EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Measurement of the ^{235}U(n,f) cross section relative to n-p scattering up to 1 GeV

May 06, 2017

L. Audouin[1], M. Barbagallo[2], N. Colonna[2], L. Cosentino[2], M. Diakaki[3,4], I. Duran[5], P. Finelli[6,2], P. Finocchiaro[2], J. Heyse[7], S. Lo Meo[8,2], C. Massimi[2,6], P.F. Mastinu[2], P.M. Milazzo[2], F. Mingrone[3], A. Musumarra[2,9], R. Nolte[10], C. Paradela[7], D. Radeck[10],P. Schillebeeckx[7], L. Tassan-Got[1], G. Vannini[2,6], A. Ventura[2]

[1] CNRS-IN2P3, Univ. Paris-Sud, Univ. Paris-Saclay, Orsay, France
[2] INFN - sezioni di Bari, Bologna, LNL, LNS and Trieste, Italy
[3] CERN, Switzerland
[4] National Technical University of Athens, Greece
[5] Universidad de Santiago de Compostela, Spain
[6] Dipartimento di Fisica e Astronomia, University of Bologna, Italy
[7] European Commission, Joint Research Centre - Geel, Belgium
[8] ENEA - Bologna, Italy
[9] Dipartimento di Fisica e Astronomia, University of Catania, Italy
[10] Physikalisch-Technische Bundesanstalt, Braunschweig, Germany

Spokespersons: C. Massimi (massimi@bo.infn.it), R. Nolte (ralf.nolte@ptb.de) and L. Cosentino (cosentino@lns.infn.it)
Technical coordinator: O. Aberle (oliver.aberle@cern.ch)

Abstract

Fission cross sections are typically measured relative to ^{235}U(n,f), since the neutron-induced fission cross section of ^{235}U is a standard at thermal energy and between 0.15 MeV and 200 MeV. However, above this energy no data are available, so that evaluations can only be based on theoretical calculations. The n_TOF facility offers a good opportunity to measure the ^{235}U(n,f) cross section relative to the ^{1}H(n,n)^{1}H reaction up to 1 GeV. We propose to perform such a measurement using a parallel plate counter and a fission ionization chamber for the detection of fission events in combination with recoil proton telescopes to measure the reference incident neutron flux.

Requested protons: 4×10^{18} protons on target
Experimental Area: EAR1

1. Introduction and scientific motivations

As already discussed in the letter of intent (LoI) submitted to INTC (52th meeting in February 2016) [1], the ^{235}U(n,f) cross section is one of the most important cross-section standards at the thermal neutron energy point (0.025 eV) and between 0.15 MeV and 200 MeV [2]. For instance, fission detectors equipped with a ^{235}U sample are used for the measurement of the neutron fluence for various applications, ranging from the investigation of the biological effectiveness of high-energy neutrons to the measurement of high-energy neutron cross-sections of relevance for accelerator-driven nuclear systems. Typically the ^{235}U(n,f) cross section is used as a reference for fission cross-section measurements, *e.g.* in ref. [3].

Despite its widespread use in the energy range between 20 MeV and 200 MeV, the recommended ^{235}U(n,f) cross-section is based on two measurements only [4, 5]. At energies above 200 MeV, the ^{235}U(n,f) cross section plays an important role also for fundamental nuclear physics. In particular, data on proton-induced fission have recently indicated that at high excitation energy (several hundred MeV) fission may be hindered with respect to particle emission due to its longer time-scale. The time delay of the fission process at high excitation energy is an important topic, as it is related to fundamental quantities of excited nuclear matter such as the viscosity. So far, as pointed out in [6], no data exist on neutron-induced fission above 200 MeV, and one has to rely on theoretical estimates using the ^{235}U(p,f) reaction above 600 MeV as guidance. This work showed up the inconsistency of the JENDL/HE evaluation [7] as well as the one recently released by the International Atomic Energy Agency (IAEA) Nuclear Data Section [8] in the energy range from 200 MeV to 500 MeV. The data shown in figure 1, together with a new theoretical calculation [6] based on the intranuclear cascade model INCL++ [9] coupled to the deexcitation model GEMINI++ [10], clearly indicate that the ^{235}U(n,f) cross section above 200 MeV may be substantially different from what was previously thought.

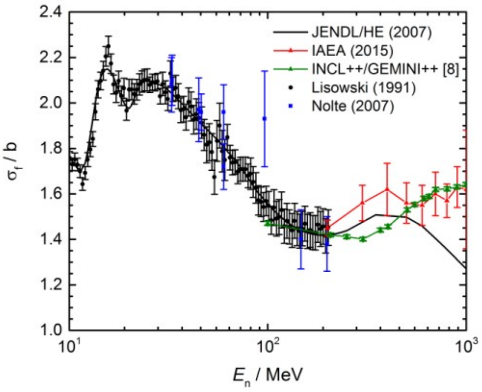


Fig. 1: The ^{235}U(n,f) cross section from the JENDL/HE evaluation, the recent IAEA evaluation (2015) and the calculated data reported in [6]. The available experimental data [4,5] measured relative to the n-p scattering cross section for E_n > 20 MeV are shown by the black and blue symbols.

Hence, there is a clear and long-standing demand from the International Atomic Energy Agency (IAEA) to improve this situation between 20 MeV and 1 GeV. In particular, there is a quest [8] for new fission experiments to be performed relative to n-p scattering that is the primary reference. We remind that phase-shift solution VL40 obtained by Arndt and collaborators [11] is accepted as a primary standard for cross section measurements in the 20 to 350 MeV range [12]. More recent phase shift solutions extend up to 3 GeV [13, 14]. In the energy region between 350 MeV and 600 MeV, and for center-of-mass proton emission angles larger than 40°, these solutions yield rather similar results. At energies above the Delta baryon production threshold at around 280 MeV above the nucleon mass, other channels besides elastic scattering are open and therefore the differential elastic scattering cross-section cannot be directly related to the total cross-section. As a consequence, a precise evaluation requires further studies, which will be carried out by the authors of this proposal and their collaborators. In particular, this is important to better understand the uncertainty of the differential n-p scattering cross section obtained from the phase shift solutions.

2. Proposed experimental set up

The proposed experiment aims at a measurement of the ratio of the ^{235}U fission cross-section to the differential n-p scattering cross-section in the energy range from about 20 MeV up to 1 GeV. Since fission fragments and recoil protons will be measured with different detection set-ups, the detection efficiencies are required to be well-known as they do not cancel out in the cross-section ratio. The experiment is to be carried out in EAR1 because here the neutron spectrum extends up to the GeV region. It will also profit by the good energy resolution at high neutron energies. The main challenge is to deal with the intense γ flash, as neutrons of 500 MeV will only be separated by 200 ns from the flash. A sketch of the proposed set-up is shown in figure 2.

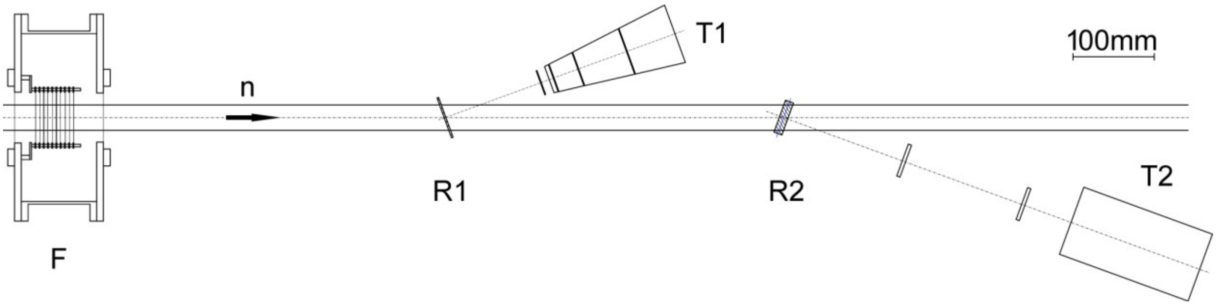


Fig. 2: Layout of the ^{235}U(n,f)/^{1}H(n,n)^{1}H cross section ratio measurement to be carried at EAR1. The setup will be positioned in the experimental hall at a distance between 1 m and 2.5 m from the entrance wall of the neutron beam. Due to the limited space available, a compact design is required. The neutron beam will be shaped by the capture collimator. The fission detector and the CH_2 samples are denoted by F, R1 and R2, respectively. Two possible RPT design alternatives T1 and T2 are shown here. The final design will be chosen after the second test run in 2017.

The fission cross section will be measured using detectors for fission fragments emitted from a set of ^{235}UF layers with a mass per unit area of about 300 μg/cm^2. The samples are produced by electrodeposition on 30 μm thick aluminum foils. In total, 15 samples will be made available by JRC Geel for this experiment. The fission fragment detectors will be stacks of parallel plate avalanche counters (PPAC) and parallel plate ionization chambers (PPFC). The PPAC is operated at low gas pressure and can be made almost massless and photon insensitive [15]. Therefore, it is well suited for the measurement of fission cross sections at neutron energies of several hundred MeV, *i.e.* very close in time to the γ flash [3]. However, it detects only fission fragments emitted into a forward cone with an opening angle of about 60° and its fragment-detection efficiency is not easily evaluated. In contrast, the fragment-detection efficiency of a PPFC is about 0.97 for 300 μg/cm^2 thick samples. However, the increased photon sensitivity due to the operation at ambient gas pressure could prevent the use of a PPFC close to the γ flash. Therefore, it will be used to study and eventually calibrate the PPAC fragment-detection efficiency in the energy range below 100 MeV.

The number of neutrons impinging on the ^{235}U samples will be measured simultaneously to fission by detecting recoil protons emitted from polyethylene (CH_2) samples. To this end, recoil proton telescopes (RPT) consisting of several fast plastic scintillation detectors will be located out of the neutron beam at proton emission angles of 20° - 30°. For each given neutron energy, recoil protons stopped in the RPT give rise to a well defined peak in the pulse height distribution of the last detector (see figure 3 below). Events produced by light charged particles (lcp) other than protons can be identified and discarded by measuring the differential energy loss in the transmission detectors and the remaining kinetic energy in the stop detector (ΔE-E method) of ΔE-E scatter plots. Spurious events, produced by stray particles not coming directly from the radiator target or by scattered neutrons, are suppressed by requiring multiple coincidences between different telescope layers. For example, such events can result from ^{12}C(n,lcp) interactions in the CH_2 samples. Additional measurements with graphite samples of matched thickness will be required only to subtract proton events produced via ^{12}C(n,p) reactions. These measurements will also be necessary for the highest neutron energies when the recoil protons can no longer be stopped in the last detector of the RPT and the separation of lcp-induced events and recoil proton events is therefore no longer possible.

The neutron beam will be shaped using the capture collimator which results in a neutron beam with an FWHM of about 18 mm at the location of the CH_2 samples. This small beam spot size is required to limit the angular divergence and the corresponding kinematical energy spread of the recoil protons detected in the RPTs. The recoil proton detection efficiencies of the RPTs are determined by the differential cross section for n-p scattering and the geometry of the instrument. In the present designs, the effective solid angle is fixed by the size of the transmission detectors instead of material aperture, which would cause excessive slit scattering and grey-edge effects at higher proton energies. Detailed Monte Carlo simulations using MCNPX and Geant4 and additional experimental work will be carried out to model the transport of protons in the RPT and investigate the uncertainty of the effective solid angle of the RPTs. The decision on the final design will be taken after the second test run in 2017 (see below).

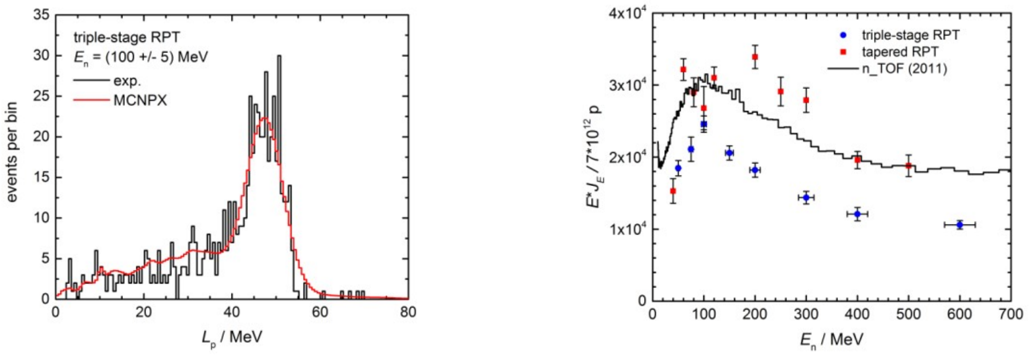
3. Results from the first test run and further preparatory work

The final RPT design will include experience gained in test runs performed in the context of the LoI [1]. The first one, carried out in 2016, mainly aimed at testing the detection of the n-p scattering under the operating conditions of EAR1 – in particular the intense γ flash – and the second, scheduled for 2017, addressing the final optimization of the design, including the detection of ^{235}U fission events.

Two slightly different concepts of the RPT for such a measurement [16] were simultaneously tested in the first run at 20° and 30° with respect to the neutron beam. One set-up was similar to the design shown in the lower half of figure 2. This triple-stage RPT had a front transmission detector consisting of a 2 mm thick EJ 228 scintillator. As second transmission detector, 2 mm and 5 mm thick EJ 228 scintillators were tested. The stop detector was 80 mm thick EJ 204 scintillator. The main design goal for this instrument was to achieve directional sensitivity for background suppression and reduced influence of angular straggling for an accurate definition of the solid angle. The neutron-energy range of interest can be varied by adapting the thicknesses of the CH_2 sample, together with the one of the transmission detectors and stop detector. The other set-up consisted of a tapered structure of four BC408 plastic scintillators, 5 mm, 30 mm, 60 mm and 60 mm in thickness. This set-up is shown in the upper half of figure 2. Here, the main design goal was to reduce wall effects in the stop detectors, by choosing a pyramidal shape, and to achieve a compact design which can be used over a large energy range. In addition to thin scintillation detectors, silicon diodes were tested as first transmission detectors in both RPTs.

The tests carried out with both instruments showed that they could be used at least up two about 600 MeV, *i.e.* the γ flash did not pose significant problems. Even operation of silicon diodes as first stage transmission detectors was possible with thin radiators 1 mm to 2 mm in thickness. With the triple stage RPT shown in figure 2, good suppression of background with a triple coincidence requirement was obtained. The possibility to separate proton- and deuteron-induced events using the ΔE-E method was proven in the first test run. The technique has afterwards been improved by using a scintillator with a larger mean emission wavelength and by improving the transport of the optical photons to the photomultiplier tube.

The left panels of figure 3 show calculated and measured pulse height distributions in the stop detector of the two RPTs for a neutron energy window centered at 100 MeV. The right panel exhibits the neutron energy distributions obtained with the triple-stage RPT and the tapered RPT for mean proton emission angles of 23° and 30°, respectively. The shape of the evaluated n_TOF neutron energy distribution is reproduced by the experimental data obtained with the triple-stage RPT. Most likely, the mismatch in normalization is due to an imperfect alignment of the RPTs. However, also such issues as the possible loss of coincident events in the pulse shape analysis or the particular n-p cross section data used in the simulation have to be scrutinized further.



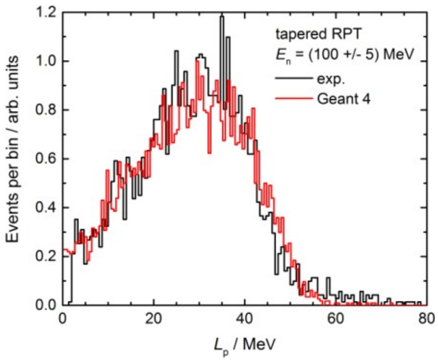


Fig. 3: Calculated and measured pulse height distributions in the stop detector of the triple-stage RPT (upper left panel) and the tapered RPT (lower left panel) for $E_n = 100$ MeV. The electron-equivalent kinetic energy of the recoil protons is denoted by L_p. The right panel shows the neutron energy distribution per 7×10^{18} protons measured with the two RPT configurations using a 10 mm CH_2 sample. The proton emission angles were about 23° for the triple-stage RPT and 30° for the tapered RPT. The vertical error bars show statistical uncertainties only.

A more quantitative test of the instruments will be performed during the second test run in 2017, and particular attention will be given to the improvement of the mechanical support structure and the alignment of the RPTs with respect to the neutron beam. The influence of edge effects in the ΔE detectors defining the effective solid angle covered by the RPTs will be studied using single-proton beams available from the PTB micro ion beam facility and proton beams from the Catania cyclotron. Moreover, efforts will be put in upgrading the detection of high energy protons. With a plastic scintillator of reasonable length, e.g. 150 mm, recoil protons produced by neutrons of about 170 MeV can be stopped. Based on the experience gained by the TAPS collaboration [17], it will be tried to extend the maximum energy of fully stopped events by using a BaF_2 stop detector equipped with an optical filter to suppress the slow scintillation component.

As previously mentioned, in the 2017 test run fission detectors will also be studied. The set-up will consist of two PPAC and two PPFC stages in a common vacuum chamber equipped with thin Kapton windows. The design of the PPACs will be a simplified and more compact version of that described in [15], since the ^{235}U samples available do not allow the detection of both fragments released in a fission event. Moreover, tracking of the fragments is not required for a measurement of the cross section only. The PPFC stages will be made as massless as possible. Monte Carlo simulations using the calculated n_TOF γ flash data [18] showed that the charge produced by the γ flash in a fission chamber stage operating at ambient pressure should be similar to the charge produced by one fission event. Hence, the main focus will be to investigate problems with the operation of the PPACs and the verification of the expected γ flash sensitivity of the PPFCs.

4. Beam time request

The proposed experiment aims at reaching a relative statistical uncertainty of 2% in the fission and n-p branches within neutron energy regions of 5% relative width. For the RPTs CH_2 samples 1 mm, 2 mm, 5 mm and 10 mm in thickness will be used. An overhead of 30% of the protons will be used for measurements with matched graphite samples and empty-target runs. For the fission branch, fifteen 300 μg/cm^2 ^{235}U samples will be employed, ten for the PPAC and five for the PPFC detector. The fragment detection efficiency of the PPAC and the PPFC is assumed to be 60% [15] and 97%, respectively. Table 1 shows the number of protons required to reach 2% statistical uncertainty for different neutron energies. The ^{235}U fission cross-section measurement is the limiting factor, and therefore 4×10^{18} protons are required to reach the desired accuracy. The RPT measurements will demand less neutrons. Hence, several combinations of CH_2 samples and transmission detectors of different thickness can be measured to adapt the recoil proton energy spread induced by the energy loss in the CH_2 samples and the transmission detectors to the energy of the incident neutrons in all neutron energy regions.

5. Conclusions

The experiment proposed here could provide first experimental results on the ^{235}U(n,f) cross section at energies beyond 200 MeV. These results could discriminate between various theoretical models on high-energy neutron-induced fission reactions, and contribute to establishing a reliable reference cross section above 200 MeV. On the low energy side (20 MeV to 200 MeV), it will be important to provide fission data relative to the differential n-p scattering cross section in order to allow a smooth match with the energy region where more data with small uncertainties are already available.

As a final remark, the measurement of the neutron fluence relative to n-p scattering can improve the knowledge of the n_TOF neutron flux for neutron energies higher than 200 MeV. As a consequence, other neutron-induced fission cross-sections measured or to be measured at n_TOF at these energies will benefit from the results of the proposed measurement.

Table 1: Estimated number N_p of protons required to achieve a statistical uncertainty of 2 % in the n-p and fission branches. The relative width of the neutron energy regions centered around E_n is 5 %. The number of neutrons per unit energy interval and per beam pulse (7×10^{12} protons) available with the capture collimator is denoted by J_E. The thickness of the CH_2 samples is denoted by t_{PE} The fragment detection efficiency ε_f of the PPAC and the PPFC was assumed to be 60% and 97%, respectively. The calculation for the RPT was carried out using the triple stage configuration shown in figure 3. It includes a 30% overhead for measurements with matched graphite samples and without CH_2 samples. The PPFC and PPAC detectors were assumed to have five and ten ^{235}U layers with a mass per unit area of 300 µg/cm^2, respectively.

E_n / MeV J_E / MeV^{-1}	23 861	30 720	40 628	60 466	100 310	150 193	230 110	300 73	400 49	600 30
	RPT									
t_{PE} / mm	1	2	5	5	10	10	10	10	10	10
N_p / 10^{18}	0.64	0.78	0.36	0.50	0.39	0.62	0.94	1.10	1.09	0.95
	PPFC									
N_p / 10^{18}	3.0	2.8	2.5	2.5	2.6	3.0	3.4	3.6	3.9	4.5
	PPAC									
N_p / 10^{18}	2.4	2.2	2.0	2.0	2.1	2.4	2.7	2.9	3.2	3.6

Summary of requested protons: 4×10^{18} simultaneously used by the PPAC/PPFC and the RPT detectors.

References

[1] Letter of Intent CERN-INTC-2016-002 I-165

[2] A.D. Carlson, *et al.*, Nuclear Data Sheets **110** (2009) 3215

[3] D. Tarrio, *et al.*, (The n TOF Collaboration), Phys. Rev. C **83** (2011) 044620

[4] A.D. Carlson *et al.*, in Proc. of the Int. Conf. on Nuclear Data for Science and Technology, Juelich 1991, p.518 (1991), Germany

[5] R. Nolte, *et al.*, Nuclear Science and Engineering, **156** (2007) 197

[6] S. Lo Meo, *et al.*, Nucl. Phys. A **933** (2015) 43

[7] Y. Watanabe, *et al.*, Journal of the Korean Physical Society **59** (2011) 1040

[8] B. Marcinkevicius, *et al.*, INDC(NDS)-0681, IAEA 2015

[9] A. Boudard, *et al.*, Phys. Rev. C **87** (2013) 014606

[10] D. Mancusi, *et al.*, Phys. Rev. C **82** (2010) 044610

[11] R.A. Arndt, *et al.*, Phys. Rev. D **35** (1987) 128

[12] A.D. Carlson, *et al.*, INDC(NDS)-368, IAEA 1997

[13] R.A. Arndt, *et al.*, Phys. Rev. C **76** (2007) 025209

[14] R.L. Workman, *et al.*, Phys. Rev. C **93** (2016) 045201

[15] D.Tarrio, *et al.*, Nucl. Instr. and Methods A **743** (2014) 79-85

[16] V. Dangendorf, *et al.*, Nucl. Instr. and Methods A **469** (2001) 205

[17] S. Diehl, *et al.*, J. Phys. Conf. Ser. **587** (2015) 012044

[18] C. Guerrero, *et al.*, (The n TOF Collaboration), Eur. J. Phys. A **49** (2013) 27