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THE ^{232}Th(γ, n)^{231}Th CROSS SECTION NEAR THRESHOLD

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Abstract: The partial photoneutron cross section of ^{232}Th was measured by activation methods using monochromatic γ-rays of energies from 5.43 to 10.83 MeV. The measured cross section shows structure similar to the one observed previously in the photofission cross section. The implications of these observations on the photoabsorption structure of ^{232}Th are discussed.

E NUCLEAR REACTIONS $^{232}_{90}$Th(γ, n), $^{232}_{90}$Th(γ, f), $^{238}_{92}$U(γ, n), $^{238}_{92}$U(γ, f), $E = 5.43$–10.83 MeV, measured $\sigma(\gamma, n)$, $\sigma(\gamma, f)$.

1. Introduction

Photofission experiments near threshold can provide particularly valuable information on the properties of fission channels because in photoabsorption the electric dipole contribution is dominant, although electric quadrupole and magnetic dipole can also contribute.

In order to get this type of information, a number of experiments has been performed using bremsstrahlung [1, 2]), monochromatic γ-rays from proton capture reactions [3]), monochromatic γ-rays from neutron capture reactions [4, 5]), and Compton scattered γ-rays [6, 7]) from the reaction ^{58}Ni(n, γ)^{59}Ni. Since these sources have different resolutions, one must be careful in comparing their results.

In the case of ^{238}U and ^{232}Th the measured cross sections of all the authors show the presence of an intermediate structure, which indicates that the density of states available for energy dissipation in fission near threshold is considerably smaller than the total density of states. This can be understood by associating the vibrational states with the second well of an octupole deformation (fig. 1) as discussed by Albertson *et al.* [8]).

Since these resonances are associated with entrance channels the measurement of the total photoabsorption cross section is very important. Photoneutron emission is practically the only process competing with fission since $\sigma_{\gamma\gamma}$ is very small just above the fission threshold. Consequently the measurement of the (γ, n) cross section is important in helping to understand the intermediate structure observed in photofission cross sections.

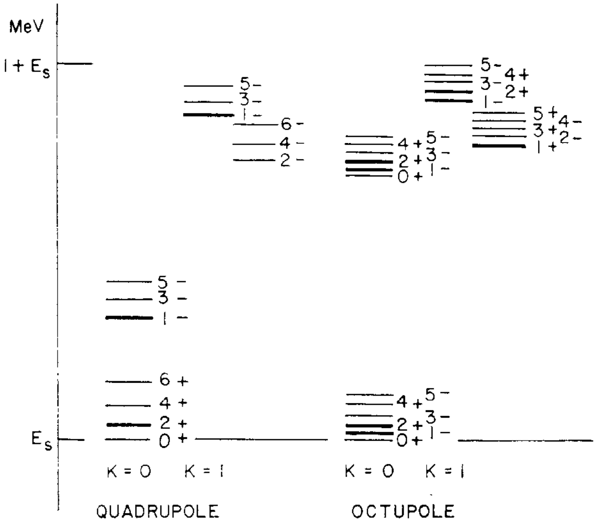

Fig. 1. Energy levels of stable deformation.

In a previous paper [5]) we have established that the photoneutron and photofission cross sections in ^{238}U and ^{232}Th have similar structures. In that experiment, however, the total photoneutron emission cross section was measured using a long counter and the photoneutron cross sections were extracted from it assuming the average number of neutrons per fission, ν, to be 2.5. This hypothesis was quite a strong one, since ν could in principle change rapidly with the increasing γ-energies.

In order to clarify this point we have measured the (γ, n) cross section [9]) in the case of ^{238}U and ^{232}Th using activation methods. In addition to that, a reanalysis of our previous data is also presented based on new results [10]) on the known variation of the number of neutrons emitted in photofission as a function of the excitation energy.

The results are presented in this paper and support our previous results presented in ref. [5]).

2. Experimental arrangement

The γ-radiation employed was produced in several elements used as targets and placed near the IEA-R1 2 MW reactor core. This experimental arrangement produces monochromatic γ-radiation from 5.43 to 10.83 MeV, and has been described in detail elsewhere [5]). The targets and respective energies and the flux obtained at the irradiation position are listed in table 1.

The thorium was employed in the ThO$_2$ form and before irradiation was radiochemically treated in order to clean the samples from their descendents. The thorium was then transformed into pellets; for each measurement two pellets were used; one was irradiated and the other was used to subtract the background. The irradiated pellet was shielded with paraffin mixed with boric acid and wrapped in a cadmium foil in order to minimize the contribution of (n, 2n), (n, γ) and (n, f) reactions in ^{232}Th. The only significant disturbing effect was due to fast neutrons through the $^{232}Th(n, 2n)^{231}Th$ reaction. For this reason the neutron flux above 6.34 MeV was monitored in all cases with pure aluminum foils through the $^{27}Al(n, \alpha)^{24}Na$ reaction and its contribution calculated and subtracted. The fast neutron flux has to be measured in each irradiation because different (n, γ) targets produce different neutron flux depression and scattering.

At the irradiation position that can be seen in fig. 2 the γ-flux was of the order of 10^8 γ/cm^2 · min.

TABLE 1

Targets employed, principal γ-ray energies and flux incident on the samples

Target	Energy (MeV)	ϕ (10^8 γ/cm^2 · min)
^{89}Y	6.07	2.5 ± 0.2
^{40}Ca	6.42	1.7 ± 0.2
^{48}Ti	6.73	4.4 ± 0.3
^{9}Be	6.83	1.1 ± 0.1
^{55}Mn	7.23	2.2 ± 0.2
^{207}Pb	7.38	3.2 ± 0.3
^{56}Fe	7.64	5.5 ± 0.6
^{27}Al	7.73	3.2 ± 0.2
^{64}Zn	7.88	$(9.7 \pm 0.9) \times 10^{-1}$
^{63}Cu	7.91	2.4 ± 0.2
^{58}Ni	9.00	2.5 ± 0.2
^{53}Cr	9.72	$(7.7 \pm 0.8) \times 10^{-2}$
^{14}N	10.83	$(1.7 \pm 0.2) \times 10^{-1}$

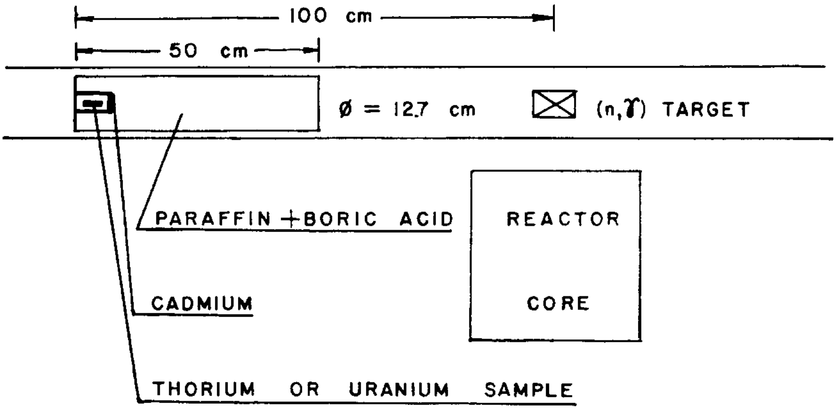

Fig. 2. Experimental arrangement for samples irradiation.

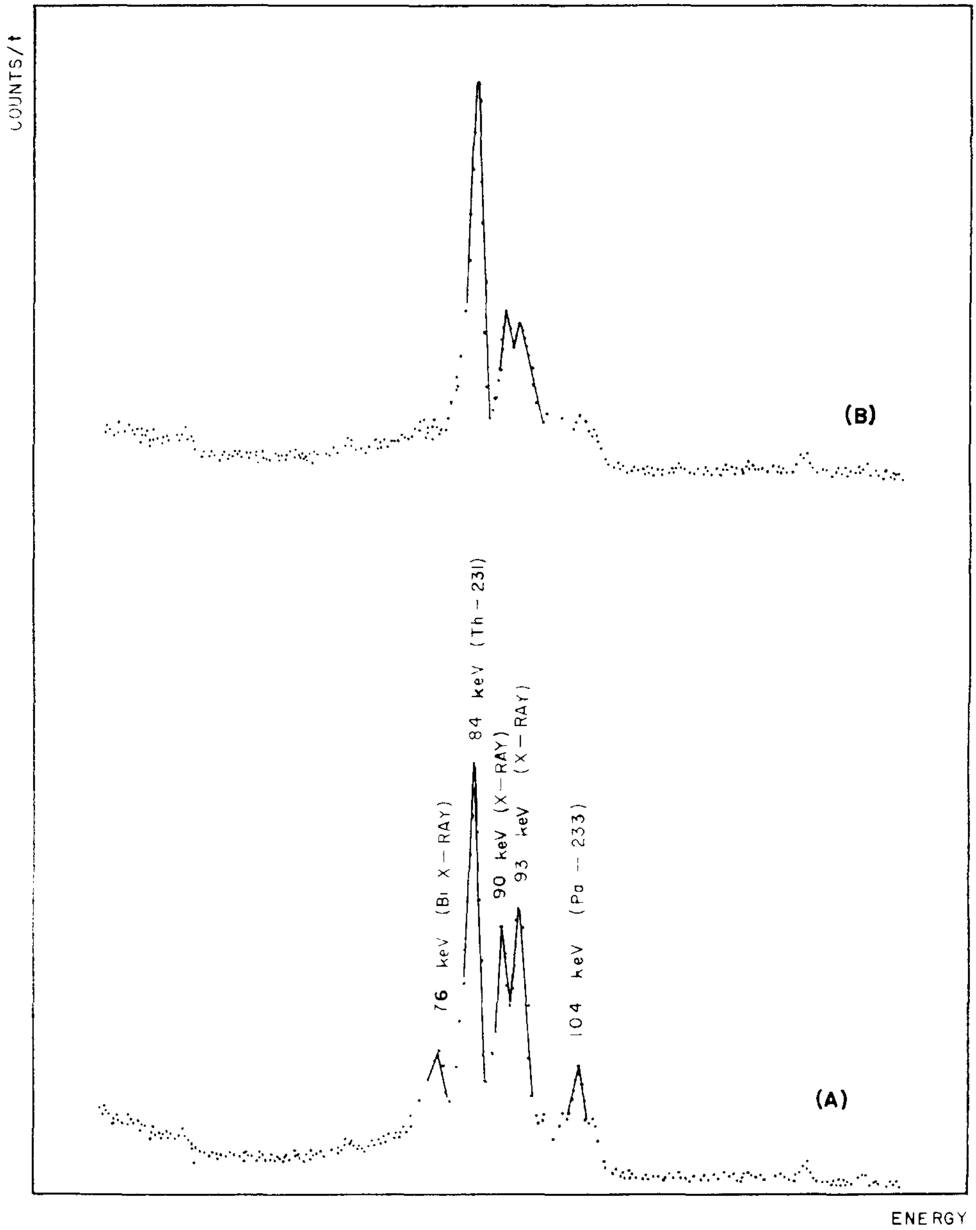

Fig. 3. Thorium spectrum before and after subtracting the background.

After the irradiation the γ-spectrum of the thorium was measured with a 18.2 cm^3 Ge(Li) detector. A typical spectrum of an irradiated sample with the lines identification is shown in fig. 3A. The background was subtracted using a non-irradiated sample and the final resulting spectra is shown in fig. 3B.

TABLE 2

^{231}Th lines, half-life and relative intensity

$T_{\frac{1}{2}}$	E_γ (MeV)	Relative yield
25.52 h	0.026	2 %
	0.084	10 %

The γ-lines, half-life and relative intensity of ^{231}Th are shown in table 2.

The activity of the ^{231}Th was measured by the 84 keV line after waiting 16 h in order to allow the ^{233}Th that can be formed, to decay. This isotope emits a 87 keV γ-ray with a half-life of 22.12 min.

The fission products eventually formed could be eliminated after the irradiation by making a new radiochemical treatment, but this would bring problems related to the loss of material during the processing, geometry variations for the various samples and weight variations due to different drying conditions.

With the Ge(Li) detector the ^{231}Th lines can be very well identified. For each irradiated sample the counts under the photopeak were integrated and all the corrections for irradiation time and decay were made.

The γ-flux was measured outside the beam hole by a 7.6 cm × 7.6 cm NaI(Tl) crystal during each irradiation. After the corrections for absorption and geometry, the γ-flux was found to have the values of table 1.

3. Results

Since the neutron capture targets employed emit also some secondary lines, although with low relative intensity with respect to the principal line, it is necessary to correct their contribution, solving a linear equation system of the type

$$\sum_i r_i \sigma_i = \sigma_{\text{exp}},$$

where r_i are the relative intensities, σ_i the cross sections for each line and σ_{exp} the experimental values.

Using a computer program for successive approximations it was possible to obtain the cross sections at each energy.

The data obtained by activation for ^{232}Th, and also for ^{238}U in a previous paper ^{11}), are shown in table 3. The errors include those arising from statistics and calibrations. These data are not absolute, they have been normalised since it was very difficult to get the overall efficiency.

Using the equations presented by Caldwell ^{10}) for the variation of the average number of neutrons as a function of the excitation energy:

$$\bar{\nu}_p = 1.619 + 0.1240\, E_\gamma \quad \text{for } ^{238}\text{U},$$

$$\bar{\nu}_p = 1.595 + 0.0449\, E_\gamma \quad \text{for } ^{232}\text{Th},$$

we have recalculated the photoneutron cross section for ^{238}U and ^{232}Th measured before through the total photoneutron emission cross section and the results are in table 4. With this reanalysis the (γ, n) cross sections have not changed drastically and the new values agree with the previous ones ^{5}) if we take into account the experimental errors.

TABLE 3

Principal γ-ray energies and thorium and uranium photoneutron cross section measured by activation

E (MeV)	$\sigma_{\gamma,n}$ (mb)	
	^{238}U	^{232}Th
6.07	18.0 ±4.8	3.4 ± 0.9
6.42	2.4 ±0.8	1.4 ± 0.4
6.73	22.7 ±1.4	27.8 ± 1.7
6.83	2.35±0.09	1.38± 0.05
7.23	1.3 ±0.7	0 ± 4
7.38	14.5 ±0.2	16.1 ± 0.2
7.64	13.3 ±4.4	10.4 ± 3.4
7.73	15.3 ±2.8	13.0 ± 2.4
7.88	23.6 ±5.0	18.0 ± 3.8
7.91		18.0 ± 3.9
9.00		28.6 ± 8.5
9.72		53.4 ±14.8
10.83		158.3 ±78.8

TABLE 4

Photoneutron cross section of ^{238}U and ^{232}Th recalculated using Caldwell's results for $\bar{\nu}_p$

E (MeV)	$\sigma_{\gamma,n}$ (mb)	
	^{238}U	^{232}Th
6.07	10.6± 3.2	
6.42	2.4± 1.2	6.7± 2.6
6.73	23.2± 6.4	30.5± 4.8
6.83	3.8± 1.2	
7.23	6.3± 3.9	6.3± 3.0
7.38	21.9± 5.4	17.7± 2.9
7.64	21.9± 6.9	24.8± 6.1
7.73	18.8± 4.1	21.4± 4.3
7.88	25.4± 6.4	25.9± 4.6
7.91		
9.00	85.1±23.2	73.8±17.4
9.72		
10.83		

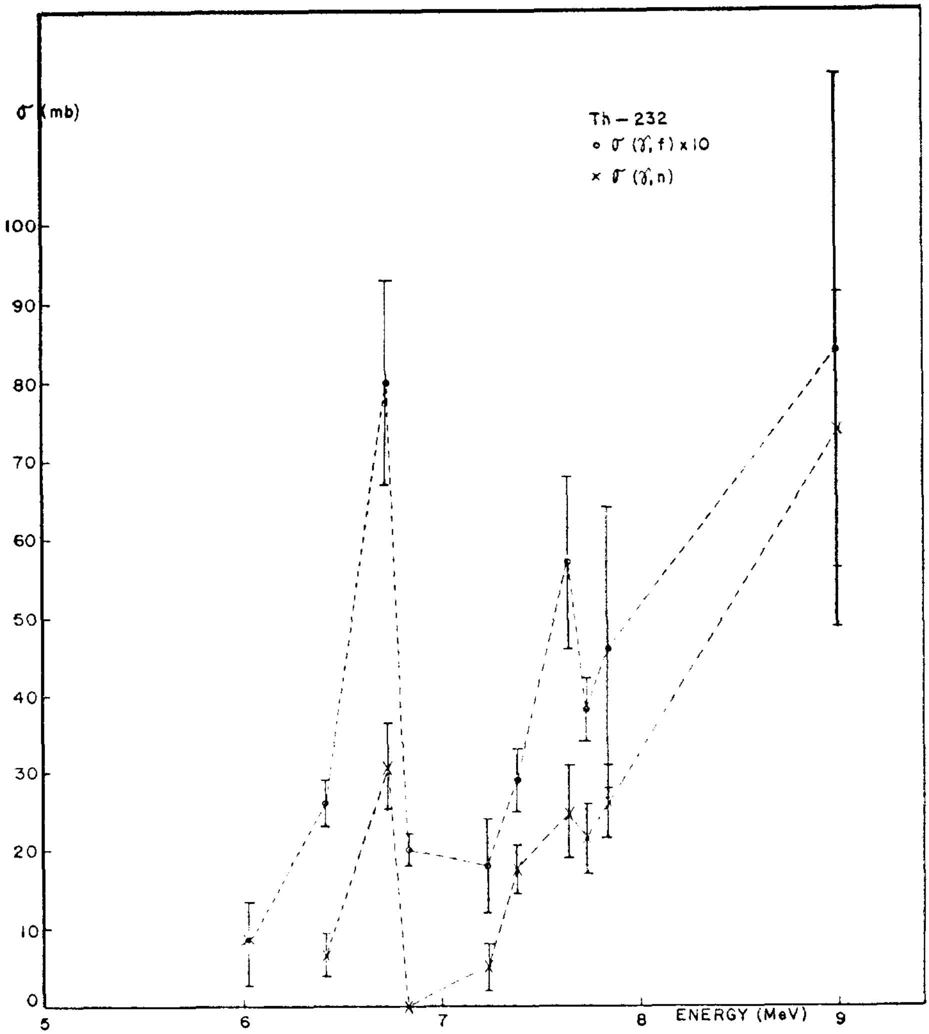

Fig. 4. Comparison of the photofission and photoneutron cross sections of ^{232}Th.

In fig. 4 we can see a comparison of the photofission and photoneutron cross sections in ^{232}Th. As in the case of ^{238}U the same structure is present in both cross sections.

Since the energies we are working with are below the (γ, 2n) threshold and the $\sigma_{\gamma\gamma}$ cross section is very small it is possible to calculated the $\sigma_{(\gamma,\mathrm{F})}/\sigma_{\gamma\mathrm{tot}}$ ratio as a function of the excitation energy for the two nuclei (fig. 5). For this purpose we have used the (γ, n) data from table 4. Our results for $\sigma_{\gamma,\mathrm{F}}/\sigma_{\gamma\mathrm{tot}} = \Gamma_\mathrm{F}/\Gamma_\mathrm{tot}$ were compared with the data of Veyssière ^{12}) obtained for excitation energies above 9 MeV. The agreement is good and one can notice a tendency of the ratio to increase as one approaches the threshold.

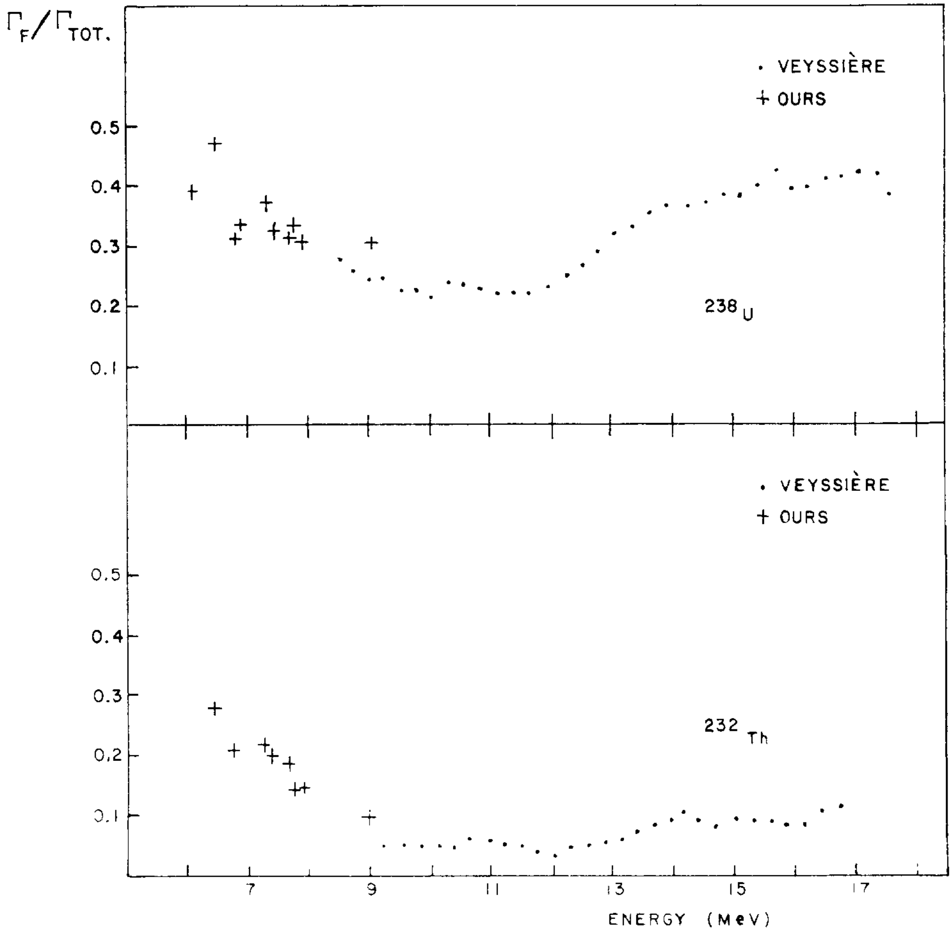

Fig. 5. Behaviour of the ratio Γ_F/Γ_{tot} as a function of the excitation energy E.

4. Discussion

The similarity of structure in both cross sections cannot be attributed to our experimental arrangement or to resonance effects due to the high resolution of the (n, γ) lines. The same experiment was repeated by Knowles ^{13}) using a long counter detector and Compton scattered γ-radiation. Although the results of Knowles do not agree in detail with ours, probably because the energy resolutions of the two experiments are quite different, the observation of a strong similarity between the (γ, n) and (γ, f) cross sections in ^{238}U has been also made by him.

The results obtained for the titanium line (6.73 MeV), which gives particularly high cross sections, have been checked a number of times. A high value of the (γ, f) cross section at this energy has been also found by Manfredini ^{4}) and so far as we can see there is no possibility that the γ-flux at the titanium point was underestimated because the γ-flux is measured in the same way for all the targets.

Since the cross section is small above the neutron separation energy ^{14}), our finding of the same structure in the (γ, n) and (γ, f) cross sections indicates that this structure is a characteristic of the photo-absorption cross section. However this conclusion conflicts with the information one has on the angular distribution of the photofission fragments $^{8, 15}$): these data show that it is possible to associate the peaks in the photofission cross section near threshold with the (1^-, 0), (1^-, 1) and (2^+, 0) saddle point states. If the same resonances are present in the photo-absorption and photofission cross section this fact implies that photo-absorption and fission fragments emission are not independent processes. The need to reconcile these two independent pieces of experimental data is the main result of this and our previous paper on the subject.

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