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ANL/NDM-96

Ratio of the Prompt-Fission-Neutron Spectrum of Plutonium-239 to that of Uranium-235

by

M. Sugimoto, A.B. Smith, and P.T. Guenther

September 1986

**ARGONNE NATIONAL LABORATORY,
ARGONNE, ILLINOIS 60439, U.S.A.**

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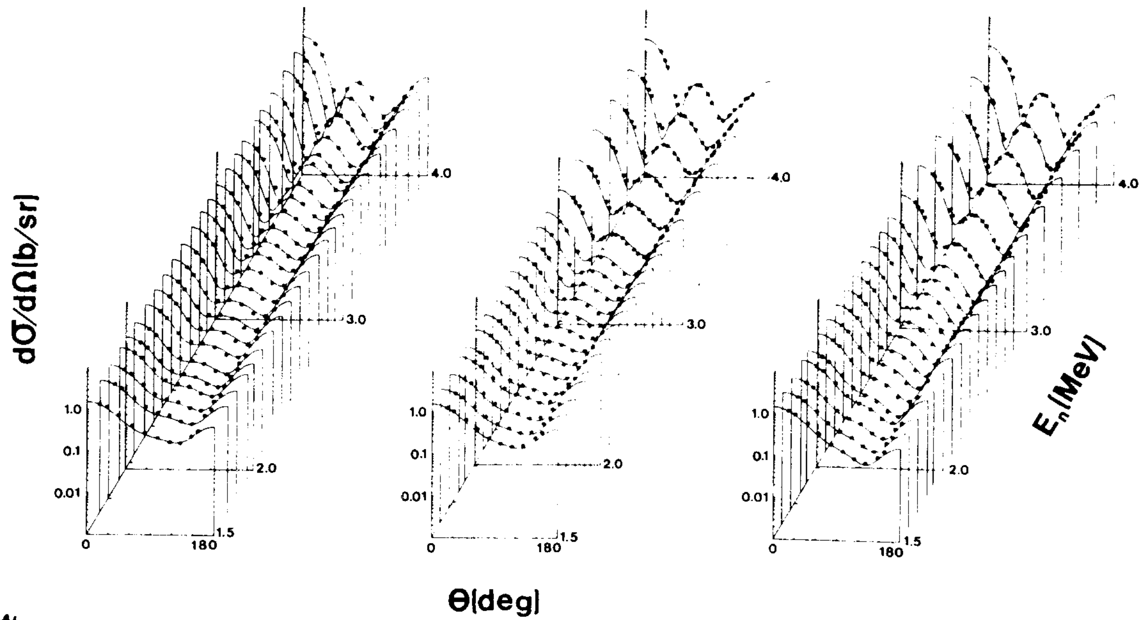

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ANL/NDM-96

RATIO OF THE PROMPT-FISSION-NEUTRON SPECTRUM OF PLUTONIUM 239 TO THAT OF URANIUM 235*

by

Masayoshi Sugimoto[+], Alan B. Smith and Peter T. Guenther

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Keywords:

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[+]Permanent address: Japan Atomic Energy Research Institute, Tokai-Ibaraki, Japan.

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Applied Physics Division
Argonne National Laboratory
9700 S. Cass Avenue
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RATIO OF THE PROMPT-FISSION-NEUTRON SPECTRUM OF PLUTONIUM 239 TO THAT OF URANIUM 235*

by

Masayoshi Sugimoto+, Alan B. Smith and Peter T. Guenther

Applied Physics Division
Argonne National Laboratory

ABSTRACT

The prompt-fission-neutron spectrum resulting from ^{239}Pu fission induced by 0.55 MeV incident neutrons is measured from 1.0 to 10.0 MeV relative to that of ^{235}U fission induced by the same incident-energy neutrons. The measurements employ the time-of-flight technique. Energy-dependent ratios of the two spectra are deduced from the measured values over the energy range 1.0 to 10.0 MeV. The experimentally-derived ratio results are compared with those calculated from ENDF/B-V, revision-2, and with results of recent microscopic measurements. Using the ENDF/B-V ^{235}U Watt parameters for the ^{235}U spectrum, the experimental measurements imply a ratio of average fission-spectrum energies of $^{239}Pu/^{235}U = 1.045 \pm 0.003$, compared to the value 1.046 calculated from ENDF/B-V, revision 2.

+Permanent address: Japan Atomic Energy Research Institute, Tokai-Ibaraki, Japan.

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I. INTRODUCTION

Precise knowledge of the prompt-fission-neutron spectrum of ^{239}Pu is important in fast-reactor neutronic studies as it is an essential property of the neutron-source term. Therefore, the process has been extensively investigated using a variety of experimental methods (1). A convenient index for the comparison of ^{239}Pu fission spectra is the average fission-neutron energy, $\bar{E}$; or the ratio, R, of $\bar{E}$ to that of ^{235}U assumed as a standard, where

$$R = \bar{E}\,(^{239}Pu) \;/\; \bar{E}\,(^{235}U). \tag{1}$$

The ratios R obtained from the older microscopic measurements scatter about the value 1.08 compared to 1.03 - 1.04 suggested from integral measurements (2). More recent microscopic measurements, using fission-inducing neutron energies of either thermal or ≈ 0.55 MeV, generally tend toward R values close to those deduced from integral studies (3,4,5). Sophisticated methods have been developed for the calculation of prompt-fission-neutron spectra (6) and these have been used to calculate the spectrum given in the ^{239}Pu ENDF/B-V, revision-2, evaluation (7).

Recent integral measurements at Lawrence Livermore National Laboratory using ^{239}Pu and ^{235}U spheres having radii of 3.22 cm and 3.145 cm, respectively, provide a new and very detailed integral basis for the comparative assessment of evaluated ^{239}Pu and ^{235}U prompt-fission neutron spectra, as well as of integral calculational methods (8). The measurements involved incident, fission-inducing, neutron energies of 0.3 - 2.7 MeV and fission-neutron-emission energies of 3.0 - 10.0 MeV. The interpretation of these integral measurements has raised some questions and their quality permits new precision in integral interpretation. Therefore the present microscopic measurements were initiated in an effort to test the results of the integral measurements and to provide an improved basis for their interpretation. This study is a direct measurement of the ratio, R, of the ^{239}Pu to ^{235}U fission-neutron spectra resulting from fission induced by ≈ 0.55 MeV incident-energy neutrons, over a spectrum-energy range of 1.0 - 10.0 MeV. The experimental results are compared with those calculated from ENDF/B-V (revision-2) and with the results of other microscopic measurements. The ratio determination does not require any knowledge of absolute detector efficiency, and the experimental results require no assumption as to the shape of the ^{239}Pu fission-spectrum.

II. EXPERIMENTAL METHODS

The prompt-fission-neutron spectra were measured using a 10-angle time-of-flight system (9) long employed in fast-neutron spectral measurements at Argonne. A pulsed-proton beam with a burst duration of approximately 1 nsec and a repetition rate of 2 MHz was incident on a metallic lithium target. The proton energy and the lithium target thickness were adjusted so that neutrons produced by the $^{7}Li(p,n)^{7}Be$ reaction (10) had an energy of 0.55 ± 0.05 MeV at the zero-degree reaction angle. The uranium and plutonium samples were both solid metal cylinders of identical size, 2 cm in diameter and 2 cm long. The uranium sample was enriched to 94.6% in ^{235}U, with the remainder distributed among the other uranium isotopes. The plutonium sample was 95.5% ^{239}Pu, and the remainder ^{240}Pu. Chemical impurities in the samples were negligible. In these experiments we ignore the approximately 5% abundance of the minor isotopes in either sample. This is a valid assumption as the minor isotopes have negligible fission cross sections at the incident-neutron energies of the present experiment, and their abundance is small and essentially equal in each sample. The uranium sample was not clad and the plutonium sample was enclosed in a 0.1 mm thick welded-steel can. Throughout the measurements an identical empty steel can was used for background determinations. The samples were placed at a zero-degree reaction angle, 15 cm from the neutron source, at the focus of the ten flight paths. Neutrons emitted from the samples, and proceeding down the flight paths, were detected by NE213 liquid scintillators 12.7 cm in diameter and approximately 2 cm thick. The low-energy-sensitivity cutoff of these detectors was set at approximately 800 keV so that the maximum energy range of detection was obtained while at the same time certainly avoiding neutrons elastically scattered from the samples. Pulse-shape-sensitive circuitry discriminated against unwanted gamma-ray backgrounds. The flight paths varied in length from 4.976 - 5.077 m and were measured to ± .5 cm, including the mean detector thickness. The detectors were approximately uniformly distributed over the angular range 25 - 150 degrees. An eleventh and independent time-of-flight channel and detector, arranged to directly observe the source, served as a monitor and also controlled dead-time effects. The monitor response was reasonably consistent with the integrated proton-beam current, given the inevitable variations in the lithium target.

The eleven measured flight times between the neutron burst and the neutron detection were multiplexed into a computer and sorted into spectra of 500 channels, each with a channel width of approximately 1 nsec. The time scale was determined using a set of calibrated delay lines, and the time zero established by the observation of the prompt-fission gamma-ray with the detector gamma-suppression circuitry turned off. Conversion of time-to-energy scale throughout the measurements was relativistically. The experiments were carried out in cyclic sets, rotating uranium, no-sample, plutonium and empty-can measurements for approximately an hour each through many cycles. The results obtained for the separate cycles were consistent. A set was continued until there were at least a 3×10^3 events per channel at the spectrum maximum, and 2×10^2 at a channel corresponding to approximately 10 MeV neutrons. This procedure properly subtracted the ambient room background and the background due to the neutron source. However, there is a small additional background due to the presence of a fissle sample. Tests showed that this was essentially time uncorrelated. Therefore it was subtracted by linearly extrapolating from low energies, well below the detector bias point, to regions corresponding to neutron energies of 15 MeV or above (e.g., extrapolations between regions where there are no significant fission-spectrum contributions). This extrapolation also would tended to correct for non-linear dead-time effects (11) but these were negligible in the present experiments. The background treatment is further described in the Appendix.

III. ANALYSIS

First the measured time spectra were integrated from 1 to 10 MeV and then normalized to make that integral unity. The normalized spectrum of ^{239}Pu was divided by that of ^{235}U channel-by-channel to obtain

$$R_j = \frac{F_{1j} - B_{1j}}{F_{2j} - B_{2j}} \times \frac{I_2}{I_1} , \qquad (2)$$

where j is the channel range of the spectrum extending from 1 to 10 MeV, F_{kj} and B_{kj} are respectively foreground and background in channel j, and

$$I_k = \sum_{j=min}^{max} (F_{kj} - B_{kj}) \ . \qquad (3)$$

If correlations between uncertainties in F_{kj}, B_{kj} and I_k are negligible, the uncertainty in R_j is

$$\delta R_j = R_j \left\{ \frac{\delta_{1j}^{\ 2} + \delta B_{1j}^{\ 2}}{(F_{1j} - B_{1j})^2} + \frac{\delta F_{2j}^{\ 2} + \delta B_{2j}^{\ 2}}{(F_{2j} - B_{2j})^2} + \frac{\delta I_1^{\ 2}}{I_1^{\ 2}} + \frac{\delta I_2^{\ 2}}{I_2^{\ 2}} \right\}^{1/2} \qquad (4)$$

where δXs represent the uncertainties in quantities X. The resulting R_j values have relatively large uncertainties near the 1 and 10 MeV energy extremes due to low counting statistics. The energy dependence of R was described by a power-series expansion of the form

$$R(E) = \sum_{n=0}^{m} a_n E^n \; . \tag{5}$$

Least-square fits were carried out with m increasing from 1 to 3 (i.e., linear to cubic fits). The cubic fit (m = 3) was abandoned as nonsignificant since the uncertainty in the a_3 coefficient exceeded its value for the results obtained with each of the ten detectors. An example of both linear and quadratic fits to R(E) obtained for one of the detectors is shown in Fig. 1. The quadratic fit gave a better description of the measured values below approximately 5 MeV, an important region for the calculation of the average fission-spectrum energy. However, it systematically deviated from the measured values above about 6 MeV. Typical chi-square values for both orders of fit were in the range 1.1 - 1.3. The final expression for R(E) was the weighted average of the parameters a_0, a_1 and a_2 from the R(E) least-square fits to results from the ten independent detectors. The resulting values, their standard deviations and their correlations are shown in Table 1. With these parameters, assuming the Watt ^{235}U fission-neutron spectrum (ENDF/B-V, revision-2), the average energy of the ^{239}Pu fission neutron spectrum over the 1-10 MeV energy range can be calculated as discussed in the following section.

Multiple-scattering of the fission neutrons in the fissile samples softens the observed fission spectra. However, in the present measurements the effect is small due to the small size of the samples, and the very similar inelastic-scattering cross sections, leading to appreciable energy transfer, of ^{235}U and ^{239}Pu (12). These similar and small effects are essentially completely cancelled in the ratio interpretation used in this study. This conclusion is supported by Monte-Carlo calculations carried out in a similar context (12).

IV. RESULTS

In Fig. 2 the ratio of the ^{239}Pu fission-neutron spectrum relative to that of ^{235}U is compared with the values obtained from ENDF/B-V (revision-2). Generally, the agreement is good, though the experimentally derived values imply a harder ^{239}Pu spectrum. The average energy between E-min and E-max can be calculated from

$$\bar{E} = \int_{E_{min}}^{E_{max}} N(E)\, E\, dE \Big/ \int_{E_{min}}^{E_{max}} N(E)\, dE, \qquad (6)$$

where N(E) is the fission-neutron spectrum, with an uncertainty in the quadratic case of

$$\delta\bar{E} = G^{+}MG \qquad (7)$$

where $G^{+} = \left(\frac{d\bar{E}}{da_0}, \frac{d\bar{E}}{da_1}, \frac{d\bar{E}}{da_2}\right)^{+}$, and M = the covariance matrixes of a_0, a_1 and a_2, and where

$$\begin{aligned} M_{ii} &= (\delta a_i)^2, && i = 0,1,2 \\ M_{ij} &= \delta a_i \delta a_j && Corr(a_i, a_j) \end{aligned} \qquad (8)$$

with $i = 1,3$
$j = 1,3$
and $i \neq j$.

The respective values are given in Table 1. When N(E) for ^{235}U is assumed (e.g. from ENDF/B-V which has the Watt form), then N(E) for ^{239}Pu follows from the experimentally-derived parameters and the N(E) of ^{235}U. The resulting $\bar{E}$ values for the experimental energy range of 1 - 10 MeV are given in Table 2, together with the comparable quantities derived from ENDF/B-V (revision-2). The table also gives the same values derived by extrapolating over the energy range 0 to 20 MeV, as well as some values deduced from recent experimental results. The agreement with the results of the present measurements and the evaluation is very good.

V. CONCLUSIONS

The energy dependence of the prompt-fission-neutron spectrum of ^{239}Pu

(induced by ≈ 550 keV incident neutrons), relative to that of ^{235}U, was determined over the emitted-neutron energy range of 1 to 10 MeV with uncertainties of less than 4%. The energy dependence of this ratio is reasonably described by either linear or quadratic energy dependences. With either description, assuming the ENDF/B-V ^{235}U fission spectrum, the average fission-neutron energy of ^{239}Pu is defined to 0.15% accuracy. Our result is in excellent agreement with that predicted from ENDF/B-V (revision-2) and with values deduced from recent measurements. The agreement with ENDF/B-V is marginally better when the experimentally deduced results are extrapolated over the full 0 - 20 MeV energy range. There are small differences between results calculated with ENDF/B revision-1 and revision-2. The present experimental results tend to support the latter. The recent integral measurements (8) span a somewhat different and wider incident-neutron energy range (0.3 - 2.7 MeV) than the present work and imply a slightly harder fission-neutron spectrum of ^{239}Pu (relative to that of ^{235}U) than indicated by the present work, or the ENDF/B-V evaluation, over approximately the same emission-energy range. The difference is beyond the respective uncertainties. We also examined the fission spectra of ^{239}Pu and ^{235}U given by ENDF/B-V (revision-2) for a number of values of the incident energy between 10^{-5} eV and 2.0 MeV and found that ^{239}Pu is always significantly harder than that of ^{235}U by similar amounts. Thus the variation in the incident energy in the integral experiments is probably not an important factor in their interpretation.

The ^{239}Pu average fission-neutron energy derived from the present work depends upon the assumed ^{235}U spectrum but the ratio values can be very useful for precise testing of relative fission-neutron energies. The present test suggests that the values given in ENDF/B (revision-2) are correct to remarkably high accuracy and suitable for the analysis of integral benchmark measurements.

Appendix

The linear backgrounds of the time-of-flight spectra were determined by least-square fitting the spectral regions before and after the fission-neutron spectra (i.e., below 0.6 MeV and above 15 MeV). The fitting function was

$$B(j) = c + d(j - J_0) \qquad \text{(A-1)}$$

where $J_0 = \frac{1}{M} \left(\sum_{M_1}^{M_2} j + \sum_{M_3}^{M_4} j \right)$, $M = M_2 - M_1 + M_4 - M_3 + 2$, and (M_1, M_2)

and (M_3, M_4) are the respective low- and high-energy regions selected for the background determination. The uncertainty in the background determination is given by

$$\delta B(j) = ((\delta c)^2 + (\delta d)^2 (j - J_0)^2)^{1/2} \quad \text{(A-2)}$$

and the correlation between δc and δd is proportional to

$$\sum_j W_j (j - J_0) \approx \bar{W} \sum_j (j - J_0) = 0 \quad \text{(A-3)}$$

when all weights, W_j, are not very different from their average, $\bar{W}$. The correlations calculated from the fitted results were always close to zero. In this case the uncertainties in the fitted parameters are simply expressed as

$$\delta c \approx (\sum_j W_j (j - J_0)^2)^{-1/2}$$

and (A-4)

$$\delta d \approx (\sum_j W_j)^{-1/2} \quad .$$

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Table 1. Linear and Quadratic Parameters Derived from Least-Square Fits to the Energy Dependence of the Ratio of $^{239}Pu/^{235}U$ Fission-Neutron Spectra.

Parameter	Linear ($a_0 + a_1E$)	Quadratic ($a_0 + a_1E + a_2E^2$)
a_0	0.902 ± 0.006	0.868 ± 0.001
a_1	0.035 ± 0.002	0.059 ± 0.008
a_2	-------------	-0.004 ± 0.001
Corr(a_0,a_1)	-0.913	-0.960
Corr(a_0,a_2)	-------------	0.875
Corr(a_1,a_2)	-------------	-0.965

Table 2. Average Energies of the Prompt-Fission-neutron Spectra of ^{235}U and ^{239}Pu.

Source	$\bar{E}(^{235}U)$a	$\bar{E}^{239}Pu$	Ratio
1 - 10 MeV Energy Range			
Present (lin. fit)	(2.652)b	2.727 ± .004	1.028
Present (quad. fit)	(2.652)b	2.715 ± .004	1.024
ENDF/B-V (rev. 2)	2.652	2.711	1.022
0 - 20 MeV Energy Range			
Present (lin. fit)	(2.031)b	2.123 ± .005	1.045
Present (quad. fit)	(2.031)b	2.119 ± .006	1.043
ENDF/B-V (rev. 2)	2.031	2.125	1.046
ENDF/B-V (rev. 1)	2.031	2.112	1.039
Smith et al. (2)[d]	1.973 ± .024	2.054 ± .029	1.041
Johansson et al. (3)	2.028 ± .030c	2.118	1.044
Integral Benchmark			
Haight et al. (8) approx. energy range = 3 - 10 MeV	------	-----	1.040 ± .005

a. Average energies in MeV.
b. ENDF/B-V values assumed.
c. As corrected by Adams (4) and fitted with a Watt form over the energy range 0.975 - 10.439 MeV.
d. Relative to ^{252}Cf.

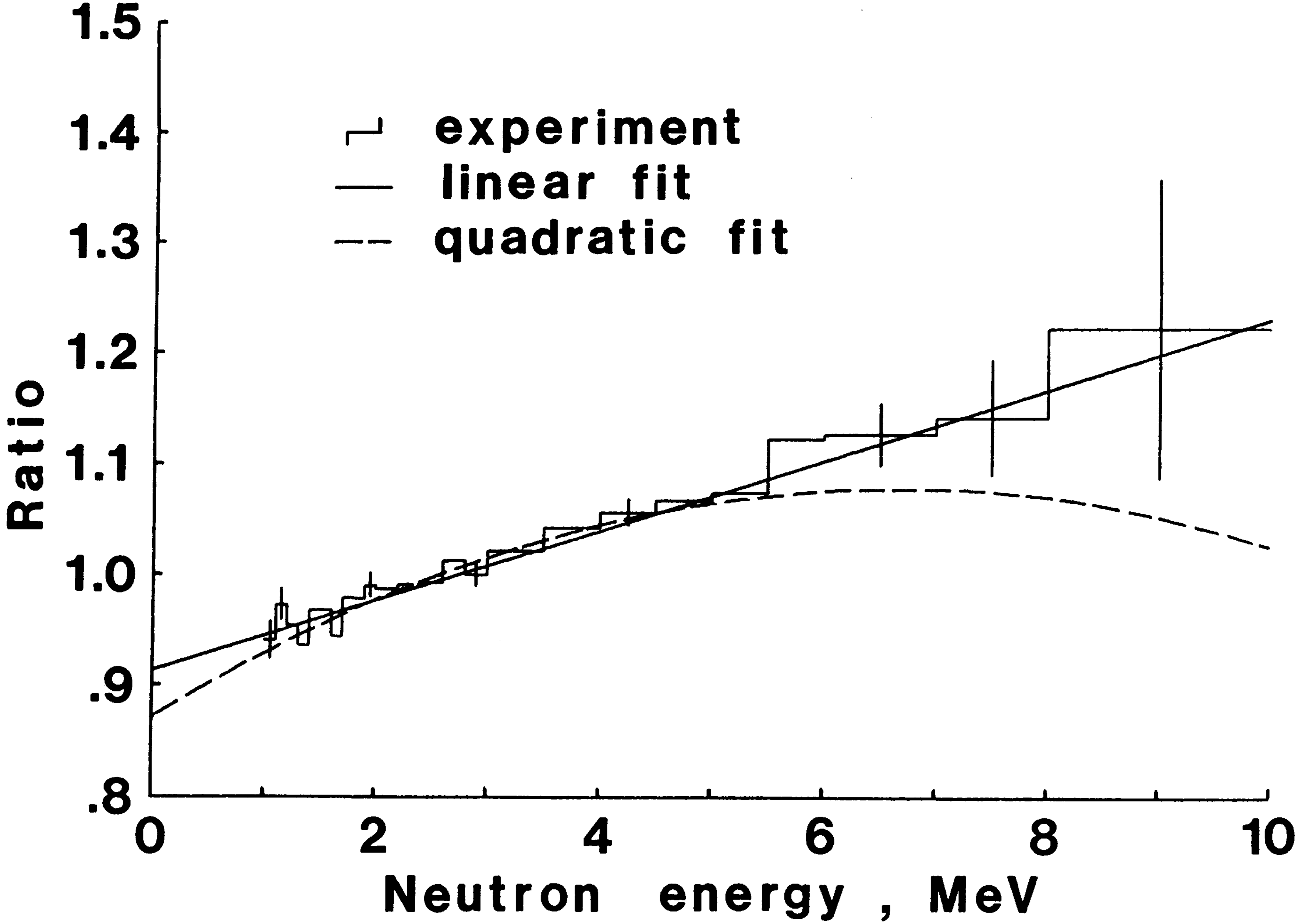

Figure 1. An example of linear and quadratic fits to measured spectral ratios. Measured values have been binned for clarity.

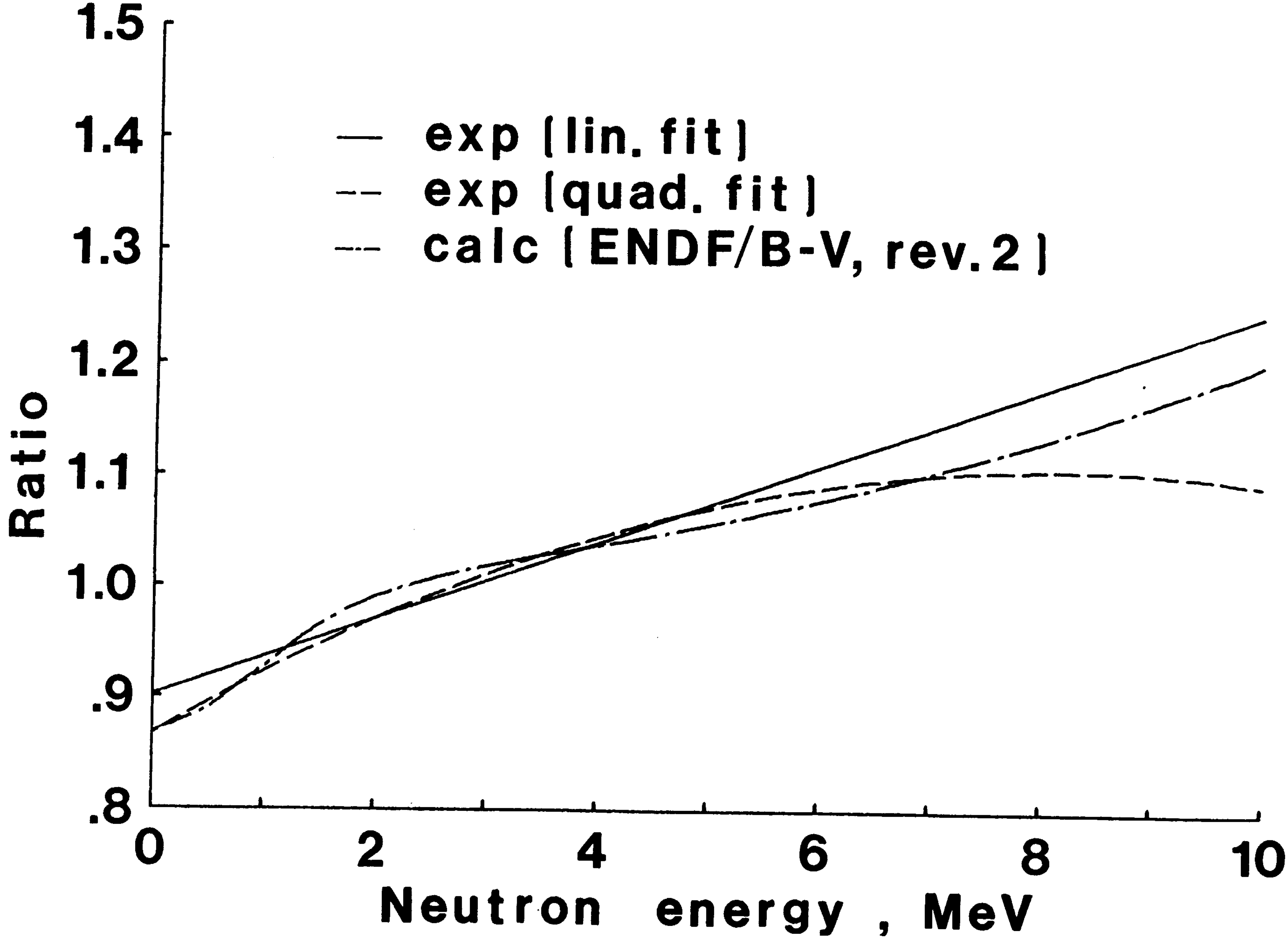

Figure 2. Comparison of measured and evaluated ratios of the $^{239}Pu/^{235}U$ prompt-fission-neutron spectra.