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AUTHOR(S): Leona Stewart
Los Alamos Scientific Laboratory

and

Robert J. Howerton
Lawrence Livermore Laboratory

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MASTER

WHAT HAPPENS TO THE FISSION PROCESS ABOVE THE 2ND- AND 3RD-CHANCE THRESHOLDS?

Leona Stewart

Los Alamos Scientific Laboratory, University of California
Los Alamos, New Mexico 87545

Robert J. Howerton

Lawrence Livermore Laboratory
Livermore, California 94550

ABSTRACT

Although the multiple fission process is important at high neutron energies, most of the evaluations available today do not include these individual fission cross sections or their associated fission spectra. The representations used in the Los Alamos and Livermore libraries are described and calculations compared with 14-MeV integral experiments available on ^{235}U, ^{238}U, and ^{239}Pu. Further work is needed to clearly delineate the specific problems in order to propose unique solutions.

INTRODUCTION

For several decades, experimentalists have reported a significant increase in the total fission cross section for all fissionable nuclides above the 2nd- and 3rd-chance fission thresholds. As late as the 1970's, however, most exaluators have consistently ignored the individual fission channels (n,n'f and n,2nf) in their analyses of the energy-dependent cross sections and the spectra of the neutrons associated with the fission process. For example, explicit representations of the n,n'f and n,2nf cross sections are omitted in all of the ENDF/B-IV evaluations except for ^{235}U, ^{238}U, ^{239}Pu, and ^{240}Pu.† The evaluations of Howerton included in the LLL-ENDL files [1] represent these process implicitly by presenting a total fission cross section with pre-processed tabular energy distributions derived from consideration of the individual fission channels. While the LASL and LLL evaluations differ in form of presentation, both laboratories take into account the 2nd-, 3rd-, and 4th-chance fission processes. On the other hand, the evaluations of Konshin [2] and Sowerby et al. [3] deal only with the total fission cross sections and thereby ignore the multiple-chance fission processes.

It is thus appropriate at this time to bring the needs of the evaluators and users to the attention of the experimentalists and theorists involved in the study of the fission process. Because little is known about 2nd- and 3rd-chance fission, except that the competing channels exist, the evaluator must make estimates in order to present hopefully reasonable spectral information of the fission neutrons. The first known attempts to represent these processes were made and published by Howerton [4] and, in fact, the representations used today are not changed very much from their original attempt.

THE MULTIPLE FISSION PROCESS

In much of the discussion which follows, ^{238}U has been chosen as an example. Our conclusions, however, apply to all of the fissionable nuclides.

Figure 1 is a schematic showing the reaction channels available when neutrons are incident on ^{238}U. Although the diagram is simplistic, it is not intended to limit the interactions to compound nuclear processes. For example, the $(n,n'\gamma)$ channel includes both pre-equilibrium and compound nuclear reactions. Note that first-chance fission defines the fissioning of the aggregate nucleus ^{239}U; second-chance, ^{238}U; third-chance, ^{237}U, etc.

Figure 2 shows the fission cross sections for ^{238}U for each individual fission channel. While this representation is taken from ENDF/B-IV, the ENDL library is quite similar in all of the aspects discussed here. Note that first-chance fission is assumed to be constant upon the onset of second-chance fission. This is in contradiction to the evaluation of Tuttle [5] who reduced the first-chance fission cross section to approximately zero immediately upon the onset of second-chance fission.

In most of the evaluations used today, the emission of charged-particles is assumed to be zero due to the high Coulomb barrier and the reportedly low charged-particle yields for the few experiments available. With this assumption, the only channels available to the system below the (n,2n) and (n,n'f) threshold (6.07 MeV for ^{238}U), are the elastic, (n,γ), $(n,n'\gamma)$ and (n,f). At 11.51 MeV, the (n,3n) and (n,2nf) channels open and lend to the confusion of separating the competition into individual channels.

Although the total fission cross section $(\sigma_{n,F})$†† may be well determined, the spectra of the neutrons associated with the fission process are not, especially in the MeV range. The problem is often related to the method used in the determination of the spectra; for example, most measurements are made of the total neutron emission cross section, that is

$$\sigma_{emis} = \sigma_{n,n} + \sigma_{n,n'} + 2\sigma_{n,2n} + 3\sigma_{n,3n} + \bar{\nu}\sigma_F + \ldots, \qquad (1)$$

and usually restricted to data taking at one angle, only. Unfolding the measurements in order to obtain the fission spectra is subject to large errors due to the many assumptions which must be introduced. Following a suggestion of Batchelor et al. [6], Howerton and Doyas [7,8] investigated fission temperatures in 1969 and 1971. The main thrust of the Batchelor et al. suggestion was that the value of $\bar{\nu}$ used in the well-known Terrell relationship [9] should be appropriate only to that fraction of the neutrons which comes from the direct fission process. The practical consequence of this suggestion is that assumptions must be made in the separation of the direct, 2nd-, and 3rd-chance fission processes above the n,n'f; n,2nf; and n,3nf thresholds. After attributing these factions, a quantity $\bar{\nu}_f(E)$ can be deduced that is more appropriate for application in the Terrell relationship.

It is readily apparent from Eq. (1) that few of the cross sections are well known at energies near 14 MeV. Almost nothing is known about the angular or energy distributions of the emitted neutrons, with the possible exception of the elastic (plus some inelastic) cross section. Even though we know that the angular distributions of the fission fragments are often very anisotropic and we include the fact that the neutrons emitted at the scission point are emitted from the moving fragments, all of the evaluations in use today contain the assumption that the fission neutrons are emitted isotropically in the laboratory reference frame. Therefore, both the evaluated spectrum and angle of emission of the fission neutrons are often incorrect.

The final sine qua non of the fission process that must be supplied by the evaluator is $\bar{\nu}(E)$. For several of the most important fissionable isotopes, this quantity has been determined by experiment [10]. In 1964 Schuster and Howerton [11] addressed the problem for uranium with a plausibility argument for the derivation of an empirical relationship between $\bar{\nu}$ and E_n. In 1971, Howerton [12] extended the previous work to provide a method for predicting $\bar{\nu}(E)$ for thorium, uranium, and plutonium isotopes in cases where this quantity has not been determined by measurement. Essentially the same assumptions about the energy dependence of the multiple-chance fission processes were made by Vasil'ev et al. [13] who also introduced the plausibility of nonlinear variation of $\bar{\nu}(E)$ above the 2nd-chance fission threshold. These authors, however, provided no quantitative estimates of $\bar{\nu}(E)$.

Although not the subject of this paper, it should be noted that the (n,n'), (n,2n), and (n,3n) cross sections are rarely well determined experimentally at high neutron energies and the spectra have not been measured at all. Minimal information can be obtained from the observation of the total emission spectra, at least for the contribution of the pre-equilibrium processes since these stand out above the various fission channels at the high-energy end. The only recent detailed experiments are those of Kammerdeiner [14], who measured the spectra at several angles for 14-MeV neutrons incident on ^{235}U, ^{238}U, and ^{239}Pu.

The main purpose of this paper is to call attention to the fact that the evaluator must supply much more information on fission than a measure of the total fission cross section. For the fissile and fertile materials, measurements of the other cross sections are also very important, especially at the higher energies.

In most of the evaluations in use today, the fission process is treated in one of the following ways:

1. Only the total fission cross section is represented; the fission neutron energy distribution is assumed to be Maxwellian in shape with the average energy increasing with incident neutron energy.

2. The total fission cross section is separated into its various parts; the choice made in ENDF/B-IV is shown in Fig. 2. Then, the neutron (or neutrons) which precedes scission is assumed to be emitted with a spectrum far softer than allowed for the scission neutrons. For example, at 14 MeV for ^{238}U, the two neutrons which come off before scission would have energies between zero and 2.49 MeV (the total energy available to the pre-scission neutrons). Therefore, it is apparent that the treatment of the competition of the first- and second-chance fission process should be an important part of each evaluation.

COMPARISON WITH SOME 14-MeV INTEGRAL EXPERIMENTS

Two different types of integral experiments have been carried out, one at LASL by Ragan et al. [15] which was made on ^{235}U with a multiplication of approximately 10-11, and one at LLL by Wong et al. [16] on ^{235}U, ^{238}U, and ^{239}Pu which are more differential in nature with a multiplication of approximately 0.9 for ^{238}U and 1.4 for ^{235}U and ^{239}Pu. In both experiments, spherical shells of the target surround a 14-MeV neutron source and the neutron spectra emerging from the sphere are recorded at one or more angles with respect to the incident neutron direction.

Figures 3a and 3b compare the spectrum of the neutrons as measured by Ragan et al. [15] and with the calculation using the ENDF/B-IV data file (MAT-1262) and the ENDL evaluation by Howerton [1]. Note that the energy scale in Figs. 3a and 3b is changed near 4 MeV in order to show all of the data on the same graph.

While ENDF/B-IV shows fairly good agreement with experiment except for the energy bins between 6 and 10 MeV, the differences between the ENDF and ENDL evaluations are much larger than one would expect from a perusal of the data in the files, themselves. These differences are better illustrated in Fig. 4, which shows the ratio of the calculated to experimental measurements (C/E) for both the ENDF and ENDL evaluations.

Figure 5 shows the comparable experiment performed on ^{235}U at LLL. To complete the analysis on the available data, the LLL experiments on ^{238}U and ^{239}Pu have been compared with calculations in Figs. 6 and 7. Table 1 gives tabular values of the integrals of the calculated and experimental spectra in three energy domains of the emitted neutrons.

SUMMARY AND CONCLUSIONS

In summary, this paper points out various problem areas in the evaluation of the cross sections and parameters associated with the fissionable nuclides. In addition, the comparison of the ENDF and ENDL libraries with experiment may even suggest errors in the files or in the calculational procedures presently employed. While all of the calculations shown were made using Monte Carlo techniques and thereby suffer somewhat from statistical accuracy, they did include all of the geometrical factors of the experiments. Further work will be undertaken to elucidate these problem areas.

At the same time, however, experimental information above 8-10 MeV is urgently required. For example, a measurement of the fission spectrum at several angles using fragment coincidence techniques would be very useful, especially if carried out at several incident neutron energies. (A need for (n,2n) and (n,3n) experiments using coincidence and anti-coincidence techniques is also apparent as are determinations of the direct and/or pre-equilibrium components of the (n,n'γ) reaction.) At several energies below 9 MeV, the shape of the fission spectrum should be measured at several angles; again a fragment coincidence experiment is required. Similar experiments should be repeated in the 14-Mev range.

Finally, theorists could lend great insight into determining how to treat the fission process, especially in the region above the second- and third-chance fission thresholds. Most of the calculations available today are limited to the study of only a few of the many available channels, while others which are more complete studies of the cross sections do not treat the spectral distributions of any of the emitted neutrons. In addition to the fission cross sections for the individual channels, $\bar{\nu}$ (E), and the energy and angular distributions of the neutrons are important input for the evaluator who must provide these data for neutronics calculations.

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FOOTNOTES

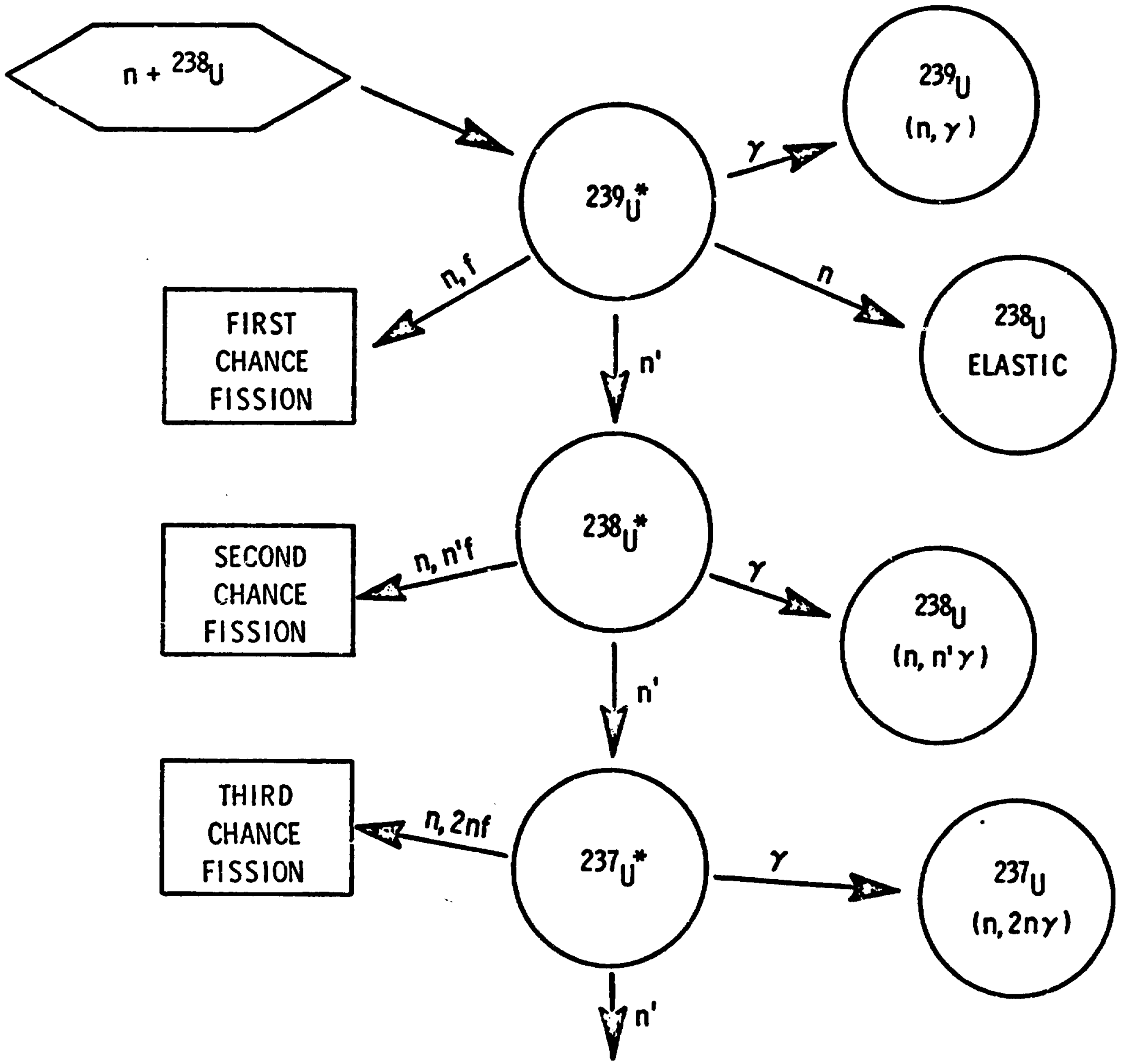
†. Those evaluations currently in ENDF were provided by LASL.

††. Where σ_{nF} is the sum of all the partials.

TABLE I

Comparison of Integrals Under Elastic Peak, Total Integrals, and Integrals from the Elastic Peak to ~2.0 MeV

Nuclide	Evaluated Library	$\int_0^{401} N(t)dt$			$\int_0^{161} N(t)dt$			$\int_{161}^{401} N(t)dt$		
		Exp.	Calc.	$\frac{\text{Calc-Exp}}{\text{Exp}}$	Exp.	Calc.	$\frac{\text{Calc-Exp}}{\text{Exp}}$	Exp.	Calc.	$\frac{\text{Calc-Exp}}{\text{Exp}}$
U^{235}	ENDF/B-IV	1.436	1.345	- 6%	.644	.687	+ 8%	.792	.648	-18%
	ENDL	1.436	1.330	- 7%	.644	.672	+ 4%	.792	.658	-17%
U^{238}	ENDF/B-IV	.907	.869	- 4%	.643	.655	+ 2%	.264	.214	-19%
	ENDL	.907	.892	- 2%	.643	.642	-0.2%	.264	.250	- 5%
Pu^{239}	ENDF/B-IV	1.421	1.381	- 3%	.648	.704	+ 9%	.773	.677	-12%
	ENDL	1.421	1.372	- 3%	.648	.736	+14%	.773	.636	-18%



n + 238U
239U*
239U
(n, γ)
γ
n, f
n
FIRST CHANCE FISSION
n'
238U ELASTIC
238U*
SECOND CHANCE FISSION
n, n'f
γ
238U
(n, n'γ)
n'
THIRD CHANCE FISSION
n, 2nf
237U*
γ
237U
(n, 2nγ)
n'

Fig. 1.

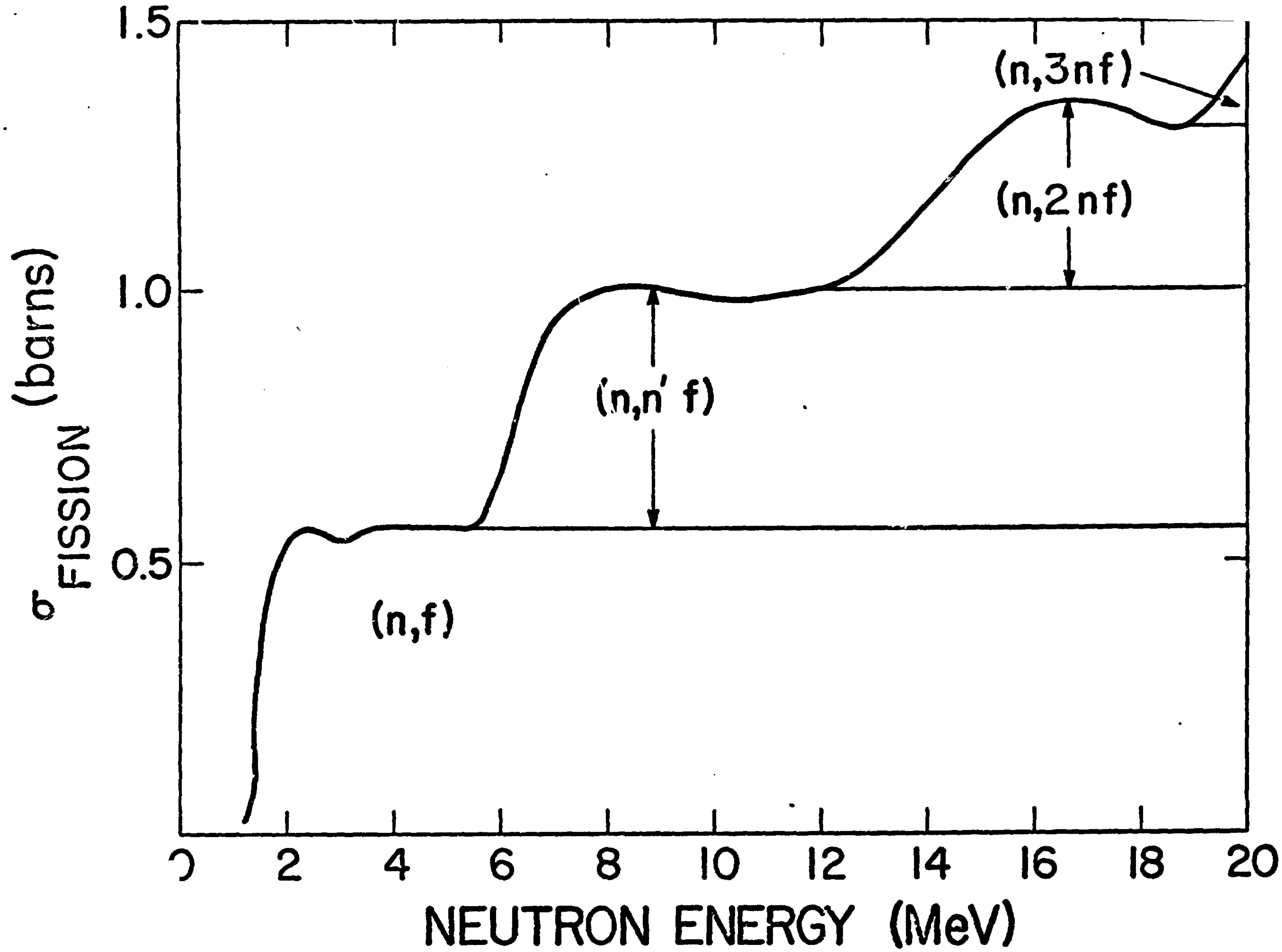
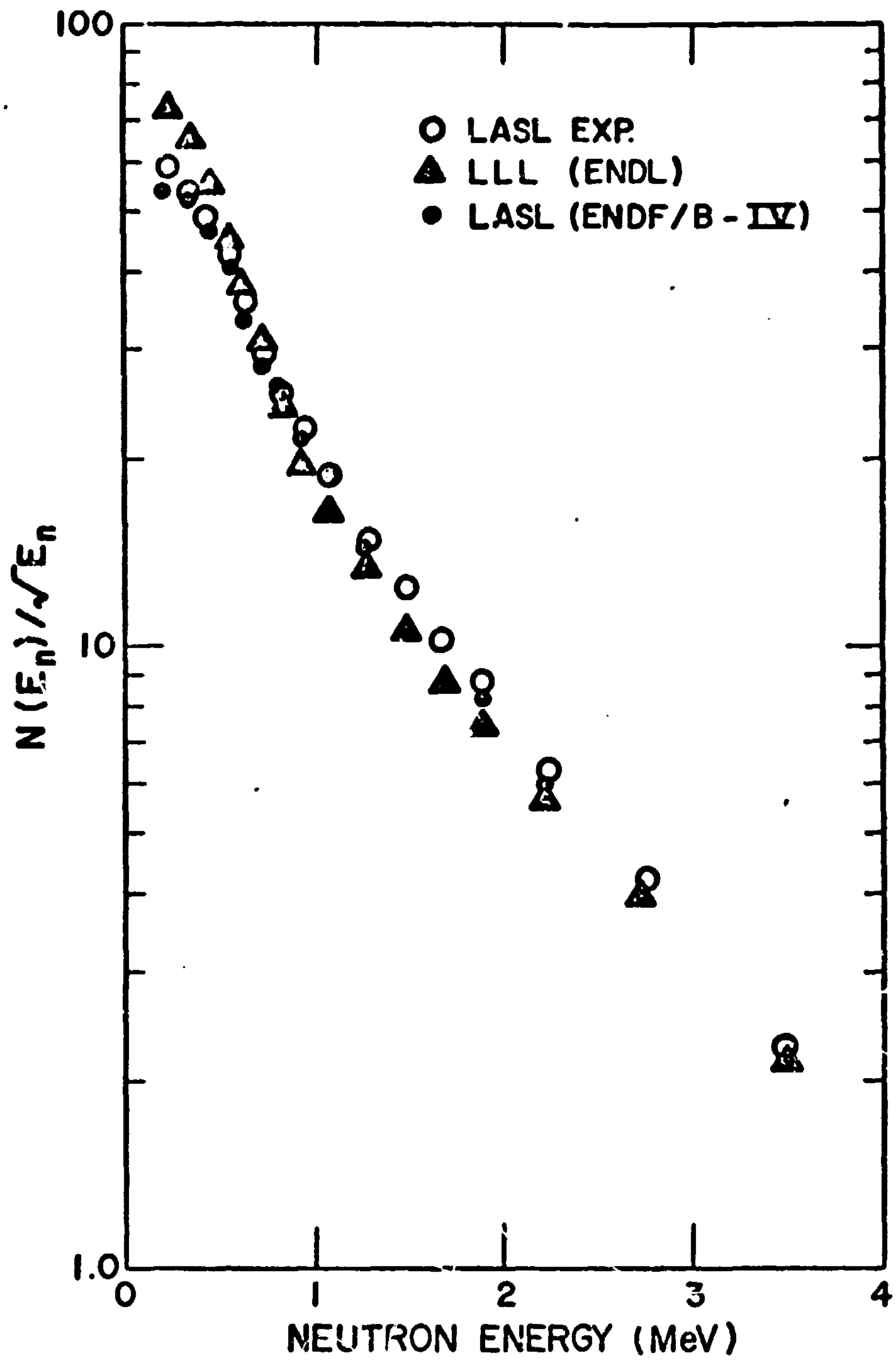

1.5
1.0
0.5
σ FISSION (barns)
(n,3nf)
(n,2nf)
(n,n′ f)
(n,f)
0
2
4
6
8
10
12
14
16
18
20
NEUTRON ENERGY (MeV)

Fig. 2.



LASL EXP.
LLL (ENDL)
LASL (ENDF/B-IV)
100
10
1.0
$N(E_n)/\sqrt{E_n}$
0
1
2
3
4
NEUTRON ENERGY (MeV)

Fig. 3a

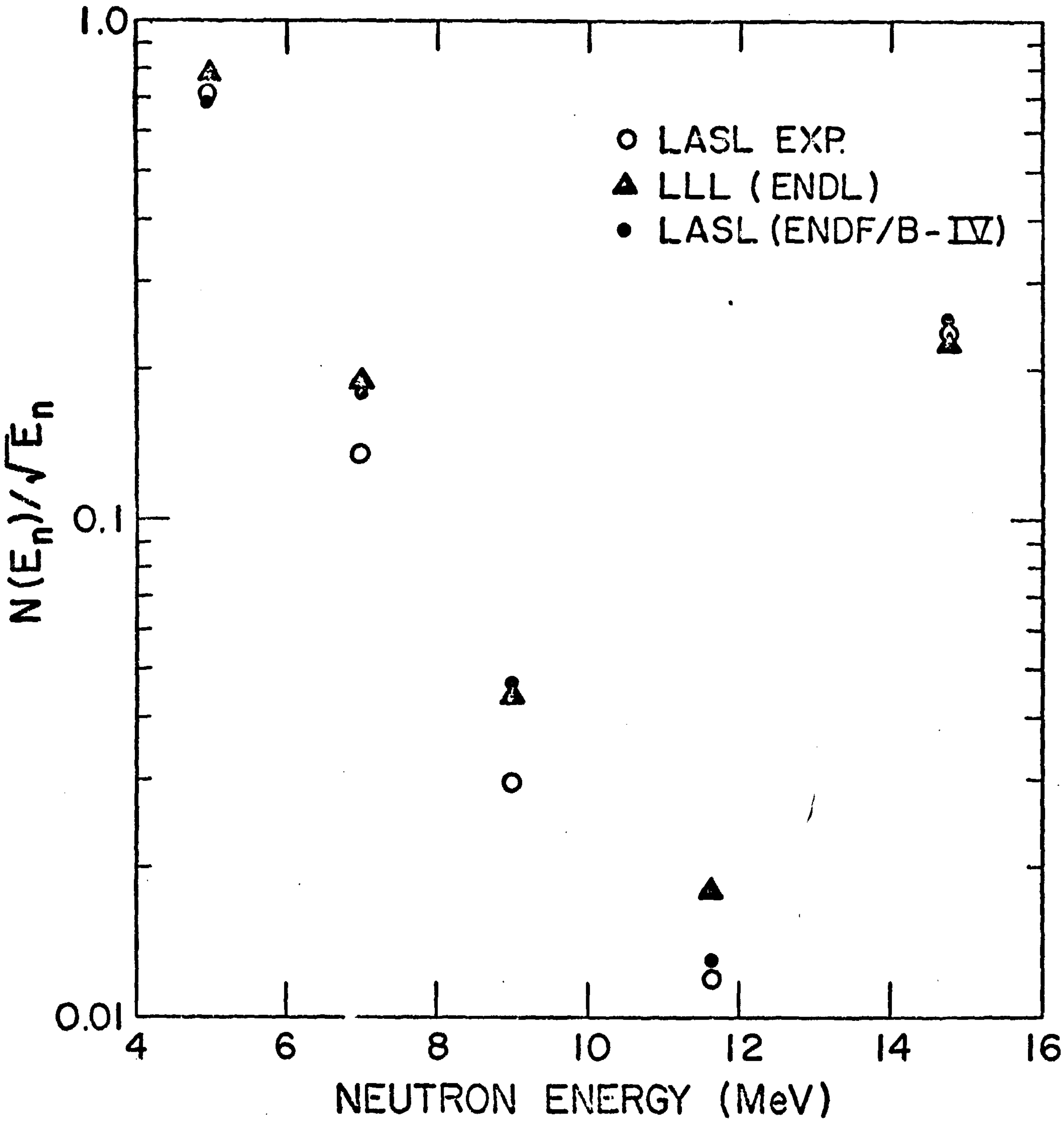
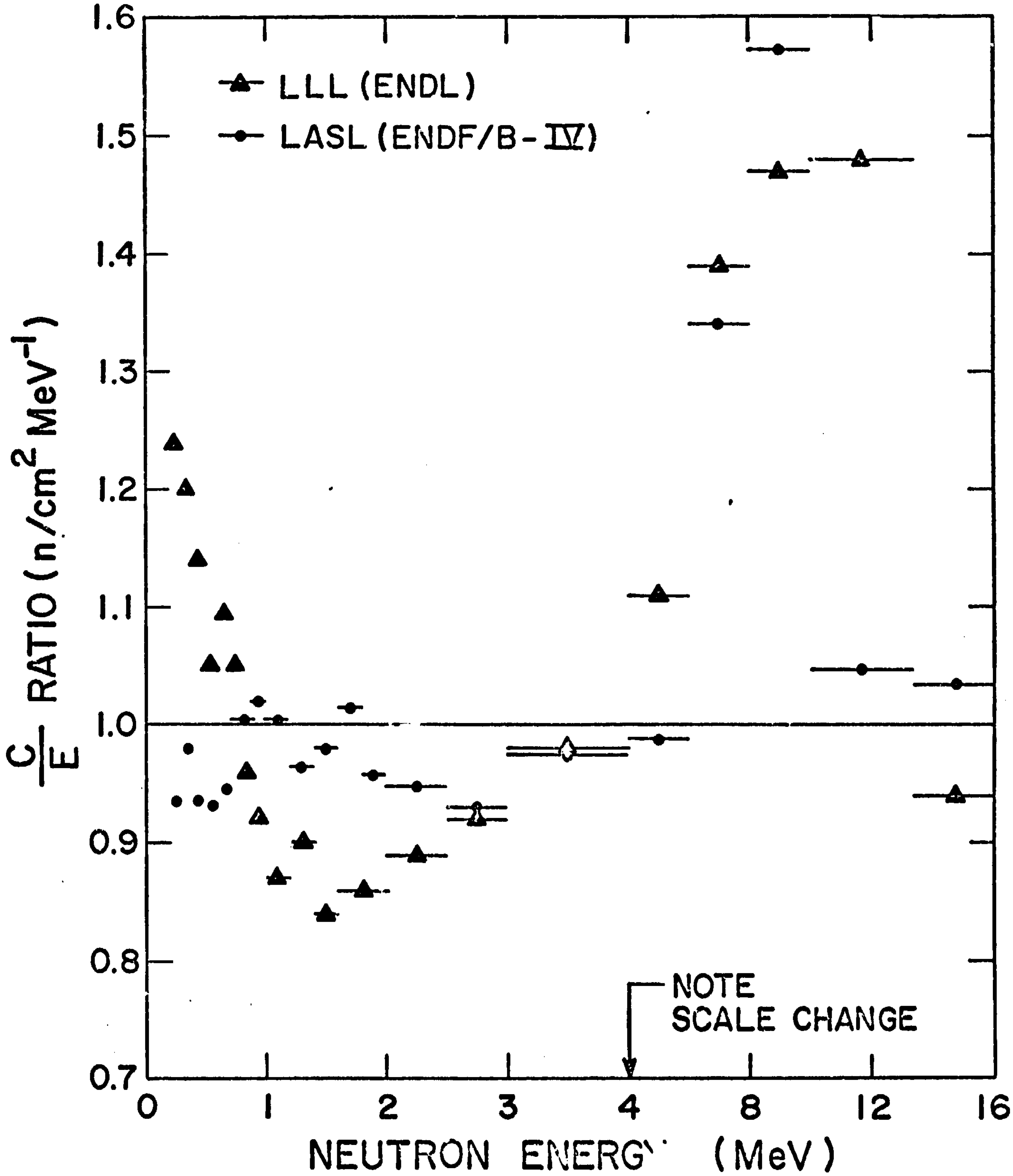

1.0
0.1
0.01
4
6
8
10
12
14
16
N(E_n)/√E_n
NEUTRON ENERGY (MeV)
LASL EXP.
LLL (ENDL)
LASL (ENDF/B-IV)

Fig. 3b



LLL (ENDL)
LASL (ENDF/B-IV)
$\frac{C}{E}$ RATIO (n/cm^2 MeV^{-1})
1.6
1.5
1.4
1.3
1.2
1.1
1.0
0.9
0.8
0.7
0
1
2
3
4
8
12
16
NOTE
SCALE CHANGE
NEUTRON ENERGY (MeV)

Fig. 4.

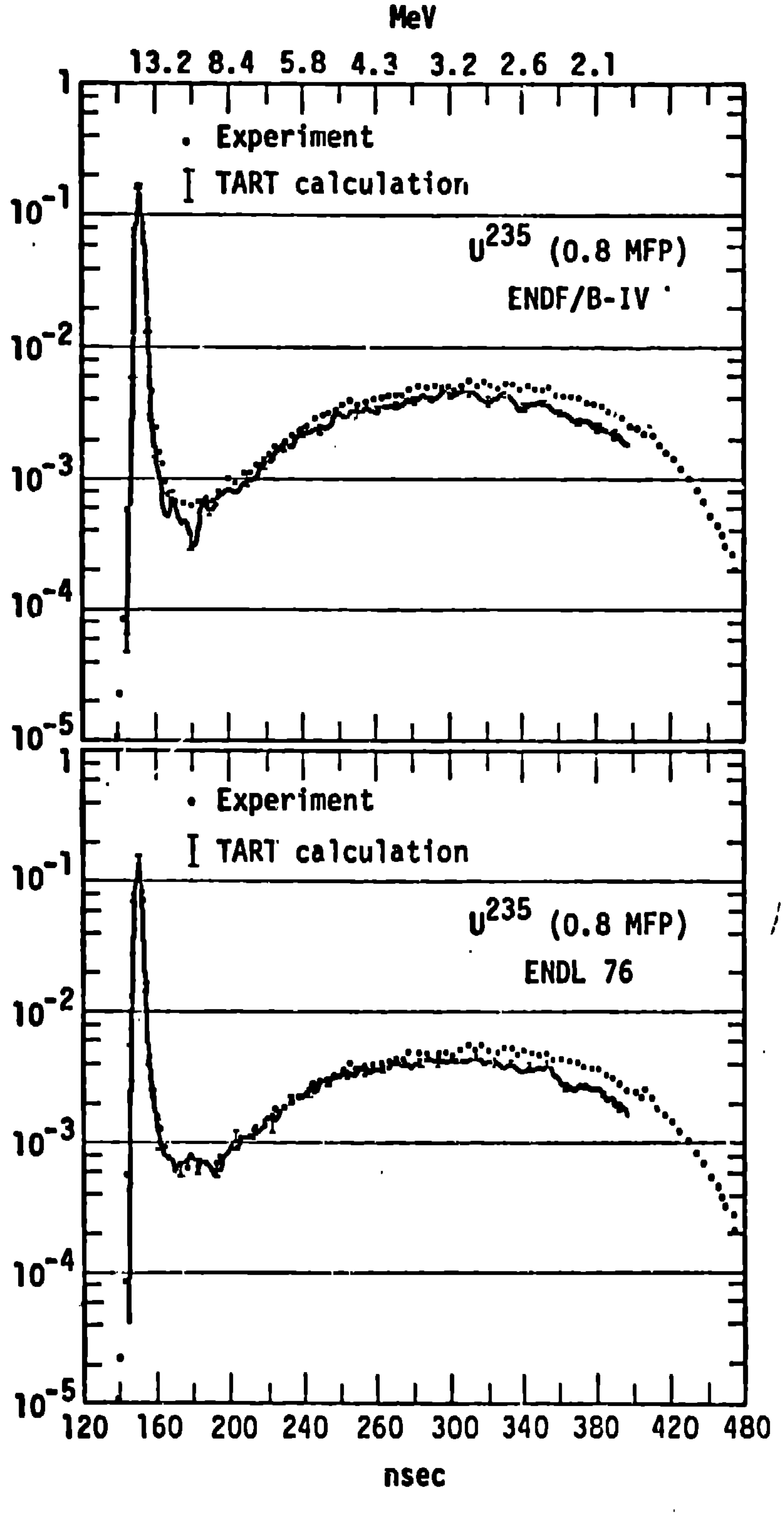

MeV
13.2 8.4 5.8 4.3 3.2 2.6 2.1
1
10^-1
10^-2
10^-3
10^-4
10^-5
• Experiment
I TART calculation
U235 (0.8 MFP)
ENDF/B-IV
• Experiment
I TART calculation
U235 (0.8 MFP)
ENDL 76
120 160 200 240 260 300 340 380 420 480
nsec

Fig. 5.

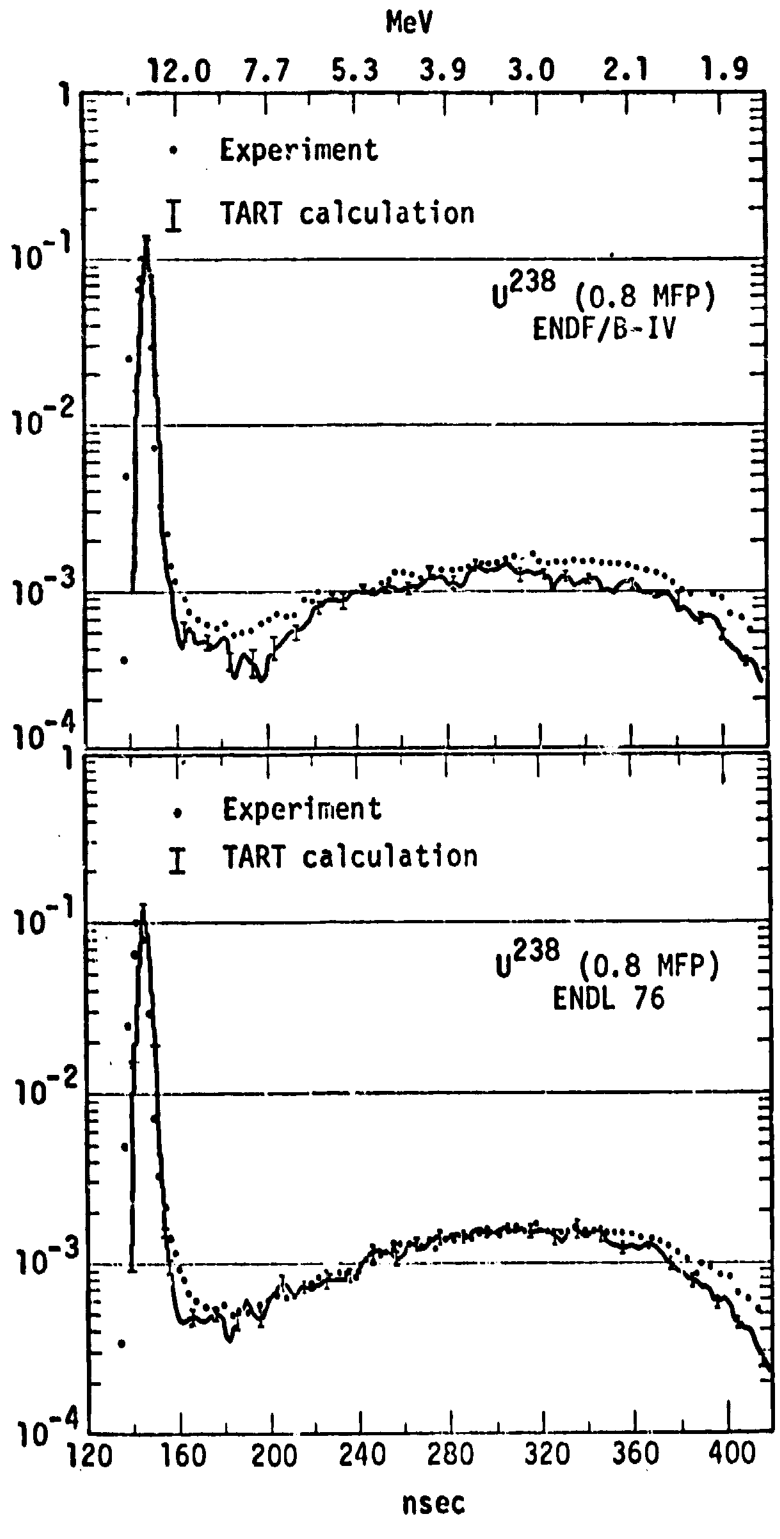
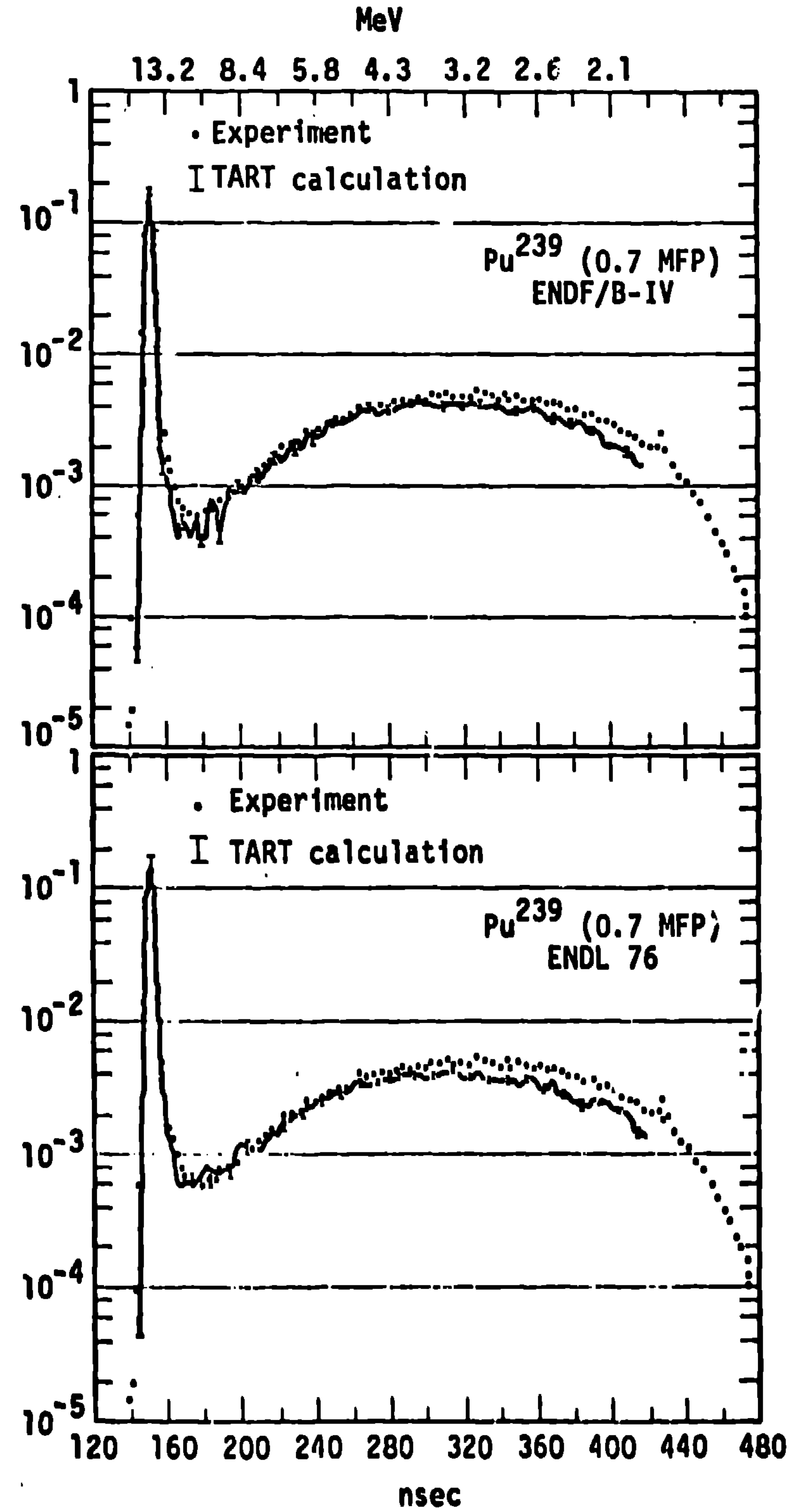

MeV
12.0
7.7
5.3
3.9
3.0
2.1
1.9
Experiment
TART calculation
U^{238} (0.8 MFP)
ENDF/B-IV
Experiment
TART calculation
U^{238} (0.8 MFP)
ENDL 76
1
10^{-1}
10^{-2}
10^{-3}
10^{-4}
120
160
200
240
280
320
360
400
nsec

Fig. 6.



MeV
13.2 8.4 5.8 4.3 3.2 2.6 2.1
• Experiment
I TART calculation
Pu239 (0.7 MFP)
ENDF/B-IV
1
10-1
10-2
10-3
10-4
10-5
• Experiment
I TART calculation
Pu239 (0.7 MFP)
ENDL 76
120 160 200 240 280 320 360 400 440 480
nsec

Fig. 7.

FIGURE CAPTIONS

Fig. 1. Schematic showing the reactions considered for neutrons incident on ^{238}U.

Fig. 2. The Evaluated fission cross sections for ^{238}U taken from ENDF/B-IV. The top curve represents the total fission cross section.

Figs. 3a and 3b. Spectrum of the neutrons from 14-MeV neutrons incident on an oralloy sphere. The experimental points, taken from the experiment of Ragan et al. [15], are compared with calculations using the ENDF/B-IV evaluated library and using the ENDL library. Note that the largest discrepancies between calculation and experiment occur in the energy bins where the flux is down by two to three orders of magnitude.

Fig. 4. The same results shown in Figs. 3a and 3b are plotted as calculated/experimental ratios for each of the neutron energy bins. The differences below 2.5 MeV in the calculations using the two evaluated libraries are not well understood.

Fig. 5. Comparison of calculated and experimental neutron spectra from a 0.8 mean-free-path hollow sphere of ^{235}U with a nominal 14-MeV neutron source at the center. The TART 175 group Monte Carlo neutronics code was used.

Fig. 6. Comparison of calculated and experimental neutron spectra from a 0.8 mean-free-path hollow sphere of ^{238}U with a nominal 14-MeV neutron source at the center. The TART 175 group Monte Carlo neutronics code was used.

Fig. 7. Comparison of calculated and experimental neutron spectra from a 0.7 mean-free-path hollow sphere of ^{239}Pu with a nominal 14-MeV neutron source at the center. The TART 175 group Monte Carlo neutronics code was used.