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ANGULAR DISTRIBUTION OF PHOTOFISSION FRAGMENTS FROM URANIUM *

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ABSTRACT

The angular distribution of photofission fragments from uranium at X-ray energies of 6.9; 8.1; 9.4; 15.5 and 20 MeV were measured using a 24 MeV betatron. The anisotropy was found to increase with decreasing the X-ray energy and to be mainly consistent with $a+b \sin^2\theta$ electric dipole photon absorption distribution. The quadrupole contribution is shown to be small. The ratios of the anisotropic dipole absorption to the isotropic fission yields at the first three energies above are 2.80±0.44, 1.18±0.14, and 0.62±0.12 and at 15.5 and 20 MeV the distribution is almost isotropic.

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INTRODUCTION

The anisotropy of the angular distribution of photofission fragments is a property of the heavy and strongly deformed even even nuclei. In 1952 Winhold, Demos and Halpern [1], found, by the catch foil technique, that the angular distribution of photofission of fragments from thorium has a maximum at right angles to the beam of X-rays. It was found also that the anisotropy decreased with the increase in maximum energy of the bremsstrahlung radiation and that within the experimental errors, the angular distribution appears to be of the form $a + b \sin^2 \theta$, resulting from a dipole photon absorption.

Katz, Baerg and Brown [2] in 1958, using ionization chambers, measured the angular distribution of photofission fragments from U-238 at nine different energies between 6 and 20 MeV and it was found to be consistent with a dipole photon absorption of the form $1 + \sin^2 \theta$, however Lazareva [3] et al have found by means of the photo plate technique, that the angular distribution of the fragments of photofission of U-238, contains a term in $\sin^2 \theta \cos^2 \theta$, implying some quadrupole absorption which is mainly observed at the maximum bremsstrahlung radiation energy of 9.4 MeV in disagreement with Katz's results at the same energy.

In the experiment described in the present paper the angular distribution of photofission fragments from uranium was obtained from careful measurements made at six different angles and at five different energies using the nuclear emulsion technique.

METHOD

The measurements were performed on the fission fragment tracks produced by the gamma rays in Ilford 100 and 200 microns KO nuclear emulsions loaded with natural uranium. From a systematic search the best method for uranium loading of nuclear emulsion was chosen to be the following: the solution for loading was prepared mixing equal volumes of a molar solution of sodium citrate with a 0.5 molar solution of uranyl nitrate and adjusting the PH to about 5 PH units with sodium hydroxide. The plates were immersed in this solution for 20 or 60 minutes according to the thickness of emulsion (100 microns or 200 microns) and put to dry at room temperature.

Natural uranium was used as U-238 target nuclide. The other isotopes in natural uranium at the X-ray energies used have a photofission contribution less than 1,8% of the total number of fissions produced. Furthermore the U-235 is an odd-even nuclide, therefore it is expected to give a nearby isotropic angular distribution.

The X-ray dose in roentgens received by the stacks of plates was measured using a Victoreen ion chamber placed in a cavity at the center of an 8 cm lucite cube. The energy scale was calibrated detecting the threshold of the following reactions: $Cu^{63}(\gamma,n)$ 10.6 MeV and $O^{16}(\gamma,n)$ 15.85 MeV, $C^{12}(\gamma,n)$ 18.75 MeV. The energy scale is believed to be accurate to ± 0.1 MeV.

The plates (one by three inches) were exposed longitudinally at 0° angle in the axial part of the beam of X-rays at different maximum energies at a distance of 45 centimeters from the betatron target. Only the central axial part of the plates was used in the scanning work. Due to absorption of X-rays in the plates the X-ray spectrum was somewhat hardned along the plates.

The thickness of the loaded emulsions were measured just before the exposures at four points, with a dial micrometer, around a small hole cut in the emulsion, so as to allow the surface of the glass backing to be taken as the zero of the micrometric measurements. The shrinkage factor was obtained measuring the thickness of the four points in the developed plates with the same dial micrometro adapted to the microscope.

The development method used was the following [4]. The loaded and exposed plates were washed with cold distilled water (5°C) for 45 minutes before immersion in the developing bath: boric acid - 3.5g, sodium sulfite - 4.5g, potassium bromide - 1.5, amidol - 0.45g, distilled water to complete 2 liters of solution.

The developing time was adjusted to the condition of the X-ray dose and uranium loading desensitization, in such a way that only the fission tracks were well developed. This made the scanning of plates considerably easier since no alpha particle tracks were visible.

After the developing bath the plates were washed in cold distilled water for 2 hours and fixed with a very cold 30% hypo solution that had been used before so that it contained a few grams of silver in solution. This diminishes the production of background grains during the fixing process.

In order to keep the shrinkage factor low the plates after fixing and washing in cold tap water, were immersed in a 10% glycerine solution and submitted to a slow drying, thus yielding a shrinkage factor of about 1.6, and so increasing the precision of the measurements of depth with the microscope.

The fraction of fission due to fast neutrons from the betatron, slow neutrons and scattered X-rays present in the background of the experimental area, was measured by the recoil proton method with E1 Ilford nuclear emulsion placed in and out of the X-ray beam and KO Ilford uranium impregnated plates placed out of the beam during the exposure time. The fraction was found to be less than 2% of the total number of fissions at the X-ray energies used.

Since the point or main of the tracks could not be determined the angles θ and $\pi - \theta$ were indistinguishable and the obtained angular distribution is actually $Y(\theta) = I(\theta) + I(\pi - \theta)$.

For the indicated angle intervals in table I the relative yields per unit of solid angle are given by

$$\overline{Y}(\theta) = \int_{\theta_1}^{\theta_2} \left[I(\theta) + I(-\theta) \right] d\Omega \Big/ \int_{\theta_1}^{\theta_2} d\Omega$$

The scanning was performed with Leitz Ortholux microscopes with 100x objective and 6x ocular. The dip angles of all tracks were obtained by measuring the horizontal projection and the vertical depth of the tracks. As mentioned above, the shrinkage factor was reduced by adding glycerine and it was measured four times a day to correct for the daily relative humidity variations.

The dip angle α of the fragment with the horizontal plane and the angle β of the horizontal projection of fragment and the direction of X-ray beam were measured and the actual angle θ of the fragment with the axial direction of the beam was obtained from the relation $\cos\theta = \cos\alpha \cos\beta$.

The fission yield per roentgen and per nucleus decreases sharply with the X-ray energy and at 6.5 MeV it is about 100 times smaller than at 16 MeV, therefore for the same dose of X-rays near the photofission threshold, where the anisotropy is high, the number of tracks available for measurement is very small. At energies higher than 9 MeV due to the abundance of tracks it is possible to select only the tracks oriented at angles smaller than 15° to the plane of the nuclear emulsion plates. However at energies smaller than 9 MeV due to the paucity of tracks it was decided to apply a different criterion in order to permit the use of a greater number of tracks. If the beam of gamma rays is not polarized the measurements of the differential cross sections at a particular average angle with the beam direction (but at different angles with the plane of emulsion) must yield the same differential cross section since there is an axial symmetry around the beam. Therefore measurement of this kind shows the overall effect of distortion and microscope scanning losses on the measurement of the angular distribution at a particular angle with the beam direction and at a particular angle with the plane of emulsion. The analysis of the data made it possible to determine the effect of distortion and scanning losses and to set a proper cut off at angles greater than the 16° used previously (at energies greater than 9 MeV) so to accept the

U-238 ANGULAR DISTRIBUTION

Maximum X-ray Energy E_o (MeV)	Relative yield for angle interval (per unit of solid angle)						Value of $\frac{b}{a}$ in $Y(\theta)=a+b\sin^2\theta$
	0°- 15° 180°-165°	15°- 30° 165°-150°	30°- 45° 150°-135°	45°- 60° 135°-120°	60°- 75° 120°- 105°	75°- 90° 105°-90°	
6.9	1.00 ± .21	1.28 ± .14	1.70 ± .13	2.45 ± .13	3.30 ± .18	3.28 ± .17	2.80 ± .44
8.1	1.00 ± .15	1.22 ± .09	1.52 ± .08	1.79 ± .08	2.06 ± .10	2.10 ± .09	1.18 ± .14
9.4	1.00 ± .10	1.10 ± .08	1.33 ± .10	1.46 ± .10	1.54 ± .10	1.55 ± .10	.62 ± .12
15.5	1.00 ± .09		1.01 ± .09		1.08 ± .09		0,09 ± 0,14
20	1.00 ± .10	.98 ± .10	.96 ± .10	.98 ± .10	1.03 ± .10	1.00 ± .10	0,08 ± 0,13

TABLE I

maximum number of tracks.

EXPERIMENTAL RESULTS AND DISCUSSION

The angular distribution of fission fragments from the photofission reaction produced by dipole and quadrupole gamma ray absorption has the following general form:

$$I(\theta) = a + b\ (\sin\theta + d \sin\theta \cos\theta)^2$$

With the method used in the present work it is impossible to distinguish the heavy fragment from the light one, thus the measured distribution is $Y(\theta) = I(\theta) + I(\pi - \theta)$ and, as a result, the term with forward backward asymmetry, due to electric dipole--electric quadrupole interference is cancelled out.

The following expression remains:

$$Y(\theta) = a + b \sin^2\theta + c \sin^2\theta$$

or

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

The coefficient "a" determines the isotropic part of the angular distribution, the coefficients "b" and "c" the anisotropic part related to dipole and quadrupole absorption of the gamma rays respectively.

Two different methods, yielding pratically the same results, were used in the computation of the coefficients a, b and c from the experimental data: the least square method, and a cosine Fourier series very appropriate to this particular case of angular distribution (dipole and quadrupole photon absorption), and also very easy to calculate. This permits the computation of the errors of the coefficents a, b and c from the statistical errors of the angular distribution measurements.

The measurements were made at six intervals of solid angles defined by the angles θ with the gamma rays beam direction i.e. 0, 15, 30, 45, 60 and 90. For the computations of the coefficients it is necessary to determine an average angle $\bar{\theta}$ for each solid angle interval. This may be done taking the arithmetical mean: $\bar{\theta} = (\theta_1 + \theta_2)/2$ or averaging θ in the solid angle interval used:

$$\bar{\theta} = \int_{\theta_1}^{\theta_2} \theta \sin \theta \, d\theta \Big/ \int_{\theta_1}^{\theta_2} \sin \theta \, d\theta$$

The last method was used in computing of the coefficients by means of the least square method, because it introduced corrections to the first solid angle intervals. Using the Fourier cosine series

$$Y(\bar{\theta}) = a_1 - (b/2) \cos 2 \theta - (c/8) \cos 4 \theta$$

where $a_1 = a + (b/2) + (c/8)$, $\bar{\theta}$ was taken as the arithmetical mean, so as to yield in the interval 0 to 180°, twelve equal angle intervals each of $(\pi/12)$.

The coefficients a_1, b and c were obtained by the usual Fourier series method as follows:

$$a_1 = (1/6)\Sigma\, y_o,\ b = (2/3)\Sigma\, y_o \cos 2\,\theta,\ c = (8/3)\Sigma\, y_o \cos 4\,\theta$$

where the y_o is the measured angular distribution. Since each measured point has a statistical error $\pm\,\epsilon$, the mean square errors of the coefficients are calculated straightforwardly from the statistical errors $\pm\,\epsilon$ as follows:

$$\Delta a_1 = \pm\,(1/6)\sqrt{\Sigma\epsilon^2},\ \Delta b = \pm\,(2/3)\sqrt{\Sigma\epsilon^2 \cos^2 2\,\theta}$$

$$\Delta c = \pm\,(8/3)\sqrt{\Sigma\epsilon^2 \cos^2 4\,\theta}$$

therefore this method yields errors in a form more reliable than the one usually obtained from the least square method, in which the probable errors computed for the constants result from the closeness of the fit and is not related to the statistical error of the measurements. This point is emphasized here to illustrate the effect of the statistical errors on the computation of the quadrupole coefficient in which the error is about four times greater than on the dipole coefficient.

The angular distribution obtained with the statistical errors at all energies measured are given in table I.

The calculated coefficients are given in table II.

Maximum X-ray Energy (MeV)	$\frac{b}{a}$	$\frac{c}{a}$
6.9	2.80±0.44	0.34±0.85
8.1	1.18±0.14	0.52±0.52
9.4	0.62±0.12	0.60±0.56
15.5	0.09±0.14	-
20.0 MeV	0.08±0.13	-

TABLE II

a) Dipole photon absorption

The ideas put forward by A. Bohr [5] on the collective model of the nucleus allows an explanation of the observed anisotropy in fission fragment emission in terms of the excitation spectrum at fission barrier. For excitation energies only slightly in excess of the lowest fission barrier the nucleus when passing over the barrier is "cold", meaning that a great deal of the excitation energy is spent in potential energy of deformation in reaching the barrier. This implies that the level spectrum at the saddle point should be rather simple if the excitation energy is only slighty greater than the barrier height. It is assumed also that the spectrum of the nucleus at the barrier sould resemble the spectrum of the deformed nucleus in the ground state.

In Bohr's theory one assumes that nucleus retains axial symmetry throughout the fission process and that the fission fragments are emitted in the direction of symmetry axis of the nucleus, therefore the orientation of the symmetry axis corresponds to the angular distribution of the fragments. The distribution is given by the square of the symmetric top wave function for a compound state of angular momentum I, and Z component along the beam M, which pass the barrier in a state of excitation with component K of angular momentum along the symmetry axis.

The anisotropy of photofission of even-even nuclei (spin zero) resulting from a dipole absorption is explained as follows: when the photon is absorbed in the electric dipole mode, the compound state has $I = 1, M = \pm 1$. The low-lying states are characterized by pairing of nucleons to states with $K = 0$. For photofission resulting from the low-lying 1^- states with $K = 0$ at the barrier, the angular distribution would be proportional to a positive $\sin^2 \theta$ term, with emission of fragments dominantly in the 90° directions from the beam. The dipole absorption corresponding to the $K = 1$ states would give an anisotropy of the opposite character, peaked at 0° corresponding to a negative $\sin^2 \theta$ term.

Near the threshold the lowest-lying saddle point is the only one available and the resulting anisotropy is peaked at 90°, but at a few MeV above the threshold both states $K = 0$ abd $K = 1$ are available and the anisotropy from both states tends to cancel yielding a more isotropic distribution. At higher energies there will be many more channels corresponding to other different states and the average angular distribution will then be isotropic.

As illustrated in Fig. 1 the ratio (b/a) measured at five different energies is in a very good agreement with the results of Katz and Lazareva. The ratios b/a are represented in a Katz plot corresponding to the following empirical formula:

$$\log (b/a) = (1.676 \pm 0.024) - (2.66 \pm 0.10) \log (E_0 - 4)$$

calculated by the least square method from the overall data available. (Where E_0 is the peak bremsstrahlung energy in MeV). This is represented in Fig. 1 by the solid straight line.

b) Quadrupole photon absorption

For quadrupole absorption of a photon the compound state will have quantum number $I = 2$, $M = \pm 1$ and the lowest corresponding excitation at the barrier would be a 2^+ state with $K = 0$, the second member of the even parity rotational band.

If at the barrier this state lies sufficiently low compared with the 1^- state, then there might exist a range of photon energies over which the smaller probability of quadrupole absorption is counter balanced by smaller probability of fission through the 1^- state. The resulting angular fission fragment distribution would have a component proportional to a $\sin^2 2\theta$ term with a maximum at 45°, and the same happens for the $K = 2$ state. However the $K = 1$ state has a negative $\sin^2 2\theta$ acting to cancel out the $K = 0$ and $K = 2$ quadrupole angular distributions.

If appropriate proportions of quadrupole were invoked and appropriate K values are assumed it is possible to explain the variation of quadrupole absorption with the energy.

As mentioned above, at 9.4 MeV, the angular distributions obtained by Katz with ionization chamber and by Lazareva with nuclear emulsion technique differ very much regarding the quadrupole coefficient c/a. The results from Katz, Lazareva and the present work, normalized at angle of 7.5°, are represented in Fig. II for comparison. It may be observed that the Katz measurements at 0°, 45° and 90° fit a pure dipole curve (2) with $b/a = 0.46$ but the Lazareva experimental points correspond to a fitting $b/a = 0.5$ and $c/a = 0.9$; the present results correspond to $b/a = 0.6$ and $c/a = 0.6$. Curve (1) represents a Fourier best fitting of Lazareva and the present results together corresponding to $b/a = 0.47$ and $c/a = 0.80$. In conclusion it appears that there is a measurable quadrupole photon absorption at 9.4 MeV with a c/a of about 0.8 ± 0.4.

As was pointed out by Lazareva c/b gives a certain estimate of the relation between the cross sections for electrical dipole and electrical quadrupole photon absorption but due to the high statistical errors (see table II) and other sources of errors not considered, the measurements of quadrupole coefficients, in the range of energy investigated, are at present very poor indeed and do not allow us to draw definite conclusions. However assuming that the measurements of Lazareva and of the present work are correct the general trend of variation of c/b with energy is illustrated in Fig. III.

In order to obtain a better quadrupole coefficient measurement it is necessary to perform more accurate experiments using an indicator of the origin of fission tracks like a 1 μ diameter grain of U_3O_8, so as to distinguish the light fragment from the heavy and thus to obtain the angular distribution in the interval 0 to 180° and from that to compute the interference term, which is removed when it is impossible to distinguish the heavy fragment from the light one.

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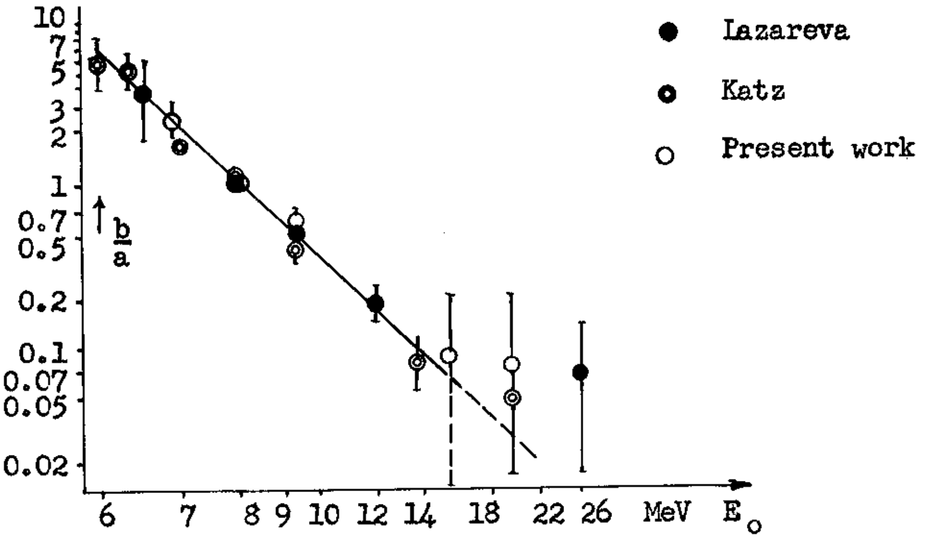

Fig. 1. Variation with the maximum bremsstrahlung energy E_o (in MeV) of ratio b/a, between the coefficients "a" related to the isotropic part of the angular distribution and "b" the anisotropic part related to the dipole photon absorption. The solid line represents the empirical equation $\log (b/a) = (1.676 \pm 0.024) - (2.66 \pm 0.10) \log (E_o - 4)$ obtained from a least square fitting of the results of Katz, Lazareva and present work.

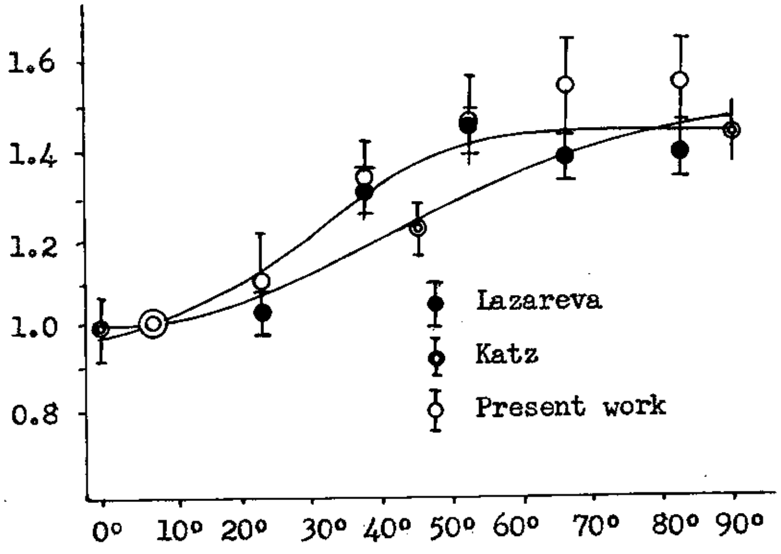

Fig. 2. Angular distribution for 9.4 MeV bremsstrahlung showing the data of Katz, Lazareva and present work, normalized to 7.5°. Curve 1 corresponds to the best fitting of the data of Lazareva and the present work together $(1+0.47 \sin^2\theta+0.80 \sin^2\theta \cos^2\theta)$, curve 2 is a fitting of the Katz results with a pure dipole photon absorption curve $(1+0.46 \sin^2\theta)$.

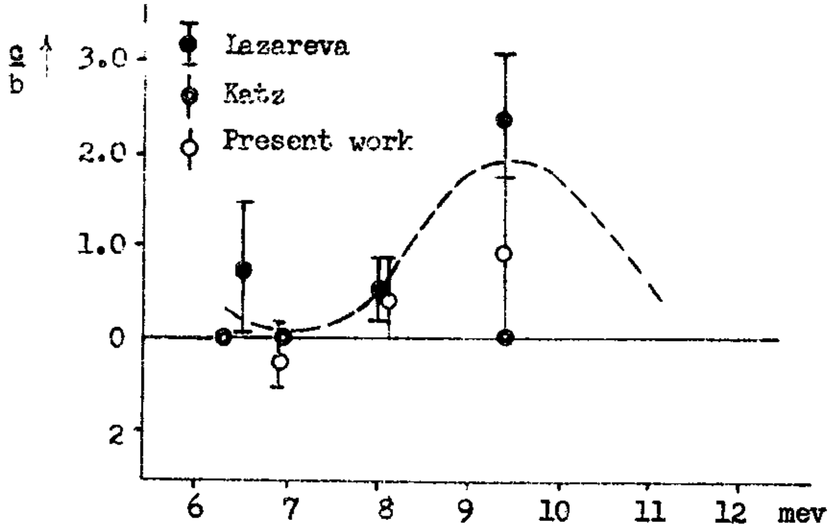

Fig. 3. Variation of the ratio a/b with the peak bremsstrahlung energy.