Method for Measuring Multiple Scattering Corrections between Liquid Scintillators

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Abstract

A time-of-flight method is proposed to experimentally quantify the fractions of neutrons scattering between scintillators. An array of scintillators is characterized in terms of crosstalk with this method by measuring a californium source, for different neutron energy thresholds. The spectral information recorded by the scintillators can be used to estimate the fractions of neutrons multiple scattering. With the help of a correction to Feynman's point model theory to account for multiple scattering, these fractions can in turn improve the mass reconstruction of fissile materials under investigation.

Keywords: liquid scintillators, crosstalk, multiple scattering, neutron multiplicity, neutron correlation, fissile materials

1. Introduction

In this paper, we are primarily studying the effect of fast neutrons scattering between different scintillators, a phenomenon also known as neutron crosstalk. The reason why this study is important is that if a neutron scatters and deposits enough energy in multiple liquid scintillators to record multiple counts, it will not only artificially increase the count rate but also the numbers of two- and three-neutron correlations. For neutron multiplicity counting (NMC) applications, masses of nuclear materials undergoing fissions can be determined using ^3He tubes measuring thermal neutrons [1, 2, 3, 4]. Cifarelli and Hage's [2] theoretical mass reconstruction is based on moments of count distributions, which are very sensitive to two-, three- and higher order correlations. The theory underlying the mass reconstruction assumes neutrons are only counted once, which is a correct assumption for ^3He tubes. However, because fast neutrons can scatter multiple times between scintillators [5, 6, 7, 8] and register artificially correlated counts, this traditional moment method unfortunately fails for scintillators. Using modified expressions for the moment method to theoretically account for multiple scattering, it was shown [9, 10] that the calculated fractions of neutrons scattering multiple times can be used to apply corrections to the masses of nuclear materials undergoing fission.

A summary of this paper is as follows. In Sec. 2, we describe the experimental setup, and characterize the scintillators. In Sec. 3, we discuss the time-of-flight method and the modified moment equations to determine the multiple scattering fractions for

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29 different neutron kinetic energies. Experimental measurements of these fractions are
 30 given in Sec. 4. In Sec. 5, we explain how the spectrum of energy deposited by the fast
 31 neutrons in the scintillators can be used to determine the multiple scattering fractions,
 32 and in turn to improve the fissile material mass and multiplication reconstruction.
 33 A list of symbols used throughout the text is available in the appending nomenclature.
 34

35 2. Detector characterization

36 Fig. 2 shows the geometrical configuration of the detectors, which are composed of
 37 different scintillating materials: EJ-301 and EJ-309 [11] cylindrical cells 4" diameter
 38 by 3" thick; backed by two kinds of photomultiplier tubes: Photonis [12] and Hama-
 39 matsu [13].

Figure 1: (color online) Geometry used to measure the multiple scattering fractions with a ^{252}Cf source. The ^{252}Cf source (in green) is located 5 cm from the trigger detector (in red, EJ-301/photons PMT) and 254 cm from the front face of the wall of timer detectors: EJ-301/photons PMT (yellow), EJ-301/hamamatsu PMT (orange), EJ-309/photons PMT (magenta) and EJ-309/hamamatsu PMT (cyan).

Figure 2: (color online) Photographs of wall of timer detectors, with lone trigger detector sitting on top for calibration. Data was taken on a low mass floor to minimize neutron scattering off concrete surfaces.

The liquid scintillators were calibrated in energy using a ^{137}Cs source placed in the middle of the line between the trigger detector and the wall of timer detectors. To reconstruct the spectra of energies deposited by the photons in the scintillators from the integral of the ADC counts I_{ADC} recorded by the PMT pulse digitizer, the following linear expression was used

$$\text{LO} = a \times I_{\text{ADC}} \quad [\text{keVee}] \quad (1)$$

where a is a coefficient that depends on the scintillator/PMT assembly and is in units of keV/(integral of ADC counts). Given the energy of the gamma ray emitted by the ^{137}Cs , the spectra exhibited a Compton edge at 477 keV. This edge was detected by an algorithm described in Ref. [14]. Fig. 3(a) shows the measured energy spectra for all scintillators for the ^{137}Cs source. The quasi-straight line between the orange and yellow regions in Fig. 3(b) is the location of the ^{137}Cs Compton edge.

Figure 3: Energy calibration of the liquid scintillators using a ^{137}Cs source.

To determine whether the detector responses are indeed linear, we fit four Compton edges from three sources: 477 keV Compton edge from ^{137}Cs which emits 661.7 keV photons; 1062 and 341 keV Compton edges from ^{22}Na which generates 1274.5 keV and 511 keV photons, and 2382 keV Compton edge from ^{232}Th with its 2615 keV photons. For LS01 for instance, the coefficients a obtained are

- 0.308321 (2382 keV Compton edge of ^{232}Th),
- 0.310952 (1062 keV Compton edge of ^{22}Na),
- 0.314823 (477 keV Compton edge of ^{137}Cs) and
- 0.311387 (341 keV Compton edge of ^{22}Na).

Normalized by the mean $\langle a \rangle$ for LS01, they become 0.9902, 0.9986, 1.0111 and 1.0001, which indicates a small spread in the coefficient a . Repeating this procedure for each one of the scintillator/PMT assemblies, we determine how the coefficients are distributed for all detectors. Fig. 4(a) shows the distribution of these coefficients relative to their mean for the different Compton edges. The spread of the distributions are very small (standard deviation $< 1\%$). To the extent that such a small error can be neglected

for our application, these distributions prove that the detector responses are indeed linear over the energy range 341 through 2382 keV. Fig. 4(b) shows the detector response of a liquid scintillator detector to a ^{22}Na source. The two Compton edges can clearly be identified.

Figure 4: Energy calibration of liquid scintillators.

The neutrons are discriminated from the photons using pulse shape discrimination (PSD) described in Ref. [15]. Fig. 5(a) shows neutron scores computed by the PSD algorithm for different detection events, as a function of the electron-equivalent energy deposited by the event. We can clearly distinguish two bands: the upper one filled with neutrons and the lower one with photons. For electron-equivalent energies greater than 2 MeVee, the two bands do not overlap significantly, leading to good discrimination. Below 2 MeVee, discrimination worsens, it becomes more difficult to distinguish neutrons from photons. The magenta outline in this plot defines a region where events are most likely neutrons, and will be tagged as such by the data acquisition system, whereas the black outline defines a region within which events are most likely photons. The ^{252}Cf source emitted so few neutrons above 5 MeVee that it was difficult to define regions of positive neutron identification with great confidence. Therefore, events lying in the extension of the neutron band above 5 MeVee are treated as particles of unknown type. The region of positive photon identification slowly curves up above 5 MeVee. This effect is attributed to saturation effects in the electronics. Indeed, large enough pulses run into the dynamic range limit of the digitizers. This causes either the PMTs to saturate or the tops of the digitized pulses to be flattened or chopped off and the corresponding charge does not get integrated. This charge deficiency decreases the denominator in the neutron score metric [15]. This effect depends on the voltage settings of the PMTs, settings which vary from scintillator to scintillator to get the same response for the same keVee¹. For these high energies, the detector response

¹This is primarily due to PMT differences and variations of how much light a PMT sees due to optical coupling variations.

looses its linearity. This is not critical for our experiment, because we only consider electron-equivalent energies below 5 MeVee.

The misidentification rates for this array of liquid scintillators were computed in Ref. [15]. The number of events misidentified as neutrons is of the order of 20 ± 4 per million events for a 100 keVee energy threshold, 13 ± 4 ppm for 200 keVee, 11 ± 3 ppm for 300 keVee, 9 ± 3 ppm for 400 keVee, and 7 ± 3 ppm for 500 keVee. The number of events misidentified as photons was not estimated, because it is of little relevance for the experiment.

Figure 5: Neutron score versus scintillation light output for one of the liquid scintillators. The upper band corresponds to neutrons, the lower one to photons. The magenta outline delimits region of positive neutron identification.

As we will see below, it is essential that the liquid scintillators be synchronized with each other to calculate neutron energies precisely. The liquid scintillators were synchronized using a method described in Ref. [16] to obtain sub-nanosecond timing resolution.

3. Methodology

Fig. 2 shows a ^{252}Cf source (green) located between the trigger detector (red) and the wall of timer detectors. Neutrons and photons are emitted simultaneously from that point source. Until one of these two particles is detected by one of the liquid scintillators, the detection system is unaware a spontaneous fission has occurred. In practice, photons will arrive to the detectors first because they travel at the speed of light, approximately 10 times faster than the fastest neutrons emitted by ^{252}Cf . The detection system finds out that a radioactive decay occurred (whether it be fission is not known) in the source as soon as the detection system fires with the first gamma ray. If a fast neutron is detected by a timer liquid scintillator within some time interval, we record the time interval between the arrivals of the trigger and timer particles.

To determine the neutron speed for each pair of photon and fast neutron detected, one needs to solve a few equations. The gamma ray is detected in the trigger scintillator centered on $(x_\gamma, y_\gamma, z_\gamma)$, the fast neutron is detected in a timer scintillator centered on (x_n, y_n, z_n) . The fast neutron is detected at a time ΔT later than the gamma ray.

113 For the sake of simplicity, we ignore for now the spatial extent of the two liquid
 114 scintillators, and assume them to be points. This is an important assumption because
 115 the spatial extent of the liquid scintillators limits the resolution of this technique. In-
 116 deed, with the technology current employed, it is not possible to determine exactly
 117 where the particle is detected within a liquid scintillator, and this can result in a uncer-
 118 tainty on the neutron speed of the order of the size of the liquid scintillators, which is
 119 currently four inches.

Let us further assume that the spontaneous fission source is located in $(x_{\text{src}}, y_{\text{src}}, z_{\text{src}})$. We can write the following equation to determine the time interval between the detected gamma ray and the detected fast neutron following it:

$$\Delta T = \frac{1}{v_n} \sqrt{(x_{\text{src}} - x_n)^2 + (y_{\text{src}} - y_n)^2 + (z_{\text{src}} - z_n)^2} - \frac{1}{c} \sqrt{(x_{\text{src}} - x_\gamma)^2 + (y_{\text{src}} - y_\gamma)^2 + (z_{\text{src}} - z_\gamma)^2} \quad (2)$$

where c is the speed of light and v_n is the speed of the fast neutron. v_n can be isolated so that

$$v_n = \frac{\sqrt{(x_{\text{src}} - x_n)^2 + (y_{\text{src}} - y_n)^2 + (z_{\text{src}} - z_n)^2}}{\Delta T + \frac{1}{c} \sqrt{(x_{\text{src}} - x_\gamma)^2 + (y_{\text{src}} - y_\gamma)^2 + (z_{\text{src}} - z_\gamma)^2}}, \quad (3)$$

$$E_{k,n} = \frac{1}{2} m v_n^2, \quad (4)$$

120 so that we can filter neutrons based on their speed or energy, and thus plot quantities as
 121 a function of the neutron kinetic energies $E_{k,n}$.

122 The algorithm to determine the multiple scattering fractions works on the principle
 123 that californium emits both spontaneous fission photons and neutrons simultaneously.
 124 It works as following: whenever a photon (spontaneous fission photon) is emitted by
 125 ^{252}Cf and detected by the close-by trigger detector (in red in Fig. 2), a time window
 126 is opened until one of the timer detectors in the wall gets a neutron count (from spon-
 127 taneous fission of ^{252}Cf). Eq. (3) can then be used to determine the speed v_n of the
 128 neutron and consequently its kinetic energy. As soon as this neutron is detected by one
 129 of the timer detectors, another time window is opened for a short time to see if any
 130 neutron count is recorded in any of the timer detectors. Such counts would likely be
 131 due to the original neutron scattering multiple times from the first timer detector.

Using the moments of the count distributions for a given incident neutron energy, we can determine the fractions of double scattering and triple scattering using the equations of Ref. [9, 10] reproduced here for a non-multiplying object and $\alpha T \gg 1$:

$$\begin{cases} \bar{C}(T) &= (1 + f_2 + 2f_3) R_1^* T \\ \lim_{\alpha T \gg 1} Y_2(T) &= (f_2 + 3f_3) R_1^* T + (1 + f_2 + 2f_3)^2 \epsilon D_{2s} \\ \lim_{\alpha T \gg 1} Y_3(T) &= f_3 R_1^* T + 2(1 + f_2 + 2f_3)(f_2 + 3f_3) \epsilon D_{2s} + (1 + f_2 + 2f_3)^3 \epsilon^2 D_{3s} \end{cases} \quad (5)$$

f_2 and f_3 are the fractions of single neutrons recording two and three counts, respectively, and R_1^* is the hypothetical count rate which one would measure if individual

neutrons could not be counted multiple times, as opposed to the measured count rate R_1 which includes two counts instead of one for each double scatter and three counts instead of one for each triple scatter. If we neglect the terms in ε and ε^2 for now, we obtain:

$$\begin{cases} \bar{C}(T) = (1 + f_2 + 2f_3) R_1^* T \\ \lim_{\alpha T \gg 1} Y_2(T) = (f_2 + 3f_3) R_1^* T \\ \lim_{\alpha T \gg 1} Y_3(T) = f_3 R_1^* T \end{cases} \quad (6)$$

This system of three equations has three unknowns f_2 , f_3 and R_1^* . The fractions of doubles f_2 and triples f_3 as well as the hypothetical count rate R_1^* can be determined by taking the slopes of Eqs. (6). These equations are better illustrated by looking at the quantities $\bar{C}(T)$, $Y_2(T)$, and $Y_3(T)$ for a californium source. The second and third moments of the count distributions obtained for incident neutrons of kinetic energy between 5 and 5.5 MeV are shown in Figs. 6(b) and 6(c). The slopes of lines through

Figure 6: Moments $\bar{C}(T)$, $Y_2(T)$ and $Y_3(T)$ of count distributions for incident neutrons of energy between 5 and 5.5 MeV. Scintillation light output threshold $LO_{\text{threshold}}$ set to 0 keVee. ^{252}Cf of intensity $151\mu\text{Ci}$ measured for 24 hours.

the data points determine the fractions of double and triple scattering. As explained in Ref. [9], the first data point of $Y_2(T)$ is not collinear with the other ones, because it takes several nanoseconds for a neutron to travel from one detector to another one. The same is true for the first data point of $Y_3(T)$.

4. Experimental results

A distance of 254 cm between the californium source and the wall of detectors was chosen so as to minimize the contribution of spontaneous fission correlated terms while keeping a reasonable neutron detection efficiency.

Fig. 7 shows the hypothetical neutron count rate R_1^* from Eq. 6 as a function of the source neutron energy. For a scintillation light output threshold of 100 keVee, the black curve resembles the ^{252}Cf fission spectrum (Fig. 7(b)) for neutron kinetic energies between 2 MeV all the way up to 10 MeV.

When the scintillation light output threshold is set to 250 keVee (red curve), we lose overall many fast neutrons that produce less than 250 keVee in the liquid scintillators,

(a) Hypothetical neutron count rates R_1^* as a function of incident neutron energy, for different scintillation light output thresholds. ^{252}Cf of intensity $151\mu\text{Ci}$ measured for 5 days. (b) Simulated ^{252}Cf spontaneous fission neutron spectrum. Simulation was performed using the LLNL Fission Library/FREYA package [17].

Figure 7: (a) Experimental measurement of energy distribution of ^{252}Cf spontaneous fission neutrons using time-of-flight. (b) ^{252}Cf fission neutron spectrum.

even if these neutrons have kinetic energies of multiple MeVs to start with. Indeed, using the light output function² in Ref. [18], the scintillation light output from neutrons of kinetic energy 2 MeV is at the most 456 keVee. Several of these 2 MeV neutrons will produce less than 250 keVee, and not be detected. As the neutron kinetic energy decreases, the maximum scintillation light produced by the neutrons rapidly falls. In fact, neutrons become undetectable when their kinetic energy falls below 1.4 MeV, because the maximum scintillation light output from 1.4 MeV neutrons is 250 keVee [18]. This explains why the rates of neutrons with kinetic energies below 2 MeV rapidly decrease.

For kinetic energies below 1.2 MeV, we observe an anomalous rise in the count rate. The rise in count rate for decreasing source neutron energies is due to (a) the finite detection efficiency of the trigger detector³ and (b) neutron scattering off neighboring surfaces. Indeed, regarding (a), imagine that a timer detector records a spontaneous fission neutron, but the trigger detector fails to detect the spontaneous fission. If a prior spontaneous fission was however detected by the trigger detector, the time interval ΔT in Eq. (3) would then measure the time interval between a detected spontaneous fission neutron and the time — not of the spontaneous fission generating the detected neutron but — of a prior spontaneous fission reaction. Given Eq. (4), the detected neutron will pass as a neutron with a much lower kinetic energy. Regarding (b), the effect is similar, if a neutron scatters off material in the surroundings to then record a count in the liquid scintillators, its time of flight will be artificially longer, and it will pass for a neutron of lower kinetic energy. This effect was minimized by running the experiment on a low-mass floor.

² $\text{LO}(E_p) = 0.817E_p - 2.63(1 - e^{-0.297E_p})$. The non linear light output function strongly depends on the geometry of the scintillators, the PMT settings and the method to integrate the charge under the pulses. The light output function listed here is for a scintillator whose geometry is close to the ones used in this paper.

³In average, spontaneous fissions emit 3.757 neutrons [20] and 8.32 photons [21]. Failure to detect a spontaneous fission is thus low.

As the neutron detector threshold increases, the valley between the first and second peaks shifts to the right, because the neutron quenching effect suppresses more and more fast neutrons.

Plots of the double and triple scattering fractions are given in Figs. 8(a) and 8(b) for scintillation light output thresholds of 100 keVee to 750 keVee. Comparing these re-

Figure 8: Multiple scattering probabilities of neutrons as a function of incident neutron energy. Varying scintillation light output thresholds $LO_{\text{threshold}}$. ^{252}Cf of intensity $151\mu\text{Ci}$ measured for 5 days.

sults with the ones obtained by simulation (Fig. 8 of Ref. [9] with a 250 keVee neutron detection threshold), we observe a good qualitative agreement for neutrons of kinetic energies above 1.5 MeV. The differences in magnitude can be attributed to the different liquid scintillator array configurations (experiment configuration in Fig. 2 versus simulated configuration in Fig. 1 of Ref. [9]). Below 1.5 MeV, there is a discrepancy between the experimental and simulation results. For the same reasons as the rise in the count rate (shown in Fig. 7) for neutron kinetic energies below 1.2 MeV, this discrepancy is related to the aforementioned neutrons scattering off neighboring materials and to the finite detection efficiency of the trigger detector.

The values of the multiple scattering fractions along with their uncertainties can be read in Tables 1- 2. For a given scintillation light output threshold, multiple scattering probabilities f_2 and f_3 for source neutron kinetic energies lower than the location of the valley should be set to 0, because these are not due to neutron multiple scattering but to the experimental errors.

Table 1: Fractions f_2 and f_3 of neutrons double- and triple-scattering between liquid scintillators, for neutrons incident on the array shown in Fig. 2. The liquid scintillator energy thresholds are 100 and 250 keV. ^{252}Cf of intensity $151\mu\text{Ci}$ measured for 5 days.

Neutron energy range [keV]	$LO_{\text{threshold}} = 100 \text{ keVee}$		$LO_{\text{threshold}} = 250 \text{ keVee}$	
	f_2 [%]	f_3 [%]	f_2 [%]	f_3 [%]
0-250	0.74 ± 0.00	0.0038 ± 0.0003	below threshold	below threshold
250-500	0.64 ± 0.00	0.0029 ± 0.0003	0.39 ± 0.00	0.0008 ± 0.0002
500-750	0.65 ± 0.01	0.0031 ± 0.0004	0.39 ± 0.01	0.0012 ± 0.0003
750-1000	0.44 ± 0.00	0.0020 ± 0.0003	0.41 ± 0.01	0.0017 ± 0.0004
1000-1250	0.23 ± 0.00	0.0010 ± 0.0002	0.38 ± 0.01	0.0018 ± 0.0005
1250-1500	0.14 ± 0.00	0.0004 ± 0.0001	0.16 ± 0.00	0.0005 ± 0.0000
1500-1750	0.13 ± 0.00	0.0004 ± 0.0001	0.08 ± 0.00	0.0002 ± 0.0000
1750-2000	0.17 ± 0.00	0.0003 ± 0.0001	0.07 ± 0.00	0.0001 ± 0.0001

Neutron energy range [keV]	LO _{threshold} =100 keVee		LO _{threshold} =250 keVee	
	f_2 [%]	f_3 [%]	f_2 [%]	f_3 [%]
2000-2250	0.24 ± 0.00	0.0005 ± 0.0001	0.09 ± 0.00	0.0001 ± 0.0000
2250-2500	0.35 ± 0.00	0.0008 ± 0.0001	0.12 ± 0.00	0.0002 ± 0.0001
2500-2750	0.51 ± 0.00	0.0009 ± 0.0001	0.15 ± 0.00	0.0001 ± 0.0000
2750-3000	0.70 ± 0.00	0.0016 ± 0.0002	0.20 ± 0.00	0.0003 ± 0.0001
3000-3250	0.92 ± 0.01	0.0025 ± 0.0003	0.29 ± 0.00	0.0005 ± 0.0001
3250-3500	1.16 ± 0.01	0.0032 ± 0.0003	0.41 ± 0.00	0.0006 ± 0.0001
3500-3750	1.40 ± 0.01	0.0048 ± 0.0004	0.57 ± 0.00	0.0012 ± 0.0002
3750-4000	1.69 ± 0.01	0.0054 ± 0.0005	0.78 ± 0.01	0.0008 ± 0.0002
4000-4250	1.93 ± 0.01	0.0091 ± 0.0007	0.98 ± 0.01	0.0029 ± 0.0004
4250-4500	2.18 ± 0.01	0.0109 ± 0.0009	1.20 ± 0.01	0.0033 ± 0.0005
4500-4750	2.37 ± 0.01	0.0139 ± 0.0011	1.39 ± 0.01	0.0042 ± 0.0006
4750-5000	2.59 ± 0.02	0.0150 ± 0.0012	1.61 ± 0.01	0.0048 ± 0.0006
5000-5250	2.71 ± 0.02	0.0182 ± 0.0014	1.75 ± 0.01	0.0062 ± 0.0008
5250-5500	2.87 ± 0.02	0.0217 ± 0.0017	1.93 ± 0.02	0.0070 ± 0.0010
5500-5750	2.99 ± 0.02	0.0272 ± 0.0025	2.08 ± 0.02	0.0102 ± 0.0016
5750-6000	3.09 ± 0.03	0.0233 ± 0.0021	2.17 ± 0.02	0.0085 ± 0.0013
6000-6250	3.20 ± 0.03	0.0288 ± 0.0025	2.30 ± 0.03	0.0120 ± 0.0017
6250-6500	3.26 ± 0.03	0.0292 ± 0.0029	2.37 ± 0.03	0.0136 ± 0.0020
6500-6750	3.39 ± 0.04	0.0317 ± 0.0031	2.52 ± 0.03	0.0162 ± 0.0023
6750-7000	3.42 ± 0.04	0.0393 ± 0.0038	2.56 ± 0.03	0.0147 ± 0.0025
7000-7250	3.47 ± 0.04	0.0390 ± 0.0044	2.64 ± 0.04	0.0218 ± 0.0036
7250-7500	3.39 ± 0.05	0.0356 ± 0.0044	2.53 ± 0.04	0.0200 ± 0.0035
7500-7750	3.48 ± 0.05	0.0522 ± 0.0061	2.72 ± 0.05	0.0235 ± 0.0047
7750-8000	3.44 ± 0.06	0.0397 ± 0.0059	2.70 ± 0.05	0.0192 ± 0.0043
8000-8250	3.49 ± 0.06	0.0482 ± 0.0071	2.75 ± 0.06	0.0225 ± 0.0047
8250-8500	3.54 ± 0.07	0.0426 ± 0.0068	2.80 ± 0.06	0.0171 ± 0.0045
8500-8750	3.55 ± 0.07	0.0437 ± 0.0074	2.80 ± 0.07	0.0247 ± 0.0059
8750-9000	3.32 ± 0.08	0.0390 ± 0.0073	2.67 ± 0.07	0.0211 ± 0.0056
9000-9250	3.43 ± 0.09	0.0470 ± 0.0094	2.77 ± 0.08	0.0331 ± 0.0085
9250-9500	3.41 ± 0.09	0.0452 ± 0.0094	2.75 ± 0.08	0.0322 ± 0.0080
9500-9750	3.30 ± 0.10	0.0414 ± 0.0088	2.65 ± 0.09	0.0265 ± 0.0074
9750-10000	3.32 ± 0.11	0.0602 ± 0.0129	2.86 ± 0.10	0.0333 ± 0.0107

Table 2: Fractions f_2 and f_3 of neutrons double- and triple-scattering between liquid scintillators, for neutrons incident on the array shown in Fig. 2. The liquid scintillator energy thresholds are 500 and 750 keV. ^{252}Cf of intensity $151\mu\text{Ci}$ measured for 5 days.

Neutron energy range [keV]	LO _{threshold} =500 keVee		LO _{threshold} =750 keVee	
	f_2 [%]	f_3 [%]	f_2 [%]	f_3 [%]
500-750	0.14 ± 0.00	0.0003 ± 0.0074	below threshold	below threshold
750-1000	0.15 ± 0.01	0.0004 ± 0.0003	0.08 ± 0.01	0.0000 ± 0.0000
1000-1250	0.15 ± 0.01	0.0000 ± 0.0000	0.06 ± 0.01	0.0000 ± 0.0000
1250-1500	0.16 ± 0.01	0.0000 ± 0.0000	0.09 ± 0.01	0.0000 ± 0.0000
1500-1750	0.14 ± 0.01	0.0005 ± 0.0000	0.09 ± 0.01	0.0000 ± 0.0000
1750-2000	0.09 ± 0.01	0.0000 ± 0.0000	0.08 ± 0.01	0.0000 ± 0.0000
2000-2250	0.03 ± 0.00	0.0000 ± 0.0000	0.05 ± 0.01	0.0000 ± 0.0000
2250-2500	0.02 ± 0.00	0.0000 ± 0.0000	0.04 ± 0.01	0.0000 ± 0.0000
2500-2750	0.03 ± 0.00	0.0000 ± 0.0000	0.02 ± 0.00	0.0000 ± 0.0000
2750-3000	0.04 ± 0.00	0.0001 ± 0.0000	0.01 ± 0.00	0.0000 ± 0.0000
3000-3250	0.05 ± 0.00	0.0001 ± 0.0001	0.01 ± 0.00	0.0000 ± 0.0000
3250-3500	0.06 ± 0.00	0.0000 ± 0.0000	0.01 ± 0.00	0.0000 ± 0.0000
3500-3750	0.08 ± 0.00	0.0000 ± 0.0000	0.02 ± 0.00	0.0000 ± 0.0000
3750-4000	0.10 ± 0.00	0.0000 ± 0.0000	0.03 ± 0.00	0.0000 ± 0.0000
4000-4250	0.11 ± 0.00	0.0000 ± 0.0000	0.03 ± 0.00	0.0000 ± 0.0000

Neutron energy range [keV]	LO _{threshold} = 500 keVee		LO _{threshold} = 750 keVee	
	f_2 [%]	f_3 [%]	f_2 [%]	f_3 [%]
4250-4500	0.15 ± 0.00	0.0001 ± 0.0000	0.05 ± 0.00	0.0000 ± 0.0000
4500-4750	0.19 ± 0.00	0.0002 ± 0.0001	0.06 ± 0.00	0.0001 ± 0.0001
4750-5000	0.27 ± 0.01	0.0004 ± 0.0000	0.07 ± 0.00	0.0000 ± 0.0000
5000-5250	0.34 ± 0.01	0.0006 ± 0.0003	0.07 ± 0.00	0.0002 ± 0.0000
5250-5500	0.41 ± 0.01	0.0005 ± 0.0003	0.08 ± 0.00	0.0000 ± 0.0000
5500-5750	0.51 ± 0.01	0.0010 ± 0.0004	0.09 ± 0.00	0.0000 ± 0.0000
5750-6000	0.60 ± 0.01	0.0004 ± 0.0000	0.13 ± 0.01	0.0000 ± 0.0000
6000-6250	0.70 ± 0.01	0.0010 ± 0.0000	0.15 ± 0.01	0.0000 ± 0.0000
6250-6500	0.79 ± 0.02	0.0019 ± 0.0008	0.18 ± 0.01	0.0000 ± 0.0000
6500-6750	0.92 ± 0.02	0.0020 ± 0.0010	0.23 ± 0.01	0.0000 ± 0.0000
6750-7000	0.99 ± 0.02	0.0004 ± 0.0000	0.28 ± 0.01	0.0000 ± 0.0000
7000-7250	1.07 ± 0.03	0.0043 ± 0.0016	0.33 ± 0.01	0.0000 ± 0.0000
7250-7500	1.08 ± 0.03	0.0046 ± 0.0018	0.37 ± 0.02	0.0007 ± 0.0000
7500-7750	1.21 ± 0.03	0.0041 ± 0.0000	0.48 ± 0.02	0.0000 ± 0.0000
7750-8000	1.25 ± 0.04	0.0032 ± 0.0016	0.51 ± 0.02	0.0000 ± 0.0000
8000-8250	1.26 ± 0.04	0.0058 ± 0.0027	0.52 ± 0.03	0.0034 ± 0.0000
8250-8500	1.39 ± 0.04	0.0000 ± 0.0000	0.62 ± 0.03	0.0000 ± 0.0000
8500-8750	1.44 ± 0.05	0.0000 ± 0.0000	0.64 ± 0.03	0.0000 ± 0.0000
8750-9000	1.33 ± 0.05	0.0032 ± 0.0023	0.66 ± 0.04	0.0017 ± 0.0017
9000-9250	1.46 ± 0.06	0.0058 ± 0.0000	0.71 ± 0.05	0.0044 ± 0.0000
9250-9500	1.51 ± 0.06	0.0021 ± 0.0021	0.73 ± 0.05	0.0000 ± 0.0000
9500-9750	1.49 ± 0.07	0.0028 ± 0.0000	0.77 ± 0.05	0.0000 ± 0.0000
9750-10000	1.58 ± 0.08	0.0028 ± 0.0028	0.88 ± 0.06	0.0000 ± 0.0000

Regarding the terms linear in ϵ and ϵ^2 in Eqs. (5), let us assess their magnitudes. A distance of 254 cm between the californium source and the wall of detectors was chosen so as to minimize the contribution of spontaneous fission correlated terms while keeping a reasonable neutron detection efficiency. Given this distance, the solid angle spanned by the 40 detectors is approximately 0.4%. Assuming each detector detects at the most 15% of the neutrons (detection efficiency peaks for neutrons with kinetic energies between 3 and 4 MeV) for a detector energy threshold of 100 keVee, the neutron detection efficiency ϵ is 0.06%. This efficiency decreases with higher neutron energy thresholds. Let us enumerate the correlated terms due to spontaneous fission in Eqs. (5):

- (a) Two spontaneous fission neutrons from ^{252}Cf can be counted within the short time window, and introduce a correlation similar to a double scattering correlation. Such occurrences contribute the term $(1 + f_2 + 2f_3)^2 \epsilon D_{2s}$ to $Y_2(T)$. This term is approximately equal to 10^{-3} in our geometry, which corresponds to the very first tick mark on the y-axis in Fig. 8(a). The effect is thus clearly negligible.
- (b) The term $2(1 + f_2 + 2f_3)(f_2 + 3f_3) \epsilon D_{2s}$ in $Y_3(T)$ is the result of two spontaneous fission neutrons detected, one of which is detected twice. This effect introduces a maximum increase of 5×10^{-5} for the 100 keVee threshold curve in Fig. 8(b). This is again a small effect.
- (c) Three spontaneous fission neutrons from ^{252}Cf can be counted within the short time window, and introduce a correlation similar to a triple scattering correlation, leading to the term $(1 + f_2 + 2f_3)^3 \epsilon^2 D_{3s}$ in $Y_3(T)$. In our detector geometry, this effect is of the order of 5×10^{-7} , which is negligible on the scale of Fig. 8(b).

216 An experiment conducted with a californium source 127 cm away from the wall of
 217 detectors (instead of 254 cm) confirmed that the effects (a) and (b) raise the curves in
 218 Fig. 8. The effects are more pronounced for the 100 keV curves where the neutron
 219 detection efficiency ε is highest.

220 5. Using spectrum of energy deposited by neutrons to apply correction to system 221 parameters

222 When measuring nuclear materials emitting neutrons with our array of liquid scin-
 223 tillators, moments of count distributions can be used in conjunction with the measured
 224 deposited energy spectrum to apply corrections to the estimates of the masses of the
 225 sources under measurement, the system multiplication M , and the detection efficiency
 226 ε [9, 10].

For liquid scintillators, neutrons can scatter multiple times between scintillators.
 The moments of the count distributions are given as

$$\begin{cases} \bar{C}(T) = \left[(1 - f_2 - f_3) + \binom{2}{1} f_2 + \binom{3}{1} f_3 \right] \varepsilon q(M) M \bar{v}_{sp} F_s T \\ Y_{2F}(T) = \frac{f_2 + 3f_3}{1 + f_2 + 2f_3} + (1 + f_2 + 2f_3) \varepsilon q(M) M [D_{2s} + (M - 1)D_2] \left(1 - \frac{1 - e^{-\alpha T}}{\alpha T} \right) \\ Y_{3F}(T) = \frac{f_3}{1 + f_2 + 2f_3} + 2(f_2 + 3f_3) \varepsilon q(M) M [D_{2s} + (M - 1)D_2] \left(1 - \frac{1 - e^{-\alpha T}}{\alpha T} \right) \\ \quad + (1 + f_2 + 2f_3)^2 (\varepsilon q(M) M)^2 \\ \quad \left[[D_{3s} + (M - 1)D_3] \left(1 - \frac{3 - 4e^{-\alpha T} + e^{-2\alpha T}}{2\alpha T} \right) + [2(M - 1)D_{2s}D_2 + 2(M - 1)^2 D_2^2] \left[1 - \frac{2 - (2 + \alpha T)e^{-\alpha T}}{\alpha T} \right] \right] \end{cases} \quad (7)$$

227 This system of equations (7) can be solved for the source strength F_s , the system mul-
 228 tiplication M and the detection efficiency ε , but the multiple scattering probabilities f_2
 229 and f_3 need be estimated. Estimating f_2 and f_3 can be achieved using the spectrum of
 230 energy deposited by the source neutrons in the liquid scintillators.

231 The measured spectra of energies deposited by fast neutrons are shown in Fig. 9 for
 232 various neutron kinetic energy ranges. We can reconstruct the neutron energy spectrum
 233 measured by our array of liquid scintillators using this set of basis spectra shown in
 234 Fig. 9. Let us use the subscript E_i to distinguish each one of the basis functions $g_{E_i}(E_d)$
 235 shown in Fig. 9. The subscript E_i denotes the initial source neutron energy range, from
 236 1000 keV to 1250 keV, all the way up to 9750 keV to 10000 keV, in increments of
 237 250 keV.

Let's call $g^r(E_d)$ a reconstructed energy deposition spectrum. $g^r(E_d)$ is defined as

$$g^r(E_d) = \sum_{i=1}^N w_{E_i} g_{E_i}(E_d) \quad (8)$$

Given a measured spectrum of deposited energies $\tilde{g}(E_d)$, the goal for the reconstruction
 is to find the optimal set of weights w_{E_i} for which the difference

$$g^r(E_d) - \tilde{g}(E_d) \quad (9)$$

Figure 9: Probability density functions of energy deposited by neutrons as they traverse the liquid scintillators for different incident neutron energies.

is minimized.

The weights w_{E_i} are physically to be interpreted as the strengths of the neutron source in the energy range E_i . If the distribution $\tilde{g}(E_d)$ is normalized, the weights w_{E_i} will be the relative strengths of the different neutron sources in the energy range E_i .

The fractions of double scattering f_2 and triple scattering f_3 are estimated from weighing the fractions $f_2(E_i)$ and $f_3(E_i)$ for different source energy ranges E_i by w_{E_i} :

$$f_2 = \sum_{i=1}^N w_{E_i} f_2(E_i) \quad (10)$$

$$f_3 = \sum_{i=1}^N w_{E_i} f_3(E_i) \quad (11)$$

where the functions $f_2(E_i)$ and $f_3(E_i)$ are the curves shown in Figs. 8. Knowing the multiple scattering fractions f_2 and f_3 from Eqs. (10)-(11), the source strength F_s , the system multiplication M and the detection efficiency ε can now be determined using the system of equations (7).

6. Conclusion

The Feynman variance to mean approach has been used for decades to characterize fissile materials using helium-3 tubes. Whereas helium tubes capture neutrons, liquid scintillators scatter them. This difference leads to expressions for the moments of count distributions that include scattering corrections. These corrections need to be known to characterize fissile materials in terms of source strength F_s and multiplication M . In this paper, we experimentally measured the fractions of neutrons double- and triple-scattering between different liquid scintillators for a series of neutron kinetic energies. The neutron kinetic energies were measured using a neutron time-of-flight approach.

255 Deposited energy spectra measured by the liquid scintillators are used to determine
256 the double- and triple- scattering fractions. These fractions in the corrected moment
257 equations lead to better estimate of the fissile material mass and its multiplication.

258 Measuring the spectrum of fast neutrons depositing energy in the liquid scintillators
259 also reveals spectral information about the source of neutrons.

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265 Nomenclature

LO	=	electron-equivalent energy deposited by fast neutrons and measured by scintillators.
I_{ADC}	=	integral of ADC counts recorded by the PMT pulse digitizer.
a	=	proportionality coefficient between LO and I_{ADC} .
$(x_{\text{src}}, y_{\text{src}}, z_{\text{src}})$	=	euclidean coordinates of the spontaneous fission source.
$(x_{\gamma}, y_{\gamma}, z_{\gamma})$	=	euclidean coordinates of trigger detector.
(x_n, y_n, z_n)	=	euclidean coordinates of a timer detector.
ΔT	=	time interval between gamma ray detection and fast neutron detection.
c	=	speed of light.
v_n	=	speed of the fast neutron.
$E_{\text{k,n}}$	=	kinetic energy of fast neutron.
T	=	time gate duration.
$b_n(T)$	=	probability to get n counts in a random time gate of duration T .
$\bar{C}(T)$	=	number of counts averaged over all time gates of duration T .
R_1^*	=	the hypothetical count rate which one would measure if individual neutrons could not be counted multiple times.
$Y_{2\text{F}}(T)$	=	the excess over unity of the variance to mean ratio of $b_n(T)$, or physically speaking the correlated pairs relative to the counts, sometimes referred to as the Feynman correlated moment.
$Y_2(T)$	=	$Y_{2\text{F}}(T)$ multiplied by $\bar{C}(T)$.
$Y_{3\text{F}}(T)$	=	the skewness to mean ratio of $b_n(T)$, or physically speaking the correlated triples relative to the counts.
$Y_3(T)$	=	$Y_{3\text{F}}(T)$ multiplied by $\bar{C}(T)$.
f_2	=	probability of counting individual neutrons twice.
f_3	=	probability of counting individual neutrons thrice.
D_n	=	n^{th} combinatorial moment of induced fission multiplicity distribution.
$D_{n \text{ sp}}$	=	n^{th} combinatorial moment of spontaneous fission multiplicity distribution.
E_p	=	energy of proton recoil.
$\tilde{g}(E_d)$	=	measured liquid scintillator spectrum.
$g_{E_i}(E_d)$	=	probability that a source neutron of initial energy E_i will deposit an electron-equivalent energy within bin E_d . Basis functions for spectral reconstruction.
$g^r(E_d)$	=	reconstructed liquid scintillator spectrum using basis functions $g_{E_i}(E_d)$.
267		
268 Greek:		
ε	=	neutron detection efficiency, or probability to detect a neutron.
$\bar{V}$	=	average number of neutrons produced in induced fission.
$\bar{V}_{\text{sp}}$	=	average number of neutrons produced in spontaneous fission.
α	=	inverse fission chain evolution time scale.
269		

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