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A Measurement of the Fission Cross Section of Plutonium-241 Relative to Uranium-235

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ERRATA

In the chapter "Samples" on page 125 Table 1 shows erroneously old, preliminary mass values. These are now replaced by the correct, final values which were used in all cross section computations shown later in the paper. The description of the mass analysis is given in more detail, too.

The assay was carried out in Geel. The masses of the ^{235}U-samples were determined from low geometry alpha-counting of the layers. The specific alpha activity was known from isotope dilution mass spectrometry analysis[x] and an accurate alpha activity measurement[xx] of the base material. The uncertainty of 1 % contains 0.5 % from alpha counting, 0.2 % from the specific alpha activity and 0.3 % from possible losses during the transport. (In fact no indication for such losses were found in the runs with the two different samples). The ^{241}Pu-sample used in the experiment broke before analysis. Therefore no direct destructive analysis was possible. The masses were determined from a comparison with the alpha-activity of reference samples which were prepared simultaneously. These additional samples have been analyzed by a complicated scheme (repeated isotope and isotope dilution analysis, ^{241}Am separation, spiking, mixing and alpha-counting). Due to this difficult method an overall uncertainty of 1.8 % had to be given by CBNM[xxx].

The sample masses and the isotopic composition are listed in Table I. Quantitative measurements were carried out with the samples marked by crosses.

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x P.J. De Bièvre and G.H. Debus, Nucl. Instr. Meth. 32, 224 (1965).

xx Spernol et al., internal communication.

xxx K.F. Lauer, Del Bino and G.H. Debus, unpublished results.

TABLE I

^{235}U Sample Mass (mg)	^{241}Pu Sample Mass (mg)
2.315 ± 1.0 %	0.438 ± 1.8 %
+2.073 ± 1.0 %	+0.471 ± 1.8 %
+2.002 ± 1.0 %	0.468 ± 1.8 %
	0.513 ± 1.8 %
Isotopic Composition (at. %)	
^{234}U 0.169	^{239}Pu 1.83
^{235}U 99.499 ± 0.05	^{240}Pu 7.25
^{236}U 0.027	^{241}Pu 89.50 ± 0.08
^{238}U 0.353	^{242}Pu 1.42
	^{241}Am 6.94 ± 1.4

+Samples which were served for quantitative measurements.

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The fission cross-section ratio ^{241}Pu:^{235}U has been measured in the neutron energy range from 5 keV to 1.2 MeV with an accuracy between 3 and 4%. The neutron energy was determined by the time-of-flight method. Two identical gas scintillation chambers were used to detect the fission events. The present measurements were carried out at sufficiently fine energy intervals to show for the first time the detailed shape of the fission cross-section ratio. In contrast to the results of other authors, a distinct structure was found. A comparison with an evaluation by Davey of previous data shows a systematic difference in shape up to 10% between 30 and 400 keV. At higher energies the evaluation generally agrees with the new data except in the region between 900 and 1000 keV, where the values of this work indicate a significant dip in the cross-section ratio.

INTRODUCTION

The importance of accurate data for the design of fast reactors motivated this measurement of the fission cross-section ratio ^{241}Pu:^{235}U. In the neutron energy region between 10 keV and 1 MeV, only a few measurements existed previously. Therefore, it seemed worthwhile to establish a new data set with improved accuracy. The present work was performed with the experimental technique described in a previous paper on measurements of the fission cross sections of ^{239}Pu and ^{233}U relative to ^{235}U.[1] Therefore, only a general survey of the technique is given together with the modifications required by the high beta activity of ^{241}Pu.

EXPERIMENTAL METHOD

Two identical gas scintillation chambers containing the samples of ^{241}Pu and ^{235}U were exposed to the neutron flux in symmetric positions. Continually flowing argon was used as scintillator gas. The chambers were divided by the opaque samples into two optically decoupled halves. From every fission event both fragments could be detected because the backings of the samples were so thin that the fragments could penetrate them. A coincidence requirement between the halves of each chamber provided good discrimination against alpha background. In the case of low alpha activity, as for ^{241}Pu and ^{235}U, it is possible to set the thresholds as low as the pulse height caused by alpha particles. The coincidence width was 40 nsec.

The neutrons were produced via the ^{7}Li$(p,n)^{7}$Be reaction with the Karlsruhe 3-MV-pulsed van de Graaff accelerator. The pulse width of the accelerator was 1 nsec and the repetition frequency 10^6 Hz. The time-of-flight technique served to discriminate against background and to determine the neutron energy. The scintillation chambers had a time resolution of 3.3 nsec, and with a flight path of 37.7 cm, the time resolution of 9 nsec/m was accurate enough for the energy determination below 300 keV. At higher energies a ^{6}Li-glass detector 1.82 m from the target and with 1.4 nsec/

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[1] E. PFLETSCHINGER and F. KÄPPELER, *Nucl. Sci. Eng.*, **40**, 375 (1970).

m time resolution was used. The thickness of the metallic ^{7}Li targets varied between 20 and 90 keV for points between 140 keV and 1.2 MeV. Lithium-7 targets of more than 130-keV thickness were used from 280 down to 10 keV. Thus a broad overlap exists between data obtained with thin and thick ^{7}Li targets; this confirms that there is no systematic difference in the results of the two types of measurements.

The zero point of the time-of-flight spectra was calculated from the position of the gamma peak produced by the gamma flash of the accelerator. A second neutron group corresponding to the ^{7}Li$(p,n'\gamma)^{7}$Be reaction could be distinguished from the main group in the time-of-flight spectra but did not distort the results.

The electronics were the same as described in Ref. 1. The only change was the elimination of the pulse height discrimination trigger parallel to the time determination trigger, because the coincidence between both halves of a chamber was sufficient for discrimination against alpha background.

The activity of the ^{241}Pu sample was about 70 mCi. Because of the very thin backing we could not risk damage of the sample by interchanging it between the scintillation chambers by means of the removable transport chamber. Therefore, it was not possible to eliminate remaining asymmetries in the neutron flux and the electronic thresholds by a measurement with interchanged samples. A very careful adjustment relative to the ^{7}Li target and the direction of the incoming beam defined by a system of slits and diaphragms was made to arrange the chambers in symmetric positions. The symmetry could be checked with good accuracy by comparing the time-of-flight spectra of both chambers at the relatively low neutron energy of 80 keV. No differences between the flight times to the two chambers (~100 nsec) could be observed. If there was any difference, it was less than 0.4 nsec, corresponding to an uncertainty in the neutron flux of 0.8%. Moreover, in a test run before the ^{241}Pu measurement, the ratio ^{235}U:^{235}U was determined to be 1.0 within the statistical error of 1.5%, including all other uncertainties, e.g., those from mass determination and electronic thresholds.

Asymmetries in the electronic thresholds were determined as indicated in Fig. 1. With the moderated neutrons of a ^{241}Am-Be source, pulse height spectra were obtained from both sides of each sample. An extrapolation to zero pulse height gives the undetected fraction of fission events. Because of the low thresholds the resulting correction is comparably small. The deviations of the pulse height spectra from one another and from the real energy distribution of fission fragments is due to the characteristics of the photomultipliers and to the chamber geometry. For the accuracy of the results, these effects do not have any importance.

SAMPLES

The samples were prepared at the CBNM/EURATOM laboratory in Geel, Belgium. On a 90-μg/cm^2-thick Vyns[2] backing metallized with 30 μg/cm^2 of aluminum, a uranium or plutonium acetate layer was deposited by electrospraying as described by Verdingh and Lauer.[3]

The mass determination was carried out in Geel by destructive analysis, using the isotopic dilution method described by de Bièvre.[4] The uncertainty of the mass analysis was 1.0% for the ^{235}U samples and 1.8% for the ^{241}Pu samples. The isotopic composition and the sample masses are listed in Table I. The quantitative measurements were carried out with the samples marked by crosses.

TABLE I

Mass and Isotopic Composition of ^{235}U and ^{241}Pu Samples

Mass (mg)	
^{235}U Samples	^{241}Pu Samples
2.345 ± 1.0% 2.099 ± 1.0%[a] 2.027 ± 1.0%[a]	0.522 ± 1.8% 0.562 ± 1.8%[a] 0.559 ± 1.8% 0.612 ± 1.8%

Isotopic Composition (at.%)			
^{235}U Samples		^{241}Pu Samples	
^{234}U	0.169	^{239}Pu	1.83
^{235}U	99.45 ± 0.05%	^{240}Pu	7.25
^{236}U	0.027	^{241}Pu	89.50 ± 0.08%
^{238}U	0.353	^{242}Pu	1.42
		^{241}Am	6.94 ± 1.4%

[a]Samples which served for quantitative measurements.

[2]Vyns consists of 85% polyvinyl chloride and 15% polyvinyl acetate. It was delivered by Union Carbide Europe, Brussels.

[3]V. VERDINGH and K. F. LAUER, *Nucl. Instr. Methods*, **21**, 161 (1963).

[4]P. J. de BIÈVRE and G. H. DEBUS, *Nucl. Instr. Methods*, **32**, 224 (1965).

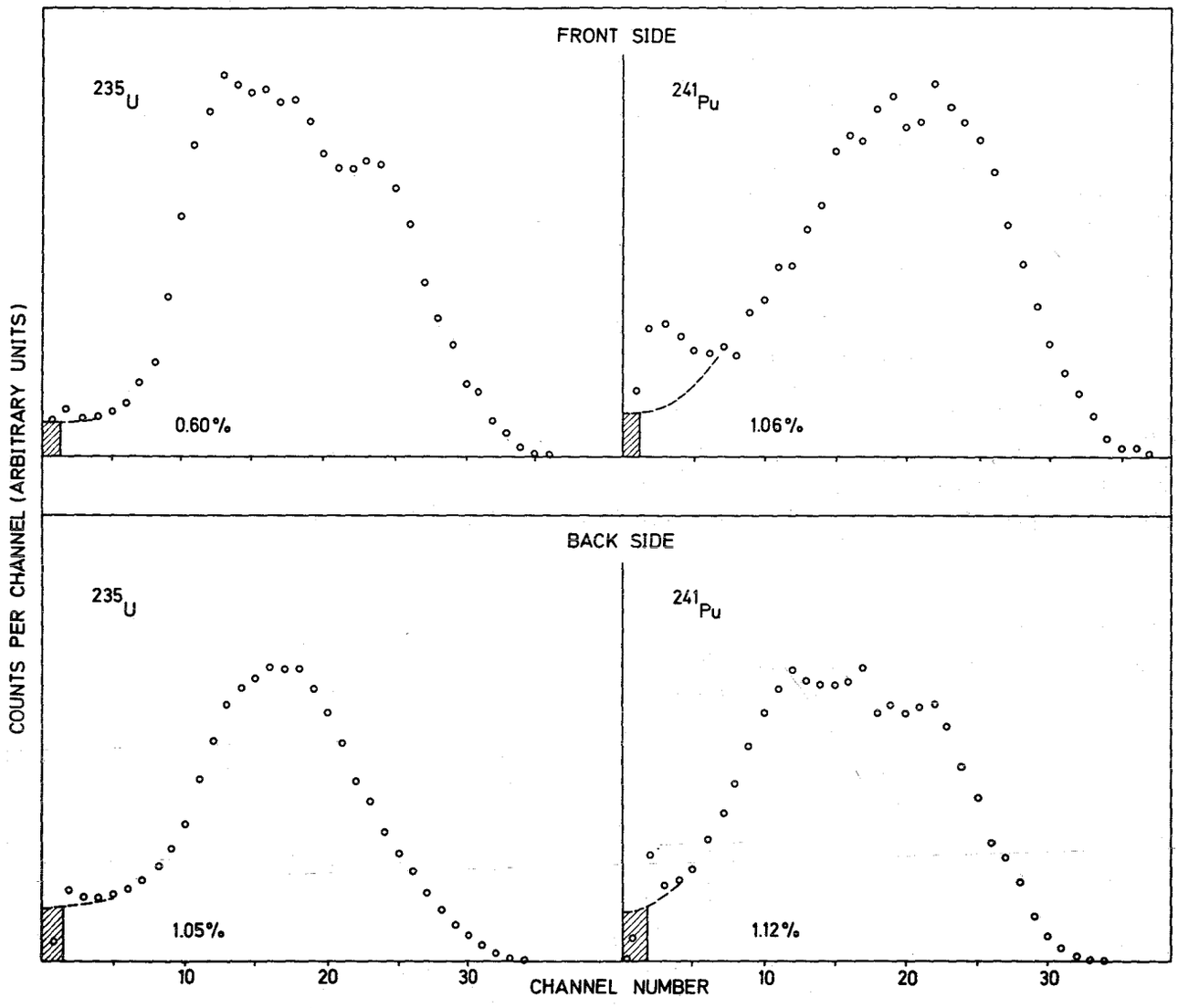

Fig. 1. Pulse height distributions. The dashed areas correspond to the extrapolated fractions.

CORRECTIONS AND UNCERTAINTIES

The corrections and uncertainties which result from this experiment are described in detail in Ref. 1. Therefore, only the characteristic values for this measurement are given here.

Background

The time-independent background was found to be about 2% for ^{241}Pu and 0.6% for ^{235}U. The resulting uncertainty is included in the statistical error and is always smaller than 0.9% except at energies below 20 keV. The time-correlated background, measured by withdrawing the fissile samples from the detectors into the transport chambers, was observed in the ^{235}U detector only. It does not exceed 2%, thus introducing an uncertainty of ~0.4%.

Finite Foil and Backing Thickness

The energy loss in the fissile layer and the backing increases with increasing angle between the direction of the fission fragments and the incident neutron beam. Therefore, not all fission events could be detected. Following the calculation of Rossi and Staub,[5] the fraction of fragments which cannot escape from the samples was determined. The values necessary for this calculation

[5] B. B. ROSSI and H. H. STAUB, *Ionization Chambers and Counters*, p. 227, McGraw-Hill Book Company, Inc., New York (1949).

are the thickness of the various layers of the sample (uranium acetate, aluminum, and Vyns) and the range and energy loss of fission fragments in these materials.[6-12] The result was averaged between light and heavy fragments and shows that 5.1% of the ^{241}Pu and 6.2% of the ^{235}U fission fragments were absorbed in the samples. The main error sources of this correction are thickness uncertainties of the aluminum (20%) and Vyns layers (20%). The Vyns backings were made by a standard method but not at the same time. Therefore, these uncertainties are taken into account. Because they influence the calculation in the same way for both samples, uncertainties in the fragment ranges cancel when the cross-section ratio is formed. The absorption correction is $0.989 \pm 1.5\%$.

The electronic threshold correction, determined by extrapolating the pulse height distributions from both sides of the samples to zero pulse height (see Fig. 1), was found to be $1.005 \pm 0.2\%$.

Electronic Corrections

Several times during the experiment the electronics were checked with a pulse generator to

TABLE II

Constant Uncertainties

Source of Uncertainty	$\sigma_f^{241}Pu/\sigma_f^{235}U$	
Correction for (n,γ) background	^{235}U	0.4%
Absorption losses in the samples	^{235}U ^{241}Pu	1.2% 1.0%
Electronic threshold correction	^{235}U ^{241}Pu	0.1% 0.2%
Sample mass	^{235}U ^{241}Pu	1.0% 1.8%
Asymmetry in neutron flux		0.8%

[6]F. NASYROV and S. V. LINEV, *At. Energ.*, **20**, 464 (1966).

[7]R. W. LAMPHERE, in *Fast Neutron Physics I*, p. 456, J. B. MARION and J. L. FOWLER, Eds., Interscience Publishers, New York (1960).

[8]G. KAHN and V. VORGUE, *Nucl. Sci. Eng.*, **23**, 8 (1965).

[9]J. B. CUMMING and V. P. CRESPO, *Phys. Rev.*, **161**, 287 (1967).

[10]E. K. HYDE, *The Nuclear Properties of the Heavy Elements III*, p. 195, Prentice-Hall, Englewood Cliffs, New Jersey (1964).

[11]J. NIDAY, *Phys. Rev.*, **121**, 1471 (1961).

[12]H. MÜNZEL, M. HOLLSTEIN, and T. ISHIMORI, in *Physics and Chemistry of Fission*, p. 573, International Atomic Energy Agency, Vienna (1965).

TABLE III

Fission Cross-Section Ratio $\frac{\sigma_f^{241}Pu}{\sigma_f^{235}U}$

Energy (keV)	$\frac{\sigma_f^{241}Pu}{\sigma_f^{235}U}$	Statistical Uncertainty[a] (%)
13.7	1.252	3.0
15.7	1.277	2.7
17.6	1.318	2.5
19.5	1.205	2.4
21.5	1.306	2.3
23.9	1.222	1.8
26.8	1.343	1.7
29.7	1.258	1.6
32.5	1.243	1.7
35.4	1.241	1.6
38.9	1.279	1.5
42.7	1.290	1.4
46.7	1.280	1.3
51.7	1.241	1.3
56.9	1.238	1.3
63.2	1.225	1.2
69.8	1.305	1.3
77.4	1.314	1.3
84.6	1.306	1.4
93.6	1.312	1.2
103.8	1.338	1.2
113.5	1.345	1.3
125.2	1.373	1.4
137.3	1.353	1.5
151.1	1.337	1.6
166.9	1.360	1.7
192 ± 16	1.382	1.8
230 ± 19	1.360	1.4
273 ± 20	1.396	1.5
324 ± 29	1.380	1.9
388 ± 18	1.261	1.7
436 ± 22	1.257	1.6
486 ± 25	1.315	1.4
535 ± 32	1.305	0.9
584 ± 23	1.265	1.1
633 ± 29	1.320	1.5
678 ± 33	1.287	1.3
740 ± 31	1.353	1.8
790 ± 38	1.320	1.9
870 ± 32	1.320	1.7
945 ± 40	1.229	1.7
1035 ± 40	1.263	2.1
1133 ± 49	1.267	2.1

Note: The data on which these results are based are available on request from the data bank of the NEA Neutron Data Compilation Centre, 91 Gif-sur-Yvette, B.P. 9, France.

[a]The errors of the isotopic corrections are included in the statistical uncertainty. The overall constant uncertainty is 2.6%.

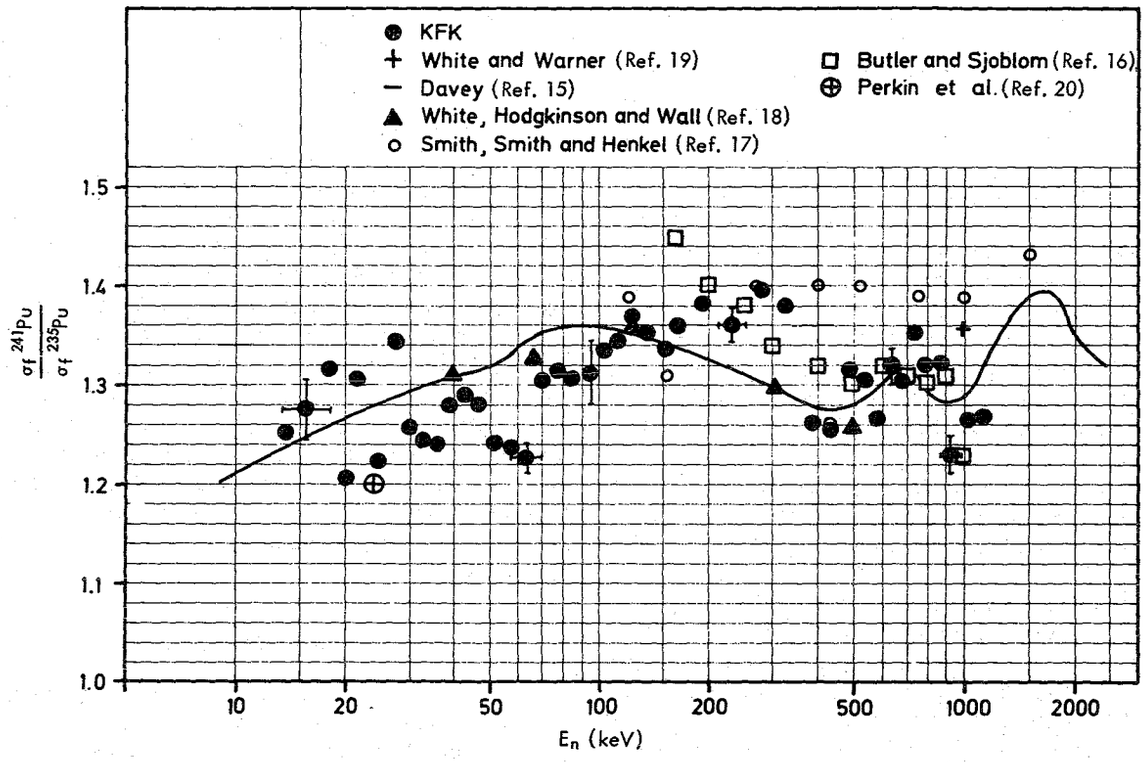

Fig. 2. Fission cross-section ratio ^{241}Pu:^{235}U. Full circles denote present work, while the solid line represents evaluated curve from older measurements (see Ref. 15).

confirm that no pulses were lost or incorrectly routed. The measurement at 535 keV was repeated at constant intervals to ensure that there was no systematic drift in the electronics. A dead time correction was necessary for neither ^{241}Pu nor ^{235}U.

Isotopic Composition

For ^{241}Pu a correction of the fission count rate has to be made because of fission events from the other plutonium isotopes and from ^{241}Am. For ^{241}Am the fission cross sections were taken from Seeger, Hemmendinger, and Diven[13] and from Bowman and Auchampaugh[14]; for the plutonium isotopes the evaluated data of Davey[15] were used. The correction for the plutonium isotopes is 1.2 ± 0.1% at 13 keV and increases to 8.9 ± 0.9% at 1.1 MeV. The correction for ^{241}Pu decreases from a value of 1.9 ± 0.5% at 13 keV to nearly zero between 50 and 500 keV; at higher energies the correction increases again up to 4.7 ± 1.2% at 1.1 MeV. The uncertainties of these corrections are included in the statistical error. All other constant uncertainties are listed in Table II.

DISCUSSION

The results of this measurement are listed in Table III. Only the statistical error is given for each energy point because it characterizes the uncertainty in the shape of the cross-section ratio. The total uncertainty of the absolute values can be obtained by adding the statistical uncertainty and the overall constant uncertainty, which is 2.6% according to Table II. The total uncer-

[13] P. A. SEEGER, A. HEMMENDINGER, and B. C. DIVEN, "Fission Cross Sections from Petrel," LA-3586, Los Alamos Scientific Laboratory (1966).

[14] C. D. BOWMAN and G. F. AUCHAMPAUGH, in *Nuclear Data for Reactors II*, p. 149, International Atomic Energy Agency, Vienna (1967).

[15] W. G. DAVEY, *Nucl. Sci. Eng.*, **32**, 35 (1968); also, *Nucl. Sci. Eng.*, **26**, 149 (1966).

[16] DANIEL K. BUTLER and RUTH K. SJOBLOM, *Phys. Rev.*, **124**, 1129 (1961).

[17] H. L. SMITH, R. K. SMITH, and R. L. HENKEL, *Phys. Rev.*, **125**, 1329 (1962).

[18] P. H. WHITE, J. G. HODGKINSON, and G. J. WALL, in *Physics and Chemistry of Fission*, p. 219, International Atomic Energy Agency, Vienna (1965).

[19] P. H. WHITE and G. P. WARNER, *J. Nucl. Energy*, **21**, 671 (1967).

[20] J. L. PERKIN, P. H. WHITE, P. FIELDHOUSE, E. J. AXTON, P. CROSS, and J. C. ROBERTSON, *J. Nucl. Energy*, **19**, 423 (1965).

tainty therefore lies between 3 and 4%. The neutron-energy spread at the points below 200 keV is about 10% of the neutron energy. In Fig. 2 our values are plotted together with those of several authors[16-20] and the evaluated curve of Davey.[15]

From Fig. 2 it can be seen that the older data do not show a very detailed structure. The values are either not close enough in energy to reproduce the structure (White, Hodgkinson, and Wall[18]), or they are averaged over a large energy interval (Butler and Sjoblom[16]; Smith, Smith, and Henkel[17]). By the values of this work, the shape of the fission cross-section ratio between 10 keV and 1.2 MeV is described in better detail.

A distinct structure in the cross-section ratio can be seen below 70 keV, which was reproduced by several runs. From a comparison of the shape of the ratio ^{241}Pu:^{235}U with that of the high-resolution fission cross section of ^{235}U (Refs. 21 and 22) (after smoothing to the same energy resolution as the present measurement), it can be seen that the structure in the ratio is mainly due to the fission cross section of ^{241}Pu. The same is valid for the dip between 300 and 400 keV. The decrease of the ratio between 900 and 1000 keV, however, is caused by an increase in the fission cross section of ^{235}U, as it is also present in the ratios ^{239}Pu:^{235}U and ^{233}U:^{235}U of Ref. 1. Very recent measurements of the fission cross section of ^{235}U (Ref. 23) also show a step of about 10% in this energy region.

As far as the absolute values are concerned, there is agreement within the given uncertainties between this work and the data of White, Hodgkinson, and Wall[18] and those of Butler and Sjoblom,[16] if one takes into account the broader energy intervals in the latter case. No agreement is found with the data of Smith, Smith, and Henkel,[17] either in the absolute values or in the shape of the cross-section ratio. When compared with the results of this work, the evaluated curve of Davey[15] is rather different in shape below 500 keV and shows a less pronounced structure at higher energies.

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[21] J. L. LEMLEY, G. A. KEYWORTH, and B. C. DIVEN, *Nucl. Sci. Eng.*, **43**, 281 (1971).

[22] C. D. BOWMAN, in *Neutron Standards and Flux Normalization*, p. 246, U.S. Atomic Energy Commission Symposium Series 23 (1971).

[23] F. KÄPPELER, to be published in *Nucl. Sci. Eng.*