Search of scission neutrons in the measurements of angular and energy distributions of the prompt fission neutrons for ^{233}U, ^{235}U, ^{239}Pu and ^{252}Cf

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Motivation

The main purpose is this experimental investigation of the prompt fission neutron emission mechanism by the coincidence measurements of angular and energy distributions of neutrons and fission fragments.

It is known from previous experimental works:

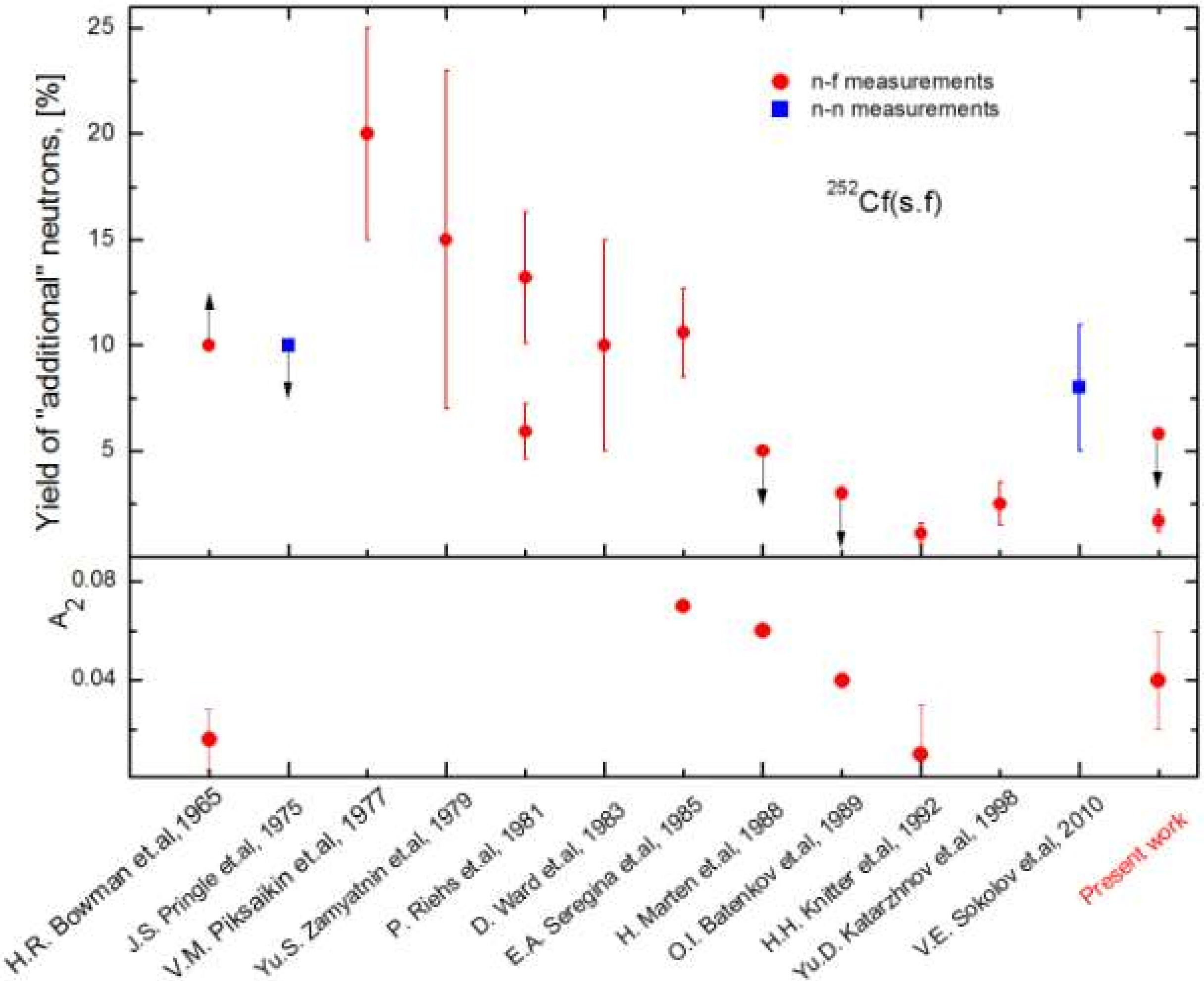
- The main source of prompt fission neutrons (PFNs) is fully accelerated fission fragments.
- The angular anisotropy of neutron emission in the center-of-mass system of fission fragment is not established.
- The contribution of neutrons with other emission mechanism ("additional" neutrons) to the total yield of PFNs ranges 1% - 20%.

The experimental data needed for such investigation, ideally, should be obtained using the same set-up and data processing for many fissioning nuclei and different excitation energies.

Therefore, using the same experimental set-up and data processing a few experiments have been carried out 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.

Motivation

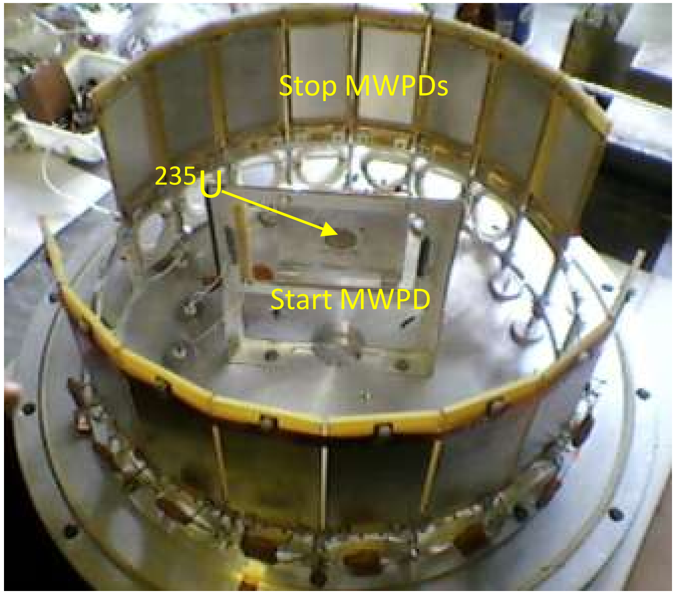
All experimental work where the yield of "additional" neutron was deduced.



Top – the "additional" neutron yield obtained by different experimental group.

Bottom – anisotropy of fission neutron angular distribution in the center-of-mass system of fission fragment.

Schematic view of the experimental set-up



Reaction Chamber:

Actinides target (Ø15mm) onto 70 μg/cm^2 Ti backing;

start MWPD (68 x 92 mm^2) located within 7 mm range from the target;

stop MWPD (72 x 38 mm^2) located at a distance of 140 mm from the chamber axis.

Neutron detectors:

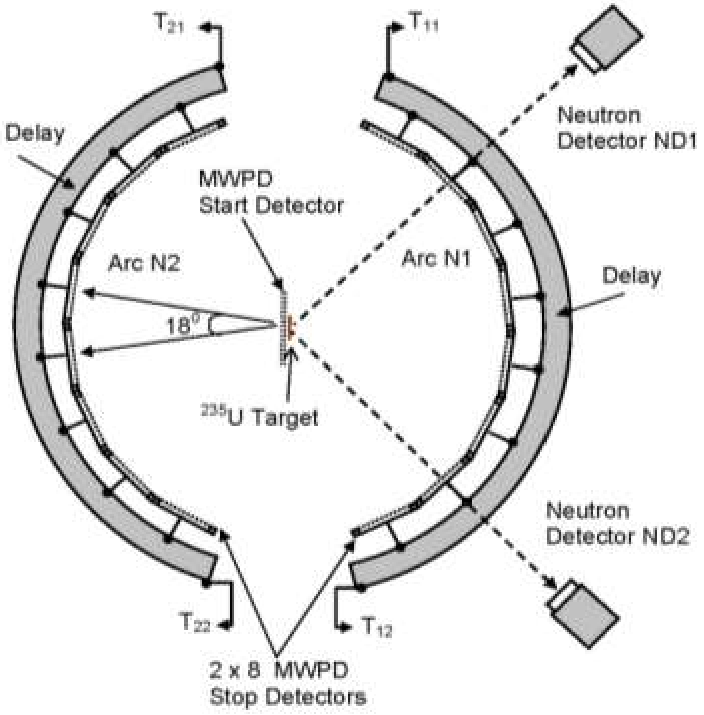
stilbene crystals (50 x 50 mm^2 and 40 x 60 mm^2 mounted on the Hamamatsu - R6091)

neutron registration threshold – 150 ÷ 200 keV;

double-discrimination method – pulse shape and time-of-flight criteria

time-of-flight distance from target – ~ 50 cm

timing resolution – 1.0÷1.2 ns



Analysis of the data

The spectra of prompt fission neutrons of ^{233}U, ^{235}U, ^{239}Pu and ^{252}Cf were measured at 11 angles relative to light fission fragment direction: 8.8°, 19.9°, 36.8°, 54.5°, 72.2°, 90°, 107.8°, 125.5°, 143.2°, 160.1° и 171.2°.

During data processing the following corrections have been taken into account:

- *Time uncertainties in TOF measurement:*
  - Pulse-height dependent time walk in neutron and fission fragment channels
  - Different fission fragments TOF to start MWPD
- *Time Neutron detector background:*
  - a double-discrimination method (TOF and pulse shape analysis)
  - true coincidence due to the neutron and fission fragment belonging different fission event registered was subtracted
  - the linear approximation of the remaining part of background was used
- *Fission fragment detectors efficiency*
- *Complementary fission fragment contribution*
- *Angular and neutron energy resolution* (timing resolution : 1.0 - 1.2 ns)
- *Bin-width correction*
- *Normalization correction* arising from the fact that experimental angular histograms were used in the measurements instead of continuous distributions
- *Neutron detector efficiency*

Analysis of the data

General features of the model used to describe measured spectra

- All prompt fission neutrons are emitted from fully accelerated fragments.
- Two fragments approximation is used. It is supposed that prompt neutrons are emitted from two (light and heavy) fragments with average mass and kinetic energy instead of real mass and kinetic energy distributions of fission fragments.
- The model prompt neutron spectra in the center-of-mass system of fission fragments are calculated using experimental data measured for 8.8°, 19.9°, 36.8° angles relative to the direction of movement of fission fragments .
- The angular and energy distributions of the prompt fission neutrons in the laboratory system are calculated on the basis of the obtained model spectra for light and heavy fragments.
- The contribution of “additional” neutrons (incompleteness of the model) is determined as difference between experimentally observable variables and model calculation in the laboratory system.

Analysis of the data

- Used equations :

The neutron spectra in the laboratory system:

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

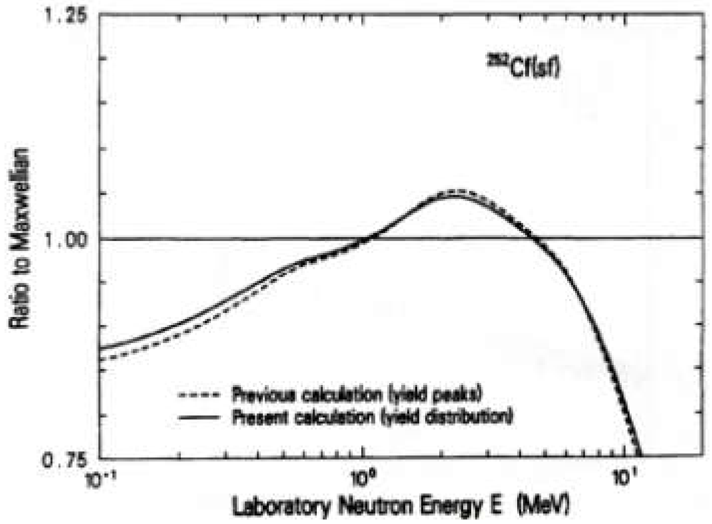
The neutron energy spectra in the c.m.s of the fission fragments:

$$E_{c.m.} = E_n + E_f - 2 \cdot \cos(\Omega) \cdot (E_n \cdot E_f)^{1/2}$$

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

where anisotropy $A_2(E_{c.m.}) = E_{c.m.} \cdot 2b/(3+b)$ and $b = (\phi(1,0^0) / \phi(1,90^0) - 1) \geq 0$.

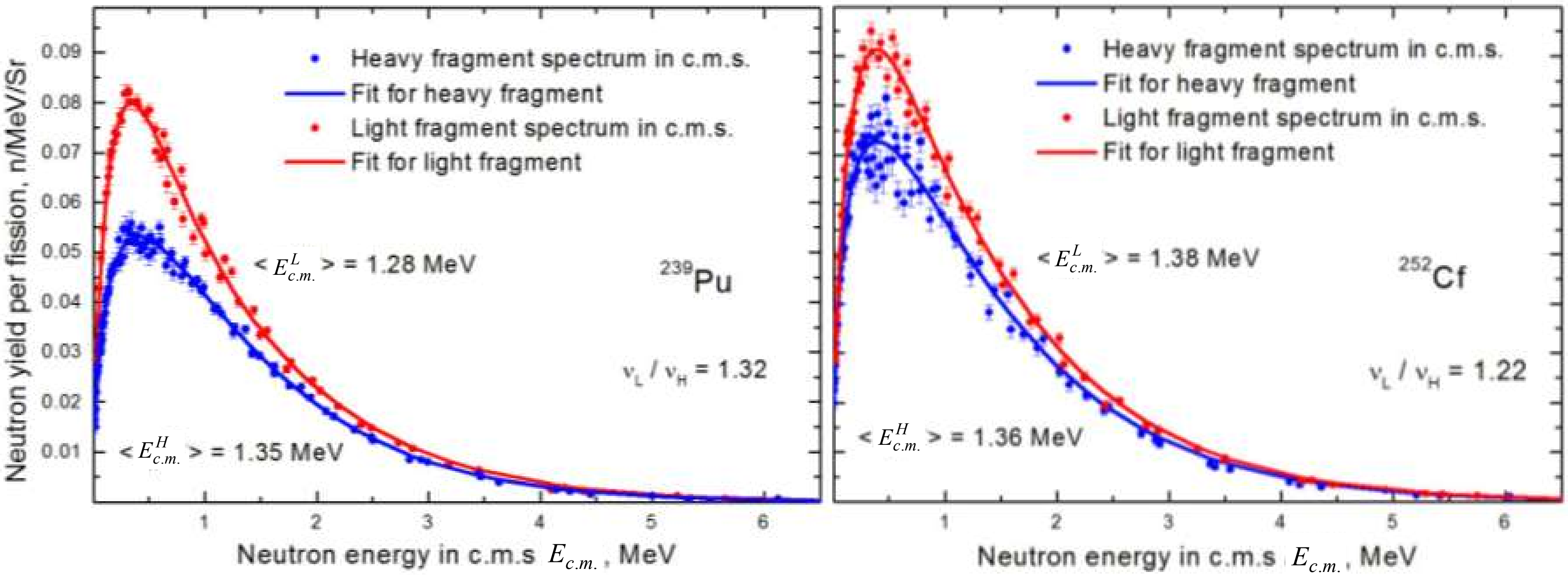
- Two fragments approximation gives a very good result, since, as it was shown for ^{252}Cf(*sf*)** , it has a minor influence on the total neutron energy spectrum.



**D.G. Madland, IAEA Report INDC(NDS) – 251, Vienna, 1991, p. 201

Analysis of the data

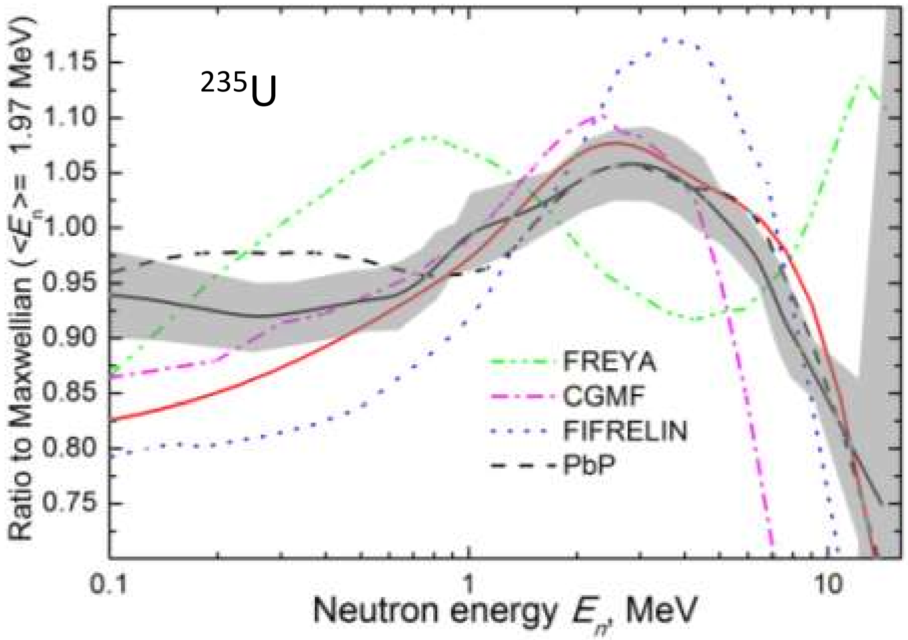
The obtained spectra in the c.m.s. of fission fragments



$$n_{c.m.}(E_{c.m.}) \cong Fit(\varpi, T_1, T_2, E_{c.m.}) = \frac{<\nu>}{4\pi} \cdot \left[\varpi \cdot \frac{E_{c.m.}}{T_1^2} \cdot \exp\left(-\frac{E_{c.m.}}{T_1} \right) + (1-\varpi) \cdot \frac{2 \cdot \sqrt{E_{c.m.}}}{\sqrt{\pi \cdot T_2^3}} \cdot \exp\left(-\frac{E_{c.m.}}{T_2} \right) \right]$$

Analysis of the data
(*Reliability of the calculation*)

Total prompt fission neutron spectrum of ^{235}U calculated by means of different groups with the same input parameters**



<u>PbP</u> (Point by Point) - deterministic method developed by the University of Bucharest and JRC-IRRM team and is an extended version of LAM (Los-Alamos or Madland-Nix model).

<u>FREYA</u> (Fission Reaction Event Yield Algorithm) – Monte-Carlo fission model developed through a collaboration between LLNL and LBNL (USA)

<u>CGMF</u> – Monte-Carlo code developed at LANL (USA)

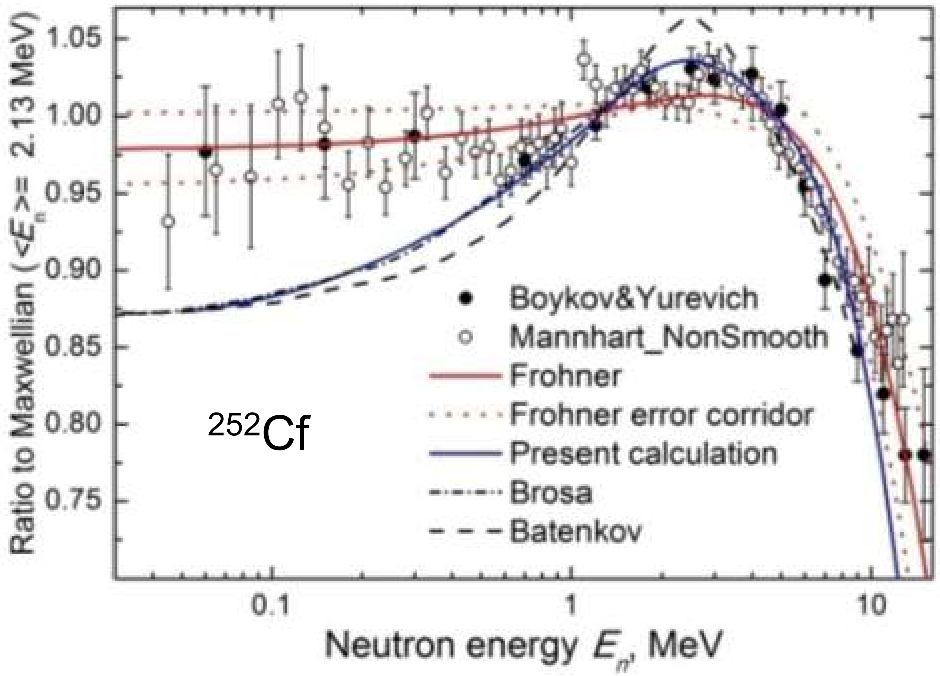
<u>FIFRELIN</u> (FIssion FRagment Evaporation Leading to an Investigation of Nuclear data) - Monte-Carlo code developed at CEA-Cadarache (France) with the aim of calculating the main fission observables

- ✓ Comparison of the total prompt fission neutron spectrum for thermal-neutron induced fission demonstrates that the existing calculation methods used in practice do not provide necessary accuracy.
- ✓ The method realized in this work gives the result in agreement with experimental data.

R. Capote, Y.-J. Chen, F.-J. Hambsch, *et al*., Nuclear Data Sheets, **131 (2016) 1.

Analysis of the data
(*Reliability of the calculation*)

The calculation performed by Brosa *et al*.** and in the work of Batenkov *et al*.** were performed using a complete set of fission fragments for mass and energies unlike to this work where the approximation of two fragments with average mass and kinetic energies was used.



- ✓ The results obtained by the different groups using different experimental data and data processing but the same calculation methods are agree with each other.
- ✓ The calculation method realized in this work (two fragment approximation) gives an accuracy not worse than those used in practice.

U. Brosa and H.H. Knitter, Z. Phys. **A343 (1992) 39.
O.I. Batenkov, A.B. Blinov, M.V. Blinov, S.N. Smirnov, IAEA Report INDC(NDS) – 220, Vienna, 1989, p. 207

Results

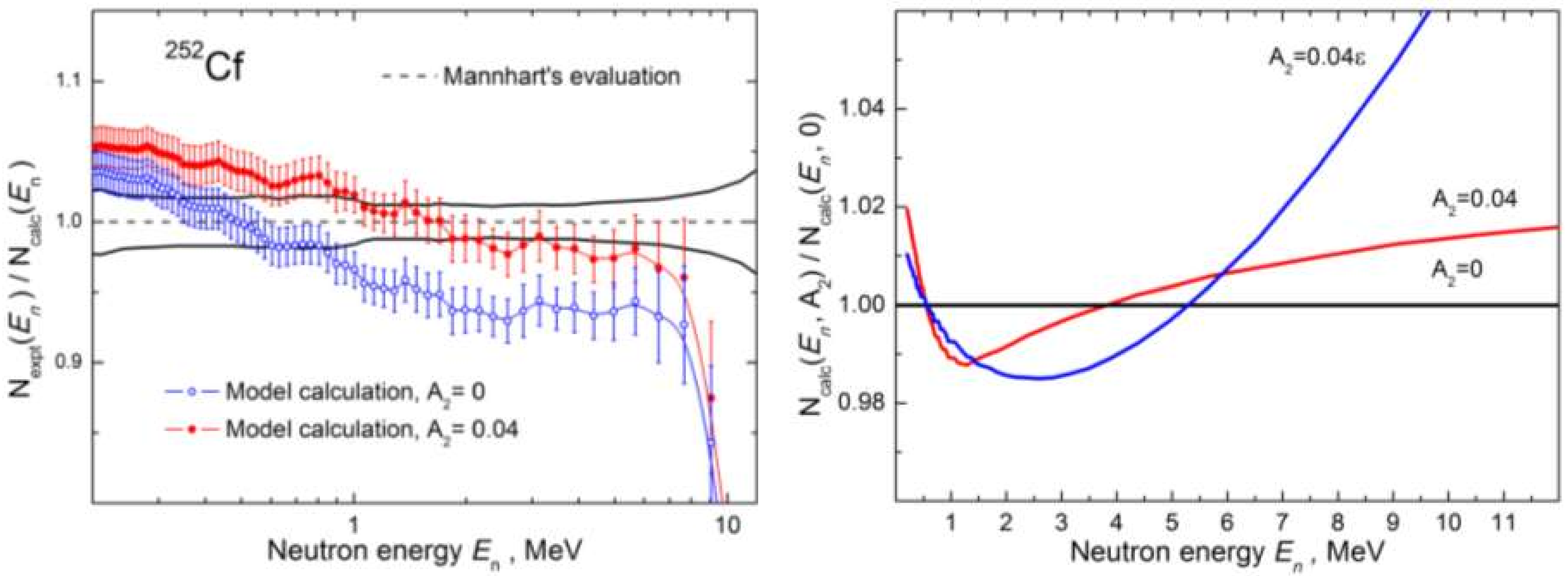
Total prompt fission neutron spectra

Target	ν_{prompt} (Neutron / fission)			
	Calculated		Experiment	Recommended
	$A_2 = 0$	$A_2 = 0.04$		
^{252}Cf(sf)	3.86	3.73	3.77 ± 0.03	3.7610 ± 0.0051
^{235}U(n_{th}, f)	2.56	2.45	2.44 ± 0.05	2.4184 ± 0.0021
^{233}U(n_{th}, f)	2.60	2.49	2.54 ± 0.06	2.4904 ± 0.0040
^{239}Pu(n_{th}, f)	2.93	2.85	2.89 ± 0.05	2.8778 ± 0.0050

- ✓ The average number of neutrons per fission event obtained by summing measured neutron distributions over angles and energies coincides with evaluated data within experimental errors.
- ✓ In the assumption that prompt neutrons are emitted isotropically from accelerated fragments, the model calculations (two fragments approximation) gives overestimated value of fission neutron yield as compared with experimental data.
- ✓ The including of anisotropy (A_2 = 0.04) of prompt neutron emission in c.m.s. of fission fragment into the model calculation improves agreement with experimental data.

Results

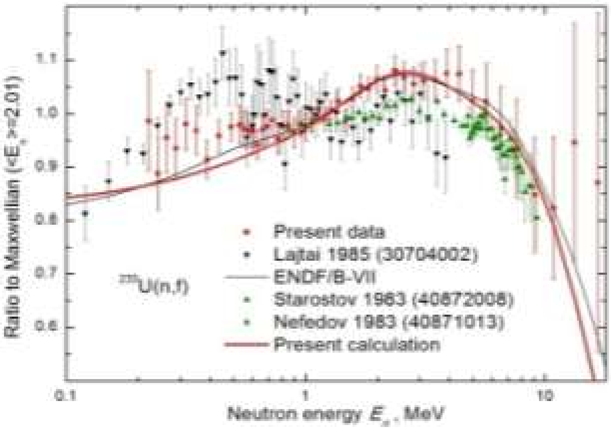
Total prompt fission neutron spectra
(sensitivity of the calculated/model spectra to anisotropy of neutron emission)

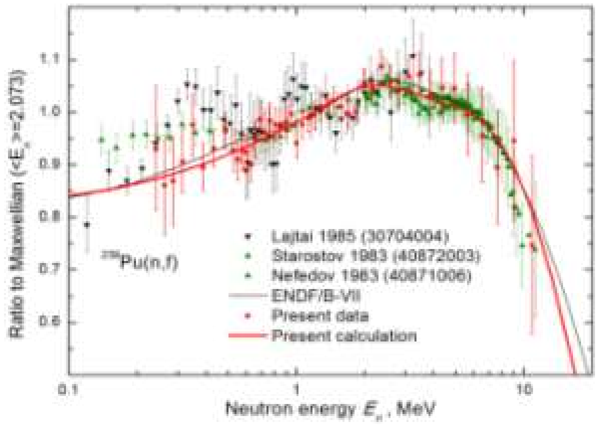


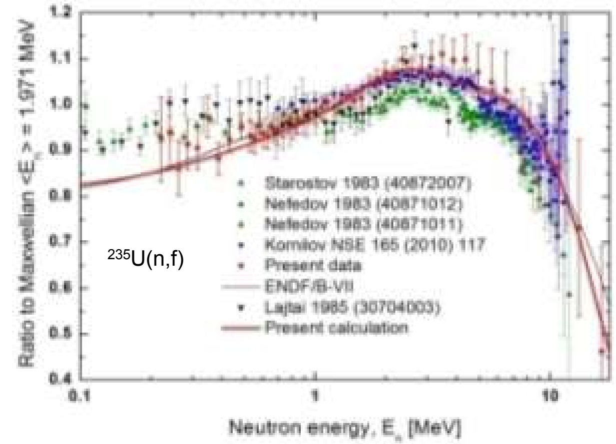
✓ The calculation performed using experimental data obtained for 8.9°, 19.8° and 36.9° angles relative to the direction of movement of fission fragments reproduces the total prompt fission neutron spectra both the shape and the average multiplicity.

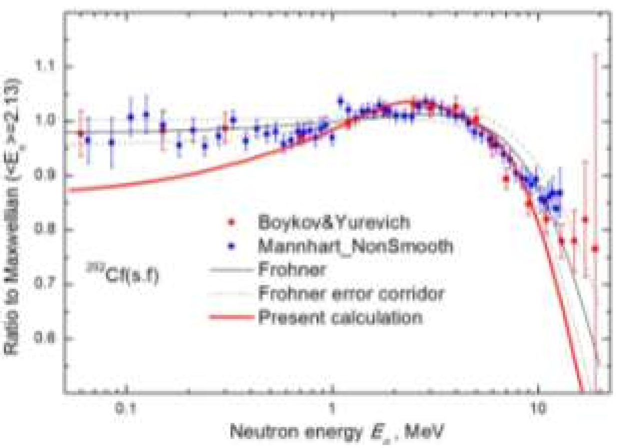
Results

Total prompt fission neutron spectra





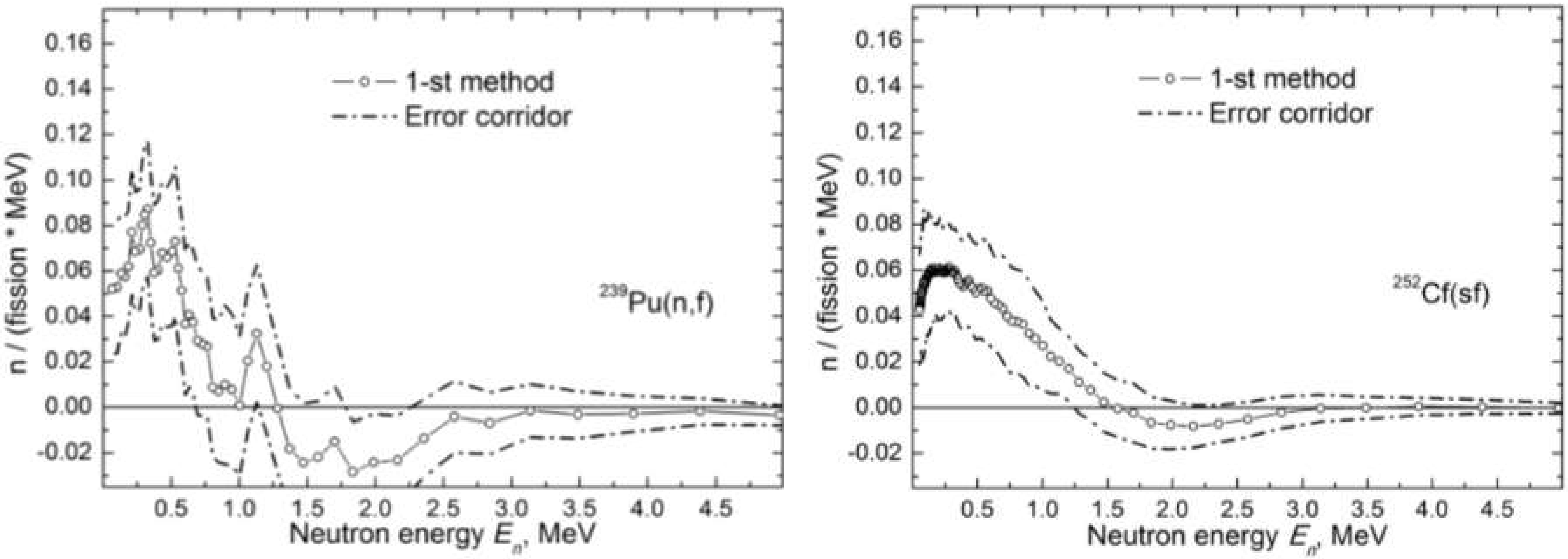




- ✓ There is an agreement between different experimental data within experimental uncertainties.
- ✓ The systematic difference of calculated spectrum from measured one is visible in the neutron energy range lower than 0.6 MeV.
- ✓ The observed difference may be interpreted as a manifestation of “additional” neutrons and, therefore, the average energy of these neutrons and their yield can be estimated.

Results

Spectrum of "additional" neutron

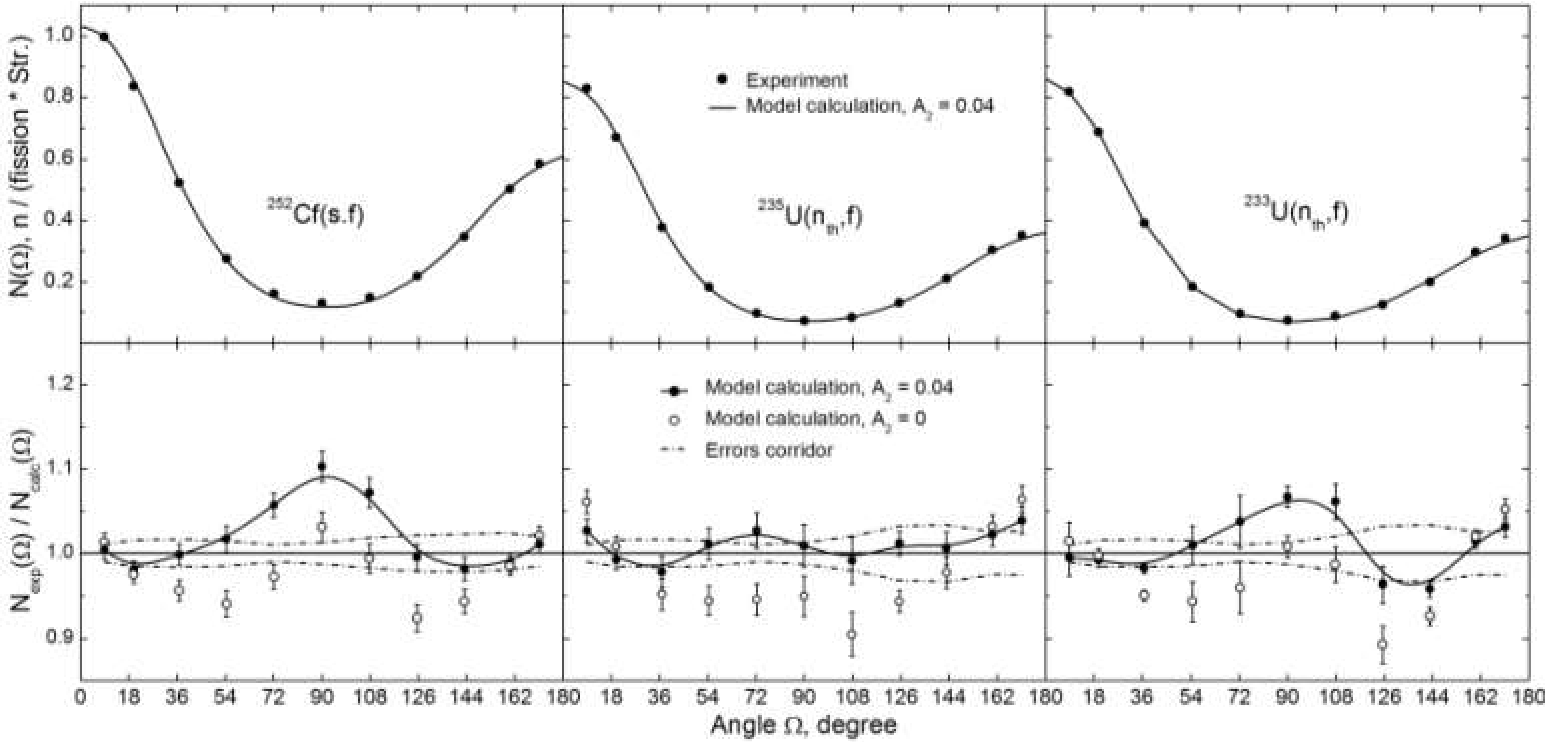


***Circles* – (1-st method)** the difference between total PFNS obtained by experiment (recommended data and it's errors) and calculated one assuming that all prompt neutrons are evaporated from accelerated fragments.

For all investigated nuclei the average energy and yield of the "additional" neutrons are ~0.4-0.6 MeV and ~1–2% of the total prompt fission neutron yield, respectively.

Results

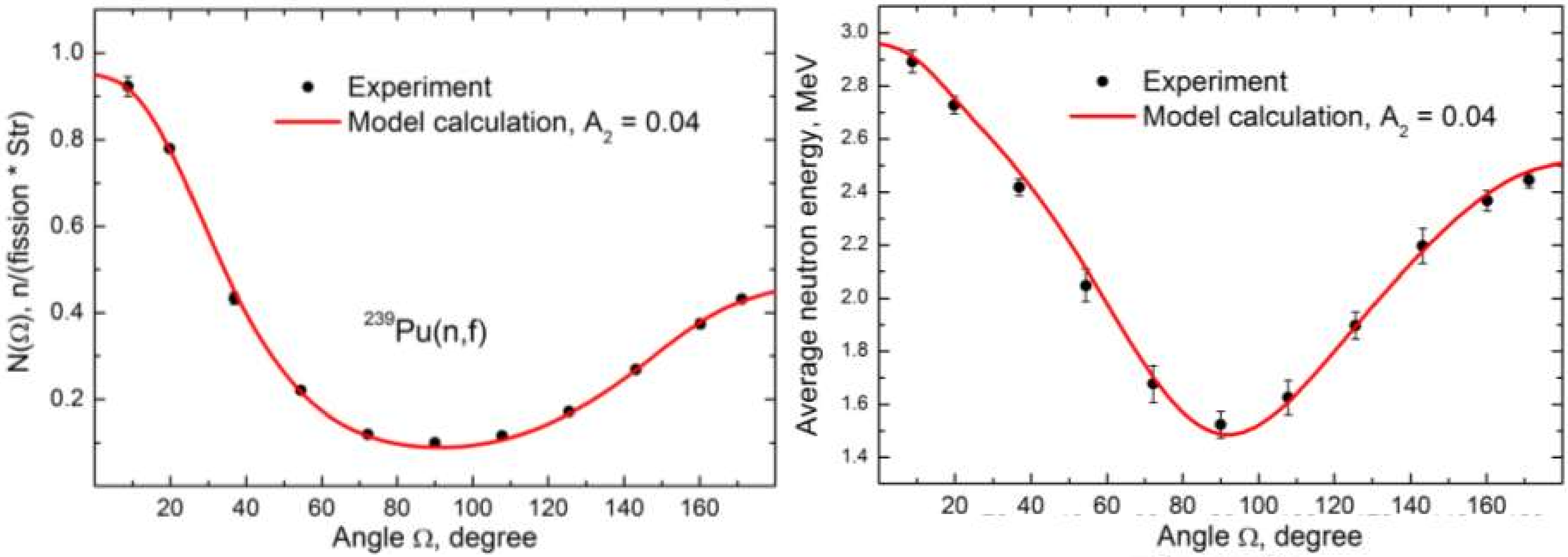
The prompt fission neutrons yield as a function of angle relative to the light fission fragment direction



- ✓ Introduction of anisotropy with $A_2 = 0.04$ into the calculation improves agreement with obtained experimental data.
- ✓ At that, there is some surplus of measured yield over calculated at angles near 90^0.

Results

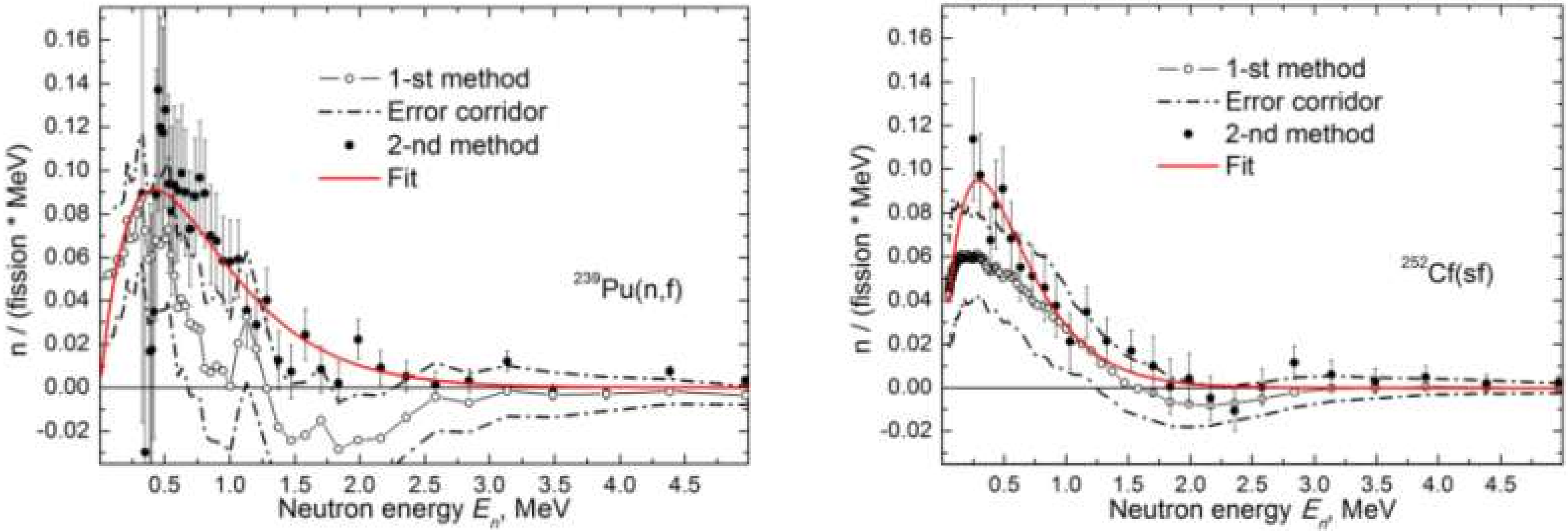
The yield and average energy of the prompt fission neutrons as a function of angle relative to the direction of movement of light fission fragments of ^{239}Pu



- ✓ There is a general agreement between experimental data and calculation performed in the assumption that prompt neutrons are emitted from accelerated fragments.
- ✓ At that, there is some surplus of measured yield over calculated at angles near 90^0.

Results

Spectra of "additional" neutrons for ^{239}Pu(n,f) and ^{252}Cf(sf)



***Circles* – (1-st method)** the difference between total PFNS obtained by experiment (estimated data and it's errors) and calculated one assuming that all prompt neutrons are emitted from accelerated fragments (A_2 = 0.04).

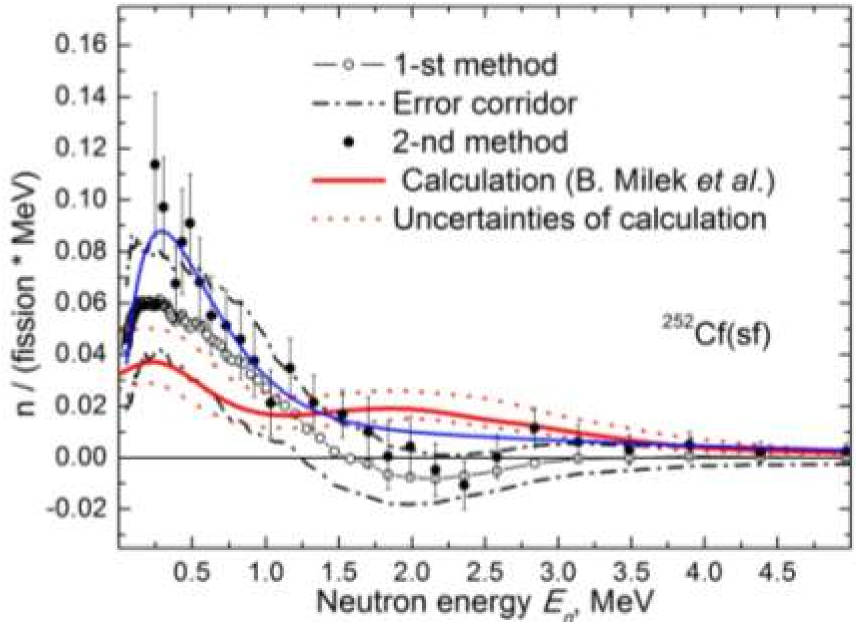
***Points* – (2-nd method)** the difference spectrum obtained using spectra measured at 72.2°, 90° and 108.8° relative to the light fission fragments and the corresponding ones calculated in the assumption that all prompt neutrons are emitted from accelerated fragments (A_2 = 0.04).

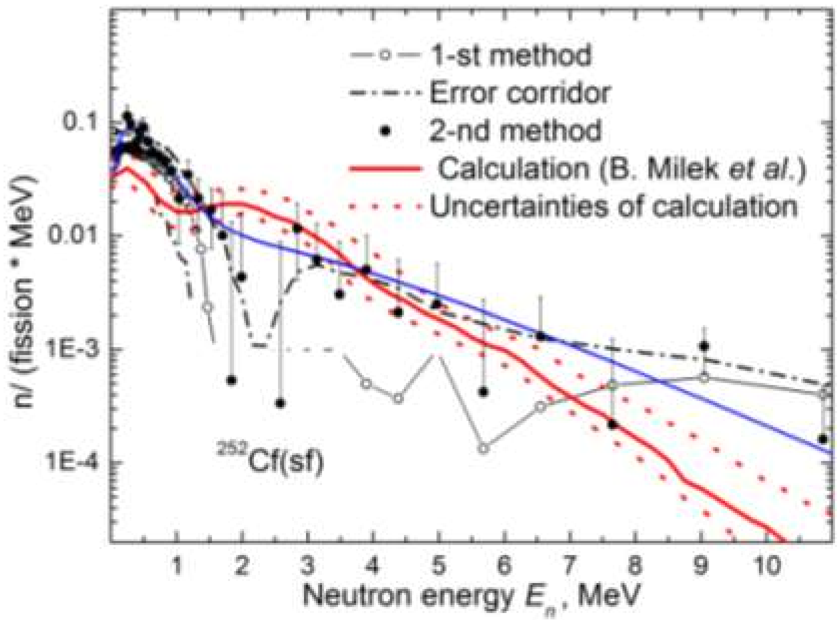
Red line – fit of experimental data marked with ***points*** by following equation: $$p_S(E) = p_0 \cdot \frac{E}{{T_S}^2} \cdot \exp\left(-\frac{E}{T_S}\right)$$

There is an agreement within experimental uncertainties between spectra of "additional" neutrons obtained by two different ways.

Results

Comparison of the obtained spectra of "additional" neutrons for $^{252}Cf(sf)$ with the model calculation taking into account nonadiabatic effects performed by Milek et al.





Blue line **–** fit of experimental data marked with ***points*** by following equation:

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

Red line **–** from the work of *B. Milek, R. Reif, and J. Révai, Phys. Rev. C **37**, 1077 (1988).

Results

Main characteristics of "additional" neutrons

	$^{233}U(n,f)$	$^{235}U(n,f)$	$^{239}Pu(n,f)$	$^{252}Cf(sf)$
Fit 1 of "additional" neutrons spectra				
Yield	1.5 ± 0.6 %	1.8 ± 0.6 %	3.6 ± 0.5 %	2.0 ± 0.6 %
Average kinetic energy	0.53 ± 0.08 MeV	0.47 ± 0.05 MeV	0.91 ± 0.19 MeV	0.58 ± 0.06 MeV
Fit 2 of "additional" neutrons spectra				
Yield	2.7 ± 0.8 %	2.6 ± 0.8 %	4.5 ± 0.9 %	3.0 ± 0.8 %
Average kinetic energy	1.7 ± 0.2 MeV	1.4 ± 0.2 MeV	1.6 ± 0.2 MeV	1.5 ± 0.2 MeV

Conclusion

The angular and energy distributions of the prompt fission neutrons for ^{233}U(n,f), ^{235}U(n,f), ^{239}Pu(n,f) and ^{252}Cf(sf) have been measured.

As a result of the comparative analysis of the obtained distributions and calculated ones, it was established that:

- The angular anisotropy of the neutron emission in the fragment center-of–mass system, which is alike to $\sim 1 + (0.06 \pm 0.02) \cdot E_{c.m.} \cdot \cos^2(\Omega_{c.m.})$, should be included into any calculation of prompt neutron properties in nuclear fission;
- There are some surplus of measured neutron yield above calculated one for the total neutron spectra as well as for neutron spectra at fixed angles near 90^o (relative to fission fragments direction);
- This difference can be eliminated by assuming that the yield of "additional" neutrons is equal to 2 - 4 % of total neutron yield per fission event;
- Probably, these "additional" neutrons are emitted from fissioning nuclei due to the dynamical effects analogous to ones discussed by Milek et al.