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Abstract

Using existing experimental data for neutron-induced total, elastic, inelastic, reaction and fission cross sections, as well as results from nuclear model calculations and evaluations from nuclear reaction data libraries, we derived an estimate for the cross sections for the ^{235}U(n,2n) and ^{239}Pu(n,2n) reactions for the neutron energy range from threshold to approximately 12 MeV. In effect, our approach is based on subtracting the fission and inelastic cross sections from the total reaction cross section where the difference is expected to yield the (n,2n) cross section. In addition to this *subtraction* approach, a *ratio method* and a *differential method* have also been explored. For ^{235}U(n,2n), as a test case, we arrive at a cross section consistent with previous measurements, and for ^{239}Pu(n,2n) we obtain a peak value of 400 ± 60 mb for the incident neutron energy range of $10 \le E_n \le 12$ MeV.

I. Introduction

Recently a series of measurements using the GEANIE detector have been carried out to determine the partial cross section for the ^{239}Pu(n,2nγ) reaction. Currently nuclear theory and modeling calculations are applied to these experimental results to derive the ^{239}Pu (n,2n) total cross section, a reaction of relevance to applications in the Defense and Nuclear Technologies Program.

Previous measurements together with model calculations and evaluations of the ^{239}Pu(n,2n) cross section have been widely discussed during the past months. For an overview refer to presentations by Becker (ref.1) and an internal report by Chadwick and Young (ref.2). In summary, there exist discrepancies between the various sets of experimental data, and the various calculations and evaluations. The purpose of the GEANIE experiments and the subsequent theoretical/modeling calculations is to clarify this matter and explain the existing discrepancies. Our effort described in this report is to pursue an alternative path, based on using data already in the literature and in evaluated data libraries and on recent inelastic scattering measurements, to arrive at an estimate for the ^{239}Pu (n,2n) total cross section over the neutron energy range between threshold (near 6 MeV) and the onset of the ^{239}Pu (n,3n) contribution (near 13 MeV).

The GEANIE experiment measures the gamma rays produced from ^{238}Pu as it decays to its ground state, thus yielding a partial cross section for ^{239}Pu (n,2nγ). If the experiment could measure all the decay gamma rays, one could effectively deduce the (n,2n) total cross section above threshold directly. However, this is not experimentally possible, and therefore theory is needed to predict the factor with which one has to multiply the measured partial gamma-ray (n,2nγ) cross section to arrive at the desired (n,2n) total cross sections. This theoretical effort is currently in process and status reports have been given by Chadwick (ref. 2) and Dietrich et al. (ref. 3). To avoid the difficulties encountered in this theoretical effort, our so-called "alternate approach", based primarily on existing data and evaluations, is applied to arrive at an estimate for the (n,2n) cross section independent of the recent theoretical modeling effort used in the interpretation of the data.

II. Calculations

Our calculations depend heavily on existing high-quality data that are available for ^{235}U, ^{238}U and ^{239}Pu which can be used to independently constrain the ^{239}Pu (n,2n) cross section. Our calculations will be limited to the energy region above threshold and below the onset of other competing processes, such as (n,3n). The reason that measurements with other neighboring actinide nuclei can provide useful information on the ^{239}Pu (n,2n) cross section is that in many regards these nuclei are similar and can often be simply related to each other. On several occasions we shall choose ^{238}U (for which most experimental data exist) as a surrogate for the neighboring nuclei. In particular, we assume that the ^{238}U+n direct interaction is a good model for the ^{239}Pu+n direct interaction.

IIA. Subtraction Method

The simplest statement one can make regarding neutron-induced reaction cross sections is that the total cross section, σ_T, is the sum of all its parts. Because of the large Coulomb barrier in actinide nuclei, charged-particle exit channels can be ignored (reaction cross sections <10 mb). Also the capture cross section (n,γ) is known to be negligible. Hence, we can write

$$\sigma_T = \sigma_{elast} + \sigma_{D.I.} + \sigma_{(n,f)} + \sigma_{(n,n')} + \sigma_{(n,2n)} \quad (1)$$

where σ_T is the total neutron cross section,

σ_{elast} is the elastic scattering cross section,

$\sigma_{D.I.}$ is the direct inelastic scattering cross section,

$\sigma_{(n,f)}$ is the fission cross section

$\sigma_{(n,n')}$ is the inelastic cross section

$\sigma_{(n,2n)}$ is the (n,2n) cross section.

Defining the reaction cross section, σ_{react}, as

$$\sigma_{react} = \sigma_T - \sigma_{elastic} \tag{2}$$

From Eq. 1 and 2 one can derive the equation for the (n,2n) process as

$$\sigma_{(n,2n)} = \sigma_{react} - \sigma_{D.I.} - \sigma_{(n,f)} - \sigma_{(n,n')} \tag{3}$$

Thus, if we know the quantities on the right-hand side of Eq. 3, we can calculate the (n,2n) cross section of the respective nucleus by using this so-called "Subtraction Method" for the energy range from threshold to the onset of the (n,3n) reaction. In the following we shall discuss the quantities we need for the input to this subtraction method.

Let us assume that we know the three main contributors to the right-hand side of Eq. 3, i.e., σ_{react} , $\sigma_{(n,f)}$ and $\sigma_{(n,n')}$, to an accuracy of about 5 % each; thus we can expect to achieve an accuracy for $\sigma_{(n,2n)}$ of slightly better than 10 % (assuming $\sigma_{D.I.}$ and its error are small compared to the other terms). The fission cross sections for ^{235}U and ^{239}Pu are available at the desired accuracy. In fact, we take the values for $\sigma_{(n,f)}$ from recent evaluations in ENDL, where evaluators claim an accuracy of ±2 %.

The reaction cross section is the largest contributor in Eq. 3. It is of the order of 2.5 to 3.0 barn in the energy range of interest. For our calculation we used the surrogate ^{238}U since we expect only small variations with respect to neighboring nuclei. Using a simple Ramsauer model we can derive an estimated reaction cross section given by $\sigma_{react} = \pi (R + \lambda\!\!\!^{-})^2$ where R is the nuclear radius derived from the total neutron cross section (see Ref. 4) and $\lambda\!\!\!^{-}$ is the reduced wavelength of the incoming neutron. Results of this estimate as a function of energy are shown in Fig. 1.

A second estimate of the reaction cross section was obtained using Optical Model calculations (Ref. 5). Again the surrogate nucleus ^{238}U has been used. Results from two separate fits to the total cross section have been obtained, referred to as FLAP 1.5 and FLAP 2.2, respectively. From inspection of the two separate Optical Model calculations, the parameters for the FLAP 2.2 fit appear to be the preferred values (ref.5). Results from the FLAP 2.2 fit are shown in Fig. 1, together with the average derived from the Ramsauer model and the Optical Model FLAP 2.2.

The Optical Model calculations were also applied to calculate the direct inelastic scattering. The cross sections for D.I. scattering by both the 2+ and the 4+ states have been included. Typically this cross section is about one order of magnitude smaller compared to σ_{react}. This quantity $\sigma_{D.I.}$ is subsquently to be subtracted from the reaction cross sections, which were arrived at either by way of the Ramsauer or the Optical Model methods. In effect, this difference $(\sigma_{react} - \sigma_{D.I.})$ is plotted in Fig. 1.

As the next step, the fission cross section is subtracted from the difference $(\sigma_{react} - \sigma_{D.I.})$. The results for ^{235}U and ^{239}Pu are shown in Fig. 2. For this illustration the average derived from the Ramsauer model and the Optical Model FLAP 2.2 (see above) has been used for the reaction.

As the last step, we have to subtract the inelastic (n,n') cross section. To arrive at this quantity we rely on recent gamma-ray measurements from GEANIE. Gamma-ray intensities have been measured in the decay of ^{235}U(n,n' γ) and ^{239}Pu(n,n' γ), respectively. We have the choice of many gamma rays in the two nuclei. However, we have chosen the most intense gamma transitions for this purpose to be able to keep the statistical uncertainty to a minimum. We normalize this gamma intensity at an energy below threshold (at approx. 5.0 to 5.5 MeV) in order to set $(\sigma_{react} - \sigma_{D.I.} - \sigma_{(n,f)})$ equal to $\sigma_{(n,n')}$, i.e., to force the (n,2n) cross section below threshold to zero (refer to Eq. 3). We further assume that the ratio of gamma intensity to the total (n,n') excitation is constant over the energy range of investigation $(5 \leq E_n \leq 12$ MeV). For the case of ^{235}U we have chosen the 129-keV gamma transition from the $5/2^+$ excited state to the $7/2^+$ ground state. For ^{239}Pu we have chosen two gamma rays (228 keV and 278 keV) which deexcite the 5/2+ state at 285 keV. (The sum of two gamma rays was used to improve the statistics.) Other gamma transitions in ^{235}U and in ^{239}Pu have also been used in the analysis obtaining similar results, but with poorer statistics.

The normalized gamma-ray intensity per neutron flux for the gamma transitions in ^{235}U and ^{239}Pu for the cases chosen above are illustrated as a function of energy in Fig. 3 and 4, respectively. As pointed out above, normalizing this gamma intensity per neutron flux below threshold to yield zero for the (n,2n) process, and subtracting this normalized gamma intensity (which is assumed to be equivalent to $\sigma_{(n,n')}$) from (σ_{react} - $\sigma_{D.I.}$ - $\sigma_{(n,f)}$) according to Eq. 3, yields the estimates for the (n,2n) cross sections for ^{235}U and ^{239}Pu. These estimates are shown in Fig. 5 and 6, respectively.

It is to be noted that this approach yields an (n,2n) cross section only between threshold and the onset of the (n,3n) process. But it should give a reliable estimate across the maximum near 10-11 MeV. We also recall two severe limitations of this approach: One is due to the fact that we subtract large numbers from large numbers, i.e., (σ_{react} - $\sigma_{(n,f)}$ - $\sigma_{(n,n')}$). The fission cross section is the best known of the three quantities to an accuracy of about 2%. The reaction cross section as shown in Fig. 1 is uncertain to about ± 100 mb. A sensitivity study has shown that this uncertainty is partially cancelled by our method of normalizing the (n,n') cross section to (σ_{react} - $\sigma_{D.I.}$ - $\sigma_{(n,f)}$) between 5.0 to 5.5 MeV. The predominant error, we believe, is introduced by uncertainties in the (n,n') cross section. It is important that the gamma intensities are to be corrected for "wrap-around" in the time-of-flight spectrum. Relatively high neutron rep rates and short flight paths cause neutrons from adjacent pulses to interfere with each other. These effects are currently under investigation. Furthermore, we assumed that the ratio of the intensity of the chosen gamma ray to the (n,n') cross section is constant over the energy range under investigation, although different reaction processes come into play. At 8 to 12 Mev the pre-equilibrium process starts to make a significant contribution to inelastic scattering. This has been clearly observed by Kammerdiener et al (ref. 6) and Baba (ref.7). These results can provide a guide to the difference in pre-equilibrium contributions for ^{235}U, ^{238}U and ^{239}Pu.

IIB. Ratio Method

Assuming that the (n,2n) cross section for a reference nucleus (either ^{235}U or ^{238}U) is known, we can use this information to arrive at an estimate for $\sigma_{(n,2n)}$ for ^{239}Pu. This method should eliminate some of the uncertainties discussed above. From Eq. 3 one can derive the following relation:

$$\sigma_{(n,2n)}(239) = \left(\frac{\sigma_{react}(239) - \sigma_{D.I.}(239) - \sigma_{(n,f)}(239) - \sigma_{(n,n')}(239)}{\sigma_{react}(ref) - \sigma_{D.I.}(ref) - \sigma_{(n,f)}(ref) - \sigma_{(n,n')}(ref)}\right)\sigma_{(n,2n)}(ref) \tag{4}$$

where (239) refers to the ^{239}Pu nucleus and *(ref)* to either ^{235}U or ^{238}U used as reference nucleus.

The advantage of using Eq. 4 is that the uncertainty in $\sigma_{(n,n')}$ which was discussed above, has very little impact on the ratio, since $\sigma_{(n,n')}$ is considerably smaller than the other terms on the right-hand side of Eq. 3. However, we still have to know the reaction cross section to within a reasonable accuracy since it is the largest contribution to the numerator and denominator. For (σ_{react} - $\sigma_{D.I.}$) the average values of the Ramsauer model and the Optical Model FLAP 2.2 have been used for our calculation (see Fig.1). There is also a correction to be considered when comparing ^{239}Pu with ^{238}U, since the latter is an even-even nucleus with a higher threshold for the (n,2n) reaction compared to ^{239}Pu.

As an illustration of this method, we show results of this ratio approach comparing ^{239}Pu with ^{235}U as a function of energy in Fig. 7.

IIC. Differential Method

Again, assuming that the (n,2n) cross section for a reference nucleus (either ^{235}U or ^{238}U) is known, we can use this information to arrive at an estimate for $\sigma_{(n,2n)}$ for ^{239}Pu by using a differential approach. This method should also eliminate some of the uncertainties discussed above. From Eq. 3 one can derive the following relation:

$$\sigma_{(n,2n)}(239) = \sigma_{(n,2n)}(ref) + \Delta\sigma_{react} - \Delta\sigma_{D.I.} - \Delta\sigma_{(n,f)} - \Delta\sigma_{(n,n')} \tag{5}$$

where Δ designates the difference of cross sections of ^{239}Pu nucleus and the reference (*ref*) nucleus which, as in Eq. 4, is usually ^{235}U or ^{238}U. Otherwise the same notation applies as above.

The advantage of using Eq. 5 is that, though σ_{react} is not too well characterized and contributes to a large degree to the final error in the two methods described above, in this differential method the value for the difference $\Delta\sigma_{react}$ is known to within 30 mb. Also $\Delta\sigma_{D.I.}$ continues to make only a slight contribution to the total error. In addition the difference $\Delta\sigma_{(n,f)}$ is very well determined. In a series of time-of-flight experiments the fission cross sections of transuranic elements have been measured with great precision relative to the fission cross section of ^{235}U (see ref.8). On the other hand, the difference $\Delta\sigma_{(n,n')}$ is not well characterized. For this reason it is advantageous, in addition to the differential method (Eq. 5) also to apply Eq. 4, the ratio method, for an additional estimate for the $\sigma_{(n,2n)}$ cross section for ^{239}Pu.

Using the cross section data from ENDF/B-VI we have made an attempt to calculate the (n,2n) cross section for ^{239}Pu at the peak value at the neutron energy near 11 MeV using Eq.5. The values and estimated errors for the individual terms in Eq. 5 are listed in Table 1. As seen in the table we arrive at 400 ± 60 mb for the estimated value for $\sigma_{(n,2n)}$ for ^{239}Pu at the neutron energy of 11 MeV.

As an illustration of the differential method, we present results in Fig. 8 for the (n,2n) cross section of ^{239}Pu compared to ^{235}U over the neutron energy range from 5 to 15 MeV. For the purpose of this comparison we have chosen to use the cross sections as listed in the ENDF/B-VI files.

IID. High Energy Approximation

It is to be noted that at energies well above threshold the statistical contribution to the (n,n') cross section goes rapidly to zero. Thus for energies above 10 MeV we can get an additional estimate of the (n,2n) cross section by subtracting the pre-equilibrium (n,n') cross section from the reaction cross section on the right-hand side of Eq. 3. At 14 MeV there are two independent measurements of the pre-equilibrium cross section reported by Kammerdiener (ref.6) and by Baba (ref.7). From these measurements we deduce a pre-equilibrium cross section of 300 mb at 14 MeV for ^{239}Pu.. In order to obtain estimates for the pre-equilibrium process at lower energies we use a $E^{-1/2}$ scaling. As shown in Fig. 9 our normalized gamma ray cross section is only 230 mb. So it is necessary to include an additional 70 mb in the reaction cross section, so that our (n,n') cross section is renormalized to 300 mb at 14 MeV.

The (n,2n) cross section $\sigma_{(n,2n)}$ for ^{239}Pu as determined by this high energy approximation is shown in Fig. 10. At the peak energies from 10 to 12 MeV, this approximation yields a value for of 360 ± 60 mb . This is in agreement with the results obtained by the other methods reported in this paper. It is also to be noted that above 12 MeV the data derived by this high energy approximation are indicated as upper limits only (see Fig. 10), because above 12.5 MeV they also contain contributions from the (n,3n) process.

III. Summary

We have used existing experimental data, results from nuclear model calculations and evaluations from nuclear data libraries for neutron-induced cross sections to derive estimates for the ^{239}Pu (n,2n) cross section for the energy range from threshold to approximately 12 MeV where the (n,3n) reaction sets in. Three interrelated methods were used to arrive at the estimated cross sections, each having a certain degree of advantage over the other two. The *subtraction method* (in Section IIA) suffers from the necessity of having to subtract relatively large numbers from large numbers, thus yielding large errors. However, it does not require a neighboring nucleus to be used as a reference nucleus, though ^{238}U is utilized as a surrogate on several occasions.

The *ratio method* (in Section IIB) uses a (n,2n) cross section of a reference nucleus, either ^{235}U or ^{238}U. Care must be exercised when comparing an odd-even nucleus such as ^{239}Pu with an even-even nucleus because of differences in reaction thresholds, such as (n,f) and (n,2n) reactions. However, some of the larger errors encountered in the subtraction method are avoided in the ratio method, especially for $\Delta\sigma_{(n,n')}$ since this cross section, though not very well characterized, is significantly smaller relative to the others and its contribution to the overall error is less significant in the ratio method.

The *differential method* (in section IIC) also uses the (n,2n) cross section of a reference nucleus and the same considerations apply as for the ratio method. The advantage of the differential method is that the difference $\Delta\sigma_{react}$ is very small and well defined and has a small error. Furthermore, the fission cross section difference $\Delta\sigma_{(n,f)}$ is very well characterized and has a small error because of the relative measurements of the cross sections discussed above.

Our "Alternate Approach" to arrive at an estimate for the ^{239}Pu(n,2n) has been successful and has been used to independently constrain the cross section in the incident neutron energy range of $10 \leq E_n \leq 12$ MeV. Our simplified methods are currently being supplemented by a theoretical physics approach using detailed nuclear reaction and nuclear structure model calculations to arrive at the ^{239}Pu(n,2n) cross section over the whole energy range from threshold to the onset of the (n,3n) reaction.

The techniques described in this report have also recently been refined using more experimental data and sophisticated statistical analysis. The optimum results using the alternate approach discussed here have been presented in a recent report by Navratil and McNabb (ref. 9).

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Figure Captions

Fig. 1. The difference of the reaction cross section minus the direct inelastic scattering cross section (σ_{react} - $\sigma_{D.I.}$) for the surrogate nucleus ^{238}U plotted for the Ramsauer model and the Optical Model FLAP 2.2 calculations as well as for the average of the two models. The three curves illustrate the degree of uncertainty in σ_{react} used in Step 1 of the Subtraction Method .

Fig. 2 The difference of the reaction cross section minus the direct inelastic scattering cross section (σ_{react} - $\sigma_{D.I.}$) for the surrogate nucleus ^{238}U plotted for the average of the Ramsauer model and the Optical Model calculation FLAP 2.2 calculations (upper curve), minus the fission cross section $\sigma_{(n,f)}$ for ^{235}U and ^{239}Pu (two lower curves). The two lower curves are used in Step 2 of the Subtraction Method.

Fig.3 The normalized gamma-ray intensity per neutron flux plotted for the 129-keV gamma transition in ^{235}U. The data have not been corrected for possible "wrap-around" in the time-of-flight spectrum. In Step 3 of the Subtraction Method, the measured gamma intensity has been normalized at 5.0 to 5.5 Mev (below the (n,2n) threshold) to yield zero for the (n,2n) process at this energy.

Fig.4 The normalized gamma-ray intensity per neutron flux plotted for the sum of two gamma rays (228 keV and 278 keV) which deexcite the same state in ^{239}Pu. The data have not been corrected for possible "wrap-around" in the time-of-flight spectrum. In Step 3 of the Subtraction Method, the measured gamma intensity has been normalized at 5.0 to 5.5 Mev (below the (n,2n) threshold) to yield zero for the (n,2n) process at this energy.

Fig.5 Illustration of the Subtraction Method: The (n,2n) cross section for ^{235}U as determined using Eq. 3. For comparison, the existing experimental data and theoretical predictions for ^{235}U(n,2n) are shown in Fig. 5a.

Fig.6 Illustration of the Subtraction Method: The (n,2n) cross section for ^{239}Pu as determined using Eq. 3. For comparison, the existing experimental data and evaluations for ^{239}Pu(n,2n) are shown in Fig. 6a.

Fig.7. Illustration of the Ratio Method: The (n,2n) cross section for ^{239}Pu as determined using Eq. 4. The measured (n,2n) cross section for ^{235}U is used as reference in this example.

Fig.8. Illustration of the Differential Method: The (n,2n) cross section for ^{239}Pu as determined using Eq. 5. The evaluated (n,2n) cross section for ^{235}U is used as reference in this example.

Fig.9. The normalized gamma-ray intensity per neutron flux plotted for the sum of two gamma rays (228 keV and 278 keV) which deexcite the same state in ^{239}Pu, the same as Fig. 4, except the normalization is to a reaction cross section reduced by 70 mb. The points marked (x) are estimated for the pre-equilibrium process using $E^{-1/2}$ scaling (see Sect.IID. High Energy Approximation).

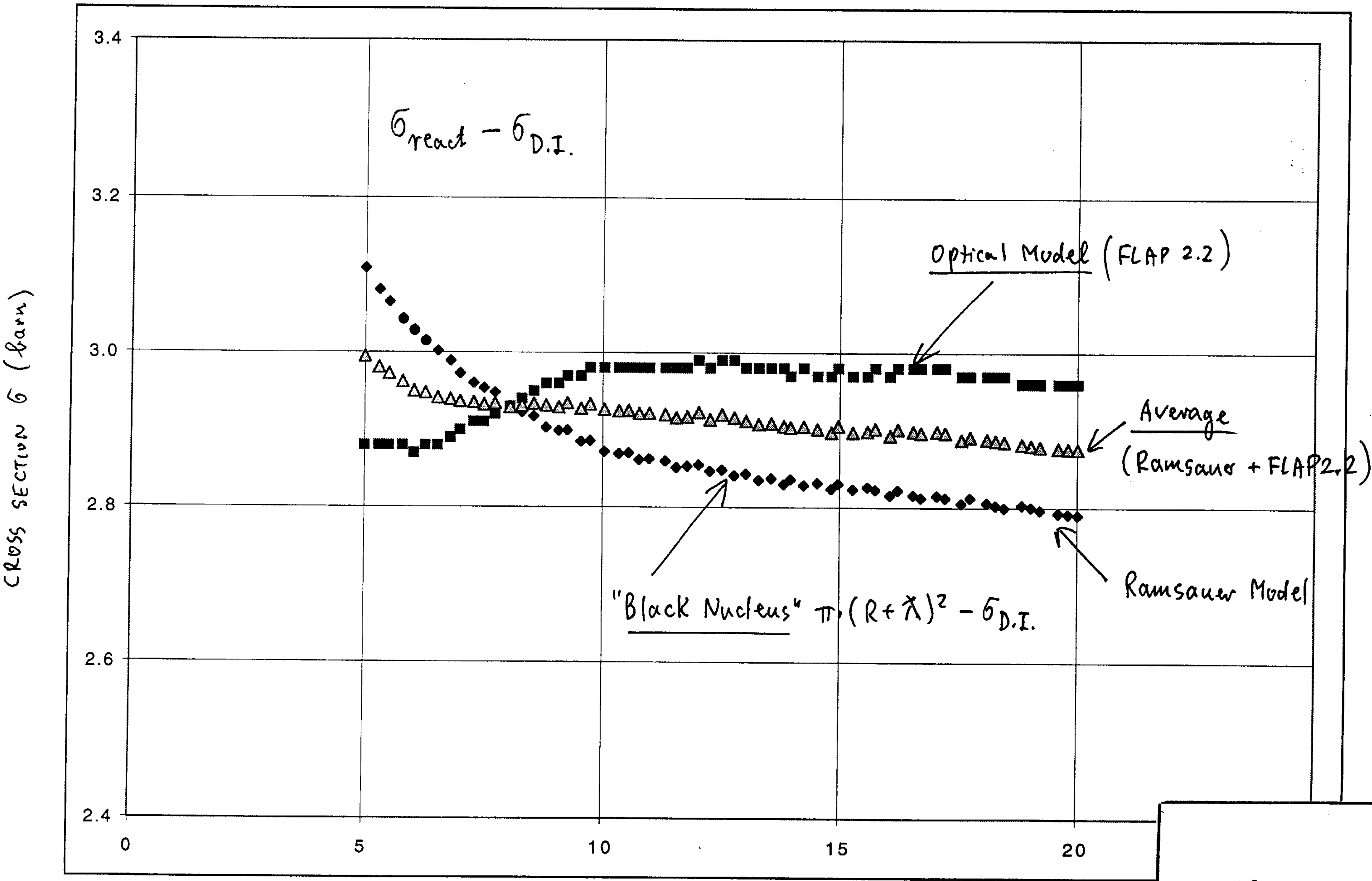
Fig.10 Illustraion of the High Energy Approximation: The (n,2n) cross section for ^{239}Pu as determined using the Subtraction Method (Eq.3), including a pre-equilibrium cross section of 300 mb (see Sect. IID). The points marked (x) are the estimates based on the pre-equilibrium $E^{-1/2}$ scaling.

Table I. Values of the cross sections and their estimated errors to be inserted in Eq. 5 for an estimate of the (n,2n) cross section for ^{239}Pu at the maximum near the neutron energy of 11 MeV. For this illustration, the cross section values used have been taken from the cross section library ENDF/B-VI. Δ designates the difference of cross sections of ^{239}Pu minus ^{235}U.

Cross Section	Value
$\sigma_{(n,2n)}$ (^{235}U)	830 ± 40 mb
Δ (σ_{react} - $\sigma_{D.I.}$) ($^{239}Pu - ^{235}U$)	30 ± 30 mb
$\Delta\ \sigma_{(n,f)}$ ($^{239}Pu - ^{235}U$)	500 ± 25 mb
$\Delta\ \sigma_{(n,n')}$ ($^{239}Pu - ^{235}U$)	-40 ± 25 mb *
$\sigma_{(n,2n)}$ (^{239}Pu)	400 ± 60 mb

* The inelastic cross section for ^{235}U is larger that that for ^{239}Pu per ENDF/B-VI.

92-U-238-non-el-2a-t1 Sheet1 Chart 2 T-U, AD-AE 12-148 9.21.00



 → E_n (MeV)

Fig. 1

92- U- 238- non- el - 2a3 - t   Sheet1 Chart 2   T-U, Z-AA / 12-148   9.21.00

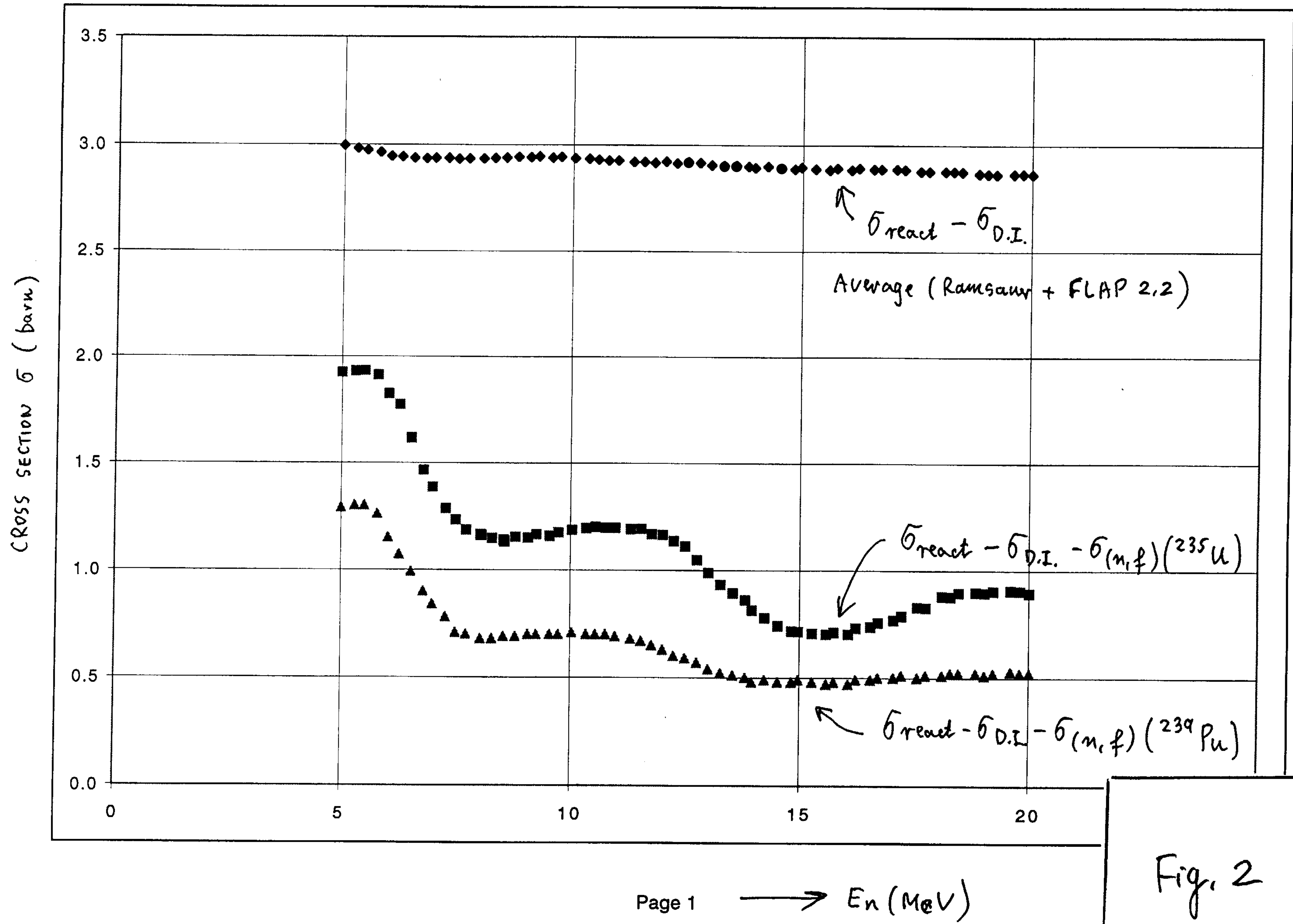


Fig. 2

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K-N
42-63

9.21.00

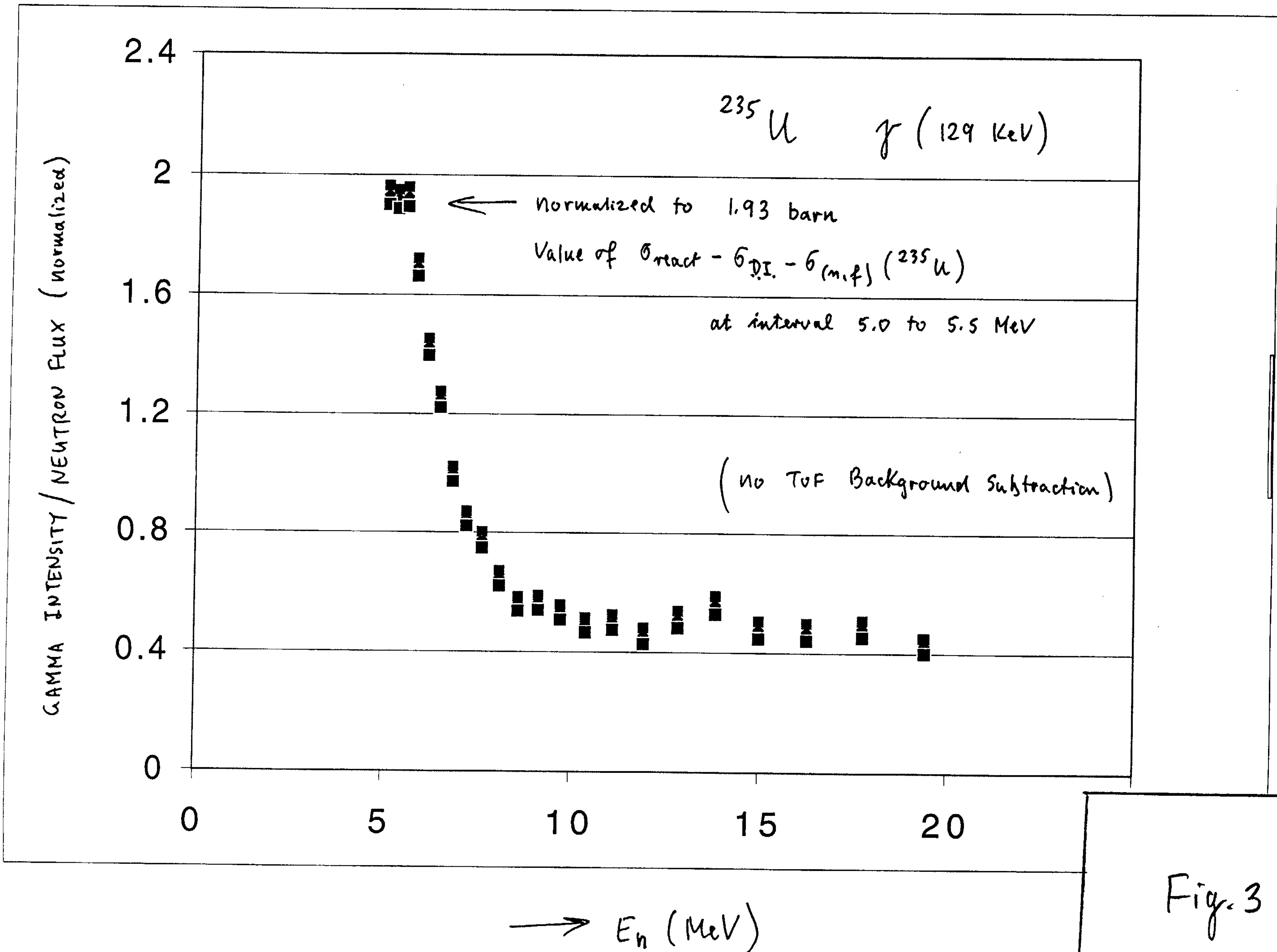
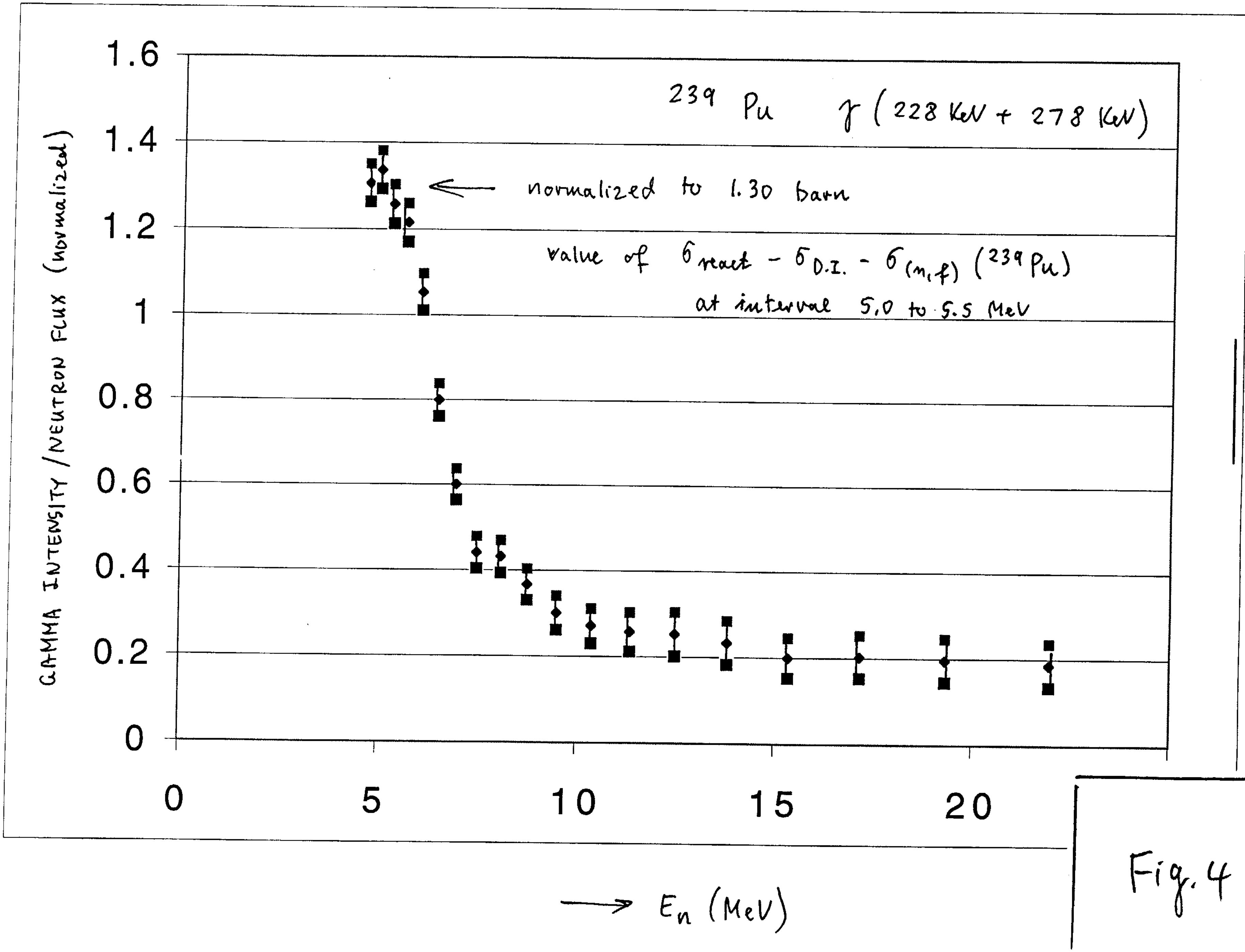


Fig. 3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1.4
1.2
1
0.8
0.6
0.4
0.2
0
0
5
10
15
20
GAMMA INTENSITY / NEUTRON FLUX (normalized)
239 Pu γ (228 KeV + 278 KeV)
normalized to 1.30 barn
value of σ react - σ D.I. - σ (n,f) (239 Pu)
at interval 5.0 to 5.5 MeV
→ En (MeV)

Fig. 4

JAB- 1fc2-238 foil- 129-sa

$\frac{Q-T}{42-63}$

9.21.00

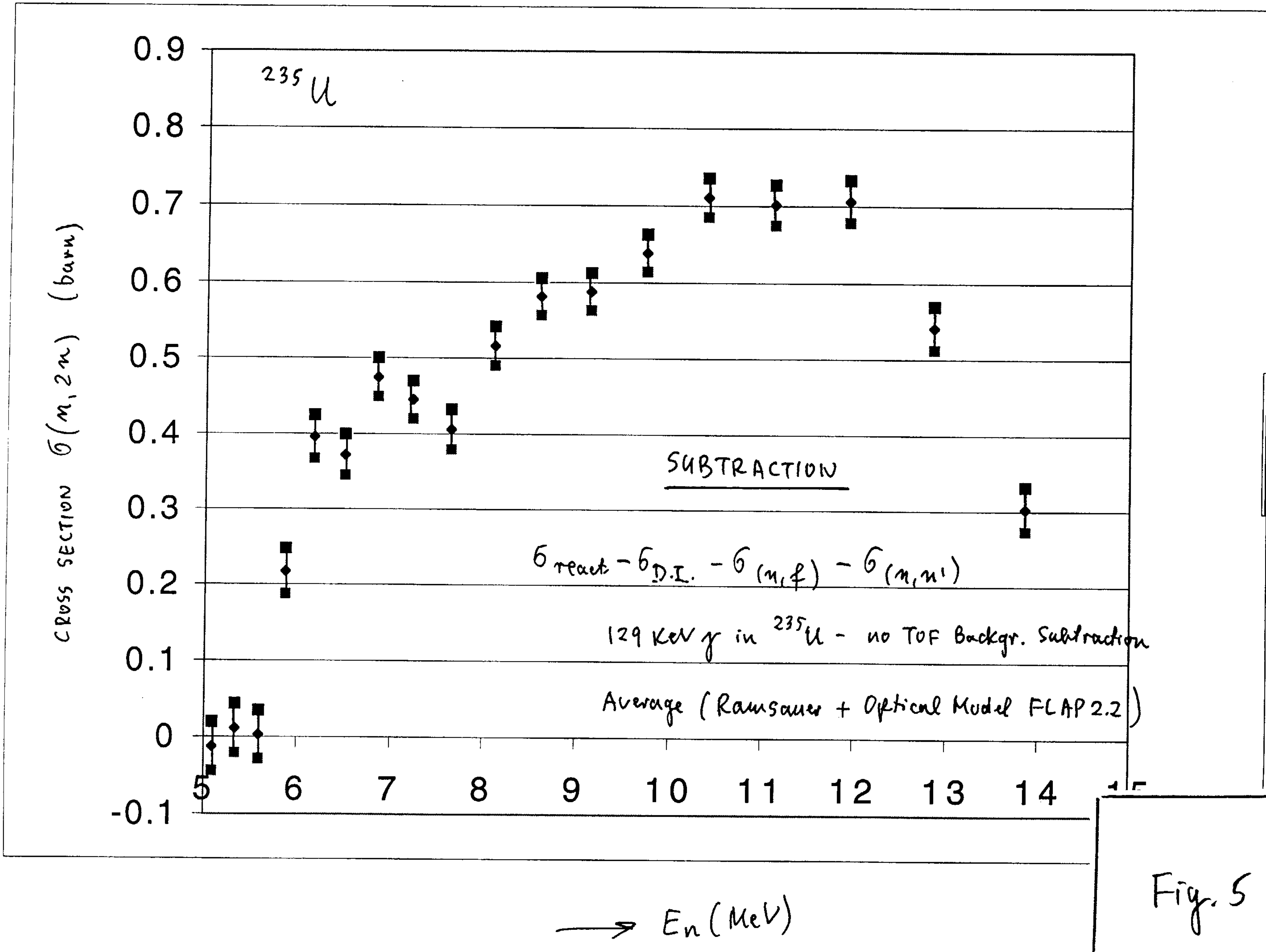
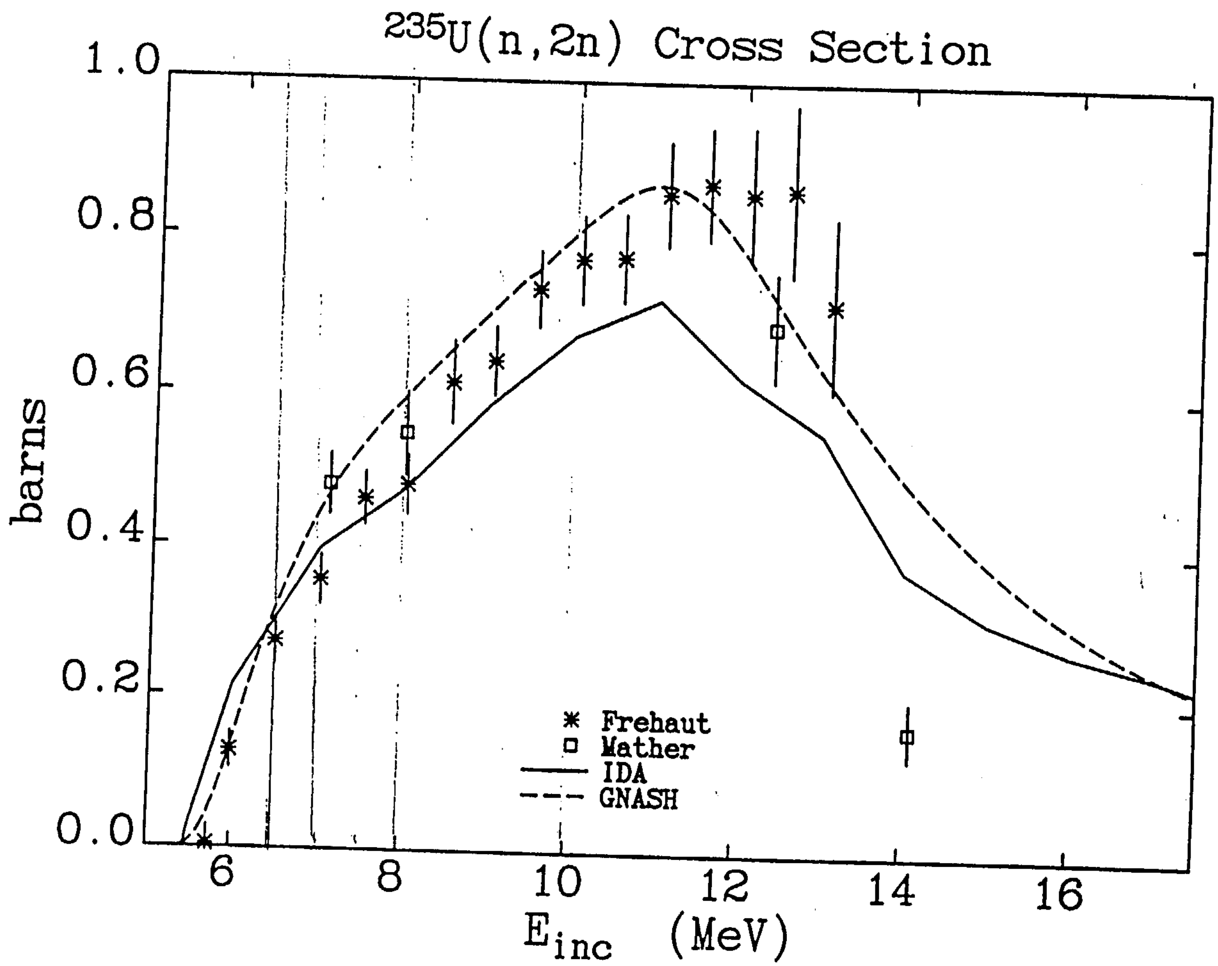


Fig. 5



Total ^{235}U$(n,2n)^{234}$U cross section, as a function of incident neutron energies, calculated by IDA and measured by Frehaut and Mather.

Fig. 5a

LAB- 239 Pu -285- aa4

Q - T

39 - 57

9.21.00

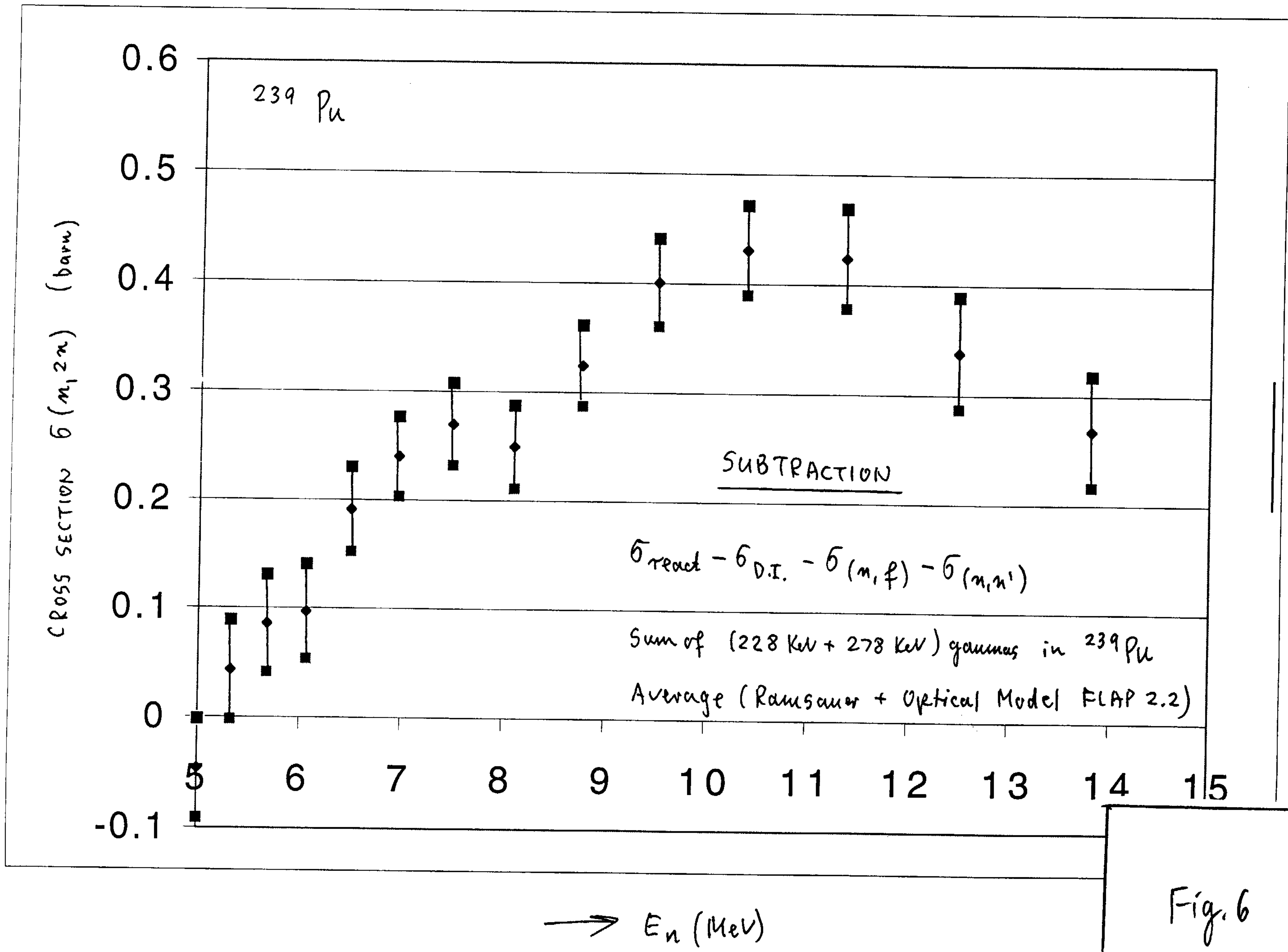
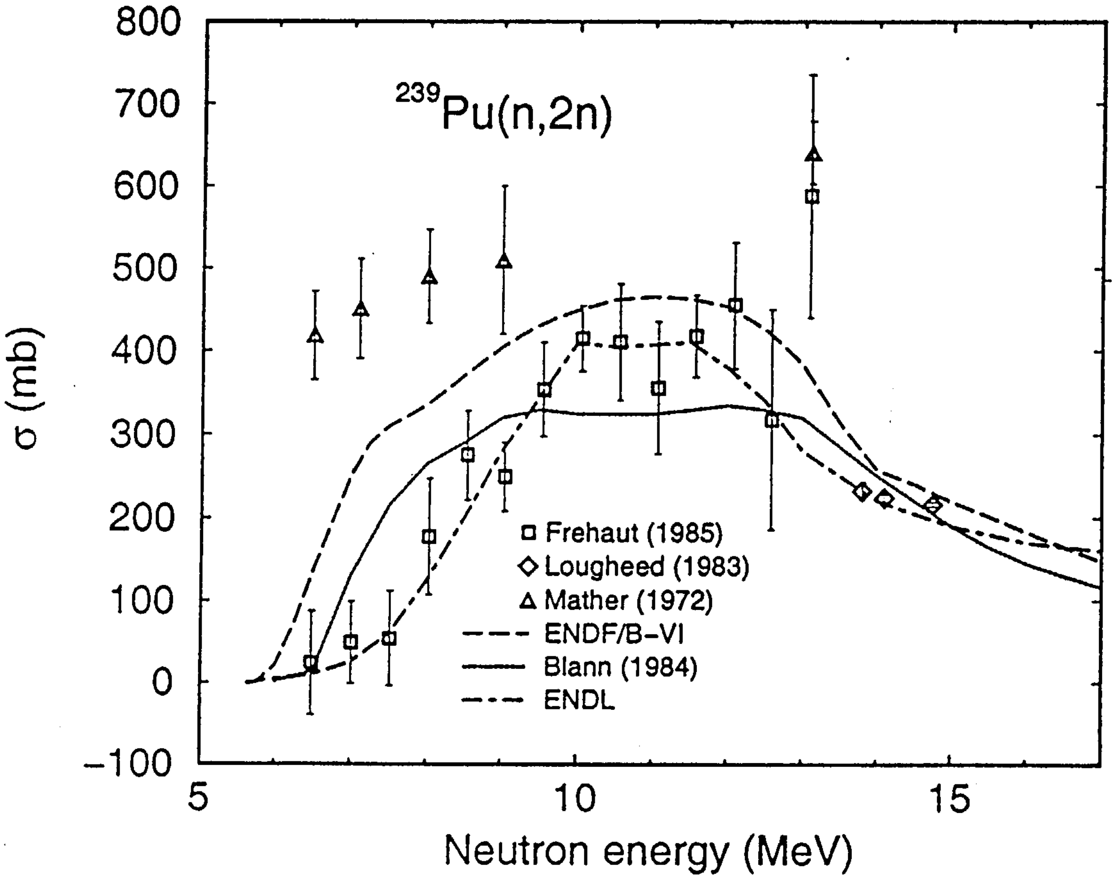


Fig. 6



The ^{239}Pu($n, 2n$ excitation function. Previous measurements, calculations, and evaluations, are shown.

Fig. 6a

9.22.00

JAB- 1fc2- 238 foil- 129- sb1

X - AA

42 - 63

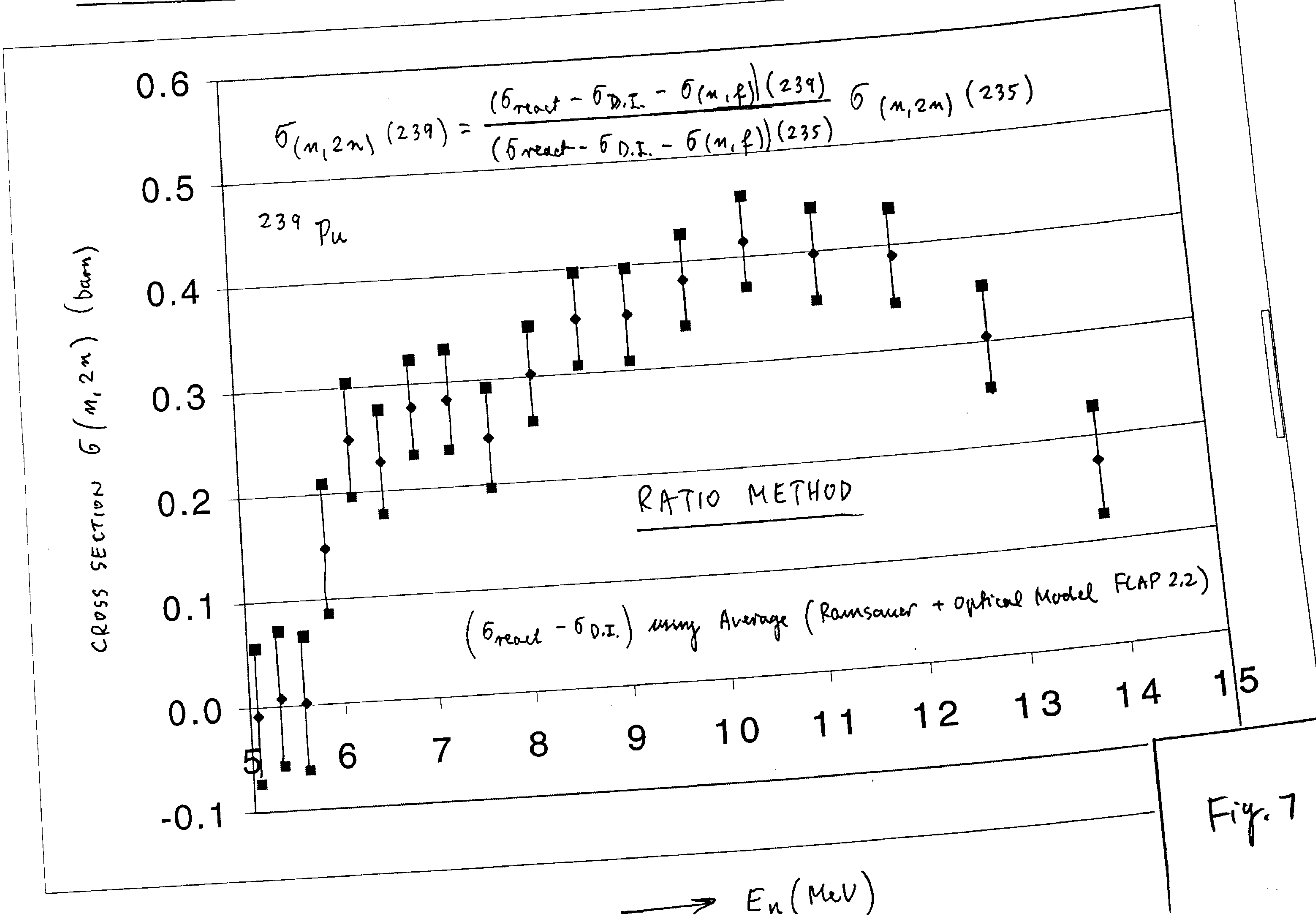


Fig. 7

9.26.00

239- 235 (n, 2n) diff - a

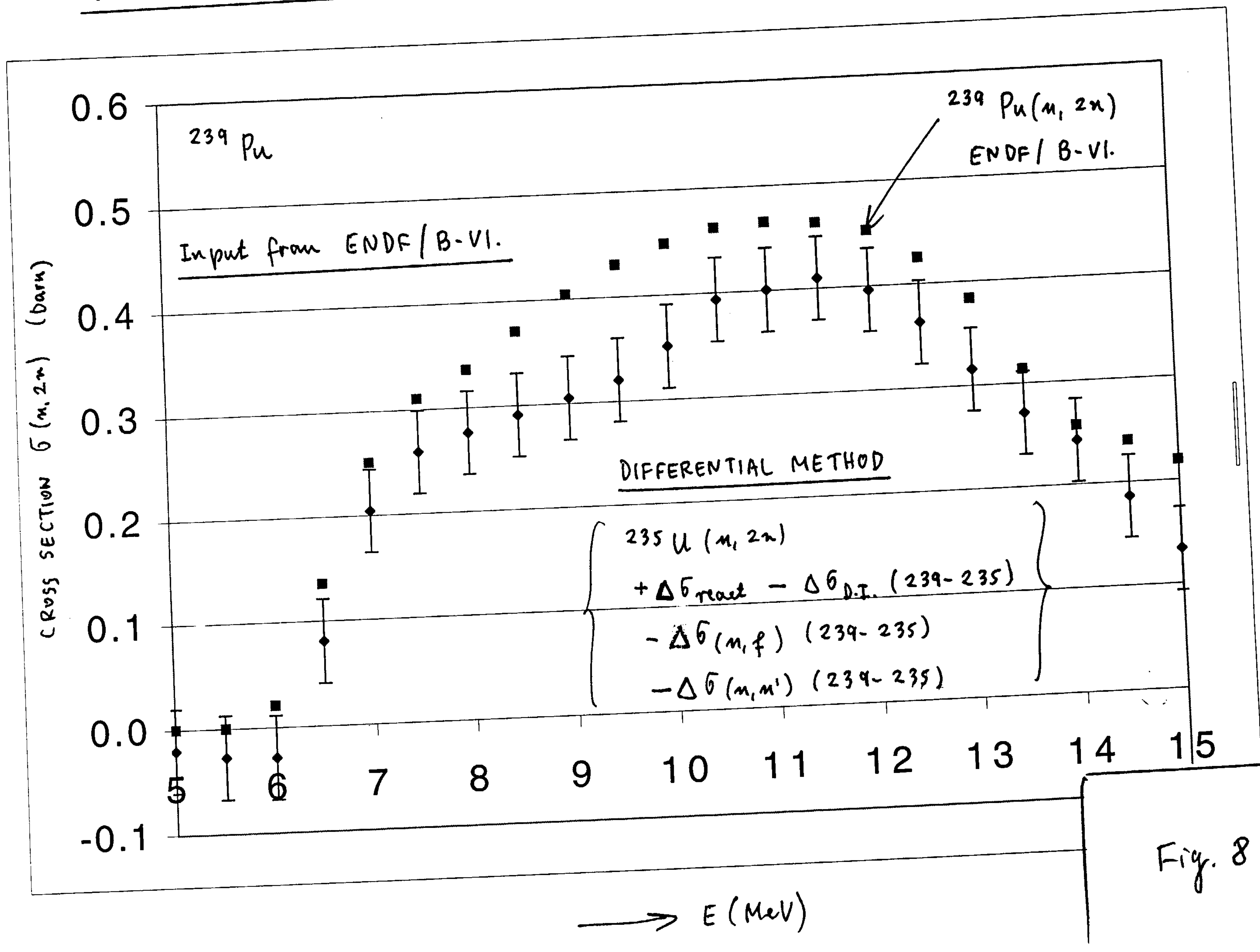


Fig. 8

LAB- 239 Pu- 285- aa5

K - 0
39- 57

9.22.00

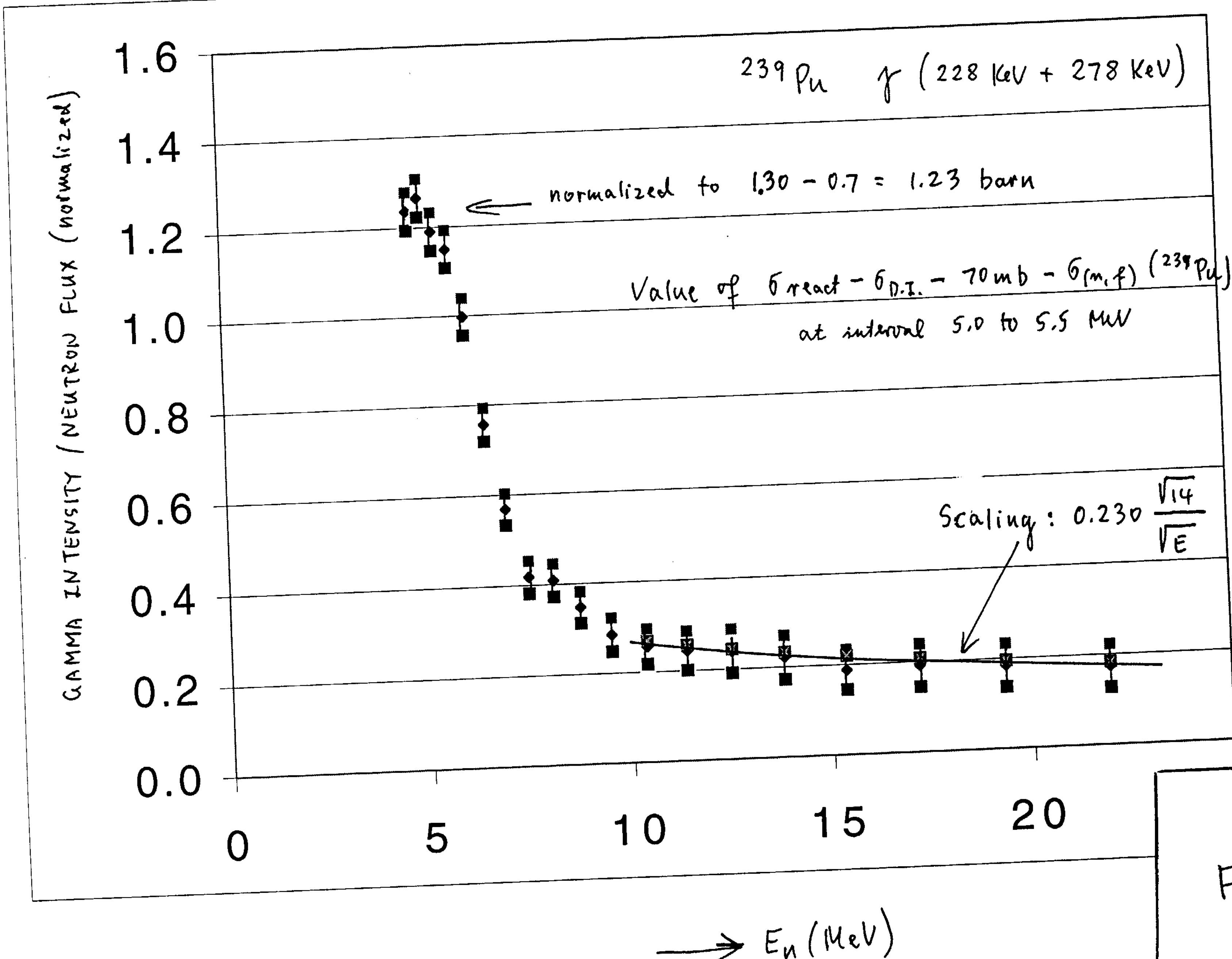


Fig. 9

LAB- 239 Pu - 285 - aa 5

R-V

39- 57

9.22.00

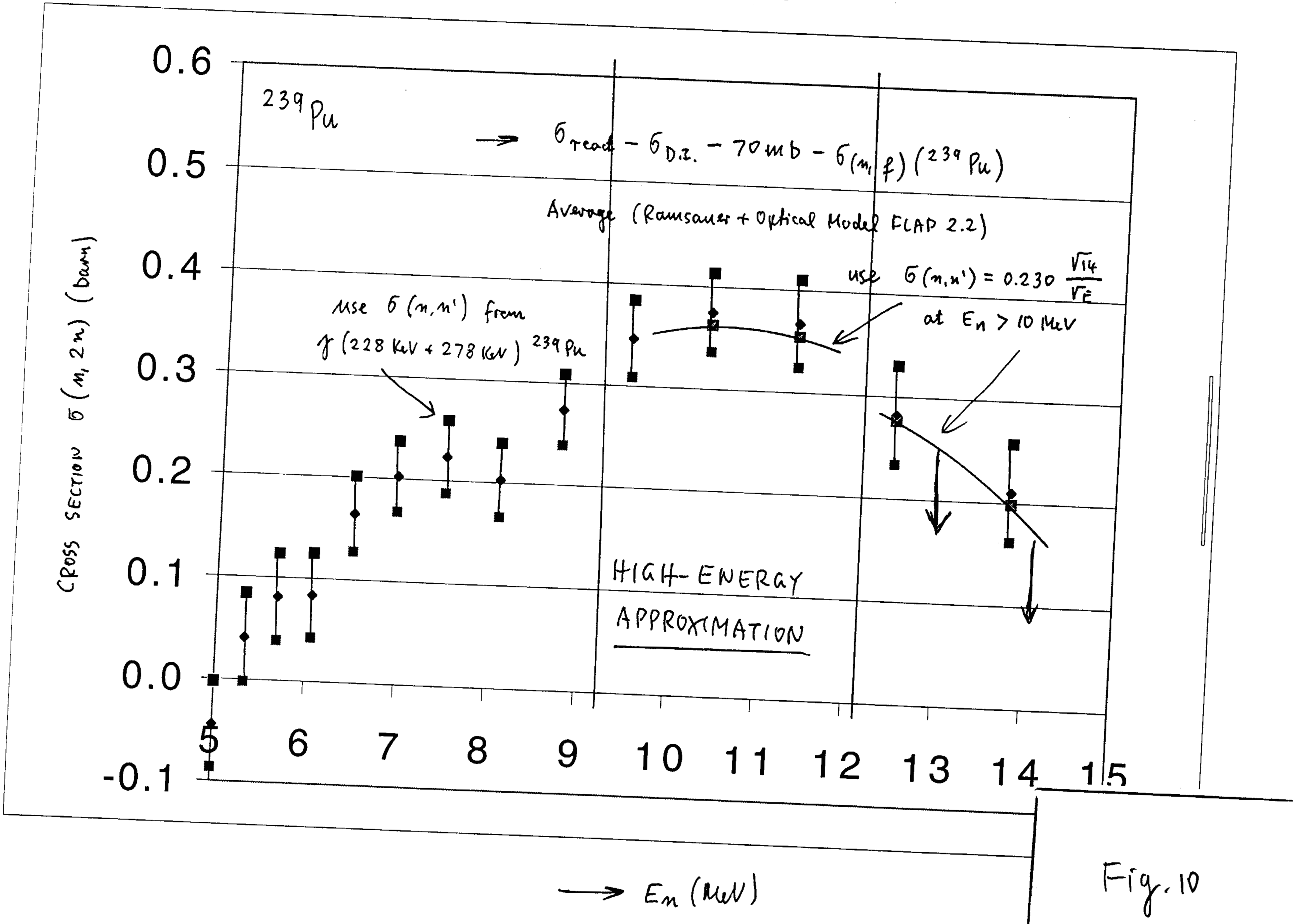


Fig. 10