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Neutron-induced capture-to-fission cross section ratio measured at LANSCE

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Abstract. Capture-to-fission cross section ratios are used as an alternative to absolute cross section measurements. This is due to the simplification on the calculations and the reduction of the uncertainties with respect to an absolute measurement of the cross section by eliminating experimental complications like self-absorption, beam/target overlap and non-uniformities. Different capture-to-fission reactions have been measured through the years at the Los Alamos Neutron Science Center (LANSCE) at Los Alamos National Laboratory (LANL) using the Detector for Advanced Neutron Capture Experiments (DANCE) combined with different fission detectors: a Parallel Plate Avalanche Counter (PPAC) to detect fission fragments (FF), and the NEUtron detector array at dANCE (NEUANCE) to detect fission neutrons. As DANCE detects the γ-rays produced in capture and fission reactions, the fission instrument placed inside the DANCE cavity is used to tag the fission γ-rays for background identification and subtraction. Some examples of capture-to-fission ratio measurements performed with DANCE in the last years are the ^{233}U, ^{235}U and ^{239}Pu. The measurement technique, the different setups, and other potential applications of the instruments will be described.

1 Introduction

There is a need of reduced uncertainties for applications in nuclear technology, including the design of advanced reactors. Uncertainties in the capture cross section impact the understanding of the criticality of U/Pu systems and transmutation rates. Fissile isotopes are key in the Th-U and U-Pu fuel cycles, and their cross sections must be accurately known. Radiative capture measurements on fissile nuclei are complicated due to the difficulty in separating capture γ rays from the high fission γ-ray background. For this reason and in order to reduce uncertainties in the calculations, for fissile isotopes, a relative cross section calculation, measuring the capture-to-fission ratio, is an attractive alternative to absolute capture cross section measurements. The capture-to-fission ratio eliminates the systematic uncertainties derived from the neutron flux, self-shielding and sample mass.

At LANSCE, an 800 MeV proton beam is ejected from the Proton Storage Ring (PSR) and collides with a Tungsten target producing neutrons by spallation reactions that are moderated in water before being ejected into the beam pipe [1]. The Detector for Advanced Neutron Capture Experiments (DANCE) [2] has been used in combination with a fission detector to measure the capture-to-fission cross section ratio of the fissile isotopes: ^{235}U, ^{239}Pu and ^{233}U. Fission γ-rays detected with DANCE are detected in coincidence with neutrons using NEUANCE [3] or FFs using a PPAC [4]. Each fission detector requires the use of a particular type of sample.

2 Experimental setup

The DANCE instrument is a ~3.5π γ-ray calorimeter composed by 160 BaF$_2$ crystals. It is permanently located in Flight Path 14 (FP14) at the Lujan Neutron Scattering Center (LANSCE), at ~20 m from the spallation target, producing a white neutron spectrum with energies from meV to hundreds of keV.

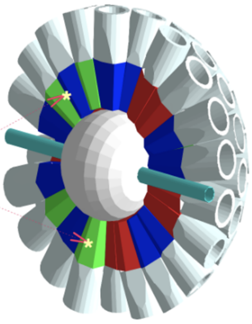

Figure 1. DANCE instrument semisphere showing the neutron absorber shell in its inner cavity.

When measuring fission in coincidence with DANCE, a fission detector is installed inside the 17 cm diameter

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DANCE cavity surrounded by a neutron absorber shell to reduce the neutron scattering from the detector material and sample backing into the DANCE crystals, see Fig. 1. The fission tagging method consists of detecting fission products using the fission detector and identifying which of these events are found in coincidence with the γ rays detected with DANCE. There are different target requirements depending on the type of fission product to be detected. The use of thick targets present an advantage over thin targets regarding beam time requirements, as more events are produced per second. However, the downside of this type of targets is that the scattering background and self absorption effects are considerably higher. The detection of fission fragments requires the use of thin targets, to avoid self absorption of the fragments in the sample material. Neutrons, being uncharged particles, can travel through thick material and therefore they can be detected using a thick sample.

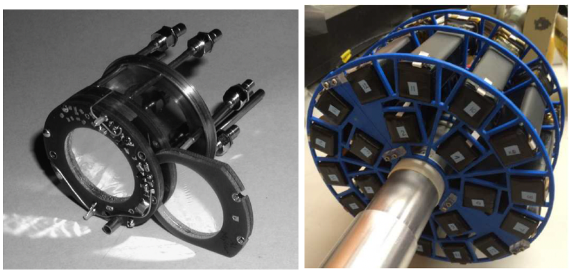

Figure 2. PPAC detector (left) and NEUANCE instrument (right).

The PPAC is a dual parallel plate avalanche counter detector with a removable target/cathode foil, see Fig. 2. The sample is electro-deposited in the center of the cathode layer, and the two anodes are located at both sides of the cathode. It is filled with Isobutane gas operating at 6-10 Torr pressure and it has a charged particle detection efficiency of 65%. More details can be found in [5]. The PPAC is placed inside the DANCE inner cavity to detect FF coming from neutron-induced fission reactions in the sample. This detector is sensitive to the α-decay background originating from the sample material. The α-particle signals have a low pulse height, and the α-decay background is partially eliminated through a threshold in the signals. This type of experiment requires the use of two targets: a thin target placed inside the PPAC for fission tagging, and a thick target with no PPAC to increase the statistics.

NEUANCE is a neutron detector array composed by 21 stilbene crystals in cylindrical geometry around the beam pipe inside which the sample is placed, see Fig. 2. It detects neutrons with energies above a 500 keV threshold which correspond to the fission neutron energies, discriminating the low energy scattered neutrons. This detector has a single fission neutron efficiency of 12.5%. A more detailed description of this instrument is given in [3].

3 Data Analysis

The data analysis involves event building, neutron energy calculation, energy calibration, fission event identification, background subtraction, and calculation of the capture-to-fission cross section ratio. An important concept in the DANCE analysis is the definition of a "cluster", DANCE crystals are grouped into “clusters” of neighboring crystals that all saw a γ ray. This concept is used during analysis due to the large segmentation of the DANCE array [6]. DANCE measures the incident neutron energy (E_n), the total energy of the γ cascades deposited in the crystals (E_{tot}), and the cluster multiplicity (M_{cl}).

The first step on the analysis is the calibration of the DANCE crystals, it is performed using γ-ray sources and the α-decay chain of the ^{226}Ra present in the BaF_2. The BaF_2 scintillation light has a fast and a slow component which ratio is very different for γ rays and α particles. This allows particle identification by pulse shape discrimination (PSD), see Fig. 3.

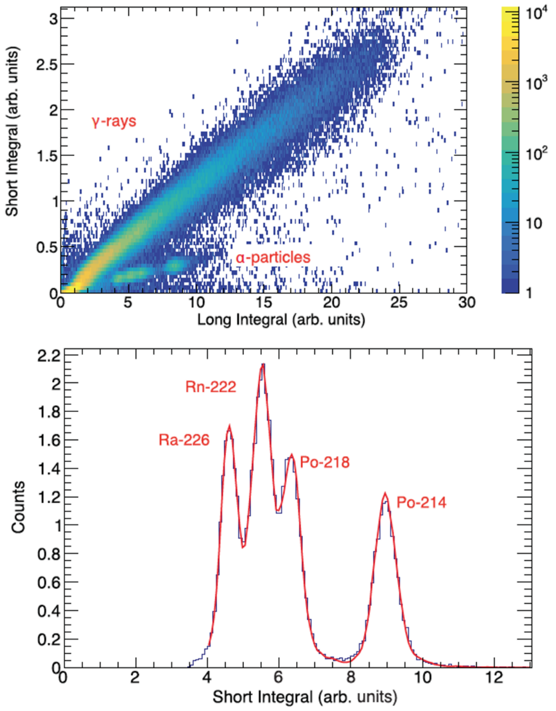

Figure 3. Pulse shape discrimination (PSD) histogram showing the short versus the long component of the signals used to distinguish γ rays and α particles (top). The α-particle spectrum is selected and used for the DANCE crystals energy calibration. Alpha-decay spectrum for one of the DANCE crystals showing the calibration peaks from ^{226}Ra, ^{222}Rn, ^{218}Po and ^{214}Po (bottom).

One of the main parts of the analysis is the identification and subtraction of the background events. The background is dependent on the incident neutron energy and has to be studied and subtracted for each neutron energy.

The fission background is identified by coincidences between the DANCE γ rays and the fission products measured with the fission detector, and those DANCE γ rays are tagged. This analysis method is the same independently of the type of both: fission detector and detected fission particles. Figure 4 shows the cluster multiplicity as a function of the total energy of the γ rays deposited in the DANCE crystals. The top figure shows the tagged events, as the efficiency of the PPAC is not 100%, not all the fission events are tagged, and the remaining fission background needs to be identified and subtracted. This is achieved by selecting a fission window in the figure, where only fission events are found in both the tagged and untagged spectra, normalizing the tagged distribution to the untagged, and subtracting it.

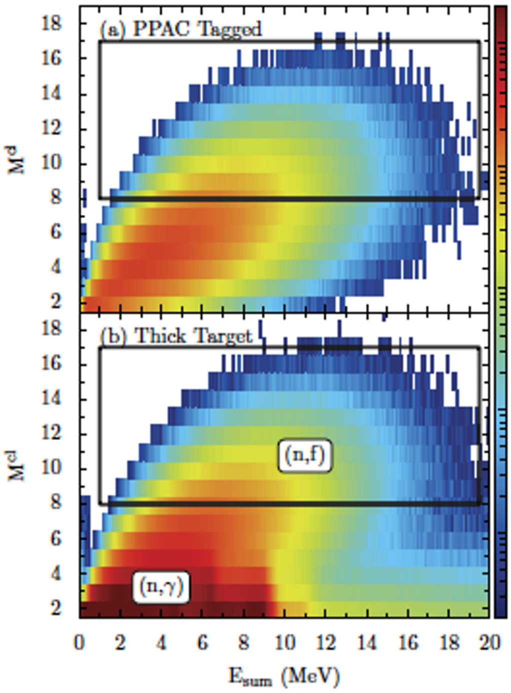

Figure 4. Cluster multiplicity as a function of the total energy of the γ rays showing the events tagged with the PPAC (a) and untagged (b). The regions where fission and capture events are dominant are labeled in the figure. The black rectangle defines the fission normalization window.

Another component of the background is due to scattering reactions. The scattering background is measured using a ^{208}Pb sample and it is normalized to the fission subtracted spetrum using an energy window around the scattering peak, around 8.5 MeV, in the E_{tot} distribution and subtracted. Figure 5 shows the different components of the background and its subtraction.

4 Capture-to-fission ratio

The capture-to-fission cross section ratio α is measured using Eq. 8. Experimentally, a number of events C_i is measured as a function of neutron energy that is associated with process i (fission or capture in this case).

$$C_i(E_n) = \varepsilon_i Y_i(E_n) \tag{1}$$

and

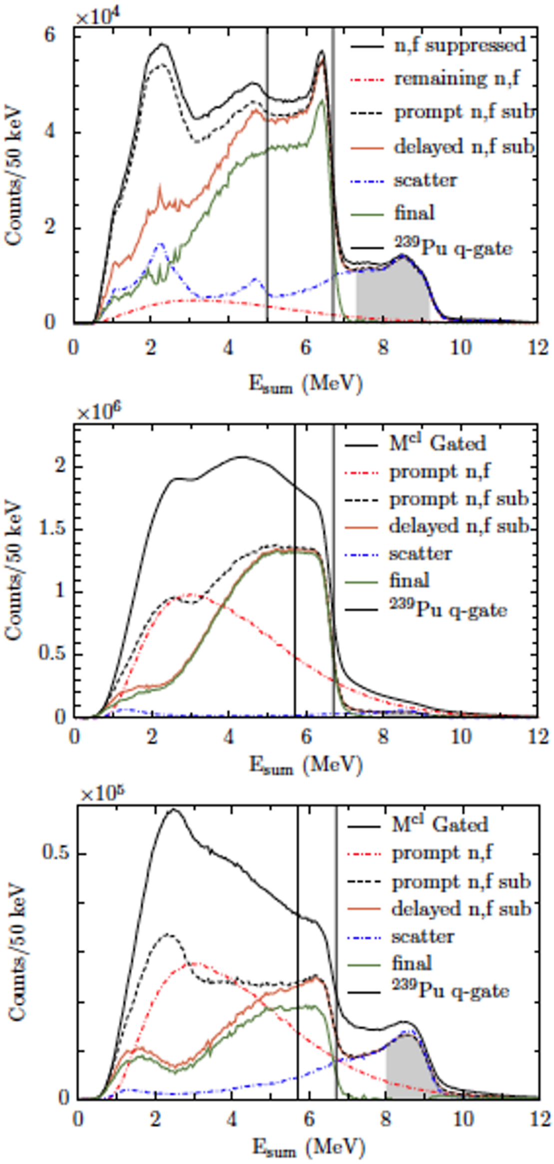

Figure 5. Total energy of the detected γ rays showing the different components of the background for different incident neutron energies. The Q-value gate defining the limits of the capture reactions for the isotope under study, in this case ^{239}Pu, is defined by the vertical black lines.

$$Y_i(E_n) = \sigma_i(E_n) N \Phi_n(E_n) \tag{2}$$

where ε_i is the efficiency of detecting an event of type i, $Y_i(E_n)$ is the total yield of events of type i, N is the total number of atoms in the sample, and $\Phi_n(E_n)$ is the total number of neutrons per unit area the sample was exposed to over the course of the experiment. Then to determine α we have

$$\begin{aligned} \frac{C_\gamma(E_n)}{C_f(E_n)} &= \frac{\varepsilon_\gamma Y_\gamma(E_n)}{\varepsilon_f Y_f(E_n)} && (3) \\ &= \frac{\varepsilon_\gamma \sigma_\gamma(E_n)}{\varepsilon_f \sigma_f(E_n)} && (4) \\ & && (5) \end{aligned}$$

$$= k\frac{\sigma_\gamma(E_n)}{\sigma_f(E_n)} \tag{6}$$

$$= k\alpha(E_n) \tag{7}$$

therefore

$$\alpha(E_n) \equiv \frac{1}{k}\frac{C_\gamma(E_n)}{C_f(E_n)}, \tag{8}$$

where, assuming the detection efficiencies for capture and fission do not depend on the neutron energy, k is an energy-independent constant. If the capture and fission cross sections are known at a particular energy or over an energy range, k can be determined from the ratio of the detected events over that energy range, and the ratio $\alpha(E_n)$ can be directly determined from the measured data over the full energy range. Experimental advantages of the capture-to-fission ratio are that it is much simpler and more reliable to determine experimentally as many of the systematic questions, as sample mass, self-shielding and neutron exposure will cancel out in an appropriately designed experiment.

5 Results

The capture-to-fission cross section ratios of ^{233}U, ^{235}U and ^{239}Pu have been measured with DANCE in the recent years. The details of the experiments, data analysis and results are provided in [4, 6–9].

The ^{235}U capture-to-fission cross section ratio was the first of the experiments, it was performed using DANCE combined with the PPAC, and provided results for neutron energies from 4 eV to 1 MeV. Three independent measurements were done: a measurement using a thick sample to increase the statistics at high neutron energies, a measurement with a thin sample for fission tagging, and a measurement of the neutron scattering background using a ^{208}Pb sample. The thick target was a 26 mg/cm^2 sample of 94% enriched ^{235}U, the thin sample was 130 ug/cm^2 of 99.9% enriched ^{235}U, and the scattering sample was 120 mg/cm^2 of 99% enriched ^{208}Pb. Details on the experiment are given in [4]. The results were normalized to the evaluated cross sections in the neutron incident energy region from 45 to 100 eV. The alpha-derived capture cross section was calculated by multiplying the capture-to-fission cross section ratio by the ENDF/B-VII.0 fission cross section, the version available at the time of the measurement. The broadened cross section was used in the Resolved Resonance Region. Significant discrepancies compared to ENDF/B-VII.0 as large as 30% were observed, especially in the region between 1 keV and 2.5 keV, a better agreement was found in this region compared to JENDL-4.0, see Fig. 6.

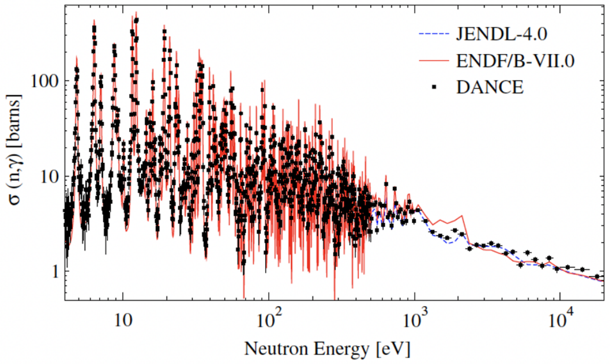

Figure 6. Alpha-derived ^{235}U(n,γ) cross section (black marks) compared to the ENDF/B-VII.0 (red line) and the JENDL-4.0 (blue line) evaluations.

The ^{239}Pu capture-to-fission ratio was measured using the DANCE and PPAC setup from 10 eV to 1.3 MeV. Three independent measurements were performed using the same method previously applied for the ^{235}U alpha-ratio measurement. The three targets used, were a thin 937 μg of 99.97% enriched ^{239}Pu, a thick 50 mg sample of ^{239}Pu, and a ^{208}Pb 200 mg/cm^2 target to measure the scattering background. More information of this experiment can be found in [7–9]. The relative capture cross section is shown in Fig. 7 in the incident energy region from 10 eV to 1 keV. The results in this energy region were published in [7].

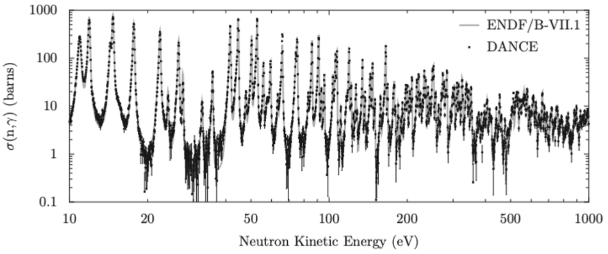

Figure 7. ^{239}Pu alpha-derived capture cross section from 10 eV up to 1 keV (black markers) compared to the ENDF/B-VII.1 evaluation (grey line).

Comparing to previously measured data, the background subtraction used in this work seems to be more advanced. For instance, in the energy region between 30 and 40 eV, the present cross section drops in line with evaluations and runs almost an order of magnitude lower than the datasets [10, 11]. In addition, the mentioned datasets report a trio of resonances at 18, 21, and 46 eV which were identified as tungsten impurities in the sample. The pure sample used in the DANCE measurement provided data clean of these contaminant resonances.

A further analysis of the data was later on performed in order to extend the energy region up to 1.3 MeV. Figure 8 shows the DANCE results from 1 keV to 1.1 MeV compared to previous experiments and the evaluations at the time of the measurement. The results show a lower relative capture cross section than JENDL and ENDF/B-VII.1 in around 10 keV, and become consistent in the vicinity of 40 keV. The peak in the ENDF/B-VII.1 cross section near 300 keV was not reproduced. The cross section is consistent with the data from Hopkins et al.. Individual points near 20 keV and 100 keV are not reported due to their proximity to the most significant Aluminum resonances, which perturbed the physics analysis such that the results at those specific energies were questionable. The results were published in [9].

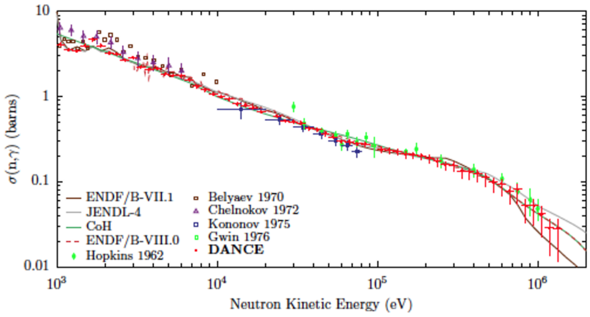

Figure 8. ^{239}Pu relative capture cross section from 1 keV up to 2 MeV (red marks) compared to the experimental data available in EXFOR, the evaluations and the CoH calculations.

The design and construction of the neutron detector NEUANCE opened the possibility for a combined measurement with DANCE to study the fission background using only one thick sample. The ^{233}U capture-to-fission ratio was recently measured using DANCE and NEUANCE from 0.7 eV to 250 keV. The ratio was normalized to the ENDF/B-VIII.0 broadened cross section ratio in the neutron energy region from 8.1 to 14.7 eV. This is the first measurement provided for incident neutron energies between 2 and 30 keV. In the resolved resonance region, the evaluations are in good agreement with this work, while the results show a lower capture-to-fission ratio in the unresolved resonance region from 10 to 150 keV, where large discrepancies were found within the evaluations. Above 10 keV and up to 150 keV, a lower capture-to-fission ratio was obtained in this work compared to the evaluations and the experimental data from Hopkins and Diven [12]. The alpha-derived capture cross section is shown in Fig. 9 compared to the evaluated data libraries. The analysis and results were published in [6].

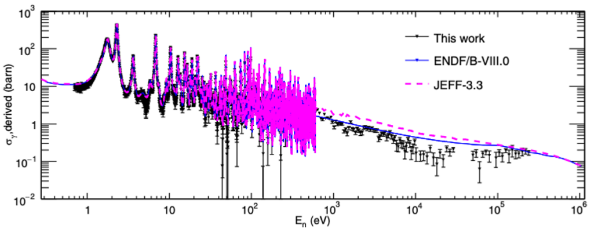

Figure 9. ^{233}U relative capture cross section from 0.7 eV to 250 keV (black markers) compared to the ENDF/B-VIII.0 (blue line) and the JEFF-3.3 (magenta line) evaluations.

Statistical model calculations were performed using the CoH3 code [13] from 1 keV to 5 MeV that is the energy for which only the first fission chance is involved. The CoH3 combines the coupled-channels optical model and the statistical Hauser-Feshbach model calculations by using the Engelbrecht-Weidenmüller transformation of the penetration matrix. Different values of the average γ-ray width were tried by adjusting the M1 γ-ray strength function for the scissors mode. Mughabghab provided a value of 40 meV. The value had to be reduced to 24 meV to reproduce the data from Hopkins and Diven. A smaller value would be needed to reproduce this work.

6 Conclusions

The capture-to-fission cross section ratio eliminates the uncertainties associated to the neutron flux, sample mass and self-shielding. It can be measured at LANSCE combining DANCE and a fission detector; a PPAC and NEUANCE have been used to detect FF and fission neutrons in each case. The relative cross sections have been calculated multiplying the alpha-ratio by the evaluated fission cross section. Measurements of the ^{235}U, ^{233}U and ^{239}Pu in the neutron energy region from eV to ~100 keV/1 MeV have been performed in the last years. Other detectors could be used in combination with DANCE to tag fission events in the future.

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