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Cross-section measurement of ^{235}U(n, f) at n_TOF from thermal energy to 170 keV

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Summary. — In the scenario of neutron data standards needed to measure neutron fluences and cross-sections of several kinds of reactions, the ^{235}U(n,f) reaction plays a key role. Motivated by significant inconsistencies found in measurements in the 10–30 keV energy range, we measured the energy-dependent cross-section of ^{235}U(n,f) at the n_TOF facility from thermal energy to 170 keV, with high energy resolution and uncertainty below 2%. This measurement, based on the two reference reactions ^{6}Li(n,t) and ^{10}B(n,α), allowed to ascertain that the origin of the discrepancies is an overestimation in average of 5% of the cross-section between 9 and 18 keV in major nuclear data libraries.

1. – Introduction

Nuclear data standards are used as high reliability references, needed to measure the neutron-induced cross-sections of a wide range of reactions such as the fission reactions of actinides, as well as to measure neutron fluences in neutron source facilities. A prerogative of the standard cross-sections is their accurate evaluation that requires high-quality data, because their uncertainty limits the precision of the measurements referred to them and hence an average uncertainty below a few percent should be ensured [1]. Many efforts have been made in the last decades to produce reliable nuclear cross-section data libraries. These are mostly based on available experimental data sets stored in appropriate databases as EXFOR (EXperimental FORmat), that are suitably treated with complex evaluation procedures [2]. With the aim to continuously improve their reliability and refine the standards by introducing new available data, these databases are periodically revised by dedicated agencies as IAEA (International Atomic Energy Agency). Moreover, new evaluations of the data are performed to update the dedicated libraries as ENDF/B-VIII [2] (Evaluated Nuclear Data File) when needed. The reaction ^{235}U(n,f) represents one of the most important reactions, that is currently employed for a wide range of applications and has been extensively studied over the decades [3, 4]. In particular the cross-section of this reaction is a standard at thermal energy and from 150 keV to 200 MeV. Moreover, the integral of the cross-section in the range between 7.8 eV and 11 eV is a standard as well. Although below 150 keV the cross-section is not standard, it is usually employed as a reference, even though some inconsistency came to light in last years when compared with other neutron data standards. In particular, significant discrepancies have been found in the energy range 10–30 keV, in the measurements of some physical quantities using both the ^{235}U(n,f) reaction and other standard data as references [5, 6]. Considering for instance the neutron beam flux at n_TOF among these, by comparing the flux measured with detectors based on the reactions ^{6}Li(n,t) and ^{10}B(n,α) (whose cross-sections are standard until 1 MeV) to that measured with the ^{235}U(n,f), a difference of about 5% was found between 10 keV and 30 keV. This significant discrepancy was suggested to be due to an overestimation of the ^{235}U fission cross-section by the major libraries. The need to clarify the origin of this problem, motivated the n_TOF Collaboration to measure with high accuracy the energy-dependent cross-section of ^{235}U(n,f).

2. – Experimental set-up

n_TOF is a neutron beam facility located at CERN, featuring a spallation neutron source and two beam lines [7]. The beam is pulsed in short bursts of period 1.2 s and the neutron energy ranges from thermal to ∼1 GeV. The ^{235}U(n,f) measurement has been performed in the experimental area 1, where the long flight path $L = 183.49(2)$ m entails a beam energy resolution around 10^{-4}. The detector specially assembled for this measurement was designed to detect fission fragments from the ^{235}U(n,f) reaction and at the same time to detect the reaction products of the ^{6}Li(n,t) and ^{10}B(n,α) reactions, that were used as references [8]. This technique, known as ratio method makes the detector totally self-consistent and does not require any other equipment, allowing to measure the neutron flux by means of the reference detectors. In details, the detector set-up was arranged as a stack of six silicon detectors $5 \times 5\,\text{cm}^2$ and 200 μm thick, each one facing a sample placed at 1 mm distance. Each detector can only collect the reaction products from its own sample, $2\times{}^{6}$LiF, $2\times{}^{10}$B, $2\times{}^{235}$U, emitted, respectively, in the forward and in the backward direction. In order to apply the ratio method, the main requirement of the experimental apparatus is that all the samples be exposed to the same neutron flux, and in fact the stack was carefully aligned to the neutron beam. The neutron flux was measured by collecting the events produced by the ^{6}Li(n,t) and ^{10}B(n,α) reactions, selecting for each the lighter and more energetic products (*i.e.*, tritium and alpha respectively), by setting suitable selection thresholds in the energy spectra. The behaviour of these thresholds against the neutron energy is conditioned by the kinematic effects induced by the neutrons and by the energy loss of the fragments in the dead layers, the last depending on how the angular distributions of the fragments change with the neutron energy. In fact the emission of the fragments for the ^{6}Li and ^{10}B reactions is not isotropic, because a larger fraction of the products are emitted in the forward angles as

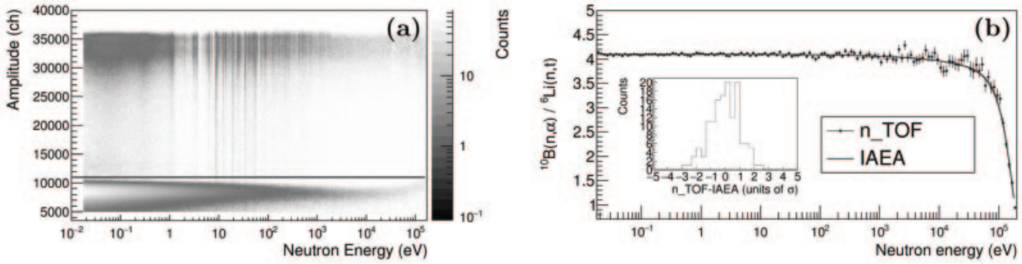

Fig. 1. – 2D scatter plot of the energy spectra *vs.* neutron energy of a ^{235}U detector (a). Ratio of the count rates between the reference reactions ^{10}B(n,α) and ^{6}Li(n,t) (b), corrected for detectors efficiency and beam attenuation. The continuous line is the ratio between the two standard cross-sections.

the neutron energy increases. For ^{235}U(n,f) the emission of the fission fragments in the investigated energy range is isotropic. In fig. 1(a), the 2D scatter plot of the detected energy *versus* the neutron energy for a ^{235}U detector is shown; the separation of the fission fragments from the α background is very clear.

The time to neutron energy calibration to assign the neutron energy from the measured time of flight was carried out by exploiting the well-known resonances of the ^{235}U(n,f) cross-section. The flight path was estimated by minimizing the χ^2 between experimental data and ENDF-B/VIII library. The beam flux was calculated for each reference detector-sample pair by using eq. (1), where C is the total counts of the selected events normalized to the number of protons in a pulse, f is the beam fraction on the sample, ε the detection efficiency, ρ the sample areal density and σ the neutron cross-section. Accurate Monte Carlo simulations performed with GEANT4 have allowed to calculate the beam fraction f and the detector efficiency ε of all the reference detectors. The beam fraction on each sample was affected by the average absorption of the neutrons in all the layers before the sample itself, with characteristic dips due to the absorption resonances of the elements present in the layers. The anisotropy of the two reference reactions with increasing neutron energy was taken into account by using the angular distributions evaluated in ENDF/B-VIII, even though the almost total solid angle covered in the forward and backward direction minimizes possible discrepancies. To prove the reliability of the experimental data, as well as the analysis and simulations, we calculated the ratio between the corrected yield of the ^{6}Li(n,t) and ^{10}B(n,α) reactions. Figure 1(b) shows the good agreement between the experimental values and the ratio of standard cross-sections, well within the experimental uncertainty, as clearly indicated by the distribution of the normalized residues shown in the inset:

$$\Phi(E_n) = \frac{C(E_n)}{f(E_n)\varepsilon(E_n)(1-e^{-\rho\cdot\sigma(E_n)})} \tag{1}$$

$$\sigma_{^{235}\mathrm{U}} = K\frac{C_{^{235}\mathrm{U}}(E_n)}{\Phi(E_n)f_{^{235}\mathrm{U}}(E_n)}. \tag{2}$$

3. – Results

After having calculated the neutron flux separately from B and Li data and combined them in weighted average, the ^{235}U(n,f) cross-section was calculated up to 170 keV with

Table I. – *Comparison between n_TOF and IAEA2020.*

Energy range	n_TOF/IAEA	Δ (σ)
4–9 keV	0.986 ± 0.010	-1.5
9–18 keV	0.949 ± 0.011	-4.8
18–30 keV	0.986 ± 0.013	-1.1
30–60 keV	0.991 ± 0.013	-0.7
60–100 keV	1.004 ± 0.015	0.2
100–150 keV	0.960 ± 0.014	-2.8
150–170 keV	1.001 ± 0.025	0.0

eq. (2). The normalization constant K has been calculated normalizing the experimental data to the standard cross-section integral between 7.8 eV and 11 eV, that allowed to calculate the absolute value of the cross-section with small uncertainty. In order to make a further validation check of the data, the ratio between the cross-section at thermal energy and this integral value was compared to the same quantity obtained using the standard values by IAEA. The experimental value obtained for this ratio was 2.352 ± 0.013 (stat.) ± 0.007 (syst.) eV^{-1}, that is in good agreement with the one provided by IAEA of $2.373 \pm 0.029\,\text{eV}^{-1}$. In order to investigate the interval between 10 keV and 30 keV, we integrated the data in sub-intervals and compared the experimental values with the ones provided by IAEA. Table I shows the ratio and the normalized residues calculated between the n_TOF data and the IAEA values. In the interval 9–18 keV the average difference is about 5% and 4.8σ, thus underlining a statistically significant overestimate of the evaluated cross-section in this energy region. In the other ranges the integral values are in agreement with the evaluations, in particular for the higher energies (>150 keV), where the evaluated data are standard. A possible flaw in the IAEA evaluation procedure [9], produced by a single point at 9.5 keV, was deemed responsible for such a discrepancy.

4. – Conclusions

The ^{235}U(n,f) cross-section measurement performed at n_TOF provideded a new set of high resolution data, with uncertainty of 1–2% with data binned at 20 bin/decade. In particular in the range 9–18 keV, where the cross-section exhibits several structures, an average overestimation of 5% has been found. The EXFOR library has been recently updated with these new reliable data set, for future refinements of the neutron data evaluation libraries.

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