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Recent Developments in Fast Neutron Detection and Multiplicity Counting with Verification with Liquid Scintillator

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0. Abstract

For many years at LLNL we have been developing time-correlated neutron detection techniques and algorithms for many applications including Arms Control, Threat Detection and Nuclear Material Assaying.

Many of our techniques have been developed specifically for relatively low efficiency (a few %) inherent in man-portable systems. Historically we used thermal neutron detectors (mainly ^{3}He), taking advantage of the high thermal neutron interaction cross-sections, but more recently we have been investigating the use of fast neutron detection with liquid scintillators, inorganic crystals, and in the near future, pulse-shape discriminating plastics which respond over 1000 times faster (nanoseconds to tens of microseconds) compared to thermal neutron detectors. We have discovered considerable detection advantages with fast neutron detection as the inherent nano-second production time-scales of fission and neutron induced fission are preserved and measured instead of being lost in the neutron thermalization of thermal neutron detectors. We are now applying fast neutron technology (new fast and portable digital electronics as well as new faster and less hazardous scintillator formulations) to the safeguards regime and faster detector response times and neutron momentum sensitivity show promise in measuring, differentiating and assaying samples that have modest to very high count rates as well as mixed fission sources (*e.g.* Cm and Pu). We report on measured results with our existing liquid scintillator array and progress on design of nuclear material assaying system that incorporates fast neutron detection including the surprising result that fast liquid scintillator becomes competitive and even surpasses the precision of ^{3}He counters measuring correlated pairs in modest (kg quantity) samples of plutonium).

1. Introduction

The low natural background rates and the penetrating nature of neutron radiation make neutron detection (and in particular time-correlated neutrons) a good method for quantifying and accounting for large amounts of special nuclear material capable of neutron induced fission and fission chains. Almost from the beginning of the atomic age, the measurement of time-correlated neutrons has been used to detect and quantify fission and fission processes. Feynman himself proposed a method, the grandfather of what we use today, to compare correlation rates of neutron flux in a fixed time to that which would be expected from a random neutron source in order to prove that fission was taking place in early reactor experiments. Fission is one of the few natural processes that produces time-correlated neutrons, the others are spallation-type processes (*e.g.* (n,xn), cosmic induced background, *etc*) which have low but measurable rates in common terrestrial material. The high rates of most transuranic spontaneous fission sources (like plutonium) of even a gram or less usually swamp any cosmic induced background rate but even kilogram quantities of natural uranium produce neutrons only on the same order as that of the cosmic generated background flux. However the primary characteristic of special nuclear material is its ability to fission and to support fission chains through ***neutron*** (and particularly *slow* neutron) induced fission. This means that neutrons produced from fission are not produced randomly (and singly) but rather in time-correlated bursts (the origin of Feynman's method) but even more importantly with the time-scale of the neutron transit time through the fissionable material which can be very long due to the ability of slow neutrons (possibly even thermal neutrons) to create more fast neutrons from induced fission. This means that the bursts of neutrons can be measured by fast neutron detectors as slow-bursts of fast neutrons which is simply not possible with thermal neutron detectors.

2. Measurements with ^{3}He Thermal Neutron Detectors

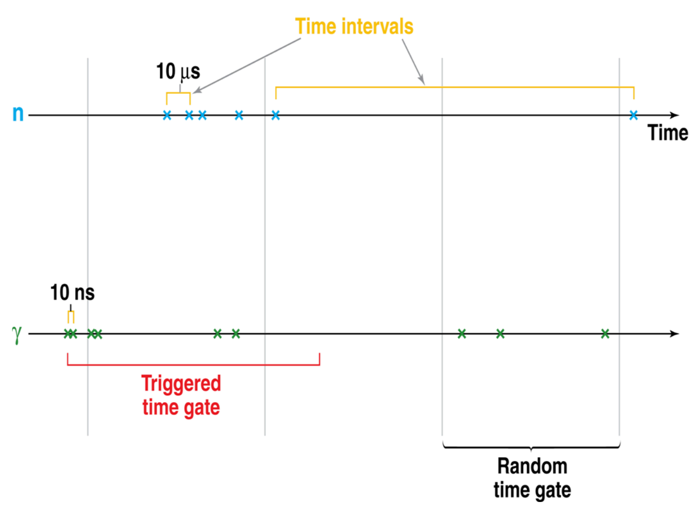


Figure 1. Three different ways to measure a time series 1) The time between arrivals 2) Counts in a fixed time gate triggered by an arrival and 3) Counts in a randomly triggered fixed time gate (all data shown here)

There are many ways to measure time correlations, three examples are illustrated in Figure 1. One can measure the time intervals between arrivals, one can measure of a fixed time interval from a detected neutron or one can count the number of neutrons occurring in a randomly triggered event. We have a long history of developing such methods based on counting detected neutrons in randomly triggered time-gates. Figures 2 and 3 are data from portable ^{3}He thermal neutron detector that can detect and assay

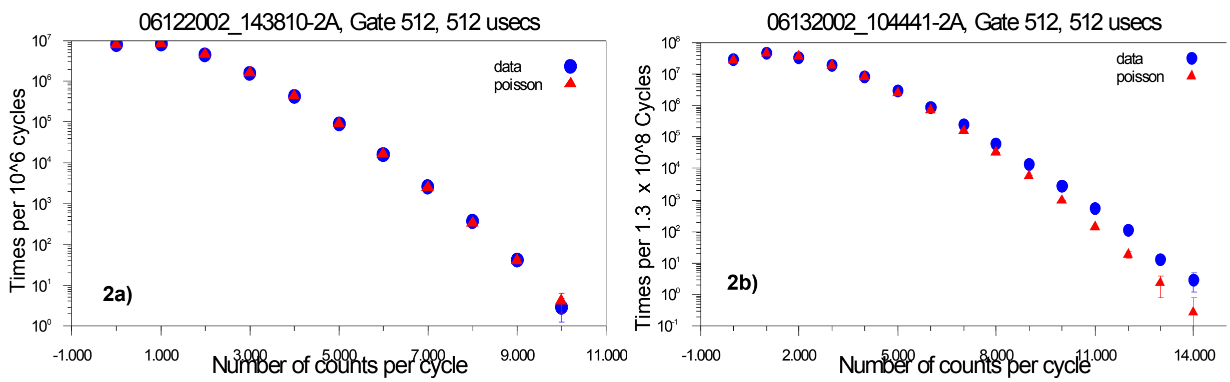


Figure 2. Data from two sources 2a) an AmBe source (2.1K cts/s) and 2b) a ^{252}Cf source (3.1K cts/s 3% efficiency). The Data are the number of neutrons detected (x axis) in 0.512 ms vs. log number of times (Y axis). Wider Variance of data from Poisson distribution of same count rate is indication of Fission

fissioning sources even though it is only a 1-3% efficient. The data shown are from a single time gate of 512 microseconds. In practice we vary the time gate from 1-512 microseconds and compare all the data to a theoretical model to find a best solution. Figure 2 shows how comparing a measured count distribution from a random (AmBe -alpha-n) source can be easily visually distinguished from a spontaneous fission (^{252}Cf) simply by comparing the measured distribution (blue) from that expected from a random source of the same count rate (red). In the case of the fissioning ^{252}Cf source the variance of the distribution is significantly wider than that expected if the source were purely random. This difference is accentuated in the case of multiplying objects such as a highly multiplying plutonium ball shown in Figure 3a. This also works even for weak sources like the modestly multiplying uranium shown in Figure 3b. In this case the actually count rate is very low (only a couple net counts/second on a background of 4 counts/second) but those few originating in the uranium are highly correlated.

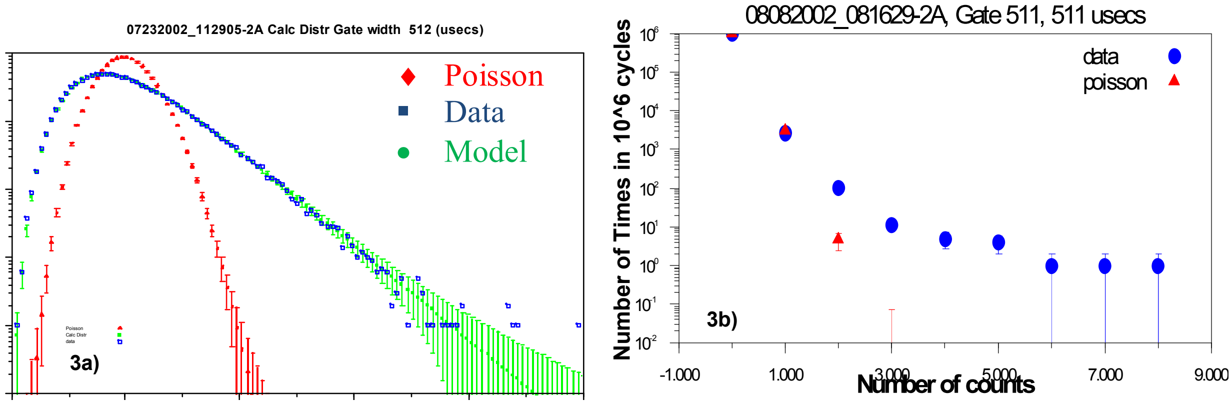


Figure 3. Data from 3a) a strong source, plutonium ball (52s data, 42.5K cts/s, M=12.5, efficiency 2%) source (2.1K cts/s) and 3b) a weak source source uranium shell (500s data, 2.1 net cts/s M=2.5). Data is still highly correlated.

In any case methods that we have been perfecting also allow assaying of the material. We do this by predicting the distributions (e.g. in the above cases from 1-512 microseconds) we would see given a source-strength, a multiplication, an alpha ratio, a detection efficiency and a time of correlation (for bare objects measured with a thermal neutron detector this will be the detector thermalization time) and finding the best fit of the data to our predictions. In cases where only the neutron source may be measured directly, the neutron source strength can be then combined with material isotopics gathered from other means (such as gamma ray spectroscopy) to determine a total mass. The data required for a few percent assay can be obtained in a minute or less for strong significantly multiplying sources (like the plutonium source shown in Figure 8 to perhaps a hour or more in the case of a weak source (like uranium) or a completely non-multiplying source like ^{252}Cf.

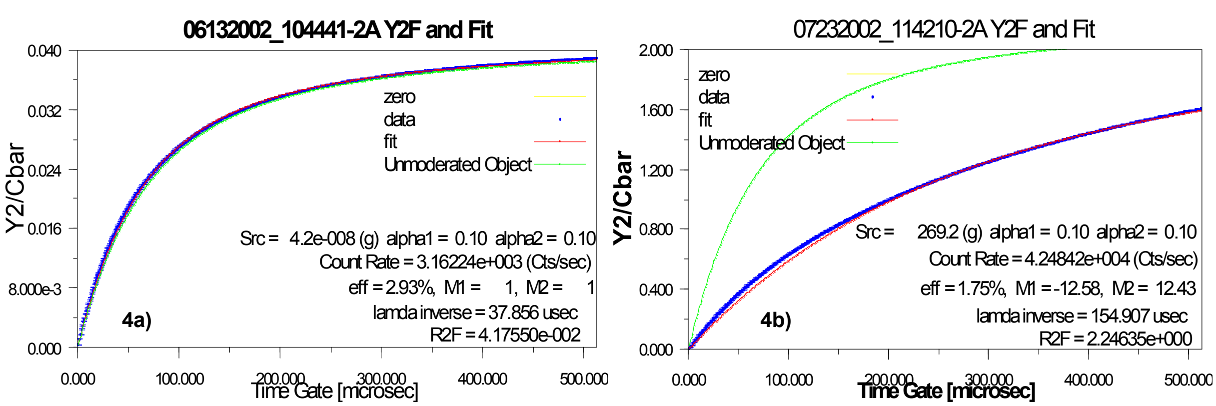


Figure 4. Second Moment Y2 vs Gate width 1-512 microseconds for 4a) unmoderated ^{252}Cf and 4b) heavily moderated plutonium ball.

We also find it useful to plot the moments as a function of the varying time gate width. The evolution of the normalized moments vs. the gate width can give indications of moderation which is illustrated in Figure 4) with data from an unmoderated ^{252}Cf source and a heavily moderated plutonium ball. The green curve is a prediction for the evolution with the known detector time constant (~40 microseconds). The match with the ^{252}Cf data in Figure 4a confirms little or no moderation and the much slower approach to its asymptotic value is an indication of heavy moderation in Figure 4b.

3. Measurements and Assaying with Fast Liquid Scintillation Detector Array

Recently we have begun to apply our neutron analysis and assaying techniques to fast neutron detection with liquid scintillators. In Figure 5 we have configured an array of liquid scintillators so they physically cover about 2π of the solid angle off a cylindrical chamber at the center of the array. Fast neutron detection relies on the recoil of a charged particle (most likely a proton) from collision with a neutron. The clear disadvantages of fast neutron detection are 1. the lower neutron scattering cross-sections (on the order of a barn at best) 2. the minimal neutron energy required for a recoiling proton to be seen (around MeV) and 3. the added difficulty of accurately identifying a neutron recoil proton from a gamma ray. Fast neutron detectors cannot possibly be as efficient as the most efficient thermal detectors simply because of the minimum energy threshold of detection for fast neutron detectors and correspondingly they can be nearly as efficient per unit weight. Efficiency is of course a very important factor when considering measuring correlated events as the probability of detecting n neutrons goes as the nth power of efficiency.

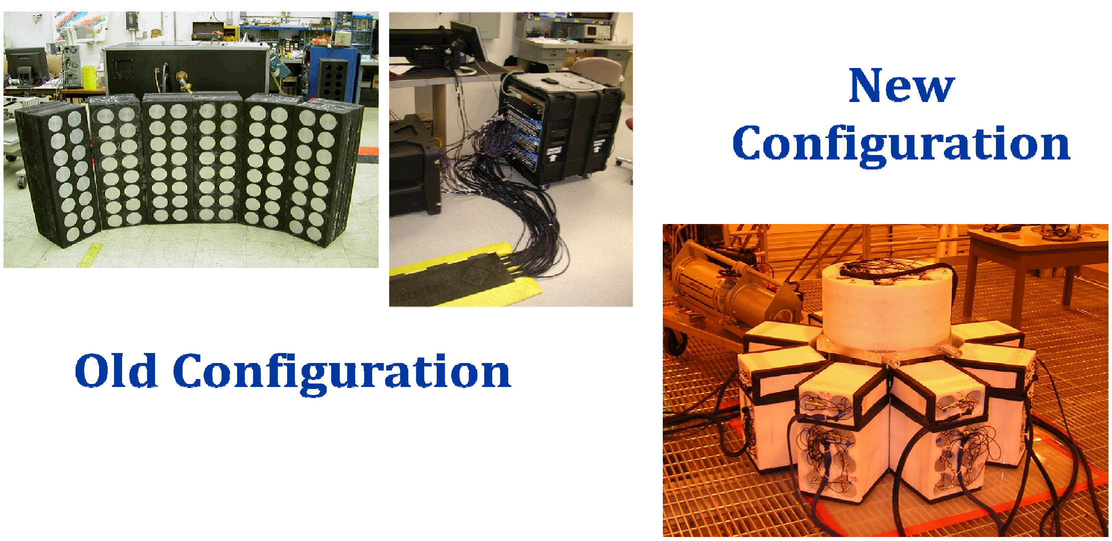


Figure 5. Liquid Scintillator Arrays 5a) older configuration and 5b) newer more efficient (approximately 2π solid angle) configuration.

However there are several very important advantages to fast neutron detection which can be paramount, especially in areas where thermal neutron detection has broken down, as in high flux. In high fluxes the probability of random correlations increases geometrically which makes the ability to detect fission correlations increasingly difficult. Consider the measurement of ^{252}Cf in the Figure 2b. ^{252}Cf only rarely fissions with a neutron multiplicity greater than 8 and yet with a detector with 3% efficiency the detector saw a non-negligible amount of times when more than 10 neutrons were detected in the detector. This clearly means that with a modest source flux of a 100,000 neutrons/second (detecting 3,000 neutrons/second) there is a significant amount of overlapping of fission events within the gate window shown in figure 2b). The single most important characteristic of fast neu tron detection is that it happens fast. Fast neutron detection allows the relevant detection time to shrink from ten's of microseconds (detector thermalization time) to nanoseconds which equivalent to reducing the effect flux by a factor of 10,000.

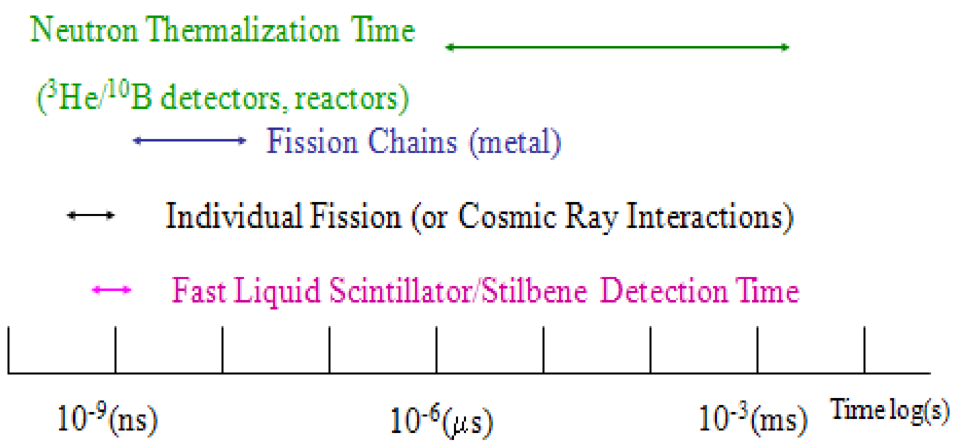


Figure 6. Illustration of timescales Fission and Fission chains occurs at a much shorter time scale than thermal neutron detector can detect. Fast Liquid and crystal scintillators are fast enough to discriminate time scales of individual fission from fiss ion chains

Secondly the fast detection preserves the timescale of the original neutron production. The prompt production of fission neutrons (from a single fission) and spallation-type processes occurs on a nanosecond timescale while the neutron produ ction of a multiplying body occurs in the neutron transit time which can be 10's of nanoseconds for pure metallic systems and up to many microseconds for moderated systems (Figure 6). Thermal neutron detection (e.g. ^3He, BF_3 etc.) requires moderation of the neutrons for efficient detection and this occurs on a ten's of microsecond timescale which smears all the timescale details of the original neutron production.

Finally fast neutron detection also preserves some of the o riginal neutron energy spectral information since it works on recoil energy. Nearly all neutrons detected by thermal detection are by definition thermal neutrons so almost all initial energy information is ultimately lost. Also more subtle especially when combined with the introduction of in particular low energy (below detection threshold) neutrons measuring the *change* in neutron flux can be most illuminating with respect to the source of neutrons in a sample.

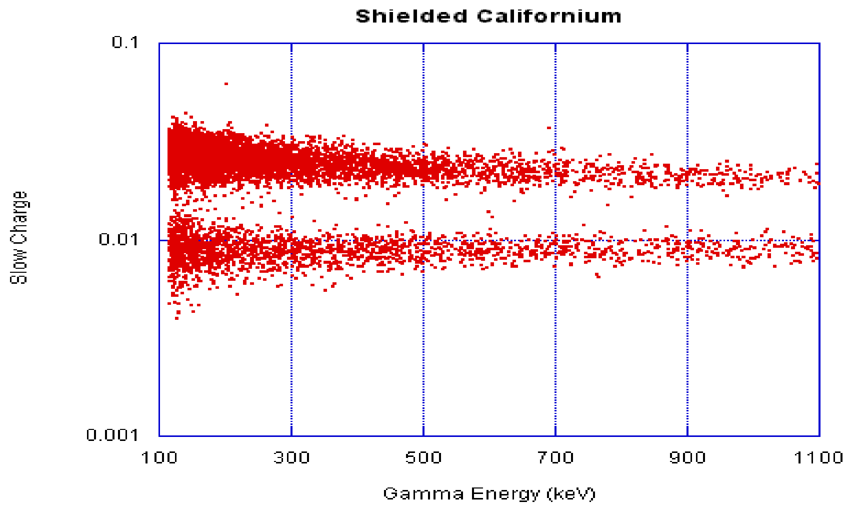
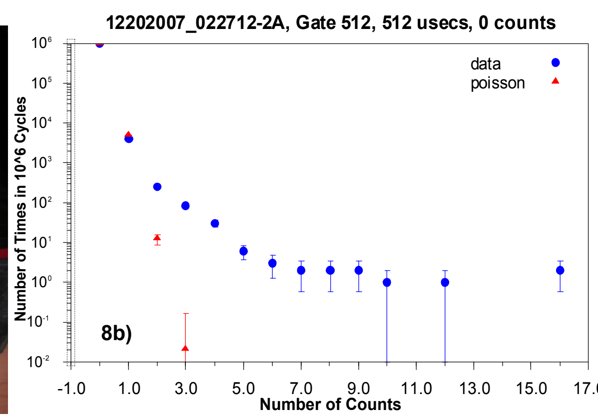


Figure 7. illustrates the now very clean ability to discriminate neutron events from gamma rays due to the advent of modern fast digital electronics. Photon rejection can be done nearly perfectly if fully digitization of a pulse is employed as the normal confusion of a later arriving gamma ray can actually be seen and rejected.

The last figures are meant to illustrate the power of fast neutron detection system with data taken with the existing liquid scintillator array. In Figure 8 an approximately 1 ton pile of lead bricks was measured with thermal neutron detector approximately 4% efficient. Very large neutron correlations were seen at the in the 10's of microsecond time scale. Compare this to Figure 3b of a multiplying uranium system and one would be very hard pressed to tell the difference. In fact assuming the pile of lead were a multiplying uranium system one could get neutron distribution which looked quite close.



8a)

Figure 8. Thermal Neutron Data taken on a ~1 Ton Pile of Lead (Compare to Figure 3b).

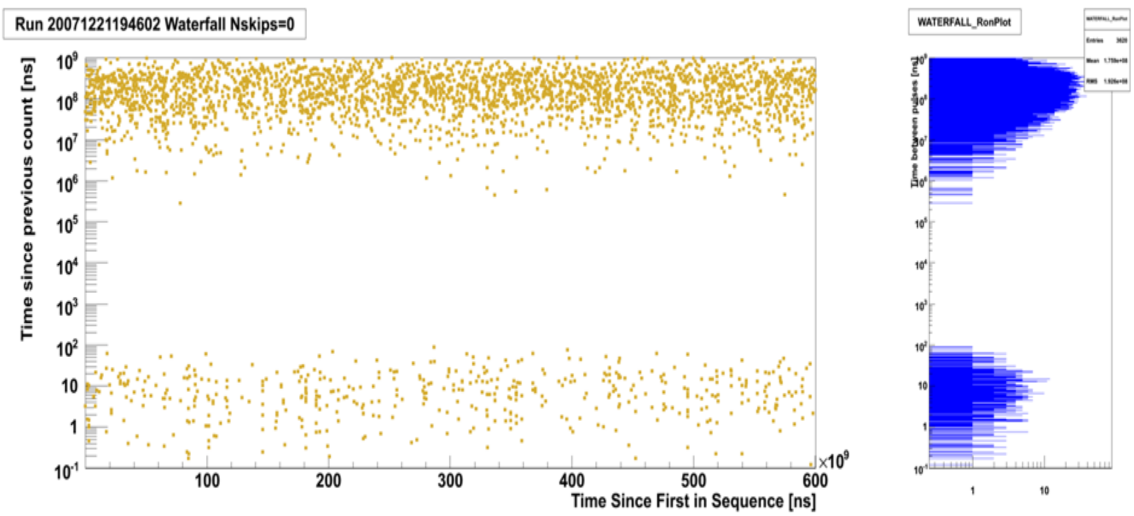


Figure 9. Fast Liquid Scintillator Neutron Data taken on ~1 Ton Pile of Lead (old configuration). 600s of data plotted neutron arrival time (x axis) vs. log (time) to next arrival (y axis). Note the absence of correlations in the 40 nanosecond to 1 microsecond time scale.

We measured the pile of lead with our 1-2% efficient liquid scintillator array (old configuration in Figure 5) in Figure 9. Here the y-axis is the log of the time interval between neutron counts (in nanoseconds and each tick is a power of nanoseconds) and the x-axis is linear running time (for 600 seconds). What you see is the top wide band which happens on the 10 microsecond to second time scale which is the average count rate (6 counts per second). The lower band is fast time correlations which are generated by cosmic interactions with the lead pile. They occur and are over in less than 10 or 20 nanoseconds. There are no time correlations occurring in the time scale between a few tens of nanoseconds and one microsecond.

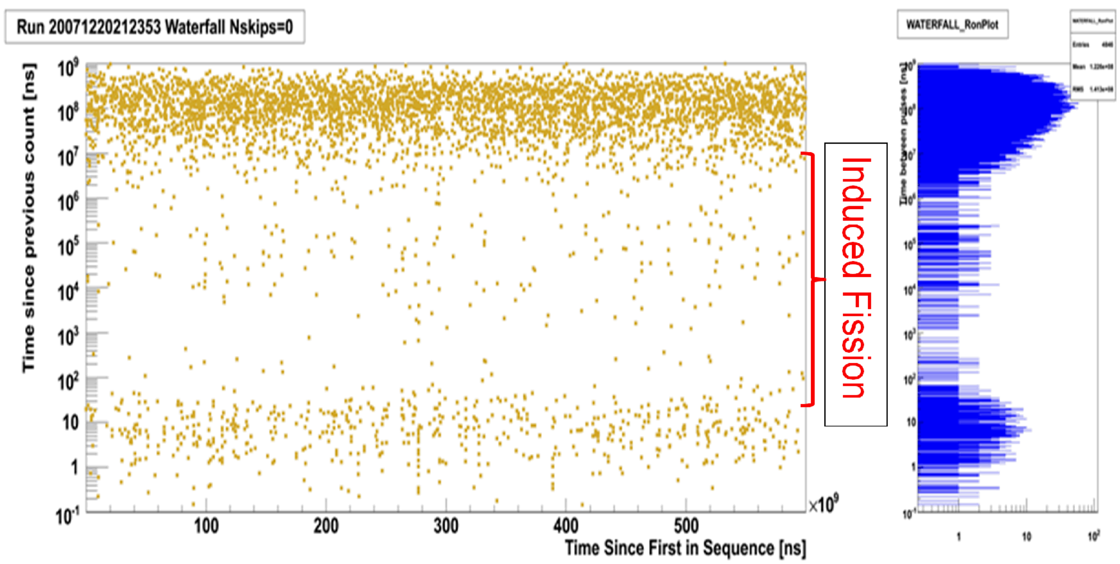


Figure 10. Fast Liquid Scintillator Neutron Data taken on ~1 Ton Pile of Lead (old configuration) plus hidden multiplying Uranium. Note the correlations in the 40 ns to 1 microsecond time scale this is a clear indication of the presence of induced fission.

Figure 10 shows data from the same pile of lead with some multiplying uranium hidden inside. You would see no gamma ray signatures and the thermal detector would look no more correlated than the data in Figure 8, but the measurement reveals time correlations of *fast neutrons* occurring in the intermediate time scale where nothing occurred before. This is a clear indication of the presence of nuclear material as this intermediate time scale can only occur because of slower neutrons inducing fission in the uranium and producing more fast neutrons which cannot happen in lead alone.

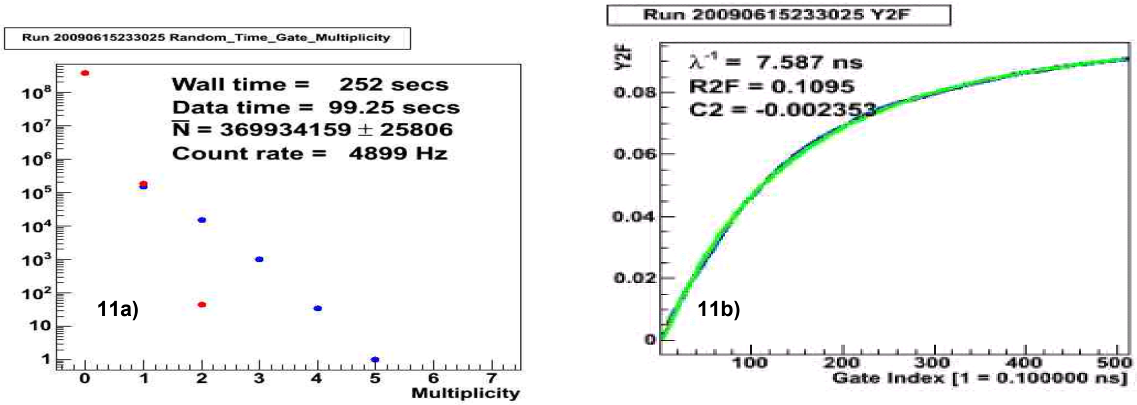


Figure 11. Fast liquid scintillator neutron data taken on ^{252}Cf source 99.25 s data with a 511 ns gate (4.9K cts/s ~6% efficiency). Note the clean separation from Poisson distribution with same count rate because of short time gate possible from fast timing.

The last two figures are demonstrations of assaying done with the liquid scintillators on real objects and the advantage of the fast timing in reducing the random correlations. In Figure 11, a ^{252}Cf source was measured in the new configuration liquid scintillator array that is about 6% efficient (note this overall true efficiency, ~6% of the neutrons emitted from the source were detected). Even with a higher count rate than was measured in the thermal neutron measurement of Figure 2b), there is clearly very little random correlations and with 4 minutes of real time (100 seconds of data) as assay was easily accomplished compared to the 8.5 hours of data shown in Figure 2b).

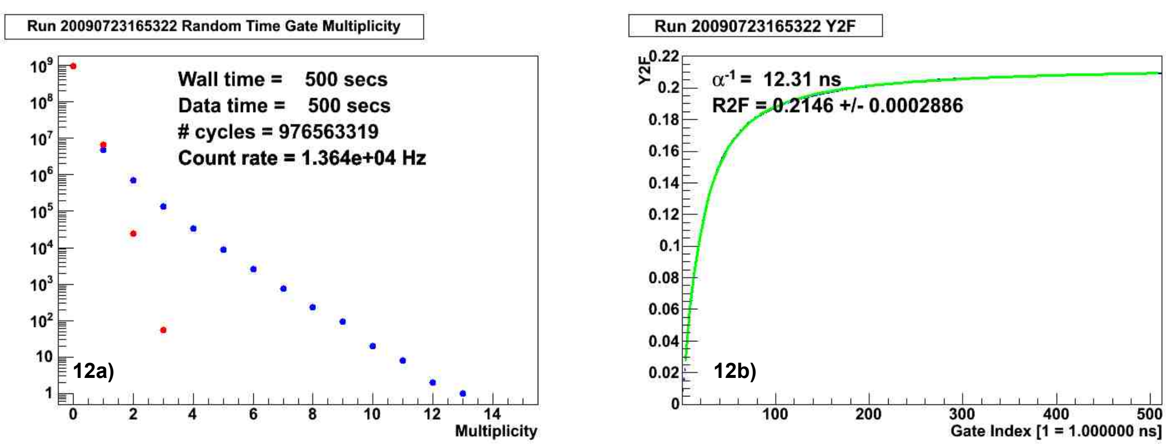


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Figure 12. Fast liquid scintillator neutron data taken on plutonium source 500s data with a 511 ns gate (13.4K cts/s ~5% efficiency). Note the clean separation from Poisson distribution with same count rate because of short time gate possible from fast timing.

This also extends to plutonium systems. A small multiplying plutonium ball was measured in Figure 12 and an assay good to a few percent accuracy was completed with 5 minutes of data. It is also important to note that the time constants seen in Figure 11b of 7ns in the ^{252}Cf source (a non-multiplying fission source) and the 12ns seen in Figure 11b for the metal plutonium source. This implies that the measured timescales of a system may be the most significant detectable difference between a system with significant material able to support induced fission from slower neutrons and those without.

4. Use of Liquid Scintillators in Safeguards

The much lower random correlation rates in the faster (shorter time bin) liquid scintillator vs. thermal ^{3}He is seen in both the ^{252}Cf data and the Pu data of figures 2 and 3 vs figures 11 and 12. This lower rate of random correlations, strictly because of the shortened time bins required, has a profound implication for safeguards measurements in a world without ^{3}He detectors. In safeguards most measurements take advantage of the net neutron pairs coincidences (employing shift-register logic) to "measure" the net amount of spontaneously fissioning material to verify quantities present, essentially ensuring that the net amount of neutron pair correlations present matches the amount expected, given the neutron efficiency of the detector system and the amount of material present. This is accomplished by measuring the amount of correlations seen in an optimized time gate (usually 1.2 times the time constant of the detector) triggered by a neutron detected, and then subtracting off the amount of correlations seen a long time later in order to correct for the random neutron occurrences. This is a very robust way to measure as long as the multiplication of the system and efficiency of the detector is known and the overall count rate is low enough so the random correlations are small compared to the true correlations. When the random rates become comparable to the true signal then the subtraction cancels out the significance of the measurement. So even though the ^{3}He based detectors can be 5-10 times more efficient than fast liquid scintillator detectors, there comes a count rate where the inherently shorter time gates of the faster time-scale correlations measured in a liquid scintillator detector begin to win in precision. This cross-over point is of course source and detector dependent (depending on how significant the source correlations actually are). For our current array vs. a 50% efficient well counter, this occurs at about 1e6 neutrons/sec for ^{252}Cf and 1e5 neutrons per second for ^{240}Pu for pure samples (Figure 13). Crossovers are lower when in oxide form and including the amount of alpha-n random neutrons which degrades the significance of the correlated spontaneous fission sources.

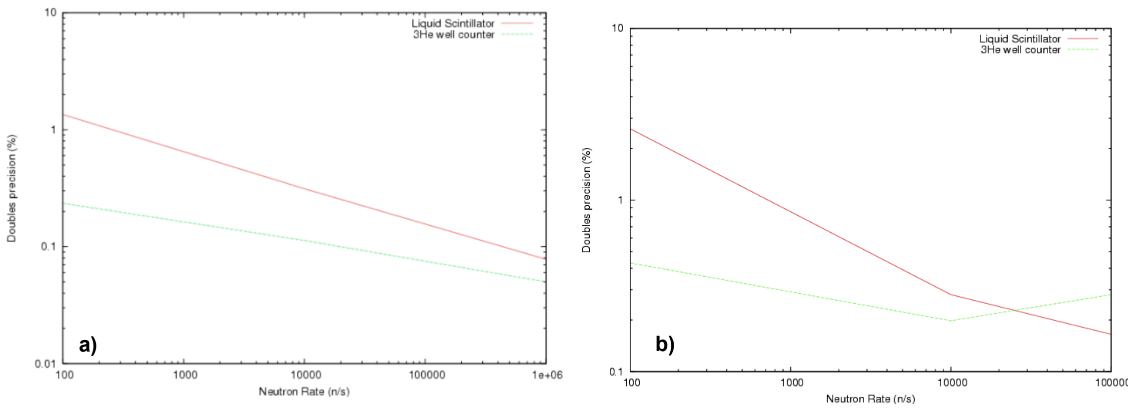

Figure 13. Doubles coincidence precision as a function of neutron rate for a) measured ^{252}Cf sources and b) simulated ^{240}Pu. Note that, even though helium-3 is 5 to 10 times as efficient as the liquid scintillator, at high rates the precision of the liquid scintillator is better than the helium-3 for ^{240}Pu above 1e4 n/s, and is approaching that of the helium-3 for the ^{252}Cf at 1e6 n/s.

5. Imaging with Fast Neutrons and Gamma Rays

Another very interesting characteristic of fast detection is to exploit the joint production of gamma rays and neutrons from the same fission and use the difference in arrival to times to "image" the fissioning source. This is possible because the inherent time scale of the detector system is nanoseconds and so it is possible to separate an individual fission from the following induced fission. Figure 14 illustrates this concept from data. Seeing how it is easy to separate individual fissions from the following induced fission and separate fission-chain bursts from each other with nanosecond timing the following simple algorithm follows easily. We use a pair of fast time-correlated neutrons (within 10ns) to tag a single fission event (as there is almost no background for this) and we look for a preceding minimal MeV gamma ray. We take the time difference between the gamma ray and the neutron arrival at the speed of the measured neutron energy deposited in the detector and translate that into distance from the detector. Collect a large number of such events and image the source. This is seen in Figures 15-17. We assume that the gamma ray is infinitely fast (compared to the neutron) and our biggest error is actually our assumption that the measured energy in the detector was the neutron energy (it is of course the true minimum energy of the neutron as it is due to a proton recoil in the scintillating material). Even so we get reasonable images with on the order of cm resolution. With Pu sources these images can be made in seconds.

It should also be pointed out that these images can be made with neutron interrogation sources as well as from intrinsic neutron sources. The neutron source can be pulsed, steady state, or our preferred way with a low –energy neutrons source (in which case the source is invisible to our fast neutron detectors).

The additional neutron flux will speed up the measurement times for neutron poor systems but the ability to see fission and more importantly induced fission from events will allow evaluation of the quantity of fissionable nuclear material present by the change of the system from purely passive to interrogated, regardless of the strength of the intrinsic source.

We also believe that spatial resolution can be greatly improved by considering events where the neutrons multiply scatter in the detector and we can then use geometry as well as deposited energy to determine the true neutron energy and reduce our largest error. This will likely lengthen the required measurement time considerably but may give more precise positional resolution.

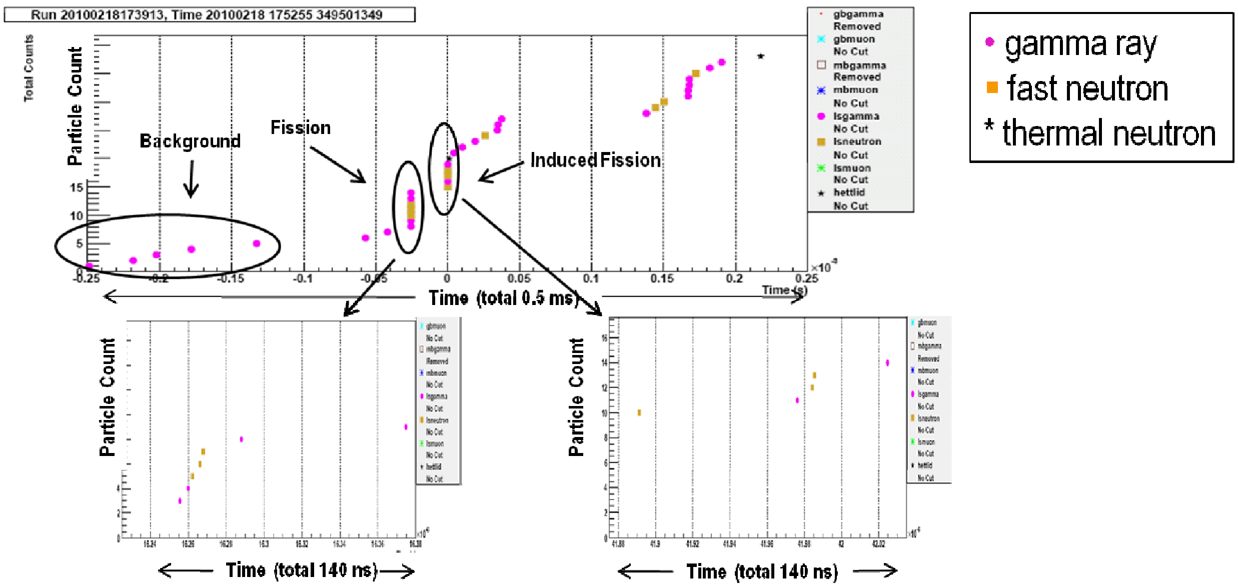

Figure 14. Fast liquid scintillator neutron data taken on plutonium source. The upper figure is 0.5 ms of data in linear running time (X-axis, Y-axis just the order of particles detected). Two Fission Bursts are easily seen separated from each other and background. Blowing up th e each fission burst in time one can see the separation of individual fissions and following induced fissions. The gamma rays closely preceding the neutrons are from the same fission.

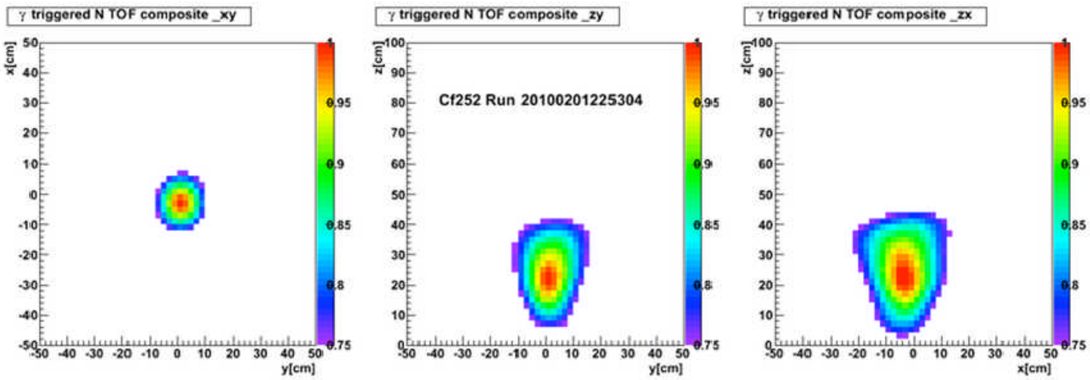

Figure 15. Image of a ^{252}Cf source made with gamma ray neutron time-correlations in our new detector system. The image is made by tagging on a neutron pair and translating the difference in arrival time of that pair to a closely preceding gamma ray. The resolution (Cf should look like point source) is mostly due to the uncertainty in the neutron energy (asymmetry in the yz and xz plains due to our detector geometry). Approximately 5 minutes of data

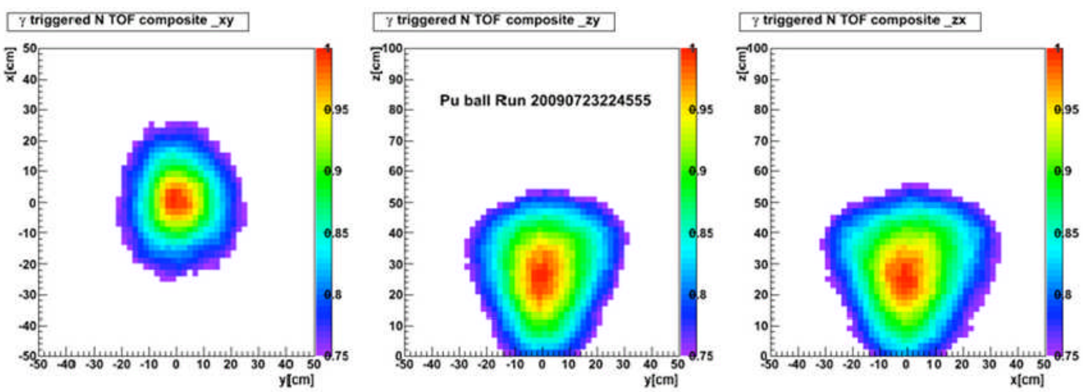

Figure 16. Image of a slightly multiplying Pu source made with gamma ray neutron time-correlations in our new detector system. The image is made by tagging on a neutron pair and translating the difference in arrival time of that pair to a closely preceding gamma ray. The difference in size between Figure 15 and figure 16 is the difference between a point and extended source of a few cm's diameter (asymmetry in the yz and xz plains due to our detector geometry). Approximately 10 seconds of data

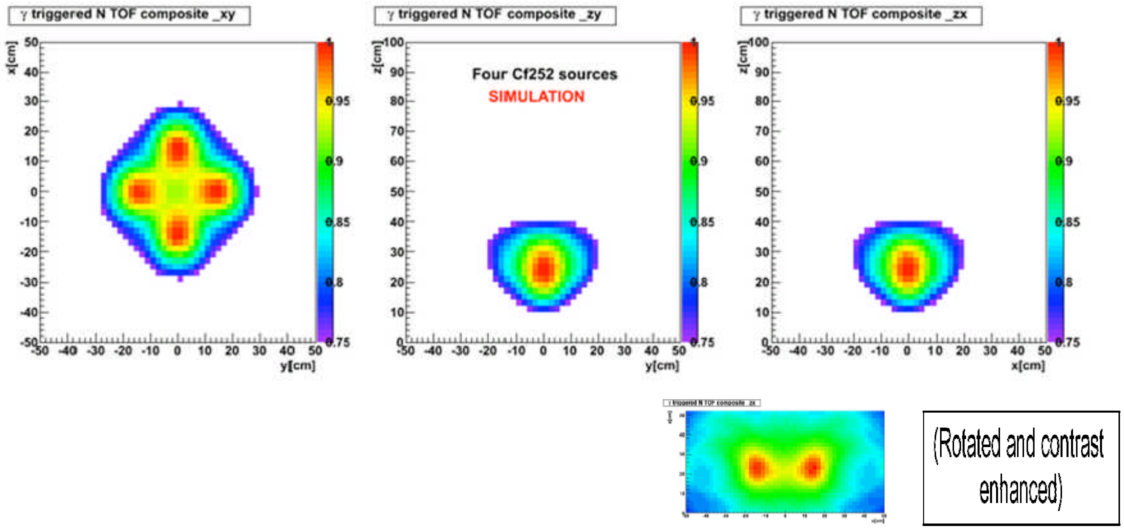

Figure 17. Simulation: Image of a 4 X ^{252}Cf sources made with gamma ray neutron time-correlations in our new detector system. The image is made by tagging on a neutron pair and translating the difference in arrival time of that pair to a closely preceding gamma ray. The last lower figure

6. Summary and Conclusions

We have taken fast neutron data and performed assays with our liquid scintillation array good to a few % with a few minutes of data and been able to achieve 5 to 6% total efficiency with an approximately 2π solid angle detector by applying algorithms developed originally for low efficiency portable thermal detectors. The intrinsically fast detection time of the liquid scintillator arrays greatly reduce random time-correlations inherent in time-correlation measurements and also permits direct measurement of the time scale differences between individual fission or fission-like processes (such as cosmic induced background) and those of fission chains which are unique to nuclear material able to support induced fission from lower energy neutrons. We have shown that the intrinsically faster liquid scintillator is already competitive with the highest efficiency well counters in measuring pairs correlations with shift-register logic for only modest sized samples of Pu and will be superior in precision for measuring multi Kg sized samples of Mox fuel rods to thermal ^{3}He detectors. This makes Liquid Scintillator a viable replacement in the short term for ^{3}He coincidence counters proposed for in line measurements at storage facilities provided that open questions of long term stability can be answered satisfactorily. In the longer term usage of plastic scintillators with doped for pulse-shape discrimination (PSD) usage (currently under development at LLNL) can be expected to have even better field performance. Also we believe that it is possible, with further development, to differentiate with direct measurement between complicated samples with different fractions of spontaneous fission sources for example Cm vs. Pu or Cf which all have different averages of neutrons produced, (a previously unsolvable problem in the safeguards community). We also believe that there is a wealth of information in the detailed timing information (including imaging the samples with fast neutron-gamma ray time-correlations) that can help with MC&A especially in samples with high fluxes and complicated neutron sources. Also the ability to see the detailed time structure to fission and induced fission makes neutron interrogation (particularly with low-energy neutrons) a very attractive technique that can certainly speed up measurement times when intrinsic neutron sources are lacking and give useful information even when there is a strong intrinsic source. Fast neutron detection also has the potential neutron to deliver energy information that is not available to thermal detectors.

The inherently lower cross-sections for fast neutron detection (and the inherently bulkier accompanying electronics required for faster timing) will probably prevent fast neutron detection from replacing thermal neutron measurements in all cases but scintillator detection shows great promise for attacking some of the more difficult problems in the safeguards arena such as dealing with high flux from spent fuels for accountability, verifying that fresh fuel has the right amount of nuclear material or to deal with the possibility of nuclear material diversion. Particularly with the advent of possible new crystal scintillators or plastic scintillators with PSD which can replace the flammable liquid scintillators which can have problematic safety issues in nuclear material storage facilities.

For the Arms Control and Treaty Verification arena we have already shown that we have the ability to determine just about any characteristic of an object that would prove it were from a weapon or weapon parts. We can measure the amount, count sources and show extent of the source (through imaging) and we can even show true density and thickness of material by examining the ratios of gamma rays from fission to neutrons (determined by their close time proximity). These all have profound implications for the quality of arms control verification. The only real questions remaining what are the exact (non-expert) algorithms to be used and how much information reveled may be too much. These will be determined by the agreements made and the careful balancing of the exact way measurements, data are collected (for example adjusting the efficiency and time of the measurement) and the exact algorithms employed.

There are many other as yet incompletely determined and developed applications for fast neutron technology. For example, we have long expounded the insensitivity of fast liquid scintillator as a positive feature for use of liquid scintillator with active or semi-active neutron interrogation in the search for HEU. The introduction of slow neutrons to a target system can be used to detect even small quantities of hidden HEU because the liquid scintillator are insensitive to slow neutrons and so any extra fast neutrons produced by the introduction of slow neutrons are easily detected and identified as hidden HEU. This technique has many implications for both search and material assays when dealing with unknown objects. It is safe to say that we have only begun to tap the wealth of information available with detection and evaluation employing fast neutron technology and the time scales of the measured-time correlations.

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