ANGULAR DISTRIBUTION OF U^{238} PHOTOFISSION FRAGMENTS NEAR THE FISSION THRESHOLD

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Submitted to JETP editor March 31, 1965

J. Exptl. Theoret. Phys. (U.S.S.R.) **49**, 476-484 (August, 1965)

The angular distributions of fragments emitted in photofission of U^{238} near the threshold have been measured by recording the fission events in glass. The photons were produced by electrons accelerated in the 12-MeV high-current microtron of the Institute for Physics Problems, U.S.S.R. Academy of Sciences. The work was carried out with the aim of detecting the component proportional to $\sin^2 2\theta$ in the angular distribution, which should be due to the 2^+, $K = 0$ channel in quadrupole photon absorption. "Quadrupole" absorption has been detected only in an excitation region which is much lower than the barrier for "dipole" fission. This is in good agreement with the usual assumption that the quadrupole absorption cross section is much smaller than that for dipole absorption. The experimental data confirm Bohr's hypothesis regarding the similarity of the fission-channel spectrum and lower-excited-level spectrum near the ground state of the equilibrium nucleus. The distance between the thresholds of the fission channels for 2^+ and 1^-, $K = 0$ as well as 1^-, $K = 0$ and 1^-, $K = 1$ is not less than 0.5 MeV.

INTRODUCTION

OUR understanding of the mechanism of nuclear fission at low excitations is closely connected with the concepts of fission channels, the foundations for which were laid by A. Bohr.[1] These concepts, initially developed in connection with fission of even-even nuclei, are based on the idea of a major "cooling off" of the nucleus at the saddle point and on the similarity between the fission-channel spectrum and the spectrum of nuclear levels close to the ground state. In regard to the characteristics of the spectrum of the lowest levels of even-even nuclei, the most concrete results of the model apply to just this class of fissioning nuclei. The main features of the fission-channel spectrum expected on the basis of the model of Aage Bohr reduce to the following: a) in the spectrum of internal excitations there is an energy gap of $\gtrsim 1$ MeV, b) in this gap are located a relatively small number of channels of a collective nature, c) the lowest-lying channels are based on a band of states of positive parity 0^+, 2^+, 4^+, ... and a band of negative parity 1^-, 3^-, 5^-, ... which is located above the first by a few hundred keV; these bands correspond to $K = 0$ (K is the projection of the total angular momentum on the fission "axis").

In spite of the fact that Aage Bohr's model is used successfully to interpret many aspects of the fission process, there are as yet no direct experimental proofs of the specific structure of the fission-channel spectrum predicted by it. This situation is due to the fact that the widely used means of nuclear excitation—neutrons and charged particles—lead to excitation of compound nuclei, as a rule, with a large number of angular momenta. This circumstance strongly hinders the manifestation of individual effects of specific fission channels. Information on fission by resonance neutrons, valuable in principle, still contains a considerable element of uncertainty due to the fact that the spins of the resonances are known only in rare cases. The most suitable means for study of the discrete structure of the fission channels of even-even nuclei is the use of photons with energy near the fission threshold. At these energies (5—7 MeV) the photons undergo only dipole and quadrupole absorption, which leads in the case of even-even target-nuclei to formation of compound nuclei only in two possible states: 1^- and 2^+. The contribution of the specific fission channels 1^-, $2^+(K = 0)$ and $1^-(K = 1)$ is easy to determine from the shape of the angular distribution of fragments, $\sigma_{1^-,0} \sim \sin^2\theta$; $\sigma_{2^+,0} \sim \sin^2 2\theta$; $\sigma_{1^-,1} \sim 1 - (1/2) \sin^2 \theta$.

Fission by dipole photons has been studied in detail in the work of the Katz group.[2,3] Analysis of the angular distributions of fragments in fission of even-even nuclei[3] has shown that the barrier for the $1^-(K = 1)$ channel is several hundred keV higher than the barrier of the $1^-(K = 0)$ channel. The data on excitation of the $2^+(K = 0)$ channel are contradictory. In most of the studies,[2-6] which have been carried out both in monoenergetic photons (6—7 MeV) and in bremsstrahlung spectra (E_{max} = 6—20 MeV), "quadrupole" fission has been found absent, with experimental errors of ~5—10%. An appreciable admixture of a quadrupole component $\sim \sin^2 2\theta$ has been observed only in two studies[7,8] of the angular distribution of fragments from photofission of U^{238}. However, the results of these measurements have not been confirmed in subsequent experiments by other authors.[3,6] This fact permitted some of us[6] to draw the conclusion that for photofission the contribution of the quadrupole component to the angular distribution of the fragments does not exceed a few per cent over the entire photon-energy region from 6 to 20 eV. This conclusion is in good agreement with the electrodynamic estimate of the cross-section ratio for dipole and quadrupole absorption in this energy region, $\sigma_{1^-}/\sigma_{2^+} \sim (\lambda\!\!\!^{-}/R)^2 \approx 20$. However, the relative contribution of the quadrupole fission cross section, insignificant in this excitation region, can become quite noticeable in fission at the threshold of the $1^-(K = 0)$ channel, if the distances between the lowest lying channels 2^+ and 1^- are sufficiently large.[9,10] The present paper is devoted to an experimental study of this possibility of observing quadrupole fission. A brief communication has been submitted previously.[11]

EXPERIMENTAL DETAILS

a) Source of γ radiation. The study of fragment angular distributions from photofission in the photon-energy region of interest here presents serious difficulties. The main hindrance to the realization of these experiments up to the present time has been the absence of sufficiently intense photon beams. Measurements in bremsstrahlung beams from betatrons and synchrotrons have not permitted lowering the energy below E_{max} = 6 MeV. Recently De Carvalho et al.[5] have suggested a new means for studying photofission by monoenergetic photons, based on use of γ rays from radiative capture of thermal neutrons in a reactor. However, the group of photon energies with which we can accomplish photofission near threshold is very limited. Furthermore, judging by the γ-ray intensity achieved in the Ti(n, γ) reaction, experiments in the energy region appreciably below the barrier can be carried out only with great difficulty.

The present work utilized as a photon source the bremsstrahlung from electrons accelerated in the high-average-current microtron of the Institute of Physics Problems of the U.S.S.R. Academy of Sciences.[12,13] The microtron has important advantages over betatrons and synchrotrons, which are the usual sources of bremsstrahlung in the study of photonuclear reactions. The main advantage of the microtron lies in the much greater γ-ray intensity, comparable with the intensity of a linear accelerator, but in a microtron achieved with considerably greater monochromaticity of the beam and with the possibility of continuous adjustment of the energy from 5 to 12 MeV for instantaneous currents of ~50 mA. The length of the current pulse is 2.5 μsec and the repetition frequency 400 cps. The transverse dimension of the electron beam is roughly 2 × 4 mm. The bremsstrahlung target is a cooled tungsten disc of thickness 1 mm. At an energy of 10 MeV the bremsstrahlung dose at a distance of 1 m from the target was ~3000 R/min.

The electron energy was varied both discretely (by transfer to different orbits) and continuously (by variation of the magnetic field strength H (the parameter Ω). In practice the value of Ω could be changed within the limits 1.1—1.3, for the resonator geometry used, without appreciable decrease of intensity. The instability of the magnet current determines the uncertainty in the absolute energy value, which is less than 100 keV. It should be noted that there is the possibility of considerable further reduction of this energy spread, which it is proposed to achieve in further experiments.

b) Experimental method and measurement procedure. In order to utilize most effectively the powerful photon beam produced in the microtron target, it was necessary to use fission-fragment detectors which have very low sensitivity to γ radiation and are capable of withstanding long exposures. Our equipment was based on the technique of counting fragments by means of glass.[14] Glass as a detector of fragments successfully combines the above requirements with the possibility of preparing a compact experimental device for measurement of fission-fragment angular distributions, which can be placed directly in the accelerator vacuum chamber in the immediate vicinity of the internal target.

Figure 1 shows a drawing of the experimental apparatus, which was mounted inside the microtron chamber. It consists of a cassette in the center of which is fastened a double layer of natural uranium of thickness 1 mg/cm^2, and around the edge of

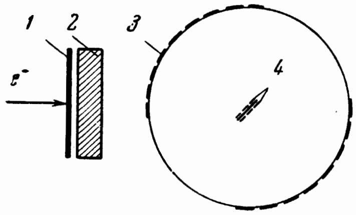

FIG. 1. Drawing of experimental equipment and geometry: 1 – 1-mm tungsten target, 2 – 10-mm aluminum absorber, 3 – glass, 4 – U^{238} layer.

which, every 15° between 0 (180) and 90° with respect to the photon beam, were placed two sets of photographic-glass plates. The dimensions of the cassette (diameter 8.0 cm), the glass plates (0.8 × 2 cm), and the uranium layer (diameter 1.0 cm) were chosen so that the equipment could be placed inside the microtron chamber and would not intercept the previous electron orbit. (The distance between orbits for a wavelength $\lambda \sim 10$ cm is about 35 mm.) The distance between the centers of the uranium layer and the target was 6.5 cm. The apparatus was firmly mounted with respect to the tungsten target so that the plane of the uranium layer was located at an angle of 45° to the photon-beam axis, and could be displaced from orbit to orbit. This arrangement of the uranium layer is the most suitable one from the point of view of the variation of fragment detection efficiency with emission angle.

Electrons scattered in the target and hitting the experimental apparatus can result in undesirable heating of the glass plates. On heating of the unetched glass plates to a temperature of ~200°C, part of the tracks disappear.[14] To avoid this effect we placed behind the target an aluminum absorber 1 cm thick, sufficient to completely slow down electrons with energies up to 10 MeV. Control experiments showed that within experimental error the absorber does not affect the shape of the fission-fragment angular distribution.

We made an evaluation of the background from photoneutrons and scattered γ rays. For this purpose at the maximum bremsstrahlung energy $E_{max} = 9.25$ MeV the apparatus was displaced beyond the limit of the photon beam. The background amounted to less than 0.1% of the effect observed in the direct beam.

The plates were scanned by two observers. The minimum spread in results (~0.5%) was for about 4000 tracks per plate. For a smaller number of tracks spurious counts and omissions contribute a larger relative error, and for less than 100 tracks per plate the error amounts to 2% on the average. Scanning of plates with more than 5000 tracks is more tiring, which also leads to increase in the counting error. The main part of the measurements were made with the optimum number of tracks per plate.

c) Analysis of the measurements. The angular dependence of the fission probability of even-even nuclei by dipole and quadrupole photons can be represented in very general form by the relation

$$W(\theta) = a + b\sin^2\theta + c\sin^2 2\theta. \tag{1}$$

The aim of the experiment was to determine for the various values of E_{max} the coefficients entering into this formula. It is obvious that the experimentally obtained distribution of number of counts per plate N_j cannot be simply identified with the angular distribution $W(\theta)$. The true function $W(\theta)$ is distorted as the result of the finite dimensions of the detector and of the fissioning sample, and also by the angular dependence of the fragment detection efficiency, which is connected with the nonuniform value of fragment energy loss in the layer for different emission angles. The effect of these factors can be excluded with sufficient accuracy in an appropriate analysis of the data. In the present work we subjected to mathematical analysis not the number of events N_j in the j-th plate but their ratio to the corresponding number of events N_j^T obtained on thermal-neutron irradiation of the natural uranium layers used. These measurements were carried out in the thermal column of a reactor with a cadmium ratio of the order of 10^6. The ratio N_j/N_j^T is related to the angular distribution of the fragments as follows:

$$\frac{N_j}{N_j^T} = C\int_S\int_{S_j}\frac{dS\,dS_j}{r^2 r_j^2}W(\theta)\cos(\mathbf{r},\ \mathbf{n})\cos(\mathbf{r}_j,\ \mathbf{n}_j)\eta(\psi)$$
$$\times\left[\int_{S_j}\frac{dS_j}{r_j^2}\cos(\mathbf{r}_j,\ \mathbf{n}_j)\eta(\psi)\right]^{-1}, \tag{2}$$

where (see Fig. 2) S and dS are respectively the area of the fissioning layer and the element of this area; S_j and dS_j are the area and element of area of the detector; $\mathbf{r}$ is the vector from the point of emission of the photon to the point of the layer in

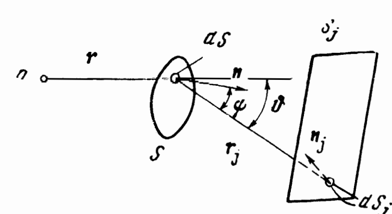

FIG. 2. Diagram illustrating the significance of the quantities in formula (2).

which the fission occurs; $\mathbf{r}_j$ is the vector from the point of fission to the point of detection of the fragments; θ is the angle between the directions of the photon and the fragment; $\mathbf{n}$ is the normal to the surface of the fissioning layer; $\mathbf{n}_j$ is the normal to the surface of the j-th detector; $\eta(\psi)$ is the detection efficiency as a function of the angle between the fragment direction and the normal to the surface of the layer; C is a constant depending on the source intensity and on the cross sections of natural uranium for fission by photons and by thermal neutrons. In the thermal region $W(\theta)$ = const, and $r \to \infty$.

In expression (2) it is assumed that the intensity of the γ rays per unit solid angle is uniform over the cone (4.5°) defined by the layer. This assumption is evidently well fulfilled for thick targets.[15] In view of the fact that $\eta(\psi)$ is a slowly varying function, according to the theorem of the mean we can take it out from under the integral sign and take the average values $\bar{\eta}$ and $\bar{\eta}^T$ to be equal without appreciable error. The variation of $\eta^T \sim N_j^T/J_j^T$ from plate to plate can be judged from the data plotted in Fig. 3, where the experimental values of N_j^T are compared with

$$J_j^T = \int\limits_{S_j} \frac{dS_j}{r_j^2} \cos(\mathbf{r}_j, \mathbf{n}_j). \quad (3)$$

For a specific function $W(\theta)$ Eq. (2) has the form

$$N_j / N_j^T = a\alpha_{aj} + b\alpha_{bj} + c\alpha_{cj}. \quad (4)$$

The quantities α_{aj}, α_{bj}, and α_{cj} are fourfold integrals; they were computed by the Monte-Carlo method. The coefficients a, b, and c in Eq. (4) were found by the method of least squares. In order to have some control of the accuracy of the calculation, a determination of the coefficients was also made from the number of events in the central half of the plates.

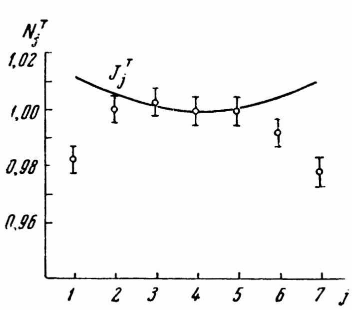

FIG. 3. Number of events in the j-th plate N_j^T in fission of the natural U^{235} impurity in the natural uranium layer used (thickness $\sim$ 1 mg/cm²) by thermal neutrons compared with the number of events in the j-th plate J_j^T for a detection efficiency $\eta^T(\psi)$ = const in an experiment with an isotropic angular distribution, determined by computation.

EXPERIMENTAL RESULTS AND DISCUSSION

Measurements of the fragment angular distributions $W(\theta)$ were made for seven values of E_{max}: 5.2, 5.4, 5.65, 5.9, 6.4, 6.9, and 9.25 MeV. In the very interesting and not previously studied region E_{max} < 6 MeV, in spite of the fact that photons from energy 0 to E_{max} are present in the bremsstrahlung spectrum, the spectrum of excitation energies of the fissioning nuclei, which is the product of a rapidly rising function (the fission cross section) and a rapidly falling function (the bremsstrahlung spectrum), has the shape of a rather narrow line with a width of ~ 0.4 MeV.

Figure 4 shows the fragment angular distributions obtained experimentally. The solid lines show the results of analysis of the data by the method of least squares for the normalization condition W(90°) = a + b = 1. The broken lines show the isotropic and quadrupole components of $W(\theta)$ separately. The experimental points are given for those values of the effective angle θ_{eff} for which the fragment emission probability per unit angle for a point source and a point detector $W(\theta_{eff})$ is equal to the probability per unit angle averaged over all directions of fragment emission allowed by the geometrical dimensions of the uranium layer and the glass plate.

Values of the coefficients a, b, and c are listed in the table. They are in good agreement with the corresponding values determined from the number of events in half of the plate (see above).

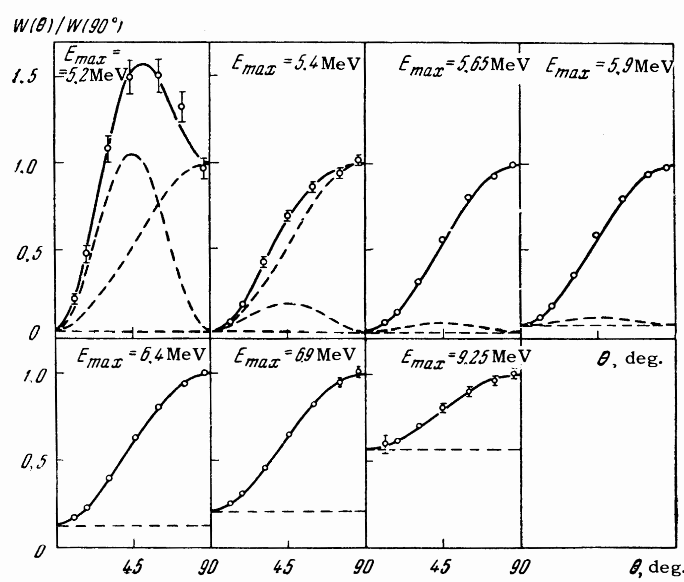

FIG. 4. Angular distributions of fragments in photofission of U^{238}.

Coefficients obtained as the result of analyzing the experimental data by the method of least squares, with representation of the angular distributions in the form $W(\theta) = a + b\sin^2\theta + c\sin^2 2\theta$

E_{max}, MeV	a	b	c
5.2	0.042±0.035	0.958±0.050	1.018±0.068
5.4	0.038±0.009	0.962±0.017	0.155±0.021
5.65	0.034±0.005	0.966±0.011	0.040±0.010
5.9	0.078±0.005	0.922±0.014	0.039±0.014
6.4	0.127±0.004	0.873±0.009	0.034±0.008
6.9	0.213±0.004	0.787±0.008	0.047±0.008
9.25	0.570±0.006	0.430±0.007	0.013±0.007

In Fig. 5 we compare the coefficient ratio $b/a = W(90°)/W(0°) - 1$ obtained in the present experiment with those of Baerg et al.[3]. Our data lie systematically higher. It is natural to explain this discrepancy by the difference in the bremsstrahlung spectra, arising from the difference in the thickness of the targets (the experiments of Baerg et al.[3] were performed with a thinner betatron target). A control experiment at E_{max} = 9.25 MeV with a 0.05 mm target, results of which are shown in Fig. 4 and are in good agreement with the work of Baerg et al.,[3] convinced us of the correctness of this explanation.

Let us clarify the physical significance of the coefficients entering into expression (1). If the photon energy is sufficient for excitation of only the very lowest fission channels 2^+ and 1^- with $K = 0$ and if we neglect the transmission of channels with $K \neq 0$, then the coefficient $a = 0$. In this case the ratio of the other two coefficients completely determines the relative probability of fission by dipole and quadrupole photons, $\sigma_{2^+}/\sigma_{1^-} = 4c/5b$. The isotropic part of the fragment angular distribution receives contributions from the higher lying channels with $K = 1$, which usually are connected with internal excitations of the nucleus. If we can neglect the contribution of quadrupole fissions, then the quantity $(b/a + \frac{1}{2})$ is equal to the ratio of cross sections for fission by dipole photons through the state 1^- with $K = 0$ and $K = 1$.

Figure 6 illustrates the dependence of the coefficient ratios a/b and c/b on E_{max}. The broken line shows the ratio a/b deduced from data obtained in experiments with monoenergetic photons.[5,8] From the sharp rise in the curves c/b (for bremsstrahlung) and a/b (for monoenergetic photons) we can conclude that the barrier height for the channel $2^+(K = 0)$ does not exceed 5.2 MeV, and for the channel $1^-(K = 1)$ is at least 6.5 MeV. Analysis of the energy dependence of the U^{238} fission cross section[2] shows that the barrier height for the channel $1^-(K = 0)$ is approximately 5.8 MeV.

Thus, the most important results of the present work: a) good agreement of the experimental data with the fragment angular distribution predicted by theory, b) high anisotropy of photofission ($b/a \approx 25$–30) for low E_{max}, and c) the appreciable distance between the thresholds of the fission channels 2^+ and $1^-(K = 0)$ on the one hand and $1^-(K = 1)$ on the other hand, are in agreement with the basic postulates of Aage Bohr's model for

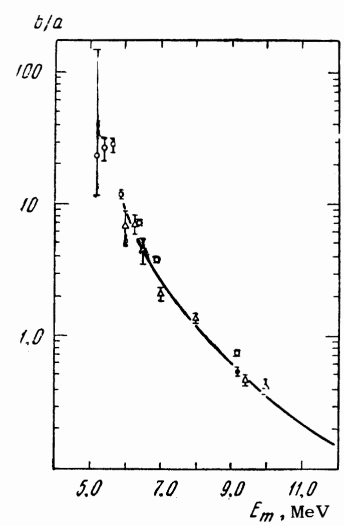

FIG. 5. Fission anisotropy $b/a = W(90°)/W(0°) - 1$ as a function of bremsstrahlung maximum energy E_{max}: Δ – results of Baerg et al.[3], ○ – results of the present work, obtained with a target 1 mm thick, ● – point obtained with a target 0.05 mm thick.

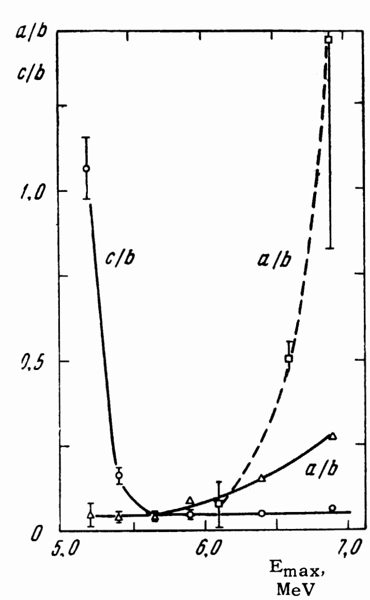

FIG. 6. Coefficient ratios a/b and c/b as a function of bremsstrahlung maximum energy.

fission channels of even-even nuclei. It follows from our discussion that the quantity K is a rather good quantum number, which is conserved in all stages of the fission process from the peak of the barrier to the breakup. It should also be noted that the appearance of an appreciable contribution by the quadrupole component only well above the threshold of the channel $1^-(K = 0)$ qualitatively agrees with the conclusion previously drawn[6] regarding the relative magnitude of quadrupole fission. A more detailed and quantitative analysis will be made subsequently on the basis of the data obtained and on data for photofission of Th^{232} and Pu^{240} which are being analyzed at the present time.

The authors are deeply grateful to L. N. Usachev and N. S. Rabotnov for their interest in the work and for helpful discussions, to P. L. Kapitza for supporting the research, and to M. K. Golubeva, L. D. Gordeeva, and N. E. Fedorova for participation in the work.

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Translated by C. S. Robinson
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