SEARCH FOR SCISSION NEUTRONS IN THE MEASUREMENT OF ANGULAR AND ENERGY DISTRIBUTIONS OF THE PROMPT FISSION NEUTRONS FOR ^{233}U, ^{235}U, ^{239}Pu AND ^{252}Cf

A.S. Vorobyev, O.A. Shcherbakov, A.M. Gagarski, G.V. Val’ski

B.P. Konstantinov Petersburg Nuclear Physics Institute of National Research Centre “Kurchatov Institute”, Gatchina, Leningrad district, 188300, Russia

Abstract

The measurements of angular and energy distributions of the prompt fission neutrons from thermal neutron-induced fission of ^{233}U, ^{235}U, ^{239}Pu and from spontaneous fission of ^{252}Cf were carried out at the WWR-M research reactor in Gatchina, Russia. Some peculiarities were found in the angular distribution of the prompt fission neutrons. It is possible to explain them by assuming that in the center-of-mass system of fission fragment the neutrons are more likely emitted along fission direction than in the perpendicular one. The value of anisotropy of neutrons emission in the center-of-mass system of fission fragment was found to be equal to 6-8% for all nuclei under investigation. The yields of “scission” neutrons have been estimated: 1.5÷2.7% (^{233}U), 1.8÷2.6% (^{235}U), 3.6÷4.5% (^{239}Pu) and 2.0÷3.0% (^{252}Cf) with the average uncertainty 0.8%.

Introduction

As a result of experimental studies of the emission of prompt fission neutrons (PFNs) [1, 2], it was found that neutrons are emitted primarily in the direction of motion of fragments and that the proposed hypothesis of evaporation of neutrons from fully accelerated fragments [3] provides the general description of observed features [4]. For the detailed description of angular and energy distributions of PFNs, it is necessary to assume the existence of “scission” neutrons, i.e., neutrons whose emission mechanism differs from evaporation of neutrons from fully accelerated fragments (emission of neutrons before or at the time of scission of a fissioning nucleus or in the process of acceleration of produced fission fragments). In particular, for the most studied case of spontaneous fission of ^{252}Cf(sf), estimates of the contribution of “scission” neutrons obtained from the analysis of independent experimental data range from 1 to 20% of the total number of neutrons per fission event (Fig. 1 upper part). The information on the anisotropy of the PFNs emission in the center-of-mass system of fission fragments obtained from the experiment is even more scarce than information on the yield of “scission” neutrons (Fig. 1 lower part).

The main purpose of this work was the experimental investigation of the emission mechanism of PFNs by the coincidence measurements of angular and energy distributions of neutrons and fission fragments. The experimental data needed for such investigation, ideally, should be obtained using the same set-up and data processing for many nuclei at different excitation energies. Therefore, using the same experimental set-up and data processing, a few experiments have been carried out at NRC KI PNPI (Gatchina, Russia) to measure the angular and energy distributions of prompt neutrons from thermal neutron-induced fission of ^{233,235}U, ^{239}Pu and spontaneous fission of ^{252}Cf [5-9]. In this paper, some results of this investigation are presented.

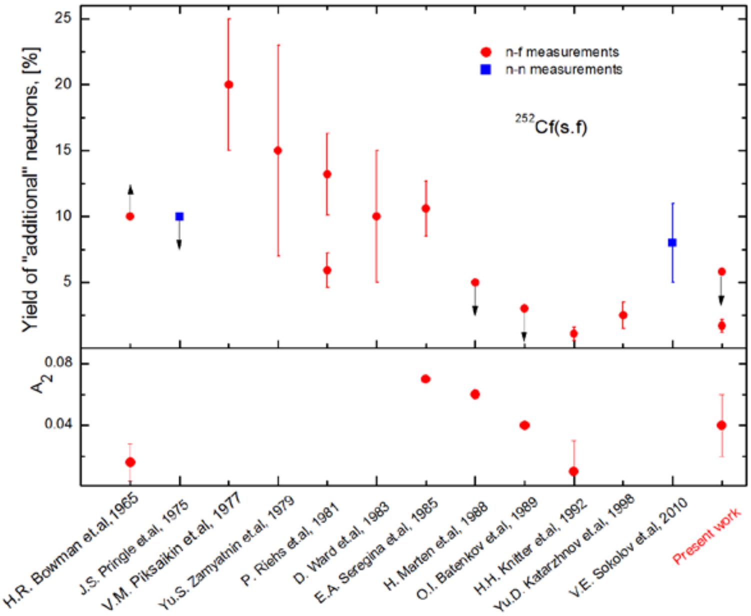

Fig. 1. Main results of previous investigations of PFN emission mechanism for ^{252}Cf: upper part - yield of "scission" neutrons (downward and upward arrows indicate the upper and lower bounds, respectively); lower part - anisotropy of the angle distribution of PFNs in the center-of-mass system of fission fragments.

1. Experiment overview

The measurements of angular and energy distribution of PFNs were carried out at the research reactor WWR-M of PNPI. The fission fragments and prompt neutrons time-of-flights were measured simultaneously for 11 fixed angles, θ, between the axis of neutron detector and normal to the surface of stop multi-wire proportional detectors (MWPDs) (coming through its center) in the range from 0° to 180° in 18° intervals. The schematic view of the experimental set-up is shown in Fig. 2. The neutron beam was coming along the chamber axis normally to the Fig. 2 plane.

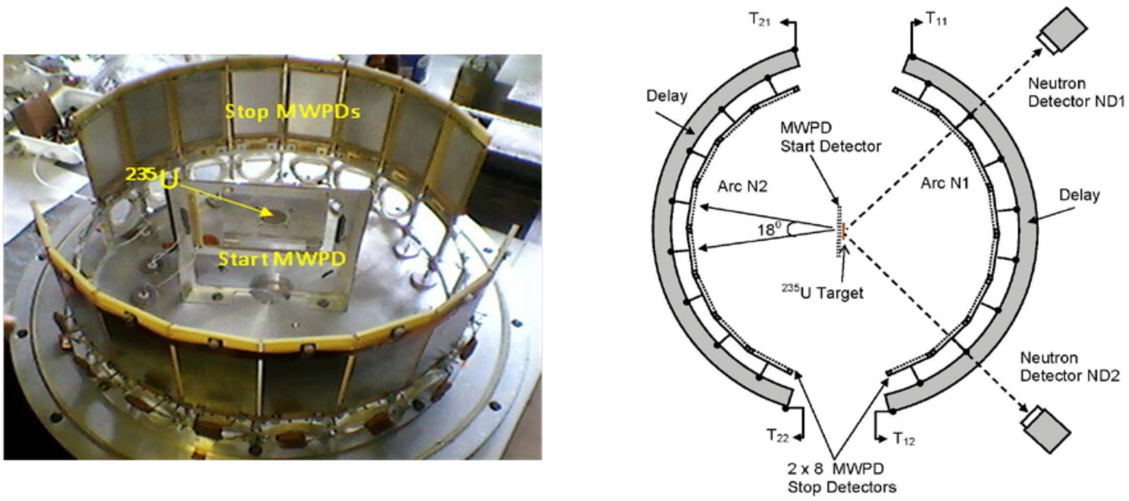

Fig.2. The experimental setup: left – the photo of reaction chamber with MWPD detectors; right - schematic view of the experiment.

It should be noted that realized scheme of the experimental set-up guarantees identity of conditions of the neutron spectra measurements at various angles relative to the fission axis, namely: the magnitude and composition of the background, the efficiency of the neutron detectors, and neutron re-scattering by the parts of experimental set-up. Also, the use of two neutron detectors with slightly different characteristics enables to estimate probable systematic errors of the data obtained.

The prompt neutrons were detected using two stilbene crystal detectors (Ø 50 mm x h 50 mm and Ø 40 mm x h 60 mm) positioned at a 90° angle between their axes at a distance of (47.2±0.2) cm and (49.2±0.2) cm from the fissile target, respectively. The axes of neutron detectors ND1 and ND2 came through the centers of two stop MWPDs located on the Arc N1. Both neutron detectors were surrounded by a cylindrical shield made of 30 mm thick layer of lead and 40 mm thick layer of polyethylene (not shown in Fig. 2). The neutron registration threshold was 150 – 200 keV. To separate events corresponding to neutrons and γ-quanta, a double discrimination by the pulse shape and time-of-flight was applied. The full time uncertainties were defined from FWHM of the "fragment - γ-quantum" coincidence curve which was equal to 1.0÷1.2 ns.

The fission fragments were detected by MWPDs in conjunction with the TOF technique. The 8 rectangular MWPDs were located in the Arc N2 in the reaction chamber at the operating gas (isobutane) pressure of 4 ÷ 6 Torr.

After all necessary correction were taken into account for 11 fixed angles between neutron and light fragment directions, the energy distributions of PFNs emitted from fixed pair of fission fragments were obtained. A description of the experimental method and the used data processing procedure are omitted since a full treatment was given in ref. [5, 7, 8].

2. Model

Since "scission" neutrons in experiment cannot be separated from neutrons emitted from fully accelerated fragments, estimates of the yield of "scission" neutrons and possible anisotropy of prompt fission neutrons in the center-of-mass system of fragments were obtained by comparing the measured distributions of PFNs with model calculations under the assumption that all prompt fission neutrons are emitted from fully accelerated fragments.

In the model calculation it is used the assumption that PFNs are emitted from fully accelerated fragments. In this case, the angular and energy distributions of PFNs in the laboratory system can be calculated using known spectra of PFNs in the center-of-mass system of fragment. The spectra of PFNs in the center-of-mass system of fragment were calculated using experimental data for small angles (8.9°, 19.8° and 36.9°) relative to the fission direction. During this calculation, it was assumed that prompt neutrons are emitted by two fragments with average mass and kinetic energy. The average energies per nucleon for light and heavy fragments for investigated nuclei were taken from ref. [10]. Further, the spectra obtained in the center-of-mass system were used for calculation of neutron angular and energy distributions in the laboratory system. These distributions were compared with the experimental distributions to estimate contribution and properties of "scission" neutrons.

It should be noted that the calculated spectra are free of any assumption about the prompt neutron spectra in the center-of-mass system (the number of neutrons emitted by heavy and light fragments, the neutron spectrum shapes, and so on). There is only one free parameter the anisotropy of PFNs in the center-of-mass system of fragment, which is adjusted so as to describe in the best way all experimental data obtained in this investigation. The value of anisotropy of neutrons emission in the center-of-mass system of fission fragment was found to be equal to 6-8% for all nuclei under investigation. The details could be found in ref. [5, 11, 12].

The shape of the neutron spectrum and the number of neutrons obtained in the center-of-mass system both depend on the fragment velocities (or E_L and E_H for fission event). Therefore, strictly speaking, the analysis performed above is not valid, because it was assumed that the prompt neutrons are emitted only from two fragments (light and heavy) characterized by the average parameters. Fortunately, as it was demonstrated for total PFN spectrum of ^{252}Cf in Ref. [11], a transition from the velocity distributions of fragments to the model of two fragments with average parameters has only a minor influence, and for angles near 90° the neutron yield changes within 4% [7].

At the same time, the existing calculation methods used in practice to describe angular and energy distributions of PFNs do not provide necessary accuracy. For example, the total PFNs of ^{235}U calculated by different commonly used codes [10] are presented in Fig. 3, where the spectra calculated assuming that PFNs are emitted from fully accelerated fragments and using the same input parameters are shown as a ratio to Maxwell distribution. It is seen that the existing calculation methods do not provide necessary accuracy to describe experimental data while the method realized in this work gives accuracy not worse than those of commonly used codes and does not require knowledge of a large number of input parameters.

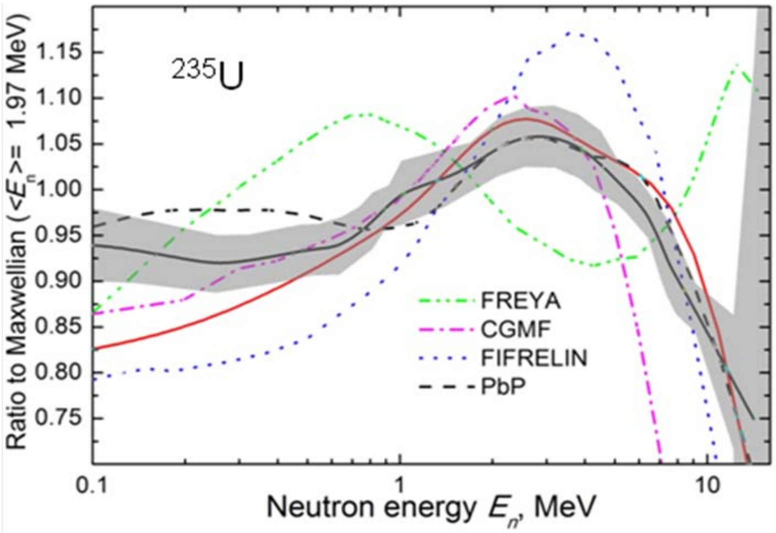

Fig. 3. Total PFN spectrum of ^{235}U(n_{th}, f): curve inside the shaded region – evaluation of experimental data within error corridor (GMA – generalized least square fit [10]); line – model calculation (two fragments approximation) [2]; **PbP** (Point by Point) - deterministic method developed at the University of Bucharest and JRC-IRRM team, which is an extended version of LAM (Los-Alamos or Madland-Nix model); **FREYA** (Fission Reaction Event Yield Algorithm) – Monte-Carlo fission model developed through a collaboration between LLNL and LBNL (USA); **CGMF** – Monte-Carlo code developed at LANL (USA); **FIFRELIN** (FIssion FRagment Evaporation Leading to an Investigation of Nuclear data) - Monte-Carlo code developed at CEA-Cadarache (France) with the aim of calculating the main fission observables.

3. Results and discussion

On the whole, the calculated model energy and angular distributions agree rather well with the experimentally obtained distributions. It is also possible to describe the total PFN spectrum in the laboratory system above 1 MeV and their average number of PFNs per fission event. However, there is a minor distinction which is observable for all investigated nuclei [6, 10-12]. For example, in Fig. 4, the PFN spectrum measured for angle 90^o relative to fission fragment direction and the total PFN spectrum obtained by summing over angles for ^{239}Pu(n,f) are compared with the corresponding calculated values. The observed differences may be interpreted as a manifestation of “scission” neutrons and the average energy of these neutrons and their yield can be estimated.

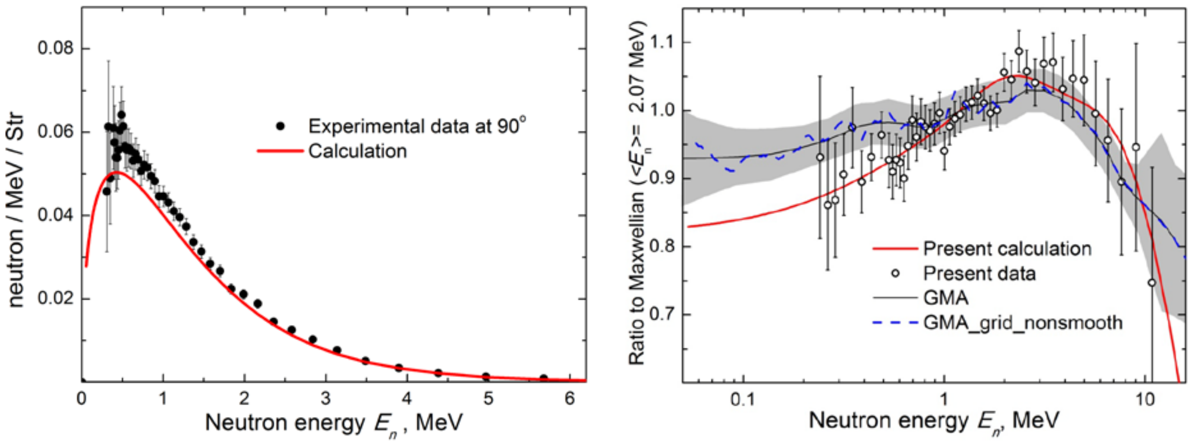

Fig. 4. Left – the PFN spectrum measured for angle 90^o relative to the fission fragment direction. Right – the total PFN spectrum of ^{239}Pu(n,f) obtained by summing over angles θ are shown as a ratio to Maxwell distribution. GMA – generalized least square fit of PFN spectra measured by different experimental groups (non-model evaluation) – taken from ref. [10].

The systematic difference between calculated total PFN spectra and total PFN spectra measured by different experimental groups (evaluated spectrum – GMA fit) is visible in the neutron energy range lower than 0.6 MeV. The “scission” neutron spectrum can obtained as a difference between evaluated total PFN spectra (GMA fit) and model calculation. To verify this statement, the PFN spectra measured for angles close to 90° relative to the direction of the light fragments’ movement, were compared with calculated PFN spectra at the same angles. The “scission” neutron spectrum was obtained with the use of the difference spectra obtained as the difference between the measured and model spectra for angles of 72.2°, 90° and 107.8° with respect to the direction of motion of the light fragment. To compare the two estimates, the “scission” neutron spectrum obtained in the first way was multiplied by 4π (it was assumed that the distribution of “scission” neutrons in the laboratory system was isotropic). A comparison of the spectra obtained in this manner shows the agreement (within the errors of the experimental data) between the results from estimates performed in different ways. For example, in Fig. 5 these difference spectra for ^{252}Cf and ^{239}Pu are presented.

Since the relative contribution from “scission” neutrons should be largest at angles Ω close to 90°, the yield of these neutrons from the fission of the investigated nuclei was estimated using the spectrum obtained in the second way: with least squares approximated by functions (1) and (2):

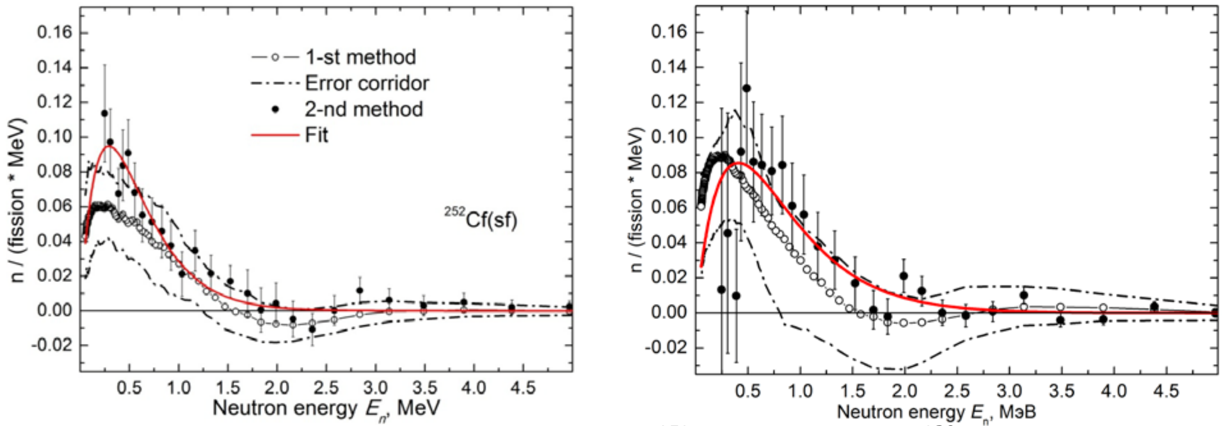

Fig. 5. Spectrum of "scission" neutrons: left - for ^{252}Cf(sf); right - for ^{239}Pu(n,f). Points - the difference spectrum obtained using spectra measured at 72.2°, 90° and 107.8° relative to the direction of motion of the light fission fragment and the corresponding ones calculated under the assumption that all prompt neutrons are emitted from the accelerated fragments. Circles - the difference between total PFN spectrum obtained by experiment (estimated data and it's errors) and calculated assuming that all prompt neutrons are evaporated from accelerated fragments. Solid line - fit of experimental data marked with points by the equation (1).

Table 1. Main characteristics of "scission" neutrons.

	^{233}U(n,f)	^{235}U(n,f)	^{239}Pu(n,f)	^{252}Cf(sf)
Approximation using function (1)				
Yield, %	1.5 ± 0.6	1.8 ± 0.6	3.6 ± 0.6	2.0 ± 0.6
Average energy, MeV	0.53 ± 0.08	0.47 ± 0.05	0.91 ± 0.19	0.58 ± 0.06
Approximation using function (2)				
Yield, %	2.7 ± 0.8	2.6 ± 0.8	4.5 ± 0.9	3.0 ± 0.8
Average energy, MeV	1.7 ± 0.2	1.4 ± 0.2	1.6 ± 0.2	1.5 ± 0.2

$$p_S(E) = \frac{p_0}{4\pi} \cdot \frac{E}{T_0^{\,2}} \cdot \exp\left(-\frac{E}{T_0}\right) \tag{1}$$

$$p_S(E) = \frac{p_0}{4\pi} \cdot \frac{E}{T_0^{\,2}} \cdot \exp\left(-\frac{E}{T_0}\right) + \frac{p_1}{4\pi} \cdot \frac{E}{T_1^{\,2}} \cdot \exp\left(-\frac{E}{T_1}\right) \tag{2}$$

The parameters p_0, T_0, p_1 and T_1 were varied. The results of these approximations are given in Table 1.

Conclusion

To estimate the yield of "scission" neutrons in fission the measurements of angular and energy distributions of PFNs from thermal neutron-induced fission of ^{233,235}U, ^{239}Pu and spontaneous fission of ^{252}Cf have been carried out at the WWR-M research reactor in NRC KI PNPI (Gatchina, Russia). The analysis of these data demonstrates a general agreement between experimental data and model calculations performed assuming that PFNs are emitted from fully accelerated fission fragments. But there are some differences which cannot be explained within the model of neutron emission from fully accelerated fragments. These differences can be eliminated by assuming that there were ~2–4% of “scission” neutrons. It should be noted that these estimations of properties of “scission” neutrons were performed assuming isotropic emission of “scission” neutrons in the laboratory system. Probably, this assumption is very close to the real situation, because in the measurements of the angular dependency of the neutron-neutron coincidence curves, which are very sensitive to isotropic component in the laboratory system, the same values of “scission” neutron yield were obtained within experimental errors [13].

The nature of the observed neutron excess can be determined after a thorough comparison of the experimental data and the calculations using theoretical models that allow for possible PFNs emission mechanisms in fission.

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