] the two effects of second chance fission, lowering the excitation energy and mass of the fissioning nucleus, are opposite and can possibly effect our results quantitatively. However, second chance fission is not expected to change the qualitative conclusions of this study.

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