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FISSION OF  $^{232}\text{Th}$  NEAR THRESHOLD: ANGULAR  
DISTRIBUTION. -

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L. Fiore<sup>(o)</sup>, A. Manfredini, C. Ramorino<sup>(x)</sup>, W. Wolfli<sup>(+)</sup>: PHOTOFIS-  
SION OF  $^{232}\text{Th}$  NEAR THRESHOLD: ANGULAR DISTRIBUTION<sup>(\phi)</sup>.

ABSTRACT. -

Measurements on angular distribution of photofission fragments of  $^{232}\text{Th}$  from monochromatic  $\gamma$ -rays at energies near threshold are reported. The data are compared with measurements of other authors obtained with a bremsstrahlung  $\gamma$ -ray source.

RIASSUNTO. -

Si riportano le misure di distribuzione angolare dei frammenti di fotofissione del  $^{232}\text{Th}$ . I risultati sono confrontati con le misure di altri autori ottenute con una sorgente di gamma di bremsstrahlung.

1. - INTRODUCTION. -

The angular distribution of photofission fragments from  $^{232}\text{Th}$  has been measured using monochromatic  $\gamma$ -rays in the energy range  $5.4 \leq E_\gamma \leq 9.0$  MeV;

The monochromatic  $\gamma$ -rays have been obtained from the  $(n, \gamma)$  reaction of thermal neutrons on particularly chosen target elements (see table I, column 1).

The fission tracks have been recorded in nuclear emulsions. Pellicles of nuclear emulsions loaded either with thorium or uranium

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complex, separated one from the other by thin mylar sheets to avoid diffusion of fissile nuclei from one pellicle to the other, have been exposed at the end of the transversal beam hole of the nuclear reactor "Saphir" of EIR (Würenlingen, Switzerland).

Results on measurements of  $^{238}\text{U}$  photofission cross section and angular distribution of fragments, as well as photofission cross section for  $^{232}\text{Th}$  have already been published<sup>(1 + 4)</sup>, and we refer to our previous papers for all details about the technique employed, the target elements chosen for the  $\gamma$ -ray irradiations, the geometrical conditions, the measurement and the evaluation method.

## 2. - EXPERIMENTAL RESULTS. -

For the analysis of our experimental data we followed the same procedure discussed in detail in our paper on the angular distribution of the photofission fragments from  $^{238}\text{U}$  (ref. 3).

We give here only a short account of the more significant results. The measured angular distribution have been fitted either to the functions

$$W(\theta) = a + b \sin^2\theta + c \sin^2\theta \cos^2\theta$$

or

$$W'(\theta) = a' + b' \sin^2\theta$$

A  $\chi^2$  test for each  $\gamma$ -ray energy was then made and the  $\chi^2$  values for either the  $W(\theta)$  or the  $W'(\theta)$  functions were plotted.

The distributions obtained are very similar to those shown in Fig. 3 of ref. (3) for the uranium analysis, allowing us to conclude that also in this case the constant  $c$  has to be generally considered different from zero, although the statistical error makes it compatible with zero for many energies.

The angular distribution  $W(\theta)$ , after correction for neutron background, is due to the contribution of all the lines of the  $\gamma$ -ray spectrum of the element used as a target. The background correction, which generally is small or negligible, was performed and a similar conclusion as in the case of  $^{238}\text{U}$  can be given. There is only the exception of the S target; the cross section measurements<sup>(4)</sup> showed that the yield of the fission tracks produced by the 5.43 MeV line on thorium is no longer negligible as in the case of the uranium photofission but of the same order of magnitude as the yield of fission tracks from the two lines at 7.78 and 8.64 MeV. The irradiation with the S target was therefore used in order to obtain some information at 5.43 MeV, although the final stati

stical error is very large, due to the large uncertainty in the cross section at 5.43 MeV and due to the severe correction.

In Table I, the values of the constants a, b, c of the  $W(\theta)$  distribution, normalized by means of

$$\int_0^{\pi/2} W(\theta) \sin\theta d\theta = 1$$

are reported both before and after the correction for the low intensity  $\gamma$ -ray lines. The values of the constants before the  $\gamma$ -ray correction are attributed to a mean energy  $\bar{E}_\gamma$  (column 2), obtained weighting the energies of the lines in the spectrum by their intensities and fission cross sections. The energy  $E_\gamma$  of the main line is reported in column 7.

The error quoted in Table I takes into account the statistical error due to the number of measured tracks and to all the corrections introduced.

### 3. - DISCUSSION. -

Fig. 1 shows the evaluated  $b/a$  distribution together with the most recent results of Rabotnov et al. (5). The two distributions are not directly comparable because the measurements of Rabotnov et al. are plotted at the maximum energy in the bremsstrahlung spectrum. We can say however that, while the behaviour is very similar in both measurements, the absolute values are so different that no physical reason can be found for the discrepancy. A look for systematic error seems unavoidable.

In our measurements the angle of each fission track recorded in the nuclear emulsion was measured assuming the edge of emulsion to be parallel to the  $\gamma$ -ray beam. However, controls have been made, recording for each angle its direction towards the glass or the surface of the emulsion and recording the angles from  $-\pi/2$  to  $+\pi/2$ . Any mistake in the alignment of the emulsion, and therefore in the assumption that the edge of the emulsion was parallel to the  $\gamma$ -ray base, would show itself in an asymmetric distribution in respect to the assumed zero. Only in a few cases was a small correction necessary.

The geometry of the exposures, background neutrons and the efficiency of scanning and its correction has already been discussed elsewhere (3).

So we cannot account in any manner for a flattening of the asymmetry in our measurements. Moreover as the alignment was set up "ex novo" for each energy exposure. It is hard to find a cause for a systematic lowering of the asymmetry as shown in Fig. 1.

TABLE I - Values of the a, b and c parameters of the angular distribution.

Target	$\bar{E}_\gamma$ (MeV)	a	b	c	Target	$\bar{E}_\gamma$ (MeV)	a	b	c
Dy	5.61	0.099 $\pm 0.067$	1.121 $\pm 0.053$	1.152 $\pm 0.435$	S	5.43	0.478 $\pm 0.631$	0.087 $\pm 0.541$	3.471 $\pm 2.595$
Y	6.14	0.103 $\pm 0.031$	1.224 $\pm 0.022$	0.607 $\pm 0.231$	Dy	5.58	0.099 $\pm 0.073$	1.114 $\pm 0.058$	1.186 $\pm 0.470$
Ca	6.42	0.086 $\pm 0.025$	1.164 $\pm 0.018$	1.034 $\pm 0.184$	Y	6.07	0.098 $\pm 0.037$	1.209 $\pm 0.027$	0.724 $\pm 0.276$
Ti	6.66	0.116 $\pm 0.018$	1.287 $\pm 0.014$	0.200 $\pm 0.153$	Ca	6.42	0.073 $\pm 0.027$	1.184 $\pm 0.020$	1.033 $\pm 0.199$
Be	6.80	0.286 $\pm 0.069$	0.952 $\pm 0.103$	0.587 $\pm 0.490$	Ti	6.75	0.128 $\pm 0.032$	1.361 $\pm 0.025$	-0.265 $\pm 0.263$
Pb	7.28	0.264 $\pm 0.074$	0.918 $\pm 0.062$	0.931 $\pm 0.611$	Be	6.82	0.286 $\pm 0.069$	0.952 $\pm 0.103$	0.587 $\pm 0.490$
S	7.33	0.590 $\pm 0.171$	0.389 $\pm 0.147$	1.128 $\pm 0.693$	Pb	7.28	0.288 $\pm 0.088$	0.839 $\pm 0.073$	1.143 $\pm 0.721$
Cu	7.60	0.495 $\pm 0.031$	0.659 $\pm 0.022$	0.490 $\pm 0.234$	Fe	7.63	0.512 $\pm 0.036$	0.685 $\pm 0.026$	0.231 $\pm 0.271$
Fe	7.63	0.499 $\pm 0.027$	0.694 $\pm 0.019$	0.291 $\pm 0.197$	Cu	7.91	0.624 $\pm 0.059$	0.456 $\pm 0.043$	0.544 $\pm 0.447$
Ni	8.27	0.742 $\pm 0.032$	0.381 $\pm 0.023$	0.033 $\pm 0.236$	Ni	8.86	0.886 $\pm 0.049$	0.218 $\pm 0.035$	-0.236 $\pm 0.362$

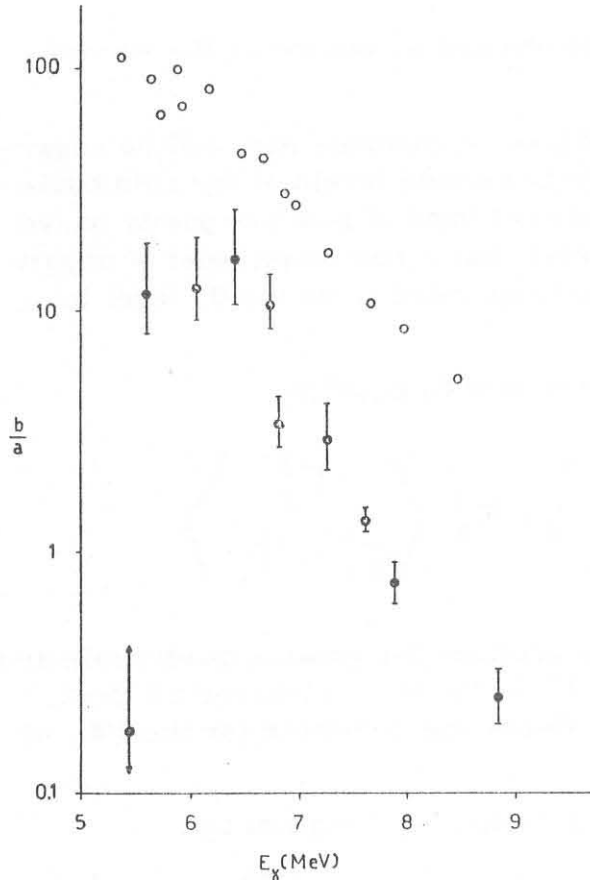


FIG. 1 - Experimental ratio  $b/a$  as a function of  $E_\gamma$ . Black points are our experimental data; white points are Rabotnov et al. (5) results and are plotted as a function of  $E_{\max}$  of the bremsstrahlung spectrum.

A systematic inefficiency in the recording of fissions at zero degrees in the measurements of Rabotnov et al. could be the only error that would account for a higher asymmetry than the real one, but from the statement of the authors, they slightly over, rather than underestimated the value of the constant  $a$ .

However, the actual value of the asymmetry has not been taken into account in the discussion of Rabotnov et al.; only the position of the maximum of the  $b/a$  function has been considered. In the single barrier configuration the fission threshold and the energy of the  $1^{-}K = 0$  level should be approximately the same; in the double barrier configuration, the fission threshold is determined by the higher of the two, while the angular distribution will depend on the relative height of the barriers A and B.

In fact, (see for instance ref. (6, 7)) if the second well is deep, the nucleus will stay there for a considerable time; therefore it may forget the  $K$  value with which it passed

over the first barrier, and the angular distribution will correspond to the distribution of  $K$  values at the second barrier.

If the second barrier is  $1 \div 2$  MeV lower than the first one, this distribution corresponds nearly to a statistical distribution of  $K$ , because many channels are open over the second barrier even for energies close to the fission barrier (in the present case the barrier A).

Therefore the angular distribution is smoother than one would expect for near-barrier fission.

If the barrier B is higher than the first one (A) the usual picture of the channel structure in near-barrier fission should be valid. In this case moreover, it is the second barrier that corresponds to the effective energy threshold. A very high value of the asymmetry in the angular distribution should be therefore an indication of the barrier B being higher than the first one. The analysis of the angular distribution should there-

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fore correspond to the analysis of the channel structure at the second barrier.

In this case the spectrum of fission channels can still be regarded similar to the spectrum of low lying excited levels of the cold nucleus. One expects therefore (i) a rotational band of positive parity based on the  $0^+$  level (containing no  $1^+$  levels); (ii) a rotational band of negative parity beginning with a  $1^-$  level and separated from the  $0^+$  level by a distance  $\Delta$ .

The photofission cross section is then equal to

$$(1) \quad \sigma_{\gamma f} = \sigma_{\gamma}^{2+} \left\langle \frac{\Gamma_f^{2+}}{\Gamma_f^{2+} + \Gamma_c} \right\rangle + \sigma_{\gamma}^{1-} \left\langle \frac{\Gamma_f^{1-}}{\Gamma_f^{1-} + \Gamma_c} \right\rangle$$

where  $\sigma_{\gamma}^{2+}$  and  $\sigma_{\gamma}^{1-}$  are the cross sections for gamma quadrupole and dipole absorption, respectively and  $\Gamma_c$  is the total width for all decay process in competition with fission. Below the threshold for the  $(\gamma, n)$  reaction  $\Gamma_c = \Gamma_{\gamma}$ .

The differential angular distribution can be given by

$$2\pi \frac{d\sigma_{\gamma f}}{d\Omega} = \sigma_{\gamma}^{1-} \sum_0^1 \frac{\Gamma_{f,K}^{1-}}{\Gamma^{1-}} W_K^1(\theta) + \sigma_{\gamma}^{2+} \sum_0^2 \frac{\Gamma_{f,K}^{2+}}{\Gamma^{2+}} W_K^2(\theta)$$

with

$$W_K^J(\theta) = \frac{2J+1}{4} \left\{ \left| D_{1K}^J \right|^2 + \left| D_{-1K}^J \right|^2 \right\}$$

where  $D_{MK}^J(\theta)$  represent the spherical Wigner functions. Therefore

$$4\pi \frac{d\sigma_{\gamma f}}{d\Omega} = a + b \sin^2 \theta + c \sin^2 \theta \cos^2 \theta$$

Taking into account the normalization mentioned above

$$\int_0^{\pi/2} W(\theta) \sin \theta d\theta = 1$$

the constants a, b and c are related to the width of states and the  $\sigma_{\gamma}$ , by

$$(2) \quad \frac{2}{3} a = \frac{1}{1 + \alpha} \frac{\Gamma_{f1}^{1-}}{\Gamma_f^{1-}} + \frac{5}{3} \frac{\alpha}{1 + \alpha} \frac{\Gamma_{f1}^{2+}}{\Gamma_f^{2+}}$$

$$(3) \quad \frac{2}{3} (b + \frac{1}{2} a) = \frac{1}{1 + \alpha} \frac{\Gamma_{f0}^{1-}}{\Gamma_f^{1-}} + \frac{5}{6} \frac{\alpha}{1 + \alpha} \frac{\Gamma_{f2}^{2+}}{\Gamma_f^{2+}}$$

$$(4) \quad \frac{2}{15} c = \frac{\alpha}{1 + \alpha} \left( 1 - \frac{5}{3} \frac{\Gamma_{f1}^{2+}}{\Gamma_f^{2+}} - \frac{5}{6} \frac{\Gamma_{f2}^{2+}}{\Gamma_f^{2+}} \right)$$

with

$$(5a) \quad \frac{1}{1 + \alpha} = \frac{\sigma_{\gamma}^{1-}}{\sigma_{\gamma f}^{1-}} \frac{\Gamma_f^{1-}}{\Gamma^{1-}}$$

and

$$(5b) \quad \frac{\alpha}{1 + \alpha} = \frac{\sigma_{\gamma}^{2+}}{\sigma_{\gamma f}^{2+}} \frac{\Gamma_f^{2+}}{\Gamma^{2+}}$$

From (4) one has that the condition

$$\frac{\alpha}{1 + \alpha} \geq \frac{2}{15} c$$

has to be satisfied for all the energies; the "equals" holds only when the  $2^+K \neq 0$  channels are not excited.

From (3) the condition for neglecting the term from quadrupole absorption is given by

$$\frac{1}{1 + \alpha} > a - \frac{1}{2}$$

which, since  $1/(1 + \alpha) > 0$ , is always fulfilled near threshold; one can therefore put, with negligible error and with no restrictive hypothesis on the behaviour of  $1/(1 + \alpha)$

$$(6) \quad \frac{2}{3} (b + \frac{1}{2} a) = \frac{1}{1 + \alpha} \frac{\Gamma_{f0}^{1-}}{\Gamma_{f0}^{1-} + \Gamma_{f1}^{1-}}$$



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for the whole range of energy considered.

On the contrary, from (2), the conditions for either the term from quadrupole or dipole absorption being negligible, are restrictive and would oblige one to make a definite hypothesis about the behaviour of  $\sigma_{\gamma}^{1-}$  and  $\sigma_{\gamma}^{2+}$  with gamma energy.

No reliable direct data are available at present for the absolute value and energy dependence of  $\sigma_{\gamma}$  and its partial components corresponding to various multipolarities; therefore a definite hypothesis has to be introduced in order to be able to evaluate the experimental data. One has to keep in mind that any conclusion would be reliable strictly under the condition of the validity of the hypothesis introduced.

The relation (6) can be written as

$$(7) \quad \frac{\Gamma_{f0}^{1-}}{\Gamma_{f1}^{1-}} = \frac{\frac{2}{3} (b + \frac{1}{2} a)}{\frac{2}{15} c + \frac{2}{3} a - \frac{\alpha}{1 + \alpha}}$$

Taking into account the Bohr and Wheeler expression for the width of states

$$\frac{\Gamma_{f0}^{1-}}{\Gamma_{f1}^{1-}} = \frac{1 + \exp \frac{B_1 - E}{E_{0p}}}{1 + \exp \frac{B_0 - E}{E_{0p}}}$$

the constants  $B_0$ ,  $E_{0p} = (E_{0 \text{ curv}})/2\pi$ ,  $B_1$ ,  $E_{1p} = (E_{1 \text{ curv}})/2\pi$  can be obtained by means of a fit of the function (7) to the experimental data, introducing a set of hypothesis for the behaviour of  $\alpha/(1 + \alpha)$  with gamma energy.

The particular hypothesis  $\alpha/(1 + \alpha) = \frac{2}{15} c$  would change (7) to

$$\frac{\Gamma_{f0}^{1-}}{\Gamma_{f1}^{1-}} = \frac{b}{a} + \frac{1}{2}$$

This function is the one which has generally been used to obtain information on the behaviour of the nucleus near the fission threshold, on the assumption that this function is not affected by any hypothesis on the variation of  $\sigma_{\gamma}^{1-}$  and  $\sigma_{\gamma}^{2+}$  with energy. On the contrary one sees that the validity of such an equation is a consequence of the well defined hypothesis that the quantity  $\alpha/(1 + \alpha)$  given by eq. (5b) has the same behaviour as the measured constant  $c$ ; one supposes therefore that the lowering of the constant  $c$  with increasing gamma energy is not a consequence of the even

tual excitation of the  $2^+K \neq 0$  levels. Rabotnov et al. (5) start from the observation that in the single barrier configuration the maximum of the function  $\Gamma_{f0}^{1-} / \Gamma_{f1}^{1-}$  occurs at an energy value slightly higher than the fission threshold. On the contrary, their experimental values of  $b/a$  for the double even nuclei measured have the maximum consistently at energies lower than the fission threshold. They therefore conclude this to be a consequence of the double potential barrier; they suppose the first barrier to be the higher one and to be responsible for the position of the fission threshold, while the angular distribution is determined by the channels at the second barrier. The position of the maximum of the relation  $b/a$  would therefore be roughly situated at the energy of the level  $1^-K = 0$  at the second barrier.

We consider, however, that such conclusions are not reliable enough, as the  $B_0$  value from eq. (7) can be very easily changed with reasonable hypothesis on the behaviour of  $\alpha/(1+\alpha)$  with energy.

Our experimental data have been fitted in eq. (7) by means of a program of minimisation of  $\chi^2$ . Introducing the two hypothesis

$$1) \frac{\alpha}{1+\alpha} = \frac{2}{15} c \quad \text{and} \quad 2) \frac{\alpha}{1+\alpha} = \text{const.}$$

the values reported in Table II have been obtained.

TABLE II

Values of the  $B$  and  $E_p$  parameters obtained by means of a fit of eq. (7), with the two hypothesis of column 1 for  $\alpha/(1+\alpha)$ .

$\frac{\alpha}{1+\alpha}$	$B_0$ (MeV)	$E_{0p}$ (MeV)	$B_1$ (MeV)	$E_{1p}$ (MeV)
$\frac{2}{15} c$	6.48	0.16	7.77	0.27
0.156	6.64	0.24	7.57	0.29

The hypotheses on the  $\alpha/(1+\alpha)$  function, correspond to suppose that either 1) the percentage contribution of quadrupole absorption becomes lower for higher energies and the  $2^+K=1$  and  $2^+K=2$  levels are not excited in the considered interval of energy; or 2) the observed lowering of the term  $\{2/15\}c$  is entirely due to the contribution of the  $2^+K=1$  and  $2^+K=2$  states, while the percentage contribution of quadrupole absorption to the fission process is constant.

In Fig. 2 the experimental data for the function.

$$\frac{\frac{2}{3}a + \frac{2}{15}c - \frac{\alpha}{1+\alpha}}{\frac{2}{3}(b + \frac{1}{2}a)}$$

are plotted. The full line gives the  $\Gamma_{f1}^{1-} / \Gamma_{f0}^{1-}$  function calculated by means of the constants given in Table II.

#### 4. - CONCLUSIONS. -

The experimental data on angular distribution of photofission fragments from  $^{232}\text{Th}$  has been analysed. A critical discussion on the interpretation of the experimental data leads us to the conclusion that the analysis is strongly influenced by the hypothesis on the behaviour of the function  $\alpha/(1+\alpha) = (\sigma_{\gamma}^{2+}/\sigma_{\gamma f})(\Gamma_f^{2+}/\Gamma^{2+})$ .

A comparison of the values given in Table II shows that the hypothesis  $\alpha/(1+\alpha) = \text{const.}$  shifts the  $B_0$  values at higher energies, as we expected. We notice however that our experimental data would put the  $B_0$  level position higher than the fission threshold, also with the hypothesis  $\alpha/(1+\alpha) = (2/15)c$ . Our experimental data are therefore much in favour of an interpretation of the second barrier higher than the first one, in agreement with the Lynn discussion<sup>(8)</sup>, that is of a picture where the photofission behaviour of  $^{232}\text{Th}$  is determined both for the fission threshold and for the angular distribution mainly by the second barrier.

We remark moreover, looking at Fig. 2, that the hypothesis  $\alpha/(1+\alpha) = (2/15)c$  is obviously wrong at high energies. The  $\Gamma_{f1}^{1-} / \Gamma_{f0}^{1-}$  ratio can not be higher than one while the experimental value at 8.86 MeV is well over one and well outside the statistical error.

No reliable information seems to be obtainable on both the position and height of the first potential barrier, as long as no information is available on the behaviour of the function  $\alpha/(1+\alpha)$  given by eq. (5b).

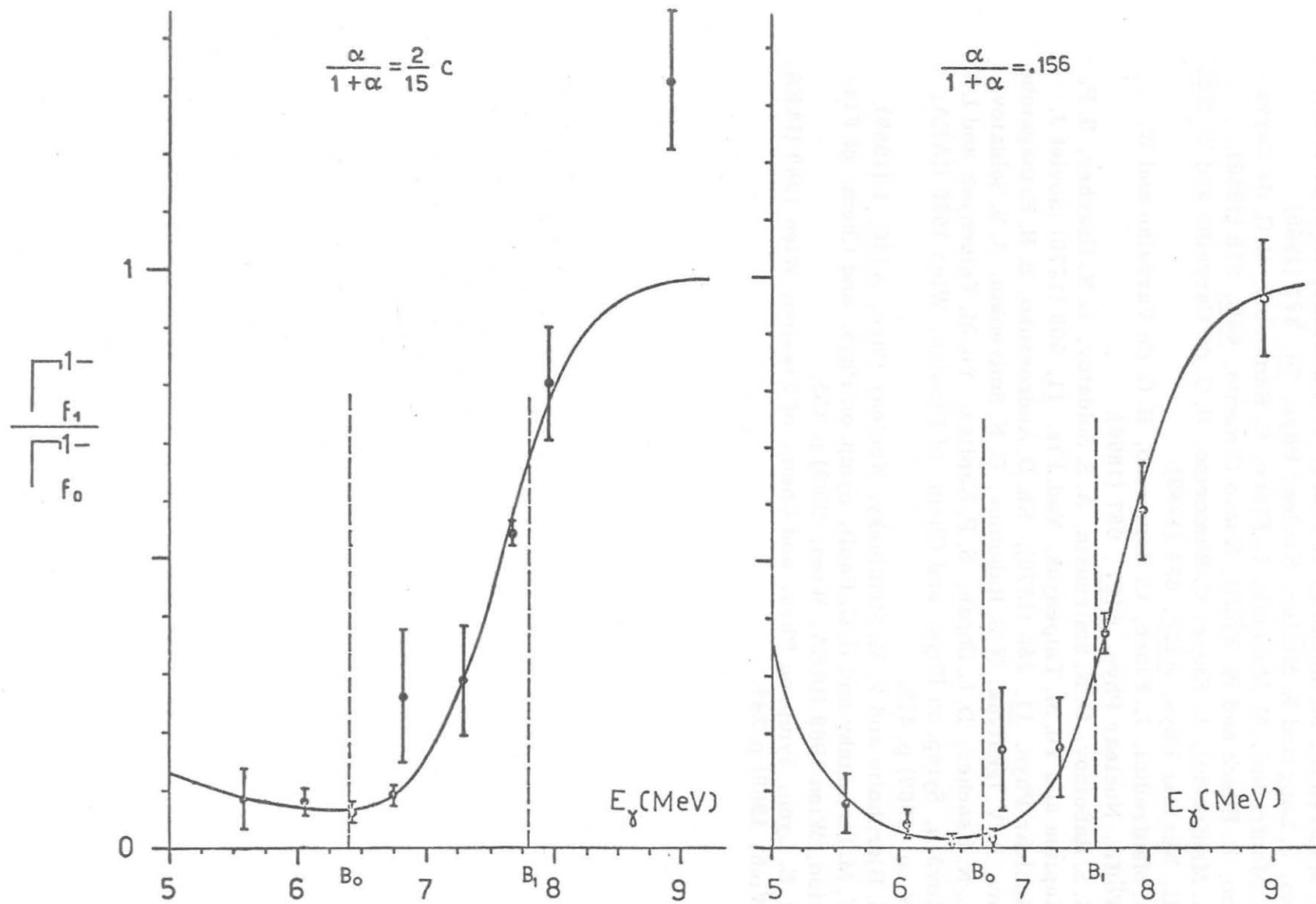


FIG. 2 -  $\Gamma_{f_1}^{1-} / \Gamma_{f_0}^{1-}$  ratio as a function of  $E_\gamma$ . The full lines are calculated by means of the parameters given in Table II. The  $B_0$  and  $B_1$  values are also shown.

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