Spectroscopy of neutron rich nuclei using cold neutron induced fission of actinide targets at the ILL: The EXILL campaign

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Abstract. One way to explore exotic nuclei is to study their structure by performing γ-ray spectroscopy. At the ILL, we exploit a high neutron flux reactor to induce the cold fission of actinide targets. In this process, fission products that cannot be accessed using standard spontaneous fission sources are produced with a yield allowing their detailed study using high resolution γ-ray spectroscopy. This is what was pursued at the ILL with the EXILL (for EXOGAM at the ILL) campaign. In the present work, the EXILL setup and performance will be presented.

1. Introduction

Nowadays a large fraction of nuclear studies is devoted to the question how nuclear potential parameters change when neutrons are added to the nucleus. By adding neutrons the characteristics of the nuclear surface will change, leading to a modification of the spin-orbit interaction and the pairing forces. These parameter changes have consequences on the ordering of the nucleon orbitals and on the sequence of magic numbers compared to appearance in stable nuclei. This knowledge is also essential to understand the astrophysical rapid neutron-capture process responsible for the creation of heavy elements in the Universe.

One way to explore exotic nuclei and to gain insights into these questions is to study their structure by performing γ-ray spectroscopy. To date the most successful method to study the low-lying excited states of neutron-rich nuclei in the mass range A = 85–160 has been by placing spontaneous

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fission sources inside efficient arrays of germanium detectors, such as EUROGAM/EUROBALL [1] or GAMMASPHERE. This method allows the structure of around 250 fission fragments to be studied concurrently. Decay schemes are created by performing γ-ray coincidences [2], which allow a unique pair of fission fragments to be selected. Unfortunately only two spontaneous fission sources are available for such experiments, ^{252}Cf and ^{248}Cm.

An excellent method for cleanly producing very neutron-rich fission fragments, suitable for study using large arrays, is to induce fission reactions in actinide targets using thermal or cold neutrons, from a neutron guide. As neutrons at these energies bring just enough energy into the reaction to produce fission, the fission fragments remain very neutron-rich as there is little prompt-neutron evaporation. About ten different fissile targets are available for use with this reaction and of particular interest are targets of ^{235}U and ^{241}Pu. These two targets give access to many nuclei where currently nothing or little is known, especially in the regions north-east of ^{78}Ni and beyond ^{132}Sn. These two regions of the nuclear chart are especially important for testing the interactions used in shell-model calculations far from stability. The use of different fission systems gives access to the structure of many new nuclei.

The combination of intense cold neutron beams available at the Institut Laue-Langevin (ILL), together with the high detection efficiency of a large array of germanium detectors has offered a unique opportunity for a set of experiments devoted to nuclear spectroscopy at the ILL: the EXILL (for EXOGAM at the ILL) campaign. A combination of EXOGAM segmented clover detectors [3], large volume GASP coaxial detectors [4] and standard clover detectors has been used to achieve a large solid angle coverage around the sample and a good detection efficiency. γ-rays from neutron capture on stable targets and from the decay of neutron-rich nuclei produced in the fission process of ^{235}U and ^{241}Pu samples have been acquired in a trigger-less mode. The experimental setup and performance measured with a ^{235}U target will be presented.

2. Experimental setup

2.1 Beam and target environment

The EXILL campaign took place at the ILL PF1b beam line at the end position of the H113 ballistic neutron guide [5]. The capture flux at the guide exit is about 2.10^{10} neutrons cm^{-2} s^{-1} with a beam size of 20 cm $\times$ 6 cm. In order to allow closer geometry for the Ge detectors and avoid the production of additional γ background from neutron capture on the structural materials, the beam size has been reduced to match a diameter of about 1 cm at the sample position. To do so, a series of boron and lithium collimators have been introduced upstream.

The neutron beam collimation system consisted of two ceramic B_4C collimators 10 mm thick and three ^{6}LiF ceramics collimators (5 mm thick) backed with 2.5 cm thick borated plasic disks. The collimators were placed at a distance of about 50 cm from each other in long pipe sections covered with 3 mm borated plastic form the inside to absorb scattered neutrons. The collimators were backed of Pb to absorb γ-rays from boron neutron capture. The resulting flux at the target position is about 10^8 neutrons cm^{-2} s^{-1}, consistent with the simulations of the collimation system.

The target chamber was made of an aluminium pipe of 50 mm diameter and 2 mm wall thickness to minimise γ absorption. The chamber was surrounded with a 1 mm thick ^{6}LiF rubber to absorb scattered neutrons and reduce neutron induced background on germanium crystals. Finally, a beam-stop consisting of a ^{6}LiF absorber was placed at about 2 m from the sample position (see Fig. 1). It was placed inside a long pipe section covered with B_4C and surrounded by a 20 cm thick Pb shielding.

Three actinide target were used for fission fragment spectroscopy: ^{235}U (99.7% enriched, 575 μg cm^{-2}), ^{235}U (99.7% enriched, 675 μg cm^{-2}) and ^{241}Pu (78.6% enriched, 300 μg cm^{-2}). All of them were sandwiched between dense backing (15 μm Zr, 25 μm Be and 25 μm Be respectively) for rapid stopping of fission fragments allowing an almost Doppler-free measurement of the emitted γ-rays.
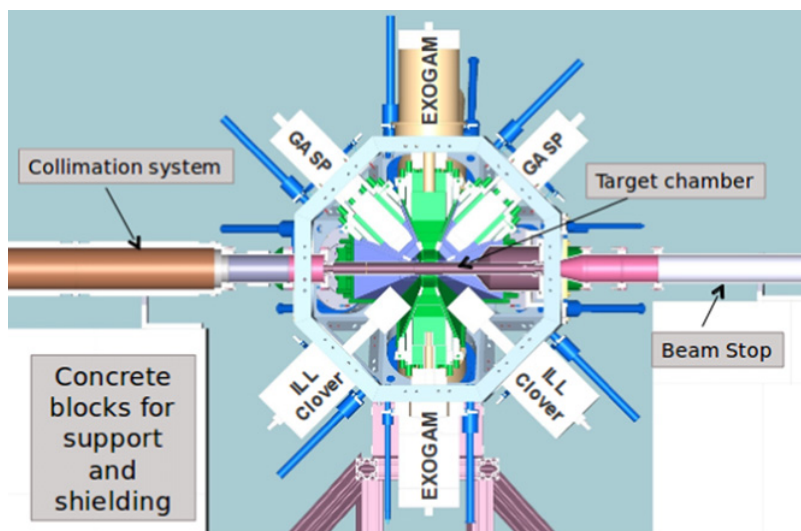
Figure 1. Schematic side view (parallel to the beam axis) of the EXILL experimental setup. The setup with the inner chamber used for the ^{241}Pu measurement is drawn.

A target holder made of Teflon was inserted inside the chamber to keep all (n,γ) and ^{235}U samples in reproducible position. In order to minimise neutron scattering, the complete collimation line, the target chamber and the beam stop were connected and kept under a vacuum of about 10^{-2} mbar. For safety reason this configuration has been modified for the ^{241}Pu runs. To ensure an additional physical barrier it was placed inside an inner chamber under a vacuum of the order of 1 mbar, the external chamber being filled with He gas at 50 mbar. The inner chamber was made of a aluminium tube of 35 mm inner diameter and 2 mm wall thickness sealed with 200 μm thick Zr windows to minimise neutron scattering.

2.2 Detector array

Figure 1 displays the different detectors forming the EXILL detector array and their placement in space around the target chamber. It consists in total of 16 germanium detector units:

- 8 EXOGAM segmented clover detectors from GANIL. They were placed at 45°, in a single ring, in the perpendicular plane with respect to the beam axis. Each detector consists of four co-axial n-type Ge crystals arranged in the configuration of a four leaf clover and housed in the same cryostat. Note that, as mentioned in section 2.1, the actinide targets were made in such a way that all fission fragments stopped shortly after emission. As a result, there was no need for a complex Doppler correction and the segmentation of the EXOGAM detectors was not used and only the core signal from each crystal was recorded.
- 2 non-segmented clover detectors usually in use at the Lohengrin mass separator at the ILL. They were placed at 45° below the sample, parallel to the beam axis. Those clovers have a thinner entrance window. As a result they are more efficient for detection of low energy γ-rays.
- 6 GASP n-type 80% relative efficiency coaxial detectors from LNL Legnaro filling the 6 remaining positions at 45°.

All detectors, with the exception of the two ILL clovers, were equipped with active BGO (bismuth germanate) Compton-suppression shields. Heavy metal collimators were placed in front of the BGO shields to reduce the background and the cross-talk between adjacent detectors. All the signals from
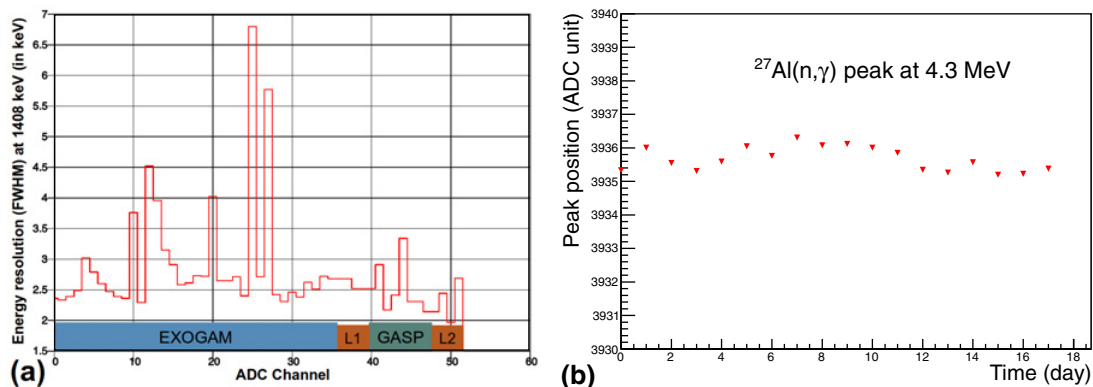
Figure 2. (a) Energy resolution (FWHM) at 1408 keV as a function of the germanium crystal. (b) Position (in ADC unit) of the ^{27}Al(n, γ) peak at 4.3 MeV as a function of time for the first crystal of the EXOGAM clover 7 (channel 28 on (a)).

the BGO detectors were recorded together with those from the Ge crystals and the anti-coincidence discrimination has been performed off-line.

In addition, part of the EXILL campaign has been devoted to fast-timing measurements using LaBr$_3$Ce detectors. These fast response scintillation detectors have an energy resolution of about 3% and an excellent time response ($<$200 ps). They are particularly suited for lifetimes measurements in the range of a few picoseconds to several nanoseconds [6, 7]. Therefore for these measurements, the GASP detectors and the ILL clovers have been replaced by 16 cylindrical LaBr$_3$Ce scintillators from the FATIMA collaboration. All crystals were 1.5″ in diameter, half of them being 2″ long and the other half being 1.5″ long. They were all mounted on Hamamatsu R9779 photomultiplier tubes.

For the EXILL campaign, a fully digital, triggerless, acquisition system capable of handling event rates up to almost 1 MHz has been developed [8]. The analogue signals from the detector preamplifiers were sampled by ten V1724, 8-channel, 14-bit, 100-MS/s CAEN digitiser cards [9]. The numerical information about the amplitude of the analogue signals and their arrival time were collected by two RIO3 PowerPC-based VME single board computer (data concentrator) [10] before being recorded on a computer hard disk. Depending on the detector setup, the data acquisition system was handling from 68 (spectroscopy) to 72 (fast timing) separate channels.

3. Performance

The EXILL campaign was spread over two ILL reactor cycles, for a total of around 100 days. The first reactor cycle was devoted to spectroscopy measurements using both stable and ^{235}U targets. The second reactor cycle was shared amongst fast timing measurements (see section 2.2) with stable, ^{235}U and ^{241}Pu targets, and γ spectroscopy with the ^{241}Pu target. During the entire campaign, in addition to the 3 actinide targets, 21 different $((n, \gamma))$ samples have been studied and less than 5% of the beam time has been lost due to technical problems or configuration change over. A total amount of about 60 TB of triggerless data has been recorded. Before and after each target change over, calibration measurements have been performed using a ^{60}Co source, a calibrated ^{152}Eu source (170 kBq) and a BaCl$_2$ sample for a high energy calibration. In addition, capture of scattered neutrons on structural materials provides known and intense γ-rays (mainly from ^{27}Al(n, γ) and ^{27}Al$(\beta\gamma)$) during the measurements.

The energy resolution of each germanium crystal of the EXILL detector array was measured using both ^{60}Co and ^{152}Eu sources. Figure 2a shows the energy resolution (FWHM) at 1408 keV as a function
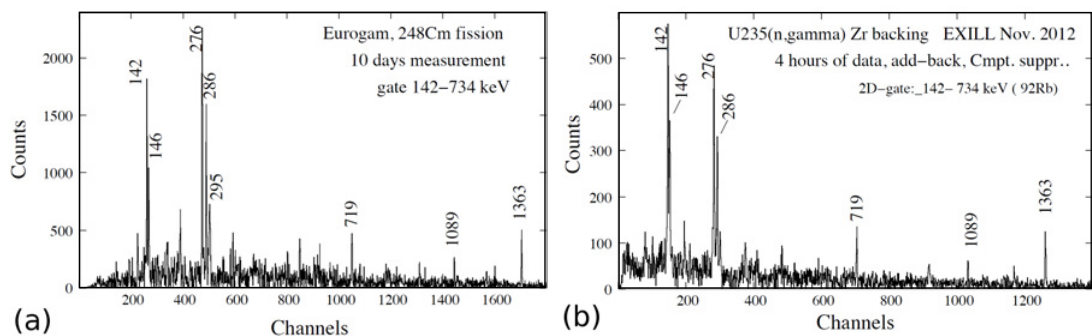
Figure 3. γ-ray spectrum gated on the ^{92}Rb low-lying transitions at 142 keV and 734 keV from ^{248}Cm EUROGAM data (a)[1] and ^{235}U(n, f) EXILL data (b).

of the germanium crystal for a ^{152}Eu calibration run recorded during the first reactor cycle, before the ^{235}U spectroscopy measurements. It shows a mean energy resolution of around 2.5 keV. However, the energy resolution of some crystals is worse than 3 keV. This is partly due to historically damaged germanium crystal and to a rather noisy environment from the electronic point of view.

The ^{28}Al γ-rays from the neutron capture on the structural materials allow to follow the stability of the germanium crystals during several weeks of measurement regardless of which target is being irradiated. As an example, figure 2(b) displays the peak position (in ADC unit) of the $^{27}Al(n, \gamma)$ peak at 4.3 MeV as a function of time for the first crystal of the EXOGAM clover 7 (channel 28 on Fig. 2a). It shows very good stability (better than 0.015%) over the 18 days of the two ^{235}U(n, f) measurements of the first reactor cycle. All EXOGAM and ILL clovers crystals show similar results. However some of the GASP crystals show slightly higher deviation, up to 0.04%.

Data evaluation performed during the various runs or immediately after, indicates the quality of the data sets. As an example, Figure 3b shows the γ-ray spectrum gated on two ^{92}Rb low-lying transitions (142 keV and 734 keV) obtained from a 4 hours dataset of the ^{235}U(n, f) measurement. All the γ lines observed come from the transitions in ^{92}Rb. These data have to be compared with a previous measurement performed using the EUROGAM array with a ^{248}Cm spontaneous fission source. Both setup have a comparable γ efficiency. The data from [1] (see Fig. 3a) have been collected in 10 days while our data plotted on Figure 3b are extracted from a very small dataset of 4 hours. The whole ^{235}U(n, f) campaign lasted 18 days. This result illustrates the very good quality of the data taken during the EXILL campaign.

4. Conclusion

The EXILL campaign ran for about 100 days of beam time, 95% of it being used for data taking. A dedicated collimation system has been developed to produce a halo-free pencil (1 cm^2) neutron beam of about 10^8 n cm^{-2}s^{-1} at the ILL PF1b line. Up to 50 Ge crystals, 14 BGO and 16 LaBr$_3$(Ce) fast scintillators have been operated simultaneously. A fully digital, trigger-less, acquisition system has been developed and it has proven to be able to handle considerable event rates up to almost 1 MHz. 24 different samples have been irradiated, including ^{235}U and ^{241}Pu samples and a total amount of about 60 TB of data has been produced. The data storage is shared between ILL, CC-IN2P3 and LPSC Grenoble and a grid-based distributed system (IRODS) allows user friendly access to the data. The potential physics output from such a campaign is considerable. This is clearly manifested in the very large (80) number of scientific proposals which have been submitted within this campaign.

The EXILL campaign has also been an important step for the development of the new ILL project FIPPS (FIssion Product Prompt γ-ray Spectrometer) [11] based on a combination of a large germanium

array and a gas-filled magnetic spectrometer. The great success of this campaign demonstrates that ILL is ready to operate the first part of the FIPPS setup, including the safe operation of a complex Ge detector array close to an intense neutron beam.

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