Measurement and Simulation of Neutron/Gamma-Ray Cross-Correlation Functions from Spontaneous Fission

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Abstract

We present an application of a digital pulse shape discrimination (PSD) technique for the novel measurement of cross-correlation functions from a spontaneous fission source. The measurement method allows for the collection of fast coincidences within a time window of the order of a few tens of nanoseconds. The use of PSD allows for the accurate acquisition of the coincidences in all particle combinations. Specifically, separate neutron–neutron, neutron–gamma-ray, gamma-ray–neutron, and gamma-ray–gamma-ray coincidences are acquired with two liquid scintillation detectors. The measurements are compared to results obtained with the MCNP-PoliMi code, which simulates neutron and gamma-ray coincidences from a source on an event-by-event basis. This comparison leads to relatively good qualitative agreement.

Simulations of the separate neutron and gamma-ray contributions to the total cross-correlation function help to further improve the performance of experimental systems that aim at accurate identification of nuclear materials. This research has direct applications in the areas of nuclear nonproliferation and homeland security.

Key words: nuclear nonproliferation, pulse shape discrimination, liquid scintillator detectors, cross-correlation functions, coincidences.

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1 Introduction

Several techniques used in the field of nuclear nonproliferation rely on the measurement of neutron multiplicity distributions to assess and characterize fissile materials in various forms. These techniques are based on the thermalization of neutrons from fission in polyethylene moderators and subsequent detection using He-3 counters [1].

The detection of correlated neutron and gamma-ray events has proven very useful for the identification of various nuclear materials. Indeed, since for a given material and geometry the time distributions of the correlated events are very characteristic, they are representative for and can be used as distinctive signatures of different material-geometry configurations.

Recent applications in the areas of homeland security and nuclear nonproliferation use organic scintillation detectors in liquid or plastic form. These detectors are sensitive to both fast neutrons and gamma rays [2]. Signatures that rely on coincidence measurements of neutrons and gamma rays from fission have been shown to be useful in the detection and characterization of nuclear materials [3]. The coincidence distributions are of interest because they contain information that can be used to accurately identify and characterize fissile isotopes.

Recently, we have described the application of a digital pulse shape discrimination (PSD) technique [4] to the identification of shielded neutron sources by visual inspection of measured pulse height distributions. In this paper, a similar PSD technique is used for the analysis of correlated neutron and gamma-ray events from a Cf-252 spontaneous fission source using two liquid scintillation detectors, and a 12 bit, 250 Mhz waveform digitizer. The correlations are performed in a time window of a few tens of nanoseconds. The experiments are performed with and without lead (Pb) shielding for a symmetric position of the source with respect to the detectors, and without lead for an asymmetric position. For the first time, we show the use of the PSD for acquisition of separate neutron–neutron, neutron–gamma-ray, gamma-ray–neutron, and gamma-ray–gamma-ray coincidences in the two liquid scintillation detectors. The knowledge of these separate contributions is essential for further improvement of the existing measurement systems based on detection of correlated events.

2 Description of Experimental Setup

The setup consists of two liquid scintillation detectors. The size of the active volume hosting the liquid scintillator is 25 by 25 by 8 cm. The detectors are placed on a steel cart, at a distance of 60 cm from each other. The Cf-252 source is placed at the center of the assembly. Figure 1 shows the photograph of the experimental setup. The voltages used at the detectors were determined by standard energy calibration performed using a Cs-137 source.

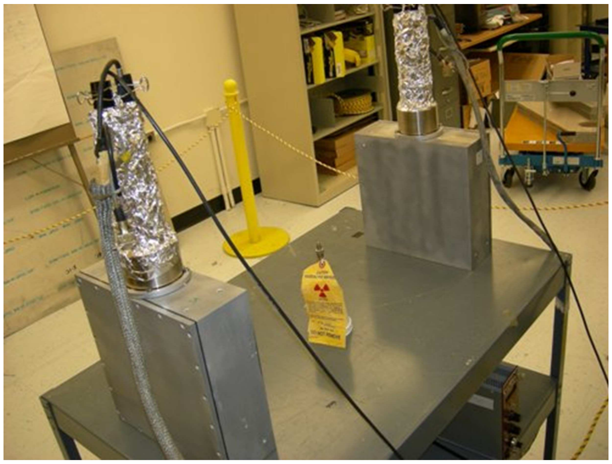

Fig. 1. Measurement setup.

The analog signals from the photo multiplier tubes were fed into a 2 channel 250 MHz digitizer (4 ns time resolution for each channel). Due to the 12 bit vertical resolution, the data could be interpolated down to 1 ns. To investigate the feasibility of using cross-correlation measurements for the identification of nuclear materials, a number of experimental setups were used. Various source-detector distances and both symmetric and asymmetric configurations were investigated. In addition, the effect of Pb shielding on the measurements was assessed. The following configurations were investigated:

- 30 cm source-detector distance, no shielding,
- 30 cm source-detector distance, 2.2 cm Pb shielding,
- 15 and 45 cm source-detector distance, no shielding,
- 40 cm source-detector distance, no shielding,
- 50 cm source-detector distance, no shielding.

The measurement time was adjusted according to the source-detector distance, to account for the geometrical detector efficiency is lowered. Using shielding such as Pb makes detection of gamma rays from the sample less frequent, while the neutrons will still reach the detectors mostly unaffected. In the asymmetric setup the effect of non negligible time of flight (TOF) for neutrons is investigated.

3 Pulse Shape Discrimination

For PSD the ratio of tail-to-total integrals was used. This method utilizes the fact that gamma-ray pulses have shorter tails than neutron pulses when interacting with liquid scintillator detectors. The ratio of tail-to-total integrals is calculated as:

$$R \equiv \frac{\text{Tail integral}}{\text{Total integral}}. \tag{1}$$

The time intervals over which the total and tail integrals are calculated are parameters that can be modified to increase the performance of the PSD method. In the experiments, the tail integral was calculated from 40 ns to 250 ns after the pulse maximum. Figure 2 shows the typical performance of our PSD system for the two detector channels.

In the measurements, many factors can affect the particle identification, for example background cosmic rays can easily interact with the detector material and create very large pulses. This type of background events often lead to pulses that are not in the range of the digitizer and can therefore easily be sorted out. Other examples are small pulses that are usually difficult to identify and particles that interact in the detector shortly after another particle was already detected by the same detector. For the latter, the tail integral gives an uncharacteristically high value, which could lead to misclassification of the particles (see Fig. 2).

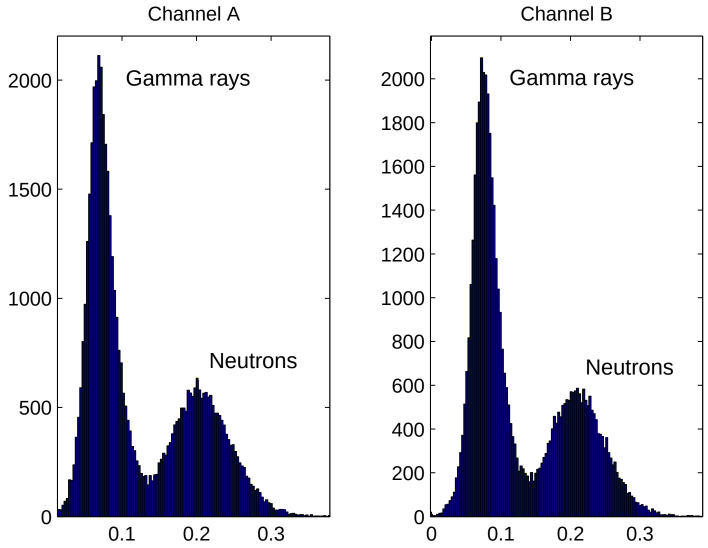

Fig. 2. Typical histogram of the ratio of tail-to-total integrals. The gamma ray and neutron peaks are very well separated. The overlap area between the peaks contain pulses that are difficult to classify.

Timing is a very important factor for accurate cross-correlations measurements. Due to the nature of light acquisition in the detector, pulses of different amplitudes will have different rise time. Therefore using pulse peaks as a trigger for timing would lead to difficulties. For this reason, the algorithm simulates a constant fraction discriminator where the timing of pulses is taken at 20 % of the peak value. The large vertical resolution of the digitizer (12 bits) enables accurate interpolation down to a timescale smaller than that of the digitizer sampling frequency (4 ns step).

4 Measurement Results and Discussion

The measurements were carried out using a Cf-252 spontaneous fission source with an activity of 3.8 μCi. The threshold in both the simulations and the measurements was set to 130 keVee.

All measured pulses that are sufficiently close (80 ns window) are separated out and classified (neutron or gamma ray). Each particle pair (doublet) contributes to the total cross-correlation distribution with one data point. To analyze the total distribution, the total curve is subdivided into four categories: gamma-ray–gamma-ray (γ, γ), neutron–neutron (n,n), gamma-ray–neutron (γ,n), and neutron–gamma-ray (n,γ) doublets. For all correctly measured symmetrical configurations, the cross-correlation functions must show a symmetry around time zero (see Fig. 3).

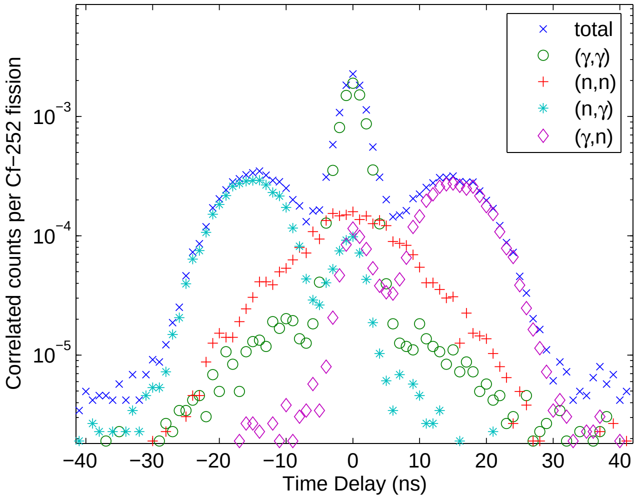

Fig. 3. The measured cross-correlation functions for the 30 cm symmetric bare case.

In Fig. 3, three distinct peaks can be observed in the total cross-correlation function. One peak corresponds to the (γ, γ) peak, which has only a very small time spread caused by the measurement electronics. Symmetrically on each side of the central peak there are the peaks for (n,γ) and (γ,n) doublets, respectively. In addition, there is the (n,n) peak ; this distribution has a much larger spread than the (γ,γ) distribution, because neutrons in addition to the measurement uncertainty are also affected by a notable variation in TOF (energy).

When analyzing the cross-correlation events it appears that a number of particles are misclassified. This is observed in Fig. 3 where the (n,γ) and the (γ,n) curves show extra peaks at times close to 0 ns. It should be noted that these spurious peaks are not observed in the Monte Carlo simulations (see section 5). This fact is, however, due to data handling rather than physics. The reason for the spurious peaks are not as easy as just misclassifications though, much of the data points in those curves are coming from a special type of event which is described below.

In the measurements, a (γ,γ) doublet is the most common one. However, when a (γ,γ) doublet is detected there is a certain probability that an additional particle, such as a neutron, will hit one of the detectors. The pulses from the event when both a gamma-ray and a neutron hit the same detector often appear as only one larger peak with the gamma ray time characteristics, but it has the tail behaviour of a neutron pulse since the neutron induced a delayed light production in the detector. Therefore, the ratio of tail-to-total integrals will indicate it as a neutron, but in the cross-correlation measurement it will have a timing similar to a (γ,γ) event. This event is visible in some of the measurements. Larger distance lowers the geometrical detector efficiency, and double particle events in one single detector reduce quickly in frequency. Support of this conjecture can be seen in Fig. 4, where the cross-correlation for the 30 cm case with 2.2 cm Pb shielding is shown. The lead shields the gamma rays very effectively, while the neutrons are mostly unaffected. This results in a lower number of gamma rays measured, thus (γ,γ) doublets are less likely. Consequently, there are less (γ,γ) events that could be affected by an additional (slower) neutron. In the case of lead shielding the most notable peak is now the broader (n,n) peak. Good performance of the PSD can be observed in the still well defined (γ,γ) peak, which is now reduced in magnitude by approximately a factor 20 (see Fig 3) but is still not exhibiting much statistical noise before low count rates are encountered.

If the detectors do not have the same distance to the source, the arrival time of neutrons will significantly change, while the gamma rays will still be more or less instantaneous with respect to the source event. As can be seen in Fig. 5, there is a notable difference both in the total cross-correlation distribution and the different components, compared to the symmetric case. The width of the (γ,n) curve is larger than that of the (n,γ) curve since one detector has a larger source distance (45 cm vs. 15 cm) and the variation in neutron energy

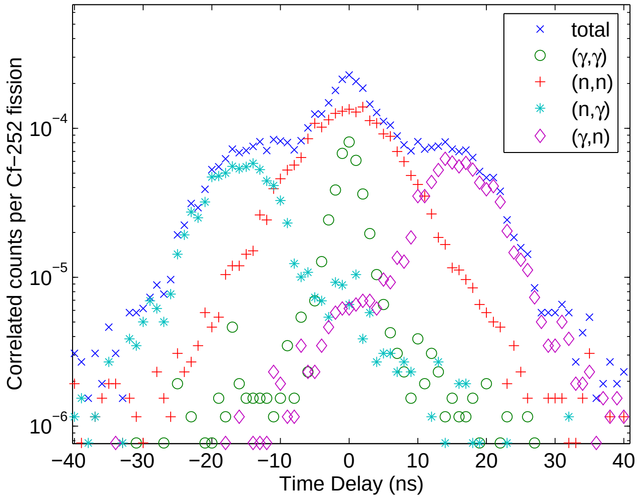

Fig. 4. Measured cross-correlation functions for the 30 cm case with 2.2 cm lead shielding.

will create a larger spread in time for greater distances. Very good performance of the timing interpolation can be seen in Fig. 5, where the one nanosecond step is still behaving very well. In addition the (γ, γ) peak correctly shows the expected shift of approximately one nanosecond, corresponding to the difference in distance of the detectors with respect to the source.

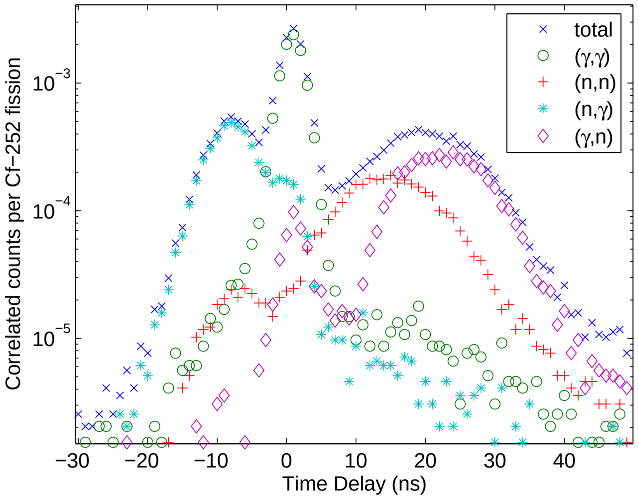

Fig. 5. Measured cross-correlation functions for the asymmetric setup with the source-detector distances 15 and 45 cm, respectively.

When increasing the distance between the source and detectors to 40 and 50 cm, respectively, the number of detected doublets per fission is reduced. Therefore, to achieve the same statistical confidence the measurement time needs to be increased. Since the measurements are made in real time and most of the data shown were acquired within a few minutes, increased data acquisition time increases the total measurement time by a few minutes. However, if the distances were increased further it would be advisable to use either larger detectors, or more than two detectors to compensate for the decreasing geometrical detector efficiency. Further, it is observed in Fig. 6 that increased distances result in wider cross-correlation distributions due to the larger TOFs, especially for neutrons.

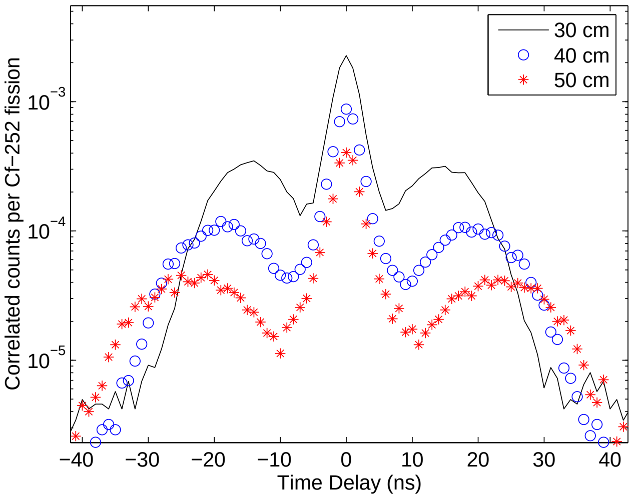

Fig. 6. Measured cross-correlation functions of the total doublets for various detector distances.

5 Comparisons with Monte Carlo

The experiments described in the previous section were simulated using the MCNP-PoliMi code [5,6]. This code is capable of correctly simulating correlations between neutrons and gamma rays from a spontaneous fission, thus it it is very well adapted for simulating the cross-correlation measurements. The detector response is simulated by a specialized postprocessing code [7], which computes the cross-correlation functions. Figure 7 shows the comparison of the experimental data with Monte Carlo simulations. Very good qualitative agreement in the total cross-correlation functions is observed. In the case of the partial cross-correlation functions, however, there are some differences, especially in the shape of the (γ, γ) peaks. Specifically, in the simulations the (γ, γ) peak is sharper than in the measurements, which is caused by the measurement uncertainty that is not present in the (ideal) simulations. Note that absolute comparisons are presented. The (n,n) peak is narrower but also higher in the simulations compared to the measurements, which partly could be explained by measurement uncertainty.

Figure 8 shows the comparison for the lead shielded case. The agreement for

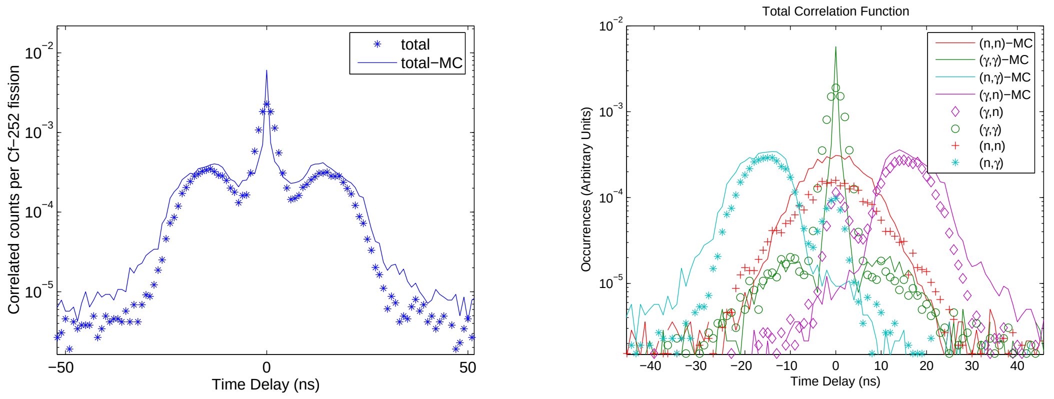

Fig. 7. Comparisons of experimental and Monte Carlo results for the total and the partial contributions to the cross-correlation functions in the case of 30 cm source-detector distance. The most notable differences can be seen in the shape of the (γ, γ) and (n,n) peaks.

the partial cross-correlation functions is still good. For the (n,n) doublets, however, there is a difference between the measurements and the simulations, where the simulation predicts them to be more frequent, compared to what was observed in the measurement. The very different triggering mechanism used in the experiment might explain a part of the difference. The initial agreement, however, is very encouraging for further experimental work on the cross-correlation signatures of various nuclear materials, beyond the Cf-252 source used in these measurements.

Further investigation and fine tuning of the experimental setup is needed to explain the unresolved differences between the measurements and simulations. One of the main differences between the simulations and the measurements is that there is no particle misclassification in the simulations. Also certain simplifications in light production and collection are currently present in the simulations. Regarding the measurements, multiple detections within a single data acquisition window are of importance. Those events can lead to particle misclassification. Therefore, it would be beneficial to ‘show’ those events also in simulations.

Figure 8 (right) shows the comparison of measurement and simulation for the asymmetric case. The agreement between measurement result and Monte Carlo is encouraging. The biggest differences can be found in the (n,n) events, which are more frequent in the simulations compared to the experiments. In the cross -correlation functions statistical errors will be more visible for low counts especially in the outer parts of the peaks, while the peaks themselves have high counts, visible in the even peaks compared ti the more jagged sides. Uncertainties in the PSD method will lead to possible misclassifications, and there is a minor overlap of neutrons and gamma rays in the ratio of tail-to-total integrals, this overlap is larger for small energy pulses, which are more

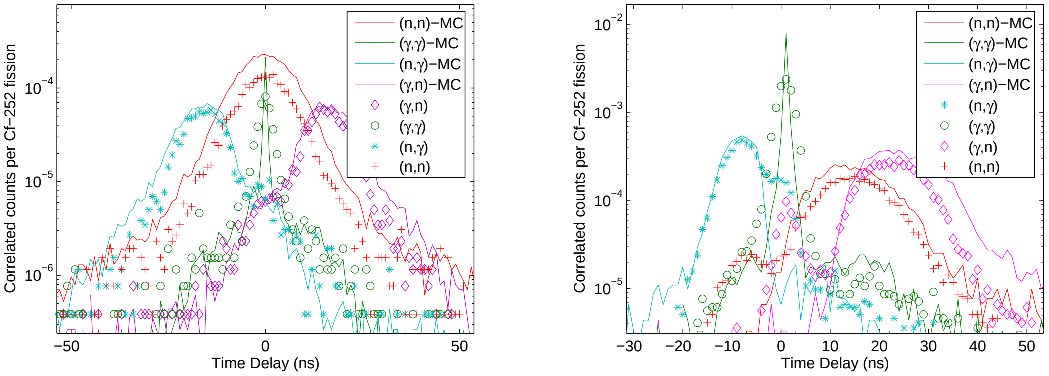

Fig. 8. The comparison between measurements and Monte Carlo of the partial cross-correlation functions for the case using lead shielding (left) and the asymmetric case (right).

difficult to classify, This can explain the wider base of the (γ, γ)-peaks visible in the experiments. Calculation of cross-correlation errors associated with the PSD method as well as the statistical uncertainties will be further investigated in the future.

6 Conclusions

This paper describes the measurement and the simulation of cross-correlation functions from a spontaneous fission source. An offline, digital PSD technique was used on the particles detected with two liquid scintillation detectors. For the first time, the PSD technique was used to measure separate neutron–neutron, neutron–gamma-ray, gamma-ray–neutron and gamma-ray–gamma-ray coincidences to be acquired and analyzed. These separate and total cross-correlation functions offer a unique insight into the investigated material sample. The knowledge of the separate contributions helps to further improve existing measurement systems relying on detection of correlated events from fissionable material. The cross-correlation functions can be used as material-geometry “signatures”, and are thereby of interest for applications in nuclear nonproliferation and homeland security.

The measured cross-correlation functions were compared with Monte Carlo simulations using the code MCNP-PoliMi and a good agreement was obtained.

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