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Prompt emission in fission induced with fast neutrons

J.N. Wilson[a,*], M. Lebois[a], P. Halipré[a], S. Oberstedt[b], A. Oberstedt[c]

[a]*Institut de Physique Nucléaire d'Orsay, Bat. 100, 15 rue G. Clemenceau, 91406 Orsay cedex, France*
[b]*European Commission, Joint Research Centre (IRMM) , 2440 Geel, Belgium*
[c]*Fundamental Fysik, Chalmers Tekniska Högskola, 41296 Göteborg, Sweden*

Prompt gamma-ray and neutron emission data in fission integrates a large amount of information on the fission process and can shed light on the partition of energy. Measured emission spectra, average energies and multiplicities also provide important information for energy applications. While current reactors mostly use thermal neutron spectra, the future reactors of Generation IV will use fast neutron spectra for which little experimental prompt emission data exist.

Initial investigations on prompt emission in fast neutron induced fission have recently been carried out at the LICORNE facility at the IPN Orsay, which exploits inverse reactions to produce naturally collimated, intense beams of neutrons. We report on first results with LICORNE to measure prompt fission gamma-ray spectra, average energies and multiplicities for ^{235}U and ^{238}U. Current improvements and upgrades being carried out on the LICORNE facility will also be described, including the development of a H_2 gas target to reduce parasitic backgrounds and increase intensities, and the deployment of ^{11}B beams to extend the effective LICORNE neutron energy range up to 12 MeV. Prospects for future experimental studies of prompt gamma-ray and neutron emission in fast neutron induced fission will be presented.

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* Corresponding author. Tel.: +33-1-6915-7980;
E-mail address: Wilson@ipno.in2p3.fr

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1. Introduction

The prompt gamma and neutron decay of newly formed fission fragments occurs in the first nanosecond after scission and accounts for around 7% of the total energy released in fission. The spectral shape, average multiplicity and energy of prompt gamma ray emission in fission are important observables in the fission process which state-of-the-art theoretical modeling attempts to reproduce (Litaize et al., 2010; Schmidt et al., 2010). Measurements can shed light on the generation of angular momentum, the energy sharing between fission fragments and the competition between neutron and gamma emission in the fission process. Furthermore, these emission spectra are the source terms for a large part of the gamma heat released in a reactor core, where gamma rays can transport far from the site of its production into non-fissile reactor core components (i.e. structural materials, blankets, instrumentation, etc.)

The first measurements of prompt gamma emission were made about 40 years ago for only two fissile nuclei using thermal neutron induced fission (Verbinski et al., 1973; Pleasonton, 1973). However, over the last three years there has been a renewed interest in the fission prompt gamma emission (Billnert et al., 2013; Oberstedt et al. 2013; Chyzh et al., 2013; Ullman et al, 2013). Integral experiments at the EOLE experimental reactor in Cadarache and other experiments demonstrated that there were observed discrepancies of up to 30% in gamma heating phenomena due to deficiencies in the spectral data (Luthi et al., 2001; Rimpault et al., 2012). The advent of new Lanthanide-Halide scintillation materials such as $LaBr_3$ has opened up the possibility for a completely new kind of experimental investigation where high resolution gamma spectroscopy is possible while exploiting the sub nanosecond timing resolution of these crystals to discriminate the emitted prompt gamma rays from prompt neutrons via time of flight at short distances. The scintillator crystal thus becomes simultaneously a gamma ray and a neutron detector giving access to information on both the prompt gamma and prompt neutron decay of the fission fragments.

Since many of the concepts for the next generation of nuclear reactors will use fast neutron spectra it is clear that there are implications for the safety of innovative reactors of the future. Gamma rays are the principal energy transfer mechanism between a reactor core and its surrounding environment (reflectors, blankets, etc.). These exterior components also require their own cooling circuits to maintain safe operation, yet the emitted energy is poorly quantified since the in-core gamma spectra are poorly known. Currently, almost no prompt gamma emission data exist for fast neutron induced fission. This situation exists because it is very difficult to produce high fluxes of fast neutrons with a corresponding directionality to the beam. In addition, the fission cross sections for fast neutrons are typically three orders of magnitude lower than those for thermal neutrons. An additional experimental problem is the discrimination between prompt gamma rays and prompt neutrons which are effectively emitted at the same time. The method best adapted to achieve this is the time-of-flight technique. However, to get good separation between PFGS and PFNS requires a detector which is far away and thus efficiency is necessarily low. The intrinsic time resolution of the gamma detector to be used must be extremely good to minimize the distance and maximize the efficiency for a given degree of PFN and PFG separation.

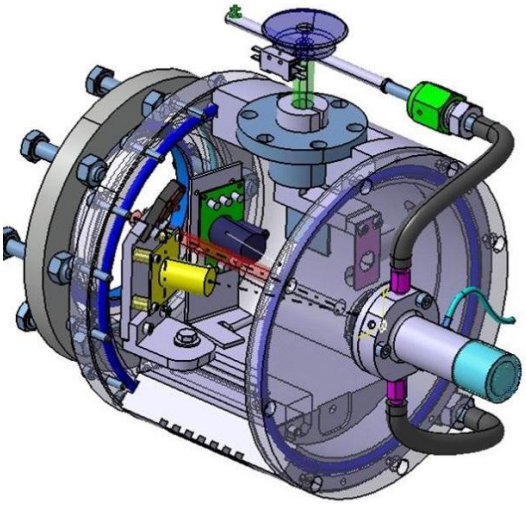

Fig. 1. (Color online) The design of the new LICORNE convertor, including optical and infrared cameras for monitoring the focussing of the beam on the thin tantalum window separating the vacuum from the gas cell.

2. The LICORNE neutron source

At the IPN Orsay we have recently developed LICORNE (Lithium Inverse Cinematiques ORsay NEutron source), a unique, directional neutron source which is based on fast neutron production using the p(^{7}Li,n) reaction inverse kinematics (Wilson et al., 2013; Lebois et al., 2014). Hydrogen targets are bombarded with a ^{7}Li beam to produce high fluxes of fast neutrons which are emitted in narrow cones. These neutrons can be used to induce fissions in different actinide samples very close to the neutron source. The innovative aspect of LICORNE is the directionality of the emitted neutrons which allows placement of gamma detectors around the irradiated sample, something which is impossible for standard isotropic neutron sources which would blind the gamma detectors with source neutrons. The fluxes available at LICORNE are very high (up to 10^7 n/s/cm^2), since like other accelerator-based sources samples can be placed very close (a few cm) to the source. However, the natural directionality of LICORNE is a key feature which is missing from other conventional sources and has opened up the possibility to perform detailed studies of prompt emission in fission.

The first experiment carried out with LICORNE was to make a comparative measurement of the PFGS emitted from ^{235}U and ^{238}U for fast neutrons produced by LICORNE with an average energy of 1.7 MeV. These spectroscopic samples of around 10 mg of each isotope were placed back-to-back at the central cathode of an ionization chamber which served as the start signal. PFGs from fission events were detected in an array of gamma detectors comprising 14 large volume hexagonal BaF_2 crystals (10 cm × 14 cm) and three cylindrical $LaBr_3$ scintillators (5 cm × 5 cm).

Fig. 2. (Color online) Left: The new LICORNE neutron source convertor; Right: The hydrogen gas pressure and flow control system.

3. Energy partition in fission

The energy budget in fission is shared between the total kinetic energy (TKE) of the fragments, the prompt fission neutrons, the prompt fission gamma rays and the subsequent delayed gammas, betas and anti-neutrinos. If the energy of the incident neutron increases, more energy becomes available to the excited compound nucleus and the energy partition in fission will change. Experiments have shown that the fragment TKE of typically 150 MeV is very insensitive to the incident neutron energy, and only varies by as little as 0.5 MeV on average over for incident neutrons in the range from 0 to 9 MeV (Meadows et al., 1978). However, the mass distribution of the fragments becomes more and more symmetric as more energy becomes available (Nagy et al., 1982). The extra energy must go somewhere, and clearly a large portion goes to the prompt neutrons since neutron multiplicities are measured to increase fairly linearly with incident neutron energy. However, the effects of changing the incident neutron energy on the PFG spectra are not yet measured and almost no data exists. The PFG spectra will change in shape due to two principal effects: Firstly, since fission becomes more symmetric the yields will change and a different set of nuclei will be populated. Secondly, with higher incident neutron energy, extra angular momentum will be brought into the compound system which will find its way into the fragments. After the emission of neutrons becomes energetically unfavorable, the excited fission fragments will decay via the emission of several gamma rays. The resulting multiplicity of gamma rays will be highly correlated with the fragment angular momentum since the prompt neutrons take away hardly any angular momentum when they are emitted.

It would therefore be particular interesting to measure the evolution of the spectral shape, mean energy and multiplicity of the PFGs as a function of the incident neutron energy, since this would be a probe of the role of angular momentum in the fission process. Such measurements over a wide range in energy should be possible using LICORNE with ^{11}B beams which will extend the range of energies produced up to 12 MeV (see section 5).

Fig. 3 (Color online) The exchangeable gas cells of different lengths (7.5cm, 3.5cm and 2cm).

4. The LICORNE hydrogen gas target

The first version of the LICORNE inverse kinematics source used rotating polypropylene disks as the hydrogen target (Wilson et al., 2013; Lebois et al., 2014). This had the advantage of fine control over the thickness of the target and hence the number of hydrogen atoms in the path of the ^{7}Li beam. Thickness could be varied between 4μm and 100μm. However, the major drawback was the production of unwanted parasitic neutrons from fusion evaporation reactions of ^{7}Li with the ^{12}C in the polypropylene polymer. In addition, the radiation damage of the polypropylene at the highest intensities caused the rapid loss of hydrogen from the polypropylene and targets needed to be changed every few hours which required a break of the vacuum. The presence of secondary unwanted neutrons is a particular problem if Germanium detectors are to be used with LICORNE, since these are sensitive to neutron damage and they will be irradiated with the secondary neutron field.

To solve these problems, a new gas target system for LICORNE has been developed. The goal is to suppress all secondary reactions via the use of high Z materials in the construction of the collimator; thin entrance window and beam stop which are made of tungsten, tantalum and lead respectively. In early 2015 LICORNE will be coupled with the MINIBALL spectrometer which is currently on loan at the IPN from the ISOLDE facility at CERN.

To achieve fine control over the number of hydrogen atoms in the path of the beam with the gas target, four aluminium cells (0.5 mm thick) have been made of lengths 2, 3.5, 5.5 and 7.5 cm respectively. The pressure of the hydrogen gas in the cells can be varied between 1 and 2 atmospheres, giving and additional flexibility to control the shape of the resulting neutron spectrum. The number of hydrogen atoms per cm^2 in the target can thus be varied very precisely over almost an order of magnitude between 1.0×10^{20} and 8.0×10^{20}. The Tantalum entrance window is 5mm in diameter and between 2μm and 4μm thick and is designed to resist in the most extreme range of pressure and beam current (2 atm. and 100 nA). To prevent air (and oxygen) entering the gas cell, a flow of hydrogen through the gas control system is maintained via a flow meter and needle valve. The flow is very low and typically 30cm^3 per minute. The hydrogen is evacuated outside the accelerator buildings to prevent potentially explosive build-ups of gas. In the event of a rupture of the cell window, two safety valves will close rapidly when sensors detect any minor changes in pressure and the flow of hydrogen from the gas bottle is cut off.

The neutron fluxes with the hydrogen gas cell LICORNE convertor are higher than those available with the rotating polypropylene disc system. The reason is that the stopping power per unit loss of ^{7}Li energy for hydrogen is less than that for polypropylene, so more hydrogen atoms can be placed in the path of the beam for a given energy loss. Figure 4 shows calculations for achievable fluxes at varying distances from the front face of the cell (0

cm) as a function of the incident beam energy of the ^{7}Li. As the energy becomes nearer the threshold, the neutron cone angle becomes smaller and the diminution of the flux becomes less severe as a function of distance.

5. Extension of the energy range with heavier beams such as ^{11}B

The principle of LICORNE, using inverse kinematics is not just restricted to beams of ^{7}Li. Indeed any reaction on a hydrogen target with a heavy ion beam which has a favourable Q-value for production of neutrons will work. The next most obvious reaction after ^{7}Li for directional inverse kinematic neutron production is p(11B,n). Since the first excited state of ^{11}B is much higher than that of 7Li (2.2 MeV compared to 0.48 MeV), correspondingly higher energies per nucleon, and a greater velocity of the centre of mass frame can be achieved without ejecting the 11C recoil in its first excited state. This implies that the range of LICORNE neutron energies can be extended to as high as 12 MeV. However, the energy of the slower neutron group will vary correspondingly between E_n = 2 MeV for 33 MeV ^{11}B at the reaction threshold and E_n = 500 keV for 58 MeV ^{11}B, which produces the highest energy (12 MeV) neutrons. If PFG and PFN spectra are to be measured with LICORNE using ^{11}B beams then pulsation of the beam is necessary to discriminate between the low and high energy neutron groups via the double time-of-flight technique. This should be relatively easy for samples at very short distances (~ 10 cm) given that the time resolution of the pulsed beam is around 2 ns and there is a typical TOF difference between high and low energy groups of 4-8 ns.

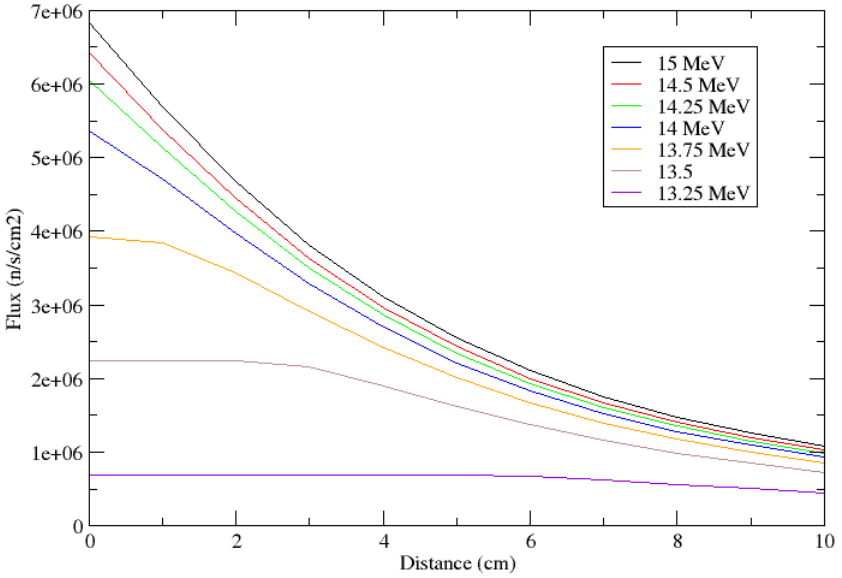

Fig. 4 Available neutron fluxes at zero degrees as a function of the distance of the gas cell

6. Conclusion

The LICORNE neutron source is a high-flux, naturally directional neutron source based on the p(7Li,n) inverse reaction and is ideal for measurements of the characteristics of prompt gamma and neutron emission in fission. Future experiments are planned to measure prompt gamma and neutron emission characteristics for various actinide nuclei over a range of incident neutron energies

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