

Experimental Measurement of Energy Correlations of Prompt Fission Neutrons from ^{252}Cf

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Abstract

The energy correlation among prompt fission neutrons has not been fully considered in coincidence and multiplicity measurement techniques. To accurately measure and systematically verify this correlation characteristic, this project conducted a dedicated experiment lasting approximately 1200 hours. The experiment utilized a total of 8 CLYC detectors and 16 EJ309 liquid scintillation detectors, achieving precise tagging of fission event times through the detection of prompt fission gamma rays. This study measured the relative ratio between the correlated neutron energy spectrum for neutrons of different energies and the ^{252}Cf fission neutron spectrum, which can provide critical data support for related fission physics research and nuclear technology application development.

Full Text

Preamble

Measurement of the Energy Correlations between Two ^{252}Cf Prompt Fission Neutrons

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Energy correlations among prompt fission neutrons have not yet been incorporated into coincidence and multiplication measurement techniques. To measure and verify these energy correlations, we performed an experiment with a total measurement duration of approximately 1200 hours. The experiment utilized

eight CLYC detectors and sixteen EJ309 liquid scintillation detectors, with the fission moment tagged by measured fission γ -rays. We obtained the relative ratios of the energy spectra of neutrons correlated with different-energy neutrons to the standard ^{252}Cf fission neutron energy spectrum. These results may prove valuable for studies of fission physics and nuclear technology applications.

Keywords: Energy correlations, Prompt fission neutrons, Energy spectrum, Fission γ -rays

Introduction

Neutron coincidence and multiplication measurement techniques have been developed over recent decades as nondestructive assay methods for special nuclear materials and nuclear fuels [1-5]. The underlying physical principle is that more than one prompt fission neutron may be emitted from a spontaneous or induced fission event, resulting in the detection of time-correlated neutron events. Beyond temporal correlation, the energies of neutrons emitted in a single fission event are interdependent. On one hand, the neutron energy distributions depend on the actual number of emitted neutrons [6]. On the other hand, neutron energies are affected by the direction and velocity of the corresponding fission fragments, as most prompt fission neutrons are evaporated from fast-moving fission fragments. Consequently, the energies of neutrons emitted from the same fission fragment or from different fragments moving in opposite directions are connected [7]. However, despite the widespread application of neutron coincidence and multiplication measurement techniques and their status as standard methods in some fields [8], the energy correlations of prompt fission neutrons have not been considered due to the lack of available data. This omission may introduce measurement deviations, particularly for fast neutron coincidence and multiplication techniques, since detection efficiency is energy-dependent.

Several codes capable of simulating neutron correlations in fission reactions on an event-by-event basis have been developed, including MCNPX-POLIMI, CGMF, and FREYA [9-13]. These codes require reliable experimental data for model validation [14, 15]. In 2019, P. F. Schuster et al. conducted the first and only measurement of prompt fission neutron energy correlations in ^{252}Cf . Their experiment employed the Chi-Nu detector array, comprising 54 EJ309 liquid scintillation detectors (17.78 cm diameter and 5.08 cm thickness), to detect prompt fission neutrons, along with a fission chamber containing a ^{252}Cf source (fission rate of $2.98 \times 10^5 \text{ s}^{-1}$) to determine the fission event timing [14, 16]. Their measurements revealed negative and positive correlations between the average energies of neutron pairs emitted at angles of 85° and 175° , respectively. However, as the authors noted, these results were inconclusive because the experimental uncertainties were substantial, with the calculated slopes falling within 2σ of zero [14].

To obtain more detailed data on the energy correlations of ^{252}Cf prompt fission neutrons, we conducted an experimental study. Prior to measurement, we de-

veloped a Monte Carlo simulation to predict the energy correlations and guide the experimental design, with neutron energies determined via time-of-flight measurement.

In most existing measurements, the fission moment (i.e., the start time of neutron flight) has been tagged by detecting fission fragments with a fission chamber [17, 18]. However, this technique may be unsuitable for measuring neutron energy correlations. In a fission chamber, the ^{252}Cf source is typically embedded in a metal foil or plate substrate, causing most fission fragments to lose energy before entering the chamber's sensitive region—particularly those emitted at large angles relative to the substrate normal. This results in significant differences in detection efficiency for fission fragments with different emission angles, potentially introducing measurement deviations since prompt fission neutron energy correlations are associated with the moving direction of the corresponding fission fragments. For ^{252}Cf , approximately 11.6 γ -rays are emitted per spontaneous fission [19]. Because the time interval between emission of nearly all fission γ -rays with energies exceeding 0.3 MeV and the fission moment is less than 0.5 ns, and because γ -ray flight speed is well known, the fission moment can be precisely determined from the detection time of fission γ -rays if the measurement threshold is set above 0.3 MeV [20]. This indicates that the fission moment can be accurately tagged using fission γ -rays. Therefore, in the present experiment, we tagged the fission moment using fission γ -rays detected by scintillation detectors rather than fission fragments detected by a fission chamber.

This manuscript presents our experimental study of energy correlations between neutrons emitted at 90° and 180° relative to each other. We measured prompt fission neutrons using 16 small EJ309 liquid scintillation detectors (5.08 cm diameter and 5.08 cm thickness) due to their superior pulse-shape discrimination performance, and detected fission γ -rays with eight CLYC detectors (2.54 cm diameter and 2.54 cm thickness) [21]. We present measured neutron energy spectra correlated with different-energy neutrons for the first time in the 1–5 MeV range and compare them with Monte Carlo simulation results.

II. Monte Carlo Simulation

Since most prompt fission neutrons are evaporated from fully accelerated fission fragments, their energy and direction in the laboratory system are significantly influenced by the energy and direction of the associated fission fragments. Therefore, predicting energy correlations requires knowledge of fission fragment yields and energies, as well as the number of neutrons evaporated from specific fragments. Figure 1 [Figure 1: see original paper] shows the flowchart of our Monte Carlo simulation. We simulated fission events individually, dividing the procedure into three stages: generation of the two fission fragments, generation of prompt fission neutrons, and neutron statistics.

A. Generation of the Two Fission Fragments

We randomly sampled the mass of the light fission fragment (M_L) with probability proportional to the yields shown in Fig. 2 [Figure 2: see original paper] [22]. The mass of the paired heavy fission fragment (M_H) is $252u - M_L$. We then determined the total kinetic energy (TKE) and its uncertainty ($\sigma\{TKE\}$) using the total kinetic energy distribution and corresponding uncertainty distribution as functions of light fragment mass, as shown in Fig. 3 [Figure 3: see original paper]. The specific total kinetic energy of the sampled fission fragment pair (E_T) was specified as $TKE + \Delta TKE$, where ΔTKE was randomly sampled from a normal distribution with standard deviation $\sigma\{TKE\}$. The kinetic energy of the light fission fragment (E_L) was determined as:

$$E_L = E_T \times \frac{M_H}{M_L + M_H}$$

The kinetic energy of the heavy fission fragment (E_H) equaled $E_T - E_L$. Finally, we sampled the direction of the light fission fragment in the laboratory system from an isotropic distribution, with the heavy fragment direction opposite to that of the light fragment.

B. Generation of the Prompt Fission Neutrons

Neutron evaporation was simulated via a cascade process for the light fission fragments, followed by the heavy fragments. Figure 4 [Figure 4: see original paper] shows the number of neutrons emitted from fission fragments of different masses and the corresponding uncertainties [22]. Since $\sigma_{\{TKE\}}$ is known, σ_v can be calculated. The number of neutrons N evaporated from the sampled light fission fragment was $v + \Delta v$, where Δv was randomly sampled from a normal distribution with standard deviation σ_v . In almost all cases, N was non-integer. Therefore, N was decomposed into an integer part I_N (the largest integer smaller than N) and a decimal part D_N (where $D_N = N - I_N$). A random number e in $[0, 1]$ was then sampled and compared with D_N . If e was smaller than D_N , the actual number of neutrons R_N emitted from the light fission fragment was $I_N + 1$; otherwise, R_N equaled I_N .

Neutrons were evaporated one by one using this process until R_N neutrons had been evaporated from the light fission fragment. After generating neutrons from the light fragment, we processed neutrons from the heavy fragment similarly.

According to standard nuclear evaporation theory, the fission neutron energy spectrum Φ in the center-of-mass system of the fission fragment can be described as [22]:

$$\Phi(E) \propto E^\lambda \exp(-E/T_{\text{eff}}) = E^\lambda \exp(-1.09E/T)$$

where λ is the cascade neutron emission coefficient and T denotes the temperature of the fission fragment, as shown in Fig. 5 [Figure 5: see original paper] [22]. Based on the energy distribution calculated using Eq. (2), we sampled the energy of a neutron in the center-of-mass system. The neutron direction was sampled from an isotropic distribution. We then calculated the energy and direction of the neutron in the laboratory system, along with those of the light fission fragments, under constraints of energy and momentum conservation. The mass of the light fission fragment equals $M_L - n$, where n represents the number of neutrons evaporated from the light fragment up to that point. Finally, we stored the neutron energy and direction in a queue.

C. Statistics of the Neutrons

After generating prompt fission neutrons, we tracked them to obtain their energy correlations. As shown in Fig. 6 [Figure 6: see original paper], we positioned 36 surface detectors (radius 5 cm) on a circle (radius 60 cm), with a ^{252}Cf source at the center. The angle between adjacent surface detector normals was 10° . We tracked the stored fission events individually, using the energy and directional information of emitted neutrons. If a neutron reached any detector, it was counted as a single event. As shown in Fig. 7 [Figure 7: see original paper], the simulated energy spectrum of single events agrees well with the ^{252}Cf prompt fission neutron energy spectrum recommended by IAEA, with relative deviation smaller than 0.6% [23]. After tracking all neutrons, if multiple neutrons from the same fission event reached detectors, we paired every two neutrons as a group. In this process, the neutron reaching the detector with the smaller serial number was designated as the first neutron. In the present work, we counted correlation events at angles from 0° to 180° with 30° intervals (the angle between the two neutron directions).

As presented in Fig. 8 [Figure 8: see original paper], the distributions of correlated events at 90° and 180° are noticeably different. The neutron energies corresponding to the highest counts are 2.6 MeV at 90° and 3.0 MeV at 180° . To present the energy correlation more clearly, we normalized the counts at each angle in two steps, as shown in Fig. 9 [Figure 9: see original paper]. First, we normalized the total counts in each column with neutron energies ranging from 1–5 MeV. Second, we normalized the count distributions in each column to the simulated ^{252}Cf prompt fission neutron energy spectrum. As Fig. 9 shows, neutron energy correlations vary with angle. The second neutron energy trends toward lower values for correlated events near 90° , whereas it trends toward higher values for correlated events near 0° or 180° . These trends increase with the first neutron energy. Because the crosstalk effect in measurement is non-negligible for correlated events at angles smaller than 75° [14], we measured energy correlations at 90° and 180° as representative cases in the present work.

III. Experiments

A. Experimental Setup

The experimental setup is illustrated in Fig. 10 [Figure 10: see original paper]. We used a ^{252}Cf source with a fission rate of approximately $1.3 \times 10^5 \text{ s}^{-1}$, sealed in a stainless steel capsule (2 mm diameter and 5 mm height) and suspended vertically 1.5 m above the floor using a fine nylon thread to minimize neutron scattering effects on detection by the EJ309 liquid scintillation detectors. We mounted all detectors on aluminum supports, combining every four EJ309 liquid scintillation detectors into an array. The angle between normals of adjacent arrays was fixed at 90° . The distance from the ^{252}Cf source to the front face of the eight CLYC detectors was $6 \pm 0.1 \text{ cm}$, and that to the front face of the sixteen EJ309 liquid scintillation detectors was $60 \pm 0.1 \text{ cm}$. Since the thickness of the EJ309 liquid scintillation detector is 5.08 cm, the uncertainty of the neutron flight path length σ_L is 2.15 cm, calculated as:

$$\sigma_L = \sqrt{\left(\frac{H}{2\sqrt{3}}\right)^2 + \sigma_p^2}$$

where H is the thickness of the EJ309 liquid scintillation detector, and σ_p represents the uncertainty in the distance from the ^{252}Cf source to the front faces of the EJ309 liquid scintillation detectors.

We acquired detector signals using two CAEN VX1730B digitizers, each with 16 input channels and a sampling frequency of 500 MS/s. One digitizer connected to the eight CLYC detectors, and the other to the sixteen EJ309 liquid scintillation detectors. Both digitizers operated in list mode, saving the timestamp (T), and long and short gate integrated charges (Q_L and Q_S) of every event for off-line analysis. The pulse height (PH) and pulse shape discrimination (PSD) parameter can be calculated as [24, 25]:

$$PH = C_A Q_L$$

$$PSD = C_P \left(\frac{Q_S}{Q_L}\right)$$

where C_A and C_P are calibration factors.

B. Calibration of the Detectors

Before measurement, we calibrated the eight CLYC detectors and sixteen EJ309 liquid scintillation detectors using ^{60}Co (1.33 and 1.17 MeV) and ^{22}Na (1.28 and 0.55 MeV) γ sources, as shown in Fig. 11 [Figure 11: see original paper]. We adjusted the working high voltages to achieve nearly equivalent signal gains for

the eight CLYC detectors, and applied similar adjustments to the sixteen EJ309 liquid scintillation detectors.

In calibrating the eight CLYC detectors, we used both photo peaks and Compton edges, while for the sixteen EJ309 liquid scintillation detectors we utilized only Compton edges. The method for determining Compton edges was described in Ref. [26, 27]. The corresponding Compton electron energy E_e is:

$$E_e = \frac{E_\gamma}{1 + \frac{m_0^2 c^2}{2E_\gamma}}$$

where E_γ denotes the γ -ray energy and $m_0^2 c^2$ represents the electron rest mass energy (0.511 MeV).

In Fig. 11(a), the Compton edge corresponding to a Compton electron energy of 1.12 MeV was obscured because it overlapped with the tail of the 1.17 MeV photo peak. The position of the Compton edge corresponding to 0.38 MeV could not be determined accurately due to significant measurement threshold effects. Consequently, we did not use these two Compton edges in calibrating the eight CLYC detectors.

C. Measurement

The experiment ran for a total measurement duration of approximately 1200 hours. Because two VX1730B digitizers acquired detector signals, they had to operate in time-synchronized mode to obtain accurate TOF data. To achieve this, the two digitizers shared the internal clock of the unit connected to the eight CLYC detectors, with synchronization configured via the data acquisition software COMPASS, as described in the COMPASS User Manual [28]. Based on our synchronization test with a DT5810 pulse generator, the synchronization accuracy was better than 0.1 ns. Although this accuracy contributes to experimental time resolution, its influence is negligible because it is much smaller than the overall experimental time resolution of 1.5 ns.

In the measurement, triple coincidence was required, resulting in very low detection efficiency. Based on tests, the time resolution was approximately 1.5 ns. Therefore, we set the neutron flight path length to 62.5 cm to balance neutron detection efficiency and energy resolution when using the TOF (time-of-flight) technique.

IV. Results and Discussions

A. Data Analysis

From the experimental data, we obtained PH-PSD two-dimensional spectra for the eight CLYC detectors and sixteen EJ309 liquid scintillation detectors, as

illustrated in Fig. 12 [Figure 12: see original paper]. By applying optimal discrimination thresholds, we could identify event types (γ -rays or neutrons).

In the present measurement, the thresholds for CLYC detectors were approximately 250 channels (0.3 MeV for γ -rays), and those for EJ309 liquid scintillation detectors were approximately 50 channels (0.5 MeV for protons). Although the low threshold for EJ309 detectors could lead to misidentification of event types using PSD for low pulse-height events, the probability of classifying a γ -ray event as a neutron event was low due to the significant TOF difference between γ -ray and neutron events, as shown in Fig. 13 [Figure 13: see original paper].

Figure 13 illustrates the TOF distribution of single neutron events detected by the 16 EJ309 liquid scintillation detectors. To suppress accidental coincidence interference, we adopted triple time correlation coincidence in data analysis, requiring a neutron event to correlate with at least two γ -rays detected by either CLYC or EJ309 detectors. The start time of TOF (T_0) is:

$$T_0 = \frac{\sum_k T_k PH_k}{\sum_k PH_k}$$

where T denotes the timestamp, and subscript k represents the serial number of the corresponding detected γ -rays.

The coincident time window was -200 to 200 ns. We took the average count S_b in the time window from -200 to -50 ns as the accidental coincident background. The net single neutron events S_{net} can be derived by subtracting S_b from the TOF distribution shown in Fig. 13. Theoretically, S_{net} was proportional to the detection efficiency and neutron flux :

$$S_{\text{net},i} = \phi_i \varepsilon_i$$

where subscript i represents the serial number of the divided TOF bin.

Neutron coincident events at 90° and 180° are shown in Fig. 14 [Figure 14: see original paper]. We employed triple time correlation coincidence in data analysis, requiring two neutrons and at least one γ -ray to define a coincident event. Five types of interference events exist. The first is the γ -ray event shown in the purple box. These events do not noticeably affect measurement of neutron events in the white box because the TOF difference between γ -rays and neutrons is significant. The second type, shown in the green box, indicates that the first neutron is an accidental coincident neutron. Related interference can be deducted by subtracting the average counts of each column in the region with TOF ranging from -150 to -50 ns for the first neutron from the counts of each column in the white box. The third type, shown in the red box, is where the second neutron is an accidental coincident neutron; related interference can be subtracted using a similar approach. The fourth type, shown in the black frame, is where the γ -ray is an accidental coincident γ -ray. The corresponding

interference can be deducted in two steps. First, we scanned the region with TOF ranging from -150 to -50 ns for the first neutron along the diagonal line using a scanning box identical to the white box shown in Fig. 14, calculating the average count of every bin in the scanning box. Second, we deducted the corresponding interference by subtracting these average counts from the counts in the white box. The fifth type comprises crosstalk events, where the neutron detected by the second detector is the same neutron that was scattered and detected by the first detector. Although accurate subtraction of this interference is difficult, it can be significantly mitigated by restricting TOF to the range of 19–46 ns, a conclusion drawn from Monte Carlo simulation performed by JMCT, as shown in Fig. 15 [29].

After subtracting the interference events mentioned above, we obtained the net neutron coincident events D . To determine the relative deviation between the neutron energy spectrum of ^{252}Cf prompt fission neutrons and that of neutrons correlated with different-energy neutrons, we executed three calculation steps. First, we compared the net neutron coincident events D with the net single neutron events S_{net} to obtain the relative ratio R :

$$R_{i,j} = \frac{D_{i,j}}{S_{\text{net},j}}$$

where subscripts i and j of R and D denote the TOF bin serials of the first and second detected neutrons, and subscript j also represents the TOF bin serial of the net single events for S_{net} . It should be mentioned that the influence of detection efficiencies for different-energy neutrons is canceled out in this step. Second, we normalized the relative ratios R over the TOF_2 region ranging from 19 to 46 ns as follows:

$$\text{Nor}_R_{i,j} = \frac{R_{i,j}}{\sum_j R_{i,j}} \times N$$

where N is the bin number in the TOF region ranging from 19 to 46 ns. Third, we converted neutron TOF into neutron energy E_n as shown in Fig. 16 and Tables 1 and 2 using [30, 31]:

$$E_n = \left(\frac{1}{\sqrt{1 - \frac{L^2}{\text{TOF}^2 c^2}}} - 1 \right) m_n c^2$$

where L is the neutron flight length, c is the speed of light in vacuum, and m_n denotes the neutron mass. In the present measurement, relativistic effects can be neglected because the detected neutron velocity is much smaller than the speed of light, allowing simplification of the E_n calculation as [32]:

$$E_n = \frac{5228.16L^2}{\text{TOF}^2}$$

where the units of L and TOF are meters and nanoseconds, respectively.

Measurement uncertainties (1.5%-3.2% at 90°; 1.6%-4.1% at 180°) include contributions from D and S_{net}. The uncertainties of D (1.5%-3.2% at 90°; 1.6%-4.1% at 180°) comprise statistical uncertainties (1.5%-3.1% at 90°; 1.6%-3.9% at 180°) and uncertainties from subtracting accidental coincidences involving the first neutron (<0.1% at 90° and 180°), the second neutron (<0.1% at 90° and 180°), and the γ -ray (<0.1% at 90° and 180°). The uncertainties of S_{net} (0.2%-0.3% at 90°; 0.2%-0.4% at 180°) consist of statistical uncertainties (0.2%-0.3% at 90°; 0.2%-0.4% at 180°) and uncertainties from subtracting accidental coincident backgrounds (<0.1% at 90° and 180°). The uncertainty of E_n (9.5%-17.2%) is contributed by the experimental time resolution of 1.5 ns (6.5%-15.8%) and the uncertainty of the neutron flight path length of 2.15 cm (6.9%).

B. Discussions

As shown in Figs. 9 and 16 and Tables 1 and 2, the results indicate that the influence of a neutron on the energy of its correlated partner is more pronounced in the high-energy region, and this effect appears more noticeable at 180° than at 90°. Beyond differences in magnitude, the correlated neutron energy shows opposite trends between events at 90° and 180°. The energy of a neutron correlated with a relatively high-energy neutron tends toward lower values at 90°, while it tends toward higher values at 180°. Using the measured relative ratios, the related neutron energy spectra can be derived by multiplying the relative ratios by the 252Cf prompt fission spectrum recommended by IAEA [23].

Compared with the measurement results obtained by Schuster et al. [14] and the simulation results introduced previously, the present results show the same tendency: the energy correlation between coincident neutrons at 180° is positive, while that between coincident neutrons at 90° is negative. This consistency enhances the credibility of our measurement results. Furthermore, the agreement between measurements and simulations regarding energy correlation tendency suggests that the dominant mechanism may be the impact of fission fragment velocities and directions on neutron energy. As noted previously, most neutrons are emitted from fast-moving fission fragments, meaning the moving direction angle between a high-energy neutron and its corresponding fission fragment tends to be small, whereas that between a low-energy neutron and its fragment tends to be large. For correlated neutrons at 90°, if the first neutron energy is relatively high, the second neutron energy tends to be low. This occurs because the high energy of the first neutron indicates a small moving direction angle between this neutron and its fission fragment with high probability. Therefore, regardless of whether the two neutrons are emitted from the same fragment

or different fragments, considering the opposite directions of the two fission fragments, the moving direction angle between the second neutron and its corresponding fragment is likely large, resulting in relatively low energy for the second neutron. Conversely, for correlated neutrons at 180° , a relatively high energy for the first neutron tends to result in low energy for the second neutron if they are emitted from the same fragment, but high energy if emitted from different fragments. Since the probability of detecting neutrons from the same fragment is noticeably smaller than that from different fragments, the overall tendency shows that a high-energy neutron is more likely to be correlated with another high-energy neutron. For example, if each fission fragment emits two neutrons and detection efficiencies are equal, the probability of detecting neutrons from the same fragment is approximately half that from different fragments. Additionally, it should be noted that energy correlation may also be influenced by factors beyond fragment velocities and directions, such as the energy competition relationship among neutrons emitted from the same fragment, as pointed out in Ref. [14].

V. Conclusions

We measured for the first time the relative ratios of energy spectra of neutrons correlated with different-energy neutrons to the ^{252}Cf prompt fission spectrum in the region from 0.96 to 5.7 MeV. The results demonstrate that neutron energies emitted from a single fission reaction are correlated. The largest deviation between the measured energy spectra of neutrons correlated with 5.7 MeV neutrons and the fission neutron energy spectra approaches 20%. Furthermore, based on measured and simulated energy correlation trends, the deviation is expected to become more significant in higher neutron energy regions. This indicates that neutron energy correlations should be considered in coincidence and multiplication measurement techniques to obtain more reliable results, particularly for fast neutron coincidence and multiplication measurement techniques.

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Note: Figure translations are in progress. See original paper for figures.

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