Measurement of Fission Cross Section and Angular Distribution of Fission Fragments from Neutron-Induced Fission of ^{242}Pu in the Energy Range 0.3-500 MeV

A.M. Gagarski, A.S. Vorobyev, O.A. Shcherbakov, L.A. Vaishnene, A.M. Tiagelskaia, N.M. Olkhovich

Petersburg Nuclear Physics Institute of NRC "Kurchatov Institute", Gatchina, Russia

A.L. Barabanov

NRC "Kurchatov Institute", Moscow, Russia

T.E. Kuz'mina

Khlopin Radium Institute, St.-Petersburg, Russia

PNPI, Gatchina

NATIONAL RESEARCH CENTER "KURCHATOV INSTITUTE"

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Motivation for measurements of the fission cross section of ^{242}Pu

- High accuracy neutron induced fission cross section data on the even-even isotopes are required to make nuclear technology safer and more efficient and to meet the upcoming needs for the future generation of nuclear power plants (GEN-IV and ADS)
- The practical implementation of plans for both the creation of a closed fuel cycle based on fast nuclear reactors and the disposal of radioactive waste is impossible without reliable and accurate nuclear data. For example, the required accuracy of the fission cross section of $^{242}Pu(n,f)$ is 2-5 times higher than currently available

	Initial versus target uncertainties (%)		
Energy Range	*Initial*	*SFR*	*ADMAB*
6.07 - 19.6 MeV	37	15	
2.23 - 6.07 MeV	15	5	7
1.35 - 2.23 MeV	21	5	5
0.498 - 1.35 MeV	19	4	4
183 - 498 keV	19	9	

Table from NEA
Nuclear Data High Priority Request List

SFR – sodium-cooled fast reactor

ADMAB – accelerator-driven minor actinide burner reactor

[WPEC-26, NEA No. 6410. OECD-NEA, 2008]

- ^{242}Pu is the longest-lived plutonium isotope in spent nuclear fuel ($T_{1/2}$ = 375 000 y) and hence it is important for nuclear transmutation
- The data available on the fission cross section of ^{242}Pu are mainly limited to the neutron energies below 20 MeV. Most of these data were obtained using monoenergetic neutrons obtained in various reactions at accelerators. The available experimental data reveal a significant scatter. There are only two data sets for neutron energies above 20 MeV. **New measurements of the fission cross section of ^{242}Pu must be taken in a wide neutron energy range on neutron beams with a continuous spectrum using the TOF method**

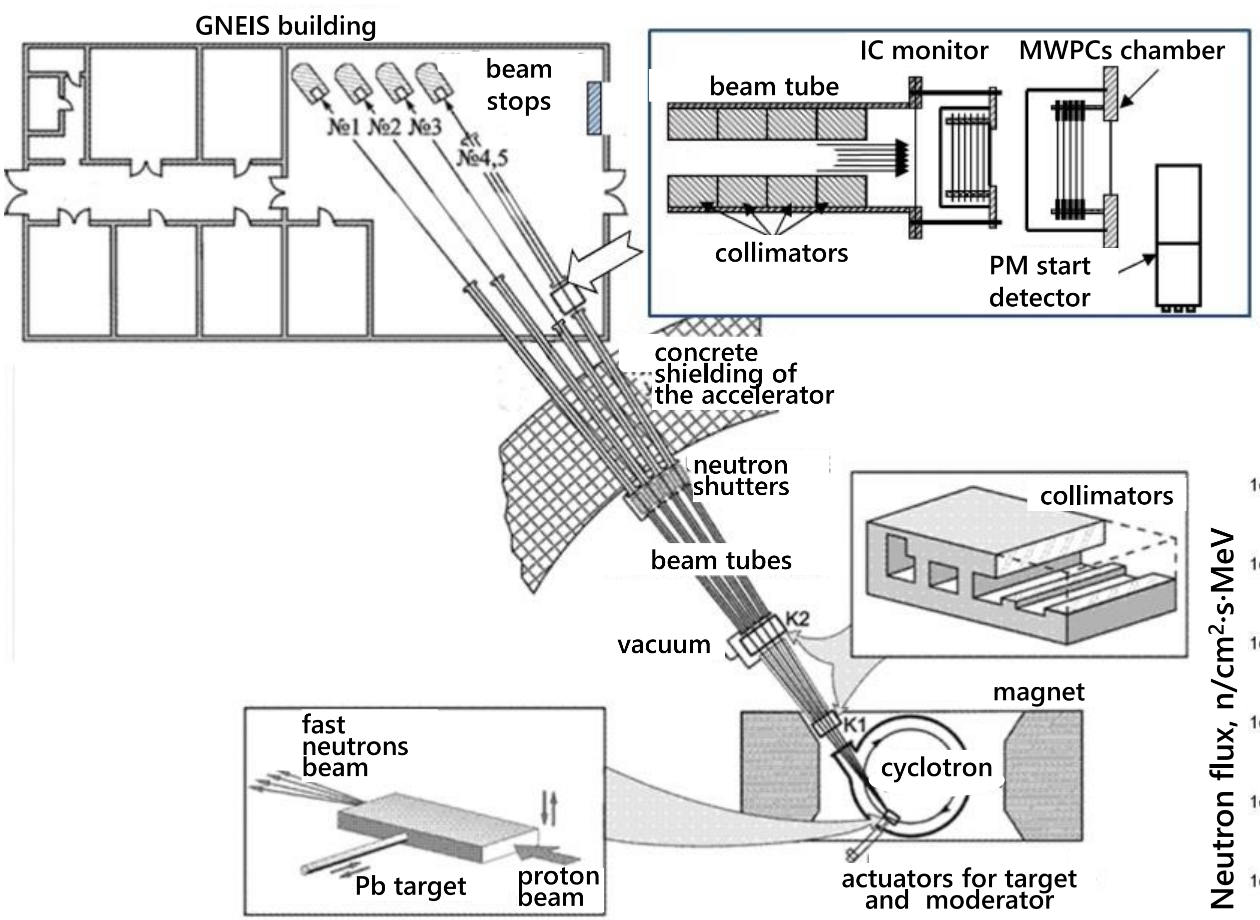
Motivation for studying the angular distributions of fission fragments

The angular distributions of fission fragments appear due to the action of two factors: 1– the ensemble of spins of fissile nuclei must be aligned; 2– the distribution of transition states over the K-projection of the nuclear spin onto the fission axis must be non-uniform. The processes preceding fission determine the first factor, while the second factor is set by the fission mechanism itself.

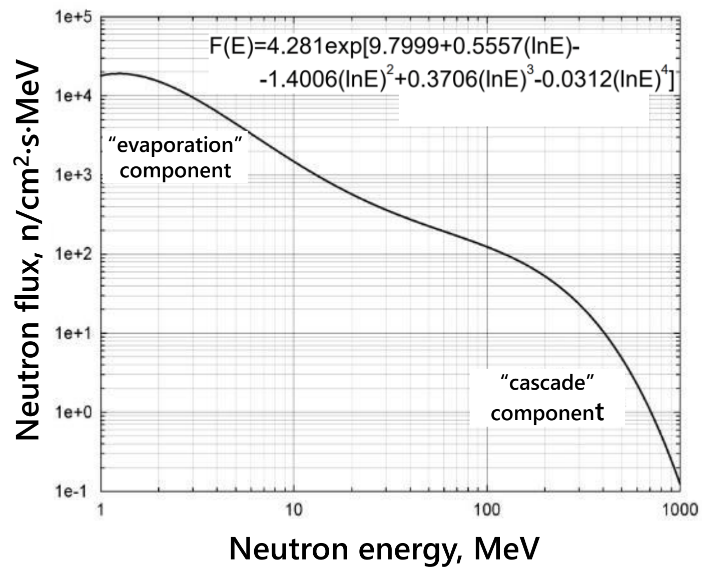
Information on fission barriers and transition state spectra on barriers. (states of highly deformed fissioning nucleus at the fission saddle point)

- Verification and developing of models for adequate description of processes in nuclei at high excitations (relative contribution of equilibrium and non-equilibrium processes into the dynamics of highly excited nuclei)
- The angular distributions data are important for precise measurements of the fission cross-sections, because it should be taken into account as efficiency correction for non 4π detectors
- Such an information for highly excited nuclei is important for development of new technologies, such as Accelerator-Driven Systems for nuclear power, nuclear waste transmutation, and etc.
- **Only two data sets are available to date for ^{242}Pu and there are no such data above ~ 8 MeV**

Neutron TOF-spectrometer GNEIS



Neutron spectrum of GNEIS: from thermal to 1GeV



Main parameters:

$E_{protons} = 1$ GeV; Lead target; $\Delta t \approx 10$ ns;

$f \approx 50$ Hz; $\Phi \sim 3 \times 10^{14} \frac{\text{n}}{\text{s}}$;

$L = 36.5$ m

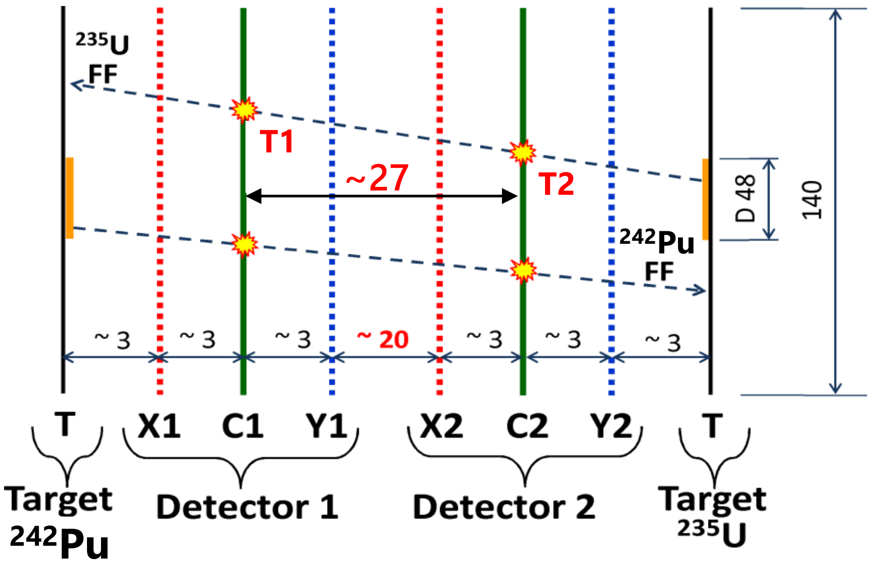
$$\frac{\Delta E}{E}(1\ MeV) \approx 1\% \ ; \ \frac{\Delta E}{E}(200\ MeV) \approx 12\%$$

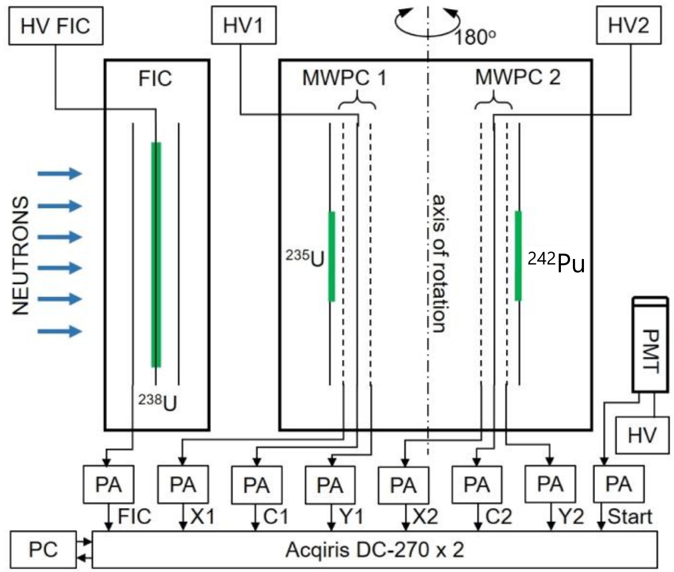
Experimental setup

Most often, the fission cross section of the nucleus under study is measured relative to the cross section of a reaction that is known with a high accuracy (standard cross section) such as n–p scattering, …, and the neutron induced fission of ^{235}U.

To do this, it is necessary to place the main target and the reference target

- with an exactly known ratio of number of nuclei (N_{Pu2}/N_{U5})
- in the same neutron flux and
- register fission fragments with detectors with the same (or well known) efficiencies.





The setup consists of two position-sensitive low pressure multi-wire proportional counters (140×140). Targets are located on opposite sides of them.

Waveforms from 6 electrodes and from "Start" PM are recorded with 500 MHz 8 bit digitizer. →

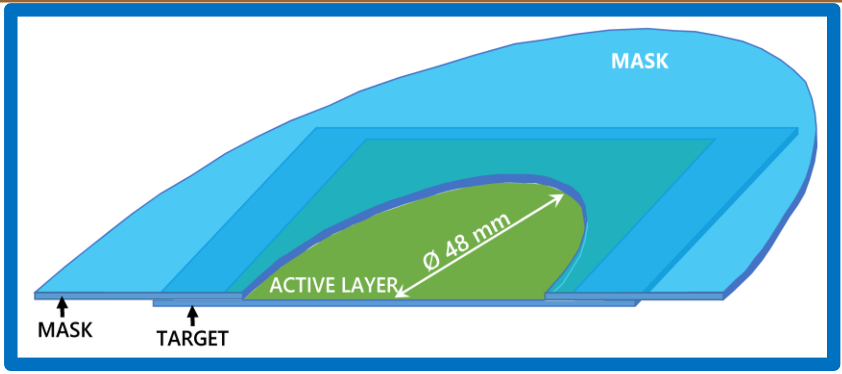
7 timestamps and pulse heights → x1, x2, y1, y2, $T_{cathode1}$, $T_{cathode2}$, T_{start} →

$$\cos(\theta) = \frac{d}{\sqrt{(x1 - x2)^2 + (y1 - y2)^2 + d^2}}$$

$$L/(T_{cathode} - T_{start}) \rightarrow E_n$$

Targets

Targets from ^{242}Pu and ^{235}U were fabricated at the Khlopin Radium Institute (St. Petersburg, Ru) by the "painting" method on aluminum substrates 0.1 mm in thickness. The initial shapes and sizes of the active layer were different (50×100 mm for ^{235}U and Ø 82 mm for ^{242}Pu).



To ensure identical conditions for measurements of fission cross sections, namely, small and equal shape samples in wide homogeneous neutron beam, 0.1-mm-thick aluminum masks were placed on the active layers of the both targets for to separate equal circular regions with a diameter of (48.0 ± 0.1) mm on the active layers.

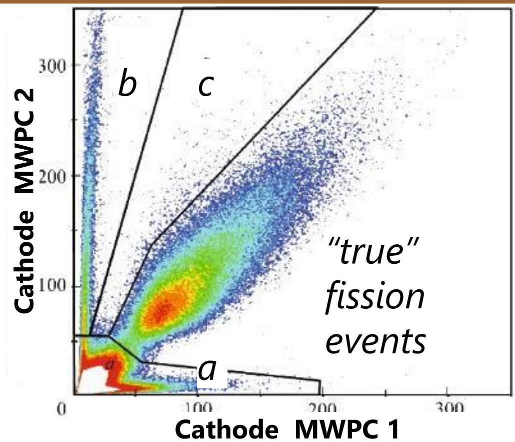
For determine the number of nuclei, we made α-spectrometry of the active spots with SB-detector in precisely known geometry.

Main isotope	^{235}U	^{242}Pu
Thickness of active layer (μg/cm2)	203(11)	281(14)
Homogeneity of active layer	10%	10%
Main isotope mass inside mask Ø 48 mm (mg)	3.480(48)	5.35(5)
Target activity inside the mask Ø 48 mm (Bq)	295	9.34×10^5
Scaling factor (N_{Pu2}/N_{U5})	**1.493(25)**	

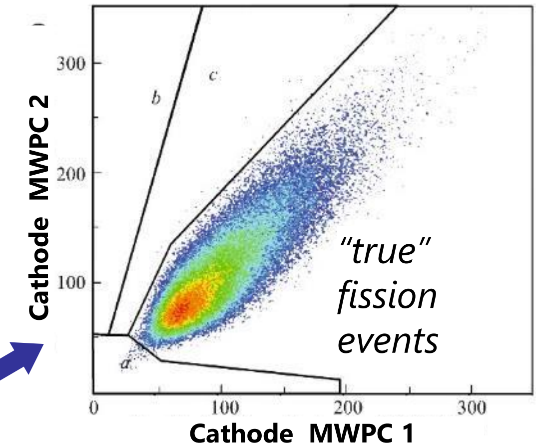
	^{235}U	^{242}Pu
Isotope	Mass percentage (%)	
^{235}U	99.9920(10)	
^{234}U	0.0020(5)	
^{236}U	0.0040(5)	
^{238}U	0.0020(5)	
^{242}Pu		99.65
^{239}Pu		0.25
^{240}Pu		0.092
^{238}Pu		0.0013
^{241}Am		0.0054

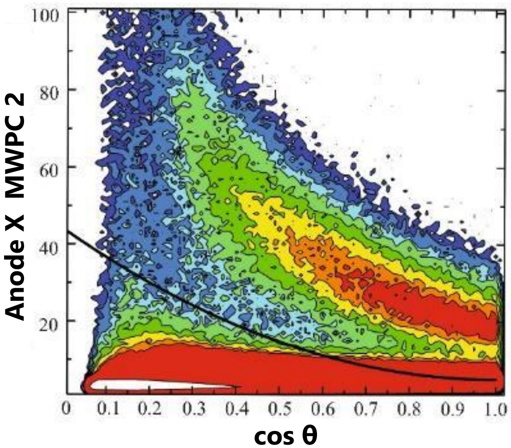
Black- parameters given in manufacturer's certificates
Blue- more accurate parameters obtained by us using α-spectrometry

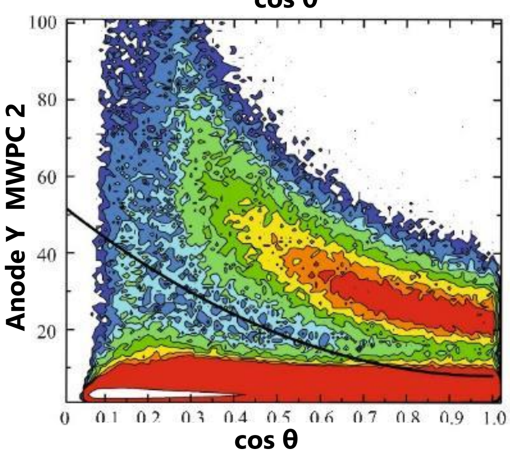
Method for selection of "true" fission events

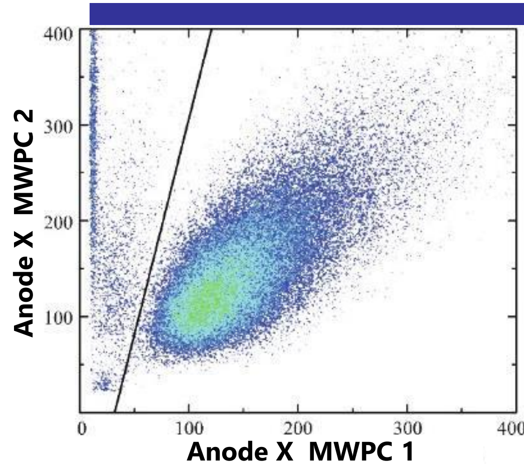


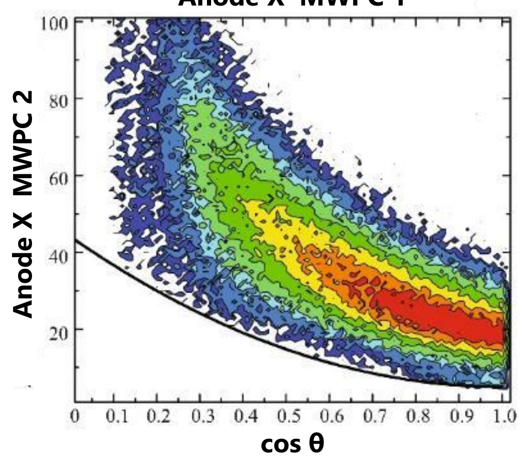
a. non-fission reactions in backing and α-partricles and noises
b. FFs "died" in MWPC 2 (first from the target)
c. FFs "died" on the cathode of MWPC 1 (second from the target)









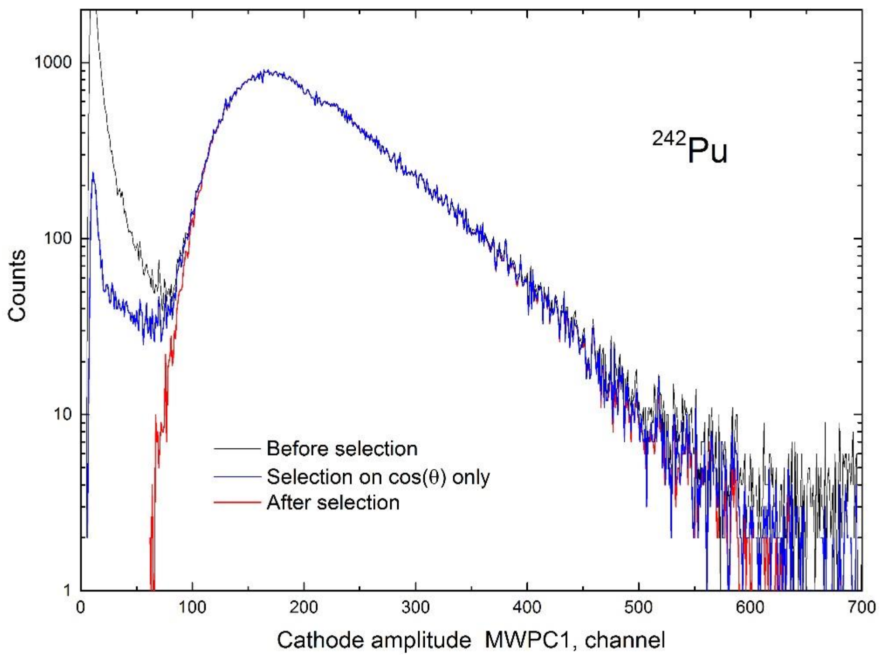


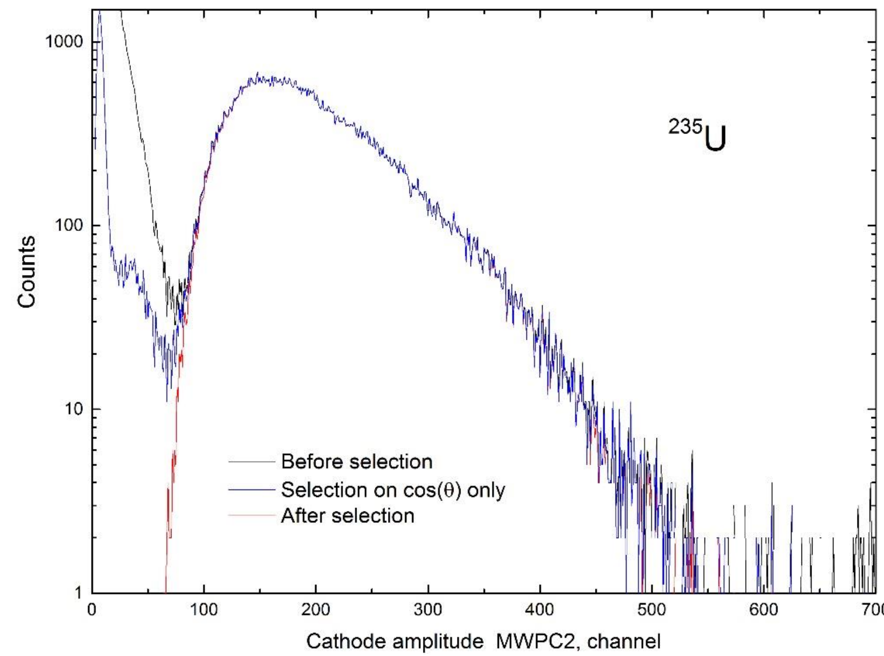
By applying a "proper" set of selection "cuts" to 2-dimentional datasets –

(AnodeX_MWPC2 × cos(θ),
AnodeY_MWPC2 × cos(θ),
AnodeX_MWPC1 × AnodeX_MWPC2)

we are able to achieve complete rejection of non-fission events.

Amplitude spectra of signals from the MWPCs cathodes before the selection of “true” fission events (black) and after (red)





- It is remarkable that “true” fission events are completely separated from neutron-induced background reactions in the substrate of the target and in other materials of the detector, from α-particles and noise signals.
- Fission fragments are registered without any threshold cutoff

Admixture of spontaneous fission of ^{242}Pu target

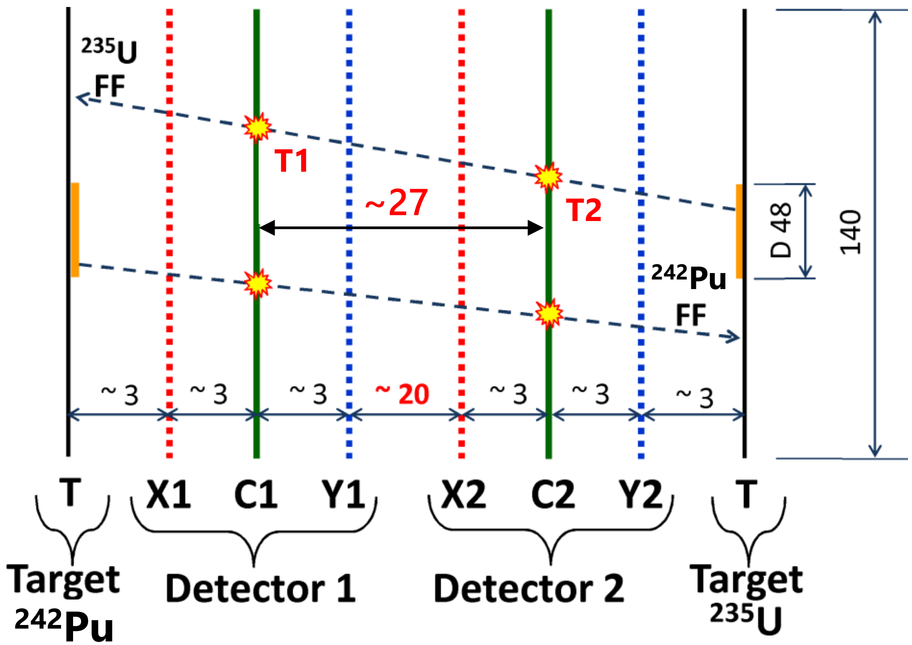
In addition to the α-decay mode, the ^{242}Pu also has a spontaneous fission decay mode with a probability 5.5×10^{-6}. This creates a non-correlated background of spontaneous fission events. The background from spontaneous fission was 1.75±0.04 1/s.

It was calculated based on the efficiency of detection of fission fragments, the spontaneous fission half-life for ^{242}Pu, and the mass of ^{242}Pu precisely determined in this work.

Energy	Share of spontaneous fission in the total fission fragments counts rate
0.2 MeV	~ 10 %
0.3 MeV	< 2 %
>1 MeV	< 0.02 %

The spontaneous fission background was subtracted from the time-of-flight spectra and from the measured angular distributions.

Separation of events from ^{235}U and ^{242}Pu



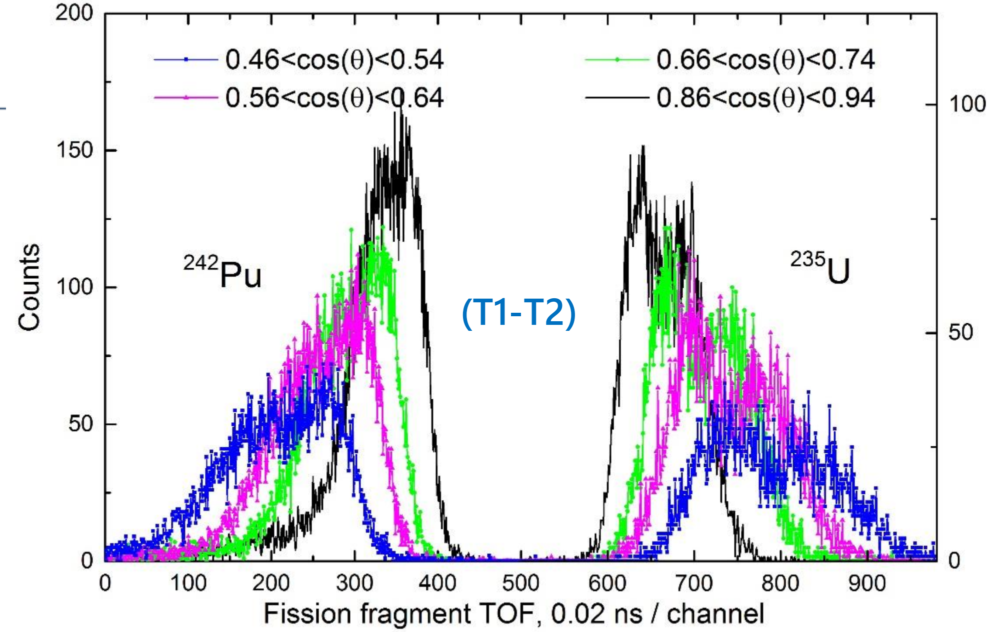
Time stamps T1 and T2 are derived from the signal waveforms from the two cathodes.

If the time mark T1 comes earlier than T2, then fragment from ^{242}Pu passed the MWPCs from left to right.

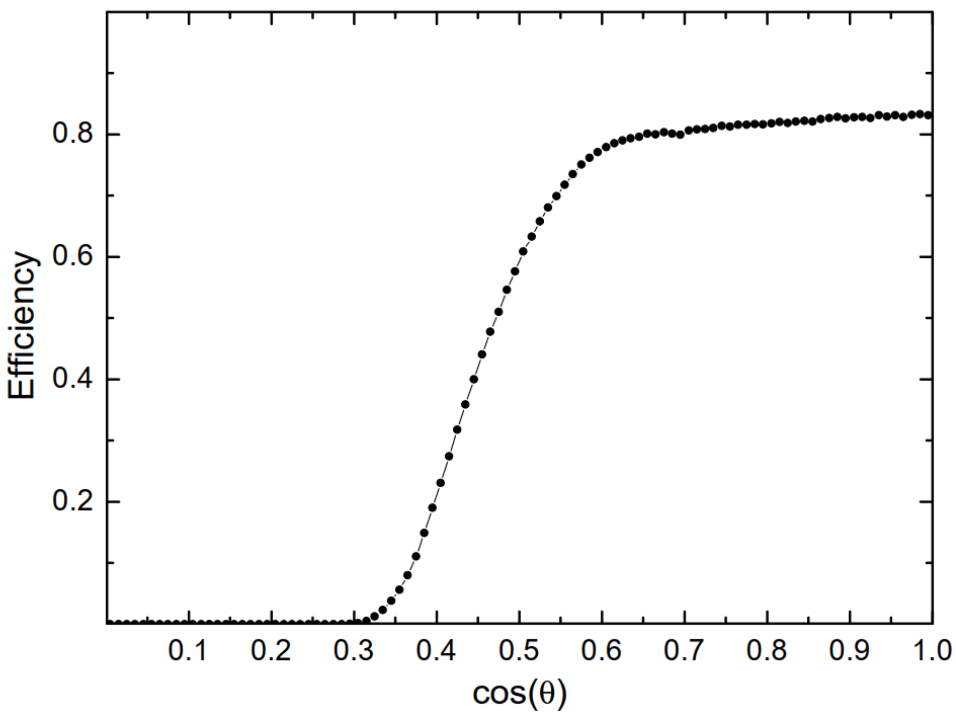
If the time mark T1 arrives later than T2, then fragment from ^{235}U fragment passed the MWPCs from right to left.

In Time-of-flight spectrum (T1-T2) events from ^{242}Pu and ^{235}U can be separated.

Time-of-flight spectrum of fission fragments of (left part) ^{242}Pu and (right part) ^{235}U from the 500th channel at various angles θ



Monte-Carlo efficiency simulation in real setup geometry



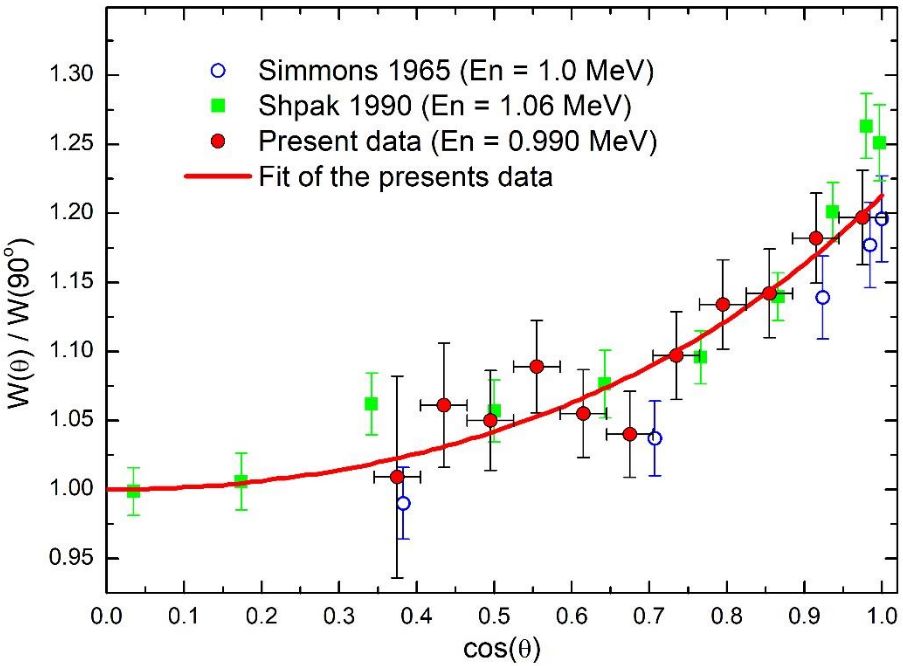
$$W_{correct}(\theta) = \frac{W_{exp}(\theta)}{\text{Efficiency}}$$

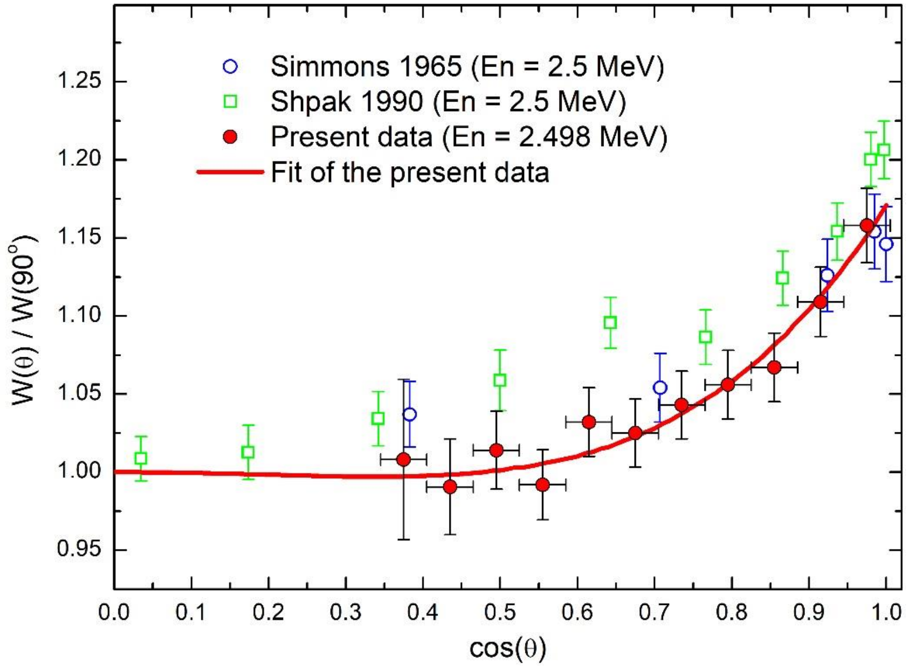
The geometrical efficiency of detection of fission fragments by the array of two MWPCs was calculated using the Monte Carlo method taking into account

- the actual geometry of the MWPCs,
- the size of the active spot on the target separated by the “mask”,
- the spatial resolution of the MWPCs.

The fission fragment detection geometrical efficiency was ~43%, and the maximum fragment detection angle relative to the normal to the MWPC electrode plane was ~72°.

Results: angular distributions of FF from $^{242}Pu(n,f)$





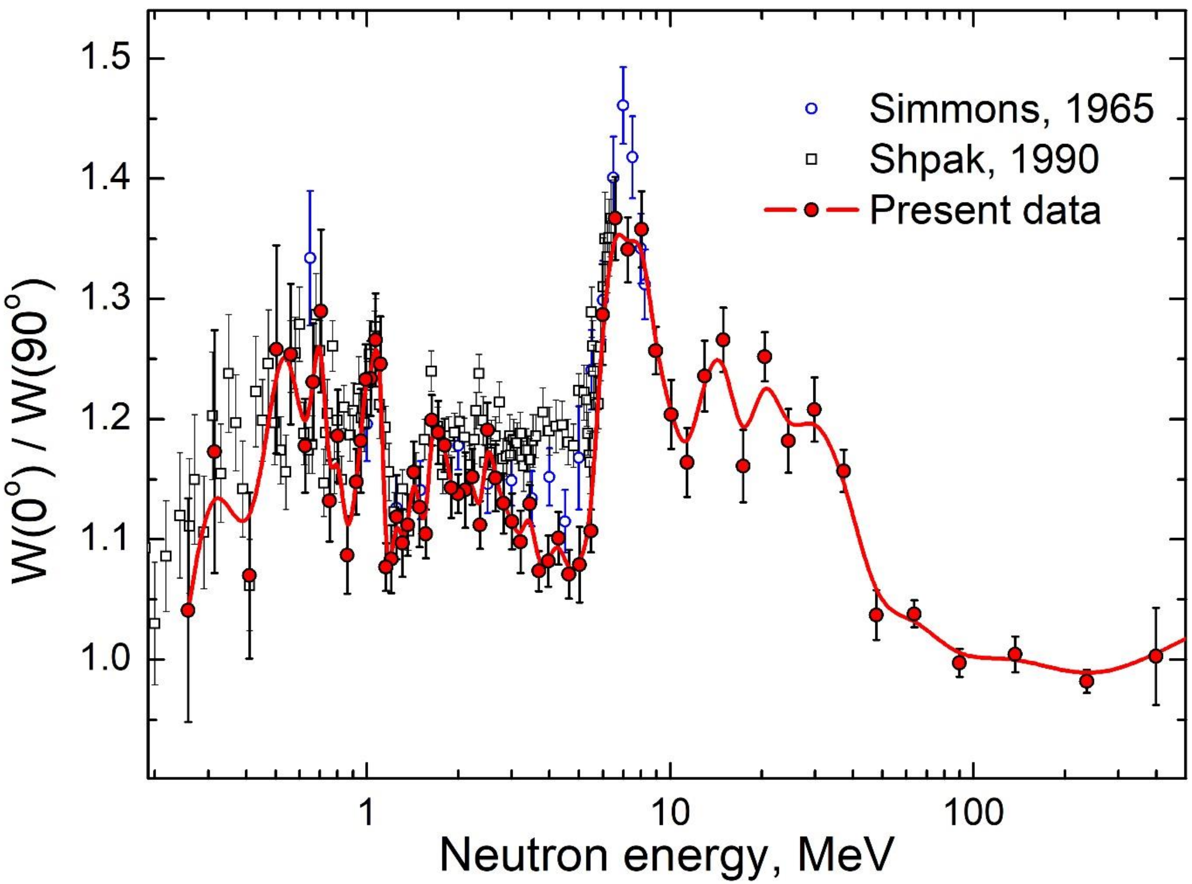
For each neutron energy point E_n, the angular distributions $W(\theta)$ measured at cos(θ) > 0.35 were approximated by the function of the sum of even Legendre polynomials up to the 4th degree :

$$W(\theta) = A_0 \left[1 + \sum_{n=1}^{2} A_{2n} P_{2n}(\cos(\theta)) \right]$$

The anisotropy of the angular distribution of fission fragments is determined using the coefficients A_2 and A_4 for the corresponding Legendre polynomials :

$$W(0°)/W(90°) = \frac{1 + A_2 + A_4}{1 - \frac{1}{2}A_2 + \frac{3}{8}A_4}$$

Results: anisotropy of FFs emission in ^{242}Pu



$$W(\theta) = A_0 \left[1 + \sum_{n=1}^{2} A_{2n} P_{2n}(\cos(\theta)) \right]$$

$$W(0^\circ)/W(90^\circ) = \frac{1 + A_2 + A_4}{1 - \frac{1}{2}A_2 + \frac{3}{8}A_4}$$

The indicated errors are statistical.

The systematic error in determining the anisotropy in this experiment, which is related to the finite angular resolution of the MWPC arrays and the uncertainty in the geometry of the experiment, is ~0.5%.

The systematic error associated with the approximation used for fitting is 1-1.5%

- ✓ Only two data sets are available to date: by Simmons et al. (Van der Graaff, Los Alamos, USA), and Shpak et al. (Van der Graaff, IPPE, Russia), where the anisotropies in the fission of ^{242}Pu were measured for discrete energies up to ~ 8 MeV.
- ✓ One can say that the results of our measurements agree satisfactorily with the previous data. Above 8 MeV measurements were performed for the first time.

Status of the anisotropy of FFs emission measurements for 2024

	GNEIS, PNPI	n-TOF, CERN	NIFFTE, WNR, LANSCE
^{232}Th	JETP Letters, 102, 203 (2015) EXFOR #41608002	Nucl. Data Sheets, 119, 35 (2014) EXFOR #23209	
^{233}U	JETP Letters, 104, 365 (2016) EXFOR #41616006		
^{235}U	JETP Letters, 102, 203 (2015) EXFOR #41608003	EPJ Web of Conf. 111, 10002 (2016)	D. Hensle et al., Phys. Rev. C 102, 014605 (2020) EXFOR #14660002
^{236}U	Phys.Rev.C 108, 014621 (2023) EXFOR #41757002		
^{238}U	JETP Letters, 102, 203 (2015) EXFOR #41608004	EPJ Web of Conf. 111, 10002 (2016)	D. Hensle et al., Phys. Rev. C 102, 014605 (2020) EXFOR #14660003
^{237}Np	JETP Letters 110, 242 (2019) EXFOR #41686002		
^{239}Pu	JETP Letters, 107, 521 (2018) EXFOR #41658003		
^{240}Pu	JETP Letters, 112, 323 (2020) EXFOR #41737002		
^{242}Pu	Measurements completed		
^{243}Am	Proc. of the ISINN-29, 236 (2023) Preliminary results		
^{nat}Pb	JETP Letters, 107, 521 (2018) EXFOR #41658004		
^{209}Bi	JETP Letters, 104, 365 (2016) EXFOR #41616007		

12 nuclei are measured
10 are published and presented in EXFOR

Ratio of the fission cross sections of ^{242}Pu and ^{235}U – corrections and uncertainties

We have determined precisely the scaling factor N_{Am3}/N_{U5} - the ratio of the number of ^{242}Pu and ^{235}U nuclei in the targets.

Both of these targets were placed in a wide and uniform neutron flux, and fission fragments were registered by the same pair of the detectors with the same efficiency.

Corrections were applied:
– for the geometrical efficiency dependence on angular anisotropy of fragment emission,
– for the isotopic composition of the targets,
– and for the background from spontaneous fission events.

Statistical uncertainties	60–3 % (0.2-0.9 MeV) 2–3 % (above 0.9 MeV)
Attenuation of the neutron flux	<0.3 %
Anisotropy	~1 %
Purity of targets (isotope composition)	~ 0,1 %
Efficiency of MWPCs (geometrical uncertainty)	0.3 %
Scaling factor (N_{Am3}/N_{U5})	1.7 %
Total error	~3.2 %
Uncertainty of the ^{235}U "standard" cross section (Carlson et al., Nuclear Data Sheets 148, 143, 2018)	
$\sigma_f(^{235}U)$	1.3–1.5 % (below 20 MeV) 1.5–4.8 % 20–200 MeV 5–7 % (above 200 MeV)

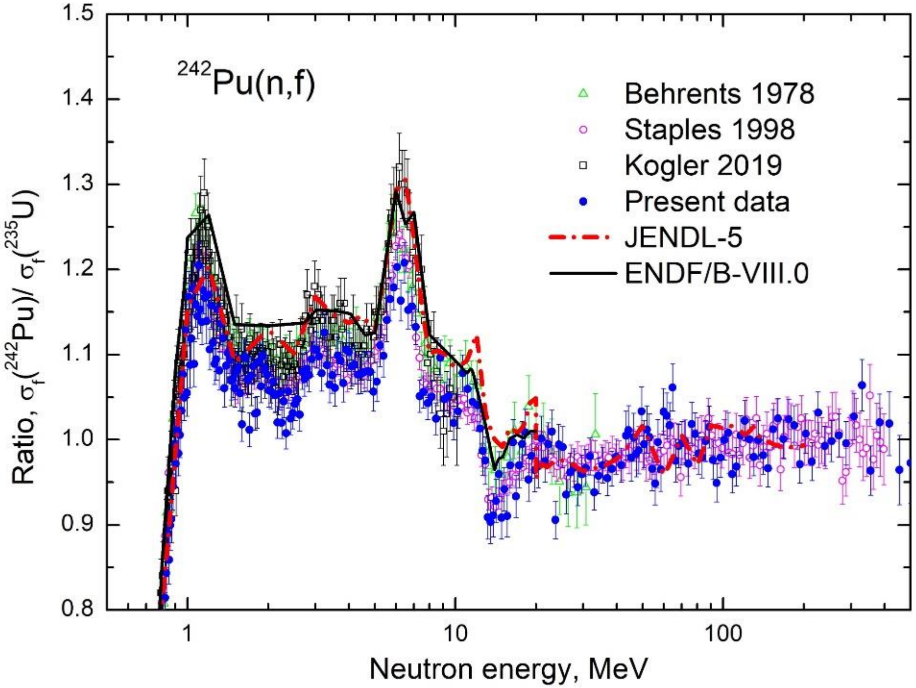
The total average systematic measurement error is ~2% and it is mainly determined by

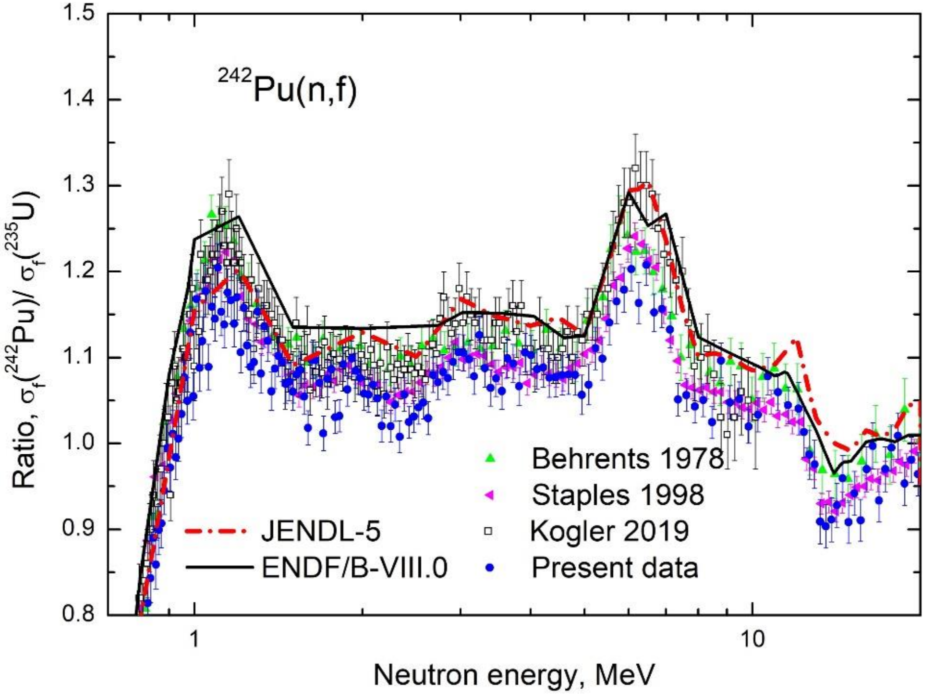
the uncertainty of the correction for the anisotropy of fragment emission ~1.0%

and the uncertainty of the scaling factor determination ~1.7%.

Total error is estimated as ~3.2%.

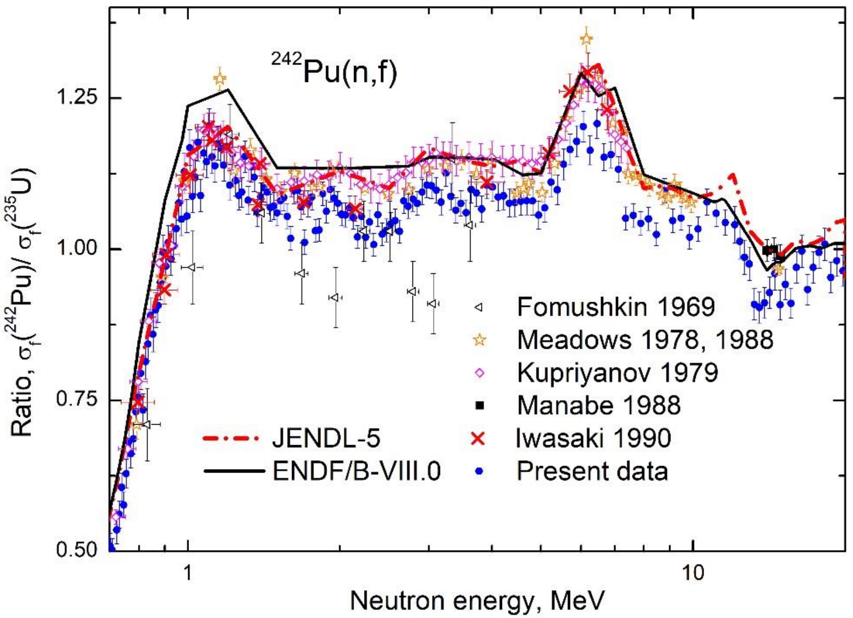
Ratio of the fission cross sections of ^{242}Pu and ^{235}U according to our measurements and other experimental data (TOF) taken from the EXFOR database

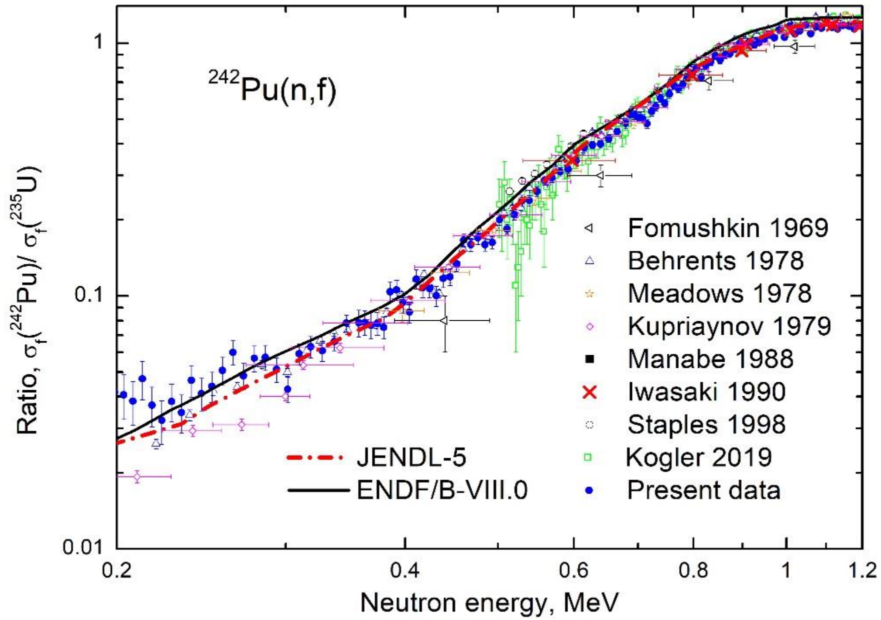




- The results of time-of-flight measurements of ^{242}Pu to ^{235}U cross sections ratio are presented.
- One can notice that JENDL-5 and especially ENDF/B-VIII.0 estimates are higher than most experimental data
- The shapes of the experimental energy dependences obtained in the time-of-flight measurements are very similar
- One can say that the results of all time-of-flight measurements relative to ^{235}U coincide within the experimental uncertainties.

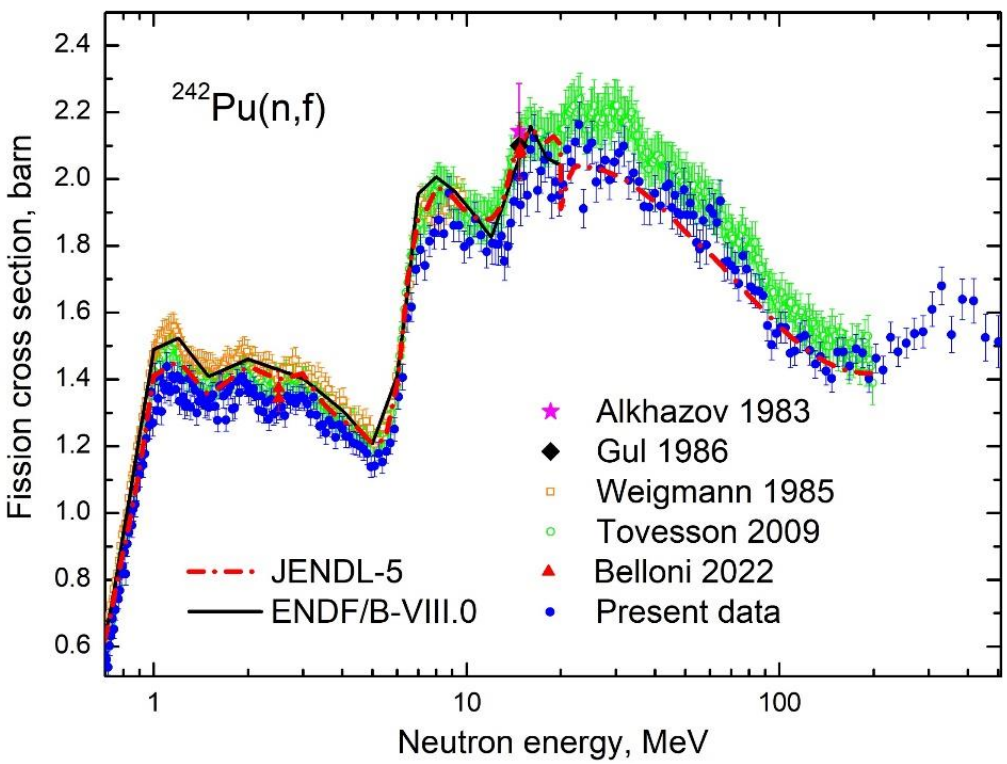
Ratio of the fission cross sections of ^{242}Pu and ^{235}U according to our measurements and other experimental data (Discrete Energies on Accelerators) taken from the EXFOR database





- The results of ^{242}Pu to ^{235}U cross sections ratio measurements obtained using monoenergetic neutrons produced in various reactions at accelerators are presented on the left figure
- On the right figure all data sets available for comparison in the energy region <1.2 MeV are shown
- One can see that JENDL-5 evaluation follows strictly through results of Kupriyanov_79 (IPPE, Obninsk, Russia)
- Results of our TOF measurements above 1 MeV are ~5 % lower than the nowadays library evaluations

Fission cross sections of ^{242}Pu obtained in this work and from other experiments (total errors are shown). The solid and dashed line consist of the estimates from the ENDF/B-VIII.0 and JENDL-5 libraries

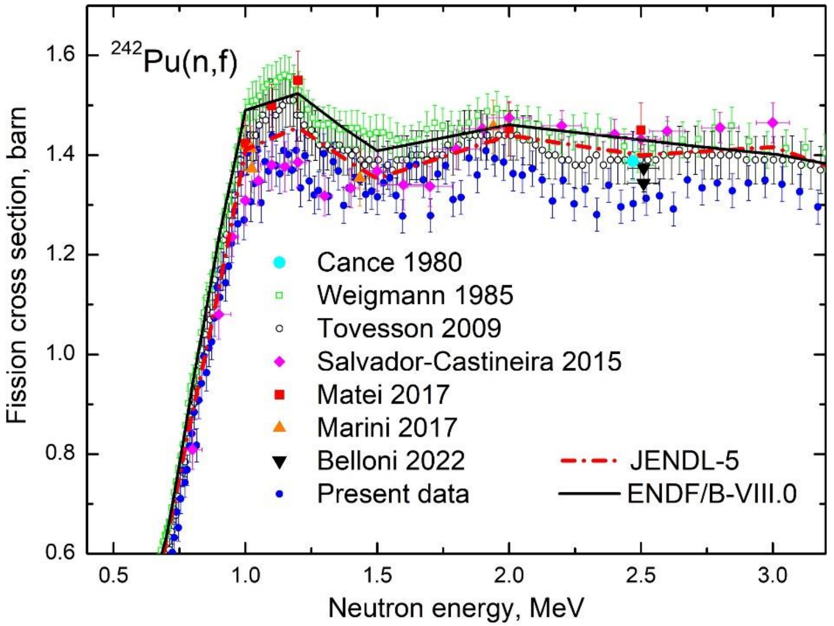


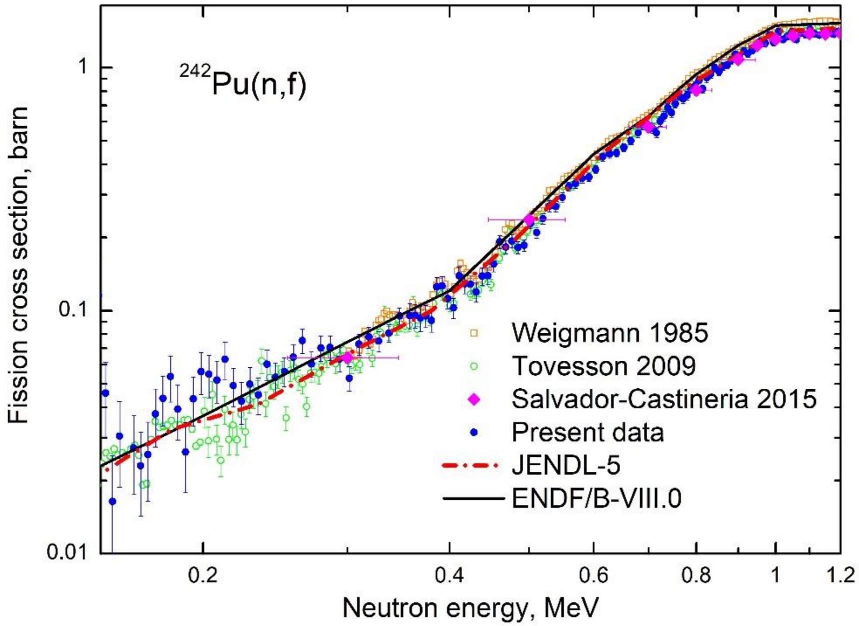
For our data shown in the figure, the ^{242}Pu cross section was determined as the product of the measured ratio R and the $\sigma_f(^{235}U)$– existing standard of the ^{235}U(n,f) fission cross section.

Another cross-section data presented here were obtained in experiments of various types. For example: Alkhazov_1983 and Gul_1986 – method of accompanying particles (14 MeV);
Tovesson_2009 – measurement of the ratio of cross section relative to ^{239}Pu (TOF);
Weigmann_1985 – measurement of the ratio of cross section relative to ^{239}Pu (TOF);
Belloni_2022 – measured relative to n-p scattering and relative to ^{238}U(n,f) (acc-or)

- One can see that ENDF/B-VIII.0 evaluation follows Weigmann_1985 data which were the only available, and, probably, the evaluation can be revised nowadays
- Our data are in reasonable agreement with Tovesson_2009 results

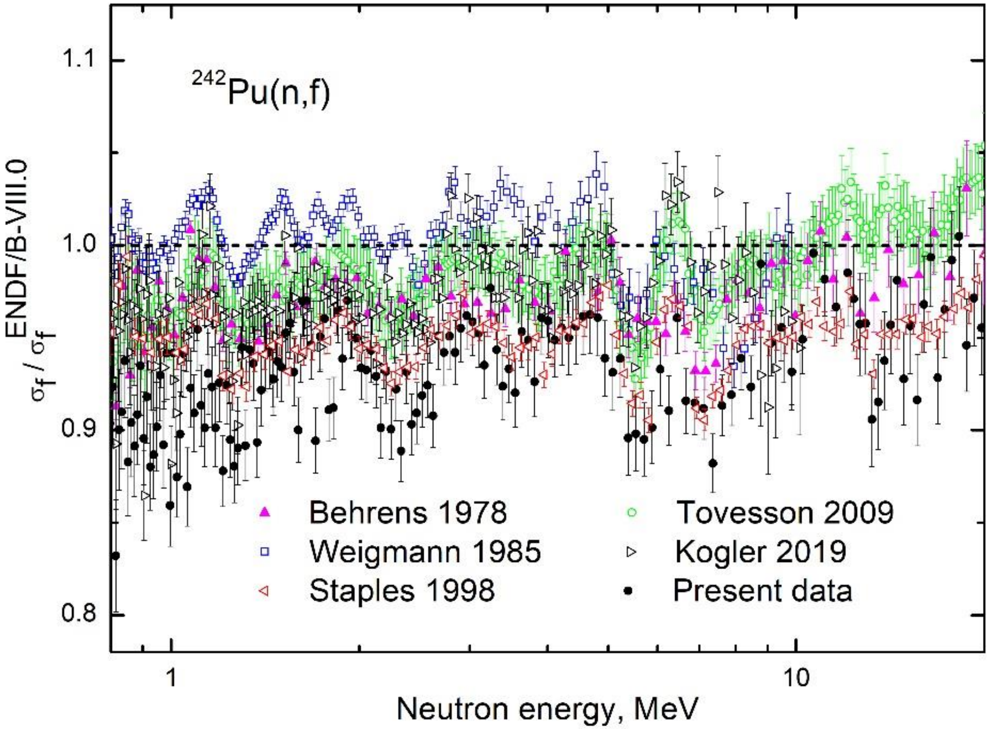
Fission cross sections of ^{242}Pu obtained in this work and from other experiments (total errors are shown). The solid and dashed line consist of the estimates from the ENDF/B-VIII.0 and JENDL-5 library – low neutron energies





- Fission cross sections from our and another experiments for low energy neutrons are shown
- The results of recent discrete energies accelerator measurements (Salvador-Castineira_2015, Marini_2017) are consistent with our data in the region En<1.5 MeV
- The shapes of cross section energy dependence in all TOF measurements are very similar, while the shapes of energy dependence in discrete energies accelerator measurements are different

Ratio of the ^{242}Pu fission cross sections obtained in this work and in other TOF measurements to the estimate for this cross section from the ENDF/B-VIII.0 library



In the neutron energy range 1–20 MeV, the ratio between the TOF experimental data and the estimate from ENDF/B-VIII.0 is approximately constant.

The average deviation for all data doesn't exceed the experimental accuracy of determining the scaling factor associated with the target masses, the detection efficiency and the neutron flux.

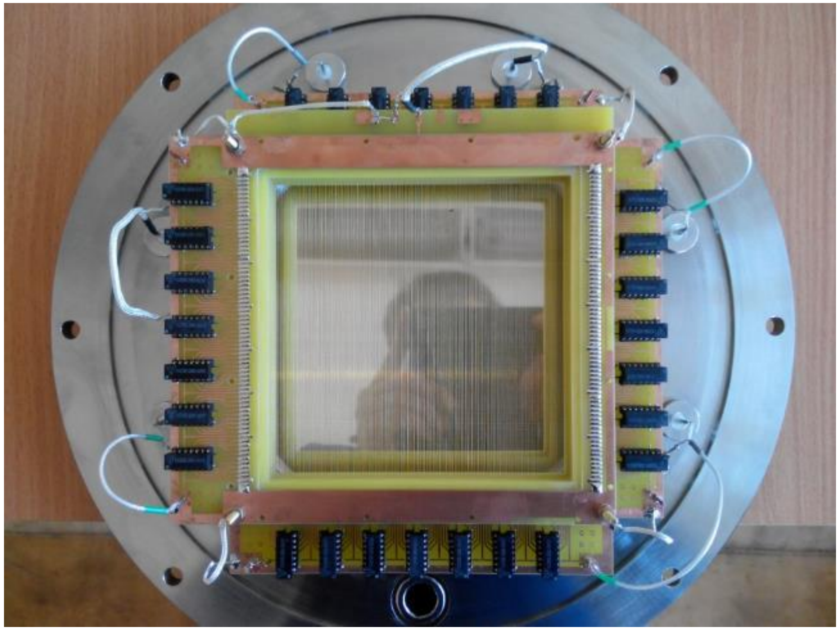
This behavior indicates that the shape of the ^{242}Pu fission cross section from the ENDF/B-VIII.0 library quite correctly describes the available experimental data, and the observed difference in some data is apparently due to the inaccuracy of the absolute normalization of the measured ratios of the fission cross sections of ^{242}Pu and ^{235}U in these works.

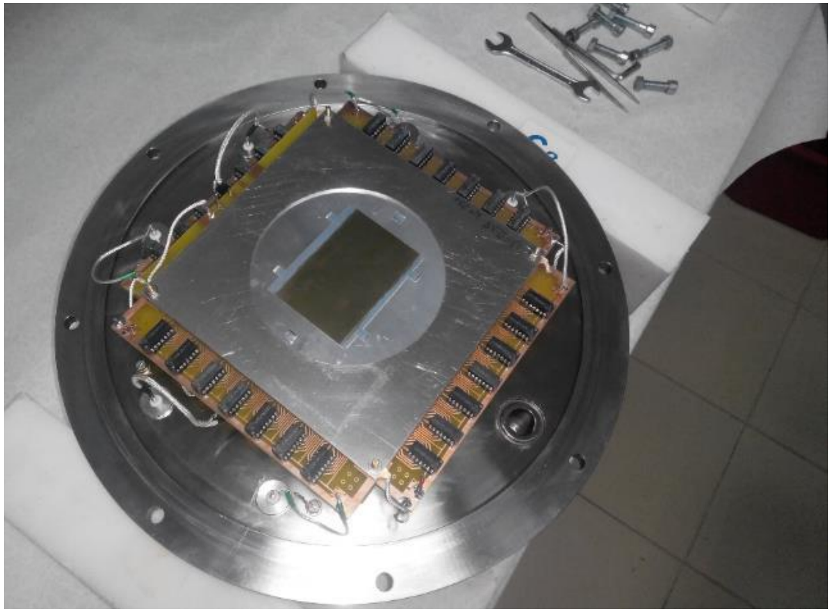
Summary

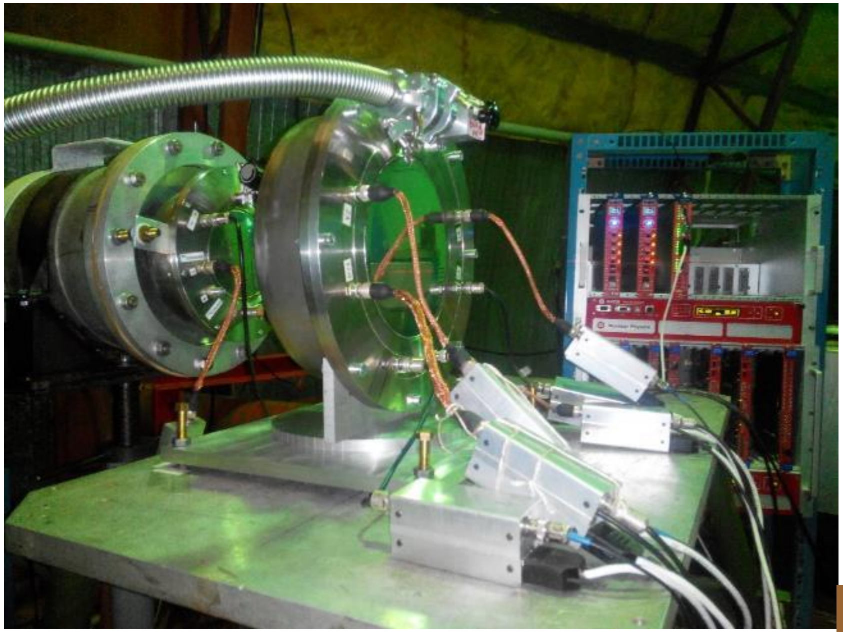
- In this work the fission cross section of ^{242}Pu is determined by the ratio method using ^{235}U as a reference. The measurements were carried out on the neutron TOF-spectrometer GNEIS at Petersburg Nuclear Physics Institute of National Research Centre «Kurchatov Institute» in the neutron energy range up to 500 MeV.
- The neutron induced fission cross section of ^{242}Pu was obtained in a wide energy range with the experimental uncertainty 3-4%.
- The shape of the fission cross section energy dependence obtained in this work is mostly consistent with the results of all earlier data obtained in TOF experiments.
- The differences between all existing TOF experimental data seem to be mostly related to uncertainties in the detection efficiency of the fission fragment detectors used, the neutron flux, and the target masses.
- The shapes of the fission cross section energy dependence in discrete energies accelerator measurements are different. This can be attributed to systematical errors of a different nature.
- The angular distributions of ^{242}Pu fission fragments were measured in the energy range 0.3-500 MэB, and above 8 MeV they were measured for the first time.

Thank you for attention

Experimental setup

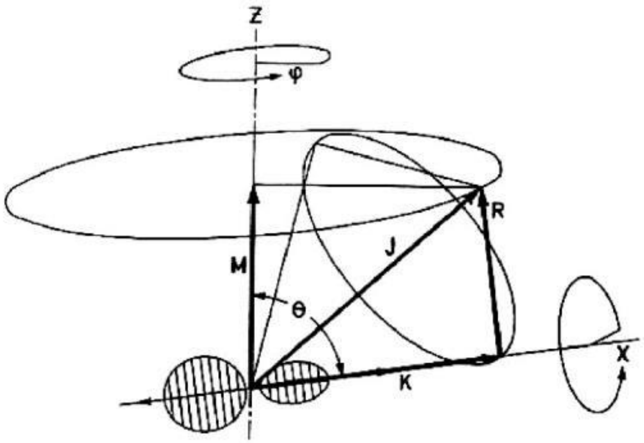






Transition states at the saddle point of highly deformed fissioning nuclei:

$$W^J_{M,K}(\theta) = \frac{2J+1}{2}\left|d^J_{M,K}\right|^2$$ (wave function of axial top)



For low excitation energies we need a proper sum over non-uniform *M* distribution, and few available *J*, *K* (fission channels) and finally:

$$W(\theta) \sim \sum_{n_{even}} A_n P_n(\cos\theta)$$

At high excitations with many opened fission channels one can use statistical model for the *K* projection distribution – $\rho(K)$:

$$\rho(K) \sim \exp\left(-\frac{E_{rot}}{T}\right); \quad E_{rot} = \frac{\hbar^2[J(J+1)-K^2]}{2J_\perp} + \frac{\hbar^2 K^2}{2J_\parallel}; \quad T = \sqrt{E^*/a_f}$$

$$\rho(K) \sim \exp\left(-\frac{K^2}{2K_0^2}\right)$$, where $K_0^2 = \frac{J_{eff}T}{\hbar^2}$, and $J_{eff} = \frac{J_\perp J_\parallel}{J_\perp - J_\parallel}$

In statistical model:

$$W(\theta) \sim 1 + A\cos^2\theta \qquad \frac{W(0^o)}{W(90^o)} = A + 1 \approx \frac{\langle J^2\rangle}{4K_0^2} + 1$$