Event-by-event Neutron-Gamma Multiplicity Correlations in ^{242}Pu(sf)

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We analyzed the emission of neutrons and gamma rays by a spontaneously fissioning source of ^{242}Pu. We performed the experiment at the Los Alamos Neutron Science Center (LANSCE) using the Chi-Nu liquid scintillator array; a neutron spectrometer consisting of 54 organic scintillation detectors. This detector array is suitable for both neutron and gamma-ray detection and provides an excellent system for the study of neutron-photon correlations accompanying fission. Fission events were separated from α decays using an ionization chamber, which provided clear separation between decay modes as well as precise timing for the fission event. In this paper, we present the results of our investigation of neutron-photon correlations in the form of neutron-photon multiplicity covariance. We determine and unfold this parameter for the ^{242}Pu(sf) reaction and compare it with the analogous result from ^{252}Cf(sf). Our preliminary results show that the covariance is comparable in magnitude to the ^{252}Cf(sf) results, indicating that the correlations are not proportional to the neutron multiplicity or initial excitation energy. This result is explained physically by considering the negative correlations as predominantly existing for the last neutrons and the first photons in the emission cascade.

I. Introduction

Neutrons and gamma rays are emitted accompanying the nuclear fission. Since neutrons and gamma rays often represent the only detectable signature of a fissionable sample, the correlations of these particles with one another and with the fissioning nuclei are important in both nonproliferation and scientific applications. The event-by-event neutron-gamma emission correlations, hypothesized to exist due to energy conservation as well as the suspected energy-dependent spin generation mechanism, has been investigated repeatedly over the past decades for ^{252}Cf(sf) [1]–[3]. While these results were not in agreement with one another in the published versions, in a previous publication [4] we have shown that corrections can be applied to the experimental results and achieve an approximate agreement. We have determined that there exist significant negative neutron-gamma emission correlations in ^{252}Cf(sf). Furthermore, we have investigated the energy dependence of these emission correlations [5] and have observed structure of enhanced neutron-gamma correlations attributed to spin-energy correlations in the formation of fission fragments.

In this work, we extend the discussion of emission correlations to another spontaneous fission reaction, ^{242}Pu(sf). The primary goal of our analysis is to confirm the hypothesized nature of the event-by-event emission correlations. Specifically, we suspect that the dominant source of neutron-photon negative emission correlations is related to the residual intrinsic excitation energy after the emission of the last neutron. If this assumption were correct, because ^{252}Cf emits more neutrons than ^{242}Pu, we would find that the correlations *per emission* is higher in plutonium than in californium. Another important goal of this analysis is to observe the energy-dependent structure observed in californium, thus strengthening the hypothesis of positive spin-energy correlations.

This paper is structured as follows. In Section II, we begin by describing the mechanism for the generation of negative emission correlations due to competition over the fragment intrinsic excitation energy. In the same section, we also present the normalized covariance; the parameter we will employ to describe the correlations. In Section III, we describe the Chi-Nu experimental setup and compare the experimental data to simulation. In Section IV, we analyze the multiplicity distribution to extract the neutron-gamma emission correlations. The result is discussed and compared to the determination based on ^{252}Cf(sf). Lastly, in Section V we draw the main conclusions on the origins of the observed emission correlations and propose avenues of future research.

II. Origin of Emission Correlations

There exist several mechanisms for the generation of neutron-gamma emission correlations in fission. For the present work we discuss only the two dominant sources of negative and positive correlations. The combination of these mechanisms gives rise to structure.

The dominant source of negative correlations is the competition between the last neutron emitted by each fragment and the subsequent statistical gamma-ray emission cascade. It is generally assumed that the neutron emission will proceed if the intrinsic excitation energy, *i.e.,* the energy not stored in collective nuclear modes, is above the neutron separation energy. After the last neutron emission, the fragment will in general have some portion of its initial intrinsic excitation energy. This intrinsic energy will be dissipated by statistical gamma-ray emission. It is then apparent that negative correlations will be introduced between the last neutron in the cascade in the emission and the statistical gamma rays.

After statistical gamma-ray emission, collective gamma-ray emission from discrete states dissipates the remaining energy stored in the collective modes of the fragments. The multiplicity and properties of these collective emissions are determined primarily by the angular momentum of the fission fragments after neutron emission and statistical gamma-ray emission. The multiplicity and properties of neutrons are, on the other hand, related most closely to the excitation energy of the fission fragments. It is then possible to introduce indirect neutron-gamma correlations based on underlying correlations between the fragments' excitation energy and angular momenta, or spin-energy for short. Enhanced correlation structure, possibly associated with positive spin-energy correlations, have already been observed for ^{252}Cf(sf) in a previous experiment [5].

III. Experiment

The experiment was performed at the Chi-Nu liquid-scintillator detector array at the Los Alamos Neutron Science Center (LANSCE) in February of 2020. This detector array consists of 54 EJ-309 liquid organic scintillators with 17.78 cm diameter and 5.08 cm thickness. The detectors are arranged in hemispherical configuration, with each detector at approximately 1 m from the center of the detector array. An ionization chamber was placed at the geometric center of Chi-Nu. The chamber was loaded with a thin deposit of ^{242}Pu, of approximately 5 mg. The Chi-Nu array and the ionization chamber are shown in Fig. 1.

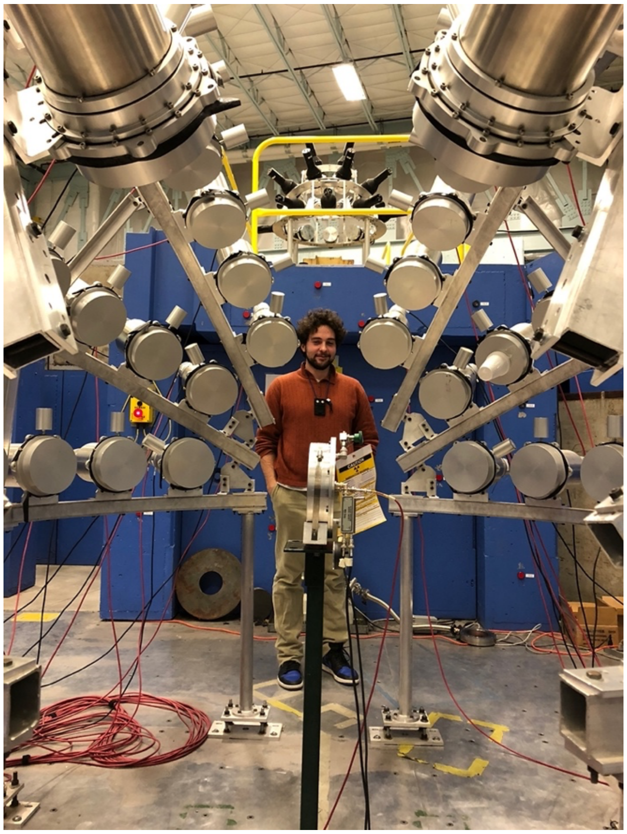

Figure 1: Photograph of the experimental setup.

Thanks to the large difference in kinetic energy of alpha particles and fission fragments, the ionization chamber can discriminate alpha particles and fission fragments on an event-by-event basis using the integrated charge induced in the ionization chamber. No coincidence logic was applied during the measurement; however, in data analysis only detections in the EJ-309 detectors which were in time coincidence with a valid fission trigger from the ionization chamber were analyzed. In total, 1.2×10^7 fission events were recorded over a month of measurement time.

For each detected event we record: the time of the interaction, the total integral of the pulse, and the ratio of the integrated tail of the pulse to the total integral, i.e., the pulse shape parameter (PSP). The time of flight of the particle is taken with respect to the signal from the ionization chamber. The discrimination between neutrons and gamma rays is performed considering all of the above properties, as shown in Fig. 2. In the top panel of the figure, we observe that neutrons and gammas are separated by their PSP, where the neutrons possess a larger tail/total compared to gamma rays, due to the interaction mechanics they have with the organic scintillators. In the bottom panel of the figure, we see that particles are well separated in time of flight as well. A light output threshold of 0.2 MeVee is applied to all detection, corresponding to approximately 1.3 MeV proton recoil energy.

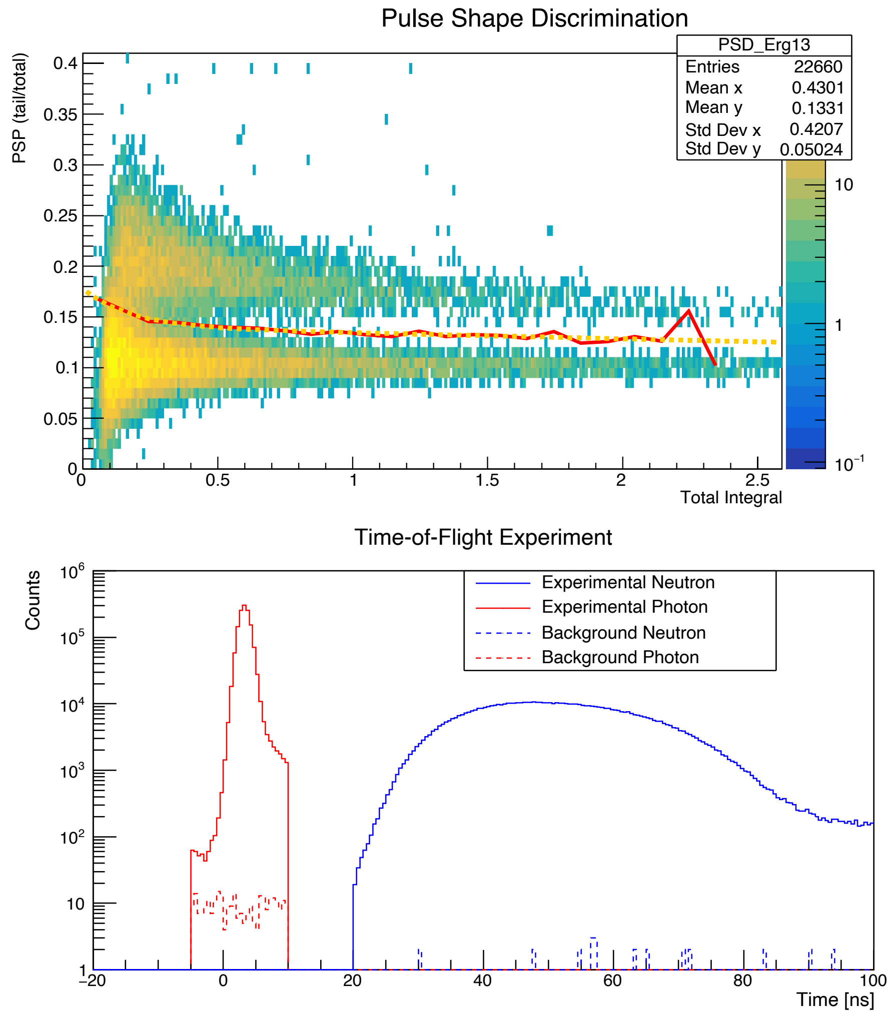

Figure 2: Discrimination of neutrons and gamma rays is performed using pulse shape discrimination (top) and time of flight (bottom)

For every signal trigger from the ionization chamber, we open a collection window in the liquid organic scintillators before the fission to collect the background. The time gates and energy thresholds of this anticipated collection are exact duplicates of the ones opened after the fission signal to collect neutrons and gamma rays from fission. This estimation method for background is well-suited in applications where the source of background is predominantly time-independent and time-uncorrelated with the fissions. In this experiment, the background is predominantly from the alpha decay of the target, which gives rise predominantly to a gamma-ray background. From Fig. 2 we see that background has a negligible impact on the distributions, thanks in large parts to the coincidence enforced with the ionization chamber.

IV. Neutron-Gamma Correlations

We investigate the neutron gamma correlations following the same procedure outlined in Refs.[4], [5]. We evaluate the normalized differentiated covariance, $\boldsymbol{C}_{\boldsymbol{E_n},\boldsymbol{E_\gamma}}$, which is defined as

$$\boldsymbol{C}_{\boldsymbol{E_n},\boldsymbol{E_\gamma}} = \frac{\partial^2 \mathbf{cov}(\boldsymbol{N_n}, \boldsymbol{N_\gamma})}{\partial \boldsymbol{E_n} \partial \boldsymbol{E_\gamma}} \left(\frac{\boldsymbol{d}\langle \boldsymbol{N_n} \rangle}{\boldsymbol{dE_n}} \frac{\boldsymbol{d}\langle \boldsymbol{N_\gamma} \rangle}{\boldsymbol{dE_\gamma}} \right)^{-\mathbf{1}} \tag{1}$$

In the definition of Eq. (1), $\boldsymbol{N_n}$ and $\boldsymbol{N_\gamma}$ are the emitted neutron and gamma-ray multiplicities, $\boldsymbol{E_n}$ and $\boldsymbol{E_\gamma}$ are the spectra of these particles. It should be noted that the quantities in Eq. (1) are all related to the emission and are thus not directly accessible in experiment. In order to reconstruct these quantities from the measured multiplicities and measured energies, we apply the same unfolding procedure outlined in Ref. [5]. Due to the low number of counts in the reaction, we do not apply the unfolding of the neutron response. For these data, we take the neutron time of flight energy to be the emitted energy. This will leave us with a bias especially at lower energies where scattered high-energy neutrons will be interpreted as low energy neutrons.

The results for $\boldsymbol{C}_{\boldsymbol{E_n},\boldsymbol{E_\gamma}}$ are shown in Fig. 3. For comparison, we have also performed calculations using CGMF[7]. The result of these calculations is labeled "Sim" in the figures.

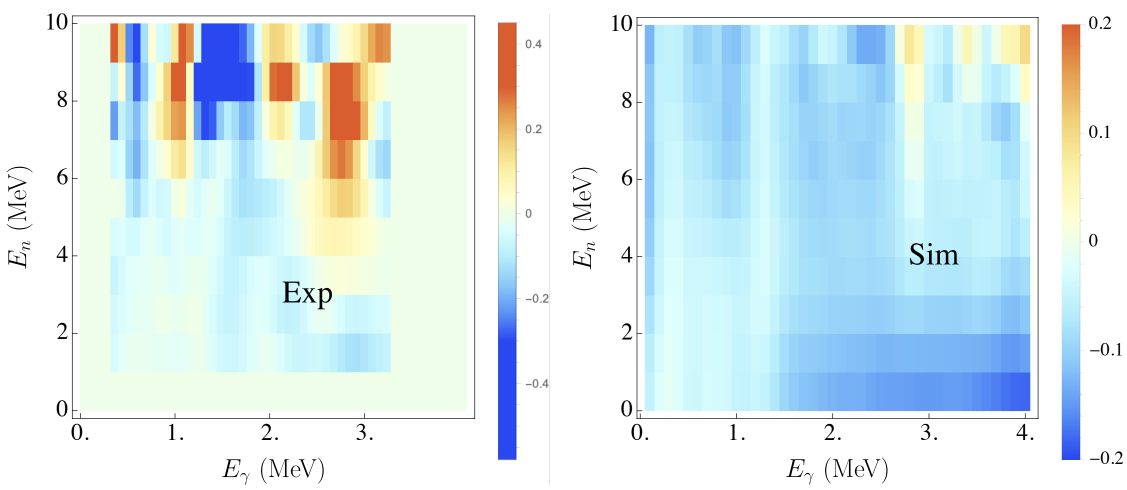

Figure 3: The normalized differentiated neutron-gamma correlations from experiment and from CGMF calculation.

It should be noted that due to the relatively low statistics and the uncertainties introduced by the unfolding procedure, the results presented here should be interpreted as qualitative, and more data should be collected. We see from Fig. 3 that structure is present in the correlations, with most of the correlations across the neutron and gamma spectra slightly negative and several peaks of enhanced positive correlations arising at $E_\gamma \approx 0.9, 2.4,$ and 3.0 MeV, with the latter being the strongest correlations.

By comparison with past experimental data and model calculations, we find that the enhanced correlation structure is due to two distinct factors. The positive spin-energy correlations result in the presence of enhanced correlations across all neutron energies. On the other hand, the behavior with increasing E_n and the increasing magnitude of some of the structure is due to biasing of the fission towards more symmetric fissions, which tends to emit higher energy neutrons[8]. Fissions near the shell closure of ^{132}Sn have high first excited states, thus producing the observed high-energy correlated structure at $E_\gamma \approx 2.4$ and 3.0 MeV. To highlight the spin-energy correlations, we show a slice of the correlation matrix C_{E_n,E_γ} for fixed neutron energies in Fig. 4. At this energy, we see peaks associated with positive spin-energy correlations. As we had previously done for the ^{252}Cf(sf) data, we again observe stronger correlations than modeled in CGMF. This effect could be caused by either stronger spin-energy correlations in the mechanisms that produce angular momentum, or a much smaller removal of angular momentum by neutrons. We also note that CGMF predicts enhanced correlations at the smallest of gamma-ray energies, $0.1\ \text{MeV} < E_\gamma < 0.5\ \text{MeV}$, which are however absent in the experimental data.

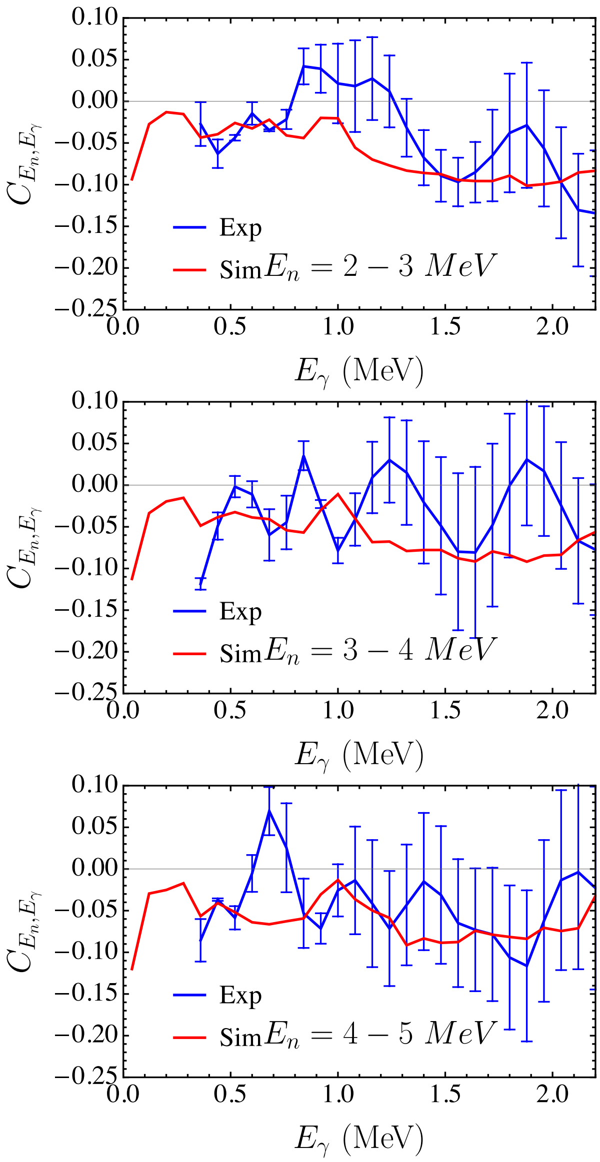

Figure 4: Slice of the neutron-gamma correlations for experiment and CGMF calculation, for fixed neutron energies.

Lastly, we compare the value of the neutron-gamma correlations integrated over all accepted energies, i.e., $1.5 < E_n < 8$ MeV and $0.36 < E_\gamma < 2.4$ MeV. We find

$$\frac{\mathrm{cov}(N_n, N_\gamma)}{\langle N_n\rangle\langle N_\gamma\rangle} \approx -0.031 \pm 0.005 \tag{2}$$

This value of the scaled covariance is approximately twice as large as that measured for ^{252}Cf(sf)[4], although the energy acceptance for the ^{252}Cf(sf) extended to lower energies. To correct for this, we have re-processed the ^{252}Cf(sf) data to artificially restrict the acceptance region. We have obtained a value of -0.01 ± 0.002 for the scaled covariance, once

This agrees with what is expected if the correlations are generated primarily by the last neutrons in the cascade emission. The lower neutron multiplicity of ≈ 2.15 in ^{242}Pu(sf) compared to ≈ 3.75 in ^{252}Cf(sf) results in increased correlation per neutrons in the former reaction.

V. Conclusions

We have shown that the structure of enhanced neutron-gamma correlations, previously observed in ^{252}Cf(sf) are present in the reaction ^{242}Pu(sf) as well. While not definitively confirming the presence of positive spin-energy correlations, these experimental results strengthen the hypothesis and aid in their quantification. Comparison with model calculations show that the data has significant unreproduced correlations enhancements at large $\boldsymbol{E_\gamma}$, most likely associated with discrete level transitions in the spherical nuclei near the shell closure at ^{132}Sn. Comparison of the neutron-gamma covariance integrated over all neutron and gamma-ray spectra is consistent with a model in which the most dominant source of correlations is determined by the last neutron emitted in the cascade of each fragment competing with the subsequent statistical gamma rays.

Future work should focus on examining the correlation structure for other types of fission, including neutron-induced and photon-induced fissions. Differential experiments capable of measuring the properties of fragments in coincidence with their emission should also be employed to isolate and quantify the spin-energy correlations.

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