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

Measurement of fission cross section and fission-fragment angular distribution of ^{231}Pa

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Abstract: We propose to measure the fission cross section and the fission-fragment angular distribution of ^{231}Pa. ^{231}Pa is a nucleus of interest in the ^{232}Th/^{233}U cycle. It is produced through (n,2n) and (n,γ) reactions within the cycle. Its high radiotoxicity makes it a relevant candidate for incineration. This nucleus is also of interest as a test of the surrogate technique in the neighboring of ^{233}Pa. From a more fundamental point of view, ^{231}Pa presents vibrational resonances in the threshold region: information about the spin states populated in the fission process can be extracted by measuring, precisely, the fission-fragment angular distribution in the region surrounding these resonances. We propose to measure, simultaneously, the (n,f) cross section and the fission-fragment angular distribution of ^{231}Pa in a range of energy from tens of meV up to hundreds of MeV, taking full advantage of the intense neutron beam of n_TOF-EAR2 and its wide energy range. The measurement will be performed using a suited setup based on parallel plate avalanche counters.

Requested protons: $2 \cdot 10^{18}$ protons on target
Experimental Area: EAR2

1 Motivations

^{231}Pa is a relevant isotope for the ^{232}Th/^{233}U cycle. If reactors based in this cycle are operational, ^{231}Pa will be produced by (n,2n) reactions on ^{232}Th that build up mainly in fast reactors, and by (n,γ) reactions on the natural abundance of ^{230}Th, present along ^{232}Th in the Th ore. It plays a role similar to ^{237}Np in the ^{238}U/^{239}Pu cycle, but it is much more radiotoxic owed to its shorter half-life of 32 kyr (instead of 2 Myr for ^{237}Np). If ^{231}Pa would be considered for incineration, its neutronic properties should be known with a suitable accuracy, especially its fission cross section. At the moment, the experimental information on this cross section is very limited: only 4 measurements [1, 2, 3, 4, 5] extend over a significant energy range and they show pronounced discrepancies between each other (see the discussion below).

The fission cross section shows marked structures in the threshold. They reveal the effect of strong vibrational resonances typical of the neighborhood of the thorium isotopes. A recent measurement of the fission probability in the resonance region from the surrogate reaction ^{231}Pa(d,pf) [6] suggest an hyperdeformed scenario, not confirmed due to the lack of data on fission-fragment angular distribution. Up to now only 2 sets of data exist [7, 8] for this angular distribution. They give contradictory anisotropies, and were obtained with mono-energetic neutrons. A precise measurement of the angular distribution of fission fragment is a good tool to determine the spin of such resonant states.

The n_TOF facility is perfectly suited to strongly improve the knowledge of the ^{231}Pa fission. The proposed measurement will cover an unprecedented energy range, and the high flux result in a good statistics even though the experiment must be performed with very low mass samples.

2 Status of the ^{231}Pa(n,f) cross section

Figure 1 displays the measurements performed in the past along with some evaluations. Among the few measurements available so far, only 4 of them cover a relative large energy range and present large discrepancies. The measurement done by Plattard et al. [1] is a high-quality measurement but its overall normalization is uncertain. Fursov's [2] follow Plattard's data. The data set from Jurado et al. [3] has been obtained by the surrogate reaction ^{232}Th(^{3}He,tf). The energy dependence is different from the previous data and it is lower by 10% beyond 3 MeV and by 20% around the opening of the 2nd chance fission. The recent measurement by Oberstedt et al. [5] is found in agreement with the surrogate method, but departs from it below 2 MeV. In addition, it is highly discrepant with Plattard's data above 15 MeV which is the only other existing measurement at this energy. Last but not least, both ENDF/B-VII.1 and JEFF-3.2 evaluations show similar behavior in the full energy range, but they present 15 % of difference respect to JENDL-4.0u and TENDL-2015 around 10 MeV, and discrepancies larger than 30% respect to EAF-2010. These evaluations, except JENDL-4.0u, extend to high energy range with no data support. Direct measurement of the fission cross section covering the broad energy range available at n_TOF (from thermal to several hundred MeV) will help to constrain the evaluations.

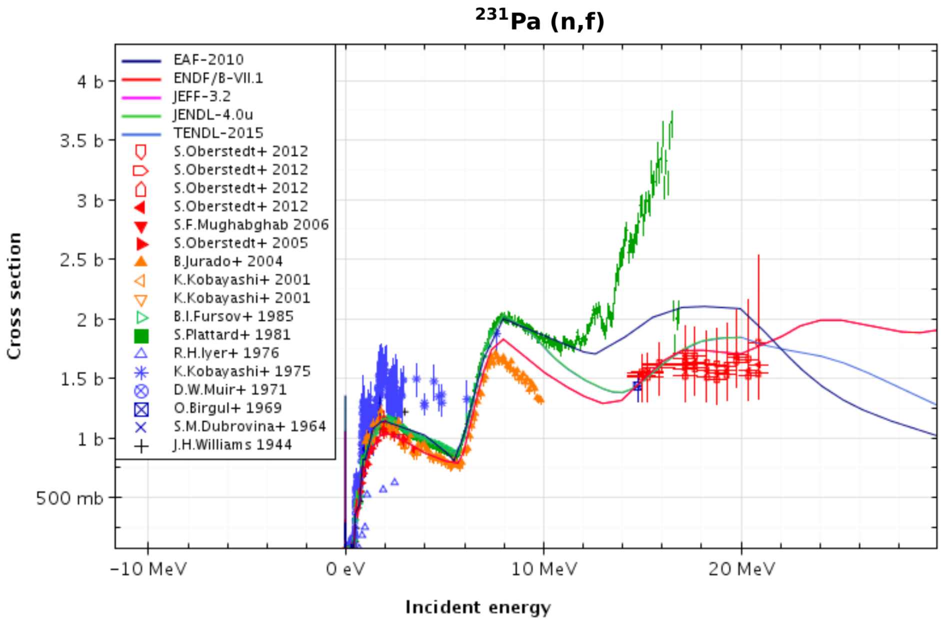

Figure 1: Existing measurements and evaluations of the ^{231}Pa(n,f) cross section.

In addition, this direct measurement will be a good test of the surrogate method in order to extend its validity to isotopes where a direct measurement is not possible, in particular ^{233}Pa, a key nucleus in the ^{232}Th/^{233}U cycle. Given the short half-life of ^{233}Pa (27 d) the surrogate method is the most accurate to measure its (n,f) cross section. However, the surrogate method is not really validated in this area. An accurate, direct measurement of ^{231}Pa(n,f) cross section and its comparison to the data obtained by surrogate will help to assess the method validity, and such a check would be directly transposable to ^{233}Pa as the spin-parity of the ground-state is the same ($\frac{3}{2}^{-}$).

3 Status of the fission fragment angular distribution

Only 2 measurements are reported. Both were performed with mono-energetic neutron beams, thereby on discrete energy values. The older one is from Vorotnikov et al. [7], covering the range 0.16 to 3.5 MeV, and the more recent, from Sicre et al. [8], spans a narrower range: 0.16 to 0.35 MeV. In the former the anisotropy was strongly affected by several corrections, indicating that it was a difficult experiment. Moreover, both sets are not consistent in the region of overlap, possibly reflecting the fact that the anisotropy changes quickly with the energy around the vibrational resonances, which can be hardly traced by discrete neutron energies (see the Plattard data in figure 1 at the threshold). In that respect a measurement at n_TOF would produce much more robust and accurate data, over a wider energy range, thus substantially improving the information on the vibrational resonances.

4 Experimental system and targets

We intend to perform the experiment at n_TOF-EAR2 using a stack of 10 Parallel Plates Avalanche Counters (PPAC), with 9 interleaved targets. The PPACs are gaseous detectors operated at very low pressure (around 5 mbar). Therefore the whole assembly is enclosed in a vacuum-tight vessel, which also acts as a confinement device. Unlike most fission detection systems, which rely on the detection of a single fission fragment, our system is based on the use of thin targets deposited on ultra-thin backing foils, allowing to detect both fission fragments. This coincidence provides an unambiguous tagging of fission events, allowing an excellent rejection of the α activity, and a very good selection against spallation-type reactions caused by the high-energy neutrons. This feature, along with the very fast signals delivered by these counters (10 ns FWMH) makes the PPAC assembly the best suited detection system to extend its measurements in the region of the hundreds of MeV at EAR2.

Any cross-section measurement requires a normalization. For this, we use 2 targets of ^{235}U and 2 of ^{238}U among the 9 available slots, since these isotopes are considered as standards in several energy regions. These targets also provide an accurate time-of-flight calibration: the resonances of ^{235}U are used to determine the length of the neutron flight-path, and the threshold of the ^{238}U is used to determine the ToF offset.

PPACs are position-sensitive detectors, with a resolution of approximatively 2 mm. By measuring the position of both fission fragments, it is possible to determine the position of the fission on the target as well as the angle of emission of the fragments.

The overall detection efficiency is in the order of 50%. It is 100% for fragments emitted perpendicularly to the target, but as the emission angle increases, fragments go through more and more material and their range is so small that the efficiency drops. By measuring the angle between the surface of the detectors and the direction of the fragments, it is possible to determine, in a purely self-consistent way, the efficiency of the system and therefore to correct the number of counts, target by target [9, 10].

The emission of the fragments is not isotropic with respect to the neutron direction. This is actually a quantity we are interested in measuring. In order to achieve this, the measurement of the angle relative to the neutron direction must be independent of the angle relative to the surface of the targets and detectors (as already explained, the latter is related to the efficiency). If the targets are perpendicular to the neutron beam, these angles are similar, therefore the efficiency and the anisotropies can not be disentangled. A simple solution is to tilt the whole assembly by 45° with respect to the neutron beam. The resulting system can be seen on the left picture in figure 2.

This detection system has been used in previous experiments at n_TOF, in order to measure ^{232}Th, ^{234}U and ^{237}Np [9, 10, 11, 12]. As an example we show in the right picture of figure 2 the anisotropy parameter obtained for ^{232}Th(n,f) [9], in a very good agreement with existing data at low energy and extending the knowledge over a large energy domain, reaching 600 MeV.

The IPN group has the skills and ressources to produce thin deposits of ^{231}Pa on thin aluminum backings (2 μm). The electrodeposition method has already been tested and validated. However, the radio-protection rules impose that any batch of ^{231}Pa should not exceed 1 mg. Therefore, any target could contain at most 1 mg. We will prepare

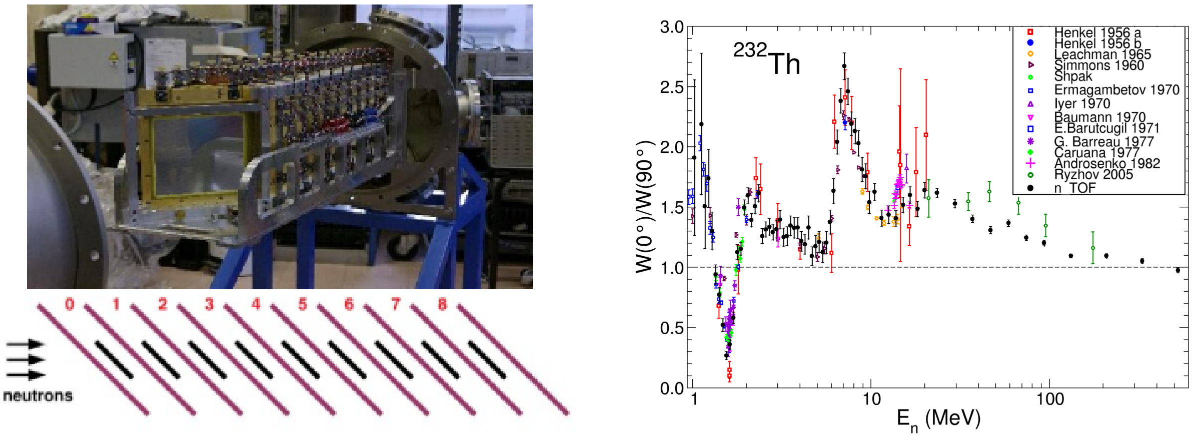

Figure 2: Left picture: view of the detectors assembly and scheme of principle. Right picture: anisotropies in the neutron-induced fission of ^{232}Th; the data obtained at n_TOF using the PPAC assembly are compared to previous measurements.

5 targets using samples of 1 mg each. We assume hereafter, conservatively, that the 5 targets contain a total of 3 mg of ^{231}Pa

5 Experiment and beam time request

Given the small amount of ^{231}Pa, the experiment has to be performed at EAR2 in order to take advantage of the higher flux. Concerning the counting rate, the most stringent condition is the good statistics for the angular distribution around the fission threshold, particularly in the region of the vibrational resonances.
From our previous experiments, we estimate that we need about 1000 counts to obtain an angular distribution with reasonable accuracy (± 0.1 on the anisotropy). On the other hand, the width of these resonances needs an energy binning of at least 100 bins per decade. For the low and narrow resonances below the threshold, 200 bins per decade would be more suited.
We report in figure 3 the expected fission rate represented with a resolution of 100 bins per decade, for 3 mg of ^{231}Pa and $2 \cdot 10^{18}$ protons, using the cross section evaluated in ENDF/B-VII.1 as reference. The angular distributions for the resonances above 300 keV are expected to be well reproduced. The narrow resonance at 156 keV will be less accurately determined but still measurable.
We request $2 \cdot 10^{18}$ protons that will allow a valuable measurement of the angular distribution and the cross section over a hitherto unreached energy domain.

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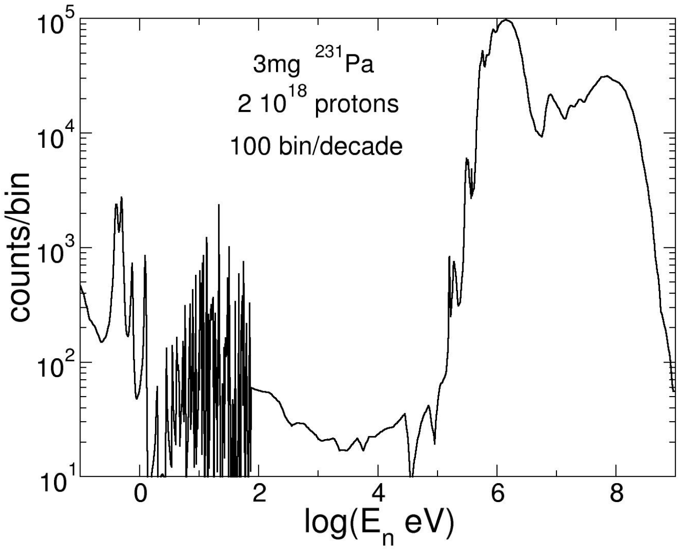

Figure 3: Expected number of events for a total of 2.10^{18} protons and 3 mg of Pa.

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