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by

A. Manfredini, M. Muchnik, M. Severi, R. Bosch,
W. Wölfli and H. G. de Carvalho

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II. - ANGULAR DISTRIBUTIONS AT 6.61 MeV

A. Manfredini, M. Muchnik *, M. Severi
Istituto Nazionale di Fisica Nucleare - Sezione di Roma
Istituto di Fisica dell'Università - Roma

R. Bösch and W. Wölfli
Laboratorium für Kernphysik, Eidg. Tech. Hochschule - Zürich **

H. G. de Carvalho ***
Centro Brasileiro de Pesquisas Físicas, Rio de Janeiro, Brazil

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SUMMARY. The angular distribution of the photofission fragments of uranium and thorium has been measured at 6.61 MeV, by combining a new type of monochromatic γ-ray source with some new developments of the nuclear emulsion technique. Using the results of part I of this paper [1], values are given (in millibarns) for the isotopic, dipole and quadrupole absorption cross-sections in the photofission process.

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* Present address: Istituto Nazionale di Fisica Nucleare, Sezione di Napoli.

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1. INTRODUCTION

The property of heavy, even-even, nuclei of exhibiting an anisotropic angular distribution of photofission fragments near the threshold is a well established experimental fact [2]. It was observed by means of bremsstrahlung γ-ray bombardment that the anisotropy decreases with increasing energy and practically disappears at about 10 MeV above what is commonly considered the threshold (5.81 MeV for uranium). The anisotropy seems to consist mainly of a dipole term and, to dipole-order terms, may be expressed as

$$W(\theta) = a + b \sin^2\theta .$$

The ratio b/a has been measured for uranium and thorium [3,5] from 6 MeV up to about 30 MeV maximum bremsstrahlung energy, and a decrease of about 3 orders of magnitude has been ascertained in that interval.

The anisotropy cannot be explained on the basis of a simple liquid-drop model of the nucleus. Correct qualitative predictions issue from the collective model: by application of the Franck-Condon principle, it envisages the anisotropy as the result of a sizeable oscillatory motion of the compound nucleus [6]. The more successful unified model of Bohr and Mottelson [2] gives more quantitative predictions which can be tested experimentally. This theory considers the nucleus going over the saddle point "cold", that is: the nuclear excitation levels would remain almost unchanged. At the saddle point each fission channel would be correlated with a set of quantum states of the nucleus, widely separated. A simple consideration of symmetry and total angular momentum conservation predicts a series of collective excitation states, each associated with a particular angular distribution. As the incident photon energy increases, the contribution of the various fission channels to the angular distribution thus changes, and rough predictions can be advanced as to the type of asymmetries to be expected. Unfortunately the position of the energy levels has not been measured yet, and the predictions of the theory contain an element of uncertainty.

A further element of uncertainty is introduced by the fact that, at about one MeV above the fission threshold the phenômenon of neutron emission sets on. Neutron emission itself takes place through the absorption of quanta whose multipolarity varies with energy.

The study of photofission near threshold, when carried out by means of monoenergetic γ-rays not only can give important information concerning the nature of the angular distributions and serve as a test of the theory, but seems to be the only available way, as pointed out by previous authors [7], of obtaining separate cross-sections corresponding to dipole and to quadrupole photon absorption processes.

The present paper is the second part of a report on an exposure of uranium and thorium to 6.61 MeV γ-rays; it consti tutes the beginning of a systematic investigation of the photofission process from the threshold up to energies near the giant resonance ($\sim$ 15 MeV), making use of two new techniques described below. Although Parts I and II refer only to a single energy, we feel that the present results have an interest, also from the technical point of view, so as to be published at the present time.

2. EXPERIMENTAL METHOD

As pointed out in the introduction, it is important to study the photofission process near threshold by means of monochromatic γ-rays. The very fact that, as the energy increases, new anisotropies are introduced in the angular distribution, and that the onset of these successive anisotropies takes place at intervals of one MeV or less, indicates that the use of bremsstrahlung beams is hardly suited for this kind of investigation.

Monochromatic γ-rays have been used in the past[7], originating in the F(p,γ) reaction. This reaction produced three lines, at energies of 6.14, 6.93 and 7.14, whose intensities depend both on the energy of the bombarding proton and on the thickness of the F target employed. Due to temperature effects, the target thickness itself may vary during the experiment, introducing further uncertainties.

The method used by us, making use of reactions of the type A(n, γ) where the thermal neutron flux is that of a nuclear reactor, has been already described in Part I of this paper. It is to be emphasized at this point that the three lines used by us for this exposure, belonging to the Ti(n, γ) reaction, cover an energy interval of only 340 keV and that their relative intensities are constant. Furthermore, the intensities have been measured with good accuracy.

Another point of importance, which has a bearing on the uncertainties with which an angular distribution is measured, is the fact that the collimation system employed at the SAPHIR reactor is such that the emulsions are placed at more than four meters from the γ-ray source, so that there is practically no angular dispersion of the beam. Once the emulsions have been aligned, the direction of the incoming γ-rays is extremely well defined and no appreciable error is introduced in the measurements by neglecting the indetermination of the 0^o direction.

Special care was taken in the preparation of the loaded emulsion so as to introduce as few sources of distortion as possible. The following are some of the precautions introduced in our usual method of loading by mixing the solution of the element with the emulsion in gel form, melting and pouring.

1) Diluted gel was used, of the "KO × 2 gelatin" type, in order to reduce the shrinkage factor.

2) The loading solution's pH was fixed at 5, as near as

possible to the isoelectric point of the gelatin, where the swelling is smallest.

3) Rather large emulsions were prepared - of the order of 7 cm × 15 cm - in order to avoid edge effects; only a small central area was scanned.

4) Both pellicles and plates were prepared, so as to have a choice of the less distorted material for scanning.

5) Processing was carried out at low temperature ($\sim 5^{\circ}C$) and at constant pH as far as possible. Fixing was at nearly $0^{\circ}C$.

6) The emulsions, developed without glass supports, were carried back almost to their original dimensions by slowly adding to the washing bath, a solution of ethylic alcohol with 3% of glycerin so as to reach a 65% alcohol concentration in two hours, and leaving the emulsion for another two hours in this solution. Temperature was kept around $2^{\circ}C$ throughout the process.

7) Once the emulsions were mounted on glass supports, drying was carried out very slowly.

The exposure was the same as for Part I, but the plates were placed at grazing incidence (angle of incidence = 89.5°). In order to avoid the effects (however small in principle) of the scattering of γ-rays and consequent variation of energy and loss of collimation, a distance of nearly one centimeter was interposed between one plate (or emulsion) and the next.

Fission tracks in K-0 diluted emulsions processed by our method - which eliminate all background - are not heavily ionized in aspect. Thus any fading could significantly alter the aspect of the tracks, perhaps to the point of introducing difficulties in the measurement of angles. Care was taken to expose the plates at $0^{o}C$ throughout, and to store them at that temperature and at low humidity till the moment of processing, thus minimizing fading.

A very high density of fission events was obtained in the plates, as said in Part I. This was particularly advantageous since, for the determination of the angular distribution, it is convenient to limit the measurements to tracks inclined at not more than a given angle β_0 with the plane of the emulsion. Our high density of fissions allowed us to confine the measurements to tracks with $\beta \leqslant 15^{o}$ and still obtain a good statistical sample.

All angle measurements were carried out in Koristka MS2 microscopes, whose dip measurement is very accurate. Optics were 10 × 100, with a calibrated eye-piece micrometer and an eye-piece external goniometer accurate to 15'.

3. ANALYSIS AND ELABORATION OF DATA

If the photofission reaction takes place only through dipole and quadrupole absorption of γ-rays, the angular distribution will be of the form

$$Y(\theta) = a + b(\sin\theta + d \sin\theta\cos\theta)^2. \qquad (1)$$

Since in nuclear emulsion measurements of fission events one does not distinguish between θ and $\theta + \pi$, the observed angular distribution will be of the form

$$W(\theta) = a + b \sin^2\theta + c \sin^2\theta \cos^2\theta . \qquad (2)$$

The space angle θ between the fission track and the beam direction - taken as $\theta = 0^\circ$ - is determined by the measurement of two angles: α, the projected angle on the plane of the emulsion, and β, the angle between the fission track and the plane of the emulsion.

When, as in the present case, the $\theta = 0^\circ$ direction lies on the plane of the emulsion, the angles α, β and θ are connected by the simple relation:

$$\cos\theta = \cos\alpha\cos\beta. \qquad (3)$$

The error on the angle θ thus determined is given by

$$d\theta = \frac{\cos\alpha \ \cos\beta}{\sqrt{1 - \cos^2\alpha\cos^2\beta}} \sqrt{\mathrm{tg}^2\alpha(d\alpha)^2 + \mathrm{tg}^2\beta(d\beta)^2}, \qquad (4)$$

where, in the case of the present work, $d\alpha = 0.25^\circ$ while $d\beta$ is a function of β itself and is given by

$$d\beta = 0.107 \sqrt{1 - 5.22\ 10^{-3}(d_2 - d_1)^2 - 4.53\ 10^{-7}(d_2 - d_1)^4}, \qquad (5)$$

where $(d_2 - d_1)$ is the difference of the z-coordinates of the end points of the fission track. The numerical factors are calculated from the shrinkage factor of the plates and the precision and magnification of the microscope used in the measurements.

It is thus seen that the measurement error for low values of θ is considerable. It follows that the $\Delta\theta$ intervals taken as the basis of accumulation of data, and consequently the corresponding solid angle, as well as the mean value of θ within that interval, are all affected by errors which are strongly dependent functions of θ itself (and which could be considered negligible compared with the statistical error on the number of tracks in the interval $\Delta\theta$ only for values of θ larger than 60° or 70°).

In order to diminish such measurement error, one may use eq. (3) to transform $W(\theta)$ into a $W(\alpha)$, by integrating over β from 0° to β_o. One thus obtains

$$W(\alpha,\beta\leqslant\beta_o) = \left[A + B\sin^2\alpha + C\sin^2\alpha\cos^2\alpha\right]\sin\beta_o, \qquad (6)$$

where

$$A = a + \frac{b+c}{3}\sin^2\beta_o - \frac{c}{5}\sin^4\beta_o,$$

$$B = b - \frac{b+c}{3}\sin^2\beta_o + \frac{c}{5}\sin^4\beta_o,$$

$$C = c\left(1 - \frac{2}{3}\sin^2\beta_o + \frac{1}{5}\sin^4\beta_o\right).$$

Here the uncertainty on the difinition of the $\Delta\alpha$ intervals is constant, as $d\alpha$ is not dependent on the values of α and is quite small ($d\alpha = 0.25^\circ$); while $d\beta$ depends only on the cut-off angle β_o and, a priori, may be as small as desired. Besides, the mean value $\bar{\alpha}$ corresponding to each interval is well defined and each interval has the same statistical weight so that no normalization is necessary. On the other hand, it is evident from (6) that the function $W(\alpha, \beta \leqslant \beta_o)$ is less dependent on the actual values of a, b, c than the function $W(\theta)$, so that the cut-off angle has to be determined on the basis of two opposite criteria:

1) to accumulate the maximum number of tracks for a given scanning time;
2) to minimize the errors on the coefficients of the angular distribution.

Considering that, for densities of fission events not greater than a certain limit, the number of measured events is obviously proportional to the area scanned in the given time interval, the number of measured events per day increases with the chosen cut-off value β_o.

In the present paper the value of β_o chosen is 15^o: the high density of events in our plates allows nevertheless the accumulation, within this limiting angle of dip, of a sufficient number of tracks to provide a good statistics. Furthermore, at such low values of β_o distortion effects are negligible [8].

Finally, another source of error arises from the eventuality that the scanner's efficiency be a function of the angle of the track. This was put to test by having various scanners measure the angular distribution (of the projected angle α) corresponding to the fission events found in the plates used for the determination of the photofission cross-section (see Part I), which were exposed normally to the beam. This distribution is obviously expected to be isotropic; however, to render any unexpected anisotropy null for the present test, the plates were rotated on the microscope stages by about 15° to 20° for every 1000 measured tracks. The distributions thus obtained, with the $\alpha = 0^\circ$ direction taken invariably as the x-axis of the microscope stage, shows for all our scanners that the efficiency is a function of α, with a minimum for $\alpha = 0^\circ$. Figure 1 shows the result of this test: the curve represents the efficiency function used by us to correct the experimental points. A 3% error was attributed to the correcting factor for all values of α.

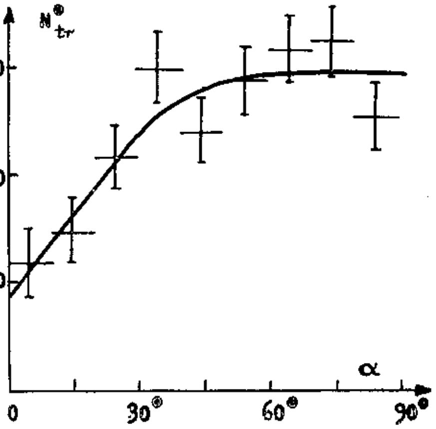

FIG. 1. - Efficiency of detection of fission tracks for an isotropic distribution. Abscissae: angle α of the track with respect to the axis of the microscope stage. Ordinate: actual number of measured tracks. The solid line represents the function used for the correction of the measured angular distributions.

To minimize the effects of such correction, however, the angular distributions were measured by having the plates aligned with the beam direction along the x-axis of the microscopes for half the time, and with the beam direction along the y-axis for the other half; all other conditions of measurement remaining the same. In this way the scanning efficiency influences in a different manner the angular distribution to be measured; the agreement within statistical errors of two such distributions corrected for the efficiency factor gives a plausibility test for the efficiency correction curve which has been used. Figure 2 shows two distributions thus measured, with and without the correction for the efficiency. The quoted errors are only statistical.

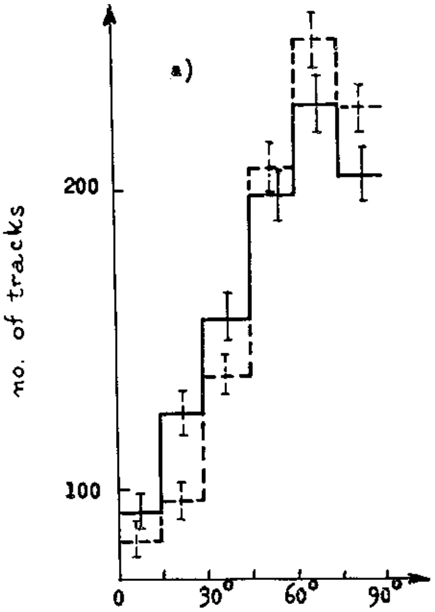

FIG. 2a. - Comparison of the angular distributions measured on the same emulsion, placed in two different positions on the stage. Solid lines direction of the γ beam, parallel to the x axis of the stage; dashed lines direction of the γ beam, perpendicular to the x axis of the stage. Histogram normalized to 1000 tracks.

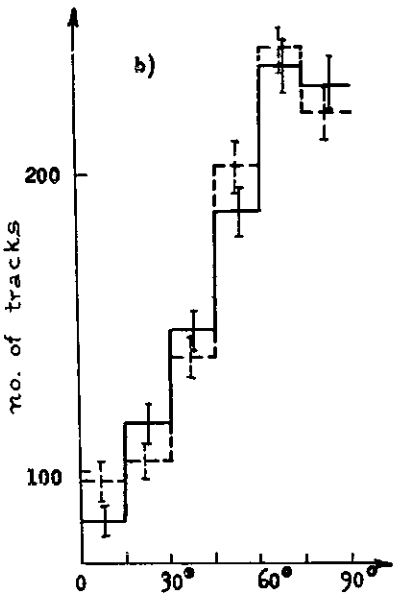

FIG. 2b. - The same distributions as in Fig. 2a, but corrected by means of the efficiency curve of Fig. 1. Histogram normalized to 1000 tracks.

We conclude that, with our precautions in this experiment, the instrumental errors, with the possible exception of the efficiency error, are negligible with respect to the statistical one.

4. EXPERIMENTAL RESULTS

Figures 3, 4 show the angular distributions $W(\alpha,\beta \leqslant 15^{o})$ measured both for uranium and thorium. The errors comprise both the statistical errors and the error on the efficiency. The total number of measured tracks of 5600 for uranium, and 3100 for thorium. The experimental points divided into 18 intervals of width $\Delta\alpha = 5^{o}$, have been analysed either by the method of least squares applied to the function

$$A_o + B_o \sin^2\alpha + C_o \sin^4\alpha$$

or by a Fourier analysis of the function

$$A_1 + B_1 \cos 2\alpha + C_1 \cos 4\alpha \text{ *}$$

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* For $\beta_o = 15^{o}$ the coefficients of the 2 distributions are related to those of eq. (2) by

$$\begin{cases} A_o = a + 0.0224\,b + 0.0215\,c \\ B_o = 0.9776 + 0.9348\,c, \\ C_o = -0.9651\,c, \end{cases} \qquad \begin{cases} A_1 = a + 0.511\,b + 0.130\,c, \\ B_1 = -0.489\,b + 0.0107\,c, \\ C_1 = -0.120\,c. \end{cases}$$

For the least squares analysis the errors on the coefficients A_o, B_o, C_o were obtained from the dispersion of the experimental points from the calculated function; from this the values of a, b and c of eq. (2), as well as the corresponding errors, were found, while, for the Fourier analysis, the values of A_1, B_1, and C_1 as well as their errors were found as explained in (4). The values of b/a, c/a and c/b as well as their errors are shown in Table I. The agreement between the two methods of elaboration is evident, and we take it as

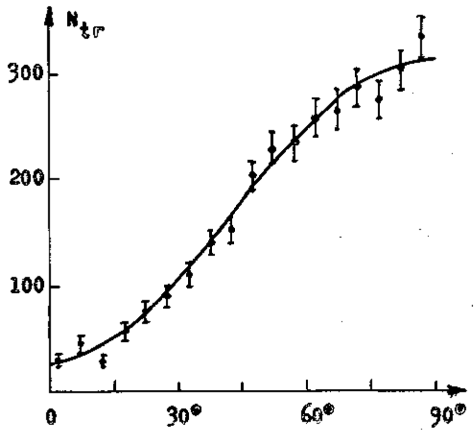

FIG. 3. - Angular distribution $W(\alpha, \beta = 15°)$ of fission fragments from ^{232}Th. E_γ = 6.61 MeV.

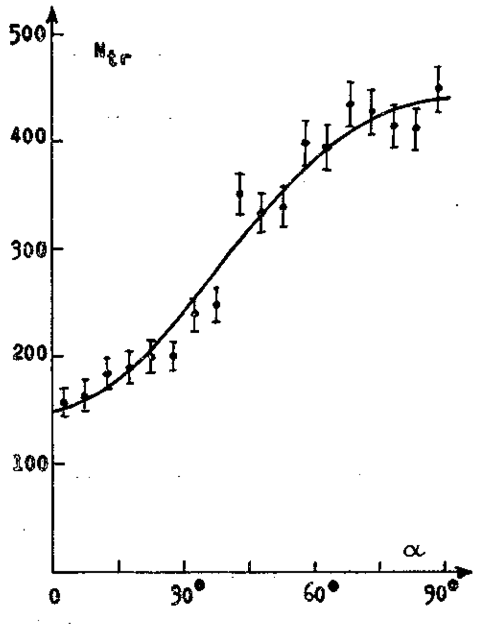

FIG. 4. - Angular distribution $W(\alpha, \beta_o = 15°)$ of fission fragments from ^{238}U.

TABLE I

Nucleus	Method	b/a	c/a	c/b
^{232}Th	Least squares	13 ± 4	1 ± 2	0.1 ± 0.1
	Fourier	11 ± 3	1 ± 1	0.1 ± 0.1
^{238}U	Least squares	2.1±0.5	0.6±0.4	0.3 ± 0.2
	Fourier	2.0±0.2	0.6±0.3	0.3 ± 0.2

TABLE II

	Eγ^* MeV	γ-ray source	b/a	c/a	c/b
Th	6.5	bremms.[2]	> 25	-	-
	6.61**	Ti(n,)[7]	11 ± 3	1 ± 1	0.1 ± 0.1
	7.0 ***	bremms.[2]	10.6±2.1	-2.1±1.6	-0.20±0.16
	6.1	F(p,γ)[7]	14 ± 14	21±21	1.4 ± 0.2
	6.5	bremms.[2] bremms.[3]	4.4 ± 1.0 4.2 ± 1.8	- 3.1 ± 3.2	- 0.75±0.71
	6.61**	Ti(n,γ)	2.0±0.2	0.6 ± 0.3	0.3 ± 0.2
	6.9	F(p,γ)[7] bremms.[4]	0.7±0.3 2.80±0.44	0.2 ± 0.8 0.34±0.85	0.3 ± 1.1 0.12 ± 0.30
	7.0 ***	bremms.[2]	2.11±1.24	0.96±0.88	0.45 ± 0.49

* Eγ indicates either the maximum bremsstrahlung energy of the energy of the monoenergetic beam, as the case may be. Only those points of other authors are quoted whose energy lies within 0.5 MeV from our energy (6.61 MeV).

** We give the values obtained by means of the Fourier analysis of the data (see Table I).

*** For purpose of comparison we quote the values given in the paper of Baerg et al.[2], including the coefficient c, although the authors assume in their discussion c = 0.

an indication that the experimental points fluctuate as expected for a normal distribution*. Table II summarizes the comparison of our results with those of previous papers, which we have limited to the parameters b/a, c/a and c/b as measured by Katz et al.[3] for U and Th using bremsstrahlung, Baz et al.[4] and de Carvalho et al.[5] for U using bremsstrahlung, and Forkman et al.[7] for U using the F(p,γ) reaction. However, the comparison is not very significant. For one thing the angular distributions found using bremsstrahlung beams are the result of the summed contributions of all fission channels whose energies lie below the maximum energy of the continuous spectrum.[5] In our case the spectrum consists of three very narrow lines (of width of the order of a few eV and energies of 6.41, 6.55 and 6.75 MeV). Since the position of the channels is not known, nor their width, it could be that the angular distribution found by us agrees with the angular distributions found by means of bremsstrahlung only because, by chance, our lines excite the same mixture of channels excited by the continuous spectrum. Or else, if the channel widths are rather large, it could be that the spectrum shape becomes less important as regards the observed angular distribution. Whether the agreement - or lack of disagreement - between our results and those of the authors who have used bremsstrahlung is a chance event or

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* Note that, because of the algebraic form of the coefficients in the least squares and Fourier analysis methods, in the first case the error on b is large while in the second case the error on a is large.

is connected with the channel widths cannot be established at the present stage of our work; measurements at other energies with our monochromatic source will give information on this matter.

It is clear that the integral over the whole solid angle of W(θ) should be proportional to the total photofission cross section. Integration of W(θ) - remembering that in nuclear emulsions the measured distributions does not distinguish between θ and θ+π -yields

$$\sigma_1 = H(a + 2b/3 + 2c/15) , \tag{7}$$

where H is a proportionality constant. This constant can be determined by introducing in (7) the value found by us in Part I of this paper. We find $H_{Th} = 5.5\ 10^{-3}$ and $H_U = 1.4\ 10^{-3}$. With these two values and obvious definitions, we find the cross-sections corresponding to the isotropic, dipole and quadrupole absorption processes shown in Table III.

In this table σ_d and σ_q are called cross-sections for dipole and quadrupole absorption, although, for several reasons, their physical nature might not be so clear-cut. If one considers Bohr's theory of fission, the level spacing is probably so large that we may assume that our incident energy will not excite K = 2 channels *. The K = 1 channels, according to the

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* K is the projection on the symmetry axys of the nucleus, of the total angular momentum. In an even-even nucleus, K = 0 corresponds to a configuration with all nucleons coupled by pairs; K = 1 to at least one uncoupled pair of nucleons; K = 2 to at least two uncoupled pairs.

theory, should yield angular distributions whose asymmetries are of opposite sign to those of the K = 0 channels. Thus, the values given for σ_d and σ_q in Table III represent an upper limit for the actual dipole and quadrupole absorption cross-sections.

TABLE III *

Nucleus	σ_f(mb) (from part I) **	σ_{is} (mb)	σ_d(mb)	σ_q(mb)
^{232}Th	5.5 ± 0.6	0.7 ± 0.2	4.8 ± 0.6	0.1 ± 0.1
^{238}U	9.2 ± 0.9	3.7 ± 0.4	5.1 ± 0.6	0.3 ± 0.2

* The values of σ_{is} σ_d and σ_q have been computed using the Fourier analysis results for a, b and c (see Table I).

** An error on the dose measurement has been detected since the publication of Part I of this paper. Instead of ($2.0\ 10^{11} \pm 10\%$) quanta/cm the dose was ($1.72 \cdot 10^{11} \pm 10\%$) quanta/cm ; accordingly the cross-sections reported in Part I should be changed to the values shown in the present Table III.

On the other hand, the isotropic term could appear either by the contribution of fission through barrier penetration [2], or by a mixture of levels, for instance K = 0 and K = 1 levels.

Supposing only the first hypothesis to be true, and only K = 0 levels to contribute to the angular distribution, then the numbers quoted in Table III, would in fact represent the cross-section for fission through barrier penetrations (σ_{is}), through dipole absorption via the $(1^-)K = 0$ channel (σ_d), and quadrupole absorption via the $(2^+)K = 0$ channel (σ_q).

This extreme hypothesis is evidently not true as our energy is well above threshold, and therefore contribution through barrier penetration should be sensibly lower than 50%.

Assuming, on the other hand, no barrier penetration at all at our energy - which is also an extreme hypothesis - and assuming that all fissions go through the $(1^-)K = 0$, $(1^-)K = 1$ and $(2^+)K = 0$ channels, Bohr's theory would give, for the first two channels, respectively, the distribution $\sin^2\theta$ and $2 - \sin^2\theta$. If one weighs these two distributions in such manner as to obtain, from their sum, our experimentally determined one, one gets the following two channels contributions

$\sigma_{Th} = 5.0$ mb; $\sigma_U = 6.3$ mb for $(1^-)K = 0$ level,
$\sigma_{Th} = 0.5$ mb; $\sigma_U = 2.5$ mb for $(1^-)K = 1$ level.

These considerations are, of course, only tentative, for they are based on measurements at a single energy. Cross-sections and angular distributions at monochromatic energies of 7.38 and 5.43 MeV, obtained from the Pb(n,γ) and S(n,γ) reactions, will be soon published and will give more information on which to base a more sound discussion.

* * *

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Errata corrige

In Table III of Part I, the value reported for the ratio as measured by Katz et al. [2] has been erroneously calculated without weighing the contributions of three differently intense lines of titanium. Instead of 0.92 it should be 0.85.

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