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Isomeric ratio measurements with the ILL LOHENGRIN spectrometer

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Abstract. The modelling of γ heating and neutron damage inside a nuclear reactor is essential to design the next generation of nuclear reactors. The determination of the fission fragment momentum is a key element to perform accurate calculations of the γ heating. One way to assess this information is to look at the isomeric ratio of different nuclei. According to the lifetime of the isomeric state, different experimental techniques were developed at the LOHENGRIN spectrometer. A focus on the measurement of isomeric ratios of ^{136}I in neutron induced fission of ^{241}Pu is presented. A discussion with the current assumptions used in the evaluation process for isomeric ratio is also shown.

1 Introduction

Seven decades ago, nuclear fission was discovered [1, 2]. Even though the understanding of fission has improved over all these decades, some aspects such as the fission fragment angular momentum were barely studied. Today, this quantity is major for the determination of the prompt γ spectra which are essential in the calculation of γ heating and damage of nuclear reactor components [3]. In order to design the next generation of nuclear reactors with a higher level of confidence, a high accuracy of the prompt γ spectra is required. One probe to study fission fragment angular momentum is to look at isomeric ratio (IR) [4–6]. In this work, a new experimental method is presented to measure IRs at the LOHENGRIN spectrometer [7]. The validation of the assumptions used in the modelling of fission fragment angular momentum in the evaluation process is also discussed.

2 Experimental setup and analysis path

The measurement of IRs were done at the LOHENGRIN mass separator of the Institut Laue-Langevin (ILL). The different parts of the LOHENGRIN spectrometer are shown on Fig. 1 (left). The fissile isotope target is placed near the core of the reactor under a thermal neutron flux of $\approx 5\times10^{14}$ n.cm^{-2}.s^{-1}. The combination of a magnetic and electric field allows a separation of the produced fission fragments according to their ratios mass A over ionic charge q and kinetic energy E_k over ionic charge. Finally a refocusing magnet [8] permits to increase the particle density at the focal position 2.

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According to the lifetime of the isomeric state, different experimental methods were developed. Ref. [9] presents the case of μs isomers whereas in this article a focus on min isomers is shown.

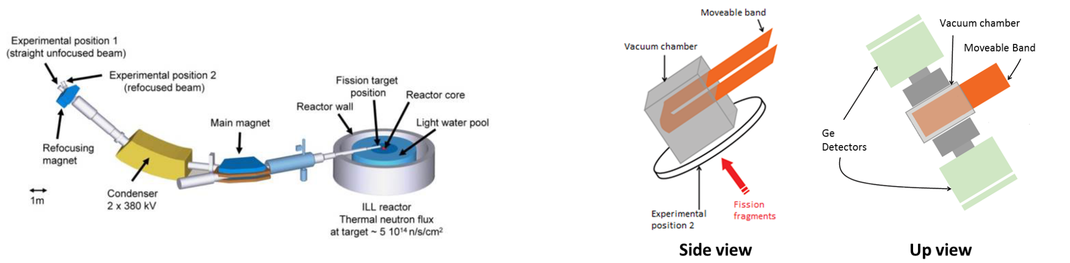

Figure 1. Scheme of the LOHENGRIN spectrometer (left). Detection system placed at the focal position 2 (right).

2.1 Beamcut method

The incoming fission fragments are implemented into a tape surrounded by two clover detectors consisting of four high purity germanium crystals, placed at the focal position 2 of the spectrometer. The band can be moved after each measurement in order to reduce the ambient background coming from the impure selection of the LOHENGRIN spectrometer. Figure 1 (right) shows the scheme of the detection system.

However, for certain nuclei, the detected γ-ray cascade is the same for the ground and the isomeric states. To remove the degeneration, for a given setting of the mass spectrometer, two measurements are performed. When the beam is on, the γ-rays are fed by both states, whereas when the beam is off, because of the difference in the lifetime between states, the γ-rays are fed mostly by one state. Figure 2 summarizes the beamcut method and its principle.

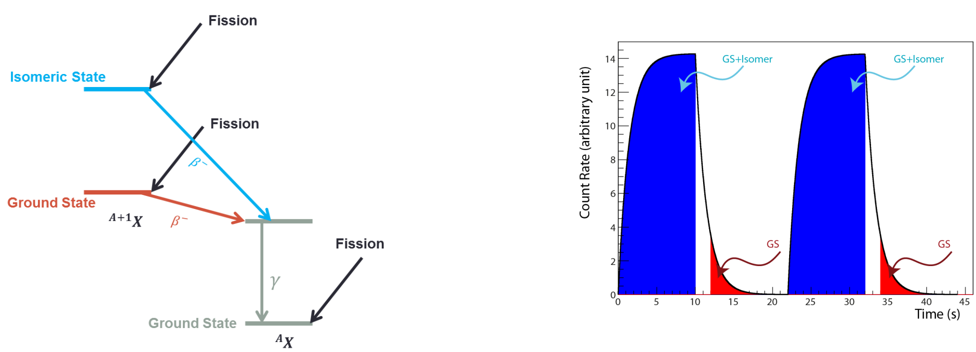

Figure 2. Principle of the beamcut method. When a γ-ray is fed by different states, the combination of two measurements permits to determine the feeding from both states. The blue area is associated to the beam on and the red area corresponds to the beam off.

To test this new procedure, the IR of ^{136}I in neutron induced fission of ^{241}Pu was measured. Indeed, some γ-rays of the isomeric state are contaminated by the isomeric state of ^{136}Xe. A double analysis, one using independent γ-rays and the other using the common γ-rays through the beamcut method was done. Table 1 shows the different γ-rays used in both methods.

Table 1. List of γ-rays used in both methods.

Isotope	Independent γ-rays (keV)	Common γ-rays (keV)
^{136m}I	369.8 / 750	197.3 / 381.4 / 1313
^{136}I	1321.08 / 2289.6 / 2414.6 / 2634.2	1313
^{136m}Xe	$\emptyset$	197.3 / 381.4 / 1313

In this work, the isomeric ratio is defined as following:

$$IR = \frac{\eta(^{m}X)}{\eta(^{m}X) + \eta(^{gs}X)} \tag{1}$$

with $\eta(X)$ the fission rate of the isomeric state (m) or the ground state (gs) respectively. The determination of the IRs from the γ-ray count rate involved several corrections. The current analysis path was presented in Ref. [9]. IRs were measured as a function of the fission fragment ionic charge because the final objective is to determine the isomeric yield. Figure 3 shows the result of both analysis and the correlation matrix calculated for the mean IR. Both methods are in good agreement, which permits to validate the beamcut technique.

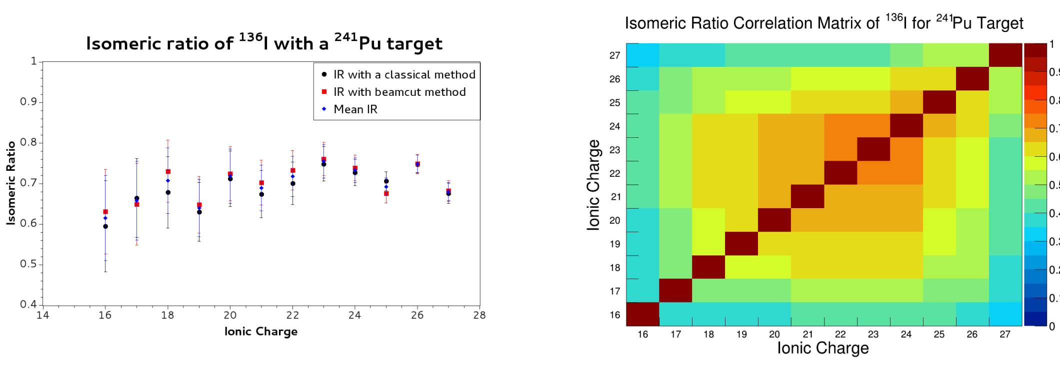

Figure 3. Validation of the beamcut method to determine the IR as a function of the ionic charge (left). Experimental correlation matrix which shows the weight of the systematic uncertainty over the total uncertainty.

3 Results and discussion

Currently, the isomeric ratio determination in the evaluation process is achieved by using the approach developed by Madland and England [10]. Their assumptions are the following : the isomeric ratio depends only on the spin of the isomeric and ground states. Also, all fission fragments have the same spin distribution characterized by a spin-cutoff $J_{rms} = 7.5\ \hbar$. Figure 4 presents the comparison of our result, corresponding to the averaged IR over ionic charge, with previous experiments performed with different fissioning systems, the JEFF evaluation based on the England-Madland hypothesis and a calculation from the code GEF [11].

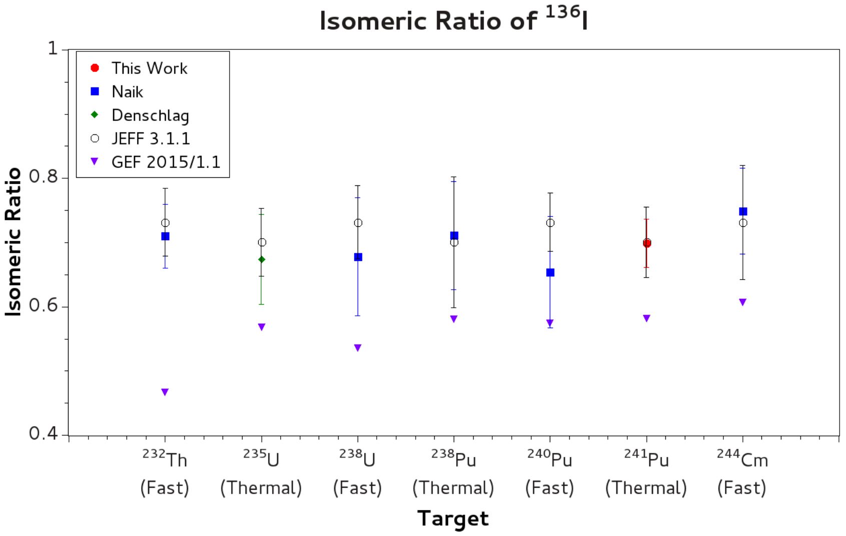

Figure 4. Comparison of the isomeric ratio of ^{136}I with other experimental measurements, the JEFF-3.1.1 evaluation and a GEF calculation. The IR measured doesn't seem to be sensitive to the fissioning system. Also, the Madland-England modelling involved in the evaluation process is in good agreement with experimental values.

The first observation is that the Madland-England modelling seems to be in agreement with all experimental measurements. The second observation is that the isomeric ratio of ^{136}I doesn't seem to be sensitive to the fissioning system, which is in disagreement with the prediction of the GEF calculation.

To go further, an investigation of the isomeric ratio of ^{132}Sn and the extraction of its angular momentum distribution was performed. To determine the spin-cutoff of this nucleus, we used a γ de-excitation code : FIFRELIN [12, 13]. For each state (E^*, J^π) an IR was calculated. More details about the analysis are presented in Ref [9]. Then an average according to the most commonly used spin distribution was performed :

$$IR_{calc}(E^*) = \sum_J \sum_\pi P(\pi)P(J)IR_{calc}(E^*, J^\pi) \tag{2}$$

with $P(\pi) = \frac{1}{2}$ and $P(J)$ following [10] :

$$P(J) \propto (2J+1)\exp\left(-\frac{(J+1/2)^2}{J_{rms}^2}\right) \tag{3}$$

with J_{rms} a free parameter also called spin cutoff. Its value is determined through a Bayesian comparison between experimental isomeric ratios and calculated ones. Finally we extracted the mean $\overline{J_{rms}}$ corresponding to the mean isomeric ratio $\overline{IR}$. The result is compared with a GEF calculation and the Madland-England modelling in Tab. 2. This work shows the limit of the current modelling of the isomeric ratio in the evaluation process. Indeed if all fission fragments have the same spin-cutoff, then in the framework of the Madland-England modelling, the isomeric ratio of ^{132}Sn should be equal to

Table 2. Extraction of a spin-cutoff from an isomeric ratio.

^{132}Sn	$\overline{IR}$	$\overline{J_{rms}}$
This work	0.054 ± 0.006	4.7 ± 0.2
GEF		7.1 ± 0.2
Madland-England a	0.67 ± 0.03	7.5 ± 0.5
Madland-England b	0.054 ± 0.006	2.6 ± 0.1

0.67 (Madl. a) which is incompatible with our measurement. On the contrary if the isomeric ratio is fixed to the experimental value, then the spin-cutoff of ^{132}Sn should be equal to 2.6 (Madl. b) which is incompatible with the extraction done with the FIFRELIN code. Finally, the spin-cutoff calculated by GEF is also in disagreement with the proposed value.

These limits show how necessary it is to build an IR database in order to test and validate the models involved in the evaluation process. This also questions on the mechanism of angular momentum generation [6, 14–17] which is the weakness of the nuclear data evaluation.

4 Conclusion

In this work, a new experimental method was developed in order to extract the isomeric ratio for nuclei where the identification is difficult. The beamcut method was validated on the test case of ^{136}I with a double analysis. The limits of the current modelling of isomeric ratios and angular momentum distribution involved in the evaluation process were pointed out. New measurements are planned in order to bring new information on the mechanism of angular momentum generation of fission fragments. A better understanding of the mechanism would permit to get more accurate nuclear data and satisfy the recommendation of the nuclear data evaluation community.

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