SAND2016-11224C

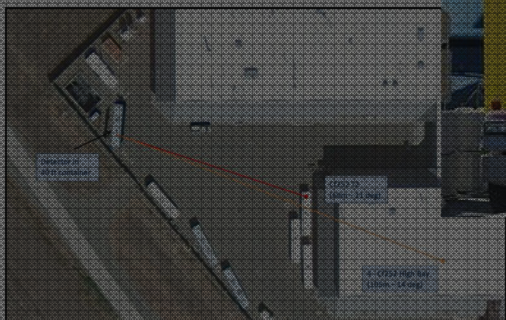
Measurement of Time Distribution of Fissions in a Chain with Fast Scintillators

Mateusz Monterial, Peter Marleau, Sara A. Pozzi

IEEE Nuclear Science Symposium
Strasbourg, France
November 1st , 2016

Special Nuclear Material - detection/imaging

Standoff detection



Cargo screening

SNM detection/localization

- Low signal rate
  - Need large area detectors!
- Low signal to background
  - Need background discrimination!

Arms control treaty verification

emergency response

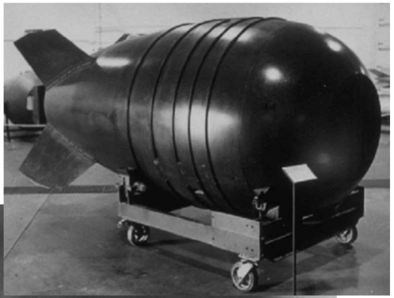
NNSA Joint Technical Operations Team

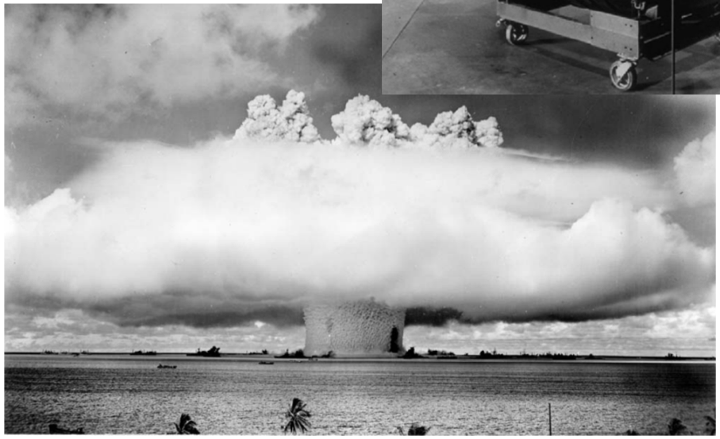
SNM characterization/imaging

- Material properties
  - Mass, multiplication, isotopics
- High resolution required
  - Fine detector segmentation
- Multiple or extended sources

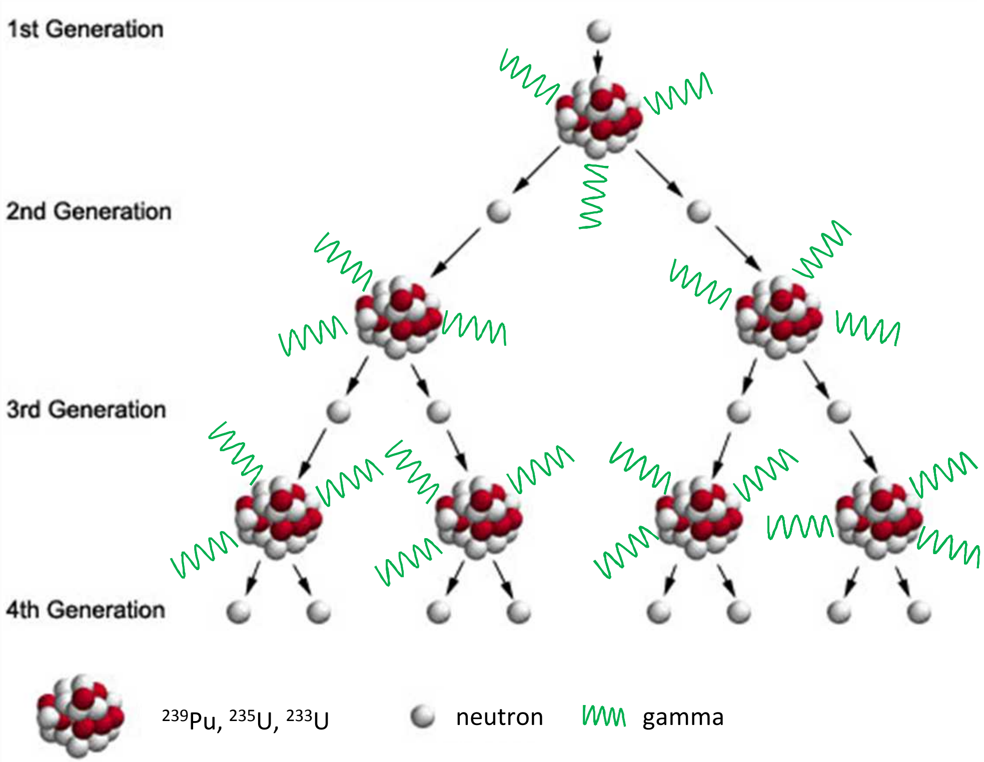
Motivation
Timing as a Unique Signature

- Applications
  - Treaty verification (e.g. New START)
  - Emergency response
- Requirements
  - Portability
  - The primary variables are mass, density, morphology of nuclear material and presence of shielding and moderators
- Method
  - Time-correlated particles are a unique signature of the fission process and therefore the nuclear material content
  - SNM, by definition, exhibit these unique properties





Principles of Fission and Chain Process



http://www.atomicarchive.com/Fission/Fission2.shtml

Fission by-products:

- **2-4** neutrons per fission ($\bar{\nu}_n$)
- **7-9** gammas per fission

Quantities of Interest:

$$k = \frac{neutrons\ in\ one\ generation}{neutrons\ in\ preceeding\ generation}$$

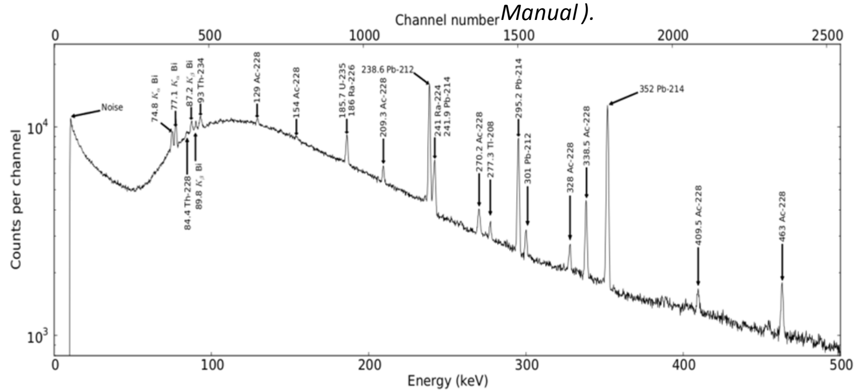
Subcritical multiplication – *total* neutrons created per source neutron:

$$\boldsymbol{M = \frac{1}{1-k}}$$

Current Methods
Measurable Unique Signatures of Fissile Material

1. Gamma spectrum
   - Isotopic content
2. Total neutron rate
   - Assay of the contents of specific materials
3. Correlated counts
   - Multiplicity analysis (singles, doubles, triples): fission rate, multiplication, (α, n) component
   - Relative multiplication:
     - Rossi-alpha distribution
     - Feynman variance technique

Plutonium Scrap Multiplicity Counter, used for accurate assays of plutonium metal, oxide, mixed oxide, or scrap *(LANL PANDA Manual).*



Current Methods
Drawbacks and Limitations

1. Gamma spectrum
   - Attenuation and self-shielding
2. Total neutron rate
   - Requires administrative controls
   - Spontaneous & induced fission and (α,n) sources are indistinguishable
3. Correlated counts
   - Requires high efficiency, necessitates large detection system
   - Efficiency has to be well known
   - Detector die-away time of 10-30 μs ("superfission concept")
   - Neutron energy information is lost due to moderation

→ He-3 based technologies

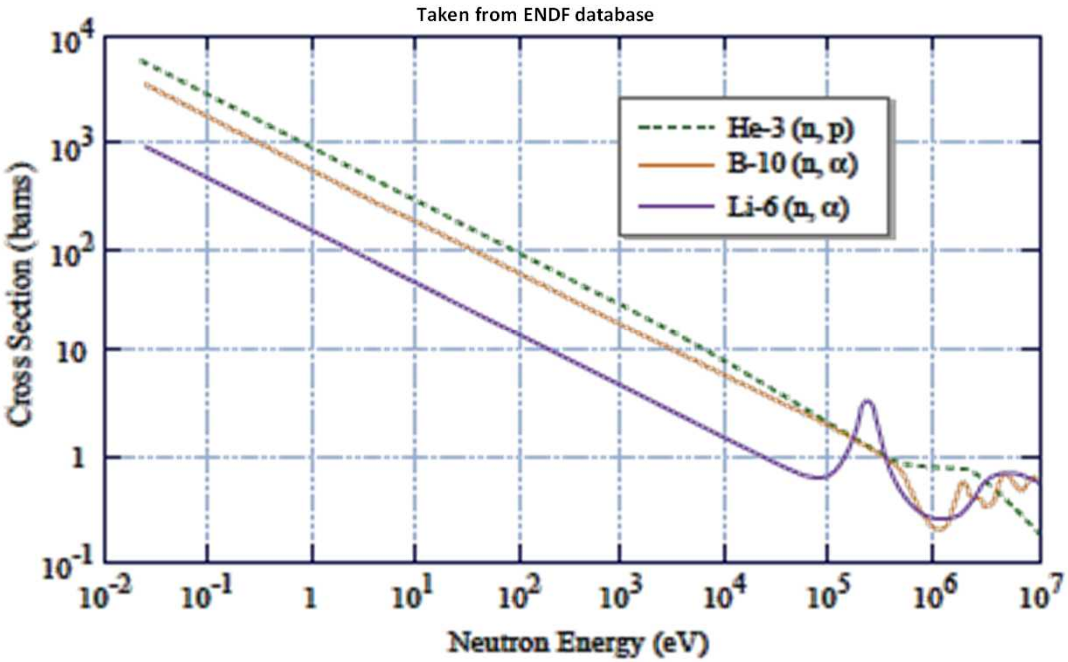
Thermal Neutron Detection

- **High thermal cross section (efficiency)**
- **High Q-value (discrimination)**

$n + {}^{3}He \rightarrow {}^{3}H + {}^{1}H + 0.764\ MeV$

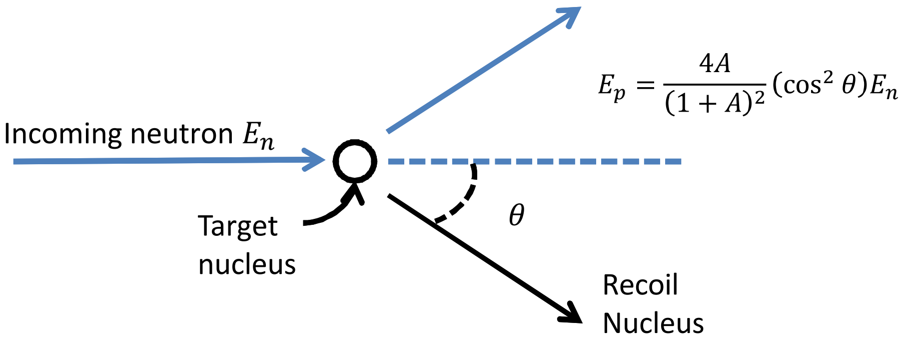
$n + {}^{10}B \rightarrow {}^{7}Li + \alpha + 2.31\ MeV$

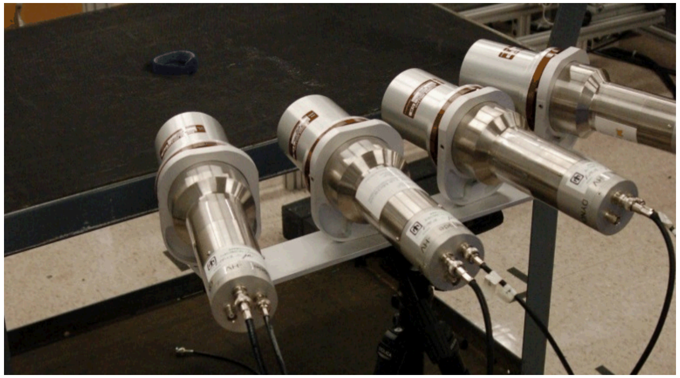
$n + {}^{6}Li \rightarrow {}^{3}H + \alpha + 4.78\ MeV$



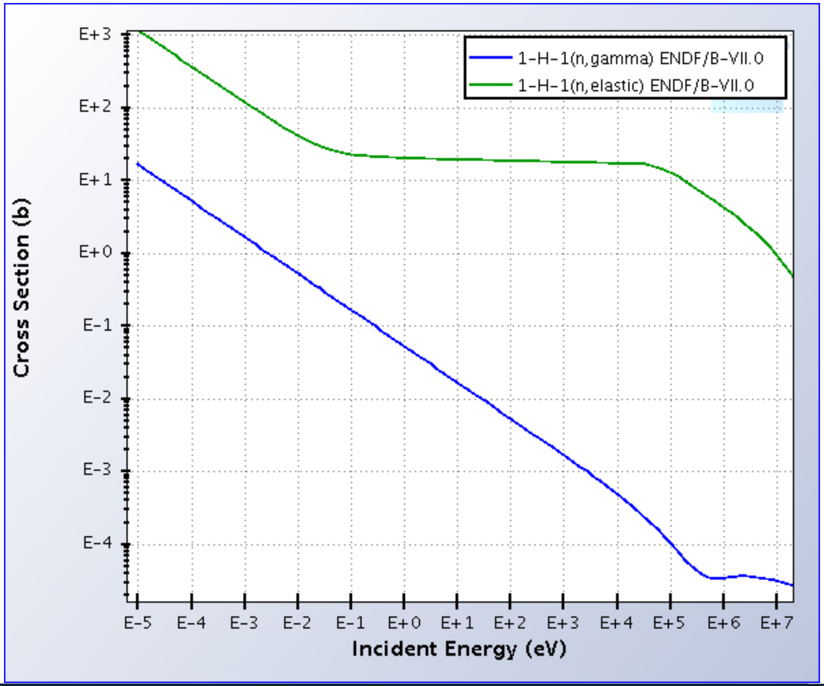
Fast Neutron Detection - Elastic

Deposited Neutron Energy





Nucleus	Q_{max}/E_n
$^{1}_{1}H$	1.000
$^{2}_{1}H$	0.889
$^{4}_{2}He$	0.640
$^{9}_{4}Be$	0.360
$^{12}_{6}C$	0.284
$^{16}_{8}O$	0.221
$^{56}_{26}Fe$	0.069
$^{118}_{50}Sn$	0.033
$^{238}_{92}U$	0.017



Pulse Shape Discrimination

Pulse Shape Dependence on Interacting Particle

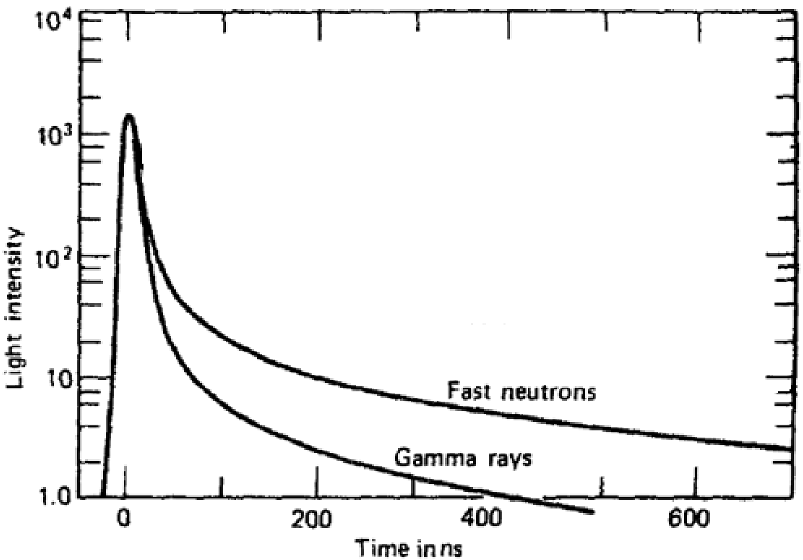
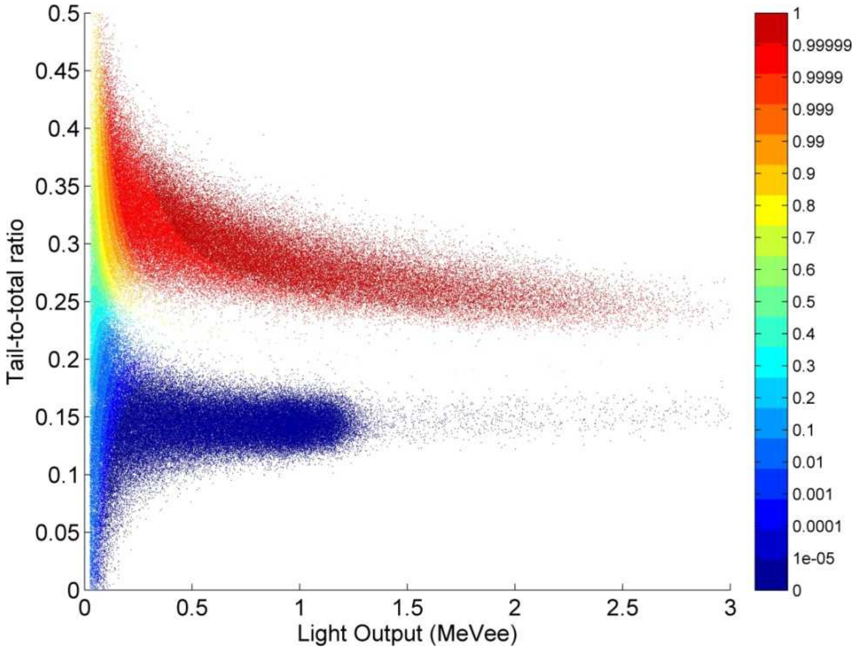


Figure from Glenn Knoll *Radiation Detection & Measurement 3rd Edition*

Bayesian Probability Map



Fast Correlation Discrimination Capable Organic Scintillators

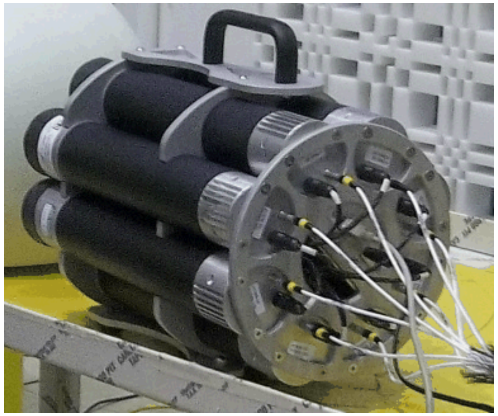
New Approach

System advantages:

1. Can be low efficiency
2. Efficiency can be ignored in calculations
3. Detection systems can be portable
4. Neutron energy information is preserved
5. Timing is within the resolution time of a fission chain

Potential:

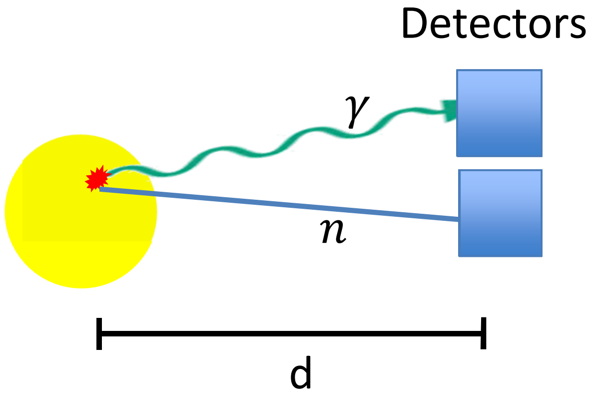
1. Differentiate contributions from spontaneous fission, induced fission (fission chains), and (α, n) sources
2. Simultaneously solve for mass, multiplication, and shielding



“The 8-shooter”
Array of Stillbene crystal scintillator

✓ PSD Capable
✓ Fast Timing

Neutron-Gamma Correlation – Same Fission



$$\Delta t_{n-\gamma} = \left(R_n \sqrt{\frac{m}{2E_{n0}}} - \frac{R_\gamma}{c} \right)$$

Time of arrival difference between γ and neutron	Time-of-flight difference between γ and neutron
Measured	Estimated

Observed:

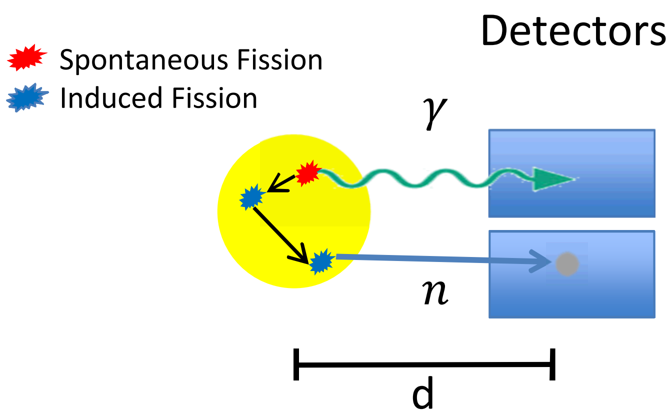
$$E_p \leq E_n$$

proton recoil

$$\Delta t_{n,\gamma} - d\left(\sqrt{\frac{m}{2E_p}} - \frac{1}{c} \right) \leq 0$$

$$\Delta t_{n,\gamma} - \Delta \mathrm{t_p} \leq 0$$

Neutron-Gamma Correlation – Fission Chain



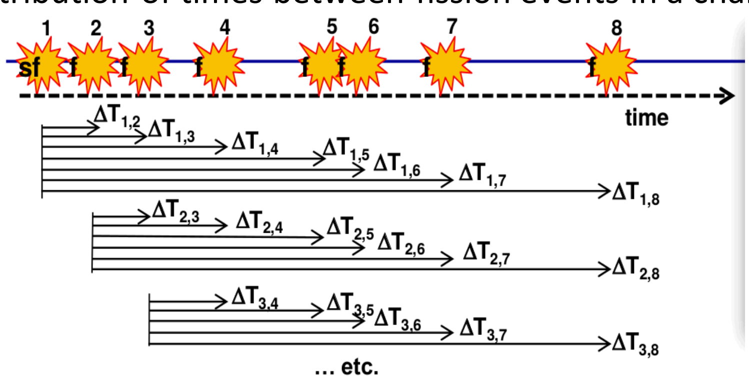
Time difference between fission events in a chain

$$\Delta t_{n-\gamma} - \Delta t_p \leq \Delta T_{f_1,f_2}$$

Measured time difference between neutron and nearest γ

Estimated travel time difference between neutron and gamma

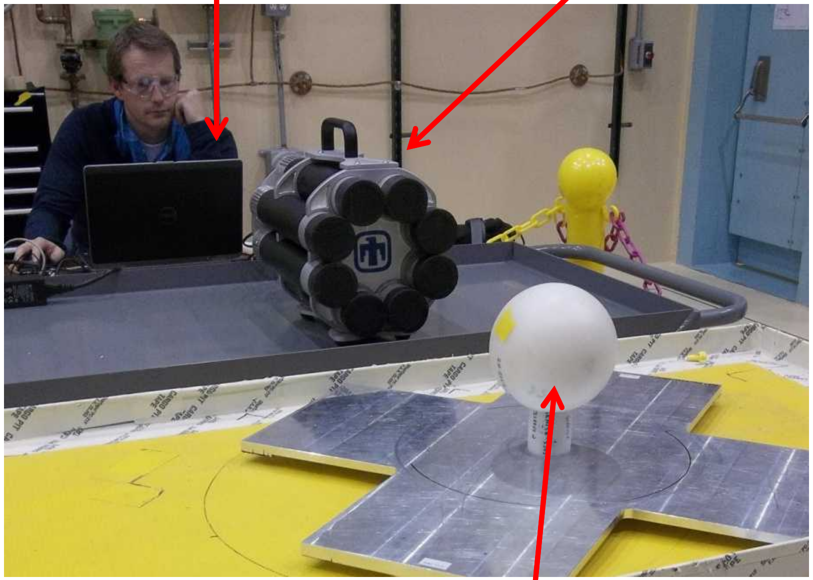
Distribution of times between fission events in a chain



$$\Delta T_{f_1,f_2} > 0 \text{ if } f_1 = \gamma \;\&\; f_2 = n$$

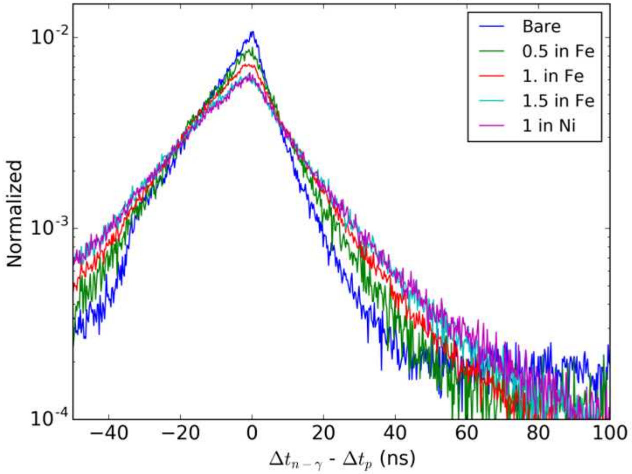
$$\Delta T_{f_1,f_2} < 0 \text{ if } f_1 = n \;\&\; f_2 = \gamma$$

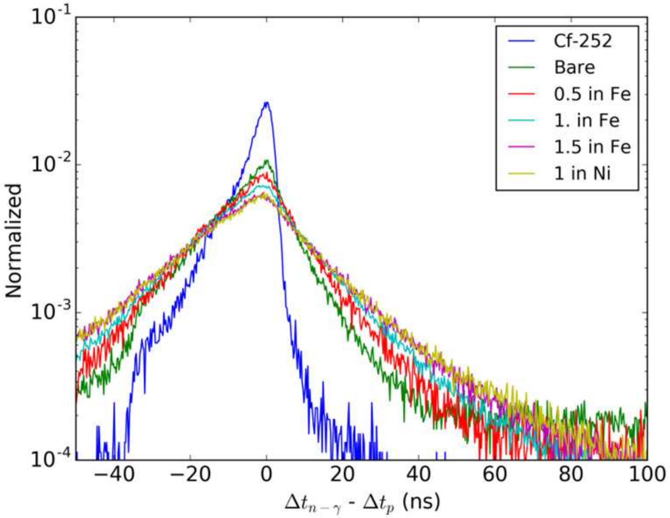
Experiments



Source	Distance (cm)	Total Time (minutes)	Rate of gamma-Neutron Pairs (Bq)
BeRP	34	1730	136
BeRP + 0.5 in Steel	34	600	211
BeRP + 1 in Steel	34	1800	280
BeRP +1 in Nickel	34	1200	239
BeRP + 1.5 in Steel	34	1200	243

Time Of Flight Fixed by Energy Estimation (TOFFEE) Distributions



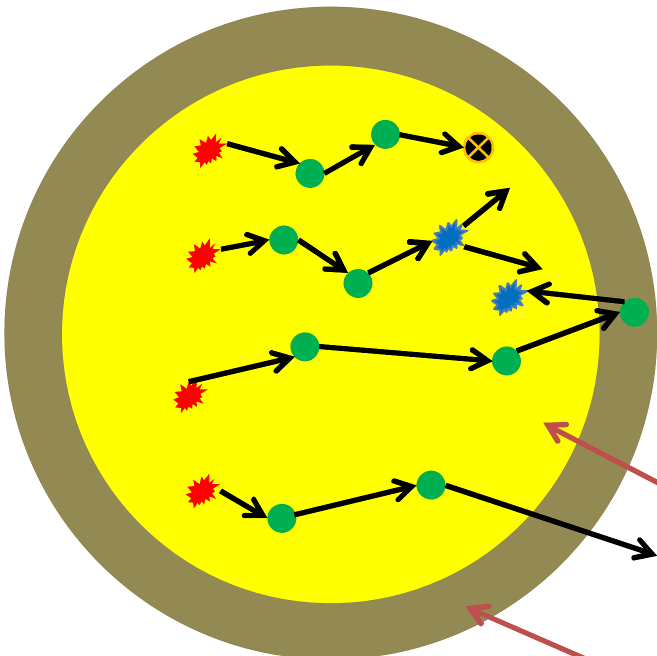


Source	Multiplication
BeRP	4.433 ± 0.001
BeRP + 0.5 in Steel	5.716 ± 0.002
BeRP + 1 in Steel	6.679 ± 0.002
BeRP +1 in Nickel	7.512 ± 0.002
BeRP + 1.5 in Steel	7.879 ± 0.002

- Shape of the distribution changes with type of reflector and amount of reflector
- The spread of the distribution changes with multiplication
- Contains more information than simple rate of gamma-neutron correlations
- Correcting for distance makes it independent of geometry (source-to-detector distance)

0-D Monte Carlo Model

Neutron Walk in Subcritical Assembly



Fissile Material

Moderator (low Z) / Reflector (high Z)

Neutron Interaction Possibilities:

- Spontaneous Fission
- Induced Fission
- Capture
- Scatter

Outcome

1) Capture

2) Fission

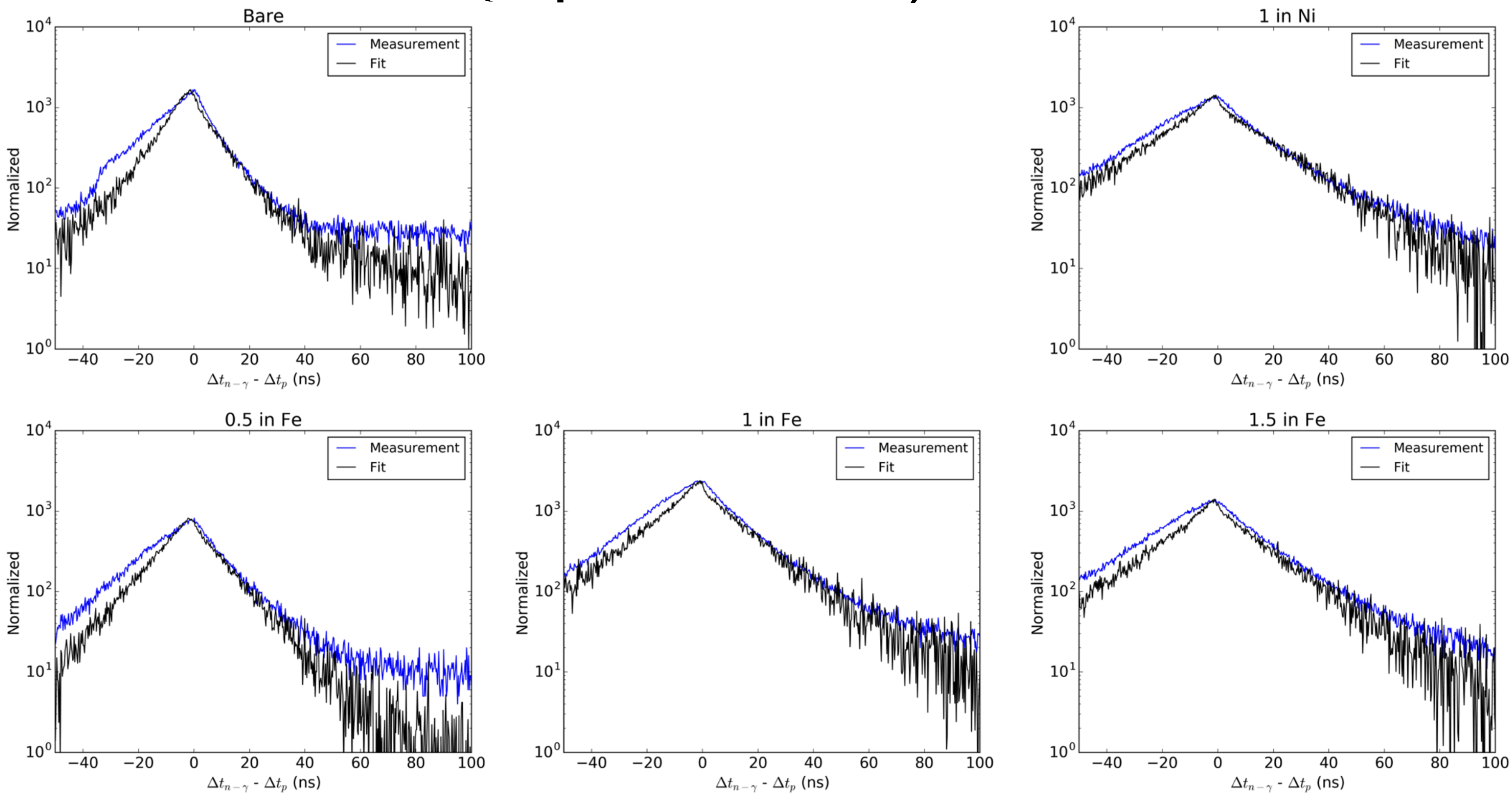
3) Reflections/Moderation

4) Escape Leakage

Factors

- Coupling to surrounding material
- Type of surrounding material (moderator/reflector)
- Mass & Density of nuclear material
- Cross-sections, probabilities of interaction
- Geometry

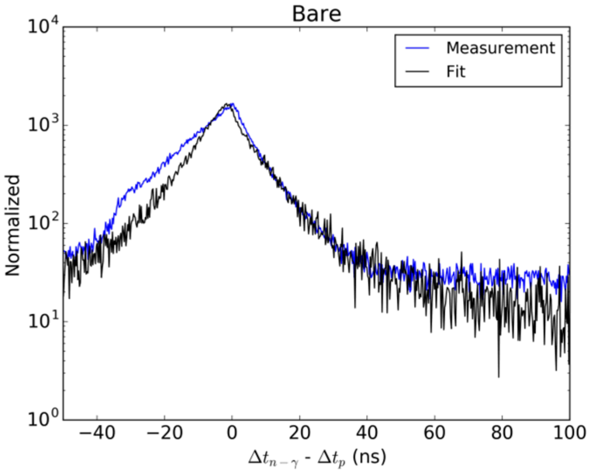
Fit Solutions (4 parameters)

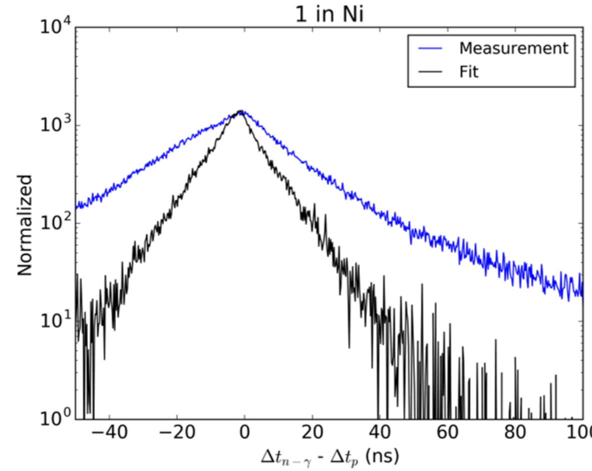


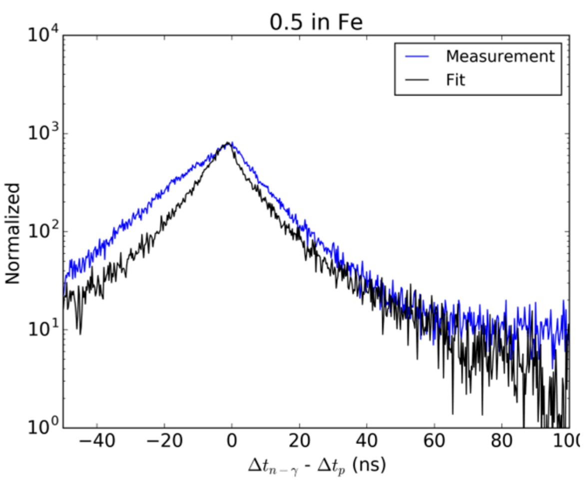
Fit Solutions (4 parameters)

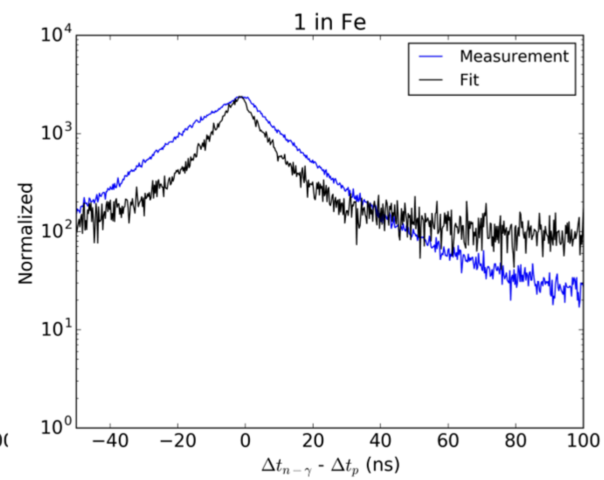
Configuration	Fission Prob	Fission Time (ns)	Return Prob.	Return Time (ns)	Multiplication
Bare	0.2615	2.188	0.0104	60.04	4.433
0.5 in Fe	0.2846	2.11	0.00224	60.49	5.675
1 in Fe	0.2850	2.989	0.0043	63.79	5.886
1.5 in Fe	0.2949	2.801	0.00219	57.40	6.897
1 in Ni	0.2926	3.121	0.003528	25.94	6.703

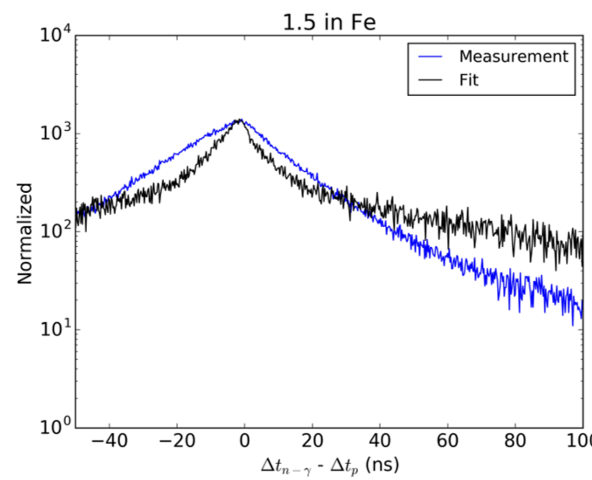
Fit Solutions (4 parameters)











Fit Solutions (4 parameters)

Configuration	Fission Prob	Fission Time (ns)	Return Prob.	Return Time (ns)	Multiplication
Bare	0.2615	2.188	0.0150	57.72	4.433
0.5 in Fe	0.2615	2.188	0.0171	23.52	5.675
1 in Fe	0.2615	2.188	0.0512	85.30	5.886
1.5 in Fe	0.2615	2.188	0.0612	34.02	6.897
1 in Ni	0.2615	2.188	0.0009	35.85	6.703

Conclusions

- Demonstrated a new approach for SNM characterization with organic scintillators
- Developed a simple model for understanding fission chain dynamics
- Performed parametric study of a subcritical assembly under different configurations
- Fit physical parameters to measured data with limited success

Acknowledgements

This research was performed under the Nuclear Forensics Graduate Fellowship Program, which is sponsored by the U.S. Department of Homeland Security, Domestic Nuclear Detection Office and the U.S. Department of Defense, Defense Threat Reduction Agency

This work was funded in-part by the Consortium for Verification Technology under Department of Energy National Nuclear Security Administration award number DE-NA0002534.

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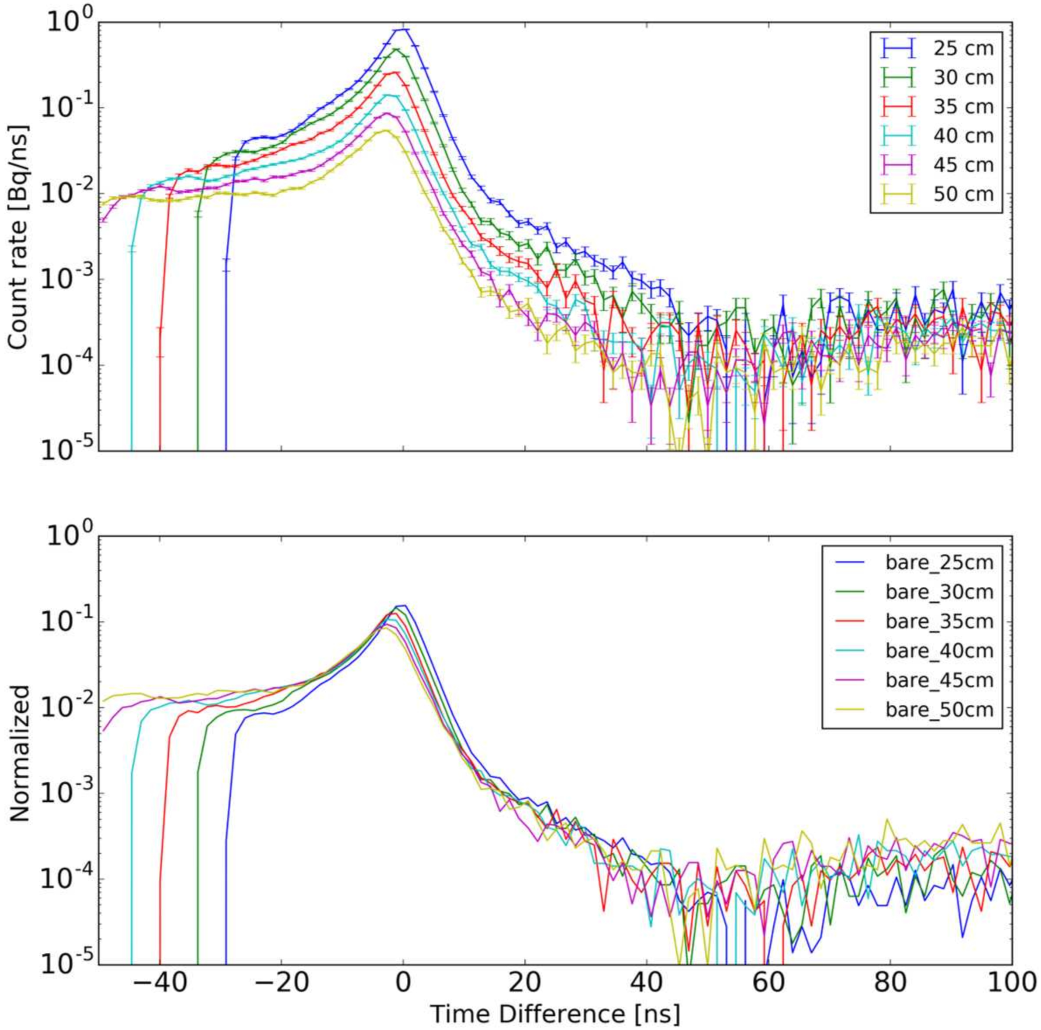
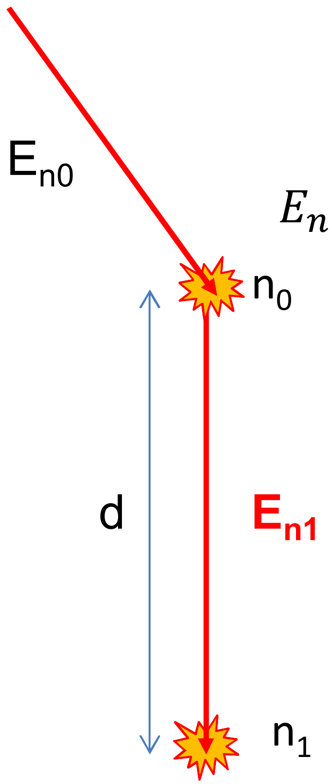
EXTRA SLIDES

TOFFEE for Cf-252

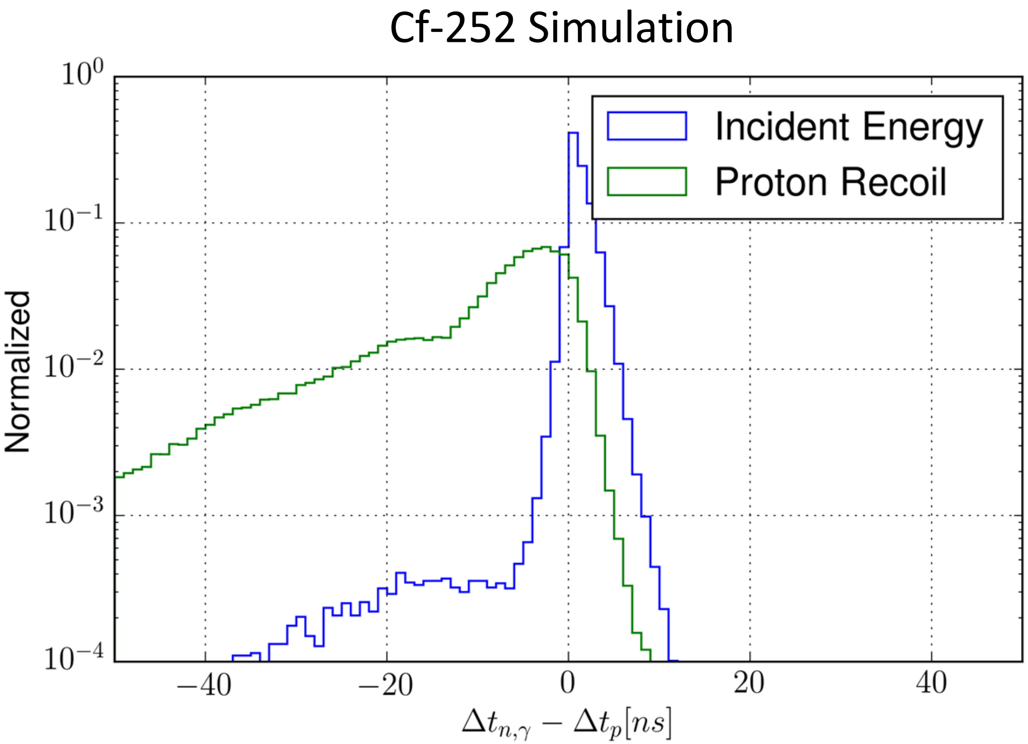


Neutron-neutron Scatter
Objective 2

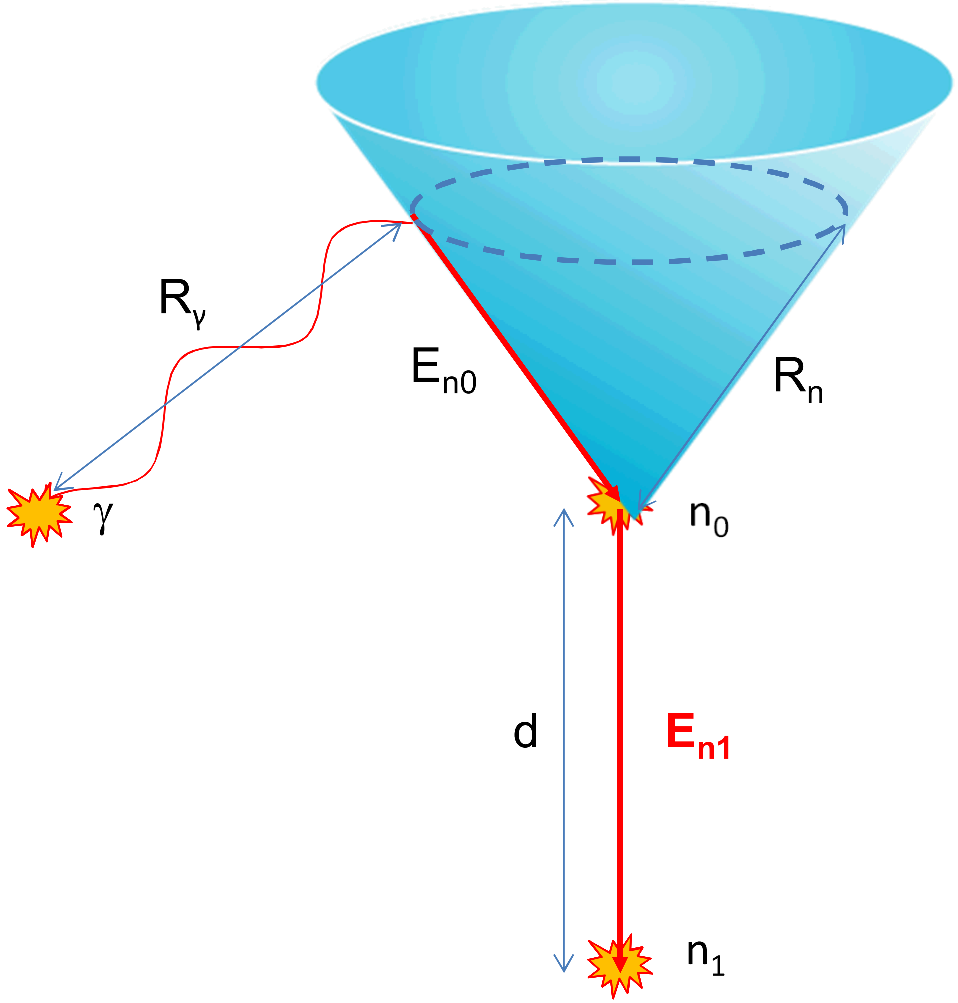


$$E_{n1} = \frac{m_n}{2}\left(\frac{d}{\Delta t_{n_0,n_1}}\right)^2$$

$$E_{n0} = E_p + E_{n1}$$



γ-n-n Coincidence Imaging



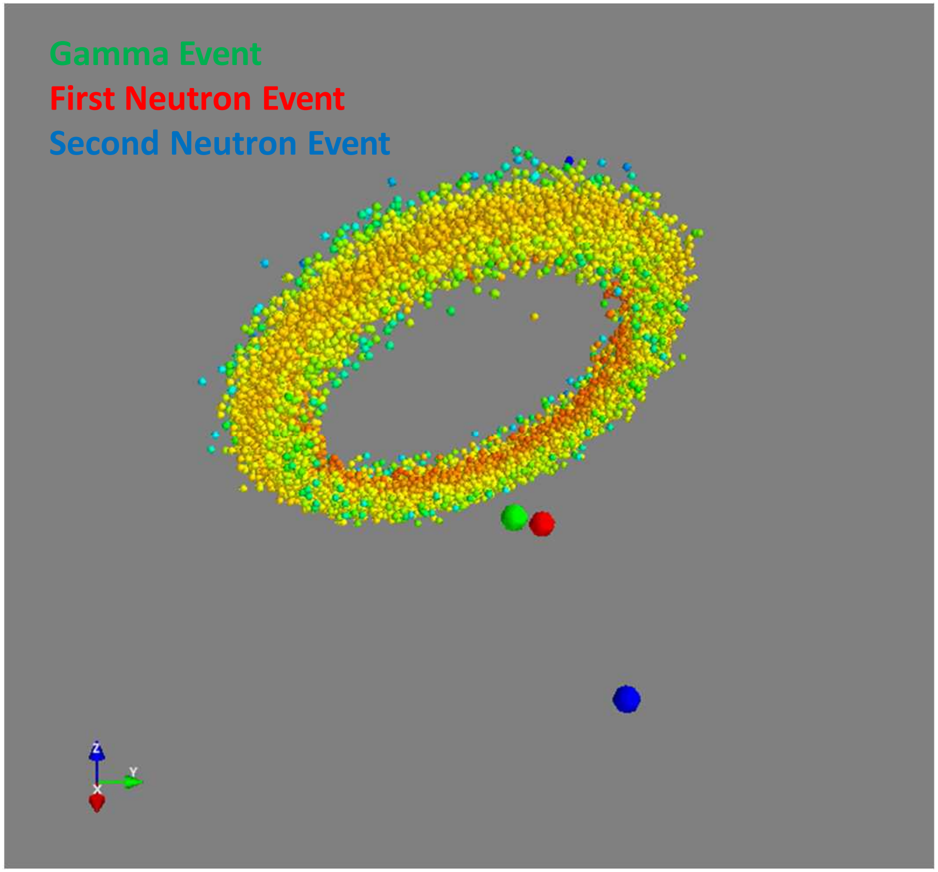
$$E_{n1} = \frac{m_n}{2}\left(\frac{d^2}{\Delta t_{n_0,n_1}}\right)$$

$$E_{n0} = E_d + E_{n1}$$

$$cos^2\theta_{n1} = \frac{E_{n1}}{E_{n0}}$$

$$\Delta t = \frac{R_n}{v_n} - \frac{R_\gamma}{c}$$

γ-n-n 3D Reconstruction

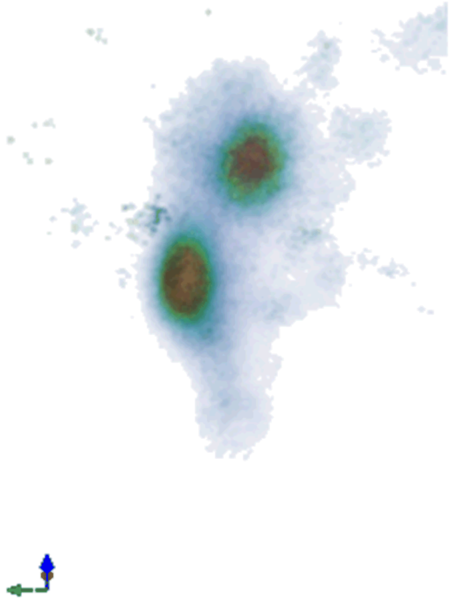


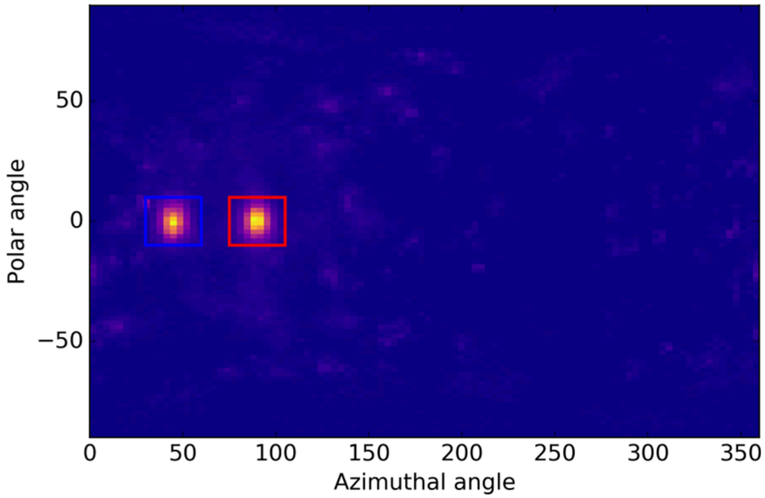
$$cos^2\theta_{n1} = \frac{E_{n1}}{E_{n0}}$$

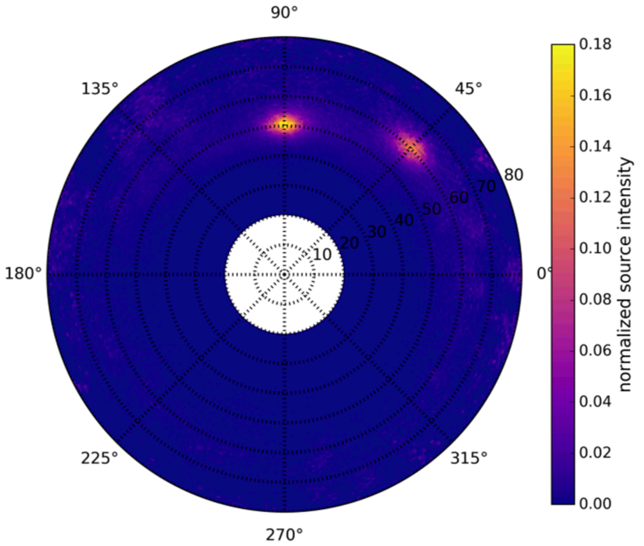
+

$$\Delta t_{n-\gamma} = \frac{R_n}{v_n} - \frac{R_\gamma}{c}$$

Preliminary Reconstruction of Cf-252







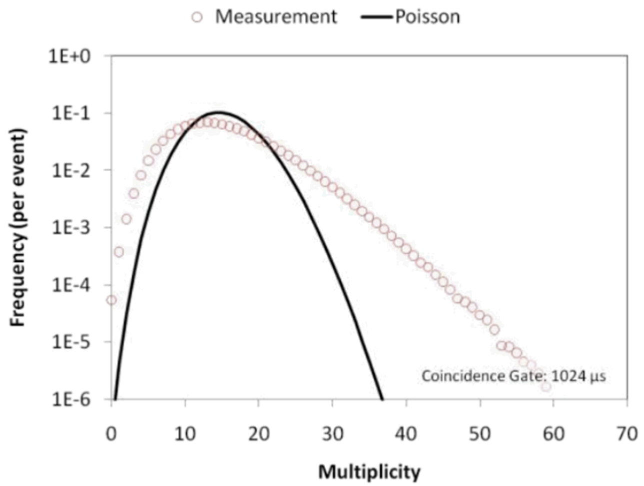
Neutron Coincidence Counting Equations

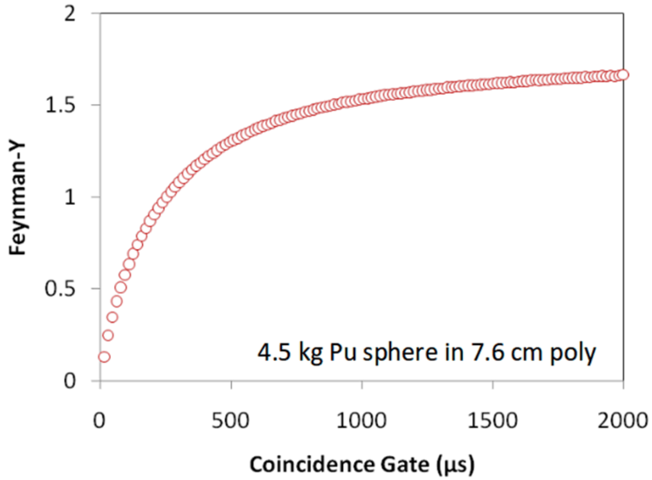
1. $S = F\epsilon M v_{s1}(1+\alpha)$
2. $D = \frac{F\epsilon^2 f_d M^2}{2}\left[v_{s2} + \left(\frac{M-1}{v_{i1}}\right) v_{s1}(1+\alpha)v_{i2}\right]$
3. $T =$
$\frac{F\epsilon^3 f_t M^3}{6}\left[v_{s2} + \left(\frac{M-1}{v_{i1}-1}\right)[2v_{s2}v_{i2} + v_{s1}(1+\alpha)v_{i3}] + 3\left(\frac{M-1}{v_{i1}}\right)^2 v_{21}(1+\alpha)v_{i2}^2\right]$

- F = spontaneous fission rate
- ϵ = neutron detection efficiency
- M = neutron leakage multiplication,
- $\alpha = (\alpha, n)$ to spontaneous fission neutron ratio
- f_d= doubles gate fraction
- f_t= triples gate fraction
- v_{s1}, v_{s2}, v_{s3}= factorial moments of the spontaneous fission neutron distribution
- v_{i1}, v_{i2}, v_{i3}= factorial moments of the induced fission neutron distribution

Feynman-Y Approach

- Measures correlated counts in a fixed gate
- Fission chains create variance in excess of Poisson distribution
- $\frac{\sigma^2}{\mu} = 1 + Y$
  - σ^2: variance
  - μ: mean





Figures courtesy of John Mattingly: http://web.ornl.gov/sci/nsed/outreach/presentation/2011/Mattingly.pdf

Neutron Sources in Nuclear Fuel Cycle

- Spontaneous Fission
  - Pu-238/240/242, Cf-252
  - Energy spectrum is Maxwellian (~2 MeV mean):
    - Sqrt(E) exp(-E/1.43)
- Induced Fission
  - U-233/235, Pu-239
  - Spectrum depend on the energy of incident neutron
- (alpha, n) reactions
  - Alpha + O-18 → Ne-21 + n
  - Alpha + F-19 → Na-22 + n
  - Spectrum depends on target isotope to second order alpha energy

Note on TCPH Sensitivity

- Correlation measurements are sensitive to multiplying sources because of the increasing chance of correlating particles from different chains
- Plot: Correlation probability vs. fission chain length

MC-Fiss Overview

Assumptions:

- No special consideration, infinite medium
- Leakage and absorption are combined into single term
- No energy dependence
  - Cross-sections
  - Moderation

Input variables:

- $\bar{\nu}_n = 3$: mean number of neutrons per fission
- $p_f = 0.24$: probability of fission
- $\mu_f = 1.58\ ns$: mean time between fission chains

Sample distributions:

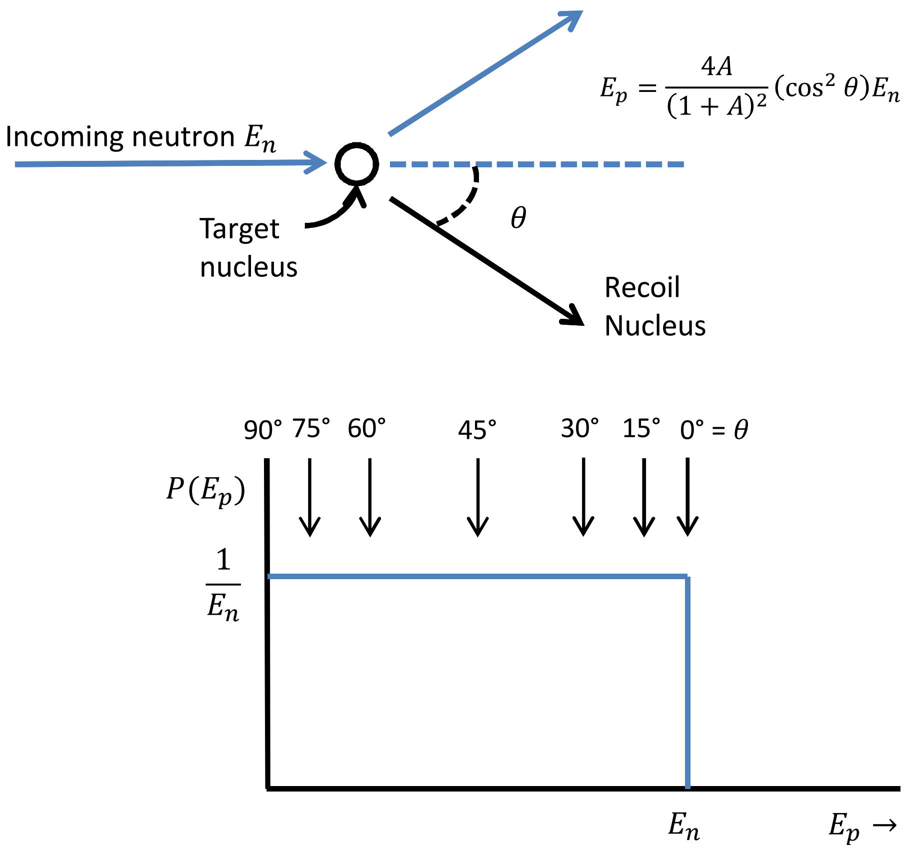
- Binomial(n=$\bar{\nu}_n$, p=p_f)
  - Number of fissions born in the next generation.
- t_f = Exponential(mean = μ_f) + t_0
  - Time from 0th generation fission

Pseudo code flow for each history:

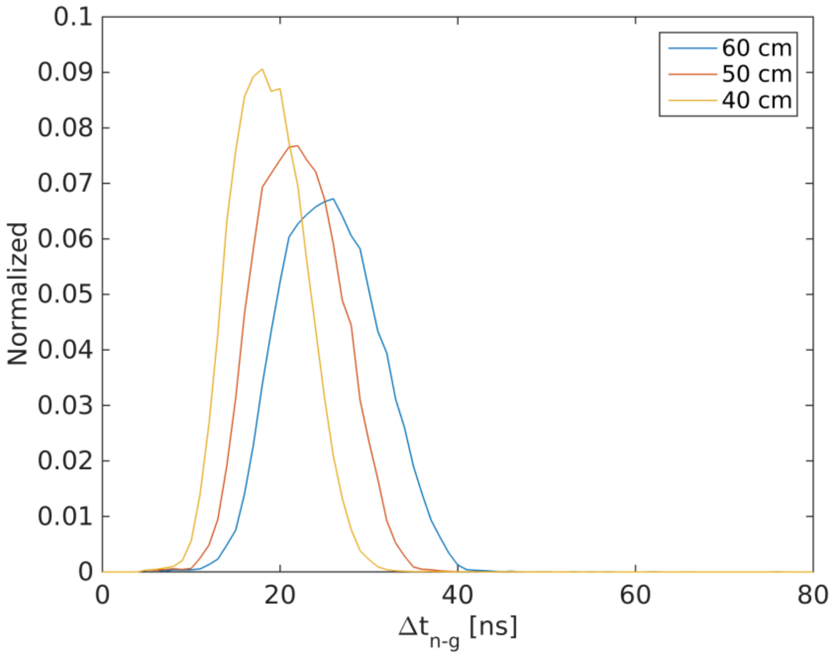
1. Sample number of fissions and time for each successful fission
2. Set $t_0 = t_f$
3. Call function recursively for each fission time.

Measured Quantities from Organic Scintillators

Deposited Neutron Energy



Neutron-Gamma Timing

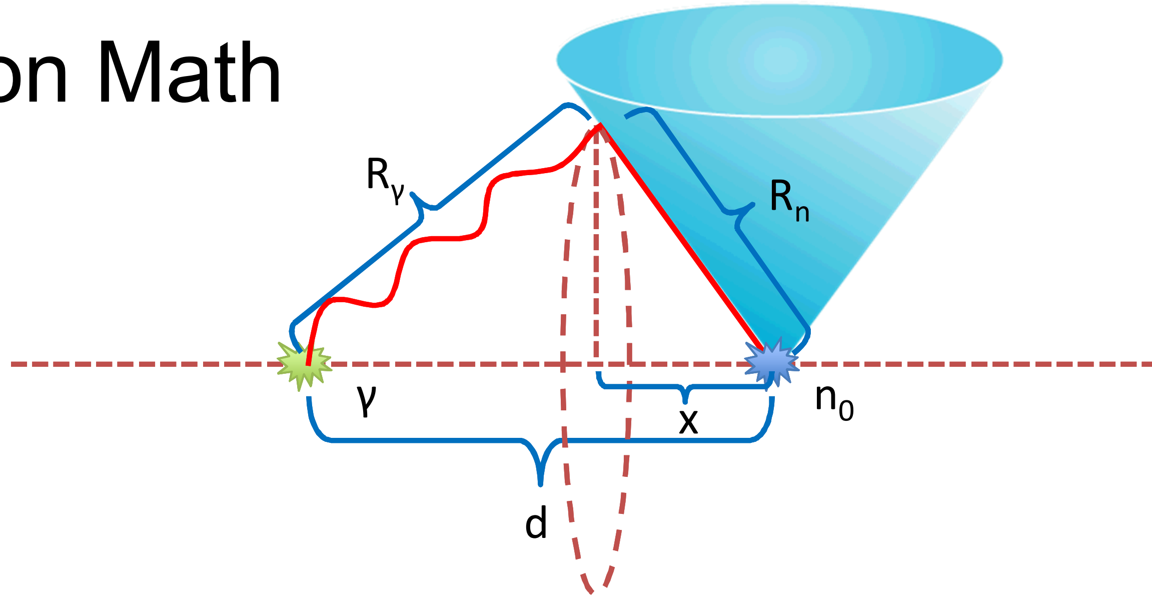


3D Reconstruction Math

$$\Delta t = \frac{R_n}{v_n} - \frac{R_\gamma}{c} \qquad [1]$$

$$x = \frac{(d^2 + R_n^2 - R_\gamma^2)}{2d} \qquad [2]$$

$$x = R_n(\widehat{R_n} \cdot \hat{x}) = R_n\mu \qquad [3]$$



$\widehat{R_n}$ traces the surface of the cone
Substitute [1] and [3] into [2] and solve for R_n:

$$\boldsymbol{R_n = \frac{c^2 \Delta t v_n - d v_n^2 \mu + \sqrt{v_n^2(c^2 d^2 - 2c^2 d \Delta t v_n \mu + (c v_n \Delta t)^2 + (d v_n \mu)^2 - (d v_n)^2)}}{c^2 - v^2}}$$

The solution is a parametric equation for R_n in terms of μ, the cosine between cone surface and unit vector between n_0 and γ.

Equation [2] is part of a solution of the intersection of two spheres