International Conference on Nuclear Data for Science and Technology 2007
DOI: 10.1051/ndata:07735

New experimental and theoretical results for the ^{235}U fission neutron spectrum

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Abstract. The prompt fission neutron spectrum (PFNS) was measured at 0.5 MeV incident neutron energy. An angular anisotropy in the prompt neutron emission was found. The neutron yield is ∼10% higher and the average energy ∼80 keV smaller at 90 degree than at 120 degree neutron emission angle. The spectral difference diminishes with increasing prompt neutron energy. The experimental data were successfully described with the semi-empirical "three sources" model.

1 Introduction

Neutron multiplicity, mean neutron energy and emission spectrum are crucial nuclear data with very stringent requests on their accuracy. Up to now for some important isotopes like ^{235}U, ^{239}Pu a rather high accuracy of ∼0.2% for neutron multiplicity and of ∼3% for the average energy of prompt neutrons was reached for low incident neutron energy. A theoretical model, developed in 1982 [1], has been adjusted to describe the experimental PFNS of ref. [2]. It was based on the well known assumption that all fission neutrons are emitted from accelerated fragments. This model or its recent modification [3] has been tested with integral experiments [4] and reasonable well predicts benchmark experiments [5].

However, between 1980 and 2000 new PFNS measurements have been carried out [6–10]. These microscopic experiments being in reasonable agreement with each other result in an average energy of the promptly emitted fission neutron smaller than required by benchmark experiments, macroscopic experimental data and the predictions by the Madland-Nix model [1]. This problem was highlighted in ref. [11] and stimulated a new interest for PFNS investigations. Since this is a major issue for evaluations, several subgroups of the OECD-Nuclear Energy Agency (NEA), Working Party on International Evaluation Cooperation (WPEC) requested new high precision investigations of prompt neutron emission in neutron induced fission of ^{235}U [12].

2 Experiment

The measurements were carried out at the 7-MV Van de Graaff accelerator of the Institute for Reference Materials and Measurements in Geel, using the fast neutron time-of-flight technique. A pulsed proton beam of about 1 ns FWHM at a repetition rate of 1.25 MHz and an average current of 0.5 μA was used. Mono-energetic neutrons of 0.52 MeV average energy were produced using the ^{7}Li(p,n) reaction with a LiF target of 596 μg/cm^2 thickness. The metallic ^{235}U sample (93.15% enrichment) weighting 161.28 g consists of 10 concentric rings canned in an aluminium container of 0.2 mm wall thickness. The outside diameter of the rings is 3.00 cm, the inside diameter 2.12 cm and the height 2.41 cm. A similar sized lead sample was applied for background measurements. The sample was placed at 5.0 cm from the neutron producing target at a 0 degree angle with a low-mass holder. The actual neutron energy range due to the LiF target thickness and the geometrical factor extended from 0.41 MeV to 0.58 MeV.

Two NE213 equivalent liquid scintillation detectors of 10.2 cm diameter and respectively 5.1 cm and 2.5 cm length, coupled to XP4312/B photomultipliers were used in the measurement. Both detectors were placed in large collimating shields made of lithium carbonate, paraffin, and lead. The distances between the center of the scintillation detectors and the sample were 273.0 ± 0.5 cm for the detector at 90 degree and 240.0 ± 0.5 cm for the one at 120 degree relative to the proton beam axis. An additional copper cylinder was placed between target and detectors to reduce the direct gamma-ray and neutron flux from the target into the detectors. The traditional neutron-gamma-ray discrimination technique was applied for gamma-ray reduction. A third detector based on a PilotU plastic scintillator (diameter 3 cm, height 2 cm) mounted to an XP2020Q photomultiplier was applied as a monitor of the proton pulses.

The detector efficiencies were measured for both neutron detectors simultaneously, relative to the ^{252}Cf standard spectrum. For this purpose a ^{252}Cf source inside a low mass, fast ionization chamber developed at IRMM [13] was put at the place of the U-sample keeping the same geometry as during the experiment. The total time resolution (FWHM) for a neutron threshold of about 0.5 MeV, as estimated from the ^{252}Cf efficiency measurement, is 1.5 ns for the detector at 90 degree and 1.3 ns for the one at 120 degree. The gamma-ray suppression was about 1/200 for both detectors. For each run data were collected in list mode and transformed off-line to an energy scale. The energy distributions were corrected for detector efficiencies, for neutron multiple scattering in the sample, and for time resolution. A detailed description of the experimental procedure will be published elsewhere [14].

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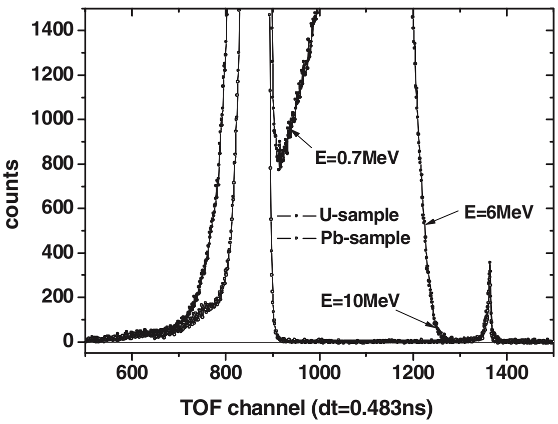

Fig. 1. A TOF spectrum of the U- and Pb-sample runs after subtraction of the time independent background.

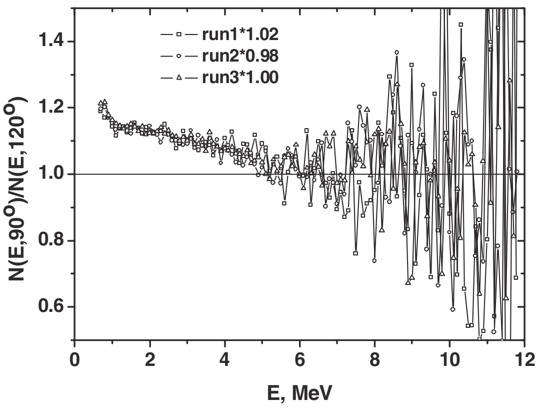

Fig. 2. The spectral ratio N(90°, E)/N(120°, E).

Table 1. Total amount of fission neutrons collected in each run, run duration, and average neutron energies.

Name	Neutrons	T, h	⟨E⟩, MeV 90-deg	⟨E⟩, MeV 120-deg
Run1	$8.0 \cdot 10^5$	37.0	2.018	2.087
Run2	$1.1 \cdot 10^6$	42.1	2.022	2.103
Run3	$8.5 \cdot 10^5$	39.2	2.014	2.092

3 Results and discussion

The time-of-flight spectra for the uranium and lead runs for the detector at 120 degree are given in figure 1. Both spectra were normalized to the elastic peak. For both detectors no time dependent peculiarities were found in the background run in the energy range of interest from 0.7 MeV to 12 MeV. The energy spectra were calculated for the full data set as well as separately for each of the three independent runs. A Maxwellian spectrum was fitted in the energy range of 0.71.5 MeV and 9–11 MeV to the measured spectrum and the calculated values were used for the extrapolation to zero and to 20 MeV.

The average energies for both detectors and for each independent run are given in table 1. An obvious difference in the average neutron energy at the different angles exists. Possible sources of uncertainty on the average energy are the calculation of the zero-time, the detector efficiency (standard neutron spectrum), and any of the other corrections mentioned above. For example, a 1 ns time shift, changes the average energy by 60 keV for our experimental conditions. The analysis of the prompt gamma-ray position allows us to estimate an uncertainty of the zero-time determination to less than 0.4 ns. Having in mind this value and the spread of the data in table 1 we concluded that for our conditions the average energy was estimated with an accuracy of ±0.020 MeV. Finally, for the total data set ⟨E⟩ = 2.018 ± 0.020 MeV for the 90 degree detector and 2.094 ± 0.020 MeV for the 120 degree detector. In general, the spectrum at 120 degree is closer to a Watt distribution whereas the spectrum at 90 degree resembles a Maxwellian shape. The ratio N(90°, E)/N(120°, E) for each measured run is shown in figure 2. Small absolute differences (as given in the inset of figure 2) between runs may be explained with the uncertainty of the sample position and proton beam position of ±2 mm. This conclusion was confirmed by MCNP simulations. Below 5 MeV a definite difference between the spectrum taken at 90 degree and at 120 degree is visible. The average ratio for the energy interval 6–10 MeV is ⟨R⟩ = 1.022 ± 0.015.

An angular dependence of the fission neutron emission was already discussed in refs. [15–19]. Most of these experiments were carried out at ~2 MeV incident neutron energy, which diminishes the effect. It seems that due to this fact and rather large experimental errors (10–15%) the final conclusion was that a possible angular anisotropy is small. Neither the spectrum shape nor an average energy dependence was discussed. No direct evidence of the discussed effect was found in ref. [20]. The measurement was made at an angle of 120 degree. Extrapolation to zero neutron energy using a Watt spectrum resulted in a much better value for the neutron multiplicity than with a Maxwellian distribution. In refs. [15, 16] the PFNS was measured at 0.4 MeV for ^{235}U at angles of 45, 90 and 135 degree. However, the data were not analysed separately. All three spectra were summed and the spectrum shape fitted with a Maxwellian distribution. The T parameter based on the fit in the energy range 0.55–7 MeV gave an average energy ⟨E⟩= 1.5 T= 2.06 ± 0.05 MeV. We can assume that the present 120 degree spectrum corresponds to the 45 and 135 degree runs of ref. [15], and can construct the spectrum $N(E) = 0.33 \cdot N_{90}(E) + 0.67 \cdot N_{120}(E)$. The resulting Maxwellian T-parameter from this constructed spectrum, fitted in the energy range 0.7–7 MeV, gives as average energy $\langle E \rangle = 1.5\ T = 2.055$ MeV, in perfect agreement with the data of ref. [15].

The PFNS at 90 degree agrees with all previous measurements [2,6–10] (see fig. 3), available in numerical form within an accuracy of smaller 10% in the prompt neutron energy range from 1–7 MeV. Even more, our data agree inside the error bars with the data of refs. [6,7] in the energy interval 0.7–5 MeV, with the data of Johansson et al. [2] in the energy interval 3–12 MeV, and with the data of Staples et al. [8] in the energy interval 1–8 MeV inside our error bars.

The fission neutron spectra for ^{235}U in refs. [2,8–10] were measured at 90 degree relative to the incident neutron beam. The measurement angle used in refs. [6,7] is unknown. Both

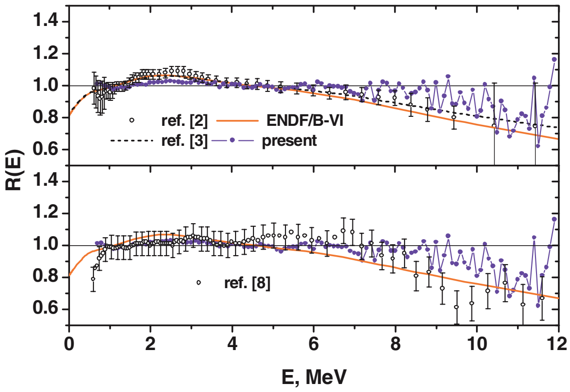

Fig. 3 (a). The present 90 degree data (full symbols) compared to literature data (open symbols) [2,8] and plotted as a ratio to a Maxwellian dsitribution with $\langle E \rangle = 2.018$ MeV. The data of ref. [2] was corrected for multiple scattering in the sample.

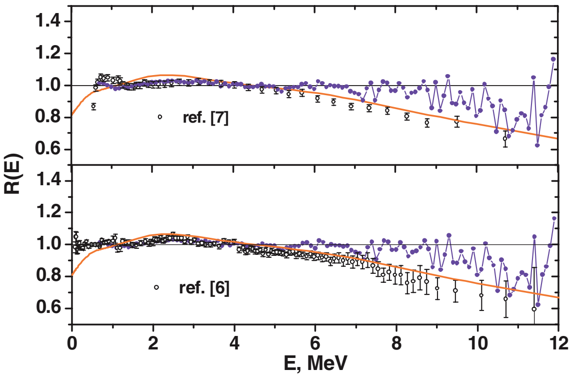

Fig. 3 (b). The same as in figure 3 (a), but compared to refs. [6,7].

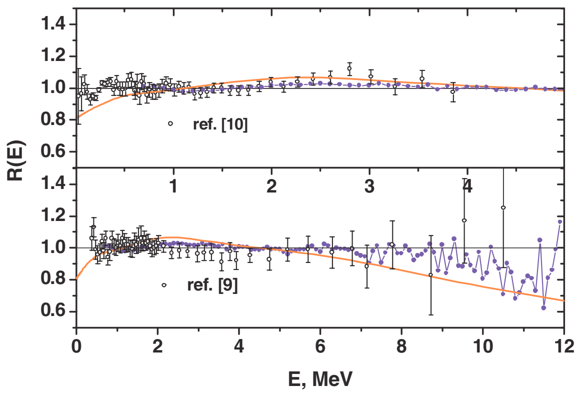

Fig. 3 (c). The same as in figure 3 (b), but compared to refs. [9,10].

thermal [6,7,9] and about 0.5 MeV [2,8,10] incident neutron energies were investigated. Hence, one may conclude that our result at 90 degree agrees with all previous measurements.

The present PFNS is systematically higher by ~10–20 % in the high energy range >6 MeV compared to some of the literature data and the smooth curve predicted by the theoretical model (fig. 3(b)). The following peculiarities should be taken into account when we are discussing this point.

1. The total energy release is ~25 MeV, so each fragment gets on average ~12 MeV only. The high energy part of the fission neutron spectrum (>5 MeV) can not be described by an evaporation process of neutrons from excited fragments only, but will be modulated by the fission fragment yield at low kinetic energy and by the energy release (Q-value) available for a given mass split.
2. A larger difference is visible for the data measured with ionisation chambers (refs. [6,7]). It may be connected with the experimental method itself. Rather thick fissile layers (low counting efficiency) were applied in the ionisation chamber to provide reasonable count rates. Hence, some fission fragments with low kinetic energy (high excitation) may be lost, leading to a systematic difference in the neutron spectrum which increases with neutron energy. A special effort was made in ref. [9] to minimize this effect, which may explain the much better agreement with the present result in the high energy part of the spectrum (see fig. 3 (c)).

In general one may conclude that the present experiment at 90 degree together with nearly all other available numerical data does not confirm the Madland-Nix model [1] or its latest modifications [3]. At presence no model can explain the experimental finding of an anisotropic PFN emission. We have to assume that an additional mechanism like an emission at the scission point (SCN) has to be incorporated.

4 Theoretical model

If the assumption of SCN emission is true fission neutrons should be emitted from three sources:

1. Neutrons from fragments after fission of the compound nucleus A_c:

$$N_A(E) = (1-\alpha)\cdot W_A(E), \tag{1}$$

where α is the share of scission neutron emission and W_A is the spectrum which describes the neutron emission from accelerated fragments;
2. Neutrons from accelerated fragments after fission of the nucleus A_c-1, which is formed after the emission of one SCN:

$$N_{A-1}(E) = \alpha\cdot(\nu-1)\cdot W_{A-1}(E)/\nu. \tag{2}$$

3. Scission neutrons itself:

$$N_{scn}(E) = \frac{\alpha}{\nu}\cdot E\cdot\left(\frac{\varsigma}{T_1^2}\exp\left(-\frac{E}{T_1}\right)+\frac{1-\varsigma}{T_2^2}\exp\left(-\frac{E}{T_2}\right)\right), \tag{3}$$

where ζ is the share of the low energetic component.

The spectra W_{A}, $W_{\mathrm{A}-1}$ were calculated with a Watt distribution for light and heavy fragments. The ratio of the neutron multiplicity for light and heavy fragments was $\nu_l/\nu_h = 0.58$. Temperature parameters were found on the basis of the Fermi-gas relation and the thermal-equilibrium assumption with an additional correction of cor = 0.9 for the excitation of the heavy fragment $U_{\mathrm{h}} = U_{0\mathrm{h}}\cdot$cor, ref. [21]. The level density parameter was calculated as $a = A/c$.

The $N_A(E)$, $N_{A-1}(E)$ spectra should have an isotropic distribution. The high energy component of $N_{\mathrm{scn}}(E)$ according

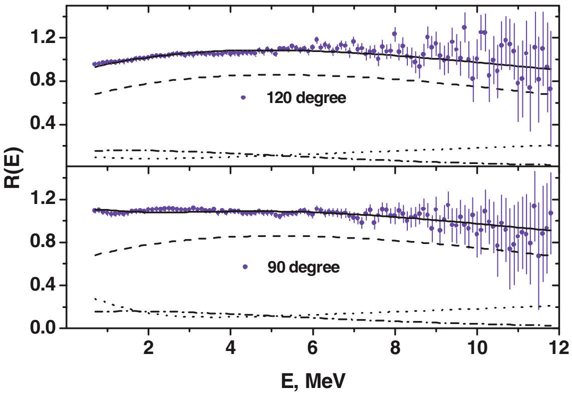

Fig. 4. Spectral description with the "three sources" model. The total spectrum (solid line), $N_A(E)$ – dashed, $N_{A-1}(E)$ – dashed-dotted, $N_{scn}(E)$ – dotted lines, respectively.

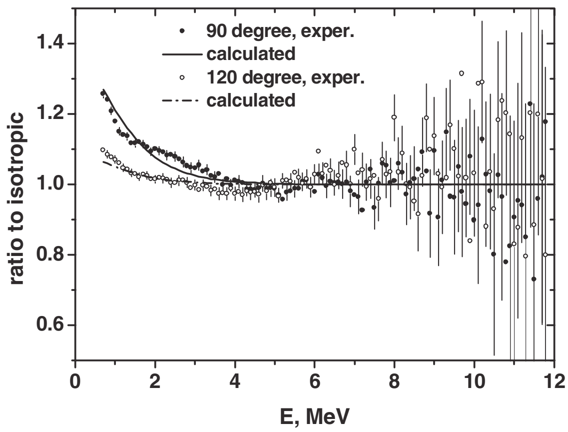

Fig. 5. Ratio of the measured (symbols) and calculated (lines) spectra to the isotropic component.

to the present result should be also isotropic. So, an angular effect may be only connected with the low energy component of the SCN. Applying the above equations one may construct the isotropic part:

$$N_{iso}(E) = 0.5 \cdot (1-\alpha) \cdot W_A(E) + 0.5 \cdot \frac{\alpha}{\nu} \cdot \left(E\frac{1-\varsigma}{T_2^2} \exp\left(-\frac{E}{T_2}\right) + (\nu-1) \cdot W_{A-1}(E) \right), \quad (4)$$

and the anisotropic part:

$$N_{ani}(E) = \frac{(1-|\mu|)^p \, (p+1)}{2 \cdot \nu} \cdot E \cdot \frac{\varsigma}{T_1^2} \exp\left(-\frac{E}{T_1}\right). \quad (5)$$

The equation for $N_{\rm scn}$(E), and the corresponding parameters ζ, T_1, T_2 were taken from ref. [22] introducing minor changes: $\zeta = 0.40$, $T_1 = 0.4$ MeV, $T_2 = 1.5$ MeV, $\langle E_{\rm scn} \rangle = 2.08$ MeV. Parameters α, and c were fitted by the least-squares method. The best result for both angles was found with $\alpha = 0.25$, $p = 2$, and $c = 8.0$, $\chi^2/\text{n} = 1.14$ (90 degree) and 0.98 (120 degree). The different components calculated with equations (1–3) for the 90 degree and 120 degree angles are presented in figure 4 as a ratio to Maxwellian distribution. The ratio of the experimental data and the calculated spectra to the isotropic component equation (4) are given in figure 5. One may conclude that the experimental data was described with the suggested semi-empirical model reasonably well.

The measurement will be continued covering more angles to verify the effect and estimate the full angular dependence. It would be very interesting to look also as a function of incident neutron energy, for different fissile targets and investigate the relation between neutron and fission fragment anisotropy.

Two of authors (N.K., I.F.) would like to thank the European Commission for a fellowship during which this work was performed. We would also like to thank the Van de Graaff accelerator team G. Lövestam, T. Gamboni, C. Chaves de Jesus, W. Geerts and R. Jaime Tornin for the high-quality pulsed beam. We are also indebted to A. Moens from the target preparation group of IRMM for the high quality U-target.

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