### EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

# High accuracy measurement of the <sup>235</sup>U(n,f) reaction cross-section in the 10-30 keV neutron energy range

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#### Abstract

The analysis of the neutron flux of n\_TOF (in EAR1) revealed an anomaly in the 10-30 keV neutron energy range. While the flux extracted on the basis of the <sup>6</sup>Li(n,t)<sup>4</sup>He and <sup>10</sup>B(n, $\alpha$ )<sup>7</sup>Li reactions mostly agreed with each other and with the results of FLUKA simulations of the neutron beam, the one based on the <sup>235</sup>U(n,f) reaction was found to be systematically lower, independently of the detection system used. A possible explanation is that the <sup>235</sup>U(n,f) cross-section in that energy region, where in principle should be known with an uncertainty of 1%, may be systematically overestimated. Such a finding, which has a negligible influence on thermal reactors, would be important for future fast critical or subcritical reactors. Furthermore, its interest is more general, since the <sup>235</sup>U(n,f) reaction is often used at that energy to determine the neutron flux, or as reference in measurements of fission cross section of other actinides. We propose to perform a high-accuracy, high-resolution measurement of the <sup>235</sup>U(n,f) cross section in EAR1@n\_TOF, to clarify this issue.

**Requested protons** :  $1.5 \cdot 10^{18}$  protons on target **Experimental Area** : EAR-1

# 1. Introduction

#### 1.1 Motivation

The n\_TOF Collaboration has recently published the results of a high-accuracy determination of the n\_TOF neutron flux in EAR1 for Phase-II, covering the years 2009-2011 [1]. To reach the high-accuracy request, four different detection systems based on three different neutron converting reactions were used. The Silicon-based SiMon device, relying on the <sup>6</sup>Li(n,t)<sup>4</sup>He converting reaction, was used to cover the range between thermal and 100 keV. The same range was also covered with the MicroMegas detector with the <sup>10</sup>B(n, $\alpha$ )<sup>7</sup>Li reaction as converter. For the higher energy range we relied on the <sup>235</sup>U(n,f) reaction. In this case, two different systems were employed for detecting the fission fragments: MicroMegas and a calibrated ionization chamber from PTB. Finally, the experimental results were compared with the predictions of FLUKA simulations.

The analysis of the n\_TOF flux revealed a discrepancy between results based on the <sup>235</sup>U(n,f) reaction and all other ones, in the neutron energy range between 10 and 30 keV. In particular, the flux extracted on the basis of the <sup>235</sup>U(n,f) reaction, determined with both the PTB and Micromegas detector, is 6-8% lower than the one extracted from the <sup>6</sup>Li, <sup>10</sup>B e FLUKA simulations, as shown in Figure 1. A possible explanation is that the evaluated fission cross section in that neutron energy region is overestimated by several percent. Although the <sup>235</sup>U(n,f) between in that energy range is not a cross section standard, such a large difference was clearly unexpected, in particular since most evaluations assign an uncertainty on this cross section of less than 1% (see for example the compilation of standard cross sections in Ref. [2]).



**Figure 1:** The neutron flux determined from <sup>6</sup>Li and <sup>10</sup>B reactions ("evaluated") and from FLUKA simulations is compared with the one extracted by means of the <sup>235</sup>U(n,f) with two different detection systems (MGAS and PTB). A 6-8% difference can be observed in the 10-30 keV neutron energy range (figure from Ref. [1]).

Figure 2 shows the evaluated fission cross section in ENDF/B-VII.1 in this neutron energy region, together with the available EXFOR data and their ratio with respect to ENDF/B-VII.1 evaluation. The figure shows that evaluations tend to overestimate most

datasets. Also, it is interesting to notice the presence of structures in the cross section, most probably related to the grouping of unresolved resonances, similar to those determined at n\_TOF for the Au $(n,\gamma)$  reaction in a recent measurement [3].



**Figure 2:** Measured and evaluated cross section of the  $^{235}U(n,f)$  reaction in the 5 to 40 keV neutron energy range. For each data set the ratio with respect to ENDF/B-VII.1 is shown.

A further hint of the existence of a problem in the  $^{235}$ U(n,f) cross section between 10 and 30 keV is shown in Figure 3, where the evaluated uncertainty on the fission cross section is shown for two consecutive versions of the latest ENDF-B library. In the most recent version, VII.1, the uncertainty has increased, only in that energy region, by almost a factor of four relative to the VII.0 as well as relative to the one quoted in the evaluation of cross section standards of Ref. [2]. The new value is around 4%, which seems compatible with the level of discrepancy observed at n\_TOF. The reason for such a sudden large change is not clear, although it seems to be related to a different method in error analysis based on existing data (it should be emphasized, however, that the cross section have not changed between the two versions).

Finally, in a recent measurement of the capture cross section of  $^{235}$ U(n, $\gamma$ ), published on Phys. Rev. Lett., Jandel et al. [4] state that their capture data are 10% larger, between 10 and 30 keV, relative to the evaluated cross section. Since capture cross section in that work is extracted relative to the evaluated fission cross section, the observed discrepancy could well be attributed to the overestimate in evaluated fission cross section, rather than on the capture one.

We propose to perform a high-accuracy, high-resolution measurement of the  ${}^{235}$ U(n,f) cross section, relative to both  ${}^{6}$ Li(n,t) ${}^{4}$ He and  ${}^{10}$ B(n, $\alpha$ ) ${}^{7}$ Li reactions, in order to clarify the anomaly observed in the 10-30 keV neutron energy range. We remark that this energy range has a rather low impact on current thermal reactor technology, while it could have significant impact on fast critical reactors and sub-critical Accelerator Driven Systems. Furthermore, since fission cross sections of  ${}^{235}$ U is used very frequently as reference in measurements of fission cross section of major and minor actinides, a more precise determination of the  ${}^{235}$ U(n,f) cross section implies in turn a higher precision on the cross

section of many other actinides, in the 10-30 keV energy range. Last, but not least, fission chamber based on the  $^{235}$ U(n,f) reaction are often used for measurements of the neutron flux in a wide energy range. In this case as well, a new, accurate determination of the fission cross section between 10 and 30 keV would be helpful.



**Figure 3:** Uncertainty on the evaluated fission cross section of <sup>235</sup>U, as reported in the two most recent versions of the ENDF-B library. The large increase between 10 and 30 keV in the newest version is another hint of a problem in that region.

Together with the measurement here proposed for EAR1@n\_TOF, a similar highprecision measurement will also be performed at GELINA (JRC-IRMM), with a different detection system, with the aim of combining the results and achieve an overall uncertainty around 1%.

# 2. Experimental setup

As reported in Ref. [1], the analysis of the neutron flux between 10 and 30 keV was affected by a rather large uncertainty of 4-5%, because of two problems related to the  ${}^{6}Li(n,t)$  and  ${}^{10}B(n,\alpha)$  data. On the one hand, the kinematical boost due to the neutron kinetic energy starts to be important in this energy region. The effect is to shift the spectrum of tritons (from  ${}^{6}Li$ ) and  $\alpha$ -particles (from  ${}^{10}B$ ) towards higher energies, making particle identification more difficult (see Ref. [1] for more details on this point). On the other hand, above 1 keV particle emission in both reactions is affected by angular anisotropy. Since only forward-emitted particles were detected in that work, a correction had to be introduced for the angular anisotropy, resulting in a relatively large uncertainty on the reconstructed neutron flux.

To avoid this problem, we propose to measure the reference reactions both in the forward and backward direction. Furthermore, to optimize particle identification, sufficiently thin deposits should be used, and the resolution on the deposited energy in the detector should be optimized.

Since the requirement of high-resolution in the cross section determination is crucial in this measurement, in order to characterize the resonant structures previously observed, we propose to perform the measurement in EAR1.

#### 2.1 The detectors

A Silicon-based detector (SiMon) is being used, since the start of the n\_TOF project, to monitor the neutron flux via the <sup>6</sup>Li(n,t) reaction [5]. The stability and resolution shown by this device over the years makes Silicon detectors the most suitable ones for measuring this reaction. For this particular measurement, we propose to use a series of Sisandwiches, with <sup>6</sup>Li, <sup>10</sup>B and <sup>235</sup>U deposits. A stack of 6 detectors of 4x4 cm<sup>2</sup> area and 200 µm thickness will be inserted in the beam. Simulations indicate that the total amount of material in the beam (1.2 mm Si) does not pose particular problems in terms of neutron attenuation and/or generated background. The measurement should be performed with the capture collimator (1.9 cm aperture). Although never used at n\_TOF for the detection of fission fragments, solid-state detectors should be appropriate for this aim as well. The main problem of these detectors, i.e. their relatively high sensitivity to the prompt  $\gamma$ -flash, which limits the analysis to a few MeV, is not a problem in this case, considering that the energy range of interest is quite low. The use of a <sup>6</sup>Li-sandwich directly inserted in the neutron beam will be tested during the commissioning phase. For the final experiment, the stack of Si-detectors will be inserted in a specifically built vacuum chamber, with thin windows.

An alternative solution would be to use gaseous detectors, of the MicroMegas type. Such detectors are being used at n\_TOF since many years, in combination with both  ${}^{10}B(n,\alpha)$  and  ${}^{235}U(n,f)$  reactions. However, while there are no problems with fission fragments, the poorer energy resolution, relative to Si-detectors, represents a complication in this measurement, where a high degree of particle separation is required for reaction products from both  ${}^{6}Li$  and  ${}^{10}B$  neutron-converting reactions. Nevertheless, in case the tests during the commissioning evidence problems with the use of Si detectors in the neutron beam, the backup solution is to use MicroMegas chambers, with optimized gap, voltage and front-end electronics. In order to detect reactions products emitted both in the forward and backward direction, a total of six chambers would be required.

### 2.2 Samples

The samples will be prepared at LNS, CERN and IRMM. The thickness of the <sup>6</sup>Li should not exceed 300  $\mu$ g/cm<sup>2</sup>, for an optimal separation between tritons and  $\alpha$ -particles. A metallic <sup>6</sup>Li deposit will be sandwiched between two very thin C layers (10 µg/cm<sup>2</sup>) to ensure the stability of the deposit. Similarly, <sup>10</sup>B<sub>4</sub>C deposit of 0.5 micron thickness should be enough for reaching the desired  $\alpha$ -<sup>7</sup>Li separation. For <sup>6</sup>Li and <sup>10</sup>B reactions, we will investigate the possibility to direct deposition on the Silicon detectors. In alternative, two different deposits on the same backing, for forward/backward detection, will be used to avoid straggling of reaction products in the backing. For the <sup>235</sup>U sample, the thickness is limited by the range of fission fragments. Areal density of 500  $\mu$ g/cm<sup>2</sup> have been used in the past at n\_TOF, resulting in only minor losses of fission fragments emitted at large angles. It should be considered that neither the precise knowledge of the areal density, nor the homogeneity of the deposit are an issue in this measurement, since the data for <sup>6</sup>Li, <sup>10</sup>B and <sup>235</sup>U will be normalized to each other at thermal energy, where all three reactions are standard. Similarly, it is not crucial to determine the neutron flux impinging on the detectors, as far as all three samples are well aligned relative to each other. Since the measurement here proposed aims at determining with high accuracy the shape of the fission cross section, the only important corrections will regard the neutron-energy dependence of the efficiency to the <sup>6</sup>Li(n,t) and <sup>10</sup>B(n, $\alpha$ ) reactions, as well as the energydependent attenuation of the neutron beam, which can be determined by means of MonteCarlo simulations (being anyway of the order of 2% at most). Finally, the chosen thickness for all three samples ensured negligible pile-up corrections in the whole range from thermal neutron energy to a few MeV.



The use of the <sup>235</sup>U sample in the "work-sector type A" EAR1 will be under Radio-Protection supervision according to standard n\_TOF practice.

### 2.3 Electronics and data acquisition

The front-end electronics consists in preamplifiers and shaping amplifiers for the Silicon detectors. We are considering several options among the modules used in the past, as well as new commercially available low-noise preamplifiers. The use of a 16-channel integrated fast preamplifier/shaping amplifier module specifically built in the past for n\_TOF will also be considered. The analogue detector signals will be digitized with the standard n\_TOF Data Acquisition System based on flash-ADCs.

### 3. Beam request

Count-rate estimates shown in Figure 4 indicate that with the proposed samples, a total of  $1.5 \times 10^{18}$  protons on target are requested for this measurement, in order to reach a 1% statistical uncertainty in the energy region 10-30 keV.

### 4. Summary

We propose to measure the fission cross section of  $^{235}$ U relative to the  $^{6}$ Li(n, $\alpha$ ) and  $^{10}$ B(n, $\alpha$ ) standard cross sections in EAR1@n\_TOF, with a stack of Si-sandwiches, or in alternative, with a multistack MicroMegas detector. The aim of this work is to determine with high accuracy the cross sections in the 10-30 keV region, where sufficient hints exist pointing to an overestimate of the current cross section by as much as 6-8%. Data collected at n\_TOF will be combined with results of a similar measurement being planned at GELINA.

## Summary of requested protons:

Protons on target  $1.5 \cdot 10^{18}$ ; EAR1

## **References:**

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