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PHOTOFISSION OF U AND Th BETWEEN 300 AND 1000 MeV

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Abstract: Loaded nuclear emulsions have been exposed to the bremsstrahlung beam of the Frascati 1 GeV electron synchrotron to measure natural uranium and thorium fission cross-sections in the energy range 300–1000 MeV. The cross-sections "per photon" are constant within the experimental errors, and amount to 67 ± 7 and 37 ± 4 mb, for U and Th, respectively. These results are briefly discussed.

E | NUCLEAR REACTIONS ^{232}Th, ^{238}U(γ, f), E_γ = 300–1000 MeV; measured σ_f. Deduced fissility. Natural targets.

1. Introduction

The photofission reaction is a particular case of the interactions of photons with complex nuclei. These interactions have been widely studied and a general picture of their principal characteristics is now available. In the 10–30 MeV energy range there is a large and wide maximum of the cross-sections known as the giant resonance; it is attributed to a dipole absorption process, in which the incoming photon interacts with the nucleus as a whole [1]). An intermediate region follows ($\approx$ 30–150 MeV) in which the cross-sections are small and show small variations; this behaviour can be explained in terms of the quasi-deuteron model which was first suggested by Levinger [2]). Finally, around 150 MeV, the cross-sections show a sharp rise, which can be safely attributed to the onset of meson-production processes. The high-energy photofission has been studied by a number of authors [3]), who, by investigating the nuclear disintegrations induced in nuclear emulsions by photons of various energies, developed the present well-established picture which has been described in detail by Peterson and Roos [4]).

This picture can be summarized as follows: to the incoming photon, whose wavelength is very short, the nucleus appears very much as an assembly of free nucleons.

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Some pions can be initially photoproduced; the cross-section of this process is easily calculated from the cross-section for meson photoproduction on free nucleons, taking due account of the motion of the nucleons in the nucleus. This initial act can be followed by subsequent interactions (re-absorption or scattering) of one or more of the produced pions and/or of the recoil nucleon. Such interactions may give rise to a "star", in which two or more charged particles are emitted or to a fission event, with a probability ("fissility") which is a strongly varying function of the photon energy.

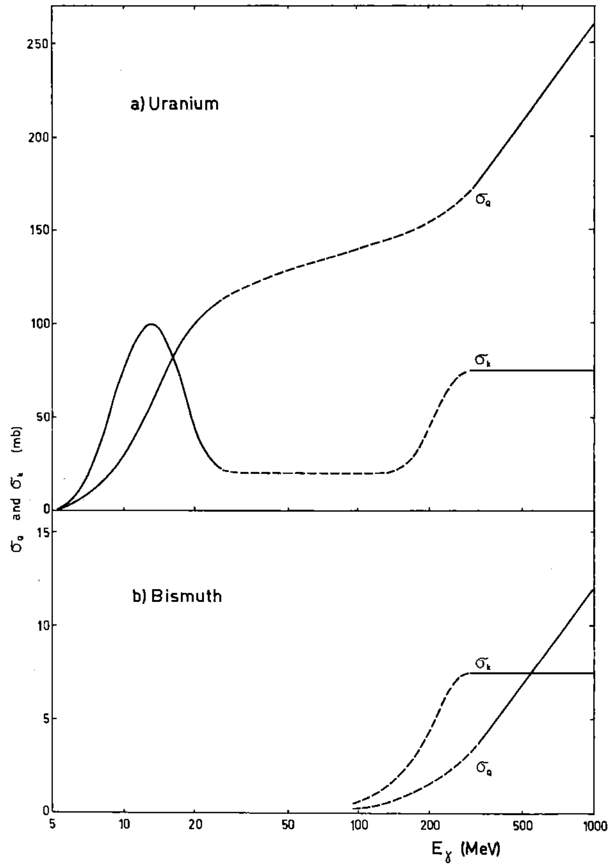

Fig. 1. General behaviour of the fission cross-section per equivalent quantum σ_Q and the fission cross-section per photon σ_K with respect to the maximum energy of the bremsstrahlung beam in the case of U and Bi.

Photofission at high energy has been studied by several authors [5]) up to 500 MeV. We have already published preliminary results on the Bi, Th and U photofission [6]) from 300 to 1000 MeV, and more complete results on Bi, W, and AgBr photofission within the same energy interval [7]). In the present paper detailed results are given concerning U and Th.

In all these works the photon-difference method is used to obtain the fission cross-section per photon σ_K from the cross-section "per equivalent quantum" σ_Q measured as a function of the maximum bremsstrahlung energy E. (In the "square" spectrum approximation $\sigma_K = d\sigma_Q/d \ln E$; for more details see the appendix). The general behaviour of these cross-sections is qualitatively described in fig. 1 for U and Bi. Due to its high fission threshold, the Bi cross-section unlike U (and the Th cross-section 8)), does not show the giant-resonance maximum.

In the cases of U and Th the low-energy part of the gamma-ray spectrum gives rise to a large number of background events which makes the measurement of σ_K more difficult (see fig. 1). This point was already stressed by Jungerman and Steiner5), who measured σ_K for Au and Bi between 150 and 500 MeV, while for ^{235}U, ^{238}U and ^{232}Th they restricted themselves to ascribe to σ_K a value between 25 and 50 mb in the 200–500 MeV energy range. Therefore some particular precautions were taken in the present measurements.

2. Experimental Procedure

The experiment has been carried out by exposing nuclear emulsions loaded with the fissioning nuclei to a variable-energy bremsstrahlung beam from the Frascati

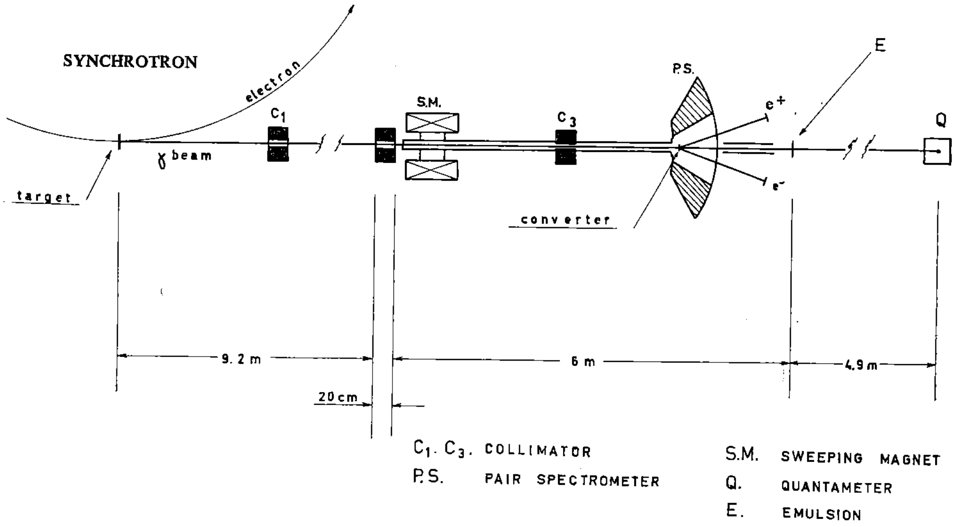

Fig. 2. Experimental layout. C_1, C_3 — collimator, P.S. — pair spectrometer, S.M. — sweeping magnet, Q — quantameter, E — emulsion.

1 GeV electron synchrotron. The experimental lay out is shown in fig. 2. During four runs a total of 64 U emulsions and 31 Th emulsions were exposed. As we explain later, only the last two runs were satisfactory from the point of view of the adequacy of the experimental conditions, and they involved 39 U emulsions and 15 Th emulsions.

In calculating the final results only the data obtained from these exposures have been used.

2.1. QUANTITATIVE EMULSION LOADING

The loading procedure and the processing method have been already described [9]. We load the emulsion by mixing quantitatively the emulsion in gel form with a suitable solution of the given element, melting at 55°C and pouring the mixture into Perspex containers from which, once dry, the pellicle is easily stripped and cut to the required size. For the processing of the emulsions we employ a procedure such that fission fragment tracks only are visible.

The use of a precision balance when preparing the emulsions ensures a good accuracy in the evaluation of the total amount of the loading element in a given pellicle. However this accuracy seems to be lost as soon as smaller pellicles are cut from the large one. A series of discrepant experimental data obtained in preliminary runs indicated that the assumption of a uniform distribution of the loading element through the parent pellicle (diameter $\varnothing = 40$ cm) was valid only to within $\approx 10\,\%$. Some measurements of the beta radioactivity of U and Th loaded emulsions confirmed this point.

On the other hand the distribution of the loading element in smaller parent pellicles ($\varnothing = 5$ cm) was uniform to within $\approx 2\,\%$; therefore the latter were preferred. From each of these we cut a single daughter pellicle ($\varnothing = 4.5$ cm), weighing about 75 % of the parent.

As a further control all results of the last two exposures were obtained from pairs of pellicles exposed together; the statistical analysis of the data shows that the total error due to the loading process is of the order of 3 %. In calculating the final results this error has been combined with the statistical one.

2.2 SCANNING

Since no alpha particle tracks are visible in our plates, fission events are identified by visual inspection; their ranges distinguish them unambiguously from nuclear recoils.

Special care was taken to maintain a control of scanning efficiency which did not interfere with routine working conditions and did not lengthen scanning time. During scanning a sampling technique has been used; in each plate 1000 to 5000 events were counted representing $\approx 20\,\%$ of the total number of fissions. With the exception of a few plates, which have been entirely re-scanned, efficiency measurements[†] have shown that the most likely value for the efficiency is 99 %; the lower limit, as obtained from standard quality control formulae [10]), is 96 %.

2.3. DOSE MEASUREMENT

The dose measurement was always carried out by means of a quantameter of the Wilson type [11]). The constant used was 4.79×10^{18} MeV/C., following Gomez

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[†] Some more details on these points will be published elsewhere.

et al. [12]). The beam was "sharply" collimated so that it hit the plates with a small cross-sectional diameter (⌀ ≈ 1.5 cm). The possible ion recombination within the quantameter (for the case of short pulses, see subsect. 2.4) was tested by comparing quantameter readings with the radioactivity simultaneously induced in copper disks, at various pulse lengths. No detectable effect was found within 0.2 %.

Intercalibration was also made (only for "long" pulses) between the quantameter readings and those of a pair spectrometer: the comparison was satisfying as a given dose was reproducible to better than 1 %.

The same pair spectrometer was used for determining to what extent the gamma-ray absorption by an interposed thickness of material would alter the quantameter readings. Quantameter and spectrometer data were compared by interposing various thicknesses of nuclear emulsion (between zero and 3.6 mm). The maximum effect at maximum thickness was 4 % at $E = 1000$ MeV and 8 % at $E = 500$ MeV. No correction was found necessary for our standard stacks (2 pellicles 300 μm thick).

2.4. ENERGY MEASUREMENTS

To check the electron energy sudden the acceleration radiofrequency was cut off at different times after the injection. The short duration of the pulses did not give rise to inconveniences from ion recombination (subsect. 2.3). The acceleration time was accurately measured by displaying the ejection pattern given by scintillator counters in an oscilloscope and simultaneously triggering a calibrated oscillator whose pulses appeared superimposed on the same screen. The Polaroid photograph of this image constituted the measuring device, whose error can easily be made as low as ≈ 100 μs (to be compared with a total acceleration time of ≈ 20 ms at $E =$ 1000 MeV).

2.5. BACKGROUND SOURCES

Beside the high-energy photons, the following fission sources have to be considered.

(i) *Neutrons from "extended" sources.* A general background of diffused low-energy neutrons, as well as a neutron beam possibly generated in the collimators have to be considered as possible sources of spurious fission events. The total angular aperture of the bremsstrahlung beam used is $\approx 1 \times 10^{-3}$ rad (subsect. 2.3), thus the density of the fissions induced in the plate by the photons is expected to be a very "square" function of the distance from the spot centre. Diffused low-energy neutrons, on the other hand, would give rise to a constant background throughout the plate; while collimated neutrons would, in any case, give rise to a "penumbra" around the gamma-ray area.

Since our beam profile as determined by fission counting is square indeed (see fig. 3), an appreciable neutron contribution to the total number of fissions is ruled out.

(ii) *Secondaries from interactions within the emulsion.* This contribution has been shown to be negligible both by a simple calculation and by comparing fission yields in stacks of different thickness.

(iii) *Low-energy photons.* The σ_Q values of the first two runs show some disagreement; the two sets of experimental points fit rather well two parallel straight lines. In the case of U the displacement is $\approx$ 30 mb, corresponding to a difference of $\approx$ 10 % at $E = 1000$ MeV. The reason for this difference was thought to be some possible variation from one run to the other of the large contribution to σ_Q of the giant-resonance photons. A check of this suggestion was devised to settle also the question of whether this contribution could vary with E. Therefore in the two last runs some copper disks were exposed and their β-activity was measured.

The ^{63}Cu(γ, n)^{62}Cu reaction, which is identifiable through the 9.8 min half-life of the β-active ^{62}Cu, takes place with a cross-section involving almost exclusively the giant-resonance photons. Moreover, the σ_K curves for the ^{63}Cu(γ, n)^{62}Cu reaction [13]) and for the U photofission in the giant resonance region are very similar. The Th fission cross-section σ_K is also similar apart from a scaling factor [8]).

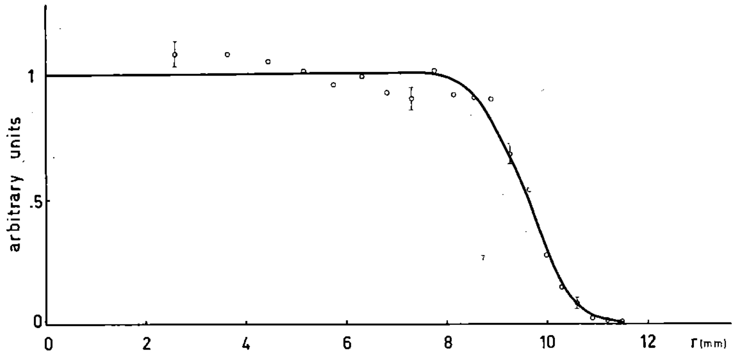

Fig. 3. Beam profile obtained in emulsions by counting the fission events as a function of the distances r from the centre of the beam spot. The density of fission, is normalized to the average value in the distance range $0 \leqq r \leqq 7.75$.

Therefore the Cu activity may be used as a good relative measure of the giant-resonance contribution. The following effects have been found:

(i) an increase of (6.0 ± 1.5)% of the Cu activity from 300 to 1000 MeV; such an effect is correlated mainly with the non-square form of the bremsstrahlung spectrum (see the appendix).

(ii) an increase of 3.5 % from the run of October 1963 to the January 1964 run. This variation hardly exceeds the experimental errors and has been neglected.

3. Experimental Results

As will be seen, our experimental results are compatible with a linear variation of σ_Q as a function of ln E. Since we cannot resolve the finer details of this dependence by our technique, we will *assume* that the dependence is linear. Therefore the resulting σ_K has a constant value.

It is convenient to analyse separately the data from the last two runs (October 1963 and January 1964). They are considered to be the most reliable for the following reasons:

(i) only emulsions obtained from *small* parent pellicles have been used, exposed in pairs (see subsect. 2.1);

(ii) Cu activation measurements have been carried out (see subsect. 2.5);

(iii) the two straight lines representing the best fit of σ_Q data as a function of ln E, show a small parallel displacement, i.e. $\approx 4\,\%$ at $E = 1000$ MeV.

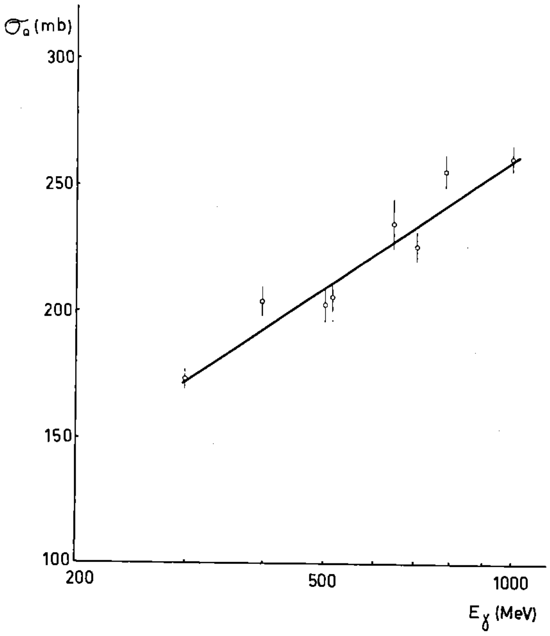

Fig. 4. The experimental data for the U fission cross-section per equivalent quantum σ_Q are plotted against the maximum energy of the γ beam. The solid line is the result of a least-squares calculation.

The σ_Q values from these two runs were combined together to obtain the weighted mean value at any energy. The errors were calculated in two different ways (from the statistical errors of the single measurements and from their dispersion); with only one exception the two errors were practically coincident. The results are shown in fig. 4, the errors quoted are the larger of the calculated ones; the solid straight line was obtained by means of the least-squares method. The slope of this line is σ_K^0, i.e. the value of σ_K, in the square spectrum approximation.

The result is σ_K^0 (U, F) $= 75\pm5$ mb with a chi-square value $\chi^2 = 47.7$ and $P(\chi^2) = 11\,\%$. The σ_K value would not be appreciably changed if the data from all four runs were combined; one would obtain $\sigma_K^0 = 76\pm4$ mb.

If the actual, non-square form of the spectrum is taken into account some corrections are needed, resulting in a decrease of σ_K by (10±2) % (see the appendix). Therefore the final result of the uranium measurements is $\sigma_K(\mathrm{U, F}) = 67 \pm 7$ mb.

The Th data have been treated in the same way. Since in the October 1963 run, no Th loaded plates had been exposed, the data of only the last run are shown in fig. 5. The least-squares straight line gives $\sigma_K^0 = 42 \pm 4$ mb, the χ^2 value being $\chi^2 = 16.8$ with $P(\chi^2) = 22\,\%$. Applying a similar correction, one obtains the final result $\sigma_K(\mathrm{Th, F}) = 37 \pm 4$ mb.

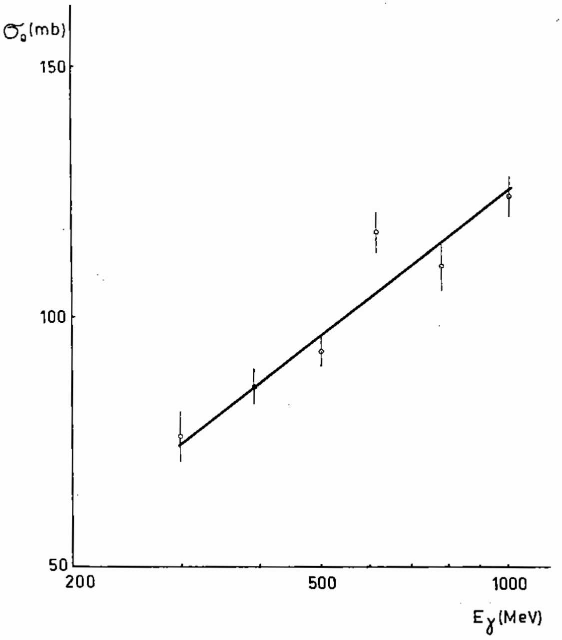

Fig. 5. The experimental data for the Th fission cross-section per equivalent quantum σ_Q are plotted against the maximum energy of the γ beam. The solid line is the result of a least-squares calculation.

These values are not very different from the preliminary results [5]). However, the present data are considered to be much more reliable because of the particular precautions taken in carrying out the measurements.

4. Discussion

From the physical point of view, the interesting parameter is not σ_K but rather the fissility, i.e. the probability

$$f = \sigma_K(X, F)/\sigma_K(X, T) \tag{1}$$

that the nucleus undergoes fission after a photon has been absorbed.

The best known values for $\sigma_K(X, T)$, are those obtainable from nuclear emulsion work which provides the cross-section σ_K (Ag Br, S) for star production in emulsion nuclei. Peterson and Roos 4) have closely studied the ratio between $\sigma_K(S)$ and $\sigma_K(T)$ for Ag and Br and found that, between 300 and 1000 MeV the following relation holds:

$$\sigma_K(\text{Ag Br}, T) = 1.25 \ \sigma_K(\text{Ag Br}, S). \tag{2}$$

In order to obtain $\sigma_K(T)$ for ^{238}U and ^{232}Th, it suffices to take into account the different number of nucleons. Indeed, as stated in sect. 1, the initial act is essentially the photoproduction of pions on single nucleons. The weighted mean value of the star production cross-section, referred *to the single nucleon* $\mathcal{N}$, as obtained from refs. 3,4), is

$$\sigma_K(\mathcal{N}, S) = 268 \pm 33 \ \mu\text{b}. \tag{3}$$

Combining eqs. (2) and (3) one obtains, for a nucleus of mass number A:

$$\sigma_K(X, T) = 1.25 \ A\sigma_K(\mathcal{N}, S) = (335 \pm 40)A \ \mu\text{b}. \tag{4}$$

In this way the small 14) difference between meson photoproduction cross-sections on protons or neutrons is neglected. From eq. (4) one obtains $\sigma_K(\text{U}, T) = 80 \pm 10$; $\sigma_K(\text{Th}, T) = 78 \pm 10$ mb and, consequently $f(U) = 0.84 \pm 0.13$; $f(\text{Th}) = 0.47 \pm 0.10$. These data are in good agreement with our previous 15) results. We have previously shown that f is a very steep function of Z^2/A but that it does not change very much when the bombarding particles change 16) from photons between 300 and 1000 MeV to protons of 600 MeV. These remarks dealt with the general behaviour of the fissility, which varies very much from Ag to U (more than three orders of magnitude).

TABLE 1
Some fissility values

Particle	Energy (MeV)	Nucleus		
		^{238}U	^{232}Th	^{209}Bi
Photons	300÷1000	0.84±0.13	0.47±0.10	0.12±0.02
Protons	600 a)	0.65	0.40	0.13
Protons	200 b)	0.80	0.47	0.09

a) Ref. 16). b) Ref. 17).

However, as far as heavy nuclei are concerned, a more detailed comparison may now be carried out and an appreciable difference emerges between protons and photons, as seen in table 1. In the case of 600 MeV protons the fissility is lower both for U and Th than the one observed for 300–1000 MeV photons. One might expect an inverse trend considering that, in the case of the positively charged particles, the fissioning nucleus could have a higher Z^2/A. On the other hand, it should be remembered that the cross-sections for U and Th proton-induced fission vary appreciably with the energy of the incident protons and show a large maximum 17) between 40 and 200 MeV. If a comparison is made with protons in this energy range (table 1, third line, the agreement is good. This comparison is suggested by considering that at high proton energies the decrease of the cross-sections may be attributed to progressively larger nuclear cascades [18]), a phenomenon which is absent or very scarce in the case of photons.

We are very grateful to Professor G. Cortini for many useful discussions and for his invaluable help and criticism. Acknowledgements are due also to Professor M. Muchnik for his contribution during many stages of the work. We are also indebted to members of the Frascati electron synchrotron staff for their friendly cooperation in performing the exposures; to the members of the Centro di Calcolo of the University of Naples and of the Frascati Sezione Calcolatore for the computer calculations.

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Appendix

Let $n(k, E)\,\mathrm{d}k$ be the number of photons in the energy interval $(k, \mathrm{d}k)$ of a bremsstrahlung beam originated by electrons of energy E.

This spectrum function $n(k, E)$ is given by the usual formula [19])

$$n(k, E) = \frac{Q}{k}\, b\left(\frac{k}{E}\right), \tag{A.1}$$

where Q is a constant and $b(u)$ is a known function, normalized according to

$$\int_0^1 b(u)\mathrm{d}u = 1. \tag{A.2}$$

The total energy in the beam, which is the quantity directly measured by the Wilson quantameter, is given by

$$\mathscr{E} = \int_0^E kn(k, E)\mathrm{d}k = Q\int_0^E b\left(\frac{k}{E}\right)\mathrm{d}k = EQ\int_0^1 b(u)\mathrm{d}u = EQ. \tag{A.3}$$

One can see from eq. (A.3) that, due to eq. (A.2), the "number of equivalent quanta" is given by

$$Q = \mathscr{E}/E \tag{A.4}$$

In the "square spectrum" approximation, the following relations hold:

$$\begin{aligned} b(u) &= 1, \qquad 0 < u < 1, \\ b(u) &= 0, \qquad u > 1, \end{aligned} \tag{A.5}$$

and eq. (A.1) becames

$$n(k, E) = Q/k, \qquad k < E. \tag{A.6}$$

If σ_K is the cross-section for a given process induced by a photon of energy k in a given nucleus, the total number of events is given by

$$\mathscr{N} = N\int_0^E \sigma_k n(k, E)\mathrm{d}k = NQ\int_0^E \sigma_k(k)b\left(\frac{k}{E}\right)\frac{\mathrm{d}k}{k}, \tag{A.7}$$

N being the number of nuclei per square centimeter of the target. In order to evaluate σ_k, one has to measure $\mathscr{N}$ as a function of E and solve the resulting integral equation. The cross-section per equivalent quantum σ_Q is defined by

$$\sigma_Q = \frac{1}{Q}\int_0^E \sigma_k n(k, E)\mathrm{d}k = \int_0^E \sigma_k b\left(\frac{k}{E}\right)\frac{\mathrm{d}k}{k}. \tag{A.8}$$

In the square spectrum approximation the values of σ_K, at any energy, can be obtained in a straightforward way by means of:

$$\sigma_k = \mathrm{d}\sigma_Q/\mathrm{d}\ln E. \tag{A.9}$$

On the other hand, if the difference

$$c(u) = b(u)-1$$

has to be taken into account, the right side of (A.9) does not depend only on σ_K at the considered energy, but also on its values at lower energies.

Therefore, one has to make some reasonable assumptions on the behaviour of σ_K in the low and medium energy range; in order to do this, the energy range $0 \leqq E \leqq 1000$ MeV has been divided into three different regions.

In the first one ($0 < k < 25$ MeV), the giant resonance is the most relevant phenomenon; in order to easily take it into account, it is reasonable to write σ_K^1 as

$$\sigma_k^1 = 15S\delta(k-15), \tag{A.10}$$

where S is an unknown constant and δ is the Dirac function. In the energy range $25 < k < 200$ MeV, the fission cross-section σ_k^m can be deduced from the quasi-deuteron [20]) model, which implies

$$\sigma_k^m = \frac{8NZ}{A}\sigma_L f, \tag{A.11}$$

where σ_L is the Levinger quasi-deuteron cross-section and f is the fissility. Eventually, for k values between 200 and 1000 MeV, we can assume (see sect. 3) an unknown constant value of σ_k^h:

$$\sigma_k^h = \text{const.} \tag{A.12}$$

Therefore, we obtain for the experimental cross-section per equivalent quantum:

$$\sigma_Q(E) = \int_0^E \sigma_k\left[1+c\left(\frac{k}{E}\right)\right]\frac{\mathrm{d}k}{k}, \tag{A.13}$$

which, under the assumptions described above, becomes

$$\sigma_{\mathrm{Q}}(E) = \sigma_{\mathrm{k}}^{\mathrm{h}} \int_{200}^{E} \frac{\mathrm{d}k}{k} + \sigma_{\mathrm{k}}^{\mathrm{h}} \int_{200}^{E} c\left(\frac{k}{E}\right) \frac{\mathrm{d}k}{k} + \int_{25}^{200} \sigma_{\mathrm{k}}^{\mathrm{m}} \left[1 + c\left(\frac{k}{E}\right)\right] \frac{\mathrm{d}k}{k} + 15S\left[1 + c\left(\frac{15}{E}\right)\right] \frac{1}{15}. \tag{A.14}$$

Using eq. (A.14) for $E = 300$ and $E = 1000$ MeV, subtracting the two expressions and solving for $\sigma_{\mathrm{k}}^{\mathrm{h}}$, one gets

$$\sigma_{\mathrm{k}}^{\mathrm{h}} = \frac{\sigma_{\mathrm{Q}}(1000) - \sigma_{\mathrm{Q}}(300) - B - SC}{\ln \frac{1000}{300} + A}, \tag{A.15}$$

where the constants A, B and C are defined by:

$$A = \int_{0.200}^{0.667} c(u) \frac{\mathrm{d}u}{u}, \tag{A.16}$$

$$B = \int_{25}^{200} \sigma_{\mathrm{k}}^{\mathrm{m}} \left\{ c\left(\frac{k}{1000}\right) - c\left(\frac{k}{300}\right) \right\} \frac{\mathrm{d}k}{k}, \tag{A.17}$$

$$C = c(0.015) - c(0.050) = b(0.015) - b(0.050). \tag{A.18}$$

The value of the constant A comes out to be ≈ 0.01 and can be safely neglected.

The calculation for B is more uncertain because of the uncertainty of the fissility f (eq. A.11). The calculation has been carried out in two different ways:

(i) on the assumption f = constant = 0.2, a value strongly suggested by a comparison between the star [4]) and fission [5]) cross sections;

(ii) on the more drastical assumption $\sigma_{\mathrm{k}}^{\mathrm{m}}$ = const = 20 mb. The obtained values are 8.5 and 7.7 mb, respectively, which support the validity of the assumptions used. As far as the term SC is considered, the two factors must be considered separately. The S value can be deduced from the giant resonance fission cross-section $\sigma_{\mathrm{k}}(E)$, as measured for instance by Katz *et al.* [8]) by calculating $S = \int_{\sigma\mathrm{k}}(E)\mathrm{d}k/k$. From the quoted data, one gets $S = 62$ mb. On the other hand such direct measurements by various authors are not in very good natural agreement [8]). Another procedure is to deduce the S value from our data at 300 MeV by subtracting the contribution of the quasi-deuteron region; in this way we get $S = 77$ mb. We have used an average value $S = 70$ mb.

The C value deduced from the $b(u)$ curve is 0.05 This is in agreement with the $(6.0 \pm 1.5)\,\%$ increase found in the Cu activity (see subsect. 2.5 (iii)) (small contributions to this increase can also be due to high and medium energy photons). Eventually we have $SC = 70 \times 0.05 = 3.5$ mb. Combining the different corrections, one gets for the final value of the fission cross-section in the case of U

$$\sigma_{\mathrm{k}}(\mathrm{U, F}) = 67 \pm 7 \text{ mb}.$$

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