Left-right and angular asymmetry of fission neutron emission

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Following a recommendation of the NEA Working Party on Evaluation cooperation (WPEC) [1] the prompt fission neutron spectrum (PFNS) was measured at ~ 0.5 MeV incident neutron energy.

1. Experimental procedure and results

Three experiments were carried out at the 7 MV Van de Graaff accelerator of the IRMM in Geel, Belgium, using the fast neutron time-of-flight technique. A pulsed proton beam of about 1.0 - 1.5 ns FWHM at 1.25 - 2.5 MHz repetition rate and 0.2 - 0.8 μA average current was used. Mono-energetic neutrons of 0.52 MeV average energy were produced using the ^{7}Li(p, n) reaction. A metallic ^{235}U sample (93.15 % enrichment, 161.28 g) and a similar sized lead sample were applied for foreground and background measurements, respectively.

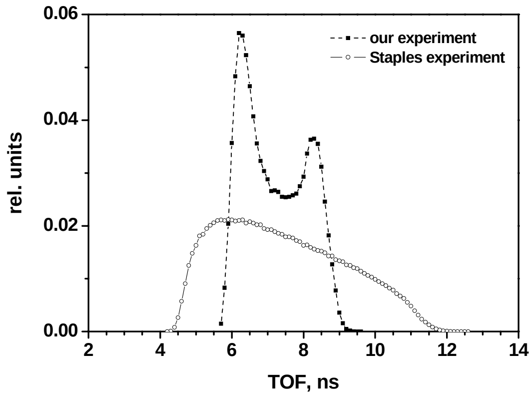

Fig. 1. Comparison between the TOF distribution of the input neutrons inside the sample for the present experiment (full symbols) and the one of Ref. [6] (open symbols). The data are from a Monte Carlo simulation.

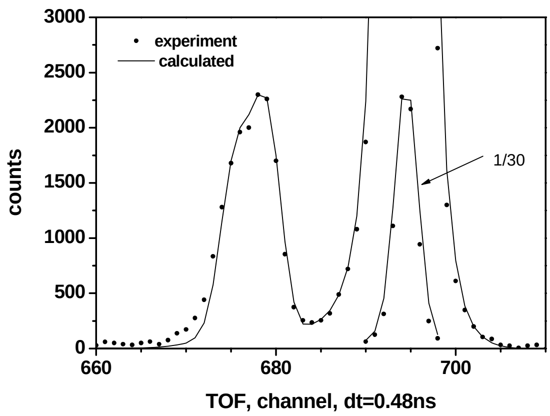

Fig.2 Prompt-gamma ray peaks measured by detector R90 (Jan08 run). The threshold is 1.2 MeV. The channel width is 0.47 ns. The convoluted result is also given. The target gamma-rays (left) peak gives the detector resolution and proton pulse width. The prompt fission gamma-rays give the total time resolution including neutron spread inside the sample (Fig. 1).

In a first run (Jul06) an angular dependent effect was found. The neutron yield is ~ 10% higher and the average secondary neutron energy ~ 80 keV higher at 120 degree compared to 90 degree. The result was discussed at the Nice ND2007 conference [2]. This unusual finding stimulated new investigations to verify and to estimate the nature of this effect. In a second experiment (Apr07) we used three identical neutron detectors at a flight path of 2.24 ± 0.01 m placed at 90, 150 and 120 degrees. The distance from the neutron production target to the sample was ~ 8 cm.

In a third experiment (Jan08) the same detectors were applied. Two of them were placed at 90 degree to the left and right side relative to the proton beam direction. The third detector was at 150 degree at the right side. Flight paths were of 2.25 ± 0.01 m. The sample was placed at 8.5 ± 0.2 cm from the neutron target (0 position) and was moved also along the axis between detectors R90 and L90 at ± 3 cm and ± 7 cm. The plus sign means that the sample was moved towards the R90 detector and the minus sign in opposite direction towards the 90L detector. The third detector can see the sample only in the 0-position. Results for the sample in the 0-position only are discussed in this report. In every experiment the neutron detectors were shielded against direct and room-scattered neutrons.

Table 1. Average energies of the PFNS for all angles and runs. The letters shows left-L and right-R sides of the detector relative to the proton beam, ΔE = 0.010 MeV.

Angle, degree	<E>, MeV Jul06	<E>, MeV Apr07	<E>, MeV Jan08
R90	2.004	2.002	2.021
L90			2.007
L120	2.076	2.050	
R150		2.026	1.975

The traditional pulse-shape analysis was applied to reduce the gamma-ray background. A small Pilot-U scintillator was used as a proton pulse shape monitor. The data were collected in list mode for offline analysis. The detector efficiencies were measured relative to the ^{252}Cf standard spectrum. A specially designed low mass, fast ionization chamber [3] was put at the place of the U-sample keeping the same geometry as during the experiments. The energy spectra were corrected for detector efficiency, for neutron multiple scattering in the sample, and for time resolution. A detailed description of the experimental procedure will be published elsewhere [4].

The pulse mode operation of the VdG was not the same during these experiments. The FWHM was ~ 1 - 1.5 ns in all experiment. However, some tailing did exit which could not be removed completely. The worst tailing was observed during the Jul06 experiment. The best beam quality was realized during the third experiment, with a FWHM ~ 1 ns and a FW(1/1000)M < 10 ns. We recalculated the time resolution correction for the measured spectra from Jul06 published before [2, 5]. An additional energy dependence in the neutron detector efficiency at E > 4 MeV has been taken into account, too. This factor slightly reduced the PFNS in the energy range 5 - 8 MeV and the average secondary neutron energy by up to ~ 15 keV for the first run.

The time resolution of the TOF spectrometer consists of the following components: detector resolution, pulse shape of the accelerator and neutron

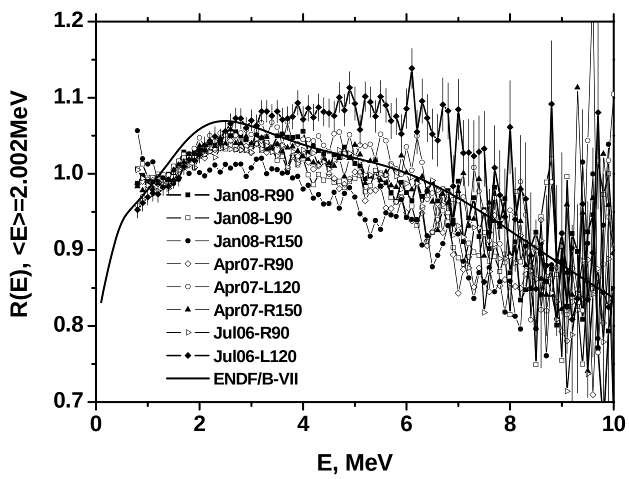

Fig. 3. Comparison between all of our results (full symbols). The ENDF/B-VII spectrum is given as a full line.

distribution inside the sample. The last factor is very important. Fig. 1 shows the TOF intensity distribution of the incident neutrons inside the sample for our experiment compared to the one in Ref. [6]. Clearly the double peaked intensity structure of our ring shaped sample is visible in contrast to the very broad distribution of the very large sample (7.7 cm diameter) of Ref. [6]. This factor together with a shorter flight path (1.63 m) in spite of a very short proton burst of 0.6 ns in Ref. [6] is very important for data comparison. Therefore, the experimental data of Ref. [6] were corrected for this time resolution assuming the same contribution of the detector resolution as in our experiment. The present experimental time resolution together with the calculated dependences including all factors is given in Fig. 2.

The experimental PFNS were normalized to unity and the average secondary neutron energy was calculated. A Maxwellian spectrum was fitted in the energy range of 0.7 - 1.5 MeV and 9 - 11 MeV to the measured spectrum and an extrapolation to zero and to 20 MeV was done. Based on our detailed analysis of all incorporated corrections and possible uncertainties, we conclude that the average energy is estimated with an accuracy of ± 0.010 MeV. The average energies measured in all experiments are given in Table 1.

The PFNS at all investigated angles and for all runs are shown in Fig. 3 as a ratio to a Maxwellian distribution with the average energy <E> = 2.002 MeV.

The following peculiarities may be highlighted:

1. The data demonstrate the variety of the spectrum shape. A difference exists not only for various detector angles but for detectors at 90 degree placed at left and right sides (see Jan08 90R, 90L in Fig 2 and Tables 1, 2);

Table 2. Average spectral ratios <R> = N(E,90R)/N(E,90L) and their errors for different energy intervals.

E_1-E_2, MeV	<R> ± δR	E_1 -E_2, MeV	<R> ± δR
0.8 - 2	0.999 ± 0.003	5 – 6	1.009 ± 0.005
2 - 3	1.010 ± 0.002	6 – 8	1.051 ± 0.006
3 - 4	1.020 ± 0.005	8 – 10	0.970 ± 0.032
4 - 5	1.034 ± 0.004		

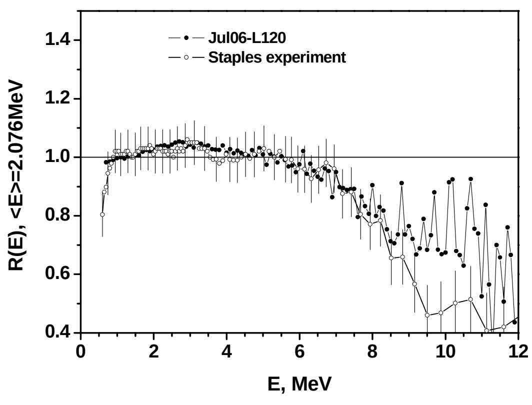

Fig. 4. Comparison between our result (Jul06 L120 detector) and data from Ref. [6].

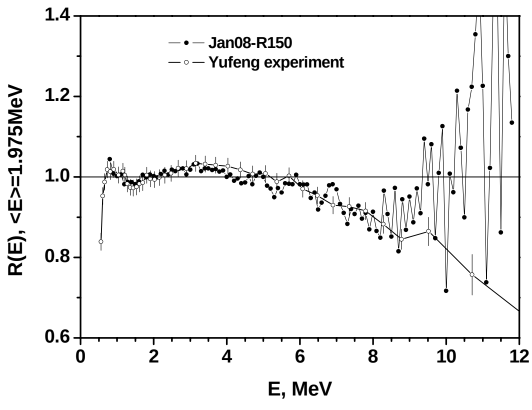

Fig.5. Comparison between our result (Jan08, R150 detector) and data from Ref. [7].

2. The normalized spectra are fixed at low and high energies (see Fig. 3). The integrals between 1.3 - 2.3 MeV and 8 – 10 MeV are constant. The standard deviations of 8 spectra are 0.2 % and 3 %, respectively;

3. Among these data one may find a result which agrees perfectly with an old experiment or evaluation.

A comparison between the different experiments and literature values are given in Figs. 4-8. The spectra measured at thermal energy were normalized to a Maxwellian with reduced average secondary energy - $\langle E_{\mathrm{th}} \rangle = \langle E_{0.5} \rangle * 0.995$.

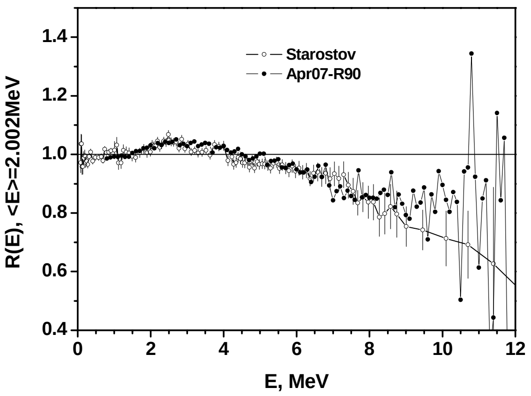

Fig. 6. Comparison between our result (Jul06, R90 detector) and data from Ref. [8].

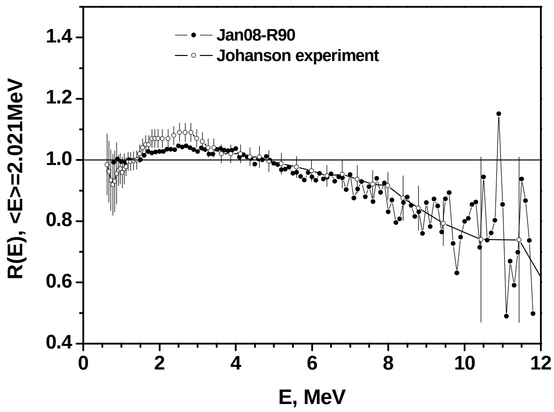

Fig. 7. Comparison between our result (Jan08, R90 detector) and data from Ref. [9] corrected with multiple scattering and angular distribution from the T(p,n) reaction taken from Drosg's evaluation [10].

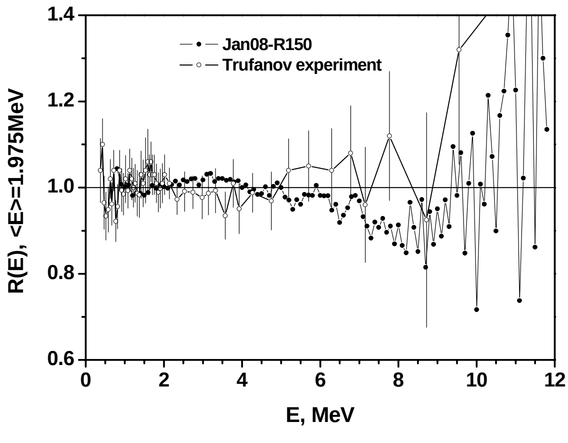

Fig. 8. Comparison between our result (Jan08, R150 detector) and data from Ref. [11].

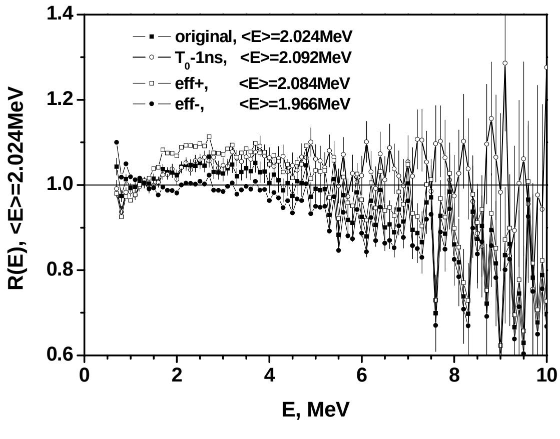

Fig. 9. The original spectrum and the ones estimated using different perturbation factors as given in the legend.

Before starting any scientific discussion about the nature of this strange behavior of the PFNS one should answer the main question: is this a real effect or an experimental artifact?

2. Possible experimental problems

The experiments were carried out relative to the standard ^{252}Cf spectrum measured in the same experimental conditions. Therefore a lot of systematic uncertainties such as: flight path, uncertainties in the time channel width, a possible time reference shift (T_0 value) connected with the detector operation, a distortion of the spectrum due to scattering in the collimator are drastically reduced or even canceled.

The shift of T_0 versus pulse height was investigated. After an additional correction as a function of pulse height, the shift was < 0.1 ns (ADC channel width was 0.117 ns).

We investigated a possible change of the ^{252}Cf spectrum due to different emission angles of the neutrons relative to the electrode plates in the ionization chamber. The ionization chamber was rotated relative to its vertical axis and the neutron spectra were measured by two detectors at 90, and 120 degrees [4]. No influence was found.

The spectrum shape may be distorted due to the proton pulse shape (VdG pulse mode operation) and a possible mistake in the time resolution correction. In this case the high energetic part of the spectrum (most sensitive to the time resolution) should be distorted. Since we observe the same integrals for the energy interval 8 -10 MeV this argument is not valid. In addition, this factor is common for all detectors and cannot explain the observed difference between them.

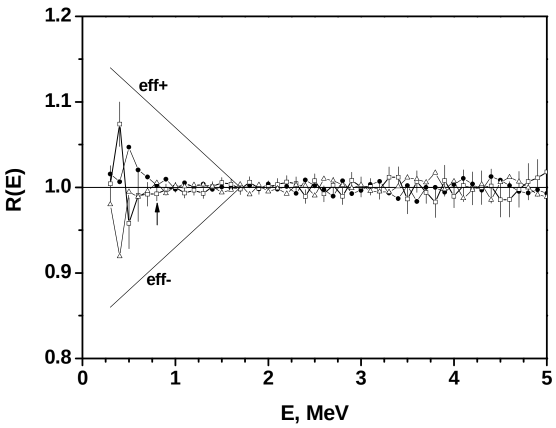

Fig. 10. The ratio of the detector efficiencies to the average value measured during the Jan08 experiment at the beginning, in the middle and at the end of the experiment. The distortion factors are shown by the full line. An arrow shows the cut-off energy in the data analysis.

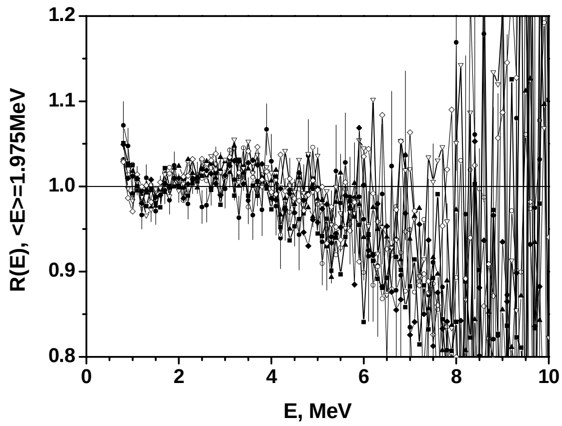

Fig.11. The spectra measured by the R150 degree detector during the Jan08 run.

So the most sensitive factor is the stability of the detectors and the correct estimation of the T_0 value. The detector efficiency might be arbitrary changed in between the Cf and U measurements. As one can see in Fig. 2 the prompt fission peak (the zero time (T_0) is determined relative to the prompt peak position) is very well separated from the main component due to prompt gamma rays from the target and T_0 can be deduced with an accuracy of ~ 0.1 ns. In addition to provide a measured difference between the spectra we should shift T_0 in the opposite direction depending on the neutron detector.

We simulated the influence of both factors. The results are given in Fig. 9. We calculated the spectrum with the nominal parameters, with a shifted T_0 by 1 ns and with a distorted detector efficiency by the function $1 \pm 0.1 * (1.7 - E)$, $E < 1.7$ MeV. The influence of these factors may provide an effect comparable with the data spread shown in Fig. 3, the average secondary energy varied by ± 70 keV. However, a shift of T_0 by 1 ns changed the integral in the energy range 8 – 10 MeV by 28 % which is ~ 10 times higher then the real data spread in Fig. 3, so we can also exclude this. Another possibility would be that the distortion factor is connected with instabilities of the threshold and neutron-gamma discrimination parameters. The detector efficiencies were measured before, in the middle, and after the U run in each experiment. The U-spectra shown in Fig. 3 are sums of several (5-7) runs measured during 10 - 20 hours, so the direct comparison of the separate spectra may answer this question about the detector stability. According to the results given in Figs. 10, 11 there is no evidence for a detector instability which may provoke the change in the measured results. In addition the detector efficiencies are in very good agreement with calculated results using the NEFF7 code [12], see Fig. 12.

These arguments are valid for each of the experiments, and the present conclusion is that we measured a real effect and no experimental artifact!

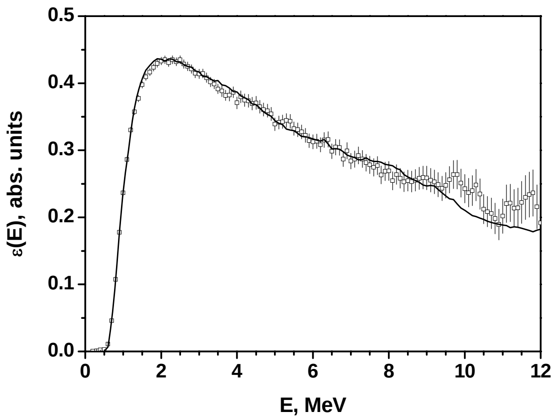

Fig. 12. The efficiency of one detector (Jan08, R150 detector) measured relative to ^{252}Cf and calculated with the NEFF7 code (full line).

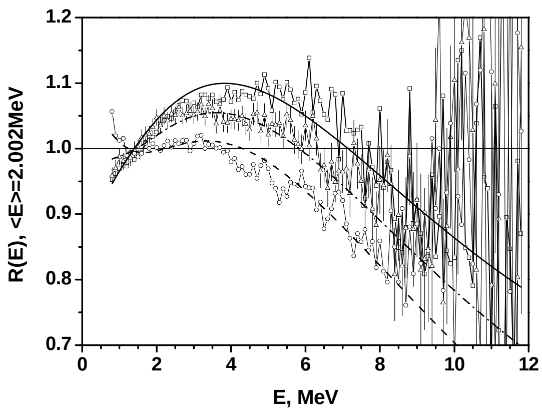

Fig.13. Some experimental data and their description with a "3 source model". Dashed line - $\zeta = 0.6$, dashed-dotted line – $\zeta = 0.4$, full line – $\zeta = 0.2$ (for details see text).

3. What does this experimental fact mean?

On the basis of the above discussion one may conclude that a factor exists which has a rather strong influence on the PFNS shape and asymmetry effects but was not fixed in our investigations and in all available experiments performed during the long history of fission investigations. One may assume that this factor is the neutron polarization. In the preparation stage of any PFNS experiment it was assumed that this factor is not important or by definition should be equal to zero. If this explanation is true, the transmission mechanism of the information from the incident neutron to the secondary fission neutron should be found. The only possibility might be scission neutron emission, a fast process without formation of the compound nucleus. This may provide the link between the incident neutron and the secondary fission neutron. We should have in mind that three particles (two fission fragments and a scission neutron) are emitted at the same time which complicates the problem a lot.

The information about scission neutron emission is very poor. It was estimated in Ref. [13] that the probability of fission with scission neutron emission is ~ 40 % (which corresponds to a total of 15 % in total multiplicity), that the spectrum of scission neutrons consists of a low (~ 0.8 MeV) and a high (~ 2.5 MeV) energy components. In Ref. [14] evidence was given that scission neutrons are emitted by fission fragments with high total kinetic energy (TKE) (compact system). From the results of this paper we may estimate a high energy limit in the spectrum for scission neutron emission of ~ 8.5 MeV. The question is now, which parameters should be changed to provide the variety of results given in Fig. 3.

In case of scission neutron (SCN) emission, fission neutrons should be emitted from three sources:

1. Neutrons from fragments after fission of the compound nucleus $A+1$

$$N_{A+1}(E) = (1-\alpha)\cdot W_{A+1}(E)\,, \qquad (1)$$

where α is the share of scission neutron emission and W_{A+1} is the spectrum which describes the neutron emission from accelerated fragments;

2. Neutrons from accelerated fragments after fission of the nucleus *A*, which is formed after the emission of one SCN:

$$N_A(E) = \alpha\cdot(\nu-1)\cdot W_A(E)/\nu\,. \qquad (2)$$

3. Scission neutrons itself:

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

where ζ is the share of the low energy component and ν is the neutron multiplicity.

The spectra W_A, W_{A-1} were calculated with a Watt distribution for light and heavy fragments with masses A_h = 140 and $A_l = A$-140. The ratio of the neutron multiplicity for light and heavy fragments was $\nu_l/\nu = \nu_h/\nu = 0.5$. Temperature parameters were found based on the Fermi-gas relation and the thermal-equilibrium assumption with an additional correction of cor = 0.9 for the excitation of the heavy fragment $U_h = U_{0h}\cdot$ cor [13]. The level density parameter was calculated as $a = A/c$, c = 8.4, TKE = 170.5 MeV, ν = 2.45.

The equation for $N_{scn}(E)$, and the corresponding parameters T_1, T_2 were taken from Ref. [13] introducing minor corrections: T_1 = 0.4 MeV, T_2 = 1.35 MeV. Changing only ζ from ζ = 0.2 to ζ = 0.6 allowed us to describe the spectrum shape with reasonable accuracy from the highest average secondary neutron energy <*E*> = 2.070 MeV to the lowest <*E*> = 1.967 MeV (Fig.13). The spectrum with ζ = 0.31 extrapolated to thermal energy describes the integral experiments. The average ratio of the calculated cross sections to the experimental ones (Ref. [15], IRDF-2002) is <*R*> = C/E = 0.997 ± 0.008. The average energy of the PFNS at thermal energy is <*E*> = 2.038 MeV.

4. Conclusion

In conclusion, a very unusual result, not observed before was found in the present investigation. Presently, there is no model, able to explain this result. We may assume that a different mechanism of the fission process and of neutron emission should be incorporated.

For the moment we may only conclude, that the measured effect is not an experimental artefact. We should assume the existence of an additional factor (parameter), for example the neutron polarisation which may be responsible for the measured peculiarities. However, we did not demonstrate the direct link between this unknown parameter and the fission neutron spectrum, the left-right and angular asymmetry. At present we can not answer the very important question, why the parameters of prompt fission neutrons changed so drastically and what is happening inside nuclear reactors.

So, new experimental efforts are urgently needed. It seems experiments with polarised thermal neutrons would be very interesting.

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