Determination of neutron-induced fission fragment spin distribution after neutron evaporation

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Abstract
Nuclear fission consists of splitting a nucleus into smaller nuclei. Several observables are available to study the fission process such as fission yields or fission fragment angular momentum. Currently, fission models cannot predict all the observables with an acceptable accuracy for nuclear fuel cycle studies for instance. Improvement of fission models is an important issue for the knowledge of the process itself and for the applications. In this work, we take an interest in fission fragment angular momentum distribution. Isomeric ratios (IRs) are a common observable giving access to investigate these distributions. We measured accurate IRs for ⁸⁸Br, ¹³²Sn and ¹³²Te with the fission fragment separator LOHENGRIN and developed a new analysis method to assess the mean value and uncertainty of the IR. An evaluation of the angular momentum distribution of ¹³²Sn was also performed with the FIFFRELIN code.

1. Introduction
Although nuclear fission was discovered seven decades ago [¹][²], the fission process has still some characteristics barely understood such as the fission fragment angular momentum. However, this observable is critical for the determination of the prompt γ spectra, which are central in the calculation of γ heating and damage of nuclear reactor components [³]. A high accuracy of the prompt γ spectra is required in order to design the next generation of nuclear reactors with a higher level of confidence. In this work, the assessment of the fission fragment angular momentum through isomeric ratio measurements is presented [⁴][⁵][⁶]. In a first section, the experimental setup is exposed. Then the data analysis procedure to determine the isomeric ratio and its uncertainty is explained. The angular momentum distribution of ¹³²Sn is then derived. Finally a brief discussion and interpretation of our results are proposed.

2. Experimental set-up
The experiments were performed at the LOHENGRIN mass separator [⁷] located at the high flux reactor of Institut Laue-Langevin (ILL). Figure 1(left) is a scheme of the LOHENGRIN spectrometer. The target of a fissile isotope is placed near the core of the reactor in a thermal neutron flux of ≈ 5 × 10¹⁴ n.cm⁻².s⁻¹. In order to reduce the self-sputtering, the target may be covered by a sputtered Ta layer or a Ni foil (see Table 1) [⁸]. The produced fission fragments are then separated according to the ratios of their mass A over their ionic charge q and their kinetic energy E_k over their ionic charge by the combination of a magnetic and an electrostatic sector. The refocusing magnet [⁹] increases the particle density at the focal position 2.
To identify the incoming fission fragments, an ionisation chamber (IC) surrounded by two clover detectors consisting of four high purity germanium (Ge) crystals were placed at the focal position 2 of the spectrometer [see Fig. 1(right)]. Since signals from the IC and Ge detectors were recorded with a triggerless digital acquisition, the data were analysed off-line.

3. Analysis path

The required observable is the isomeric ratio $IR$:

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

with $\eta(X)$ the fission rate of the isomeric state ($m$) or the ground state ($gs$) respectively. Several corrections are needed to assess this quantity from the raw data. In this section, the method developed to determine the IR mean value and uncertainty is presented. In this paper, only $\mu s$ isomeric states are studied.

3.1 Count rate extractions from $\gamma$ spectra

Since the time of flight within the LOHENGRIN spectrometer is about 1 – 2 $\mu s$, the detection of $\mu s$ isomeric states is possible. All of the isomeric states studied in this work decay through an isomeric transition whereas ground states disintegrate through $\beta^-$ decay. The unique $\gamma$ signature for each of both states allows to identify them unambiguously.

To measure the isomeric state population, a time coincidence of $\approx 10T^{1/2}_{1/2}$ was performed between IC and Ge signals. The ion-gated $\gamma$ spectra were very clean and permitted to extract the isomeric state count rates with a high accuracy (see Fig. 1).

The measurement of the ground state was done by extracting its $\gamma$ lines from the ungated spectra.
Because of the background coming from different separated nuclei and the ambient background, the uncertainty of the ground state count rate was larger than for the isomeric state.

Fig. 2: Ion-gated spectra for $^{88}$Br (left). The identification of the isomeric states is unambiguous. Ungated spectra for $^{88}$Br (right). The extraction of the ground state is more complex because of the $\gamma$ background.

3.2 Total Monte Carlo Method to extract the isomeric ratio

To obtain the isomeric and ground state fission rate $\eta^{(m)X}$ and $\eta^{(g)X}$ from the count rates extracted, different corrections are needed such as the $\gamma$ lines intensity, detector efficiencies and the solution of the Bateman equation for the moment of fission taking into account the lifetimes and branching ratios.

Some parameters, like the lifetime of the isomeric and ground states, appear in different correction factors. The uncertainty propagation is then quite complex. In order to evaluate the uncertainty of the measurement without bias, a Monte Carlo method was developed. The principle is to draw every independent parameters according to a Gaussian distribution and calculate all the correction factors to evaluate the isomeric ratio. This procedure is then repeated (typically $10^6$ times) and permits to obtain the probability density function (pdf) of the isomeric ratio. From this distribution the mean value and the uncertainty of the isomeric ratio is derived. The pdf is globally well reproduced by a Gaussian function, but for low number of counts with an important statistical uncertainty, the distribution is asymmetric. With a classical uncertainty propagation, this observation would not be possible.

3.3 Building of experimental correlation matrices

For a given experiment and nucleus, a set of IRs as function of kinetic energy $E_i$, was measured. Some parameters, $\gamma$ intensity for instance, are common for all of the IR measurements. The experimental covariance matrix can then be built from the method described above. Since the common parameters are independent from one another, the covariance matrix is the product of the sensibility $S_{ik}$ with a term linked to the independent parameters $a_k$ for each measurement:

\[
IR(E_i) = f(\{a_k\})
\]

\[
S_{ik} = \frac{\partial f}{\partial a_k} \left(\frac{\pi}{\pi_i}\right)
\]

\[
\text{Cov}(IR(E_i),IR(E_j)) = \sum_k S_{ik} S_{jk} \sigma_k^2 \frac{\pi_j}{\pi_i} \]

with $f$ the function which relates the parameters and the isomeric ratio. The sensibilities are calculated from the Monte Carlo method. Indeed, for a given parameter, the other being fixed to their mean value,
a drawing was done according to a Gaussian distribution. A distribution of IRs was then obtained as a function of the parameter. The extracted slope around the mean value (the sensibility is a local parameter) permits to determine the sensibility.

4. Determination of the spin distribution for $^{132}$Sn

Fission models can determine the fission fragment angular momentum distribution. To derive this value from the isomeric ratio a $\gamma$ de-excitation code is required. In this work, we used FIFRELIN (Fission FFragment Evaporation Leading to an Investigation of Nuclear data), which is a Monte Carlo code simulating the prompt fission neutron and $\gamma$-ray emission [10, 11]. Its particularity is to describe the fission fragment nuclear structure through the combination of the experimental level scheme and models of nuclear level density. The probability to decay from an initial state to a final state is related to models of $\gamma$ strength function. In this article, CTM (Constant Temperature Model) [12] and EGLO (Enhanced Generalized LOrentzian model) [13] were used as nuclear level density and $\gamma$ strength function models.

For a given excitation state and angular momentum $(E^*,J^\pi)$, called an entry state, FIFRELIN is able to produce a $\gamma$ de-excitation and then compute the probability to feed both isomeric and ground states. In other words, for each entry state, FIFRELIN calculates an isomeric ratio.

In this work, only the angular momentum distribution of $^{132}$Sn was studied. Its entry states were divided by bins of 200 keV from the isomeric state energy, to the neutron binding energy. Indeed, since measurements were done after neutron evaporation, the angular momentum distribution after neutron emission is the more accurate. The binning in the $J^\pi$ axis is $1\ h$ from $0^\pi$ to $30^\pi$. Figure 3 presents the principle of the FIFRELIN calculation (left) and the IR as a function of the entry state (right).

The calculated IRs are then averaged:

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

(3)

with $P(\pi) = \frac{1}{2}$ and $P(J)$ following [14]:

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

(4)
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. Figure 4 presents the probability to reproduce experimental results as a function of the excitation energy and the spin cutoff (left). The warmer the colors, the more probable the initial distributions are. In this example, an angular momentum distribution parametrized by a spin cutoff of $J_{rms} \approx 5 \hbar$ is more probable than with $J_{rms} \approx 10 \hbar$. This assessment is only valid if the calculated IRs (with the most probable spin cutoff value) are in agreement with the experimental results, which is the case as shown in Fig. 5.

Fig. 4: Exclusion plot for $^{132}$Sn at $E_k = 75$ MeV obtained from the comparison between experimental and calculated IRs (left). Spin cutoff as a function of the measured kinetic energy (right). A clear dependence is shown. The mean excitation energy is obtained without any prior in the analysis.

5. Results and discussion

An angular momentum distribution was extracted from our measurement of the isomeric ratio of $^{132}$Sn. Figure 5 presents the experimental results, compared with the calculations performed by FIFRELIN (with the most probable spin cutoff determined through a comparison with experimental IRs) and the experimental correlation matrix. A clear dependence of the IRs and the spin cutoff [see Fig. 4 (right)] as a function of the kinetic energy is shown. This observation would lead to exclude the common mechanism of generation of fission fragment angular momentum, the so-called bending and wriggling modes [15]. Indeed in this model, a deformed pair of fission fragments is required. Otherwise the angular momentum is expected to be equal to zero [16, 17, 18]. Since the $^{132}$Sn is supposed spherical at the scission point, our results are in contradiction with these modes. New theoretical calculations must be performed in order to interpret this result. It seems that $^{132}$Sn is particularly appropriate to test the limits of angular momentum generation models. However, for deformed nuclei, the role of the deformation energy must be investigated in order to describe the mechanism of angular momentum generation.

In addition, results on $^{132}$Te and $^{88}$Br tend to show the significance of the covered target role in the kinetic energy dependence of IRs. A thick target washes out the correlation between these two quantities whereas the cover shifts the kinetic energy. Figure 6 illustrates this phenomenon by comparing IRs with differently covered targets.

6. Conclusion

The measurement of IRs as function of kinetic energy can provide useful information for theoretical works on the fission process and more exactly on the generation of the fission fragment angular mo-
Fig. 5: Comparison between experimental and calculated IRs as a function of the measured kinetic energy for $^{132}$Sn (left). The experimental correlation matrix shows the weight of the systematics to the total uncertainty (right).

Fig. 6: IRs as a function of the measured kinetic energy. The slope depends drastically on the thickness of the target. Results are shown for $^{88}$Br (left) and $^{132}$Sn (right).

The role of the covered target was emphasized. Thus the use of a thin target with a thin and regular cover is required to investigate properly the dependence of the IRs with kinetic energy. A new method of analysis was developed and permitted to obtain the probability function of IRs. Experimental correlation matrices were built and shed light on the leading role of systematics in the total uncertainty. To go further and reduce the uncertainties, an emphasis on the nuclear structure of the studied nuclei must be done.

A complete set of data, with different fissioning system and nuclei, will permit to validate the different models describing the fission fragment angular momentum. In this framework, $^{132}$Sn is a cornerstone to test in depth the model robustness.

References