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Neutron Correlations in Special Nuclear Materials, Experiments and Simulations

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Summary

Fissile materials emit neutrons with an unmistakable signature that can reveal characteristics of the material. We describe here measurements, simulations, and predicted signals expected and prospects for application of neutron correlation measurement methods to detection of special nuclear materials (SNM).
The occurrence of fission chains in SNM can give rise to this distinctive, measurable time correlation signal. The neutron signals can be analyzed to detect the presence and to infer attributes of the SNM and surrounding materials. For instance, it is possible to infer attributes of an assembly containing a few kilograms of uranium, purely passively, using detectors of modest size in a reasonable time. Neutron signals of three radioactive sources are shown to illustrate the neutron correlation and analysis method. Measurements are compared with Monte Carlo calculations of the authenticated sources.

Basic physics and concept

The key fact about fissile material is that a sufficient quantity of the material can produce chains of fissions, including some very long chains. A chain of fissions will give rise to a detected burst of neutrons with longer chains generally producing larger bursts. These bursts produce distinctive time correlations in a detector near the multiplying material. These correlations are measurable and can be analyzed to infer attributes of the fissile material including fissile material mass, assembly neutron multiplication, characteristic neutron slowing-down time scale and the presence of absorbing or thermalizing material. The correlation signal is very robust with respect to background and to neutron absorbing material.

Fissions, chains and detections

A fission chain is a chain reaction where neutrons induce fissions in atoms such as uranium and plutonium. As these atoms split, they emit several neutrons which in turn induce themselves subsequent fissions and more neutrons. Some of the neutrons produced in the fission chain will induce a detected signal pulse in the nearby detection system, some will be absorbed, while some others will escape the system.
In the case of SNM in the form of metal, the time scale for the fission chain is very short compared to the time scale for neutron slowing down and detection, so that the burst of fissions from a chain is, for all practical purposes, instantaneous. This instantaneous approximation may fail for fissile material dispersed in a thermalizing medium (e.g. uranyl fluoride dissolved in water).
In addition to the neutrons from fission chains there may also be single neutrons coming in from the outside and neutrons from non-fission processes in the assembly (e.g. (α,n) reactions). These neutrons may be detected, contributing a Poisson component, or may

initiate fission chains. Environmental neutrons that are a bothersome background for a neutron multiplicity counter (NMC) are a source of signal for the more sophisticated techniques described here. Some other background sources such as rare cosmic-ray showers can generate a large number of correlated neutrons that are difficult to distinguish from fission chains.

Correlations

The simplest sort of radioactive decay produces a single decay quantum, which is completely independent of all past (or future) quanta. This kind of uncorrelated process is called a Poisson process and the distribution of the number of decays in a fixed time interval follows a Poisson distribution.

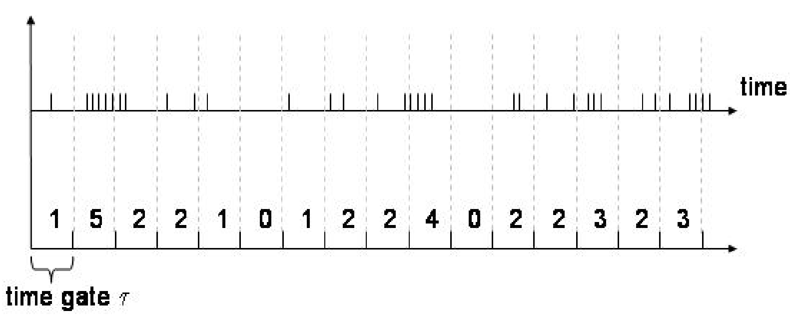
In contrast to a Poisson process a fissile material assembly will produce correlated neutrons. From a detection perspective, instead of each detection event being independent of all past or future events, detections will tend to occur in bunches (see Fig. 1), where several neutrons from a single fission chain will be detected over a short time frame, of the order of the neutron thermalization time scale.

The key feature is that high multiplicity events (many pulses in a short time period) are much more likely for fissile material than for the corresponding Poisson process with the same total pulse rate.

Data analysis

There are several ways to analyze and present correlation results, including correlation functions, moment methods and others. Here we will focus on the discrepancy between measured and expected (Poisson) multiplicity probabilities or counting rates.

The basic form of the data is the time of arrival of neutron counts, and is processed into random time gates imposed on the data stream. A simple cartoon illustrating a correlated stream of pulses is shown in Fig. 1. The table reports the number of times there were ν counts in the time gate τ. Normalized to the number of time gates (e.g. 16), these numbers form the count distribution $C(\tau,\nu)$. With increasing measurement time, this distribution improves statistically.



ν	C(τ,ν)
0	2/16
1	3/16
2	7/16
3	2/16
4	1/16
5	1/16

Figure 1: The number of time tags within a time gate τ is reported for each time interval.

Instead of using a single time gate τ as in Fig. 1, the data stream is analyzed with 512 different time gates, accumulating the number of counts in the first 1 μs, 2 μs, …, and

512 μs, and this is repeated every 512 μs. In fine, 512 count distributions $C(\tau,\nu)$ are computed.
The discrepancy between a random neutron source and a multiplying source is illustrated by the measured count distribution shown in Fig. 5 for time gate 512 μs. This data is from a measurement on a Pu assembly and shows how dramatically different a fissioning system is from what would be expected from a Poisson process with the same total neutron output.
Time-dependent moments of the 512 count distributions are also computed. The first moment $\overline{C}$ determines the average number of counts in that time gate. $\overline{C}$ is only a function of τ. Other moments shown below are proportional to the probability to count more than one neutron from the same fission event within the time gate. The variance of the count distribution $C(\tau,\nu)$ for a fission source is greater than that of a random Poisson distribution of the same count rate: $\left\langle \left(C(\tau,\nu)-\overline{C}\right)^2 \right\rangle = \overline{C} + 2Y_2(\tau)$

The variance of a Poisson distribution is equal to the mean $\overline{C}$. The increased variance, is proportional to $Y_2(\tau)$, which has the physical meaning of the probable number of pairs of counts from the same fission event. Plotted below will be the time dependent ratio $Y_{2F}(\tau) = Y_2(\tau)/\overline{C}$.

Simulation tools/methods

The simulations presented here were done using the Monte Carlo (MC) radiation transport COG (LLNL report UCRL-TM-202590). For use in correlation applications COG has been upgraded. The first upgrade was to the induced fission physics. Prior to the upgrade the data bases for fission physics contained only an average fission neutron multiplicity $\overline{\nu}$. At each induced fission event COG would create an integer number of neutrons randomly alternating between the integer values just above and just below $\overline{\nu}$ to produce the correct average value. Now a new type of data set has been implemented for use with COG which describes the detailed probability for the range of values of ν. The database has been populated with data for ^{239}Pu and ^{235}U using measured data from "Energy Dependence of Neutron Multiplicity P_ν in Fast-Neutron-Induced Fission for ^{235,238}U and ^{239}Pu," M.S. Zucker, N.E. Holden, BNL-38491 (1986), and "A Reevaluation of the Average Prompt Neutron Emission Multiplicity Values from Fission of Uranium and Transuranium Nuclides," N.E. Holden, M.S. Zucker, BNL-NCS-35513.
The correlation simulations depend on special purpose, user-defined radiation source and detector models. With COG a user can create arbitrary source and detector algorithms that can be incorporated into a simulation without rebuilding COG itself. This capability is crucial to our correlation simulations.
We have implemented a spontaneous fission source module that samples the multiplicity and energy distributions of spontaneous fission for ^{236}U, ^{238}U, ^{236}Pu, ^{238}Pu, ^{240}Pu, ^{242}Pu, ^{242}Cm, ^{244}Cm and ^{252}Cf. In order to produce simulated correlation data a special detector module was developed. This module exports data from every ^{3}He(n,p) reactions in the system into a set of files that are suitable for application of the correlation analysis.

Measurements and simulations

To illustrate our capability to identify SNM in storage containers, we show examples of experimental data, and comparison to MC simulations. These examples span a range of possible sources:

a) ^{252}Cf, a non-multiplying, spontaneous fission source. The agreement between the measured and predicted data sets demonstrate that the detector simulation and neutron transport are working properly. The measured data are non-Poisson as expected from a non-multiplying spontaneous fission source with $\bar{\nu}$=3.7.
b) a highly multiplying 4.4 kg ball of weapons grade plutonium moderated by 3" thick polyethylene
c) a bare 22 kg shell of highly enriched uranium (HEU), a very weak neutron source with low multiplication.

^{252}Cf spontaneous fission source

The ^{252}Cf sample, of mass less than .1 μg is a spec, a spontaneous fission source that does not multiply the spontaneously created neutrons. A spontaneous fission, though, can emit no neutrons, or as many as about eight, with a probability distribution. The average number of emitted neutron is about 3.7. The count distribution for the ^{252}Cf experiment is shown in Fig. 2. The increased variance of the measured count distribution compared to a Poisson distribution is indicative of a fission source.

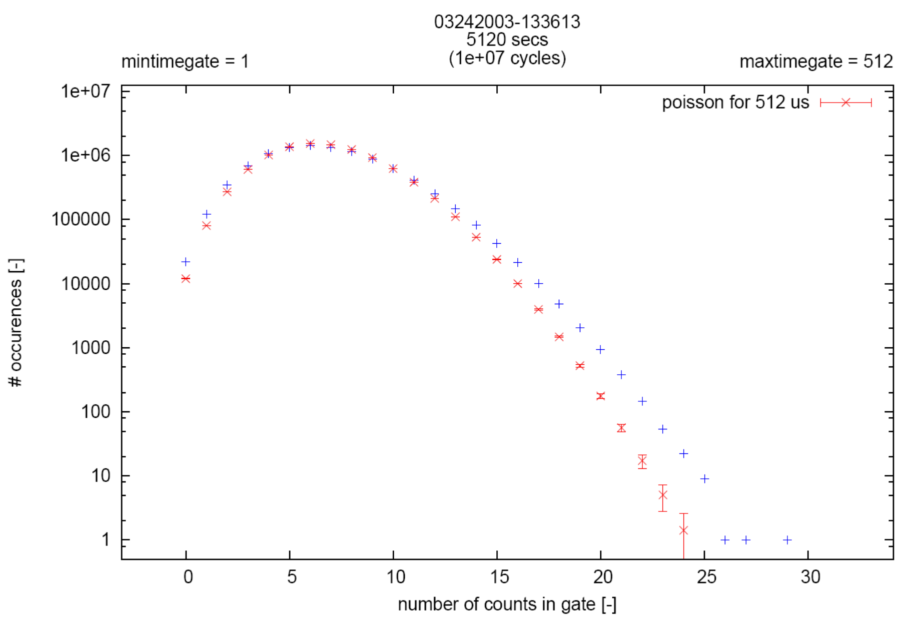


Figure 2 Measured ^{252}Cf count distribution for time gate 512 μs (blue), compared to a Poisson distribution (red) of the same count rate.

The mass of a spontaneous fission source was measured from the ratio of the count rate to the long time gate asymptote of $Y_{2F}(\tau)$, shown in Fig. 4. The proportionality is ($2.34\ 10^6$ / D_{2S}) m_{Cf} [μg], where D_{2S}=1.5 is a parameter determined from the probability distribution that the spontaneous fission creates different numbers of neutrons (number of pairs to the average number). This mass was used for the MC simulation and the resulting count distribution is shown in Fig. 3.

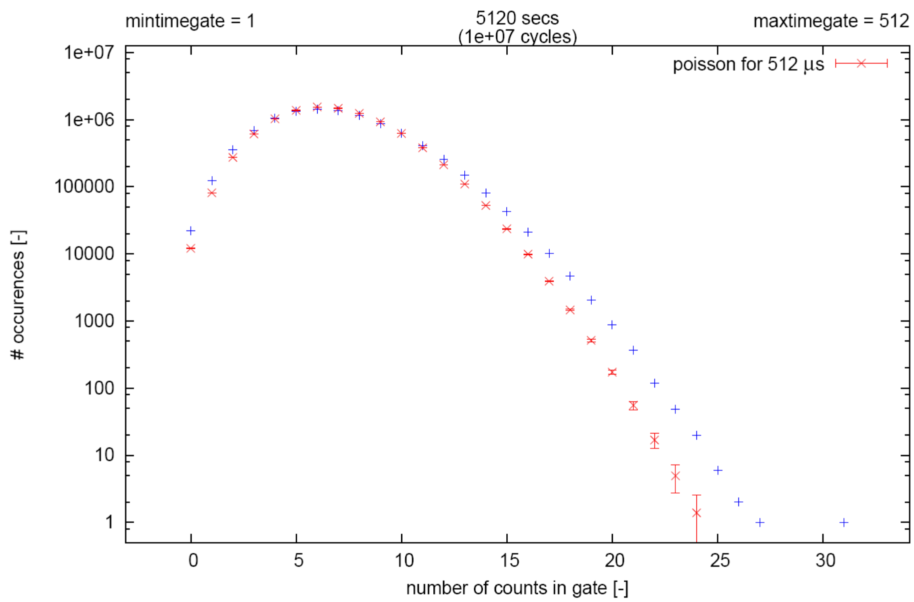


Figure 3 Simulated ^{252}Cf count distribution for time gate 512 µs (blue), compared to a Poisson distribution (red) of the same count rate. This count distribution is to be compared to the experimental distribution of Fig. 2.

The Feynman variance to mean $Y_{2F}(\tau)$ is shown in Fig. 4. $Y_{2F}(\tau)$ would be zero for all time gates τ for a random (non-fissioning) neutron source.

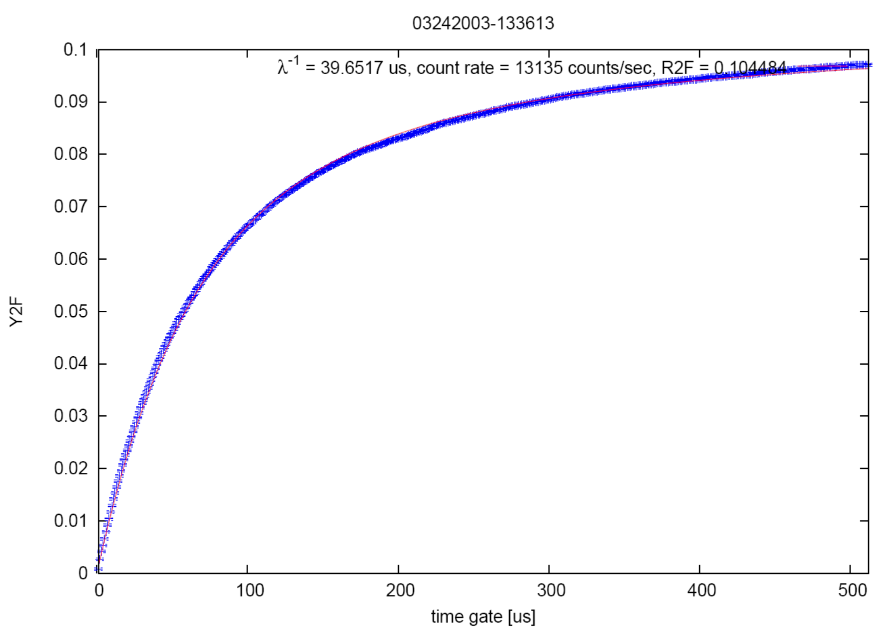


Figure 4 Time dependent Feynman variance to mean for ^{252}Cf spontaneous fission source (blue), determined from 512 experimentally measured count distributions varying from 1 µs to 512 µs. The red curve is a fit using a theoretical formula for the time dependence. The fit determines a diffusion time constant, about 40 µs, and the long time gate asymptote.

The theoretical fit determines a diffusion time constant, here 40 µs. The diffusion process is associated with the detector. The ^3He tubes are embedded in a matrix of polyethylene that thermalizes the neutrons. The ^3He has extremely high efficiency for capturing, and therefore counting neutrons of thermal energies, compared to the high energy with which the neutron is created in fission.

$Y_{2F}(\tau)$ from the simulation is virtually indistinguishable from the measured data. Overall, the simulated data for spontaneous fission of ^{252}Cf compares very well to the measured data.

Moderated plutonium ball

The second example is a highly multiplying 4.4 kg plutonium ball, moderated by 3" thick polyethylene. The spontaneous fission rate for ^{240}Pu (489,000 fissions/sec/kg) is much larger than for other isotopes of U or Pu. This means that it is possible to acquire sufficient correlated data from a Pu-containing source very quickly (a few seconds) compared to a uranium source of similar weight. The count distribution in Fig. 5 is for a 51.2 sec experiment, i.e. 10^5 observations.

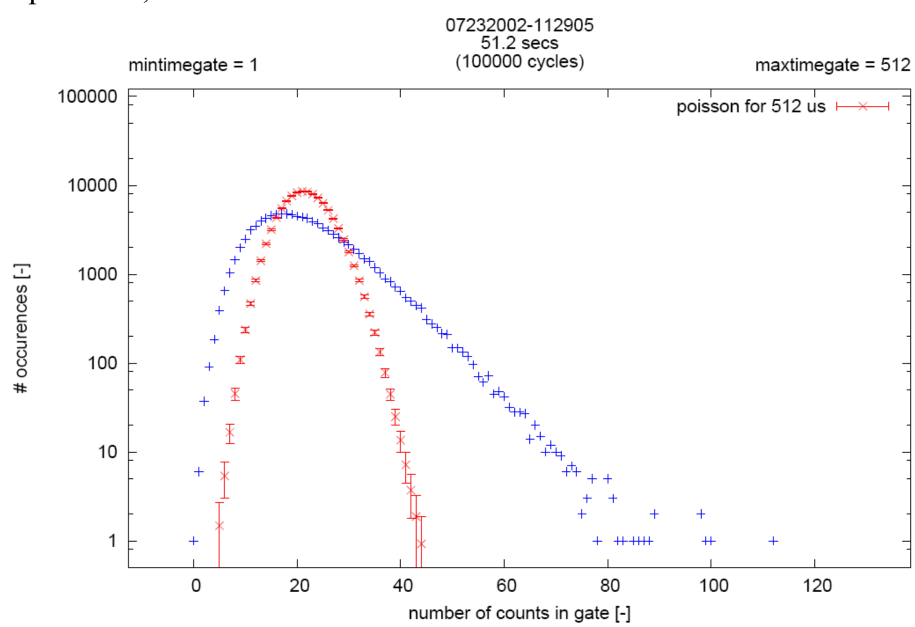


Figure 5: Experimental data (blue) from a moderated Pu ball for the number of occurrences of specific numbers of counts, from zero to 112, within a 512 µs time gate, after 10^5 observations. The red points are from a theoretical Poisson distribution of the same count rate.

It shows a dramatic increase in variance compared to a Poisson distribution. This is because of enormous fluctuations in the number of neutrons created by fission chains, from one chain to the next. This system has multiplication about M = 14. The simulated count distribution in Fig. 6 is statistically essentially the same as the measured data.

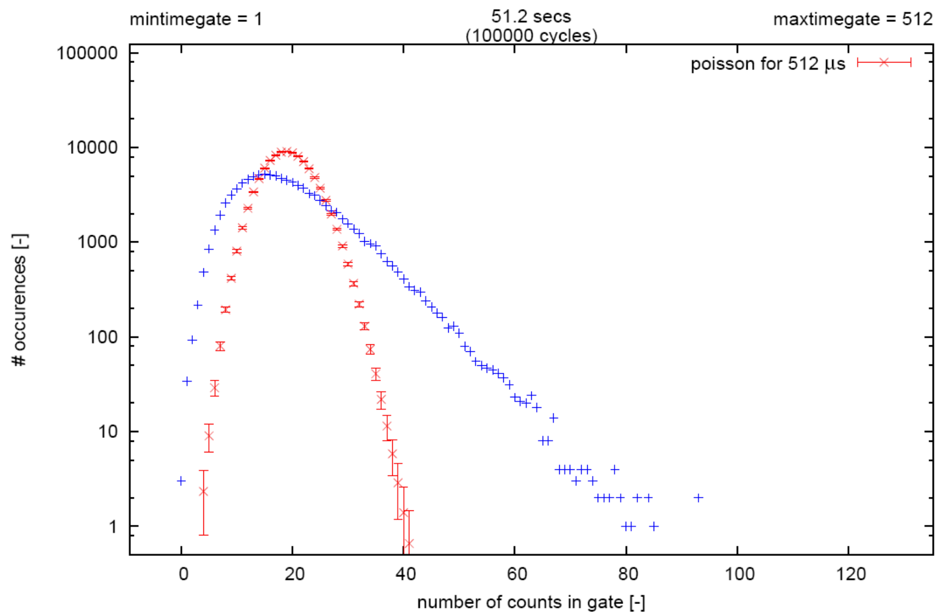


Figure 6 Simulation data (blue) from a moderated Pu ball for the number of occurrences of specific numbers of counts, after 10^5 observations. The red points are from a theoretical Poisson distribution of the same count rate.

Bare highly enriched uranium shell

The third example is passive neutron data from low multiplying HEU. Because of the very low rate of spontaneous fission it was previously thought that there was no significant passive neutron signature of low multiplication HEU. If the multiplication, the average number of neutrons created starting from a single neutron, were indicative of the typical number of neutrons created, then there would be no signal.

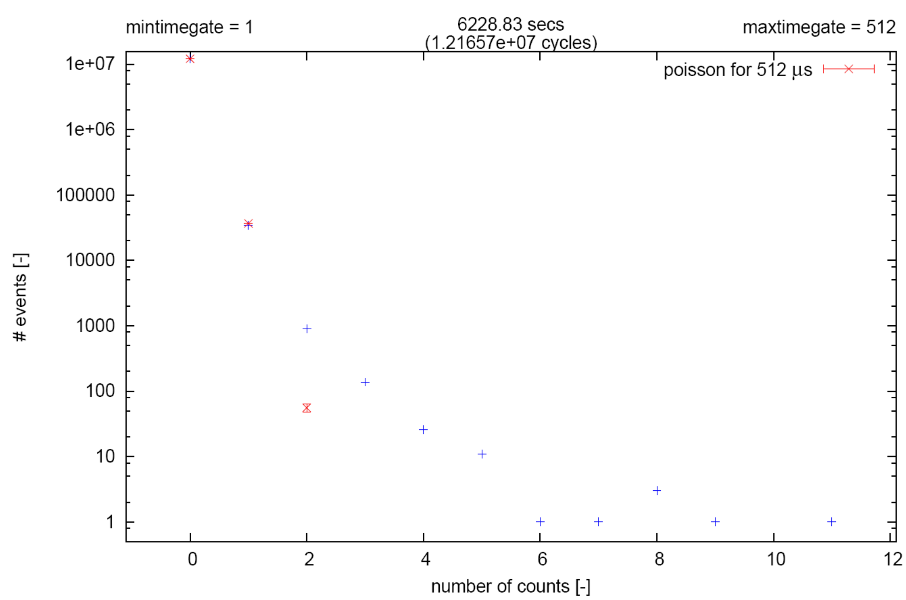


Figure 7 Experimental count distribution from HEU shell for 512 µs time gate (blue), compared to Poisson distribution of the same count rate (red). The count time was about 100 minutes, and the count rate was about 6 counts per second.

The data in Fig. 7 illustrates, however, the fundamental feature of fission chains, that even for low multiplication the number of neutrons created by individual fission chains fluctuates enormously from one chain to the next. The rare very long chains create the detectable signal.

Fig. 8 shows the simulated count distribution for the same HEU shell. The count rate of the simulation is much less than the measured data because the simulation has no cosmic rays built into the model. The count rate from the HEU fission, less than 2 counts per second, is much less than the cosmic ray background count rate, which was measured to be 2 to 4 counts per second for the detectors in the building where the data for Fig. 7 was taken. Nevertheless the correlated signal is mostly from fission, since fission chains in HEU are induced both by spontaneous fission and cosmic rays. The measured count distributions for HEU always show contributions from cosmic ray induced fission chains. The few high multiplicity events in the count distribution are probably due to cosmic ray events interacting with the object.

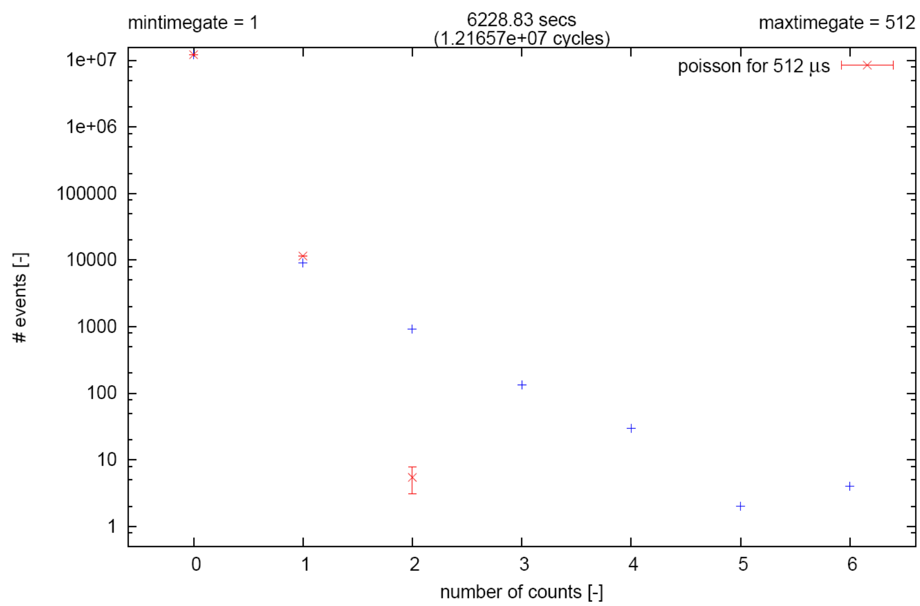


Figure 8 Simulated count distribution of HEU shell for 512 µs time gate (blue), compared to Poisson distribution of the same count rate (red). The count time is the same as the experimental data of Fig. 7, about 100 minutes, and the count rate in the simulation is about 2 counts per second.

Conclusions

Key lessons from these measurements and simulations are that even low multiplying systems with low spontaneous fission activity can be detected passively using neutron correlation techniques, and that simulations can faithfully model the physical features of fissioning systems such as ^{252}Cf, Pu and U sources. For systems with low spontaneous fission activity (e.g. HEU) counting times may be relatively long, on the order of one hour. For an assembly with very low neutron output cosmic ray background is at the origin of most of the detected neutron counts. Based on the results one can judge the capability of simulations to faithfully reproduce as yet unmeasured objects.