EPJ Web of Conferences **146**, 04041 (2017) DOI: 10.1051/epjconf/201714604041
ND2016

Study of fission fragment de-excitation by gamma-ray spectrometry with the EXILL experiment

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Abstract. A large array of Ge detectors installed at ILL, around a ^{235}U target irradiated with cold neutrons, (EXILL) allowed measurement of prompt gamma-ray cascades occurring in fission fragments with an unambiguous determination of fragments. Here we present preliminary results of a systematic comparison between experimental γ-ray intensities and those obtained from the Monte-Carlo simulation code FIFRELIN, which is dedicated to the de-excitation of fission fragments. Major γ-ray intensities in the ^{142}Ba and ^{92}Kr fission products, extracted from EXILL data, were compared to FIFRELIN, as well as to reported values (when available) obtained with EUROGAM2 in the spontaneous fission of ^{248}Cm. The evolution of γ-ray intensities in ^{92}Kr versus the complementary partner in fission (i.e. versus the total number of evaporated neutrons by the fission pair) was then extracted and compared to FIFRELIN.

1. Introduction

Nuclear fission is a complex process, still not well described by microscopic models, and its observables are difficult to measure accurately. Following the capture of a thermal neutron, the compound nucleus fissions predominantly into two fragments. Most of the energy of the process is transferred to kinetic energy, whereas the nuclei are left with few tens of MeV deposited in excited states. This excitation energy is then released, the majority of it within less than some nanoseconds, by the evaporation of neutrons and the emission of γ rays in cascade. To overcome the low accuracy of microscopic models in the prediction of fission observables, nuclear technology relies on libraries of evaluated data and semi-empirical models, like the GEF model [1]. Such a strategy requests systematic and accurate experimental data on the few possible observables. Here we present the benefit of studying the de-excitation of fission fragments using an array of high-resolution γ-ray detectors placed around an actinide target irradiated by a thermal or cold neutron beam. Such a setup (EXILL) was installed at the Institut Laue-Langevin (ILL) in 2012 and campaigns were performed with ^{235}U and ^{241}Pu targets [2]. The γ-ray cascade is directly linked with the angular momentum of the fragments after scission, which is one of the least understood properties. With the development of new simulation codes for the neutron evaporation and γ-ray cascade like FIFRELIN [3], systematic studies and comparisons with the large amount of experimental data resulting from double and triple γ-ray coincidence analyses become possible. Preliminary results obtained with the ^{235}U target on the well-produced Kr and Ba fission products are presented and compared to FIFRELIN simulations (here, only binary fission is considered in the simulation process). This study is largely motivated by the future installation of a permanent γ-ray detector array at the ILL, which will be coupled to fission fragment detectors in a first construction phase and a fragment separator in a second phase (FIPPS) [4].

2. Experimental details

A large array of sixteen HPGe detectors was placed at the end of the PF1b cold neutron guide, which has the highest neutron flux at the ILL. It included eight EXOGAM clovers, two smaller clovers from LOHENGRIN and six large efficiency detectors from GASP. The acronym EXILL refers to EXOGAM@ILL. All detectors, except the two LOHENGRIN clovers, were surrounded by a BGO active shielding to reduce Compton background. Overall, the array had a total efficiency of 6.2% for γ rays of 1 MeV. The neutron beam was reduced by collimators to about 12 mm diameter spot at the target position. Its capture flux

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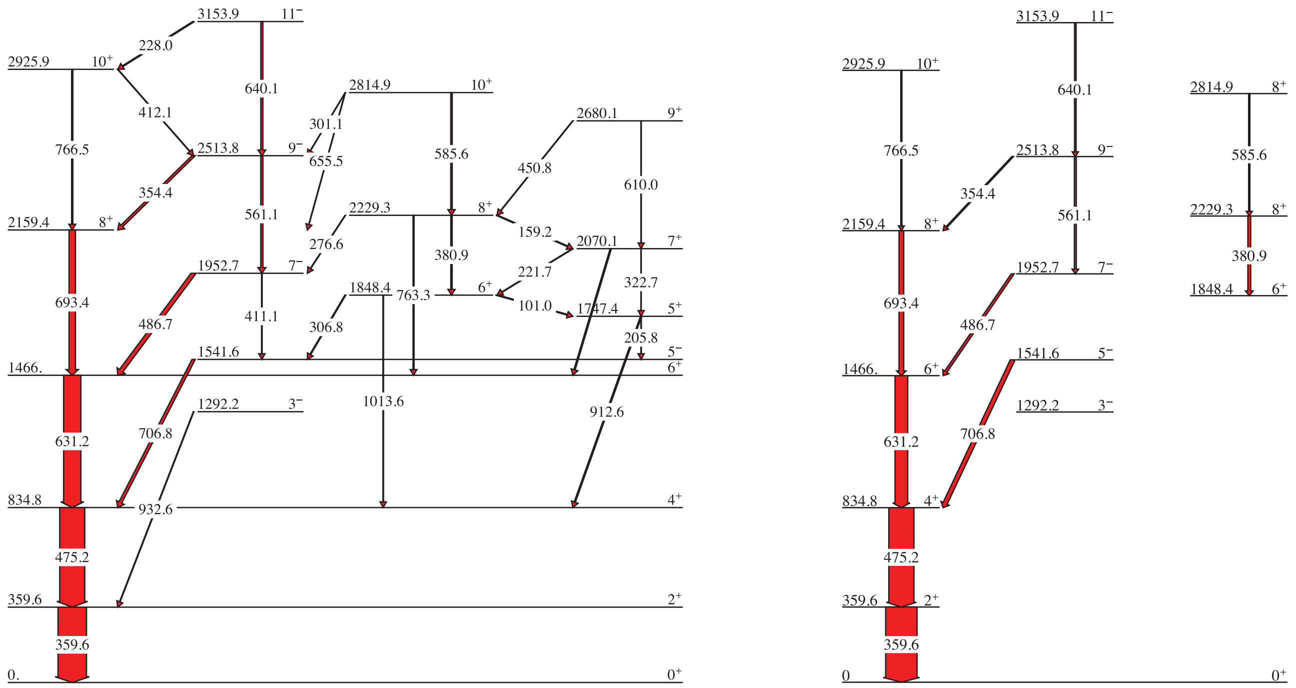

Figure 1. Partial level scheme and γ-ray cascade simulated with FIFRELIN in ^{142}Ba in the $^{235}U(n_{th}, f)$ reaction (left) and partial experimental cascade measured in the EXILL experiment (right).

there was around 10^8 n/s/cm^2. The fission rate in the targets was estimated to around 10^5 fissions/s. Data were collected in a triggerless mode using a 100 MHz digital acquisition system for over 20 days with a ^{235}U target and 15 days with a ^{241}Pu target. More details on the setup, the target assembly and the shielding can be found in [2,4].

3. Analysis

All the data were calibrated, run by run, to account for energy and time shifts in the whole set of 46 germanium crystals. Then a three-dimensional histogram with the energies of events in triple coincidence ($\gamma\gamma\gamma$ cube) was built in a similar way to the one described in [5]. In that process, coincident events occurring in the crystals of the same clover detector are summed to reduced Compton background (add-back process). A 200-ns coincidence time window was chosen. It is long enough to account for most of the cascades occurring in the fragments without adding too many accidental events. The final $\gamma\gamma\gamma$ cubes contain over 10^{11} events (^{235}U target) and 3×10^{10} events (^{241}Pu target). We concentrate our study on fission fragments with a well-known level scheme and a high production yield. The goal is to study transitions between discrete levels, occurring in the de-excitation of the fragments, after the neutron evaporation process. A first step was to compare these transition intensities to the ones predicted by the current version of FIFRELIN simulation code. Then we look for any systematic correlations between the cascade in a fragment and its fission partner. At this stage of work, there was no adjustment on FIFRELIN parameters to match our results. The analysis was performed on the $\gamma\gamma\gamma$ cube in a semi-automatic way using a home-made software developed on ROOT. To construct e.g. the cascade in ^{142}Ba, we searched for events in coincidence with a strong transition (at the energy E_1) in one of its fission partner (e.g. ^{92}Kr) and, in coincidence with a strong transition (at the energy E_2) in ^{142}Ba. Usually we selected transitions from the first excited state to the ground state. This operation is similar to the cut of the $\gamma\gamma\gamma$ cube with two gates centered respectively on E_1 and E_2, and to analyze the resulting γ-ray spectrum, i.e. to identify the peaks belonging to ^{142}Ba and to fit their area. Background, resulting mainly from Compton events, is taken into account by cutting the cube with additional gates at energies slightly above E_1 and E_2, and by removing their contribution to the peak area.

4. Preliminary results

^{142}Ba is a good study case. It is well produced in the thermal fission of ^{236}U (fission yield = 2.8%) and its level scheme is well known. According to RIPL-3 database [6], the level scheme is complete up to 1848.4 keV and 38 levels are known up to 5284 keV. In addition, the structure of this nucleus was studied by Urban and coworkers [7] in the spontaneous fission of ^{248}Cm. In that last case however, the exact gating conditions, and especially the fission partner used to build the level scheme, were not reported. Figure 1 shows the level scheme and γ-ray cascade simulated by FIFRELIN in ^{142}Ba, when this fission product (i.e. after neutron evaporation) is obtained in coincidence with ^{92}Kr (after neutron evaporation as well), compared to the experimental cascade observed within the EXILL data in the same conditions. The agreement is rather good. For clarity, we removed the weakest transitions (and associated levels) in the simulated cascade and we organized the level scheme in the same bands as in Fig. 4 in [7]. For the experimental cascade, we extracted the most intense γ rays and a selection of γ rays in side bands. Level energy, spin and parity as well as transition energies are taken from RIPL-3.

Table 1 details the obtained γ-ray intensities as well as the ones measured in the spontaneous fission of ^{248}Cm.

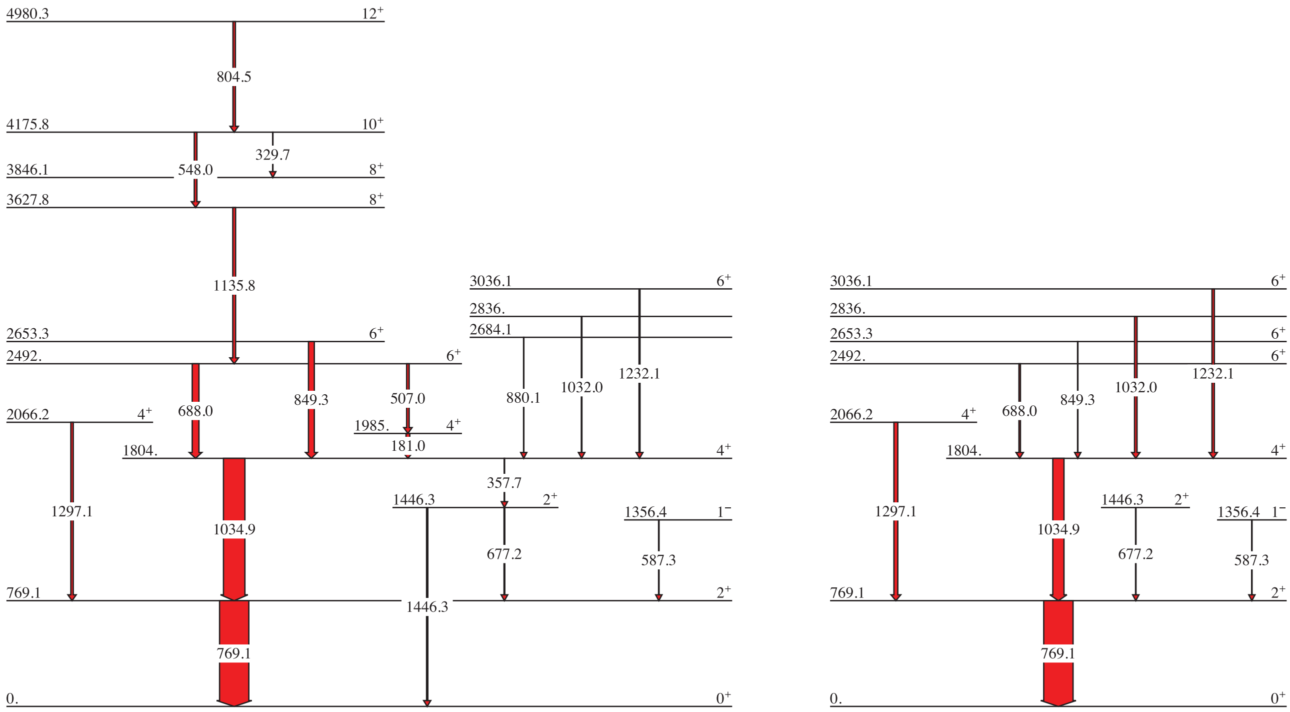

Figure 2. Partial level scheme and γ-ray cascade simulated with FIFRELIN in ^{92}Kr in the $^{235}U(n_{th}, f)$ reaction (left) and partial experimental cascade measured in the EXILL experiment (right).

Table 1. Gamma-ray intensities in ^{142}Ba measured in the spontaneous fission of ^{248}Cm [7], simulated by FIFRELIN in the thermal fission of ^{236}U and, extracted from the EXILL data. All values are normalized on the 359.5-keV intensity (set to 100).

E (keV)	J1→J2	I_γ ^{248}Cm	EXILL	FIFR.
359.6	$2^+ \to 0^+$	100 (5)	100 (3)	100.0 (1)
475.2	$4^+ \to 2^+$	85 (5)	80 (2)	91.4 (2)
631.2	$6^+ \to 4^+$	40 (3)	42 (2)	66.3 (1)
693.4	$8^+ \to 6^+$	13 (2)	12 (1)	26.0 (1)
766.5	$10^+ \to 8^+$	2.3 (3)	2.4 (7)	3.60 (3)
706.8	$5^- \to 4^+$	6.0 (6)	10 (1)	10.17 (5)
486.7	$7^- \to 6^+$	18 (2)	8 (1)	15.60 (6)
354.4	$9^- \to 8^+$	5.1 (4)	4.4 (5)	8.40 (4)
561.1	$9^- \to 7^-$	4.5 (4)	4.0 (5)	7.52 (4)
640.1	$11^- \to 9^-$	5 (1)	2 (1)	6.22 (3)
380.9	$8^+ \to 6^+$	6.5 (8)	4.0 (6)	5.06 (3)
585.6	$10^+ \to 8^+$	5.5 (5)	4 (1)	5.66 (3)

We observed that the intensities measured with EXILL are very close to the ones measured by [7], with the exception of E1 transitions linking the rotational negative parity band to the ground state positive parity band (transitions at 706.6 and 487 keV), which request more investigations. One notices that transition intensities from high spin states are systematically overestimated in FIFRELIN results, which may be related to an excessive value in the simulation inputs for the spin cut-off of fragments (before neutron evaporation) in this region.

The same study was done for ^{92}Kr. Figure 2 shows the cascade simulated with FIFRELIN and the observed one with EXILL data, both obtained in coincidence with ^{142}Ba. The match is not as good as in ^{142}Ba but the global picture at low spin is respected. Transitions from high spin states (at 688.0 and 849.3 keV) are visibly too intense in the simulation. This is somehow expected; the spin and parity of many high-energy levels are unknown. In a previous FIFRELIN simulation, an old version of RIPL3 was used as input: simulated results [8] were significantly worse, with e.g. an inversion of the 1034 keV and 1297 keV

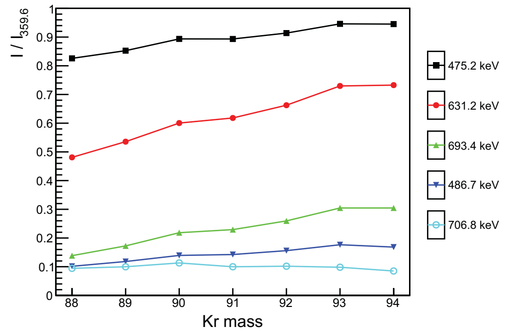

Figure 3. Evolution of main transition γ-ray intensities in ^{142}Ba versus possible Kr partners, according to FIFRELIN simulations. Intensities are normalized on the 359.6-keV intensity.

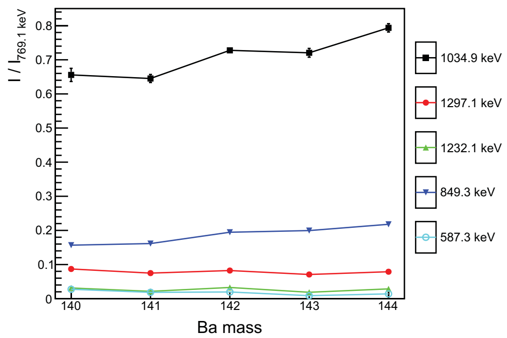

Figure 4. Evolution of main transition γ-ray intensities in ^{92}Kr versus possible Ba partners, according to FIFRELIN simulations. Intensities are normalized on the 769.1-keV intensity.

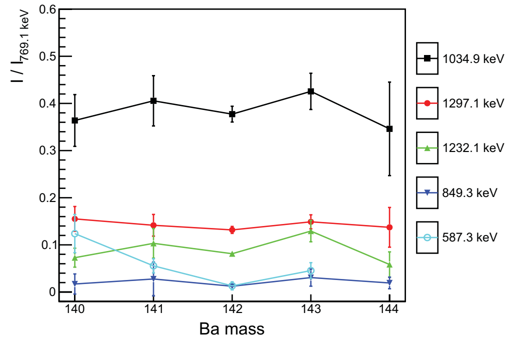

Figure 5. Evolution of main transition γ-ray intensities in ^{92}Kr versus possible Ba partners, according to EXILL data. Intensities are normalized on the 769.1-keV intensity.

intensities. The present FIFRELIN simulation uses the 2015 updated version of RIPL-3, which includes many new values or changes of level spin and parity for the ^{92}Kr. The partial experimental cascade obtained from EXILL data seems consistent with the results of Rzaca-Urban et al. (Fig. 4 in [9]) measured in the spontaneous fission of ^{248}Cm. However the values of the transition intensities were not reported in [9]).

The EXILL data will allow systematic studies of the cascades to be performed. One question we are studying is whether and how the deexcitation cascade in a fragment evolves when its fragment partner in fission changes. FIFRELIN simulations were done to test such systematic trends. Figure 3 shows the (simulated) intensity of main transitions in ^{142}Ba versus possible Kr partners. Here, in the simulation, all fission products are taken after neutron evaporation. One sees a clear decrease in the intensity from high spin states when the mass of Kr decreases, i.e. when more neutrons are evaporated. In FIFRELIN, emitted neutrons carry away a certain amount of angular momentum, depending on the optical model potential parametrization used for neutron transitions. Simulations in other fragments give similar results. Figure 4 shows the simulated intensity of main transitions in ^{92}Kr versus possible Ba partners. A decrease with neutron evaporation is visible for one of the $4^+ \rightarrow 2^+$ transition (at 1034.9 keV). We studied this evolution with EXILL data. The result is plotted in Fig. 5. There is no visible change in the transition intensities, even in the 1232.2-keV transition. However, the uncertainties are too large to conclude on this effect. The same trend will be studied in other nuclei in the near future.

5. Conclusion and perspectives

The preliminary results on ^{142}Ba and ^{92}Kr extracted from EXILL data were compared to FIFRELIN simulations without any adjustment of the input parameters (like the spin cut-off) to fit EXILL data. The general agreement between the experimental cascade and the simulated one in ^{142}Ba is however already very promising. EXILL data seems to be an excellent way to test the validity and limits of FIFRELIN models and nuclear level structure data. A systematic study of more fragments is planned in the near future as well as a comparison between the cascades produced in ^{235}U(n_{th},f) and ^{241}Pu(n_{th},f) reactions.

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