239 Pu neutron cross section evaluation

from the Kev region to 20 Mev

Preliminary results

E.FORT

Abstract

In the energy range of interest the bulky experimental data base of 239 Pu has two main characteristics:

_The data related to the neutron channels are of good quality permitting a correct Optical Model parametrization for the derivation of reliable neutron penetrabilities to be used in the H.F statistical calculations.

-Concerning the reaction x-sections the reality is much more contrasted. Among the accumulation of data it seems difficult to extract reliable informations with an accuracy better than 5% especially in the range 30 Kev to 1 Mev.

This situation has suggested the use of all the possible sources of information.

The presented evaluation expresses the convergence of the experimental, theoretical and integral informations.

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I INTRODUCTION

The importance of 239Pu for energetic applications does not need to be justified anymore:the design of LMFBR and of reactor with pitch lattice involves reactivity and fuel cycle calculations which in turn require data covering the energy range between thermal energy and 20 Mev.Because of these needs a large experimental effort has been concentrated on Plutonium . Since the first measurement (referenced in the year 1943 (TASHEK-LANL 90kev-1.5 Mev)) on fission, all the x-sections of 239 Pu have been measured. Even the elastic and inelastic x-sections, although it is difficult to separate both processes because of the closeness of the ground and the first excited states ,have been measured or deduced through angular distribution or "pseudo elastic scattering"cross section measurements. There are also numerous experimental data for microscopic parameters like **alpha** or **eta** which are related to the integral parameter **k+**.

From this inventory of data it could be inferred that all the information needed for a meaningful evaluation is available. Actually all the successive major evaluations of this nucleus differ significantly from one another.

A good example is given by the two more recent evaluations which have been released within a short interval of time. .These evaluations are due to **ANTSIPOV** et al (1)released in 1981 and to **ARTHUR** et al (2) released in 1984.They are very similar in what concerns the neutron channel x-sections but they differ significantly by the fission x-section up to 2 Mev.Exactly at the timewher e ARTHUR's evaluation was released,**WESTON** (3)published fission x-section experimental values which agree with the LOS-ALAMOS data set.If this agreement is really fortuitous it is of extreme importance because the american evaluation seems to be a purely theoreticalwork. The apparearance of WESTON's data damaged the situation of consensus implied by the russian evaluation or by a review analysis by **PATRICK** (4).As a consequence the 239 Pu fission x-section is now in the NEANDC-INDC discrepancy list.

Globally the present situation for what concerns the experimental microscopic data is judged as follows:

-the information on the total x-section (26 measurements) is of good quality. Thanks to data recently obtained at ORELA and analyzed by **DERRIEN** (5), correct data for the "s" wave neutron strengh function and the scattering radius are available,so that a valuable o.m.p can be derived in the frame of the well known SPRT method. From this parametrization, correct predictions of the compound nucleus formation cross section, of the shape elastic x-section, and of the direct reaction contribution to the inelastic scattering are expected.

-Among the capture x-section data(33 measurements) those obtained at OAK-RIDGE should be considered favorably.

-The knowledge of the total inelastic x-section (5 measurements), although it gives a valuable piece of information for the validation of the optical parameter parametrization, provide a value of the fission x-section (which of similar amplitude) with an accuracy not better than 10%.

-There are so many experimental data (215 measurements) for the fission x-section that there is a good probability that the correct values are among them. But the location of the "TRUTH" is difficult to find because of the very large discrepancies observed in both absolute and relative measurements. It is worth noting that the major discrepancies occur between 30kev and 1Mev ,an energy range of major interest for LMFBR.

-The alpha values are in reasonnable agreement but are'nt accurate enough.The information can be concentrated within 10% . That is not sufficient for a good determination of sigmaf.

-There is one set of eta values relevant to this energy range.These data are very valuable at low energies but they appear to be largely erroneous for energies above 1 Kev.

-There is one set of nu-sigmaf microscopic data hopefully useful.

II EVALUATION METHOD

Considering the possible quality of the optical model parametrization the difficulty in the present evaluation lies in the performance of a correct splitting of the compound x-section into its various components.Since it appears extremely difficult to make a precise choice among the reaction x-section data,especially those of the fission x-section just by using arguments of experimental technique,and (that would be a hudge task being given the number of experiments to be reviewed),the adopted method is to follow the sequential steps roughly described below:

-The first operation is to perform averages of values using a weighting based on the accuracies.

-The second one is to tentatively reproduce the averaged x-sections using the most sophisticated formalisms for the relevant reaction mechanisms and the well known related parameter data.

-The final one is to compare the evaluated data to selected clean integral informations.

In other words a good evaluation will be obtained when the three types of following informations namely -microspic, theoretical and integral-,will converge. Because of the character uncertain of the fission x-section data and the existence of reliable integral data it is particularly appropriate to apply this methodology to 239 Pu.

Assuming that the quality of the measurements increases with the time, the experimental data base has been actually limited to the data released after 1970 for what concerns fission.

III THEORETICAL FORMALISMS _ EVALUATION RESULTS

Neutron Channel

A coupled channel optical model is used to describe the contribution of the direct interaction process.It provides also neutron penetrabilities for the statistical model used to calculate the contribution of the compound nucleus process to the reaction mechanism. A phenomenological model is employed to account for the preequilibrium effects which appear at incident energies greater than 5Mev.

The neutron penetrabilities, the shape elastic x-section and the direct reaction contributions to the inelastic scattering from the first excited states are those derived by **LAGRANGE** and **MADLAND**(6) from semi-microscopic calculations.The ground state and four excited states are coupled so that the coupling basis is:

1/2+,3/2+,5/2+,7/2+,9/2+.

Similar results concerning the x-sections are obtained with a phenomenological model keeping the same coupling basis, but the angular distributions are slightly better described by the semi-microscopic model.

For a better agreement with the experimental data of the total x-section small adjustements (within 2%) have been applied,in some energy ranges, to the neutron penetrabilities. Because of the very small amplitude of the modification this solution has been preferred to a generation of a new complete set of parameters.

The calculations have been performed with the code FISINGA (7) when the statistical theory is involved and with the codes FISINGA and SI4N(8) when the preequilibrium effects are present.

Although it is not totally appropriate for this type of a nucleus the FERMI gas law given by **GILBERT-CAMERON** is adopted to describe the level density.The preequilibrium effects are taken into account by adding to the GILBERT-CAMERON law a correction term based on the exciton model by **GRIFFITH** where constants can be adjusted according to the projectile and the target. It is assumed that the equilibrium component decreases linearly with energy from a threshold value set up at 5 Mev for the actinide nuclei.More details on this formalism can be found in reference (9).

The energy of appearance of preequilibrium effects coincides with the threshold of the (n,Xn) processes described by a formalism reported elsewhere(10) and whose basis is as follows:

$$\sigma^{A}_{n,xn_i}(E) = \sigma_c^{A+1} \cdot P_{ne}^{A+1} \cdot P(E^*, xn_i)^{A+1} \qquad (1)$$

with E*=En+Bn, Bn being the neutron binding energy.

In the R.H.S term , σ_c stands for the compound nucleus formation x-section ,

-Pne for the total neutron emission probality,

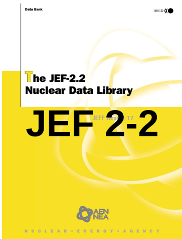
-P(E+Bn,Xn) for the probability of the emission of X neutrons, the residual nucleus decaying by gamma emission or by fission . The neutron cascade is described by the recurrence relationship:

$$P(E^*,Xni)^{A+1} = \left\langle \frac{\Gamma_n}{\Gamma_T}^{A+1} \rho^{A} P(E,(x-1)n_i) \right\rangle_{n}^{A+1} \qquad (2)$$

where the superscript refers to the isotope under consideration; the brackets mean an averaging over the neutron spectrum emitted by the (A+1) nucleus.

In this formalism the spin and parity dependencies are omitted in the neutron channels leading from the nucleus A to the nucleus A-1. This approximation, of little consequence throughout the energy scale except in the region near the threshold, permits a large gain in computing time.

The probabilities of neutron emission are obtained from the



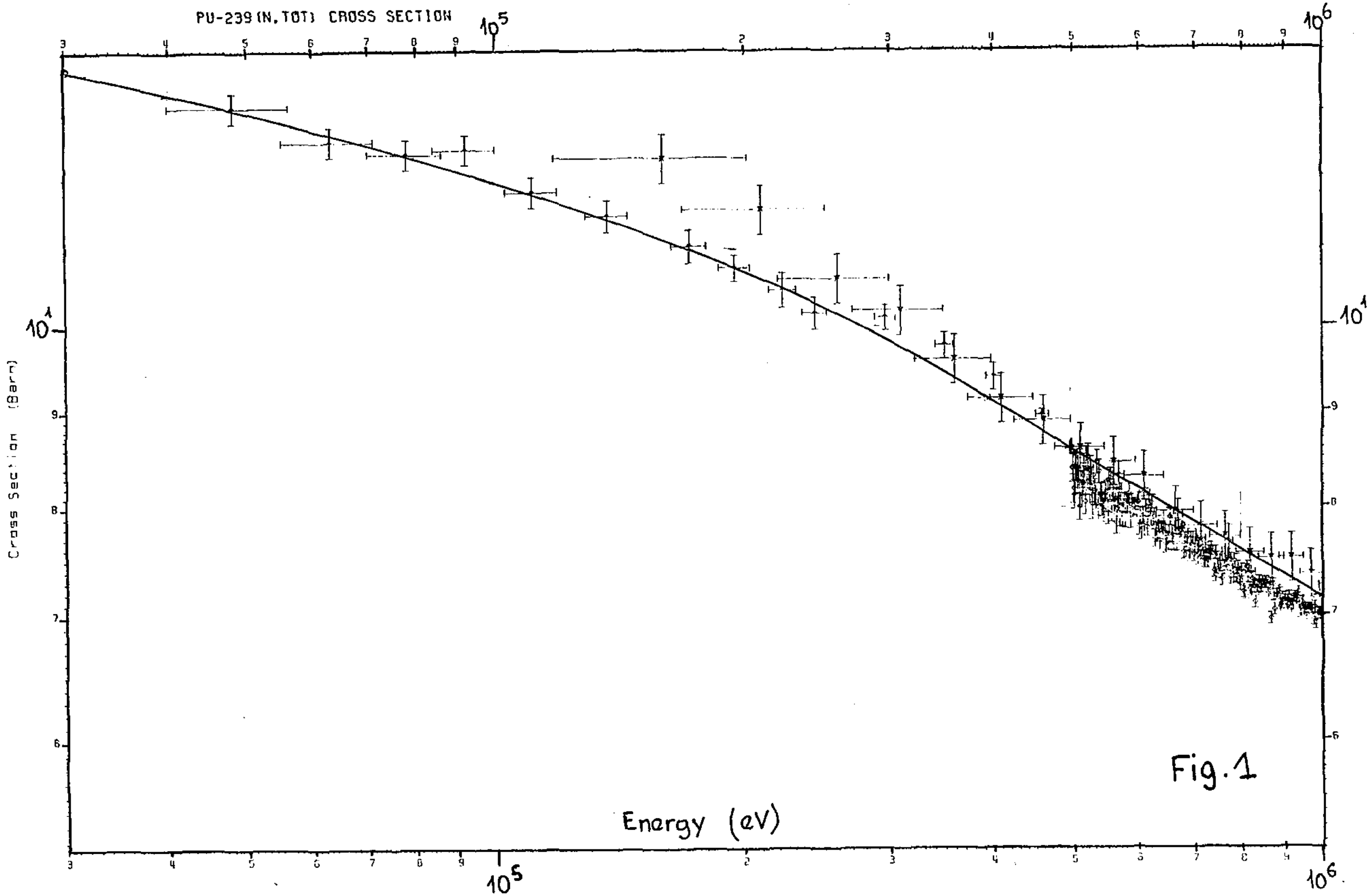


Fig. 1

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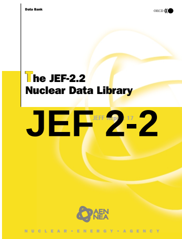

PU-239 (N,TOT) CROSS SECTION

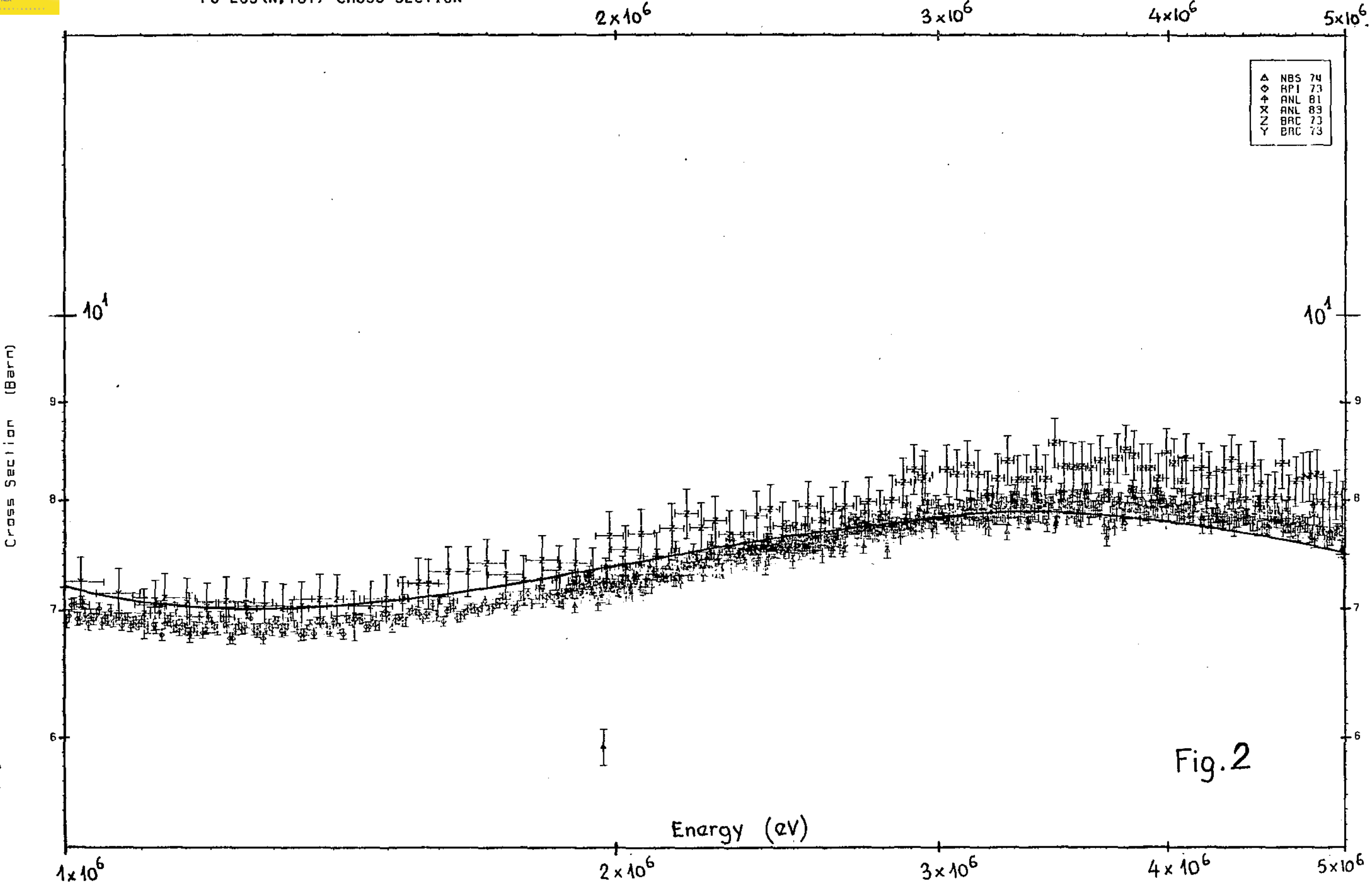

Fig.2

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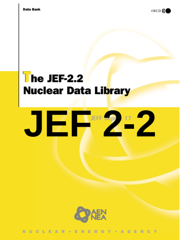

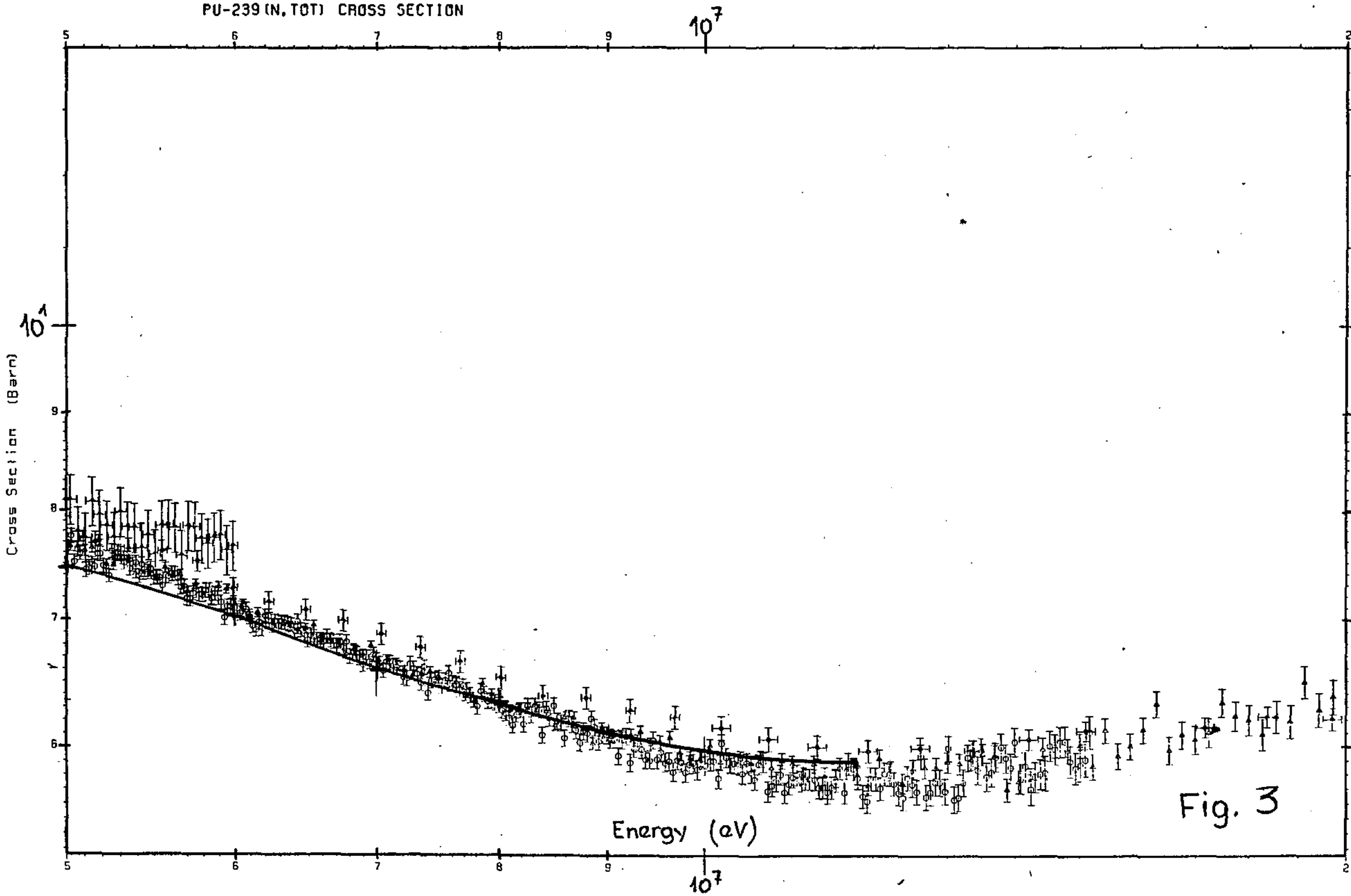

Fig. 3

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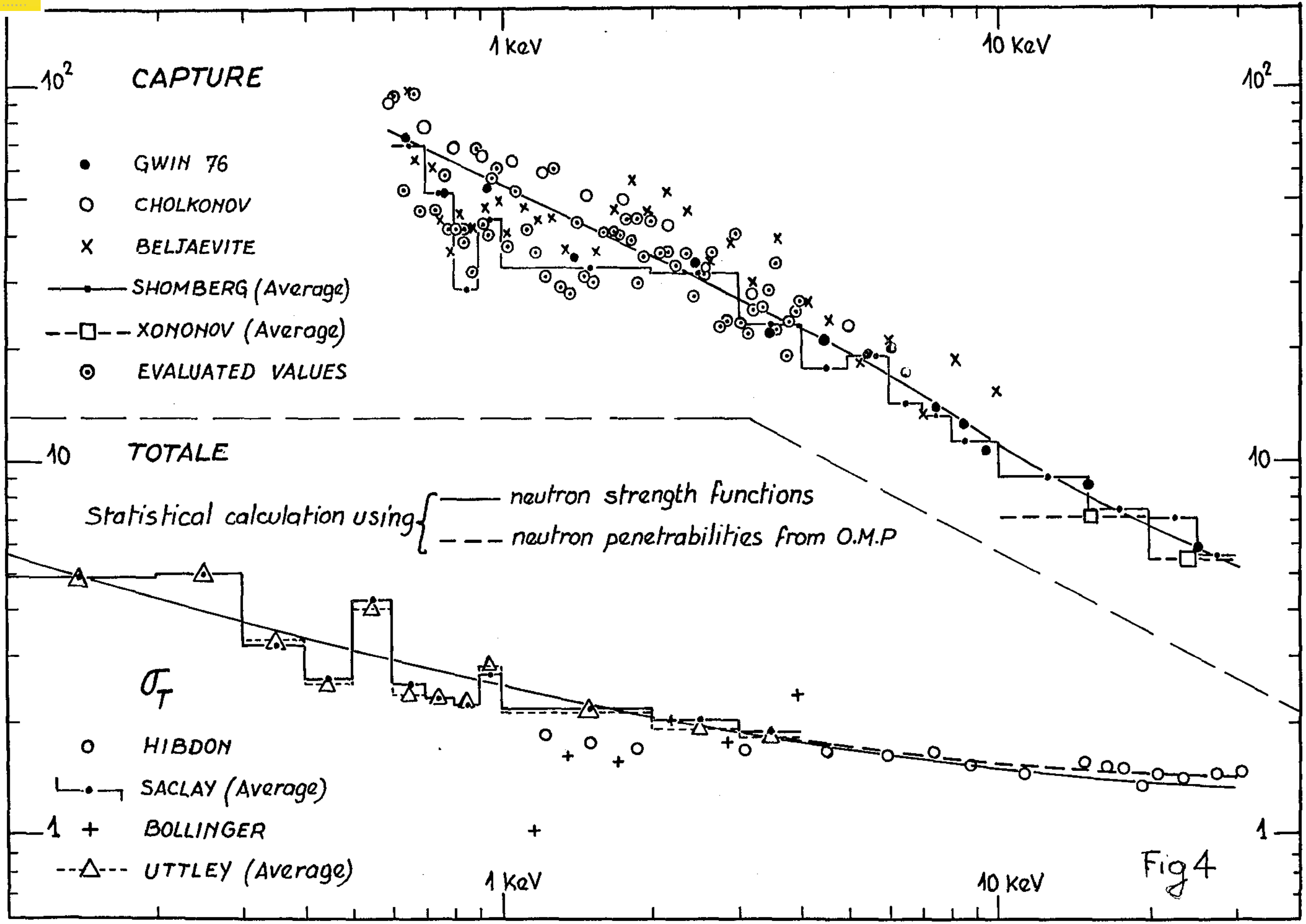
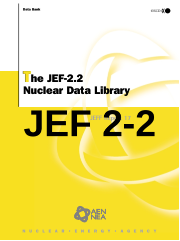

1 KeV
10 KeV
10^2
CAPTURE
GWIN 76
CHOLKONOV
BELJAEVITE
SHOMBERG (Average)
XONONOV (Average)
EVALUATED VALUES
10
TOTALE
Statistical calculation using
neutron strength functions
neutron penetrabilities from O.M.P
σ_T
HIBDON
SACLAY (Average)
BOLLINGER
UTTLEY (Average)
1

Fig 4



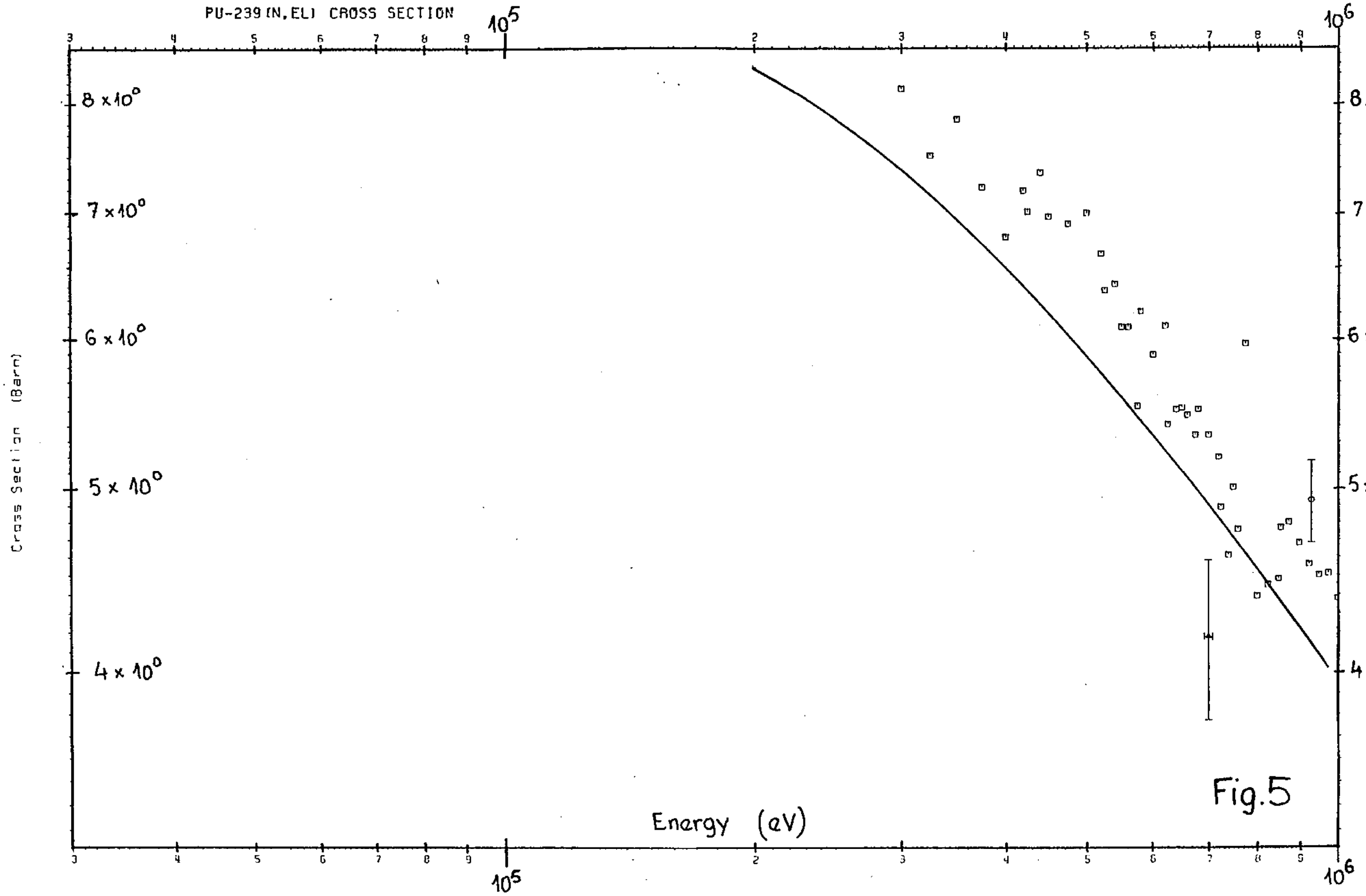


Fig.5

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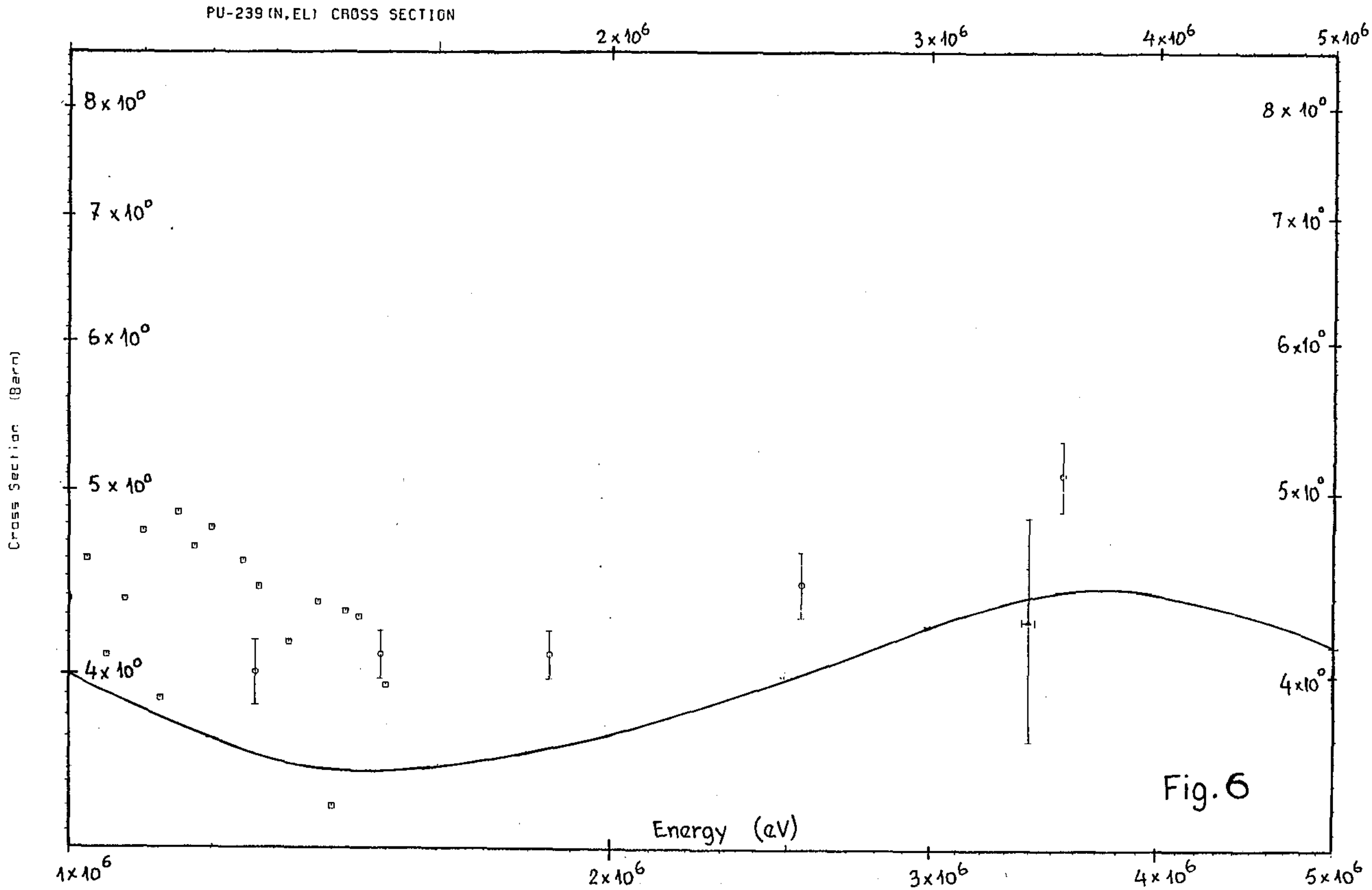
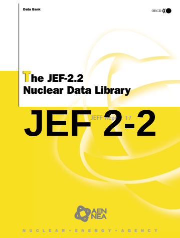
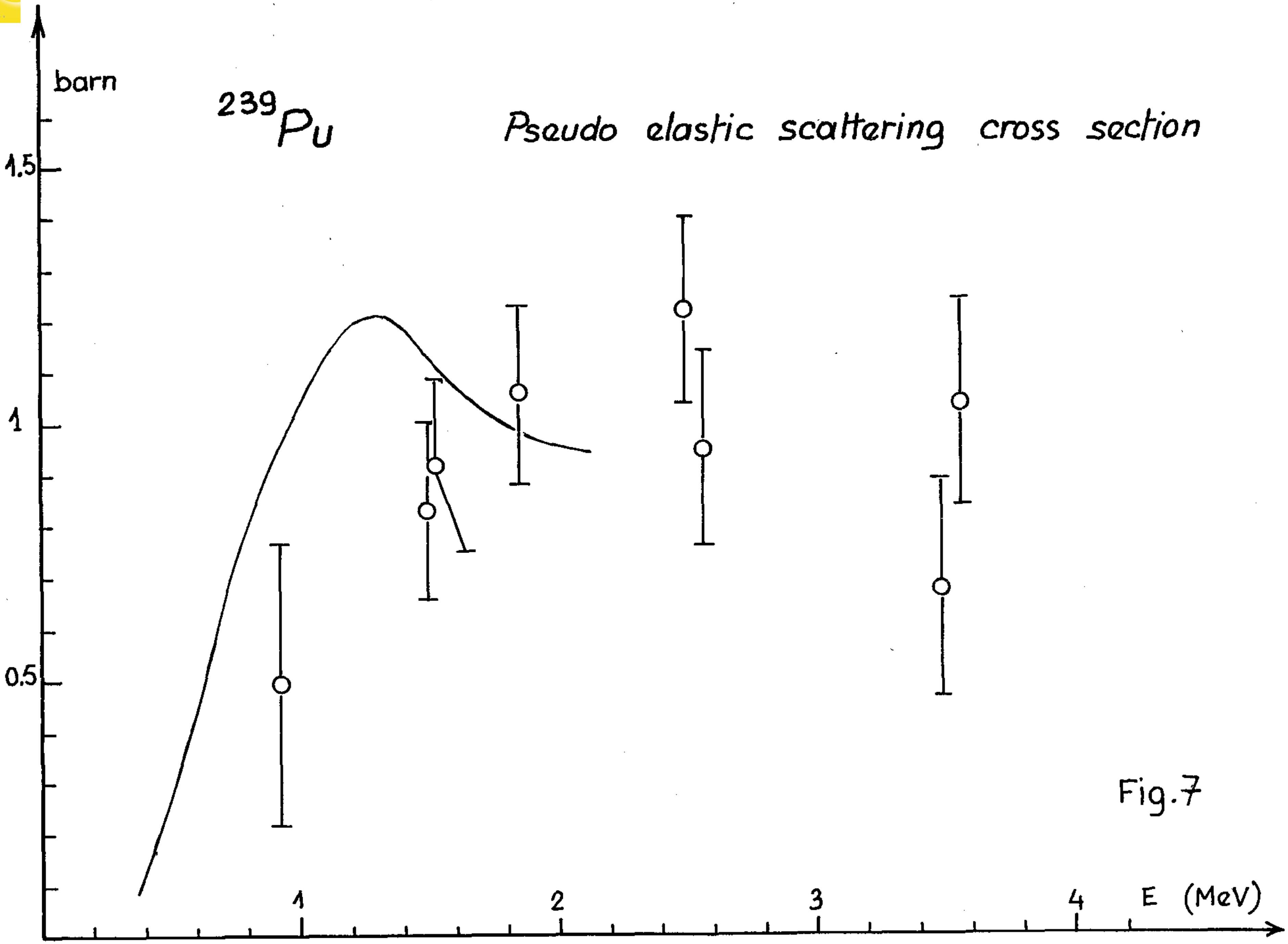

PU-239 (N,EL) CROSS SECTION
Cross Section (Barn)
Energy (eV)
8 x 10°
7 x 10°
6 x 10°
5 x 10°
4 x 10°
1 x 10^6
2 x 10^6
3 x 10^6
4 x 10^6
5 x 10^6

Fig. 6

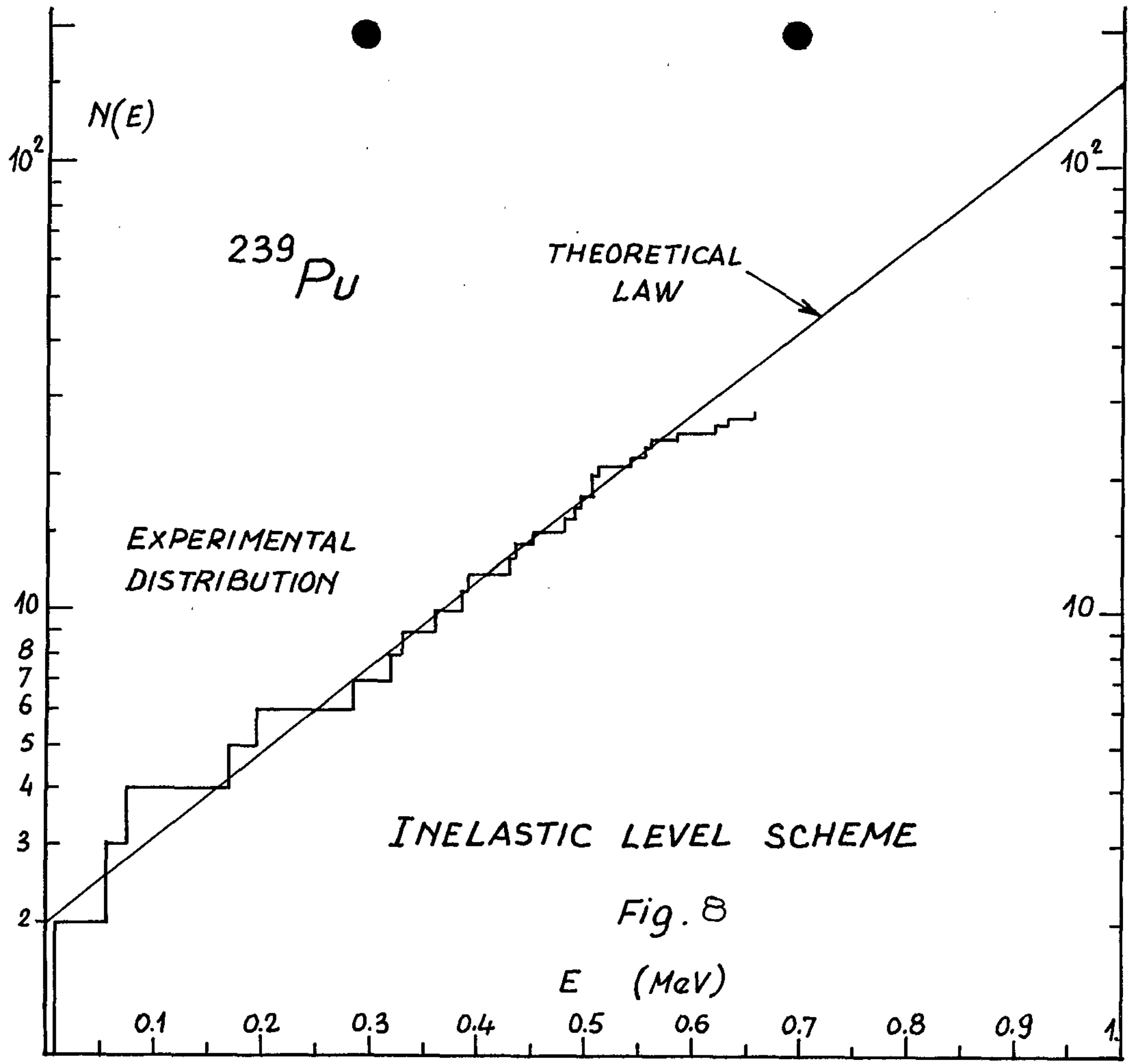


The JEF-2.2
Nuclear Data Library
JEF 2-2



barn
^{239}Pu
Pseudo elastic scattering cross section
1.5
1
0.5
1
2
3
4
E (MeV)

Fig.7


N(E)
10^2
^{239}Pu
THEORETICAL LAW
EXPERIMENTAL DISTRIBUTION
10
8
7
6
5
4
3
2
INELASTIC LEVEL SCHEME
Fig. 8
E (MeV)
0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.

neutron induced reaction x-section of every isotope involved in the cascade which have been evaluated separately. The (n,Xn) x-sections of the nucleus A have to be considered as tests of consistency with the x-section data of the isotopes Ai of the cascade. In a reciprocal way the (n,Xn) x-sections can be predicted with a reasonnable quality, especially the (n,2n) x-section.

Figures (1), (2), (3) show a comparison of the evaluated data for the total x-section in the range covered by the present work with the experimental values, while in the figure (4) one judges the consistency of these results with the calculations performed by DERRIEN (11) using neutron strength fonction values derived from the resolved range.

Figures (5) and (6) show a systematic underestimation of the elastic x-section by the calculation.

There are two reasons in this state of affairs:

-A probable slight contamination by inelastically scattered neutrons.

-A too small scattering radius value used by LAGRANGE and MADLAND: 9.15 fm to be compared with 9.46 fm obtained by DERRIEN. It results an underestimation by about 6% of the direct component. In the energy ranges where the total x-section is not underestimated, the elastic x-section has been modified by the amount necessary to produce a good fit to the experimental data. Such a modification was less than 3%, ie within the limits suggested by the scattering radius ratio.Correspondigly,the reaction x-sections have been reduced by the same quantity.

.One should refer to the paper by LAGRANGE and MADLAND to judge on the performance of the O.M.P used in this evaluation to represent the angular distributions of the elastic and inelastic processes.

The total inelastic scattering x-section above a given excitation energy threshold inferred by **SMITH** and **GUENTHER** (12) from"pseudo elastic scattering" x-section measurement is shown on the figure (7).As stated by ARTHUR and coworkers one sees that the inelastic scattering to states above the ground state rotationnal band corresponds to a compound nucleus process essentially. The calculation correctly fits into the experimental data.The inelastic level scheme has been taken from N.D.S 1977. A discrete configuration has been considered up to 600 Kev.Above this energy a continuous description has been adopted whose energy dependent term exp((E+1.. _ /0.48)) has been derived from the analysis at low energy and the level density at Bn(See fig (8).

Fission Channel

One hundred eleven sets of data have been released after 1970 and considered in our data base. They have been performed on 3 different types of experimental set-ups.

a) The slowing down spectrometers(russian measurements essentially)

b) The electrostatic accelerators

c) The linacs

The relative measurements have been made with respect to 235U fission. The x-section ratios have been converted into x-section data by using the 235U fission x-section values of ENDF-BV as reference. The technique of the center of gravity was used to derive averaged values.

For a given category, first the averaged data of absolute or relative origin have been treated separately and intercompared. No major inconsistencies have been observed. After this encouraging result the data of a given category have been mixed together with no consideration to the origin. The averaged data of the three categories have beenintercompared.Once again it has been observed a good consistency of the data within 3% to 5%. From this situation it can be stated that a large part of the systematic experimental effects have been eliminated by selecting the data sets produced after 1975 and for that reason it has been decided to merge all the data sets in a single data base. The "total" averaged curve obtained in such a way exhibits fluctuations up to about 1 Mev. At this stage of the work it is intersting to make the following remarks, considering the total average curve as a reference(see fig 9)

1) In the range 30 Kev-1 Mev ,JEF1 is higher by 3% to 5%.

2) on the energy range 30 Kev-5 Mev the ANTSIPOV's evaluation is quasi identical(see Fig 9 and 10)

3) WESTON's data agree very well up to about 30 Kev.Upwards they are lower by 7% to 10%.

This situation is a real dilemna because of opposite arguments of similar weight:

-The average curve expresses the statistical consistency of all the experimental values in the data base.

-On the other hand there are strong arguments in favor of WESTON's data at low energy (very small background in the resolved and unresolved ranges (11)). Are the data of the average curve insufficiently corrected for background? In the frame of this hypothesis the experiment by WESTON would appear as a new type of fission measurement characterized by low background. On the contrary if one thinks to an overcorrection in this last experiment it is not understood why correct values would have been obtained for 235 U and not for 239 Pu. Actually,in the experimental conditions, there is no real difference between these two nuclei with the exception of the important alpha radioactivity of 239 Pu which is classically treated by the use of fast electronics.

In any case it does not seem appropriate to treat WESTON's data in the same way as the others and to incorporate them in the data base. Provisionnally it has been decided to give a heavy weight to this experiment, so that the target for theoretical calculations lies between WESTON's and the total average curves.

Concerning these theoretical calculations a double humped barrier ,the outer barrier being well below the inner one, is used to calculate the fission penetrabilities assuming a complete damping for the coupling of the class I and the class II states.If the two barriers are labelled A and B the total fission probability for given spin and parity is written as:

$$T^{J,\pi} = \frac{T_A^{J,\pi} \cdot T_B^{J,\pi}}{T_A^{J,\pi} + T_B^{J,\pi}} \qquad (3)$$

with

$$T_{A,B}^{J,\pi} = \sum_i T_{i_{A,B}} + \int_{E_{cf}}^{\infty} \rho_F^{J,\pi}(\epsilon)\, T_f^{0}(E+B_n-\epsilon)\, d\epsilon \qquad (4)$$

In the R.H.S part of equation (4) the sum represents the

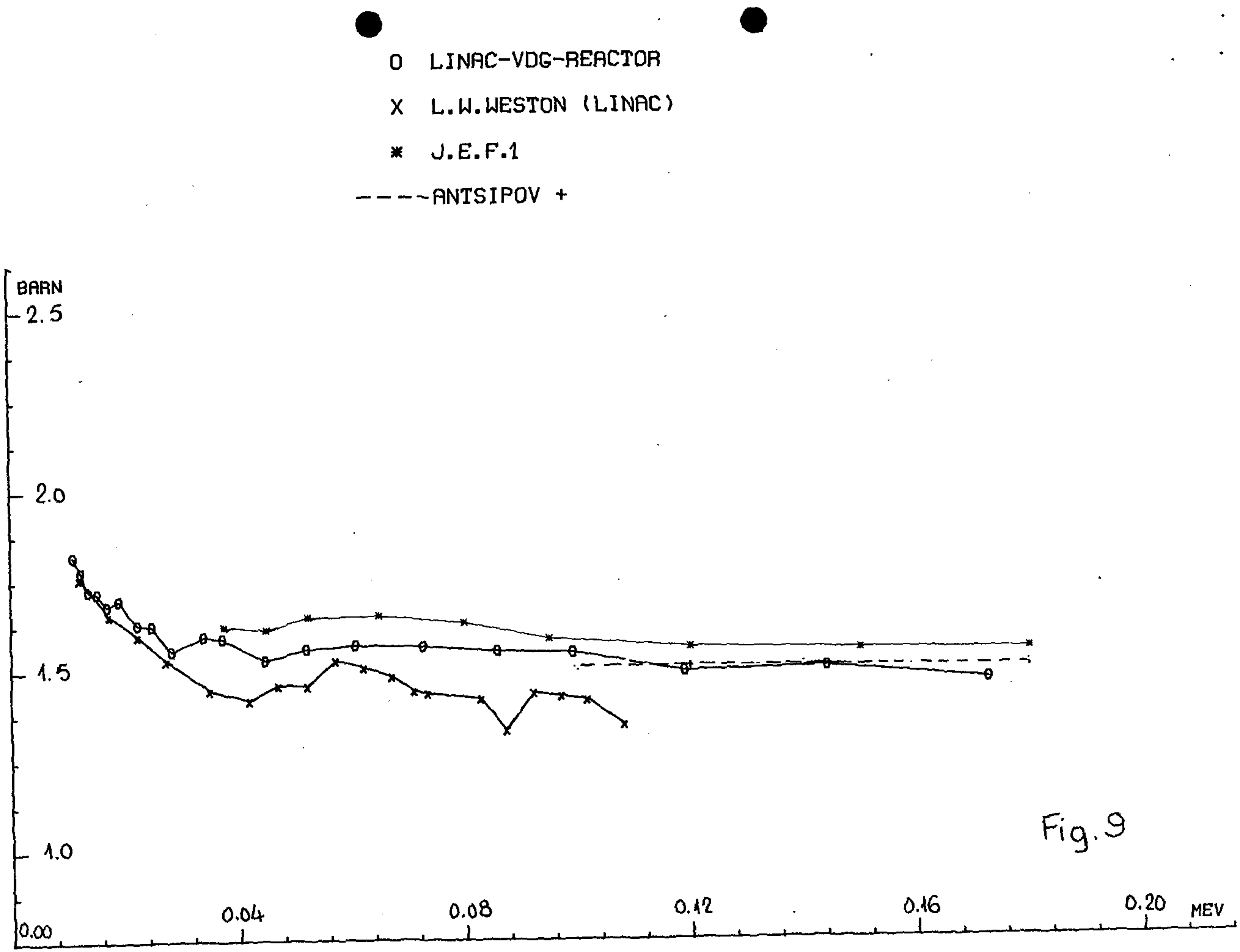
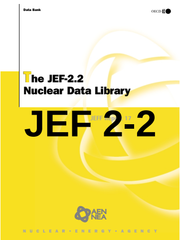

O LINAC-VDG-REACTOR
X L.W.WESTON (LINAC)
✱ J.E.F.1
----ANTSIPOV +
BARN
2.5
2.0
1.5
1.0
0.00
0.04
0.08
0.12
0.16
0.20
MEV

Fig. 9



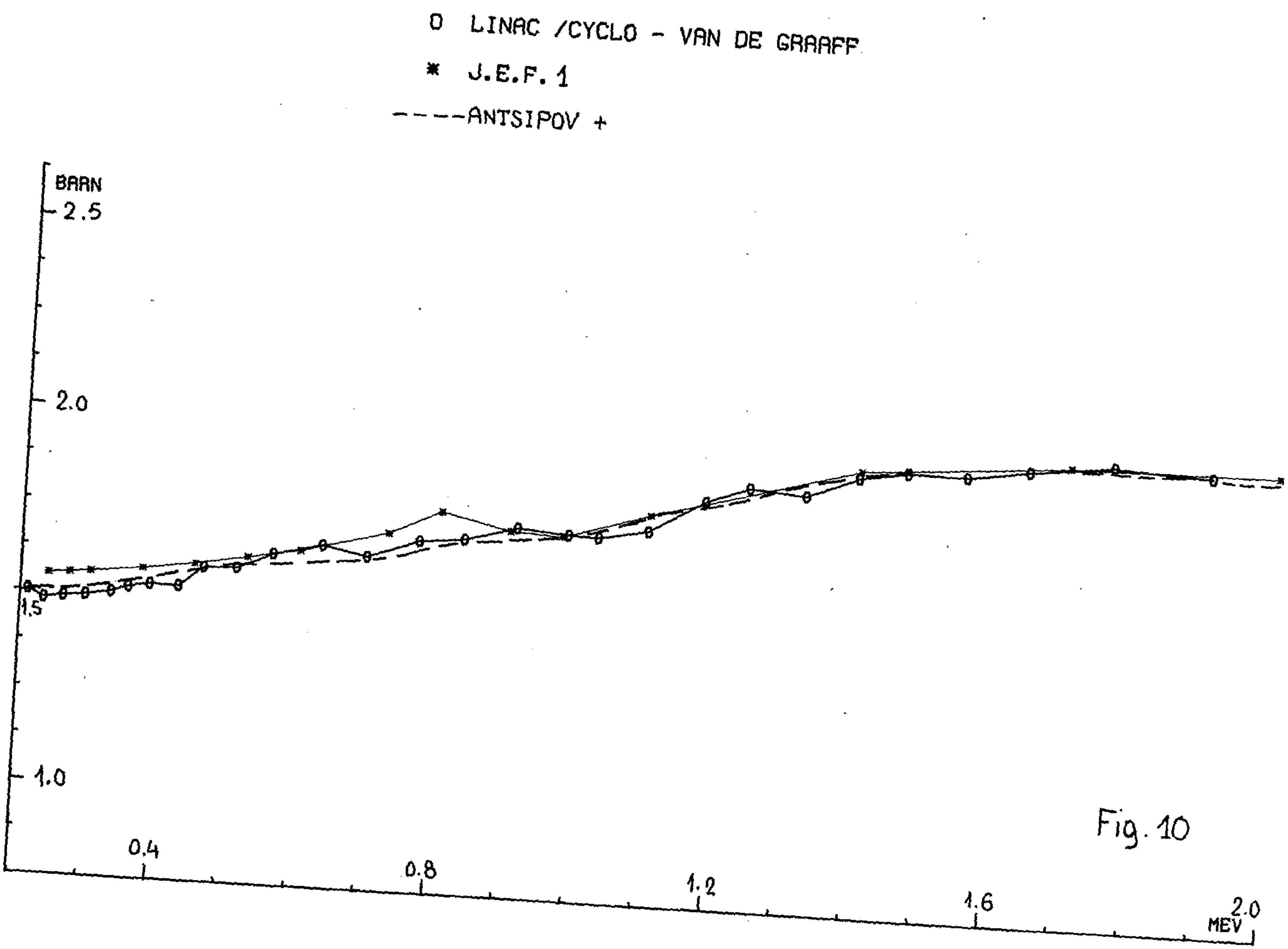


Fig. 10

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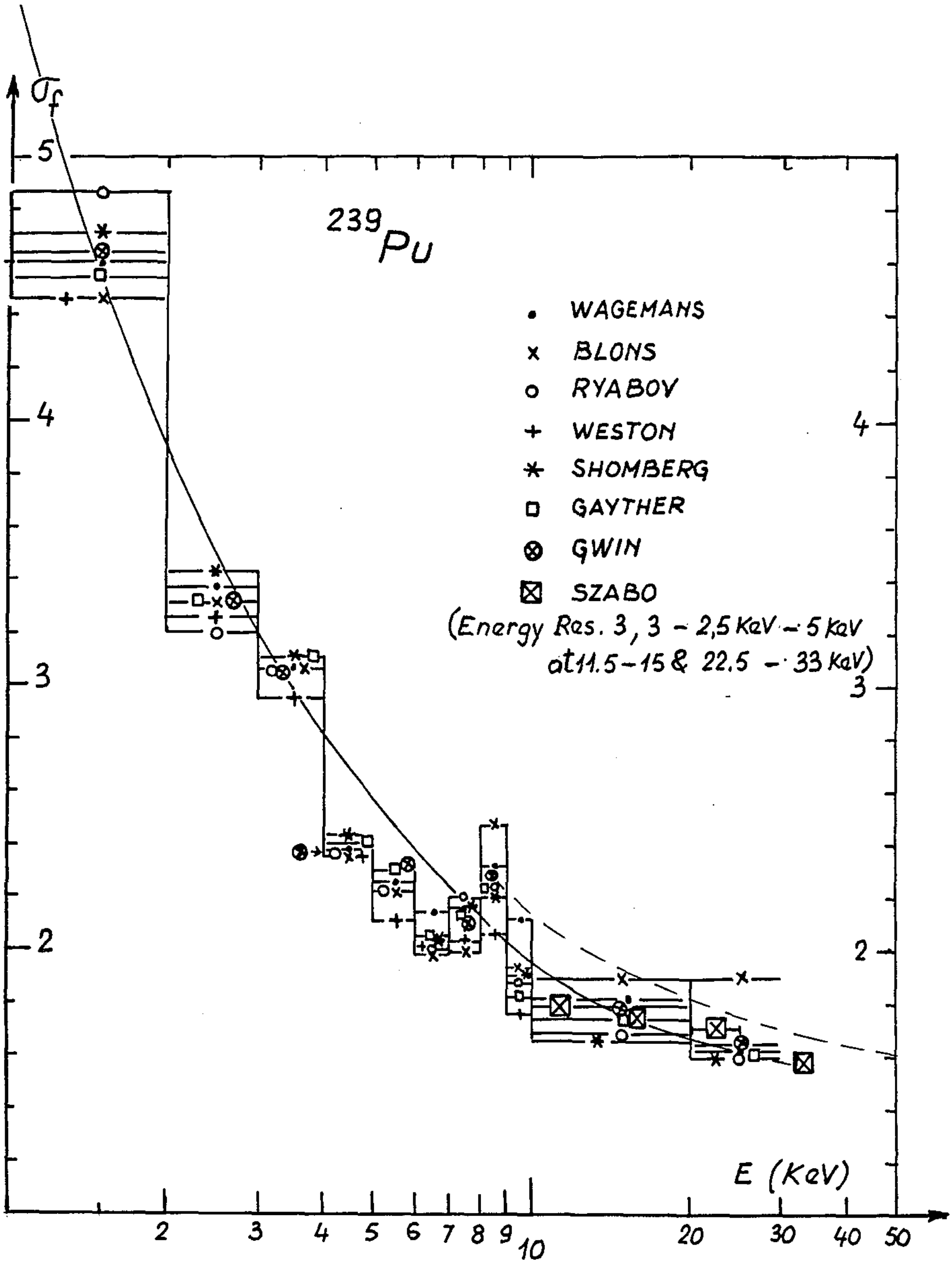

Fig. 11

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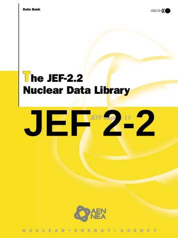

PU-239 FISSION CROSS SECTION (1975-)

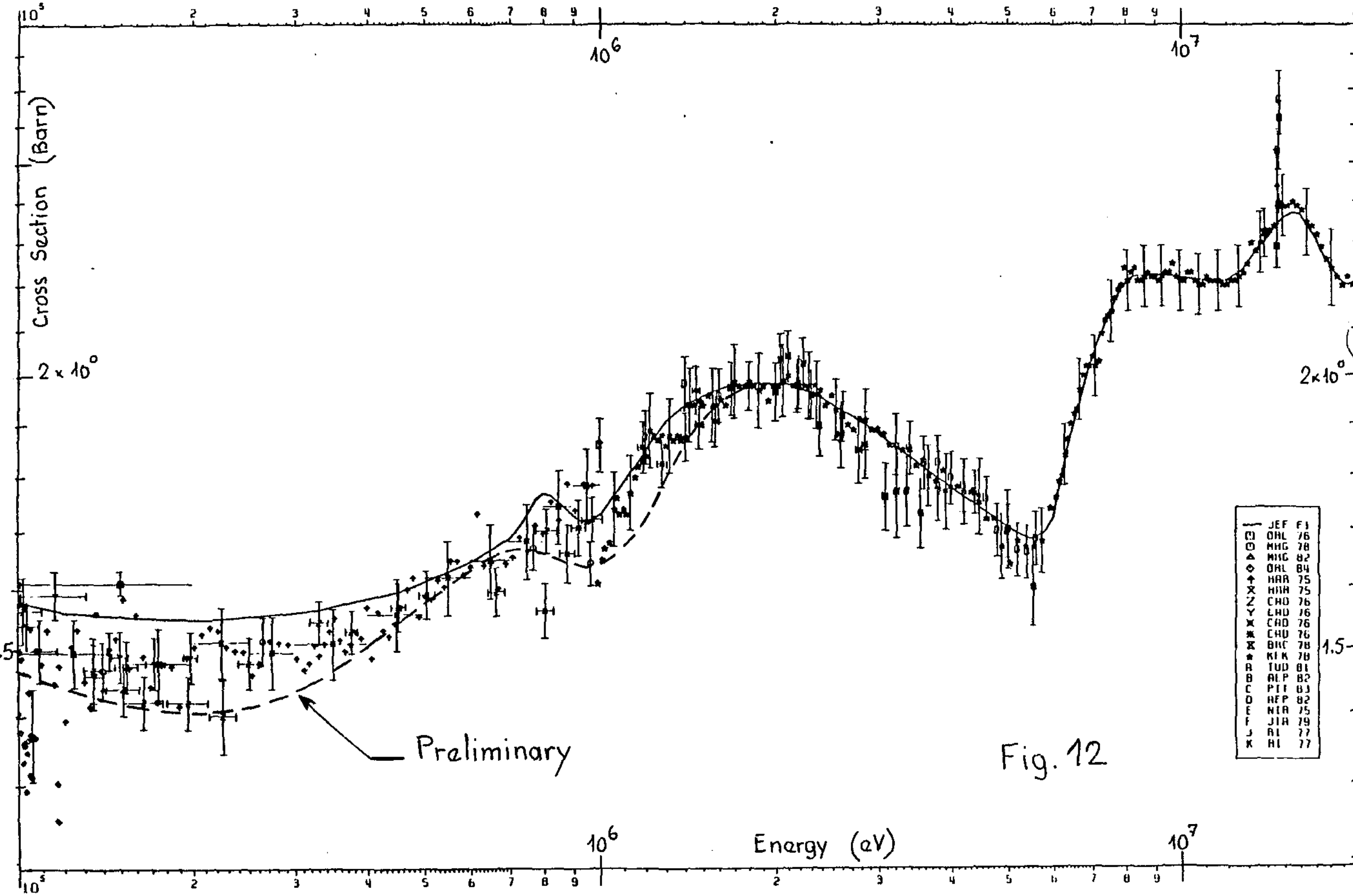

Fig. 12

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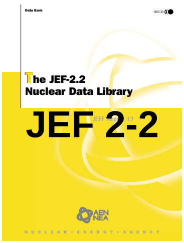

contribution of the i discrete fission channels and the integral the one of the continuum.

T_f° is the tunneling probability for the fundamental barrier, while ρ_F^J is the fission channel density for a given spin and parity at the deformation corresponding to the barriers A and B.

The discrete fission channels above the fundamental were the first class I states of 240 Pu .Their excitation energies were modified to reproduce the fission x-section at energies lower than 100 kev.

In the continuum the fission density probability has the following expression:

$$\rho^J(E^*) = C_{1i}(2J+1)\, e^{-\frac{(J+1/2)}{2\sigma^2}}\, e^{E^*/\Theta_{Fi}} \qquad E^* \leq B_n \tag{5}$$

$$\rho^J(E^*) = C_2(2J+1)\, e^{-\frac{(J+1/2)^2}{2\sigma^2}}\, \frac{e^{2\sqrt{a_f(E^*-\Delta)}}}{(E^*-\Delta)^{3/2}} \tag{6}$$

For E* located in the range 0-Bn the constants C_{1i} and Θ_{Fi} have been defined in different subranges according to LYNN's work(13) after a slight modification of their values.

The constraint to reproduce the averaged values in the range 1Kev-30Kev (see Fig11) clearly imposes low values of fission x-section up to the minimum at 200 Kev.Above this energy the correlation with the situation in the tens of Kev rapidly decreases.The fact that there are difficulties in reproducing the total averaged experimental curve justifies a posteriori the importantweight given to the WESTON's data(see Fig12).

Gamma Channel

Up to now no deep analysis of the capture experimental data has been performed and this x-section has been considered as a by-product in the theoretical calculations.This is more or less justified since the radiative capture x-section is a small fraction of the compound x-section in the range of interest.In the unresolved range where this x-section is more significant,the calculation has been performed(10) with the parameters $\Gamma_\gamma^{l=0} \simeq 40$ mev deduced from the resonance analysis and $\Gamma_\gamma^{l=1}$ =30 mev adjusted for a good fitting into the experimental data. A theoretical background to that can be found in the frame of the electric dipolar transitions and the BRINK-AXEL profile function ,since the radiative width can be expressed as:

$$\Gamma_\gamma^l(E) = K \int_0^{B_n+E} \Phi(\varepsilon)\, \rho^l(E+B_n-\varepsilon)\, d\varepsilon \tag{7}$$

The inequality $\Gamma_\gamma^{l=1} < \Gamma_\gamma^{l=0}$ results from the difference in the low lying levels densities in the 240 Pu as it can be observed in the figure (13).In our calculations based on the N.D.S indications (14) and D_{obs} determination we have found $\Gamma_\gamma^{l=1}/\Gamma_\gamma^{l=0}$ = . The value of $\Gamma_\gamma^{l=1}$ deduced from this ratio and the experimental indication for $\Gamma_\gamma^{l=0}$ has been used in our calculations.

On the figure (14) the calculated curve is compared with JEF1 and the experimental data.

PU-239 CAPTURE CROSS SECTION

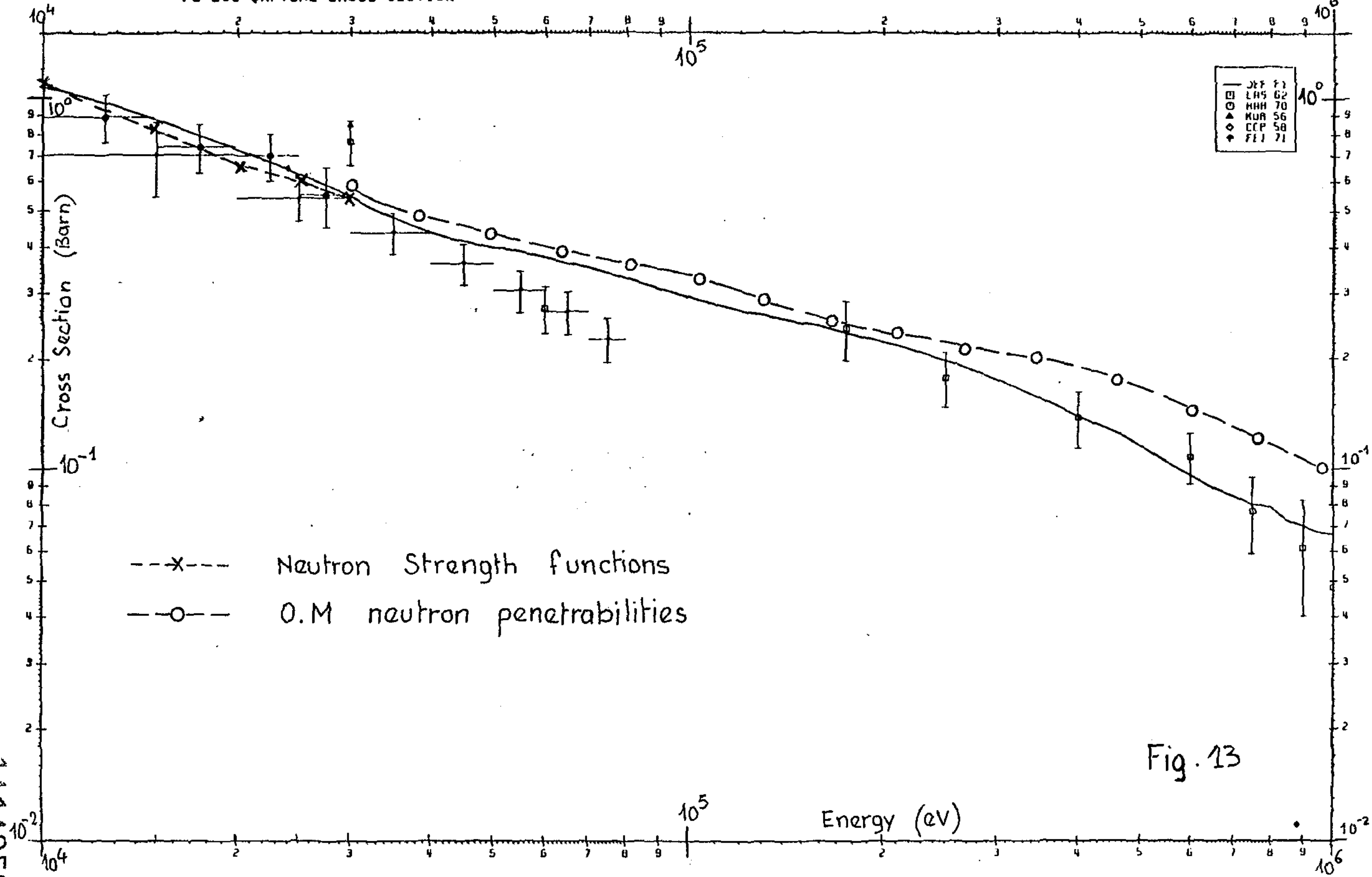

Fig. 13

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