ANGULAR AND ENERGY DISTRIBUTIONS OF PROMPT NEUTRONS FROM THERMAL NEUTRON-INDUCED FISSION OF ^{233,235}U(n_{th}, f)

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Abstract

The energy spectra and angular distributions of prompt fission neutrons from thermal neutron-induced fission of ^{233,235}U(n_{th}, f) have been measured for fragments with given mass and kinetic energy. The prompt neutron energy and fission fragment characteristics were obtained by a conventional time-of-flight method. To separate fission neutrons and γ-quanta the double discrimination by the pulse shape and the time-of-flight was applied. The first results are presented and briefly discussed in comparison with calculation performed on the basis of a simple evaporation model. So it was found that the agreement between measured distributions and calculation could be improved if anisotropy of the fission neutron angular distribution in the center-of-mass system of fission fragments (about 4-8%) is taken into account. At that, there is some surplus of measured yield over calculated at angles near 90^0. On the assumption that these "additional" neutrons are emitted isotropically in the laboratory system their yield could be obtained as less than 5% of the total neutron yield.

Introduction

In spite of the fact that a large number of experiments was devoted to investigation of the properties of the prompt fission neutrons, up to now only the average characteristics of the neutron emission process could be ascertained for sure (average neutron multiplicities per fission event, total neutron multiplicity distribution, the shape of integral neutron energy spectra, an average number of neutrons as a function of energy and mass of the fission fragments.

Experimental studies dedicated to ascertain the mechanism of the prompt neutron emission are limited to spontaneous fission of ^{252}Cf and thermal neutron-induced fission of ^{235}U. The result of these investigations can be formulated as a conclusion that the main part of prompt fission neutrons are emitted from the excited fragments fully accelerated in the Coulomb field of nuclei. At that, a part of fission neutrons originated due the emission mechanism of another nature (for example, emission before and during the rupture of fissioning nucleus or at the initial stage of acceleration of the fragments in the Coulomb field) is varied from 30% down to the total absence of such neutrons [1]. At the same time, only four of these publications (three works dealing with ^{252}Cf and one with ^{235}U) contain numerical information that can be used for independent self-consistent analysis of the experimental data. Using the data of these works, it was ascertained [2] that about 0.4 neutrons per fission can not be described within the framework of a simple evaporation model. The energy spectra of such neutrons consist of two components with average energies 0.9 MeV and 3 MeV. According to the author's opinion, the most likely mechanism of emission of these additional neutrons is emission from fissioning nucleus at the stage of its descent to the scission point (the so-called "pre-scission" neutrons,) [3]. The direct answer to the question on the nature of emission of these additional neutrons could be obtained from the measurements of angular and energy distributions of prompt fission neutrons in the laboratory system in the correlation with the fission fragment characteristics. The measurements of this type were performed recently at the PNPI RAS. In the present paper we present first results of these measurements.

Also, it ought to mention that up to now the energy spectra of the prompt fission neutrons are calculated using the semi-empirical systematic where an absence of information about the mechanism of emission of additional neutrons is compensated by the artificial variation of the nuclear model parameters. This circumstance significantly complicates production of the evaluated data files for the nuclei and energy ranges where experimental data are absent. That is why a new experimental investigation of the mechanism of fission neutrons emission will provide a good basis for future evaluations and enable to increase their accuracy and reliability.

1. Experiment overview

The measurements were carried out at the radial neutron beam N7 of the research reactor WWR-M of the PNPI RAS in Gatchina equipped with a neutron guide 3 m in length. The flux density of neutrons of wavelength $\lambda \sim 1.5$ Å from the neutron guide outlet slit (3×40 mm^2 in cross-section) was $\sim 2 \cdot 10^7$ cm^{-2}·sec^{-1}. 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 stop MWPD surface (coming through its center) in the range from 0^0 to 180^0 in 18^0 intervals. The schematic view of the experimental set-up is shown in Fig. 1.

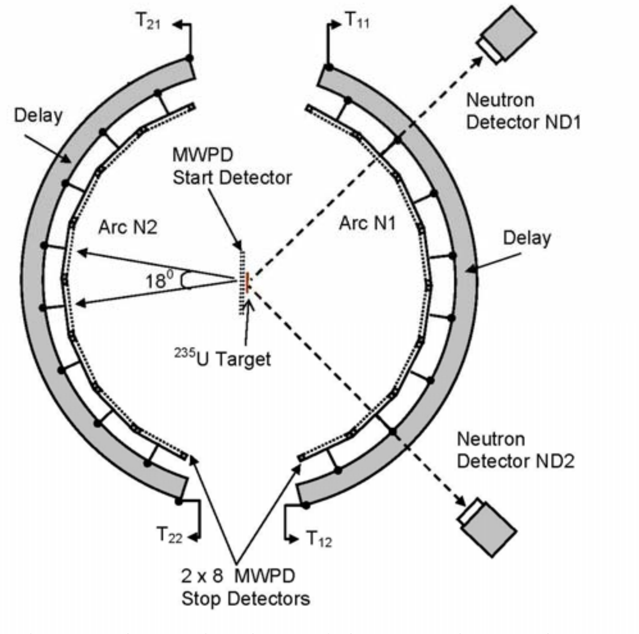

Fig.1. Schematic view of the experimental setup

The neutron beam was coming along the chamber axis normally to the Fig.1 plane. It should be noted that realized scheme of 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 allows 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 90^0 angle between their respective axes at a distance of (47.2±0.20) cm and (49.2±0.20) cm, respectively, from the fissile target. The axes of neutron detectors ND1 and ND2 come through the centers of two stop MWPDs located on the Arc N1. The angular acceptance calculated using the MWPD dimensions of 72×38 mm^2 and the MWPD-target distance of 140 mm: $\Delta\varphi = 28.8^0$ and $\Delta\theta = 15.5^0$. Both neutron detectors were shielded by a cylindrical shield made of 30 mm thick layer of lead and 40 mm thick layer of polyethylene (not shown in Fig. 1). 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 1.2 ns.

The fission fragments were detected by multi-wire proportional detectors (MWPDs) in conjunction with the TOF technique. The 16 rectangular fragment detectors were located in the form of 2 arcs (8 detectors in every one) diametrically opposite each other in the reaction chamber at the operating gas (isobutane) pressure of $4 \div 6$ Torr. A detail description of the experimental set-up and some preliminary results can be found in Ref. [4, 5].

As a result, for 11 fixed angles between neutron and light fragment directions the energy distributions of prompt neutrons emitted from fixed pair of fission fragments were obtained. During data processing, the following corrections were taken into account: for detector efficiency, for neutron detector background, for angular resolution, for the fragment detector efficiency and for incomplete separation of light and heavy group of fission fragments. The detector efficiency was determined by comparing the total neutron energy spectrum (in present experimental set-up it corresponds to the neutron yield integrated over all angles) in laboratory system with the evaluated total neutron spectrum of ^{235}U from Ref. [6].

2. Simple evaporation model

It is well established that the prompt neutrons in low energy fission are emitted mainly from fully accelerated fragments and the yield of neutrons with other emission mechanism may be no more than 30% of the total prompt neutron yield. The wide scatter of the published data on such neutron yield is caused probably by the different shape of the neutron spectrum in the center-of-mass system used in analysis. It arises from the fact that yield of these neutrons is usually determined by comparing experimentally observable variables in the laboratory system with those calculated using known center-of-mass spectra on the basis of the assumption that neutrons are emitted only from accelerated fragments. We used a more constructive approach which consists in obtaining the neutron spectrum in the center-of-mass system without resort to any model representation (the number of neutrons emitted by heavy and light fragments, the neutron spectrum shapes and so on), using only experimental data obtained for small angles relative to the fission direction.

A circumstance of considerable importance is the fact that for such angles it is possible to obtain a neutron spectrum in the center-of-mass system, which is practically unrestricted in the low energy range. Therefore, in this case it is possible to produce not only the response functions but also the neutron yield (in absolute units) which is one more additional reliability criterion of the data obtained.

It is established that the fission fragments have a large angular momenta (~ $7\hbar$ on average), which is usually considered to be normal to the direction of motion of the fragments (for example, Ref. [7]). Due to this fact, the neutron emission anisotropy in the center-of-mass system of fragment may be given by Eq. (2) [8] and the neutron spectrum in the center-of-mass system is related to the neutron spectra in the laboratory system by the following equations:

$$n_{lab}(E_n, \Omega) = (E_n / E_{c.m.})^{1/2} \cdot \varphi(E_{c.m.}, \Omega_{c.m.}) \cdot n_{c.m.}(E_{c.m.}) , \tag{1}$$

$$\varphi(E_{c.m.}, \Omega_{c.m.}) = 1 + A_2 \cdot E_{c.m.} \cdot (3 \cdot \cos^2(\Omega_{c.m.}) - 1) / 2 , \tag{2}$$

where the function $\varphi(E_{c.m.}, \Omega_{c.m.})$ is the angular distribution of neutrons in the center-of-mass system and the parameter $A_2 \geq 0$ defines the value of angular anisotropy; $n_{lab}(E_n, \Omega)$ and $n_{c.m.}(E_{c.m.})$ are the corresponding neutron yields in laboratory and center-of-mass system, per unit energy range and solid angle; E_n, Ω and $E_{c.m.}$, $\Omega_{c.m.}$ are the energy and angle in the laboratory and center-of-mass system, respectively.

At the first stage of calculations, the neutron contribution to the neutron energy spectrum from complementary fragment was calculated (Fig. 2) under assumption that neutrons registered at 0^0 and 180^0 angles relative to the light fission fragment direction were emitted solely by one light and one heavy fragments, respectively. While doing so, the specific energy per nucleon for light and heavy fragments obtained in our measurements was used. These values were taken as $\langle E_L \rangle = 1.046$, 1.025 MeV and $\langle E_H \rangle = 0.471$, 0.476 MeV for $^{233,\,235}$U, respectively. As it can be seen from Fig. 2, for small angles relative to the fission fragment direction the number of neutrons emitted from complementary fragment is a negligible quantity and could leave out of account without any serious consequence in most applications. This assertion becomes stronger if it is taken into account that the calculated spectra were deduced from corresponding neutron spectra in laboratory system above ~1 MeV for light and ~0.5 MeV for heavy fragment.

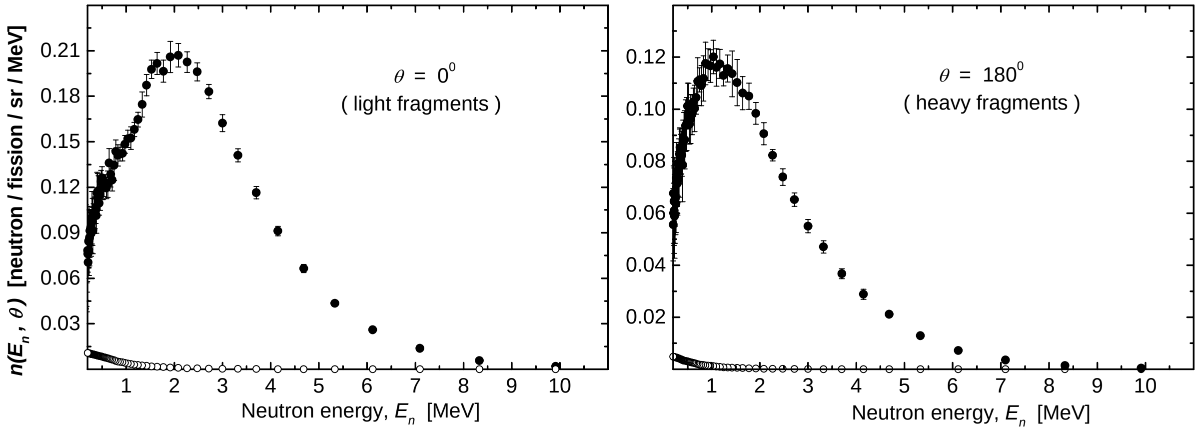

Fig. 2. Spectrum of fission neutrons in laboratory system for ^{235}U(n_{th},f): experiment – full circles with error bars; contribution of fission neutrons from complementary fragment (model calculation) – hollow circles without error bars.

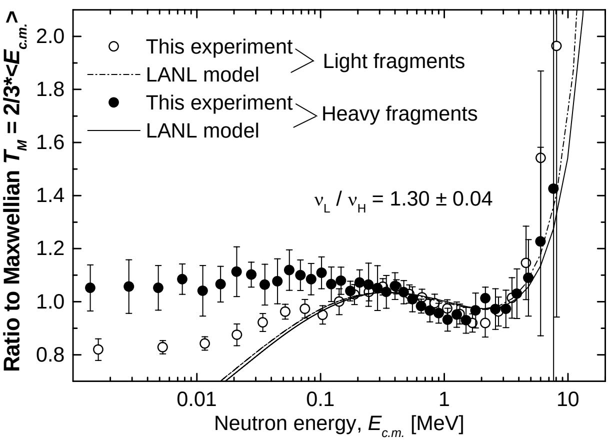

Fig. 3. Ratio of the prompt fission neutron spectrum from light and heavy fragments in the center-of-mass system to the Maxwellian spectrum with adjusted parameters ($\langle E^L_{c.m.} \rangle = 1.17 \pm 0.3$ MeV, $\langle E^H_{c.m.} \rangle = 1.26 \pm 0.2$ MeV).

At the second stage, to obtain the "true" experimental energy spectra of neutrons from the light and heavy fragments for small angles in the laboratory system, the neutron contribution from the complementary fragment was subtracted.

Further, using these energy spectra for small angles in the laboratory system, the neutron energy spectra for light and heavy fragments were obtained in the center-of-mass system. For example, the neutron spectra obtained in the center-of-mass system of ^{235}U are presented in Fig. 3 as a ratio to the Maxwellian in comparison with spectra calculated on the basis of LANL model formalism [9, 10]. In the framework of LANL model, the neutron energy spectrum in the center-of mass system is given by:

$$n_{c.m.}(E_{c.m.}) = (2 \cdot E_{c.m.} / T_m^{\,2}) \cdot E1(E_{c.m.} / T_m)$$

$$E1(x) = \int_x^{\infty} \frac{\exp(-u)}{u}\,du \qquad\qquad T_m = (<E^*> / a)^{1/2} \qquad (3)$$

where $E1(x)$ is exponential integral and T_m is maximum nuclear temperature obtained by the use of the Fermi gas model; E^* and a are the excitation energy and nuclear level density parameter. The Eq.(3) was obtained using standard nuclear evaporation theory under the assumption that the distribution of fission fragment residual nuclear temperature is triangular in shape and the cross section for the inverse process of compound-nucleus formation is constant. The ratio of deduced spectra in the center-of-mass system of fission fragment to Maxwellian is very close to 1 at low neutron energy, but a power index of energy is larger than 0.5 for light fragments and less than 0.5 for heavy fragments.

Finally, the spectra obtained in the center-of-mass system are used for calculation of neutron angular and energy distributions in the laboratory system. These distributions are compared with the experimental distributions.

3. Results and discussion

The number of fission neutrons and their average energy for fixed angles in the laboratory system (obtained experimentally and calculated using an assumption about neutron emission from accelerated fragments) are shown at the top of Figs. 4, 5.

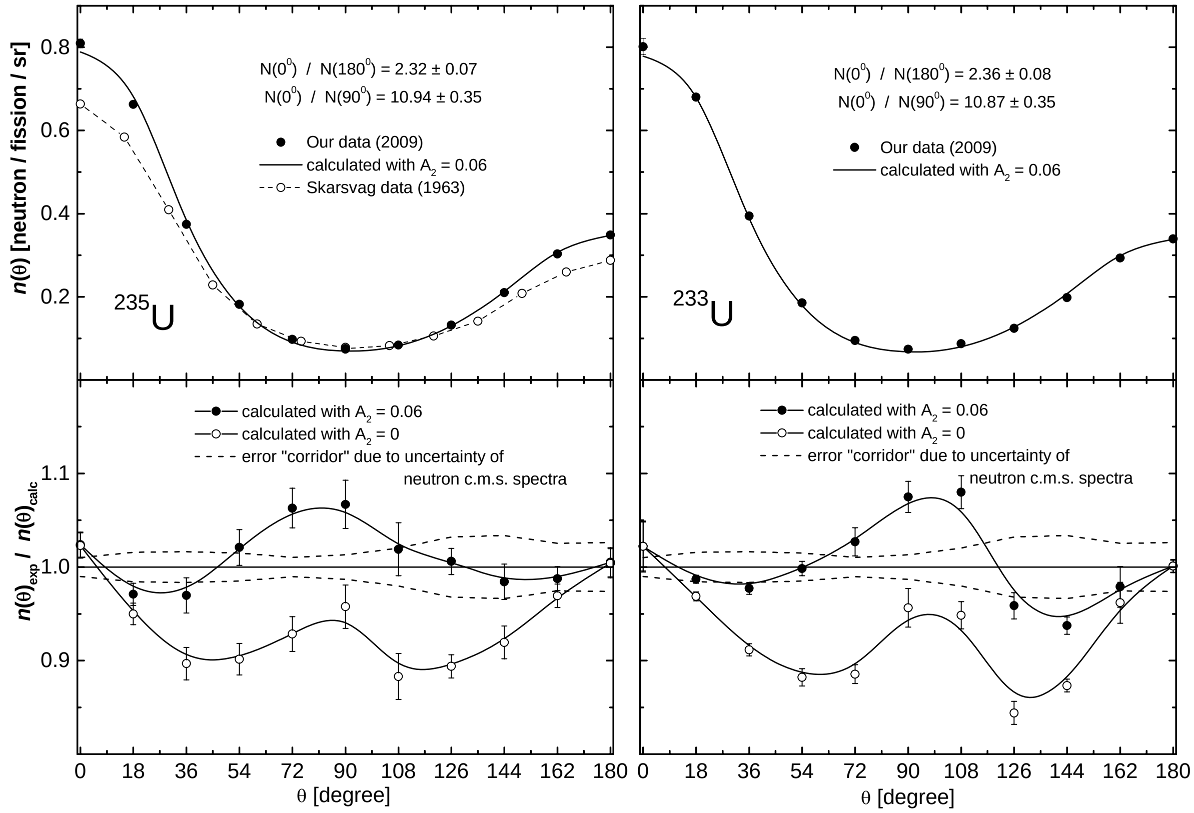

Fig. 4. Fission neutron yield as a function of the angle between neutron flight direction and the direction of motion of the light fragment.

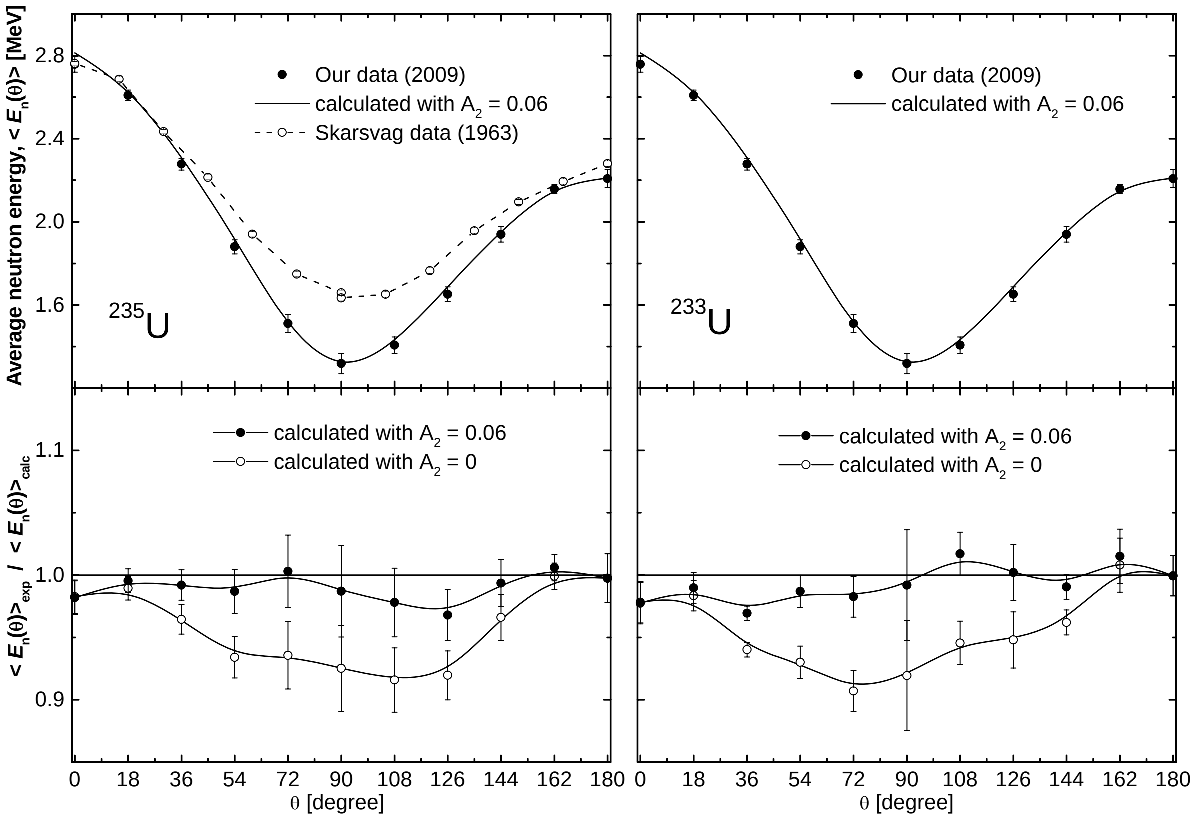

Fig. 5. Angular dependence of the average neutron emission energy in the laboratory system.

Both experimental and model neutron spectra have been compared in 0.2–10 MeV energy range. The errors of the obtained experimental data are comparable with the point's size. In these figures, the experimental results of Skarsvag and Bergheim [11] are also shown. These authors came to a conclusion that about 15% of neutrons are emitted during the fission process itself. In our case, on the whole, the calculated model energy and angular distributions agree rather well with the experimentally obtained distributions.

However, there is a minor distinction which is most clearly demonstrated at the bottom of Figs. 4, 5 where the angular dependence of the ratios of experimentally obtained neutron yield and average energy to calculated values are shown. The error "corridors" presented in these Figs are the standard deviation of ratio obtained for each cycle of measurements to the average ratio obtained for all cycles of measurements. Special attention must be given to the fact that the model calculation gives overestimated values of fission neutron yield and average energy as compared with the experiment for all measured angles. Our model calculation shows that such discrepancy may be related to the presence of anisotropy of the fission neutron angular distribution in the center-of-mass system. Introduction of anisotropy with $A_2 = 0.06$ into the model calculation improves agreement between experimental data and calculation. At that, the total neutron spectra (integrated over all angles) for both investigated isotopes in the energy range above 2 MeV is described with a χ^2 – value close to 1 but in the energy range 0.3 – 2 MeV it shows some surplus of measured yield over calculated (Fig. 6). Under the assumption that these "additional" neutrons are emitted isotropically in the laboratory system, their yield is deduced as about 4% of the total neutron yield.

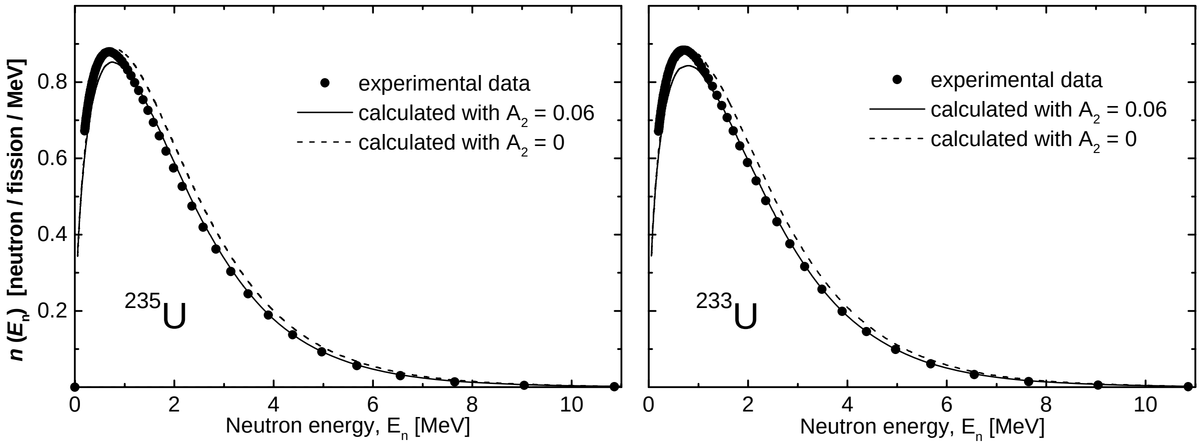

Fig. 6. The total prompt neutron spectra for ^{233,235}U(n_{th}, *f*): experiment – full circles; calculated – lines (see legends).

The shape of the neutron spectrum and the number of neutrons obtained in the center-of-mass system both depend on the fragment velocities. 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, a transition from the velocity distributions of fragments to the model of two fragments with average parameters has only a minor influence (~ 2%) on the total neutron energy distribution [12]. The sensitivity of the model to used average parameters of fission fragments was evaluated by using different value of energies per nucleon for the heavy (0.47-0.49 MeV) and light (1.01-1.04 MeV) fragments. It was found that the ratio value (the bottom of Fig. 4) at 90^0 changes within 4%, that was twice larger than the total experimental uncertainties.

Due to the fact that we are considering the neutron spectrum ratio, our conclusion about the discrepancies between measured and calculated prompt neutron yields, in a systematic sense, is weakly dependent (~3-5% in equatorial emission) on the choice of the standard neutron spectrum used for calibration of neutron detectors. We obtained this value numerically by using various standard neutron spectrum shapes (Watt distribution, LANL model [9], Kornilov – Ref. [6], Maxwellian). To exclude this uncertainty, we are planning to modify slightly our experimental set-up and perform the total neutron spectra measurements relative to that of ^{252}Cf(*sf*) which is a standard of the prompt neutron spectra measurements.

It should be mentioned that the results presented above differ from our preliminary results reported on Ref. [5]. Under the assumption that additional neutrons are emitted isotropically in the laboratory system, their yield was deduced as about 7% (now about 4%) of the total neutron yield. At the same time, the anisotropy of the fission neutron angular distribution in the center-of-mass system of fission fragments was supposed to be equal to 0.04 (at present analysis $A_2 = 0.06$). This difference of the results is connected to the fact that it is necessary to take into account some additional corrections. So, for determination of prompt neutron energy spectrum from the measured time-of-flight spectrum, a relativistic equation is used. Additionally, the following effects have been taken into account: neutron recoil, fragment transmission of the start and stop MWPDs, normalization correction connected to the fact that in the measurements we have the experimental histogram distributions instead of continuous distributions. Also, the angular resolution correction was applied by a more consistent approach than in Ref. [5]. Since the discrepancy between obtained model distributions and experimental data is small, in the first approximation the angular distribution at different energies can be described by model distribution obtained from experimental data without angular resolution correction. The angular resolution can be taken into account by folding model distribution with angular resolution function for each fixed angle θ, which was calculated using the dimensions of fragment and neutron detectors. As a result, model neutron distributions corrected for angular resolution were found. The experimental spectra have been corrected by multiplying each their point by the ratio of corrected model neutron distribution to uncorrected.

The effects of neutron multiple scattering from elements of the experimental apparatus or/and the room have been taken into account by fitting the neutron background shape with linear function. Our calculations demonstrate that other neutron background shapes (constant equal to counting rate before the gamma-peak, constant equal to counting rate on the right wing of the TOF spectrum) leads to practically the same result within the given experimental errors (statistic + systematic) shown in Figs. 4, 5. In other words, the scattered neutrons can't significantly increase the yield at 90^0. Nevertheless the neutron dependencies presented above may be different due to the fact that real scattering effect could be described by function other than applied linear one. In order to estimate an upper limit of the possible deviation, the measurements with shadow cone have been performed recently for ^{233}U and now it is under processing.

4. Future improvements

As concluded above, the yield of neutrons emitted due to the mechanism(s) other than from fully accelerated fragments is small. To obtain surely the yield and to ascertain an emission mechanism of these neutrons, it is necessary to perform a more careful analysis of measured angular and energy distributions of $^{233,\,235}$U(n_{th},f). It can be done in the same manner as we used, but in this case the transformation of neutron spectrum in the laboratory system (at small angles relative to fragments direction) to the center-of-mass system should be performed for each fragment fixed mass and energy. At present, such analysis is in progress.

Since the average number of prompt fission neutrons is well known [13-15], the average number of prompt fission neutrons has been calculated (Fig. 7, 8) using our experimental data with the aim to examine a reliability of the used model. The total average number of neutrons for fission event was found to be about 3% lower than a recommended value [16]. A good agreement is also observed between the average number of prompt neutrons as a function of

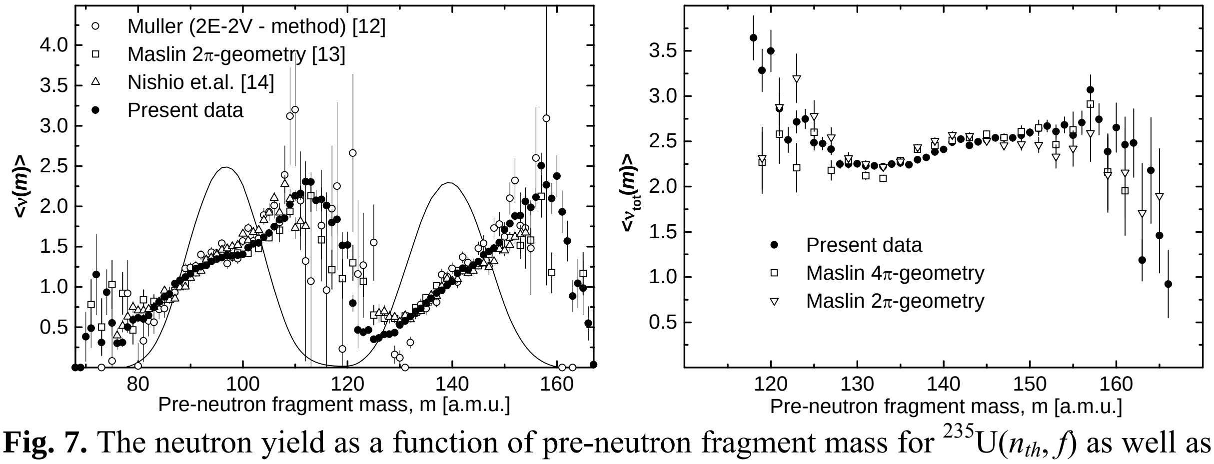

Fig. 7. The neutron yield as a function of pre-neutron fragment mass for ^{235}U(n_{th},f) as well as pre-neutron fission fragment mass distribution obtained by these measurements.

fission fragment mass obtained by this experiment and other authors (Fig. 7, left). It's ought to note that the total number of prompt neutrons as a function of fission fragment mass calculated from measured data on a basis of evaporation model (Fig. 7, right) is practically coincident with this dependence obtained from the direct measurement. At that, the total number of prompt neutrons for fixed mass split in our case was calculated as a sum of numbers of prompt neutrons emitted by light and heavy fragments while in direct measurement it was measured using large gadolinium-loaded liquid scintillation counter in 4π - geometry with registration efficiency about 85%, which is practically not dependent on the fission fragment properties. These facts, probably, are an evidence of absence of large (more than 5%) other mechanism(s) besides the evaporation from fragments.

We observed that for decreasing TKE the neutron yield increases approximately linear as expected by energy conservation (see Fig. 8). This is at least true down to fragment energies of 130 MeV. Most probably, the approximate constancy is simply due to the scattered events since at these energies the count rate is extremely low and a few scattered events will have a big influence. This interpretation is underlined by the results of Maslin [14] and Nishio [15]. Their experiments, probably, suffer from some common systematic drawback which caused below about 150 MeV an "anomaly" in conflict with the energy conservation.

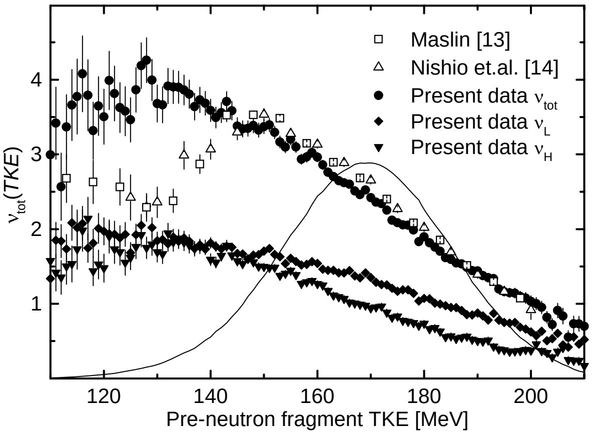

Fig. 8. The neutron yields as function of pre-neutron fragment TKE for ^{235}U(n_{th}, f) as well as pre-neutron fission fragment TKE distribution obtained by these measurements.

Conclusion

The energy spectra and angular distributions of fission neutrons have been measured for thermal neutron-induced fission of ^{233,235}U. A comparison of the measured angular distributions of fission neutrons with calculation, carried out using the model of emission of neutrons from accelerated fragments, enables : 1) to estimate the contribution of "additional" neutrons as not to exceed 5% of total neutron yield in an assumption of isotropic evaporation in the laboratory system; 2) to conclude that the angular anisotropy of the neutron emission in the fragment center-of–mass system ($A_2 = 0.06$) should be included into any calculation of prompt neutron properties in the nuclear fission.

It is necessary to perform a complete self-consistent analysis of neutron energy and angular distributions for each fixed pair of fission fragments which should be analogous to that carried out above under approximation of two fragments with average parameters. Only in that case one has a chance to determine for sure a degree of difference between calculated and measured distributions and in that way to clarify a mechanism of neutron emission. Now this analysis is carrying on.

In the future we are planning to carry out the same experiment for ^{239}Pu(n_{th}, f) and ^{252}Cf(sf). Also, to exclude any uncertainty of neutron detector efficiency, the measurements of the total neutron energy spectra of ^{233,235}U relative to that of ^{252}Cf(sf) are planned.

Acknowledgements

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