## Neutron-rich isotopes from <sup>238</sup>U(n,f) and <sup>232</sup>Th(n,f) studied with the v-ball spectrometer coupled to the LICORNE neutron source

Jonathan Wilson<sup>1,\*</sup>, Matthieu Lebois<sup>2</sup>, and Liqiang Qi<sup>1</sup>

<sup>1</sup>Institut de Physique Nucleaire d'Orsay, CNRS-IN2P3, Université Paris–Sud, Université Paris–Saclay, 91406 Orsay Cedex, France

Abstract. We have recently successfully demonstrated a new technique for production and study of many of the most exotic neutron-rich nuclei at moderate spins. LICORNE, a newly developed directional inversekinematic fast neutron source at the ALTO facility of the IPN Orsay, was coupled to the MINIBALL  $\gamma$ -ray spectrometer to study very neutron rich nuclei using the <sup>238</sup>U(n,f) reaction. This reaction and <sup>232</sup>Th(n,f), are the most neutron-rich fission production mechanisms achievable and can be used to simultaneously populate hundreds of neutron-rich nuclei up to spins of ~16h. High selectivity in the experiment was achieved via triple  $\gamma$ ray coincidences and the use of a 400 ns period pulsed neutron beam, a technique which is unavailable to other reaction mechanisms such as spontaneous fission. The pulsing allows time correlations to be exploited to separate delayed  $\gamma$  rays from isomeric states and supresses unwanted  $\gamma$ -rays from beta decay. In Autumn 2017, the v-ball array will be operational at the ALTO facility of the IPN Orsay. This high efficiency hybrid Ge-LaBr3 spectrometer based around 24 clover Ge detectors, 10 co-axial Ge detectors and 20 LaBr3 scintillators will help to further refine the technique and achieve a large increase in the current observational limit.

## **1** Introduction

Theories of atomic nuclear stability suggest that up to ~7000 isotopes can exist in nature. However, only around 4000 of these have been successfully synthesized in the laboratory. For some of these isotopes very little is known about them other than that they exist and their half-life is measured. Often, no knowledge of excited states and their structure exists. The problem is particularly acute on the neutron-rich side of the nuclear chart, where the neutron drip line is often very far away from the edge of current knowledge. Information on excited states is important for those nuclei where none is currently available, and for those nuclei where only a handful of excited states are known, more complete information is needed on their nuclear structure in order to constrain nuclear models at the extremes of isospin and better understand the fundamental forces which hold nuclei together.

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<sup>\*</sup> Corresponding author: wilson@ipno.in2p3.fr

Very neutron-rich nuclei are fabricated in nature in extreme astrophysical environments such as supernova and hence they are the precursors of the stable nuclei in our solar system. However, the reason that we have difficulty studying them in the laboratory is that we are at the mercy of the available production mechanisms that can be used on earth.

One mechanism to populate neutron-rich nuclei is nuclear fission at low excitation energy, where the curvature of the valley of stability guarantees that when a heavy nucleus splits in two fragments they will be produced, on average, with N/Z ratios significantly larger than those of the corresponding stable isotopes for these elements. However, not all fission reactions are equivalent. Figure 1 shows a comparison of various fission reactions which have been used to populate neutron-rich and subsequently study the prompt decay of the excited fission fragments via  $\gamma$ -ray spectroscopy. During the 1990's and 2000's the spectroscopy of prompt decay of neutron-rich fission fragments was studied extensively using <sup>252</sup>Cf and <sup>248</sup>Cm spontaneous fission (SF) sources and the Euroball and Gammasphere arrays[1][2].



Fig. 1. Different fission reactions used to study the prompt  $\gamma$ -decay of neutron-rich nuclei. The spontaneous fission sources are circled in green. Nuclei fissioned with thermal neutrons during the EXILL campaign are circled in blue. The new reaction mechanisms currently only available at the IPN Orsay with LICORNE/v-ball are circled in red.

More recently, the EXILL campaign, where the Exogam spectrometer was moved from GANIL to the ILL reactor in Grenoble, studied thermal neutron induced fission of <sup>235</sup>U and <sup>241</sup>Pu during a campaign lasting several months[3][4]. At the ALTO facility of the IPN Orsay in 2015, we successfully demonstrated a new technique to study prompt and delayed  $\gamma$ -decay of fragments from the most neutron-rich fission reactions available <sup>238</sup>U(n,f) by coupling the LICORNE directional neutron source [5][6] with the high efficiency MINIBALL spectrometer [7][8][9]. The LICORNE fast neutron beam can be pulsed with a 400 ns period, allowing easy access to time correlations (clean detection of delayed  $\gamma$ -rays from isomers) and suppression of random events from beta-decay and other unwanted

processes. This allows an additional selectivity that is unavailable to previously-used techniques (SF and thermal neutron induced fission).

In autumn 2017 we intend to build on the valuable experience gained during this experiment and push the technique to its limits in order to study nuclei that cannot be populated and studied at medium/high spin by any other reaction mechanism. To do this we are constructing the v-ball spectrometer to which we will couple to the LICORNE source. v-ball. A first experiment will attempt to perform spectroscopy above and below the fission isomer in <sup>238</sup>U via (n,n' $\gamma$ ), which is the reaction mechanism with the highest isomer population cross section (> 0.1 mb for 3-4 MeV neutrons). The main v-ball/LICORNE experiment will address a wide range of physics in neutron-rich nuclei and is a collection of 16 sub-proposals from various nuclear physics groups around the world. There at least 4 major themes: Spectroscopy above <sup>78</sup>Ni and search for neutron radioactivity; shape co-existence around N=60; the study of neutron-rich nuclei around <sup>132</sup>Sn and the onset of deformation in Xe, Ba, Ce and Nd isotopes.



**Fig. 2.** Simulated spectrometer efficiency curves for three 30 g  $^{238}$ U metal samples with different densities. The lowest curve demonstrates the efficiency losses from self-shielding if a compact metal sample is used similar to the one used in the first MINIBALL/LICORNE experiment.

## 2 Experimental Details

The LICORNE neutron source uses the high intensity <sup>7</sup>Li beams available from the tandem accelerator of ALTO to perform neutron production in inverse kinematics by bombarding a hydrogen gas cell. The LICORNE source will be placed close to the centre of the v-ball array and used to bombard massive samples (around 100 g) of <sup>238</sup>U and <sup>232</sup>Th. Simulations show that fission rates of around 75 kHz and 15 kHz for the respective samples should be reasonably easy to achieve with corresponding primary <sup>7</sup>Li beam intensities of 50 nA. The actinide sample densities are decreased from 19.1 g/cm<sup>3</sup> and 11.5 g/cm<sup>3</sup> for <sup>238</sup>U and <sup>232</sup>Th respectively to between 0.5 g/cm<sup>3</sup> and 1 g/cm<sup>3</sup>. This is achieved by filling an aluminium

casing with uranium metal turnings in the first case, and encapsulating a stack of eight 1mm thick thorium disks in the second case. It can be shown that the reduced effective thickness,  $x_I$ , of material for gamma rays to traverse, for a sample of the same mass, is in proportion to a power of the ratio of new and old densities:

$$x_1 / x_0 = (\rho_1 / \rho_0)^{2/3} \tag{1}$$

So in our case, reducing the average density from 19.1 g/cm<sup>3</sup> to 0.5 g/cm<sup>3</sup> correspondingly reduces the effective thickness of uranium metal for gamma rays to pass through by a factor of 11.3 and hence dramatically reduces the self-shielding and increases the effective spectrometer efficiency (see figure 2), particularly for lower gamma-ray energies.

The actinide sample can be extended in space (up to  $\sim 10$  cm in length and diameter) since the fragment velocity is zero for gamma-rays emitted after fragments stop (picoseconds), so no Doppler correction is required and hence the reaction does not have to be confined to a point-like zone in the centre of the array.

The pulsed <sup>7</sup>Li primary beam will have ~17 MeV of incident energy and a time structure of 400 ns period and 1.5 ns pulse width giving access to decays from isomeric states in the 0.5 ns  $-1 \mu s$  range.



**Fig. 3.** Drawing of the setup with LICORNE and v-ball spectrometer using the fast directional neutrons to bombard <sup>238</sup>U or <sup>232</sup>Th samples with an average energy 2 MeV. The Ge clover and coaxial detectors (centre and left) have anti-Compton shields and are employed with LaBr<sub>3</sub> scintillators at forward angles (right) to cover around 70% of the total solid angle.

The v-ball array itself (see figure 3) consists of 24 clover Ge detectors and their anti-Compton shields at ninety degrees, currently on loan at the IPN Orsay from the Gammapool collaboration, 10 co-axial Ge detectors and their BGO shields and 20 LaBr3 detectors on loan from the FATIMA collaboration. The array has 590 individual crystals and 184 channels of digital electronics. The anti-Compton shields will be run without collimators to allow event-by-event measurement of sum energy and multiplicity. This, along with timing information relative to the beam pulse, will be used to uniquely identify fission events via gamma-ray detection alone. It is expected that uncorrelated events from beta decay will be heavily suppressed by a factor of perhaps 400 or more). A major increase in resolving power is expected over the previous setup with MINIBALL. The resolving power of the array, R.P., is proportional to the peak-to-total, PT, to the power of the event fold, F.

$$R.P. \alpha \left( PT/\Delta E \right)^F \tag{2}$$

For the case of v-ball, the peak-to-total ratio at 1.3 MeV is expected to be  $\sim 0.5$ , improving on that of the previous setup with MINIBALL  $\sim 0.2$ , with a similar photo-peak efficiency and energy resolution. The resolving power is thus expected to improve by a factor of 15 for triple coincidence events. This increase coupled to the reduction in self-shielding of the samples, event calorimetry, better array timing properties, increased fission rates and longer running times ought to produce very high quality data on the decay of very neutron-rich fission fragments.

Previous experience with the safe coupling of LICORNE/MINIBALL for 11 days effective beam time showed no detectable neutron damage to the Ge detectors after the experiment, thus corroborating what we measured online, and our prior neutron scattering simulations. Simulations and experimental experience agree that no observable neutron damage is expected during the v-ball campaign, provided metal samples are used.  $UO_2$  and  $ThO_2$  oxide powder samples were simulated but were found to increase neutron scattering from the sample into detectors at 90 degrees by a factor of around 3 over metal samples of similar mass.

The success of the v-ball experimental campaign depends on gathering high-statistics data sets for analysis of double  $\gamma$ - $\gamma$  and triple  $\gamma$ - $\gamma$ - $\gamma$  coincidences. It is estimated that in 6 weeks of beam time at ALTO we can acquire a data set of a similar size to that obtained during the EXILL campaign (> 10<sup>10</sup>) triple  $\gamma$ -ray or higher fold coincidences. This is possible thanks to the very high fluxes available (> 10<sup>7</sup> n/cm2/s) a few centimetres from the end of the LICORNE gas cell (see figure 4).

The <sup>232</sup>Th(n,f) and <sup>238</sup>U(n,f) reactions will be used to preferentially populate different regions in mass. A particular advantage of the  $^{232}$ Th(n,f) reaction is the possibility to study at high/medium spins of neutron-rich nuclei in the Zn, Ga and Ge isotopes which are very difficult to populate via other means. For this reaction there will also be a synergy with the radioactive beams part of the ALTO facility since studies in this region via beta-decay is an active part of current research. The advantage of the <sup>238</sup>U(n,f) reaction is the high achievable fission rate (~75 kHz), some 5 times greater than is possible with <sup>232</sup>Th(n,f) due to its higher cross section. However, it is important to study both reactions simultaneously because of the synergy from complementary data sets. For some nuclei where no information is currently known, new  $\gamma$ -rays will have to be assigned on the basis of coincidences with known binary partners and intensity arguments compared to other isotopes in the chain. The ability to double check the presence of the new  $\gamma$ -rays with a different binary partner from the other reaction is extremely important for making solid isotope assignments. In the past, there are examples where newly observed  $\gamma$ -rays from a single SF source have subsequently found to be assigned to the wrong isotope [10][11] underlining that cross checking from a different data set is essential.



**Fig. 4.** Distribution of the LICORNE neutron flux in the z,y plane simulated using GEANT IV. A primary beam current of 100 nA, a gas cell length of 3.5cm length at 1.5 atmosphere pressure and a beam energy of 15 MeV after the LICORNE gas cell entrance window are assumed. Flux is given in units of neutrons/cm<sup>2</sup>/s.

For the EXILL campaign, the complementary study of two different fissile systems ( $^{235}$ U and  $^{241}$ Pu) was an important synergy. We fully expect this to be the case for the v-ball campaign involving  $^{232}$ Th(n,f) and  $^{238}$ U(n,f).

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