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**Generalised Campbell formulae for compound  
Poisson processes with applications in nuclear  
safeguards**

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## Abstract

Multiplicity counting is a widely used non-destructive assay method for estimating unknown parameters (primarily the mass) of samples of spontaneously fissioning materials (e.g. plutonium). Traditionally, measurements are performed with thermal neutron detectors operating in pulse counting mode. The method is based on determining the first three lowest order moments of the number of particles emitted simultaneously from the sample, through measuring the so-called singles, doubles and triples detection rates from the counting statistics of the detectors, from which the sought sample parameters can be unfolded with algebraic inversion. The main difficulty with multiplicity counting is its inherent sensitivity to dead time effects, which poses a major constraint on the ability to extract correlated neutron counts.

To overcome this difficulty, a new method of multiplicity counting has been developed, which is based on the statistics of the time-resolved signals of detectors operating in current mode. Specifically, the method utilizes information in the auto and cross cumulants of the stationary signals of different groups of detectors. Based on a stochastic theory of fission chamber signals, expressions were derived for the one-, two- and three-point (in time) cumulants of the detector currents. It was shown how the traditional multiplicity count rates can be recovered from the detector currents with the help of these relationships. Although the new approach needs a more involved calibration, its main advantage is that it is insensitive to dead time effects. As a result, no dead-time corrections are required and the sample parameters can be extracted from three (or even fewer) detectors.

Keywords: nuclear safeguards; fissile material assay; passive non-destructive assay; multiplicity counting.



# List of publications

This thesis is an introduction to and a summary of the work published in the following papers.

## Papers included in the thesis:

### PAPER I

I. Pázsit, L. Pál and L. Nagy. “Multiplicity counting from fission chamber signals in the current mode.” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 839 (2016): 92-101.

### PAPER II

L. Nagy, I. Pázsit and L. Pál. “Multiplicity counting from fission detector signals with time delay effects.” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 884 (2018): 119-127.

### PAPER III

L. Nagy, I. Pázsit and L. Pál. “Two- and three-point statistics of fission chamber signals for multiplicity counting with thermal neutrons.” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* (to be submitted).

## Papers not included in the thesis:

L. Nagy, I. Pázsit and L. Pál. “An extended theory of multiplicity counting from fission chamber signals in the current mode.” *International Conference on Mathematics and Computational Methods Applied to Nuclear Science and Engineering*, (Jeju, Korea), April 16–20 2017.



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Lajos Nagy, Gothenburg, December 2018





dedicated to Éva  
*for serving as a mirror to me*



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# Chapter 1

## Introduction

*The topic of this chapter is some general practical considerations regarding neutron multiplicity counting. Section 1.1 gives a short overview on neutron multiplicity counting. Section 1.2 summarizes the process of neutron emission in spontaneously fissioning materials. In Section 1.3 the operation modes of detector systems and the significance of dead time is discussed. Section 1.4 highlights the motivation behind the research presented in this thesis.*

### 1.1 Overview of multiplicity counting

In nuclear safeguards, nuclear material assay refers to a group of measurement techniques applied in fuel-cycle facilities for material accounting, process control, criticality control, and perimeter monitoring. There are two fundamental groups of assay techniques. Destructive techniques involve sampling the material and analysing the sample with chemical procedures. Nondestructive techniques, on the other hand, measure radiation from the nuclear material. The main advantage of nondestructive techniques is that they are faster, simpler and they reduce operator exposure; they are, however, less accurate than destructive methods. Nondestructive assay techniques can be characterized as passive or active depending on whether they measure radiation from the spontaneous decay of the nuclear material or radiation induced by an external source.

*Neutron multiplicity counting* is a passive nondestructive assay technique used to gain quantitative information on samples of spontaneously fissioning heavy-nuclide materials (e.g.: uranium, plutonium, californium, etc.) by measuring spontaneously emitted neutron radiation [1]. In a general measurement setup, the sample is surrounded by a large number (usually several dozens) of neutron detectors; such an arrangement is shown on Figure 1.1. A fraction of all the neutrons emitted by the sample is detected, and the registered signals of the detectors are processed in order to determine the complete heavy-nuclide mass (or some other equivalent quantity) of the sample. The two fundamental processes in the measurement are the emission and detection of neutrons.

The rate and multiplicity of the neutron emission is directly related to the qualitative and quantitative characteristics of the sample [1, 2], and hence, their determination from the corresponding rate and multiplicity of the neutron detection is the primary objective of the assay. Since these measurements are traditionally performed with detectors operating in pulse mode, that is, with pulse counting techniques, the method

was named *multiplicity counting* [1].

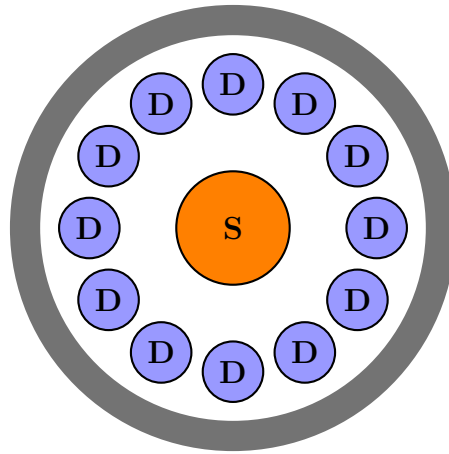


Figure 1.1: A schematic view of the experimental setup of a multiplicity counting measurement, in which a heavy-nuclide sample (S), emitting neutrons spontaneously, is surrounded by a large number of neutron detectors (D).

## 1.2 Emission of neutrons

In a heavy-nuclide sample, spontaneously emitted neutrons might originate from several sources [3]. Most heavy nuclei, including the isotopes of plutonium, decay both by spontaneous fission and by alpha-particle emission. In practice, samples of these nuclides are usually impure and contain light matrix materials (e.g.: oxygen, water, fluorine, etc.) as well, and the typical energy of the alpha-particles is in a suitable range for causing  $(\alpha, n)$  reactions on these light nuclides. On the other hand, besides the neutron-capturing reactions like  $(n, \gamma)$ , heavy isotopes are also capable of different neutron-multiplying reactions such as induced fission or  $(n, 2n)$ .

As a net result of the above reactions, the process of neutron emission in a heavy-nuclide sample goes as follows. Initially, neutrons are produced at random time instants in decay. Some of them might then undergo several levels of internal multiplication, before they all leave the sample. The complete process is illustrated on Figure 1.2.

## 1.3 Detection of neutrons

In most detectors and in particular in fission chambers, the interaction of a neutron with the detector material creates a burst of charge. Using a static electric field, the charge is collected and delivered as a current impulse to the electrodes of the detector. As a result of continued exposure to radiation, a transient current signal made of subsequent (and possibly overlapping) pulses appears at the output of the detector. This current signal is then fed to a chain of signal processing electronics which convert it to a form suitable for acquisition and analysis. Based on the actual components of the processing chain, we distinguish between several modes of operation. Two of them

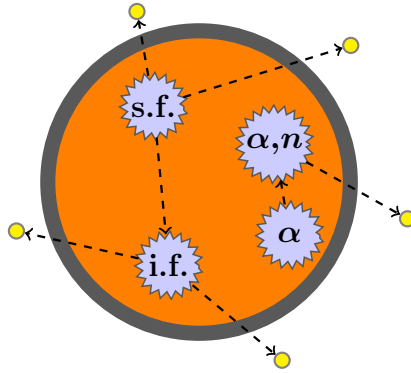


Figure 1.2: Illustration of the neutron emission process in a heavy-nuclide sample. A small number of neutrons are produced spontaneously via spontaneous fission of a heavy isotope or via  $(\alpha, n)$  reaction of a light matrix isotope following the alpha-decay of a heavy isotope. These neutrons (and the induced ones as well) might either leave the sample, get captured (e.g.: in  $(n, \gamma)$  reaction), or undergo multiplication (e.g.: induced fission). The dashed lines represent the path of the neutrons.

are particularly relevant to our discussion: current mode and impulse mode. They are illustrated on Figure 1.3.

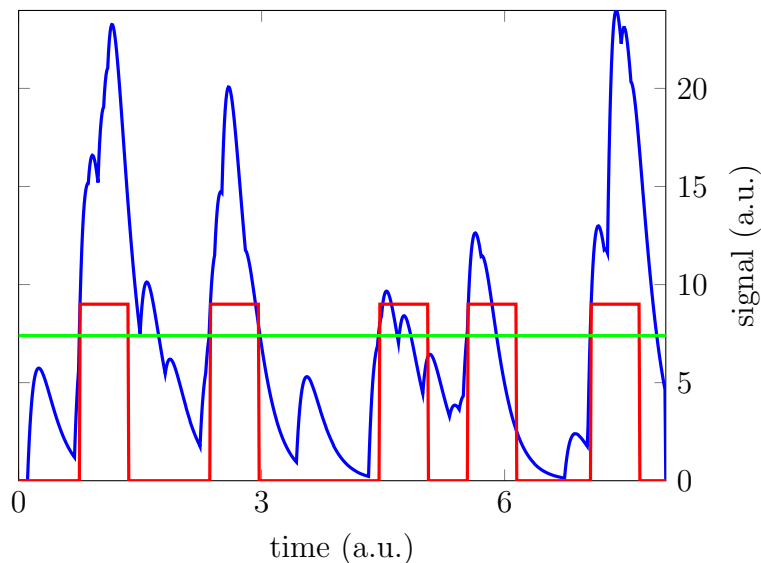


Figure 1.3: An illustration of the signals of detectors in the two modes of operation. The amplified voltage signal is shown with blue lines. The logic pulses generated by an integral discriminator (with threshold represented by the green line) is shown with red.

In almost all applications, the current signal is sent to an amplifier which converts current pulses into linear voltage pulses. Through their presence and amplitude, linear pulses carry information not only on the time of detection, but on the radiation energy as well. In current mode operation the amplified voltage signal is sampled at discrete points in time with suitable frequency and this time-resolved signal is analysed. In some applications, the linear voltage signal is sent to various pulse selecting circuits

that convert selected pulses into logic pulses. Since logic pulses have standard sizes and shapes, they carry information only on the time of (selected) detection events. The pulse selection criteria varies between different circuits. An integral discriminator, for example, selects pulses above a certain threshold. In pulse mode operation the logic signal is analysed which usually involves counting pulses in a certain period of time.

In multiplicity counting measurements, detectors are traditionally operated in pulse mode and sample characteristics are determined from the intensity of coincident detection events. A more detailed discussion of the mathematical basis of the methodology can be found in Section 2.2.

## 1.4 Motivation behind the research

A major limiting factor in the applicability of multiplicity counting is its sensitivity to dead time losses. Every pulse selecting system is characterized by a minimum time called *dead time* that must separate two pulses in order to distinguish them. When a pulse is selected, the information on every other pulse following it within the dead time is lost. There are two fundamental types of dead time losses: nonparalizable and paralyzable; both can be observed on Figure 1.3. The nonparalizable dead time loss originates from the width of the logic pulse: while the logic pulse is on, selected linear pulses will not be converted into logic pulses. In case of an integral discriminator, the paralyzable dead time loss originates from the threshold level: while the voltage signal stays above the threshold, linear pulses will not be selected at all. Clearly, with increasing detection intensities dead time losses also increase, therefore any accurate pulse counting system must include some correction for these losses [4]. Nevertheless, such correction methods are applicable only up to a certain extent.

Multiplicity counting is primarily based on the detection of neutrons which were emitted simultaneously from the sample. Since simultaneously emitted neutrons are likely being detected very close in time, multiplicity counting is particularly sensitive to dead time losses even when measurements are performed on small samples with low emission intensities. Besides applying standard correction techniques [5, 6], dead time losses are traditionally reduced by using several detectors with independent signal processing chains. Nevertheless, with increasing emission intensities, dead time losses become high, and measurement results become inaccurate.

The primary motivation behind the work of this thesis is to provide an alternative version of multiplicity counting which is inherently free of dead time problems. Obviously, such a method can be based only on detectors operating in current mode. Fission chambers are particularly suitable for this type of application because of their advantages over other detector types [7]. The main advantages of the newly proposed technique are that the measurement can be performed with only few detectors and no complicated correction techniques need be applied to the measurement results. For the same reasons, the suggestion of using fission chambers in the current mode in pulsed subcriticality measurements was recently put forward as well [8].



# Chapter 2

## Multiplicity counting in pulse mode

*The topic of this chapter is the theory behind multiplicity counting measurements. Section 2.1 introduces a statistical model of neutron emission in samples of spontaneously fissioning materials. Section 2.2 gives a short summary of the theory of traditional multiplicity counting which is performed with detectors operating in pulse mode.*

### 2.1 Statistical model of neutron emission

A well-known mathematical model of the emission of neutrons in a heavy-nuclide sample is the theory of superfission [2]. A brief overview of this theory will be given here using the terminology of [9].

The emission of neutrons from the sample is the result of a cascade of events. This cascade is initiated by one of the following two reactions. The majority of source neutrons arise from *spontaneous fission* producing neutrons with intensity  $F$  and with a number distribution  $p_{\text{sf}}(n)$ , whose factorial moments of order  $k$  are denoted by  $\nu_{\text{sf},k}$ . In addition, single neutrons are generated via  $(\alpha, n)$  *reactions* with intensity  $Q_\alpha$ . All the initially emitted neutrons (and those arising in induced fission as well) may then undergo one of two possible events: assuming that capture is negligible, they either leave the sample with probability  $1 - p$  or cause induced fission with probability  $p$  producing neutrons with a number distribution  $p_i(n)$ , whose factorial moments of order  $k$  are denoted by  $\nu_{i,k}$ . Since the total time required for such a cascade to complete is negligibly small compared to the average time between subsequent cascades, it is assumed that the cascade takes place instantaneously: the initiating spontaneous fission or  $(\alpha, n)$  reaction is followed by the immediate and simultaneous emission of neutrons from the sample.

Mathematically, the sample emission is described as a compound Poisson process, in which emission events occur with intensity  $Q_s$  each resulting in the emission of neutrons with a number distribution  $P(n)$  whose probability generating function is defined as

$$G(z) = \sum_{n=0}^{\infty} P(n) z^n \quad |z| \leq 1. \quad (2.1)$$

Based on the above description of the cascade process, both  $Q_s$  and  $G(z)$  can be expressed with the intensities and distributions introduced earlier. The total source in-

tensity  $Q_s$  is immediately written as

$$Q_s = F(1 + \alpha \nu_{sf,1}), \quad (2.2)$$

where  $\alpha$  is the so-called  $\alpha$ -ratio defined as

$$\alpha = \frac{Q_\alpha}{F \nu_{sf,1}} \quad (2.3)$$

which equals the expected ratio of the number of neutrons generated in  $(\alpha, n)$  reaction and spontaneous fission. For  $G(z)$  a backward type master equation is written down and solved. Using  $G(z)$ , the distribution  $P(n)$  or its factorial moment of order  $k$  denoted by  $\nu_k$  can easily be reconstructed. In multiplicity counting the first three of these moments have particular importance. In order to simplify the upcoming formulas, let us introduce the modified factorial moments

$$\tilde{\nu}_i = \nu_i(1 + \alpha \nu_{sf,1}). \quad (2.4)$$

The first three modified moments can then be written as

$$\tilde{\nu}_1 = M \nu_{sf,1}(1 + \alpha), \quad (2.5a)$$

$$\tilde{\nu}_2 = M^2 \left[ \nu_{sf,2} + \left( \frac{M-1}{\nu_{i,1}-1} \right) \nu_{sf,1}(1 + \alpha) \nu_{i,2} \right], \quad (2.5b)$$

$$\begin{aligned} \tilde{\nu}_3 = M^3 & \left[ \nu_{sf,3} + \left( \frac{M-1}{\nu_{i,1}-1} \right) [3\nu_{sf,2}\nu_{i,2} + \nu_{sf,1}(1 + \alpha)\nu_{i,3}] \right. \\ & \left. + 3 \left( \frac{M-1}{\nu_{i,1}-1} \right)^2 \nu_{sf,1}(1 + \alpha) \nu_{i,2}^2 \right], \end{aligned} \quad (2.5c)$$

where  $M$  is the so-called net leakage multiplication defined as

$$M = \frac{1-p}{1-p\nu_{i,1}} \quad (2.6)$$

which equals the expected number of neutrons that leave the sample per one initial neutron.

Equations (2.2) and (2.5) show that the emission intensity and the moments of the number of emitted neutrons are functions of a large number of parameters. Since the fissile composition of the sample is assumed to be known when performing a multiplicity counting measurement, the factorial moments  $\nu_{i,k}$  and  $\nu_{sf,k}$  can also be assumed to be known. Determination of the three remaining unknown parameters,  $F$ ,  $\alpha$  and  $M$  (among which  $F$  is the most important as it is directly related to the fissile mass) is the goal of multiplicity counting.

## 2.2 Detection rates

The traditional method of multiplicity counting utilizes the counting statistics of detectors operating in pulse mode. Specifically, the detection intensities of the first few

$k$ -tuplets ( $k$  neutrons originating from the same sample emission) are determined. The theoretical basis of the method is discussed here briefly.

Consider a large array of detectors. Let  $\varepsilon$  denote the detection efficiency of the entire system, that is,  $\varepsilon$  equals the probability of detecting a neutron by any of the detectors in the array. Now, let  $C_k$  denote the intensity of detecting  $k$  neutrons from the same emission. It can be shown that

$$C_k = F \frac{\tilde{\nu}_k \varepsilon^k}{k!}, \quad (2.7)$$

where  $F$  is the spontaneous fission intensity and  $\tilde{\nu}_k$  are the modified factorial moments, both introduced in the previous section. In the practical application of multiplicity counting, only the first three coincident detection rates play a role: these are called the singles ( $S$ ), doubles ( $D$ ) and triples ( $T$ ) rates. Based on (2.7), they are defined as

$$S = F \tilde{\nu}_1 \varepsilon, \quad (2.8a)$$

$$D = F \frac{\tilde{\nu}_2 \varepsilon^2}{2}, \quad (2.8b)$$

$$T = F \frac{\tilde{\nu}_3 \varepsilon^3}{6}. \quad (2.8c)$$

The above simple model disregards an important practical feature of the measurement. Due to the finite distance between the sample and the detector, the detection of particles takes place with a certain time delay (referred to as the “detector die-away time” in safeguards parlance) with respect to the instant of their emission. Moreover, the time delay is random in nature for at least two reasons: first, the spatial transport and slowing down of the neutron is a random process; second, the emitted neutron has a random energy. As a consequence, even neutrons originating from the same emission will be detected non-simultaneously. Clearly, the exact distribution of the delay depends primarily on the experimental set-up and the (unknown) sample properties. In thermal detection systems, however, it is primarily determined by the slowing down of neutrons. Therefore, in case of the most frequently used thermal neutron based multiplicity counter devices, an exponential distribution is assumed.

In order to account for the non-simultaneous arrival of neutrons originating from the same sample emission, a detection time gate is applied in the measurement. Specifically, the first detection triggers a timer, after which further counts are collected only for a fixed period of time. The length of the gate is chosen in a way that it is short enough to distinguish between detections from subsequent source events, but long enough to collect as much counts from one source event as possible by covering a large fraction of the detector die-away time. Nonetheless, application of such a time gate will inevitably lead to some loss of double and triple coincident counts. As a consequence, the measured doubles and triples rates will be smaller than those given by (2.8). This effect is taken into account by introducing the so-called doubles and triples gate factors  $f_d$  and  $f_t$  and

reformulating the detection rates as

$$S = F \tilde{\nu}_1 \varepsilon, \tag{2.9a}$$

$$D = F \frac{\tilde{\nu}_2 \varepsilon^2}{2} f_d, \tag{2.9b}$$

$$T = F \frac{\tilde{\nu}_3 \varepsilon^3}{6} f_t. \tag{2.9c}$$

A detailed discussion on the gate factors and their determination is given in [1].

By inserting the modified factorial moments (2.5) into the expression of the detection rates, a system of algebraic equations is obtained for the three unknown sample parameters,  $F$ ,  $M$  and  $\alpha$ . The values of the sample parameters can then be obtained from the measured values of the detection rates by algebraic inversion. The details of the inversion procedure are discussed in [9].

As it was already mentioned in Chapter 1, the main difficulty with operating detectors in pulse mode (which is the basis of traditional multiplicity counting) is their sensitivity to dead time effects which leads to a loss of coincident count information. This dead time problem can be eliminated by developing an alternative method based on detectors operating in current mode. The theory of such a method will be the topic of the rest of this thesis.

# Chapter 3

## Multiplicity counting in current mode

*The topic of this chapter is the theory behind the new method of multiplicity counting which is based on the moments of the signals of detectors operating in current mode. Section 3.1 introduces the stochastic theory of the detector currents. Based on Paper I, Section 3.2 presents the calculation of one point moments assuming an instantaneous detection of neutrons. Based on Paper II, Section 3.3 discusses the effect of delayed detection on the use of one-point moments. Based on Paper III, Section 3.4 presents the calculation of two- and three-point moments. Finally, Section 3.5 gives some general recommendations on the practical use of the proposed method.*

### 3.1 Stochastic theory of detector currents

To overcome the limitations of traditional multiplicity counting arising from the dead time effect, an alternative form of the measurement method is proposed. The new approach utilizes the temporal statistics of the time-resolved signals of detectors operating in current mode. Specifically, various moments of the signals are determined which are expected to carry information on the sample. The theory of the proposed method is based on a formalism developed recently for describing the fluctuating signals of neutrons detectors [10]. The key elements of this formalism are summarized here briefly.

The detector is characterized with a detection efficiency  $\varepsilon$ . Each detection generates a response in the form of a pulse described with a deterministic shape  $f(t)$  and a random magnitude  $a$ . The distribution of the magnitude is specified by its probability density function  $w(a)$ . We introduce the  $n$ th raw moment of  $a$  defined as

$$\langle a^n \rangle = \int_0^\infty a^n w(a) da. \quad (3.1)$$

Clearly, the complete signal of the detector will be the sum of the pulses induced by the individual detections. Let us denote this signal by  $y(t)$ . Recalling that the emission of neutrons is a stationary process, it is easy to see that  $y(t)$  is a stationary process as well. Among other things, this means that the signal has a constant asymptotic mean

denoted by  $\langle y \rangle$  and defined as

$$\langle y \rangle = \lim_{t \rightarrow \infty} \mathbb{E}[y(t)]. \quad (3.2)$$

In the following we shall consider three detectors labelled with 1–3. Correspondingly, their signals will be denoted by  $y_1(t), y_2(t)$  and  $y_3(t)$ . We shall assume that all the detectors are identical: they are characterized by the same efficiency  $\varepsilon$ , pulse shape  $f(t)$  and amplitude distribution  $w(a)$ . Our ultimate goal is to determine some of the low order moments of the signals. These will include auto and cross cumulants derived from the distribution of the process in one or more (two and three) points in time (called *finite dimensional distributions*). We shall see that the cumulants form simple expressions with the detection rates (2.8) of traditional multiplicity counting. This shows that the moments of the detector currents contain the same information on the sample as the coincident count rates of the traditional method. As a consequence, sample parameters can be extracted from the moments of the detector current using an unfolding procedure similar to the one developed for the traditional method. Due to its inherent insensitivity to dead time effects, however, the newly proposed method has the benefit of requiring at most three detectors and not needing any dead time corrections.

## 3.2 One point moments with instant detection

The one point distribution of the detector signals characterizes their statistical properties at a single time instant  $t$ . Several moments can be derived from the one point distribution of the signals, but only few of them are of interest to our particular application. In case of one detector, we shall examine the (stationary) *mean*, *variance* and *skewness* of its signal defined as

$$\kappa_1 = \lim_{t \rightarrow \infty} \mathbb{E}[y_1(t) - \langle y_1 \rangle], \quad (3.3a)$$

$$\kappa_2 = \lim_{t \rightarrow \infty} \mathbb{E}\{[y_1(t) - \langle y_1 \rangle]^2\}, \quad (3.3b)$$

$$\kappa_3 = \lim_{t \rightarrow \infty} \mathbb{E}\{[y_1(t) - \langle y_1 \rangle]^3\}. \quad (3.3c)$$

In case of two and three detectors, we shall consider the (stationary) *covariance* and *bi-covariance* of their signals defined as

$$\kappa_{1,1} = \lim_{t \rightarrow \infty} \mathbb{E}\{[y_1(t) - \langle y_1 \rangle][y_2(t) - \langle y_2 \rangle]\}, \quad (3.4a)$$

$$\kappa_{1,1,1} = \lim_{t \rightarrow \infty} \mathbb{E}\{[y_1(t) - \langle y_1 \rangle][y_2(t) - \langle y_2 \rangle][y_3(t) - \langle y_3 \rangle]\}. \quad (3.4b)$$

The above cumulants can be calculated from their corresponding cumulant-generating functions with simple differentiation. These cumulant-generating functions are derived from master equations written for the one point probability density functions of the detector signals. The main building block of the equations is the probability distribution function  $h(y, t)$  of the detector response to the detection of *one single neutron*. Assuming that emitted neutrons are detected instantaneously, the density function reads as

$$h(y, t) = \int_0^\infty \delta[y - af(t)] w(a) da. \quad (3.5)$$

Once the cumulant-generating functions are known, the sought moments can be calculated with simple differentiation. The details of this lengthy derivation can be found in Paper I; here only the final results are presented. The cumulants of the signal of one detector read as

$$\kappa_1 = S \langle a \rangle I_1, \quad (3.6a)$$

$$\kappa_2 = [S \langle a^2 \rangle + 2D \langle a \rangle^2] I_2, \quad (3.6b)$$

$$\kappa_3 = [S \langle a^3 \rangle + 6D \langle a \rangle \langle a^2 \rangle + 6T \langle a \rangle^3] I_3. \quad (3.6c)$$

The cumulants of the signals of two and three detectors read as

$$\kappa_{1,1} = 2D \langle a \rangle^2 I_2, \quad (3.7a)$$

$$\kappa_{1,1,1} = 6T \langle a \rangle^3 I_3. \quad (3.7b)$$

In the above formulas, we introduced the integral expression

$$I_n = \int_0^\infty f^n(t) dt. \quad (3.8)$$

Expressions (3.6)–(3.7) form a system of algebraic equations between the moments (3.3)–(3.4) of the detector signals as well as the detection rates (2.8) of traditional multiplicity counting. The elements of the matrix of this system depend on the moments (3.1) of the pulse amplitude and on the integrals (3.8) of the pulse shape. These latter can be determined from calibration, as has been demonstrated using fission chambers in reactor measurements [11]. As a consequence, when the proper moments of the measured detector signals are calculated, the singles, doubles and triples detection rates can be obtained by algebraic inversion. Once these are known, the exact same procedure can be used to unfold the sample parameters as the one applied in traditional multiplicity counting (see Section 2.2).<sup>1</sup>

Recall that the above results are based on the rather unrealistic assumption that neutrons are detected instantly (hence simultaneously) after their emission. However, as it was pointed out in Section 2.2, detection of neutrons takes place with a random time delay, hence even neutrons originating from the same emission will be detected non-simultaneously. The effect of this phenomenon on the statistics of the detector current is investigated in the following subsection.

### 3.3 One point moments with delayed detection

To account for the non-instantaneous detection of neutrons, a random variable  $\tau$  is introduced representing the arrival time to the detector with respect to the time of emission. We shall assume, that this delay is independent and identically distributed for every neutron, including those originating from the same emission.<sup>2</sup> Denoting the

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<sup>1</sup>Note that this step requires the knowledge of the detector efficiency as well which is incorporated into the expressions of the detection rates. The efficiency, however, is also easily determined from calibration.

<sup>2</sup>It is worth noting that in principle, the detection times of neutrons from the same source emission may not be totally independent due to the correlations between their energies. Such correlations in the energy (hence in the detection time as well) are nevertheless expected to be small, in particular after the internal multiplication of neutrons in the sample.

density function of  $\tau$  by  $u(\tau)$ , the density function  $h(y, t)$  of the detector response to a single detection will read as

$$h(y, t) = \int_0^\infty \int_0^\infty \delta[y - af(t - \tau)] w(a) u(\tau) da d\tau. \quad (3.9)$$

Note that by substituting  $u(\tau) = \delta(\tau)$ , which corresponds to the instant detection of neutrons, the above expression of  $h$  reverts back to (3.5) from the previous subsection.

The values of the cumulants corresponding to the case of delayed detection can be calculated by performing the exact same steps as in the case of instant detection. The only difference is that the above form of  $h(y, t)$  is used instead of (3.5). The details of this straightforward but lengthy derivation can be found in Paper II; here only the final results are presented. The cumulants of the signal of one detector read as

$$\kappa_1 = S \langle a \rangle I_1, \quad (3.10a)$$

$$\kappa_2 = [S \langle a^2 \rangle + 2D \langle a \rangle^2 \xi_{1,1}] I_2, \quad (3.10b)$$

$$\kappa_3 = [S \langle a^3 \rangle + 6D \langle a \rangle \langle a^2 \rangle \xi_{1,2} + 6T \langle a \rangle^3 \xi_{1,1,1}] I_3. \quad (3.10c)$$

The cumulants of the signals of two and three detectors read as

$$\kappa_{1,1} = 2D \langle a \rangle^2 \xi_{1,1} I_2, \quad (3.11a)$$

$$\kappa_{1,1,1} = 6T \langle a \rangle^3 \xi_{1,1,1} I_3. \quad (3.11b)$$

Here we introduced the integral expressions

$$\xi_{1,1} = \frac{1}{I_2} \int_0^\infty [I_1(t)]^2 dt, \quad (3.12a)$$

$$\xi_{1,2} = \frac{1}{I_3} \int_0^\infty I_1(t) I_2(t) dt, \quad (3.12b)$$

$$\xi_{1,1,1} = \frac{1}{I_3} \int_0^\infty [I_1(t)]^3 dt, \quad (3.12c)$$

where the function  $I_n(t)$  is defined as

$$I_n(t) = \int_0^\infty f^n(t - \tau) u(\tau) d\tau. \quad (3.13)$$

It is seen that the above expressions of the moments are almost identical to (3.6)–(3.7) corresponding to the case of instantaneous detection. The only difference is the presence of the integral factors defined by (3.12). These are analogous to the doubles and triples gate factors of traditional multiplicity counting in two ways. First, they appear at the same position: they multiply the doubles and triples detection rates. Second, they depend on  $u(\tau)$ , which essentially characterizes the temporal separation of neutrons, just like the die-away time used to calculate the traditional gate factors. Because of this analogy, we shall refer to (3.12) as gate factors as well. Note, however, that while in the traditional method, the gate factors were introduced empirically, in the newly proposed method, they appear straightforward from the theory. If in the



given measurement setup the form of  $u(\tau)$  is known<sup>3</sup>, the detection rates (and the unknown sample parameters) can be determined from the measured signals the same way as described in the previous section for the case of instant detection.

In order to investigate the effect of the random time delay on the practical applicability of the proposed method, let us calculate the values of the gate factors (3.12) for actual analytic forms of the detector pulse shape  $f(t)$  and the time delay density  $u(\tau)$ . For the detector pulse shape, a simple exponential function with a time scale parameter  $\alpha > 0$  (characterizing the “width” of the pulse) is chosen:

$$f(t; \alpha) = \begin{cases} e^{-t/\alpha}, & \text{if } t \geq 0 \\ 0, & \text{if } t < 0, \end{cases} \quad (3.14)$$

For the distribution of the time delay, the exponential distribution with expected value  $\beta > 0$  (which also characterizes the variance, i.e. “spread” of the delay) is chosen:

$$u(\tau; \beta) = \begin{cases} \frac{1}{\beta} e^{-\tau/\beta}, & \text{if } \tau \geq 0 \\ 0, & \text{if } \tau < 0. \end{cases} \quad (3.15)$$

With these choices, the gate factors can be written as the following functions of  $x = \beta/\alpha$ :

$$\xi_{1,1}(x) = \frac{1}{1+x}, \quad (3.16a)$$

$$\xi_{1,2}(x) = \frac{2+3x}{2+6x+4x^2}, \quad (3.16b)$$

$$\xi_{1,1,1}(x) = \frac{2}{2+5x+2x^2}. \quad (3.16c)$$

Figure 3.1 shows that the gate factors are monotonically decreasing functions of  $x$  – or  $\beta$ , when a fixed value of  $\alpha$  is considered. When the width of the distribution of the delay is in the same order as the width of the pulse, the values of all three gate factors are close to unity. In this case it is still possible to extract the multiplicity rates from the detector currents. However, when the distribution of the delay is much wider than the pulse, the gate factors become negligibly small, therefore the doubles and the triples rates will enter the formulae with very small weights. As a consequence, the moments of the detector current will carry information only on the singles rates.

Taking into account that the characteristic pulse width of an ionization chamber is typically in the order of hundreds of nanoseconds, a narrow time delay distribution can only be achieved in fast detection systems, where neutrons are detected directly from the sample, without slowing down. In case of the most frequently used thermal detection systems, the time delay distribution will be several orders of magnitude wider than the detector pulse.

This leads to the conclusion, that multiplicity counting based on the one point moments (3.3)–(3.4) of detector signals is possible only when fast detection is applied. A major disadvantage of this approach is the low detection efficiency of these systems compared to the more frequently used thermal systems. To exploit the benefits of thermal detection, the methodology needs to be extended to the use of temporal correlations of the signals. The possibility of this approach is investigated in the next subsection.

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<sup>3</sup>Since the density function of the time between the emission and detection of particles is difficult to measure, it is most likely to be obtained either from simulation or by assuming an analytic function.

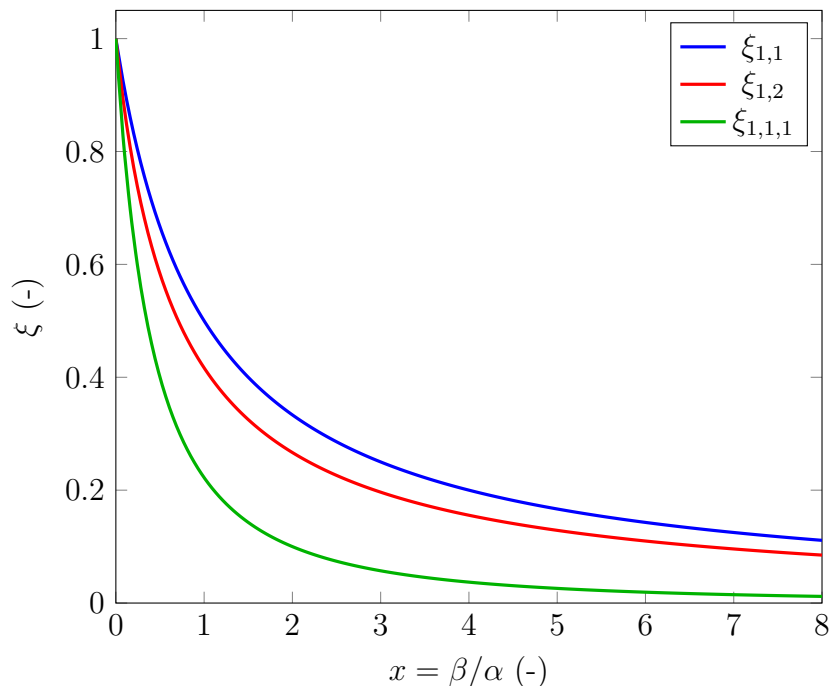


Figure 3.1: Dependence of the gate factors (3.12) on the ratio of the spread of the time delay distribution and the pulse width.

### 3.4 Two- and three-point moments with delayed detection

In order to explore the temporal correlations in the signals of the detectors, their distribution must be described at more than one point in time. Besides time  $t$ , a second time instant  $t - \theta$  is chosen for two-point distributions, whereas for three-point distributions an additional third time instant  $t - \theta - \rho$  is considered as well. We shall assume, that  $\theta$  and  $\rho$  are both nonnegative, that is,  $\theta, \rho \geq 0$ .

Although there are a few cumulants that can be derived from the multi-point distributions of the detector signals, it is reasonable to examine the two- and three-point analogies of those defined by (3.3) and (3.4). In case of one detector, we shall consider the (stationary) *auto-covariance* and *auto-bi-covariance* functions of its signal defined as

$$\text{Cov}_2(\theta) = \kappa_2(\theta) = \lim_{t \rightarrow \infty} \mathbb{E} \{ [y_1(t) - \langle y_1 \rangle] [y_1(t - \theta) - \langle y_1 \rangle] \}, \quad (3.17a)$$

$$\begin{aligned} \text{Cov}_3(\theta, \rho) = \kappa_3(\theta, \rho) = \lim_{t \rightarrow \infty} \mathbb{E} \{ [y_1(t) - \langle y_1 \rangle] [y_1(t - \theta) - \langle y_1 \rangle] \\ \times [y_1(t - \theta - \rho) - \langle y_1 \rangle] \}. \end{aligned} \quad (3.17b)$$

In case of two and three detectors, the (stationary) *cross-covariance* and *cross-bi-*

covariance functions of their signals defined as

$$\text{Cov}_{1,1}(\theta) = \kappa_{1,1}(\theta) = \lim_{t \rightarrow \infty} \mathbb{E} \{ [y_1(t) - \langle y_1 \rangle] [y_2(t - \theta) - \langle y_2 \rangle] \}, \quad (3.18a)$$

$$\begin{aligned} \text{Cov}_{1,1,1}(\theta, \rho) = \kappa_{1,1,1}(\theta, \rho) &= \lim_{t \rightarrow \infty} \mathbb{E} \{ [y_1(t) - \langle y_1 \rangle] [y_2(t - \theta) - \langle y_2 \rangle] \\ &\times [y_3(t - \theta - \rho) - \langle y_3 \rangle] \} \end{aligned} \quad (3.18b)$$

are chosen.

Similarly to the one-point moments discussed in the previous sections, the two- and three-point cumulants are calculated from the corresponding cumulant-generating functions. These are also derived from master equations which, in this case, are written for the two- and three-point probability density functions of the detector signals. Besides the one-point density function  $h(y, t)$  of the detector response given by (3.9), the two- and three-point density functions will also appear in the derivations. Assuming delayed detection of neutrons, these can be written as

$$\begin{aligned} h_2(y_1, y_2, t, \theta) &= \int_0^\infty \int_0^\infty \delta[y_1 - af(t - \tau)] \\ &\times \delta[y_2 - af(t - \theta - \tau)] w(a) u(\tau) da d\tau \end{aligned} \quad (3.19)$$

and

$$\begin{aligned} h_3(y_1, y_2, y_3, t, \theta, \rho) &= \int_0^\infty \int_0^\infty \delta[y_1 - af(t - \tau)] \\ &\times \delta[y_2 - af(t - \theta - \tau)] \\ &\times \delta[y_3 - af(t - \theta - \rho - \tau)] w(a) u(\tau) da d\tau. \end{aligned} \quad (3.20)$$

Once the cumulant-generating functions are calculated, the cumulants (3.17)–(3.18) can be obtained by simple differentiation. The details of this derivation can be found in Paper III. We must mention, however, that for the purposes of multiplicity counting not the cumulants themselves, but their integrals with respect to the time variables will be of primary interest. These are defined as

$$\text{Cov}_2 = \int_0^\infty \text{Cov}_2(\theta) d\theta, \quad (3.21a)$$

$$\text{Cov}_3 = \int_0^\infty \int_0^\infty \text{Cov}_3(\theta, \rho) d\rho d\theta \quad (3.21b)$$

and

$$\text{Cov}_{1,1} = \int_0^\infty \text{Cov}_{1,1}(\theta) d\theta, \quad (3.22a)$$

$$\text{Cov}_{1,1,1} = \int_0^\infty \int_0^\infty \text{Cov}_{1,1,1}(\theta, \rho) d\rho d\theta. \quad (3.22b)$$

By substituting the calculated cumulants into the above integrals, after considerable algebra, whose details are found in Paper III, we obtain

$$\text{Cov}_2 = \frac{1}{2} [S \langle a^2 \rangle + 2D \langle a \rangle^2] I_1^2, \quad (3.23a)$$

$$\text{Cov}_3 = \frac{1}{6} [S \langle a^3 \rangle + 2D \langle a \rangle \langle a^2 \rangle (\xi_A + \xi_B + \xi_C) + 6T \langle a \rangle^3] I_1^3, \quad (3.23b)$$

for one detector, and

$$\text{Cov}_{1,1} = D \langle a \rangle^2 I_1^2, \quad (3.24a)$$

$$\text{Cov}_{1,1,1} = T \langle a \rangle^3 I_1^3, \quad (3.24b)$$

for two and three detectors. Here, we introduced the following gate factors:

$$\xi_A = \frac{6}{I_1^3} \int_0^\infty \int_0^\infty \int_0^\infty I_1(t-\theta) I_{1,1}(t, t-\theta-\rho) dt d\theta d\rho, \quad (3.25a)$$

$$\xi_B = \frac{6}{I_1^3} \int_0^\infty \int_0^\infty \int_0^\infty I_1(t) I_{1,1}(t-\theta, t-\theta-\rho) dt d\theta d\rho, \quad (3.25b)$$

$$\xi_C = \frac{6}{I_1^3} \int_0^\infty \int_0^\infty \int_0^\infty I_1(t-\theta-\rho) I_{1,1}(t, t-\theta) dt d\theta d\rho, \quad (3.25c)$$

where the function  $I_{1,1}(t, s)$  is defined as

$$I_{1,1}(t, s) = \int_0^\infty f(t-\tau) f(s-\tau) u(\tau) d\tau. \quad (3.26)$$

Expressions (3.23) and (3.24) are very similar to the one-point moments (3.10) and (3.11) from the previous section. The most apparent difference is that, except in the second term of  $\text{Cov}_3$ , all the gate factors have disappeared. For this reason, (3.23) and (3.24) are actually more reminiscent to (3.6) and (3.7), the one-point moments obtained by assuming an instant detection of neutrons. Again, when the detection efficiency, the pulse shape and the time delay distribution is known, the detection rates (and the unknown sample parameters) can be determined from the measured signals using the inversion procedure mentioned in Section 3.2.

The presence of the gate factors (3.25) in  $\text{Cov}_3$  is somewhat counter-intuitive. Based on an analogy with  $\text{Cov}_2$ , one would expect that  $\xi_A$ ,  $\xi_B$  and  $\xi_C$  are all equal to 1 hence their sum equals to 3. Nevertheless, it can be worth investigating their values in a similar way than the values of the gate factors in the previous section. Again, assuming an exponential pulse shape with a time scale parameter (or “width”)  $\alpha > 0$  and an exponentially distributed time delay with expected value (or “spread”)  $\beta > 0$ , the gate factors (3.25) can be written as the following functions of  $x = \beta/\alpha$ :

$$\xi_A = \frac{1 + 3x + 3x^2}{1 + 3x + 2x^2}, \quad (3.27a)$$

$$\xi_B = \frac{2 + 18x + 58x^2 + 63x^3 + 18x^4}{6 + 36x + 78x^2 + 72x^3 + 24x^4}, \quad (3.27b)$$

$$\xi_C = \frac{1 + 3x}{1 + 3x + 2x^2}. \quad (3.27c)$$

Figure 3.2 shows that neither the individual gate factors, nor their sum is constant, and they do not add up to 3. On the other hand, it is seen that the sum of the three gate factors is quite close to the expected value 3, and it changes relatively little as the ratio of the pulse width to the width of the time delay density changes.

To conclude, the non-vanishing character of the sum of the gate factors in  $\text{Cov}_3$  and their absence from all the other expressions in (3.23) and (3.24) has a positive consequence on the practical applicability of the method. Unlike in the case of the one-point moments (3.10) and (3.11) a thermal detection system can be used to determine the values of (3.23) and (3.24).

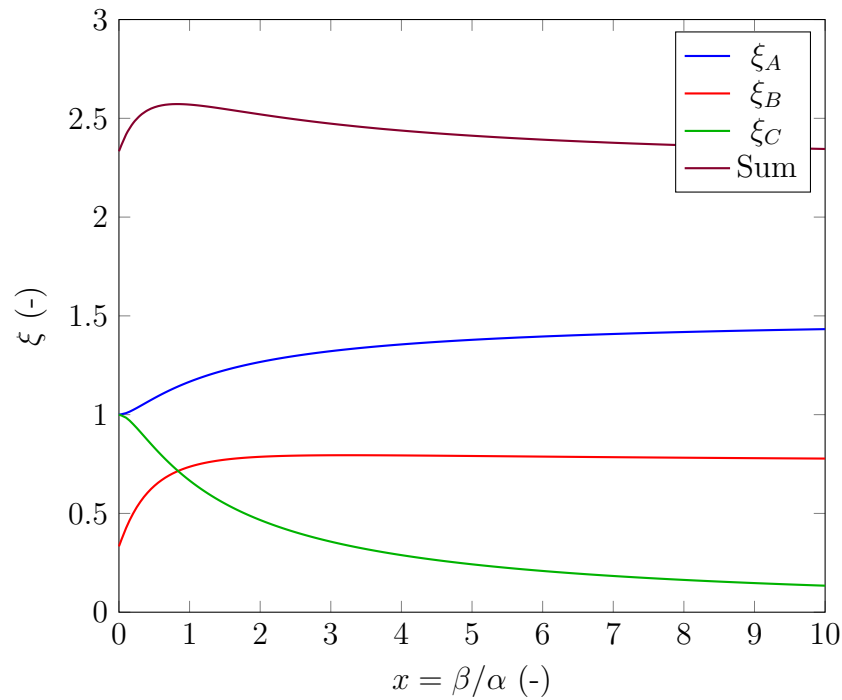


Figure 3.2: Dependence of the gate factors (3.25) on the ratio of the spread of the time delay distribution and the pulse width.

### 3.5 Practical considerations

In the view of the results of sections 3.3 and 3.4, it is possible to design a measurement procedure based on quantities which are independent of the time delay distribution and hence on the measurement setup.

One can use the mean  $\kappa_1$  to determine the singles rates, the covariance  $\text{Cov}_{1,1}$  or  $\text{Cov}_2$  to determine the doubles rate, and the bi-covariance  $\text{Cov}_{1,1,1}$  to determine the triples rate. The suggested moment–rate combinations are summarised in Table 3.1. Using these cumulants, only the detection efficiency  $\varepsilon$  and the detector pulse shape  $f(t)$  needs to be determined from calibration.

cumulant	rate
$\kappa_1$	$S$
$\text{Cov}_{1,1}$	$D$
$\text{Cov}_{1,1,1}$	$T$
$\text{Cov}_2$	$S, D$



# Chapter 4

## Summary

Neutron multiplicity counting is a commonly used non-destructive assay method for estimating unknown parameters (primarily the mass) of samples of spontaneously fissioning materials. The method is based on determining the first three lowest order moments of the number of particles emitted simultaneously from the sample, through measuring the so-called singles, doubles and triples detection rates from the counting statistics of the detectors. The measurements are usually performed with thermal neutron detectors operating in pulse counting mode. A major problem with pulse mode of operation is its sensitivity to dead time effects, which poses a constraint on the ability to extract correlated neutron counts.

To overcome this difficulty, a new method of multiplicity counting has been developed, which is based on the statistics of the time-resolved signals of detectors operating in current mode. Specifically, the possibility of extracting the traditional multiplicity count rates from the auto and cross cumulants of the stationary signals of different groups of detectors was investigated.

In Section 3.2 some of the low order one-point cumulants of the detector signals were calculated. It was assumed that neutrons emitted simultaneously from the sample were also detected simultaneously, hence each neutron inducing a detector pulse of finite width at the same time. From the analytical formulae obtained, it was shown that the singles, doubles and triples count rates could be retrieved from the measured cumulants of the detector current by algebraic inversion. In this process, certain parameters of the detector, such as its pulse shape and amplitude distribution have to be known.

It is though clear that in reality the detection of neutrons of common origin does not take place simultaneously. Even in the case of fast detection systems where neutrons are detected directly from the sample, there will be differences in the arrival times due to the energy spectrum, and hence different velocities of the source neutrons. Much larger differences are expected in the case of thermal systems where neutrons are slowed down before detection. To account for this phenomenon, in Section 3.3 it was assumed that multiple neutrons from an initial source event arrive to the detector with a random, individual time delay, described by an independent, identically distributed density function. It was found that more complicated, but still closed analytical formulae exist between the multiplicity rates and the cumulants. However, in these expressions, in addition to the detector pulse shape and amplitude distribution, the probability density of the delay time also appears. Unlike the detector pulse shape, this density is not

a unique function of the detector, rather it depends on the (a priori unknown) source parameters and the experimental setup (geometry and possible moderating material), whose effects on the time delay density are usually not known in advance. Another, more problematic difficulty is that when the width of the density function of the time delay is much wider than the pulse width (which, considering the typical pulse width of a few tens of nanoseconds, is often the case), the coefficients multiplying the doubles and triples rates in the cumulants vanish hence only the singles rate can be unfolded from the cumulants.

To remedy these shortcomings, in Section 3.4 the theory was extended to the use of two- and three point distributions in time. It was shown that the integrals of suitably chosen moments with respect to their variables become independent of the probability density function of the detection delay. This is a very gratifying result, because it means that at least in principle, the singles doubles and triples detection rates can be extracted from the measurements even thermal neutron detectors are used.

Although some very promising theoretical results were found regarding the newly proposed method of multiplicity counting, it remains to prove its applicability in practice. To assess the potentials of the method, both pilot measurements, as well as extensive simulations and a sensitivity and uncertainty analysis are necessary, which are currently under preparation.



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