Extraction of angular momenta from isomeric yield ratios

Employing TALYS to de-excite primary fission fragments

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Abstract. The generation of angular momentum in fission is difficult to model, in particular at higher excitation energies where data are scarce. Isomeric yield ratios (IYR) play an important role in deducing angular momentum properties of fission fragments (FF), albeit this requires some assumptions and simplifications. To estimate FF angular momentum, fission codes can be used to calculate IYRs and compare them to experimental data. Such measurements have systematically been performed at the IGISOL facility using novel experimental techniques. In conjunction, a new method has been developed to infer the angular momentum of the primary FF using the nuclear reaction code TALYS. In this work, we evaluate this new method by comparing our TALYS calculations with values found in the literature and with results from the GEF fission code, for a few well-studied reactions. The overall results show a consistent performance of TALYS and GEF, as well as of many reported literature values. However, some deviations were found, possibly pinpointing the need to re-examine some of the reported literature values. A sensitivity analysis was also performed, in which the role of excitation energy, neutron emission, discrete level structure and level density models were studied. Finally, the role of multiple chance fission, of relevance for the reactions studied at IGISOL, is discussed. Some literature data for this reaction were also re-analyzed using TALYS, revealing significant differences.

1 Introduction

As in all processes, conservation of angular momentum, $\mathbf{J}_{\text{tot}}$, has to be obeyed in fission. The total angular momentum of the compound nucleus is shared between the two fragments and their relative motion

$$\mathbf{J}_{\text{tot}} = \mathbf{J}_1 + \mathbf{J}_2 + \mathbf{L}, \quad (1)$$

where $\mathbf{J}_{1,2}$ are the respective fragment angular momenta and $\mathbf{L}$ is the orbital angular momentum of the relative fragment motion [1].

Thus, even for a compound nucleus with zero angular momentum, as in the spontaneous fission of ^{252}Cf , the fission fragments (FFs) can carry considerable amounts of angular momenta, something which has proven difficult to account for in contemporary fission modeling [2]. Many questions concerning the generation of $\mathbf{J}_{1,2}$ are still open. For instance there are conflicting experimental results concerning a possible saw-tooth trend in the average $J(A)$ [3]. Another important question is how angular momentum is shared among individual and collective degrees of freedom and how that eventually depends on the compound nucleus and excitation energy.

Experimentally, different techniques can be used to derive initial angular momentum populations in fission fragments. The most commonly used are prompt-fission γ -ray multiplicity studies [4,5] and measurements of isomeric yield ratios (IYR) [6,7]. At the IGISOL facility in Finland, a novel technique for measurements of IYR in fission, based on direct ion counting, has been developed. Systematic studies of the proton-induced fission of ^{235}U , at incident particle energy of 25 MeV, have been initiated [7,8]. From the measured values of IYRs a method has been proposed, which uses the TALYS reaction code to infer the initial fragments spin distributions [7]. The method has been successful in extracting angular momenta in good agreement with other literature works. However, due to the high entrance energy, the reaction is further complicated by a high probability for multi-chance fission and a high entrance angular momentum. In comparison to cold reactions (*e.g.*, spontaneous and thermal neutron induced), rather large values of J_{rms} are extracted from these measurements and sometimes with large fluctuations between different nuclei [7,9]. Therefore, we focus here on the methodology itself and compare it with other calculations. The main goal is to systematically assess the overall performance of the method and investigate its advantages

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Fig. 1. Simplified depiction of the de-excitation of fission fragments. The contour plot shows the distribution of excitation energy and spin of the fragments for ^{136}Xe in $^{235}\text{U}(n_{\text{th}}, f)$. In a first step most of the energy is exhausted through the evaporation of prompt fission neutrons (until reaching the neutron separation energy, S_n) followed by prompt γ -rays. Discrete γ -rays remove the remaining energy along with most of the angular momentum. The positions of the GS and isomer are only indicative.

and limitations. We discuss the procedure of deducing the spin distribution root-mean-square (rms) from IYRs, utilizing the TALYS reaction code. TALYS is chosen as it has a sophisticated evaporation model and because it allows the user to vary the nuclear spin distribution. It is also very well documented and widely used. The cases chosen for this validation work are listed in table 1 and originate from the cold fissioning systems $^{235}\text{U}(n_{\text{th}}, f)$ and $^{252}\text{Cf}(sf)$ where data are readily available. Finally, we briefly discuss the cases relevant for the IGISOL measurements, where multi-chance fission is energetically possible. We also use our method to re-analyze older IYR data.

2 Calculations

2.1 Fragment de-excitation

When the neck ruptures at scission, the nuclear shape changes and the fragments gain some relaxation energy [10]. Due to the Coulomb repulsion the fragments quickly acquire their terminal velocity, and most of the de-excitation process occurs from fully-accelerated fragments. The resulting fission fragments are neutron-rich, excited, deformed and rotating nuclei. Figure 1 depicts a simplified de-excitation scheme. The contour plot shows the event-by-event population of excitation energies and spins right after scission (pre-neutron emission) obtained from GEF (V.2017-1.2) [11]. The fragment's de-excitation starts with statistical neutron emission, followed by, and in competition with, prompt fission γ -ray emission (mainly from dipole transitions). During these processes it is fair to assume that the change in spin is minimal (on average). Once lower excitation energies are reached, near the yrast band, γ -rays are emitted from well-defined discrete states (primarily quadrupole polarity). Most of the possessed spin is removed in these latter steps (*e.g.*, through rotational or vibrational band cascades).

TALYS uses the statistical Hauser-Feshbach evaporation model which takes into account the competition between neutrons and γ -rays, in addition to the RIPL-3 data-base for discrete levels. In the GEF code, the de-excitation model is simpler. An explicit neutron/ γ -ray competition is included above the neutron separation. Statistical neutrons are emitted based on the Weisskopf's evaporation theory, and γ -rays are emitted until reaching the yrast line or a long-lived isomeric state. Further down in the excitation energy, discrete γ -rays are emitted until reaching the ground state.

2.2 Angular momentum

Fission fragments can possess many different modes of rotation, *e.g.* bending, twisting, tilting and rigid rotation around the centre of mass [1, 12]. Early on it was assumed that the angular momentum distribution of the fragments is the same as the one describing the nuclear level density [13, 14],

$$P(J) \propto (2J + 1) \exp \left(-\frac{(J + 0.5)^2}{2b^2} \right), \quad (2)$$

which is a Rayleigh distribution shifted by one half unit. The scale parameter b^2 , sometimes referred to as the “spin cut-off” parameter, defines the mean and width of the distribution. Often, the denominator of the exponent in eq. (2) is replaced by $B^2 = 2b^2$. Figure 2(a) shows the distribution for different values of B . In many studies of fission angular momentum distributions, the value derived for the parameter B is reported as the root-mean-square angular momentum, $J_{\text{rms}} = \sqrt{\langle J^2 \rangle}$. However, this assumption is only valid for a non-shifted Rayleigh distribution. For the distribution in eq. (2) it can be shown that

$$J_{\text{rms}}^2 = 2b^2 - \sqrt{0.5\pi}b + 0.25. \quad (3)$$

By reporting B as the root-mean-square angular momentum, values of J_{rms} found in the literature are typical overestimated by $0.4\hbar$. To be able to compare our results with the values found in the literature, and to avoid any ambiguity, we choose to report values for B , rather than the true J_{rms} .

2.3 Methodology

This study follows the methodology of ref. [7], in which the de-excitation process of the fission fragments is simulated using TALYS 1.9 [15]. Since TALYS is a deterministic code, the calculations have to be performed separately for each parent nuclei that feeds a certain isomer. However, in this study we have chosen to limit the calculations to cases of one and two neutrons emitted, being the values closest to the average $\nu(A)$. As will be discussed later, the sensitivity analysis suggests that this choice, because of the small amount of angular momentum carried away by the evaporated neutrons, has a negligible impact on the results.

Fig. 2. (a) The distribution of angular momentum as generated from eq. (2) for different values of $B = \sqrt{2}b$. (b) The population of spin *versus* excitation energy which was provided to TALYS. It was produced based on the assumed energy sharing (eq. (4)) and by varying B of the spin distribution. This particular matrix shows the case of Xe-136 for $B = 12\hbar$.

For a given fission product (Z, A) the de-excitation of the primary fragments, $(Z, A + 1)$ and $(Z, A + 2)$, are calculated assuming a normal distribution of the excitation energy while the angular momentum is assumed to follow the Rayleigh distribution of eq. (2). Figure 2(b) displays the matrix of angular momentum versus excitation energy provided to TALYS for the case of ^{136}Xe . The mean excitation energy was estimated using the average excitation energy of the corresponding mass chain, $\bar{E}(A)$, and the spread is obtained from the average excitation-energy spread of the respective fission reaction.

From the TALYS calculations, IYRs can be extracted and compared to experimentally determined values. By repeating the calculations for different values of the parameter $B \approx J_{\text{rms}}$, of the Rayleigh distribution, the value which best reproduces the data can be determined, resulting in a unique set of parameters $(A, Z, \nu, E$ and $B)$ for the initial fragments.

2.4 Input from GEF

In this study, the average excitation energy of the fragments is derived, but not explicitly taken, from the GEF code [11]. The Total excitation Energy (TXE) of the fragment pair is given by the Q -value of the reaction and the Total Kinetic Energy (TKE), according to $\text{TXE} = Q - \text{TKE}$. The Q -value is defined here as: $Q = M_{\text{Target}} -$

Fig. 3. (a) The $\bar{\nu}(A)$ (blue squares) and $\bar{\nu}_{\text{tot}}(A)$ (black empty circles, dashed line) from GEF for $^{235}\text{U}(n_{\text{th}}, f)$. The ratio between the two is also plotted (red full circles, full line). (b) The $\overline{\text{TXE}}(A)$ and $\bar{E}(A)$ from GEF for $^{235}\text{U}(n_{\text{th}}, f)$. The sharing of $\overline{\text{TXE}}(A)$ based on the ratio of $\bar{\nu}$ (eq. (4)) is plotted (red squares).

$M_1 - M_2 + M_n + E_n$. The average total energy, $\overline{\text{TXE}}(A)$ and average neutron emission, $\bar{\nu}(A)$, were obtained from GEF. The sharing of the TXE between the fragments is far from straight forward. The TXE consists of intrinsic excitation (single-particle like), deformation excitation and collective excitation. The fission codes have different treatments of the energy sharing. Therefore and for simplicity, we choose to estimate the mean fragment excitation energy, $\bar{E}_{1,2}$, by the mean excitation energy of the mass chain obtained from the ratio of the average neutron-emission, $\bar{\nu}$:

$$\bar{E}_{1,2}(A) = \overline{\text{TXE}}(A) \times \frac{\bar{\nu}_{1,2}(A)}{\bar{\nu}_1(A) + \bar{\nu}_2(A)}. \quad (4)$$

The justification of this simple assumption relies on the fact that the number of emitted neutrons is directly proportional to the available excitation energy. Moreover, $\bar{\nu}(A)$ is an experimental observable, in contrast to $\bar{E}(A)$. It is relatively well-reproduced in many fission codes, because it is the first observable to constrain the fragment excitation energy [16].

The total neutron emission for every fragment pair is plotted in fig. 3(a) along with the single fragment average neutron emission $\bar{\nu}(A)$. The ratio of the two is plotted and is used to estimate the portion of energy distributed for each fragment. The average TXE is plotted in fig. 3(b) along with the single fragment energy, both from GEF and based on eq. (4). In addition to the mean excitation energy, a spread of the energy distribution had to be assumed. At first and for simplicity, the same spread was used for all fragments, based on average GEF

Table 1. Isomeric yield ratio data from the reactions $^{235}\text{U}(\text{n}_{\text{th}}, \text{f})$ and $^{252}\text{Cf}(\text{sf})$, chosen for this comparison work. The spin values in parenthesis are updated values compared with the old tabulated ones used in the respective reference. The high-spin cross section ratio, σ_{HS} , is taken relative to the total ($\sigma_{IS} + \sigma_{GS}$). The excitation energy $\bar{E}$ is tabulated for the primary fragments, $A+n$ and $A+2n$, respectively.

Reaction	REF	Isotope	J (GS) ($\hbar$)	J (IS) ($\hbar$)	σ_{HS} (err)	J_{rms} ($\hbar$)	GEF IYR	$\bar{E}$ ($A+n/A+2n$) (MeV)
$^{235}\text{U}(\text{n}, \text{f})$	[20]	Br-84	2	6	0.38 (0.08)	5.9	0.10	6.3/7.5
	[18]	Rb-90	0	3	0.897 (0.1)	5.74	0.59	9.7/9.8
	[22]	Sb-130	4 (8)	8 (4)	0.398 (0.09)	7.5	0.43	3.8/4.2
	[22]	Sb-132	8 (4)	4 (8)	0.323 (0.08)	6.9	0.31	4.6/5.6
	[18]	Te-131	3/2	11/2	0.66 (0.03)	6.95	0.58	4.2/4.6
	[18]	Te-133	3/2	11/2	0.57 (0.03)	5.8	0.57	5.6/7.0
	[22]	I-134	4	8	0.583 (0.12)	9.5	0.33	7/5.6
	[22]	I-136	2 (1)	6	0.565 (0.09)	6.6	0.53	9.1/9.8
	[18]	Xe-133	3/2	11/2	0.74 (0.03)	8.3	0.42	5.6/7
	[18]	Xe-135	3/2	11/2	0.64 (0.03)	6.75	0.58	8.7/9.1
	[20]	La-146	2	6	0.055 (0.02)	–	0.59	12.1/12.6
$^{252}\text{Cf}(\text{sf})$	[6]	Sb-128	8	5	0.534 (0.076)	10.5	0.37	5.8/6.1
	[6]	Sb-130	8	4	0.456 (0.066)	8.9	0.5	6.0/6.2
	[6]	Sb-132	4	8	0.374 (0.037)	7.5	0.37	6.8/7.9
	[17]	Te-131	3/2	11/2	0.641 (0.078)	5.3	0.641	6.2/6.8
	[17]	Te-133	3/2	11/2	0.546 (0.071)	4.4	0.62	7.9/9.1
	[19]	I-132	4	8	0.565 (0.072)	10.2	0.36	6.8/7.9
	[17]	I-134	4	8	0.549 (0.066)	9.9	0.38	9.1/10.5
	[6]	I-136	1	6	0.755 (0.055)	10.0	0.56	11.2/11.8
	[6]	Xe-135	3/2	11/2	0.651 (0.059)	5.5	0.64	10.5/11.2
	[17]	Cs-138	3	6	0.582 (0.068)	7.9	0.56	12.5/13.25

values ($\sigma_E \approx 6$ MeV for $^{235}\text{U}(\text{n}_{\text{th}}, \text{f})$ and $\sigma_E \approx 8$ MeV for $^{252}\text{Cf}(\text{sf})$). In a later stage (see sect. 3.2), the average energy and spread were systematically varied to assess their relative importance.

3 Results

Table 1 lists the literature data of IYR used in this comparison. For every isomeric ratio, two TALYS calculations were performed; accounting for 1 and 2 neutrons emitted from the fission fragment. Figures 4–6 show examples of derived IYR as a function of B ($\approx J_{\text{rms}}$) for a selection of cases. All plots show the experimentally determined IYRs in horizontal solid green lines together with experimental uncertainty bands. Solid green vertical lines, labeled “Ref”, mark the values of B as reported in the respective references. The horizontal dashed black lines show the isomeric yield ratios from GEF (for one neutron emitted) and dashed black vertical lines, labeled “GEF”, correspond to the values of B obtained from GEF. The pink shaded areas mark the span of possible B values for which the TALYS calculations match the experimental isomeric yield ratio.

The thick red vertical line, labeled “TALYS”, shows the most probable B , taken as the average between the two calculations (for one and two neutrons emitted).

Table 2 lists all B values obtained for the different IYRs. The values listed are those found in the literature (labeled as “Ref.”) and the ones obtained with TALYS from the literature values of the IYR (labeled as “Ref.+TALYS”). In addition, we list the predicted B values from the GEF code (labeled as “GEF”) and the corresponding TALYS calculation using the predicted IYR from GEF (labeled as “GEF+TALYS”). Missing values indicate that TALYS was unable to match the IYR.

3.1 Default TALYS results

The cases shown in fig. 4 are two examples of excellent agreement between all three models. In plot (a), the IYR of ^{138}Cs in $^{252}\text{Cf}(\text{sf})$ from Datta *et al.* [17] is presented. The extracted values of B from all three methods are between $7\hbar$ and $8\hbar$. In (b), a satisfying agreement is also found for ^{132}Sb , produced in $^{252}\text{Cf}(\text{sf})$ [6], yielding values around $8\hbar$.

Fig. 4. Two examples where a perfect match between TALYS, GEF and literature [17,6] could be achieved. The vertical lines show B as extracted from GEF (dashed black line), TALYS (full thick red line) and the experiment (full green line). (a) The calculations for Cs-138 assuming 1 (red circles) or 2 (blue squares) neutrons emitted. All three data-sets agree in this case pointing to a $J_{\text{rms}} \approx 7-8\hbar$. (b) The calculations for Sb-132 which give a $J_{\text{rms}} \approx 7-8\hbar$.

Fig. 5. Two examples where both GEF and TALYS disagree with literature data [6]. The vertical lines show B as extracted from GEF (dashed black line), TALYS (full thick red line) and the experiment (full green line). (a) The calculations for Sb-130 assuming 1 (red circles) or 2 (blue squares) neutron emissions separately. (b) The calculations for Xe-135.

Fig. 6. The vertical lines show B , as extracted from GEF (dashed black line) and the experiment (full green line). (a) The calculations for Rb-90 assuming 1 (red circles) or 2 (blue squares) neutron emissions separately. TALYS fails to meet the high IYR measured reported in ref. [18]. GEF agrees perfectly with the TALYS results if the IYR predicted by GEF is taken as reference. (b) The calculation for Xe-133 where TALYS agrees with the reported B value of ref. [18]. GEF predicts a lower IYR and is in perfect agreement with TALYS (regarding B) if the IYR of GEF is taken as reference.

Figure 5 on the other hand, shows two examples where the literature values deviate from the ones obtained with GEF and TALYS, outside the margins of the error bars. In plot (a), the value of B for ^{130}Sb in $^{252}\text{Cf}(\text{sf})$ derived from both codes is $7\hbar$, whereas the literature value is around $9\hbar$ [6]. In plot (b), the value of B for ^{135}Xe is $7\hbar$ according to GEF and TALYS but the literature value is $5.5\hbar$ [6]. Another example is ^{132}I in $^{252}\text{Cf}(\text{sf})$, for which TALYS

agrees with the value reported in the literature of around $10-11\hbar$ [19]. However, this is about $3\hbar$ higher than the neighbouring isotope ^{132}Sb and GEF predicts around $7\hbar$ for both nuclei. Moreover, TALYS can match the IYR as estimated from GEF for a value of $B = 7\hbar$. A third example is ^{146}La where the authors report a very small IYR [20]. As a consequence, they were not able to extract any value for B . TALYS, however, was able to deduce a

Table 2. The deduced $B \approx J_{\text{rms}}$ from TALYS for both the measured IYR (Ref.) and the predictions of GEF, respectively.

Reaction	REF	Isotope	B [Ref.]	B [Ref.+TALYS]	B [GEF]	B [GEF+TALYS]
$^{235}\text{U}(\text{n}_{\text{th}}, \text{f})$	[20]	Br-84	5.9	4.5	3.2	2.4
	[18]	Rb-90	5.7	–	3.5	3.4
	[22]	Sb-130	7.5	6.1	6.2	6.2
	[22]	Sb-132	6.9	7.7	6.7	7.1
	[18]	Te-131	7.0	7.4	5.8	6.6
	[18]	Te-133	5.8	6.3	6.3	6.6
	[22]	I-134	9.5	12.5	6.6	6.9
	[22]	I-136	6.6	–	6.9	–
	[18]	Xe-133	8.3	9.0	5.1	4.8
	[18]	Xe-135	6.8	7.6	6.2	6.9
	[20]	La-146	–	1.8	6.7	10.3
$^{252}\text{Cf}(\text{sf})$	[6]	Sb-128	10.5	6.0	6.0	–
	[6]	Sb-130	8.9	6.5	6.8	6.8
	[6]	Sb-132	7.5	8.1	7.4	7.6
	[17]	Te-131	5.3	6.4	6.2	6.9
	[17]	Te-133	4.4	5.7	6.9	7.0
	[19]	I-132	10.2	11	6.7	6.9
	[17]	I-134	9.9	10.6	7.3	7.4
	[18]	I-136	10	–	7.3	–
	[18]	Xe-135	5.5	7.3	6.9	7.6
	[17]	Cs-138	7.9	7.2	7.5	6.5

spin of about $1\text{--}2\hbar$. GEF, finally, predicts a rather large IYR and consequently an angular momentum around $7\hbar$. These cases and other similar discrepant ones, highlight a possible need for reexamination of some of the reported B values. In fact, B values found in literature are often uncertain and discrepant. Very often, the uncertainties in experimental IYR set a rather wide span of possible J_{rms} values which may extent to several $\hbar$.

Figure 6 presents the results for ^{90}Rb (a) and ^{133}Xe (b), both measured in $^{235}\text{U}(\text{n}_{\text{th}}, \text{f})$ [18]. For ^{90}Rb , TALYS fails to derive any B value from the literature IYR. Furthermore, the IYR predicted in GEF differs considerably from the experimental values. As a consequence, the extracted values for B are rather different. Note that GEF shows good agreement for other IYR for the same reaction, see fig. 69 of ref. [11]. Due to the limited and uncertain existing IYR data, it is sometimes hard to extract accurate information on fragment angular momentum from IYRs. This increases the demand for high-quality, systematic IYR data such as the ones measured at IGISOL [21].

However, comparing TALYS with GEF reveals a consistent performance and overall agreement between the two; if the predicted IYR from GEF is used, TALYS reproduces the B value from GEF. This behaviour was observed in most studied cases. Note that we only selected to plot the GEF B values corresponding to one emitted neutron, since the results were close to those of two emitted neutron.

3.2 Sensitivity analysis

To assess the sensitivity of the calculations, comprehensive tests have been performed. The calculations explore variations due to the physics (*e.g.* excitation energy or number of emitted neutrons), the models (by using different level density models) and structure data (by omitting parts of poorly known level schemes).

3.2.1 Effects of level schemes

In a few cases, TALYS was unable to match the measured IYR regardless of B . Figure 7(a) shows the example of ^{136}I for which GEF and the literature data are in agreement [22], while TALYS fails to match the measured IYR for any B . A possible reason for this is a poorly known level scheme. For the calculations TALYS includes by default 10 discrete levels (N) for the residual nucleus, and bridges the gaps to high energies by the aid of level density models. According to the RIPL-3 data-base [23], the known level scheme for ^{136}I is only complete up to the first level and many discrete levels are missing (for example between 0.6 MeV and 2.6 MeV). Therefore, our calculations might miss important transitions leading to the population of either isomeric state. To test this hypothesis, the calculations were repeated with different number of discrete levels (N) included. By decreasing N , the calculations will instead rely more on nuclear level density

Fig. 7. (a) The TALYS calculations for ^{136}I with different number of considered nuclear levels (both for one and two emitted neutrons). By reducing the use of experimentally known nuclear levels (N), a higher IYR can be reached [22]. (b) The calculations for ^{90}Rb with variable number of considered nuclear levels [18].

models. The minimum assigned value ($N = 6$) ensures the inclusion of the isomeric state whereas $N = 40$ represents a relatively large value. As observed in fig. 7(a), a lower number of discrete levels results in an increase of the population of the high spin state, although an agreement could still not be reached. This example shows that extra care has to be taken when considering exotic nuclei where the level scheme might be poorly known. For cases where important level structure is missing, the obtained B values carry much larger uncertainties.

Another example is shown in fig. 7(b) for ^{90}Rb . According to RIPL-3, the completely known part of the level scheme of ^{90}Rb contains 14 levels. TALYS fails to reach the reported IYR but agrees rather well with GEF (for $10 < N < 14$) if the GEF-predicted IYR is taken as reference. By only considering 3 levels, TALYS could match the very high IYR reported in the literature. The agreement between TALYS and GEF could potentially indicate an overestimated IYR in ref. [18].

In an earlier work, it has been noticed that TALYS could match the IYR for Cd isotopes only when all the experimentally known levels were included in the calculations. When different combinations for reduced number of levels were tried, the IYR as estimated by TALYS could not come to an agreement with the experimental result [9].

potential. Using the other models showed that the calculations were insensitive to which level density model was used (see the example of ^{132}Sb in 8(a)).

3.2.3 Variations of the number of emitted neutrons

Due to the deterministic nature of TALYS, the contribution from different primary fission fragments to a specific isomeric state in the fission product has to be calculated separately. For simplicity, we have assumed the emission of one and two neutrons only (the average neutron emission is mostly between those two values). Neutrons are normally assumed not to carry much angular momentum. To check for possible systematic errors due to this assumption, we have run TALYS with zero to three emitted neutrons from the fission fragments for a few cases. Figure 8(a) shows that the neutron emission is less important within this de-excitation framework, and that the obtained IYR is rather insensitive. This was also confirmed with GEF where the J_{rms} was rather similar for all fission fragments leading to a certain fission product (resulting from different emitted neutrons). As a conclusion, the combined effect of both varying the level density model and the number of emitted neutrons, introduce an uncertainty of $1\hbar$ in the extracted B value.

3.2.2 Variations of nuclear level density models

TALYS provides several choices of nuclear level density models. A thorough comparison of these level density models is beyond the scope of this work. However, to investigate the sensitivity of our calculations on this choice, three models have been used:

- 1) LD1: Constant Temperature (CT) + Fermi Gas Model (FGM)
- 2) LD2: Back-Shifted Fermi Gas Model (BSFGM)
- 3) LD4: Microscopic-Goriely model

The microscopic-Goriely model (LD4) was used as the default model in this study. Although, by defining our own distribution, we neglect the energy-dependence of the spin-cut-off parameter, thus not using the model to its full

3.2.4 Excitation energy dependence

The fission fragment excitation energy and spin govern the probability of neutron and γ -ray emission, and hence the isomeric yields ratios. For simplicity, we assign a Gaussian energy distribution. To investigate the IYR dependency on the excitation energy; both the mean energy, $\bar{E}$ and the spread σ_E , were systematically varied.

As mentioned earlier, σ_E was assumed to be 6 MeV for $^{235}\text{U}(n_{\text{th}}, f)$ and 8 MeV for $^{252}\text{Cf}(sf)$, based on the average fragment values taken from GEF. By varying σ_E between 0.5 and 10 MeV for the case of ^{131}Te , the impact was shown to be small as can be seen in fig. 8(b). The variation of B over the range of values for σ_E tested, is in the order of $1\hbar$. In future studies, σ_E should be considered as a function of mass and kinetic energy to possibly improve the accuracy of the initial energy assumptions.

Fig. 8. (a) The TALYS calculations for ^{132}Sb with different nuclear level density models and different assumptions on the number of emitted neutrons [22]. (b) Variation of the excitation energy spread for ^{131}Te . The literature data are from Refs. [18].

Fig. 9. (a) Variation of the average excitation energy for ^{131}Te [18] assuming an energy spread of 6 MeV. (b) Variation of the average excitation energy for ^{131}Te assuming a energy spread of 0.5 MeV.

The average excitation energy ($\bar{E}$) was varied between 2 MeV and 12 MeV for several nuclei. Figure 9(a) shows again the example of ^{131}Te , where the average energy has a small effect, because of the spread of $\sigma_E = 6$ MeV, the impact of the average energy is minimal. If we assume a small energy spread instead, this introduces a large variation in the calculated IYR and a larger dependency on the assumed level of excitation energy. This is demonstrated in fig. 9(b) where σ_E was set to 0.5 MeV.

3.3 Multi-chance fission

This work focuses on spontaneous and thermal neutron-induced fission, however it is worth emphasizing the role of multi-chance fission in the observed angular momenta from the measurements at IGISOL. At higher excitation energies, pre-fission neutrons are emitted which may affect the population of angular momentum in the primary fission fragments. Figure 10(a) shows a GEF calculation for the proton-induced fission ($E_p = 25$ MeV) of ^{238}U . The J_{rms} is plotted as a function of A , for the different multi-chance fission channels. According to GEF the fractions of multi-chance fission are 24% for first, 36% for second, 30% for third and 10% for fourth chance fission, respectively. As can be seen, the value of J depends significantly on the fission channel, according to GEF. This is observed more clearly in terms of the population of excitation energy *versus* spin, plotted for all heavy fragments in fig. 10(b), (c). The J_{rms} is about $3.5\hbar$ lower in fourth chance fission compared to first chance fission, which will influence the population of the isomers. According to GEF, the available

excitation energy decreases considerably as neutrons are emitted from the compound nucleus. Due to the strong correlation between excitation energy and spin, angular momentum is also reduced for higher fission chances. More generally, the scission shapes could change for different compound nuclei by unequal contributions of the different fission modes; the asymmetric Standard-1 and Standard-2 and the symmetric Super long [24]. This could affect the quadrupole moments and the moments of inertia of fragments. In some earlier works it has been estimated that multiple chance fission does not affect J_{rms} significantly (about $0.5\hbar$ difference between first and third chance fission) [25]. This seems to be a simplification which could contradict predictions of recent fission modelling.

3.4 Re-analysis of earlier literature data

In their paper from 1993, Tanikawa *et al.* report a measurement of 30 IYRs in 24 MeV proton induced fission of ^{238}U [25] using γ -ray spectroscopy. From these they derive the angular momentum distribution using a method based on the statistical model [14]. The angular momentum is reported as values of the parameter B . To compare their method to the one developed in this work five of their values were re-analyzed using TALYS [21]. The excitation energy was estimated based on the average neutron emission from ref. [26]. A single energy value was used without energy spread. The results are shown in fig. 11 where the B values from the original work are plotted together with the TALYS results. In all cases the TALYS calculations give larger angular momenta.

Fig. 10. (a) The distribution of angular momentum, as a function of fragment mass, simulated with GEF for the $^{238}\text{U}(\text{p}_{25}\text{MeV}, \text{f})$ reaction. The contribution of multi-chance fission is visualized in terms of average J_{rms} , plotted for compound nucleus mass of $A = 239, 238, 237$ and 236 amu. (b), (c): The excitation energy *versus* angular momentum, averaged for all heavy fragments and gated for first chance fission (b) and fourth chance fission (c).

Fig. 11. A comparison between B values reported in ref. [25] (black circles) and TALYS calculations assuming the same measured IYR (red squares).

Possible explanations for this discrepancy are: 1) differences in excitation energies of the primary fragments. 2) the lack of discrete level schemes in ref. [25]. 3) different treatment of multi-chance fission and the resulting available excitation energy. Tanikawa *et al.* assume the effect of multi-chance fission on the angular momentum to be negligible, in contrary to GEF which predicts a significant difference. We conclude that studies of angular momentum in high energy fission reactions should be carefully analyzed, including the effect of multi-chance fission.

4 Summary and conclusions

In this work we have assessed a method for deducing initial angular momentum in fission fragments from experimental data on IYR. This method is being used to extract root-mean-square of fission fragment angular momentum distributions from IYR measured at the IGISOL facility. TALYS has been used as the main nuclear de-excitation code, with a few input data from the GEF fission code. The present work has shown that the method is able to reproduce most literature values, calculated with different models, methods and approximations.

We especially note a very good agreement between GEF and TALYS. The predicted IYR from GEF, analyzed with TALYS yields similar B values (within $1\hbar$ margin) as GEF in most cases tested (*e.g.* for ^{84}Br , ^{90}Rb , $^{130,132}\text{Sb}$, $^{132,134}\text{I}$, $^{131,133}\text{Te}$, $^{133,135}\text{Xe}$, ^{138}Cs). The only situations where GEF and TALYS differ are ^{146}La and ^{136}I , among the considered samples. However, for lanthanum the uncertainty of the value of B is large due to weak dependence of the calculated IYR on B in this region. We therefore conclude that the methods in both codes are consistent.

In a few cases, the values of J_{rms} , respectively B , reported in the literature deviate from both the GEF and TALYS calculations. This is seen in $^{128,130}\text{Sb}$, ^{135}Xe , ^{133}Te , ^{90}Rb and ^{84}Br . In the case of $^{252}\text{Cf}(\text{sf})$, many literature values were as high as $10\hbar$, which is large compared to theoretical estimations of average J_{rms} of fragments in cold fission reactions.

A sensitivity analysis revealed that the choice of level density models is of less importance, at least when TALYS is fed with a user-defined energy *versus* spin matrix. When different number of neutrons are emitted, the calculated IYRs show very little fluctuation which agrees with common assumptions on how angular momentum is removed from the de-exciting nucleus.

A good estimation of the excitation energy in the fragment is generally important to obtain reliable results. However, if the energy is somewhat realistic the IYR results seem not very sensitive to the initial $\bar{E}$. If the energy spread (σ_E) is sufficiently large, the choice of average energy is less crucial. By assuming a minimal spread, $\sigma_E = 0.5$ MeV, the calculations were more sensitive to the initial mean energy assumption. As a result, a realistic energy spread is advised in order to ensure robust IYR estimates. As a future improvement, a more elaborate energy distribution will be adopted and an energy-dependent spin distribution will be implemented.

The sensitivity analysis stresses the importance of accurate level structure data. For the two cases where TALYS failed in reproducing a measured IYR, reducing the number of considered discrete levels (and thus relying more on level density models) resulted in better agreement with literature data, and with GEF. In another example from ref. [9], the IYR values for Cd could only be matched when all known levels were included.

The inclusion of multi-chance fission complicates the picture since contributions of different multi-chance fission channels could imply different angular momentum populations. It is therefore important to consider multi-chance fission (when applicable) in the calculations and to weight the results accordingly. Earlier literature works seem to assume an independence of J_{rms} on the multi-chance probability, as opposed to the GEF code. This could be attributed to a better understanding of the fission dynamics and the changes in scission shapes in contemporary fission modelling.

Finally, we have used TALYS to re-analyze some of the IYR data from Tanikawa *et al.* [25] measured for the $^{238}\text{U}(p_{25\text{ MeV}}, f)$ reaction. The TALYS calculations show larger values of B in almost all cases, contrary to what was obtained for the colder reactions in this study. This could be explained by the different treatment of multi-chance fission. Hence, in future studies of angular momentum in high energy fission reactions this effect should to be carefully considered.

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