Studies for γ -ray emission in the fission process with LICORNE

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Abstract. The LICORNE neutron source is a new device at the ALTO facility. Its use of inverse kinematics makes the production of naturally focused neutron beams possible with an energy range of 0.5 to 4 MeV. This is perfect for any studies concerning fast neutron induced reaction based on detection setup that require to be placed in a compact geometry – such as Ge based γ detection setup.

In this paper, neutron production with LICORNE is described. The development of a gas cell target and the extension of the energy range up to 7 MeV with the $p(^{11}\text{B},n)^{11}\text{C}$ are presented. An overview of the major research fields studied with LICORNE is given and two types of experiment are presented. The first one dedicated to prompt fission γ -ray emission in fission, as a function of incident neutron energy, is described. Some preliminary results are shown. The second, in the context of the MINORCA campaign, is detailed. The most recent outcomes in the data analysis process are also presented.

1 Introduction

The LICORNE project started in 2012 with the test of a new technique to produce neutrons using the $p(^7\text{Li},n)^7\text{Be}$ reaction in inverse kinematics. A commissioning experiment clearly showed the capability of LICORNE, to naturally produce fast neutrons, with energy of 0.5 - 4 MeV, in narrow cones ($< 20^\circ$).

The development of this neutron source using an hydrogen gas target has opened up possibilities for new experiments about the γ spectroscopy of fast neutron induced reactions. Since then, two research programs have been initiated. The first one concerns the emission of prompt fission γ -rays (PFG) from major actinide nuclei. This is of interest for understanding the emission of neutral particles in the fission process. Indeed, the competition between neutrons and gammas is still a puzzle that theoretical descriptions need to understand. At the same time, these measurements are directly related to nuclear energy and reactor safety, since such data are used as source terms in gamma heating calculations in reactor cores. The other axis of research of the LICORNE physics program concerns the γ spectroscopy of individual exotic fission fragments at the moment of their formation. This gives access to collective and highly excited states which will provide new interesting information

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on nuclear structure for fission fragments that are still unknown. To achieve this goal, LICORNE was coupled in March 2015 with the Miniball high efficiency gamma ray spectrometer on loan from ISOLDE at CERN. This coupling was only made possible by careful elimination of all secondary isotropic neutron fields generated by the LICORNE directional neutron source.

In the following, a description of the LICORNE neutron source and its recent developments will be given. Then, an overview of the most recent results obtained for PFG and the MINIBALL experiment will be given.

2 Presentation of the LICORNE neutron source

In 2012, at the ALTO facility [1, 2], a new project was started. The aim was to add neutron beam production capability to this facility in prevision of the NFS project [3] start at GANIL. In Europe, several neutron facilities were already running at that time: GELINA in Geel [4], ILL in Grenoble [5], or KFKI research reactor [6] in Budapest, ... It was necessary for LICORNE to be different from these installations in order to be able to explore new ways of using neutrons. In many facilities, a light primary beam impinging an heavy target is used to induce nuclear reactions which produce neutrons. In this case, the neutron emission is isotropic. As a consequence, it is mandatory, for any detection setup, to be at a great distance from the source. Thus, the neutron fluxes are quite small compare to the total amount of produced neutrons, making almost 99% of them useless and participating to the neutron background. In addition, any detector, placed in the neighbourhood of the sample, still needs to be heavily shielded to not be blinded by the source neutrons.

In order to be complementary with the other sources, LICORNE also had to be able to produce decent fluxes of fast neutrons without the use of a very intense ($< \mu\text{A}$), or very energetic ($> 100 \text{ MeV}$), primary beams. It also required to provide in the ALTO environment, where experimental caves are small compare to typical installations, a very low neutron background.

2.1 Description of the $p(^7\text{Li},n)^7\text{Be}$ reaction for neutron production

The LICORNE neutron source meets those criteria thanks to the clever use of specific reactions in inverse kinematics. In his work, M. Droszg [7] have listed a wide range of reactions that produce neutrons with energies from keV to 500 MeV. To satisfy our needs (see §.3), fast neutrons with energy close to fission neutrons were required. For this reason, the $p(^7\text{Li},n)^7\text{Be}$ reaction was chosen. The results of 2-body relativistic kinematics calculations are shown on the left side of figure 1. With beam energy in the range of 13.5 to 16.5 MeV, it is possible to produce neutron beams in the laboratory frame with two different energies corresponding to forward and backward emission of neutrons in the centre of mass frame. The relative size of the principal (high energy) peak and satellite (low energy) peak is governed by both the relativistic kinematics of the focusing and the angular distribution of emission in the centre of mass frame, which changes drastically as a function of ^7Li bombarding energy. The gain from the focusing and natural collimation can be expressed in terms of neutron flux enhancement over the non-inverse reaction (see right part of fig. 1). To obtain this curve, the reaction rates have been calculated for different ^7Li kinetic energies. The neutron flux per steradian has been normalized to the ^7Li beam intensity. At threshold the enhancement factor is maximal, since the emitted neutrons move with the centre of mass of the system, which follows the ^7Li beam direction. As a consequence, close to the threshold energy, it is possible to produce very narrow ($< 5^\circ$) conical neutron field. With the increasing ^7Li bombarding energy, the cone broadens and the number of neutrons in a given solid angle decreases, which causes the enhancement factor to drop. For a beam energy lower than 16.5 MeV, the maximum opening of the cone is smaller than 29° . In this case, an enhancement factor of

Figure 1. On the left: kinematic curves relating the angle of neutron emission to neutron energy in the laboratory frame for different ${}^7\text{Li}$ bombarding energies from 13.15 to 16.5 MeV, calculated using two-body relativistic kinematics.

On the right: the top panel shows the enhancement factor of the neutron flux between the inverse kinematic and the direct kinematic reaction as a function of ${}^7\text{Li}$ bombarding energy. The bottom panel shows the $p({}^7\text{Li},n){}^7\text{Be}$ reaction cross section over the same energy range.

Figure 2. Technical scheme of the LICORNE neutron source chamber. The right hand figure is a zoom of the gas cell.

30 can still be expected over the non-inverse reaction.

The maximum available fluxes of up to 10^7 neutrons/s/sr (assuming ${}^7\text{Li}$ beam currents of 100 nA) are comparable to those available at other conventional quasi-monoenergetic neutron sources. However, the natural collimation and neutron beam directionality provides at least two distinct advantages: (i) the placement of gamma detectors adjacent to the sample being irradiated becomes feasible; (ii) the scattered neutron background in the room will be reduced by up to a factor of 100.

2.2 Facts about the LICORNE neutron source

To produce neutrons with the Tandem ^7Li beam, a dedicated reaction chamber had to be built. At first, the reaction chamber is a cylindrical aluminum chamber of 17 cm. The exit window is made of aluminum and is only 0.3 mm thick. Inside, a very proton rich target ($\sim 10^{19}$ p/cm²) had to be placed. A mini-camera and an illuminating LED are included for beam tuning and inspecting the target from inside. At first, a simple layer of polypropylene of 4.4 μm thickness was chosen. However, the carbon content allowed parasitic fusion-evaporation reactions that produce isotropic neutrons. Added to this, this material cannot stand high intensity beam exposure. For these reasons, the choice of a gas target was made: it was possible to limit the presence of low Z material in the beam path. The exit flange was modified to be able to attach a cylindrical gas cell made of steel. A 5 mm diameter hole was drilled in the flange. The hole's contour were coated with a 50 μm thick tantalum foil to prevent any fusion-evaporation reaction with the flange material. The gas is separated from the beam line vacuum by a 4 μm tantalum foil. In case of a high intensity beam, the tantalum window is air cooled. Inside the gas cell, pure H_2 gas flows with pressure above 1 bar and up to 1.8 bar. The bottom of the gas cell is coated with 100 μm lead foil to form a beam dump. Figure 2 shows a technical drawing of the design.

Different sizes of the gas cell are available depending on the needs. The longer the gas cell, the greater is the ^7Li beam energy loss and the wider is the neutron energy dispersion. Four sizes are now available: 2, 3.5, 5.5 and 7.5 cm. The bigger ones are used when a high neutron flux is wanted: the ^7Li enter with the maximum energy and slows down to threshold energy. All the neutron energies between 0.5 and 4 MeV are produced in a broad spectrum. On the contrary, short cells are used when a precise neutron energy is required: ^7Li beam loses little energy and the neutron energy spread is small. The minimal energy spread measured was 300 keV.

Nowadays, only gas cells are used for the neutron production with LICORNE. In the near future, some upgrades are foreseen to increase the energy range.

2.3 The LICORNE energy upgrade

Getting to higher energies requires a different primary beam. By comparing M. Drosge's work [7] an the available beams at the Tandem [10], ^{11}B was considered. It is a good candidate because it can be produced with a rather good intensity (~ 100 nA) and the kinematics allow the extension of LICORNE energy range up to 12 MeV – see fig.3.a – with features similar to ^7Li kinematics. However, the cross-section (see fig.3.b) for the $p(^{11}\text{B},n)^{11}\text{C}$ reaction is, on average, four times smaller than for the ^7Li induced reaction. This beam has been used for the first time, in may 2015, during an experiment dedicated to test the PARIS detector [14] response to neutrons with energies of 4 to 12 MeV. During this experiment the reaction kinematics at 0° was the expected one. Neutrons, up to 12 MeV, were produced. However, another test experiment was conducted in november to make quantitative measurements of fluxes and source background. We observed an opening of fusion-evaporation channels when beam energy reaches 44.5 MeV. From that energy, as many neutrons are produced outside the cone as inside and the natural collimation provided by the inverse kinematics is no longer an advantage.

This test experiment demonstrated the new capabilities of LICORNE to produce neutrons, in a narrow cone, up to 7 MeV with fluxes of 10^{5-6} n/s/sr.

3 Studies of Statistical properties of Prompt fission γ -rays

As mentioned previously (see 1), the LICORNE physics program follows two major axes. The first one is dedicated to the measurement of prompt fission γ -ray properties. Prompt γ emission in nuclear

Figure 3. a. kinematic curves relating the angle of neutron emission to neutron energy in the laboratory frame for different ¹¹B bombarding energies from 33 to 58 MeV, calculated using two-body relativistic kinematics. b. Measured cross section for the p(¹¹B,n)¹¹C reaction taken from [11, 12].

fission is one of the least well-understood parts of the fission process. It has important applications for nuclear reactors. Prompt fission γ emission contributes up to 40% to the reactor core γ -ray spectrum. Induced γ heating effects, particularly at the core periphery are closely linked to reactor safety. These are still largely underestimated [13] and measurement of spectral shapes, average multiplicities and distributions, average energy per quantum and total energy are at the top of OECD/NEA high priority request list [15, 16]. Besides its importance for more detailed databases for nuclear reactors physics, understanding γ /neutron competition in the fission process is important for the modeling of post-scission phases of the nuclear fission. Unlike neutrons, prompt γ -rays remove most of the fission fragment spin. Studying the spectrum can help understanding the deexcitation process of fission fragments and the angular momentum generation. So, this type of studies can provide information on the partition of energy and angular momentum, and also on the competition between neutron and γ emission during fission fragment de-excitation.

In 2013, comparative prompt fission γ -rays characteristics for fast neutron induced fission of two key nuclei, ²³⁵U and ²³⁸U, have been measured for the first time. In this experiment, a broad neutron spectrum with an average energy of 1.5 MeV was produced using LICORNE. Fourteen BaF₂ crystals from the Château de Cristal were placed 30 cm away from an ionisation chamber. After a careful selection of γ -rays, by time-of-flight, relative shapes of the PFG spectrum from fast neutron induced fission of ²³⁵U and ²³⁸U were observed to be similar within the experimental error bars [17].

In february 2015, we participated in our CEA/DAM collaborators' experiment with LICORNE. At first, it was designed to measure prompt neutron emission for ²³⁸U fast neutron induced fission. For a week, they bombarded an ionisation chamber containing ~ 300 mg of ²³⁸U with the LICORNE neutron beam. Two energies were selected: 2.4 and 3.3 MeV. In the setup, three 2"x2" LaBr₃ crystals were placed 40 cm above the ionisation chamber to measure prompt fission γ -rays at the same time. Off beam, a similar ionization chamber containing ²⁵²Cf sample replaced the one containing ²³⁸U. For two days, the prompt γ -rays of spontaneous fission of ²⁵²Cf were measured. Analysis on this smaller data set was performed. First, fission fragments were selected in the fission chamber. They can be easily separated from α and β thanks to the higher charge collected on the anodes. With this fission

Figure 4. TOF vs measured energy for one LaBr_3 crystal. Two major horizontal structures can be distinguished. The lower one corresponds to the γ . The triangular one just above corresponds to neutrons.

Figure 5. Prompt fission γ -ray spectra for each of the LaBr_3 crystals: Thalia, Aglaea and Euphrasyne. The two other spectra are taken from [19]. The subview in the bottom left corner is a zoom of the low energy region.

tag, we measured times of flight of particles between ionization chamber and LaBr_3 crystals. Then, a matrix representing time of flight as a function of the energy measured in one crystal is generated (see fig. 4). In such matrix several structure can be distinguished. The lowest horizontal one corresponds to γ energy deposit in the crystal. A projection of this structure on the X-axis will give the experimental prompt fission γ -ray spectrum. The second triangular structure, just above, is created by neutrons. The vertical lines in this triangle correspond to (n,γ) reaction with La or Br isotopes in the crystal.

After projection, and background subtraction, the obtained spectrum needs to be unfolded according to the response function of the crystal, which have been simulated using GEANT4 [18]. Up to 200 energies of incident γ -rays were simulated in the range of 10 keV to 10 MeV, with an energy interval determined by the detector FWHM function. The simulation takes into account the detector geometry, its environment, as the different photons interactions that occur in the crystal. Then, a deconvolution algorithm, detailed in [19], is used to extract the spectrum emitted during the fission process. This analysis technique is then applied to each crystal separately to obtain three independent PFGS for ^{252}Cf . Then, the PFGS have to be normalised to the number of fission detected during the experiment. After these different steps, our first need was to validate the data analysis process. For this, our spectra were compared to R. Billnert's work[19]. Results are presented in figure 5. One can

Table 1. Statistical properties of prompt fission γ -rays measured for ^{252}Cf and compared with R. Billnert's *et al.* works

Detector Name	Total Energy (MeV)	Average Energy (keV)	Average Multiplicity
Aglaea	6.04 ± 0.25	742 ± 23	8.14 ± 0.24
Thalia	6.44 ± 0.26	761 ± 22	8.47 ± 0.24
Euphrasyne	7.08 ± 0.29	842 ± 25	8.41 ± 0.25
Average	6.47 ± 0.15	777 ± 13	8.34 ± 0.14
Billnert <i>et al.</i>	6.65 ± 0.04	800 ± 10	8.29 ± 0.03

notice the perfect agreement of our three spectra with our collaborator's work. The lower left corner subview enlarge the energy region between 60 keV and 2 MeV. Again, all the structure at low energy are also observed. From these spectral data, we can extract average energy, multiplicity and total energy emitted under the form of γ -rays in the fission process. The results are shown in table 1 and compare with R. Billnert's work. Again, the obtained results are in perfect agreement with previous measurements. As a consequence, our data analysis on ^{238}U PFGS for two neutron energies of 2.4 MeV and 3.3 MeV can be continued.

In 2016, two experiments are planned to measure prompt fission γ -ray. First, we will use higher neutron energies (up to 7 MeV) to understand γ emission when second chance of fission channel is opened. In the second one, we will induce fission of ^{239}Pu to study influence of the fissioning system on prompt fission γ properties.

4 Spectroscopy of Prompt fission γ -rays of ^{238}U

4.1 Presentation of the LICORNE experiment in the MINORCA campaign

In parallel to our PFG research activity, we started in 2014 to study the feasibility of performing fine γ spectrometry of neutron induced reaction – radiative capture, fission, ... The possibility to study the nuclear γ emission of fission fragments at the moment of their creation could provide important information about their structure. The ratio of neutron/proton is a very interesting quantity, which can be used to determine the predicted properties of a nucleus. The extreme of this ratio define the limits of existence for nuclear matter. For neutron rich nuclei, which are close to the limits of existence, specific parts of the nuclear interaction are strongly amplified or tend to vanish, which depict our understanding of the nuclear interaction between nucleons.

In the past, major campaigns of measurement, such as EXILL [20], were performed. Compared to these, fission of fertile nuclei can be induced with LICORNE. With, from start, three more neutrons and a rather cold process, one should expect to produce more exotic species. A test experiment with the ORGAM detector [21] was performed in january 2014 to prepare the MINIBALL campaign (MINORCA) in 2014-15 at ALTO. This experiment was used to understand γ background and neutron exposure of germanium crystals due to scattering in the target. After making clear there was no risk for the Ge crystals, during the campaign, LICORNE was coupled to the MINIBALL array [22] as can be seen in picture 6. A γ detection efficiency of $\sim 10\%$ was achieved. A peak-to-total ratio of $\sim 20\%$ was expected. The uranium target was placed 1.5 cm away from gas cell. It was a half cylinder of uranium metal with 1.2 cm diameter and a length of 3 cm. A pulsed ^7Li beam from the IPN tandem was used to bombard hydrogen gas in gas cell to generate neutrons. The beam pulses were 2 ns wide and separated by 400 ns with an average beam current of approximately 4 nA. The beam energy and

Figure 6. Picture of the MINIBALL setup. In the center, the LICORNE gas cell is visible. It is surrounded with lead and copper shielding. Around, the LICORNE chamber the eight MINIBALL cluster are placed around a sphere of 14 cm radius. At the center a metallic target of ^{238}U is placed to be irradiated.

gas cell length were chosen to maximize the flux for a cone of neutrons with an opening angle of not greater than 20 degrees. The length and pressure of the gas chamber (3.5 cm and 1.5 atm) assured that the beam energy slowed from 15 to 13 MeV producing maximum number of neutrons per second (order of 10^6n/s).

A limited fission rate of 50 kHz was achieved in order to maintain MINIBALL single crystal counting rate about 8 – 9 kHz. In order to tag the fission process a trigger was implemented in the data acquisition system. Three clusters had to be fired in a time window of $1.8\text{ }\mu\text{s}$. Among those three γ -rays firing clusters, two had to be prompt ($\Delta t < 50\text{ ns}$ with a beam pulse) and the third one could be either prompt, or delayed: $\Delta t < 1.8\text{ }\mu\text{s}$ with a beam pulse. A trigger rate of 5 – 6 kHz was achieved. Data were acquired for two weeks and up to 10^9 triple coincidences, and higher folds, were stored to disk.

4.2 Preliminary Results

A preliminary analysis has been performed on this data set. After calibration and alignment in time of the different crystals, add-back algorithm has been implemented. It improved peak-to-total ratio from 16% up to 21%. The result of this first step is shown on figure 7. It represents the γ -ray energy measured *versus* the time difference between a γ hit and a beam pulse in a $1.8\text{ }\mu\text{s}$ window. Because of the 400 ns beam period, several pulses can be seen on this matrix. Horizontal lines can also be spotted. They belong to isomeric de-excitation in some fission fragment isomers.

4.2.1 Prompt γ - γ matrix

The first method to extract interesting data from this matrix is to take all the coincidences between γ -rays belonging to the first pulse. These two γ -rays are considered as prompt. This technique allow the selection of a fission fragment and its partner. However, the γ - γ matrix contains many lines. Figure 8 shows the spectrum obtained with a 814 keV energy gate on the matrix. This γ line belongs to ^{96}Sr . The fission partners are Xe isotopes with masses around 141. On the spectrum, many lines are identified and belong to the expected Xe isotope. The nuclei identified are up to eight neutrons away

Figure 7. Two dimensional plot of the energy measured in one of the cluster as a function of the time difference between hit and beam pulse. The total time window is $1.8 \mu\text{s}$. The vertical structures correspond to different beam pulse.

Figure 8. Spectrum conditioned by a 814 keV γ -ray extracted for the γ - γ matrix obtained from fig.7. ^{96}Sr and $^{140,142}\text{Xe}$ γ lines are visibles.

Figure 9. Total projection of the γ - γ matrix conditioned by the isomeric decay of the 164 ns state in ^{134}Te . ^{134}Te and $^{102,104}\text{Kr}$ γ -lines are visibles.

from the last stable isotope. This preliminary result is a good proof of concept for this experimental technique dedicated to the study of fission fragment at the moment of their formation. Excited states with angular momentum up to 8^+ were identified. If one can expect higher spins, this technique is already complementary to actual β -decay measurements which are usually pioneers in accessing structure data very far from stability. Unfortunately, the low peak-to-total ratio is a major drawback making only the most produced fission fragment γ lines visible in the matrix. This way of analysing the data is not perfectly suited to explore the most exotic regions.

4.2.2 Delayed-Prompt γ - γ coincidences.

As seen previously, the peak visibility in the fold two matrix needs improvement to give access to more neutron rich fission fragment. Thus, a higher selectivity is required. Based on coincidence technique, a good way to access to exotic nucleus is to select one that is close to stability. For example, a gate on a transition on very long lived Sn isotopes could give access to Mo nuclei which are beyond the actual knowledge on structure in this region. A simple energy gate might not be selective enough to clean the conditioned spectrum. For this reason, the use of isomeric transitions was considered.

On figure 7, an horizontal line at 1279 keV can be observed between the pulses. It corresponds to the de-excitation of the isomeric state ($T_{1/2} \sim 164$ ns) decay of ^{134}Te . In addition, a projection of the region between beam pulses on the vertical axis would allow the identification of up to 20 isomeric transitions in different nuclei. If we select any two γ in the pulse in coincidence with the isomeric line, we obtain the figure 9. In this spectrum, γ lines from $^{102,104}\text{Kr}$ are visible. These nuclei are 6 and 8 neutrons away from the last stable Kr isotope. This spectrum is also very “clean” compared to figure 8.

If using triple coincidences can end in a lack of statistics, this result demonstrates that isomers can

provide a very high selectivity which is crucial to this technique. We expect that future data analysis will allow us to cross the actual frontier on structure knowledge for very neutron rich fission fragments.

5 Conclusion and Perspectives

In this paper an overview of the research activity with the LICORNE neutron source has been given. Well suited for the study of neutron induced reactions, LICORNE is now providing important results about prompt fission γ -rays. In the near future, the outcomes of the data analysis about the ^{238}U experiment with 2.4 and 3.3 MeV neutrons will be compared with theoretical calculations. Furthermore, this experimental program will be completed in 2016 with two other experiments.

In parallel, MINORCA data will be extensively analysed. The information extracted will prepare us for our future measurement campaign. In the next two years, a Ge/LaBr₃ hybrid array is expected to be mounted in Orsay. The very good time resolution of LaBr₃ will increase the selectivity of the triple coincidences technique described previously. Added to a greater γ detection efficiency and a better peak-to-total ratio, we intend to explore even exotic regions. In addition, ambitious experiments dedicated to fission isomer studies will be considered.

As a matter of fact, in the next years until NFS starts, LICORNE will pave the way by testing bench for many new experimental techniques and by producing many interesting data.

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