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EXOGAM at the ILL: the EXILL campaign

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Abstract. A combination of germanium detectors has been installed at the PF1B neutron guide of the ILL to perform the prompt spectroscopy of neutron-rich nuclei produced in the neutron-capture induced-fission of $^{235}\text{U}$ and $^{241}\text{Pu}$. Radiative capture reactions on rare targets have also been performed. LaBr$_3$ detectors from the FATIMA collaboration have also been installed in complement with the EXOGAM clovers to measure lifetimes of low-lying excited states. The measured characteristics indicate very good performances of the overall setup. Some recent results will be discussed.

1. Introduction

It is known since long that fission produces neutron-rich nuclei in a very effective way. One of the best mechanism is to induce fission of actinide targets using cold neutrons from a neutron guide. In this regime, the neutrons bring just enough energy into the system to produce fission, whilst preserving the neutron-richness of the fragments. The average spin generated in this reaction is around $6-7\hbar$ [1] and states with spins as high as $20\hbar$ can be observed. Several possible actinide targets have been envisaged and it came out that of particular interest in this context is the use of $^{235}\text{U}$ and $^{241}\text{Pu}$ fissile targets which give access to nuclei that are too weakly produced using spontaneous fission sources or another production mean to perform their prompt spectroscopy. In addition to the spectroscopy of nuclei produced in such a cold fission process, neutron capture reactions using rare targets have also been performed. In this contribution we will describe the installation of the EXOGAM array, usually standing at GANIL, at the Institut Laue Langevin (ILL), Grenoble (hence the name EXILL for EXOGAM at the ILL), and the experimental campaign which took place in 2012 and 2013.
Figure 1. Mass yield distributions measured for the spontaneous fission of $^{252}$Cf and $^{248}$Cm compared to the thermal neutron induced fission of $^{235}$U and $^{241}$Pu (data from [2]).

2. Physics case

The basic idea of EXILL was to install a large number of efficient germanium detectors around a fissile target irradiated by a well collimated cold neutron beam. Large arrays like Euroball or Gammasphere were already used to study fission fragments produced in the spontaneous fission of $^{252}$Cf and $^{248}$Cm sources but it was the first time that such a large array was installed in a reactor facility, around a fissile target irradiated by cold neutrons. We used $^{235}$U and $^{241}$Pu targets giving access to many nuclei where little was known, especially in the regions north-east of $^{78}$Ni and beyond $^{132}$Sn. This is what is shown in Figure 1 which compares the mass yield distribution from the thermal neutron induced fission of $^{235}$U and $^{241}$Pu and the yield from the spontaneous fission of $^{252}$Cf and $^{248}$Cm. The production of nuclei in the mass A~90 region is an order of magnitude larger using a $^{235}$U target compared to the two spontaneous fission sources. The same is true for nuclei in the mass A~125 region using a $^{241}$Pu target. These two mass regions were the focus of the EXILL physics case.

One main subject was the so-called Shell Model (SM) nuclei with just a few particles or holes outside a double shell closure like $^{132}$Sn. They allow very sensitive tests of the interactions that are used in this kind of calculations. One way to address this key question is to measure spectroscopic properties. Correct predictions of nuclear structure are important as nuclei here lie on, or next to, the astrophysical rapid neutron capture process. Properties, such as neutron binding energies and low-lying decaying states can change the speed and direction of the predicted r-process path. The evolution of the N=50 gap in exotic nuclei and the question of whether $^{78}$Ni is a doubly magic nucleus is highly debated. From the structure of nuclei with N=49-50 close to $^{78}$Ni, one can see how well SM interactions can reproduce excited states and to what level contributions from core excitations need to be included. In particular, we are missing the single-particle energies and the two body matrix elements for the residual proton-neutron interactions. Low lying excited states in nuclei located in the vicinity of doubly closed shell nuclei are usually dominated by the coupling of single-particle excitations to core excitations. The nature of these couplings might be responsible of basic phenomena such as the quenching of spectroscopic factors or anharmonicities in vibrational spectra. Such phenomena have been investigated in particular in the region of $^{132}$Sn with the EXILL setup. The reasons for the rapid change from a spherical to a deformed shape in the A~100 region, with the addition of just a few neutrons, remained a mystery for a long time. In recent years, studies point to the role played by deformation-driving, and resisting, Nilsson orbitals. However, such arguments
neglect any role played by the protons in such phenomena and the neutron-proton interaction is thought to be largely responsible for the onset of collectivity when moving away from closed shells. Studies of the intermediate spin states of the Kr and Rb nuclei with $N \sim 59$ will help to clarify the role played by the protons.

3. Setups

Two setups were envisaged (see Figures 2 and 3 and reference [3] for a complete description). The first one consisted in 8 EXOGAM large clovers [4], 6 large coaxial detectors from GASP [5] and the 2 clovers from the ILL. In the second setup, the GASP and ILL detectors were replaced by 16 LaBr$_3$ crystals from the FATIMA collaboration [6]. The EXOGAM and GASP detectors were mounted with their anti-Compton shield. Both configurations were instrumented with a full digital triggerless data acquisition system based on 14 bit 100MHz CAEN digitizers. The two setups were installed around the target point of the PF1B line of the ILL. The ILL reactor is the worlds brightest continuous neutron source with an in-pile flux up to $1.5 \times 10^{15}$ neutrons per s.cm$^2$. Neutrons are guided first via the H113 ballistic neutron guide [7] toward the experimental area. The neutron beam collimation is then realized by using a series of lithium and boron collimators mounted upstream the target. In order to produce around $10^4$ fissions per second in a 400 $\mu$g thick, 1cm$^2$, $^{235}$U target a flux of $10^8$ neutrons per s.cm$^2$ is required.
Details of the collimation can be found in [3]. The campaign covered two reactor cycles (93 days of effective beam time for experiments) organized as follows: 22 (14) days for spectroscopy studies with \(^{235}\text{U}\) (\(^{241}\text{Pu}\)); 13 (10) days for fast timing measurements using \(^{235}\text{U}\) (\(^{241}\text{Pu}\)) targets; during the remaining time several \((\text{n},\gamma)\) reactions were performed using \(^{36,48}\text{Ca}, \, ^{48}\text{Ti}, \, ^{68,70}\text{Zn}, \, ^{77}\text{Se}, \, ^{96}\text{Mo}, \, ^{96}\text{Zr}, \, ^{143}\text{Nd}, \, ^{155}\text{Gd}, \, ^{164}\text{Dy}, \, ^{167}\text{Er}, \, ^{194,195}\text{Pt} \) and \(^{209}\text{Bi} \): 24 (10) for spectroscopy (fast timing) studies.

In the absence of any fragment identification, it is crucial to acquire high-fold events (3 or larger) to possibly associate the observed \(\gamma\)-rays to a given fragment. This technique relies on the knowledge of the transitions deexciting the first excited states in the two fragments and the measurements of the evolution of the unknown \(\gamma\)-rays intensity gated by known transitions in a series of isotopes. \(^{235}\text{U}\) and \(^{241}\text{Pu}\) targets have been used leading for a given nucleus to two distinct complementary fragments, which greatly helps to confirm the assignment. The method relies therefore primarily on the efficient detection of high-fold data, which is achieved using a large number of efficient HPGe detectors. Such a technique is now well established and has been proven to work very well to analyze fission data [8, 9]. The total photopeak efficiency for the 16 Ge detectors setup was about 6% at \(E_\gamma=1.3\) MeV. The granularity is also crucial: even though 2/3 of this efficiency is coming from the EXOGAM clover, the gain in the number of \(\gamma-\gamma\) coincidences obtained by adding the GASP and the ILL detectors is a factor of 4.2. As an example we can compare the spectra obtained after setting two gates to select \(^{92}\text{Rb}\) in \(^{248}\text{Cm}\) fission data obtained with EUROGAM2 (10 days experiment) and our EXILL dataset. A gain of 30 in statistics is obtained. The second setup using the \(\text{LaBr}_3\) detectors was dedicated to lifetime measurements. In this configuration, the EXOGAM detectors are used to select the fragment of interest and the \(\text{LaBr}_3\) crystals give the lifetimes via the electronics fast timing technique using \(\text{LaBr}_3-\text{LaBr}_3\) coincidences [10, 11, 12, 13]. This method requires therefore Ge-LaBr\(_3\)-LaBr\(_3\) or Ge-Ge-LaBr\(_3\)-LaBr\(_3\) coincidences. Recently, a novel method to analyze such data has been developed [14] and lead to the determination of lifetimes with an accuracy of 5 ps between 40 keV and 6.7 MeV.

4. Example of results

4.1. \(^{133}\text{Sb}\): Coupling collective and single-particle states in the vicinity of a doubly closed shell nucleus

We have studied particle-core excitations in the one-valence proton nucleus \(^{133}\text{Sb}\). This study used the full complementarity of the various runs within the EXILL campaign: spectroscopy and lifetime measurements using \(^{235}\text{U}\) and \(^{241}\text{Pu}\) targets in both cases. In the spectroscopy part we search for excited states above the \(21/2^+\) isomer using the capability of EXILL to build prompt-delayed and prompt-prompt coincidences (see Figure 4). With the first condition, several transitions were found in coincidence with known, delayed transitions in \(^{133}\text{Sb}\) while the prompt-prompt coincidences, ordering of these new transitions above the isomer was possible. We also used fission events from \(^{235}\text{U}\) and \(^{241}\text{Pu}\) to ensure that the newly observed transitions were observed in both cases hence not associated to a complementary fragment of \(^{133}\text{Sb}\).

We then concentrated on lifetime measurements of the \(15/2^+\) and \(13/2^+\) states located just below the isomer. Using Ge-LaBr\(_3\)-LaBr\(_3\) coincidences, the Ge gate to select properly the decay path, the two LaBr\(_3\) ones to measure the lifetime, and the centroid shift technique [12] we could extract the lifetimes and deduce the reduced transition probabilities: \(B(M1 : 15/2^+ \rightarrow 13/2^+) > 0.24\) W.u and \(B(M1 : 13/2^+ \rightarrow 11/2^+) = 0.0042(15)\) W.u. This difference of two orders of magnitude clearly indicates a dramatic change in the structure of the two levels. In order to get insight into this puzzle a new model called the Hybrid Configuration Mixing Model was developed [15, 16] with the aim to couple collective and single-particle excitations in a fully self-consistent manner. This model suggests that the low spin states are built from a proton
Figure 4. Partial level scheme of $^{133}$Sb showing only states discussed in this paper with the newly observed $\gamma$-rays above the $21/2^+$ isomer in red and the new lifetimes in blue.

$g_{7/2}$ coupled to the $2^+~^{132}$Sn core excitation while the higher lying levels are arising from the valence proton coupled to single-neutron $h_{11/2}^{-1}f_{7/2}$ excitations. As a result the model gives 0.021 W.u and 0.001 W.u for the $15/2^+\rightarrow 13/2^+$ and $13/2^+\rightarrow 11/2^+$ transitions respectively, in fair agreement with data, which is not the case for shell model calculations [17].

4.2. Symmetry studies
$^{196}$Pt has been proposed a long time back as a good candidate to test the validity of the SO(6) limit of the sd Interacting Boson Model ([18, 19]). During the EXILL campaign, the Cologne group led by J Jolie proposed to use the fast timing technique for this purpose. In the SO(6) limit, the E2 transition operator is a generator of SO(6) and strictly speaking only transitions between states with $\Delta\sigma=0$ are allowed ($\sigma$ being one of the quantum number associated to the irreducible representations of SO(6)). Therefore, E2 transitions between different SO(6) representations ($\Delta\sigma \neq 0$) are forbidden. The states of interest have low spin and high excitation energy and are not populated in heavy-ion induced reactions. In contrary they might be well populated in $(n,\gamma)$ reactions. This was realized during the EXILL campaign using a $^{196}$Pt target and using Ge-LaBr$_3$-LaBr$_3$ coincidences to deduce the lifetimes of the third $0^+$ state at 1402 keV and the first $2^+$ state at 356 keV (which lifetime was already known at 49.2(2) ps). The measurements yielded the values of 50(5) ps for the $2^+_1$ state and an upper limit of 12 ps for the $0^+_3$ state. Using also the lower limit from Börner et al [19], it was established that $0.56$ W.u < $B(E2; 0^+_3 \rightarrow 2^+_1) < 5$ W.u and $0.05$ W.u < $B(E2; 0^+_3 \rightarrow 2^+_2) < 0.41$ W.u. The absence of collective $B(E2)$ values demonstrates for the first time the validity of the SO(6) symmetry [20].

4.3. $^{195}$Pt: the link between nuclear structure and medical physics
The structure of $^{195}$Pt was also studied and more specifically the population of the $21/2^+$ isomer using the radiative capture $^{194}$Pt(n,\gamma) reaction. This nucleus has been recognized since long as a good candidate to probe multi-j supersymmetry ([21, 22]). In addition to this fundamental interest, $^{195}$Pt is also a promising candidate for nuclear medicine since it combines therapy and diagnosis capabilities. Indeed its $T_{1/2}=4.01$ days isomer is compatible with such a usage (long enough for transportation and short enough for biology). This isomer decays via a highly converted M4 transition which provides an interesting possibility for treatment, with a very high linear energy transfer deposited within a few nm$^3$, capable to induce double strand DNA
breaks. In addition, the 99 keV gamma-ray and 65-78 keV X-rays emitted in the cascade from the isomer allow imaging using Single Photon Emission Computed Tomography and gamma cameras. During the EXILL campaign we searched for new doorway states that would significantly feed the isomer and decay to the ground state. In this study several new states have been identified in the decay from the $1/2^+$ neutron capturing state at 6.1 MeV. To envisage an abundant production of the isomer for nuclear medicine, a method to populate specifically the doorway state of interest has to be applied. For this, another experiment was realized at the HIGS facility (TUNL) where we tried to use a gamma beam at energies tuned around the newly identified states in order to measure the population of the isomer of interest. This work is still under analysis and is part of the thesis work of D. Wilmsen.

5. Conclusion
The EXILL campaign ran smoothly for two reactor cycles (~100 days) at the ILL, harvesting results from 73 proposals and gathering together 18 different laboratories. Today more than 30 papers have been published in refereed journals, showing the success of this adventure. The installation of a large and efficient gamma-ray array at a neutron beam facility is proven to be very effective in the study of neutron rich nuclei from cold neutron induced fission as well as in performing the detailed spectroscopy of close-to-stability nuclei in radiative capture reactions. Recent and new results have been discussed in the various aspects of the campaign, aiming at highlighting the diversity of the physics cases.

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