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Capture-to-fission ratio of ^{235}U in the neutron energy range from 10 to 500 keV

H. Beer and F. Käppeler

Kernforschungszentrum Karlsruhe GmbH, Institut für Angewandte Kernphysik, 7500 Karlsruhe, Postfach 3640, Federal Republic of Germany

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The capture-to-fission ratio $\alpha = \sigma_c/\sigma_f$ of ^{235}U has been determined in the neutron energy range from 10 to 500 keV with an accuracy of typically 8 to 10%. The measurement was carried out on a 3 MV pulsed Van de Graaff accelerator using an 800 *l* liquid scintillator tank for the detection of capture and fission gamma rays. A fission neutron counter in coincidence with the tank served to distinguish between capture and fission events. With the good energy resolution of 2 nsec/m achieved in this experiment, intermediate structure in the capture-to-fission ratio could be resolved up to ~50 keV neutron energy. Interpreted in terms of the double humped fission barrier, the experimental average level spacing $D_{II} = 1000 \pm 300$ eV led to an energy difference between the first and second well of $E_{II} = 3.26 \pm 0.14$ MeV.

[NUCLEAR REACTIONS $^{235}U(n,f), (n,\gamma)$, $E = 10-500$ keV; measured $\sigma(n,\gamma)/\sigma(n,f)$, deduced average level spacing of class II states and energy difference between first and second well, enriched target.]

I. INTRODUCTION

^{235}U is one of the most studied fissile nuclei. But in spite of the effort spent to elucidate the fission process for this important nucleus, the knowledge of fundamental properties concerning the states in the second well of the double humped fission barrier is still unsatisfactory. A quantity which might yield information about the second well is the excitation function for neutron induced fission. Fluctuations in the eV and keV range ascribed to subthreshold fission of an only partially open fission channel are well known.[1-4] But until now it was not possible to extract reliable average resonance widths and spacings for states in the second minimum from the observed intermediate structures. This is due to the influence of the reaction entrance channel. Its fluctuations are superimposed on the intermediate structure in the fission cross section. The measurement of the capture-to-fission ratio α of ^{235}U represents a direct way to avoid these "noise effects" as in the capture-to-fission ratio the influence of the reaction entrance channel is eliminated. The capture width is believed to vary smoothly with neutron energy and therefore any structure in the fission channel should show up intensified in α. Indications for such a behavior have already been reported[5,6] but without sufficient energy resolution to allow for a quantitative analysis.

In addition to its special importance for the interpretation of intermediate structures, the capture-to-fission ratio α of ^{235}U is one of the most important quantities for reactor design. However, in the fast neutron energy range the experimental uncertainities were still of the order of 10 to 15% at the time when the evaluation for KEDAK 3 was completed.[7] Since then, only three new measurements in the keV range were reported[5,8,9] which all show uncertainties of typically 8 to 12%. This is not sufficient to satisfy the demands of reactor physicists which are compiled in the World Request List for Nuclear Data.[10] There an accuracy of 5% is requested in the keV range.

In view of the importance of the capture-to-fission-ratio of ^{235}U, an experiment was carried out at the Karlsruhe 3 MV pulsed Van de Graaff accelerator with the aims of both improving the accuracy and achieving a better energy resolution in α. This would permit meeting the requirements for reactor physics and the investigation of intermediate structure as well. As the confidence in a measured quantity is largely a question of the agreement of independent measurements, we have employed a new experimental technique, as compared to earlier work.

II. EXPERIMENTAL METHOD

The capture-to-fission ratio has been measured in the neutron energy range from 10 to 500 keV using the time-of-flight (TOF) technique for neutron energy determination. A sketch of the experimental arrangement is shown in Fig. 1. Neutrons were produced by a pulsed proton beam via the $^7Li(p,n)$ reaction. Pulse repetition rates of 1 MHz and 500 kHz were chosen for the different energy regions in order to avoid overlap in the TOF spectra. The proton pulse width was $\lesssim 1$ nsec and the average beam current was 4 μA at 1 MHz. The Li target was surrounded by a shielding of boron loaded paraffin with a double tapered collimator insert.

An 800 l liquid scintillator tank served for detection of capture and fission events. The tank

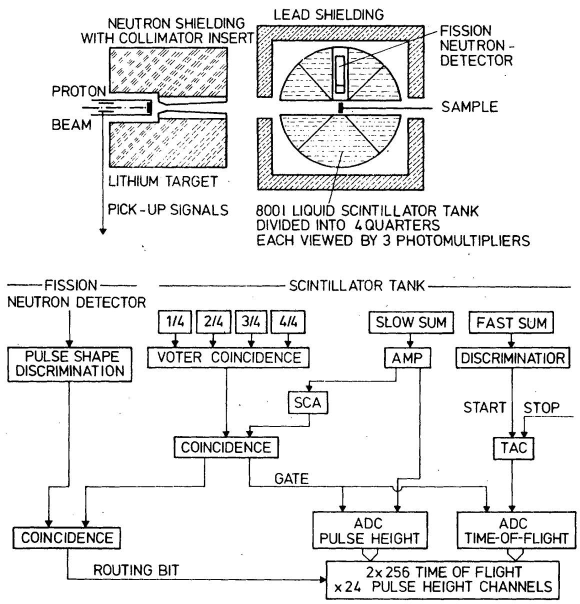

FIG. 1. Sketch of the experimental arrangement and block diagram of the electronics.

was divided into four optically decoupled quarters, each quarter being inspected by three large area photomultipliers (VALVO 60 AVP). Fast anode signals and slow signals from the 10th dynode were matched in time and pulse height and then added as indicated in the electronic block diagram of Fig. 1. The total sum of the fast signals was used for TOF determination and the sum of the slow signals for pulse height analysis. By a coincidence requirement between at least two of the four quarters, the time independent tank background could be reduced considerably. In spite of this coincidence an efficiency close to 100% could be maintained for capture and fission owing to the high multiplicity of the γ-ray cascades emitted in these events. The signals from the scintillator tank were accumulated in a two-dimensional matrix of 256 TOF vs 24 pulse height channels.

The scintillator tank alone allows only registration of sum of capture and fission events. In order to derive the capture-to-fission ratio, an additional detector is required for the identification of either capture or fission events. Differing from earlier measurements, the present technique made use of the prompt fission neutrons for the detection of fission events. In this way the signal-to-background ratio may be improved by using much thicker samples as compared to the techniques based on fission fragment detection.

The fission neutron detector was placed in a vertical tube in the center of the tank close to the ^{235}U sample. Fission neutrons were detected in a cylindrical liquid scintillator (NE 213), 5 cm in diameter and 5 cm thick, which was equipped with a pulse shape discriminator.[11] This fast and unequivocal identification of fission events helped also to keep the background at a much lower level as for instance in measurements with loaded scintillator tanks. Each signal from the fission neutron detector generated a routing bit for separate accumulation of a second two-dimensional matrix containing only fission events. From this matrix the shape of the pulse height distribution for fission events could be determined.

Finally the capture and fission component can be separated in the sum spectrum by normalizing the fission spectrum to the high energy part which contains no capture events.

III. MEASUREMENTS

The sample used in the actual measurements was a 0.15 mm thick metallic uranium disk with a diameter of 70 mm and a total mass of 10.25 g. The material was enriched to a ^{235}U abundance of 93.14%.

All measurements were performed at a neutron flight path of 2051 mm between the target and sample. The energy range from 10 to 500 keV was covered by five different runs with overlapping energy regions. Up to 300 keV continuous neutron spectra were produced by bombarding thick Li targets, whereas the two highest energies at 400 and 500 keV were measured with monoenergetic neutrons from 50 keV thick Li targets. The number of TOF channels was restricted to 256 by the available computer memory for data accumulation. Therefore, in runs I and II, which were performed with a repetition rate of 500 kHz, the energy resolution was limited by the time interval per channel of 8.7 nsec. In runs III to V the time per channel was 4.3 nsec, which is very close to the intrinsic time resolution of the scintillator tank, so that in these runs the full experimental time resolution could be exploited.

In Table I the various experimental parameters are summarized. Two examples for experimental TOF spectra are given in Fig. 2, demonstrating the signal-to-background ratio achieved.

IV. DATA ANALYSIS

In a first step integral pulse height spectra were generated for each run from both two-dimensional matrices containing the sum of fission plus capture events and fission events. The respective background was determined from the TOF region between the γ peak and the onset of the neutron induced events. As in the investigated energy range the fission γ-ray spectrum does not depend on

TABLE I. Experimental parameters.

^{235}U sample	metal disk 70 mm diameter 0.15 mm thickness total mass 10.25 g 93.14% ^{235}U, 6% ^{238}U, 0.66% ^{236}U, 0.2% ^{234}U		
Proton beam	pulse width ≤1 nsec repetition rate 500 kHz 1 MHz average current 2 μA 4 μA		
Resolution	flight path 2051 mm energy resolution 2 nsec/m 4 nsec/m	run III, IV, V run I, II	
Neutron energy regions	500 kHz	run I	10–200 keV
	8.7 nsec/channel	run II	100–300 keV
	1 MHz	run III	15–64 keV
	4.3 nsec/channel	run IV	379 ± 23 keV
		run V	482 ± 25 keV

neutron energy; the fission spectra of all runs were added together to improve statistics. The pulse height spectra determined in this way are given in Fig. 3.

The total γ-ray energy emitted after neutron capture $E_{\gamma C}^{\mathrm{TOT}}$ is the sum of neutron binding energy B_n and neutron kinetic energy E_n. For ^{235}U the neutron binding energy is 6.5 MeV so that $E_{\gamma C}^{\mathrm{TOT}}$ is always less than 7 MeV in our neutron energy range. The total γ-ray energy associated with fission events extends up to 14 MeV. This difference in the maximum γ energies offers the possibility of normalizing the shape of the integral fission γ spectrum to the total γ spectrum of each run at that high energy part of the spectrum.

The fact that only fission contributes to the high energy part of the pulse height spectrum measured with a large scintillator tank was used previously to determine the energy dependence of the capture-to-fission ratio.[12,13] In the present measurement an improved procedure was applied by which larger spectrum fractions can be used resulting in reduced statistical uncertainties. As is shown in Fig. 3 two pulse height windows—indicated by the shaded areas—were selected such that equal parts a_f were obtained in the fission γ spectrum (regions 1 and 2). In the thin sample

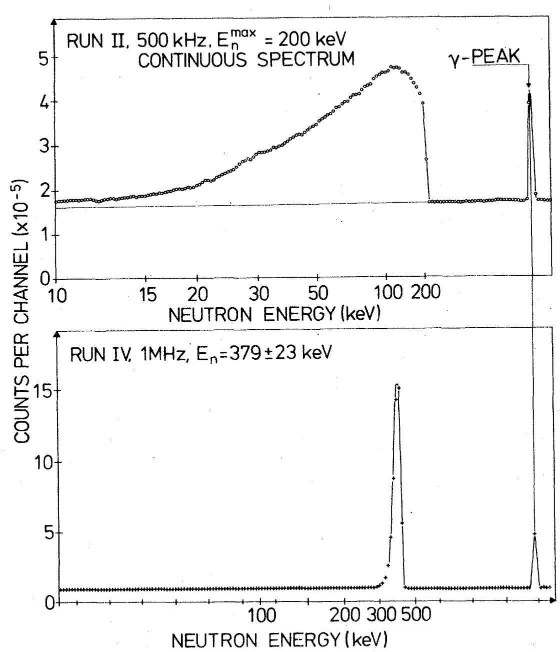

FIG. 2. Experimental TOF spectra measured in runs II and IV, demonstrating the signal-to-background ratio.

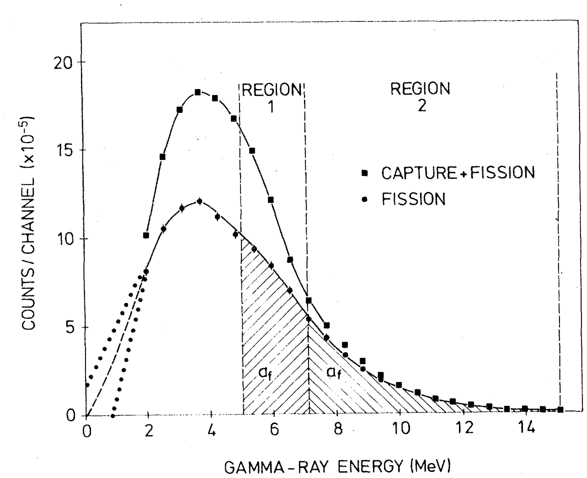

FIG. 3. Pulse height spectra of capture plus fission and fission events from run I. The shaded areas a_f indicate pulse height windows containing equal fractions of the fission γ-ray spectrum. The dotted lines give the estimated uncertainties in spectrum extrapolation below the electronic threshold at 2 MeV.

TABLE II. Summary of systematic uncertainties.

	Systematic uncertainty (%)
Spectrum extrapolation	5–6
Ratio of efficiencies	
ϵ_f/ϵ_c	3
Spectrum normalization	~3
Isotopic impurity	1
Total	~7

approximation the counting rates in regions 1 and 2 of the total γ spectrum for capture-plus-fission events can be expressed by

$$Z_{1,2}=\phi N(\epsilon_f q_f \sigma_f + \epsilon_c q_{c1,2}\sigma_c), \tag{1}$$

where ϕ is the neutron flux and N is the number of target atoms. σ_f and σ_c are the fission and capture cross sections, ϵ_f and ϵ_c denote the scintillator tank efficiency for fission and capture, and q_i stands for the spectrum fractions in regions 1 and 2. From the ratio Z_1/Z_2 one finds immediately

$$\alpha=\frac{\sigma_c}{\sigma_f}=\frac{\epsilon_f}{\epsilon_c}\,\frac{q_f}{Z_1(q_{c1}-q_{c2})/(Z_1-Z_2)-q_{c1}} \tag{2}$$

It was not necessary to construct the curves of Figs. 3 and 4 and make the extrapolations for each TOF channel because the shapes are essentially the same for all channels of one run.

The capture-to-fission ratio can now be calculated from the counting rates Z_1 and Z_2 if the remaining quantities in Eq. (2) are known. As can be seen from Eq. (2) the accuracy of α depends

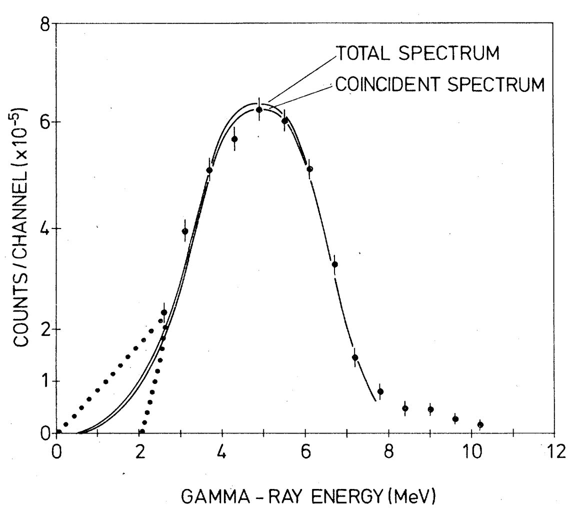

FIG. 4. Experimental (♦) and calculated pulse height spectra (solid line) for capture events from run I. The dotted lines again represent the estimated uncertainties in spectrum extrapolation.

strongly on the difference Z_1-Z_2. This has to be considered in the choice of the pulse height regions 1 and 2.

The ratio ϵ_f/ϵ_c has not been measured experimentally. It can be shown, however, that it is close to unity. The probability of detecting a capture or fission event by a coincidence between at least two of the quarters in the scintillator tank strongly depends on the multiplicity of this event. Very little experimental information is available about the γ-ray multiplicities of capture and fission events. Verbinsky *et al.*,[14] reported a mean total γ-ray energy $E_{\gamma f}^{TOT}$ of 6.51 MeV released in the fission of ^{235}U with an average energy $\overline{E}_\gamma$ of 0.97 MeV. This leads to average multiplicities >5 in the fission of ^{235}U. For neutron capture the measurements of Ottmar *et al.*[15,16] indicate multiplicities larger than 3 or 4 because only weak high energy γ transitions to low lying states were found for thermal neutron capture and at 2 keV neutron energy. Poenitz[17] and *v.* Egidy[18] developed models for the calculation of multiplicities. Although both authors give numerical values only for 150 $<A<200$, it might be plausible that the multiplicities of ~4 determined for this mass region can also be assumed for the actinides.

In addition to this information about γ multiplicities we have tried to get some hints from our experimental data. Similar to de Saussure *et al.*[19] we fitted our pulse height distributions with spectra calculated with the FORTRAN-IV code GAMOC[20] for various compositions about capture γ multiplicities. The result was the same as was found in Ref. [19]: A good fit to both the pulse height spectrum for coincident events and to the sum of all events required an average multiplicity of 5. The experimental spectrum which is the difference between the spectra in Fig. 3 is shown in Fig. 4 together with the calculated spectra. The almost identical shape of the coincidence and the total spectrum demonstrates that only a small systematic uncertainty had to be assigned because of the fact that data analysis was restricted to the coincident events. The result of our calculation was that the probability for detecting a capture event by a coincidence between at least two quarters is $\epsilon_c=97.5\pm1\%$. The corresponding number for fission events ϵ_f might be somewhat higher but as only the ratio is of importance in Eq. (2) we used a value $\epsilon_f/\epsilon_c=1.00\pm3\%$ as an approximation.

The total areas under the fission and γ pulse height spectra have to be known for the calculation of the spectrum fractions $q_i=q_i/A_i$. This required the extrapolation of the spectra from the electronic threshold at 1 MeV down to zero pulse height as is demonstrated in Figs. 3 and 4. The dashed lines give the most probable shape while the dotted lines are estimates for the possible systematic uncertainties. Knowing the total areas A_f and A_c for the pulse height spectra of each single run, one can immediately determine the spectrum fractions in regions 1 and 2. This is possible because the pulse height spectrum for capture events did not change noticeably within the limited energy range of an individual run.

Finally the counting rates Z_1 and Z_2 were integrated in the pulse height regions 1 and 2 for all TOF channels. With these quantities α could be calculated from Eq. (2) as a function of neutron energy.

V. CORRECTIONS AND UNCERTAINTIES

The corrections to the quantities in Eq. (2) and the uncertainties relevant to the accuracy of the results are discussed in the order of their importance.

The extrapolation in the pulse height spectra for fission and capture events caused the main contribution to the systematic uncertainty (Table II). As indicated in Figs. 3 and 4 by the dotted lines, an uncertainty of 5 to 6% had to be assigned to this procedure, dependent on the statistical accuracy of the respective run.

The correction related to small differences in the tank efficiency for fission and capture events was discussed in the preceding section. The ratio ϵ_f/ϵ_c was estimated to $1.00 \pm 3\%$.

The normalization of the integral fission pulse height spectrum to the total pulse height spectrum of each run was connected with an uncertainty of mainly statistical nature. It was between 2.7 and 2.9%.

The counting rates Z_1 and Z_2 had to be corrected for capture events in the isotopic impurity of the sample (6% ^{238}U. This correction is small, however, as for the determination of Z_1 and Z_2 only the high energy region of the pulse height spectrum was used. Owing to its small neutron binding energy of 4.8 MeV, the capture events in ^{238}U contributed only to region 1 with an associated correction of ~1%. A systematic uncertainty of also 1% was taken into account for this correction.

As the γ-ray spectra for fission and capture events are not significantly different (see Sec. IV) the absorption losses in the sample cancel in the evaluation of α. Anyhow, the sample was thin enough so that this effect was certainly less than 0.1%.

Multiple scattering and self-absorption corrections need also not be considered because they also cancel in the capture-to-fission ratio.

The effect of the threshold for fission neutrons of 1.3 MeV was neglected in data analysis. The total energy released in fission is composed of the kinetic energy of fission fragments E_{ff}, the energy associated with fission neutrons E_{fn}, and the total γ-ray energy $E_{f\gamma}$. It is characteristic of this partition that all three components reveal a marked energy variance[21] ~10 MeV for E_{ff}, ~8 MeV for E_{fn}, and ~2 MeV for $E_{f\gamma}$. This behavior ensures that the fission γ-ray spectrum recorded in coincidence with the detected fission neutrons is a representative average because the influence of the neutron threshold of 1.3 MeV is small compared to the energy fluctuations observed.

VI. RESULTS AND DISCUSSION

From the runs described in Sec. III five data sets were evaluated. For the continuous neutron spectra of runs I, II, and III the total uncertainty is governed by the statistics if the data are determined with the full energy resolution of 2 or 4 nsec/m. Therefore, the numerical values listed in Tables III to V are averaged over several TOF channels to achieve statistical uncertainties which are comparable to or smaller than the systematic uncertainties. A total uncertainty of typically 8% was achieved for most of the data points which is comparable to the accuracy of the best previous measurements. Numerical values of the high resolution data can be obtained from the NEA Neutron Data Compilation Centre.[22] We have not tried to establish a best set of α values from the results of Tables III to VI. This can be done by straightforward procedures. In this case the contribution of the statistical uncertainty is reduced by square root summation.

The present results as given in Tables III to VI are plotted in Figs. 5 and 6. These graphs clearly demonstrate that runs I and II as well as runs I and III yield consistent results in the respective overlap regions. This means that data processing was performed properly for all runs. Although the present data are averaged over energy intervals greater than those given by the experimental resolution, they still exhibit a better resolution than the previous results.

The comparison (Figs. 5 and 6) with the data of other authors shows excellent agreement with the values of de Saussure *et al.*[23] over the entire energy range. Most of the other measurements are consistent with the present results within their quoted uncertainties.[5,8,24,25,26] However, some discrepancies are observed with respect to the data of Bandl *et al.*,[6] who reported a gross structure in α between 10 and 40 keV which is not reproduced by this experiment. The recent results of Gwin

TABLE III. Numerical values of α from run I.

$E_n \pm \Delta E_n$ (keV)		$\alpha = \sigma_c/\sigma_f$	Uncertainty (%) Statistical	Systematic	Total
12.5	2.2	0.335	10.2		12.5
17.3	2.5	0.345	5.9		9.3
22.2	2.3	0.390	4.1		8.3
26.0	1.2	0.370	4.5		8.5
28.7	1.4	0.370	3.7		8.1
31.5	1.2	0.365	3.7		8.1
33.9	1.0	0.375	4.0		8.2
36.2	1.1	0.360	3.9		8.2
38.8	1.2	0.370	3.7		8.1
41.6	1.4	0.360	3.5		8.0
44.2	1.0	0.350	4.1	7.2	8.3
46.4	1.0	0.365	4.0		8.2
48.8	1.1	0.345	3.8		8.1
51.5	1.2	0.340	3.8		8.1
54.4	1.3	0.330	3.6		8.0
56.7	0.6	0.345	5.0		8.8
58.3	0.6	0.330	5.0		8.8
60.0	0.7	0.280	5.2		8.9
61.8	0.7	0.320	4.8		8.7
63.6	0.7	0.315	4.8		8.7
65.5	0.7	0.330	4.6		8.5
67.5	0.8	0.355	4.4		8.4
69.7	0.8	0.330	4.5		8.5
71.9	0.9	0.320	4.5		8.5
74.2	0.9	0.325	4.4		8.4
76.6	0.9	0.325	4.4		8.4
79.2	1.0	0.350	4.2		8.3
81.7	1.0	0.355	4.1		8.3
84.7	1.1	0.285	4.4		8.4
87.7	1.2	0.310	4.2		8.3
90.8	1.2	0.340	4.0		8.2
94.1	1.3	0.310	4.1		8.3
97.6	1.4	0.315	4.0		8.2
101.3	1.4	0.310	4.0		8.2
105.2	1.5	0.305	4.0		8.2
109.3	1.6	0.305	4.0		8.2
113.7	1.7	0.340	3.8	7.2	8.1
118.3	1.8	0.290	4.0		8.2
123.2	1.9	0.310	3.8		8.1
128.5	2.0	0.290	4.0		8.2
134.1	2.2	0.270	4.1		8.3
140.0	2.3	0.285	4.1		8.3
146.4	2.5	0.285	4.1		8.3
153.2	2.7	0.270	4.2		8.3
160.5	2.8	0.265	4.3		8.4
168.3	3.0	0.290	4.2		8.3
176.7	3.3	0.240	4.8		8.7
185.8	3.5	0.260	4.7		8.6
195.6	3.8	0.265	4.9		8.7

et al.[9] show a somewhat different shape compared to the data of this work. Slightly higher but below 20 keV, the data of these authors agree with the present values until 60 keV and then start to be systematically lower by up to 30%. The data of Weston *et al.*[27] show a similar but less pronounced behavior. They are in fair agreement with the present data below 100 keV but at higher energies they are also lower by ~10%.

As for a better survey the error bars of the previous data are omitted in Figs. 5 and 6, the accuracy achieved in the different measurements is

TABLE IV. Numerical values of α from run II.

$E_n \pm \Delta E_n$ (keV)		$\alpha = \sigma_c/\sigma_f$	Uncertainty (%) Statistical	Systematic	Total
110.8	8.1	0.345	11.2		13.1
125.6	8.6	0.255	10.9		12.8
142.1	8.6	0.305	9.0		11.3
156.5	8.2	0.225	9.6		11.8
172.8	9.5	0.255	7.5		10.1
188.4	8.4	0.245	7.6		10.2
203.7	9.5	0.230	6.9		9.7
218.1	7.5	0.235	7.4		10.0
230.5	8.1	0.215	7.0	6.8	9.6
240.5	5.2	0.225	8.9		11.2
247.5	5.4	0.225	8.5		10.9
254.9	5.7	0.245	7.8		10.3
262.6	5.9	0.245	7.6		10.2
270.7	6.2	0.215	7.9		10.4
279.1	6.5	0.230	7.4		10.0
288.0	6.8	0.230	7.5		10.1
297.3	7.2	0.220	7.7		10.3
307.0	7.5	0.215	8.0		10.5

given in Table VII for a representative energy around 30 keV.

The second aim of this experiment was to improve the energy resolution of the capture-to-fission ratio. In this regard difficulties emerged from the restricted number of TOF channels and from the limited statistical accuracy. In spite of these problems the present data plotted in Fig. 7 with full resolution reveal dramatic fluctuations in α as a function of neutron energy. For easier comparison with the fission cross section values of Perez *et al.*,[1] which are given below in Fig. 7, the fission-to-capture ratio $1/\alpha$ is shown. The anticorrelation between the size of the uncertainty and α follows immediately from Eq. (2) because the statistical uncertainty is strongly determined by (Z_1-Z_2). In the following section we try to analyze these strong fluctuations and to find a plausible interpretation.

TABLE V. Numerical values of α from run III.

$E_n \pm \Delta E_n$ (keV)		$\alpha = \sigma_c/\sigma_f$	Uncertainty (%) Statistical	Systematic	Total
18.8	2.1	0.350	8.8		11.3
23.5	2.6	0.355	5.6		9.0
28.4	2.6	0.380	3.9		8.1
33.4	2.6	0.370	3.6		8.0
38.4	2.6	0.375	3.7	7.1	8.0
43.4	2.8	0.395	3.4		7.9
48.7	2.7	0.355	3.7		8.0
53.4	2.4	0.330	4.3		8.3
58.0	2.7	0.310	5.2		8.8
62.7	2.6	0.340	8.3		11.0

VII. INTERMEDIATE STRUCTURE

It is apparent from Fig. 7 that the fluctuations in the fission-to-capture ratio are much more pronounced than in the high resolution fission cross section of Ref. 1. Peak-to-valley ratios of up to 3 can be observed in $1/\alpha$ while the respective ratios in σ_f are of the order of 0.5 only. In spite of this difference a correlation between the various data sets is clearly visible, at least for the structures at 14.7, 21.9, and 29.1 keV as is indicated by arrows.

The structure in the fission cross section has been confirmed by several measurements.[2-4] An explanation of intermediate structure in the fission cross section of ^{235}U has first been given by Cao *et al.*[28] in terms of the double humped fission barrier and the channel theory of fission. According to this picture the structure is associated with states in the second minimum of the fission barrier which all should have the same spin and parity. This concept was confirmed re-

TABLE VI. Numerical values of α from runs IV and V.

$E \pm \Delta E$ (keV)	$\alpha = \sigma_c/\sigma_f$	Uncertainty (%) Statistical	Systematic	Total
379 ± 23	0.182	2.0	7.1	7.4
481 ± 25	0.141	1.4	6.6	6.8

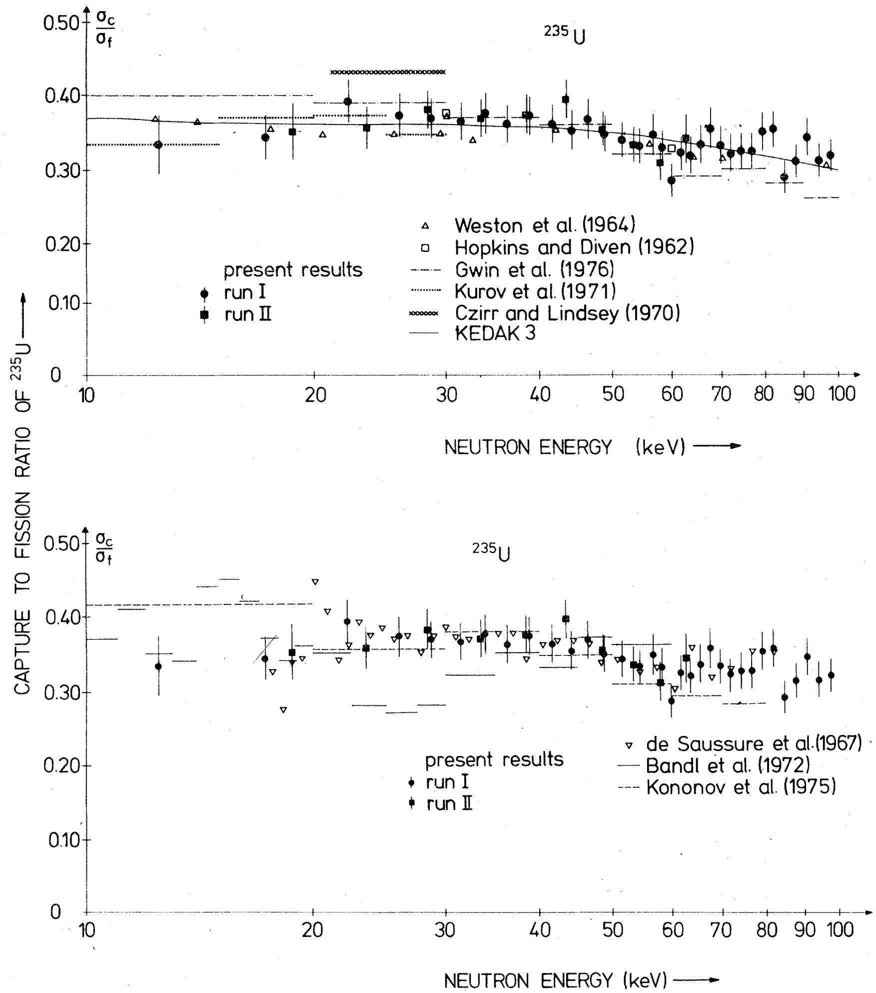

FIG. 5. Present results of the capture-to-fission ratio α of ^{235}U for neutron energies between 10 and 100 keV. For comparison, previous measurements and evaluated curves are also included.

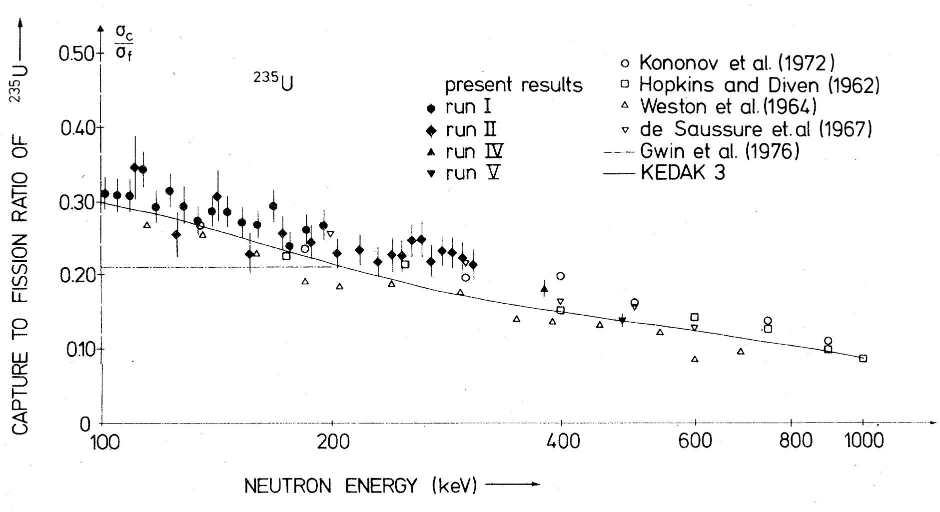

FIG. 6. Present results of the capture-to-fission ratio α of ^{235}U for neutron energies between 100 and 1000 keV. For comparison, previous measurements and evaluated curves are also included.

TABLE VII. Comparison of the accuracy achieved by the various measurements.

Authors, Ref.		Year	Total uncertainty (%) around 30 keV neutron energy
Hopkins and Diven	(24)	1962	9.6
Weston *et al.*	(27)	1964	9.0
de Saussure *et al.*	(23)	1967	8.6
Czirr and Lindsay	(26)	1970	11.6
Kurov *et al.*	(25)	1971	13.8
Bandl *et al.*	(6)	1972	12.5
Kononov *et al.*	(8)	1975	10.3
Gwin *et al.*	(9)	1976	7.3
Present work		1978	~8.0

cently by the fission cross section measurements of Keyworth *et al.*[29] with polarized neutrons and a polarized ^{235}U target which allowed distinguishing between fission channels with $J=3^-$ and $J=4^-$. It was found that the intermediate structure could be assigned to the partially open $J=4^-$ channel. Accordingly, it can be assumed that the level spacings of the structures corresponding to intermediate states should follow a Wigner distribution.

Several times efforts have been made to deduce widths and spacings of intermediate structures in the fission cross section of ^{235}U and to calculate with these quantities the energy difference E_{II} between the first and second well of the fission barrier. From an autocorrelation analysis between 6 eV and 3 keV Cao *et al.*[28] found an average level spacing $D_{II}=280$ eV leading to values of E_{II} between 2 and 3 MeV. A similar analysis was reported by Sood and Sarma,[30] who determined $D_{II}=250$ eV and $E_{II}=2.35$ MeV in the neutron energy range from 2 to 1500 eV. The approach of Perez *et al.*[31] with artificially generated fission cross sections yielded similar results. The difficulty with this kind of analysis was that strong statistical fluctuations of the reaction entrance channel are superimposed on the fission cross section. Migneco *et al.*[32] have therefore included both the total neutron cross section and the fission cross section, in their analysis. These authors also find evidence of intermediate structures in the fission cross section of ^{235}U between 6 and 20 keV with a width smaller than 1.5 keV.

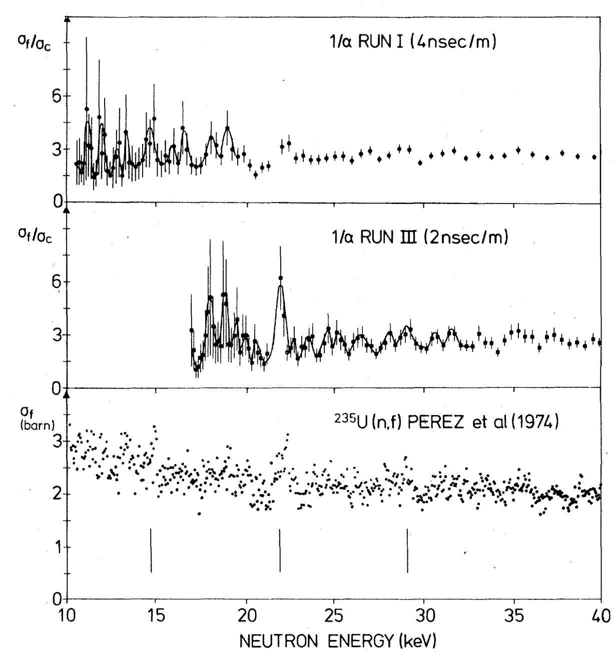

FIG. 7. Resonance structure in the present results of the fission-to-capture ratio $1/\alpha$ of ^{235}U plotted with full experimental resolution. There is some evidence for correlation to intermediate structure in the fission cross section shown in the lower part at 14.6, 21.9, and 29.0 keV.

As already mentioned, a more direct way to eliminate the influence of the entrance channel is offered by the investigation of the fission-to-capture ratio where the effects of the entrance channel cancel out. Fluctuations in the capture widths are normally small enough[33] so that the structure in $1/\alpha$ can be attributed to the fission channel.

In spite of the large statistical uncertainties it was possible to analyze the present results with respect to widths and level spacings of the intermediate structures in $1/\alpha$. First, several statistical tests[34] confirmed the high significance of these structures. The probability of a random nature was found to be less than 0.1%. In addition, the influence of Porter-Thomas fluctuations from the fission and neutron channels on $1/\alpha$ was investigated. Using average spacing and width parameters reported by Keyworth *et al.*[29] levels and widths were generated from Wigner and Porter-Thomas distributions for the given experimental energy bins. Then $1/\alpha$ was computed for each experimental data point and the resulting curve for $1/\alpha$ is shown in the upper part of Fig. 8. On a 95% confidence level the fluctuations vis-

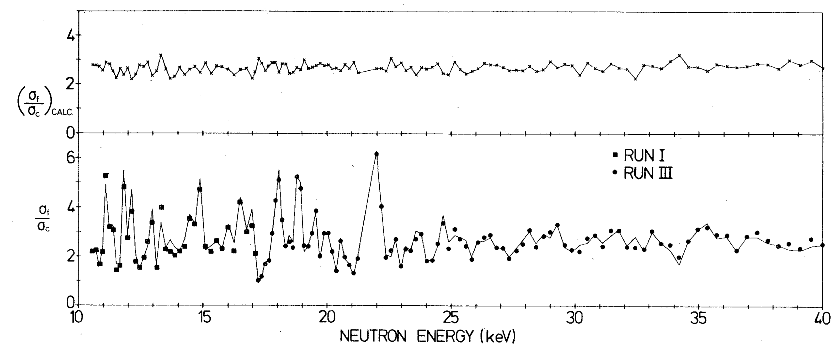

FIG. 8. (above). Mocked-up fission-to-capture ratio of ^{235}U computed by Monte Carlo techniques to demonstrate the influence of Porter-Thomas fluctuations. (below). Comparison of the experimental $1/\alpha$ data (circles and squares) with the normalized ratio of experimental data and the calculated curve (solid line). It is obvious that Porter-Thomas fluctuations do not affect the observed structures significantly.

ible in this curve are smaller than 17% at 15 keV and smaller than 9% at 40 keV. In order to check whether these Porter-Thomas fluctuations could affect the observed structure in $1/\alpha$, the ratio of the experimental data and the calculated $1/\alpha$ values was formed. The normalized ratio is shown in the lower part of Fig. 8 (solid line) together with the experimental data points. As is obvious from this comparison the Porter-Thomas fluctuations do not change the intermediate structure wiggles significantly.

Below 20 keV where the energy resolution is sufficient an average width of typically 0.5 keV is observed for the structures at 11.3, 12.0, 13.2, 14.7, 16.0, 16.4, 18.0, and 18.7 keV. If these energies are checked with Fig. 9 of Ref. 29, there is strong evidence for correlated structures in the fission cross section of the 4^- channel. Especially the cluster at 14.7 keV can clearly be identified in these data. In the energy range from 10 to 30 keV 20 resonancelike structures were determined giving an average level spacing of 1000 ± 300 eV. The corresponding level spacing distribution is shown by the histogram plot in Fig. 9 which indeed is well represented by a Wigner distribution within the available statistics. Using the level density formula with adjusted parameters reported by Dilg *et al.*[35] and the additional information that the partially open fission channel with $J = 4^-$ is responsible for the intermediate structure[29] a value of 3.26 ± 0.14 MeV was calculated for E_{II}, where the quoted uncertainty reflects the uncertainty in the average level spacing only. Compared to earlier analyses of intermediate structures in the neutron fission cross section of ^{235}U which yielded values around 2.3 MeV (Refs. 28, 30) the present result is significantly higher. This might be due to the fact that our value is determined from a more reliable data base. There is also disagreement with theoretical calculations of Nilsson *et al.*,[36] who determined $E_{II} = 2.4$ MeV for the compound nucleus ^{236}U.

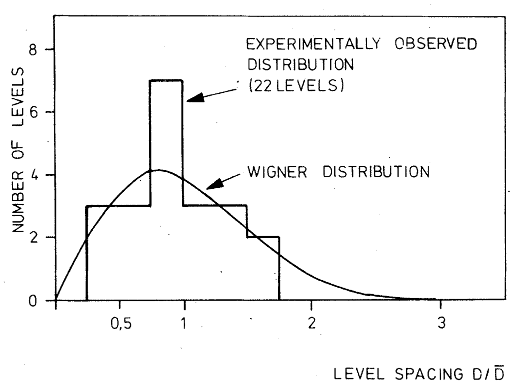

FIG. 9. Spacing distribution for 22 resonancelike structures in $1/\alpha$ of ^{235}U between 10 and 32 keV neutron energy. Within the available statistics good agreement is found with a Wigner distribution.

VIII. CONCLUSIONS

In this experiment the capture-to-fission ratio α could be determined with a new experimental technique in the neutron energy range from 10 to 500 keV with an accuracy of typically 8% which has been exceeded only by the results of Gwin *et al.*[9] but at the expense of averaging over larger energy intervals. In addition, this quantity was measured with a reasonable energy resolution of 2 nsec/m. This allowed, for the first time, observation of a strong intermediate structure in the unresolved resonance region which can be attributed to the fission channel. In this way it was possible to deduce an average level spacing D_{II} from a statistically significant sample. The interpretation of this structure in terms of the double humped fission barrier resulted in a value for the energy difference between the first and second well of $E_{II} = 3.26 \pm 0.14$ MeV.

The present measurement demonstrates that neutron induced fission can make an important contribution to the investigation of the fission barrier, even in cases such as ^{235}U where the neutron binding energy exceeds the fission barrier. The capture-to-fission ratio α is especially sensitive to structure in the fission channel because the influence of the reaction entrance channel cancels out. Provided that sufficient statistics can be achieved, further improvements in energy resolution seem possible, which certainly would allow a more detailed analysis of fission barrier parameters.

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