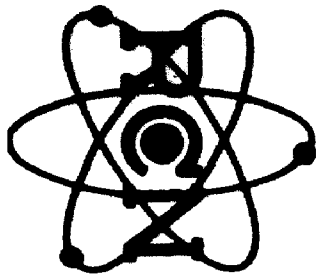

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NEUTRON SELFSHIELDING OF ACTIVATION DETECTORS USED IN SPECTRUM UNFOLDING

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30 PAGES, 10 FIGURES, 8 TABLES

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Keywords:

activation analysis
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Neutron selfshielding of activation detectors used in spectrum unfolding

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Petten, August 1975.

Abstract

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Neutron selfshielding of activation detectors used in spectrum unfolding.

Neutron selfshielding corrections for activation detectors have been studied with a computer program SELFS, which is used in combination with SAND-II, the computer program for determining neutron spectra with aid of a set of irradiated foil detectors. The method does not apply integral correction factors to the measured activities, but applies energy dependent correction factors to the group cross section values. This calculation method has the advantage that correction can be calculated only once for a given foil thickness and can be applied to each neutron spectrum with known shape. The applied formula to calculate the selfshielding is valid under limited conditions, but in practice the elementary formula provides data which are accurate enough. The results of calculations have been compared with literature data (e.g. for a 1/E neutron spectrum, and for the neutron spectra of the ΣΣ facility in Mol and CFRMF in Los Alamos). The cross section data required were taken from the SAND-II, ENDF/B and the BNL-325 data sources. The program SELFS is written in FORTRAN and was used on a CDC-6600 computer.

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CONTENTS

1. INTRODUCTION

At ECN the program SAND-II is often used to determine neutron spectra with aid of activation and fission foils, irradiated in locations of interest in the High Flux Reactor (HFR), the Low Flux Reactor (LFR), and the coupled fast thermal critical facility STEK. The SAND-II program [1] requires as input a best estimate of the neutron spectrum, a set of experimentally obtained saturation activities, and a cross section library for all reactions of interest. The calculations are performed in 620 energy groups, comprising the whole range from 10^{-10} MeV up to 18 MeV.

The experimental activation or fission rates may be influenced by relatively important neutron selfshielding effects due to the thickness of the selected target material.

This report describes a method which performs a correction for this neutron selfshielding not to the measured activities, but to the group cross sections. The method prepares a set of corrected cross section values, adapted to the target material, and this modified cross section set is then valid for each type of neutron spectrum.

To test this approach we derived also integral selfshielding correction factors and compared these with experimental selfshielding data reported by other laboratories. In this way calculations with the program SELFS have been performed for a 1/E neutron spectrum, and for the neutron spectra of the ΣΣ facility at Mol and the CFRMF at Idaho.

2. THEORY USED IN SELFS

The selfshielding factor G for an activation detector can be defined as the ratio of the experimental activation to the theoretically expected activation without selfshielding, the latter activation being determined by the appropriately chosen activation cross section and the flux density of neutrons at the surface of the detector.
The selfshielding factor defined in this way is always less than unity.
Assuming an isotropic flux density, one has generally [2]:

$$G = \frac{\int_{E=0}^{\infty}\int_{x=0}^{t}\int_{\theta=0}^{\pi/2} \sigma_{act}(E).\,\phi_E(E).e^{-N_v.\sigma_t(E).x/\cos\theta}\sin\theta.d\theta.dE.dx}{\int_{E=0}^{\infty}\int_{x=0}^{t}\int_{\theta=0}^{\pi/2} \sigma_{act}(E).\phi_E(E).\sin\theta.d\theta.dE.dx}$$

where N_v = the number of target atoms per unit volume;
σ_{act} = the activation cross section;
σ_t = the total cross section (activation plus scattering);
t = foil thickness.

The equation used in SELFS is based on the following assumptions:
- the neutron flux density can be considered as monoenergetic in each energy group;
- the cross sections can be taken constant in these energy groups;
- the incident neutron flux density is isotropic;
- there are no multiple interactions;
- the foil may be considered as a plane slab (i.e. there are no edge effects);
- the flux depression outside the foil is much less than the self-shielding in the foil;
- the Doppler broadening effect is negligible.

When we apply a rather fine group structure, like the 620 group structure of SAND-II, then the first two assumptions will give negligible errors (except when very narrow resonances occur).
When the integration in the formula above is performed first over the energy one obtains for each energy group i:

$$G^i = \frac{\int_{x=0}^{t} \int_{\phi=0}^{\pi/2} e^{-N_v \cdot \sigma_t^i x/\cos\phi} \cdot \sin\phi \cdot d\phi \cdot dx}{\int_{x=0}^{t} \int_{\phi=0}^{\pi/2} \sin\phi \cdot d\phi \cdot dx}$$

where σ_t^i denotes the total cross section for group i.

Substitution of $u = \cos\phi$

and $\tau^i = N_v \cdot \sigma_t^i \cdot t$

(which expresses the foil thickness in units of mean free path, since $\tau^i = t/\lambda_t^i$, where λ_t^i is the macroscopic total cross section in the i^{th} group) and integration over x leads to

$$G^i = \frac{1}{\tau^i} \int_1^0 u(1-e^{-\tau^i/u}) \cdot du.$$

Substitution of $s = 1/u$ gives

$$G^i = \frac{1}{\tau^i} \int_1^\infty -\frac{(1-e^{-st^i})}{s^3} ds.$$

Introducing $E_3(\tau)$, the third exponential integral defined as

$$E_3(\tau) = \int_1^\infty \left(\frac{e^{-\tau t}}{t^3}\right) dt$$

one obtains the basic expression used in SELFS:

$$\boxed{G(\tau^i) = \frac{1-2\,E_3(\tau^i)}{2\tau^i}}$$

Since one can calculate $G(\tau^i)$ values for each cross section for each energy group i relevant to the multifoil detector set, one can prepare a new and modified group cross section set which can be used in the spectrum evaluation program. The cross section modified for self-shielding can be written as

$$(\sigma_c^i)_{mod} = G(\tau^i) \cdot \sigma^i.$$

These modified cross section data with 620 energy groups for the specified foils have been used in the SAND-II program.

3. BRIEF INTRODUCTION TO SAND-II

This program has been developed by McElroy and coworkers [1] to evaluate the neutron flux density spectrum, starting from measured activities of a series of irradiated activation and fission detectors, and a zeroth approximation neutron spectrum.
One of the main features of this program is that the information of the zeroth approximation spectrum (the input spectrum) is essential in deriving the solution spectrum (the output spectrum). The program can select an input spectrum from a library of about 60 neutron spectra, or can prepare such an input spectrum from tabulated group flux density data by using interpolation and extrapolation procedures. By subsequent iterative adjustments of the neutron spectra, the program arrives at an appropriate output spectrum, which produces calculated activities which fit within experimental error as close as possible to the measured activities. The calculation is performed for 620 energy groups, ranging from 10^{-10} MeV to 18 MeV.
Also the SAND-II cross section library covers this range.
The cross section library is made from point cross section values by the subprogram CSTAPE. For more information on the SAND-II program the reader is referred to [1].

4. SELFS IN RELATION TO SAND-II

The program SELFS reads an original cross section library made by the program CSTAPE of the SAND-II package, or a library made by SELFS. It then performs the calculations required to obtain modified cross sections, and writes a new library which contains the original group cross sections and the modified group cross sections.
The library can be used in a SAND-II spectrum calculation, and can also be read by SELFS, when an extension of the library with respect to other foil thicknesses is required.
When SELFS has to calculate modified cross section data for one or more foils, one has to specify in the input a few characteristics of the foils under consideration (e.g. reaction name, thickness, atomic mass number, etc.).

The programs SELFS and SAND-II provide a means to calculate integral selfshielding factors, which lead themselves for comparison with experimental selfshielding factors.
The integral selfshielding factors which are clearly spectrum dependent can be determined as a ratio of two activity values calculated by using cross section data and neutron spectrum data; these activity calculations have to be performed for the unmodified capture cross section data as well as for the modified capture cross section data set. The calculation of the activities is easily performed with the ACTIVITY mode of the SAND-II program using the modified and the unmodified differential cross section values, and also the differential neutron spectrum data in the SAND-II group structure with 620 groups. The program offers the possibility for the calculation of several activities in one computer run, so that integral selfshielding factors for several materials and foil thicknesses can easily be obtained.

5. PREPARATION OF TOTAL CROSS SECTION DATA

The first version of SELFS, prepared by I. Kondo [3] took only into account the activation cross sections, thus neglecting the contribution of the scattering cross section to the total cross section. This version yielded rather inaccurate results. This was especially the case for reactions with an important scattering contribution (e.g. copper, manganese and cobalt). A modified version, prepared by T. Noda [4], took into account also the scattering effect by introducing the total cross sections. This approach made it necessary to have available in addition to the activation cross section library a total cross section library, again in the 620 groups structure.
The calculations started with total cross section values prepared from BNL-325 and from the ENDF/B-III file. The data from the file were converted into point values and changed into the SAND-II group structure. Recently the ENDF/B IV file with up-to-date capture and total cross section data became available. In applying this file many more cross section point values have been prepared than previously, so that, apart from a probable improvement in the quality of the data, also improvement was established for the conversion into the SAND-II group structure [5]. The conversion from ENDF/B IV data to the input data for the CSTAPE of the SAND-II program package was performed with a special program package prepared by Barsky [6].

6. TEST OF SELFS

The program SELFS was tested by comparing calculated G-values with experimental values reported by other laboratories. The comparison was performed for a few materials (e.g. the reactions $^{197}Au(n,\gamma)^{198}Au$, $^{63}Cu(n,\gamma)^{64}Cu$, $^{235}U(n,f)$) and three different neutron spectra (i.e. a 1/E spectrum, the ΣΣ spectrum, and the CFRMF spectrum). For these neutron spectra experimentally determined selfshielding factors were available, while also the shapes of the neutron spectra are well-known. The data for the ΣΣ secondary intermediate energy standard neutron field at Mol were obtained from Fabry [6]. The data for the Coupled Fast Reactivity Measurement Facility at Idaho were obtained from a LMFBR progress report [7].

6.1. Results for gold

Results for G-values for gold foils are presented in tables 1 and 2. All G-factors were calculated with aid of total cross section data from the ENDF/B III and the ENDF/B IV file. The 1/E spectrum which was applied had a cadmium cut-off energy of 0.68 eV. Here the G-values using σ_c and σ_t from the ENDF/B IV file are in good agreement with the experimental data published by Brose [8].
For the ΣΣ and CFRMF neutron spectra the G-values based on the ENDF/B III data are somewhat closer to the reference values than values based on the ENDF/B IV data.

6.2. Results for copper

The results of the calculations of selfshielding factors for the activation reaction $^{63}Cu(n,\gamma)^{64}Cu$ are presented in tables 3, 4 and 5. In case of the 1/E spectrum we observe that the G-values calculated with σ_c from the ENDF/B III file show rather large deviations from the reference values, and are worse than the G-values using σ_c from the SAND-II library. Remarkable is that in this case the SAND-II cross sections yield still better agreement with experimental data than the recent ENDF/B IV cross sections.
For the ΣΣ spectrum the agreement between calculated and experimental G-values is rather poor. Here the smallest deviations are obtained

for the ENDF/B III capture cross sections. The deviations remain large (about 5%) for these cross section data.
In the case of the CFRMF spectrum we obtain the best results using the cross sections from the ENDF/B IV file.

6.3. Results for cobalt

Calculated selfshielding factors for cobalt foils in the CFRMF neutron spectrum deviate appreciably from the experimental values, reported by McElroy (see table 6). Since it is well-known that scattering cross section of cobalt has a strong influence on the selfshielding, this point should be investigated further.

6.4. Results for ^{235}U

Some experimental data are available for the CFRMF facility. The calculated G-values for this facility, using the cross section data from the ENDF/B IV file are very close to the experimental data of McElroy.

7. CONCLUSION

Tests of the described simple mathematical model for the calculation of neutron selfshielding factors using capture cross section data and total cross section data derived from the ENDF/B IV dosimetry file, resulted in values for the selfshielding factors which often agreed with experimental data.
Discrepancies were observed for copper foils in the ΣΣ spectrum and for cobalt foils in the CFRMF spectrum.
Further tests should be perfo.med with other detector materials, preferably with relatively large scattering cross sections.
Also the influence of the energy group structure used in the calculations of the G-values should be considered.

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Table 1. Selfshieldings for gold foils in a 1/E apsectrum

Values G_3 and G_4 were obtained using σ_c and σ_t from ENDF/B III and ENDF/B IV files respectively.

Values G_2 were obtained using σ_c from SAND-II library and σ_t from the ENDF/B III file.

ΔG_i denotes the relative deviation $(G_{exp} - G_i)/G_i$.

The cadmium cut-off energy of the 1/E neutron spectrum for these calculations was 0.68 eV.

foil thickness (in mm)	G_{exp} a)	G_2	G_3	G_4	ΔG_2 (in %)	ΔG_3 (in %)	ΔG_4 (in %)
2×10^{-5}	* 0.9936	0.9957	0.9963	0.9957	-0.21	-0.27	-0.21
4×10^{-5}	* 0.9893	0.9936	0.9926	0.9914	-0.43	-0.33	-0.21
8×10^{-5}	* 0.9815	0.9833	0.9857	0.9834	-0.18	-0.38	-0.19
2×10^{-4}	0.9644	0.9630	0.9667	0.9624	+0.1	-0.24	-0.21
4×10^{-4}	0.9340	0.9340	0.9408	0.9334	-0.00	-0.73	-0.06
8×10^{-4}	0.8852	0.8896	0.9007	0.8890	-0.50	-1.75	-0.43
2×10^{-3}	0.7852	0.7946	0.8141	0.7935	-1.20	-3.68	-1.05
4×10^{-3}	0.6836	0.6909	0.7190	0.6899	-1.07	-5.18	-0.91
8×10^{-3}	0.5612	0.5655	0.6022	0.5658	-0.77	-7.31	-0.81
2×10^{-2}	0.3952	0.3996	0.4427	0.4023	-1.11	-12.02	-1.76
4×10^{-2}	0.3020	0.2997	0.3403	0.3028	+0.77	-12.68	-0.26
8×10^{-2}	0.2219	0.2272	0.2591	0.2271	-2.39	-16.76	-2.29
2×10^{-1}	0.1505	0.1640	0.1796	0.1550	-8.97	-19.34	-2.90
4×10^{-1}	* 0.1118	0.1313	0.1347	0.1160	-17.4	-20.5	-3.62
8×10^{-1}	* 0.0836	0.1048	0.0983	0.0858	-25.4	-17.6	-2.52

a) These values were taken from the experimental work by Brose [9]. Only the values denoted with a * refer to calculations by Brose.

Table 2. Selfshielding for gold foils in the [illegible] neutron spectrum.

Values G_3 and G_4 were obtained using σ_c and σ_t from ENDF/B III and ENDF/B IV files respectively.

Values G_2 were obtained using σ_c from SAND-II library and σ_t from ENDF/B III file.

ΔG_i denotes the relative deviation $(G_{exp} - G_i)/G_i$

foil thickness (in mm)	G_{exp} a)	G_2	G_3	G_4	ΔG_2 (in %)	ΔG_3 (in %)	ΔG_4 (in %)
2×10^{-3}	0.986	0.988	0.984	0.984	-0.20	+0.20	+0.21
4×10^{-3}	0.978	0.981	0.974	0.975	-0.31	+0.41	+0.31
8×10^{-3}	0.965	0.973	0.960	0.963	-0.83	+0.52	+0.20
2×10^{-2}	0.932	0.958	0.933	0.943	-2.79	-0.11	-1.20
4×10^{-2}	0.894	0.944	0.905	0.925	-5.59	-1.23	-3.36
8×10^{-2}	0.863	0.920	0.867	0.902	-7.30	-0.46	-4.29

a) These experimental values were taken from the work by Fabry [7].

Table 3. Selfshielding for gold foils in the CFRMF neutron spectrum

Values G_3 and G_4 were obtained using σ_c and σ_t from ENDF/B III and ENDF/B IV files respectively.

Values G_2 were obtained using σ_c from SAND-II library and σ_t from ENDF/B III file.

ΔG_i denotes the relative deviation $(G_{exp} - G_i)/G_i$.

foil thickness (in mm)	G_{exp} a)	G_2	G_3	G_4	ΔG_2 (in %)	ΔG_3 (in %)	ΔG_4 (in %)
1.270×10^{-3}	1.000	0.999	0.997	0.999	+0.10	+0.30	+0.14
1.270×10^{-2}	0.976	0.992	0.979	0.990	-1.64	-0.30	-1.40
5.080×10^{-2}	0.923	0.973	0.939	0.969	-5.42	+1.73	-4.77

a) These experimental values were taken from the work by McElroy [8].

Table 4. Selfshielding for copper foils in a 1/E neutron spectrum.

Values G_3 and G_4 were obtained using cross section data from ENDF/B III and ENDF/B IV files respectively.
Values G_1 were obtained using activation cross sections from SAND-II library and total cross sections from BNL-325.
Values G_2 were obtained using σ_c from SAND-II library and σ_t from ENDF/B III file.
$\Delta G_i/G_i$ denotes the relative deviation $(G_{exp}-G_i)/G_i$.
The cadmium cut-off energy of the 1/E neutron spectrum was 0.622 eV, except that for G_4 a value of 0.68 eV was used.

foil thickness (in mm)	position	a) G_{exp}	G_1	G_2	G_3	G_4	$\Delta G_1/G_1$ (in %)	$\Delta G_2/G_2$ (in %)	$\Delta G_3/G_{ref}$ (in %)	$\Delta G_4/G_{ref}$ (in %)
4.7244	A	0.910	0.933	0.928	0.897	0.948	-2.47	-1.94	+1.4	-4.0
4.798	A	0.902	0.933	0.928	0.896	0.948	-3.32	-2.80	+0.7	-4.8
4.801	B	0.930	0.939	0.927	0.896	0.948	-1.00	+0.32	+3.8	-1.9
4.851	B	0.940	0.932	0.927	0.895	0.947	+0.86	+1.40	+5.0	-0.8
1.074	A	0.868	0.880	0.872	0.817	0.904	-1.36	-0.46	+6.2	-4.0
1.128	B	0.850	0.876	0.868	0.811	0.901	-4.29	-2.12	+4.8	-5.6
1.270	A	0.828	0.862	0.853	0.791	0.892	-3.94	-3.02	+4.7	-7.1
1.270	B	0.836	0.862	0.853	0.791	0.892	-3.02	-2.03	+5.7	-6.7
2.210	A	0.780	0.809	0.798	0.713	0.842	-3.58	-2.31	+9.4	-7.3
2.248	B	0.782	0.807	0.796	0.710	0.840	-3.10	-1.79	+10.1	-6.8
5.207	A	0.686	0.695	0.681	0.551	0.732	-1.29	+0.73	+24.5	-6.2
5.207	B	0.692	0.695	0.681	0.551	0.732	-0.43	+1.62	+25.6	-5.8

a) These experimental values were taken from the work by Baumann [10].

Table 5. Selfshielding for copper foils in the CFRMF neutron spectrum.

Values G_3 and G_4 were obtained using σ_c and σ_t from ENDF/B III and ENDF/B IV files respectively.
Values G_2 were obtained using σ_c from SAND-II library and σ_t from ENDF/B III file.
ΔG_i denotes the relative deviation $(G_{exp} - G_i)/G_i$.

foil thickness (in mm)	G_{exp} a)	G_2	G_3	G_4	$\Delta G_2/G_2$ (in %)	$\Delta G_3/G_3$ (in %)	$\Delta G_4/G_4$ (in %)
1.270×10^{-3}	1.000	0.998	0.997	0.999	+0.20	+0.30	+0.14
2.540×10^{-3}	0.966	0.972	0.952	0.977	-0.62	+1.47	-1.10
1.267×10^{-3}	0.934	0.907	0.843	0.917	+2.98	+10.79	-1.84

a) These experimental values were taken from the work by McElroy [8].

Table 6. Selfshielding for copper foils in the ΣΣ neutron spectrum.

Values G_3 and G_4 were obtained using σ_c and σ_t from ENDF/B III and ENDF/B IV files respectively.
Values G_2 were obtained using σ_c from SAND-II library and σ_t from ENDF/B III file.

foil thickness (in mm)	G_{exp} a)	G_2	G_3	G_4	ΔG_2 (in %)	ΔG_3 (in %)	ΔG_4 (in %)
2.79×10^{-2}	0.900	0.977	0.957	0.981	-8.56	-6.33	-8.23
5.58×10^{-2}	0.854	0.960	0.927	0.965	-12.41	-8.55	-11.5
1.16×10^{-1}	0.822	0.931	0.877	0.939	-13.26	-6.69	-12.4
1.65×10^{-1}	0.800	0.911	0.845	0.921	-13.86	-5.63	-13.1
2.23×10^{-1}	0.778	0.891	0.813	0.902	-14.52	-4.50	-13.7

a) These experimental values were taken from the work by Fabry [7].

Table 7. Selfshielding for ^{235}U fission in the CFRMF neutron spectrum.

Values G_4 were obtained using σ_f and σ_t from ENDF/B-IV file.
ΔG_4 denotes the relative deviation $(G_{exp} - G_4)/G_4$.

foil thickness (in mm)	G_{exp} a)	G_4	ΔG_4
1.270×10^{-3}	1.000	1.000	0.0
1.270×10^{-1}	0.985	0.982	+ 0.27
2.540×10^{-1}	0.964	0.966	- 0.26

a) These experimental values were taken from the work by McElroy [8].

Table 8. Selfshielding for cobalt in the CFRMF neutron spectrum.

Values G_4 were obtained using σ_c and σ_t from ENDF/B-IV file.
ΔG_4 denotes the relative deviation $(G_{exp} - G_4)/G_4$.

foil thickness (in mm)	G_{exp} a)	G_4	ΔG_4
1.270×10^{-3}	1.000	0.922	+ 8.4
3.810×10^{-2}	0.788	0.500	+57.6
5.080×10^{-2}	0.700	0.460	+52.2

a) These experimental values were taken from the work by McElroy [8].

Annotation to the figures

The following figures show the ENDF/B IV cross section curves for the foil reactions considered in this report.
The plots were made from data in the SAND-II group structure and were derived from the ENDF/B IV dosimetry file using the programs BARSKY and CSTAPE.
Curves a) refer to the uncorrected cross section data, and curves b) refer to the cross section data corrected for selfshielding.
Curves for the corrected cross section data were made for the following foil thickness.

Figure	Foil material	Thickness t (in mm)
2	copper	0.5207
5	gold	0.800
8	uranium	0.254
10	cobalt	0.381

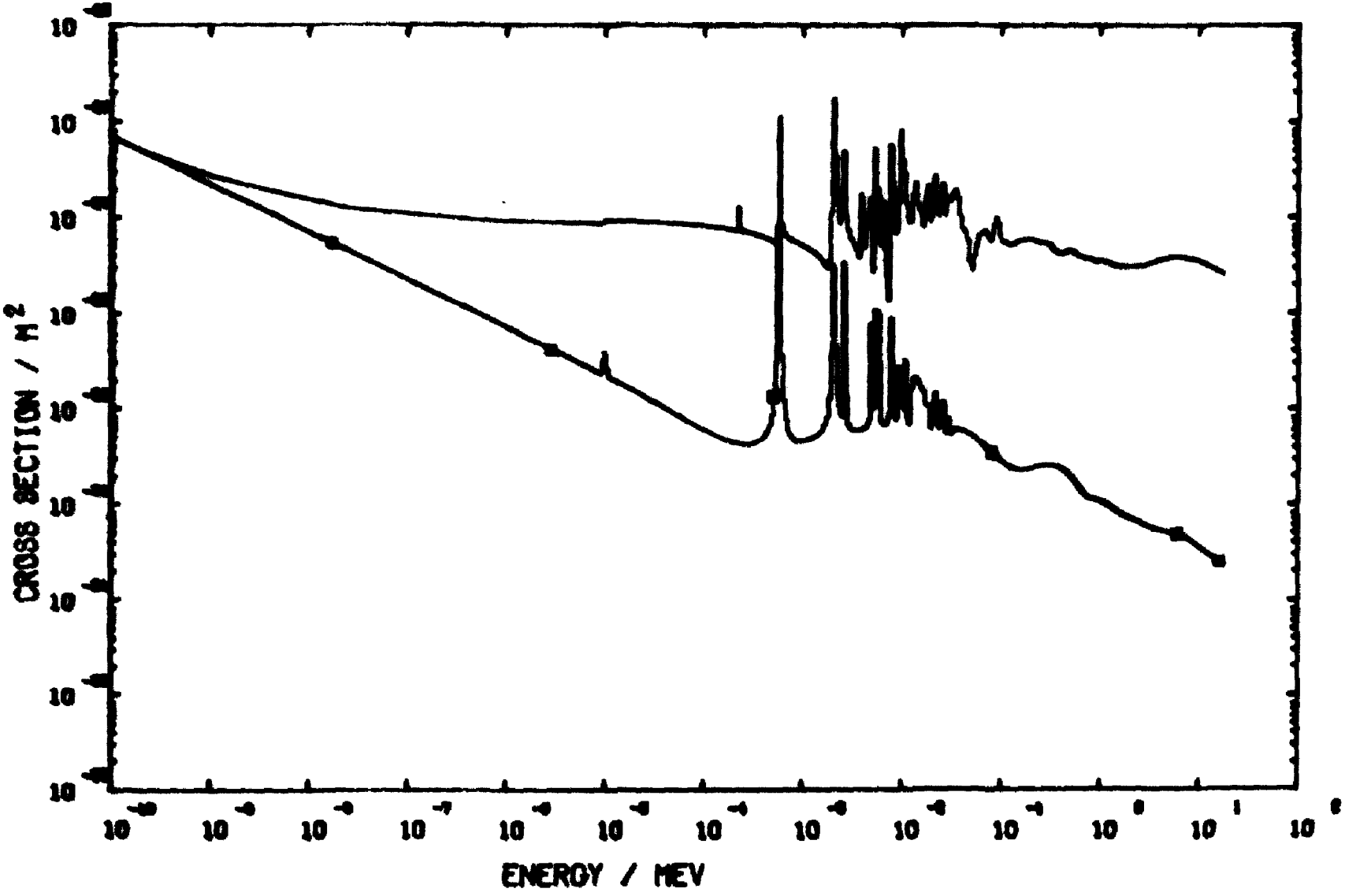

FIG. 1 CROSS SECTION CURVE FOR THE REACTION CU63(TOTAL)

CROSS SECTION CURVE FOR THE REACTION CU63(N,G)CU64 ■ ■ ■

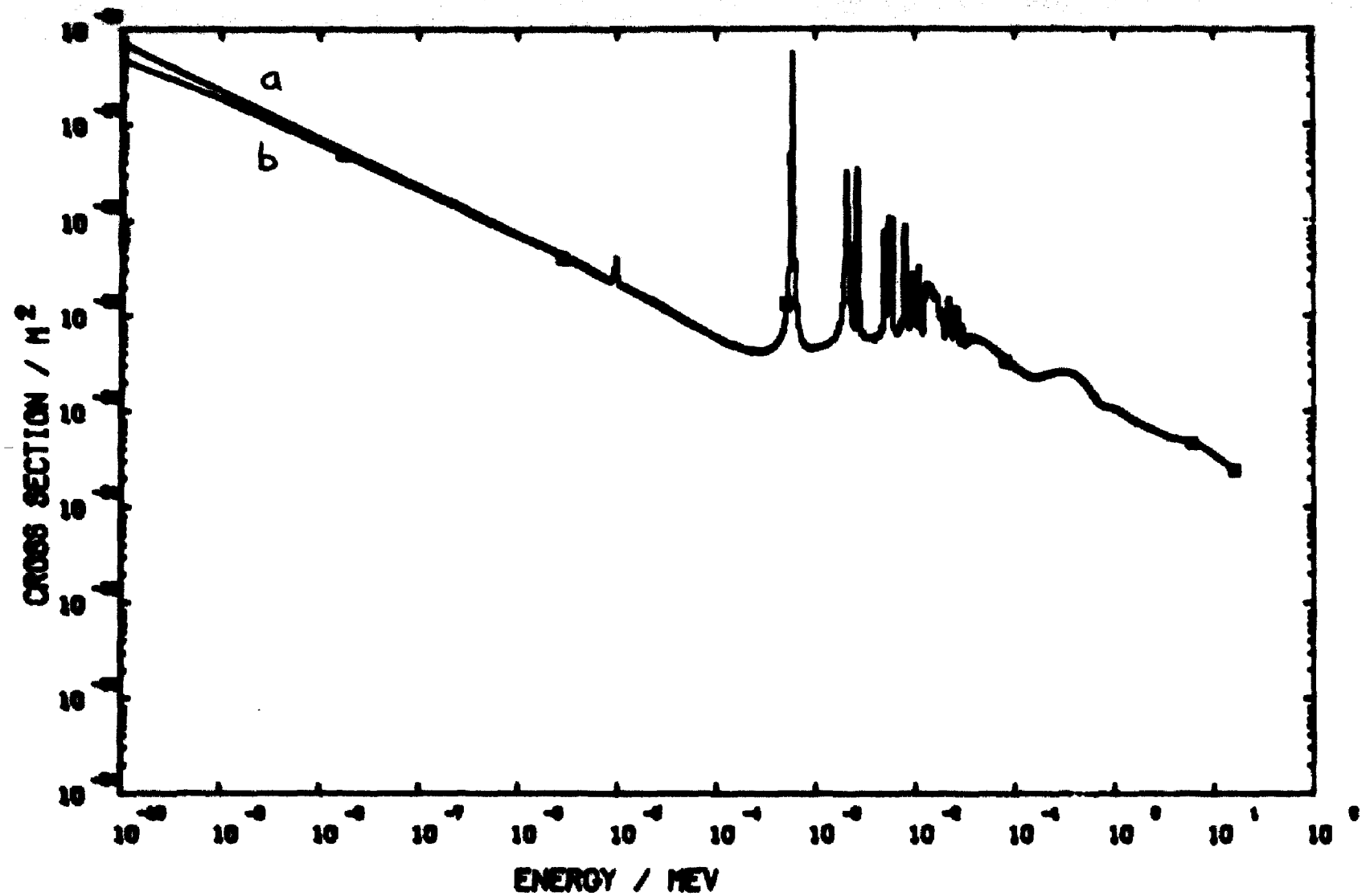

FIG. 2 CROSS SECTION CURVE FOR THE REACTION CU63(N,G)CU64

CROSS SECTION CURVE FOR THE REACTION CU63(N,G)CU64 ■ ■ ■

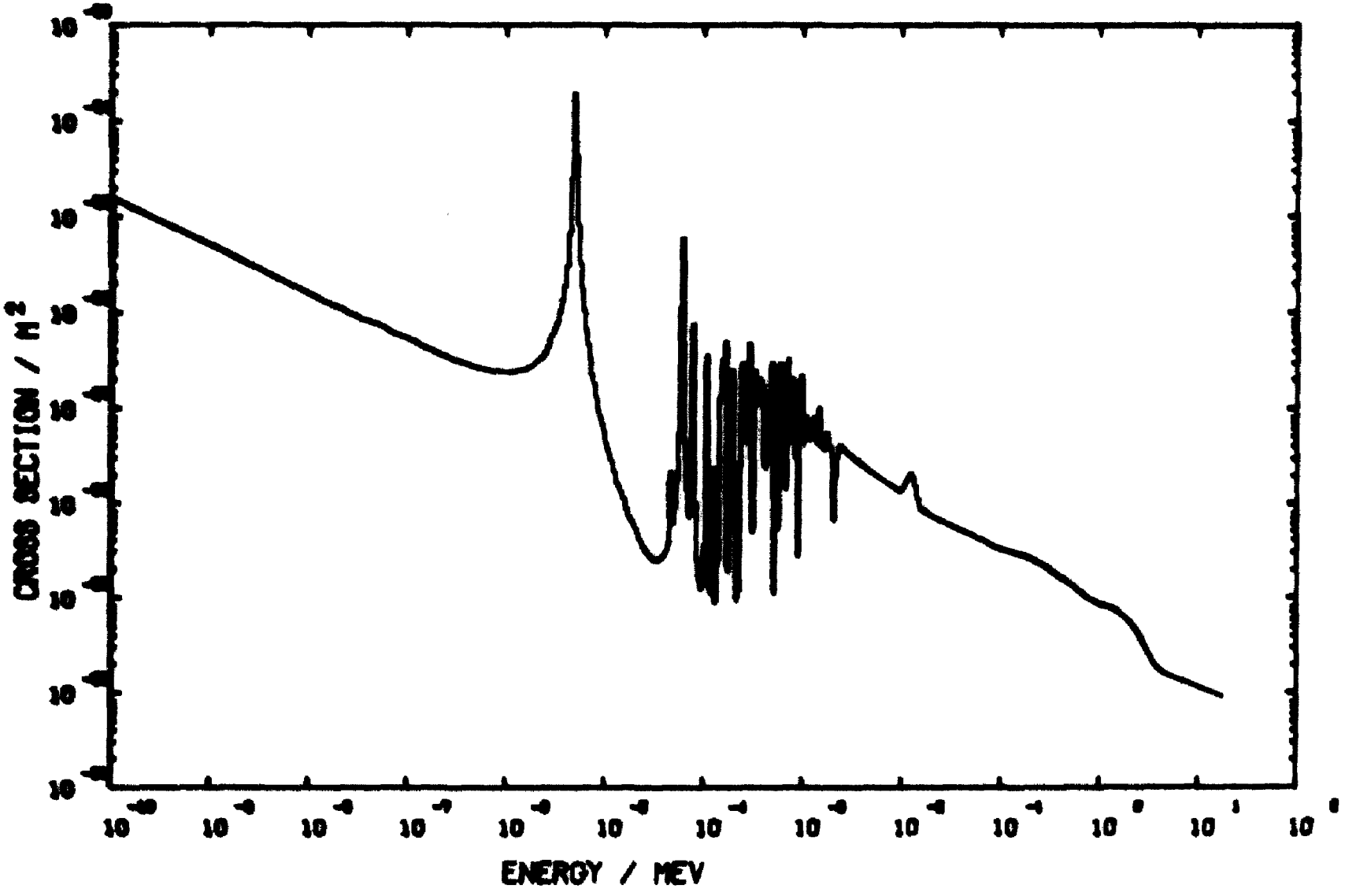

FIG. 3 CROSS SECTION CURVE FOR THE REACTION AU197(N,G)AU198

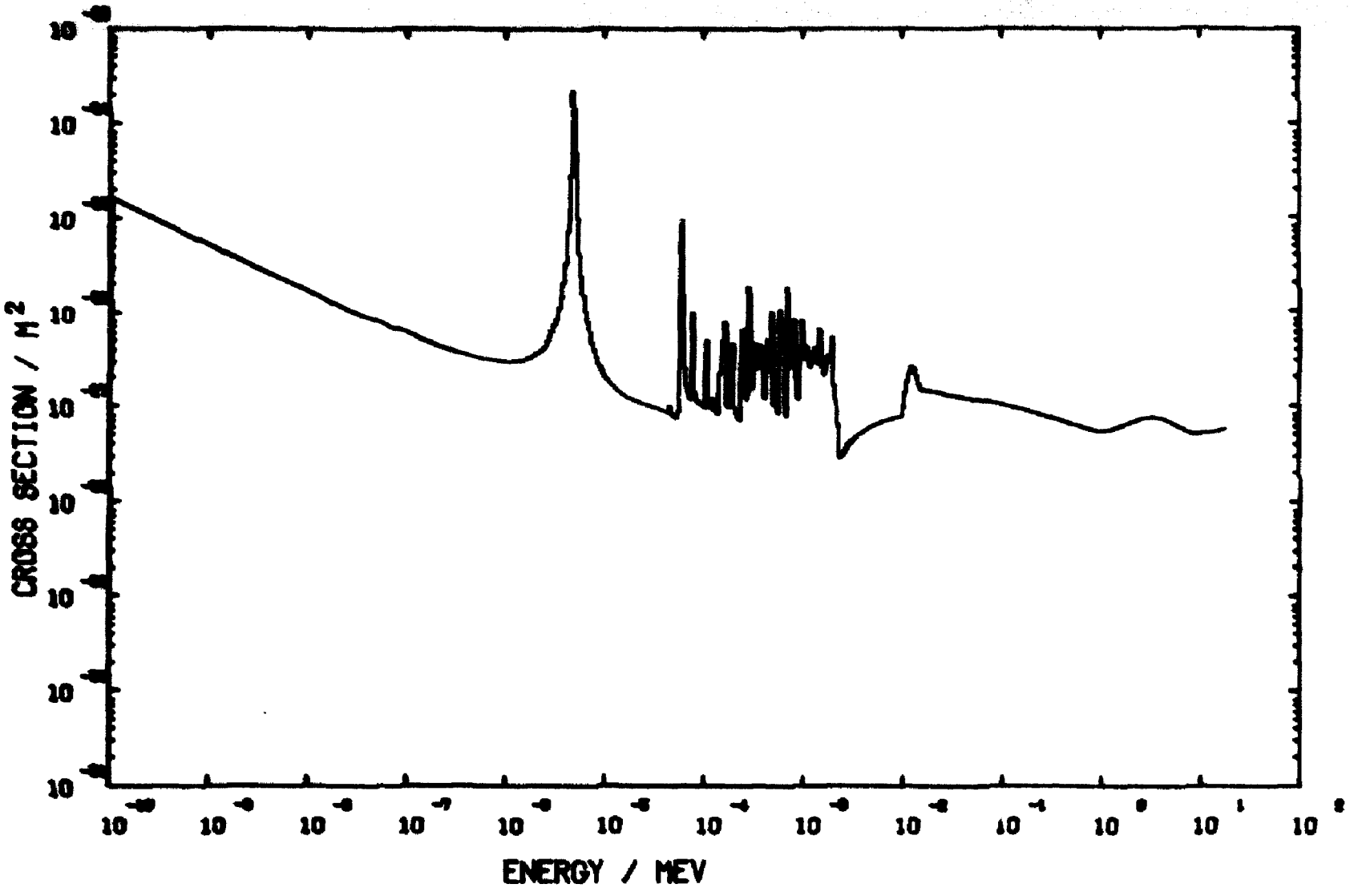

FIG. 4 CROSS SECTION CURVE FOR THE REACTION AU197(TOTAL)

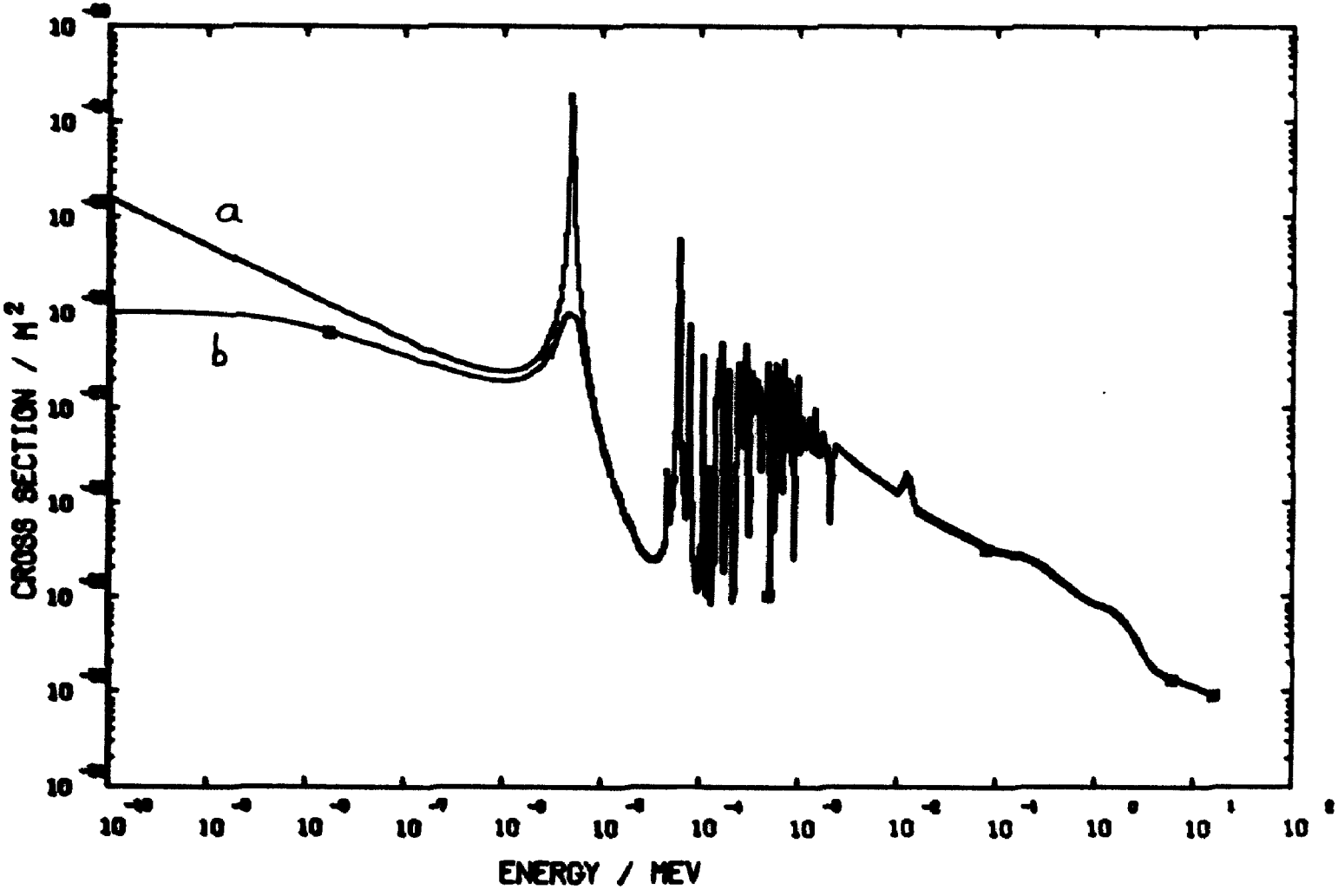

FIG. 5 CROSS SECTION CURVE FOR THE REACTION AU197(N,C)AU198

CROSS SECTION CURVE FOR THE REACTION AU197(N,G)AU198 = = =

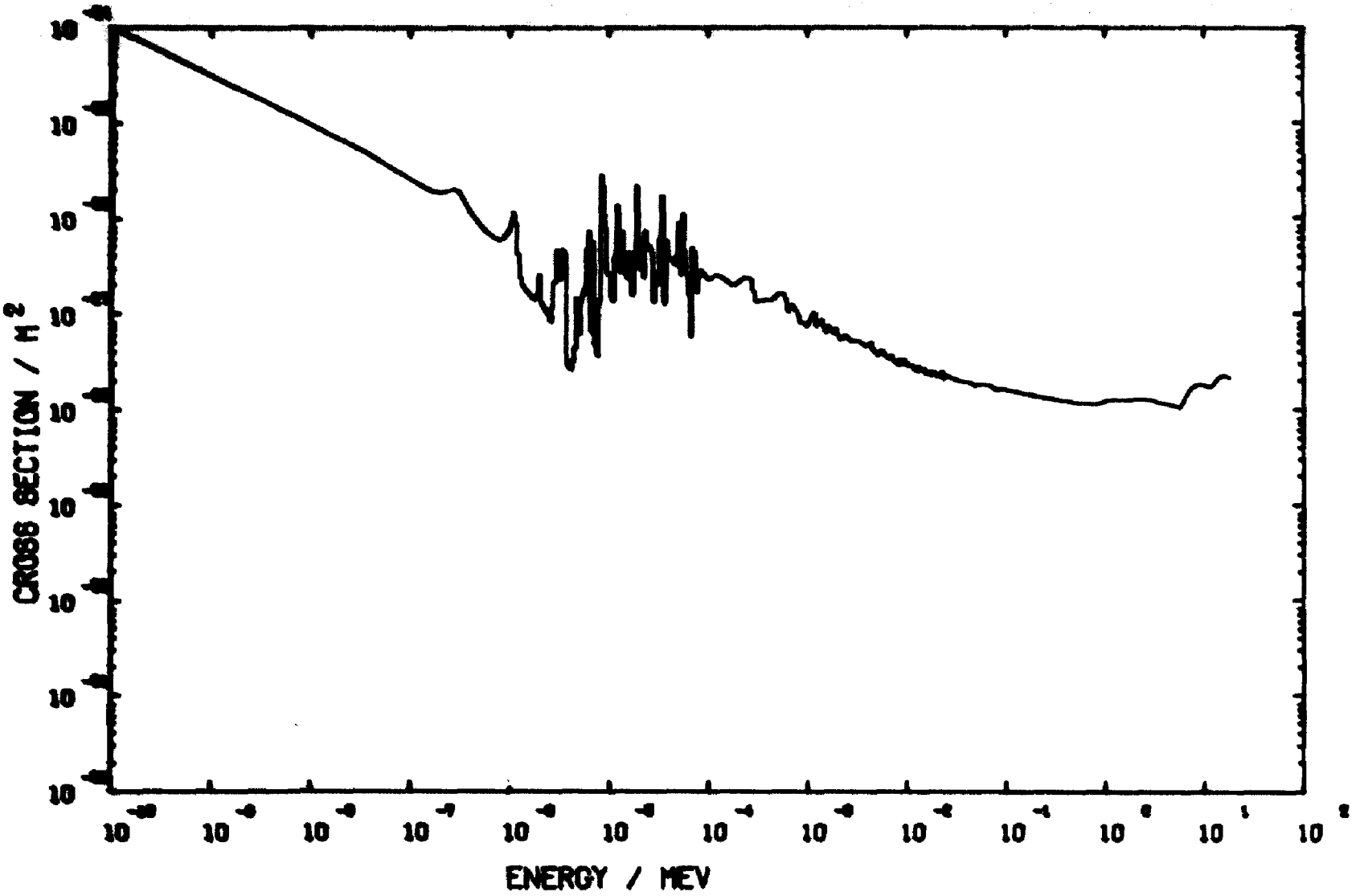

FIG. 6 CROSS SECTION CURVE FOR THE REACTION U235(N,F)FP

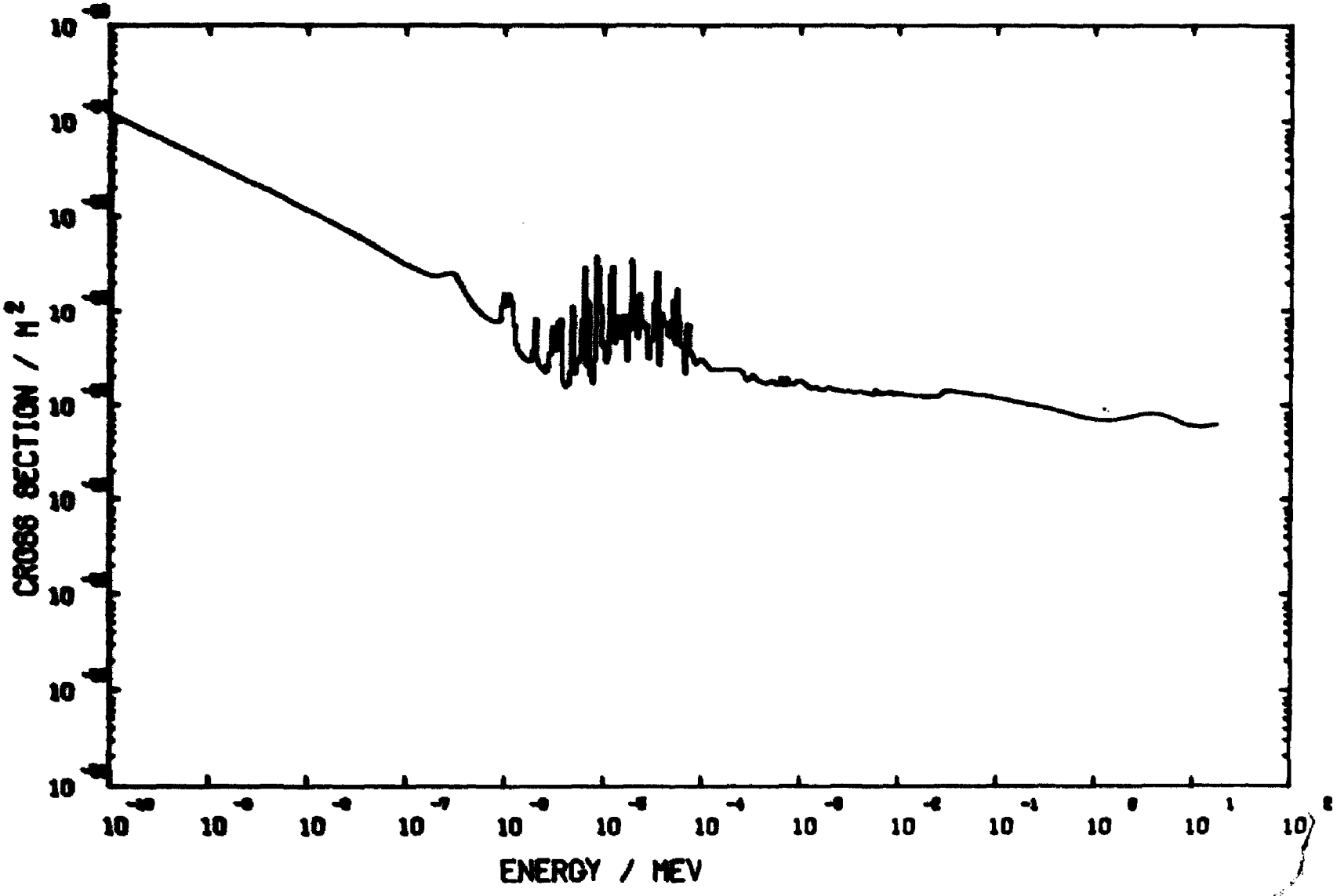

FIG. 7 CROSS SECTION CURVE FOR THE REACTION U235(TOTAL)

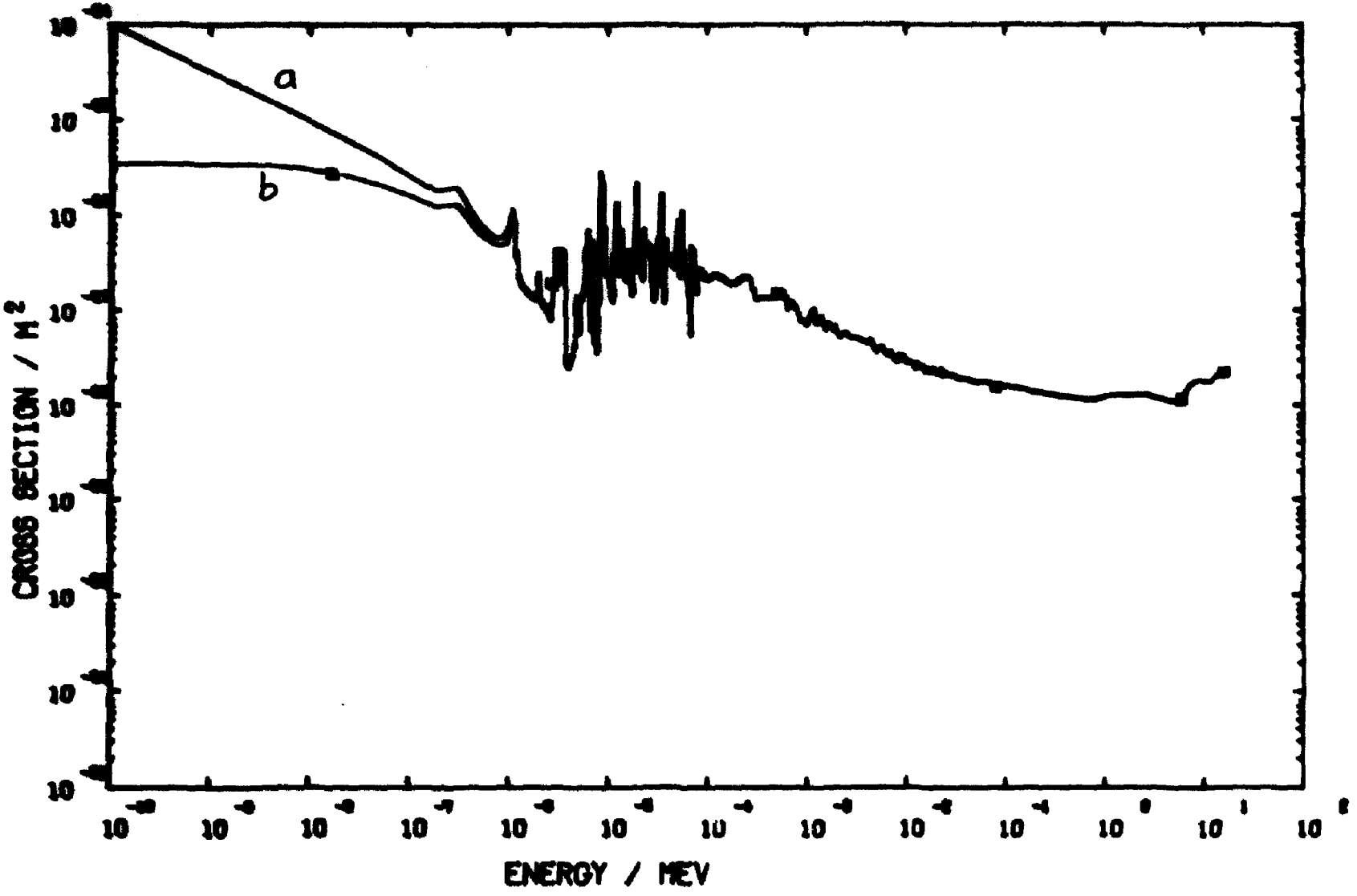

FIG. 8 CROSS SECTION CURVE FOR THE REACTION U235(N,F)FP

CROSS SECTION CURVE FOR THE REACTION U235(N,F)FP ■ ■ ■

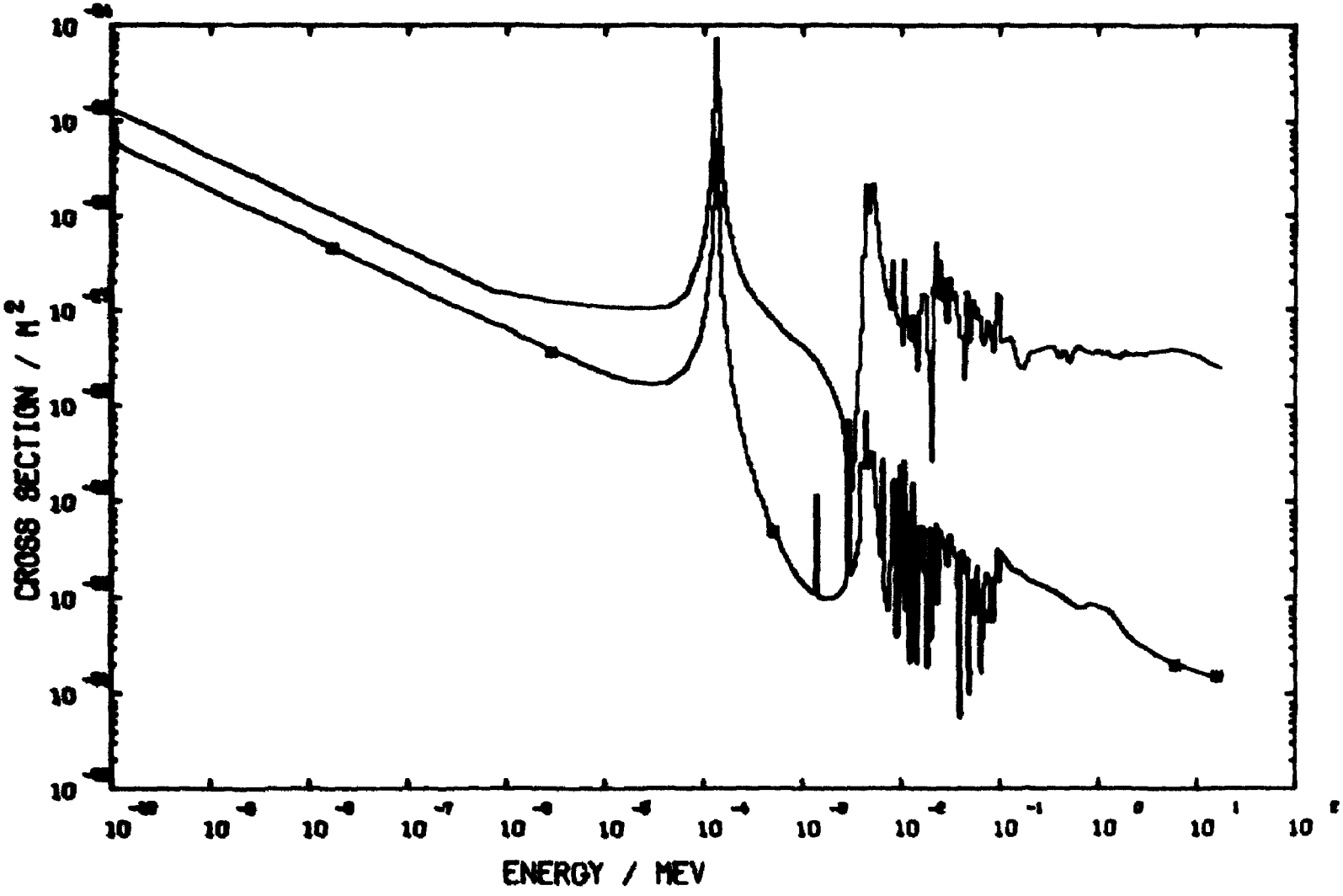

FIG. 9 CROSS SECTION CURVE FOR THE REACTION C059(TOTAL)

CROSS SECTION CURVE FOR THE REACTION C059(N.G)C060 ■ ■ ■

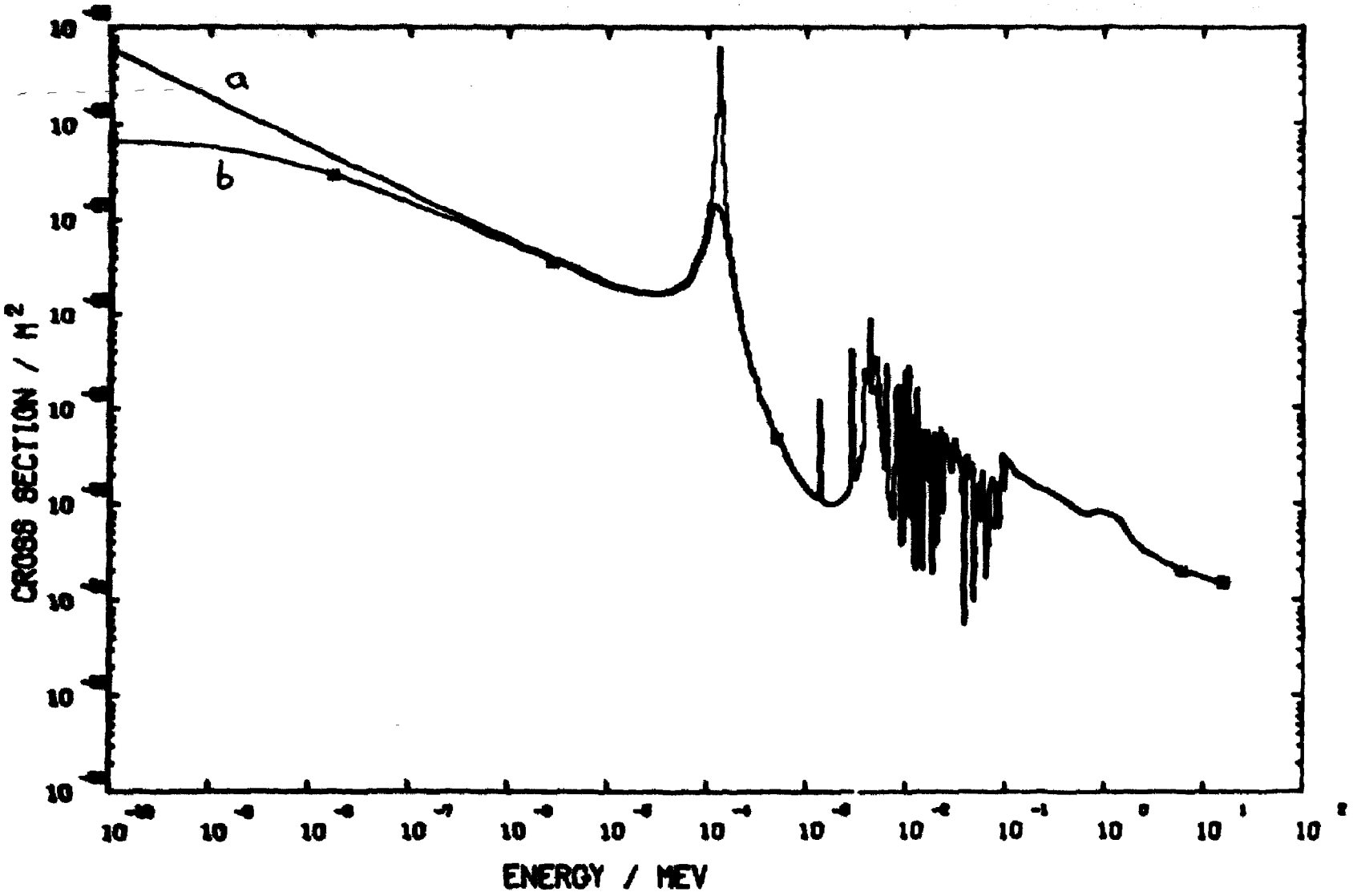

FIG. 10 CROSS SECTION CURVE FOR THE REACTION C059(N,G)C060

CROSS SECTION CURVE FOR THE REACTION C059(N,G)C060 ■ ■ ■