Measurements of prompt gamma-rays from fast-neutron induced fission with the LICORNE directional neutron source

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Abstract
At the IPN Orsay we have developed a unique, directional, fast neutron source called LICORNE, intended initially to facilitate prompt fission gamma measurements. The ability of the IPN Orsay tandem accelerator to produce intense beams of ^{7}Li is exploited to produce quasi-monoenergetic neutrons between 0.5 - 4 MeV using the p(^{7}Li,^{7}Be)n inverse reaction. The available fluxes of up to 7×10^{7} neutrons/second/steradian for the thickest hydrogen-rich targets are comparable to similar installations, but with two added advantages: (i) The kinematic focusing produces a natural neutron beam collimation which allows placement of gamma detectors adjacent to the irradiated sample unimpeded by source neutrons. (ii) The background of scattered neutrons in the experimental hall is drastically reduced. The dedicated neutron converter was commissioned in June 2013.

Some preliminary results from the first experiment using the LICORNE neutron source at the IPN Orsay are presented. Prompt fission gamma rays from fast-neutron induced fission of ^{238}U, ^{232}Th and ^{235}U were measured by two different techniques. An ionisation chamber containing 10 mg samples of ^{238}U and ^{235}U to provide a fission trigger was used in conjunction with BaF_2 and $LaBr_3$ scintillator gamma detectors to detect fission fragment-gamma-ray coincidences. In the second part of the experiment thick targets of around 50g of ^{238}U and ^{232}Th were used, with a high efficiency calorimeter to tag on fission events by requiring both high sum energy and multiplicity.

1 Introduction

Conventional quasi-mono-energetic neutron sources produce neutrons isotropically via direct reactions on light nuclei (e.g. d(d,p)n or ^{7}Li(p,n)^{7}Be). The lack of directionality means the typically less than 1 percent of the source neutrons produced can be used for irradiating samples, the vast majority instead contributing to the room background. However, natural collimation of neutron beams can be achieved if the neutrons are produced using a reaction in inverse kinematics where the projectile is much heavier than the target. Neutron production via this method thus combines the best features of white neutron sources (collimated beams) and conventional quasi-mono-energetic neutron sources (high neutron fluxes at short distances).

The LICORNE neutron source is based on the p(^{7}Li,^{7}Be) reaction in inverse kinematics[1]. It has been initially developed for performing fundamental studies of the nuclear fission process and associated nuclear data measurements related to 4th generation nuclear reactors. A first experimental program will involve the study of prompt gamma-ray emission in fission since the directional neutron beam will allows placement of gamma-ray detectors out-of-flux but adjacent to the sample to be irradiated. However, other potential uses of LICORNE span several different research fields and include gamma-spectroscopy of neutron-induced reactions, measurements of capture and inelastic scattering cross sections, non-destructive assay of nuclear waste and irradiation for the aerospace industry.

The main advantage of inverse kinematics is the natural forward collimation of the reaction ejectiles. This opens up the possibility of placing gamma detectors very close to the source without them being irradiated with source neutrons. In addition, the lack of emission at most angles means that the source is a very low background source.

For reactions which eject neutrons this will induce large enhancements of neutron fluxes at 0 degrees in the laboratory frame. The p(7Li,7Be)n reaction is one of the most commonly used in direct kinematics to produce mono energetic neutrons, especially below 0.7 MeV. However, in inverse kinematics with a Li-beam a mono-energetic neutron of 1.5 MeV is produced at bombarding energies at the reaction threshold of 13.09 MeV.

The gain from the focusing and natural collimation can be expressed in terms of neutron flux enhancement over the non-inverse reaction. Near the threshold the enhancement factor is maximal (> 100) since the emitted neutrons move with the centre of mass of the system which follows the ^{7}Li beam direction. As a consequence, close to the threshold energy, it is possible to produce very narrow (< 5 degrees) cones of neutrons. With increasing ^{7}Li bombarding energy, the cone broadens and the number of neutrons in a given solid angle decreases so the enhancement factor drops to around 20 at 16.5 MeV. However, the huge gain in intensity due to the kinematic focusing is offset by corresponding losses from two other factors. Firstly, the available beam current of ^{7}Li is much lower than that available for protons in the non-inverse reaction because of the relative difficulty of extraction of ^{7}Li-ions from the ion source. Secondly, the energy loss of ^{7}Li across a given target will be higher than that for protons due to its higher atomic number.

The current sputter source of the IPN tandem can produce ^{7}Li beam currents of up to 200 nA, but currents greater than 500 nA may be achievable with source improvements. The maximum available fluxes from LICORNE are therefore expected to around 10^{7} neutrons/second/steradian for a thin polypropylene target (4μm) and $7x10^{7}$ neutrons/second/steradian for a thick (28μm) polypropylene target. These fluxes are comparable with other accelerator-based neutrons sources, but LICORNE has the added advantages of a natural directionality and a much lower background.

The LICORNE neutron converter sits in an aluminium chamber of diameter approximately 17 cm. It is designed with a rotating polypropylene target of 4 μm thickness and a diameter of 8 cm. The rotation is necessary to increase the irradiated surface area by a factor of 25 with respect to a fixed target. Polypropylene is not very resistant to radiation damage, and therefore the rotating target prolongs the lifetime of the target by a similar factor. Even so, it has been shown that at maximum 7Li beam currents the targets loses hydrogen fairly rapidly and may need to be changed every few hours to maintain maximum neutron fluxes. Between one and 10 polypropylene targets can be stacked on the target wheel. The discs are self-supporting due to the surface tension of the polypropylene between the outer and inner zone made of epoxy, so the target wheel has no arms which have to pass through the ^{7}Li beam.

The beam current and time structure can be measured at the beam stop, which consists of a 50μm gold foil. This measurement coupled to neutron flux measurement in the experimental area can serve as an online monitoring of hydrogen content in the target and can indicate the appropriate time to change targets before significant quantities of hydrogen are lost. The exit window of the LICORNE convertor front face is made of aluminium and is only 0.3 mm thick. A mini camera, illuminating LED are included for beam-tuning and inspecting the targets from inside without having to break the vacuum of 10^{-5} bar. The beam spot is tuned by placing a retractable phosphorescent quartz target in the path of the beam to ensure that the beam spot is sufficiently diffuse (typically 8 mm diameter).

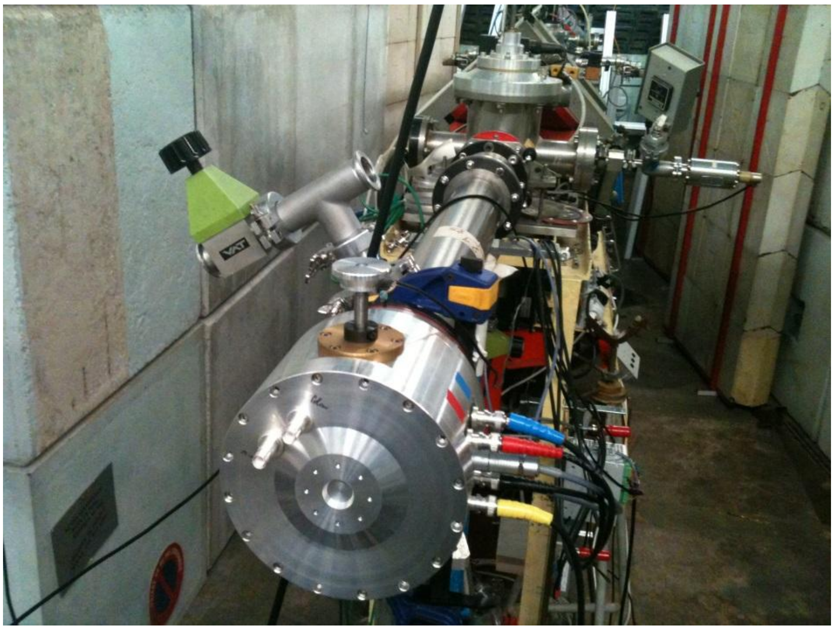

Fig. 1: The LICORNE neutron convertor

This ensures that the power density in the polypropylene is sufficiently low to keep temperature rises to only a few degrees since the polypropylene is very sensitive to heating with a melting point of 160℃. Macroscopic structural changes almost certainly occur at lower temperatures than that and it is currently an open question how much the polypropylene deforms and/or becomes thinner under a combination of surface tension, heating effects and radiation damage.

2 First experiment

A first experiment using LICORNE was conducted in July 2013 over a period of two weeks to measure prompt fission gamma ray spectra of ^{232}Th, ^{238}U, ^{235}U and ^{252}Cf. The experiment was financed by ERINDA and was split into two parts with around 100 h of beam time allocated to each part.

The first part used a cylindrical twin Frisch grid ionization chamber of 28 cm diameter and 20 cm length. The chamber was filled with P10 counting gas (90% argon, 10% methane) to detect fission fragments with an efficiency of almost 100%. Two targets of ^{235}U and ^{238}U were placed back to back at the central cathode position and signals from the cathode and anode were digitized and recorded to disk. The targets consisted of approximately 10 mg of uranium, forming circular deposits of 6.5 cm diameter on aluminium backings of 30 µm thickness. Fission fragments emitted from the surface of the targets were identified by measuring the anode and cathode signals in coincidence and placing a constraint on the minimum pulse heights recorded to reject intrinsic alpha activity.

For gamma detection 14 hexagonal BaF_2 scintillator crystals were configured into two independent clusters of seven detectors. Each crystal measured 10 cm diameter and 14 cm in length for a total mass of scintillator of 62 kg. The two clusters were placed at 29 cm from the target position at angles of +/- 62 degrees to the beam axis. In such a configuration, the total geometric efficiency of the two clusters was estimated to be ~7%. MCNP simulations of the clusters and targets show that each cluster has a high photopeak efficiency of 2.1% and a peak-to-total ratio of 75% for gamma rays of energy 1 MeV. Figure 2 shows a schematic diagram of the setup for the first part of the experiment.

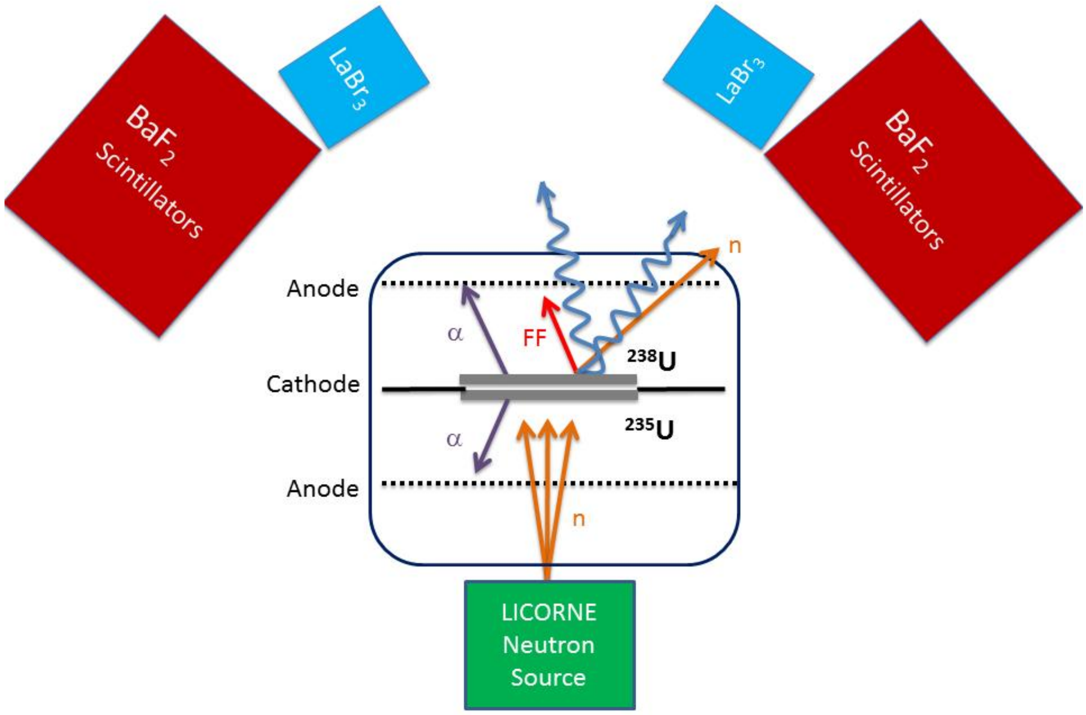

Fig. 2: Schematic diagram of the experimental setup for the first part of the experiment.

Neutrons of average energy 1.5 MeV from the LICORNE inverse kinematics neutron source were used to bombard the targets with estimated fluxes at the target position of up to 2×10^5 n/s/cm^2 . This gave maximum fission rates of around 0.3 fissions per second and 1.2 fissions per second for ^{238}U and ^{235}U targets respectively. In total 4.2×10^4 fission events of ^{238}U and 1.5×10^5 fission events for ^{235}U were recorded to disk over a period of around 3 days. Gamma rays detected in coincidence with the fission fragments were histogrammed into spectra. The analyses of the data are ongoing.

The second part of the experiment involved the same two clusters of 14 BaF_2 in a close packed geometry around thick samples of ^{238}U (38g) and ^{232}Th (50g), forming a calorimeter with a geometric efficiency of approximately 70%. The ^{238}U sample was a disc of 6.5cm diameter and the thorium sample a square of dimensions 5cm × 5cm. The cone of neutrons was produced from a ^{7}Li beam of 15 MeV bombarding energy was emitted at a maximum opening angle of 20 degrees and passed through the centre of the calorimeter to irradiate the thick samples placed at 14 cm from the neutron source. It was estimated that fission rates of ~500 and ~150 fissions per second were produced in the ^{238}U and ^{232}Th samples respectively. Neutron beams were pulsed at 2.5 MHz rate (400 ns between bunches) and bunch width of around 2 ns. This allowed timing information from the beam buncher to be used as a reference with which to measure event detection times relative to the bunch. Fission events can be discriminated from background by looking for high sum-energy and multiplicity events in the calorimeter that occurred within a short time window. The background is complex and arises from several sources: $^{7}Li+^{12}C$ fusion evaporation reactions in the polypropylene giving rise to high energy gammas and neutrons, intrinsic activity of the target itself, parasitic neutrons from the $^{7}Li+^{12}C$ reactions provoking (n,n') reactions in the scintillators, (n,n') reactions in the detectors from scattering of the primary neutron beam on the target, intrinsic activity of the scintillator crystals, and gammas from the room. Once fission events are identified, the spectrum in the $LaBr_3$ detectors in coincidence can be projected. The technique has been demonstrated to work well for a ^{252}Cf source in circumstances where the relative background is quite low. Figure 3 shows the 2D histogram of the multiplicity and sum energy of events detected in the calorimeter. The fission events are selected to the right of the black line, eliminatingbackgrounds from intrinsic activity of the BaF_2 detectors and the room.

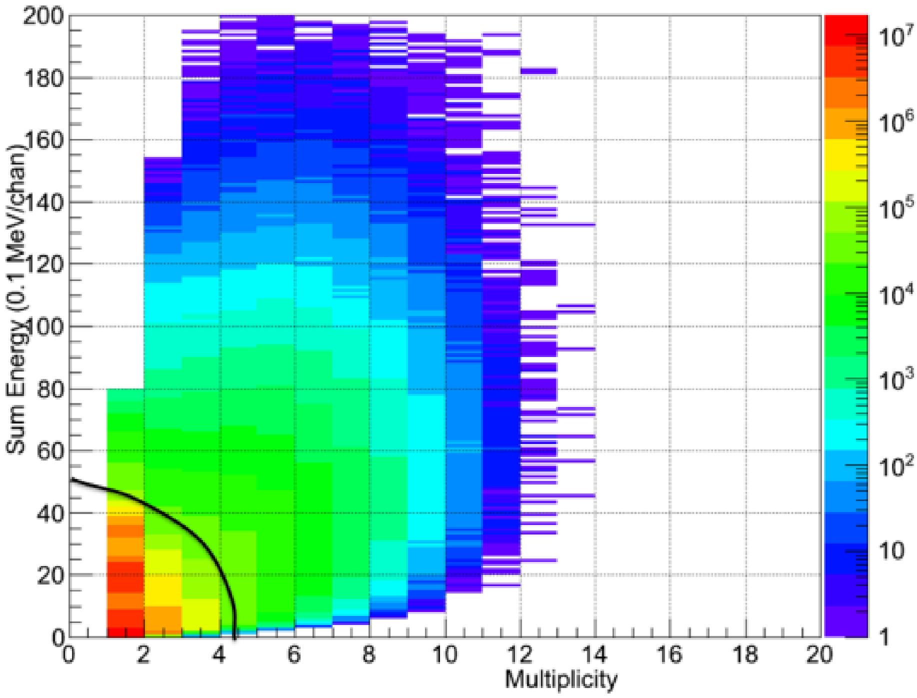

Fig. 3: Sum energy vs multiplicity of events detected in the BaF_2 calorimeter when a ^{252}Cf source is placed inside. Fission events are selected to the right of the black line.

The analysis of the in-beam data is ongoing and it remains to be seen if fission events can be discriminated from background without having to place a severe cut on the multiplicity, sum-energy and event time. Too severe selection criteria have the potential to introduce a bias in the resulting prompt fission gamma ray spectrum.

3 Conclusion

LICORNE is a dedicated facility to produce intense, naturally collimated, quasi-mono-energetic neutron beams at the IPN Orsay. The kinematic focusing of the neutron allows gamma detectors to be placed near the irradiate sample and opens up a whole host of new possibilities for the study of neutron-induced reactions, in particular nuclear fission. A first experiment, financed by ERINDA, was carried out with LICORNE in July 2013 ot measure the prompt fission gamma ray emission in fast neutron induced fission of ^{235}U, ^{238}U and ^{232}Th via two different techniques. Analysis of the in-beam data is ongoing.

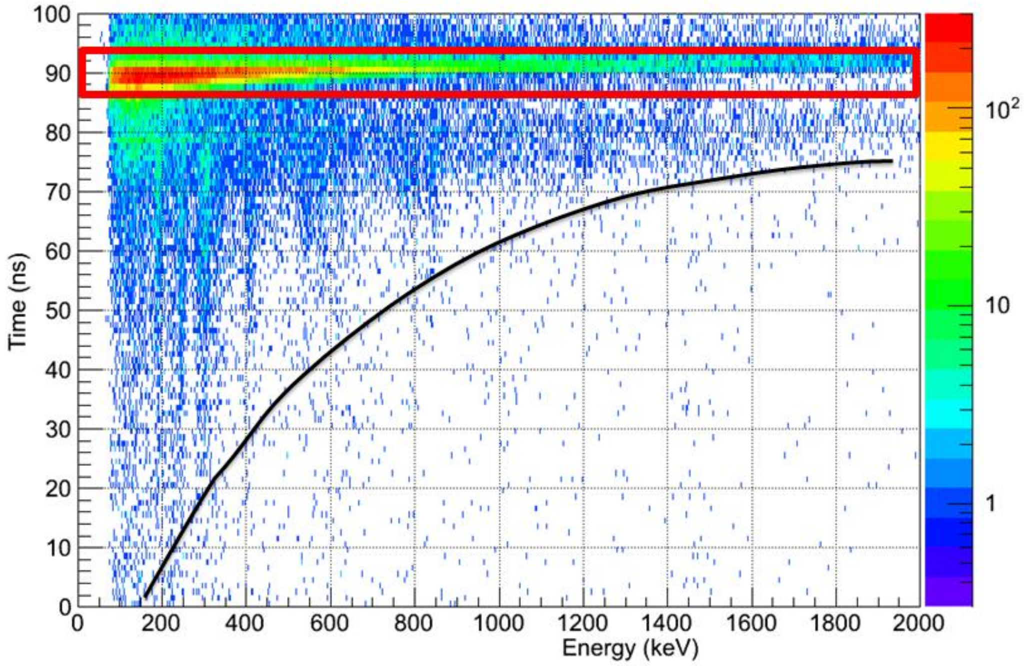

Fig. 4: Relative arrival time vs energy of gamma rays detected in the $LaBr_3$ detector in coincidence with fission events tagged in the calorimeter. Time on the y-axis goes backwards. Prompt fission gamma rays can be seen selected in the red box. The black line indicates the locus of gamma rays which can originate from (n,n') events caused by prompt fission neutrons.

References

[1] M. Lebois, J.N. Wilson et al., Development of a kinematically focused neutron source using the p(7Li,7Be)n inverse reaction. Nuc. Instr. Meth A. 735, 145 (2014)