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# Photonuclear Cross-Sections of $^{233}$U Using Neutron Capture Gamma-Rays, Near Threshold (*).

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**Summary.** — The photofission and photoneutron cross-sections of $^{233}$U have been measured by using monochromatic and discrete photons produced by thermal neutron capture gamma-rays in several targets placed at the core of the IEA-R1 reactor in the energy interval from 5.43 MeV to 9.72 MeV. The evaluation of the total photoabsorption cross-sections and the parameters: single fission barrier, relative fissionability, fission probability, neutron emission/fission competition, nuclear temperature are also presented.

PACS 25.20.Gf – Total photon cross-sections.
PACS 25.85.Jg – Photofission.

## 1. – Introduction.

Photonuclear studies for nuclei in the actinide region have been the purpose of several laboratories, during the last forty years due to the few data available for those nuclei in the literature. The main objective of these studies has been to obtain nuclear information at excitation energies in the region of the giant dipole resonance (GDR, between 10 and 20 MeV), and in the region of low energy, near the photofission and photoneutron thresholds ($(5 \div 10)$ MeV) [1].

Photonuclear experiments can be performed by using several kinds of gamma-ray sources, with resolutions ranging from a few eV to tens of keV [2]. With these markedly different energy resolutions, it is only possible to do a qualitative comparison between the results obtained with different gamma-ray sources. These comparisons are still useful because of the significance of such measurements and the lack of data available in the literature. However they are always questionable and only of qualitative interest.

(*) The authors of this paper have agreed to not receive the proofs for correction.

Our laboratory has carried out several studies of photonuclear reactions for some actinides nuclei using experimental data obtained with neutron capture gamma-rays in the low-energy region [3-8].

In the present work measurements of the photoneutron cross-sections for the $^{233}U$ nuclei, near threshold ((5.43 ÷ 9.72) MeV) are presented. The photofission data, previously reported in ref. [6], are also included for comparisons with the photoneutron results. These measurements were performed by using discrete and high-resolution photons produced by neutron capture. The evaluation of the total photoabsorption cross-sections $\sigma(\gamma, A)$ $(E)$ and the parameters $\sigma(\gamma, f)$ $(^{233}U)/\sigma(\gamma, f)$ $^{238}U$ (relative fissionability); single fission barrier; $\sigma(\gamma, f)/\sigma(\gamma, A)$ (photofission branching ratio or fission probability); $\Gamma_n/\Gamma_f$ (neutron emission-fission competition); nuclear temperature, are also presented. The correlation trends of the photofission branching ratio and the $\Gamma_n/\Gamma_f$ competition with the fissility parameter of the liquid drop model $Z^2/A$ were performed because these correlations studies are very important to verify the consistency of the experimental data as well as the consistency of the theoretical models usually employed in these studies.

The data reporting these measurements for $^{233}U$ published previously, independently of the type of gamma-ray source employed, are mainly concerned with photofission results:

*a*) in the low-energy region:

Huizenga *et al.* [9], Ostapenko *et al.* [10], Moraes *et al.* [6];

*b*) including the GDR region:

Katz *et al.* [11], Bermann *et al.* [12].

The unique photoneutron cross-sections data for $^{233}U$ reported in the literature was found in ref. [12].

## 2. – Experimental procedures.

The experimental apparatus used to produce the discrete and monochromatic gamma-rays, and the techniques employed for measuring the photofission and photoneutron cross-sections are well documented in previous publications [3, 4, 6, 7], and they will not be described here in any detail. Briefly, a collimated gamma radiation beam is produced by thermal neutron capture in several critically chosen targets placed near the core of the IEA-R1, a 2 MW, pool type research reactor. This beam passes through several filters to minimize the neutron beam contribution and after leaving the beam hole impinges on the sample under study.

The gamma fluxes incident on the sample were measured by means of a 3 in. ×3 in. NaI (Tl) crystal and are shown in table I.

The reactor powers was monitored by a self-powered detector located near the targets [7].

The photofission fragments were detected by the fission track registration technique in Makrofol KG (8 μm) [7].

Neutrons from the $(\gamma, f)$ and $(\gamma, n)$ reactions were simultaneously detected by a 4 $\pi$ «long-counter» consisting of 60 $^3He$ detectors conveniently distributed inside a moderator material (polyethene), where the neutrons are slowed down.

TABLE I. – *The targets, their principal* ($\gamma$) *line energy and fluxes used in this work.*

| Target | $E$ (MeV) | ($\gamma$/cm$^2$ s) |
|---|---|---|
| $^{32}$S | 5.43 | $(6.89 \pm 0.73) \cdot 10^4$ |
| $^{48}$Ti | 6.73 | $(2.89 \pm 0.32) \cdot 10^5$ |
| $^{55}$Mn | 7.23 | $(1.10 \pm 0.13) \cdot 10^5$ |
| $^{207}$Pb | 7.38 | $(1.49 \pm 0.16) \cdot 10^5$ |
| $^{56}$Fe | 7.64 | $(1.86 \pm 0.22) \cdot 10^5$ |
| $^{27}$Al | 7.72 | $(1.63 \pm 0.21) \cdot 10^5$ |
| $^{68}$Zn | 7.88 | $(1.17 \pm 0.13) \cdot 10^5$ |
| $^{64}$Cu | 7.91 | $(1.89 \pm 0.23) \cdot 10^5$ |
| $^{64}$Ni | 9.00 | $(1.74 \pm 0.20) \cdot 10^5$ |
| $^{52}$Cr | 9.72 | $(8.38 \pm 1.06) \cdot 10^4$ |

TABLE II. – *Masses of the* $^{233}$U *samples, including isotopic percentage and photofission* ($E_f$) *and photoneutron* ($B_f$) *thresholds.*

| Masses (mg) | Isotopic percentage (atoms/%) | $Z^2/A$ | $E_f' - B_n'$ | $E_f$(*) | $B_n$(**) |
|---|---|---|---|---|---|
| 13.9 | $^{233}$U: 99.702 | 36.33 | − 0.75 | 5.7 | 5.753 |
| 13.0 | $^{234}$U: 0.236 | — | — | — | — |
| 12.8 | $^{235}$U: 0.012 | — | — | — | — |
| 12.7 | $^{238}$U: 0.050 | — | — | — | — |
| Total: 52.4 ± 2% | | | | | |

(*) Taken from ref. [16].
(**) Taken from ref. [25].

The efficiency of the system was determined by using a calibrated $^{252}$Cf source: $E_n = (38 \pm 1)\%$ [7].

The values of the prompt neutron multiplicities $\nu(E)$ (the average number of prompt neutrons per fission) necessary to obtain the photoneutron cross-sections $\sigma(\gamma, n)$ were extracted from ref. [12].

In order to subtract the background contribution in the photofission and photoneutron measurements caused by gamma-rays coming from the reactor core (mainly aluminium capture gamma-rays) a blank target was used. To simulate the gamma attenuation inside the targets a replica of each one was placed inside the beam hole and outside the reactor.

The $^{233}$U samples were granted by the International Atomic Energy Agency (IAEA) and contained 52.4 mg deposited in the form of $U_3O_8$ on four titanium disks, each with an active diameter of 40 mm. The mass evaluation and isotopic analysis of these samples were carried out by AERE-Harwell Chemistry Division [13] using a gravimetric method and the results are shown in table II. The masses of the samples have been experimentally confirmed by using the gamma spectrometry method [14].

The error sources were: gamma flux: 10% of the systematic uncertainty in the photopeak efficiency determination plus 1.5 to 3% of statistical error; photofission events: 3.1% of the systematic error in the calibration for fission counting plus 2 to 5%

of the statistical uncertainty; photoneutron events: 1% of the systematic error in the calibration for neutron counting plus 6.0 to 10% of the statistical uncertainty; sample atom content: 2% of the systematic error. The experimental errors in $\nu(E)$ and in the relative intensity of the gamma-ray lines were not included in any error analysis. These values resulted in a overall uncertainties of about 15% for the final photofission cross-sections, about 20% for the final total neutron cross-sections, and ranged from 20 to 50% for the final photoneutron cross-sections.

## 3. – Results and discussion.

3·1. *Photofission cross-sections.* – The photofission cross-section data shown in table III were extracted from ref. [6], where a complete description of the methodology and discussion of results have been done. The main results obtained are: there is good agreement between our results and the data of the other authors [9-12]. A possible structure was observed in the energy of 7.23 MeV that may be endorsed by the data in 7 MeV obtained by Huizenga *et al.* [9], but the lack of data in this region does not permit a conclusion about it [6]. From our $^{233}$U photofission data and the photofission data obtained for $^{238}$U in ref. [3], using a similar arrangement, the relative fissionability was calculated and shown to be energy independent: FR = (2.12 ± 0.25). The single fission barrier height $H$ predicted by the liquid drop model was also calculated by using barrier transmission parameters from ref. [15, 16]: $H \doteq (5.6 \pm 0.2)$ MeV.

3·2. *Photoneutron cross-sections.* – As previously mentioned, the neutron counting obtained with the «long-counter» system represents the total neutron emission in the two photonuclear processes. This counting is associated with a total neutron production cross-section $\sigma(\gamma, \mathrm{N})$ which is related to the photoneutron $\sigma(\gamma, \mathrm{n})$ and photofission $\sigma(\gamma, \mathrm{f})$ cross-sections by the following expressions [3]:

$$\sigma(\gamma, \mathrm{N})(E) = \sigma(\gamma, \mathrm{n})(E) + \sigma(\gamma, \mathrm{f})(E) \times \nu(E) . \tag{1}$$

According to eq. (1) $\sigma(\gamma, \mathrm{n})$ can be obtained by using the experimental results for $\sigma(\gamma, \mathrm{f})$ and $\sigma(\gamma, \mathrm{N})$, if $\nu(E)$ is known. In the present study the $\nu(E)$ values

TABLE III. – *Photonuclear cross-sections of* $^{233}$U *as function of neutron capture gamma-ray energies measured in this work.*

| Targets/energy (MeV) | $\sigma(\gamma, \mathrm{f})$(mb) | $\sigma(\gamma, \mathrm{N})$(mb) | $\sigma(\gamma, \mathrm{n})$(mb) | $\sigma(\gamma, \mathrm{A})$(mb) |
|---|---|---|---|---|
| S/ 5.43 | 8.3 ± 4.1 | — | — | — |
| Ti/ 6.73 | 14.0 ± 2.1 | 44.9 ± 11.0 | 12.2 ± 6.2 | 26.2 ± 8.3 |
| Mn/ 7.23 | 29.8 ± 3.8 | 82.8 ± 14.6 | 10.7 ± 5.3 | 40.5 ± 9.2 |
| Pb/ 7.38 | 20.9 ± 2.4 | 60.5 ± 9.3 | 9.4 ± 3.4 | 30.3 ± 5.9 |
| Fe/ 7.64 | 21.8 ± 3.2 | 64.3 ± 10.7 | 10.0 ± 2.7 | 31.8 ± 5.9 |
| Al/ 7.72 | 26.8 ± 3.7 | 84.0 ± 13.1 | 17.0 ± 3.8 | 43.8 ± 7.5 |
| Zn/ 7.88 | 26.1 ± 3.7 | 81.3 ± 16.8 | 15.2 ± 7.5 | 41.4 ± 11.2 |
| Cu/ 7.91 | 29.1 ± 4.3 | 90.2 ± 12.5 | 16.5 ± 1.6 | 45.6 ± 5.9 |
| Ni/ 9.00 | 72.8 ± 11.6 | 221.6 ± 35.7 | 23.6 ± 4.3 | 96.5 ± 15.8 |
| Cr/ 9.72 | 98.5 ± 17.3 | 339.8 ± 71.9 | 60.1 ± 22.8 | 158.6 ± 40.1 |

reported by Berman *et al.* [12] have been used in order to determine the photoneutron cross-sections.

In order to calculate the total neutron cross-sections $\sigma(\gamma, N)$ from the experimental data, contributions due to the secondary gamma-rays with energies above the $(\gamma, n)$ threshold were taken into account. For this endeavour the total neutron cross-sections were represented by using the following formula:

$$\sum_i \sigma(\gamma,\, \mathrm{N})_i r_i = NN/(E_\mathrm{n} \times M \times G)\,,$$

where $\sigma(\gamma,\, \mathrm{N})_i$ = total neutron cross-sections at the energy of the $i$-th line in the gamma-ray spectrum emitted by the target element, $r_i$ = gamma-ray flux of the $i$-th line relative to the main gamma line, corrected for the attenuation in the filters of the collimation assembly; $NN$ = number of neutron counts obtained per unit of time of exposition; $E_\mathrm{n}$ = total neutron detection efficiency; $G$ = flux of the main gamma-ray; $M$ = number of atoms of the sample.

By making approximations of about 60 keV in energy a set of ten linear equations with 40 unknowns quantities may be obtained. This system of linear equations was solved using the same approximations reported in ref. [17, 18].

The total neutron cross-sections $\sigma(\gamma, \mathrm{N})$ and the final photoneutron cross-sections $\sigma(\gamma, \mathrm{n})$ are shown in table III. The photoneutron cross-sections are compared with the results of ref. [12] in fig. 1 and are in good agreement within experimental errors.

Neglecting $\gamma$-ray de-excitation $(\gamma, \gamma')$ above the photoneutron threshold the

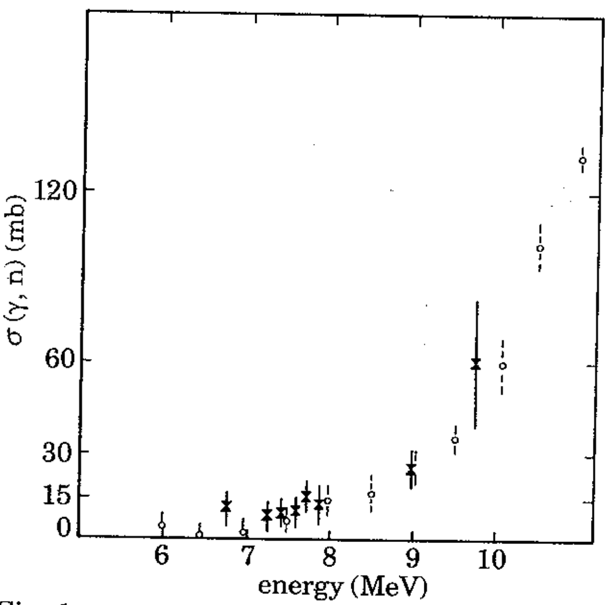


Fig. 1.

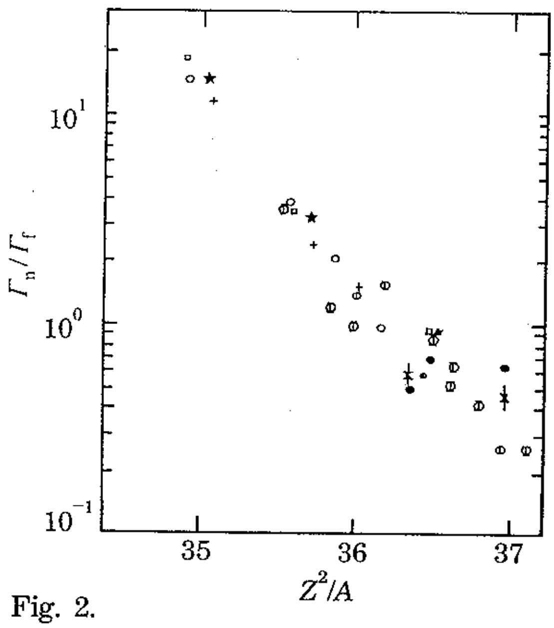


Fig. 2.

Fig. 1. – $^{233}$U photoneutron cross-sections, $\sigma(\gamma, \mathrm{n})$ in mb. The symbols mean: ○ ref. [12]; ⧗ ref. [7] (this work).

Fig. 2. – $^{233}$U logarithm of neutron emission and, fission competition values are plotted as a function of $Z^2/A$. The symbols mean: ●, $\Phi$, ○, +, □, ★, ref. [12]; ▲ ref. [18]; ⧗ ref. [7]/this work.

TABLE IV. – $^{233}$U, $\Gamma_n/\Gamma_f$ *and* $\Gamma_f/\Gamma_A$ *competions as a function of neutron capture gamma-ray energies calculated in this work.*

| Target | $E$ (MeV) | $\Gamma_n/\Gamma_f$ | $\Gamma_f/\Gamma_A$ |
|---|---|---|---|
| $^{32}$S | 5.43 | — | — |
| $^{48}$Ti | 6.73 | $0.87 \pm 0.31$ | $0.53 \pm 0.08$ |
| $^{55}$Mn | 7.23 | $0.36 \pm 0.13$ | $0.74 \pm 0.07$ |
| $^{207}$Pb | 7.38 | $0.45 \pm 0.11$ | $0.69 \pm 0.05$ |
| $^{56}$Fe | 7.64 | $0.46 \pm 0.06$ | $0.69 \pm 0.02$ |
| $^{27}$Al | 7.72 | $0.64 \pm 0.05$ | $0.61 \pm 0.02$ |
| $^{63}$Zn | 7.88 | $0.58 \pm 0.20$ | $0.63 \pm 0.08$ |
| $^{64}$Cu | 7.91 | $0.57 \pm 0.03$ | $0.64 \pm 0.01$ |
| $^{84}$Ni | 9.00 | $0.32 \pm 0.01$ | $0.76 \pm 0.01$ |
| $^{52}$Cr | 9.72 | $0.61 \pm 0.05$ | $0.62 \pm 0.04$ |
| | mean values | $0.54 \pm 0.05$ | $0.66 \pm 0.02$ |

TABLE V. – $^{233}$U, $\Gamma_n/\Gamma_f$ *values obtained by several authors by using several kinds of gamma-radiations, nuclear reactions and theoretical predictions.*

| $\Gamma_n/\Gamma_f$ | Energy (MeV) | Reactions | References |
|---|---|---|---|
| $0.54 \pm 0.05$ | $6.73 \div 9.72$ | neutron capture | this work |
| 0.49 | 11 | positron capture in flight | [12] |
| 0.50 | — | spallation reaction | [25] |
| 1.00 | $8.0 \div 12.0$ | bremsstrahlung | [16] |
| 0.44 | $(\approx 2)$ | (n, f) reactions | [21] |
| 0.67 | $(\approx 2)$ | (n, f) reactions | [22] |
| 0.66 | — | theoretical work | [22] |
| 0.61 | — | theoretical work | [24] |
| 0.72 | — | theoretical work | [23] |

competition between neutron emission and fission may be expressed by the following relationship [16]:

$$\Gamma_n/\Gamma_f(E) = \sigma(\gamma, n)/\sigma(\gamma, f)(E).$$

The $\Gamma_n/\Gamma_f$ values obtained in the 6.73 MeV to 9.72 MeV range for $^{233}$U are shown in table IV. A constant value was found: $\Gamma_n/\Gamma_f = (0.54 \pm 0.05)$ in this energy range [19]. In table V our data can be seen in comparison with the data of other authors that have used other types of gamma-ray excitation energies. Our data is also in good agreement. This competition has been correlated with the results of the other authors for other nuclei, as function of the fissility parameter $Z^2/A$, as can be seen in fig. 2. This figure shows that the values for $\Gamma_n/\Gamma_f$ decreases exponentially with the fissility of the nucleus.

The theoretical expression for $\Gamma_n/\Gamma_f$ which better explains the behaviour observed for the neutron emission and fission competition is derived from the constant nuclear temperature [16] for the level density, and is expressed by

$$\Gamma_n/\Gamma_f = (T \times 2 \times A^{2/3}/10)\exp[(E_f' - B_n')/T],$$

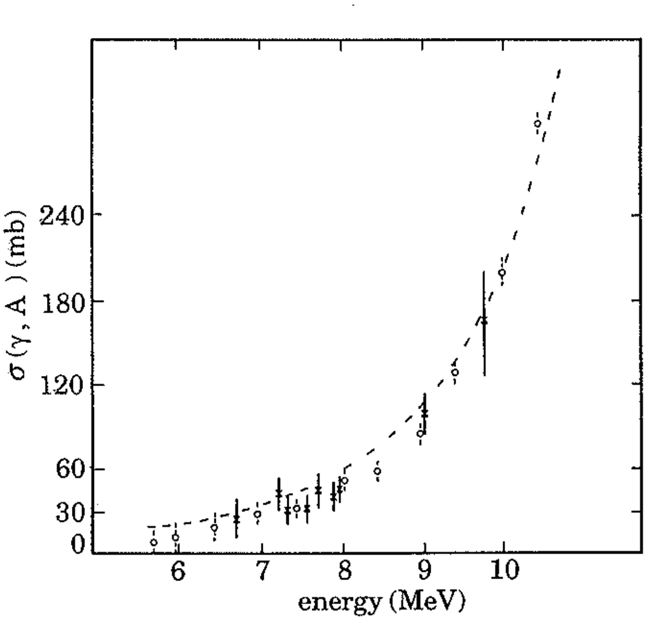


Fig. 3. – $^{233}$U photoabsorption cross-sections, $\sigma(\gamma, A)$ in mb. The symbols mean: ○ ref. [12]; ⧗ ref. [7]/this work.

where $E_f' - B_n'$ are the effective thresholds for the respective photonuclear processes and $T$ is the nuclear temperature. In the case of $^{233}$U (even-odd) nuclei, the values of $E_f'$ and $B_n'$ were determined using the following relationship [16]:

$$E_f' = E_f \quad \text{and} \quad B_n' = B_n + \Delta n\,.$$

For this study it was assumed that $\Delta n = 0.7$ MeV [27].

By using the constant result obtained for the $\Gamma_n/\Gamma_f$ competition, the following nuclear temperature for $^{233}$U was determined on basis of known model of level density:

$$T = (0.69 \pm 0.06)\,\text{MeV} \qquad \text{(constant temperature model [16])}.$$

This value is not in agreement with $T = 0.4$ MeV reported by Vandenbosch and Huizenga [16]. These authors used experimental data obtained from fast neutron and spallation reactions.

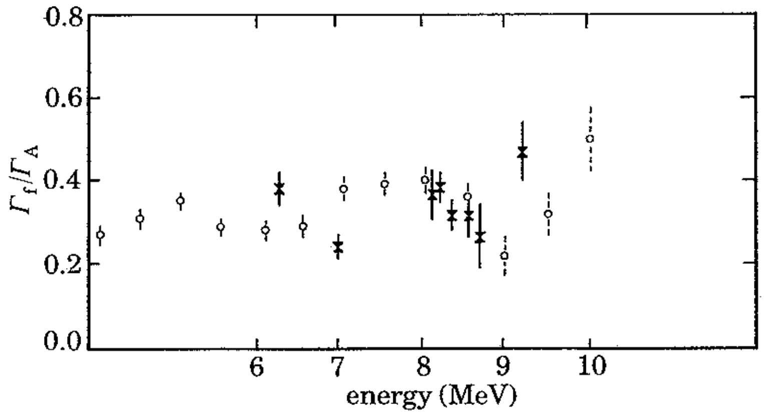


Fig. 4. – $^{233}$U photofission branching ratio ($P_f$). The symbols mean: ○ ref. [12]; ⧗ ref. [7]/this work.

TABLE VI. – $^{233}$U, $\Gamma_f/\Gamma_A$ *values obtained by several authors using several types of gamma-radiation excitation energies.*

| $\Gamma_f/\Gamma_A$ | Energy (MeV) | Gamma radiation | Reference |
|---|---|---|---|
| 0.66 ± 0.02 | 6.73 ÷ 9.72 | neutron capture | this work |
| 0.7 | 11 (asymptotic value) | positron capture in flight | [12] |
| 0.49 | 8 ÷ 12 | bremsstrahlung | [26] |

3.3. *Total photoabsorption cross-sections.* – The total photoabsorption cross-section $\sigma(\gamma, \mathrm{A})$ may be calculated by the following sum, considering the $\gamma$-ray deexcitation negligible in the (6.73 ÷ 9.72) MeV [16]:

$$\sigma(\gamma, \mathrm{A})(E) = \sigma(\gamma, \mathrm{f})(E) + \sigma(\gamma, \mathrm{n})(E).$$

The values obtained are shown in table III and are compared with the results of the ref. [12] in fig. 3. Our results are in good agreement with these, within the experimental errors.

Another important nuclear parameter is the fission probability ($P_f$), or the photofission branching ratio, which is defined as the ratio between the photofission cross-sections $\sigma(\gamma, \mathrm{f})$ and the total photoabsorption cross-sections $\sigma(\gamma, \mathrm{A})$ [14]

$$P_f(E) = \sigma(\gamma, \mathrm{f})/[\sigma(\gamma, \mathrm{f}) + \sigma(\gamma, \mathrm{n})](E).$$

The results obtained are shown in table IV. Again a constant value may be approximate: $P_f = (0.66 \pm 0.06)$. A comparison with data of ref. [12] can be seen in fig. 4 and table VI. There is good agreement between them.

According to Gell-Mann, Goldberger and Thirring [20], the photoabsorption cross-section is expected to be much the same for all actinide nuclei. In this way it is also expected a linear trend for the correlation between $P_f$ and $Z^2/A$. In fig. 5 the correlation between $P_f$ and the fissility parameter $Z^2/A$ can be seen. Our data are in better agreement with data obtained by other authors using also neutron capture gamma-rays. The trends of this figure show that the values for $P_f$ decrease

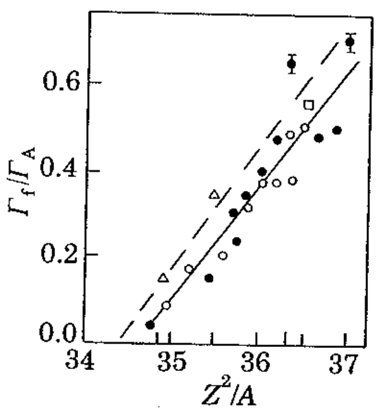


Fig. 5. – $\Gamma_f/\Gamma_A$ competition as a function of the $Z^2/A$ parameter. The symbols mean: ○, ● ref. [26] (data obtained with «bremsstrahlungs» and neutrons, respectively); △, □, ⧳ ref. [7,8] (data obtained with neutron capture gamma-rays).

exponentially with the fissionability of the nucleus thus demonstrating once again the importance of the fissility parameter $Z^2/A$ in determining the decay properties of the compound nucleus.

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