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ANGULAR DISTRIBUTION OF PHOTOFISSION FRAGMENTS  
FROM THORIUM

by

M. V. Ballariny

CENTRO BRASILEIRO DE PESQUISAS FÍSICAS

Av. Wenceslau Braz, 71

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ANGULAR DISTRIBUTION OF PHOTOFISSION FRAGMENTS  
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M. V. Ballariny

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SUMMARY. The angular distribution of photofission fragments from thorium relative to the photon beam was measured, using the bremsstrahlung of a 24 MeV betatron at the maximum energy of 12,4 MeV, which incided on films of  $\text{ThO}_2$  put upon nuclear emulsions. It was found an anisotropic distribution of the form  $W(\theta) = a + b \cdot \text{sen}^2 \theta$ , that corresponds to an electric dipole photon absorption. The ratio  $b/a$  of the anisotropic dipole fission yield to the isotropic one was found to be  $0,73 \pm 0,15$ .

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\* This work was performed at the laboratories of Centro Brasileiro de Pesquisas Físicas in 1960 as subject for a thesis presented to the Escola Nacional de Engenharia, Universidade do Brasil, for post graduation in Nuclear Engineering, under the advise of Professor H. G. de Carvalho.

## 1. INTRODUCTION

The angular distribution of photofission fragments relative to the photon beam is isotropic for odd-even and even-odd nuclei, but anisotropic for the high mass number even-even ones, the anisotropy degree being function of photon energy <sup>1, 2, 3, 4, 5, 6</sup>. It is maximum in the threshold region, around 5 MeV (5.4 MeV for Th<sub>232</sub>) <sup>7</sup>, falling off fast in the region of higher energies, and is related to the fragments mass distribution assimetry <sup>2</sup>. For Th<sub>232</sub> the angular distribution found has been that corresponding to electric dipole photon absorption, peaked at right angles with photons direction, expressed by:  $W(\theta) = a + b \cdot \text{sen}^2\theta$ . <sup>1, 2, 3, 4, 8</sup>.

The A. Bohr's collective model of the nucleus allows an explanation to the anisotropy of the even-even nuclei fragments distribution, in terms of the fission channels spectrum, as follows <sup>9</sup>.

Assuming that during the fission process the nucleus' shape retains axial symmetry, the fission channels may be characterized by the K component of the nuclear angular momentum in the direction of the symmetry axis. When the photon is absorbed in the electric dipole mode, the resulting compound state has angular momentum  $I = 1$ , and quântic magnetic number  $M = \pm 1$ . The lower stationary state has  $K = 0$ , and the photofission resulting from this state conducts to an anisotropic angular distribution of the fragments peaked at  $90^\circ$  with photons direction expressed by  $W(\theta) = a + b \cdot \text{sen}^2\theta$ . The stationary state with  $K = 1$  conducts to anisotropy peaked at  $0^\circ$  with photons direction, expressed by  $W(\theta) = a - b \cdot \text{sen}^2\theta$ . Near the threshold these spin zero, strongly deformed nuclei, pass the fission saddle point

only in the lowest lying state and so, the resulting anisotropy is peaked at  $90^\circ$  with photons direction, with the anisotropy degree at high values. ( $\text{Th}_{232}$ ,  $E = 7,0 \text{ MeV}$ ,  $b/a = 13 \pm 1$ ) <sup>3</sup>. But to photon energies a few MeV above the threshold, in addition to the channel with  $K = 0$  also the channel with  $K = 1$  is possible, and the two orthogonal anisotropies tend to cancel each other, resulting a more isotropic distribution ( $\text{Th}_{232}$ ,  $E = 10,0 \text{ MeV}$ ,  $b/a = 1,63 = 0,06$ ) <sup>3</sup>. To photon energies still higher, other fission channels become also available, and the resulting angular distribution is almost isotropic. ( $\text{Th}_{232}$ ,  $E = 20,0 \text{ MeV}$ ,  $b/a = 0,14 = 0,06$ ) <sup>3</sup>.

In this experiment the angular distribution of photo-fission fragments from  $\text{Th}_{232}$  at  $12,4 \text{ MeV}$  was measured using a betatron bremsstrahlung incident on  $\text{ThO}_2$  films put upon nuclear emulsions. In this case, for a photofission produced in the film only one fragment will have penetrated the emulsion, but as in the photofission the two fragments go in opposite directions, the angular distribution obtained is correct. As a consequence of the technique, losses of fragments at certain angles depending of the film thickness occurred. But this technique has over the classical technique of loaded emulsions the advantage of permitting higher Th concentration, higher irradiation doses, and easier development techniques, because the properties of the emulsion are not changed by the presence of the chemical components of the loading bath, and also the presence of the  $\alpha$  particles is reduced, once only during the irradiation time Th must be in contact with the emulsion.

## 2. EXPERIMENT

The experimental measurements were performed on the tracks produced in nuclear emulsions by thorium photofission fragments resulting from the section of the X-rays on the  $\text{ThO}_2$  films.

To prepare these  $\text{ThO}_2$  films, an adaptation of Novakov et al. formule to  $\text{UO}_2$  films<sup>10</sup> was used:  $(\text{NO}_3)_4\text{Th} = 200$  up  $500$  mg; ethil alcohol =  $3$  ml; 4% collodium =  $3$  ml; acetone =  $10$  ml. The solution obtained by simple mixture of these components, very volatile and with low viscosity, was sprayed on glass, that was the way we obtained the best homogeneity. After normal drying the films were put in an electric furnace at a temperature between  $500^\circ\text{C}$  and  $600^\circ\text{C}$  for about 12 minutes, to calcinate the organic material. Only an adherent layer of  $\text{ThO}_2$  remained, and the excess was whipped with soft paper. To obtain a film medium thickness of  $0,50\mu$  (thickness measured by weight) this operation was repeated about 25 or 30 times, according with  $\text{Th}(\text{NO}_3)_4$  quantity of the solution. But the increase of this quantity above  $500$  mg was ineffective, since the excess of  $\text{ThO}_2$  was not adherent after the calcination. The films so obtained had good homogeneity, which was better the more they got thick.

The nuclear emulsions used were Ilford K-0 of  $1 \times 3$  inches and  $100\mu$  thick.

The irradiation was performed in the  $24$  MeV betatron of the University of S. Paulo, at the maximum energy of  $12,4$  MeV. For this purpose, the films were put face to face with the

emulsion plates, and the assemblies were put longitudinally in the axial part of the X-rays beam at  $0^\circ$  with the X-rays direction, at 40 cm from the betatron target. The total doses received by the films, measured with a Victoreen ion chamber placed in a cavity at the center of an 8 cm lucite cube, were: 2000 R for  $0,10\mu$  films, 1500 R for  $0,25\mu$  films, and 1000 R for  $0,50\mu$  films. The diminishing of the beam intensity due to absorption along the assemblies was not considered. The energy scale was calibrated by detection of the threshold for the following reactions:  $\text{Cu}^{63} (\gamma, n) 10,6 \text{ MeV}$ ,  $\text{O}^{16} (\gamma, n) 15,85 \text{ MeV}$ , and  $\text{C}^{12} (\gamma, n) 18,75 \text{ MeV}$ , with accuracy of  $\pm 0,1 \text{ MeV}$ . According with measurements performed by H. G. de Carvalho, the fraction of fissions due to fast neutrons from the betatron beam, and to slow neutrons and scattered X-rays from background was less than 2%.

The irradiated assemblies were maintained at  $0^\circ \text{C}$  during about 12 hours for transportation to the C.B.P.F. laboratory (Rio de Janeiro), where the plates were separated from the films and put in a developing bath composed of: boric acid = 3,5 g; sodium sulfite = 4,5 g; potassium bromide = 1,5 g; amidol = 0,45 g; distilled water to complete 2 liters. The developing time was established previously in order to develop only the fission fragment tracks, and was about 20 minutes. After the developing bath the plates were put in an 1% cold solution of acetic acid ( $8 - 10^\circ \text{C}$ ) under continuous vibration, during one hour, after washed in cold distilled water ( $8 - 10^\circ \text{C}$ ) during two hours, and put again in a cold 0,5% solution of acetic acid during ninety minutes. Then they were fixed with a cold 30% hiposulfite solution ( $8 - 10^\circ \text{C}$ ) which, on account of its use before, had a large quantity of silver,

that diminished the production of background grains. After the fixing process the plates were washed in cold tap water, and in order to keep low the shrinkage factor, they were put in a 10% glicerine solution during ninety minutes and then submitted to slow drying.

The scanning, only at the central region of the plates, was performed with Leitz Ortolux microscopes, with 100 x objective and 6 x eye pieces. The following direct measurements of photo-fission fragments tracks were performed:

- 1) length of the projection on the emulsion surface;
- 2) angle  $\beta$  between this projection and the X-rays direction, reduced to the first quadrant, for it was not possible to distinguish between the heavy and the light fragment tracks, since for each fission only one fragment has penetrated the emulsion;
- 3) depth of the tracks related to the emulsion surface, for the fissions were produced at the films, and so, all the fragments tracks had their origin at the emulsion surface.

The emulsion thickness of each plate was measured just before the irradiation, at four points, around small holes cut in the emulsion, using a dial micrometer. To obtain the shrinkage factor during the scanning, the plates thickness were measured at the same points four times a day, with dial micrometers adapted to the microscopes.

### 3. RESULTS, ANALYSIS AND DISCUSSION

The dip angle ( $\alpha$ ) was calculated from the two lengths measured. The spacial angle ( $\theta$ ), of the fragments direction with axial direction of X-rays beam, was calculated by the relation:

$$\cos \theta = \cos \alpha \cdot \cos \beta$$

Assuming that the fissions were produced at the medium layer of the films, by considerations of geometry, the conclusions are that only the fragments whose directions had dip angle greater than:

$$\alpha_{\min} = \text{arc. tg} \left[ \frac{(2L - t)}{2m} \right]$$

(where: L is the total average range of the fragment, considered equal in both film and emulsion,

t is the  $\text{ThO}_2$  film thickness

m is the least distance measurable at the microscope)

penetrated the emulsion, producing there a developable track measurable by the microscope.

Besides, other factors as the emulsion distorsion during developing process, the preference of scanners by certain angles, and other occasional or sistematic errors, produced deformation in the tracks spatial distribution, which should be axially symmetric relative to X-rays direction. To exclude the angular regions where this deformation took place, the tracks were plotted having as coordinates ( $\sin \alpha$ ) and ( $\beta$ ). Upon this plot lines with  $\theta = \text{constant}$  were drawn and the tracks densities around these lines were determined. This analysis was made



separately to each plate. In another graphic, the tracks' densities around each line  $\theta = \text{const.}$  was plotted as function of  $(\text{sen } \alpha)$  also to each plate separately, and the  $\alpha$  region where this density remained constant in all the plates, that means, the region where the distribution was axially symmetric, was determined. The tracks at this region, that means, the tracks with dip angle between  $20^\circ$  and  $55^\circ$  were chosen for the calculations. Of the 1150 tracks measured, only 840 were computed.

As distinction was not made between the light and the heavy fragments, the spatial angles  $\theta$  and  $(\pi - \theta)$  were also indistinguishable, and so, the angular distribution measured was actually:  $W'(\theta) = W(\theta) + W(\pi - \theta)$ .

To the angular distribution determination, the tracks were plotted in six solid angle intervals defined by their average angle  $\bar{\theta}$ . The relative yield for  $\theta$  angle interval, per unity of solid angle, in table I, is given by:

$$N(\bar{\theta}) = \int_{\theta_1}^{\theta_2} W'(\theta) \cdot d\Omega \Big/ \int_{\theta_1}^{\theta_2} d\Omega ,$$

and was easily calculated on the graphic having as coordinates  $(\text{sen } \alpha)$  and  $(\beta)$ , where equal surfaces correspond to equal solid angles.

As average angle  $\bar{\theta}$  for each solid angle interval, it was took the arithmetical mean:  $\bar{\theta} = (\theta_1 + \theta_2)/2$ . The tracks at the interval  $(\theta_1 = 0^\circ; \theta_2 = 15^\circ)$  were not computed, for they have dip angles less than  $20^\circ$ , as:  $\alpha_{n1} \leq \theta_n$ . The first  $\theta$  interval calculated was

really  $\theta_1 = 20^\circ$ ;  $\theta_2 = 25^\circ$ . The values of the relative yields at Table I are normalized to unity at  $\bar{\theta} = 22,5^\circ$ .

TABLE I

$\text{Th}_{232}$  angular distribution at 12,4 MeV

$\theta$	$15^\circ - 30^\circ$	$30^\circ - 45^\circ$	$45^\circ - 60^\circ$	$60^\circ - 75^\circ$	$75^\circ - 90^\circ$
Interval	$165^\circ - 150^\circ$	$150^\circ - 135^\circ$	$135^\circ - 120^\circ$	$120^\circ - 105^\circ$	$105^\circ - 90^\circ$
$\bar{\theta}$					
(average)	$22,5^\circ$	$37,5^\circ$	$52,5^\circ$	$67,5^\circ$	$82,5^\circ$
relative yield per unity of solid angle	1,00	1,12	1,30	1,45	1,54
	$\pm$	$\pm$	$\pm$	$\pm$	$\pm$
	0,08	0,10	0,09	0,09	0,08

As the known results in other photons energies 1, 2, 3, 4 and the A. Bohr's theoretical previsions indicate for  $\text{Th}_{232}$  an angular distribution of photofission fragments corresponding to an electric dipole photo absorption, and besides, an analysis of the experimental results in Table I indicates a distribution peaked at  $90^\circ$  with X-rays direction, an expression of the type  $W(\theta) = a + b \cdot \text{sen}^2 \theta$  was fitted by the least squares method. The calculations were performed by a 205 Burroughs computer, and the b/a ratio of the anisotropic dipole absorption fission yield to the isotropic one was found to be  $0,73 \pm 0,15$ .

This result is in agreement with Katz, Baerg and Brown empiric formula <sup>3</sup> :  $(b/a)$  to  $\text{Th}_{232} \approx 68/(E-5)^{2,31}$ , which for  $E = 12,4$  MeV

conducts to  $b/a$  at an order of 0,69.

Fig. I shows the results of Winhold et al. <sup>1</sup> obtained by the catch foil technique, those of Katz et al <sup>3</sup> obtained using ionization chambers, that of Faissner and Gonnenswein <sup>8</sup> obtained by the loaded emulsion technique, and the obtained by the film - e-mulsion technique in the present work.

Fig. II shows the form of the angular distribution expressed by  $W(\theta) = 1 + 0,73 \text{ sen}^2 \theta$  and the  $W(\theta)$  values obtained experimentally.

Attention must be paid to the fact that the X-rays beams used in those experiments were continuous energies spectrum beams. This condition associated to the property of decreasing anisotropy of the photofission fragments distribution with photons energy increasing, allows to foresee more isotropic distributions for photofissions by monochromatic beams.

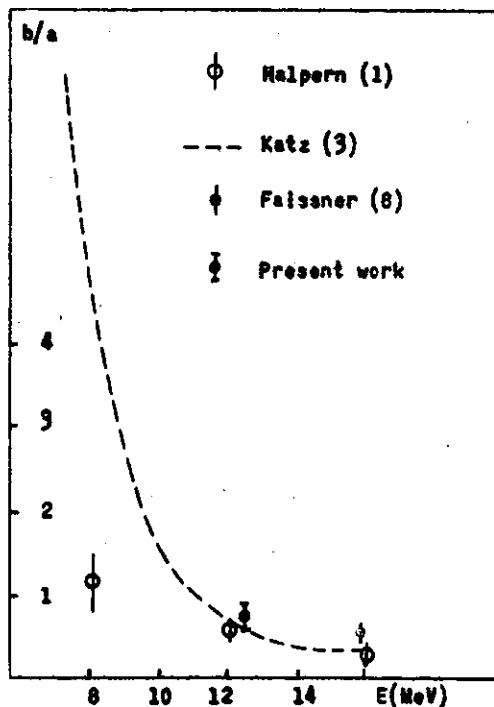


Fig. I

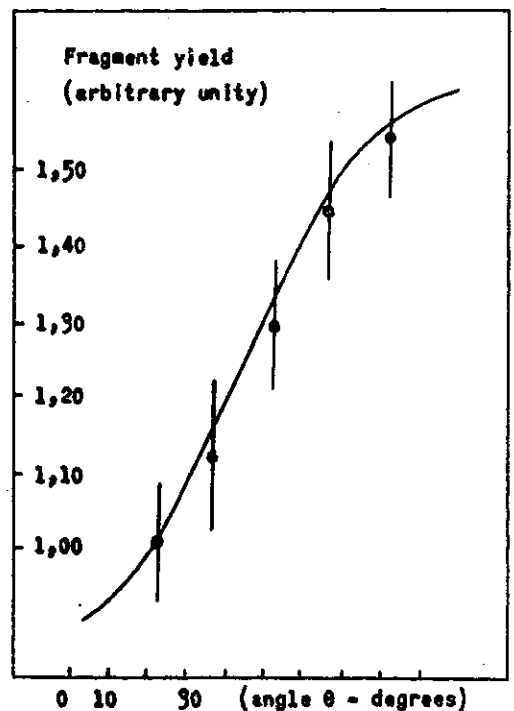


Fig. II

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