Angular distribution of Pu-239 photofission fragments near threshold, using neutron capture gamma rays

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The angular distribution of Pu-239 photofission fragments produced by thermal neutron capture gamma rays in two targets placed in the IEA-R1 (Instituto de Energia Atomica-Reator 1) research reactor were measured. Angular distributions of the form $W(\theta) = A + B \sin^2 \theta$ were observed for discrete excitation energies of 5.43 MeV (sulphur target) and 7.35 MeV (lead target). An anisotropy that corresponds to $(B/A = -0.122 \pm 0.036)$ was recorded for $E = 5.43$ MeV. The angular distribution for $E = 7.35$ MeV was practically isotropic: $(B/A = -0.01 \pm 0.02)$.

On a mesuré la distribution angulaire des fragments de la photofission de Pu-239 produite par des rayons gamma de capture de neutrons thermiques dans deux cibles placées dans le réacteur de recherche IEA-R1. Des distributions angulaires de la forme $W(\theta) = A + B \sin^2 \theta$ ont été observées pour deux énergies d'excitation, 5.43 MeV (cible de soufre) et 7,35 MeV (cible de plomb). Une anisotropie qui correspondait à $(B/A = -0,122 \pm 0,036)$ a été enregistrée pour $E = 5,43$ MeV. La distribution angulaire pour $E = 7,35$ MeV était pratiquement isotrope : $(B/A = -0,01 \pm 0,02)$.

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Introduction

According to the Bohr hypothesis regarding fission channels (1), the angular distribution of fission fragments can provide useful information about the nuclear states at the saddle point configuration. Extensive studies relative to angular distribution of fission fragments for even-*A* nuclei can be found in the literature (2–7), because the chances of discovering anisotropies are higher for these nuclei. The experimental results obtained are in excellent agreement with the Bohr theory (1).

As pointed out by Griffin (8), the best chance of observing the angular anisotropy in an odd-*A* nucleus would be with Pu-239, because this nucleus has the lowest possible ground-state spin, $J = 1/2$. The results of Rabotnov *et al.* (2) and Soldatov *et al.* (9) have in fact shown this to be true by using bremsstrahlung gamma rays with energies of 5–6 MeV produced by electron accelerators. Values of angular anisotropies as large as 20% were observed in this energy interval for the Pu-239 nucleus.

However, for odd-*A* nuclei with spin $J > 1/2$ there are some indications of anisotropic angular distributions, such results have been reported by Ivanov *et al.* (10) for U-235, $J = 7/2$, using bremsstrahlung radiation later contradicted by Zhucko *et al.* (7) and Geraldo (11) for Np-237, $J = 5/2$, using capture gamma rays at energies near threshold.

The main purpose of the present work is to measure the angular distribution of the Pu-239 photofission fragments by using *monochromatic* thermal neutron capture gamma rays, high-resolution gamma rays at two discrete energies:

(*i*) at very low energy (5.43 MeV) near the fission threshold, where the chances of observing anisotropies would be high (1). Then to compare our results with the results reported by Soldatov *et al.* (9) and Rabotnov *et al.* (2),

(*ii*) at a moderately high energy (7.35 MeV) so as to compare our results with the results reported by Baerg *et al.* (12), Soldatov *et al.* (9), and Rabotnov *et al.* (2). They have discovered isotropies near the energy of 8 MeV.

We have not found in the literature any report of an experiment of this kind using thermal neutron capture gamma rays.

TABLE 1. Energies and relative intensities of the most important gamma rays of the targets used in this work

Target	Gamma line energies (MeV)[a]	Relative intensities[a]	Photofission cross-sections (mb)[b]
Sulphur	5.43	59.08	8.9
	5.58	1.25	8.9
	7.80	3.91	43.0
	8.64	2.61	60.0
Lead	6.74	5.04	20.8
	7.37	94.06	34.5

[a] Taken from ref. 14.
[b] Taken from ref. 17.

Experimental methods

The gamma radiation used in the present work was produced by thermal neutron capture reactions in the sulphur (S) and lead (Pb) targets placed near the core of the IEA-R1 (Instituto de Energia Atomica-Reator 1), a 2 MW swimming pool type reactor. The experimental apparatus used to produce this monochromatic gamma radiation was mounted inside a radial beam and is described in detail elsewhere (11, 13). The principal gamma ray and the most important secondary gamma rays of the S and Pb targets, whose line widths are of the order of only a few electronvolts, are listed in Table 1, with their relative intensities (14). The total gamma flux incident on the samples were $\phi_S = 1.8 \times 10^{11}$ γ rays cm^{-2} for S, and $\phi_{Pb} = 2.6 \times 10^{11}$ γ rays cm^{-2} for Pb.

The fission chamber assembly (11) consists of two independent cylindrical aluminium chambers. It was aligned with the beam hole and positioned at the opposite side to the target element, outside the reactor concrete shield. In this assembly each chamber contained two samples discs joined back to back and suspended by a metallic rod. The discs in the two chambers were rotated at approximately 60 rpm during irradiation by an external rotating device in order to avoid the corrections arising from the flat geometry of the samples. The chambers were evacuated to approximately 70 mTorr (1 Torr = 133.3 Pa) to ensure that the fission fragments reached the detector. Inside each of these chambers, another cylindrical aluminium tube of 30 cm diameter was placed each with 16 holes of 2 cm diameter at different angles to the sample. These dimensions gave the best combination of fragment intensity at the detector position (11) and the smallest solid angle. The fis-

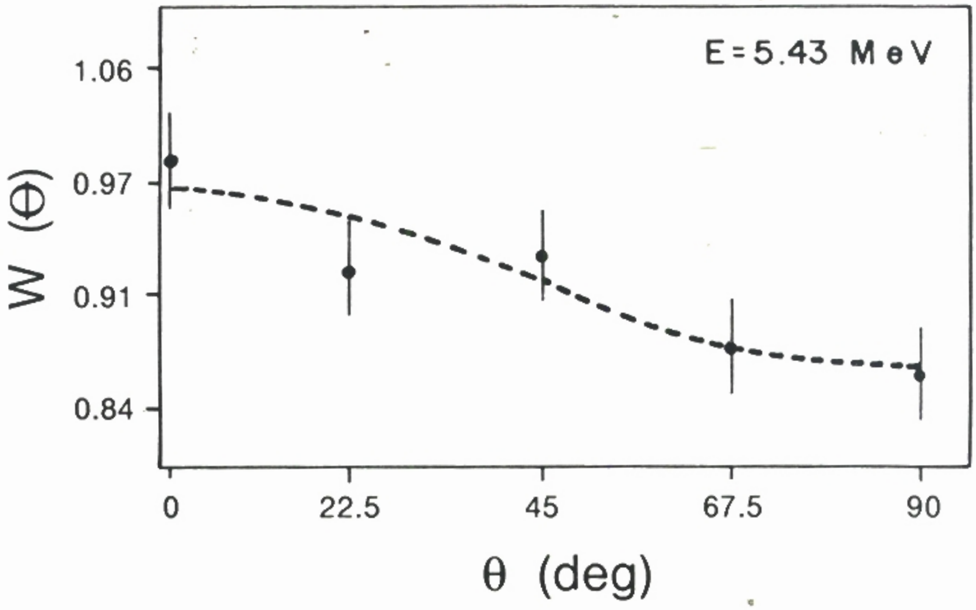

FIG. 1. Observed photofission fragment angular distributions for Pu-239 at 5.43 MeV (sulphur target).

sion fragment detectors were attached to the outside wall of this inner tube, covering the area of each hole. Two fission chambers as opposed to a single one were used in order to obtain the lowest total statistical error in the measurement. In addition, this allowed two independent and simultaneous sets of data, which gave a better handle on the systematic uncertainties.

The Makrofol Kg (8 μm) track detector foils were used to record the fission fragments. These foils after being irradiated were etched in a KOH solution (35% by weight) at 60°C for 30 min and the tracks were counted by means of an automatic sparking chamber (13).

The Pu-239 samples were supplied by the International Atomic Energy Agency (IAEA), and contained 51.4 mg of Pu-239 deposited on four titanium disks, each with an active diameter of 40 mm. The isotopic analysis of the sample are Pu-238 (0.01%), Pu-239 (99.01%), and Pu-240 (0.98%) (13).

Results and analysis

The contribution of the secondary gamma rays to the lead target was taken into account by using the weighted mean energy of the two gamma rays, this was done by considering the relative intensities (14) and the photofission cross sections at these energies, which are shown in Table 1, in a procedure similar to that used by Carvalho *et al.* (15), Manfredini *et al.* (16), and Geraldo (11).

The values of the Pu-239 photofission cross sections used in this work were measured separately by also using neutron capture gamma rays in several targets, in the energy range from 5 to 10 MeV. These results have been previously published in ref. 17 and are shown in Table 1 (these cross sections are already corrected for the secondary-gamma-ray contributions of the all targets, as described in detail in ref. 17).

The photofission cross section values considered for the Pb target were obtained with the targets of Ti (6.73 MeV) and Pb (7.38 MeV) (17). In this case, the cross sections considered were measured with the natural line width of the thermal capture gamma ray, respectively. The mean energy of this target, obtained in this way, was 7.35 MeV.

The contributions of the secondary gamma rays to the sulphur target were taken into account using a different procedure. The gamma energies and their respective intensities are shown in Table 1. Because the energies of the lines are distant, the contribution relative to energies of 7.8 and 8.6 MeV were grouped together and also weighted by the relative intensities and the photofission cross section. We have not measured the Pu-239 photofission cross sections specifically at these energies. The values considered were obtained from ref. 17 and represent average cross sections obtained at nearby energies by adjusting the best curve to the cross sections measured with the targets: Al (7.72 MeV), Zn (7.88 MeV), and Ni (9.00 MeV). The mean energy obtained is 7.93 MeV. These values are given in Table 1. The energies of 5.4 and 5.6 MeV were also grouped following the same procedure described before. The mean energy obtained was 5.43 MeV.

By taking into account these considerations, the total angular distribution obtained was attributed to the mean energies of E_1 = 5.43 MeV and E_2 = 7.93 MeV for the S target. In this way, the percentage of the total photofission reactions generated at low energy (5.43 MeV) is 62%, while at high energy (7.93 MeV) it is 38%. It is the relative magnitude estimated for this correction.

The angular distribution in high energy (above 7.3 MeV) was considered isotropic (2, 12, 18), and this relative contribution was subtracted from the total angular distribution obtained. After doing this subtraction, the corresponding angular distribution was considered relative to the mean energy of 5.43 MeV of the sulphur target.

The (γ, f) contribution relative to the gamma contribution from the structural aluminium of the reactor constitutes an undesirable background, and its correction was made by subtracting the angular distribution obtained without the target. This distribution obtained was practically isotropic, and the (n, f) contributions were found to be negligible (13).

The neutron flux was monitored by a "self-powered" detector (13).

The number of tracks at each corresponding angle were found to be nearly the same in the two fission chambers within the limits of experimental error and they were added together for

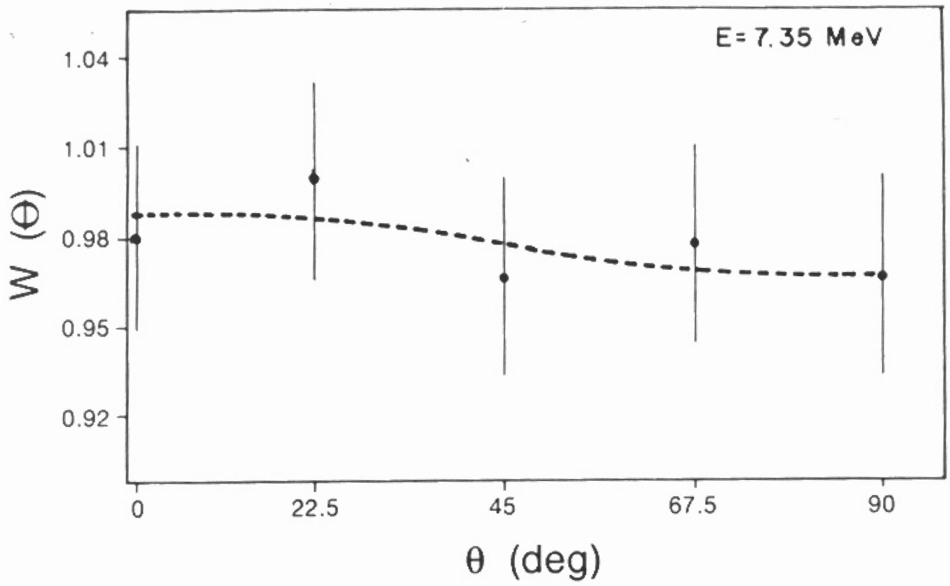

FIG. 2. Observed photofission fragment angular distributions for Pu-239 at 7.35 MeV (lead target).

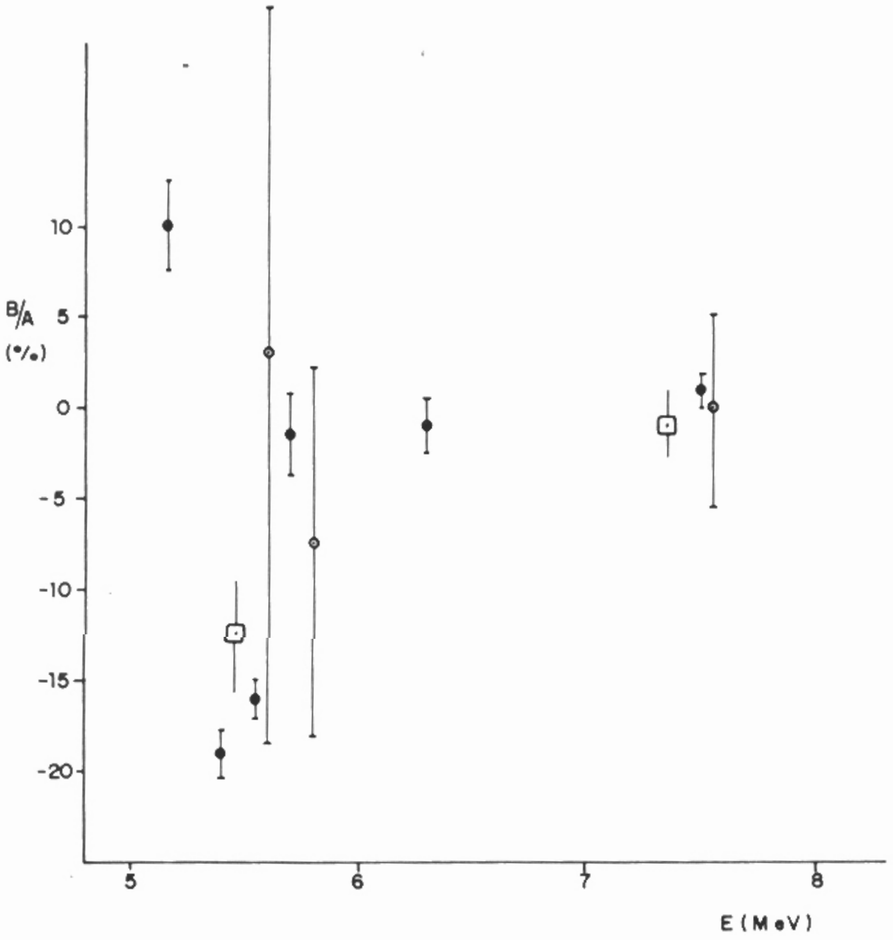

FIG. 3. Energy dependence of *B*/*A* for photofission of Pu-239. □, this work; ○, ref. 12; and ●, ref. 9.

the final analysis. The results of the angular distributions are presented in Figs. 1 and 2. The experimental points, shown with error bars were normalized at 0°. The errors quoted are standard deviations based on the reproducibility and counting statistics. The errors in the relative intensity values of the gamma-ray lines and in the photofission cross sections were not included in any error analysis.

The broken-line curve is the weighted least-squares fit of the

TABLE 2. Values of the coefficients A and B obtained by fitting the experimental data to the function $W(\theta) = A + B \sin^2\theta$

Coefficients	5.43 MeV	7.35 MeV
A	0.980 ± 0.022	0.990 ± 0.010
B	-0.120 ± 0.035	-0.010 ± 0.020
B/A	-0.122 ± 0.036	-0.010 ± 0.020

experimental data to the function $W(\theta) = A + B \sin^2\theta$. The coefficients A and B are normalized such that

$$\int_0^\pi W(\theta) \sin(\theta)\, d\theta = 1$$

and are shown in the Table 2. The angular anisotropies defined as B/A are also shown in this table and are compared with the results of other authors in Fig. 3.

The almost isotropic angular distribution observed at 7.35 MeV is in good agreement with the results of Rabotnov *et al.* (2) at 7 MeV and Baerg *et al.* (12) at 8 MeV.

The anisotropy obtained at 5.43 MeV ($B/A = -0.122 \pm 0.036$) is in reasonable agreement with the results reported by Soldatov *et al.* (9) and Rabotnov *et al.* (2).

As mentioned by Griffin (8), absorption of a dipole gamma quantum by a Pu-239 nucleus results in the formation of three degenerate compound states ($I, \pm M$) = (3/2, ± 3/2); (3/2 ± 1/2); (1/2, ± 1/2) with probabilities 1/2, 1/6, and 1/3, respectively.

According to Rabotnov (2) the angular distribution of the fragments of Pu-239 may be represented by the following formula:

$$[1] \quad W(\theta) = \frac{-1}{3}(2 - X) + \frac{1}{4}(2X - 1)\sin^2\theta$$

where X is the probability for fission with a given value of K (open fission channels (1)).

In the case of Pu-239 there are only two possible values of K: 1/2 and 3/2. In this way, if X denotes the probability for fission via $K = 1/2$, then $(1 - X)$ denotes the probability for fission via $K = 3/2$. By comparing the expression [1] with our result of the angular anisotropy at 5.43 MeV, we conclude that the probability of fission via $K = 1/2$ is 37% and via $K = 3/2$ is 63%.

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