

THE PROMPT FISSION NEUTRON SPECTRA FROM SPONTANEOUS FISSION OF ^{252}Cf AND THERMAL NEUTRON-INDUCED FISSION OF ^{233}U , ^{235}U , AND ^{239}Pu

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Abstract

The measurements of the energy and angular distributions of prompt neutrons associated with light and heavy groups of fission fragments from spontaneous fission of ^{252}Cf and from $^{233,235}\text{U}(n,f)$, $^{239}\text{Pu}(n,f)$ thermal-neutron induced fission have been done. A description of experimental set-up, data processing and analysis is presented. The analysis of the results demonstrates some deviation of experimentally observed fragment-neutron angular correlations from those calculated assuming, that the prompt neutrons were emitted solely by the accelerated fragments.

1. General description

The fission neutrons which are emitted from sources other than accelerated fission fragments at present are known as "scission" neutrons. The "scission" neutrons can't be selected from prompt fission neutrons emitted by accelerated fragments. This can be done only by comparison of the experimental fragment-neutron angular correlations with model calculations based on the assumption of neutron emission from accelerated fragments. The model calculations taking into account other emission mechanism enable to select the emission mechanism realized in practice. It is clear that the experimental data needed for such comparison, should be obtained using the same set-up and data processing for many fissioning nuclei. A series of such experiments has been performed to measure prompt neutron angular and energy distributions from spontaneous fission of ^{252}Cf and from $^{233,235}\text{U}(n,f)$, $^{239}\text{Pu}(n,f)$ thermal-neutron induced fission.

The measurements were carried out at the WWR-M research reactor in PNPI (Gatchina, Russia). The energy and angular distributions of prompt neutrons associated with light and heavy groups of fission fragments from spontaneous fission of ^{252}Cf and from $^{233,235}\text{U}(n,f)$, $^{239}\text{Pu}(n,f)$ reactions induced by 0.0363 eV neutrons have been measured. The neutrons were detected using two scintillation stilbene crystal detectors while the fragments were detected by multi-wire proportional detectors. The neutron registration threshold was 150-200 keV. A pulse shape analysis was applied to select neutron and gamma-quanta events. The neutron energy and fission fragment characteristics were obtained by the time-of-flight method.

The prompt fission neutron spectra, PFNS, were measured simultaneously for 11 angles between the neutron direction and the fission-fragment direction of motion: 0° , 18° , 36° , 54° , 72° , 90° , 108° , 126° , 144° , 162° and 180° . If a real geometry of the experimental set-up is taken into account, these angles are equal to 8.9° , 19.8° , 36.9° , 54.5° , 72.2° , 90° , 107.8° , 125.5° , 143.1° , 160.2° and 171.2° , respectively. To avoid usage of any model assumption in the analysis and data processing, the spectra were measured in a wide prompt fission neutron energy interval. It should be noted also that realized scheme of the experimental set-up guarantees identity of conditions of PFNS measurements at various angles relative to the

fission fragment direction, namely: the magnitude and composition of the background and neutron re-scattering by various parts of the experimental set-up. A detailed description of the experimental set-up and some preliminary results have been presented in our previous publication [1]. From the subsequent analysis of these data [2], it was concluded that it was necessary to consider some additional corrections and to improve the data processing. The following effects should be noted: correction for real efficiency of neutron detectors, the normalization correction arising from the fact that experimental angular histograms were used in the measurements instead of continuous distributions, neutron energy resolution correction and the neutron detector background due to accidental coincidence between fragment and neutron belonging to different fission events. In order to determine the PFNS from measured time-of-flight spectra, the relativistic equation was used rather than non-relativistic one used in earlier publication. Also, the angular resolution correction was applied in a more consistent approach than used in Ref. [1].

2. Analysis 1: the total PFNS obtained by summing up measured angular and energy distributions over angles

As a result of the data analysis and additional measurements, the absolute ratio of the total PFNS of investigated nuclei (^{235}U , ^{233}U and ^{239}Pu) to the total PFNS of $^{252}\text{Cf}(\text{sf})$ were obtained (the used procedure can be found in ref. [3]). Then, the total PFNS of investigated nuclei were calculated using the total PFNS of $^{252}\text{Cf}(\text{sf})$, which is recommended as an international standard for neutron-spectrum measurements [4]. The advantages of ratio measurement are as follows: it enables to exclude the neutron detector efficiency uncertainty from the data obtained and allows to re-determine the total PFNS of investigated nuclei in the case of an improvement of the ^{252}Cf - standard. So, the total corrections needed to include into the measured ratio, which is almost independent of the features of the experimental set-up, does not exceed 3% in the 0.2 - 10 MeV neutron-energy range. The obtained ratios and the PFNS of investigated nuclei are presented in Fig. 1-3.

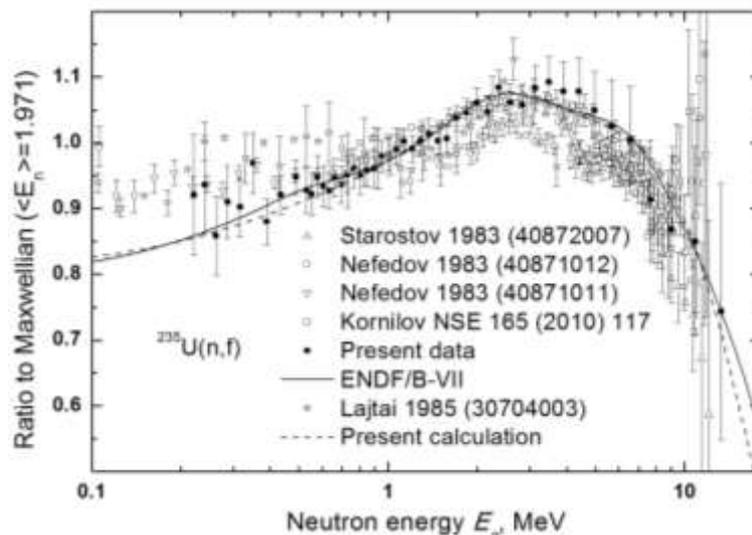


Fig. 1. Comparison of total PFNS of ^{235}U obtained by different experimental groups and ENDF/B-VII. Data are presented as ratio to Maxwellian with $T=1.314$ MeV. All experimental data were normalized to 2.42 (average number of prompt fission neutrons). The dash curve is the total PFNS (normalized to 1) calculated from prompt neutron spectra measured at small angles relative to the fission fragment direction of motion in the assumption, that neutrons are emitted by accelerated fragments (see text).

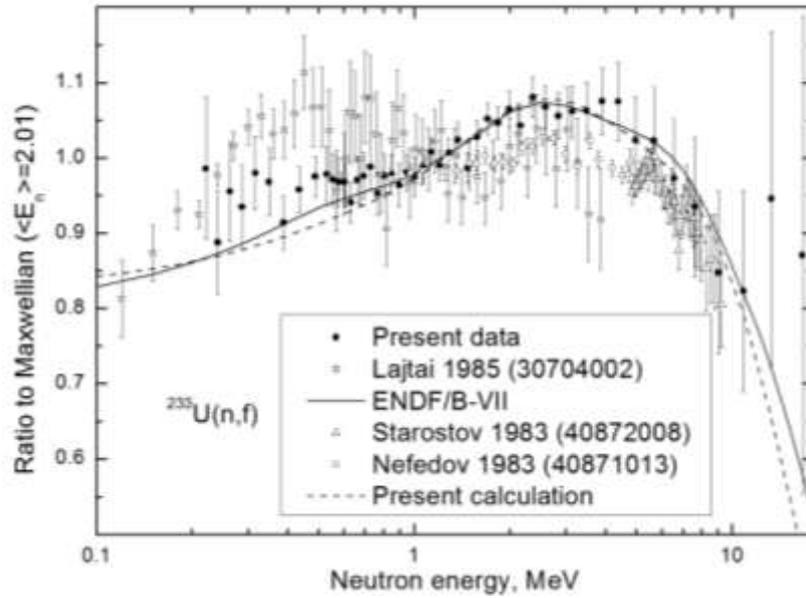


Fig. 2. Comparison of total PFNS of ^{233}U obtained by different experimental group and ENDF/B-VII. Data are presented as ratio to Maxwellian with $T=1.34$ MeV. All experimental data were normalized to 2.49 (average number of prompt fission neutrons). The dash curve is the total PFNS (normalized to 1) calculated from prompt neutron spectra measured at small angles relative to the fission fragment direction of motion in the assumption, that neutrons are emitted by accelerated fragments.

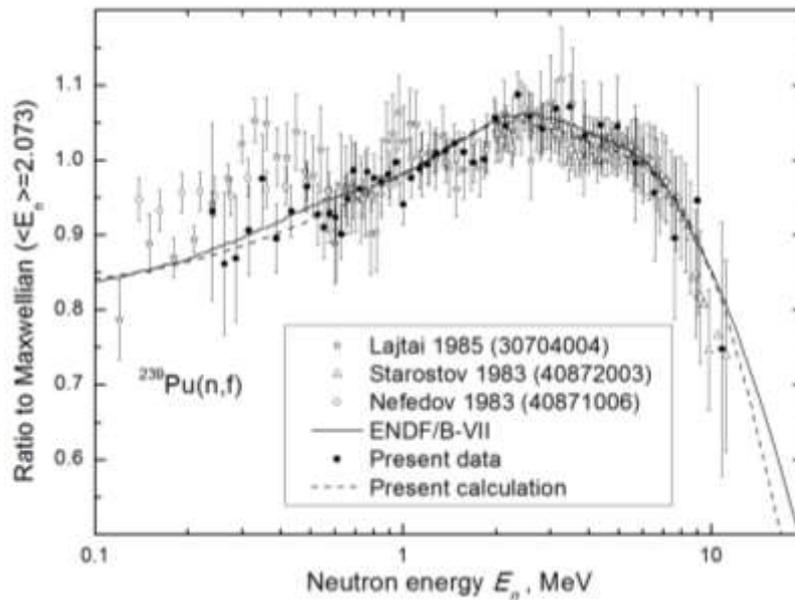


Fig. 3. Comparison of total PFNS of ^{239}Pu obtained by different experimental group and ENDF/B-VII. Data are presented as ratio to Maxwellian with $T=1.382$ MeV. All experimental data were normalized to 2.89 (average number of prompt fission neutrons). The dash curve is the total PFNS (normalized to 1) calculated from prompt neutron spectra measured at small angles relative to the fission fragment direction of motion in the assumption, that neutrons are emitted by accelerated fragments.

3. Analysis 2: the total PFNS calculated from neutron spectra measured at small angles relative to fission fragments of motion

It should be noted that the total PFNS may be calculated using neutron data measured at small angles relative to fragment direction of motion in the laboratory system (8.9°, 19.8°, 36.9° and 171.2°, 160.2°, 143.1°) on the basis of the assumption, that prompt fission neutrons are emitted from light and heavy accelerated fragments (two fragment approximation) and the anisotropy of fission neutron angular distribution in the center-of-mass system of fission fragment A_2 ($N(0)/N(90)=(1+A_2)/(1-A_2/2)$) is equal to 0.04 [1, 5]. In the course of these calculations, only the average energy per nucleon for light $\langle E_{Lf} \rangle$ and heavy $\langle E_{Hf} \rangle$ fragments, obtained from literature data by means of the equations analogous to that in Ref. [6] (Table 1), are used as input parameters.

Table 1. Data used for calculation of the average energy per nucleon for light and heavy fragments.

	$^{233}\text{U}(\text{n},\text{f})$	$^{235}\text{U}(\text{n},\text{f})$	$^{239}\text{Pu}(\text{n},\text{f})$	$^{252}\text{Cf}(\text{sf})$
$\langle \text{TKE} \rangle$, MeV	170.5 ± 0.5	171.0 ± 0.6	177.5 ± 0.7	185.3 ± 0.9
$\langle E_L \rangle$, MeV	101.5 ± 0.3	101.2 ± 0.4	103.1 ± 0.5	105.5 ± 0.6
$\langle M_H \rangle$	139.3 ± 0.2	139.7 ± 0.1	139.5 ± 0.1	143.5 ± 0.1
$\langle E_L/M_L \rangle$, MeV	1.075 ± 0.004	1.054 ± 0.004	1.029 ± 0.005	0.975 ± 0.005
$\langle E_H/M_H \rangle$, MeV	0.499 ± 0.002	0.503 ± 0.002	0.537 ± 0.003	0.560 ± 0.003
$\langle E_{Lf} \rangle = \langle v_L E_L / \langle v_L \rangle M_L \rangle$, MeV	1.033 ± 0.007	1.012 ± 0.007	0.995 ± 0.007	0.949 ± 0.007
$\langle E_{Hf} \rangle = \langle v_H E_H / \langle v_H \rangle M_H \rangle$, MeV	0.471 ± 0.004	0.474 ± 0.004	0.511 ± 0.004	0.540 ± 0.004

The calculations performed by this method have several advantages: they are very simple to use and free from any model parameters (the number of neutrons emitted by light and heavy fragments, the neutron spectrum shape, etc.). It is possible to obtain the PFNS in the center-of-mass system, which is practically unrestricted in the low energy range and, therefore, it is possible to determine the average number of neutron without any approximations or interpolation. Only the experimental data obtained in present investigation are used – minimizing uncertainties due to errors of used input parameters, which are difficult to estimate. The analysis and data processing are performed equally for all investigated nuclei – the possible systematic effect will be the same for all investigated isotopes and can be estimated.

The analysis of the data revealed, that the PFNS in the center-of-mass system of fission fragment per MeV per steradian can be fitted by means of a following equation:

$$Fit(\varpi, T_1, T_2, E_n^{c.m.}) = \frac{\langle v \rangle}{4\pi} \cdot \left[\varpi \cdot \frac{E_n^{c.m.}}{T_1^2} \cdot \exp\left(-\frac{E_n^{c.m.}}{T_1}\right) + (1-\varpi) \cdot \frac{2 \cdot \sqrt{E_n^{c.m.}}}{\sqrt{\pi} \cdot T_2^3} \cdot \exp\left(-\frac{E_n^{c.m.}}{T_2}\right) \right]. \quad (1)$$

By using fitting parameters obtained (ϖ , T_1 , T_2 , v for light and heavy fragments), the total PFNS were calculated for all investigated nuclei (see Fig. 1-4, dash curve). The detailed comparison of calculated total PFNS with that, obtained by summation of measured angular and energy distributions analogous to ref. [3] for investigated nuclei (Fig. 1-4), enables to say about the agreement between present and literature data within experimental errors in neutron energy range from 0.6 MeV to 10 MeV.

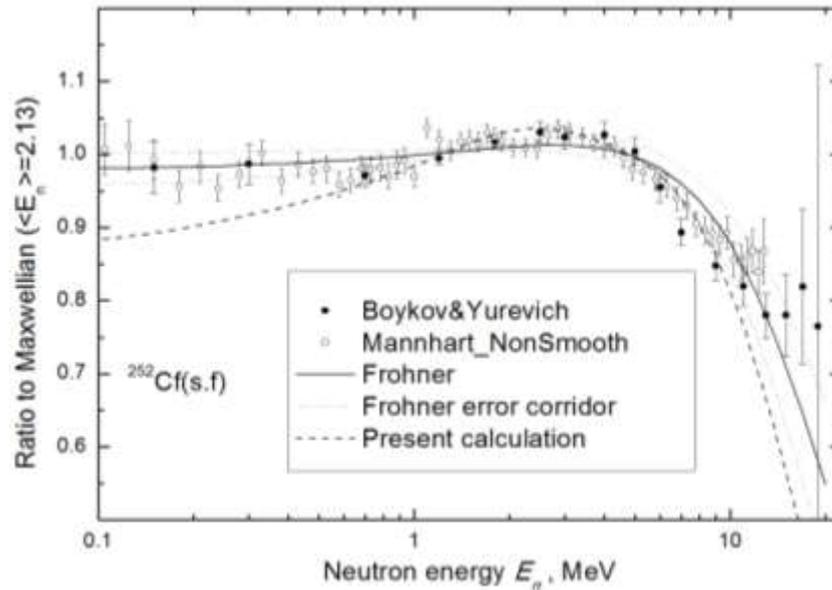


Fig. 4. Comparison of total PFNS of ^{252}Cf obtained by different evaluations, using relevant experimental data and present calculation. Data are presented as ratio to Maxwellian with $T=1.42$ MeV. The dash curve is the total PFNS (normalized to 1), calculated from prompt neutron spectra measured at small angles relative to the fission fragment direction of motion in the assumption, that neutrons are emitted by accelerated fragments.

Also, it takes place a good agreement between present calculation and ENDF/B-VII data. Therefore, it is possible to conclude that the contribution of the “scission” neutrons to the total PFNS is comparatively small. The systematic difference of calculated total PFNS (analysis 2) from total PFNS, measured by different experimental groups (including our data - analysis 1) is visible for all investigated nuclei in the neutron energy range lower than 0.6 MeV. For example, in Fig. 5 the real difference between Mannhart’s evaluation and our calculation (analysis 2) is shown instead of the normalized ratio as in Fig. 4. This difference may be interpreted as a manifestation of “scission” neutrons and, therefore, for investigated nuclei their average energy is about $0.4\div 0.5$ MeV and their yield can be estimated as large as $1\div 3\%$ of the total prompt fission neutron yield. This result coincides with conclusion made in earlier works [9, 10]. To verify this statement, the PFNS measured at angle 90° relative to the direction of the fragments’ movement, were compared with calculated PFNS at the same angle (for example, see Fig. 6).

As a result, the difference of these spectra can be described as a sum of two contributions: the low energy component with average energy and yield as that obtained from difference spectrum of total PFNS, and the high energy component with average energy about 3.1 MeV. Analogous features are seen for all investigated nuclei. They were also found in the compilation work [11]. It seems that neutrons of low energy component have isotropic distribution in the laboratory system, because the low energy component of difference spectrum, obtained both by analysis of the total PFNS and by analysis of partial PFNS at angle 90° relative to the direction of the fission fragments’ movement, coincides with each other. Since the high energy component is visible only for angles near 90° relative to the fission fragment direction of movement, the emission mechanism of these neutrons can be established only step-by-step self-consistent comparison of measured spectra with calculation, taking into account any possible additional mechanism. In an assumption, that these high

energy neutrons are emitted isotropically in the laboratory system, their yield was calculated as equal to 1%, 2%, 3% and 3% of the total neutron yield for $^{235,233}\text{U}(n,f)$, $^{239}\text{Pu}(n,f)$ and $^{252}\text{Cf}(s.f)$, respectively.

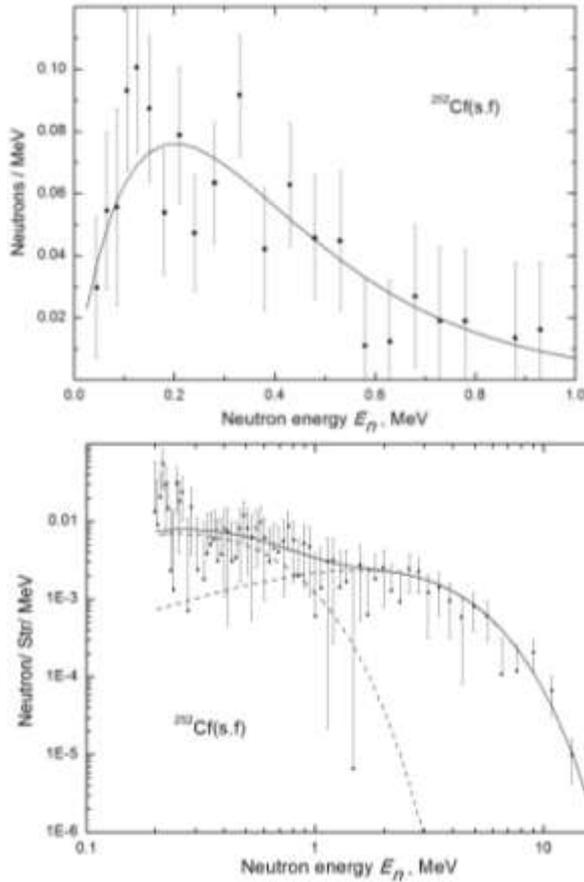


Fig. 5. The difference spectrum between Mannhart's evaluation and the total PFNS, calculated using neutron spectra measured at small angle relative to the direction of the fission fragments' movement for $^{252}\text{Cf}(s.f)$. The PFNS used were normalized to the average number of prompt fission neutrons per fission event, 3.759. The full line represents a least square fit by Weisskopf distribution (with $T = 0.20$ MeV) through the difference data points, presented as dots with error bars.

Fig. 6. The difference spectrum between the PFNS, measured at angle 90° relative to the direction of the fragments' movement, and the PFNS calculated at the same angle (analysis 2). The full line represents a fit by sum of two Weisskopf distributions (with $T_1 = 0.24$ MeV and $T_2 = 1.56$ MeV, dash lines) through the difference data points presented as dots with error bars.

4. Conclusion

The prompt fission neutron energy spectra have been measured for $^{235, 233}\text{U}(n,f)$, $^{239}\text{Pu}(n,f)$ and $^{252}\text{Cf}(s.f)$ at 11 fixed angles between the neutron and light fragment directions in the range from 0° to 180° . The comparison of experimentally obtained angular and energy distributions of prompt neutrons and calculated ones on the base of neutron emission from accelerated fission fragments enables to do some conclusions. First, the angular anisotropy of the neutron emission in the fragment center-of-mass system, $A_2 \sim 0.04$, should be included into any calculation of prompt neutron properties in the neutron-induced fission. Second, the "scission" neutron spectrum can be presented as a sum of two components. The neutron yield of low energy component is equal to about 1÷3% of the total prompt neutron yield and definitely has isotropic distribution in the laboratory system for all investigated nuclei, whereas nothing can be said about angular distribution of the second component. Third, in an assumption of isotropic emission in the laboratory system, the contribution of "scission" neutrons is not more than $3\pm 2\%$, $4\pm 2\%$, $6\pm 2\%$ and $5\pm 2\%$ of the total neutron yield for $^{235,233}\text{U}(n,f)$, $^{239}\text{Pu}(n,f)$ and $^{252}\text{Cf}(s.f)$ respectively. Probably, this assumption is very close to the real situation, because in the measurements of the angular dependency of the neutron-neutron coincidence curves [12], which are very sensitive to isotropic component in the

laboratory system, the same values of “scission” neutron yield were obtained within experimental errors.

Acknowledgments

Authors wish to thank D.O. Krinitsin, L.S. Falev and V.E. Sokolov for help in adjustment of the equipment and conducting the experiments, T.E. Kuz'mina and S.M. Soloviev from Khlopin Radium Institute for producing the targets. The authors are very grateful to I.S. Guseva and F. Gönnewein for helpful discussions.

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