ANGULAR AND ENERGY DISTRIBUTIONS OF THE PROMPT FISSION NEUTRONS FROM THERMAL NEUTRON-INDUCED FISSION OF ^{239}Pu

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Abstract

The measurements of angular and energy distributions of the prompt fission neutrons from thermal neutron-induced fission of ^{239}Pu were carried out at the WWR-M research reactor in Gatchina, Russia. Some peculiarities were found in the angular distribution of the prompt fission neutrons. It is possible to explain them by assuming that in the centre of mass system of fission fragment the neutrons are more likely emitted along fission direction than in the perpendicular to one. The value of anisotropy of neutrons emission in the center of mass system of fission fragment was obtained and was equal to ~6%. Also the yield of "scission" neutrons and their spectrum have been estimated.

Introduction

Up to now many theoretical and experimental works were performed to investigate the low energy nuclear fission. A special attention was given to the details of the prompt fission neutron (PFN) emission: spectra and multiplicities, their dependence on fission fragment (FF) characteristics and all possible correlations between reaction products. These data are used widely for the construction of nuclear reactors and applied for the development of non-destructive methods of nuclear safety and for the control of non-proliferation of nuclear materials.

In spite of a notable advance achieved in theoretical description of the prompt fission neutron properties, still there are some problems (for example, [1, 2]). The difference observed between measured and calculated data is probably due to both an inadequacy of theoretical model used for this description and a deficit of experimental data. Because the properties of neutrons emitted before fragments have been accelerated (emission before and during the rupture of fissioning nucleus or at the initial stage of acceleration of the fragments in the Coulomb field – so called "scission" neutrons) are not established experimentally, in theoretical calculations it is usually assumed that the main part of prompt fission neutrons are emitted from accelerated fission fragments. Experimental studies dedicated to ascertain the mechanism of the prompt neutron emission are limited to spontaneous fission of ^{252}Cf and thermal neutron-induced fission of ^{235}U. For ^{239}Pu these investigations are limited to works shown in Table 1. Also, it ought to mention that up to now the energy spectra of the prompt fission neutrons are calculated using the semi-empirical systematics where an absence of information about the mechanism of emission of additional neutrons is compensated by the artificial variation of the nuclear model parameters. This circumstance significantly complicates production of the evaluated data files for the nuclei and energy ranges where experimental data are absent. That is why a new experimental investigation of the mechanism of fission neutrons emission will provide a good basis for future evaluations and enable to increase their accuracy and reliability.

Therefore, to clear up how well the model calculation can describe or predict the prompt fission neutron properties it is necessary to improve the quality of data obtained by differential as well as integral experiments. A series of such experiments have been carried out in PNPI of NRC KI (Gatchina, Russia) [3-7]. In this paper some results of this investigation are presented.

Table 1. Main results of previous investigations of neutron emission mechanism for ^{239}Pu(n,f).

Author, References Experimental Set-up	Yield of "scission" neutrons	Average energy of "scission" neutrons	Anisotropy of PFN emission in c.m.s of FF, A
Investigation of (n,f)-angular correlation			
J.S. Fraser *et.al.* [8] (1965). Two plastic scint. for FFs spectroscopy (TOF with the base of 125cm and 99cm). Four neutron detectors (plastic scint.) were used, TOF (106cm). The neutron spectra measurements have been done simultaneously at 10°, 25°, 45° and 80° relative to FFs direction.	30%	~ 2 MeV	"… all results are consistent with A = 0."
Yu.S. Zamyatnin *et.al.* [9] (1979). IC with collimator used for FFs spectroscopy. One neutron detector (plastic scint.) was placed interchangeably at 0° and 90° relative to FFs direction, TOF (40cm).	20 ± 12%	---	Not investigated
Investigation of (n,n)-angular correlation			
I.S. Guseva *et.al.* [10] (2017). Two stilbene neutron detectors, n/γ pulse shape discrimination.	4.0 ± 1.5%	1.8 ± 0.2 MeV	Weakly sensitive

1. Experiment overview

The angular and energy distributions of prompt fission neutrons were measured in turn for neutron-induced fission of ^{239}Pu (beam on) and the spontaneous fission of ^{252}Cf (beam off) under identical experimental conditions. The measurements were carried out using the collimated neutron beam №1 of the research reactor WWR-M (Gatchina, Russia) with a flux of ~ 10^8 thermal neutrons/ $\text{cm}^2 \cdot \text{sec}$. The ^{239}Pu target was deposited on 100 μm thick Al backing. The target thickness was 150 $\mu\text{g/cm}^2$ and made in the form of a circular spot 15 mm in diameter. A ^{252}Cf layer 10 mm in diameter was made on a 0.18 mm thick stainless steel foil. The fission fragments and prompt neutrons time-of-flights were measured simultaneously for 11 fixed angles, θ, between the axis of neutron detector and normal to the stop multi-wire proportional detectors (MWPDs) surface (coming through its center) in the range from 0° to 180° in 18° intervals. The schematic view of the experimental set-up is shown in Fig. 1.

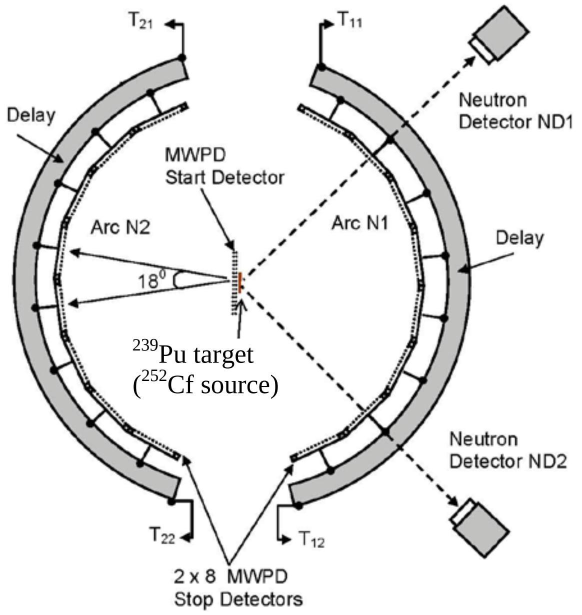

Fig.1. Schematic view of the experimental setup

The neutron beam was coming along the chamber axis normally to the Fig.1 plane. It should be noted that realized scheme of the experimental set-up guarantees identity of conditions of the neutron spectra measurements at various angles relative to the fission axis, namely: the magnitude and composition of the background, the efficiency of the neutron detectors, and neutron re-scattering by the parts of experimental set-up. Also, the use of two neutron detectors with slightly different characteristics allows to estimate probable systematic errors of the data obtained.

The prompt neutrons were detected using two stilbene crystal detectors (Ø 50 mm x h 50 mm and Ø 40 mm x h 60 mm) positioned at a 90° angle between their respective axes at a distance of (47.2±0.2) cm and (49.2±0.2) cm, respectively, from the fissile target. The axes of neutron detectors ND1 and ND2 come through the centers of two stop MWPDs located on the Arc N1. Both neutron detectors were surrounded by a cylindrical shield made of 30 mm thick layer of lead and 40 mm thick layer of polyethylene (not shown in Fig. 1). The neutron registration threshold was 150 – 200 keV. To separate events corresponding to neutrons and γ-quanta, a double discrimination by the pulse shape and time-of-flight was applied. The full time uncertainties were defined from FWHM of the "fragment - γ-quantum" coincidence curve which was equal to 1.0÷1.2 ns.

The fission fragments were detected by MWPDs in conjunction with the TOF technique. The 8 rectangular MWPDs were located in the Arc N2 in the reaction chamber at the operating gas (isobutane) pressure of 4 ÷ 6 Torr.

As a result, for 11 fixed angles between neutron and light fragment directions the energy distributions of prompt neutrons emitted from fixed pair of fission fragments were obtained. During data processing, the following corrections were taken into account:

- for the fragment detector efficiency;
- for incomplete separation of light and heavy group of fission fragments;
- for angular and energy resolution of experimental setup;
- for neutron detector background and the neutron background due to accidental coincidence between fragment and neutron belonging to different fission events;
- the normalization correction arising from the fact that experimental angular histograms were used in the measurements instead of continuous distributions;
- for the neutron detector efficiency. The detector efficiency was determined for each neutron detector independently as the ratio of the obtained total prompt fission neutron spectrum (PFNS) of ^{252}Cf to a reference standard spectrum from ref. [11]. The total PFNS, were calculated by summing up the obtained angular-energy distributions over angle in the laboratory system.

In order to determine the PFNS from measured time-of-flight spectra, the relativistic equation was used. A description of the experimental method and the used data processing are omitted here since a full treatment is presented in ref. [3, 5, 6].

2. Model

In the model calculation it is used the assumption that PFN are emitted from fully accelerated fragments. In this case the angular and energy distributions of PFN in the laboratory system can be calculated using known spectra of PFN in the center-of-mass system of fragment. Since the fission fragments have a large angular momenta (~ $7\hbar$ on average), which is usually considered to be normal to the fission axes (for example, Ref. [12]), the neutron emission anisotropy in the center-of-mass system of fragment should be included into the model calculation [13, 14]. The spectra of PFN in the center-of-mass system of fragment are calculated using experimental data for small angles (8.9°, 19.8° and 36.9°) relative to the fission direction. During this calculation it was assumed that prompt neutrons are emitted by two fragments with average mass and kinetic energy. The average energy per nucleon for light and heavy fragments were taken as $<E_L> = 0.995\pm0.007$ MeV and $<E_H> = 0.511\pm0.004$ MeV for ^{239}Pu(n_{th}, f). Further, the spectra obtained in the center-of-mass system are used for calculation of neutron angular and energy distributions in the laboratory system. These distributions are compared with the experimental distributions to estimate contribution and properties of “scission” neutrons.

It should be noted that the calculated spectra are free of any assumption about the prompt neutron spectra in the center-of-mass system (the number of neutrons emitted by heavy and light fragments, the neutron spectrum shapes, and so on). There is only one free parameter the anisotropy of PFN in the center-of-mass system of fragment, which is adjusted so as to describe in the best way all experimental data obtained in this investigation. The details could be found in ref. [3, 15].

The shape of the neutron spectrum and the number of neutrons obtained in the center-of-mass system both depend on the fragment velocities (or E_L and E_H for fission event). Therefore, strictly speaking, the analysis performed above is not valid, because it was assumed that the prompt neutrons are emitted only from two fragments (light and heavy) characterized by the average parameters. Fortunately, as it was demonstrated for total PFNS of ^{252}Cf in Ref. [15], a transition from the velocity distributions of fragments to the model of two fragments with average parameters has only a minor influence, and for angles near 90° the neutron yield changes within 4% [5].

At the same time, the existing calculation methods used in practice to describe angular and energy distributions of PFNS do not provide necessary accuracy. For example, the total PFNS of ^{235}U calculated by different commonly used codes [2] are presented in Fig. 2, where spectra calculated assuming that PFN are emitted from fully accelerated fragments and using the same input parameters are shown as a ratio to Maxwell distribution. It is seen that the existing calculation methods do not provide necessary accuracy to describe experimental data while the method realized in this work gives accuracy not worse than those of commonly used codes and does not require knowledge of a large number of input parameters.

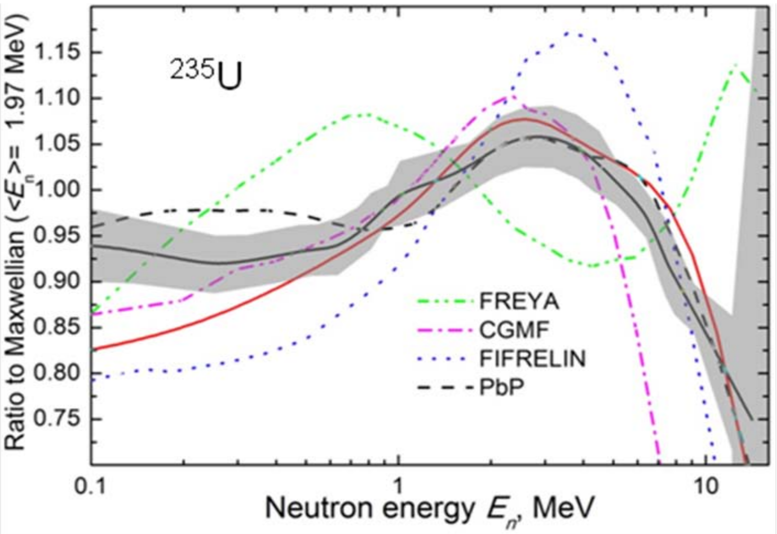

Fig. 2. Total PFNS of ^{235}U(n_{th}, f): curve inside the shaded region – evaluation of experimental data within error corridor (GMA – generalized least square fit [2]); line – model calculation (two fragments approximation) [2]; **PbP** (Point by Point) - deterministic method developed at the University of Bucharest and JRC-IRRM team, which is an extended version of LAM (Los-Alamos or Madland-Nix model); **FREYA** (Fission Reaction Event Yield Algorithm) – Monte-Carlo fission model developed through a collaboration between LLNL and LBNL (USA); **CGMF** – Monte-Carlo code developed at LANL (USA); **FIFRELIN** (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.

3. Results and discussion

The PFNS for fixed angles in the laboratory system (obtained experimentally and calculated using an assumption that PFN are emitted from fully accelerated fragments with anisotropy parameters $A_2 \approx 0.04$ ($N(0°)/N(90°)=(1+A_2)/(1-A_2/2)$) are shown in Fig. 3. The yield and average energies of these PFNS are presented in Fig. 4.

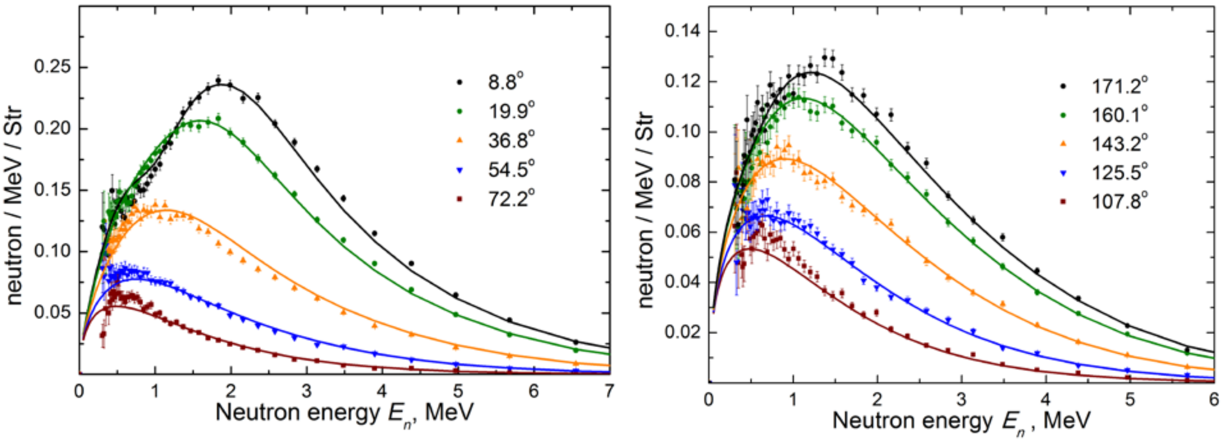

Fig. 3. The comparison of prompt fission neutron spectra measured for fixed angles relative to the direction of motion of the light fragment and calculated ones.

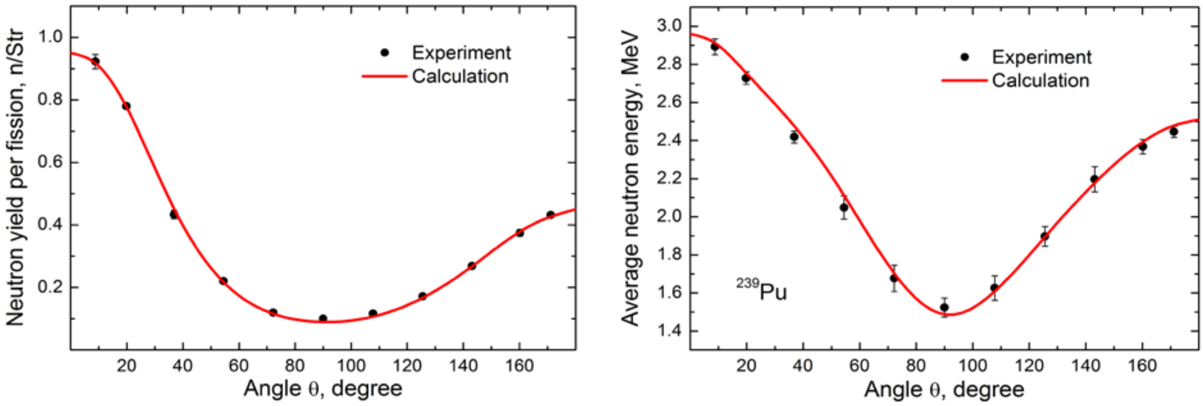

Fig. 4. Left – the prompt fission neutron yield as a function of the angle between neutron flight direction and the direction of motion of the light fragment. Right – the angular dependence of the average neutron emission energy in the laboratory system.

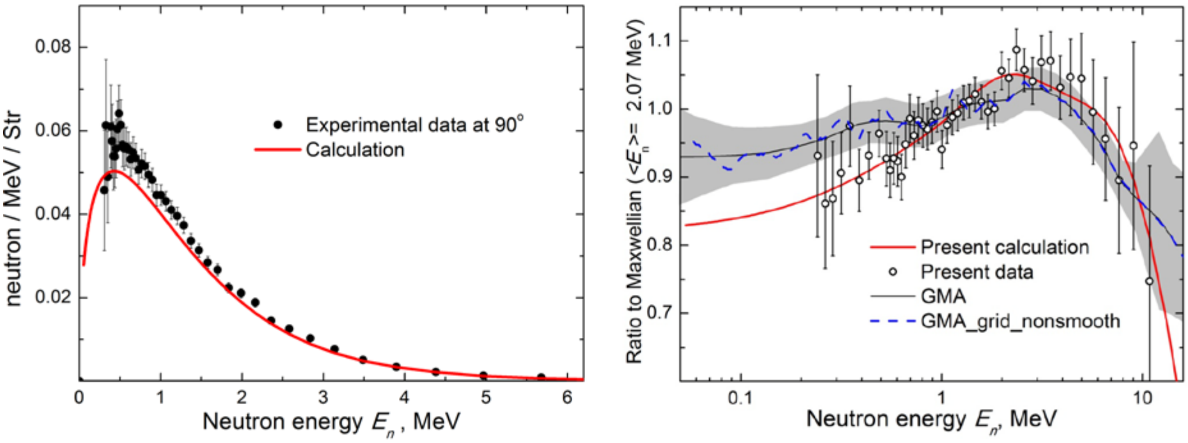

Fig. 5. Left – the PFNS measured for angle 90° relative to the fission fragment direction. Right – the total PFNS of ^{239}Pu(n,f) obtained by summing over angles θ are shown as a ratio to Maxwell distribution. GMA – generalized least square fit of prompt fission neutron spectra measured by different experimental groups (non-model evaluation) – taken from ref. [2].

The experimental and model neutron spectra have been compared in 0.2–10 MeV energy range. On the whole, the calculated model energy and angular distributions agree rather well with the experimentally obtained distributions. However, there is a minor distinction which is most clearly demonstrated at Fig. 5, where PFNS measured for angle 90° relative to fission fragment direction and the total PFNS obtained by summing over angles are compared with the corresponding calculated values. Note that the obtained total PFNS are in agreement with evaluated spectrum (GMA fit [2]) within experimental errors and, therefore, it can be said about the absence of any significant systematic measurement errors in our investigation. Then, the observed differences may be interpreted as a manifestation of "scission" neutrons and the average energy of these neutrons and their yield can be estimated.

The systematic difference of calculated total PFNS from total PFNS measured by different experimental groups (evaluated spectrum – GMA fit) is visible in the neutron energy range lower than 0.6 MeV. The “scission” neutron spectrum obtained as a difference between evaluated total PFNS (GMA fit) and model calculation is shown in Fig. 6 as a line with error corridor. To verify this statement, the PFNS measured for angles close to 90° relative to the direction of the light fragments’ movement, were compared with calculated PFNS at the same angles. And the “scission” neutron spectrum was obtained with the use of the difference spectra obtained as the difference between the measured and model spectra for angles of 72.2°, 90° and 107.8° with respect to the direction of motion of the light fragment. There is a good agreement within experimental uncertainties between spectra of “scission” neutrons obtained by two different ways (see Fig. 6). Further, the spectrum of scission neutrons found from partial data was approximated by the least squares method by two functions:

$$p_S(E) = \frac{p_0}{4\pi} \cdot \frac{E}{{T_0}^2} \cdot \exp\left(-\frac{E}{T_0}\right) \tag{1}$$

$$p_S(E) = \frac{p_0}{4\pi} \cdot \frac{E}{{T_0}^2} \cdot \exp\left(-\frac{E}{T_0}\right) + \frac{p_1}{4\pi} \cdot \frac{E}{{T_1}^2} \cdot \exp\left(-\frac{E}{T_1}\right) \tag{2}$$

The parameters p_0, T_0, p_1 and T_1 were varied. These two functions describe well the experimental data. The approximation Eq.(1) shown in Fig. 6 provides the following parameters of the spectrum of scission neutrons: the fraction of scission neutrons in the total number of prompt fission neutrons per fission event and the average energy of scission neutrons are equal to 3.6 ± 0.5 % and 0.9 ± 0.19 MeV, respectively. When Eq.2 is used, the corresponding values are 4.5 ± 0.9 % and 1.6 ± 0.2 MeV.

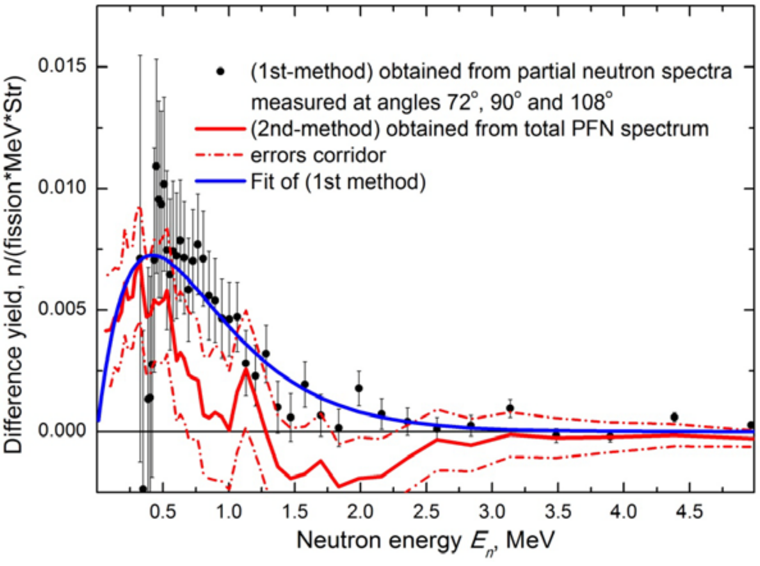

Fig. 6. Spectrum of “scission” neutrons for ^{239}Pu(n,f). Circles - the difference spectrum obtained using spectra measured at 72°, 90° and 108° relative to the direction of motion of the light fission fragment and the corresponding ones calculated under the assumption that all prompt neutrons are emitted from the accelerated fragments. Broken line - the difference between total PFNS obtained by experiment (estimated data and it’s errors) and calculated assuming that all prompt neutrons are evaporated from accelerated fragments. Solid line - fit of experimental data marked with circles by the equation (1).

It should be noted that these estimations of properties of “scission” neutrons were performed assuming isotropic emission of “scission” neutrons in the laboratory system. Probably, this assumption is very close to the real situation, because in the measurements of the angular dependency of the neutron-neutron coincidence curves (see table 1, ref. [10]), which are very sensitive to isotropic component in the laboratory system, the same values of “scission” neutron yield were obtained within experimental errors.

Conclusion

The angular and energy distribution of the prompt neutrons for ^{239}Pu have been measured.
A comparative analysis of the obtained angular and energy distributions of prompt neutrons from ^{239}Pu and calculated ones enabled to make the following conclusions:

- the angular anisotropy of the neutron emission in the fragment center-of–mass system should be taken into account;
- there are some surplus of measured neutron yield above calculated one in low energy range for the total prompt fission neutron spectrum as well as for neutron spectra at fixed angles near 90^o (relative to fission fragments direction);
- the yield of this low energy component of “scission” neutrons is equal to 3.6 ± 0.5 % of total neutron yield per fission event;
- the maximum contribution of “scission” neutrons do not exceeds 5% of the total neutron yield.

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