NEW DETAILED STUDY OF N-N CORRELATIONS IN THERMAL FISSION OF ²³³U, ²³⁵U AND ²³⁹Pu

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Abstrast

An experiment has been performed to study neutron–neutron angular correlations in slow neutron induced fission of ²³³U, ²³⁵U and ²³⁹Pu. Angular dependences of the number of neutron–neutron coincidences obtained in the experiment were compared with the results of the Monte Carlo calculations for various neutron detection thresholds in the range 490÷2080 keV. It was inferred that 2-5% of the total number of prompt neutrons from these nucleus in the laboratory system were emitted isotropically and may probably be interpreted as neutrons directly associated with the instant of scission of the nucleus. Also as a result of the analysis the energy distributions of this component have been obtained.

Introduction

Recently new precise measurements of the angular dependence of neutron-neutron coincidence count rates in thermal fission of ²³³U, ²³⁵U and ²³⁹Pu were performed in PNPI on the reactor WWR-M. The purpose of these experiments was to find more correctly the moment and place of neutron emission.

It is known that most of prompt neutrons accompanying the fission process are evaporated from the fragments fully accelerated due to Coulomb interaction [1-3]. However, some neutrons can appear directly at scission of nucleus. In contrast to alpha-particles scission neutrons do not focus by the Coulomb field. To distinguish them from the neutrons emitted after fully acceleration of fission fragments, their difference in the angular and energy distributions is normally used. In addition to neutron-fragment angular correlation the angular dependence of n-n coincidences can be also applied to determine the emission conditions of prompt fission neutrons [4-7].

Experiment

The Fig.1 presents the scheme of the experimental set-up for measurements of n-n coincidences in the neutron induced fission of ²³³U, ²³⁵U and ²³⁹Pu. Here direction of the neutron beam from neutron guiding system is shown. For prompt neutron pairs registration we have used two identical stilbene crystals with PM tubes (Hamamatsu R13107). These detectors were surrounded by polyethylene and lead. The distance from the target to each stilbene scintillator was about 51 cm. Angles between the detectors with respect to the source could be easily varied and in such a way we can get the angular dependence of neutron-neutron coincidence count rate for two detectors from 30 to 180 degrees between them.



Fig.1. Experimental setup for measurements of n-n coincidences in the neutron induced fission.

Stilbene scintillators allow neutron and gamma-ray pulse shape separation. For this purpose it is possible to use the known method of simultaneous measurement of the total charge and the part of this charge in the tail of the pulse (so-called slow component, see Fig.2).



Fig.2. *Simultaneous measurement of the total charge and the part of this charge.*



Fig.3. Two-dimensional distributions for the parameters of PM signals: CDC_0 (total integral) versus CDC_1 (slow component). The signals characterized by larger CDC_1 with identical CDC_0 correspond to the detected neutrons.

An example of two-dimensional distribution of these quantities is shown in the Fig.3, where it is seen that neutrons and gamma-rays are reliably separated down to the lower electron detection threshold.

The next Fig. 4 shows the time-of-flight spectrum for all detected events, including two neutrons and two γ -rays, as well as registrations of a neutron by one detector and a γ -ray by another.



Fig.4. Time-of-flight spectra for two neutrons and two γ -rays, as well as registrations of a neutron by one detector and a γ -ray by another.

Such form of time-of-flight spectrum on the left side is due to the fact that a delay line has been installed. In this experiment we are interested only in the count rate of neutron–neutron events as a function of the angle between the detectors. In this experiment the direction of motion of fission fragments is not fixed; therefore, the number of neutron–neutron coincidences for a particular angle between the neutrons is averaged over all orientations of fission axis and all neutron energies above the energy threshold.

Evidently, to get true parameters of scission neutrons it is extremely important to obtain the experimental angular distributions free from systematic errors. Special attention in this work was paid to the study of possible effects that can distort the experimental distributions. One such effect is the neutron scattering in the fission source and other surrounding materials. This effect leads to a flattening of the investigated curve and can simulate the number of additional events, which falsely can be considered as scission neutrons. Second effect, which certainly influence on investigated angular distribution is "cross-talk" between neutron detectors.

We have performed the series of control experiments together with their cross-check by Monte-Carlo simulations. For these experimental tests we have used also the neutron source with a composition of Pu+Be, which gives only one gamma-quantum per neutron. It allows us to have single neutron events and we can see the probability to have coincidence of one neutron registration in both detectors. All of this allowed us to estimate the experimental data uncertainty, which can be added to statistical errors, on the level of 1%.

Calculations

The task of the experimental data description in the modeling representation was limited to the determination of the angles between the neutron pairs belonging to the same fission event. As a result, in accordance with the experiment, we should obtain only the total number of pairs of neutrons recorded for each angle between them. But these values are determined by the energy and angular characteristics that can be attributed to each neutron at the moment of its emission. To find these parameters the Monte Carlo method was used. In process of calculation pairs of neutrons from 10^8 fission acts were analyzed.

At the first stage of this calculation it was simulated neutron emission The model was based on the well-founded assumption that main part of neutrons evaporates from fully accelerated fragments $(\overline{v}_L + \overline{v}_H)$. Additionally the emission of small part of neutrons was assumed just in the moment of nucleus rupture (\overline{v}_{sci}) . The relative share of scission neutrons $(r = \overline{v}_{sci}/\overline{v}_{tot})$ was considered as a fitting parameter of calculation and was adjusted so as to obtain the best agreement between the calculated and experimental angular distributions for count rate of the neutron pairs.

It was assumed that the angular dependences for the neutron pairs in the two groups will have a different shape, and variation of the contributions corresponding to these two components will enable us to determine the best ratio "r" due to comparison of calculated angular dependence with experimental one. Differentiation of the correlation forms is associated with a difference in the final angular and energy characteristics for these groups of neutrons. This exists due to the presence of additional speed for one of them, namely the speed of the fragment. This fragment velocity should be added to the velocity of a neutron in the fragment's center of mass system.

During the calculation the so-called model of the two fragments was used. This means that each of the two groups of fragments (light or heavy) has one and the same average velocity obtained due to Coulomb repulsion. These average velocities were weighted due to the fact that the amount of vaporized neutron depends on the mass of the fragment [8]. As a result, the speed of light or heavy group of fragments was selected in accordance with the kinetic energy per nucleon (see Tab.1): Table 1

²³³ U	²³⁵ U	²³⁹ Pu
E _{vL} =1.033 MeV	E _{vL} =1.012 MeV	E _{vL} =0.995 MeV
E _{vH} =0.471 MeV	E _{vH} =0.474 MeV	E _{vH} =0.511 MeV

We carefully followed to provide that all integral characteristics of the calculation for the fission neutrons coincided with the parameters known from a number of other experiments, for example, partial neutron multiplicity, their variance and covariance [9]. In order to satisfy these values, the standard procedure NORMCO [10] for obtaining random numbers corresponding to the two-dimensional normal distribution was used. Considering, however, that in our task we are dealing with the neutron multiplicity distribution, which has not a real Gaussian form, but with the "cut off" of negative values (the number of neutrons can not be

negative) and their numbers are also discrete, we conducted additional testing to more accurately selection of the parameters that should be used in NORMCO procedure.

The shape of the energy spectra of neutrons in the center of mass of the fragments was assumed to be Maxwellian form

$$N_{L,H}(E) \sim \frac{\sqrt{E}}{T_{L,H}^{3/2}} \cdot \exp(-E/T_{L,H})$$

with fixed temperature for light and heavy fragment, respectively. This type of spectrum is due to evaporation by each fragment more than one neuron per fission [11]. These temperature parameters correspond to that obtained in the experiment on n-f angular and energy correlations [9].

We took into account that the angular distribution of neutrons in the fragment system is different from an isotropic due to the presence of a large primary fragment spin. The anisotropy of the angular distribution was determined according to the calculations performed in the work [12] using the emission energy of each neutron in fragment system.

Since it was assumed that the share of "scission" neutrons will not be very large and, therefore, can not have a cascading nature, their energy distribution was described by Weisskopf spectrum

$$N_{L,H}(E) \sim \frac{E}{T_{sci}} \cdot \exp(-E/T_{sci})$$

and its temperature $T_{sci} \equiv T_{W}$ was the second fitting parameter of this calculation.

In this experiment the neutron energy was not measured. But the results of angular dependence integrated over all neutron energy values were connected with seven different energy thresholds. This allowed us to get some information not only about the contribution of



Fig.5. Geant4 simulation of the detectors efficiency (a) and angular response function (b).

additional component but also concerning its energy distribution. Before our Monte-Carlo calculations the detectors efficiency for all neutron energy thresholds and angular response function were obtained using Geant4 simulation (see. Fig.5).

Thus we had two free parameters (the contribution of scission neutrons "r" and their temperature T_{sci}) to fit experimental data of n-n angular distributions measured for seven different energy thresholds.

It should be noted that the sensitivity of calculations to the temperature parameter T_{sci} at the lower neutron energy threshold is insufficient. In this case a small variation in the temperature of Weisskopf spectrum does not leads to a big change of that part of "scission" neutrons, which is used in the analysis of pair correlations to determine the proportion "r" of the two groups of neutrons. But such conditions are the most suitable for the determination of the first parameter, i.e. the relative contribution of "scission" neutrons in the total amount of fission neutrons. The difference in the spectral composition of "scission" neutrons and



Fig.6. Experimental data and calculated results of *n*-*n* coincidences for the target of ²³⁹Pu (for the lowest energy threshold)



Fig.7. Experimental data and calculated results of *n*-*n* coincidences for the target of ²³⁹Pu. (for seven energy thresholds)

neutrons evaporated from fully fragments accelerated is most "tails" pronounced in the of distributions, i.e. this inequality is affected more at the highest neutron energy threshold. As a result it is possible to separate the searching of two parameters. Initially we can find the first of them at the lowest energy threshold, and then at the upper of the existing thresholds and with a fixed share of "scission" neutrons we can specify the form of the energy distribution, that is, to determine the temperature of Weisskopf spectrum.

The Fig.6 shows the angular dependence of n-n coincidence count rates for the target of ²³⁹Pu. These experimental data correspond to the lowest neutron energy threshold, namely E_{th}=490 keV. Here and in all figures below we present only statistical error bars. The scattered neutrons influence on these experimental data and they include also cross-talks component. The dashed line in this figure corresponds to our calculations for the angular distribution of n-n coincidences without taking into account of cross-talk component, but the scattering of neutrons and the angular resolution were already included in our model during the efficiency determination. If we take into account the presence of cross-talks then the calculated values of n-n coincidences at small angles between the detectors will rise. These corrected data were marked by the band between two solid curves. As we can see the experimental data for the target of ²³⁹Pu in case of the lowest energy threshold can be rather well described by the Monte-Carlo simulation with 4% of scission neutron contribution. The dash-dot line in the Fig.6 shows the result of Monte-Carlo calculations for the same neutron energy threshold but without the addition of scission neutron component. Note that the influence of cross-talks was taken into account. Obviously it is seen that this description is less suited to the experimental data.



Fig.8. Experimental data and calculated results of n-n coincidences for the target of ^{235}U (for seven energy thresholds).



Fig.9. Experimental data and calculated results of *n*-*n* coincidences for the target of ²³⁵U (for seven energy thresholds).

The Fig. 7 demonstrates that other six n-n angular distributions for this target related to different neutron energy thresholds can be also rather well described with the same fitting parameters that were used for the lowest energy threshold. It is necessary to mention that the model calculated curves were normalized to experimental data with one common coefficient. Successful descriptions of all experimental relations show us in addition that the correct efficiency of neutron detection in these calculations was used.

The same is truth for the targets of ²³³U and ²³⁵U. It was also possible to describe all experimental data for seven energy thresholds with the addition to neutrons evaporated from fully accelerated fragments only a small fraction of scission neutrons. In case of ²³³U this contribution was 5% and for ²³⁵U it was necessary to add only 2% (see Fig.8 and Fig.9).

It is necessary to mention that these results are very close to that obtained by A. Vorobiev [13] with coauthors due to neutron fragment correlation (see Tab. 2). If neutron spectrum corresponds to Weisskopf distribution, then $\langle E_{sci} \rangle = 2 \cdot T_{sci}$.

Table 2

_	$r = \overline{v}_{sci} / \overline{v}_{tot}$		$< E_{sci} >, MeV$	
Target	This work	A.Vorobiev et al.	This work	A.Vorobiev et al.
²³³ U	$(5.0 \pm 1.5)\%$	\leq 4%	2.0 ± 0.2	2.1 ± 0.2
²³⁵ U	$(2.0 \pm 1.5)\%$	≤ 3%	1.8 ± 0.2	1.8 ± 0.2
²³⁹ Pu	$(4.0 \pm 1.5)\%$	\leq 7%	1.8 ± 0.2	1.8 ± 0.2

Conclusions:

- Precise measurements of the angular dependence of n-n coincidence count rates in ${}^{233}U(n,f)$, ${}^{235}U(n,f)$ and ${}^{239}Pu(n,f)$ were performed.
- All possible experimental effects which can distort the experimental distributions are analyzed and taken into account with accuracy ~1%. (Control experiments and GEANT4 simulation have been done).
- The experimental data can be better described if we use besides neutrons evaporated from fully accelerated fragments, the additional component.
- These distributions can be described if all neutrons of additional component are supposed isotropically emitted in the laboratory system. This component assumed to be Weisskopf energy spectrum with $T_{sci} \approx 1$ MeV. These neutrons can be attributed as scission neutrons.
- The results obtained due to n-n angular distributions are practically coincided with those obtained in neutron-fragment correlations.

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