Experimental Investigation of Multiplicity Correlation Between Prompt Neutrons and Photons in the Spontaneous Fission of $^{252}$Cf

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Abstract

Excited nuclear fragments are emitted during nuclear fission. The de-excitation of fission fragments takes place as sequential emission of neutrons followed by photons. Particle emission dissipates the intrinsic and rotational excitation energies of the fragment until a long-lived state is reached. Both the number of neutrons and the number of photons emitted during de-excitation are dependent on the initial excitation energy, angular momentum, and parity of the radiating fission fragment. A correlation between neutron and photon multiplicities following fission is thus expected. We perform linear regression analysis of the data collected by the Chi-Nu array at the LANSCE facility in Los Alamos National Laboratory. We have developed analytic expressions to determine the bias introduced by background sources and the misclassification of particles by our pulse shape discrimination capable organic scintillation detectors. Additionally, we have developed perturbative techniques to derive corrections to the binomial-response model. We use the perturbative techniques to correct the experimentally-determined neutron-photon correlations for the effects of photon-neutron pileup. We have experimentally determined an event-by-event covariance, cov($N_n, N_\gamma$), of $-0.33 \pm 0.05$ between the neutron and photon multiplicities emitted following the spontaneous fission of $^{252}$Cf. The result suggests the existence of a small event-by-event competition of neutron and photon emission in fission. We compare our result with previous analyses and model calculations.

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I. INTRODUCTION

Since its discovery eighty years ago, nuclear scientists have been investigating the properties of fission, attempting to shed light on the dynamics of the fission process. Due to the complexity introduced by the large number of nucleons and strong forces involved, a complete and satisfactory description of fission is still lacking. Spontaneous fission offers a simpler problem to solve than induced fission, because no additional particles are introduced. Due to its widespread use as a calibration reaction and its high spontaneous fission rate, $^{252}$Cf is a baseline isotope for fission studies. However, there are still many aspects of the spontaneous fission of $^{252}$Cf that are not fully characterized or understood, most notably the correlation between the emission of particles following fission [1].

Nuclear fission is not only interesting for fundamental nuclear physics, but also has important applications in the fields of nuclear energy and nonproliferation [2–4]. Among the goals of research prompted by nonproliferation efforts is the characterization of fission signatures [5], namely the penetrating particles that are emitted following a fission reaction. Only neutrons, photons, and neutrinos are considered signatures of fission. All the other reaction products, such as fission fragments, as well as $\alpha$ and $\beta$ particles, are charged and are therefore more easily shielded [6]. Although specialized neutrino detection systems exist, conventional detection technologies for most applications are based only on the detection of neutrons and photons.

Our objective is to experimentally determine the event-by-event neutron-photon multiplicity correlation in the spontaneous fission of $^{252}$Cf. The dependence of neutron multiplicity on photon multiplicity is an important, and yet unresolved, question in the characterization of fission signatures.

The outline of this work can be summarized as follows. In Section II, we present background on neutron-photon emission and detection, as well as an overview of previous work. In Section III, we present linearly-correlated emission as a suitable model for fission de-excitation. In Section IV, we describe the experimental setup used to measure neutrons and photons from the spontaneous fission of $^{252}$Cf. In the same section, we describe how the data was collected and assess experimental biases. In Section V, we analyze the data and extract a value for the neutron-photon multiplicity covariance at emission. In Section VI, we compare our experimentally-determined covariance with previous measurements and model-based predictions. Lastly, we summarize the results and draw conclusions in Section VII. Four appendices are included. In Appendix A, we derive weighted linear regression analysis. In Appendix B, we derive the sensitivity of the covariance on uncertainties in data. In Appendix C, we derive analytic expressions for the biases introduced by background and misclassification in the detected distribution. In Appendix D, we derive the formalism of the perturbed binomial response.

II. BACKGROUND

A. Neutron and Photon Emission in Fission

As shown schematically in Fig. 1, a fission fragment with a given initial angular momentum and excitation energy reaches its ground state by emitting neutrons and photons. In the figure, a shaded ellipse represents the distribution of the initial energy and angular momentum of the fragment. As long as the excitation energy of the fragment is above the
neutron separation energy, $S_n$, neutron emission is dominant. In neutron emission, the excitation energy of the fragment is dissipated as neutron separation energy as well as kinetic energy of the neutron. Neutron emission does not significantly affect the fragment angular momentum because neutrons are emitted predominantly as $s$-waves \cite{7}. When the excitation energy of the fragment falls below the neutron separation energy, photon emission becomes the primary mechanism of de-excitation. The Yrast line defines the minimum excitation energy for states with fixed angular momentum \cite{8 9}. Fragments in states near the Yrast line have a high spin compared to their energy and thus de-excite by photons of higher multipolarity \cite{10}. Because the fragment excitation energy is dissipated through neutron and photon emission, we expect neutron-photon emission in a given fission event to be correlated. We can further expect these event-by-event correlations to affect the neutron and photon multiplicities relative to each other within a given fission event \cite{11}.

Two possible origins of multiplicity correlations exist: a competition between neutron and photon emission probabilities \cite{7}, referred to here as the decay-width effect, and a correlation between the different de-excitation paths resulting from the possible fragment splits at scission \cite{8 12}, referred to as the yield effect. The decay-width effect gives rise to a negative correlation whereas the yield effect can produce either a positive or a negative correlation. Both effects are present in the fission process. However, decay-width effects are only important in a narrow excitation energy range near the neutron separation energy (the dashed line in Fig. 1), where the probabilities of neutron and photon emission are comparable \cite{7}. The probability for the fragment to de-excite to a state within this narrow energy region is small \cite{10 13 14}. Therefore, we expect the yield effect to be the dominant cause of multiplicity correlations.

![Fig. 1. A fission fragment de-excites to its ground state by emitting neutrons and photons. This figure is adapted from Ref. \cite{10}.](image)

Physics-based fission event generators CGMF \cite{1 14 15} and FREYA \cite{1 12 16 19} predict negative event-by-event correlations in the neutron and photon multiplicities emitted following the spontaneous fission of $^{252}$Cf \cite{20}. For details on the fission models employed in CGMF and FREYA, see the review in Ref. \cite{1} and references therein.

In addition to the event-by-event analysis discussed so far, neutron and photon emission has also been correlated through fragment-by-fragment analyses. Fragment-by-fragment analyses assign a mean neutron and a mean photon multiplicity to specific fission fragments, as determined over an ensemble of measured fission events. Thus, fragment-by-fragment analyses integrate over all possible de-excitation paths that a fragment may take. The mean multiplicities are then correlated with each other through the intermediary fragments \cite{11}.
Event-by-event analyses, on the other hand, assign a neutron multiplicity and a photon multiplicity to each fission event, thereby retaining information on the de-excitation path followed by the fragment. Although fragment-by-fragment and event-by-event analyses can both produce correlations between neutron and photon multiplicities, they are two distinct types of correlation that cannot generally be expected to agree. This point is made clear in Ref. [21]: the same data produces a positive multiplicity correlation when analyzed fragment-by-fragment, and a negative multiplicity correlation when analyzed event-by-event.

B. Binomial Response

It is not possible to directly determine a correlation between the emitted neutron and photon multiplicities, because this would require $4\pi$ coverage and 100% efficiency. To determine the correlation in the emitted multiplicities, we determine a correlation in experiment that we then unfold using knowledge of the system response. The analytic model of system response used in this work is the binomial-response model developed by Diven et al. [22] and further investigated by Nifenecker et al. [23]. See also the work by Bzdak et al. [24, 25] for more recent applications of the binomial-response model. The system response relates the emitted neutron and photon multiplicities, $N_n$ and $N_\gamma$, to the detected multiplicities, $D_n$ and $D_\gamma$. Binomial response assumes that every particle is detected independently of all other particles and that each particle is detected with a constant detection efficiency, $\epsilon_n$ and $\epsilon_\gamma$ for neutrons and photons, respectively. Neutron and photon detection efficiencies are expressed in terms of the emitted and detected mean multiplicities as [24]

$$\langle D_n \rangle = \epsilon_n \langle N_n \rangle, \quad (1a)$$

$$\langle D_\gamma \rangle = \epsilon_\gamma \langle N_\gamma \rangle. \quad (1b)$$

We note here that the only explicit variables of the binomial-response model are the multiplicities, both detected and emitted, and the detection efficiencies. Therefore, this model misses some important underlying physical mechanism. For example, the dependence of multiplicity on emission energy is not treated explicitly. As a result of unfolding multiplicity correlations using the binomial-response model, we are implicitly assuming that the correlation in the detected neutron and photon multiplicities is a good representation of the correlation across the entire energy spectrum. Energy-dependent multiplicity correlations are the subject of future work.

Because the number of detectors in any detection system is finite, and detectors cannot resolve and analyze interactions occurring in short succession ($\approx 15$ ns decay time for pulses from our detectors), the detection of particles effectively depletes the total number of available detectors for subsequent detections. This behavior, which cannot be modeled by the binomial-response model, is called pileup. We define photon-neutron pileup to be the coincident interaction of both a photon and a neutron within the same detector. Although this occurrence can be rare for a low-efficiency detection system, affecting only 0.08% of all measured events in the experiment we analyze, the effects of photon-neutron pileup on multiplicity correlations are significant.
C. Previous Analyses

There have been previous experimental analyses of multiplicity correlations in the spontaneous fission of $^{252}$Cf. Experimental evidence of neutron-photon multiplicity correlations has been published by Nifenecker et al. [11], Glässel et al. [21-26], Bleuel et al. [27], Wang et al. [28], and Marcath et al. [20]. The results reported in these works, while open to interpretation, have been used to suggest the existence of a positive, negative, or effectively null event-by-event multiplicity correlation between neutrons and photons. The studies by Nifenecker et al. [11] and Wang et al. [28] were carried out on a fragment-by-fragment basis and thus are not relevant for our investigation and are not discussed further. The results of Glässel et al. [21-26], Bleuel et al. [27], and Marcath et al. [20] were reported on an event-by-event basis. Glässel et al. reported evidence for a small negative correlation, while both Marcath et al. and Bleuel et al. reported a much smaller or null correlation. We note that while Marcath et al. and Glässel et al. investigated the correlations from all fragment pairs, Bleuel et al. investigated correlations from two specific fragment pairs. Marcath et al. used a forward model of unfolding to interpret the results of the experiment, and did not draw statistically significant conclusions. The results of Glässel et al. were not quantitatively corrected for efficiency losses and pileup in Refs. [21-26]. Although experimental evidence exists, a resolution of the discrepancies between these results is needed.

The work presented here is the continuation of two papers that investigated the event-by-event multiplicity correlation in the prompt de-excitation of $^{252}$Cf [20, 29]. In Ref. [20], Marcath et al. presented the experimental findings of the presence of a multiplicity correlation in the data collected by the Chi-Nu array at Los Alamos National Laboratory. They compared the data with simulated events produced with the Monte-Carlo particle transport code MCNPX-PoliMi [3, 30] using, as simulated fission source, the neutrons and photons generated by CGMF [1, 14, 15], FREYA [12, 16–19], and the multiplicity-uncorrelated fissions source included in MCNPX-PoliMi [3, 30], referred to as PoliMi. In these simulations, the events produced by PoliMi were used as the null hypothesis of uncorrelated neutron-photon emission. Although Marcath et al. found a small negative multiplicity correlation, the result could not demonstrate the existence of a correlation with a greater than 1σ significance. In a conference publication [29], the Marcath et al. data was unfolded from the simulated detector response and compared directly to CGMF and FREYA predictions. The Pearson correlation coefficient, which quantifies the linear component of the correlation between two variables, was used to quantify the neutron-photon multiplicity correlation at emission. The resulting correlation was smaller than those predicted by CGMF and FREYA by a factor of $\approx 3 - 4$.

III. LINEARLY-CORRELATED EMISSION

In this section, we show that both experimental results and model predictions suggest approximately linearly-correlated emission for neutrons and photons following the spontaneous fission of $^{252}$Cf. In the case of linearly-correlated emission, the conditional mean neutron multiplicity changes linearly with the photon multiplicity, and vice versa. The conditional mean multiplicities are described as

$$\langle N_n|N_\gamma = n_\gamma \rangle = \langle N_n \rangle + [n_\gamma - \langle N_\gamma \rangle] \alpha_n(N_n, N_\gamma) ,$$  

(2a)  

$$\langle N_\gamma|N_n = n_n \rangle = \langle N_\gamma \rangle + [n_n - \langle N_n \rangle] \alpha_\gamma(N_n, N_\gamma) ,$$  

(2b)
where \( \alpha_n(N_n, N_\gamma) \) and \( \alpha_\gamma(N_n, N_\gamma) \) are regression slopes characterizing the correlation. The notation \( \langle X \mid Y = y \rangle \) is understood as the mean of \( X \) given that \( Y = y \). We present evidence that Eq. (2) provides a good approximation for the emission of neutrons and photons from \(^{252}\)Cf near the mean multiplicities \( \langle N_n \rangle \) and \( \langle N_\gamma \rangle \). We analyze the predictions made by CGMF and FREYA, as well as the experimental evidence of Glässel et al. [21].

### A. Fission Event Generators

For an analysis and overview of the predictions of CGMF and FREYA of the neutron and photon multiplicity distributions in \(^{252}\)Cf, see Section III of Ref. [20]. In both fission event generators, the neutron-photon multiplicity correlation is not an input parameter, but rather a prediction arising from the physical models. The fission event generators simulate energy and spin (and parity in the case of CGMF) of the fission fragments as the fragments de-excite to their ground states by emission of neutrons and photons, in a manner analogous to that presented in Fig. 1. A low energy threshold of 150 keV, in the laboratory frame, is applied to both neutron and photon emission.

CGMF and FREYA predict decreasing mean neutron (photon) multiplicity as the photon (neutron) multiplicity increases. The results of regression analysis of the simulations are shown in Fig. 2. In Fig. 2(a), data points are positioned at the conditional mean neutron multiplicity for each photon multiplicity, while the size of each point is proportional to the relative probability of that photon multiplicity. The role of neutron and photon multiplicity is switched in Fig. 2(b). The least-square linear fits indicated in the legends are shown in solid lines. We note that to find the proper linear regression slopes, one must weight each point by the probability of the given multiplicity, i.e., the point \( \langle X \mid Y = y \rangle \) is weighted by the probability that \( Y \) takes the value \( y \), \( P(Y = y) \). In Appendix A, we prove the validity of the weighted linear regression, starting from the principles of standard linear regression.

We find that the expected r.m.s. deviations (see Appendix A) of \( \langle N_n \mid N_\gamma = n_\gamma \rangle \) and \( \langle N_\gamma \mid N_n = n_n \rangle \) from the linear model are \( \approx 0.01 \) and \( \approx 0.03 \), respectively for both CGMF and FREYA. Although CGMF and FREYA are based on different models of nuclear de-excitation they both predict an approximately linearly-correlated emission. The deviations from linearity are in regions of large multiplicities compared to the mean multiplicities. The regression analysis of CGMF and FREYA suggests that a linear correlation is warranted and only a negligible error is introduced by assuming linearly-correlated emission. We note that, aside from the threshold energy imposed on both neutron and photon emission, no other restrictions are placed on the simulated events.

### B. Prior Experimental Work

Glässel et al. [21, 26] presented results showing a negative event-by-event multiplicity correlation in the spontaneous fission of \(^{252}\)Cf. In this section, we only discuss whether these data can be decribed by linearly-correlated emission and defer a discussion of the magnitude and sign of their result to Section VI. The Glässel et al. result was presented as a regression of detected mean neutron multiplicity on the detected photon multiplicity (see Section 5.3 of Ref. [24]). However, given that the detection system used by Glässel et al. achieved nearly perfect detection efficiency for photons \( \epsilon_\gamma > 98\% \) [31], the detected photon multiplicity is very close to the emitted photon multiplicity.
FIG. 2. Regression plots of neutron and photon multiplicity from $^{252}\text{Cf}$ for CGMF and FREYA, for both (a) mean photon multiplicity on neutron multiplicity and (b) mean neutron multiplicity on photon multiplicity. Linear regression lines and their equations are shown on the plot. We note that the $y$ axes on both (a) and (b) are zero-suppressed and that the axis scales are chosen to better reflect the relatively smaller neutron multiplicity.

We perform regression analysis of the Glässe et al. data, reproduced in Fig. 3, to compute the linearity of the correlation. To perform a regression analysis of the Glässe et al. data, we need to know the event-by-event photon multiplicity distribution, *i.e.*, the probability of emission of each emitted photon multiplicity. Several discordant experimental results exist for this distribution (see discussion in Ref. [32]). Fortunately, the result of the regression does not depend significantly on the choice of distribution. Here and later in Section VI we adopt the empirical negative binomial model (see Ref. [33] and Eq. (1) in [32]), which has two parameters: the mean photon multiplicity $\langle N_\gamma \rangle$ and the relative width parameter $\Delta_\gamma$.

In this context, the relative width parameter is defined as the excess width of the negative binomial model from the Poisson distribution, or

$$\frac{\sigma^2(N_\gamma)}{\langle N_\gamma \rangle} = 1 + (\Delta_\gamma - 1)\langle N_\gamma \rangle .$$

We use the measurement of $\langle N_\gamma \rangle$ by Oberstedt et al. [32], $\langle N_\gamma \rangle = 8.29 \pm 0.13$, while the
measurement of Ramamurthy et al. [34] is used for $\Delta_\gamma = 1.074 \pm 0.0154$.

FIG. 3. Weighted linear regression analysis performed on Schmid-Fabian and Glässel et al. data [21, 26]. Marker size indicates the probability of the photon multiplicity, from Valentine and Oberstedt et al. [32, 33]. Note that the y axis is zero-suppressed.

The r.m.s. deviation of $\langle D_n | N_\gamma = n_\gamma \rangle$ from linearity is $\approx 0.002$. We note that, from the data discussed here, we may only conclude that the detected neutron multiplicity $D_n$ is linear in the emitted photon multiplicity. Assuming that the system response does not suppress non-linearity in the data, we conclude that the emitted multiplicities, $N_n$ and $N_\gamma$, are also linearly correlated. Thus, we see that the regression analysis of the Glässel et al. data indicates that the neutron and photon multiplicities from $^{252}$Cf are linearly correlated.

C. Summary

When two variables are linearly correlated, their covariance fully quantifies the correlation between them [35, 36]. The covariance between the two multiplicities is defined as

$$\text{cov}(N_n, N_\gamma) = \langle N_n N_\gamma \rangle - \langle N_n \rangle \langle N_\gamma \rangle.$$  \hspace{0.5cm} (4)

Linearity is conserved when the system response can be approximated by the binomial-response model. The preservation of linearity allows us to fully characterize the correlation in the data using

$$\text{cov}(D_n, D_\gamma) = \langle D_n D_\gamma \rangle - \langle D_n \rangle \langle D_\gamma \rangle.$$  \hspace{0.5cm} (5)

IV. EXPERIMENT

We analyze the data collected and reported by Marcath et al. [20] using the Chi-Nu array at Los Alamos National Laboratory. The data were collected continuously over the course of 1.5 hours. On the day of the experiment, the fission source was calculated to be (by activity) 99.8% $^{252}$Cf, 0.2% $^{250}$Cf, and 0.005% $^{248}$Cm [20].

The Chi-Nu array comprises up to 54 EJ-309 liquid organic scintillators, each 17.78 cm in diameter and 5.08 cm inches thick. These detectors are distributed on a hemispherical surface.
centered on the fissioning source, at a mean distance of 1 m. During our measurement, 42 detectors were active. We record variations of the order of 2 – 3 cm in distance between the source and each of the detectors. The overall solid angle coverage provided by the 42 detectors was 0.083 × 4π sr, with an intrinsic efficiency of approximately 32% and 23% for neutrons and photons, respectively. We derive precise values of the absolute detection efficiency of the system by comparing the experimentally-determined mean multiplicities with published nuclear data in Section V. Due to the anisotropy in the emission of neutrons in the laboratory frame, we note that the hemispherical configuration of Chi-Nu biases towards the collection of neutrons from a single fragment, rather than homogeneously detecting neutrons from both fragments. Because photon emission is approximately isotropic in the lab frame, the geometric bias is not important for photon detection.

At the center of the scintillator array, \(^{252}\text{Cf}\) is deposited on the thin metal surface of an ion chamber that also serves as a fission fragment detector. We require each fission to be tagged by the ion chamber in order to be recorded and allow detection in the external scintillators. Fission tagging plays a triple role in the experiment: it provides a time-tag and time of flight (ToF) for all subsequent detections; it reduces the impact of background; and it restricts the set of measured fissions to the defined set of tagged fissions.

Due to data throughput limitations, only events in which the ion chamber trigger was followed by a detection in at least one of the scintillators were recorded. Consequently, we do not have a direct measurement for the 0-0 bin, i.e., a fission event tagged in the ion chamber with no subsequent detection in the scintillators.

Data were collected for 150 ns before and after the trigger from the ion chamber. Data collected before the trigger represent the background multiplicity distribution, while data recorded after the trigger represent the convolution of the background multiplicity distribution and the neutron-photon multiplicity distribution from the fission source. The data are presented in Table I.

| \(D_n\) | \(D_n = 0\) | \(D_n = 1\) | \(D_n = 2\) | \(D_n = 3\) |
|--------|------------|------------|------------|------------|
| \(D_n = 0\) | No data | \((1.9645 ± 0.0006) \times 10^{-1}\) | \((8.65 ± 0.01) \times 10^{-3}\) | \((2.309 ± 0.022) \times 10^{-4}\) |
| \(D_n = 1\) | \((2.955 ± 0.004) \times 10^{-1}\) | \((2.81 ± 0.01) \times 10^{-2}\) | \((1.190 ± 0.003) \times 10^{-3}\) | \((3.07 ± 0.05) \times 10^{-5}\) |
| \(D_n = 2\) | \((2.574 ± 0.005) \times 10^{-2}\) | \((2.309 ± 0.015) \times 10^{-3}\) | \((9.66 ± 0.05) \times 10^{-5}\) | \((2.4 ± 0.1) \times 10^{-6}\) |
| \(D_n = 3\) | \((1.663 ± 0.008) \times 10^{-3}\) | \((1.487 ± 0.03) \times 10^{-4}\) | \((5.85 ± 0.07) \times 10^{-6}\) | \((1.6 ± 0.2) \times 10^{-7}\) |
| \(D_n = 4\) | \((8.69 ± 0.11) \times 10^{-5}\) | \((7.45 ± 0.04) \times 10^{-6}\) | \((2.87 ± 0.09) \times 10^{-7}\) | \((7 ± 2) \times 10^{-9}\) |

Neutrons and photons from a tagged fission event deposit energy in liquid organic scintillators primarily through elastic scattering \([3, 37]\). In elastic scattering, only a portion of the kinetic energy of the particle is deposited in the detector material. The energy deposited in these reactions is then converted to an electrical signal, digitized at a frequency of 500 MHz by three 16-channel CAEN-V1730 digitizers with a dynamic range of 2 V \([20]\). Two types of pulses are removed immediately from further analysis: pulses with signal voltage below the signal threshold, and pulses with signal voltage higher than the maximum of the dynamic range, i.e., clipped pulses.

With a calibration of 477 keVee (electron recoil equivalent keV) at 0.3 V, the maximum allowed energy deposited by a photon is approximately 3.3 MeV. The maximum energy of neutron deposition on hydrogen is approximately 8.1 MeV \([38]\). Particles with initial
energies above the maximum energies mentioned here can be detected by our system, as long as the energy deposition is below the maximum of the dynamic range. However, we expect a bias against very energetic particles due to pulse clipping [20].

We apply a minimum light output threshold of 100 keVee. Below this threshold, pulse shape discrimination (PSD) is not reliable and the particle type cannot be assigned unambiguously. Photons with energies below 220 keV and neutrons with energies below approximately 800 keV are not recorded by our detectors. Our dynamic range is thus from 0.22 to 3.3 MeV for photons and from 0.8 to 8.1 MeV for neutrons. The detection system is most sensitive to the lower energy region, and the detection efficiency decreases with higher energies. A detailed analysis of the spectral sensitivity of the detection system is shown in Fig. 7 of Ref. [20]. Although different portions of the neutron and photon spectra may be correlated differently, the dependence of the multiplicity correlation on energy of the emitted particles will be the subject of future work.

We can assign a particle identity to each measured event in an EJ-309 scintillator through PSD, as outlined in Refs. [37, 39, 40]. Discrimination between neutrons and photons is performed in a two-step process: ToF and PSD. A particle needs to satisfy both ToF and PSD requirements for it to be considered a valid neutron or photon detection. An upper-bound estimate on the misclassification rate of 1% was determined by Marcath et al. [20].

Double pulses are removed in post-processing because the PSD method requires the examination of the tail of the pulse. This analysis cannot be performed for overlaid double pulses. By removing double pulses, we also remove events in which a photon and a neutron both deposit energy in the same detector, i.e., photon-neutron pileup events. This rare effect is a significant bias in the multiplicity correlation.

V. DATA ANALYSIS

The most direct way to compute the covariance in the data is to reconstruct the value of the missing 0-0 bin from source activity and then compute the covariance of the reconstructed distribution using Eq. (5). However, although the total number of fissions that occurred over the time of the measurement can be reconstructed with good accuracy, the number of tagged fissions includes an uncertainty due to the efficiency of the ion chamber, only approximately known. Thus, reconstructing the 0-0 bin can lead to a significant uncertainty. In Appendix B we show that uncertainties in the probability of the 0-0 bin, \( \delta [P(D_n = 0, D_\gamma = 0)] \), are translated to uncertainties in the multiplicity covariance, \( \delta [\text{cov}(D_n, D_\gamma)] \), as

\[
\frac{\delta [\text{cov}(D_n, D_\gamma)]}{\text{cov}(D_n, D_\gamma)} = -\delta [P(D_n = 0, D_\gamma = 0)] \left(1 + \frac{\langle D_n \rangle \langle D_\gamma \rangle}{\text{cov}(D_n, D_\gamma)} \right),
\]

where \( P(D = d) \) is the probability that the detected multiplicity of either neutrons or photons, depending on subscript, has value \( d \). Using the results for \( \langle D_n \rangle \), \( \langle D_\gamma \rangle \), and \( \text{cov}(D_n, D_\gamma) \) determined at the end of this section, we estimate that a 5% relative uncertainty in the fission tagging probability leads to a 100% relative uncertainty in the covariance between the detected multiplicities. Because of these limitations, we choose to compute the covariance using regression analysis, thus circumventing the problem of reconstructing the 0-0 bin.

We assume linearly-correlated emission, along with binomial response, to find the linear regression slopes of the data in the absence of complete information. We use the Pearson correlation coefficient, computed as the geometric average of the two least-square linear
regression slopes \([36]\),
\[
\rho^2(D_n, D_\gamma) = \alpha_n(D_n, D_\gamma)\alpha_\gamma(D_n, D_\gamma) ,
\]
(7)
to quantify the correlation between neutron and photon multiplicities. In Appendix A we show that the regression slopes that define the Pearson correlation coefficient are the same as obtained from performing weighted linear regression analysis of the conditional mean neutron and photon multiplicities. The Pearson correlation coefficient is related to the covariance by \([35]\)
\[
\rho(D_n, D_\gamma) = \frac{\text{cov}(D_n, D_\gamma)}{\sigma(D_n)\sigma(D_\gamma)} ,
\]
(8)
where \(\sigma(D)\) is the standard deviation of the distribution of \(D\). Using Eq. (7), we find
\[
\text{cov}^2(D_n, D_\gamma) = \alpha_n(D_n, D_\gamma)\alpha_\gamma(D_n, D_\gamma)\sigma^2(D_n)\sigma^2(D_\gamma) .
\]
(9)
We determine the regression slopes \(\alpha_n\) and \(\alpha_\gamma\) associated with the experimental result, through a weighted least-squares linear fit of the conditional means. The result of the regression analysis from the data is shown in Fig. 4. The deviation from linearity at higher detected multiplicities is not surprising, because the linearity conditions in Eq. (2) is valid only close to the mean multiplicity. We note that every detected multiplicity most likely originates from a much higher emitted multiplicity due to the low detection efficiency. Therefore the appearance of non-linearity at \(d_n \geq 3\) and \(d_\gamma > 4\) partially reflects the non-linearity of the correlation between particles at a much larger emitted multiplicity. Using Eq. (8) and the regressions shown in Fig. 4 we find
\[
\rho(D_n, D_\gamma) = -0.00418 \pm 0.00009 ,
\]
(10)
where the sign is determined from the sign of the regression slopes, which is negative for both \(\alpha_n\) and \(\alpha_\gamma\). The uncertainty is the standard error of the mean of 40 two-minutes measurements taken over the course of the experiment.

To compute the covariance using Eq. (9), we need the standard deviations of the detected neutron and photon multiplicity distributions. The standard deviations depend on the 0-0 bin and thus cannot be computed directly from the detected multiplicity distribution. However, we may approximate the value of the standard deviation in terms of quantities that we can experimentally determine. Given our low detection efficiency we have \([24, 41]\)
\[
\sigma^2(D_n) = \langle D_n \rangle + \mathcal{O}(\epsilon_n^2) ,
\]
\[
\sigma^2(D_\gamma) = \langle D_\gamma \rangle + \mathcal{O}(\epsilon_\gamma^2) ,
\]
(11a)
(11b)
where \(\epsilon_n\) and \(\epsilon_\gamma\) are the absolute detection efficiencies of the system. Simulations of the system with MCNPX-PoliMi predict that the error introduced in approximating the standard deviation using Eq. (11a) is \(\approx 3%\) for neutrons and \(\approx 0.4%\) for photons. While we note that the mean detected multiplicities \(\langle D_n \rangle\) and \(\langle D_\gamma \rangle\) depend linearly on the efficiencies, \(\epsilon_n\) and \(\epsilon_\gamma\), we defer the computation of these quantities to the end of this section. We combine the results of the regression analysis with our estimates of the standard deviations using Eq. (9).

Using Eq. (9), (10), and (11a), the covariance in the data is then
\[
\text{cov}(D_n, D_\gamma) = (-0.00418 \pm 0.00009) \sqrt{\langle D_n \rangle \langle D_\gamma \rangle} + \mathcal{O}(\epsilon_n, \epsilon_\gamma) .
\]
(12)
Background is found to have little influence on the data, due to a combination of time-gates, fission tagging, and overall low background activity. Because there is no measurement of
FIG. 4. Regression plots of neutron and photon multiplicity from $^{252}\text{Cf}$ for our experiment, for both (a) mean photon multiplicity on neutron multiplicity and (b) mean neutron multiplicity on photon multiplicity. We note that the $y$ axes on both (a) and (b) are zero-suppressed and that the scales on the $y$ axes are different.

In the 0-0 bin, the de-convolution of the background distribution from the $^{252}\text{Cf}$ signal cannot be performed directly. Fortunately, this de-convolution is not required. The multiplicity covariance of the measured source is changed by the presence of a source-independent background (see Appendix C),

$$\text{cov}(\mathbf{D}_n, \mathbf{D}_\gamma) \rightarrow \text{cov}(\mathbf{D}_n, \mathbf{D}_\gamma) + \text{cov}(\mathbf{B}_n, \mathbf{B}_\gamma) ,$$

(13)

where $\text{cov}(\mathbf{B}_n, \mathbf{B}_\gamma)$ is the covariance between background neutron multiplicity and background photon multiplicity, $\mathbf{B}_n$ and $\mathbf{B}_\gamma$ respectively. Because of the way background is collected, we have a complete background distribution, including a 0-0 frequency. We can compute the background covariance directly without using regression, finding

$$\text{cov}(\mathbf{B}_n, \mathbf{B}_\gamma) = -(6.4 \pm 0.2) \times 10^{-6} .$$

(14)

Another source of bias we identify is particle misclassification. Several factors mitigate the effect of misclassification bias. First, the energy threshold of 100 keVee improves the accuracy
of the PSD algorithm at the expense of a reduced absolute detection efficiency. Second, because photons travel at the speed of light while neutrons do not, we can also discriminate neutrons and photons based on their ToF. Although longer-lived isomeric fragment states contribute to a delay in the photon emission times for some fragments, the overall time scale of detection makes these differences negligible in the current experimental setup \[42\]. After particle discrimination, based on both PSD and ToF requirements, the possibility of misclassification is small but not negligible. The effect of particle misclassification is to transform the covariance as

\[
\text{cov}(D_n, D_\gamma) \rightarrow (1-2\theta)\text{cov}(D_n, D_\gamma) + \theta \left[ \sigma^2(D_n) - \langle D_n \rangle \right] + \theta \left[ \sigma^2(D_\gamma) - \langle D_\gamma \rangle \right] + O(\theta^2), \tag{15}
\]

where \(\theta\) is the probability of a neutron being misclassified as a photon and vice versa. The details of the derivation of Eq. \((15)\) are presented in Appendix C. As mentioned in Section IV, the estimated probability of a photon being classified as a neutron is less than 1%. However, this estimate does not consider ToF coincidences applied during the measurement that significantly reduce misclassification.

Having considered the effects of background and particle misclassification, we unfold the covariance assuming a binomial-response model of particle detection. When a binomial-response model is valid, the covariance at emission and the covariance at detection are connected through

\[
\text{cov}_{\text{Bin}}(N_n, N_\gamma) = \frac{\text{cov}(D_n, D_\gamma)}{\epsilon_n \epsilon_\gamma}. \tag{16}
\]

We infer the detection efficiencies, \(\epsilon_n\) and \(\epsilon_\gamma\), by comparing the detected mean multiplicities with published nuclear data for the mean neutron and photon multiplicities from the spontaneous fission of \(^{252}\text{Cf}\). As stated in Section I, although the event-by-event multiplicity correlation is not known, the mean neutron and photon multiplicities from \(^{252}\text{Cf}\) are known to good accuracy.

We compute the efficiencies appearing in Eq. \((16)\) using Eq. \((2)\). We find that the intercepts of the linear regressions, i.e., the extrapolated mean number of detected neutrons when no photons are detected and vice versa, contain information on the global mean of the detected probability distribution. We present this relation as

\[
\begin{pmatrix}
1 & -\alpha_n(D_n, D_\gamma) \\
-\alpha_\gamma(D_n, D_\gamma) & 1
\end{pmatrix}
\begin{pmatrix}
\langle D_n \rangle \\
\langle D_\gamma \rangle
\end{pmatrix}
= \begin{pmatrix}
\langle D_n | D_\gamma = 0 \rangle \\
\langle D_\gamma | D_n = 0 \rangle
\end{pmatrix}. \tag{17}
\]

The quantities on the right-hand-side of Eq. \((17)\) are the intercepts of the linear fit. The off-diagonal elements in the matrix on the left-hand-side are the regression slopes of the linear fit. Both the intercepts and the slopes are given in the legends of Fig. 4. Using Eq. \((17)\), we find the mean multiplicities \(\langle D_n \rangle\) and \(\langle D_\gamma \rangle\):

\[
\begin{align*}
\langle D_n \rangle &= 0.09681 \pm 0.0001, \tag{18a} \\
\langle D_\gamma \rangle &= 0.1519 \pm 0.0001. \tag{18b}
\end{align*}
\]

The uncertainties we reported on the mean detected multiplicities in Eq. \((18)\) are only statistical. We use published data for the mean emitted multiplicities \(\langle N_n \rangle\) and \(\langle N_\gamma \rangle\) from Santi \textit{et al.} \[13\] and Oberstedt \textit{et al.} \[32\], respectively (an infrared energy cutoff of 100 keV is reported for photons)

\[
\begin{align*}
\langle N_n \rangle &= 3.757 \pm 0.010, \tag{19a} \\
\langle N_\gamma \rangle &= 8.29 \pm 0.13. \tag{19b}
\end{align*}
\]
We note that several values of the mean photon multiplicity have been reported that are not all in agreement with each other [32]. Therefore, the uncertainty in the mean photon multiplicity is likely underestimated. However, the effect of a larger uncertainty does not significantly alter the conclusions drawn in this work. We compute the absolute detection efficiencies for neutron and photon detection substituting Eqs. (18)-(19) in Eq. (1),

$$\epsilon_n = 0.0258 \pm 0.0001 ,$$  \hspace{1cm} (20a)

$$\epsilon_\gamma = 0.0183 \pm 0.0003 .$$  \hspace{1cm} (20b)

The uncertainties on the efficiencies are from standard errors in fitting, statistical uncertainties, and uncertainties on the mean emitted multiplicities. We can now readily compute the unfolded covariance of Eq. (16), using the efficiencies we have just obtained with our estimates of $\langle D_n \rangle$ and $\langle D_\gamma \rangle$. We find

$$\frac{\text{cov}(D_n, D_\gamma)}{\epsilon_n\epsilon_\gamma} = \sqrt{\langle D_n \rangle \langle D_\gamma \rangle} \left( \frac{\epsilon(D_n, D_\gamma)}{\epsilon_n\epsilon_\gamma} \right) + \mathcal{O}(\epsilon_n, \epsilon_\gamma)$$

$$= -1.045 \pm 0.028 + \mathcal{O}(\epsilon_n, \epsilon_\gamma) .$$  \hspace{1cm} (21)

Photon-neutron pileup represents the largest bias in the covariance we have determined in Eq. (21). Because the probability of two particles interacting in the same detector is much smaller than the probability of the two particles interacting in different detectors (by a factor proportional to the number of detectors employed, 42 here), we treat pileup as a perturbative correction to the binomial response. In Appendix D, we show that the first order perturbative corrections to the covariance due to pileup correction can be computed from the joint probability of detecting a neutron and a photon in coincidence. Although the first particle, which we consider to be a photon without loss of generality, can be detected by $k$ detectors each with probability $\epsilon_\gamma/k$, the neutron can now only be detected by $k - 1$ detectors each with probability $\epsilon_n/k$. The probability of detecting both particles is then

$$\mathbb{P}(D_n = 1, D_\gamma = 1|N_n = 1, N_\gamma = 1) = k\frac{\epsilon_\gamma}{k} \times (k - 1)\frac{\epsilon_n}{k} = \epsilon_n\epsilon_\gamma - \frac{\epsilon_n\epsilon_\gamma}{k} .$$  \hspace{1cm} (22)

Using Eq. (D24), we see that the second term on the right-hand-side of Eq. (22) is the correction to the joint neutron-photon detection efficiency, $\tilde{C}_{1,1}$. Using Eq. (D22), we find

$$\text{cov}(N_n, N_\gamma) = \frac{\text{cov}(D_n, D_\gamma)}{\epsilon_n\epsilon_\gamma} + \frac{\langle N_n N_\gamma \rangle}{k} + \mathcal{O}(\epsilon_n, \epsilon_\gamma) .$$  \hspace{1cm} (23)

The emitted mean joint neutron-photon multiplicity, $\langle N_n N_\gamma \rangle$, depends implicitly on the covariance. Using Eq. (4) and retaining only terms at leading order, we can rewrite Eq. (23) in terms of $\langle N_n \rangle \langle N_\gamma \rangle$ and the number of detectors, as

$$\text{cov}(N_n, N_\gamma) = \frac{k}{k - 1} \frac{\text{cov}(D_n, D_\gamma)}{\epsilon_n\epsilon_\gamma} + \frac{\langle N_n \rangle \langle N_\gamma \rangle}{k - 1} .$$  \hspace{1cm} (24)

Considering the effects of background, Eq. (13), and misclassification, Eq. (15) and Eq. (11a), we express the covariance in the emitted distribution as

$$\text{cov}(N_n, N_\gamma) = \frac{k}{k - 1} \frac{1}{\epsilon_n\epsilon_\gamma} \left( \frac{\text{cov}(D_n, D_\gamma)}{1 - 2\theta} - \text{cov}(B_n, B_\gamma) \right)$$

$$+ \frac{\langle N_n \rangle \langle N_\gamma \rangle}{k - 1} + \mathcal{O}(\epsilon_n, \epsilon_\gamma, \theta^2) .$$  \hspace{1cm} (25)
Using the unfolding relation in Eq. \((25)\), we find the covariance at emission,
\[
\text{cov}(N_n, N_\gamma) = -0.329 \pm 0.053 .
\]

We estimate the uncertainties on the covariance in Eq. \((26)\) from statistical uncertainties, non-linear components in the emission models, neglected terms in Eq. \((25)\), and uncertainties on the mean neutron and photon multiplicities in Eq. \((19)\).

VI. DISCUSSION

The null hypothesis, \(i.e.,\) the hypothesis that there is no neutron-photon multiplicity correlation in the emission \(^{252}\text{Cf}(sf)\), is not supported by the present experimental result. The result of a correlation is statistically significant to the \(> 5\sigma\) level. Assuming linearly-correlated emission, we infer the complete joint neutron-photon multiplicity distribution,
\[
\mathbb{P}(N_n = n_n, N_\gamma = n_\gamma) = \mathbb{P}(N_n = n_n)\mathbb{P}(N_\gamma = n_\gamma) \left[ 1 + \text{cov}(N_n, N_\gamma) \frac{n_n - \langle N_n \rangle}{\sigma^2(N_n)} \frac{n_\gamma - \langle N_\gamma \rangle}{\sigma^2(N_\gamma)} \right] .
\]

The unfolded multiplicity distribution is presented in Fig. 5. In the figure, we use the unfolded value of the covariance given in Eq. \((26)\) and the multiplicity-uncorrelated neutron-photon distributions, \(\mathbb{P}(N_n = n_n)\) and \(\mathbb{P}(N_\gamma = n_\gamma)\), included in MCNPX-PoliMi, \(i.e.,\) PoliMi. By doing this, we introduce a correlation in the initially uncorrelated model. Superimposed on the unfolded multiplicity distribution, are the two regression slopes of the unfolded emitted distribution, given as a cone of half width of \(1\sigma\). If emission were completely uncorrelated, the regression slopes would be perpendicular to each other and parallel to the axes, as shown by the dashed lines.

Given the covariance presented in Eq. \((26)\) and the standard deviations of the emitted neutron and photon multiplicity distributions in Refs. 43 and 33, using Eq. \((8)\) with \(D\) instead of \(N\), we find
\[
\rho(N_n, N_\gamma) = -0.072 \pm 0.011 .
\]
A. Covariance of the Fission Event Generators

The unfolded neutron-photon multiplicity covariance, Eq. (26), is smaller in magnitude than the multiplicity covariances predicted by the fission event generators CGMF and FREYA. The covariances predicted by these models, integrated over the entire energy spectrum of both particles and in all directions, can be computed from the models using Eq. (4)

\[
\text{cov}_{\text{FREYA}}(N_n, N_\gamma) = -0.8200 \pm 0.0004 , \\
\text{cov}_{\text{CGMF}}(N_n, N_\gamma) = -0.839 \pm 0.004 .
\]

The uncertainties on these quantities are due purely to limited computational run time. The result in Eq. (26) shows that the experimentally-determined correlation is smaller than that predicted by both CGMF and FREYA. This conclusion is statistically significant to 5σ.

The issue of energy-biasing is relevant here. To estimate the energy nonuniformity of the multiplicity correlations predicted by the fission event generators, we use MCNPX-PoliMi and a high fidelity simulation of the Chi-Nu array, with simulated neutrons and photons generated by FREYA. CGMF was not used for this purpose because the number of simulated fission events available is insufficient to draw significant conclusions. We apply the same unfolding we used in the experiment to the simulations. Our preliminary result shows that, after taking into account the sensitivity of our system to the energy spectra and direction of the emitted neutrons and photons, a covariance closer to \(-0.50 \pm 0.02\) would have been determined in experiment if FREYA were correct. This value of the FREYA covariance is still in disagreement with our experimental observation but further studies of the energy dependence of the correlation are required and will be pursued in future work. Our preliminary results show that the energy dependence of correlations may have a significant effect on the unfolded covariance.

B. Covariance Determined in Prior Experiments

Glässel et al. [21] reported a value of \(-0.02\) (with no uncertainty) for \(\alpha_n(D_n, N_\gamma)\), the regression slope of the detected mean neutron multiplicity on the photon multiplicity in the spontaneous fission of \(^{252}\text{Cf}\). Our analysis of their data, see Fig. 3 in this work, yields a regression slope of \(\alpha_n(D_n, N_\gamma) = -0.0247 \pm 0.0007\). Using the linear regression shown in Fig. 3 and the assumed emitted photon distribution, we find a mean detected neutron multiplicity of \(\langle D_n \rangle_{\text{Glässel}} = 1.50 \pm 0.01\). Using Eq. (1) and the mean neutron multiplicity from \(^{252}\text{Cf}(sf)\) [43], we compute a detection efficiency of \(\epsilon_n = 0.400 \pm 0.003\). The Glässel et al. result, corrected for efficiency losses is

\[
\alpha_n(N_n, N_\gamma) = \frac{1}{\epsilon_n} \alpha_n(D_n, N_\gamma) = -0.062 \pm 0.002 .
\]

We use the relationship between regression slopes and covariance, namely [35, 36]

\[
\text{cov}(N_n, N_\gamma) = \alpha_n(N_n, N_\gamma) \sigma^2(N_\gamma) ,
\]

to estimate the covariance of the Glässel et al. data. We determine the variance of the photon multiplicity distribution from the width parameter \(\Delta_\gamma\) given in Ref. [34] and included in the evaluation by Valentine [33]. This yields \(\sigma^2(N_\gamma) = 13.4 \pm 1.1\). Finally, we find

\[
\text{cov}_{\text{Glässel}}(N_n, N_\gamma) \approx -0.84 \pm 0.07 .
\]
We note that this result did not take into account the effect of photon-neutron pileup. Schmid-Fabian [26] estimates pileup to account for 15% of the correlation in the measured data. Our own perturbative pileup correction, Eq. (24) when higher order terms are not truncated, predicts a similar estimate of the impact of photon-neutron pileup (≈ 13%). Correcting for photon-neutron pileup, we find

\[ \text{cov}_{\text{Glässl}}(N_n, N_\gamma) \approx -0.71 \pm 0.06. \] (33)

The covariance determined in Eq. (33) is in agreement with CGMF and FREYA, see Eq. (29). It is larger in magnitude, and of the same sign, as the covariance determined in this work. Although not in quantitative agreement, the conclusion that there exists a competition in the emitted neutron and photon multiplicities from \(^{252}\text{Cf}(sf)\) is in qualitative agreement with our conclusion.

Energy sensitivity arguments are applicable to the covariance unfolded in Eq. (33). The NaI(Tl) inorganic scintillator used in Glässl et al. [21, 26, 31] detect neutrons primarily via the inelastic scattering on \(^{127}\text{I}\) [44]. The energy sensitivity of NaI(Tl) detectors is thus not expected to agree with the energy sensitivity of the organic scintillators we employ.

The result reported in Bleuel et al. suggests that the multiplicity correlation in \(^{252}\text{Cf}\) is small or null for at least some portion of the fission fragment yields. Bleuel et al. measured event-by-event correlations by identifying the fragment using \(\gamma\)-ray spectroscopy. The mean photon multiplicity regression on the neutron multiplicity was computed by measuring the photon multiplicity distribution from fragment pairs with different neutron multiplicities, \(i.e., N_n = 2\) for \(^{106}\text{Mo} + ^{144}\text{Ba}\), and \(N_n = 4\) for \(^{106}\text{Mo} + ^{142}\text{Ba}\). Bleuel et al. did not measure neutrons, effectively eliminating the issue of photon-neutron pileup and the need for a corrected unfolding.

By measuring the detected mean photon multiplicity for the two fragment pairs, Bleuel et al. computed a regression slope of detected photon multiplicity on emitted neutron multiplicity, \(\alpha_\gamma(N_n, D_\gamma)\), and found this to be approximately 0. Therefore,

\[ \text{cov}_{\text{Bleuel}}(N_n, N_\gamma) \approx 0. \] (34)

The observation of Bleuel et al. is thus consistent with uncorrelated emission. However, we note that the results of Bleuel et al. were derived from two fragment pairs only and thus cannot be directly compared with our results that are averaged over all fission fragments. Furthermore, we note that a correlation of the magnitude indicated in this work would have likely been too small to be observed by Bleuel et al.

VII. SUMMARY AND CONCLUSION

We have presented evidence that there exists a negative event-by-event correlation in neutron and photon multiplicities in the prompt emission following the spontaneous fission of \(^{252}\text{Cf}\). The magnitude of this correlation is quantified by the covariance, \(\text{cov}(N_n, N_\gamma) = -0.244 \pm 0.046\). The discussion and comparison of our results with other experiments revealed several discordant behavior, even after efficiency and pileup corrections are performed. First, we note that it is possible that different portions of the neutron and photon spectra will have different multiplicity correlations. Our experimental result assumes that the multiplicity correlation between neutrons and photons is uniform over the entire energy range and that the spectral sensitivity of our system is a good representation of the correlation across the
entire energy range. Future work will investigate the predictions made by physical models of an energy-dependent multiplicity correlation. Next, we note that the discordant result of Bleuel et al. may be interpreted as a dependence of the multiplicity correlation on the fragment identity. In future experiments, it will be important to detect fragments in coincidence with neutrons and photons.

Aside from the results presented here, the question of a possible correlation between momenta and multiplicities, for both neutrons and photons, remains. Some work has already been performed [13, 45, 46], but no definitive conclusion has been reached. Although determining these correlations would represent an important milestone in the characterization of fission signatures from $^{252}$Cf(sf), the same treatment should be applied to other isotopes undergoing spontaneous fission, notably $^{240}$Pu, as well as for isotopes undergoing neutron-induced fission, especially at higher incident energies where multi-chance fission and pre-equilibrium contributions would most likely complicate these correlations. The complexity introduced by incident neutrons is a necessary obstacle to be overcome, given the importance of induced-fission reactions for applications involving the multiplication properties of fission [47–49].

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Appendix A: Correlation from Regression Means

The equivalence of applying weighted linear regression on the conditional mean multiplicities and of standard least-square linear regression is proved in this appendix. Consider a set of events drawn from a multiplicity distribution $(N_n, N_\gamma)$. Note that the following arguments apply equally well to the detected multiplicities $(D_n, D_\gamma)$. Every event is an ordered pair, $(n_n, n_\gamma)$, where $n_n$ and $n_\gamma$ are non-negative integers. We obtain one regression slope from the data by applying least-square regression analysis, i.e., we find the linear fit that minimizes the total sum of the residuals. We obtain the other regression slope by applying a linear fit to the same set of events, but switching the roles of dependent and independent variables. An elementary theory of statistics [35] states that the product of the two regression slopes thus obtained is the square of the Pearson correlation coefficient between the variables $N_n$.
and $N_\gamma$,  
\begin{equation} 
\alpha_n(N_n, N_\gamma) \alpha_\gamma(N_n, N_\gamma) = \rho^2(N_n, N_\gamma) , 
\end{equation}
where $\alpha_n$ is the regression slope for dependent variable $N_n$, and analogously for $\alpha_\gamma$.

Without loss of generality, consider the regression slope of $N_n$ on $N_\gamma$. Let us then suppose regression analysis of the entire set of events yields the linear fit with slope $\alpha_n$ and intercept $b$. By definition, these parameters minimize the sum of the squares of the residuals of the fit. For each event $(n_n, n_\gamma)$, the residual of the fit for that point is
\[ n_n - (\alpha_n n_\gamma + b) . \]

The sum of all residuals squared is then 
\begin{equation} 
|\text{res}(\alpha_n, b)|^2 = \sum_{n_n, n_\gamma \in \mathbb{Z}_{\geq 0}} \text{mult}(n_n, n_\gamma) \times (n_n - (\alpha_n n_\gamma + b))^2 , 
\end{equation}
where $\text{mult}(n_n, n_\gamma)$ is the number of events with identical joint multiplicity $(n_n, n_\gamma)$, and the set $\mathbb{Z}_{\geq 0}$ is the set of all possible multiplicities. We divide the number of events of a given multiplicity by the total number of events, $\mathcal{N}$, to obtain
\begin{equation} 
\lim_{N \to \infty} \frac{\text{mult}(n_n, n_\gamma)}{\mathcal{N}} \approx \mathbb{P}(N_n = n_n, N_\gamma = n_\gamma). 
\end{equation}
Furthermore, we express the right-hand-side of the above equation as
\begin{equation} 
\mathbb{P}(N_n = n_n, N_\gamma = n_\gamma) = \mathbb{P}(N_n = n_n | N_\gamma = n_\gamma) \mathbb{P}(N_\gamma = n_\gamma) . \end{equation}

We divide both sides of Eq. (A2) by the total number of events, $\mathcal{N}$, and obtain
\begin{equation} 
\frac{|\text{res}(\alpha_n, b)|^2}{\mathcal{N}} = \sum_{n_n, n_\gamma \in \mathbb{Z}_{\geq 0}} \mathbb{P}(N_n = n_n | N_\gamma = n_\gamma) \mathbb{P}(N_\gamma = n_\gamma) (n_n - (\alpha_n n_\gamma + b))^2 . \end{equation}

After performing the summation over $n_n$, we find
\begin{equation} 
\frac{|\text{res}(\alpha_n, b)|^2}{\mathcal{N}} = |\text{Res}(\alpha_n, b)|^2 + \sum_{n_\gamma \in \mathbb{Z}_{\geq 0}} \mathbb{P}(N_\gamma = n_\gamma) \sigma^2(N_n | N_\gamma = n_\gamma) ,
\end{equation}
with
\begin{equation} 
|\text{Res}(\alpha_n, b)|^2 = \sum_{n_\gamma \in \mathbb{Z}_{\geq 0}} \mathbb{P}(N_\gamma = n_\gamma) \left[ \langle N_n | N_\gamma = n_\gamma \rangle - (\alpha_n n_\gamma + b) \right]^2 . \end{equation}

The term $|\text{Res}(\alpha_n, b)|^2$ is the sum of residuals of conditional mean multiplicities weighted by the probability of the photon multiplicity. The second term in Eq. (A6) simplifies to the variance of the neutron multiplicity distribution, $\sigma^2(N_n)$. Therefore, we find
\begin{equation} 
|\text{Res}(\alpha_n, b)|^2 = \frac{|\text{res}(\alpha_n, b)|^2}{\mathcal{N}} - \sigma^2(N_n) . \end{equation}

The number of events, $\mathcal{N}$, is a positive number. The variance $\sigma^2(N_n)$ is independent of the choice of $\alpha_n$ and $b$. Therefore, the global minimum of $|\text{res}(\alpha_n, b)|^2$ is a global minimum of $|\text{Res}(\alpha_n, b)|^2$ as well. The same argument would apply for the case of the slope of $N_\gamma$ on $N_n$. 

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Thus we conclude that a linear regression analysis performed using the conditional mean multiplicities, weighted by the marginal distribution of the multiplicity of the other particle, yields the regression slopes obtained from standard regression analysis. The regression slopes can then be used to compute the Pearson correlation coefficient in the data, as done in Eq. (7). We characterize the linearity of the data using the residuals of the weighted linear regressions. From Eq. (A7), we see that the $|\text{Res}(\alpha_n, b)|^2$ is also the mean value of the squared residuals. Therefore, the root mean square (r.m.s.) value of the residuals from the linear fit is given by $\sqrt{|\text{Res}(\alpha_n, b)|^2}$. In Section III we use this metric to quantify the agreement of data with the linear fit.

**Appendix B: Sensitivity of Covariance to the 0-0 Bin**

We derive the sensitivity of the covariance on the uncertainties of the 0-0 bin, when the covariance is computed directly using Eqs. (4) and (5). Let us take the probability of observing a count belonging to the 0-0 bin to be $\omega_0$, with an associated uncertainty $\omega$

$$p(D_n = 0, D_\gamma = 0) = \omega_0 \pm \omega \quad (B1)$$

Because the probability distribution is normalized, the value of $\omega$ scales all other probabilities in the distribution. Specifically, if the probability distributions is normalized for $\omega = 0$, then all the probabilities in the distribution are scaled by $(1 \pm \omega)^{-1}$. We compute the covariance of the $\omega$-dependent distribution of the detected neutron and photon multiplicity as

$$\text{cov}(D_n, D_\gamma)(\omega) = \langle D_n D_\gamma \rangle(\omega) - \langle D_n \rangle(\omega) \langle D_\gamma \rangle(\omega)$$

$$= \frac{1}{1 \pm \omega} \langle D_n D_\gamma \rangle(0) - \frac{1}{(1 \pm \omega)^2} \langle D_n \rangle(0)\langle D_\gamma \rangle(0) \quad (B2)$$

We perform a Taylor series expansion around $\omega = 0$, retaining only leading order terms in $\omega$

$$\text{cov}(D_n, D_\gamma)(\omega) = (1 \mp \omega) \text{cov}(D_n, D_\gamma)(0) \mp \omega \langle D_n \rangle(0)\langle D_\gamma \rangle(0) + \mathcal{O}(\omega^2) \quad (B3)$$

We then let

$$\delta [\text{cov}(D_n, D_\gamma)] = \text{cov}(D_n, D_\gamma)(\omega) - \text{cov}(D_n, D_\gamma)(0) \quad (B4)$$

and

$$\left[ p(D_n = 0, D_\gamma = 0) \right] = \omega \quad (B5)$$

to obtain Eq. (6) in Section V.

**Appendix C: Covariance of Convolved Distributions**

When multiple neutron and photon sources are present, the detected multiplicity distribution is the convolution of the multiplicity distribution from each source. In this Appendix, we show how the covariance in the neutron-photon multiplicity can be determined from the convolved distribution, without performing a direct deconvolution.
1. Background

Let us consider the detected fission multiplicities of \((D_n, D_\gamma)\) and the background multiplicities \((B_n, B_\gamma)\). We use the properties of covariance to find the covariance of the sum of the two random variables

\[
\text{cov}(D_n + B_n, D_\gamma + B_\gamma) = \text{cov}(D_n, D_\gamma) + \text{cov}(B_n, B_\gamma) + \text{cov}(D_n, B_n) + \text{cov}(D_\gamma, B_\gamma) .
\] (C1)

Because, by assumption, the background multiplicities are uncorrelated with the fission multiplicities, we have

\[
\text{cov}(D_n, B_n) = \text{cov}(D_\gamma, B_\gamma) = 0 .
\] (C2)

Therefore, we obtain the covariance in the convolved distribution as in Eq. (13) in Section V,

\[
\text{cov}(D_n + B_n, D_\gamma + B_\gamma) = \text{cov}(D_n, D_\gamma) + \text{cov}(B_n, B_\gamma) .
\] (C3)

2. Misclassification

We now derive the effects of particle misclassification. We introduce \(\theta_n\) (\(\theta_\gamma\)) as the probability of a neutron (photon) being classified as a photon (neutron). Then, the observed multiplicities after misclassification, \(D_n'\) and \(D_\gamma'\), are given by

\[
D_n' = \text{Bin}(D_n, 1 - \theta_n) + \text{Bin}(D_\gamma, \theta_\gamma) ,
\]
\[
D_\gamma' = \text{Bin}(D_\gamma, 1 - \theta_\gamma) + \text{Bin}(D_n, \theta_n) ,
\]

where \(\text{Bin}(X, p)\) is the binomial distribution with \(X\) trials and probability of success \(p\). The covariance between the observed multiplicities \((D_n', D_\gamma')\) is

\[
\text{cov}(D_n', D_\gamma') = C_{n,\gamma} + c_{n,\gamma} + C_{n,n} + C_{\gamma,\gamma} ,
\] (C4)

with

\[
C_{n,\gamma} = \text{cov} [\text{Bin}(D_n, 1 - \theta_n), \text{Bin}(D_\gamma, 1 - \theta_\gamma)] ,
\] (C5a)

\[
c_{n,\gamma} = \text{cov} [\text{Bin}(D_n, \theta_n), \text{Bin}(D_\gamma, \theta_\gamma)] ,
\] (C5b)

\[
C_{n,n} = \text{cov} [\text{Bin}(D_n, 1 - \theta_n), \text{Bin}(D_n, \theta_n)] ,
\] (C5c)

\[
C_{\gamma,\gamma} = \text{cov} [\text{Bin}(D_\gamma, 1 - \theta_\gamma), \text{Bin}(D_\gamma, \theta_\gamma)] .
\] (C5d)

The misclassification processes are independent of each other, i.e., the number of misclassified neutrons is not correlated with the number of misclassified photons. The first two terms in Eq. (C4) are therefore only scaled by the rate of misclassification

\[
C_{n,\gamma} + c_{n,\gamma} = [(1 - \theta_n)(1 - \theta_\gamma) + \theta_n \theta_\gamma] \text{cov}(D_n, D_\gamma) .
\] (C6)

The third and forth terms are symmetric under index swapping. Without loss of generality, let us consider the neutron term only. Two sources of covariance exist for these terms. The first is the covariance between correctly-classified and misclassified neutrons, that is perfectly anticorrelated. The second is the covariance between the multiplicities. Because the two multiplicities are identical, the covariance is equal to the variance. Combining the two variances,
\[ C_{n,n} = \theta(1 - \theta_n) \left[ \sigma^2(D_n) - \langle D_n \rangle \right], \quad (C7) \]

and similarly for \( C_{\gamma,\gamma} \). We find the observed covariance between observed detected multiplicities,

\[
\text{cov}(D'_n, D'_\gamma) = \left[(1 - \theta_n)(1 - \theta_\gamma) + \theta_n\theta_\gamma \right] \text{cov}(D_n, D_\gamma) \\
+ \theta_n(1 - \theta_n) \left[ \sigma^2(D_n) - \langle D_n \rangle \right] \\
+ \theta_\gamma(1 - \theta_\gamma) \left[ \sigma^2(D_\gamma) - \langle D_\gamma \rangle \right]. \quad (C8)
\]

To derive Eq. (15) in Section V, we assume a common misclassification rate, \( \theta_n = \theta_\gamma = \theta \), and truncate all terms of order \( \theta^2 \).

### Appendix D: Perturbed Binomial Response

We present the mathematical framework for perturbative corrections to the binomial-response model. These corrections are used in Section V to correct the unfolded covariance for photon-neutron pileup.

1. **Single Particle Response**

We begin by considering the relationship between detected and emitted multiplicities for a single particle type. We write the detected multiplicity distribution in terms of the emitted multiplicity distribution,

\[
P(D = i) = \sum_{j \in \mathbb{Z}_{\geq 0}} P(D = i|N = j)P(N = j), \quad (D1)
\]

where the summation is taken over the support set of the probability distribution \( P(N = j) \), which is finite in practical implementations. We express Eq. (D1) as

\[
D_i = R^j_i N_j, \quad (D2)
\]

where summation over repeated indices \( j \) in Eq. (D2) is implied, and

\[
D_i = P(D = i) , \quad (D3a) \\
N_j = P(N = j) , \quad (D3b) \\
R^j_i = P(D = i|N = j) . \quad (D3c)
\]

Eq. (D2) has a simple physical interpretation. The vectors \( D \) and \( N \) represent the detected and emitted multiplicities, related to each other by the system response \( R \). We can express Eq. (D2) as a matrix multiplication

\[
\begin{pmatrix}
P(D = 0) \\
P(D = 1) \\
P(D = 2) \\
\vdots
\end{pmatrix} =
\begin{pmatrix}
P(D = 0|N = 0) & P(D = 0|N = 1) & P(D = 0|N = 2) & \cdots & P(N = 0) \\
P(D = 1|N = 0) & P(D = 1|N = 1) & P(D = 1|N = 2) & \cdots & P(N = 1) \\
P(D = 2|N = 0) & P(D = 2|N = 1) & P(D = 2|N = 2) & \cdots & P(N = 2) \\
\vdots & \vdots & \vdots & \ddots & \vdots
\end{pmatrix},
\]

\[
(D4)
\]
In the binomial-response model, we assume all particle detections are independent of each other and the efficiency of detection, \( \epsilon \), is constant. When these conditions are met, we write the response as

\[
B_j^i(\epsilon) = \mathbb{P}_{\text{Bin}}(D = i | N = j) = \binom{j}{i} \epsilon^i (1 - \epsilon)^{j-i}
\]

or, in matrix form,

\[
B(\epsilon) = \begin{pmatrix}
1 & 1 - \epsilon & (1 - \epsilon)^2 & \cdots \\
0 & \epsilon & 2\epsilon(1 - \epsilon) & \cdots \\
0 & 0 & \epsilon^2 & \cdots \\
\vdots & \vdots & \vdots & \ddots
\end{pmatrix}.
\]

2. Factorial-Moment Base

So far we expressed the elements of the vectors and matrices in what we refer to as the probability base. We introduce the factorial-moment base, defined by the base-transformation matrix

\[
g^i_j = \binom{i}{j}.
\]

The base transformation can be performed on vectors and matrices; for example, the detection vector \( \mathbf{D} \) transforms as

\[
\tilde{\mathbf{D}}_i = g^i_j \mathbf{D}_j.
\]

The components of the vector \( \tilde{\mathbf{D}} \) are the reduced factorial moment of the distribution of \( D \).

In the factorial moment base, we have

\[
\tilde{\mathbf{D}} = \begin{pmatrix}
\langle D \rangle \\
\langle D \rangle^2 \\
\vdots
\end{pmatrix} = \begin{pmatrix}
1 \\
\langle D \rangle \\
\frac{1}{2} \langle D(D-1) \rangle \\
\vdots
\end{pmatrix}.
\]

In the factorial moment base, the binomial response is diagonal and its diagonal elements are powers of the efficiency \( \epsilon \),

\[
\tilde{B}^i_j = \delta^i_j \epsilon^j = \begin{cases} 
\epsilon^j, & \text{for } i = j \\
0, & \text{otherwise}
\end{cases}
\]

The matrix \( \delta^i_j \) is non-zero only for \( i = j \). In matrix form, we have

\[
\tilde{B}(\epsilon) = \begin{pmatrix}
1 & 0 & 0 & \cdots \\
0 & \epsilon & 0 & \cdots \\
0 & 0 & \epsilon^2 & \cdots \\
\vdots & \vdots & \vdots & \ddots
\end{pmatrix}.
\]
3. Neutron-Photon Covariance

When both neutrons and photons are considered, we interpret the transformation from emitted multiplicities to detected multiplicities as

\[ D_{i,k} = R_{i,k}^{j,l} N_{j,l} , \]  
(D12)

with

\[ D_{i,k} = \mathbb{P}(D_n = i, D_\gamma = k) , \]  
(D13a)
\[ N_{j,l} = \mathbb{P}(N_n = j, N_\gamma = l) , \]  
(D13b)
\[ R_{i,k}^{j,l} = \mathbb{P}(D_n = i, D_\gamma = k | N_n = j, N_\gamma = l) . \]  
(D13c)

In the main text, we have presented \( D_{i,k} \) in Table I of Section IV. There, the two indices of the vector were used as the indices of a rectangular array. The system response is treated here as nearly binomial,

\[ R_{i,k}^{j,l} = B^j_i(\epsilon_n) B^l_k(\epsilon_\gamma) + C_{i,k}^{j,l} , \]  
(D14)

where the first term is the double binomial-response model for neutrons and photons, written as the product of the two independent binomial-response models. The perturbation term, \( C \), is treated as a correction to double binomial-response model. It describes the interdependence of neutron and photon detection. We recall the definition of covariance in the detected multiplicities, presented in Eq. (5) of Section III,

\[ \text{cov}(D_n, D_\gamma) = \langle D_n D_\gamma \rangle - \langle D_n \rangle \langle D_\gamma \rangle . \]  
(D15)

We can now express the quantities defining the covariance as the components of the vector \( \tilde{D} \),

\[ \langle D_n \rangle = \tilde{D}_{1,0} , \]  
(D16a)
\[ \langle D_\gamma \rangle = \tilde{D}_{0,1} , \]  
(D16b)
\[ \langle D_n D_\gamma \rangle = \tilde{D}_{1,1} . \]  
(D16c)

Because the binomial-response model is diagonal in the factorial-moment base, and the eigenvectors of the response are assumed to be unchanged in the first perturbative correction \[50, 51], we conclude that \( \tilde{R} \) is diagonal in the first perturbative correction. The eigenvalues of \( R \), which form the diagonal elements of \( R \), are changed in the first perturbative correction. Therefore, we find that the corrected response is

\[ \tilde{R}_{i,k}^{j,l} = \delta_i^j \delta_k^l \left( \epsilon^j \epsilon^l + \tilde{C}_{i,k}^{j,l} \right) . \]  
(D17)

Using the approximate binomial-response model, Eq. (D12) becomes

\[ \tilde{D}_{i,k} = \delta_i^j \delta_k^l \left( \epsilon^j \epsilon^l + \tilde{C}_{i,k}^{j,l} \right) \tilde{N}_{j,l} . \]  
(D18)

We use Eq. (D18) to compute the perturbed covariance, through perturbative corrections of the quantities in Eq. (D16), i.e.,

\[ \langle D_n \rangle = \left( \epsilon_n + \tilde{C}^{1,0}_{1,0} \right) \langle N_n \rangle , \]  
(D19a)
\[ \langle D_\gamma \rangle = \left( \epsilon_\gamma + \tilde{C}^{0,1}_{0,1} \right) \langle N_\gamma \rangle , \]  
(D19b)
\[ \langle D_n D_\gamma \rangle = \left( \epsilon_n \epsilon_\gamma + \tilde{C}^{1,1}_{1,1} \right) \langle N_n N_\gamma \rangle . \]  
(D19c)

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A response that strictly decreases the multiplicity of detected multiplicities is called upper triangular. For an upper triangular response, $U_{ij}^j$, the elements in the probability basis are zero for $i > j$. We use Eq. (D7) to show that the first two diagonal elements of an upper triangular response remain unchanged when the response is written in the factorial-moment base,

\begin{align}
U_0^0 &= \tilde{U}_0^0 , \quad \text{(D20a)} \\
U_1^1 &= \tilde{U}_1^1 . \quad \text{(D20b)}
\end{align}

The correction $C$ accounts only for interdependence in neutron and photon, hence the correction is non-zero only when both neutrons and photons are detected in coincidence. Therefore, $C_{i,k}^{j,l}$ vanishes for $i = 0$ or $k = 0$. Pileup corrections can only decrease the detected multiplicities, therefore we shall assume that the correction is upper triangular. (Other corrections, such as inelastic production of neutrons and photons, as well as cross talk, are the subject of future work.) Then, using Eq. (D20),

\[ \tilde{C}_{1,0}^{1,0} = \tilde{C}_{0,1}^{0,1} = 0 . \quad \text{(D21)} \]

Therefore, the covariance in the detected multiplicities is perturbed as

\[ \text{cov}(D_n, D_\gamma) = \left( \epsilon_n \epsilon_\gamma + \tilde{C}_{1,1}^{1,1} \right) \langle N_n N_\gamma \rangle - \epsilon_n \langle N_n \rangle \epsilon_\gamma \langle N_\gamma \rangle , \]

\[ = \epsilon_n \epsilon_\gamma \text{cov}(N_n, N_\gamma) + \tilde{C}_{1,1}^{1,1} \langle N_n N_\gamma \rangle . \quad \text{(D22)} \]

From Eq. (D13c), we write

\[ R_{1,1}^{1,1} = \mathbb{P}(D_n = 1, D_\gamma = 1 | N_n = 1, N_\gamma = 1) , \quad \text{(D23)} \]

so that, using Eq. (D17) and Eq. (D20), we can express

\[ \tilde{C}_{1,1}^{1,1} = C_{1,1}^{1,1} = \mathbb{P}(D_n = 1, D_\gamma = 1 | N_n = 1, N_\gamma = 1) - \epsilon_n \epsilon_\gamma . \quad \text{(D24)} \]

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