Neutron Crosstalk between Liquid Scintillators

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Abstract

A method is proposed to quantify the fractions of neutrons scattering between liquid scintillator cells. Using a spontaneous fission source, this method can be utilized to quickly characterize an array of liquid scintillators in terms of crosstalk. The point model theory due to Feynman is corrected to account for these multiple scatterings. Using spectral information measured by the liquid scintillators, fractions of multiple scattering can be estimated, and mass reconstruction of fissile materials under investigation can be improved. Monte Carlo simulations of mono-energetic neutron sources were performed to estimate neutron crosstalk. A Californium source in an array of liquid scintillators was modeled to illustrate the improvement of the mass reconstruction.

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

1. Introduction

The purpose of this work is to quantify the fractions of fast neutrons scattering between adjacent liquid scintillator cells, a phenomenon also known as neutron crosstalk. We propose a new method to quantify them. While this paper will focus on theoretical development to model neutron crosstalk, and show simulation results, the strength of this new method lies in that it can be used to determine neutron crosstalk experimentally using fissile materials as neutron sources. This will enable experimentalists to quickly characterize their array of liquid scintillators in terms of multiple scattering fractions/neutron crosstalk, the same way as instruments are calibrated in energy, and cells are synchronized in time, prior to taking data. The authors are currently working on a second paper, where an array of liquid scintillators is thus characterized experimentally. Preliminary results are available in Ref. [1].

It is well known [2, 3, 4, 5] that masses of nuclear materials undergoing fissions can be determined using ^3He tubes counting thermal neutrons. For general neutron multiplicity counting (NMC) applications, the theory usually assumes that neutrons can only be counted once. This assumption is correct for ^3He tubes, where neutrons are captured when counted. However, for liquid scintillators, a neutron can scatter and deposit enough energy in multiple liquid scintillator cells to record multiple counts. The measured count rate and the two- and three-neutron correlations will thus be adversely increased. NMC applications are very sensitive to two-, three- and higher order correlations. With fast neutrons registering multiple correlated counts, the standard Feynman

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moment method [2, 3, 4, 5] will fail. In this paper, we derive new expressions for the Feynman moment method to theoretically account for multiple scattering. In this paper, new expressions of the Feynman correlated moments are proposed to account for multiple scattering.

A summary of this paper is as follows. In Sec. 2, we derive new expressions for the Feynman correlated moments to account for individual neutrons scattering and recording multiple counts in detectors.

Neutron Since neutron crosstalk depends on the energy of the neutrons incident on the liquid scintillator cells detectors, it must thus be characterized as a function of this incident neutron energy. By measuring the energies deposited in the liquid scintillator cells, we can infer the spectrum of incident neutrons and in turn the expected multiple scattering fractions. In Sec. 3, we simulate the liquid scintillator response to different mono-energetic neutron sources, from which we infer the spectrum of incident neutrons (Sec. 4), and in turn the expected multiple scattering fractions (Sec. 5). The inferred neutron source energy spectrum reveals information about the source under investigation. This spectral information has been successfully used experimentally in Ref. [6]. This paper shows how to infer the multiple scattering fractions from measuring the spectrum of energies deposited. Secondly, it also shows that the spectrum of energy deposited by fast neutrons reveals information about the source under investigation itself, via its inferred energy spectrum. Such spectral information has been successfully used experimentally [6].

In the last part of this paper, Sec. 6, we simulate a ^{252}Cf source in a modeled array of liquid scintillators. We will show that the multiple scattering fractions inferred from the spectrum of deposited energies can be used to apply corrections to the masses of fissile materials undergoing fission.

A list of symbols used throughout the text is available in the appending nomenclature.

2. Theory

When a multiplying object is placed in the center of a liquid scintillator array such as the one shown in Fig. 1, one can experimentally measure the times of arrival of the neutrons in each of the liquid scintillator cells. Randomly splitting the sequence of time tags into N segments of length T — where T is of the order of nanoseconds to hundreds of microseconds — one can count how many neutrons arrive in the first segment, how many in the second segment, in the third one, etc. allows one to build a distribution $b_n(T)$ of the number n of neutrons arriving in the segments of length T . Randomly splitting the sequence of time tags into N segments of duration T ,¹ one can count how many neutrons arrive in the first segment, how many in the second segment, in the third one, etc. and build the distribution $B_n(T)$ of the number of neutrons n arriving in the segments of duration T . By repeating this procedure for segments of different lengths durations T , multiple count distributions $b_n(T)B_n(T)$ can be obtained.

¹ T is of the order of nanoseconds to hundreds of microseconds

Figure 1: (color online) Multiplying object in the middle of a 77 liquid scintillator array.

These count distributions $b_n(T)$ can be used to determine the strength intensity F_s of the spontaneous fission sources in the object, the efficiency ϵ of the liquid scintillator array and the multiplication M of the multiplying object. This will be shown by way of the following three equations. Defining $b_n(T)$ as the probability distribution of the count distribution $B_n(T)$,² one can show theoretically that the first moment of the count probability distribution $b_n(T)$ can be written as [3, 4]

$$\bar{C}(T) = \epsilon q M \bar{v}_{\text{sp}} F_s (1 + A) T \quad (1)$$

where F_s is the strength intensity of the spontaneous fission source (in units of spontaneous fissions per second), and A is the α -ratio, i.e. the ratio of neutrons emitted by sources emitting single neutrons to neutrons emitted by sources emitting multiple neutrons simultaneously. In this equation, let us recall that the multiplication M is defined as

$$M = \frac{1}{1 - p \bar{v}} \quad (2)$$

where p is the probability that a neutron will fission a nucleus, and $q = 1 - p$ is the probability that a neutron does not induce fission. $q(M)$ is usually referred to as the escape multiplication³ and is given by

$$qM = M - (M - 1)/\bar{v} \quad (3)$$

The symbols $\bar{v}$ and $\bar{v}_{\text{sp}}$ are the average numbers of neutrons emitted per induced and

²which is equivalent to normalizing $B_n(T)$ by the number of segments of duration T : $b_n(T) = B_n(T)/N$

³or leakage multiplication

spontaneous fissions, respectively. $\bar{\nu}$ can be calculated as

$$\bar{\nu} = \sum_{n=1}^8 n C_n \quad (4)$$

where C_n is the probability of emitting n neutrons per induced fission. The upper limit of 8 on the summation sign is the largest number of neutrons that known isotopes produce per fission. The probability distribution C_n depends on the energy of the neutron inducing fission. To obtain $\bar{\nu}_{sp}$, the multiplicity distribution C is replaced by C^{sp} .

For time-gated fast neutron counting, the Feynman correlated moment Y_{2F} , which is the excess over unity of the variance to mean ratio of $b_n(T)$, or physically speaking the correlated pairs relative to the counts, is a generalization of Eq. (132) in Ref. [4] divided by $\bar{C}(T)$ (Eq. (1) above),

$$Y_{2F}(T) = \varepsilon q M \left[\frac{D_{2s}}{1+A} + (M-1)D_2 \right] \left(1 - \frac{1-e^{-\alpha T}}{\alpha T} \right) \quad (5)$$

where α is a time constant related to the transport of the neutrons in both the measured object and the detection system. D_{2s}, D_2, D_{3s}, D_3 are combinatorial moments of spontaneous and induced fission neutron number multiplicity distributions. They depend on nuclear data, and are in general given by

$$D_n = \frac{\sum_{i=n}^8 \binom{i}{n} C_i}{\bar{\nu}}. \quad (6)$$

For spontaneous fission nuclear data, $D_{n,sp}$ is calculated similarly, replacing the multiplicity distribution C by the multiplicity distribution C^{sp} .

Y_{3F} , which is the skewness to mean ratio of $b_n(T)$, or physically speaking the correlated triples relative to the counts, is a generalization of Eq. (133) in Ref. [4] divided by $\bar{C}(T)$ (Eq. (1) above),

$$Y_{3F} = (\varepsilon q M)^2 \left[\left[\frac{D_{3s}}{1+A} + (M-1)D_3 \right] \left(1 - \frac{3-4e^{-\alpha T} + e^{-2\alpha T}}{2\alpha T} \right) + \left[2(M-1) \frac{D_{2s}D_2}{1+A} + 2(M-1)^2 D_2^2 \right] \left(1 - \frac{2-(2+\alpha T)e^{-\alpha T}}{\alpha T} \right) \right] \quad (7)$$

It is important to note that for uncorrelated random Poisson sources, Y_{2F} and Y_{3F} are identically zero and therefore provide a quantitative measure of correlation. The two values R_{3F_1} and R_{3F_2} are the two components of Eq. (7). We define R_{3F_1} and R_{3F_2} as

$$R_{3F_1} = (\varepsilon q M)^2 \left[\frac{D_{3s}}{1+A} + (M-1)D_3 \right] \quad (8)$$

$$R_{3F_2} = (\varepsilon q M)^2 \left[2(M-1) \frac{D_{2s}D_2}{1+A} + 2(M-1)^2 D_2^2 \right] \quad (9)$$

so that Eq. (7) can be rewritten as

$$Y_{3F} = R_{3F_1} \left(1 - \frac{3 - 4e^{-\alpha T} + e^{-2\alpha T}}{2\alpha T} \right) + R_{3F_2} \left(1 - \frac{2 - (2 + \alpha T)e^{-\alpha T}}{\alpha T} \right) \quad (10)$$

Fig. 2 shows an example of $Y_{2F}(T)$ and $Y_{3F}(T)$. The time constant α^{-1} is 6 ns and the asymptotical value of $Y_{2F}(T)$ is 0.1174.

Figure 2: (color online) Examples of $Y_{2F}(T)$ and $Y_{3F}(T)$. λ^{-1} refers to α^{-1} .

The slope of Eq. (1) and the asymptotes of Eqs. (5) and (7) for large time gate widths T can be written as

$$\begin{cases} R_1 = \varepsilon q M \bar{v}_{sp} F_s (1 + A) \\ R_{2F} = \varepsilon q M \left[\frac{D_{2s}}{1+A} + (M-1)D_2 \right] \\ R_{3F} = (\varepsilon q M)^2 \left[\frac{D_{3s}}{1+A} + (M-1)D_3 + 2(M-1) \frac{D_{2s}D_2}{1+A} + 2(M-1)^2 D_2^2 \right] \end{cases} \quad (11)$$

This system of 3 equations has 4 unknowns ε , M , F_s and A . In the absence of (α, n) sources which emit single neutrons at a time, the ratio A is equal to zero. In this case, the system of equations (11) reduces to

$$\begin{cases} R_1 = \varepsilon q M \bar{v}_{sp} F_s \\ R_{2F} = \varepsilon q M [D_{2s} + (M-1)D_2] \\ R_{3F} = (\varepsilon q M)^2 [D_{3s} + (M-1)D_3 + 2(M-1)D_{2s}D_2 + 2(M-1)^2 D_2^2] \end{cases} \quad (12)$$

2.1. Detector corrections

These equations rely on the assumption that each neutron in the system can be detected only once. This is certainly true for a detection system based on ^3He tubes, because ^3He captures the neutron and the neutron disappears from the system. It is different in an array of liquid scintillator cells, because neutrons in such a system deposit energy in cells by scattering with the atoms, and are not absorbed in these elastic and inelastic scattering collisions. They keep traveling, and if they still have enough energy, they can potentially deposit this energy in adjacent or even remote cells. If a neutron

scatters multiple times between liquid scintillator cells, Eqs. (1), (5) and (7) no longer hold. They can however be replaced by the count rate $\bar{C}(T)$ and the moments $Y_{2F}(T)$ and $Y_{3F}(T)$ ones that account for the multiple scattering of neutrons (the derivation of these equations is in Appendix A and assumes no more than triply counted neutrons.⁴):

$$\begin{cases} \bar{C}(T) = \left[(1 - f_2 - f_3) + \binom{2}{1} f_2 + \binom{3}{1} f_3 \right] R_1^* T \\ Y_{2F}(T) = \frac{f_2 + 3f_3}{1 + f_2 + 2f_3} + (1 + f_2 + 2f_3) \varepsilon q M \left[\frac{D_{2s}}{1+A} + (M-1)D_2 \right] \left(1 - \frac{1 - e^{-\alpha T}}{\alpha T} \right) \\ Y_{3F}(T) = \frac{f_3}{1 + f_2 + 2f_3} + 2(f_2 + 3f_3) \varepsilon q M \left[\frac{D_{2s}}{1+A} + (M-1)D_2 \right] \left(1 - \frac{1 - e^{-\alpha T}}{\alpha T} \right) \\ \quad + (1 + f_2 + 2f_3)^2 (\varepsilon q M)^2 \\ \quad \left[\left[\frac{D_{3s}}{1+A} + (M-1)D_3 \right] \left(1 - \frac{3 - 4e^{-\alpha T} + e^{-2\alpha T}}{2\alpha T} \right) + \left[2(M-1) \frac{D_{2s}D_2}{1+A} + 2(M-1)^2 D_2^2 \right] \left[1 - \frac{2 - (2 + \alpha T)e^{-\alpha T}}{\alpha T} \right] \right] \end{cases} \quad (13)$$

where R_1^* is the true count rate the hypothetical count rate which one would measure if individual neutrons could not be counted multiple times, as. As opposed to the measured count rate R_1 which includes 2 counts instead of 1 for each double scatter and 3 counts instead of 1 for each triple scatter, R_1^* is the rate of individual neutrons detected by the liquid scintillator array.⁵ f_2 is the ratio of the doubles rate to the true count rate R_1^* , f_3 is the ratio of the triples rate to the true count rate R_1^* . For multiple scattering, R_1 is defined as

$$R_1 = (1 + f_2 + 2f_3) R_1^* \quad (14)$$

78 The first equation for $\bar{C}(T)$ in the system of equations (13) is the sum of three terms, all
79 factors of $R_1^* T$. The first term is the fraction of single neutrons that are counted as such,
80 the second term is $\binom{2}{1}$ times the fraction of single neutrons that are counted twice, the
81 third term is $\binom{3}{1}$ times the fraction of single neutrons that are counted thrice. As a mat-
82 ter of consistency, let us assume the case where single neutrons always double scatter
83 but never triple scatter. In this case f_2 is 1 and f_3 is 0, so that the number of counted
84 neutrons within a time gate T is $\bar{C}(T) = \binom{2}{1} R_1^* T$, which is twice the number of real sin-
85 gle neutrons. The same could be said for the case of neutrons always triple scattering.
86 While the derivations in Appendix A could easily be generalized to neutrons scattering
87 any number of times, equations (13) assume that not a single neutron registers counts
88 in more than 3 liquid scintillator cells.

89 Let's examine the assumption that neutrons cannot register counts in more than 3
90 liquid scintillator cells. For a single neutron to deposit more than 1.2 MeV — which

⁴While the derivations in Appendix A could easily be generalized to neutrons scattering any number of times, equations (13) assume that individual neutrons do not register counts in more than 3 liquid scintillator cells on physical grounds laid out below.

⁵To illustrate R_1^* , if a neutron were to multiple scatter between different liquid scintillator cells, this neutron would contribute only a single count towards the hypothetical R_1^* , in lieu of contributing multiple counts.

is approximately the liquid scintillator threshold energy for detecting fast neutrons — in 4 different liquid scintillator cells, it would theoretically only need to have an initial energy of 4.8 MeV. In reality however, a neutron would need to have a much higher energy to record counts in 4 different liquid scintillator cells with a reasonable probability. In Fig. 3, we show that the probability of a neutron registering 4 counts in 4 different liquid scintillator cells is negligible for any neutron below 10 MeV. Since the

Figure 3: Fraction of detected neutrons registering four counts in the liquid scintillator cells, as a function of the source neutron energies.

purpose here is to study fast neutrons with energies similar to the ones found in fission spectra, we can safely neglect any multiple scattering greater than 3.

Once we know f_2 and f_3 , and if we assume that A is zero, the system of equations (13) corrected for multiple scattering can be solved for the three system parameters: the strength F_s of the spontaneous fission source, the multiplication M of the object, and the efficiency ε of the detection system. Our goal is to determine whether the multiple scattering fractions f_2 and f_3 can be determined from measuring the spectrum of energies deposited by the fast neutrons from a single measurement, that is without any prior measurement. If this were the case, we could easily use these fractions to apply a correction to the system parameters.

3. Neutron energy sensitivity study

In this section, we are studying the detector response to isotropic and mono-energetic neutron sources. The goal is to determine whether spectral information measured experimentally by the detectors can be used to infer the energy distribution of the neutrons entering the detectors⁶, and in turn to infer the multiple scattering values f_2 and f_3 . In Secs. 4 and 5, we will infer the values of f_2 and f_3 from this spectral information.

For this study, we considered the liquid scintillator array shown in Fig. 1. We assumed placed a point neutron source is located in the middle of the array of liquid scintillators depicted in Fig. 1. Neutrons are emitted isotropically and one at a time from that point source. For 19 different mono-energetic neutron beams ranging from

⁶or the energy distribution of the neutron source itself if it is not shielded

1 MeV to 10 MeV in increments of 0.5 MeV, we use the Monte Carlo radiation transport code MCNPX [7, 8] to transport the neutrons from the point source in the middle of the array through the geometry consisting of the array of liquid scintillator cells. The black curve in Fig. 4 shows the energies of the neutrons entering the liquid scintillator cells, that had an initial kinetic energy of 10 MeV. These were calculated by MCNPX using a flux tally. The other colors correspond to neutrons with different initial kinetic energies. It is interesting to first take a peek at the ensemble of the energies of

Figure 4: (color online) Distributions of numbers of fast neutrons traversing Energy distributions of fast neutrons entering the liquid scintillator cells as predicted by MCNPX tallied by a MCNPX flux tally. The different colors refer to a subset of the different initial neutron energies.

the neutrons as they traverse the liquid scintillator cells shown in Fig. 4. One notices that most neutrons have their full energy as they traverse the liquid scintillator cells, One notices that most neutrons enter the liquid scintillator cells with their full initial energy, but there is also a second relatively strong peak from the neutrons which have been thermalized by the hydrogen in the liquid scintillator cells themselves.

Figure 5: (color online) Distributions of numbers of fast neutrons as a function of the energy deposited in Electron-equivalent energy distribution measured by the liquid scintillator cells as predicted by MCNPX. The different colors refer to a subset of the different initial neutron energies.

In liquid scintillators, the reaction used for neutron detection is inelastic scattering of neutrons primarily on hydrogen, producing a recoil proton from which scintillation light is emitted. A decade ago, LLNL developed a customized version [9] of MCNPX which writes out detection events to an output file in “list-mode”. Using a

quench function, each individual proton recoil energy in this file is converted to a corresponding electron-equivalent energy E_d , in terms of scintillation light output. This electron-equivalent energy is what liquid scintillators effectively measure and has units of MeVee. The measured spectrum of energies deposited by fast neutrons is shown in Fig. 5, where multiple scattering of neutrons is included. This figure shows that source neutrons with different energies produce different responses, that is different measured spectra of deposited energies in the liquid scintillator cells. This differentiation is the basis for inferring the energy spectrum of the neutrons incident on the liquid scintillator cells, from the measured spectrum in the cells.

The last set of figures 6 shows the probability density functions for the electron-equivalent energies deposited by the neutrons in the liquid scintillator cells. To produce these distributions, each distribution in Fig. 5 was normalized, and each bin of the normalized distribution was divided by the bin width. Integrated over the energy range, each cumulative density function thus produced is equal to 1.

Figure 6: (color online) Probability density functions for the energies deposited of the energy distributions measured by mono-energetic source neutrons in the liquid scintillator cells as predicted by MCNPX. The different colors refer to a subset of the different initial neutron energies.

3.1. First method to determine multiple scattering fractions

For these 19 initial neutron energies between 1 MeV and 10 MeV, we computed the fractions of single source neutrons registering counts in two and three liquid scintillator cells. The simulations were such that (a) the system does not multiply neutrons, so that both M and $q(M)M$ are equal to 1, and (b) neutrons are emitted one at a time, so that both D_{2s} and D_{3s} are zero. In these conditions, many terms disappear in the system of equations (13), and we can write the following equations that apply for non-multiplying sources emitting single neutrons at a time:

$$\begin{cases} \bar{C}(T) = (1 + f_2 + 2f_3)R_1^*T \\ Y_2(T) = (f_2 + 3f_3)R_1^*T \\ Y_3(T) = f_3R_1^*T \end{cases} \quad (15)$$

where $Y_2(T)$ and $Y_3(T)$ are equal to $Y_{2F}(T)\bar{C}(T)$ and $Y_{3F}(T)\bar{C}(T)$, respectively. This system of 3 equations has 3 unknowns. The fractions of doubles f_2 and triples f_3 as

well as the true hypothetical count rate R_1^* can be determined by taking the slopes of Eqs. (15).

Let's look at the quantities $\bar{C}(T)$, $Y_2(T)$, and $Y_3(T)$ calculated from a MCNPX Monte Carlo simulation of a weak 8 MeV-neutron source in the middle of the 77-cell liquid scintillator array (depicted in Fig. 1). Using LLNL's customized version [9] of MCNPX, neutrons are transported from spontaneous fission to the liquid scintillators, where their times of detection are recorded. Splitting the sequence of time tags into segments as explained in Sec. 2, we build count distributions $B_n(T)$. In the top left quadrant

Figure 7: (color online) Count distribution $B_n(T=512$ ns), and first three cumulant moments $\bar{C}(T)$, $Y_2(T)$ and $Y_3(T)$ as a function of time gate width T , for T ranging between 16 ns and 512 ns. The cumulant moments $\bar{C}(T)$, $Y_2(T)$ and $Y_3(T)$ are fit using Eqs. (16). The data are from a MCNPX simulation of 8 MeV neutrons in the middle of the liquid scintillator array shown in Fig. 1. Blue is Monte Carlo data, green is theoretical fit.

of Fig. 7, the blue points are $B_n(T)$ for $T=512$ ns. In the other 3 quadrants, $\bar{C}(T)$, $Y_2(T)$ and $Y_3(T)$ were calculated from this 512 ns count distribution, as well as for 5 other Monte Carlo generated count distributions for time gate durations in geometric progression between 16 and 512 ns. The top right is $\bar{C}(T)$, the bottom left and right are $Y_2(T)$ and $Y_3(T)$, respectively. Blue is Monte-Carlo data, green is theoretical fits. The green curves are analytical (see Eqs. (16) below) fits to the blue Monte Carlo data

164 points. The slopes of $\bar{C}(T)$ in the top right quadrant, of $Y_2(T)$ and $Y_3(T)$ in the bottom
 165 quadrants of Fig. 7 are the factors of T in Eqs. (15) and (16).

Given the size of the liquid scintillator cells (10-cm-diameter), and the speed of ~ 1.5 cm/ns of 1 MeV neutrons, it would take a neutron at least 7 ns to register a count in a cell, register a count in a second cell and travel 10 cm to get to a third cell. Even for 4 MeV neutrons traveling twice as fast, it would take 3.5 ns for them to register counts in 3 different cells. From this follows that the slope of $Y_3(T)$ is not quite as steep when T is close to 0. The same can be said for $Y_2(T)$. Because of this unmodeled latency — even though short — the moments $Y_2(T)$ and $Y_3(T)$ in Fig. 7 are not exactly fit by the linear functions (15), but by affine versions of them thereof with identical slopes:

$$\begin{cases} \bar{C}(T) = (1 + f_2 + 2f_3) R_1^* T \\ Y_2(T) = (f_2 + 3f_3) R_1^* (T - T_2^0) \\ Y_3(T) = f_3 R_1^* (T - T_3^0) \end{cases} \quad (16)$$

166 The values of f_2 , f_3 and R_1^* can be determined by solving the system of equations (16).
 167 Interestingly, T_2^0 and T_3^0 give orders of magnitude for the times it takes for neutrons
 168 to register 2 and 3 liquid scintillator counts, respectively. Fig. 7 reveals that 8-MeV
 169 neutrons take in average ~ 6.6 ns to scatter from one liquid scintillator cell to another
 170 one, while they take in average ~ 12 ns to scatter from one liquid scintillator cell to
 171 two other ones, consecutively. The reason why triple-scattering does not take twice
 172 as long as double-scattering can be explained. When a neutron scatters and records
 173 counts in 2 cells, the energy of the neutron between the first and second cells can be as
 174 low as 1.2 MeV (threshold for detection of fast neutrons). For a neutron to scatter and
 175 record counts in 3 cells, the energy of the neutron between the first and second cells
 176 has to be at least 2.4 MeV. Otherwise, the fast neutron — which needs to deposit at
 177 least 1.2 MeV in the second cell to be counted, and would thus be left with at the most
 178 1.2 MeV after the second cell — would not have the required 1.2 MeV of energy to
 179 record a count in a third cell. Therefore, for three counts to be recorded, the speed of
 180 the neutron between the first and the second cell has to be at least 1.4 times greater than
 181 the speed of a neutron recording only two counts. Thus, we can conclude that the time
 182 it takes for a neutron to record 3 counts in a liquid scintillator array will be less than
 183 twice the time it takes for a neutron to record 2 counts in the same liquid scintillator
 184 array. We should point out that the quantities T_2^0 and T_3^0 in Eqs. 16 are small, and that
 185 their introduction negligibly affects the slopes. Therefore, Eqs. 15 will be used from
 186 now on.

187 The fractions f_2 and f_3 of neutrons scattering multiple times and registering two
 188 and three counts in the liquid scintillator cells are shown in Fig. 8 for a range of source
 189 neutron energies. The fraction of neutrons scattering multiple times and registering
 190 four counts in the liquid scintillator cells is shown in Fig. 3. For all simulations, the
 191 threshold for detection in the liquid scintillators was set to 250 keVee. This value is
 192 not arbitrary, and corresponds to the energy above which pulse shape discrimination
 193 for liquid scintillators works well. Below 250 keVee, neutrons and photons cannot be
 194 distinguished as reliably.

195 Because the count rate is artificially inflated by the neutrons registering multiple

Figure 8: Fractions f_2 and f_3 of detected neutrons registering two and three counts in the liquid scintillator cells, as a function of the source neutron energies.

196 counts in the liquid scintillator cells, the ~~true~~hypothetical count rate differs from the
 197 measured count rate as shown in Fig. 9. This figure shows the ~~true~~hypothetical count
 198 rate R_1^* and measured count rates R_1 as a function of the source neutron energies. Note
 199 that the 3 plotted count rates are exactly matched up to 3 MeV, and are thus hidden
 200 behind each other. Furthermore, the “Measured count rate R_1 ” and “Count rate R_1
 201 determined by method 2” are indistinguishable all the way to 10 MeV.

Figure 9: (color online) Comparison between the measured count rate R_1 , the count rate R_1^* predicted by the system of equations (13) and the count rate predicted by the second method, as a function of the different initial neutron energies. Neutron detection efficiency of liquid scintillator array, as a function of neutron energy.

202 The right graph in Fig. 9 shows the absolute neutron detection efficiency of the
 203 liquid scintillator array. The detection efficiency is maximum for neutron energies
 204 around 3.5 MeV and 4 MeV, and decreases steadily as the neutron energy increases.
 205 Fast neutrons 1 MeV and lower are undetectable. This is because 1 MeV neutrons
 206 produce quenched energies of the order of 250 keVee, which is our detection threshold,
 207 as explained above.

208 The fractions f_2 and f_3 of neutrons scattering multiple times are listed in table 1
 209 for different mono-energetic neutron sources.

Table 1: Fractions f_2 and f_3 of neutrons double- and triple-scattering between liquid scintillator cells, for mono-energetic and fissile neutrons incident on the array shown in Fig. 1. The liquid scintillator energy threshold is 250 keVee.

Neutron source	f_2 [%]	f_3 [%]
1.5 MeV	0.000e+00	0.000e+00
2.0 MeV	0.000e+00	0.000e+00
2.5 MeV	0.000e+00	0.000e+00
3.0 MeV	5.008e-02	0.000e+00
3.5 MeV	1.808e-01	0.000e+00
4.0 MeV	6.746e-01	0.000e+00
4.5 MeV	1.199e+00	0.000e+00
5.0 MeV	1.678e+00	0.000e+00
5.5 MeV	2.088e+00	1.188e-03
6.0 MeV	2.408e+00	3.843e-03
6.5 MeV	2.506e+00	6.802e-03
7.0 MeV	2.695e+00	1.129e-02
7.5 MeV	3.056e+00	1.516e-02
8.0 MeV	3.034e+00	1.763e-02
8.5 MeV	2.998e+00	2.510e-02
9.0 MeV	3.153e+00	3.195e-02
9.5 MeV	3.094e+00	3.219e-02
10.0 MeV	3.086e+00	3.355e-02
²³⁸ U	2.786e-01	0.000e+00
²⁴⁰ Pu	3.875e-01	3.407e-04
²⁵² Cf	4.245e-01	9.739e-04
2.2 MeV (Fig. 12)	0.000e+00	0.000e+00
4.4 MeV (Fig. 10)	1.113e+00	0.000e+00
7.24 MeV (Fig. 11)	2.680e+00	1.307e-02

One should point out that the multiple scattering fractions f_2 and f_3 greatly depend on the geometry of the liquid scintillator array and on the energy threshold of the liquid scintillator cells. The values in table 1 are only valid for the geometry shown in Fig. 1 and for a liquid scintillator energy threshold of 250 keVee.

3.2. Second method to determine multiple scattering fractions

To make sure the analysis using the system of equations (13) was derived properly, we derived the numbers of multiple scatterings not from a statistical perspective, but from an alternative second method. This can be done simply using the procedure described here: each fast neutron count registered by the liquid scintillator cells triggers a time window of length duration T and one counts how many fast neutrons are within that window. If there is one fast neutron in the window, then this count along with the one that triggered the window will be consolidated into a single double scattering

count. If there were two fast neutrons in the window, then these 2 counts along with the trigger count will be consolidated into a single triple scattering count. The empty windows are considered single scattering events, because they only contain the trigger event. The fractions of doubles and triples are quasi-identical to the ones shown in Fig. 8, the count rates do not differ much either, as shown by the “method 2” line in Fig. 9. These two ways of producing the same data gives us confidence that the models used to predict the unknowns R_1^* , f_2 and f_3 are correct.

The advantage of the first method laid out in Sec. 3.1 is that experimentally on the experimental side. When measuring the double and triple-scattering fractions of neutrons experimentally, the method ~~it~~ does not require a neutron source emitting single neutrons (such as AmBe or AmLi) ~~any~~. Any non-multiplying ($M=1$) spontaneous fission source (such as ^{252}Cf) of known source intensity $\bar{v}_{\text{sp}}F_s$ can be used to compute measure the multiple scattering fractions using the system of equations derived from Eqs. (13):

$$\begin{cases} R_1 = (1 + f_2 + 2f_3) \epsilon \bar{v}_{\text{sp}} F_s \\ R_{2F} = \frac{f_2 + 3f_3}{1 + f_2 + 2f_3} + (1 + f_2 + 2f_3) \epsilon D_{2s} \\ R_{3F} = \frac{f_3}{1 + f_2 + 2f_3} + 2(f_2 + 3f_3) \epsilon D_{2s} + (1 + f_2 + 2f_3)^2 \epsilon^2 D_{3s} \end{cases} \quad (17)$$

as long as it the source is not contaminated by (α, n) neutrons⁷. Using a time-tagged ^{252}Cf source for instance, we can experimentally measure R_1 , R_{2F} and R_{3F} , and solve the system of equations (17) for ϵ , f_2 and f_3 . This is an important advantage over method 2, which intrinsically does not account for the fact that spontaneous fissions emit multiple neutrons simultaneously.

If the intensity of the non-multiplying spontaneous fission source is not known, we can use the equation for R_{4F} as well:

$$R_{4F} = \left(\frac{(f_2 + 3f_3)^2}{1 + f_2 + 2f_3} + 2f_3 \right) \epsilon D_{2s} + 3(1 + f_2 + 2f_3)(f_2 + 3f_3) \epsilon^2 D_{3s} + (1 + f_2 + 2f_3)^3 \epsilon^3 D_{4s} \quad (18)$$

4. Input spectrum reconstruction

Before trying to determine the factors f_2 and f_3 , we will see if we can reconstruct the spectra of 3 different mono-energetic neutron sources, based on the set of measured spectra of energies deposited energies shown in Fig. 5. We use the subscript E_i to distinguish each one of the basis functions $g_{E_i}(E_d)$ shown in Fig. 5. The subscript E_i denotes the different initial source neutron energies energy, from 1 MeV ($i=1$) up to 10 MeV ($i=19$), in increments of 0.5 MeV. Given a measured spectrum of deposited energies $\bar{g}(E_d)$, the goal for the reconstruction is to find the set of weights w_{E_i} for

⁷This is a permissible approximation for a pure, non-sealed and unshielded source.

243 which the difference between $\tilde{g}(E_d)$ and the reconstructed liquid scintillator energy
 244 deposition spectrum is minimized. The weights w_{E_i} are physically to be interpreted as
 245 the strengths of the neutron source of energy E_i . Each weight w_{E_i} is physically to be
 246 interpreted as the intensity of the neutron source in the energy bin around E_i .⁸

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

$$g^r(E_d) = \sum_{i=1}^{19} w_{E_i} g_{E_i}(E_d) \quad (19)$$

The optimal set of weights w_{E_i} will be such as to minimize

$$g^r(E_d) - \tilde{g}(E_d) \quad (20)$$

In order to minimize Eq. (20), we use χ -square minimization algorithm implemented by Minuit in ROOT [10]. Based on this optimization, an estimate of the source neutron spectrum will be given by the energies E_i weighed by the weights w_{E_i} . For mono-energetic neutron sources, the estimated average source neutron energy E^{est} will be given by the w_{E_i} weighed average of the E_i .

$$E^{\text{est}} = \sum_{i=1}^{19} w_{E_i} E_i \quad (21)$$

Based on the solution set for the weights w_{E_i} , we can produce estimate the spectrum of electron-equivalent energies deposited by the neutrons in the liquid scintillator using

$$g^{\text{est}}(E_d) = \sum_{i=1}^{19} w_{E_i} g_{E_i}(E_d) \quad (22)$$

247 4.1. Reconstruction of mono-energetic neutron sources

248 We ran a few Monte Carlo simulations similar to the ones in Sec. 3, but with
 249 energies not among the E_i for which we have basis functions. The first energy of
 250 mono-energetic neutrons considered was 4.4 MeV. The first simulation was a mono-
 251 energetic 4.4 MeV neutron source. Fig. 10 shows the reconstructed source neutron
 252 energy spectrum and the spectrum of energies deposited by the neutron constructed
 253 from the reconstructed source neutron energy spectrum using Eq. (22). The blue his-
 254 togram in Fig. 10a shows the spectrum measured by the modeled liquid scintillators
 255 cells, along with the best reconstruction $g^{\text{est}}(E_d)$ in red. The reconstruction $g^{\text{est}}(E_d)$ is
 256 not perfect but nonetheless a very good approximation of $\tilde{g}(E_d)$. Fig. 10b shows the re-
 257 construction of the neutron source spectrum, which is basically the set of weights w_{E_i} .
 258 Because we know the neutron source was mono-energetic, we used Eq. (21) to compute

⁸If the distribution $\tilde{g}(E_d)$ is normalized, the weights w_{E_i} will be the relative strengths intensities of the different neutron sources of energy E_i .

Figure 10: (color online) (a) Measured spectrum $\tilde{g}(E_d)$ (blue) and reconstructed spectrum $g^{\text{est}}(E_d)$ (red) of energies deposited by the neutrons, evaluated from Eq. (22). (b) Reconstruction of the source neutron energy spectrum using Eq. (20). The estimated average source neutron energy is 4.406 MeV. The data is from a MCNPX simulation of a mono-energetic 4.4 MeV neutron source.

the energy of the neutron source, the reconstructed energy was estimated at 4.406 MeV. Using Eq. (21), the reconstructed average neutron source energy was estimated to be 4.406 MeV.

The second and third simulations are mono-energetic 7.24 MeV and 2.2 MeV neutron source in the middle of the same liquid scintillator array. The reconstructed source neutron energy spectra and the spectra of energies deposited by the neutron constructed from the reconstructed source neutron energy spectra are shown in Fig. 11 and 12. The

Figure 11: (color online) (a) Measured spectrum $\tilde{g}(E_d)$ (blue) and reconstructed spectrum $g^{\text{est}}(E_d)$ (red) of energies deposited by the neutrons, evaluated from Eq. (22). (b) Reconstruction of the source neutron energy spectrum using Eq. (20). The estimated average source neutron energy is 7.51 MeV. The data is from a MCNPX simulation of a mono-energetic 7.24 MeV neutron source.

blue histograms in Figs. 11a and 12a show the spectra $\tilde{g}(E_d)$ measured by the liquid scintillators, along with the best reconstructions $g^{\text{est}}(E_d)$ in red. Figs. 11b and 12b show the reconstructions of the neutron source spectra. The average energies determined from the reconstructions were 7.51 MeV and 2.22 MeV. From these few data points, we observe that the predictions of the source neutron energies are good, but not perfect. Fig. 11b indeed shows that there is a somewhat unexpected secondary peak at 8.5 MeV, while we would have expected a secondary peak at 7.5 MeV.

Figure 12: (color online) (a) Measured spectrum $\tilde{g}(E_d)$ (blue) and reconstructed spectrum $g^{\text{est}}(E_d)$ (red) of energies deposited by the neutrons, evaluated from Eq. (22). (b) Reconstruction of the source neutron energy spectrum using Eq. (20). The estimated average source neutron energy is 2.22 MeV. The data is from a MCNPX simulation of a mono-energetic 2.2 MeV neutron source.

4.2. Reconstruction of spontaneous fission sources.

The next test is to see whether the reconstruction algorithm could predict the neutron spectrum emitted by spontaneous fission sources. Of course, the caveat here is one should not expect to predict the source neutron spectrum under approximately 1.25 MeV, since liquid scintillators cannot reliably distinguish neutrons from photons detect neutrons below that energy.

We performed a simulation of a ^{252}Cf source in the middle of the liquid scintillator array shown in Fig. 4. Fig. 13a shows the measured spectrum $\tilde{g}(E_d)$ in blue and the reconstructed spectrum $g^{\text{est}}(E_d)$ in red. The reconstruction of the source neutron spectrum is shown in Fig. 13b. As expected, the source neutron spectrum reconstruction fails below 1.5 MeV for the reasons mentioned above. The estimated average source neutron energy is 3.05 MeV, but this value cannot be compared to the true average energy of ^{252}Cf spontaneous fission neutrons, because it is computed over the truncated energy distribution shown in Fig. 13b.

Figure 13: (color online) (a) Measured spectrum $\tilde{g}(E_d)$ (blue) and reconstructed spectrum $g^{\text{est}}(E_d)$ (red) of electron-equivalent energies deposited by the neutrons, evaluated from Eq. (22). (b) Reconstruction of the source neutron energy spectrum using from Eq. (20) (red), along with the measured true ^{252}Cf spectrum (blue) from Ref. [11]. The data is from a MCNPX simulation of a ^{252}Cf source.

5. Prediction of multiple scattering correction

To predict multiple scattering corrections, we first measure the spectrum of fast neutrons in the liquid scintillator cells. Then the basis functions given in Fig. 4 are used to reconstruct the measured spectrum by minimizing Eq. 20. This minimization process will produce the weights w_{E_i} in $g^r(E_d)$ (see Eq. 19). Knowing these weights, Using equations similar to Eq. (24), we can estimate the fraction of double scattering f_2 and triple scattering f_3 by weighing the fractions $f_2(E_i)$ and $f_3(E_i)$ at different source energies E_i by w_{E_i} (given by Eq. (19) when Eq. (20) is minimized):

$$f_2 = \sum_{i=1}^{19} w_{E_i} f_2(E_i) \quad (23)$$

$$f_3 = \sum_{i=1}^{19} w_{E_i} f_3(E_i) \quad (24)$$

288 The functions $f_2(E_i)$ and $f_3(E_i)$ are the curves shown in Fig. 8. Values for f_2 and f_3
 289 are also given in Table 1. For the 3 mono-energetic neutron sources of section 4.1, and
 290 the ^{252}Cf source of section 4.2, the estimated fractions f_2 and f_3 are given in table 1.

291 6. Application to NMC: Mass correction using multiple scattering correction

292 A Monte Carlo simulation was run to see if the strength intensity of a ^{252}Cf source
 293 could be more accurately determined using the multiple scattering correction factors
 294 f_2 and f_3 . For this simulation, we used the same geometry as for the previous cases.
 295 The input source strength intensity was 39,761 neutrons/s. The count distribution $b_{\bar{n}}$
 296 B_n , along with $\bar{C}$, Y_{2F} and Y_{3F} the first 3 moments are shown in Fig. 14.

Figure 14: (color online) Count distribution $b_{\bar{n}}$ $B_n(T = 20 \mu s)$ and first three moments $\bar{C}(T)$, $Y_2(T)$ and $Y_3(T)$ as a function of time gate width T . λ^{-1} denotes α^{-1} . $\bar{C}(T)$, $Y_2(T)$ and $Y_3(T)$ are fit using Eqs. (1), (5) and (7), with A set to 0. The data are from an MCNPX simulation of a ^{252}Cf source in the middle of the liquid scintillator array shown in Fig. 1.

If we assume a multiplication⁹ M of 1 and an α -ratio A of 0, the first 2 moments

⁹According to the MCNPX output, the multiplication is about 1.0002

equations of the system of equations (12) reduce to

$$\begin{cases} R_1 = \varepsilon \bar{v}_{\text{sp}} F_s \\ R_{2F} = \varepsilon D_{2s} \end{cases} \quad (25)$$

which has two unknowns F_s and ε . This system of two equations contains enough information to solve for the efficiency ε and the source strength $\bar{v}_{\text{sp}} F_s$. We can thus solve for the mass of ^{252}Cf using this simulated data set. Knowing that D_{2s} is equal to 1.595975 for ^{252}Cf , the detection efficiency ε is equal to 7.6% with a relative error of 0.2% and the ^{252}Cf source strength is $38,524 \pm 0.2\%$ neutrons/s. The source strength is thus underestimated by 3.2% with the equations with no multiple scattering correction.

If we account for multiple scattering, we will use the following system of equations instead of Eqs. (25):

$$\begin{cases} R_1 = (1 + f_2 + 2f_3) \varepsilon \bar{v}_{\text{sp}} F_s \\ R_{2F} = \frac{f_2 + 3f_3}{1 + f_2 + 2f_3} + (1 + f_2 + 2f_3) \varepsilon D_{2s} \end{cases} \quad (26)$$

How do we determine the correction factors f_2 and f_3 ? Simply by measuring the neutron spectrum in the liquid scintillators. This measured spectrum is shown in blue in Fig. 13a. In red, the reconstruction $g^{\text{est}}(E_d)$ generates the weights w_{E_i} that we need to calculate f_2 and f_3 (using Eqs. 23 and 24). The corrections f_2 and f_3 for ^{252}Cf were already calculated in Sec. 5 and are in Table 1. Using the multiple scattering corrections given in table 1, Using these corrections, the source strength now becomes 39,925 neutrons/s and the efficiency 7.3% with a relative error of 0.2%. With the multiple scattering correction, the source strength is within 2 standard deviations from the true value, which is a significant improvement over the estimate not using the multiple scattering correction.

The same checks were done We verified that Eqs. 26 also hold for mono-energetic random (i.e. uncorrelated) neutron sources emitting single neutrons at a time. In this case, we showed that the measured value of R_{2F} was equal to the first term in the equation for R_{2F} in the system of equations (26) $\frac{f_2 + 3f_3}{1 + f_2 + 2f_3}$, which is consistent with an expected D_{2s} of 0., which is the correct uncorrelated value. Of course, it is impossible to determine the detection efficiency ε , nor the mass of such sources in this case.

We should emphasize that the authors did not expect perfect reconstruction of the source strength. The reasons are multiple: (a) we have seen in Sec. 4.2 that the method does not reconstruct neutron source spectra perfectly, which leads to an error in the estimation of the multiple scattering fractions. In that respect, using a larger number of mono-energetic neutron beams with intermediate energies might help. (b) The point model theory due to Feynman [2] on which this work is based, makes several assumptions. One of them is that the neutron detection efficiency ε does not depend on the neutron energy. Whereas this is a reasonable assumption for ^3He tubes embedded in polyethylene, it is less so for liquid scintillators detecting fast neutrons, as shown in Fig. 9. Multiple scattering corrections are one of the corrections that is required to improve the reconstruction of the source mass, but it is only one of them.

331 7. Conclusion

332 With the help of theoretical expressions for the [Feynman correlated](#) moments of
 333 count distributions that account for multiple scatterings of neutrons, we showed that it
 334 is possible to determine the fractions of neutrons scattering twice and thrice between
 335 liquid scintillator cells from the first 3 [Feynman correlated](#) moments of measured count
 336 distributions. These multiple scattering fractions can be determined for any neutron
 337 source, whether mono-energetic or not, and they strongly depend on the energy of the
 338 source neutrons.

339 For nuclear materials undergoing either spontaneous or induced fission, the same
 340 theoretical expressions for the [Feynman correlated](#) moments can be used in conjunction
 341 with the measured deposited energy spectrum to apply corrections to the estimates
 342 of the detection efficiency, the system multiplication, and the masses of the sources
 343 under measurement. A simple simulation using a Californium source showed that the
 344 determination of the ^{252}Cf mass was improved using the multiple scattering correction.

345 Also, we were able to show that measuring the spectrum of fast neutrons depositing
 346 energy in the liquid scintillator cells reveals spectral information about the neutron
 347 source.

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351 Appendix A. Derivation of the [Feynman correlated](#) moment equations for neu- 352 trons scattering multiple times

353 In this appendix, we will derive the expressions for $\bar{C}(T)$, $Y_{2F}(T)$, $Y_{3F}(T)$ two
 354 different ways: using generating functions based on the work of Böhnelt [12], and a
 355 second method in the line of Hage-Cifarelli [13].

356 Appendix A.1. Generating function

Let's consider the generating function for the $e_n(\epsilon)$ distribution, which gives the
 probability of detecting n neutrons from a single fission chain. In the case when neu-
 trons never multiple scatter between liquid scintillator cells, the generating function for
 the $e_n(\epsilon)$ distribution can be written as

$$h(d(y)) = \sum_{v=0}^{\infty} e_n(\epsilon) y^n \quad (\text{A.1})$$

$$d(y) = (1 - \epsilon) + \epsilon y \quad (\text{A.2})$$

357 where $h(y)$ is the single neutron induced fission chain probability generating function
 358 defined by Eq. 12b of Böhnelt [12] and Eq. 34 of Prasad-Snyderman [4]. The generat-
 359 ing function parameter y in $h(y)$ must be replaced by detector probability generating
 360 function $d(y)$ through the compound process to account for detector efficiency ϵ .

When neutrons are allowed to multiple scatter between liquid scintillator cells, the generating function parameter y in $h(y)$ must be replaced, through the compound process, by the following detector probability generating function $f(y)$:

$$f(y) = (1 - \epsilon) + \epsilon(1 - f_2 - f_3)y + \epsilon f_2 y^2 + \epsilon f_3 y^3 \quad (\text{A.3})$$

where the first term $(1 - \epsilon)$ — which could also be written as $(1 - \epsilon)y^0$ — is the probability that a neutron is not detected; the second term (or more precisely the coefficient of y) $\epsilon(1 - f_2 - f_3)$ is the probability that a neutron is detected by a single liquid scintillator cell; the third polynomial coefficient ϵf_2 is the probability that a neutron is detected by a liquid scintillator cell, scatters and is detected once and only once more by another liquid scintillator cell; and finally the last polynomial coefficient ϵf_3 is the probability that a neutron is detected thrice by 3 different liquid scintillator cells. The powers in y count the number of times a single neutron is detected by the array: 0, 1, 2 or 3 times.

The single neutron induced fission chain probability generating function which accounts for neutrons to multiple scatter between liquid scintillator cells becomes:

$$h(f(y)) = \sum_{v=0}^{\infty} e_n(\epsilon) y^v \quad (\text{A.4})$$

Taking the first derivative of $h(f(y))$ with respect to y , and setting y to 1, we get

$$\begin{aligned} \left. \frac{\partial h}{\partial y} \right|_{y=1} &= h'|_{y=1} f' \\ &= \epsilon h'|_{y=1} ((1 - f_2 - f_3) + 2f_2 + 3f_3) \\ &= \epsilon h'|_{y=1} (1 + f_2 + 2f_3) \\ &= R_1^* (1 + f_2 + 2f_3) \end{aligned} \quad (\text{A.5})$$

The second derivative leads to

$$\begin{aligned} \left. \frac{1}{2!} \frac{\partial^2 h}{\partial y^2} \right|_{y=1} &= \frac{1}{2!} [h''|_{y=1} f'^2 + h'|_{y=1} f''] \\ &= \frac{1}{2!} h''|_{y=1} [\epsilon(1 - f_2 - f_3) + 2\epsilon f_2 + 3\epsilon f_3]^2 + h'|_{y=1} \epsilon(f_2 + 3f_3) \\ &= R_2^* (1 + f_2 + 2f_3)^2 + R_1^* (f_2 + 3f_3) \end{aligned} \quad (\text{A.6})$$

Similarly, the third derivative is written as

$$\begin{aligned} \left. \frac{1}{3!} \frac{\partial^3 h}{\partial y^3} \right|_{y=1} &= \frac{1}{3!} [h'''|_{y=1} f'^3 + 3h''|_{y=1} f' f'' + h'|_{y=1} f'''] \\ &= \frac{1}{3!} h'''|_{y=1} \epsilon^3 (1 + f_2 + 2f_3)^3 + h''|_{y=1} \epsilon^2 (1 + f_2 + 2f_3)(f_2 + 3f_3) + h'|_{y=1} \epsilon f_3 \\ &= (1 + f_2 + 2f_3)^3 R_3^* + 2(1 + f_2 + 2f_3)(f_2 + 3f_3) R_2^* + f_3 R_1^* \end{aligned} \quad (\text{A.7})$$

If needed, the fourth derivatives reads

$$\begin{aligned}
\frac{1}{4!} \frac{\partial^4 h}{\partial y^4} \Big|_{y=1} &= \frac{1}{4!} \left[h''''|_{y=1} f'^4 + 6 h'''|_{y=1} f'^2 f'' + 3 h''|_{y=1} f''^2 + 4 h''|_{y=1} f' f''' + h'|_{y=1} f'''' \right] \\
&= \frac{1}{4!} \left[h''''|_{y=1} \epsilon^4 (1 + f_2 + 2f_3)^4 + 6 h'''|_{y=1} \epsilon^3 (1 + f_2 + 2f_3)^2 (2f_2 + 6f_3) \right. \\
&\quad \left. + 3 h''|_{y=1} \epsilon^2 (2f_2 + 6f_3)^2 + 24 h''|_{y=1} \epsilon^2 (1 + f_2 + 2f_3) f_3 \right] \\
&= (1 + f_2 + 2f_3)^4 R_4^* + 3 (1 + f_2 + 2f_3)^2 (f_2 + 3f_3) R_3^* \\
&\quad + (f_2 + 3f_3)^2 R_2^* + 2 (1 + f_2 + 2f_3) f_3 R_2^*
\end{aligned} \tag{A.8}$$

370 If needed, higher-order derivatives can be calculated trivially.

Dividing Eqs. (A.6), (A.7) and (A.8) by Eq. (A.5), we get the following equations for R_{2F} , R_{3F} and R_{4F} :

$$R_{2F} = \frac{f_2 + 3f_3}{1 + f_2 + 2f_3} + (1 + f_2 + 2f_3) R_{2F}^* \tag{A.9}$$

$$R_{3F} = \frac{f_3}{1 + f_2 + 2f_3} + 2(f_2 + 3f_3) R_{2F}^* + (1 + f_2 + 2f_3)^2 R_{3F}^* \tag{A.10}$$

$$R_{4F} = \left(\frac{(f_2 + 3f_3)^2}{1 + f_2 + 2f_3} + 2f_3 \right) R_{2F}^* + 3(1 + f_2 + 2f_3)(f_2 + 3f_3) R_{3F}^* + (1 + f_2 + 2f_3)^3 R_{4F}^* \tag{A.11}$$

371 Appendix A.2. Combinatorial expansion

An alternative way to get to the same result is by starting with $\Lambda_n(T)$, the probability to count n neutrons from the same fission chain in a random time gate of duration T . One can derive the expressions for $\bar{C}(T)$ starting from the equations¹⁰ for the $\Lambda_n(T)$ including single neutrons registering multiple counts in different liquid scintillator cells

¹⁰See Eqs.(3), (54), (55) and Appendix B of Ref. [13], or Eq. A.20 in Ref. [14].

in the limit of large time gate width T :

$$\begin{aligned}
\lim_{T \gg \alpha^{-1}} \frac{\Lambda_n(T)}{F_s T} &= \sum_{v=n}^{\infty} P_v \binom{v}{n} \varepsilon^n (1-\varepsilon)^{v-n} f_1^n \\
&+ \sum_{v=n-1}^{\infty} P_v \binom{v}{n-1} \varepsilon^{n-1} (1-\varepsilon)^{v-n+1} \binom{n-1}{1} f_1^{n-2} f_2 \\
&+ \sum_{v=n-2}^{\infty} P_v \binom{v}{n-2} \varepsilon^{n-2} (1-\varepsilon)^{v-n+2} \binom{n-2}{2} f_1^{n-4} f_2^2 \\
&+ \dots \\
&+ \sum_{v=n-2}^{\infty} P_v \binom{v}{n-2} \varepsilon^{n-2} (1-\varepsilon)^{v-n+2} \binom{n-2}{1} f_1^{n-3} f_3 \\
&+ \sum_{v=n-4}^{\infty} P_v \binom{v}{n-4} \varepsilon^{n-4} (1-\varepsilon)^{v-n+4} \binom{n-4}{2} f_1^{n-6} f_3^2 \\
&+ \dots \\
&+ \sum_{v=n-3}^{\infty} P_v \binom{v}{n-3} \varepsilon^{n-3} (1-\varepsilon)^{v-n+3} \frac{(n-3)!}{1!1!(n-5)!} f_1^{n-5} f_2 f_3 \\
&+ \sum_{v=n-4}^{\infty} P_v \binom{v}{n-4} \varepsilon^{n-4} (1-\varepsilon)^{v-n+4} \frac{(n-4)!}{2!1!(n-7)!} f_1^{n-7} f_2^2 f_3 \\
&+ \dots \\
&+ \sum_{v=n-5}^{\infty} P_v \binom{v}{n-5} \varepsilon^{n-5} (1-\varepsilon)^{v-n+5} \frac{(n-5)!}{1!2!(n-8)!} f_1^{n-8} f_2 f_3^2 \\
&+ \sum_{v=n-6}^{\infty} P_v \binom{v}{n-6} \varepsilon^{n-6} (1-\varepsilon)^{v-n+6} \frac{(n-6)!}{2!2!(n-10)!} f_1^{n-10} f_2^2 f_3^2 \\
&+ \dots
\end{aligned} \tag{A.12}$$

where $\Lambda_n(T)$ is the probability of measuring n neutrons from a fission chain originated by a single spontaneous fission in a time gate of width T . where f_2 and f_3 are the probabilities that a neutron detected in one liquid scintillator cell scatters and registers one or two more counts in other liquid scintillator cells, respectively. f_1 is the probability that a neutron detected in one liquid scintillator cell does not register more counts in other liquid scintillator cells, so that the f distribution is normalized, $f_1 = 1 - f_2 - f_3$. If we were to account for higher order scattering events, we would have $f_1 = 1 - \sum_{i=2}^{\infty} f_i$.

Here is how to interpret Eq. (A.12): the first term in the series is a summation of probabilities to detect n neutrons from a single fission chain producing v neutrons where none of the v neutrons registered multiple counts. Each one of the probabilities in the sum is thus multiplied by f_1^n , the probability that none of the n neutrons detected registered multiple counts. The second term in the series is a summation of probabilities to detect n neutrons from a single fission chain producing v neutrons, where $n-2$

of the v neutrons produced by the chain did not register multiple counts in liquid scintillator cells, and one neutron from this fission chain registered two counts in different liquid scintillator cells. Each one of the probabilities in the sum is thus multiplied by f_1^{n-2} and f_2 : the probability that $n-2$ neutrons registered single detected events and the probability that one neutron was detected in two different cells. Because the neutron registering two counts can be any of the $n-1$ neutrons, we need to multiply these probabilities by the number of ways one can choose 1 neutron among $n-1$ neutrons, i.e. $\binom{n-1}{1}$.

The first term in the second group of terms in Eq. (A.12) is a summation of probabilities to detect n neutrons from a single fission chain producing v neutrons, where $n-3$ of the v neutrons produced by the chain did not register multiple counts in liquid scintillator cells, and one neutron from this fission chain registered three counts in different liquid scintillator cells. Each one of the probabilities in the sum is thus multiplied by f_1^{n-3} and f_3 : the probability that $n-3$ neutrons registered single detected events and the probability that one neutron was detected in three different cells. Because the neutron registering three counts can be any of the $n-2$ neutrons, we need to multiply these probabilities by the number of ways one can choose 1 neutron among $n-2$ neutrons, i.e. $\binom{n-2}{1}$.

If one denotes by $\Lambda_n^*(T)$ the expressions $\Lambda_n(T)$ when neutrons are not counted multiple times (that is with $f_{i>1} = 0$), we get

$$\lim_{T \gg \alpha^{-1}} \Lambda_n^*(T) = F_s T \sum_{v=n}^{\infty} P_v \binom{v}{n} \epsilon^n (1 - \epsilon)^{v-n} \quad (\text{A.13})$$

One notices that each one of the terms in the series in Eq. (A.12) is a different order of $\Lambda_n^*(T)$ multiplied by a multinomial coefficient of the form

$$\frac{(\sum_{i=1}^{\infty} j_i)!}{\prod_{i=1}^{\infty} j_i!} \prod_{i=1}^{\infty} f_i^{j_i} \quad (\text{A.14})$$

The multinomial coefficients are counting the number of ways to distribute the different populations of neutrons (those registering one count, those registering two counts, etc.).

When there are only two kinds of multiple counts (e.g. f_1 and f_2), the multinomial reduces to a binomial coefficient

$$\binom{i+j}{i} f_1^i f_2^j \quad (\text{A.15})$$

Using the $\Lambda_n^*(T)$ notation, the first term of Eq. (A.12) becomes $f_1^n \Lambda_n^*$, the second term $\binom{n-1}{1} f_1^{n-2} f_2 \Lambda_{n-1}^*$, the third one $\binom{n-2}{2} f_1^{n-4} f_2^2 \Lambda_{n-2}^*$, etc. Eq. (A.12) can thus be rewritten more compactly as

$$\lim_{T \gg \alpha^{-1}} \Lambda_n(T) = \sum_{j=0}^{j \leq n/3} \sum_{i \leq (n-3*j)/2} \frac{(n-2j-i)!}{i! j! (n-3j-2i)!} f_1^{n-3j-2i} f_2^i f_3^j \Lambda_{n-2j-i}^* \quad (\text{A.16})$$

The first few $\Lambda_n(T)$ are thus

$$\begin{aligned}
\lim_{T \gg \alpha^{-1}} \Lambda_1(T) &= \Lambda_1^* f_1 \\
\lim_{T \gg \alpha^{-1}} \Lambda_2(T) &= \Lambda_2^* f_1^2 + \Lambda_1^* f_2 \\
\lim_{T \gg \alpha^{-1}} \Lambda_3(T) &= \Lambda_3^* f_1^3 + \Lambda_2^* \binom{2}{1} f_1 f_2 + \Lambda_1^* f_3 \\
\lim_{T \gg \alpha^{-1}} \Lambda_4(T) &= \Lambda_4^* f_1^4 + \Lambda_3^* \binom{3}{1} f_1^2 f_2 + \Lambda_2^* f_2^2 + \Lambda_2^* \binom{2}{1} f_1 f_3
\end{aligned} \tag{A.17}$$

Using these expressions for $\Lambda_n(T)$, the measured count rate $\bar{C}(T)$ can be written as¹¹

$$\begin{aligned}
\bar{C}(T) &= \sum_{i=1}^{\infty} \binom{i}{1} \Lambda_i \\
&= (1 + f_2 + 2f_3) \sum_{i=1}^{\infty} \binom{i}{1} \Lambda_i^* \\
&= (1 + f_2 + 2f_3) \bar{C}^*(T)
\end{aligned} \tag{A.18}$$

405 which means that the true count rate ~~hypothetical~~ $\bar{C}^*(T)$ ¹² is increased by the multiplier factor
406 $1 + f_2 + 2f_3$. In the limiting case where f_2 is 1, that is, all neutrons register two counts
407 in the liquid scintillators, the measured count rate is twice the ~~true~~ $\bar{C}^*(T)$ count
408 rate. In the limiting case where f_3 is 1, where all neutrons register three counts in the
409 liquid scintillators, the measured count rate is thrice the ~~true~~ $\bar{C}^*(T)$ count rate.
410 These two limiting cases are consistent with our expectations.

Similarly, the moment $Y_2(T)$ can be written as [4]

$$\begin{aligned}
Y_2(T) &= \sum_{i=2}^{\infty} \binom{i}{2} \Lambda_i \\
&= (f_2 + 3f_3) \bar{C}^*(T) + (1 + f_2 + 2f_3)^2 Y_2^*(T)
\end{aligned} \tag{A.19}$$

and $Y_3(T)$ as

$$\begin{aligned}
Y_3(T) &= \sum_{i=3}^{\infty} \binom{i}{3} \Lambda_i \\
&= f_3 \bar{C}^*(T) + 2(1 + f_2 + 2f_3)(f_2 + 3f_3) Y_2^*(T) + (1 + f_2 + 2f_3)^3 Y_3^*(T)
\end{aligned} \tag{A.20}$$

Dividing both expressions by $\bar{C}(T)$, we get

$$Y_{2F}(T) = \frac{f_2 + 3f_3}{1 + f_2 + 2f_3} + (1 + f_2 + 2f_3) Y_{2F}^*(T) \tag{A.21}$$

¹¹Equation (115) for Y_1 in Ref. [4]

¹²hypothetical number of counts which one would measure if individual neutrons could not be counted multiple times

and

$$Y_{3F}(T) = \frac{f_3}{1+f_2+2f_3} + 2(f_2+3f_3)Y_{2F}^*(T) + (1+f_2+2f_3)^2 Y_{3F}^*(T) \quad (\text{A.22})$$

411 Nomenclature

412

T	=	time gate duration.
$B_n(T)$	=	number of times n neutrons were counted a random time gate of duration T .
$b_n(T)$	=	probability to get n counts in a random time gate of duration T .
p	=	probability that a neutron will induce fission in a nucleus on interaction.
q	=	probability that a neutron will not induce fission in a nucleus on interaction.
M	=	multiplication of the object.
qM	=	escape multiplication.
F_s	=	intensity of spontaneous fission source in units of spontaneous fissions per second.
A	=	α -ratio, the ratio of neutrons emitted by sources emitting single neutrons to neutrons emitted by sources emitting multiple neutrons simultaneously.
C_n & C	=	probability that a fission will emit n neutrons in induced fission & induced fission multiplicity distribution.
C_n^{sp} & C^{sp}	=	probability that a fission will emit n neutrons in spontaneous fission & spontaneous fission multiplicity distribution.
D_n	=	n^{th} combinatorial moment of induced fission multiplicity distribution.
$D_{n \text{ sp}}$	=	n^{th} combinatorial moment of spontaneous fission multiplicity distribution.
P_v	=	probability that the fission chain created n neutrons (excluding those internally absorbed to create subsequent fissions).
e_n	=	probability of detecting n neutrons from a single fission chain.
$\bar{C}(T)$	=	number of counts averaged over all time gates of duration T .
$Y_{2F}(T)$	=	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.
$Y_{3F}(T)$	=	the skewness to mean ratio of $b_n(T)$, or physically speaking the correlated triples relative to the counts.
$Y_2(T)$	=	$Y_{2F}(T)$ multiplied by $\bar{C}(T)$.
$Y_3(T)$	=	$Y_{3F}(T)$ multiplied by $\bar{C}(T)$.
R_1	=	count rate measured by the detector.
R_{2F}	=	asymptote of $Y_{2F}(T)$.
R_{3F}	=	asymptote of $Y_{3F}(T)$.
f_1	=	probability of counting individual neutrons once.
f_2	=	probability of counting individual neutrons twice.
f_3	=	probability of counting individual neutrons thrice.

413

	$h(y)$	=	generating function for the e_n distribution.
	R_1^*	=	the hypothetical count rate which one would measure if individual neutrons could not be counted multiple times.
	$R_n^*(T)$	=	the hypothetical value of $R_n = \frac{1}{n!} \frac{\partial^n h(y)}{\partial y^n}$ which one would obtain if individual neutrons could not be counted multiple times.
414	E_d	=	electron-equivalent energy measured by liquid scintillators and deposited by fast neutrons.
	$\tilde{g}(E_d)$	=	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 spectrum reconstruction.
	$g^r(E_d)$	=	reconstructed liquid scintillator spectrum using basis functions $g_{E_i}(E_d)$.
415	Greek:		
	ε	=	detection efficiency, or probability to detect a neutron.
	$\bar{\nu}$	=	average number of neutrons produced in induced fission.
	$\bar{\nu}_{\text{sp}}$	=	average number of neutrons produced in spontaneous fission.
	α	=	inverse fission chain evolution time scale.
416	$\Lambda_n()$	=	probability to count n neutrons from the same fission chain in a random time gate of duration T .
	$\Lambda_n^*()$	=	hypothetical probability to count n neutrons from the same fission chain in a random time gate of duration T , when individual neutrons are not counted multiple times.

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