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Material Characterization and Imaging using fast neutrons

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Motivation

Correlated Timing and imaging as a Unique Signatures

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IAEA
International Atomic Energy Agency
Non-proliferation

- Applications
  - Non-proliferation and safeguards
  - Arms control treaty verification
  - Emergency response
- Characterization/imaging
  - Is it Special Nuclear Material?
  - Is it weaponized?
  - Material properties
    - Mass, multiplication, isotopic
- Method
  - Measuring time correlated gammas and neutrons for non-destructive assay of nuclear material

Arms control treaty verification

NNSA
Joint Technical Operations Team

Emergency response

Bilateral nuclear stockpile reduction

Summary

- What's so special about special nuclear material (SNM)?
- Characterization of SNM
  - What signatures can be used?
  - Neutrons: slow and fast.
  - Correlated Timing
  - 3D imaging
- Characterization of other materials
  - Active interrogation
  - Inelastic interactions
  - 3D imaging

Nuclear Material?

As defined by the IAEA:

Nuclear Material – metals uranium, plutonium, and thorium in any form.

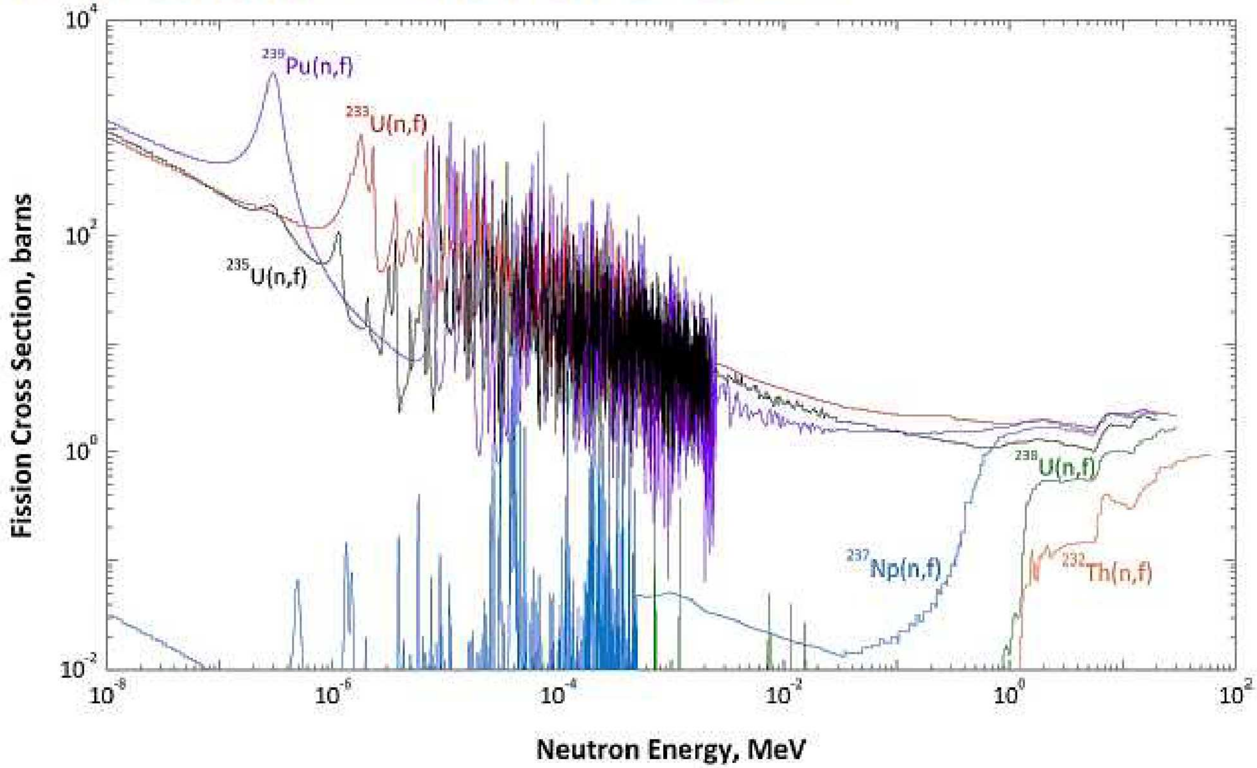
Special Nuclear (fissile) Material – U-233, U-235, Pu-239

Source Material – everything that is not special

What's so special?

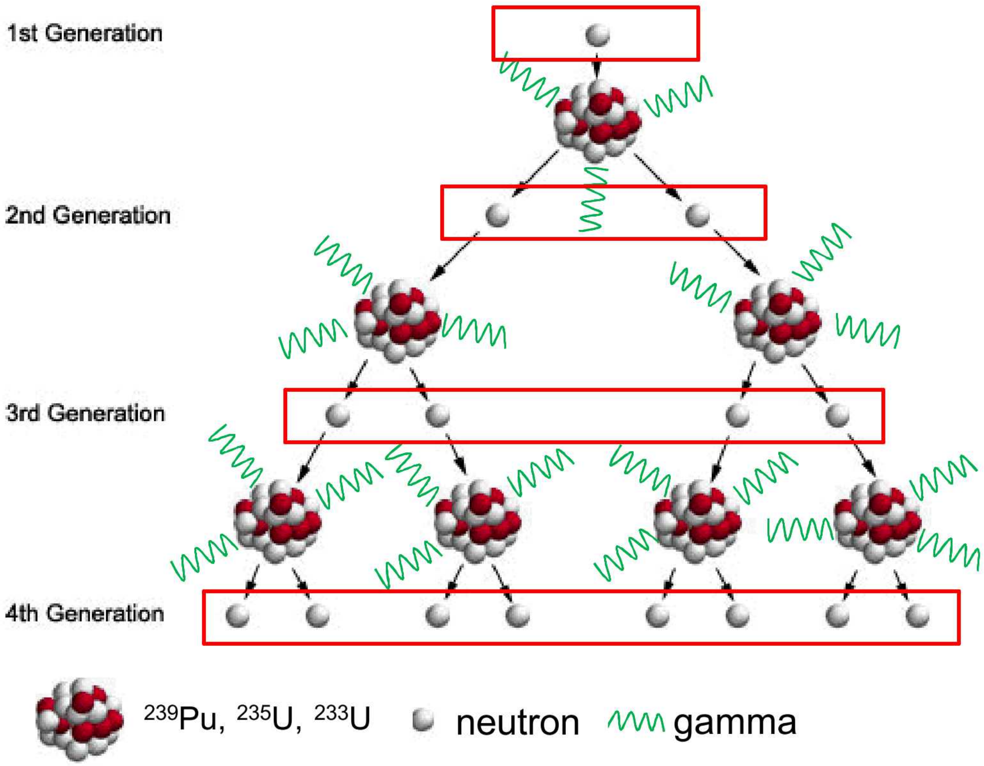
Isotope	Induced Thermal Fission Multiplicity ν
^{232}U	3.13
^{233}U	2.4
^{234}U	2.4
^{235}U	2.41
^{236}U	2.2
^{238}U	2.3
^{237}Np	2.70
^{238}Pu	2.9
^{239}Pu	2.88
^{240}Pu	2.8
^{241}Pu	2.8
^{242}Pu	2.81
^{244}Cm	3.46
^{252}Cf	4.06

The Neutron Fission Cross Sections



What is so special about SNM?

Basic vocabulary and definitions



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

Fission products:

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

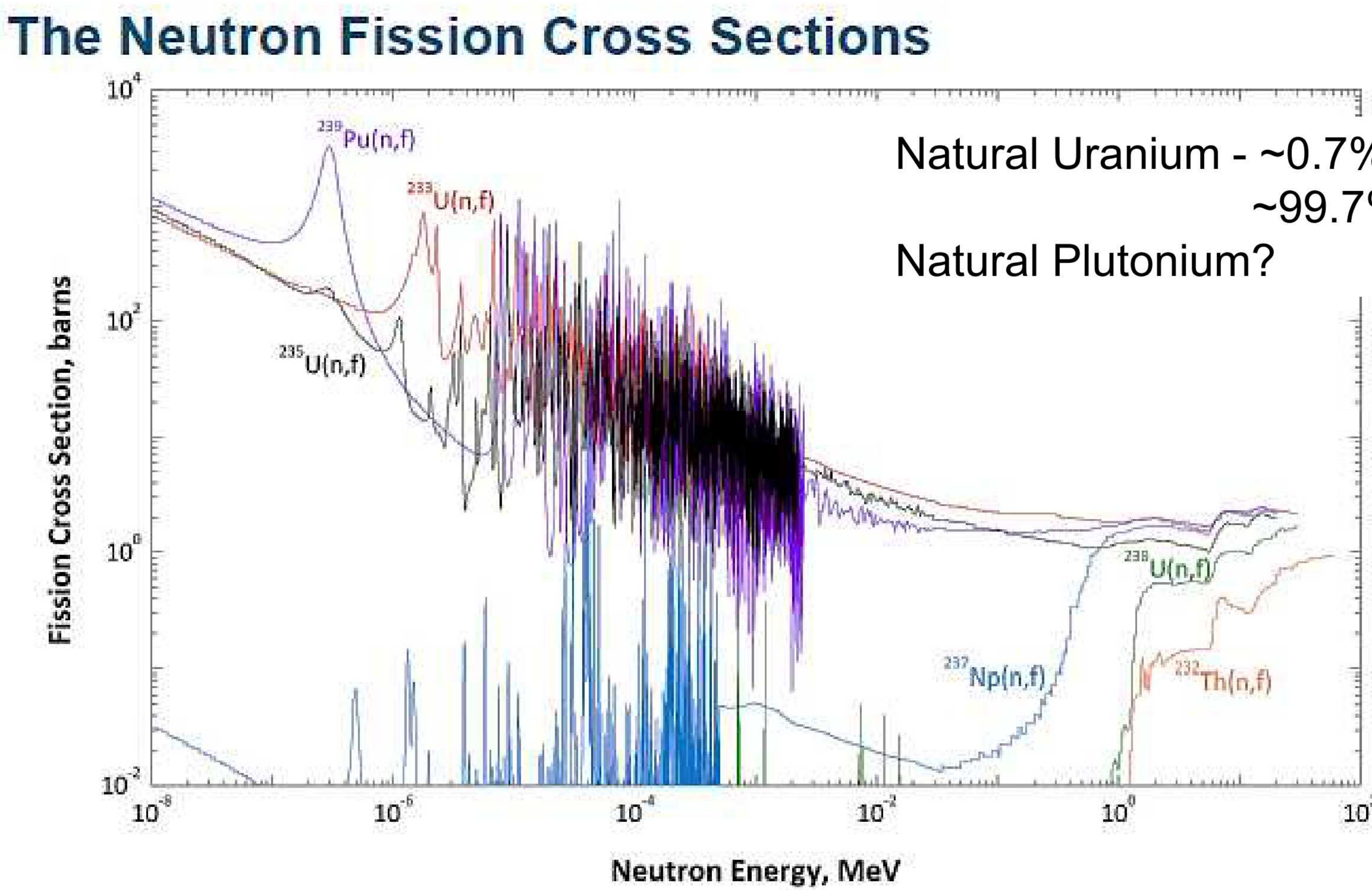
Quantities of Interest:

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

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

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

Where can I get some?



Natural Uranium - ~0.7% U-235
~99.7% U-238

Natural Plutonium?

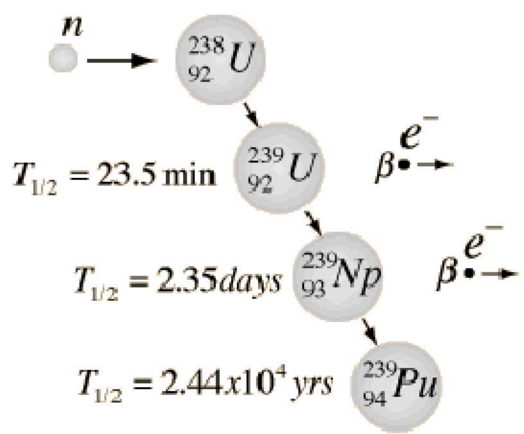
Figure courtesy of David Chichester, INL

Can I make it?

The Passive Neutron Signatures

Isotope	Half Life	Spontaneous Fission Yield (n/s-kg)	Spontaneous Fission Multiplicity ν	Induced Thermal Fission Multiplicity ν
^{232}U	71.7 yr	1,300	1.71	3.13
^{233}U	1.59×10^5 yr	0.86		
^{234}U	2.45×10^5 yr	5.02		
^{235}U	7.04×10^8 yr	0.299		
^{236}U	2.34×10^6 yr	5.49		
^{238}U	4.47×10^9 yr	13.6		
^{237}Np	2.14×10^6 yr	0.114		
^{238}Pu	87.7 yr	2.59×10^6		
^{239}Pu	2.41×10^4 yr	21.8		
^{240}Pu	6.56×10^3 yr	1.02×10^6		
^{241}Pu	14.35 yr	50 ±		
^{242}Pu	3.76×10^5 yr	1.72×10^6		
^{244}Cm	18.1 yr	1.08×10^{10}		
^{252}Cf	2.65 yr	2.34×10^{15}		

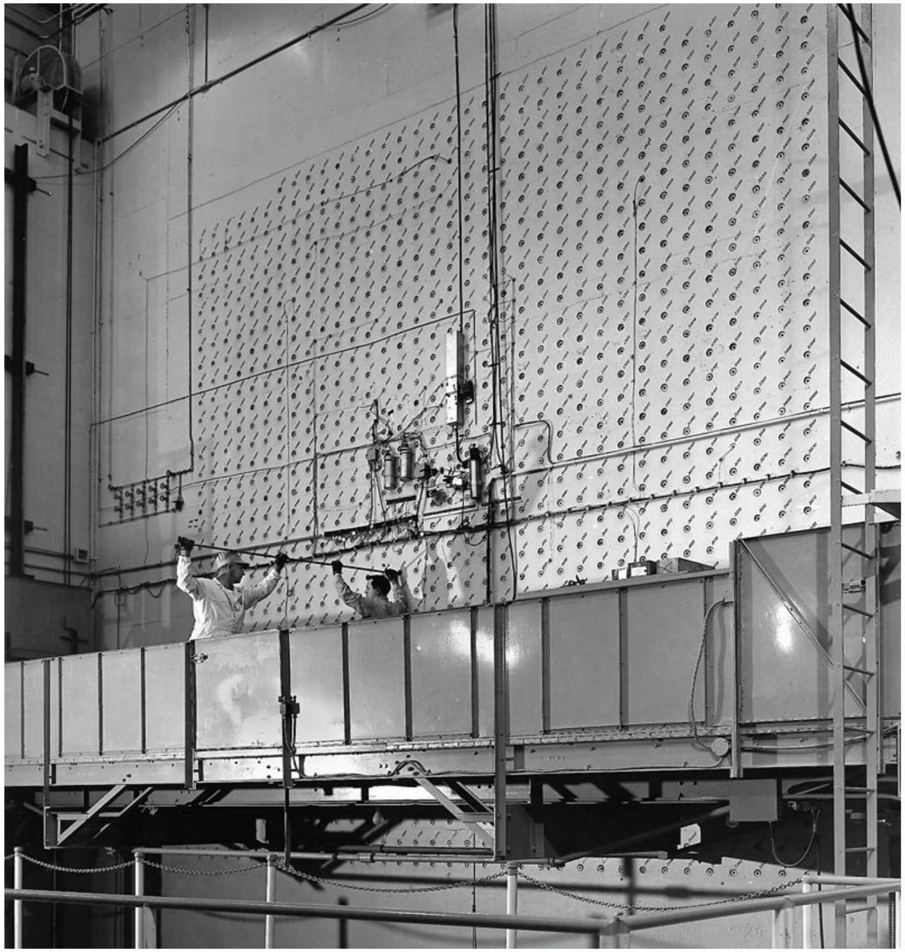
There isn't natural Plutonium to be found, but …



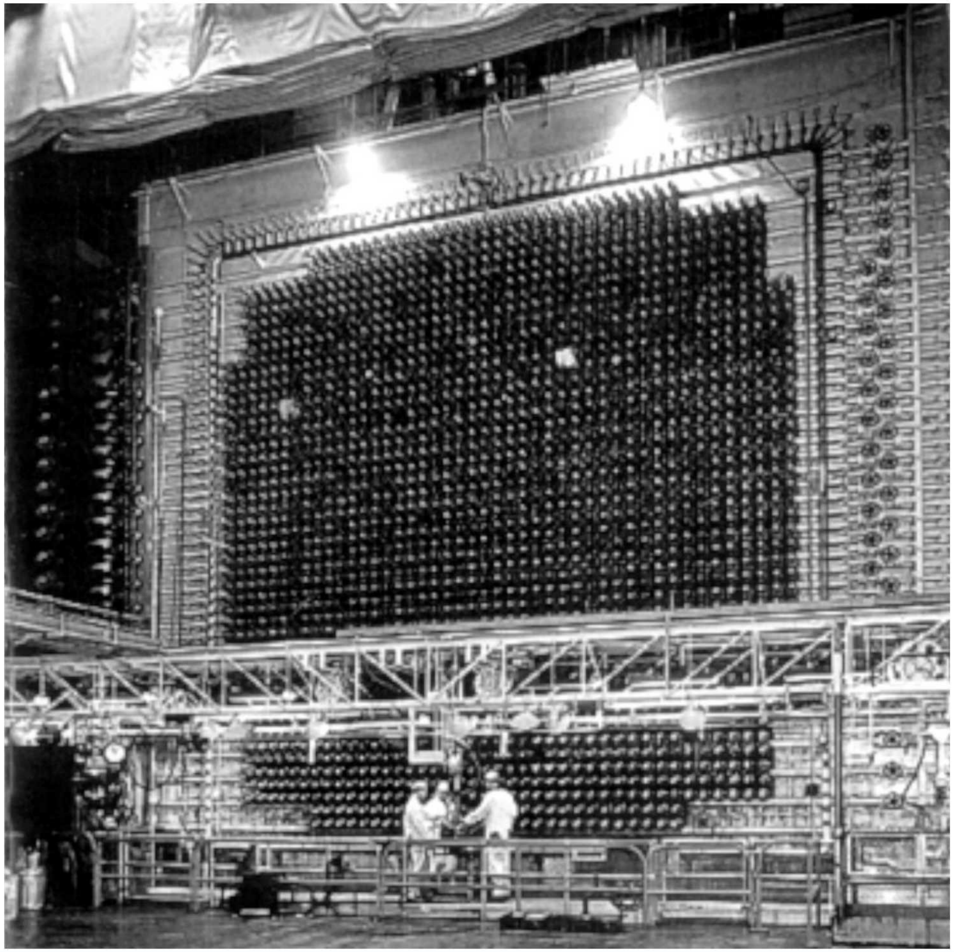
Ref: "Panda Book", values with ± have significant uncertainty

First Nuclear Reactors

ORNL X-10 Graphite Reactor
graphite moderated, air cooled

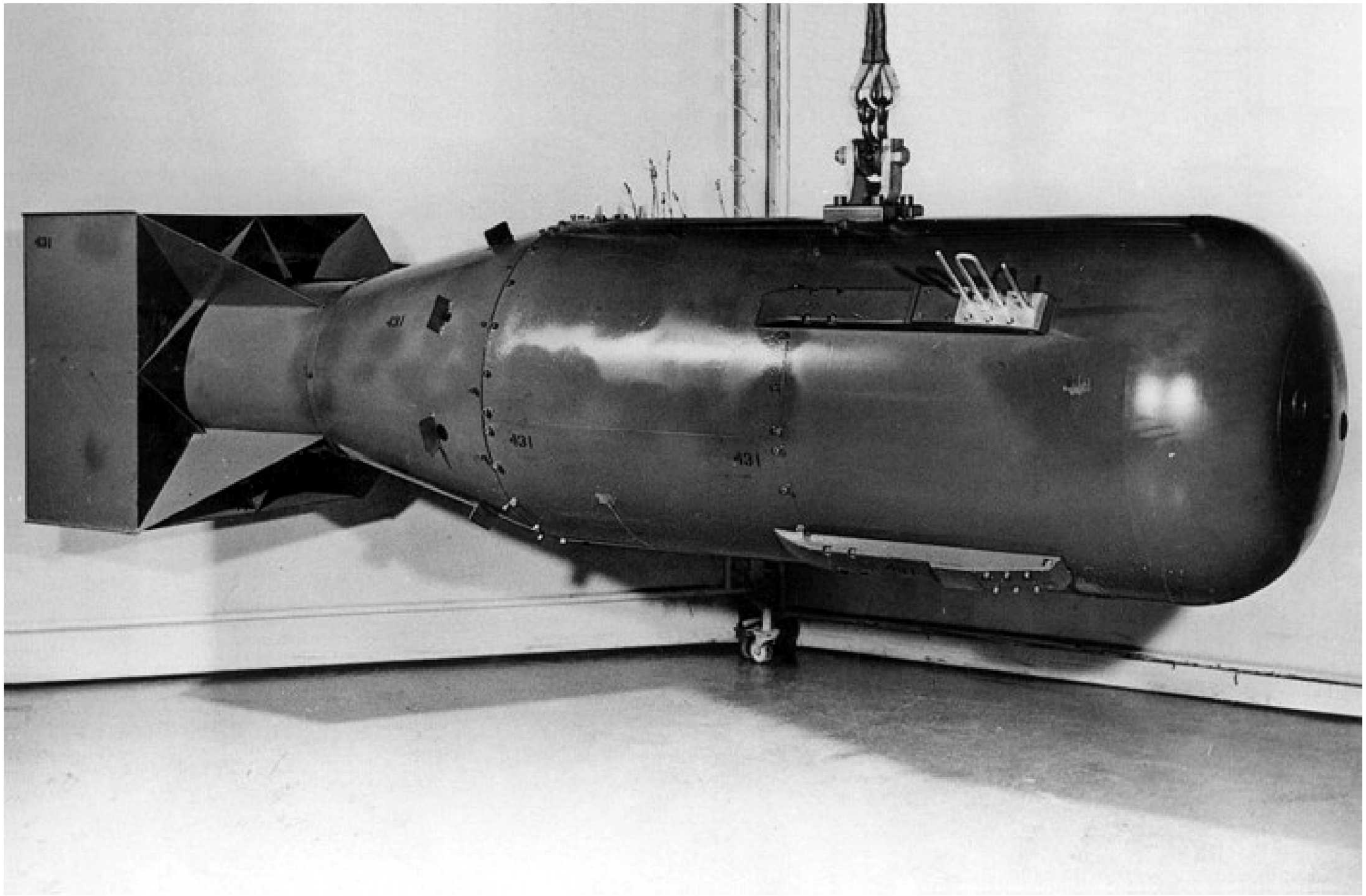


Hanford B Reactor
graphite moderated, water cooled



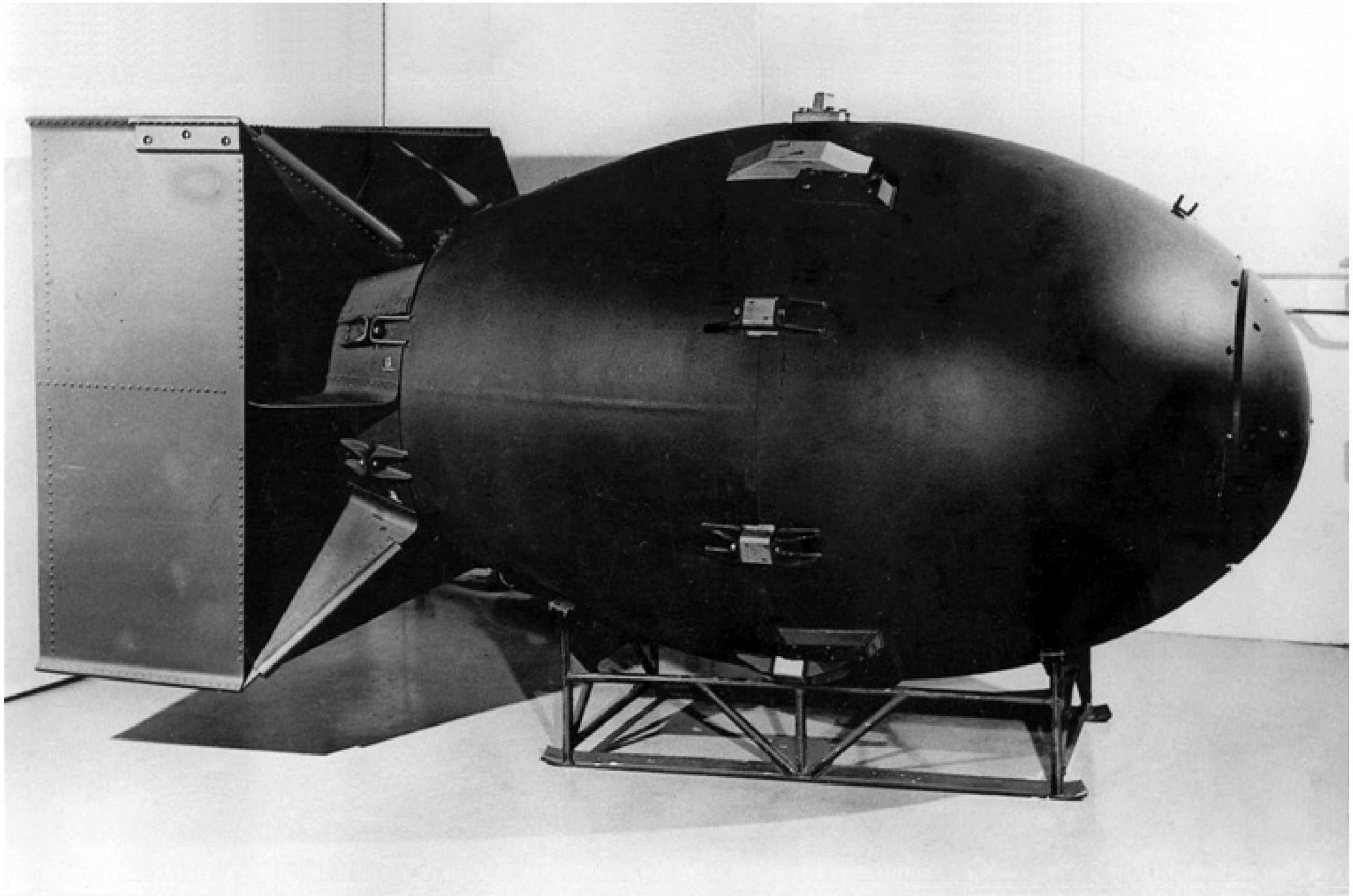
Little Boy

Highly Enriched Uranium (HEU): >20% U-235



Fat Man

Weapons Grade Plutonium (WGPu): >93% Pu-239

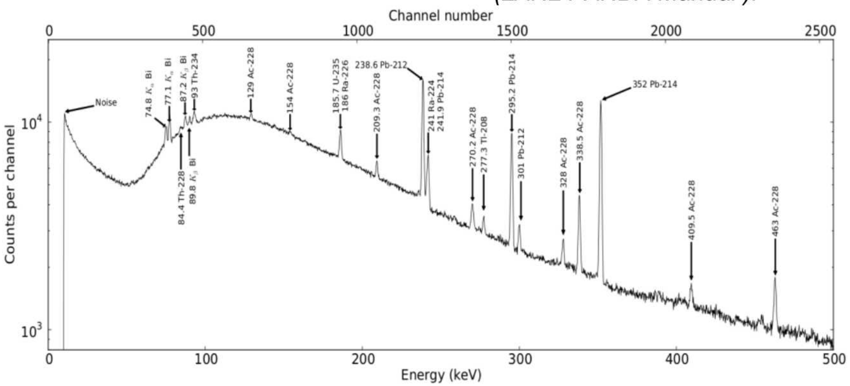


Current Methods

Measurable Unique Signatures of Fissile Material

1. Radiography
   - Geometry
2. Gamma spectrum
   - Isotopic content + surface area/shielding
3. Total neutron rate
   - Assay of the contents of specific materials
4. 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).*



Special Nuclear Material Detection

Sandia National Laboratories

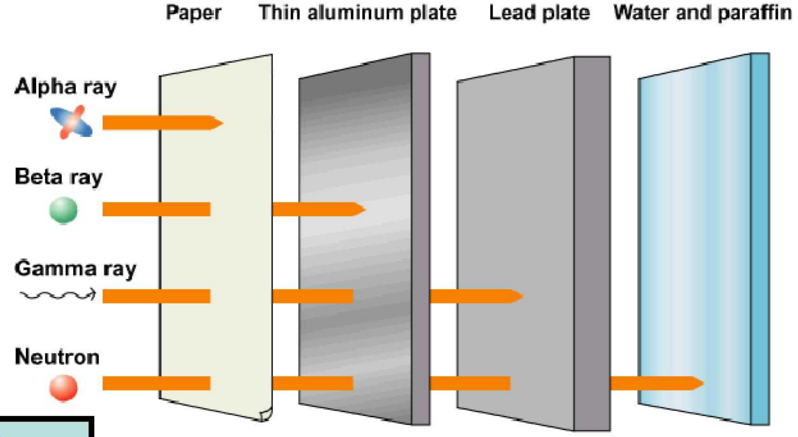
Gamma-rays

Isotope	Energy (keV)	Activity (γ/g-s)
^{234}U	120.9	9.35×10^4
^{235}U	143.8	8.40×10^3
	185.7	4.32×10^4
^{238}U	766.4	2.57×10^1
	1001.0	7.34×10^1
^{238}Pu	152.7	5.90×10^6
	766.4	1.387×10^5
^{239}Pu	129.3	1.436×10^5
	413.7	3.416×10^4
^{240}Pu	45.2	3.80×10^6
	160.3	3.37×10^4
	642.5	1.044×10^3
^{241}Pu	148.6	7.15×10^6
	208.0	2.041×10^7
^{241}Am	59.5	4.54×10^{10}
	125.3	5.16×10^6

Neutrons

Isotope	Half Life	Spontaneous Fission Yield (n/s-kg)
^{232}U	71.7 yr	1,300
^{233}U	1.59×10^5 yr	0.86
^{234}U	2.45×10^5 yr	5.02
^{235}U	7.04×10^8 yr	0.299
^{236}U	2.34×10^6 yr	5.49
^{238}U	4.47×10^9 yr	13.6
^{237}Np	2.14×10^6 yr	0.114
^{238}Pu	87.7 yr	2.59×10^6
^{239}Pu	2.41×10^4 yr	21.8
^{240}Pu	6.56×10^3 yr	1.02×10^6

Special Nuclear Material Detection – why neutrons?



www.remnet.jp

The Passive Gamma-Ray Signatures

Isotope	Energy (keV)	Activity (γ/g-s)	Mean Free Path (mm) (High-Z, ρ)	(Low-Z, ρ)
^{234}U	120.9	9.35×10^4	0.23	69
^{235}U	143.8	8.40×10^3	0.36	73
	185.7	4.32×10^4	0.69	80
^{238}U	766.4	2.57×10^1	10.0	139
	1001.0	7.34×10^1	13.3	159
^{238}Pu	152.7	5.90×10^6	0.40	75
	766.4	1.387×10^5	9.5	139
^{239}Pu	129.3	1.436×10^5	0.27	71
	413.7	3.416×10^4	3.7	106
^{240}Pu	45.2	3.80×10^6	0.07	25
	160.3	3.37×10^4	0.45	76
	642.5	1.044×10^3	7.4	127
^{241}Pu	148.6	7.15×10^6	0.37	74
	208.0	2.041×10^7	0.86	83
^{241}Am	59.5	4.54×10^{10}	0.14	38
	125.3	5.16×10^6	0.26	70

These materials are dense; self-shielding is not negligible

Ref: "Panda Book"

Table courtesy of David Chichester, INL

Current Methods
Drawbacks and Limitations

1. Geometry/Radiography
   - Insensitive to element/isotope type
   - Poor penetration
2. Gamma spectrum
   - Attenuation and self-shielding
3. Total neutron rate
   - Spontaneous & induced fission and (α,n) sources are indistinguishable
4. Correlated counts
   - Requires high efficiency, necessitates large detection system
   - Efficiency has to be well known
   - Detector lifetime of 10-30 μs is long compared to fission chain processes
   - Neutron energy information is lost due to moderation

→ He-3 based technologies

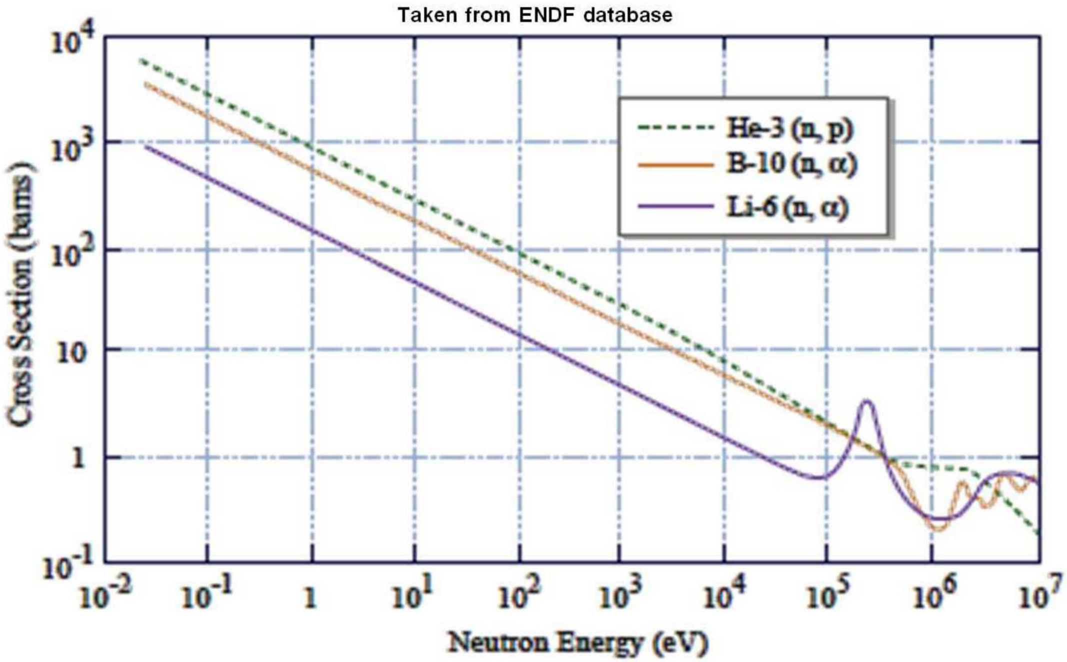
Thermal Neutron Detection

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

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

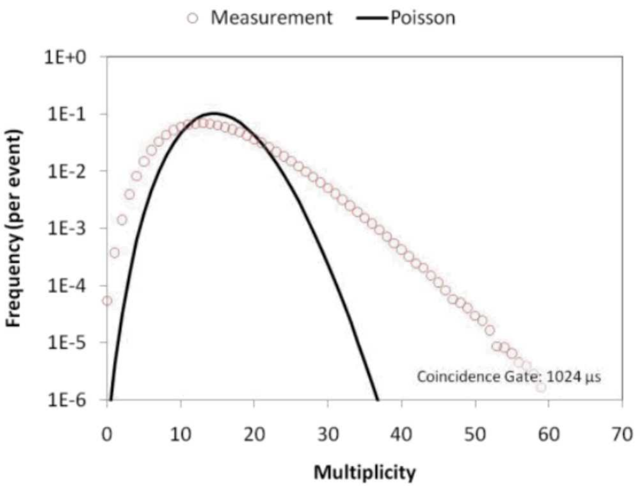
$$n + {}^{10}B \rightarrow {}^{7}Li + \alpha + 2.31 \text{ MeV}$$

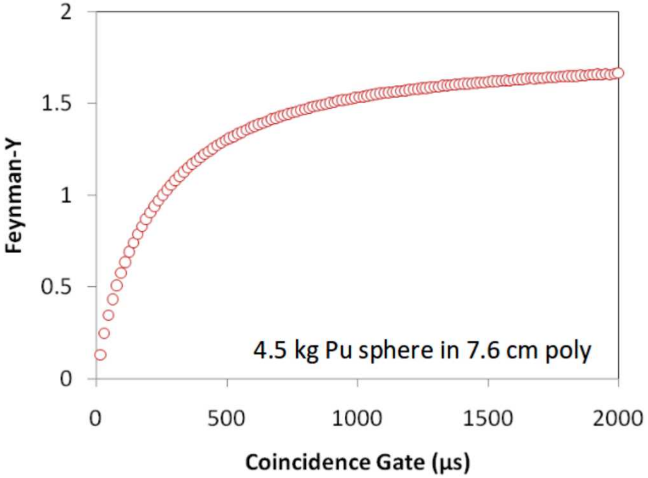
$$n + {}^{6}Li \rightarrow {}^{3}H + \alpha + 4.78 \text{ MeV}$$

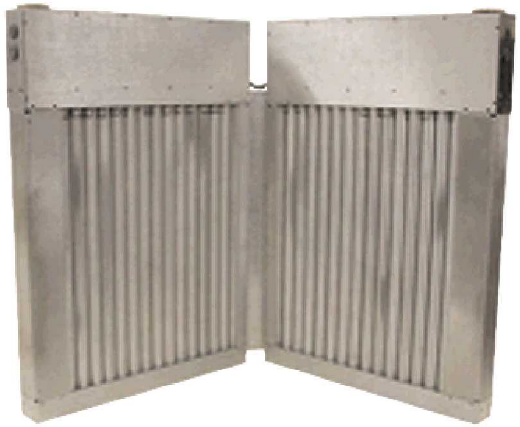


Feynman-Y Approach

- 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

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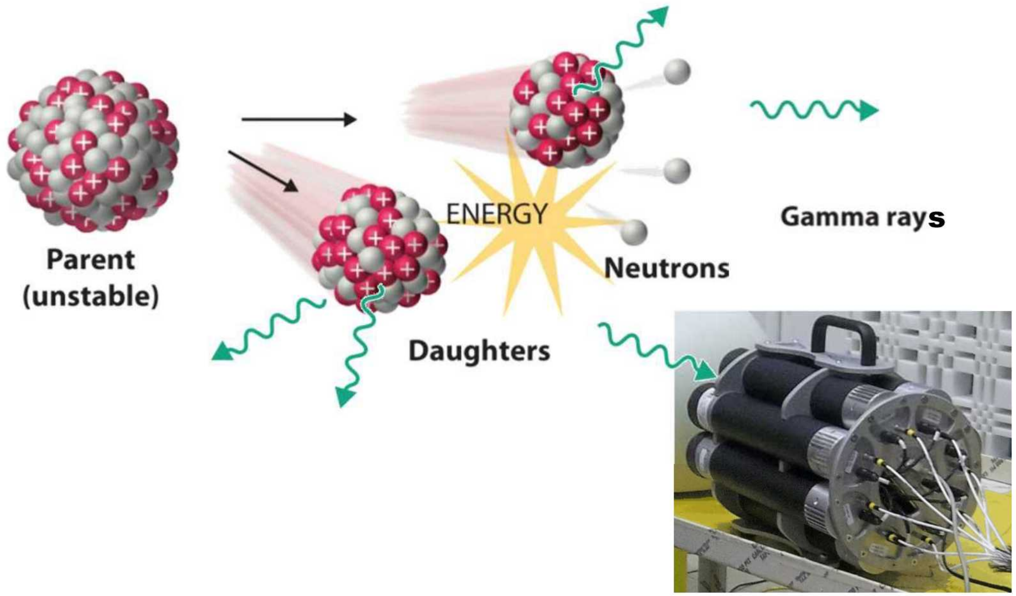
Neutron Coincidence Counting Equations

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

Targeting Multiplicity 2 correlations only

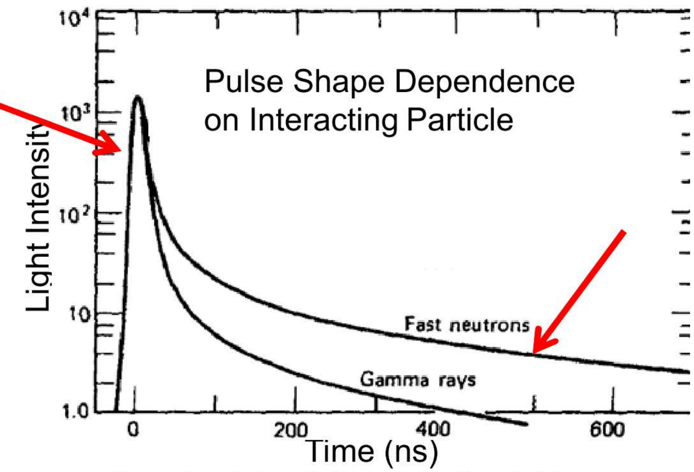


- Neutrons and gamma-rays are emitted nearly simultaneously during the fission process.
- Minimum multiplicity equals two:
  1. Gamma-gamma: great for timing, but a lot of detector cross talk and uncorrelated background.
  2. Neutron-neutron: without event by event energy, expected correlated timing is spread on the order of the spread in fission chain dynamics.
  3. Gamma-neutron: gamma starts precise clock, neutron creates certainty that fission has taken place (also more penetrating).

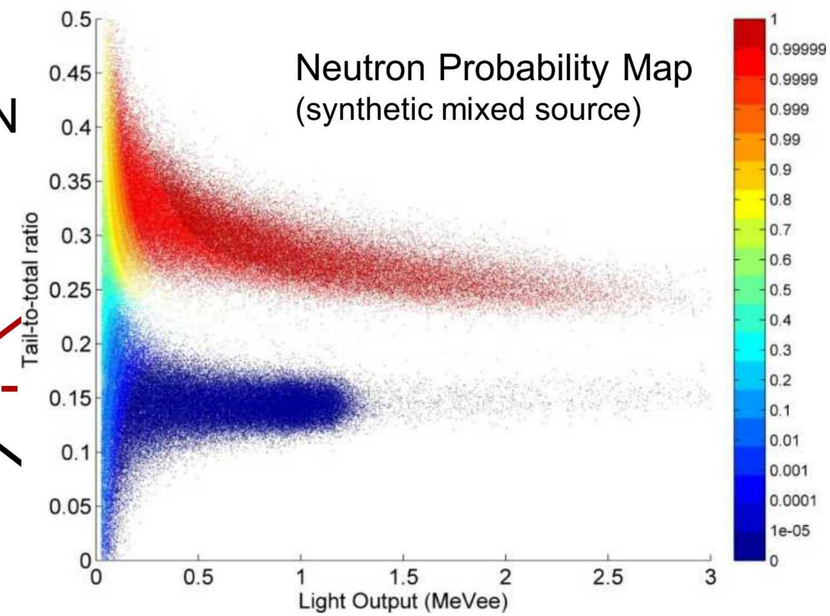
Fast Organic Scintillators

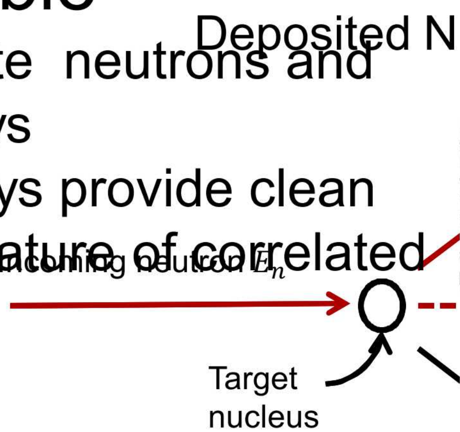
Three main new advantages

1. Fast rise time
   - Sub-nanosecond timing resolution
2. Incident neutron energy
   - Light output proportional to proton recoil
3. PSD Capable
   - Discriminate neutrons and gamma rays
   - Gamma rays provide clean timing signature of correlated event



Glenn Knoll Radiation Detection & Measurement 3rd Edition





Same Fission Gamma-Neutron Correlations

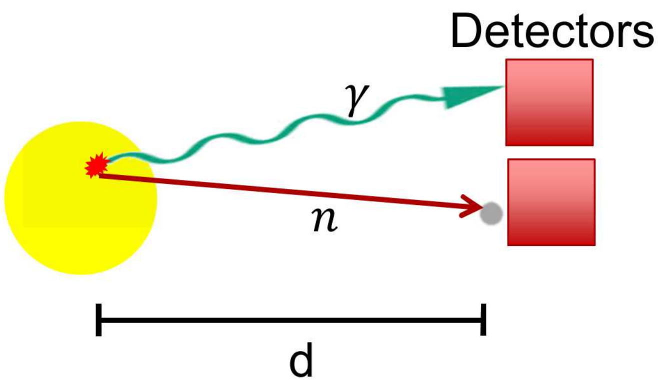
Time Of Flight Fixed by Estimated Energy (TOFFEE)

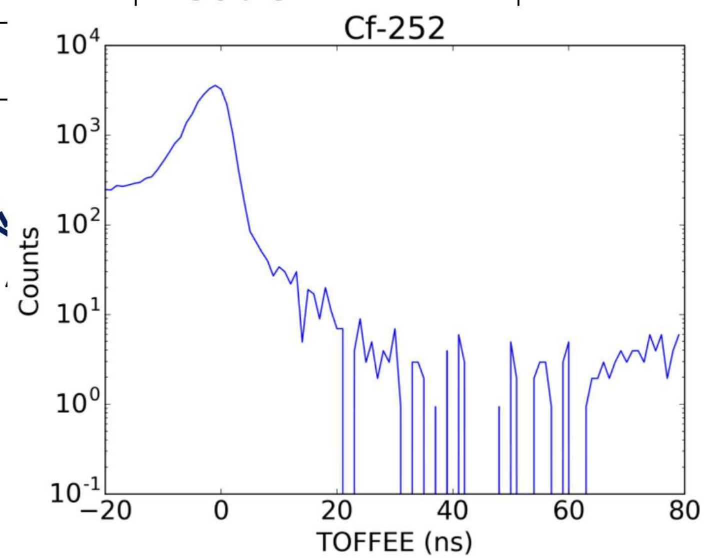
E_n: neutron energy
R_γ: distance traveled by gamma ray
R_n: distance traveled by neutron

$$\Delta T_{n-\gamma} - \left(R_n \sqrt{\frac{m}{2E_n}} - \frac{R_\gamma}{c} \right) = 0$$

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

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

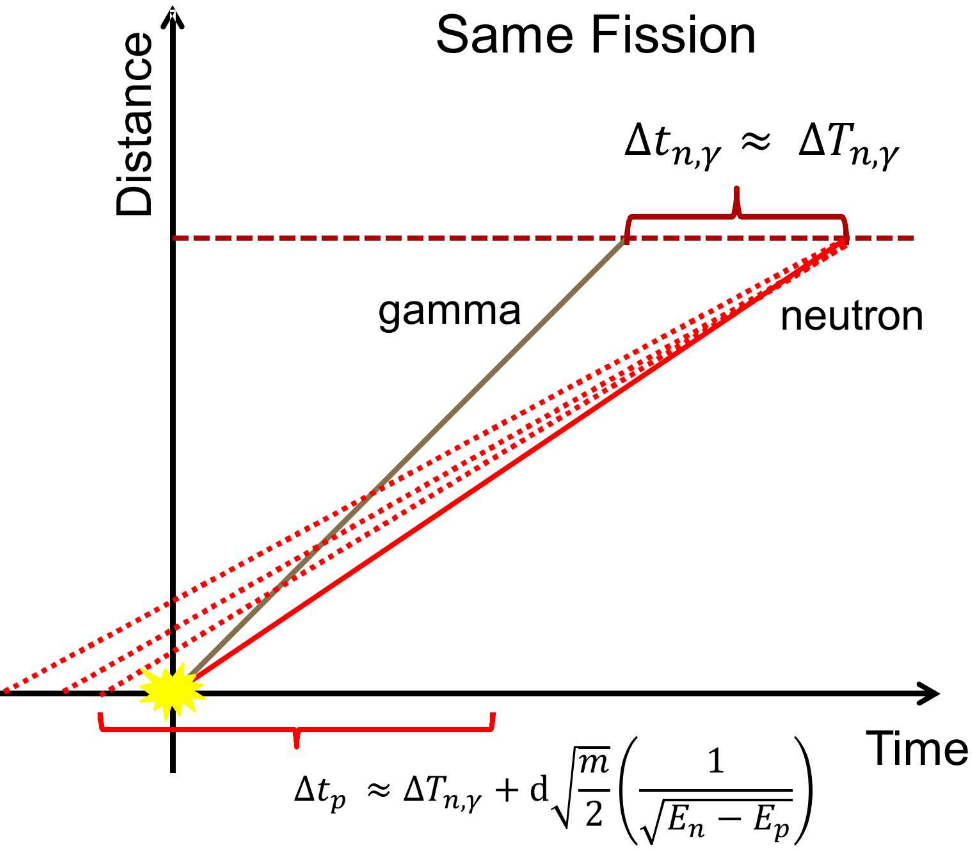




Measured Quantities:

- Time between correlated γ and neutron: $\Delta t_{n,\gamma}$
- Proton recoil energy: $E_p \leq E_n$
- Source-to-detector distance: $d \approx R_n = R_\gamma$

Same Fission Gamma-Neutron Detection Space-Time

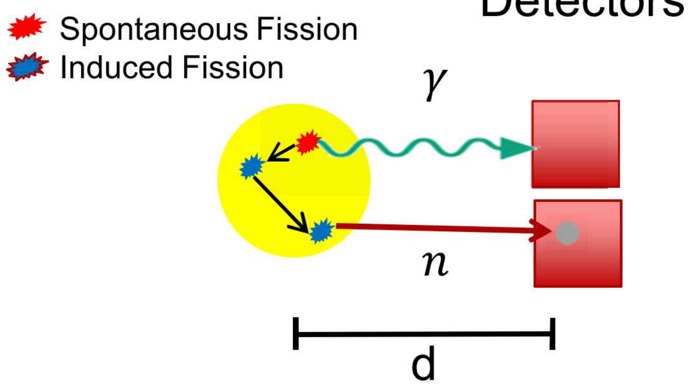


1. Neutron and gamma are born from the same fission event
2. They travel the same distance the detectors
3. The measured and actual difference in time-of-flight is equivalent
4. When subtracting estimated time of flight of the neutron, the result is less than zero

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

Neutron-Gamma Correlation – Fission Chain

Time Of Flight Fixed by Estimated Energy (TOFFEE)



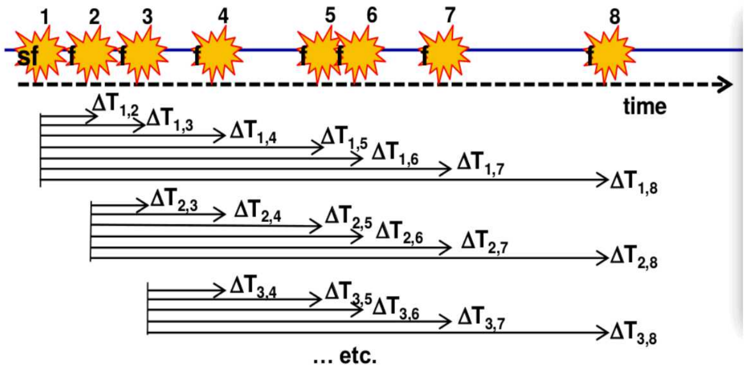
Measured time difference between neutron and nearest γ

$$\Delta t_{n-\gamma} - \Delta t_p \leq \Delta T_g$$

Time difference between fission events in a chain

Estimated travel time difference between neutron and gamma

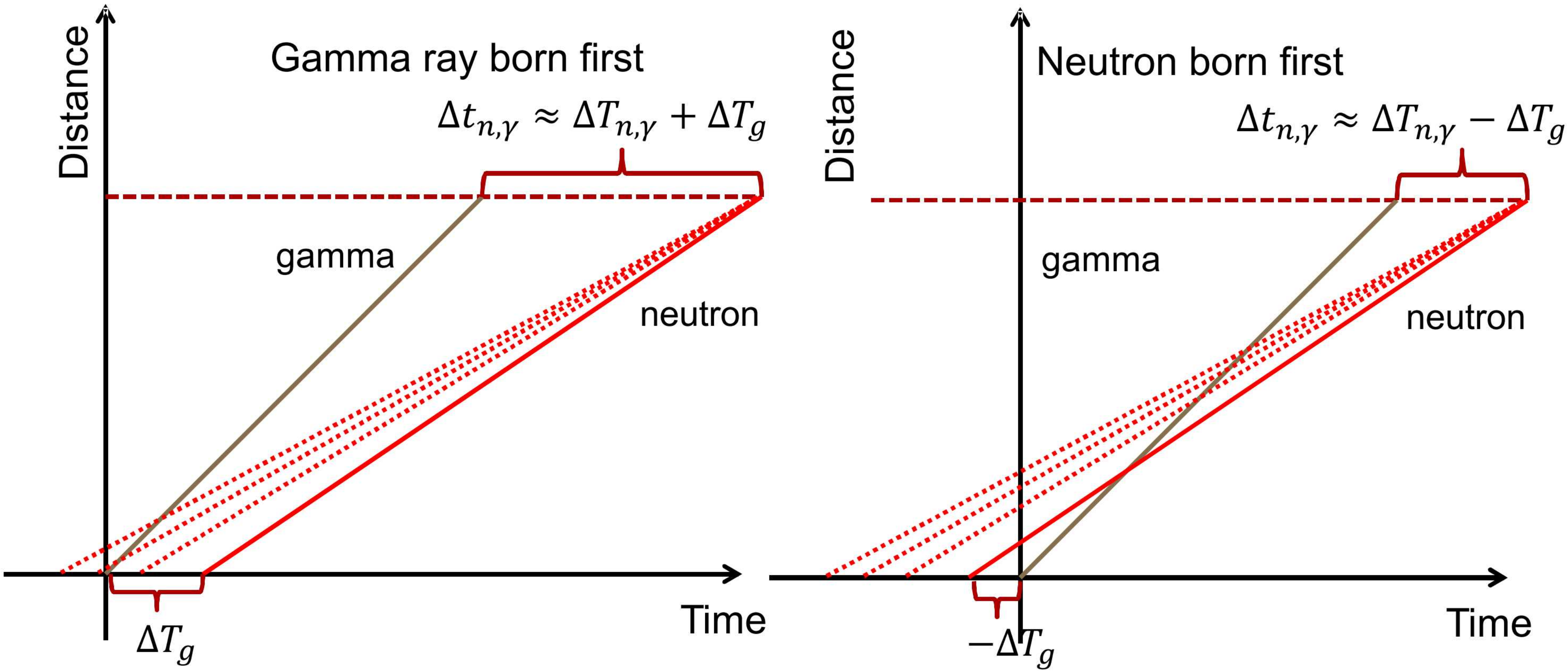
Distribution of times between fission events in a chain



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

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

Fission Chains Gamma-Neutron Space-Time



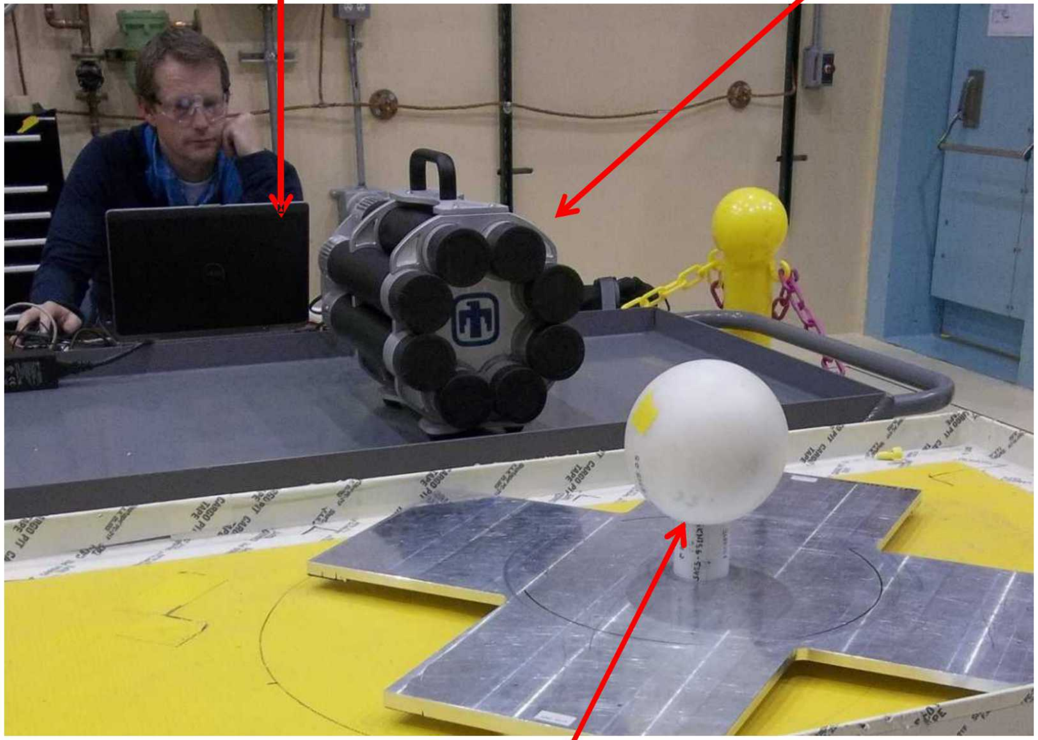
$$\Delta t_{n,\gamma} - \Delta \mathrm{t_p} \leq \Delta T_g$$

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

Correlated Radiation Signatures - Stilbene Array

Computer/digitizer

Stilbene array



Beryllium Reflected Plutonium (BeRP) ball, 4.5 kg WGPu in a 1” shell of High Density Polyethylene

TOFFEE Sensitivity to Fissile Material and Configuration

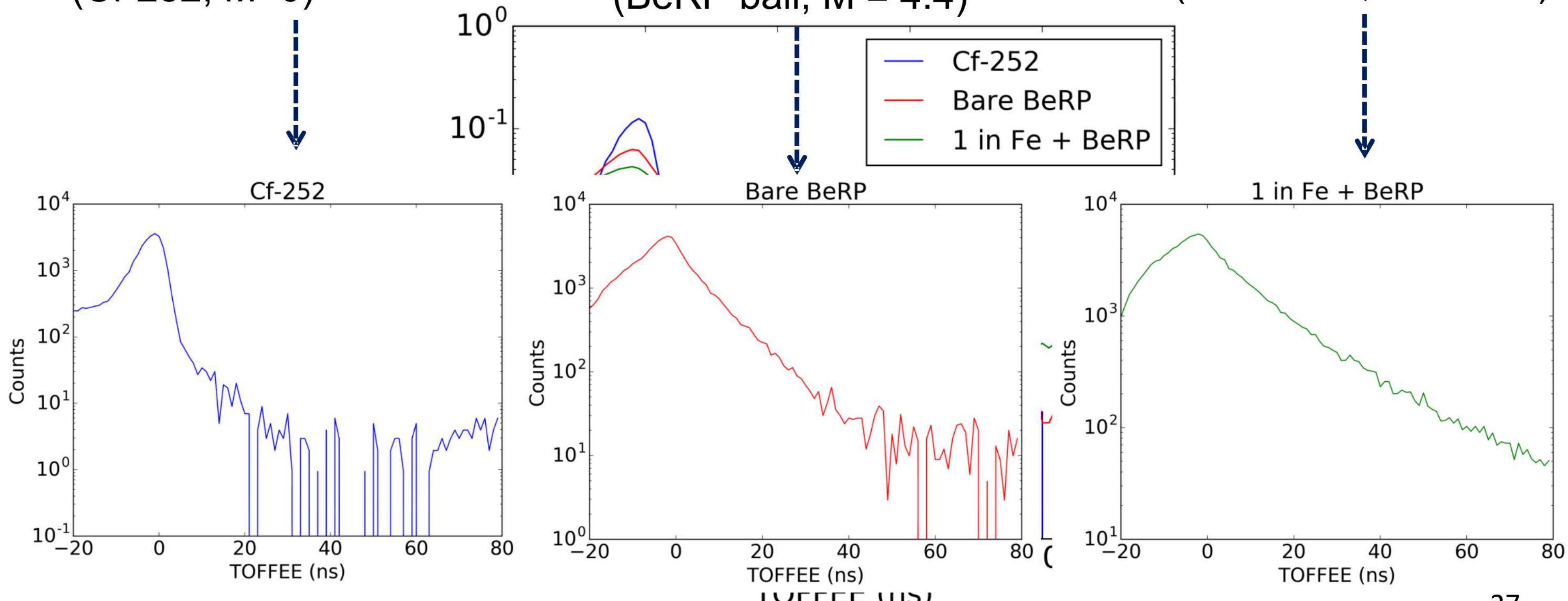
TOFFEE is sensitive to:

1. Fissile vs. fissionable
2. Fissile materials with different neutron multiplication
3. Bare vs. reflected fissile cores

Non-fissile fission source
(Cf-252, M=0)

Fissile material
(BeRP ball, M = 4.4)

Reflected Fissile
material
(BeRP ball, M = 6.48)



Extracting Physical Parameters from TOFFEE

Parametric Experiments and Simulations

- TOFFEE distribution was shown to discriminate between fissile assemblies
- A physical model is required to extract physical parameters by fitting the TOFFEE distribution
- Use point-kinetics model (energy and spatially independent), which describes the neutron population change, which tracks with the density of fissions in a fissile material

Measurements of BeRP ball

- Bare
- 0.5 in Steel
- 1.0 in Steel
- 1.5 in Steel
- 1.0 in Nickel

Simulations

- 0.5 – 6 in Steel
- 0.5 – 6 in Nickel
- 0.5 – 6 in Tungsten
- 0.5 – 6 in Aluminum

Two-region point kinetics

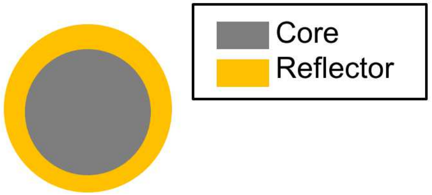
Neutron multiplication

Return from reflector

Loss in the core (abs & leakage)

Core:

$$\frac{dN_c}{dt} = \frac{k_c}{l_c} N_c + f_{rc} \frac{N_r}{l_r} - \frac{N_c}{l_c}$$



Influx from the core

Loss in the reflector

Reflector:

$$\frac{dN_r}{dt} = f_{cr} \frac{N_c}{l_c} - \frac{N_r}{l_r}$$

N_c & N_r: the number of neutrons in the fissile core and reflector regions

l_c & l_r: neutron lifetimes in the fissile core and reflector regions

k_c : multiplication factor in the fissile core region

$f = f_{cr} * f_{rc}$: fraction of neutrons that leak to reflector AND return to the core

Solution is a double exponential

$$N_c(t) = N_o[(1 - R)e^{tr_1} + Re^{tr_2}]$$

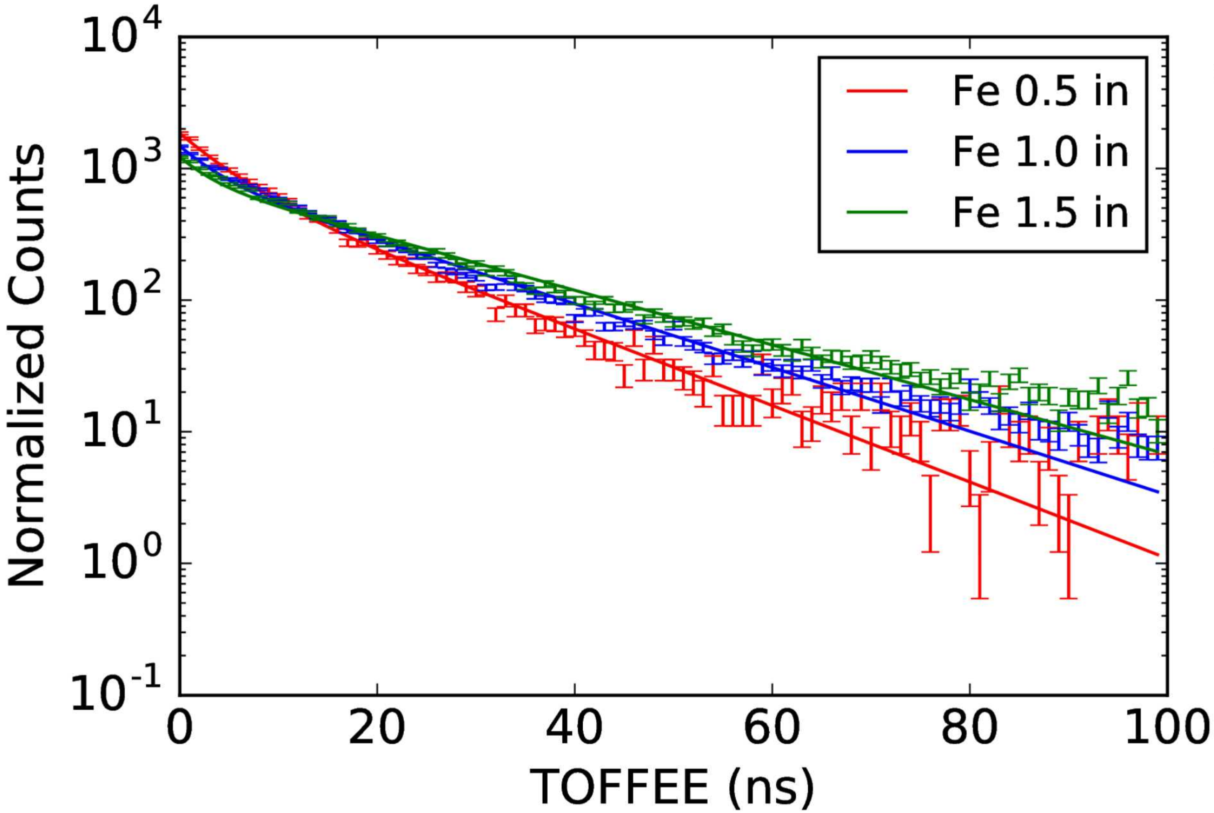
$$r_{1,2} = \frac{\pm\sqrt{4l_c l_r(f + k_c - 1) + \left(l_c - l_r(k_c - 1)\right)^2} - l_c + l_r(k_c - 1)}{2l_c l_r}$$

Scale Parameter: $R = \dfrac{r_1 - \alpha}{r_1 - r_2}$

Rate parameter
(in a bare configuration): $\alpha = \dfrac{k_c - 1}{l_c}$

Fitting Strategy for Reflected Configurations

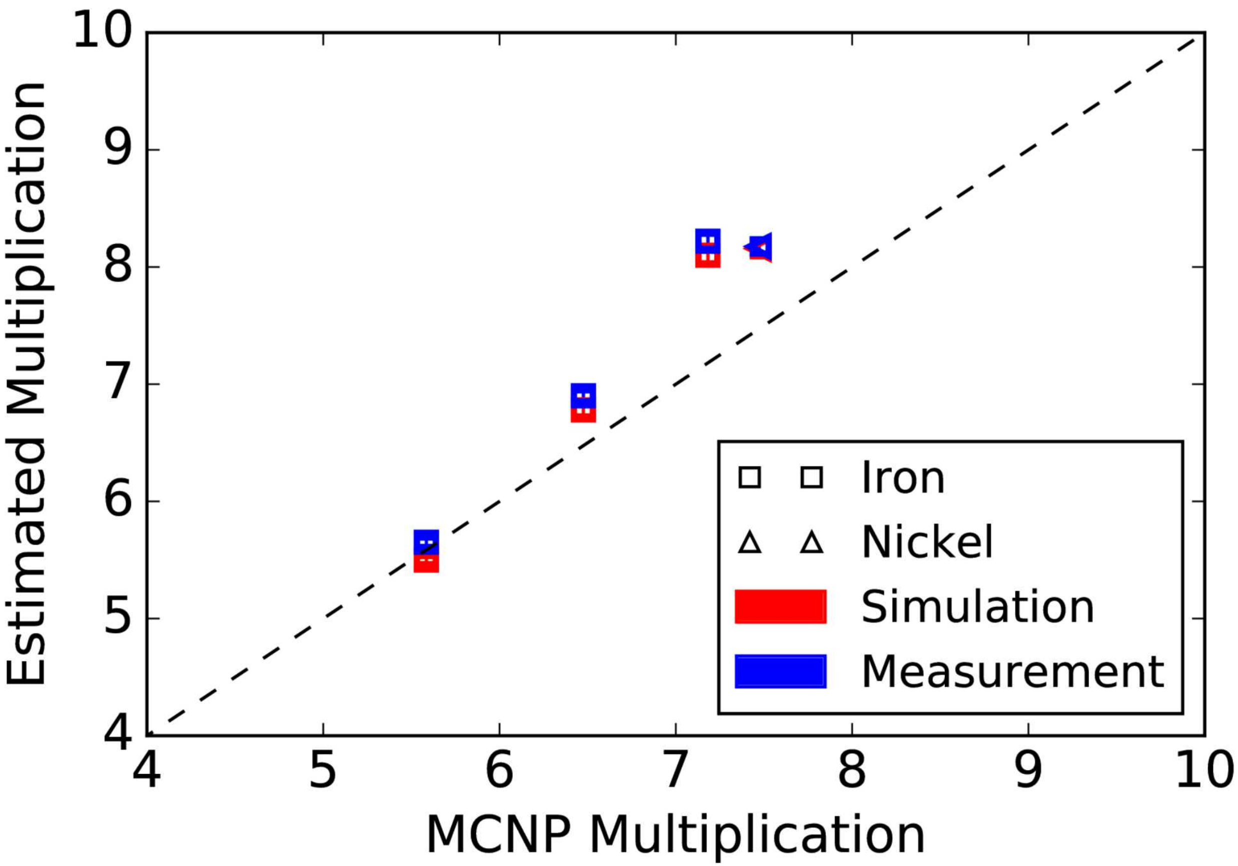
$$N_c(t) = N_o[(1-R)e^{tr_1} + Re^{tr_2}]$$



- Allowing reflection fraction (f) and neutron lifetime (l_r, l_c) produces muddled results
- Solution: fix k_c and l_c to what the core parameters are from the fit of the bare case

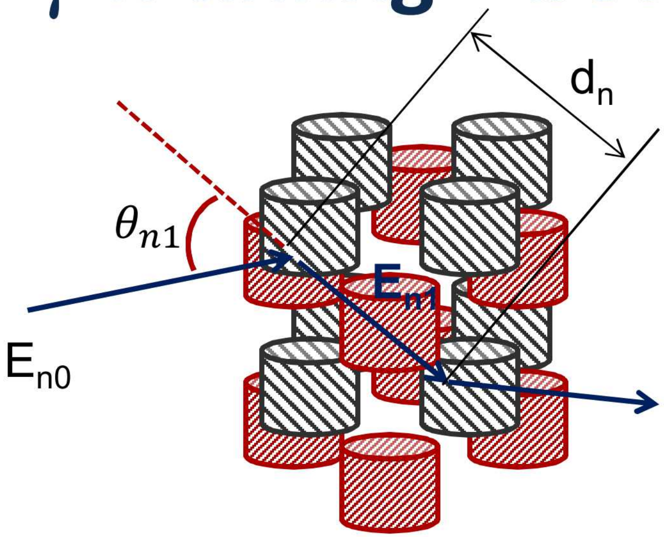
Estimating Multiplication of Reflected Assemblies

Measurement vs Simulation



$$k = \frac{k_c}{1 - f}$$

γ-n timing + better energy estimate



$$\Delta t_{n-\gamma} = \left(R_n \sqrt{\frac{m}{2E_{n0}}} - \frac{R_\gamma}{c} \right) + \Delta T_{f_1,f_2}$$

Time of arrival difference between γ and neutron	Time-of-flight difference between γ and neutron	Time difference between fissions in a chain

$$E_{n1} = \frac{m_n}{2} \times \frac{{d_n}^2}{TOF^2}$$

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

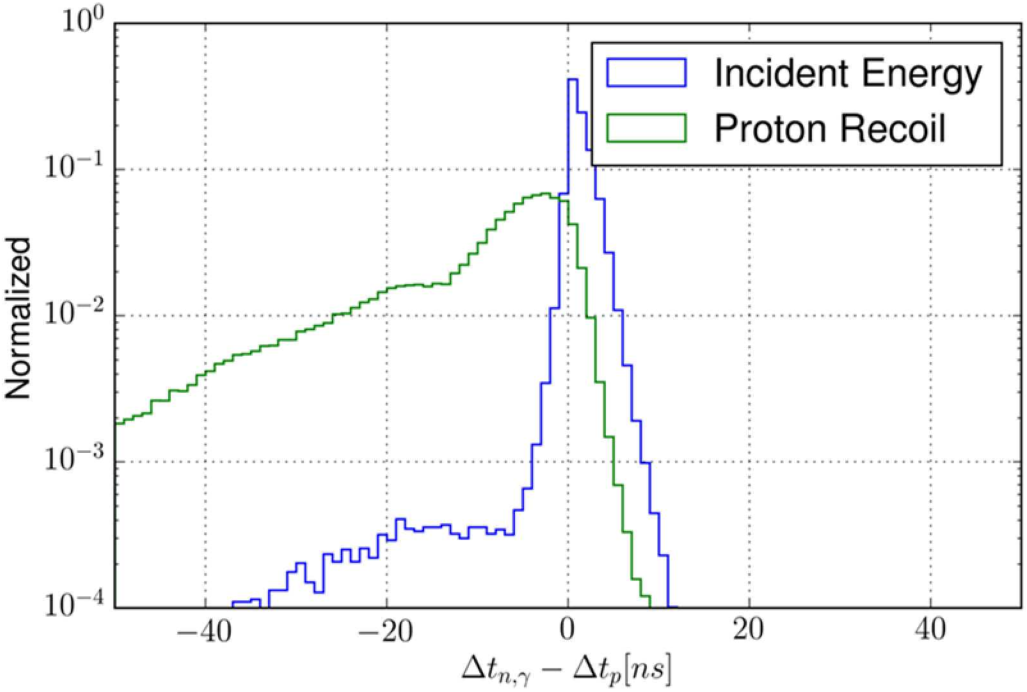
$$cos^2\theta_{n1} = \frac{E_{n1}}{E_{n0}}$$

MINER: the Mobile Imager of Neutrons for Emergency Response

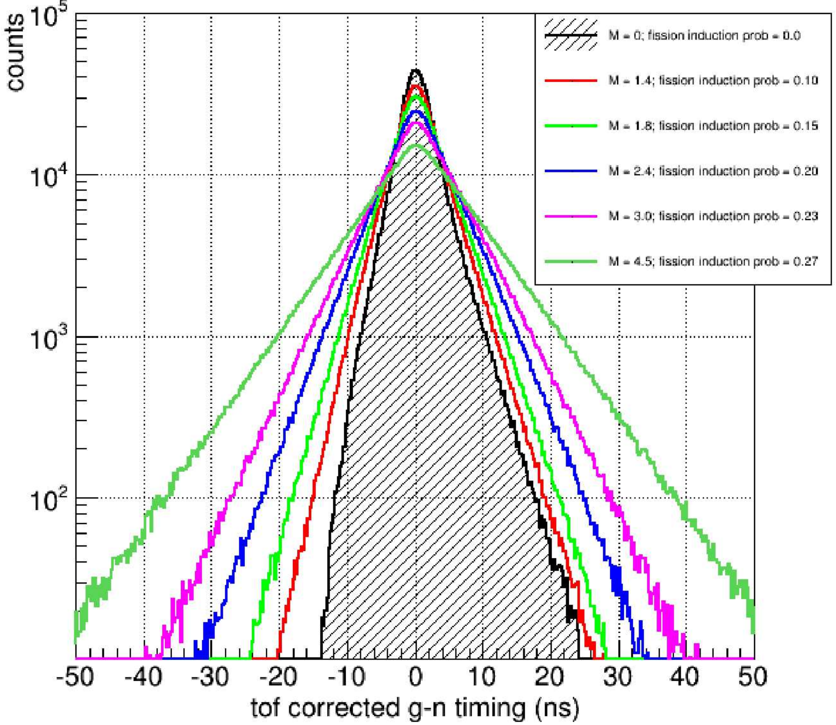
16 independent 3”x3” EJ-309 liquid scintillator cells

$(\Delta t_{n-\gamma} - \Delta t_{En0})$ Distributions

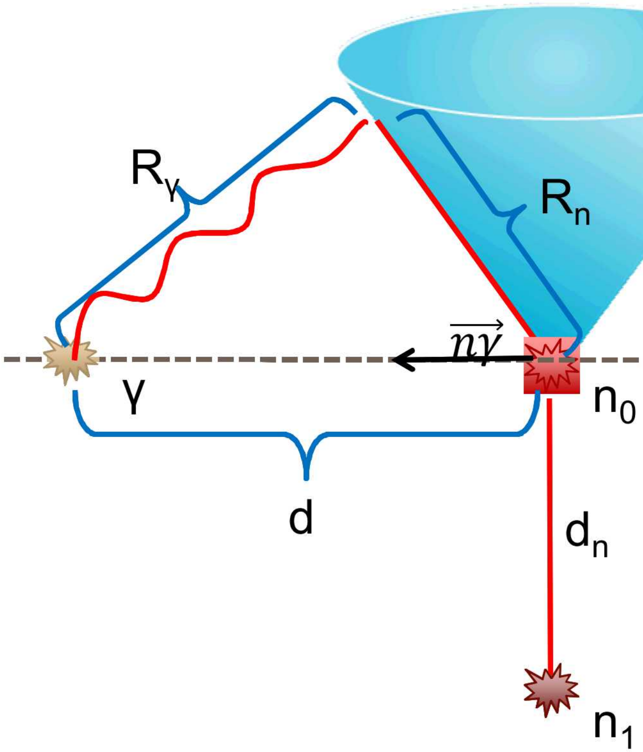
Cf-252 Simulation



Different Multiplications



γ-n-n 3-D reconstruction



Double scattered neutron:

1. Energy (or velocity) of the incident neutron

$$v_n = \sqrt{2E_p m_n + \left(\frac{d_n}{\Delta t_{n_0,n_1}}\right)}$$

2. Cone of possible source locations

$$\cos^2 \theta_{n_1} = \frac{E_{n_1}}{E_{n_0}}$$

Correlated gamma:

Distance to the source
(constrained by conical surface)

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

$$R_n = \frac{c^2 \Delta t v_n - d v_n^2 \mu + \sqrt{v_n^2\left(c^2(\Delta t^2 v_n^2 - 2 d v_n \mu \Delta t + d^2) + v_n^2 d^2(\mu^2 - 1)\right)}}{c^2 - v_n^2}$$

where

$$\mu = \frac{\overrightarrow{n\gamma}}{d} \cdot \widehat{R_n}$$

γ-n-n 3-D reconstruction – single event

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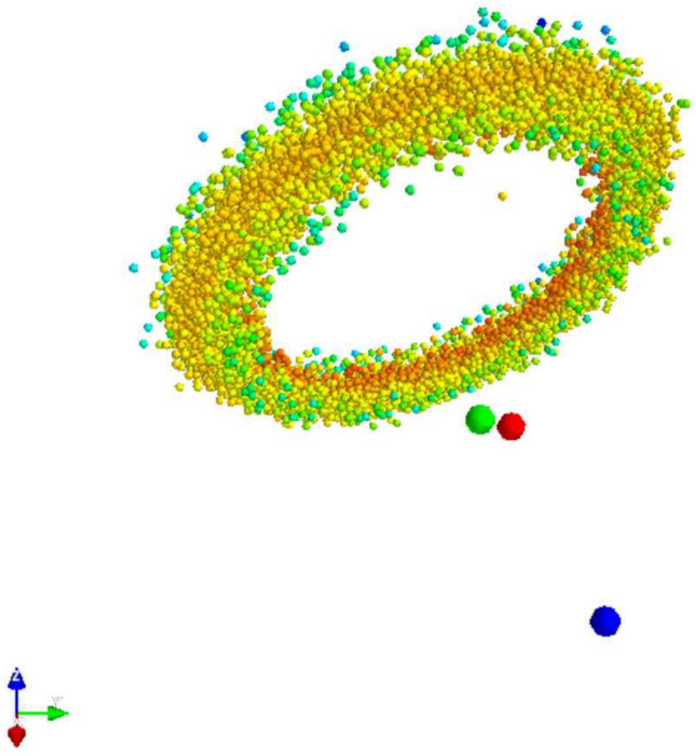
Single correlated event back-projection



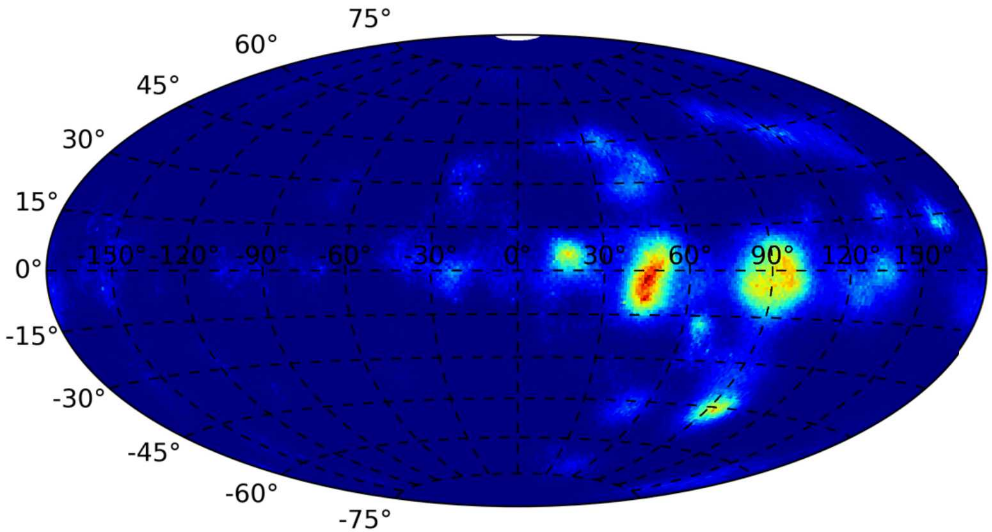
$$cos^2\theta_{n1} = \frac{E_{n1}}{E_{n0}}$$

+

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

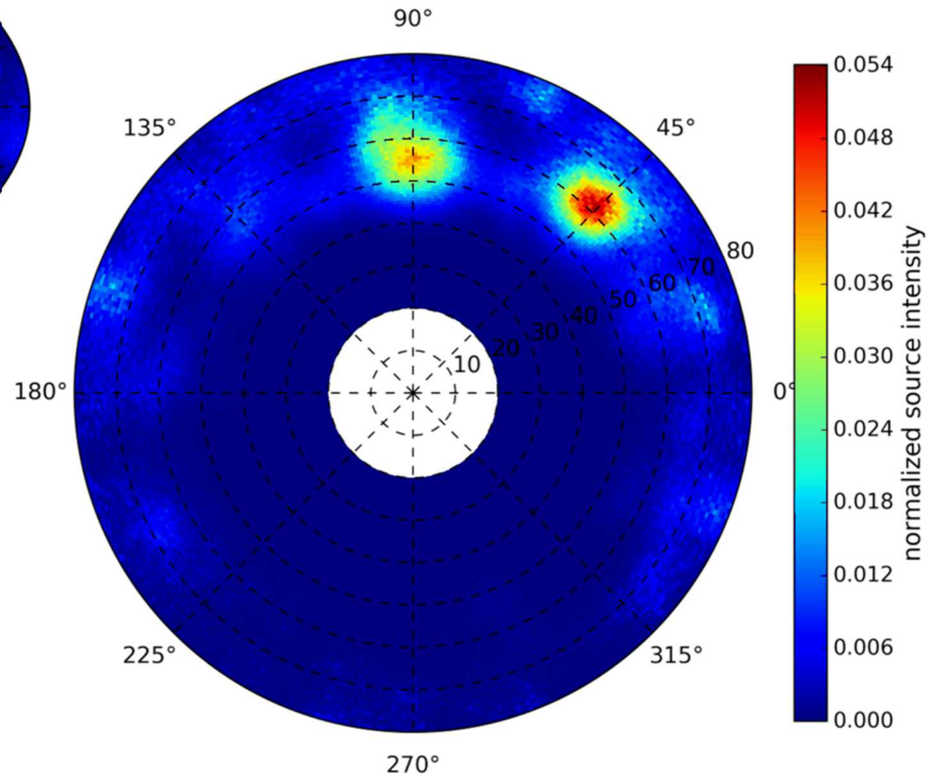
γ-n-n 3-D reconstruction

(Stochastic Origins Ensemble Reconstruction (SOE)),
Two Cf-252 fission sources, 50 & 60 cm from MINER



2D angular view

“top down” view



γ-n-n 3-D reconstruction

Stochastic Origin Ensemble Reconstruction
(~10,000 events with MINER, Cf-252 at 50cm and 60 cm)

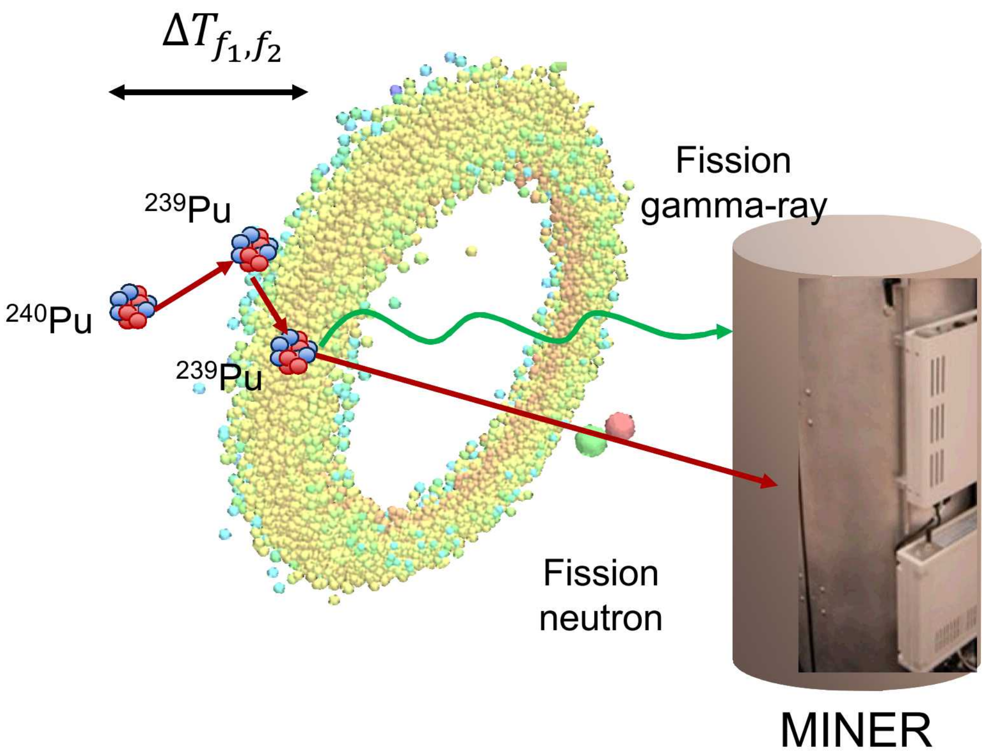
$$cos^2\theta_{n1} = \frac{E_{n1}}{E_{n0}}$$

+

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

- **Enables 3-D reconstruction.**
- **Can be achieved with a single view when accessibility and/or time is an issue.**

Event level characterization and imaging (fission chain length and 3D localization)



Material characterization:

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

(metal vs. oxide vs. fluoride vs. other (alpha,n))

3D localization and characterization:

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

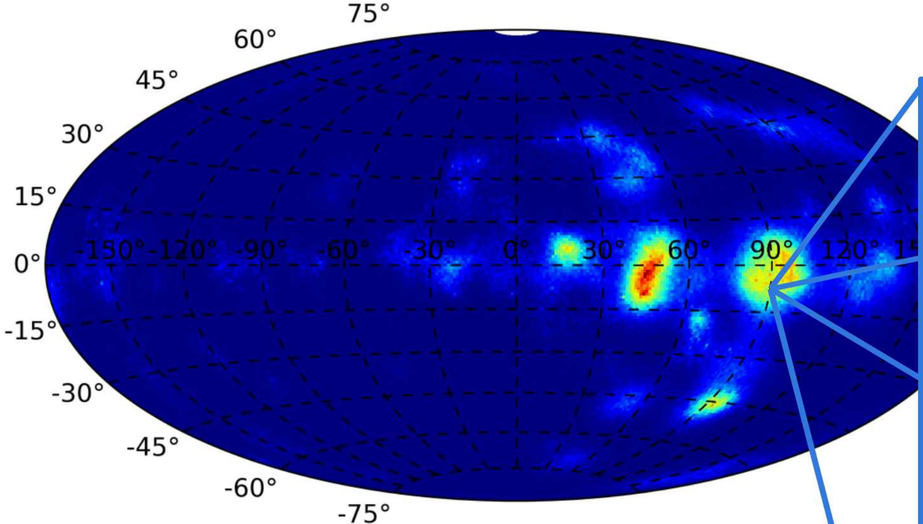
$$\Delta t_{n-\gamma} = \frac{R_n}{v_n} - \frac{R_\gamma}{c} + \Delta T_{f_1,f_2}$$

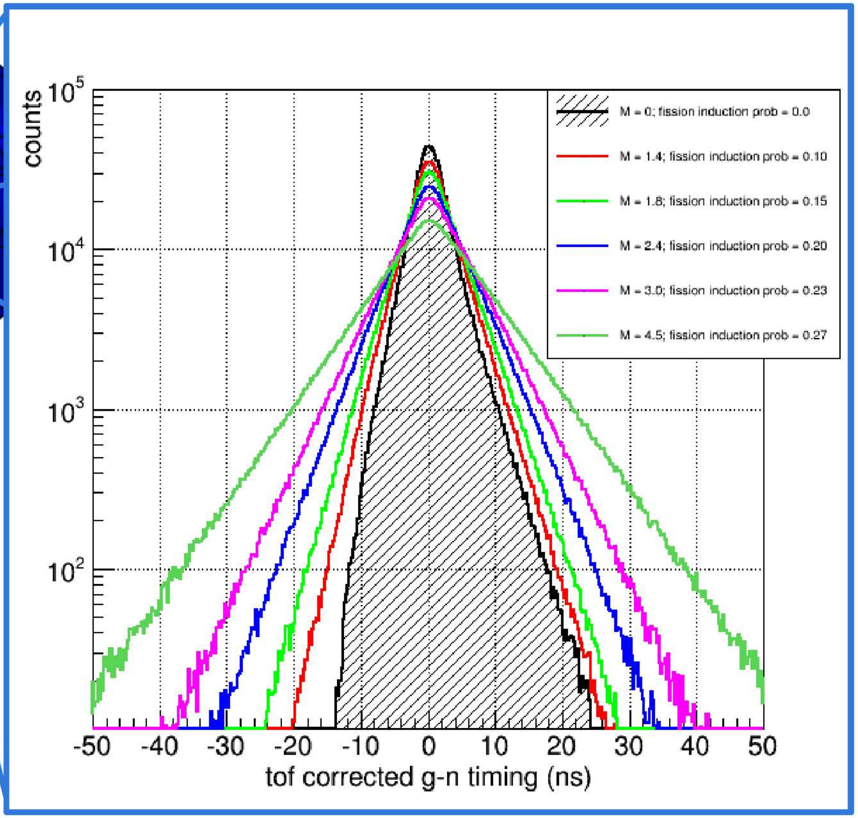
(average length of fission chains in a voxel; a.k.a. Multiplication M)

Multiplication within a voxel

$$\Delta T_{f_1,f_2} = \frac{R_n}{v_n} - \frac{R_\gamma}{c} - \Delta t_{n-\gamma}$$

is proportional to the average length of fission chains in a voxel (Multiplication M)

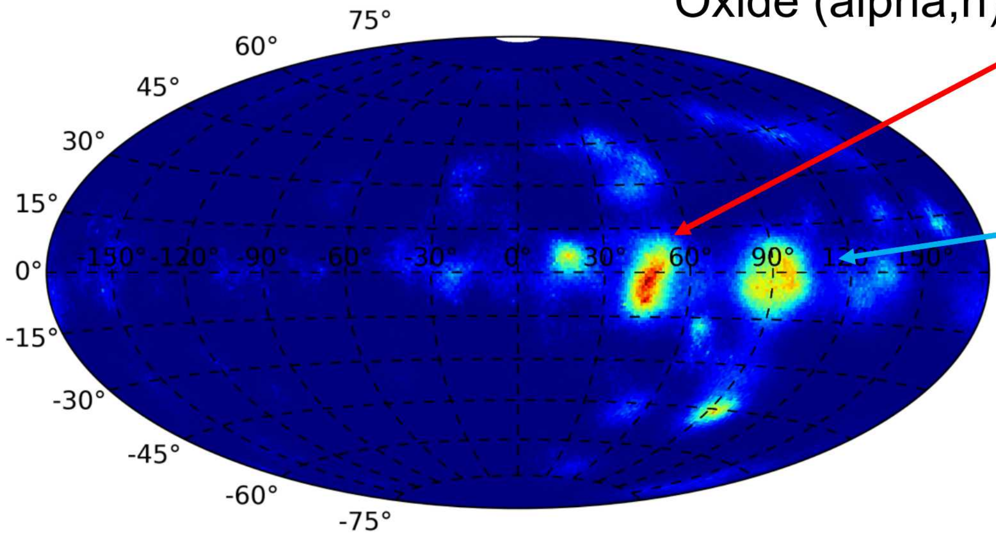




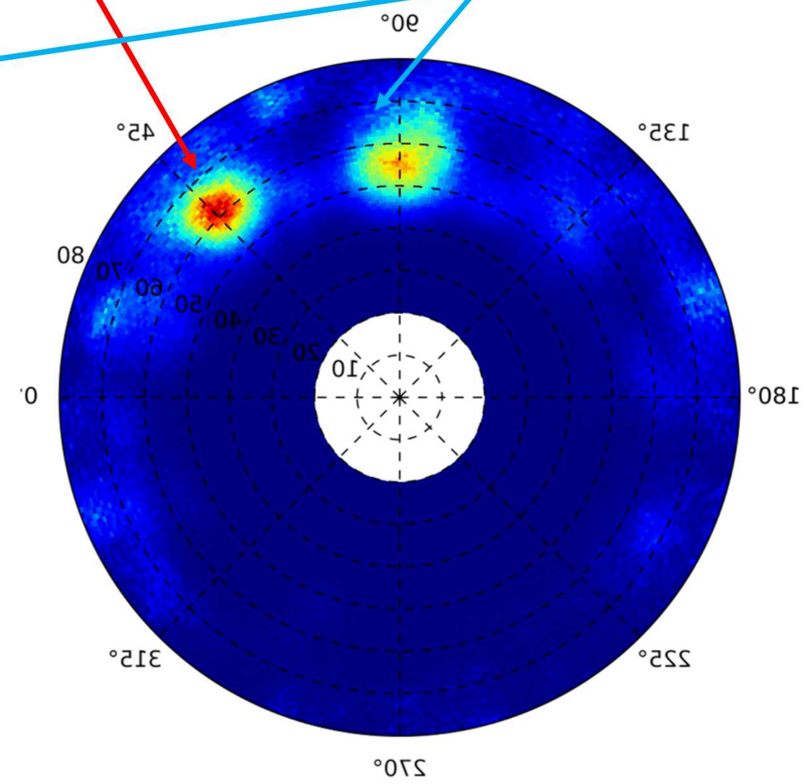
Pipe dream #1 (Multiplication and 3D localization):

Oxide (alpha,n)(Multiplication = M_1)

Fissile source (Multiplication = M_2)

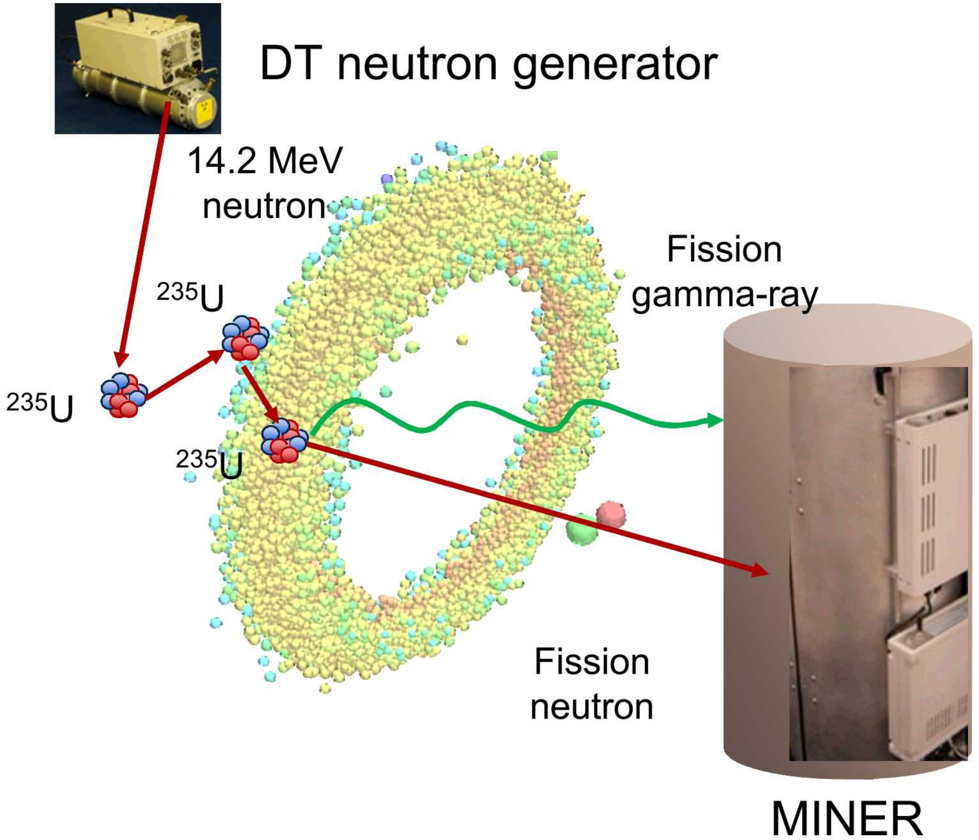


2D angular view



“top down” view

Event level characterization and imaging (Active - M and 3D Imaging for Uranium)



Material characterization:

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

(metal vs. oxide vs. fluoride vs. other (alpha,n))

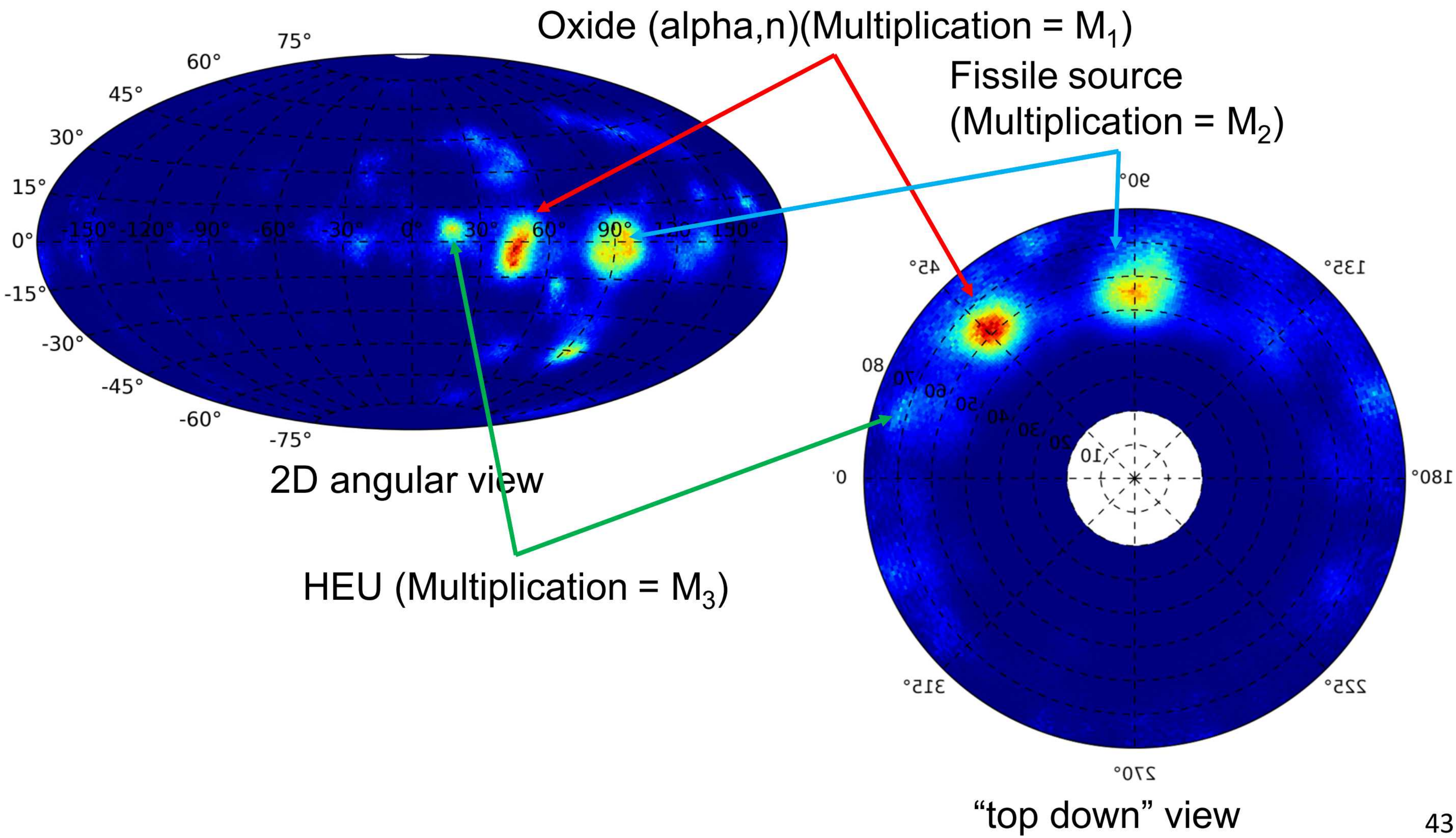
3D localization and characterization:

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

$$\Delta t_{n-\gamma} = \frac{R_n}{v_n} - \frac{R_\gamma}{c} + \Delta T_{f_1,f_2}$$

(average length of fission chains in a voxel; a.k.a. Multiplication M)

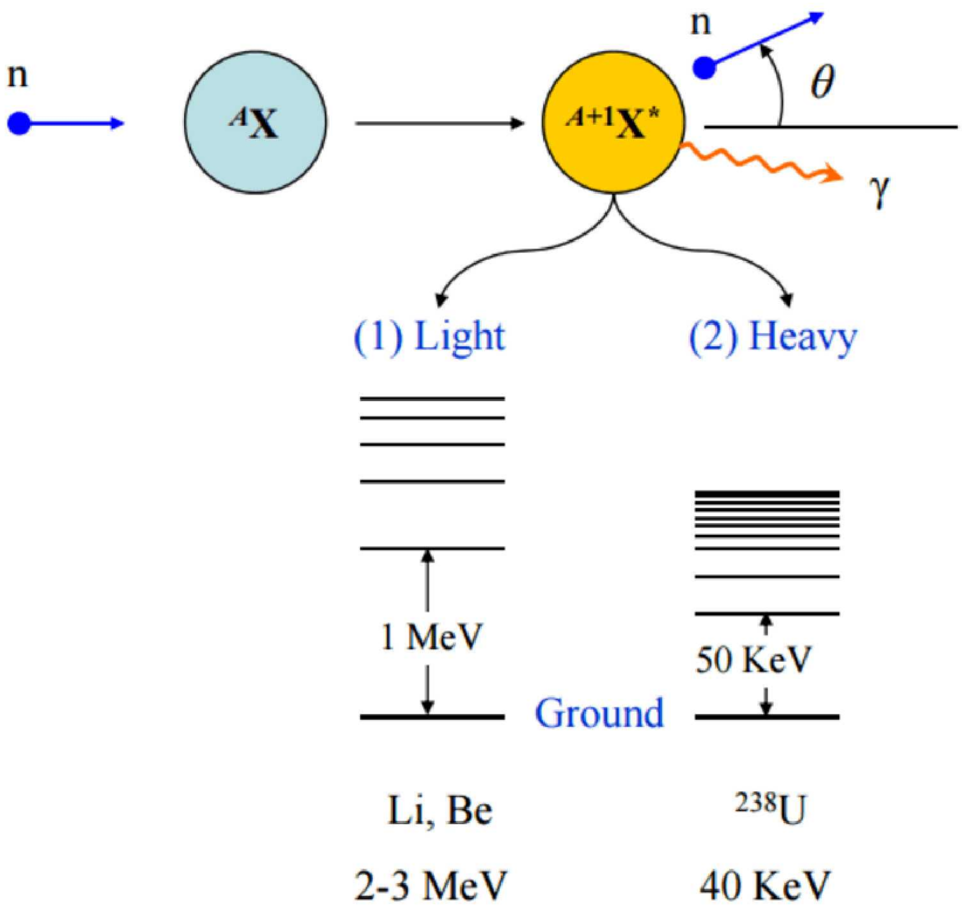
Pipe dream #2 (Active 3D Imaging and Multiplication):



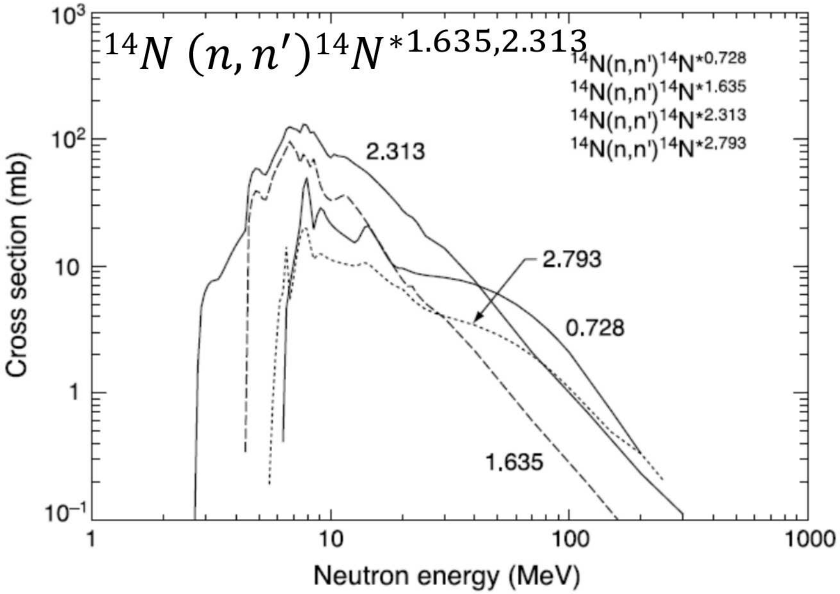
Now that we have a D-T generator, what about Inelastic Interactions?

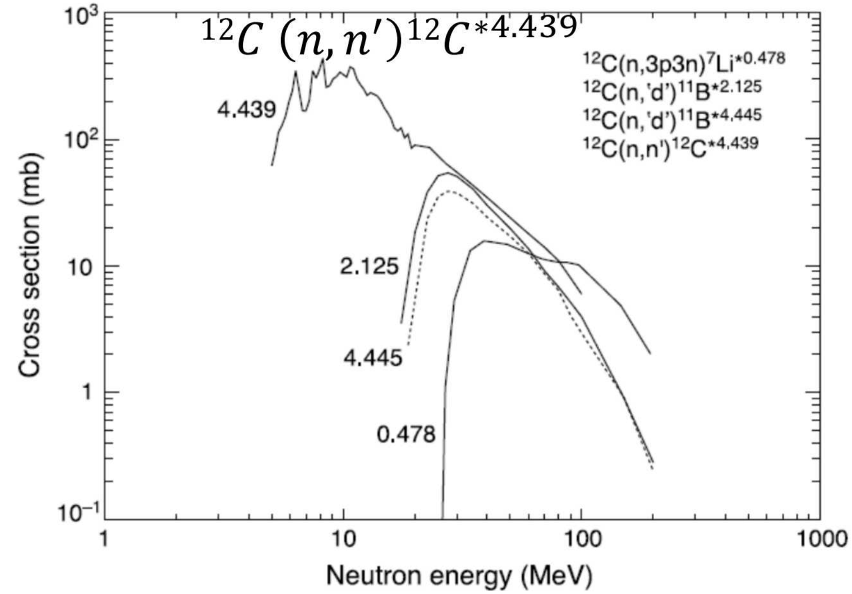
- An incident neutron is absorbed by a nucleus.
- Momentum is conserved, but kinetic energy is not.
- The compound nucleus emits a neutron of lower kinetic energy leaving the nucleus in an excited state.
- The nucleus de-excites to its ground state by emitting one or more gamma-rays.
- Inelastic scattering can only occur above a threshold energy (higher than the first excited state (ε_1)):

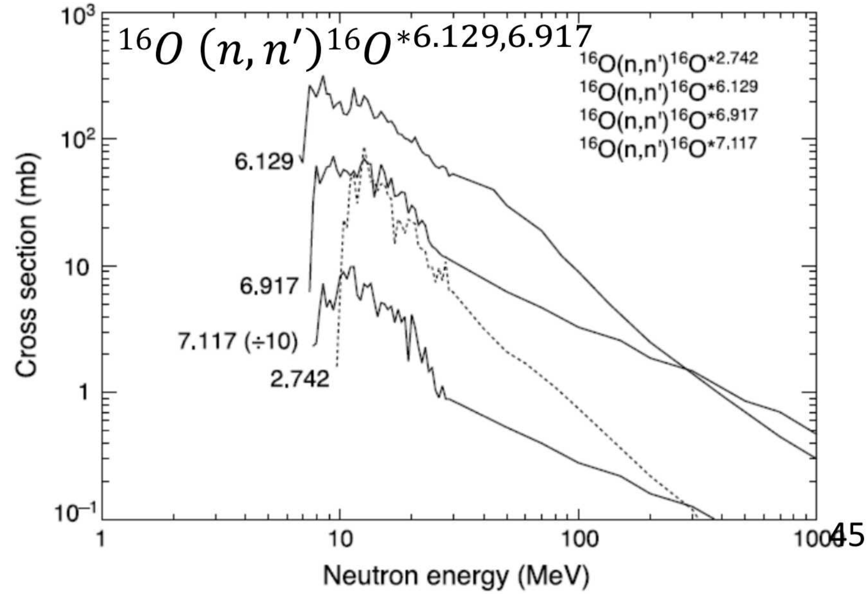
$$E_t = ((A+1)/A)^* \varepsilon_1$$



Some inelastic interactions of interest







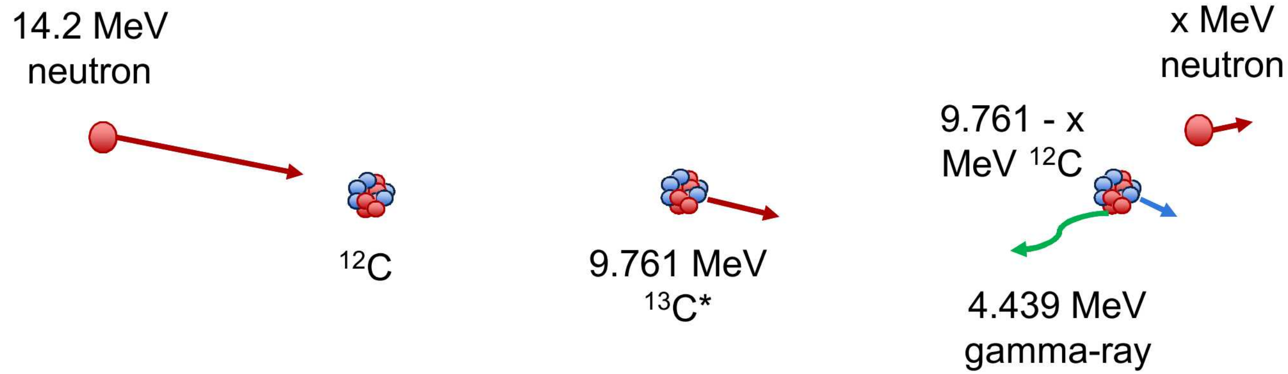
Kinetic energy of the emitted neutron?

For example, if an incident 14.2 MeV neutron inelastically interacts with ^{12}C

$$^{12}C\ (n, n')^{12}C^{*4.439}$$

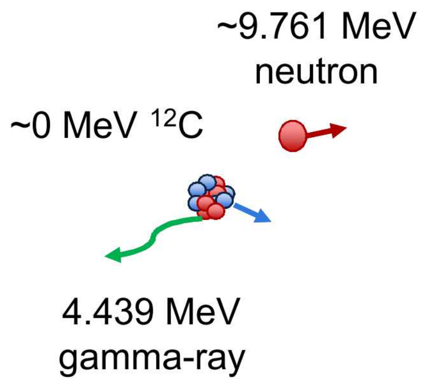
Then the compound nucleus has 9.761 MeV of kinetic energy.

The neutron and recoiling nucleus will therefore share this kinetic energy.



Kinetic energy of emitted neutron

- The emitted neutron carries most of the kinetic energy of the end state.



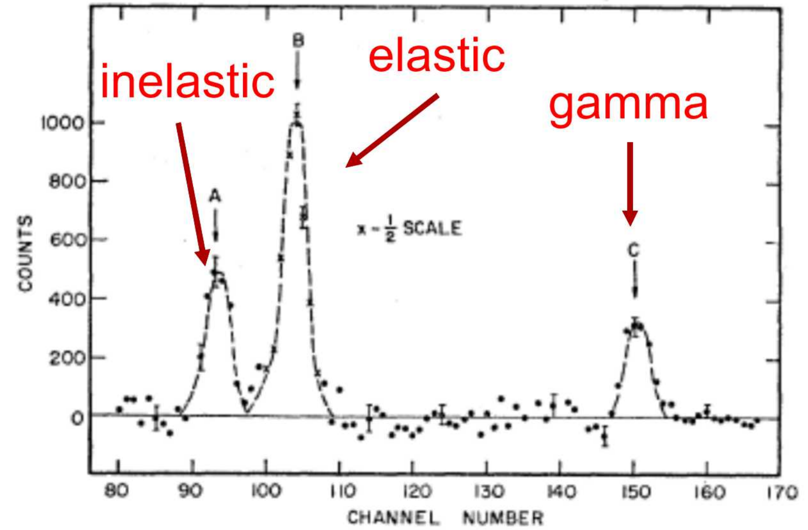


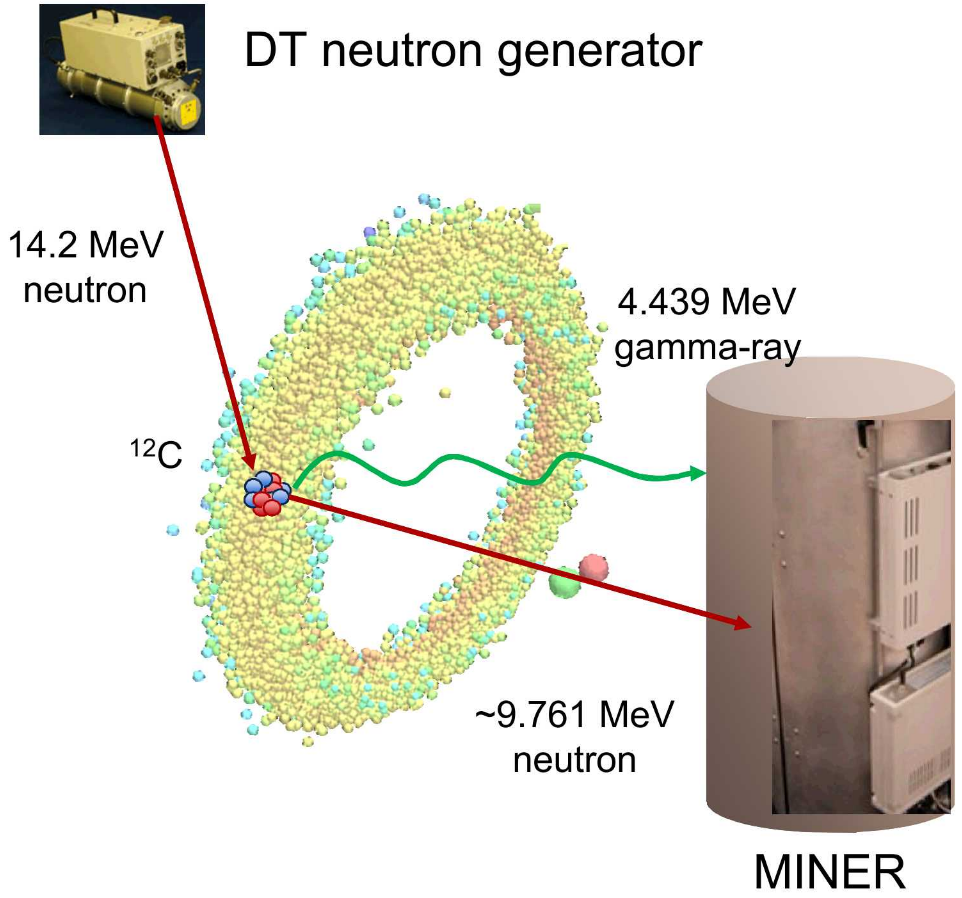
FIG. 1. Time-of-flight spectrum for carbon at a scattering angle of 40°. The time scale is 1 mμsec per channel, and increasing flight time is towards the left. *C* is due to the 4.43-Mev gamma ray from the inelastic scattering of neutrons from carbon, *B* to elastically scattered neutrons, and *A* to carbon first-level inelastically scattered neutrons.

Inelastic Scattering of 14-Mev Neutrons from Carbon and Beryllium*

J. D. ANDERSON, C. C. GARDNER, J. W. MCCLURE, M. P. NAKADA, AND C. WONG
University of California Radiation Laboratory, Livermore, California

(Received March 11, 1958)

Event level characterization and imaging



Material characterization:

$$E_{n0} = E_p + E_{n1} =$$
$$14.2\ MeV - E_\gamma = 9.8\ MeV$$

$$E_\gamma = 4.4\ MeV$$

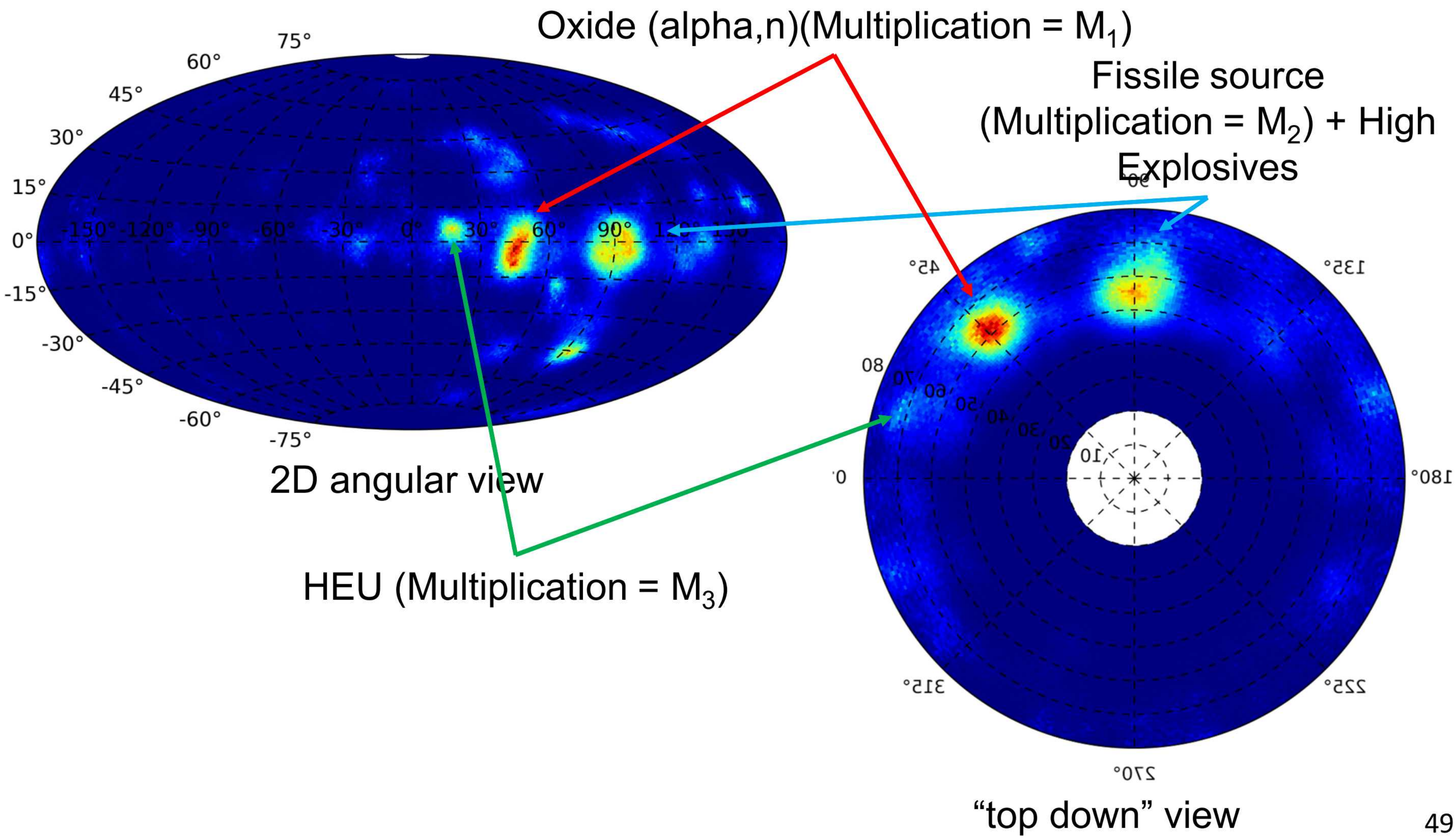
(Low-Z elements, e.g. High Explosive composition by neutron and gamma energy)

3D localization:

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

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

Pipe dream #3
(Active 3D imaging, M, and Low-Z ID):



Conclusions

- Special Nuclear Material can be characterized by fast (ns) timing of correlated gammas and neutrons.

- This can be used to determine:
    1. Geometry (both imaged and through mass-multiplication relation)
    2. Multiplication
    3. Fissile mass
    4. Presence and nature of surrounding reflectors/moderators

- We have demonstrated that the determination of multiplication and the 3-D imaging of SNM is possible using this signature.

Extra Slides

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