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NEW EXPERIMENTS FOR DETECTING EMISSIONS FROM FISSION

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ABSTRACT

One of the most pressing challenges facing our society is to ensure that nuclear science and technology are used solely for peaceful purposes. The Consortium for Monitoring, Technology, and Verification (MTV) mission is to help meet this challenge by providing advanced technology and trained talent for careers in nuclear security and nonproliferation. The study of nuclear fission is one area of interest in the MTV. We are performing new experiments for the characterization of neutron and gamma ray emissions from fission fragments. These emissions are signatures for the detection and characterization of nuclear materials. To perform these experiments, we have developed Fission Sphere (FS-3), an array of forty organic stilbene detectors operated in time-coincidence. The FS-3 is used to measure the prompt emissions of neutrons and gamma rays from Cf-252 spontaneous fission. These new data will be used to validate physics-based prediction codes, including CGMF and FREYA, and will be useful in future ENDF and ENSDF evaluations. This paper presents new results from the first experiments using FS-3 and a Cf-252 spontaneous fission source. It compares the experimental results with predictions from theory and Monte Carlo simulation. Specifically, we describe the correlations among energy, multiplicity, and angles of emitted particles. We will also discuss the application of these correlations in multiplicity counting techniques, which are widely used in nuclear safeguards.

INTRODUCTION

A unique property of special nuclear material (SNM), such as uranium and plutonium, is its propensity to fission, either spontaneously or by neutron or photon excitation. Nuclear nonproliferation and safeguards measurements frequently employ fission signatures [1–3]. However, our understanding of the process of radiation emission from fission is still incomplete [4]. Measurements of average and uncorrelated fission de-excitation signatures exist, but the dependencies between them are still unknown. Several theoretical and empirical nuclear models of de-excitation, including the physics-based CGMF [5] and FREYA [6] codes, have been used to describe properties of fission, but important differences have been observed between the model predictions and experimental data [7].

In the past, we measured the correlated neutron and gamma-ray emissions from spontaneous and induced fission for several isotopes of interest to nonproliferation [8]. Specifically, we measured the correlations among energy, multiplicity, and angles of emitted neutrons and gamma rays; the correlation between energy spectrum and multiplicity is a particularly important parameter in multiplicity counting techniques, which are widely used in nuclear safeguards. In this work, performed in collaboration with ORNL, we measured the Cf-252 spontaneous fission signatures. For this experiment, we used Fission Sphere (FS-3), an array of 40 trans-stilbene detectors and a Cf-252 ionization chamber.

We present here the measured gamma ray and neutron time of flight distributions and multiplicity distributions. The purpose of the current analysis is to show the capabilities of the system to measure neutrons and gamma-rays from spontaneous fission on an event-by-event basis.

EXPERIMENTAL SETUP

The FS-3 detection system comprises 40 scintillation detectors in a spherical configuration. The FS-3 detectors used in this work are cylindrical 5.08 cm x 5.08 cm trans-stilbene organic crystals. The distance from the face of each detector to the fission source is adjustable, and can vary between 12 to 25 cm. This range allows the system to be used as a high-efficiency multiplicity counter as well as a neutron spectrometer. Other detectors can be used in the slots to optimize the detection of other particles. Experiments with 10 NaI (Tl) and 30 trans-stilbene detectors have also been performed. Pictures from an experimental setup are shown in Fig. 1. The structure of the FS-3 holder is composed of 3 rings held by vertical bars that extend to the floor. The entire supporting structure is made of aluminum. The detector holders are mounted on the three rings. In this paper, we present results obtained using the full array of trans-stilbene detectors, placed 23.5 cm from the geometric center of the array.

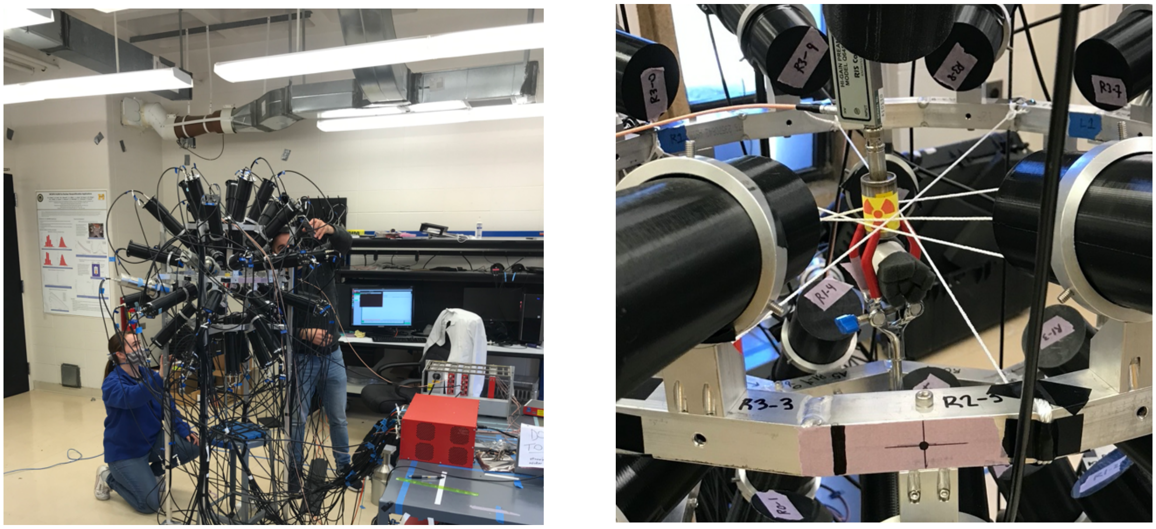

Figure 1: Photographs of the experimental setup. (Left) The FS-3 system and associated electronics, (right) interior of the FS-3 system highlighting the placement of the ^{252}Cf(sf) source.

We reproduced the geometry of the FS-3 system and the surrounding room in an MCNPX-PoliMi [3] simulation. The simulation code was used to assess systematic biases in the measurement system, and individual responses for each detector were modeled. The light output conversion is modeled using the semi-empirical Birks' model [9,10] The simulation model used in this study is shown in Fig. 2. The source of simulated particles in the model was the MCNPX-PoliMi default ^{252}Cf(sf) source, which includes energy and angular distributions of neutrons and gamma rays sampled from the included evaluated nuclear data [11,12].

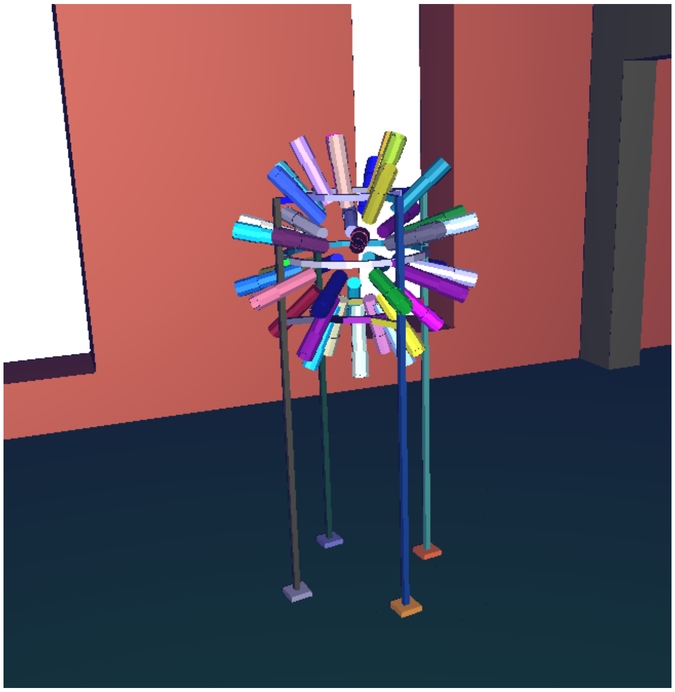

Figure 2: Simulation model of the FS-3 system

The source for this measurement was a fission chamber provided by Oak Ridge National Laboratory (ORNL). A picture of the fission chamber and preamplifier assembly is shown in Fig. 1. Charged particles induce a signal in the chamber proportional to the energy of the particle and the angle of emission with respect to the plates of the fission chamber. Due to the much larger energies of fission fragments compared to alpha particles, fission events can be discriminated on an event-by-event basis by analyzing the height of the pulse induced in the fission chamber.

In addition to the isolation of fission fragments and the subsequent suppression of background contributions, the fission chamber is used as a timing signal for time of flight (ToF) spectroscopy. The time resolution of the fission chamber was estimated to be approximately 1 ns FWHM. The signal from the fission chamber is passed through a ORNL manufactured preamplifier and is cloned through a CAEN N625 linear FAN-IN/FAN-OUT module. Three of the signals are digitized directly using three CAEN V1730 digitizers, where the pulse height cut is applied to discriminate the alpha contribution. A fourth copy of the fission chamber signal is inputted in one of the V1730 digitizer for digital CFD analysis, thus extracting the time of the fission event.

The measurement of the source in this configuration was performed quasi-continuously for two weeks, from Nov 1st to November 15th, 2020. The signal for the fission chamber was checked twice a day for consistency in the signal. Due to the age of the source, it is estimated that ~20% of the fissions are contributed from ^{250}Cf(sf) with the remaining mostly being ^{252}Cf(sf). A negligible contribution of ^{248}Cm(sf) is also expected. A total of 2×10^9 spontaneous fissions were recorded.

RESULTS

After receiving a valid signal from the fission chamber, a coincidence window is opened for the measurements of interactions in any of the surrounding trans-stilbene detectors. For each measured event, we record the total integrated charge, the integrated charge in the tail region of the pulse, and the time stamp of the interaction, as determined by digital constant fraction discrimination (CFD). The total integral of the distribution, after calibration, corresponds to the scintillation output produced in the interaction. The two integrals of the distribution can be used to perform pulse shape discrimination (PSD) of the pulses. The PSD procedure relies on the different time-dependence of the scintillation produced by electrons, set in motion by gamma-ray scattering, and protons, set in motion by neutron scattering. The histograms used for this discrimination are shown in Fig. 3.

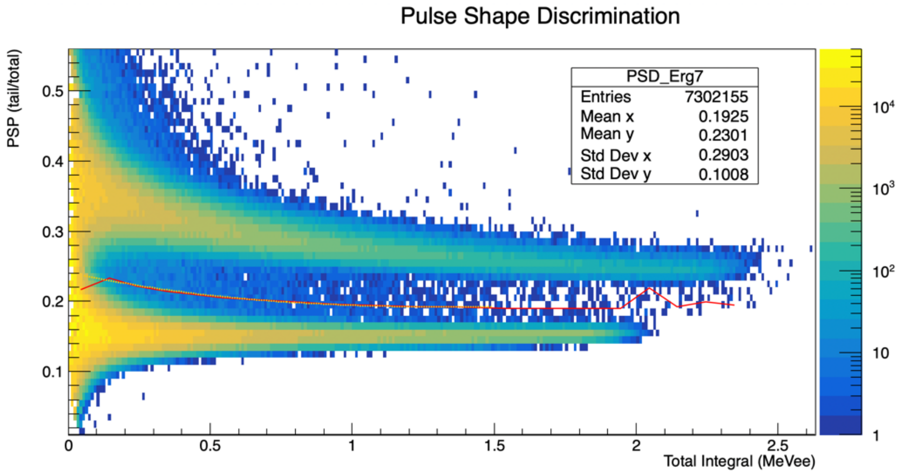

Figure 3: Particle discrimination capabilities of one detector in FS-3.

The integral of the pulses generated in the detector is proportional to the light output of the interaction. Figure 5 shows the measured light output spectra for neutrons and gamma rays measured with all detectors combined. The result shows good agreement between simulation and experiment for neutrons. The deficiency of gamma rays of high energies is currently being investigated and could be caused by geometrical effects or an overestimation of hard gamma rays from the fission source in simulation.

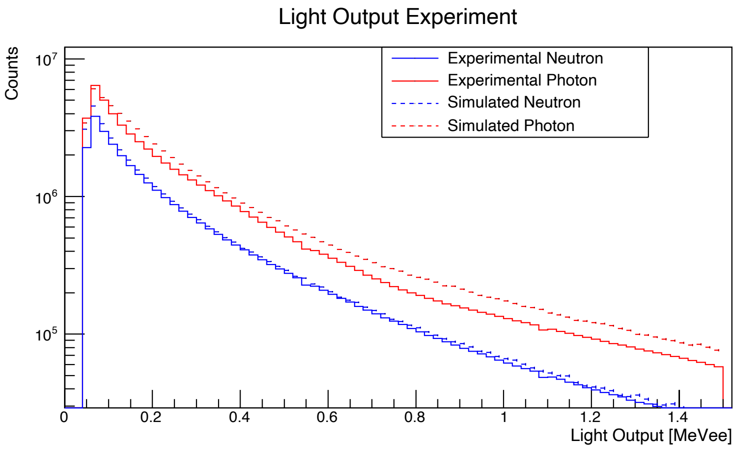

Figure 4: Light output distribution of neutrons and gamma rays in the measurement and simulation. Statistical errors are too small to be seen.

The time stamp of each interaction is used to determine the time of flight of the particle. Two time-gates are applied for the detection of the particles, with the limits of the time gates based on the kinematics of the radiation of interest. Two equivalent time gates are also opened before the signal from the fission chamber, to assess background. Particle discrimination is performed using both PSD and ToF gates. Gamma rays are collected between from 5 ns before the fission trigger until 10 ns after, while neutrons are collected between 4 ns and 50 ns after the fission trigger.

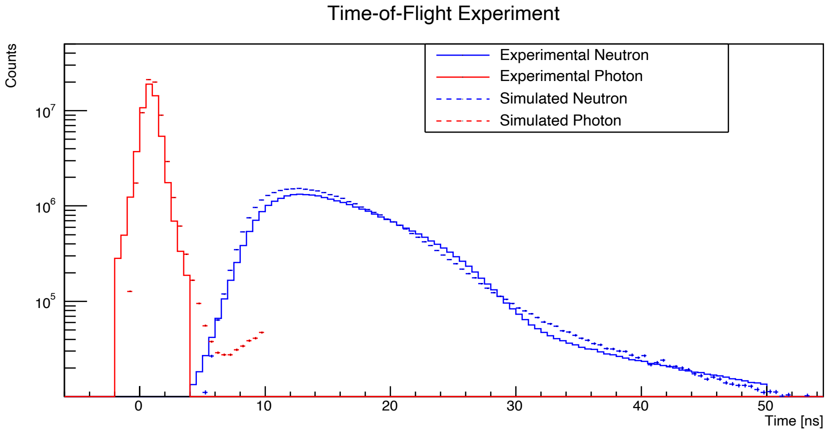

Figure 5: Time of flight distribution for all detectors of FS-3. Statistical errors are too small to be seen.

One of the primary questions of interest motivating the new experiment is the measurement of event-by-event neutron-gamma correlated multiplicities, *i.e.*, how the emission of neutrons affects the subsequent emission of gamma rays. Some studies of these correlations have already been performed using the Chi-Nu scintillator array at the Los Alamos Neutron Science Center and reported in Refs. [8,13,14]. The advantage of using trans-stilbene crystals instead of the EJ-309 organic liquid used in Chi-Nu lies in the superior light output and PSD properties of the trans-stilbene detectors, which allow us to explore neutron-gamma correlations at lower energies than was previously possible. Figure 6 shows the measured, correlated neutron-gamma multiplicity from Cf-252. Considering the light output and ToF gates applied, we have determined an acceptance energy region between 0.5 MeV and 5 MeV for neutrons and between 0.15 MeV and 2 MeV for gamma rays. At the lowest neutron energies, significant crosstalk and room return is present, but analysis techniques for the reduction of these biases are in development. The FS-3 system enabled us to measure correlated neutron multiplicities of 0 to 6, and gamma ray multiplicities of 0 to 8.

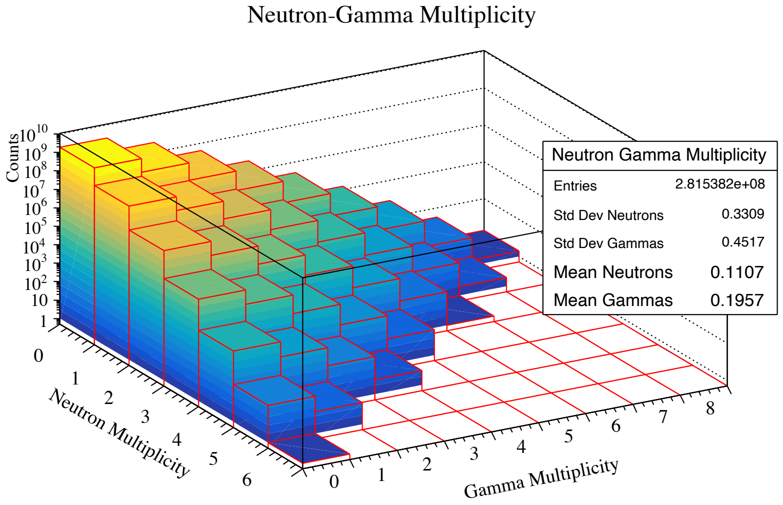

Figure 6: Measured neutron-gamma multiplicity distribution for ^{252}Cf(sf) with FS-3

We present in Fig. 7 the normalized covariance between neutrons and gamma rays if only neutrons of ToF energy T_n and gamma-rays of light output F_γ are considered, see Ref. [14] for details on this quantity. We observe the structure of positive neutron-gamma correlations superimposed on negative correlations previously observed using the Chi-Nu liquid organic scintillator array at Los Alamos National Laboratory [14]. This finding supports the hypothesis that mechanism responsible for the generation of angular momentum of fission fragments are energy dependent. Thus, with increasing internal energies of the fragments, both neutron emission and gamma-ray emission along discrete angular momentum states become more likely, and positive correlations are observed.

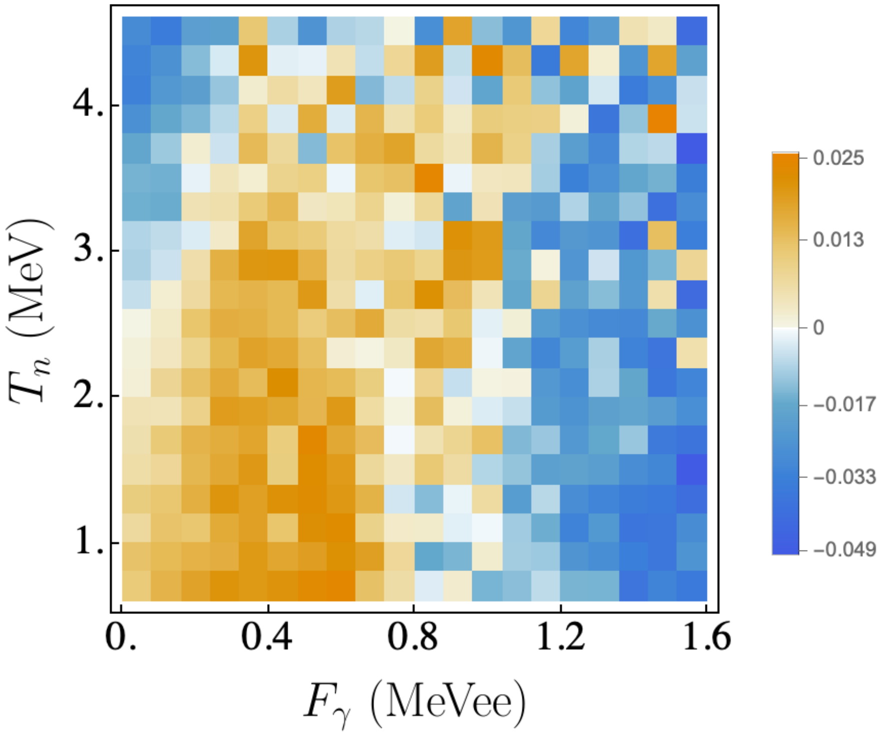

Figure 7: Spectrally dependent neutron-photon correlations in ^{252}Cf(sf) determined with FS-3.

By taking the ratio of the measured mean neutron and gamma-ray multiplicities to their known tabulated values, we have experimentally determined the absolute detection efficiencies of the FS-3 array in this configuration, 2.4% and 3.0%, for gamma rays and neutrons, respectively. The large number of detectors of the array make this system also an excellent detector of high-multiplicity events. We are currently developing statistical and event-by-event techniques for the removal of crosstalk events.

CONCLUSION

We presented new results from the FS3 array, a 40-detector array of trans-stibene scintillation detectors. We measured the correlated neutrons and gamma-rays from Cf-252 spontaneous fission. The data shows that the FS-3 array is capable of measuring coincident events from fissionable sources, and its fast organic scintillators have excellent neutron-photon discrimination. Preliminary results regarding the neutron-gamma emission correlations are consistent with previous observation, and analysis of these data is ongoing. Comparison between experiment and simulation showed good agreement across most of the observables. We are currently developing improved simulation models to improve the agreement with measurement. These models will also be used to develop energy unfolding matrices. Analytic and machine-learning based techniques are currently being developed to address systematic biases.

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