

Investigation of fission product isomeric ratios and angular momenta of ^{132}Sn populated in the $^{241}\text{Pu}(\text{n}_{\text{th}}, \text{f})$ reaction

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Abstract. During an experimental campaign performed at the LOHENGRIN recoil spectrometer of the Institut Laue-Langevin (ILL), a kinetic energy dependence of ^{132}Sn fission product isomeric ratio (IR) has been measured by inducing thermal fission of ^{241}Pu . The IRs are deduced using gamma ray spectrometry in coincidence with the ionisation chamber. To interpret these data, we use the FIFRELIN Monte-Carlo code to simulate the de-excitation of the fission fragments. Combining the IRs with the FIFRELIN calculations, the angular momentum distribution with kinetic energy of the doubly magic nucleus of ^{132}Sn was deduced. This will be compared with the angular momentum distribution obtained for the reaction $^{235}\text{U}(\text{n}_{\text{th}}, \text{f})$ for ^{132}Sn .

1 Introduction

The past decade has seen a growing energy need and thus a renewed interest in nuclear energy. Nonetheless, to make the new reactors more safe, the existing technology needs to be improved and the challenge for new and innovative fuel must be overcome. Furthermore, a precise understanding of the fission process is a noteworthy challenge faced by nuclear physicists even though eight decades have passed since its discovery in 1939 [1, 2]. Many different models and hypotheses such as the liquid drop model, shell model etc. have been developed to explain and reproduce the experimental data obtained as well as to improve the understanding of fission. However, the angular momenta of the fission fragments are a poorly known quantity. Angular momenta of fission fragments are a component of the phenomenological model used to assess nuclear observables used in applications as prompt gammas and neutrons. This angular momentum can be estimated by using models, see for example Ref. [3], or through direct measurements, see Ref. [4] and references therein. On the other hand, one can study indirectly the angular momenta of the fission fragments by measuring the isomeric ratios of the fission fragments [5–7].

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2 Experiment

The experiment was conducted at the LOHENGRIN recoil spectrometer [8] at the Institut Laue-Langevin (ILL) [9]. The LOHENGRIN recoil spectrometer was built in the 1970s and is one of the key instruments at the ILL to carry out experiments for nuclear physics and nuclear data. The LOHENGRIN recoil spectrometer enables us to study the mass, charge and kinetic energy distribution of the fission products. These fission products are obtained by exposing a fissile or fertile target to a thermal neutron flux of about $5 \times 10^{14} \text{ n/cm}^2/\text{s}$ near the core of the ILL high-flux reactor. The total length of LOHENGRIN is 23 m and has a high mass resolution ($A/\Delta A \approx 400$) [10], which is dependent on target size. By using the electric and magnetic fields of LOHENGRIN, fission products of interest are extracted and brought to the detection area. These fields are perpendicular to each other and have focussing properties in their respective planes.

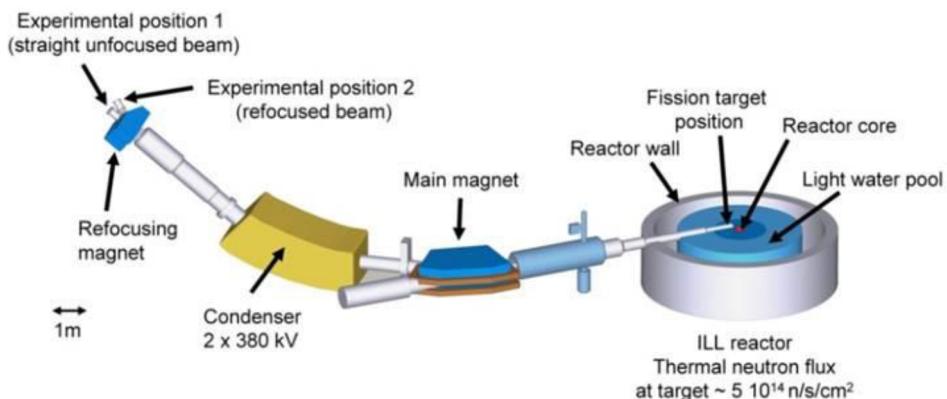


Figure 1. Schematic of the LOHENGRIN mass spectrometer showing the dipole magnet in blue (centre) and the electrostatic condenser in yellow. They are used to select and divert the charged fission products.

For this experiment, we used ^{241}Pu ($39.9 \mu\text{g}/\text{cm}^2$) as the target nuclei. By varying the electric and magnetic fields, the desired fission products are selected by the A/q and E_k/q ratio, where A is the desired mass of the fission product, q is the ionic charge and E_k its kinetic energy. As can be seen from Fig. 1, there are two experimental positions present. At experiment position 1, the beam has an energy dispersion of $7.2 \text{ cm per } 1\% \text{ energy difference}$, whereas at position 2, the beam is refocussed in the energy axis using the RED magnet [11] and, hence, the particle flux density is increased by up to a factor of seven [10] as compared to position 1. In our experiment, the detectors were set up at position 2. The detection setup consists of an ionisation chamber and two HPGe clover detectors with four germanium crystals each. This is depicted in Figure 2.

The extracted fission products are then implanted in the Al foil at the top of the ionisation chamber. The gamma rays, emitted by internal transition (IT) of the isomeric state and after the β^- decay of the ground state are detected using the two clover detectors. By using LOHENGRIN, ^{132}Sn was extracted at ionic charges 20 and 24 respectively with kinetic energy ranging from 57 MeV to 84 MeV.

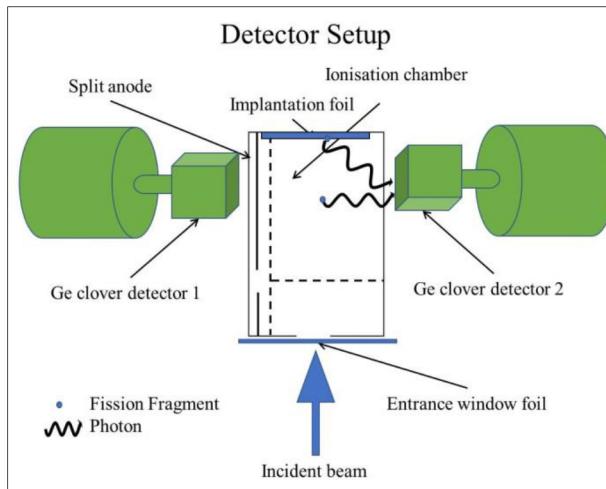


Figure 2. Detector setup showing the two HPGe clover detectors in green and the split-anode ionisation chamber in between them. The implantation foil is placed at the top of the ionisation chamber to trap the fission products.

3 Isomeric ratios from experiments

Isomeric ratios (IR) are defined as ratio of the production rate (P_i) of one isomeric state to the sum of the production rates of all the isomeric states and the ground state (GS).

$$IR = \frac{P_i}{\sum P_i + GS} \quad (1)$$

3.1 The analysis

^{132}Sn has one microsecond isomeric state at 4848.5 keV having $J^\pi = 8^+$ with a half-life of $2.08 \mu\text{s}$ [12]. The analysis was carried out off-line. To analyse the gamma rays originating from the isomeric state, a coincidence spectrum was generated. The time coincidence window was set to $20 \mu\text{s}$. The coincidence was created between the ionisation chamber and the HPGe clover detectors. By using the coincidence method to generate a spectrum, we are able to reduce the gammas originating from room background and beta decays and gammas from β^- decay of the ground state of the parent nucleus (^{132}In). Thus, we obtain a clean spectrum for the gamma rays originating from the microsecond isomeric state of ^{132}Sn as seen in the Fig. 3. For the measurement of the gamma rays originating from the β^- decay of the ground state (^{132}Sn), the ungated spectrum was used.

The gamma spectrum was analysed using the TV gamma spectrum analyser program [13]. The intensities of the gamma rays were obtained from literature [12] and the efficiencies for the germanium detectors were obtained from simulations validated with experimental data. To calculate the IR from the counts obtained from the gamma spectra, firstly, we have to calculate the production rates of the isomeric state as well as the ground state. To do this, the Bateman equations need to be solved. Along with this, corrections for the decay during flight also need to be taken into account; this is due to the fact that the distance between target and the experimental focal plane is 23 m. To accurately determine the production rates and the IR along with their respective uncertainties, a Monte-Carlo method [5, 14] is being used. This

Monte-Carlo Code (MCC) is used as compared to the analytical method as there are quite a few parameters involved in different corrections, which need to be added. Hence trying to propagate and calculate their uncertainties analytically would be a complex task.

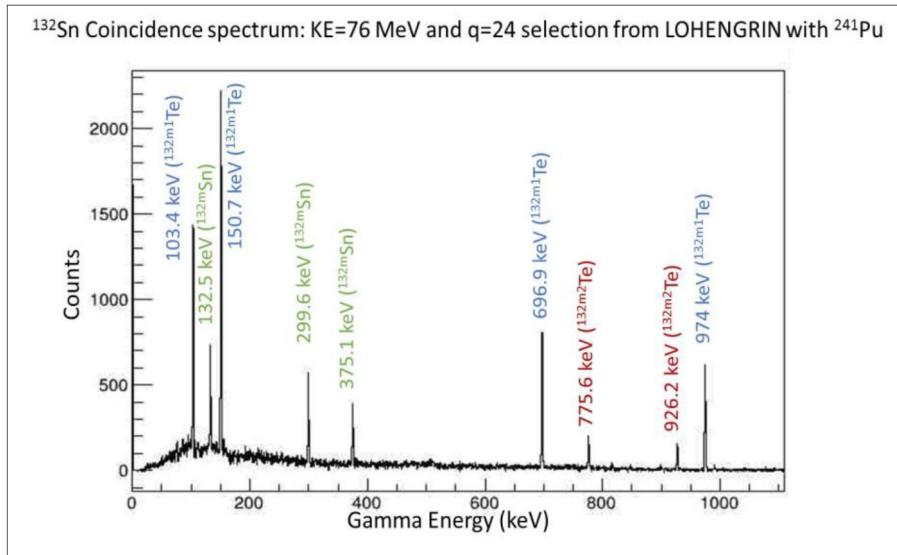


Figure 3. Gated Spectrum showing the three gamma rays of interest (in green) from the ^{132}Sn isomeric state (IS). Gamma rays from the two ISs of ^{132}Te are also observed in this spectrum.

As input to the MCC, one must provide the half-life of the states (ground and isomeric), counts and the error on counts which are extracted using TV gamma spectrum analyser, detection efficiency, the gamma intensities and the normalisation factor for the gamma intensities, the branching ratio, kinetic energy selected by LOHENGRIN and the flight path length. The MCC then calculates the average number of disintegrations (N_d), the production rates and the isomeric ratios along with their respective uncertainties. One can also obtain sensibility plots as well as covariance matrices from the MCC.

In the MCC, N_d is calculated for each of the gamma rays (N_{di}) arising from a particular state, these N_{di} are then used to calculate the mean N_d . The χ^2 test is used to verify whether a 90% level of confidence is achieved. If not, progressively, uncertainty is added [15] and the N_d is recalculated to achieve the test criteria. Thus, a final mean value of the N_d along with its uncertainty is obtained. The systematic uncertainties account for less than 5%, whereas the statistical uncertainties go as high as 50%.

The beamtime had a span of ten days and hence, not all the measurements were performed consecutively or even on the same day. For this reason, we must take into account the target evolution. To see the target evolution, several energy scans were carried out throughout the experiment schedule. The shift in the mean value of the kinetic energy between the first and the last experimental days is equal to (4.6 ± 1.0) MeV. It should be noted that the evolution of the mean kinetic energy was linear with a slope of (-0.52 ± 0.09) MeV/day. For each kinetic energy selected with the LOHENGRIN spectrometer, IRs are measured. By combining all the experimental data (weighted by the kinetic energy distribution) the mean IR can be derived and is equal to 0.0719 ± 0.0016 . It should be noted that covariance was not taken into account for the uncertainty propagation.

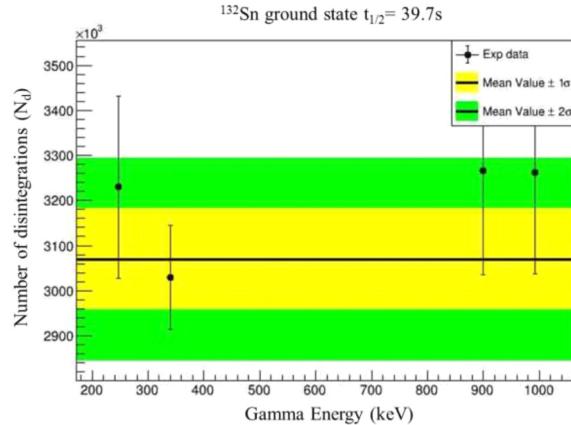


Figure 4. Number of disintegrations calculation using the MCC for GS for 72 MeV and q=24 selection of LOHENGRIN.

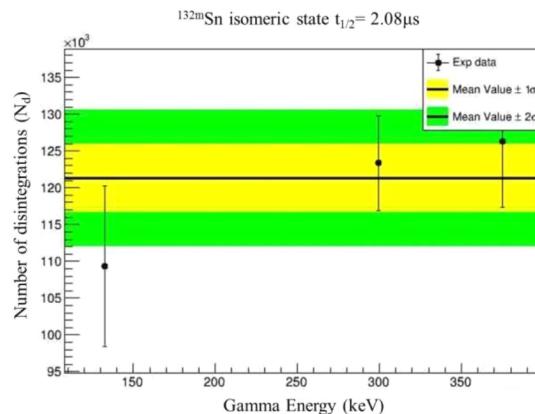


Figure 5. Number of disintegrations calculation using the MCC for IS for 72 MeV and q=24 selection of LOHENGRIN.

4 Spin extraction using FIFRELIN

Fission FRagment Evaporation Leading to an Investigation in Nuclear data (FIFRELIN) [16] is a Monte-Carlo code developed at CEA. This code simulates the fission process and gives information on fission observables such as prompt neutrons, gammas, neutron multiplicity. For this work, this code has only been used to carry out the de-excitation simulation of the nuclei of interest (¹³²Sn). The inputs provided for this code are the mass number (A), the atomic number (Z), excitation energy (E*) and spin along with parity (J^π). It also requires additional files from the RIPL-3 2015 [17, 18] library to get information on the nuclear levels and the gammas and electrons emitted at lower energies. Additional models are also required such as the Composite Gilbert-Cameron Model (CGCM), which is propositioned in RIPL-3 [17] for nuclear level densities, Back Shifted Fermi Gas Model (BSFGM) for spin cut-off and the Enhanced General Lorentzian (EGLO) model [19] for the gamma strength functions. The BRICC code [20] is used to calculate the internal conversion coefficients. FIFRELIN

uses these input libraries and assumes that the experimental level scheme provided in the RIPL-3 2015 file is complete up to cut-off energy (E_{cut}). After this E_{cut} , it is assumed that the experimental level scheme is incomplete and this is then filled by FIFRELIN using the CGCM model. The BSFGM is used to attribute a J^π value to these new levels and the EGLO model is used to get the gammas and their respective intensities originating from these levels. The probability to emit neutrons are obtained from the neutron transmission coefficients which are derived from an optical model; the Koning-Delaroche global neutron optical model potential [21] but in our case, no neutrons are emitted as the simulations were run below neutron separation energy.

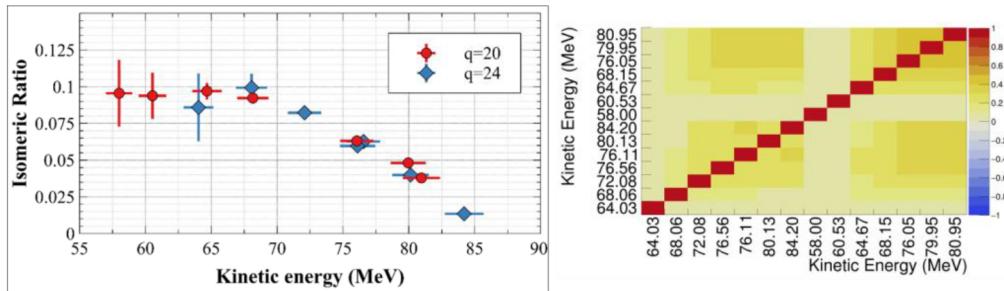


Figure 6. Isomeric Ratio of ^{132}Sn from thermal neutron induced fission of ^{241}Pu measured at two different ionic charge selections (left) and the associated covariance matrix (right). The kinetic energies are corrected from the relative evolution of the energy loss during the experimental campaign.

Once the input parameters, A , Z , E^* and J^π are provided, a cascade can start. The above-mentioned models and experimental level schemes are used to calculate the isomeric ratio for a particular E^* and J^π combination. The E^* has a range starting at the energy of the isomeric state up to the neutron separation energy. For each excitation energy, a range J^π of values (0^\pm to 30^\pm) is given. For each of these J^π values, an IR ($\text{IR}_{FIF}(E^*, J^\pi)$) is calculated by FIFRELIN. To compare these results with the experimental data, the results are averaged by the equation given below:

$$\text{IR}_{FIF}(E^*, J_{rms}) = \sum_J \sum_\pi P(\pi) P(J) \text{IR}_{FIF}(E^*, J^\pi) \quad (2)$$

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

$$\text{and } P(\pi) = P(\pm 1) = \frac{1}{2} \quad (4)$$

The Likelihood method is used to adjust the spin cut-off (J_{rms}), which in that case is a free parameter

$$L(E^*, J_{rms} | E_k) \propto \exp\left(-\frac{\left(\text{IR}_{exp}(E_k) - \text{IR}_{FIF}(E^*, J_{rms})\right)^2}{2(\sigma_{exp}^2, \sigma_{FIF}^2)}\right) \quad (5)$$

where, $\text{IR}_{exp}(E_k)$ and σ_{exp} are the isomeric ratios and their uncertainty obtained from the experiments that is dependent on the selected kinetic energy from LOHENGRIN. σ_{FIF} is the uncertainty obtained from FIFRELIN.

5 Results and conclusion

Combining the FIFRELIN calculations with the experimental results, we were able to extract the J_{rms} value as a function of kinetic energy. Figure 7 depicts the results from this work, which have been compared to the results obtained by using a ^{235}U target [5]. It can be seen that the J_{rms} value obtained from two different fissioning systems are quite similar. Also for this work, one can observe a flat plateau-like region at lower kinetic energies. Further experiments and calculations need to be carried out to explain this phenomenon. Experimental results from this work have been further compared with the calculations using the Madland-England (M.E.) model and the GEF code. This can be seen in Table 1. The M.E. model uses the assumption that the isomeric ratio is only dependant on the spin of both the ground state and the isomeric state. Furthermore, it works on the supposition that all the fission fragments are characterised by a spin cut-off value of $(7.5 \pm 0.5) \text{ h}$, which gives an isomeric ratio of 0.642 ± 0.039 for ^{132}Sn (M.E. (a)). By using the isomeric ratio from this work, which is 0.0719 ± 0.0016 , a J_{rms} of 2.8 ± 0.1 (M.E. (b)) is obtained from the M.E. model. We clearly observe a mismatch between the experimental results and those obtained from the M.E. model for ^{132}Sn .

Table 1. Comparison of results: In M.E. (a), $\overline{J_{rms}}$ is set to 7.5 h and the IR is calculated by using M.E. model. In M.E. (b), the IR is set equivalent to our experimental result and the $\overline{J_{rms}}$ is calculated using the same model.

Experiments	\overline{IR}	$\overline{J_{rms}} (\hbar)$
This work (^{241}Pu)	0.0719 ± 0.0016	4.8 ± 0.1
^{235}U [5]	0.054 ± 0.006	4.7 ± 0.2
Models		
Madland-England (a)	0.642 ± 0.039	7.5 ± 0.5
Madland-England (b)	0.0719 ± 0.0016	2.8 ± 0.1
GEF [22]	0.234	6.65 ± 0.03

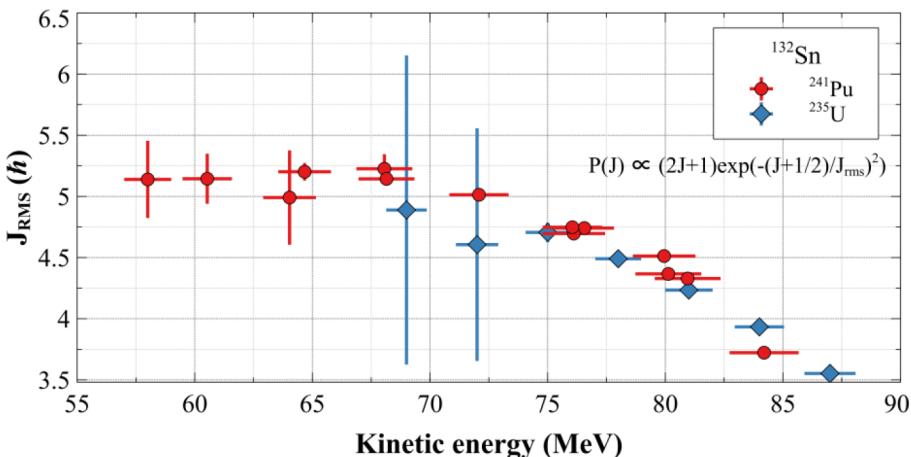


Figure 7. Spin of ^{132}Sn and its dependence on kinetic energy using ^{241}Pu and ^{235}U targets. The kinetic energies are corrected from the relative evolution of the energy loss during the experimental campaign.

In conclusion, the dependence of the isomeric ratios of the fission products on their kinetic energy was obtained. Using statistical analysis, along with FIFRELIN calculations involving level density models, gamma strength functions, spin cut-off models and internal conversion coefficients, we were able to determine the J_{rms} value for each of the isomeric ratios obtained. Furthermore, it can be observed that the J_{rms} values for two different fissioning systems are quite similar and follow a similar trend. For nuclei such as ^{132}Sn , the results from the experiments should be taken into account for the nuclear data evaluations instead of using codes or models to compute them. In this same experimental campaign, experiments were carried out on other isotopes of Sn as well. It would be very interesting to see how the J_{rms} value changes with change in mass for the same element.

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Raw data of this experiment are available via ref. [23].

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