

**THE PHOTOFISSION CROSS SECTIONS OF URANIUM AND
THORIUM NEAR THRESHOLD**

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ABSTRACT

Using monochromatic gamma rays from (n,γ) reactions, the photofission cross sections behaviour for natural Uranium and Thorium have been determined in the 5 to 10 MeV energy interval.

"Bumps" around the energies of 6 MeV and 6.6 MeV for Uranium and around 6.7 MeV and 7.6 MeV for Thorium have been found. The existence of this structure in the photofission cross section near the threshold is in accordance with Bohr's prevision about the excitation of rotational states.

I - INTRODUCTION

The photodesintegration of a lot of elements has been studied with a variety of gamma ray energies. However, the research in photonuclear reactions has always been restricted by the gamma ray sources. It is not always possible to obtain monochromatic gamma radiation with enough intensity and resolution to permit significant experimental work.

The first experiments in photodesintegration had been performed with proton capture reactions (p,γ) . This kind of source is still utilized^{1,2} although it has only a few lines available and the resolution is about a hundred KeV.

The lack of monochromatic gamma rays has forced experimental workers for many years^{3,4} to use the Bremsstrahlung spectra from Betatrons, Synchrotrons and Linear accelerators. The continuous distribution of photon energies up to the maximum energy of the incident electrons is of course an unavoidable disadvantage. This was one of the reasons for the search of new gamma radiation

sources with better resolution in energy.

Recently the annihilation of positrons in flight⁵ and neutron capture gamma reactions (n,γ) ^{6,7} have been used bringing some advantages in relation to the primary techniques permitting the obtention of more precise data.

The annihilation of positrons in flight is used mainly for research in the giant resonance region, because its intensity is very low near the threshold where the cross sections are smaller. The neutron capture gamma rays have sufficient intensity to be used near the threshold of the photonuclear reactions and can be used only there because the energies from the (n,γ) reaction do not go beyond 11 MeV.

Recently Knowles et al.⁸ have used the Compton scattered-neutron capture gamma rays obtaining a practically continuous variation of the gamma rays energies around the known (n,γ) lines which can not be obtained with the direct neutron capture radiation, but the gamma rays intensities and resolution are lower than the neutron capture gamma rays.

A comparison of the resolution and intensity for various gamma rays sources is shown in tables I and II.

The nuclear fission of heavy elements following nuclear absorption of electromagnetic radiation was first predicted by Bohr and Wheeler in 1939⁹ and the first experimental measurement were made in 1942¹⁰ by Haxby using proton capture gamma rays in Uranium and Thorium targets.

The first measurements of photofission cross section curve for Uranium and Thorium near threshold found in the literature^{4,5,11} are those made with Bremsstrahlung spectra which are low precision experiments because of the complex calculations that had to be done in order to obtain the cross sections.

In 1966 Manfredini and co-workers¹² measured the photofis

sion cross section for Uranium in the 5 to 10 MeV energy interval using neutron capture gamma rays and nuclear emulsions as detectors. They found some structure in the cross section behaviour that could not be seen in the Bremsstrahlung results.

The existence of discrepancies about the photofission cross section near threshold led us to the measurement of the cross section curve trying to get more precise results using neutron capture gamma rays as source.

In the present paper we report the measurements of the photofission cross sections for Natural Uranium and Thorium and show that peaks really exist, although we can not precise exactly their width because of the few points that can be measured.

Table I

Source	E_{γ} (MeV)	ΔE (KeV)
Reaction		
$^{19}\text{F}(p,\alpha\gamma)$	7.12	130
$^7\text{Li}(p,\gamma)$	17.6	12.2
Betatron	19 (Maximum energy)	500
Positrons annihilation	19	500
Bremsstrahlung Monochromator	19 (Maximum energy)	100
Reaction		
$^{58}\text{Ni}(n,\gamma) ^{59}\text{Ni}$	9.00	0.011
$^{207}\text{Pb}(n,\gamma) ^{208}\text{Pb}$	7.38	0.0045
Compton Scattered Neutron Capture radiation	9.00	255

Comparison between widths of gamma radiation sources.

Table II

Source	E_{γ} (MeV)	Gamma flux in $\text{cm}^{-2} \text{sec}^{-1}$
Proton Current 1 mA		30 cm from the target 1×10^5
$^{19}\text{F}(p, \alpha\gamma)$	7.12	
$^7\text{Li}(p, \gamma)$	17.6	1×10^4
Betatron Maximum Energy 31 MeV	10	50 cm from the target 1×10^6
Positrons annihilation in flight	19	1×10
Reactor $\phi = 3.7310^{11} \text{n/cm}^2 \text{sec}$ 1500 g		400 cm from the target
$^{58}\text{Ni}(n, \gamma)^{59}\text{Ni}$ 1000 g	8.997	2.49×10^5
$^{207}\text{Pb}(n, \gamma)^{208}\text{Pb}$	7.38	1.6×10^4
Compton Scattered Neutron Capture radiation		
$^{58}\text{Ni}(n, \gamma) \text{ Ni}$	8.997	2×10^{-1}

Comparison between gamma ray fluxes from different sources.

II - EXPERIMENTAL SET-UP

The gamma rays used have its origin in the neutron in targets placed near the IEAR-1 reactor core in a tangential beam-hole. The thermal neutron flux incident in the target with the reactor power at 2 Mw is about $4 \times 10^{11} \text{n/cm}^2/\text{sec}$.

The monochromatic gamma radiation thus obtained is then collimated and filtered to prevent neutron contamination. The schematic diagram of the collimator can be seen in the fig. n^o 1. The gamma rays are detected by a NaI(Tl) crystal of 3 x 3 inch

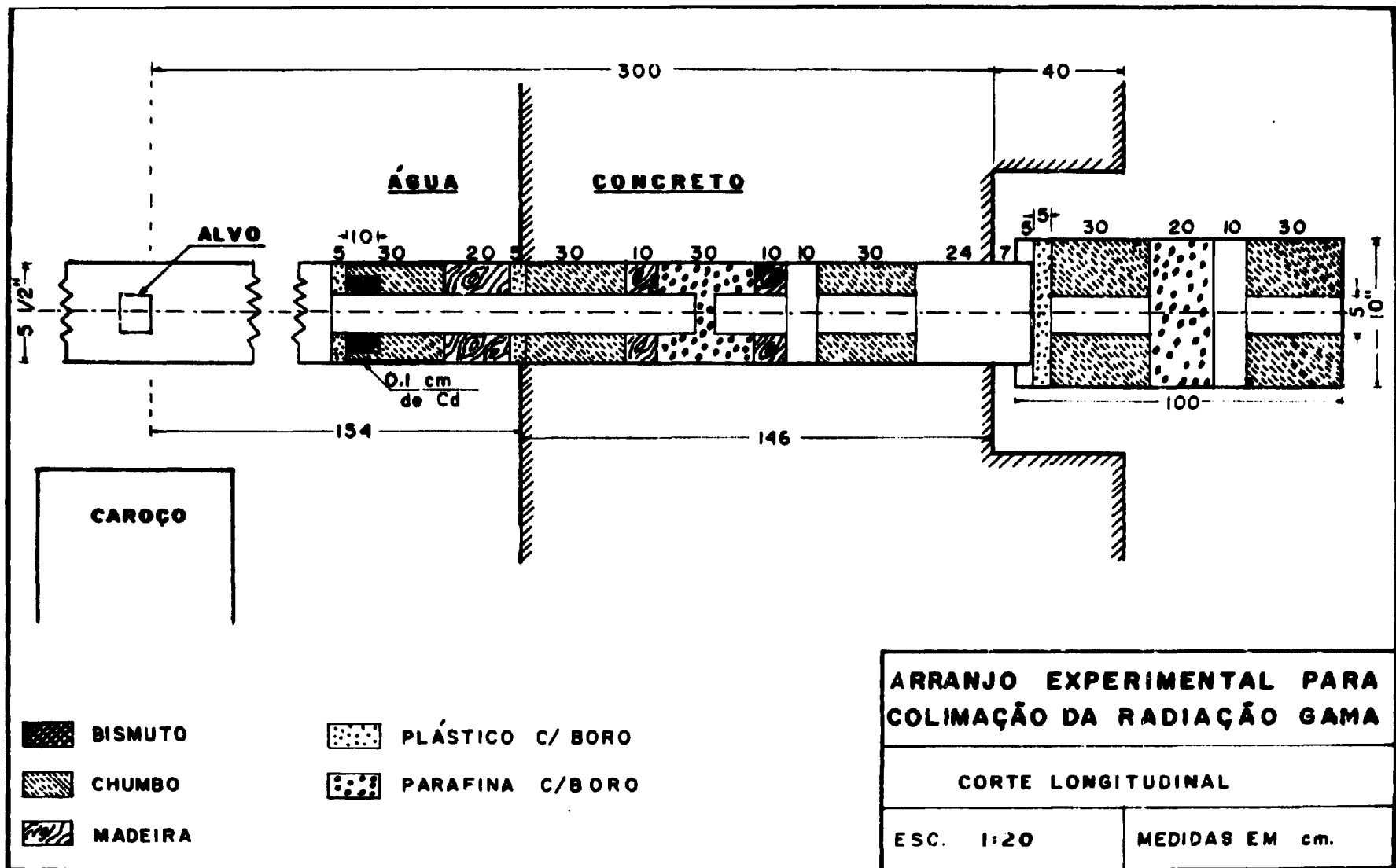


Figure 1

Schematic diagram of the collimator

and analysed by a 1024 multichannel analyser.

The detection of the fission fragments is made by ionization chambers¹³ of multiparallel plates in which Uranium and Thorium had been electrolytically deposited in an oxide form. The plates containing about 2.5 mg/cm² of UO₂ and 1.4 mg/cm² of ThO₂ are in number of 12 in each chamber, in order to increase the detector sensibility; it is known that the cross sections at these energies are of the order a few milibarns.

The pulses produced in the chamber by the fission fragments are larger than the pulses produced by the alpha particles emitted by the Uranium and Thorium so the discrimination is very easy to be done. The experimental arrangement is shown in fig. n° 2.

In order to prevent a variation caused by the reactor power fluctuations a BF₃ detector is used as a monitor. Moreover the measurements of the gamma rays intensity, fission counts in the chamber and background are alternated, having in view the possibility of reactor power variation.

III - EXPERIMENTAL DATA HANDLING

The gamma rays intensity can be obtained by integrating the counts under the photopeak assumed to be a gaussian curve and using the total efficiency for the NaI(Tl) 3" x 3" crystal

$$I \left(\frac{\text{gammas}}{\text{sec}} \right) = \frac{A}{K p (1 - e^{-\mu x})}$$

were:

A is the photopeak area in counts/sec;

p is the photofraction that can be calculated by the Monte Carlo method¹⁴ or measured experimentally¹⁵;

(1 - e^{-μx}) is the crystal intrinsic efficiency and

K is a correction factor because the intensity measured 2m away from the chamber position.

EXPERIMENTAL ARRANGEMENT

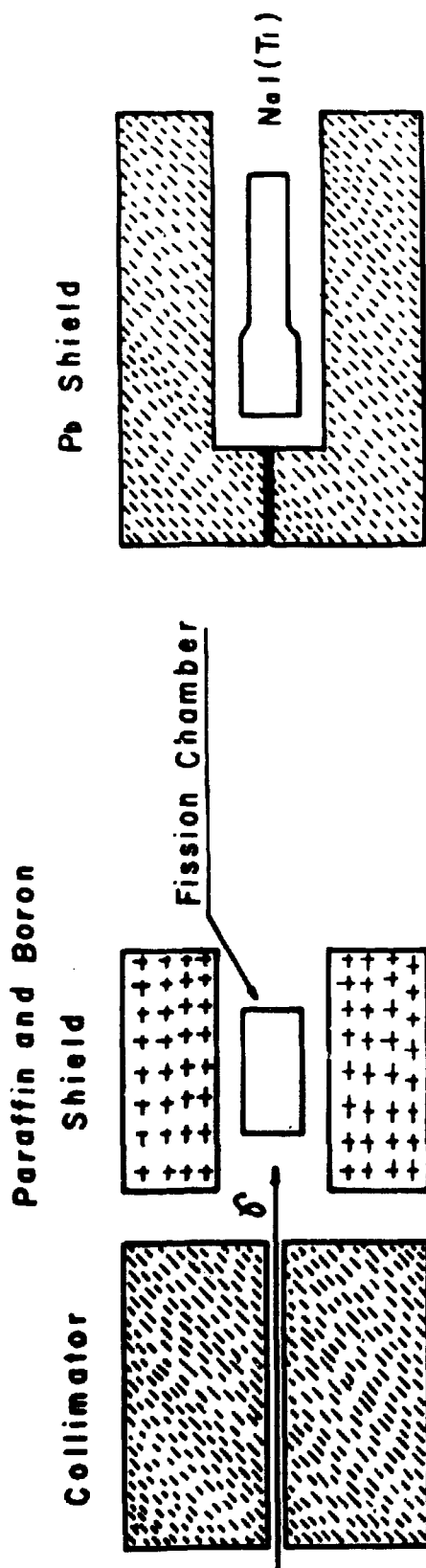


Fig. 2

Experimental arrangement

The mean gamma-ray intensity for the elements used as targets, the mean counting rate obtained with the two chambers and the statistical errors are calculated by a computer program named Carcarã and are given in table III.

The contribution due to neutron scattering in the target container as well as the room background have been measured and subtracted. The influence of neutrons scattered in the target have been calculated and also taken into account.

The gamma ray lines obtained for an element are not exactly monochromatic, but the relative intensities of the secondary to the principal lines are well known and not very high¹⁶.

For each target the counts measured in the fission chamber are due contributions from several gamma lines; however there is a more intense gamma line that is the main responsible for the counts.

If we had just one gamma ray line the photofission reaction rate in the fission chamber would be given by:

$$R \left(\frac{\text{reactions}}{\text{sec}} \right) = \frac{C}{\epsilon} = \phi \sigma NV = \phi \sigma N_t$$

where:

- C are the chamber counts per second
- ϵ is the chamber intrinsic efficiency
- ϕ is the incident gamma flux
- σ is the photofission cross section
- N_t is the number of Uranium and Thorium atoms.

$$\frac{C}{\epsilon N_t} = \frac{K A(E) \sigma(E)}{(1 - e^{-\mu(E)x}) p(E) S}$$

where S is the beam area.

$$\frac{\sigma(E)}{K'} = \frac{C (1 - e^{-\mu x}) p(E)}{A (E)} = \frac{C}{I}$$

with $K' = \frac{S}{\epsilon N_t K}$ and I the gamma ray intensity.

Since for each target there are more than one gamma line we get:

$$\frac{C}{I} = \sigma_1' + r_2 \sigma_2' + r_3 \sigma_3' + \dots \text{ where } \sigma_i'$$

are numbers proportional to the photofission cross section and r_i are the secondary lines intensity relative to the principal ones.

Table III

Elements	Uranium chamber counts/sec	Thorium chamber counts/sec	Gamma ray intensity γ/sec
Ti	7.54 ± 0.07	2.41 ± 0.02	(4.9 ± 0.5)10 ⁴
Al	0.97 ± 0.01	0.178 ± 0.002	(7.6 ± 0.8)10 ³
Ni	8.73 ± 0.09	0.944 ± 0.009	(1.54 ± 0.15)10 ⁴
Zn	1.12 ± 0.01	0.177 ± 0.002	(4.68 ± 0.48)10 ³
Cu	3.73 ± 0.04	0.673 ± 0.007	(1.71 ± 0.18)10 ⁴
Mn	2.66 ± 0.03	0.449 ± 0.004	(3.12 ± 0.37)10 ⁴
Fe	4.73 ± 0.05	0.909 ± 0.008	(2.9 ± 0.3)10 ⁴
K	—	0.0677 ± 0.0007	(2.31 ± 0.26)10 ⁴
S	0.236 ± 0.002	0.0306 ± 0.0003	(1.4 ± 0.1)10 ⁴
Ca	0.166 ± 0.002	0.0588 ± 0.0006	(4.4 ± 0.4)10 ³
Y	0.232 ± 0.004	0.00572 ± 0.00006	(2.2 ± 0.2)10 ³
Pb	0.130 ± 0.001	0.0153 ± 0.0002	(1.1 ± 0.1)10 ³
Be	0.0023 ± 0.001	0.00011 ± 0.00005	(1.1 ± 0.1)10 ²

This last equation can be written for all the targets and a linear system is obtained.

As some of the energies are not very different, approximations of the order of 50 KeV have been made to reduce the final

system.

To solve this system it was first taken the quadratic system made with the principal lines of the elements. The cross section obtained were then affected by errors corresponding to the lines that had not been considered earlier.

IV - RESULTS

The results with the errors obtained for Natural Uranium and Thorium are in table IV and fig. n° 3 and 4. The errors include statistical errors, uncertainties in the intensity measurement and the influence of the gamma ray lines that had not been considered.

The data obtained for Uranium had also been normalised to the results of Manfredini¹² showing a very good agreement (fig.5).

Looking at the cross section behaviour we see that peaks really exist near the energies of 6 MeV and 6.6 MeV for Uranium and 6.7 and 7.6 MeV for Thorium.

The distance between them is approximately 1 MeV and the width of the order of 300 KeV although this value is very rough because of the small number of experimental points available.

To show the importance of the gamma line width in the shape of the cross sections we have folded our experimental data with 1% and 5% resolution functions; it can be seen clearly that this last resolution function is sufficient to smear out the structure in the cross section (fig. n° 6). This can be an explanation why some authors¹⁷ found peaks that are poorly defined near these energies.

The existence of peaks is in accord with the Bohr¹⁸ ideas based on the liquid drop model.

According to Bohr theory the photofission in even nuclei close to the fission threshold should occur predominantly via a

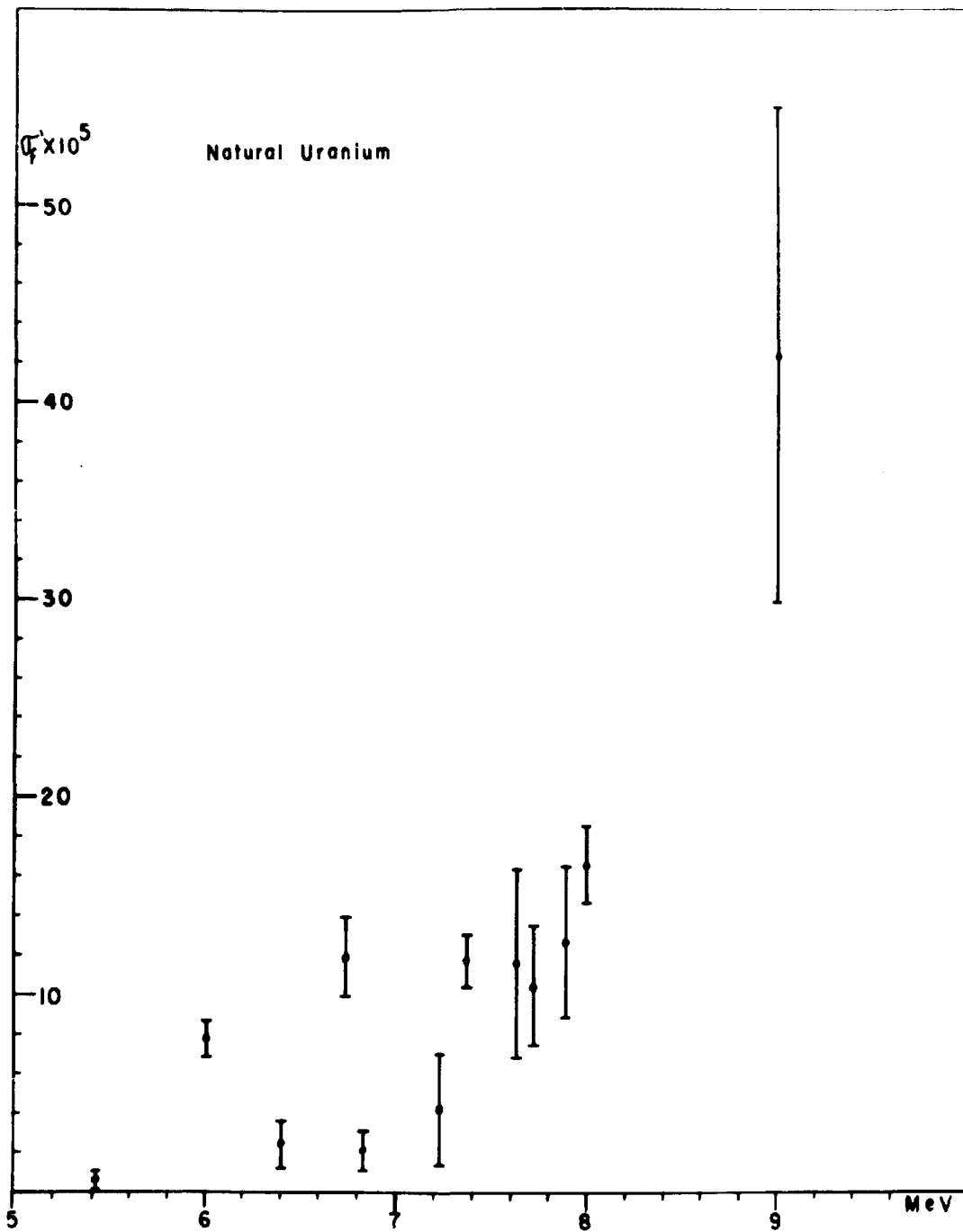


Fig. 3

Photofission cross section behaviour for Natural Uranium

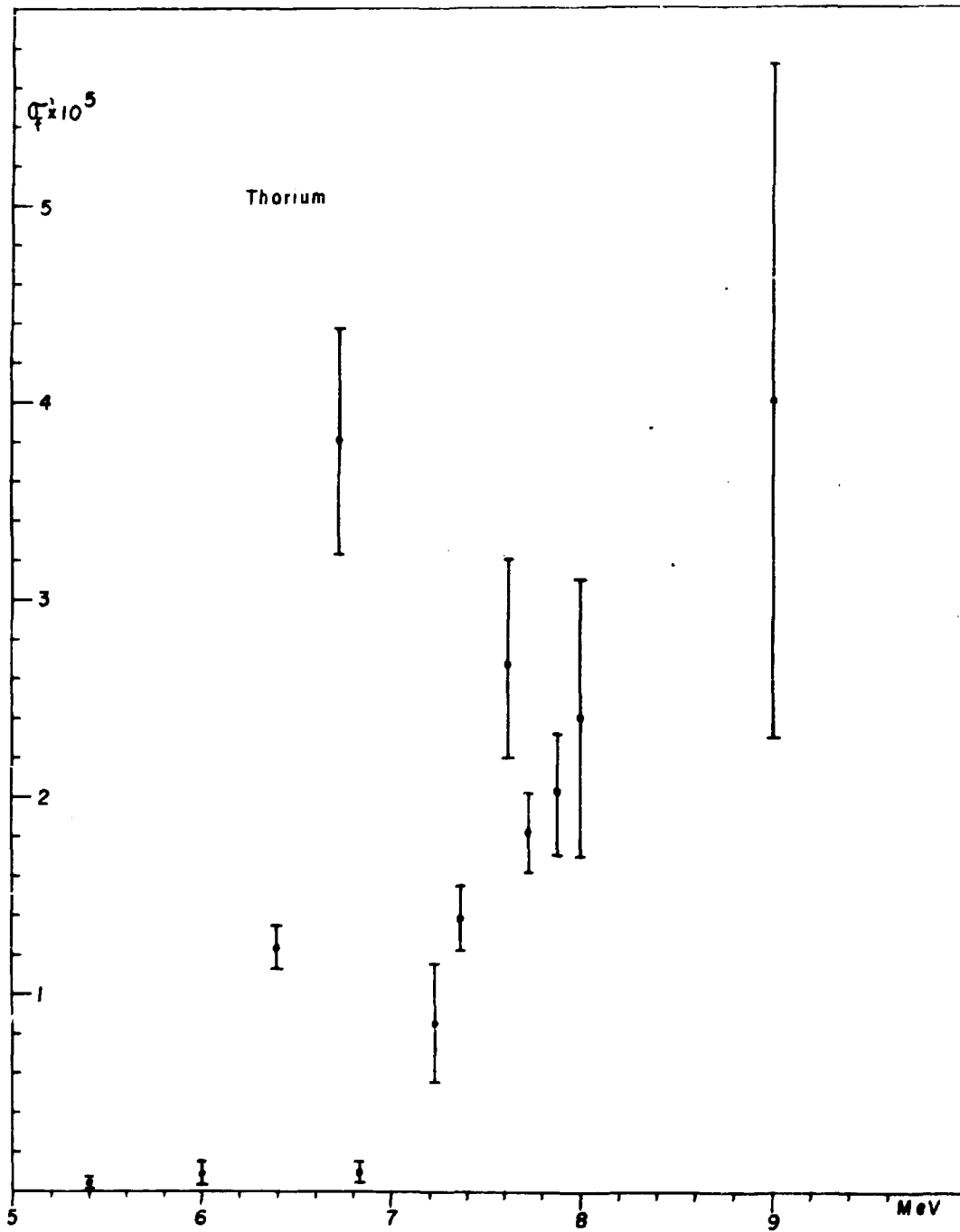


Fig. 4

Photofission cross section behaviour for Thorium

single or small number of well defined low lying excited states of the compound nucleus at the saddle point configuration. In the Bohr interpretation this excitation is an odd parity state (-1) similar to the observed low energy excitations of the ground state and represents a collective mode of excitation associate with as symmetric vibrations in the shape of the nuclear surface.

Table IV

Energy	$\sigma'_U (x 10^5)$	$\sigma'_{Th} (x 10^5)$
5.43	0.61 ± 0.49	0.05 ± 0.04
6.05	7.85 ± 0.86	0.09 ± 0.06
6.42	2.47 ± 1.20	1.23 ± 0.13
6.73	1.2 ± 2	3.8 ± 0.6
6.83	2.2 ± 1	0.1 ± 0.046
7.23	4.3 ± 2.8	0.86 ± 0.3
7.38	11.8 ± 1.3	1.4 ± 0.2
7.63	11.6 ± 5	2.7 ± 0.5
7.73	10.6 ± 3	1.8 ± 0.2
7.79	---	---
7.88	12.8 ± 4	2.2 ± 0.9
7.91	16.6 ± 1.8	2.4 ± 0.7
8.99	42.4 ± 13	4.0 ± 1.7

σ' are numbers proportional to the cross sections at the various energies.

From this we can expect that rotational states of the assymmetric liquid drop model ($K = 0, 1, 2, \dots$) with odd parity would be excited in photofission just above the threshold giving this cross section behaviour.

RESUMO

Utilizando radiação gama proveniente da reação (n, γ) foram medidos os comportamentos das secções de choque de fotofissão do Urânio Natural e Tório no intervalo de energias de 5 a 10 MeV.

Foram encontrados picos nas vizinhanças das energias de 6 MeV e 6.6 MeV para o Urânio e nas vizinhanças de 6.7 MeV e 7.6 MeV para o Tório. A existência desta estrutura na secção de choque de fotofissão junto ao limiar está de acôrdo com a previsão de Bohr sobre a excitação de estados rotacionais.

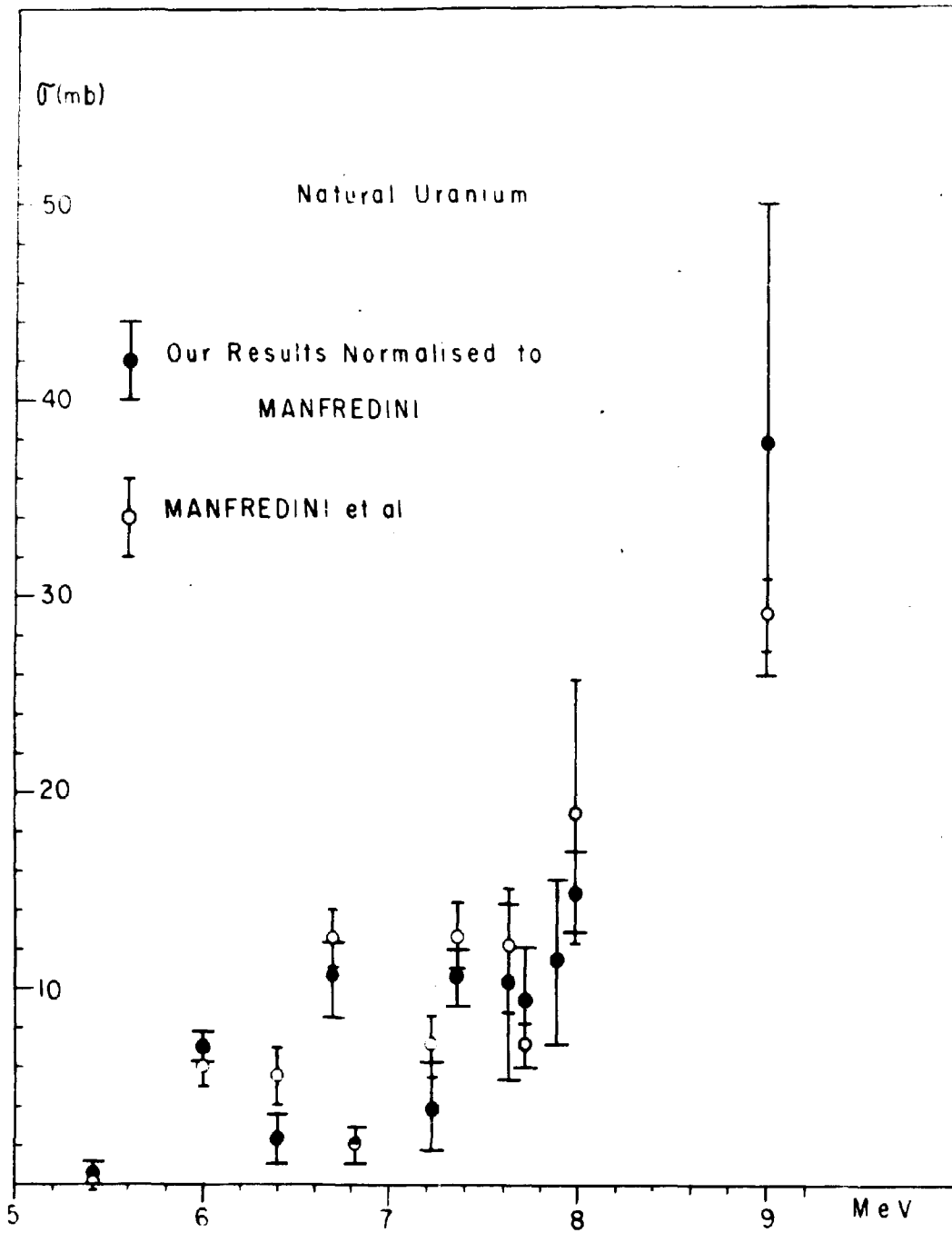


Fig. 5

Photofission cross section for Uranium
normalised to Manfredini's results

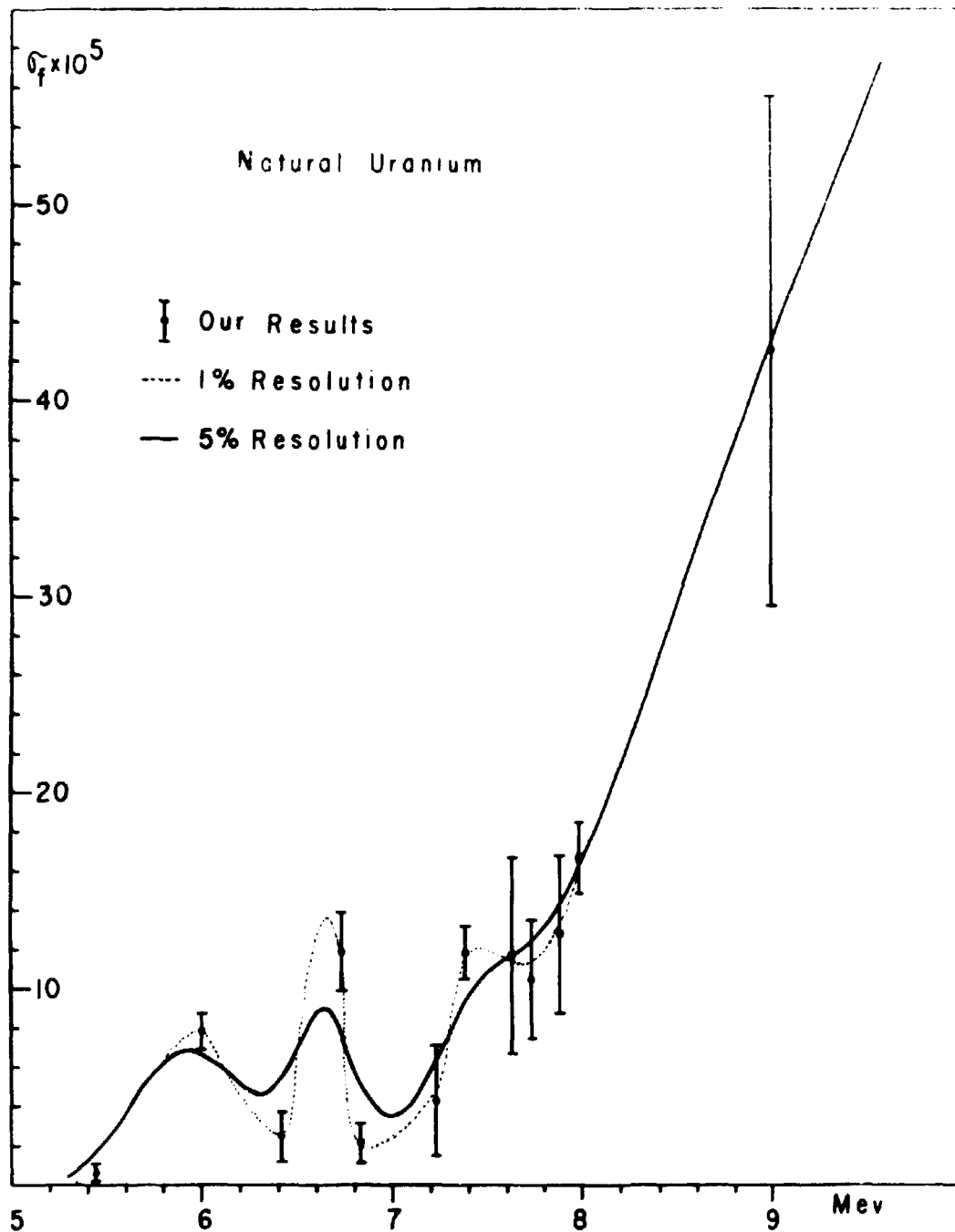


Fig. 6

Photofission cross sections folded with
1% and 5% resolution functions

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VI - REFERENCES

- 1 - K. Reibel and A.K. Mann, Phys. Rev. 118, 701 (1960)
- 2 - K.M. Clarke, ANL 5853 (1958)
- 3 - R.A. Schmitt and R.B. Duffield, Phys. Rev. 105, 1277 (1957)
- 4 - L. Katz, A.P. Baerg and F. Brown, Paper P/200 in Proceedings of the Second United Nations Conference on ... Peaceful Uses of Atomic Energy, 15, Geneva (1958)
- 5 - C.D. Bowman, G.F. Auchampaugh and S.C. Fultz, Phys. Rev., 133B, 676 (1964)
- 6 - O.Y. Mafra, Master Thesis - U.S. Paulo
- 7 - L. Jarczyk, H. Knoepfel, R. Muller and W. Wolfli, Nucl. Inst. and Meth. 13, 287 (1961)
- 8 - J.W. Knowles and N.M. Ahmed AECL 2535 (1966)

- 9 - N. Bohr and J.A. Wheeler, Phys. Rev. 56, 426 (1939)
- 10 - R.O. Haxby, W.E. Shoupp, W.E. Stephens and W.A. Wells, Phys. Rev. 59, 57 (1941)
- 11 - R.B. Duffield and J.R. Huizenga, Phys. Rev. 1042 (1953)
- 12 - A. Manfredini, Muchnik, L. Fiore, C. Ramorino, H.G. de Carvalho, R. Bosch and W. Wolfli, Il Nuovo Cimento.. XLIVB, n° 1 218 (1966)
- 13 - O.Y. Mafra and F.G. Bianchini, IEA 19
- 14 - H.R. Franzen, O.Y. Mafra and F.G. Bianchini, IEA 171 (1968)
- 15 - L. Jarczyk, H. Knoepfel, J. Lang, R. Muller and W. .. Wolfli, Nucl. Inst. and Meth. 17, 310 (1962)
- 16 - Nucl. Data Tables, Section A - vol. 5, n° 1-2 (1968)
n° 3 (1969)
- 17 - J.W. Knowles and A.M. Khan, Private Communication
- 18 - A. Bohr, Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, 151 (1955)

