Angular momentum of doubly magic $^{132}$Sn fission product: experimental and theoretical aspects

Olivier Serot$^{1,}$, Abdelhazize Chebboubi$^{1}$, Jehaan Nicholson$^{1}$, Grégoire Kessedjian$^{1}$, Olivier Litaize$^{1}$, Christophe Sage$^{2}$, Olivier Meplon$^{2}$, Mourad Ramdhané$^{2}$, Ulli Köster$^{3}$, and Yung Hee Kim$^{3}$

$^{1}$CEA, DES, IRESNE, DER, SPRC, Cadarache, 13108 Saint Paul lez Durance, France
$^{2}$LPSC, Université Grenoble Alpes, CNRS/IN2P3, 38026 Grenoble Cedex, France
$^{3}$Institut Laue Langevin, 38042 Grenoble, France

Abstract. Despite the numerous theoretical and experimental works published very recently, the way in which fission fragments acquire their angular momentum is still an open question. This angular momentum generation mechanism is important not only for improving our understanding of the fission process, but also for nuclear energy applications, since the angular momentum of fission fragments strongly impact the prompt gamma spectra and consequently the decay heat in a reactor. In this context, within the framework of a collaboration between the ‘Laboratoire de Physique Subatomique et Corpusculaire’ (LPSC, France), the ‘Institut Laue Langevin’ (ILL, France) and the CEA-Cadarache (France), an experimental program was developed on the LOHENGRIN mass-spectrometer with the aim of measuring isomeric ratio of some fission products for different thermal-neutron-induced fission reactions. This paper will be focused on the results obtained for the spherical nucleus $^{132}$Sn following thermal-neutron-induced fission of both $^{235}$U and $^{241}$Pu targets. To further challenge the angular momentum generation models, $^{132}$Sn isomeric ratio (IR) was measured as a function of $^{132}$Sn fission product kinetic energy ($KE$). The angular momentum was determined by combining our experimental data with the calculations performed with the FIFRELIN Monte Carlo code. A clear angular momentum decrease with $KE$ was observed for both reactions. Lastly, we investigate the dependence of the $^{132}$Sn angular momentum with the incident neutron energy, from thermal region up to 5 MeV (below the second-chance fission). For that, the four free available parameters in FIFRELIN are selected in order to reproduce the average prompt neutron multiplicity. In this way, the angular momentum is deduced for each neutron energy. These results are discussed in terms of the impact of the available intrinsic excitation energy at scission on the spin generation mechanism.

1 Introduction

Nuclear fission is such a complex phenomenon that despite more than 80 years of intensive research, we are still unable to predict the main observables of fission from a consistent and robust fission model. For example, the question of how fission fragments acquire their spin (total angular momentum) is still unclear, as demonstrated by the recent workshop on this topic [1]. This question is however important because the spin of the fission fragments and the available excitation energy at scission affect the de-excitation process and has therefore an impact on the characteristics of the emitted prompt neutrons and gamma rays.

In this paper, we focus on the spin of the doubly magic $^{132}$Sn nucleus produced after neutron evaporation in the case of the thermal-neutron-induced fission of $^{235}$U and $^{241}$Pu. In the first section, we will briefly recall how the spin of $^{132}$Sn was determined as a function of its kinetic energy thanks to the isomeric ratio measurements. In section 2, we discuss the role of the intrinsic excitation energy available at scission on the mechanism of spin generation.

From these considerations, we investigate the average spin of $^{132}$Sn as a function of the incident neutron energy during a low-energy fission reaction (before the second-chance fission).

2 $^{132}$Sn total angular momentum as a function of its kinetic energy

Isomeric Ratio measurements

One way to assess the spin of fission products is to measure their isomeric ratio, as proposed a long time ago by Huizenga and Vandenbosch [2, 3]. So, in the frame of the collaboration between the ‘Laboratoire de Physique Subatomique et Corpusculaire’ (LPSC, France), the ‘Institut Laue Langevin’ (ILL, France) and the CEA-Cadarache (France), experimental campaigns were performed on the LOHENGRIN recoil mass spectrometer of the Institut Laue-Langevin with the aim of measuring, after prompt neutron emission, the dependence of the isomeric ratio with the kinetic energy ($KE$). The isomeric ratio is defined by:

$$IR(KE) = \frac{R^{^{132}Sn}(KE)}{R^{^{132}Sn}(KE) + R^{^{208}Pb}(KE)}$$ (1)
Figure 1. Isomeric Ratio of $^{132}$Sn measured as a function of its kinetic energy (left scale, black points). The $^{132}$Sn kinetic energy distribution is given by the blue curve (right scale). Top: results from the $^{235}$U(n$_{th}$,f) reaction [4]. Bottom: results from the $^{241}$Pu(n$_{th}$,f) reaction [5, 6].

where $R_{^{132}Sn}$ and $R_{^{132}Sn}$ are respectively the production rate of the isomeric state and ground state. In the present study, the isomeric state is characterized by its energy $E_1$=4.847 MeV, its spin parity $J^π$=8$^+$ and its half life $τ$=2 µs, while the ground state is characterized by its energy $E$=0 MeV, its spin parity $J^π$=0$^+$ and its half life $τ$=39.7 s.

The experiment on the $^{235}$U(n$_{th}$,f) reaction was performed in 2015 and is reported in Ref.[4], while the experiment on the $^{241}$Pu(n$_{th}$,f) reaction was carried out in 2019 (see Refs.[5, 6]). Results on both reactions are plotted in Fig. 1. In this figure, the $^{132}$Sn kinetic energy ($KE$) distributions (after prompt neutron emission) measured thanks to the mass spectrometer LOHEGRIN are also shown. By weighing $IR_{KE}$ with this $KE$-distribution, we obtained the average isomeric ratio <$IR$> which is reported in Tab. 1.

**Spin assessment**

In order to assess the spin of the $^{132}$Sn fission product from our measured isomeric ratio, we use the Monte Carlo FIFRELIN code [7, 8]. This code is capable to simulate the de-excitation of a nucleus by means of the Hauser-Feshbach statistical theory [9]. Hence, for an initial state defined by its excitation energy and spin-parity ($E^*$, $J^π$) as shown in Fig. 2, the probability to feed the isomeric and the ground states can be calculated and the $IR$ can be deduced. Since we want to reproduce the measured $IR$ after prompt neutron emission, FIFRELIN calculations were limited to the following energy range (by step of 0.5 MeV): $E_1 < E^* < S_n$ where ($E_1$ is the energy of the isomeric state and $S_n$=7.3 MeV the neutron separation energy), and for $J^π$ varying from 0$^+$ to 30$^+$. Assuming that the spin distribution of the $^{132}$Sn fission product follows a Rayleigh-type distribution (see Eq. 2) and using a Bayesian statistical analysis, we were able to extract, for each kinetic energy, the best spin cutoff parameter $σ^2$, i.e. the one which best reproduces the experimental isomeric ratio.

$$P(J) = \frac{2J+1}{2πσ^2} exp \left\{ -\frac{(J+0.5)^2}{2σ^2}\right\} \hspace{1cm} (2)$$

The detailed procedure used to extract the spin of the fission product can be found in Ref. [4]. The dependence of the $^{132}$Sn average spin with its kinetic energy for both $^{235}$U(n$_{th}$,f) and $^{241}$Pu(n$_{th}$,f) reactions is shown in Fig. 3. For both reactions, a similar trend can be observed, i.e a rather flat behavior at low kinetic energy and then a decrease of the spin with increasing $KE$. The average spins <$J$> obtained by weighting results shown in Fig. 3 with the $^{132}$Sn kinetic energy distribution (blue curve of the Fig. 1)

Table 1. Average isomeric ratio and spin of the $^{132}$Sn produced after prompt neutron emission from $^{235}$U(n$_{th}$,f) and $^{241}$Pu(n$_{th}$,f) reactions

<table>
<thead>
<tr>
<th>Reaction</th>
<th>&lt;$IR$&gt;</th>
<th>&lt;$J$&gt; (ℏ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{235}$U(n$_{th}$,f)</td>
<td>0.0541 ± 0.007</td>
<td>3.44 ± 0.15</td>
</tr>
<tr>
<td>$^{241}$Pu(n$_{th}$,f)</td>
<td>0.072 ± 0.026</td>
<td>3.76 ± 0.03</td>
</tr>
</tbody>
</table>
Figure 3. Dependence of the average spin with the $^{132}\text{Sn}$ kinetic energy for $^{235}\text{U}(n_{\text{th}},f)$ (red points) and $^{241}\text{Pu}(n_{\text{th}},f)$ (blue points) reactions.

are given in Tab. 1. Two additional comments can be mentioned:

- Since the probability to emit the prompt neutrons is very low for the doubly magic $^{132}\text{Sn}$, the spin distributions before and after prompt neutron emission are close to each other;
- Our results (see Table 1), are consistent with the recent experimental data obtained by Wilson [10], where an average spin between 3.5 $\hbar$ and 4 $\hbar$ was found around the mass 130 for the three investigated fissioning systems. They are also consistent with the calculation of Marevic [11] that predicts an average spin (before prompt neutron) around 2.5 $\hbar$ in the mass region 132. The same order of magnitude was found from the theoretical work proposed by Randrup [12].

3 Discussion

3.1 Dependence of $\langle J \rangle$ with the Kinetic Energy

In order to interpret, at least qualitatively, the dependence of $\langle J \rangle$ with the kinetic energy, Fig. 4 can be helpful. On this figure, proposed by Thomas [13] and continued by Gönnenwein [14], the energy components (deformation, coulomb and intrinsic) are plotted for a given fission fragment pair, as a function of the elongation of the system at scission. On the x-axis, the deformation starts from two spherical compact fission fragments up to two deformed fission fragments configuration. These two extreme cases are called respectively “cold compact” and “cold deformed” because in both cases no intrinsic excitation energy is available. An “intermediate” case is also considered, where the light fragment is strongly deformed, while the heavy one ($^{132}\text{Sn}$) is spherical. Note that the sum of the three energy components is constant and must be equal to the Q of the reaction (horizontal line). Since the stiffness parameter which describes the resistance of a nucleus against deformation is very high for the doubly magic $^{132}\text{Sn}$ fission product, the phase space between “intermediate” and “cold deformed” configurations (black points in Fig. 4) will almost never be reached during the fission process. From the figure, we see that when the $KE (V_{\text{end}})$ increases (from the right to the left), then the intrinsic excitation energy decreases, as also observed for the average spin (Fig. 3). In other word, the average spin seems to be generated mainly by the available intrinsic excitation energy at scission.

3.2 Average spin as a function of $E_n$

In this sub-section, we try to calculate the dependence of the $^{132}\text{Sn}$ isomeric ratio with the incident neutron energy, $E_n$, for the $^{235}\text{U}(n,f)$ reaction. For that, the four free parameters available in FIFRELIN are tuned in order to reproduce the total prompt neutron multiplicity $\nu_{\text{TOT}}$ given in the JEFF-3.3 library (see upper part of Fig. 5). FIFRELIN calculations are performed for three incident neutron energies (before the second-chance fission): thermal, 2 MeV and 5 MeV. It is generally admitted that the additional energy brought by the incident neutron appears mainly under the form of intrinsic energy (not deformation energy) and goes essentially into the heavy fragment (see for example Ref.[15]). As a consequence, the average prompt neutron multiplicity of the light fission fragment group stays rather constant, while the average prompt neutron multiplicity of the heavy fission fragment group is increasing with $E_n$. This phenomenon, observed experimentally (see Ref.[16]
product was measured as a function of its kinetic energy for both $^{235}$U(n,f) [4] and $^{241}$Pu(n,f) [5, 6] reactions. By reproducing these experimental data thanks to the FIFRELIN code, we were able to deduce the dependence of the spin on the kinetic energy. Similar trends were obtained for both reactions, i.e. a clear decrease of $\langle J \rangle$ with increasing $KE$ (see Fig. 3). In the case of $^{132}$Sn fission, this behavior suggests that the spin is generated mainly from the available intrinsic excitation energy at scission. Consequently and according to FIFRELIN calculations, a strong increase of $\langle J \rangle$ with increasing $E_n$ is expected, that could be interesting to check experimentally.

4 Conclusion

To further constrain the angular momentum generation models, isomeric ratio of the doubly magic $^{132}$Sn fission product was measured as a function of its kinetic energy for both $^{235}$U(n,f) [4] and $^{241}$Pu(n,f) [5, 6] reactions. By reproducing these experimental data thanks to the FIFRELIN code, we were able to deduce the dependence of the spin on the kinetic energy. Similar trends were obtained for both reactions, i.e. a clear decrease of $\langle J \rangle$ with increasing $KE$ (see Fig. 3). In the case of $^{132}$Sn fission, this behavior suggests that the spin is generated mainly from the available intrinsic excitation energy at scission. Consequently and according to FIFRELIN calculations, a strong increase of $\langle J \rangle$ with increasing $E_n$ is expected, that could be interesting to check experimentally.

References

[1] https://indico.in2p3.fr/event/26459/