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Neutron angular distribution in plutonium-240 spontaneous fission

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ABSTRACT

Nuclear safeguards applications require accurate fission models that exhibit prompt neutron anisotropy. In the laboratory reference frame, an anisotropic neutron angular distribution is observed because prompt fission neutrons carry momentum from fully accelerated fission fragments. A liquid organic scintillation detector array was used with pulse shape discrimination techniques to produce neutron-neutron cross-correlation time distributions and angular distributions from spontaneous fission in a ^{252}Cf, a 0.84 g ^{240}Pu$_{eff}$ metal, and a 1.63 g ^{240}Pu$_{eff}$ metal sample. The effect of cross-talk, estimated with MCNPX-PoliMi simulations, is removed from neutron-neutron coincidences as a function of the angle between detector pairs. Fewer coincidences were observed at detector angles near 90°, relative to higher and lower detector angles. As light output threshold increases, the observed anisotropy increases due to spectral effects arising from fission fragment momentum transfer to emitted neutrons. Stronger anisotropy was observed in Cf-252 spontaneous fission prompt neutrons than in Pu-240 neutrons.

1. Introduction

Plutonium is a special nuclear material that can be used in the core of a nuclear weapon. The odd-numbered isotopes of plutonium, in particular Pu-239, are used to sustain a fission-chain reaction. However, bulk plutonium always contains some percentage of Pu-240, which has a relatively high spontaneous fission rate. This feature makes plutonium detectable and quantifiable by passive means, and more specifically by counting neutron doubles [1]. In fact, fission generates multiple prompt neutrons emitted in coincidence. Over 70% of Pu-240 spontaneous fissions emit more than one neutron [2].

Detection and characterization of special nuclear material often relies on knowledge of fission neutron anisotropy [3–6]. In fission, an anisotropic neutron angular distribution is observed in the laboratory reference frame because neutrons emitted isotropically in the fission fragment frame of reference carry momentum from the fully accelerated fission fragment [6,7]. Current nonproliferation and verification neutron measurement systems rely on detecting thermalized neutrons; the neutron properties at the time of emission are obscured by down-scattering. When fast neutron detectors are used, the neutron–neutron correlation from fission can be used to characterize fissile samples. Neutron–neutron correlation can distinguish metal from oxide and can estimate the fission to (α, n) rate [8]. In metals, fission neutrons are dominant, while in oxides, fission and (α, n) neutrons are both present. Fission neutrons are emitted anisotropically; (α, n) reactions, present in oxides, emit single neutrons, therefore neutron–neutron coincidences from (α, n) reactions are observed isotropically.

Many fission neutron angular correlation experiments have quantified anisotropy in fission neutron emission [4,9,10]; no published work exists, however, that investigates Pu-240 spontaneous fission neutrons. Previous experiments characterize neutron correlations in Pu-239 thermal neutron induced fission, but conclusions from this data are not directly applicable to Pu-240 spontaneous fission [11].

Experimental results are presented here on correlated Pu-240 spontaneous fission prompt neutrons, expanding on work performed by Dolan and colleagues [12]. Specifically, we present new results on the angular correlation of prompt neutrons from Pu-240 fission. These experimental results are compared to simulations performed with the MCNPX-PoliMi v2.0.0 Monte Carlo code. Cross-talk coincidence effects are estimated with MCNPX-PoliMi simulations and are removed from the experimental neutron–neutron angular distributions.

2. Experiment

The experiment consisted of two plutonium experiments, using a 0.84 g and 1.63 g ^{240}Pu$_{eff}$ sample, hereby designated PM2 and PM3. A Cf-252 sample was also used. At the time of the experiment, the plutonium metal samples had fission rates of 400 and

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760 fissions/s for PM2 and PM3, respectively; the Cf-252 sample had a fission rate of 26,000 fissions/s. The experiment was performed at the PERLA laboratory at the Joint Research Centre at Ispra, Italy [12].

As shown in Fig. 2, $16-7.62Ø \times 7.62$ cm EJ-309 organic liquid scintillation detectors were used to measure neutron–neutron coincidences from the plutonium samples. Two concentric 8-detector rings were stacked; samples were placed along the central detector ring axis. The sample was shielded with a 1 cm thick lead cylinder to reduce the gamma-ray count rate and acquisition dead time. Each detector is approximately 18 cm from the central axis; the top and bottom detector rings are separated by 10.6 cm from detector centers. Two, time-synchronized CAEN V1720 digitizers were used to collect 120-sample pulses. Pulses above a 70 keVee threshold were recorded for offline processing. A 70 keVee acquisition threshold and a 2 V dynamic range used in this experiment correspond to a range of 0.65–6.7 MeV proton recoils.

Pulse shape discrimination (PSD) by digital charge integration was used to discern neutron detections from gamma-ray detections [13,14]. The pulse tail of a gamma-ray interaction decays faster than a pulse of the same height from a neutron interaction. Waveforms that exhibit pile-up are eliminated, and then a ratio of the pulse tail integral to the total pulse integral was used to distinguish gamma-ray and neutron detections, shown in Fig. 1 for the PM2 sample. The PSD line was assigned using an algorithm described by Polack and colleagues that minimizes particle misclassification by analyzing segments of the tail-to-total integral points grouped by total integral [15]. The photon to neutron ratio for this sample is approximately 100:1 making PSD difficult at low pulse heights. The detection time differences of particles in a 60-ns coincidence window were used to create cross-correlation distributions with 2-ns bins for each detector pair.

3. Simulation with MCNPX-PoliMi

The MCNPX-PoliMi and MPPost codes were used to model the experiment geometry, the plutonium metal sample, and the detector response [5]. The experiment assembly is shown in Fig. 2a and the corresponding simulation model is shown in Fig. 2b. The detector photomultiplier tubes, detector holder, aluminum table, and concrete floor were also modeled in simulation, but were omitted from the figure for clarity.

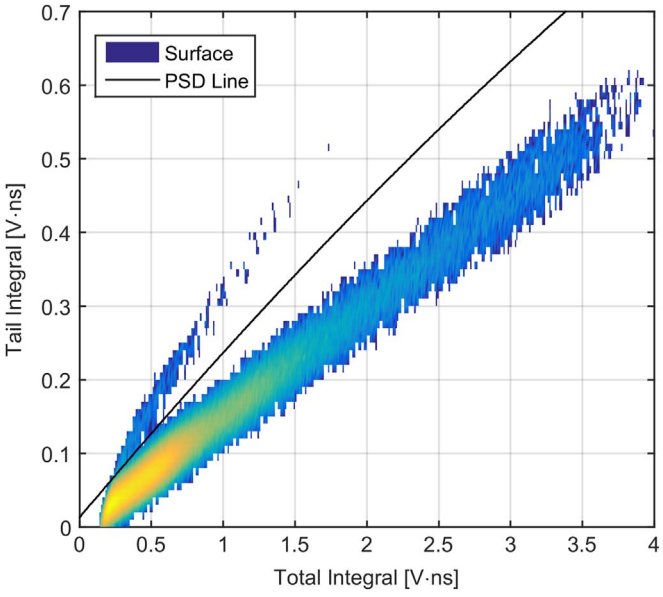

Fig. 1. Experiment pulse shape discrimination plot of tail to total pulse integrals for 290,000 pulses from the PM2 sample. Color scaling is logarithmic ascending to yellow. Neutrons lie above the discrimination line. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

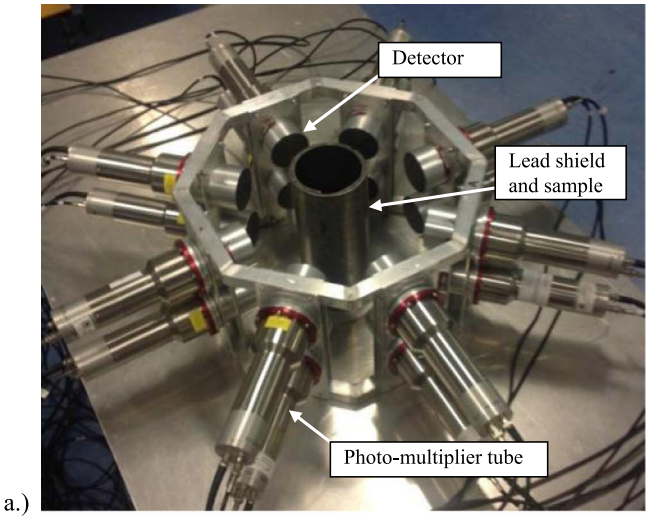

Fig. 2. (a) A photograph of the experiment setup with a plutonium metal sample centered inside a lead shield and (b) the simulated plutonium metal experiment setup with EJ-309 $7.62Ø \times 7.62$ cm organic liquid scintillator detectors, plutonium metal, and lead shield are shown.

Table 1
Plutonium metal sample isotopic composition.

Isotope	PM2 Weight fraction	PM3 Weight fraction
Ni	0.0504	0.0167
Cu	0.0319	0.0427
Pu-238	0.00004	0.00023
Pu-239	0.87367	0.85141
Pu-240	0.04076	0.07891
Pu-241	0.00028	0.00089
Pu-242	0.00013	0.00044

Each plutonium metal sample was modeled as a metal cylinder with composition described in Tables 1 and 2. The cylinder was topped by a void, and the void and cylinder were encased in aluminum.

The full isotopic composition of the samples was modeled in the material definition. Most fissions are from Pu-240 spontaneous fissions; only 0.5% and 0.9% of the spontaneous fissions are from Pu-242 for the PM2 and PM3 samples respectively. Pu-238 spontaneous fission was ignored. The PoliMi mixed-source option, IPOL (1)=99, was specified to sample from Pu-240 and Pu-242 built-in models. Using the plutonium metal sample MCNP model and the