Neutron & Gamma Multiplicity Modeling at LLNL

Manoj K. Prasad

Lawrence Livermore National Laboratory, Livermore CA 94550, USA
prasad1@llnl.gov
LLNL-CONF-722877, February 2017

Abstract - *Neutron and gamma multiplicity assay analysis attempts to determine SNM (special nuclear material) parameters such as mass and level of criticality of a fissioning system, based on inverting the information in neutron and gamma counting. We describe the various modeling capabilities developed and published at LLNL towards this effort.*

I. INTRODUCTION

Neutron assay analysis has been based on efficient counting of thermal neutrons on the μs time-scale using ^{3}He detectors. In its most fundamental form, the counting data are just the time of each neutron count. Even if many neutrons from a chain arrive at the detector approximately simultaneously (< 1 μs spread in arrival times), they will diffuse through a moderator (polyethylene) for typically tens of μs before being detected by the ^{3}He(n,p)T reaction which creates a charge pulse. The neutron detection times are therefore spread over tens of μs and the fission chains are approximately instantaneous on the detection time scale.

Fast neutron and gamma-ray counting using scintillators (liquid, stilbene) arrays can measure evolving fission chains with nanosecond time resolution. Fast counting can therefore isolate and resolve the fission chain bursts. The time dependence of correlations now comes from the time evolution of the fission chains rather than from a diffusion process in the detector. There will still be a spread in arrival times from time-of-flight to the scintillator detectors and the energy spectrum of the neutrons resulting in an effective diffusion time on the order of ten nanoseconds. For gamma counting the effective diffusion time approximates the effect of geometry on the time-of-flight to the detectors and is typically less than a nanosecond.

We describe here the neutron and gamma multiplicity modeling capabilities, for both thermal and fast counting, developed and published at LLNL.

II. Statistical Theory of Fission Chains and Generalized Poisson Neutron Counting Distributions

The neutron counting probability distribution for a multiplying medium was shown by Hage and Cifarelli to be a generalized Poisson distribution that depends on the fission chain number distribution. Assuming the fission chains are instantaneous on the timescale of the neutron detection process, an analytic formula was obtained at LLNL (Ref. 1) for this number distribution, the probability to produce a number of neutrons in a fission chain. This point model theory has been used to analyze thermal neutron counting data acquired with ^{3}He detectors.

The general formula for the count distribution generating function in terms of the fission chain generating function can be put in the form (Ref. 1),

$$\sum_{n=0}^{\infty} b_n(T) y^n = Exp\left[F_s \int_0^T \frac{dt}{(1-e^{-\lambda t})}\left(C^{spont}\left[h\left[1-\varepsilon(1-y)\left(1-e^{-\lambda t}\right)\right]\right]-1\right)\right] \tag{1}$$

where T is the measurement time gate, λ is the inverse neutron lifetime, ε is the detection efficiency, Fs is the rate of spontaneous fission source of neutrons. The fission chain is initiated by spontaneous fission, whose generating function is C^{spont} and $h(y) = \sum_{\nu=0}^{\infty} P_\nu y^\nu$ is the generating function of a fission chain initiated by a single neutron which creates ν neutrons with probability P_ν. The generating function variable y tracks the number of neutrons in the fission chain. The generating function h(y) satisfies the Bohnel functional equation:

$$h(y) = (1-p)y + p\, C[h(y)] \tag{2}$$

where p is the probability to induce a fission and C is the generating function for induced fission which creates k neutrons with probability C_k: $C(y) = \sum_{k=0}^{\infty} C_k y^k$. A formal solution for h(y) can be expressed as a sum over all combinatorial trees (q = 1-p):

$$h(y) = \sum_{\nu=0}^{\infty} P_\nu y^\nu = qy + q^\nu \sum_{n_f=1}^{\infty} \frac{p^{n_f}}{n_f!}\frac{(n_f-1+\nu)!}{\nu!} \sum_{n_0,n_1,n_2,\cdots} \frac{n_f!}{n_0!\,n_1!\,n_2!\cdots}\left(C_0^{n_0} C_1^{n_1} C_2^{n_2}\cdots\right) \tag{3}$$

An independent (α,n) source of neutrons at a rate S can be described with a corresponding generating function:

$$\sum_{n=0}^{\infty} b_n(T) y^n = Exp\left[S\int_0^T \frac{dt}{(1-e^{-\lambda t})}\left(h\left[1-\varepsilon(1-y)\left(1-e^{-\lambda t}\right)\right]-1\right)\right] \tag{4}$$

An external random source of neutrons at a rate R^{ext} is described a simple Poisson generating function Exp[R^{ext}T(y-1)]. In general one has to use a product of these various generating functions to describe a multiplying system with a spontaneous fission source rate F_s, an (α,n) source rate S, and an external random source rate R^{ext} of neutrons. The ratio $A = S/(\nu_s F_s)$, where ν_s is the average number of neutrons in a spontaneous fission, is referred to as the alpha ratio. One can easily develop empirical models of correlated background source of neutrons within this framework.

Efficient algorithms have been developed to compute the fission chain generating function h(y) for any multiplication $M = 1/(1-p\nu)$. The probabilities P_ν, along with all the other parameters, can be used to quickly generate analytical count distributions in real time. Furthermore, by sampling the analytically computed fission chain distributions we can generate a time-evolving sequence of event counts by spreading the fission chain distribution in time. This allows a real time 0-D Monte Carlo simulation of list mode data as would be acquired with a neutron detector equipped with time tagging electronics. At LLNL we have developed a code, SrcSim, which does precisely this.

An optimization code, BigFit, was developed at LLNL to efficiently compute the count distribution $b_n(T)$ that best fits a measured count distribution. This optimization constitutes an absolute assay of a system by determining its multiplication M, spontaneous fission source Fs, alpha ratio A, and possibly an external random source R^{ext} or correlated background source along with ε and λ. This optimization algorithm has been patented in several U.S. patents (US7756237, US8180013, US8194813, and US9201025) (Ref. 2, 3, 4, 5). BigFit has proven to be a robust assay tool over a wide range of SNM parameter space. Detailed description of BigFit absolute assay methodology and its various applications can be found in Ref. 2-5.

While the count distribution $b_n(T)$ is a complicated function, the moments of the count distribution are quite simple. We define the (k^{th} factorial moments)/k! $\equiv M_k$ of $b_n(T)$ as:

$$M_k(T) = \frac{1}{k!}\sum_{n=0}^{\infty} n(n-1)(n-2)\ldots(n-k+1)b_n(T) \tag{5}$$

The mean of $b_n(T)$ is $M_1(T) = R_1T$ which is the number of counts measured in time T with count rate R_1. The correlated Feynman cumulant moments Y_k are defined by:

$$\sum_{n=0}^{\infty} b_n(T)y^n = Exp[\sum_{k=1}^{\infty} Y_k(T)\,(y-1)^k] \tag{6}$$

For a random source of neutrons, $Y_k = 0$ for $k > 1$. Therefore, any significant non-zero value of Y_k for $k > 1$ is a good measure of how correlated a source of neutrons originating from fission or elsewhere is. The Feynman moments Y_k can be related to the normalized factorial moments M_k and the first three of these relations are:

$$Y_1(T) = M_1(T) = R_1T \tag{7}$$

$$Y_2(T) = M_2(T) - \frac{1}{2}[M_1(T)]^2 \tag{8}$$

$$Y_3(T) = M_3(T) - M_1(T)M_2(T) + \frac{1}{3}[M_1(T)]^3 \tag{9}$$

In the limit of instantaneous fission chains assumed in this section, $Y_2(T)$ and $Y_3(T)$ have simple analytical forms for a spontaneous fission source Fs and/or an (α,n) source S:

$$Y_{2F}(T) \equiv \frac{Y_2(T)}{Y_1(T)} = R_{2F}\left(1 - \frac{1-e^{-\lambda T}}{\lambda T}\right) \tag{10}$$

$$Y_{3F}(T) \equiv \frac{Y_3(T)}{Y_1(T)} = R_{3F}\left(1 - \frac{3+e^{-2\lambda T}-4e^{-\lambda T}}{2\lambda T}\right) \tag{11}$$

Historically, several assay techniques have been based on moments but, in our experience, are not as robust as those based on the full count distributions such as BigFit. However, moments analysis is very useful in constraining and guiding BigFit optimization.

III. Time Interval Distributions and the Rossi Correlation Function

For material spontaneously generating fission chains, the arrival times of neutron and gamma counts create a clustering pattern distinctly different from a random source. A theory for the time interval distribution between adjacent counts was developed in Ref. 6. As well as the distribution of nearest-neighbor counts, we gave the general distributions for all n^{th}-neighbor intervals. The sum of these distributions gives the Rossi correlation function. This theory directly applies to experimentally measured list mode data.

List mode data can be analyzed in a variety of ways. First, we can construct count distributions using random (Feynman) or triggered (shift-register coincidence) time gates T. For random counting we cut up the list mode data into N segments, each of length T, and define the random count distribution $b_k(T)$ = (number of segments with k events)/N. For triggered counting, at each event of the list mode data we open a segment of length T and define the triggered count distribution $n_k(T)$ = (number of segments with k events)/(total number of events). A different way of analyzing the list mode data is to look at the distribution of time intervals between neighboring events. Time interval distributions are especially useful for low count rate data where one would have to otherwise collect data for a very long time to achieve good counting statistics.

In Ref. 6 we derived an exact mathematical relation between the triggered $n_k(T)$ and random $b_k(T)$ count distributions:

$$n_k(T) = \frac{1}{R_1} \frac{d}{dT} \sum_{j=(k+1)}^{\infty} b_j(T) \tag{12}$$

and their (k^{th} factorial moments)/k! ≡ M_k:

$$M_k^n(T) = \frac{1}{R_1} \frac{d}{dT} M_{k+1}^b(T) \tag{13}$$

where R_1 is the count rate. In general, the probability distribution function $I_n(T)$ for the time interval from a trigger count to the n^{th} skipped count is given exactly by:

$$I_n(T) = \frac{d}{dT}\left[1 - \sum_{k=0}^{n} n_k(T)\right] \tag{14}$$

In particular, $I_0(T)$ is the waiting time distribution between succesive counts and is given by:

$$I_0(T) = \frac{1}{R_1} \frac{d^2 b_0(T)}{dT^2} \tag{15}$$

The Rossi correlation function for having a count at time T following a trigger count at time 0 , regardless of the number of intervening counts, is:

$$Rossi(T) = \sum_{n=0}^{\infty} I_n(T) = \frac{1}{R_1} \frac{d^2}{dT^2} M_2^b(T) \tag{16}$$

The formulas just described for counting and time interval distributions are fundamental and true regardless of the underlying model of fission chains. For the special limit of instantaneous fission chains described in Section II, the Rossi distribution above reduces to $[R_1 + R_{2F}\, \lambda\, \mathrm{Exp}(-\lambda T)]$.

A challenging aspect of analyzing list mode data is to visualize it in a useful way. One such way is the waterfall plot which plots the time to the next event versus time as shown in Fig. 1 below from Ref. 6:

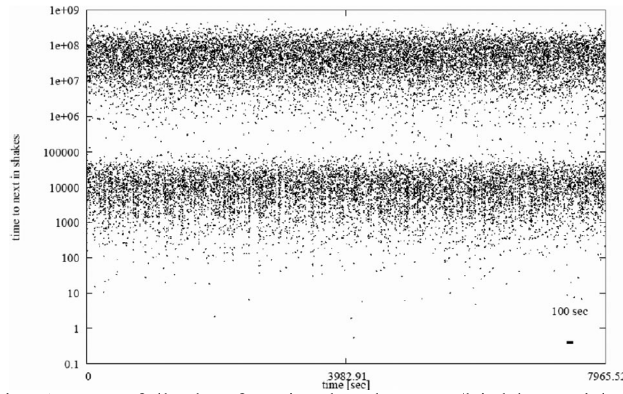


Fig. 1 Waterfall plot for simulated HEU (highly enriched uranium) count data, accumulated over 2.2 hours. The time difference between neighboring counts is plotted at the time of the first count.

In Fig. 1 we clearly see two bands corresponding to two characteristic timescales; the time intervals mostly between 0.1 and 1 second are associated with the source initiation of the fission chains, and the time intervals between 1 and 500 µs are associated with the timescale for neutron diffusion separating counts from a fission chain burst. A time projection of Fig. 1 gives the waiting time distribution $I_0(T)$ shown in Fig. 2 below using logrithmic binning to display the product $T*I_0(T)$ to accentuate the characteristic time scales.

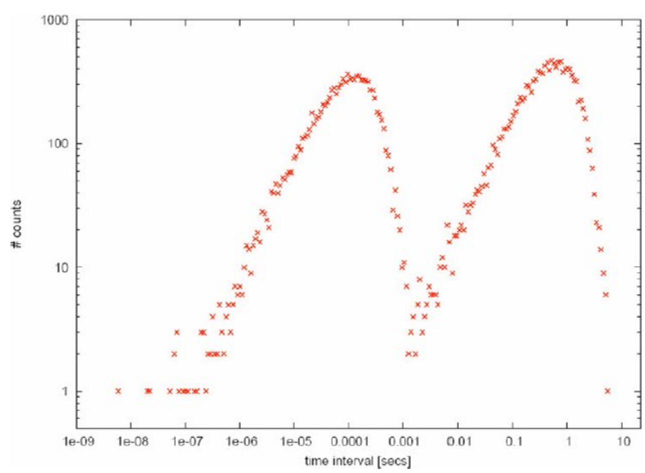


Fig. 2 Plot of $TI_0(T)$ versus time for the waterfall plot of Fig. 1.

Depending on the source rate of what is being measured, a particular way of analyzing the list mode data will be most effective. For low count rate data the time interval distributions, in particular the waiting time distribution $I_0(T)$, are most useful. At LLNL we have developed a processing algorithm to assay low count rate SNM which operates in real time as the list mode data is being collected. To assay low count rate data one must also carefully model background neutrons as described in Ref. 7. For high count rate data the standard random time gate count distributions $b_n(T)$ are most useful because of good counting statistics. For medium count rate data triggered counting distributions may be best.

IV. Time Evolving Fission Chain Theory and Fast Neutron and Gamma-Ray Counting Distributions

In Ref. 9 we solved a simple theoretical model of time evolving fission chains due to Feynman that generalizes and asymptotically approaches the point model theory described in Section II. The point model theory has been used to analyze thermal neutron counting data collected with ^{3}He detectors. This extension of the theory underlies fast counting data for both neutrons and gamma rays from metal systems. Fast neutron and gamma counting is now possible using both liquid and stilbene crystal scintillator arrays with nanosecond time resolution. In Ref. 8 we describe the various benefits of fast counting using a prototype stilbene scintillator detector array. With nanosecond time resolution, it is possible to visually see individual fission chain bursts as the data is being collected. With this detailed time resolution, we can infer more details of the SNM.

List mode data collected by liquid scintillators clearly show the time evolution of fission chains. The data shown below in Fig. 4 are for a Pu ball: (a) 1 millisecond of data of the accumulation of counts including fast neutrons (gold) and gamma rays (red) and (b) the large isolated burst evolving over 50 nanoseconds.:

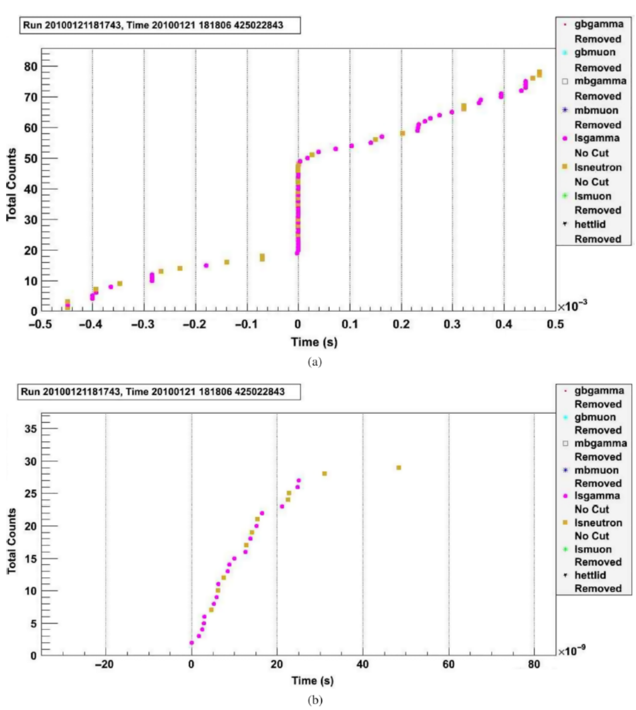


Fig. 4 The accumulation of counts in list mode data taken with a Pu ball and liquid scintillator arrays.

A time evolving fission chain is described by the probability $P_{i,\nu,\gamma}(t)$ that starting from one neutron at t=0, there are i internal neutrons in the multiplying system at time t, ν neutrons that have been created by the fission chain but have escaped from the system, and γ gamma rays that have been created by fissions in the chain. The generating function for this fission chain

$$f(t,x,y,z) = \sum_{i=0}^{\infty}\sum_{\nu=0}^{\infty}\sum_{\gamma=0}^{\infty} P_{i,\nu,\gamma}(t) x^i y^\nu z^\gamma \tag{17}$$

satisfies the Feynman rate equation (Ref. 9):

$$\tau\frac{\partial f}{\partial t} = (-x + qy + pQ(z)C(x))\frac{\partial f}{\partial x} \tag{18}$$

Where Q is the gamma multiplicity generating function for gammas created in an induced fission and τ is the total neutron lifetime. This equation can be easily solved by the method of characteristic. In Ref. 9 we show how to compute the various correlated moments of the time evolving counting distribution. The analogues of the Feynman correlated cumulant moments described in Section II are:

$$Y_1(T) = R_1 T \tag{19}$$

$$Y_{2F}(T) \equiv \frac{Y_2(T)}{Y_1(T)} = R_{2F}\left(1 - \frac{1-e^{-\alpha T}}{\alpha T}\right) \tag{20}$$

$$Y_{3F}(T) \equiv \frac{Y_3(T)}{Y_1(T)} = R_{3F}^{(3)}\left[1 - \frac{3 + e^{-2\alpha T} - 4e^{-\alpha T}}{2\alpha T}\right] + R_{3F}^{(2,2)}\left[(1+e^{-\alpha T}) - \frac{2}{\alpha T}(1-e^{-\alpha T})\right] \tag{21}$$

The formula for $Y_{2F}(T)$ is the same as in Section II except that the inverse neutron lifetime λ is now replaced by the inverse fission chain evolution time α = (1-pν)/τ. The formula for $Y_{3F}(T)$, however, now has an additional time dependent term. The different time dependencies in $Y_{3F}(T)$ are associated with different topologies of common ancestry within the fission chain. The term $R_{3F}^{(3)}$ is the probability that all three counted neutrons have a single common ancestor fission somewhere in the chain while the term $R_{3F}^{(2,2)}$ is the probability that a pair of the counted neutrons have a common ancestor fission, and that fission has a common ancestor with the third counted neutron.

The corresponding random Feynman correlated cumulant moments for gamma counting have been worked out in Ref. 9. The gamma count rate has contributions from both fission chains and from nonfission background. There may be two sources contributing to that background: from external environmental background and from alpha decay chain gamma rays originating in the fissioning material. Both contributions are essentially random (although there are a few cascade decays where multiple gamma rays are emitted).

The analogue $Y_1(T)$ and $Y_2(T)$ for gammas are:

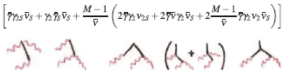
$$\bar{C}_\gamma = \varepsilon_\gamma\left(\bar{\gamma}_S + \bar{\nu}_S\frac{M-1}{\bar{\nu}}\bar{\gamma}\right)F_S T + R_{1\gamma}^{ext}T \tag{22}$$

$$\begin{aligned}\frac{Y_{2\gamma}}{F_s\varepsilon_\gamma^2} = &\left(\gamma_{2S} + \bar{\nu}_S\frac{M-1}{\bar{\nu}}\gamma_2\right)T \\ &+\left[\bar{\nu}_S\bar{\gamma}_S\frac{M-1}{\bar{\nu}}\bar{\gamma} + \bar{\nu}_S\left(\frac{M-1}{\bar{\nu}}\right)^2\bar{\nu}\bar{\gamma}^2\right. \\ &\left.+\nu_{2S}\left(\frac{M-1}{\bar{\nu}}\right)^2\bar{\gamma}^2 + \bar{\nu}_S\left(\frac{M-1}{\bar{\nu}}\right)^3\nu_2\bar{\gamma}^2\right] \\ &\times\left[T - \frac{1}{\alpha}\left(1-e^{-\alpha T}\right)\right].\end{aligned} \tag{23}$$

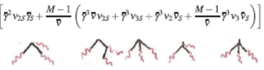
The definitions of the various symbols can be found in Ref. 9. The analogue of $Y_3(T)$ for gammas is complicated but each term can be understood using Feynman diagrams, as described in Ref. 9:

$$\frac{Y_{3\gamma}}{F_s\varepsilon_\gamma^3} = \left(\gamma_{3S} + \frac{M-1}{\bar{\nu}}\gamma_3\bar{\nu}_S\right)T$$

$$+\frac{M-1}{\bar{\nu}}\left[\bar{\gamma}\gamma_{2S}\bar{\nu}_S + \gamma_2\bar{\gamma}_S\bar{\nu}_S + \frac{M-1}{\bar{\nu}}\left(2\bar{\gamma}\gamma_2\nu_{2S} + 2\bar{\gamma}\bar{\nu}\gamma_2\bar{\nu}_S + 2\frac{M-1}{\bar{\nu}}\bar{\gamma}\gamma_2\nu_2\bar{\nu}_S\right)\right]\left[T - \frac{1}{\alpha}\left(1-e^{-\alpha T}\right)\right]$$



$$+\left(\frac{M-1}{\bar{\nu}}\right)^2\left[\bar{\gamma}^2\nu_{2S}\bar{\gamma}_S + \frac{M-1}{\bar{\nu}}\left(\bar{\gamma}^3\bar{\nu}\nu_{2S} + \bar{\gamma}^3\nu_{3S} + \bar{\gamma}^3\nu_2\bar{\nu}_S + \frac{M-1}{\bar{\nu}}\bar{\gamma}^3\nu_3\bar{\nu}_S\right)\right]\left[T - \frac{3-4e^{-\alpha T}+e^{-2\alpha T}}{2\alpha}\right]$$



$$+\left(\frac{M-1}{\bar{\nu}}\right)^2\left[\bar{\gamma}^2\bar{\nu}\bar{\gamma}_S\bar{\nu}_S + \frac{M-1}{\bar{\nu}}\left(\bar{\gamma}^3\bar{\nu}\nu_{2S} + \bar{\gamma}^3\bar{\nu}^2\bar{\nu}_S + \frac{M-1}{\bar{\nu}}\left(2\bar{\gamma}^3\nu_2\nu_{2S} + 3\bar{\gamma}^3\bar{\nu}\nu_2\bar{\nu}_S + 2\frac{M-1}{\bar{\nu}}\bar{\gamma}^3\nu_2^2\bar{\nu}_S\right)\right)\right]\left[T\left(1+e^{-\alpha T}\right) - \frac{2}{\alpha}\left(1-e^{-\alpha T}\right)\right]$$

$$+\left(\frac{M-1}{\bar{\nu}}\right)^3\bar{\gamma}^2\nu_2\bar{\gamma}_S\bar{\nu}_S\left[T\left(1+2e^{-\alpha T}\right) - \frac{5-e^{-2\alpha T}-4e^{-\alpha T}}{2\alpha}\right]$$

(24)

The correlated moment for counting one neutron and one gamma in a time gate T from the same fission chain is:

$$\frac{Y_{1n,1\gamma}}{\varepsilon\varepsilon_\gamma F_S\left(M-\frac{M-1}{\bar{\nu}}\right)} = \left[\bar{\nu}_S\bar{\gamma}_S + 2\nu_{2S}\frac{M-1}{\bar{\nu}}\bar{\gamma} + \bar{\nu}_S\frac{M-1}{\bar{\nu}}\bar{\nu}\bar{\gamma} + 2\bar{\nu}_S\left(\frac{M-1}{\bar{\nu}}\right)^2\bar{\gamma}\nu_2\right]\left(T - \frac{1-e^{-\alpha T}}{\alpha}\right) \tag{25}$$

The correlated moment for counting one (two) neutron and two (one) gammas in a time gate T from the same fission chain is quite complicated and can be found in Ref. 9.

V. Fission Chain Restart Theory

Fast nanosecond time scale neutron and gamma ray counting can be performed with (liquid, stilbene) scintillator arrays. Fission chains in metal evolve over a time scale of tens of nanoseconds. If the metal is surrounded by a moderator, neutrons leaking from the metal can thermalize and diffuse in the moderator. With finite probability, the diffusing neutrons can return to the metal and restart the fast fission chain. The timescale for this restart process is microseconds. A theory describing time evolving fission chains for metal surrounded by a moderator, including this restart process, is presented in Ref. 10. This theory is sufficiently simple for it to be implemented for real time analysis.

The Fig. 6 below shows Neutron (gold x) and γ-ray (maroon +) counts from moderated HEU. The upper panel shows the accumulation of counts in time. The lower panel shows the time intervals between adjacent counts, plotted at the time of the first count of the pair. The color is the particle type of the second count of an adjacent pair of counts. The time intervals within the entire fission chain burst range from nanoseconds (ns) to 10's of microseconds (μs). Fission chains within the HEU metal evolve over only a few 10's of ns. The fast chains in metal are being re-started over the diffusion time scale of 10's of μs. Only fast fission neutrons created in the HEU metal that escape being scattered down below 1 MeV in energy can potentially be counted by the liquid scintillator threshold detectors.

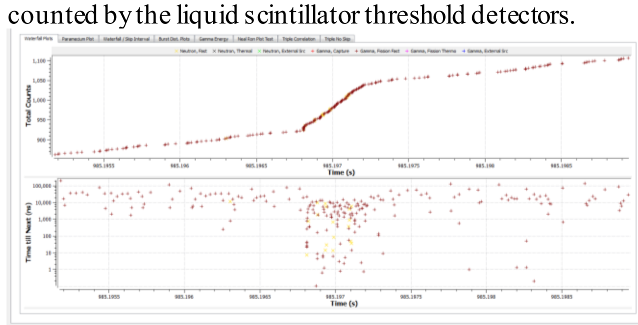


Fig. 6 Accumulated counts (upper panel) and time intervals between adjacent counts (lower panel) of neutrons and gammas.

The rate equation for a time evolving fission chain, including a moderator, is given below in Eq. (27). We consider two neutron populations, fast and thermal. There are two time scales, τ the fast neutron total lifetime and, λ^{-1} the thermal neutron lifetime in the moderator. The generating function for tracking the different populations of neutrons and gammas is defined by:

$$f(t,x,y,z_1,z_2,z_{th},w,u) = \sum_{i,i_{th},n,n_{th},f,f_{th},j_{th}=0}^{\infty} P_{i,n,i_{th},n_{th},f,f_{th},j_{th}}(t)\,x^i y^n z_1^f z_2^{f_{th}} z_{th}^{j_{th}} w^{i_{th}} u^{n_{th}} \tag{26}$$

The generating function variables track the different populations. The variable x tracks the fast internal neutron population in the multiplying SNM. This population of neutrons drives all the others. The variable y tracks those fast neutrons that leak from the system, and u tracks thermal neutrons that leak from the system. The internal thermal population is tracked by w. Fast neutrons that leak from the system with probability q=1-p can get scattered down to thermal in the moderator with probability s. The thermal neutrons in the moderator can leak, with probability $q_{th} = 1-p_{th}$, or induce fission with probability p_{th}. The thermal neutrons that are lost from the system can be lost through leakage, or by capture within the moderator and be tracked by z_{th}. The neutron capture probability in the moderator is generally significant, and can be included by partitioning q_{th} into $q_{th}c$ and $q_{th}(1\text{-}c)$, where c is the probability for neutron capture. The fission gamma-rays created by fast neutron induced fission are tracked by z_1 and the gamma-rays created by thermal neutron induced fission are tracked by z_2. The Feynman rate equation for the probability generating function Eq. (26) is (Ref. 10):

$$\frac{\partial}{\partial t}f(t,x,y,z_1,z_2,z_{th},w,u)=\frac{1}{\tau}\left(-x+q(1-s)y+qsw+pQ(z_1)C(x)\right)\frac{\partial f}{\partial x} + \lambda\left(-w+q_{th}(1-c)u+q_{th}cz_{th}+p_{th}Q^{th}(z_2)C^{th}(x)\right)\frac{\partial f}{\partial w} \tag{27}$$

This rate equation can again be easily solved by the method of characteristics, as discussed in Ref. 10.

The random time gate neutron count distribution can be computed along with its various correlated Feynman cumulant moments. In particular, the Feynman $Y_2(T)$ has a remarkably simple time dependence: it is a superposition of the point model time dependent formula Eq. (10) associated with fast, an effective slow, and time-of-flight time scales. If we neglect the time-of-flight correction, $Y_2(T)$ is a sum of just two terms with separate fast fission chain and total fission chain time dependence:

$$Y_2(T)=R_{2\,fast}\left(T-\frac{1-e^{-\alpha T}}{\alpha}\right)+R_{2\,restart}\left(T-\frac{1-e^{-\lambda_{eff}T}}{\lambda_{eff}}\right) \tag{28}$$

If we include the time-of-flight (TOF) correction, Eq. (27) has an extra point model contribution associated with λ_{TOF}. In the limit $\lambda << \alpha$:

$$\alpha=\frac{1-p\bar{\nu}}{\tau} \tag{29}$$

$$\lambda_{eff}=\lambda\frac{M_0}{M} \tag{30}$$

$$M_0=\frac{1}{1-p\bar{\nu}} \tag{31}$$

$$M=\frac{1}{1-p\bar{\nu}-qsp_{th}\bar{\nu}_{th}} \tag{32}$$

where M_0 is multiplication of the SNM metal and M is the total system multiplication.

A simple limit of this theory can be applied to the analysis of list mode data acquired on a subcritical assembly ISSA at LLNL with bare ^{3}He tubes. The assembly is a series of thin foils of HEU in a tank of water, and is described in Ref. 11. The multiplication of the HEU metal foils $M_0 \approx 1$. The fission chains are driven by thermal neutron induced fission in the HEU. The fast neutrons created thermalize in water and the diffusing neutrons can restart the chain. The thermal neutrons diffuse to either induce fission in the foils or diffuse out of the assembly to possibly be counted by the bare ^{3}He tubes placed outside the water tank. Because of the adjacency of the ^{3}He tubes there is no time of flight corrections.

The analysis is done using the $\tau \rightarrow 0$ limit of the restart theory. For $M_0 \approx 1$ ($p \approx 0$) the total multiplication becomes:

$$M \approx \frac{1}{1-sp_{th}\ \bar{\nu}_{th}} \tag{33}$$

and the time constant for the correlated moments is now

$$\lambda_{eff}{}^{-1}=M\lambda^{-1} \tag{34}$$

a time scale much longer than the diffusion time scale. For counting with bare ^{3}He no fast neutrons are counted.

A segment of the list mode data measured is shown below in Fig. 7:

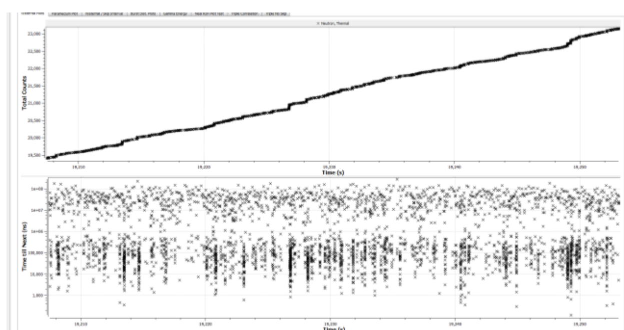


Fig. 7 Segment of experimental list mode data from HEU foils in water. The form of the data is the time of each neutron count in bare ^{3}He tubes, placed outside the assembly. The upper panel is the accumulation of counts in time, and the lower panel is the time difference between neighboring counts, plotted at the time of the first count. The large fission chains are seen as discontinuities in the accumulation of counts and as large streaks in the time interval plot.

The data can be segmented to create count distributions. For this system, making the approximations that the chains are initiated by a random source, and that the multiplication is completely due to thermal neutron induced fission, the formulas for the count distribution moments reduce to the fast fission chain formulas of the previous Section IV with the replacements of nuclear data by thermal values, and:

$$\alpha^{-1} \rightarrow \lambda_{eff}^{-1} \tag{35}$$

$$\varepsilon(M-(M-1)/\bar{\nu}) \rightarrow \varepsilon s q_{th} M. \tag{36}$$

Moments of the count distribution, compared to theory, are shown below.

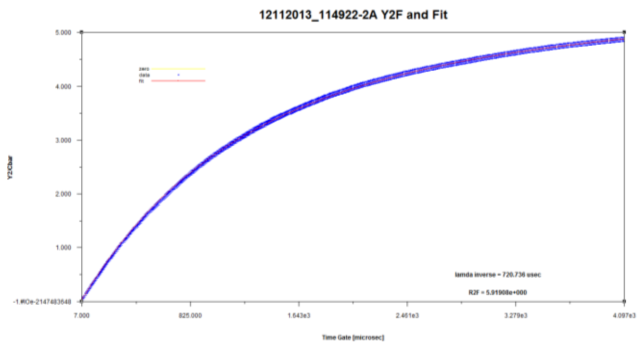


Fig. 8 Feynman random time gate correlated pair moment $Y_{2F}(T)$ versus time gate T. The data is in blue and the fit to the theory is in red.

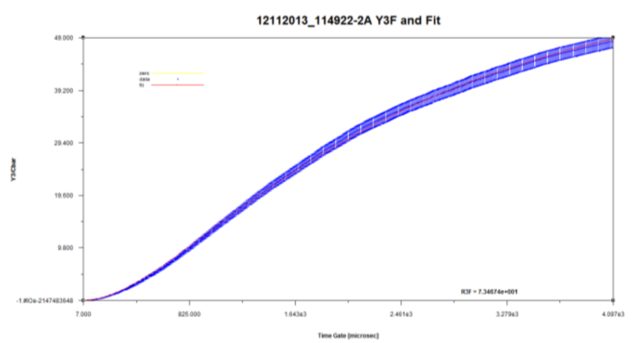


Fig. 9 Feynman random time gate correlated triple moment $Y_{3F}(T)$ versus time gate T. The data is in blue and the fit to the theory is in red.

The data quality is sufficiently good that we can also analyze the fourth Feynman random time gate correlated quad moment $Y_{4F}(T)$:

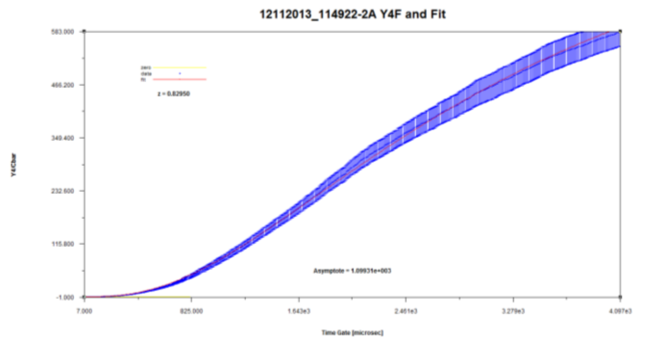


Fig. 10 Feynman random time gate correlated quad moment $Y_{4F}(T)$ versus time gate T. The data is in blue and the fit to the theory is in red.

In Section IV we gave the formulas for $Y_{2F}(T)$ and $Y_{3F}(T)$ and we now give the formula for $Y_{4F}(T)$ being used in the fit shown in Fig. 10:

$$\begin{aligned}
Y_{4F}(T) &= (\varepsilon s q_{th} M)^3 \frac{M-1}{\bar{\nu}_{th}} \nu_4^{th} \left(1 - \frac{11 - 18e^{-\lambda_{eff}T} + 9e^{-2\lambda_{eff}T} - 2e^{-3\lambda_{eff}T}}{6\lambda_{eff}T}\right) \\
&+ 5(\varepsilon s q_{th} M)^3 \left(\frac{M-1}{\bar{\nu}_{th}} \nu_2^{th}\right)^3 \left[1 - \frac{29 - 4(7 + 5\lambda_{eff}T + (\lambda_{eff}T)^2)e^{-\lambda_{eff}T} - e^{-2\lambda_{eff}T}}{10\lambda_{eff}T}\right] \\
&+ 5(\varepsilon s q_{th} M)^3 \left(\frac{M-1}{\bar{\nu}_{th}}\right)^2 \nu_2^{th} \nu_3^{th} \left[1 - \frac{71 - (81 + 42\lambda_{eff}T)e^{-\lambda_{eff}T} + 3(3 + 4\lambda_{eff}T)e^{-2\lambda_{eff}T} + e^{-3\lambda_{eff}T}}{30\lambda_{eff}T}\right]
\end{aligned} \tag{37}$$

At LLNL we have developed optimization algorithms and codes that can separately assay M_0 and M along with all the other model parameters from measurement of moderated systems. The ISSA assembly data just described has M ≈ 20 and λ_{eff}^{-1} ≈ 720 μs.

VI. Approximate Limits of Fission Chain Counting Theory

Analytic formulas were developed for neutron and gamma-ray counting distributions and time interval distributions, in the limit of high and low multiplication. Underlying the counting distributions are models for fission chains, the instantaneous Bohnel chain, and the Feynman time evolving fission chain. The theory of counting distributions is based on the Hage-Cifarelli model of randomly initiated fission chains, and its generalization for time evolving chains. The time evolving fission chain model, studied previously by numerically solving non-linear differential equations, simplifies to analytic forms for both high and low multiplication. These formulas show how fission chain information is packaged in the different counting distributions.

The random time gate count distribution simplifies dramatically in the limit of high M (Ref. 12). For thermal neutron counting with ^{3}He detector (τ→0), in the high M limit, the generating function becomes:

$$\sum_{n=0}^{\infty} b_n(T)y^n \to Exp\left[-R_1 \int_0^T dt \frac{2(1-y)}{1+\sqrt{1+4R_{2F}(1-e^{-\lambda t})(1-y)}}\right] \tag{38}$$

The integral above can be evaluated to give:

$$\sum_{n=0}^{\infty} b_n(T)y^n \to e^{\frac{-2R_1T}{\sqrt{1+4R_{2F}(1-y)}+1}(1-y)} \left[\frac{\sqrt{1+4R_{2F}(1-y)(1-e^{-\lambda T})}+1}{2}\right]^{\frac{R_1\lambda^{-1}}{R_{2F}}}$$

$$\times \left[\left(\frac{\sqrt{1+4R_{2F}(1-y)}+1}{\sqrt{1+4R_{2F}(1-y)}+\sqrt{1+4R_{2F}(1-y)(1-e^{-\lambda T})}}\right)\right]^{\frac{R_1\lambda^{-1}}{R_{2F}}\sqrt{1+4R_{2F}(1-y)}} \tag{39}$$

Because this formula depends only on R1 and R2F, only two parameters can be determined by ^{3}He counting using the point model approximation in the high M limit.

For fast neutron counting, with time evolving fission chains, the count distribution generating function becomes:

$$\begin{aligned}\pi(y,T) &= \sum_{n=0}^{\infty} b_n(T)y^n \\ &= e^{-2R_1(1-y)T/\left(1+\sqrt{1+4R_{2F}(1-y)}\right)} exp\left\{-\frac{R_1\alpha^{-1}}{R_{2F}} log\left[\frac{1}{2}\left(1+\frac{1}{\sqrt{1+4R_{2F}(1-y)}}\right)\right.\right. \\ &\left.\left.+\frac{1}{2}\left(1-\frac{1}{\sqrt{1+4R_{2F}(1-y)}}\right)e^{-\alpha T\sqrt{1+4R_{2F}(1-y)}}+\frac{R_{2F}(1-y)}{\sqrt{1+4R_{2F}(1-y)}}\left(1-e^{-\alpha T\sqrt{1+4R_{2F}(1-y)}}\right)\right]\right\}\end{aligned} \tag{40}$$

This remarkable formula shows that there is a universality limit of the count distribution for high M, analogous to the Gaussian distribution in the central limit theorem. The complete count distribution depends on only three parameters, α^{-1}, R_1, and R_{2F}. The source rate dependence appears only in the form of R_1. All three parameters depend on M. Also α^{-1} is proportional to τ, the total neutron lifetime, which includes only induced fission and neutron leakage as the processes where neutrons can be lost from the multiplying system, $\frac{1}{\tau} = \frac{p}{\tau} + \frac{q}{\tau} = \frac{1}{\tau_f} + \frac{1}{\tau_{leak}}$. Nuclear fission data $\bar{\nu}$ is contained in R_1, and both $\bar{\nu}$ and ν_2 are contained in R_{2F}.

The point model and time evolving chain have different time dependence for the moments, as was already noted in Section IV. In general, for time evolving chains, the different topologies that lead to correlations have separate time dependence. In the diffusion approximation of the point model all the different topologies give the same time dependence.

The full count distribution is more sensitive to the largest chains than the moments. The expansion of fission chains in powers of 1/M can be systematically obtained by summing successively non-leading terms in the moments formulas. We have at LLNL developed these 1/M corrections to the asymptotic formulas which are remarkably accurate from low M to high M (Ref. 12). Because these are analytical formulas they can be computed instantaneously and therefore have direct impact on our suite of analysis codes at LLNL that process list mode data as it is streaming in real time.

VII. CONCLUSIONS

We have described here the neutron and gamma multiplicity modeling efforts at LLNL. These modeling tools underlie the suite of analysis codes at LLNL designed to assay SNM.

ACKNOWLEDGMENTS

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

REFERENCES

1. M. K. PRASAD and N. J. SNYDERMAN, “Statistical Theory of Fission Chains and Generalized Poisson Neutron Counting Distributions,” Nucl. Sci. Eng., 172, 300 (2012); http://dx.doi.org/10.13182/NSE11-86.
2. M. K. PRASAD, N. J. SNYDERMAN, and M. S. ROWLAND, “Absolute Nuclear Material Assay,” US Patent #7756237, Issued July 5, 2010.
3. M. K. PRASAD, N. J. SNYDERMAN, and M. S. ROWLAND, “Absolute Nuclear Material Assay,” US Patent #8180013, Issued May 15, 2012
4. M. K. PRASAD, N. J. SNYDERMAN, and M. S. ROWLAND, “Absolute Nuclear Material Assay Using Count Distribution (LAMBDA) Space,” US Patent #8194813, Issued June 5, 2012.
5. M. K. PRASAD, N. J. SNYDERMAN, and M. S. ROWLAND, “Absolute Nuclear Material Assay Using Count Distribution (LAMBDA) Space,” US Patent #9201025, Issued December 1, 2015.
6. M. PRASAD, N. SNYDERMAN, J. VERBEKE, and R. WURTZ, “Time Interval Distributions and the Rossi Correlation Function,” Nucl. Sci. Eng., 174, 1 (2013); http://dx.doi.org/10.13182/NSE11-87.
7. MANOJ PRASAD, NEAL SNYDERMAN, and SEAN WALSTON, “Neutron Time Interval Distributions with Background Neutrons,” Lawrence Livermore National Laboratory, LLNL-JRNL-678514 (2016) to be published in Nucl. Sci. Eng.
8. MANOJ PRASAD, DAN SHUMAKER, NEAL SNYDERMAN, JEROME VERBEKE, and JAMES WONG, “Prototype Stilbene Neutron Collar,” Lawrence Livermore National Laboratory, LLNL-TR-707598 (2016).
9. K. S. KIM, L. F. NAKAE, M. K. PRASAD, N. J. SNYDERMAN, and J. M. VERBEKE, “Time Evolving Fission Chain Theory and Fast Neutron and Gamma-Ray Counting Distributions,” Nucl. Sci. Eng., 181, 1 (2015); http://dx.doi.org/10.13182/NSE14-120.

10. K. S. KIM, L. F. NAKAE, M. K. PRASAD, N. J. SNYDERMAN, and J. M. VERBEKE, "Fission Chain Restart Theory," Lawrence Livermore National Laboratory, LLNL-JRNL-704257, submitted for publication (2016).
11. D. HEINRICHS, J. BURCH, B. HUDSON, C. PERCHER, and A. WYSONG, "Design, Development, and Utilization of the New LLNL Inherently Safe Subcritical Assembly (ISSA), LLNL-CONF-689348 (2016).
12. M. K. PRASAD and N. J. SNYDERMAN, "Approximate Limits of Fission Chain Counting Theory," Lawrence Livermore National Laboratory, (2016) report in preparation.