EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Measurement of the neutron-induced fission cross section of ^{239}Pu at n_TOF

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Abstract: Accurate ^{239}Pu fission cross section data are required over a large energy range due to the prospect of new nuclear devices and the goal of optimizing nuclear waste management. Indeed, ^{239}Pu has been included in the 2017 Evaluation of the Nuclear Data Standards from 150 eV up to 200 MeV, and the IAEA has demanded more accurate fission data from thermal up to above 200 MeV, where no high-resolution data exists.
We propose an experiment, to be performed in EAR-1 at n_TOF, to measure the ^{239}Pu(n,f) cross section relative to ^{235}U(n,f). The aim of this experiment is to produce a single, accurate data-set covering the energy range from thermal up to 1 GeV. It will include a detailed measurement of the angular distribution of the fission fragments over the whole energy range. The measurement will be performed with the tilted PPACs ensemble, using six ^{239}Pu and three ^{235}U samples. The number of protons requested is driven by

the objective of having around 1% statistical uncertainty on the cross-section ratio in the 10 to 20 MeV neutron energy range.

Requested protons: 4.5×10^{18} protons on target
Experimental Area: EAR1 - fission collimator

1 Scientific Motivation

Neutron-induced fission cross sections have been playing a prominent role and have been systematically measured since the 1940s due to their importance in nuclear security and energy production. Nowadays, any development of nuclear technology is based on nuclear data libraries, carefully built from experimental datasets after an evaluation process.
Over the years, the standards required in reactor safety and efficiency calculations have become more demanding, leading to new measurements with increased precision and a wider range of neutron-induced reactions [1]. In particular, the ambition of a closed nuclear fuel cycle has motivated the studies of Generation IV Fast Neutron Reactors and Accelerator Driven Systems (ADS) [2], thus prompting an extension of the neutron energy range of interest.
The neutron-induced fission cross section on ^{239}Pu is one of the most important reactions contained in the nuclear data libraries, especially in the context of the development of new nuclear technologies. A large set of measurements of the ^{239}Pu(n,f) cross section across the energy range that is considered primarily relevant for direct nuclear applications already exists. However, above 10 MeV, little experimental data are available, and their discrepancy increases significantly.
The ^{239}Pu(n,f) cross section is also of particular importance for its role of reference. Indeed, most of the experimental datasets retrieved from EXFOR [3] come from high-resolution cross-section data measured by the time-of-flight (ToF) method, using as reference either ^{1}H(n,p), ^{235}U(n,f) or ^{239}Pu(n,f) reactions. Even in the high energy region, ^{239}Pu(n,f) is a major reference, together with ^{235,238}U(n,f), ^{209}Bi(n,f) and ^{nat}Pb(n,f) [4]. Therefore, the ^{239}Pu(n,f) reaction has been included in the 2017 Evaluation of the Nuclear Data Standards [4] from 150 eV up to 200 MeV, prompting demand for further ^{239}Pu(n,f) cross section data, to improve accuracy from 10 MeV upwards. Arguments have also been given for the extension of the measurement beyond 200 MeV neutron energy [5], where no data exist a part from an experimental measurement up to 260 MeV [6].
The interval from 8 to 10 MeV is especially relevant here, because it lays between the second and third fission-chance thresholds, an energy region in which the cross section is relatively flat - a feature common to the whole set of actinides involved in nuclear fission applications [7]. The accurate integral value of the cross section in this 8 to 10 MeV interval can be used to normalize the whole energy range above threshold with high precision.

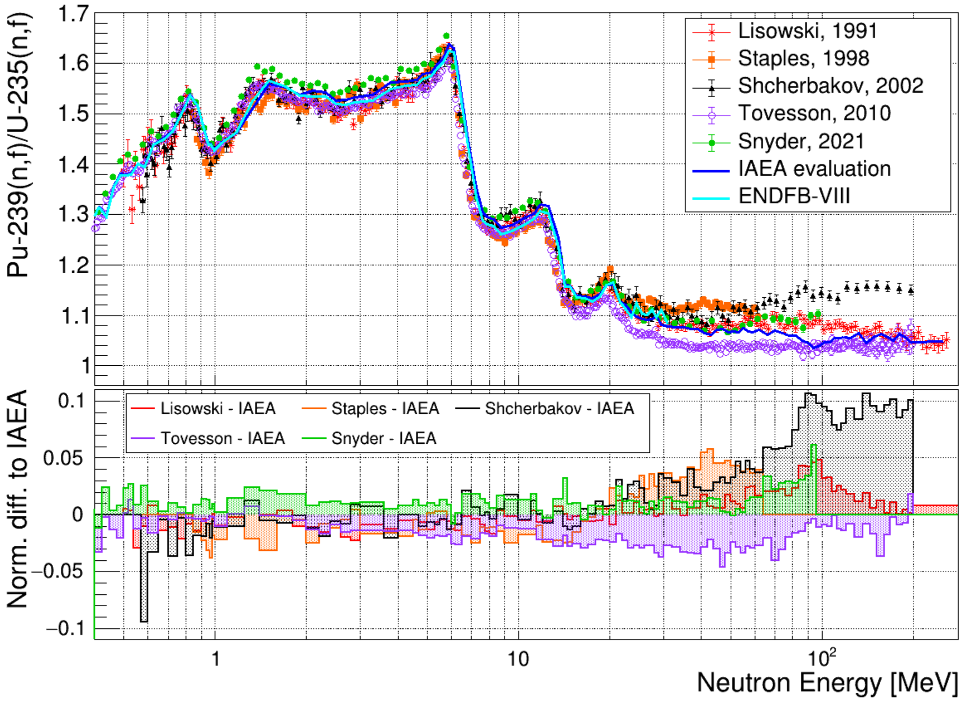

Figure 1: Top panel: more recent measurements of the ^{239}Pu(n,f)/^{235}U(n,f) cross-section ratio together with the ENDF/B-VIII.0 [8] and the IAEA evaluations [5]. The Lisowski [6], Staples [9], Shcherbakov [10], Tovesson [11] and the more recent Snyder [12] work are reported. Bottom panel: normalized difference between the data sets and the IAEA evaluation.

A collection of the data from the most recent measurements of the ratio ^{239}Pu(n,f)/^{235}U(n,f) is reported in Fig. 1. Each of the shown data sets has been declared to have both statistical and systematic uncertainties of the order of 1%. However, it can be easily noticed that the spread between each of these measurements and the IAEA evaluation (one of the most relevant evaluated nuclear data set) is notably higher than the declared uncertainties, an indication that additional and improved experimental activities are suitable.
As it can be seen from Fig. 1, even in the range between 10-20 MeV, which is still required for nuclear data evaluations, the data differs by 3% or more. The situation clearly worsens above 20 MeV. Reducing the uncertainty of the fission cross section on ^{239}Pu, especially in the fast neutron range, is of relevance due to its significance in the nuclear fuel cycle. The neutron beam characteristics (wide energy range and high resolution, amongst others), combined with low background and reduced sensitivity to samples radioactivity, make n_TOF EAR1 a suitable facility for further experimental investigation of this key nuclear reaction.

2 Experimental setup

The main challenge for this measurement is the ability to recognise and identify the fission fragments generated by neutrons with an energy of up to 1 GeV. Therefore, a detection system with very good time resolution and low sensitivity to the γ-flash is needed. Furthermore, good discrimination between α particles and fission fragments is necessary, given the high activity of ^{239}Pu ($\sim$2,3 MBq/mg).
The measurement will be performed with the PPAC ensemble, which consists in nine target slots and ten PPACs. The time properties of the output signal are related to the fast component of the signals, typically of the order of nanoseconds, ensuring a time resolution of $\sim$200 ps. Thanks to their fast time response and very low sensitivity to the γ-rays, the PPACs have already been successfully used at n_TOF to measure the fission cross section of several actinides, for neutron energies up to 1 GeV [13–16]. The difference in signal amplitudes coupled with the coincidence method between consecutive PPACs, makes it possible to efficiently reject any background, in particular the α-emission of radioactive targets.
We plan to use six target slots for ^{239}Pu and three for ^{235}U, since its neutron-induced

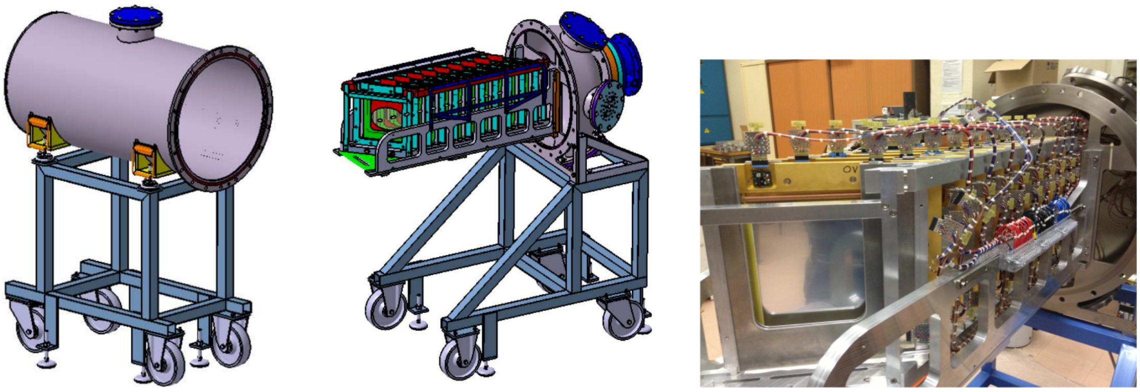

Figure 2: (a) Ensemble of 10 detectors and 9 targets tilted by 45° with respect to the neutron beam direction. (b) Picture of the detectors and targets on their support holding also the cables and connectors.

fission cross section is standard up to 200 MeV and usable as reference up to the GeV region.
The ^{239}Pu material suitable for this experiment is already available at JRC, Geel. The radiochemistry laboratory of SCK-CEN, Belgium, will perform the material purification process, which will result in a ^{241}Am/^{239}Pu purity level of the order of 1 ppm. The targets will then be produced at IJC Lab (Orsay). The ^{239}Pu and ^{235}U samples will be deposited on ultra-thin aluminum foils (2 μ thick) by electroplating, forming radioactive layers of 40 mm diameter and 160 and 250 μg/cm^2 thickness, respectively. Larger diameters were used in the past, but radio-protection considerations impose to reduce the diameter of the targets in this case.
Both ^{239}Pu and ^{235}U samples are practically pure - contaminants represent less than 0.1% in mass.

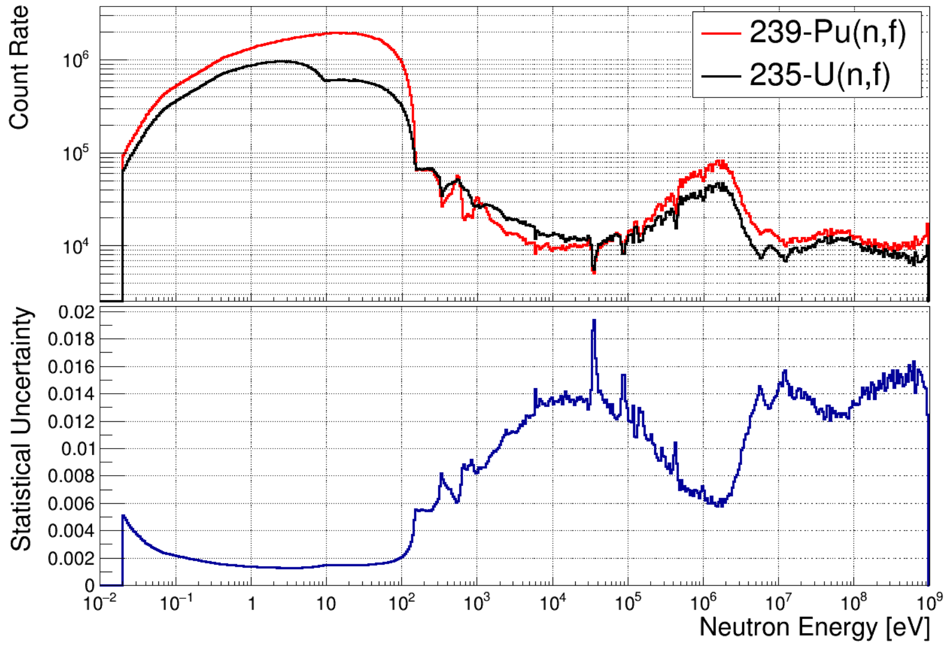

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Figure 3: (a) Expected total number of fission reactions detected in the depicted experimental setup coming from ^{239}Pu and ^{235}U samples. The number of protons on target is 4.5×10^{18}. (b) Expected statistical uncertainty of the ratio ^{239}Pu/^{235}U.

The samples will have a diameter smaller than the neutron beam profile in EAR1 with the fission collimator. Therefore, a careful mapping of the sample material will not be necessary as it will be entirely and homogeneously hit by the neutron beam. In order to determine the beam interception factor and monitoring it over the entire length of the chamber, two of the three reference samples of ^{235}U will be positioned as first and last sample in the detector stack.

3 Beam time request

Considering an effective beam interception factor of 0.35 and the typical PPAC fission fragment detection efficiency of 55%, we expect to detect the total number of fission reactions from six ^{239}Pu and three ^{235}U samples as reported in Fig. 3.
The ^{239}Pu(n,f) cross section will be measured as ratio with respect to the ^{235}U(n,f). The absolute ^{235}U cross section, standard up to 200 MeV, will be provided up to the GeV region as result of an already approved measurement at n_TOF [17].
In the bottom panel of Fig. 3, the expected statistical uncertainty on the ratio of ^{239}Pu(n,f) and ^{235}U(n,f) fission events, with 4.5×10^{18} protons on target, is reported. Both the expected counts and the statistical uncertainty are calculated with 50 bins per energy decade. The proton request is mainly driven by the low uncertainty required in the energy region from 10 to 20 MeV where a very high accuracy is required for various applications.

The measurement performed in EAR1 will additionally allow to detect fission events up to $\sim$GeV, significantly extending the range of high-precision data.

Summary of requested protons: 4.5×10^{18} protons on target - EAR1 fission collimator.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing
If relevant, write here the name of the fixed installation you will be using: Fission collimator	☒ To be used without any modification ☐ To be modified
PPAC - ISD Form: 3006565 v.1	☐ Standard equipment supplied by a manufacturer ☒ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT

Additional hazard from flexible or transported equipment to the CERN site:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	☒	PPAC: 0.004 bar, 100 l
	Vacuum	☐	
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	[fluid] [m3]
Electrical Safety	Electrical equipment and installations	☐	[voltage] [V], [current] [A]
	High Voltage equipment	☒	PPAC: 650 V
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	☐	[fluid], [quantity]
	Toxic/Irritant	☐	[fluid], [quantity]
	Corrosive	☐	[fluid], [quantity]
	Oxidizing	☐	[fluid], [quantity]
	Flammable/Potentially explosive atmospheres	☐	[fluid], [quantity]
	Dangerous for the environment	☒	C_3F_8 gas, 5 kg
Non-ionizing radiation Safety	Laser	☐	[laser], [class]
	UV light	☐	
	Magnetic field	☐	[magnetic field] [T]
Workplace	Excessive noise	☐	
	Working outside normal working hours	☐	
	Working at height (climbing platforms, etc.)	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards	Use of radioactive material	☒	^{239}Pu sample 12 mg 24 MBq ^{235}U sample 10.5 mg 1 kBq