

Prompt fission gamma-ray emission spectral data for $^{239}\text{Pu}(n,f)$ using fast directional neutrons from the LICORNE neutron source

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Abstract. Prompt fission gamma-ray spectra (PFGS) have been measured for the $^{239}\text{Pu}(n,f)$ reaction using fast neutrons at $\bar{E}_n=1.81$ MeV produced by the LICORNE directional neutron source. The setup makes use of LaBr_3 scintillation detectors and PARIS phoswich detectors to measure the emitted prompt fission gamma rays (PFG). The mean multiplicity, average total energy release per fission and average energy of photons are extracted from the unfolded PFGS. These new measurements provide complementary information to other recent work on thermal neutron induced fission of ^{239}Pu and spontaneous fission of ^{252}Cf .

1 Introduction

One of the key isotopes for present and future energy applications is the fissile nucleus ^{239}Pu . As such it occurs frequently in the NEA Nuclear Data High Priority Request List (HPRL). In conventional Light Water Reactors (LWR), up to one third of the energy released is from the fission of ^{239}Pu . ^{239}Pu is produced through neutron capture reaction on ^{238}U , followed by two beta decays. For many of the Generation IV reactors or current LWR using MOX fuel, fission of plutonium dominates the energy production.

Gamma-ray emission in nuclear fission is one of the least known fission observables and for this reason measurement requests of PFGS for ^{239}Pu in the thermal and fast neutron region are in the NEA

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Nuclear Data HPRL. Recent results on PFG emission in thermal neutron induced fission of ^{239}Pu have been published in Ref [1]. Recent reactor experiments indicate heating effects from gamma-rays are underestimated up to 28% [2] when compared with the simulations based on the currently available experimental data.

Here we report on the PFG measurement for fission of ^{239}Pu induced by fast neutrons at an energy very relevant for future Gen IV fast reactors ($\bar{E}_n=1.8$ MeV). An experiment was performed at the ALTO facility at the IPN Orsay, using the LICORNE neutron source [3]. Results from the experiment will provide important nuclear data for reactor physics, e.g. as an input for gamma heating calculations and for a better understanding of gamma emission in the fission process.

2 LICORNE

The LICORNE neutron source generates forward focused neutron cones by exploiting neutron-producing reactions in inverse kinematics. The major advantage is that detectors can be placed around the sample to be irradiated without the need for shielding from neutrons emitted by the source. A further consequence is a very low thermal neutron background in the experimental room. This feature of LICORNE makes the study of fissile isotopes in the fast neutron region possible. In the experiment reported here, PFGS characteristics of ^{239}Pu at an incident neutron energy of 1.81 ± 0.52 MeV were studied.

In order to estimate the neutron flux in the space after the LICORNE gas cell, a GEANT4 [4] code was developed to simulate the physical processes in LICORNE (see Fig. 1). The program incorporates user-defined physical processes, which are $p(^7\text{Li},n)^7\text{Be}$ and $p(^{11}\text{B},n)^{11}\text{C}$ reactions, into the standard physics list provided by GEANT4. They are based on relativistic kinematic calculations and consider the experimental differential cross section data from literature [5, 6]. Since the cross sections of these reactions are low (less than 1 barn), event biasing techniques are used to have more sampling in the region of interest to decrease the calculation time. There are mainly two categories of event biasing techniques, importance biasing [7] and multi-level splitting. Importance biasing is to increase the cross section of the process of interest. Multi-level splitting includes force collision (widely used in MCNP) and increasing the final neutron yields (encouraged by the GEANT4 example: bremsstrahlung splitting). All these un-physical techniques will need a weight to be corrected. They were implemented in the G4WrapperProcess without changing the tool-kit itself. Analogue simulation is also compared to the biased simulation. The final results agree with each other while the Figure-Of-Merit (FOM) has been improved more than 20 times with the event biasing techniques. The final strategy is to make full use of these two techniques, which is to increase the cross section of the reaction of interest and at the same time increase neutron yields once the reaction is invoked. This code has been validated by several experiments, e.g. the neutron time-of-flight (TOF) spectra measurement with liquid scintillation detectors from the EDEN array [8].

3 Experimental methods

3.1 Fission events tagging

A fission chamber, developed at JRC-Geel, was used to discriminate α emissions and fission fragments (FF) of the fissile samples. The chamber contained high purity (99.97%) ^{239}Pu samples of 3.519 mg in a compact geometry. The counting gas was pure Tetrafluoromethane (CF_4). Due to high activity of the sample (7 MBq), Pulse Shape Discrimination (PSD) was applied to have a better discrimination (see Fig. 2) by eliminating α pile-up events.

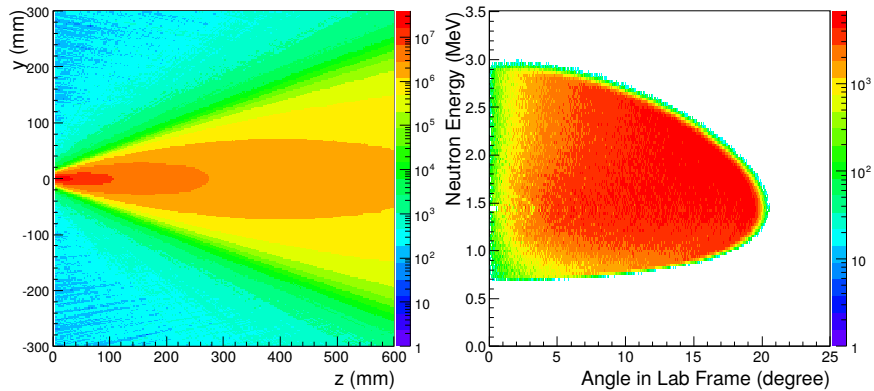


Figure 1. (a) Neutron flux distribution, normalized to the beam intensity 100 nA. The total neutron flux is of the order of 10^{6-7} n/(cm² s) a few centimeters after the LICORNE gas cell; (b) Kinematic curve correlating the outgoing neutron energy and the angle in the laboratory frame for the $p(^7\text{Li},n)^7\text{Be}$ reaction.

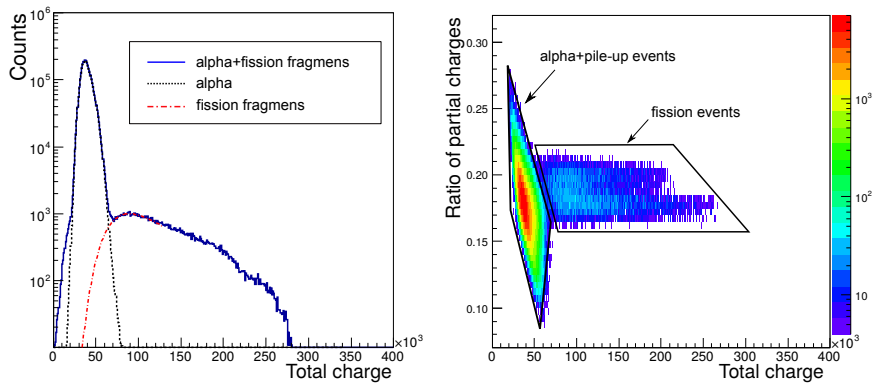


Figure 2. (a) Total integrated charge spectrum, separating α particles and FF; (b) Correlation between the ratio of partial charges and the total integrated charge. PSD improves the discrimination between α particles and FF.

3.2 PFG measurements

The prompt gamma rays were measured, in coincidence with the fission fragments tagged in the ionization chamber, by scintillation detectors: 6 individual LaBr₃(Ce) detectors with two sizes (50.8 mm × 50.8 mm and 76.2 mm × 76.2 mm in diameter and length) and a cluster of 9 phoswiches detectors from the PARIS collaboration [9]. LaBr₃ detectors were selected due to their state-of-the-art energy and time resolution. Detection efficiency for high energetic gamma-rays is favourable in terms of the PARIS phoswiches. The calibration was performed by using some conventional gamma-ray sources, including ¹³⁷Cs, ⁶⁰Co, ¹⁵²Eu and thorium series radionuclides. In addition, a practical and convenient source of 9 MeV gamma rays (see Fig. 3) [10] was used. It is able to check the linearity

of the detectors and the dependence of resolution versus energy, i.e. $(\frac{dE}{E}) = \sqrt{\alpha^2 + \frac{\beta^2}{E} + (\frac{\gamma}{E})^2}$ [11], up to 9 MeV. It also provides an opportunity to validate the simulation of the experimental setup in the high-energy region.

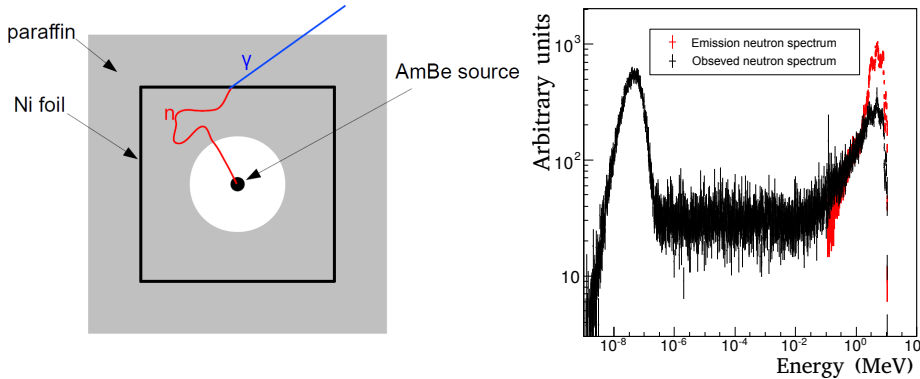


Figure 3. (a) Schematic of the 9 MeV gamma-ray source, consisting of an AmBe source, paraffin and nickel foils; (b) Emission neutron spectrum from the AmBe source and observed neutron spectrum in the nickel foils. The fast neutrons from the AmBe source are thermalized by the surrounding paraffin, and then captured in the nickel foils.

3.3 Deconvolution

Figure 4 represents the matrix of TOF between the fission chamber and the gamma detector versus the observed gamma-ray energies. The first structure in the zero time reference corresponds to the PFG and the second structure corresponds to the prompt neutron response. The prompt neutrons are well separated from the prompt gamma rays. The time resolution, expressed in terms of the full width half maximum (FWHM), of the prompt gamma-ray peak is 2 ns. The measured PFGS is extracted with the time window set to be $[-3 \text{ ns}, +3 \text{ ns}]$. This time window was chosen due to the convenience in comparison with another experiment [1], which measured the PFGS characteristics with thermal neutrons.

Since gamma rays have a complicated response in a single detector, the measured PFGS is folded by the response function. The aim is to unfold the measured PFGS with the simulated response matrix in GEANT4. The unfolding algorithm is by linear iteration [12]. The comparison of different unfolding techniques has been presented in another work [8]. The unfolding process highly depends on the simulated response matrix. In order to validate the response matrix, the efficiency of each gamma detector has to be verified with the calibration results in the range between 100 keV and 9 MeV. In the low energy region, the gamma spectrometer is very sensitive to the threshold effect, including electronic threshold and physical threshold. Preliminary results from one LaBr_3 have been extracted, with values for the mean multiplicity of $\bar{M}_\gamma = 7.71 \pm 0.09$, average total energy release per fission $E_{\gamma, \text{tot}} = 6.67 \pm 0.05 \text{ MeV}$ and average energy of photons $\epsilon_\gamma = 0.87 \pm 0.01 \text{ MeV}$. A full analysis of the data is ongoing and final results will be published as a journal article in the near future, so these numbers are liable to change.

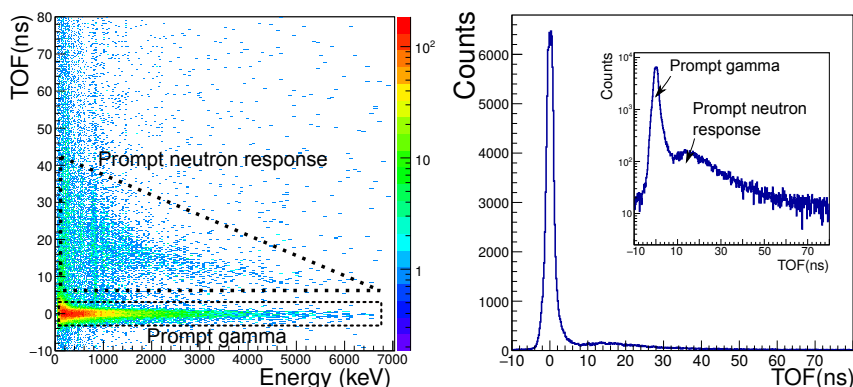


Figure 4. (a) Correlation between the gamma-ray energy detected in one gamma detector and the TOF, where the ionization chamber gives the start signal and the gamma detector gives the stop signal; (b) TOF spectra in linear scale; the inset shows the same distribution in logarithmic scale.

4 Conclusions

PFGS have been measured for the $^{239}\text{Pu}(n,f)$ reaction using fast neutrons produced by the LICORNE directional neutron source. Preliminary results from one LaBr_3 have been extracted, including the mean multiplicity, average total energy release per fission and average energy of photons. These results provide complementary information to recent thermal neutron induced fission and spontaneous fission measurements. Further analysis concerning all the detectors is still ongoing.

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