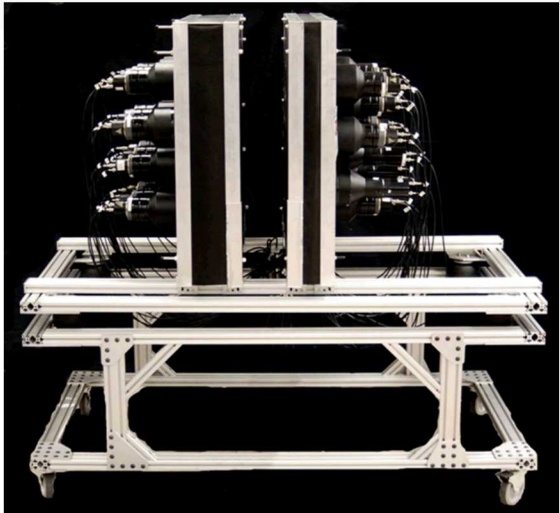
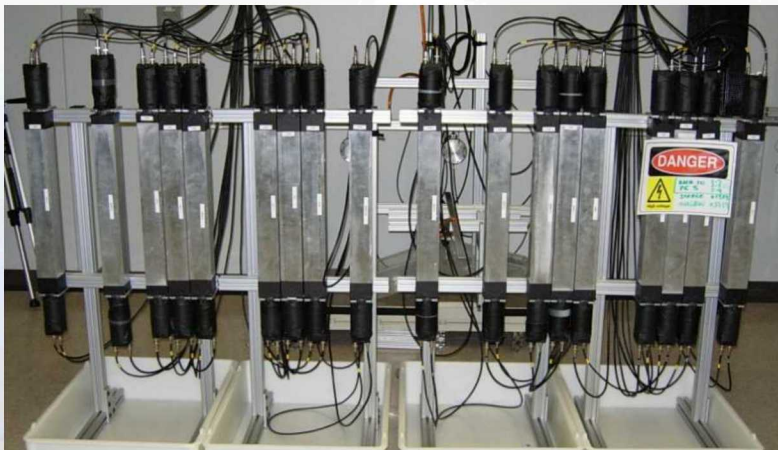
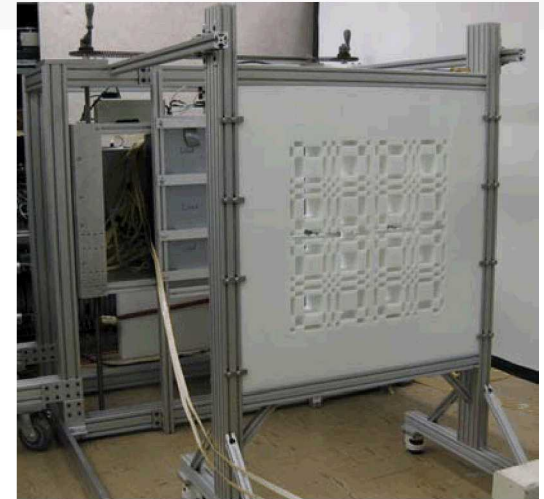


Fast Neutron Detection

SAND2018-12870PE



Peter Marleau
Sandia National Labs



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... and Carlsbad Programs
... and Nevada Projects
... and Washington
 Program Office
... and Weapons Evaluation
 Test Laboratory



Outline

■ Neutrons?

- Discovery
- Fission
- Chain reactions

■ Special Nuclear Material

- What's so special?
- Characterization methods

■ Multiplicity analysis

- He-3 detectors

■ Scintillator detectors

- dE/dx – quenching and pulse shape discrimination
- PSD methods

■ Correlated timing (TOFFEE)

Neutrons?

Sir James Chadwick - 1932

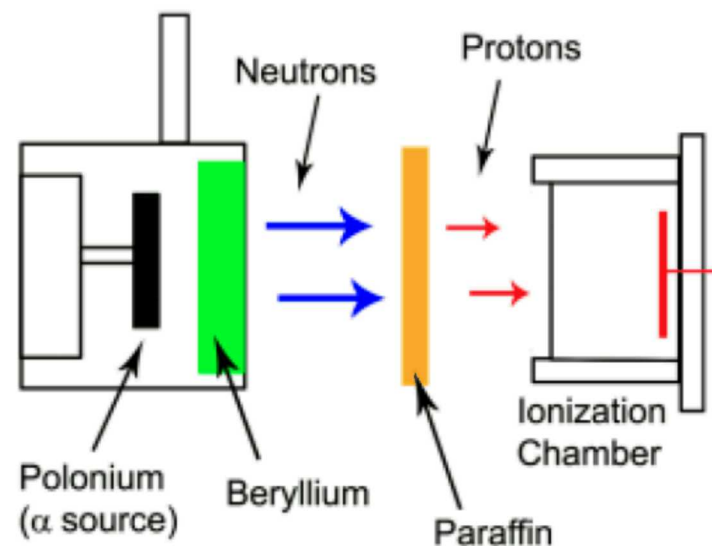
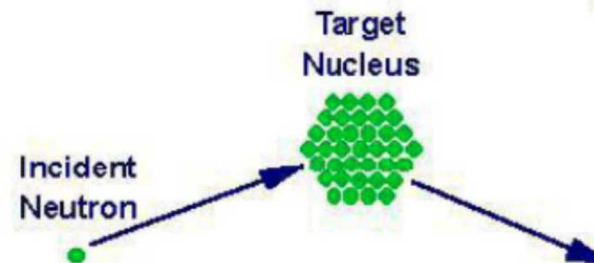


Possible Existence of a Neutron

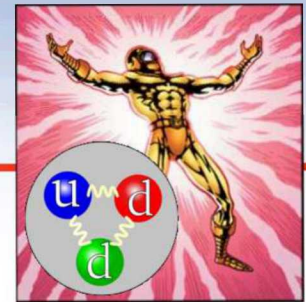
It has been shown by Bothe and others that beryllium when bombarded by α -particles of polonium emits a radiation of great penetrating power, which has an absorption coefficient in lead of about 0.3 (cm.)^{-1} . Recently Mme. Curie-Joliot and M. Joliot found, when measuring the ionisation produced by this beryllium radiation in a vessel with a thin window, that the ionisation increased when matter containing hydrogen was placed in front of the window. The

Ref.: J. Chadwick, Nature 132 (1932) 3252

- Atoms were thought to be composed only of protons and electrons.
- Chadwick explained the Curie-Joliot experiment as caused by neutrons rather than photons.
- First measurement of the neutron mass.

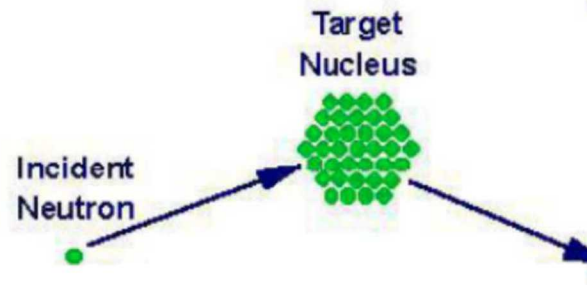
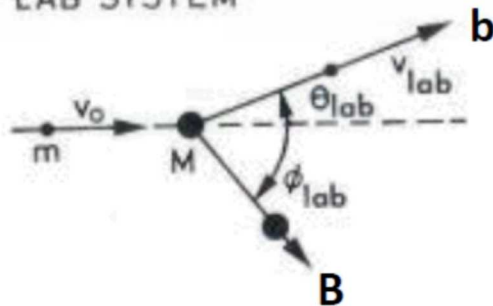


Neutron Interactions - Elastic

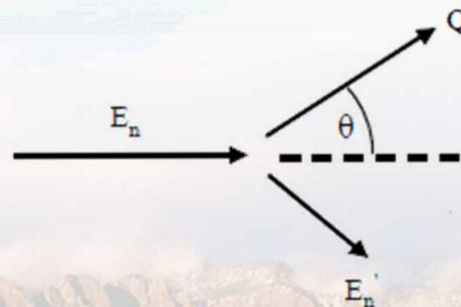


$$E_B = E_0 \frac{4A}{(A+a)^2} \cdot \cos^2 \phi_B^{\text{lab}}$$

LAB SYSTEM



$$Q_{\text{max}} = \frac{4mME_n}{(M+m)^2}$$



Nucleus

Q_{max}/E_n

^1_1H	1.000
^2_1H	0.889
^4_2He	0.640
^9_4Be	0.360
$^{12}_6\text{C}$	0.284
$^{16}_8\text{O}$	0.221
$^{56}_{26}\text{Fe}$	0.069
$^{118}_{50}\text{Sn}$	0.033
$^{238}_{92}\text{U}$	0.017

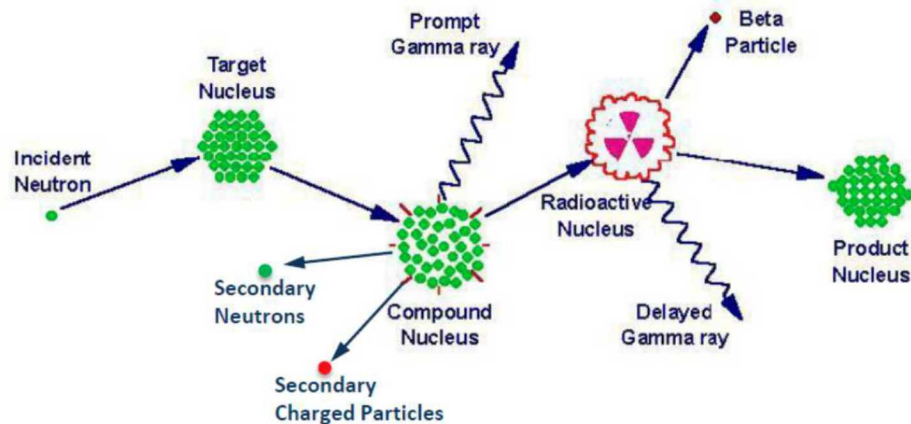


Neutron Induced Radioactivity



Enrico Fermi and students

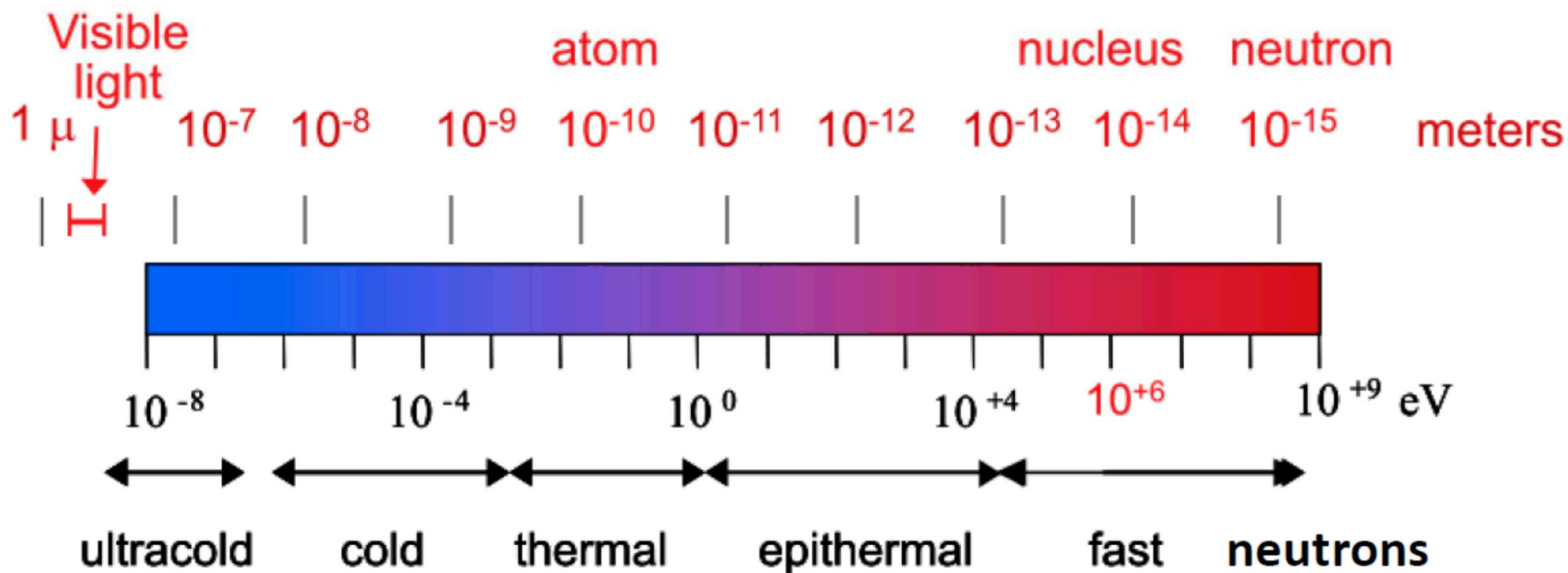
Inelastic interactions



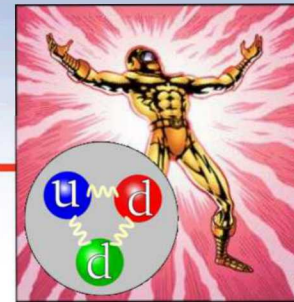
- By 1934 Enrico Fermi used neutrons to induce radioactivity in 22 different elements.
- He discovered that slowing (or moderating or thermalizing) the neutrons increased the rate of radioactivity.
- In 1938, he received a Nobel Prize in Physics for this work.

Neutron properties

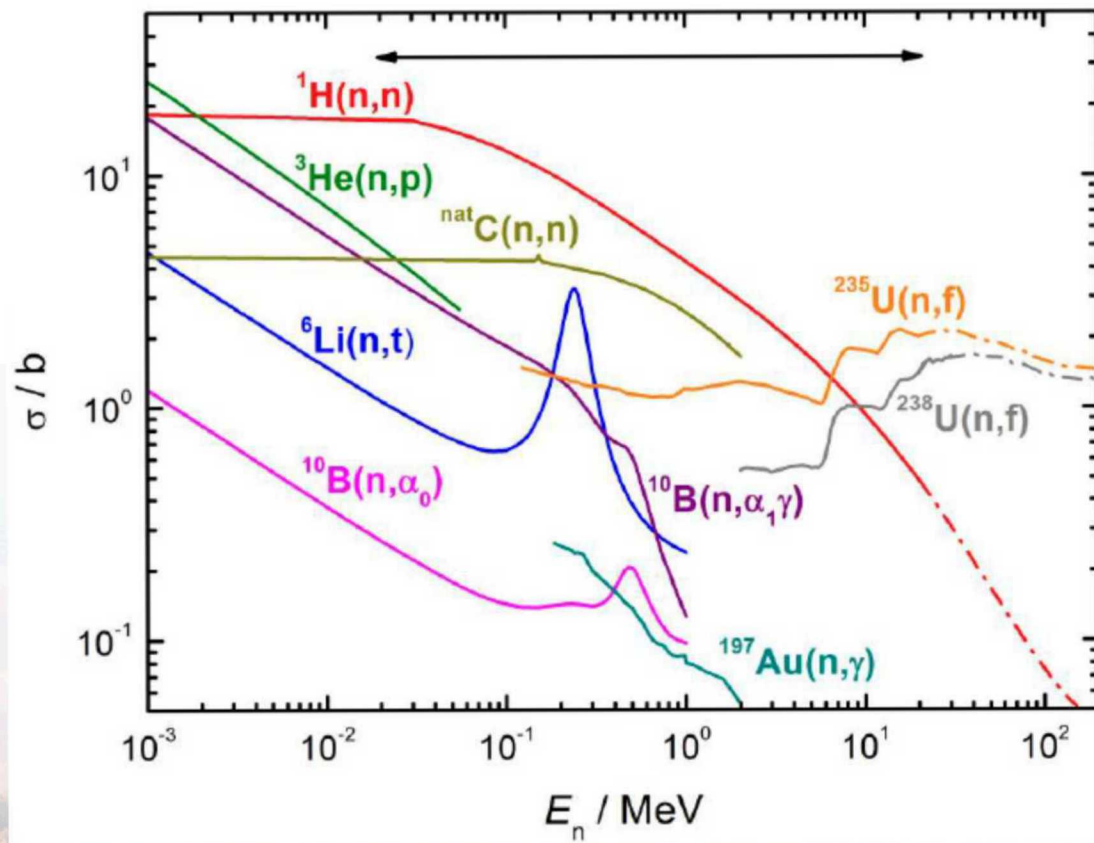
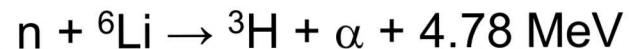
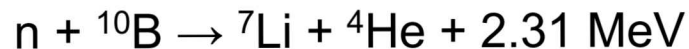
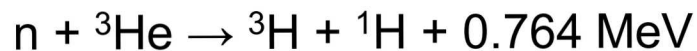
de Broglie wavelength: $\lambda = h / p$



Slow Neutron Interactions/Detection



- High thermal cross section
- High Q-value



Taken from ENDF database



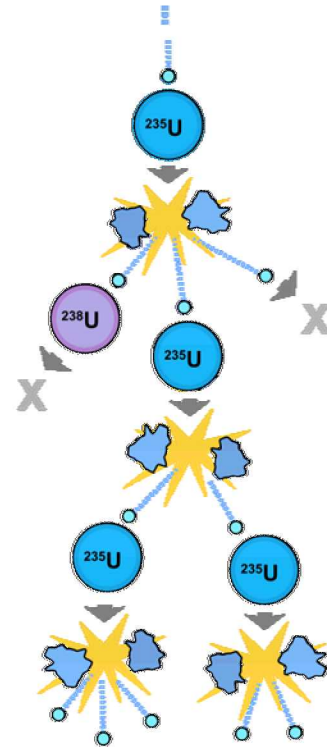
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Neutrons and fission

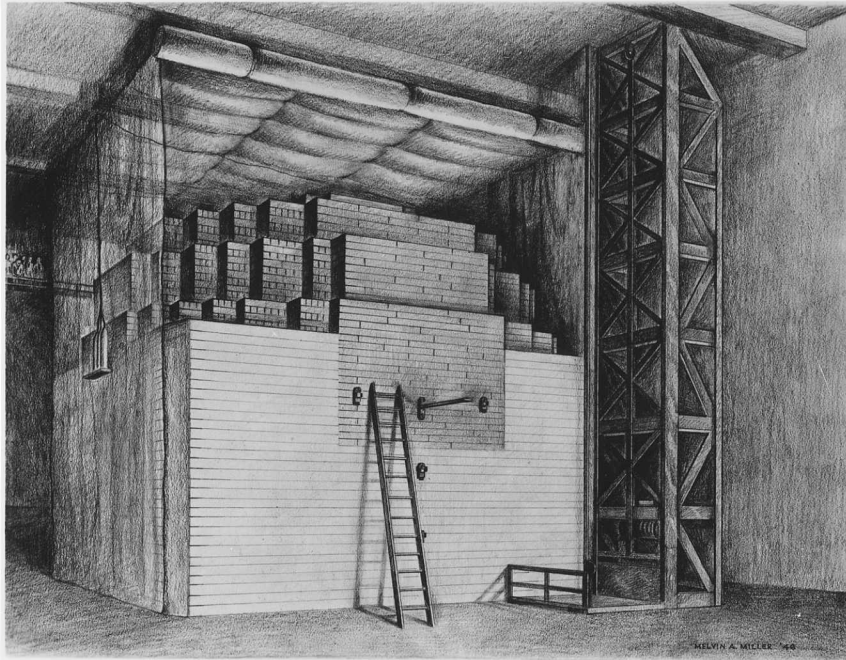


Lisa Meitner and Otto Hahn

- 1938 – Lisa Meitner, Otto Hahn, and Fritz Strassman bombarded uranium with neutrons in Berlin.
- 1939 - Nuclear fission (term coined by Otto Frisch), the neutron induced splitting of nuclei into lighter elements releasing 100s of MeV of energy, was discovered.
- Hahn and Strassman predicted the liberation of additional neutrons during fission. Joliot discovered the phenomenon of chain reactions.
- Hahn received the 1944 Nobel Prize in Chemistry for the discovery of the fission of heavy atomic nuclei.



Reactor, bombs, and the Manhattan Project



1939 – Leo Szilard proposed the possibility of sustainable and exponentially increasing neutron driven chain reactions.

1939 – Chadwick proposed an atomic bomb utilizing tons of natural uranium.

1939 – Oppenheimer proposed a cube of uranium detueride might “blow itself to hell”

1942 – the “Chicago Pile” reactor demonstrated sustained critical nuclear chain-reactions.

1942 – the Manhattan Project began.



What's so special?

As defined by the IAEA:

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

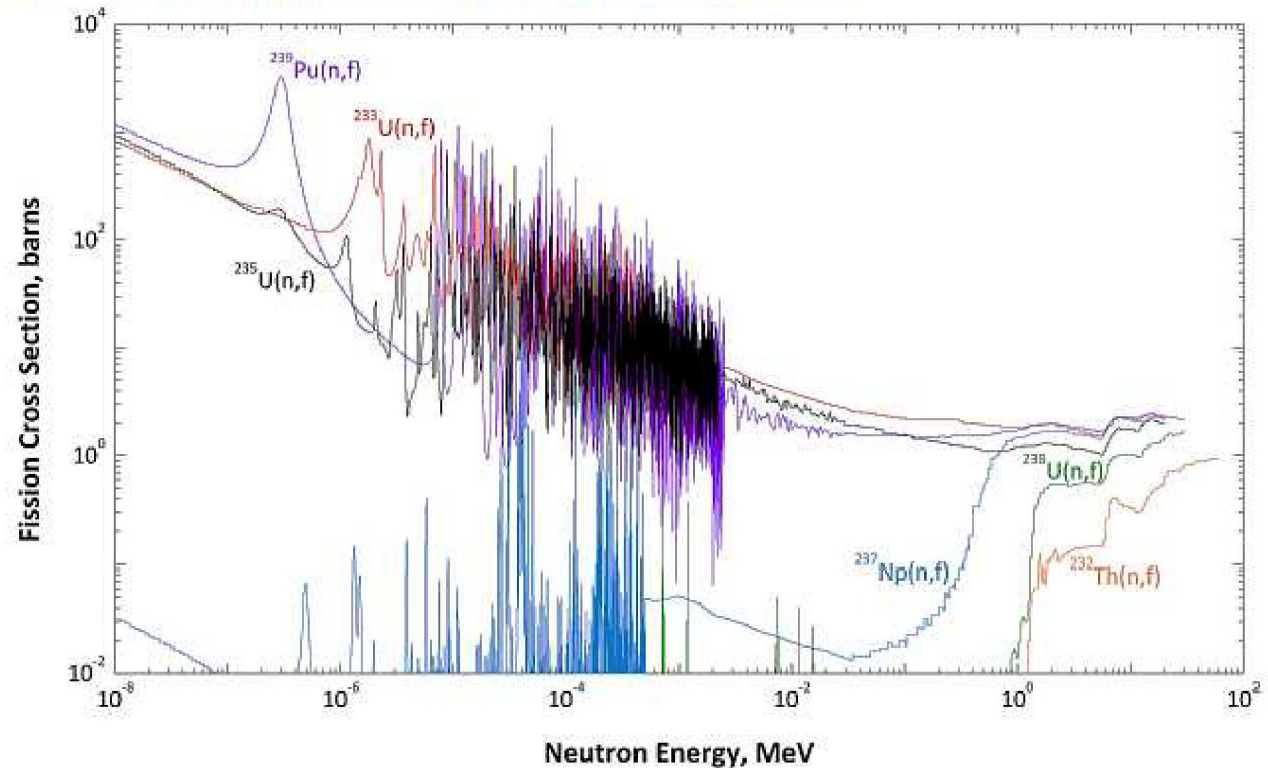
Special Nuclear Material – U-233, U-235, Pu-239

Source Material – everything that is not special

What's so special?

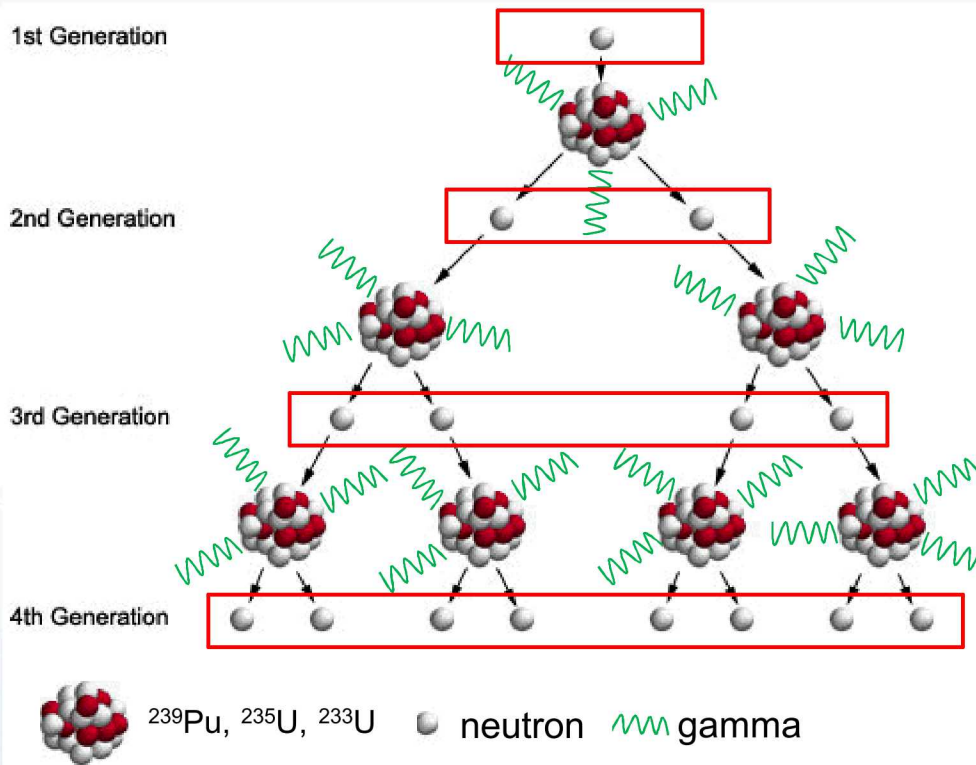
Isotope	Induced Thermal Fission Multiplicity ν
^{232}U	3.13
^{233}U	2.4
^{234}U	2.4
^{235}U	2.41
^{236}U	2.2
^{238}U	2.3
^{237}Np	2.70
^{238}Pu	2.9
^{239}Pu	2.88
^{240}Pu	2.8
^{241}Pu	2.8
^{242}Pu	2.81
^{244}Cm	3.46
^{252}Cf	4.06

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:

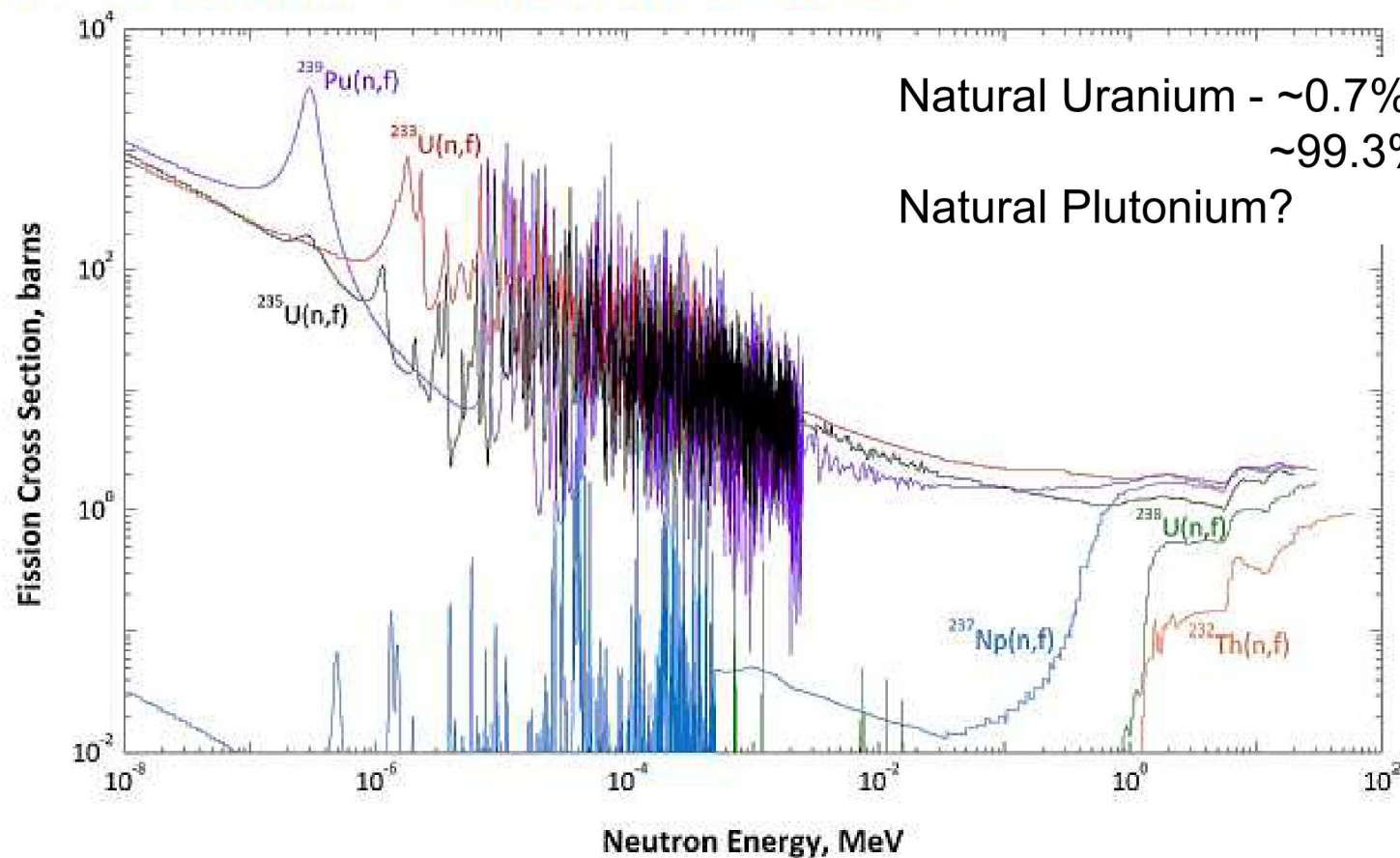
$$k = \frac{\text{neutrons in one generation}}{\text{neutrons in preceeding generation}}$$

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

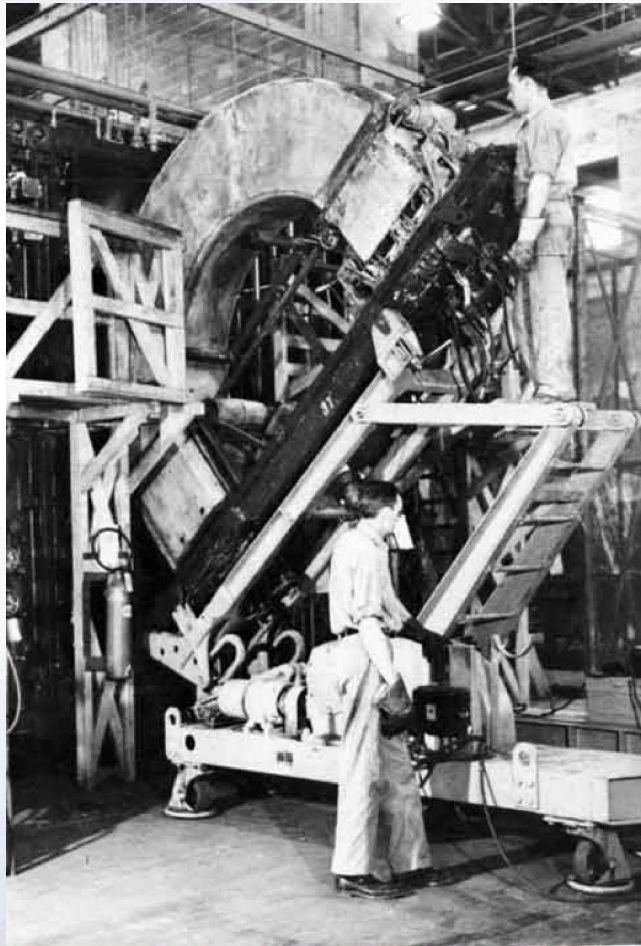
$$M = \frac{1}{1 - k}$$

Where can I get some?

The Neutron Fission Cross Sections



Uranium enrichment and Oak Ridge



- Ernest Lawrence invented the Calutron, based on the Cyclotron, at UC Berkeley.
- 1943 - The Y-12 plant in Oak Ridge, TN was stood up to enrich natural uranium.
- 1945 – Little Boy was ready.



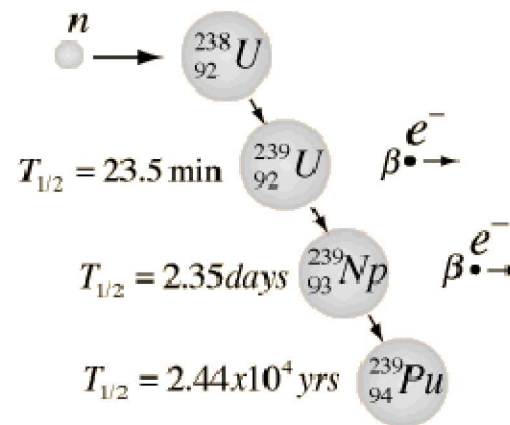
Recovery of U-235 from a Calutron

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	$50 \pm$		
^{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}		

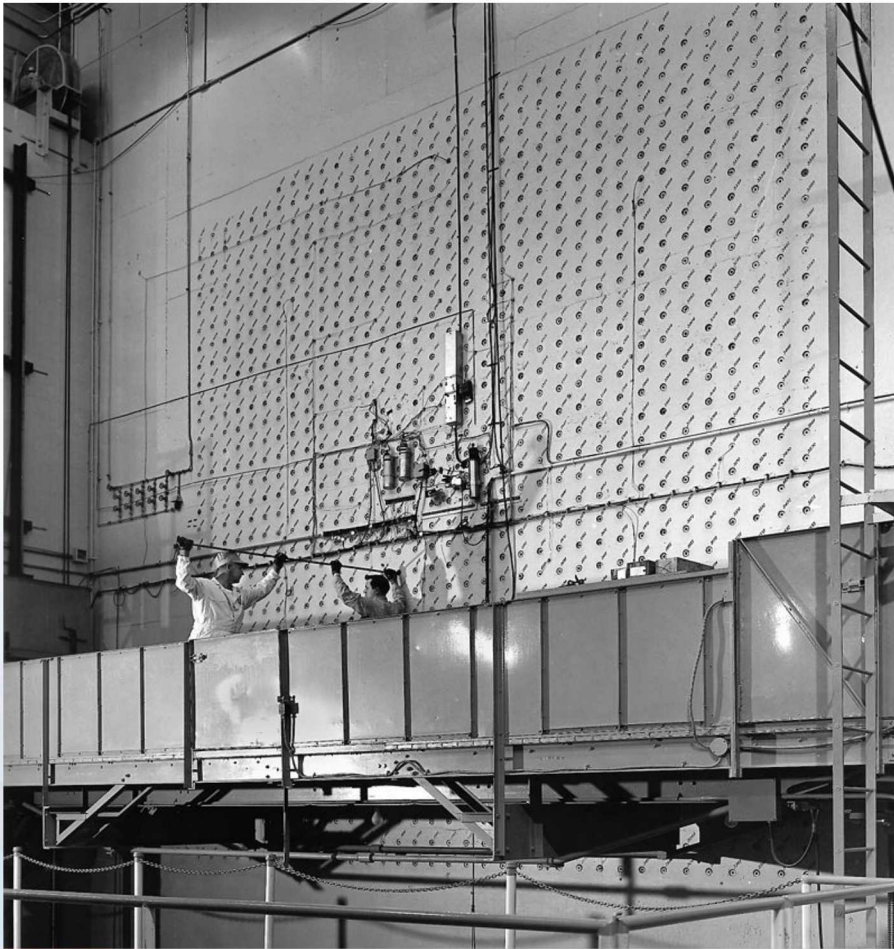
There isn't natural Plutonium to be found, but ...



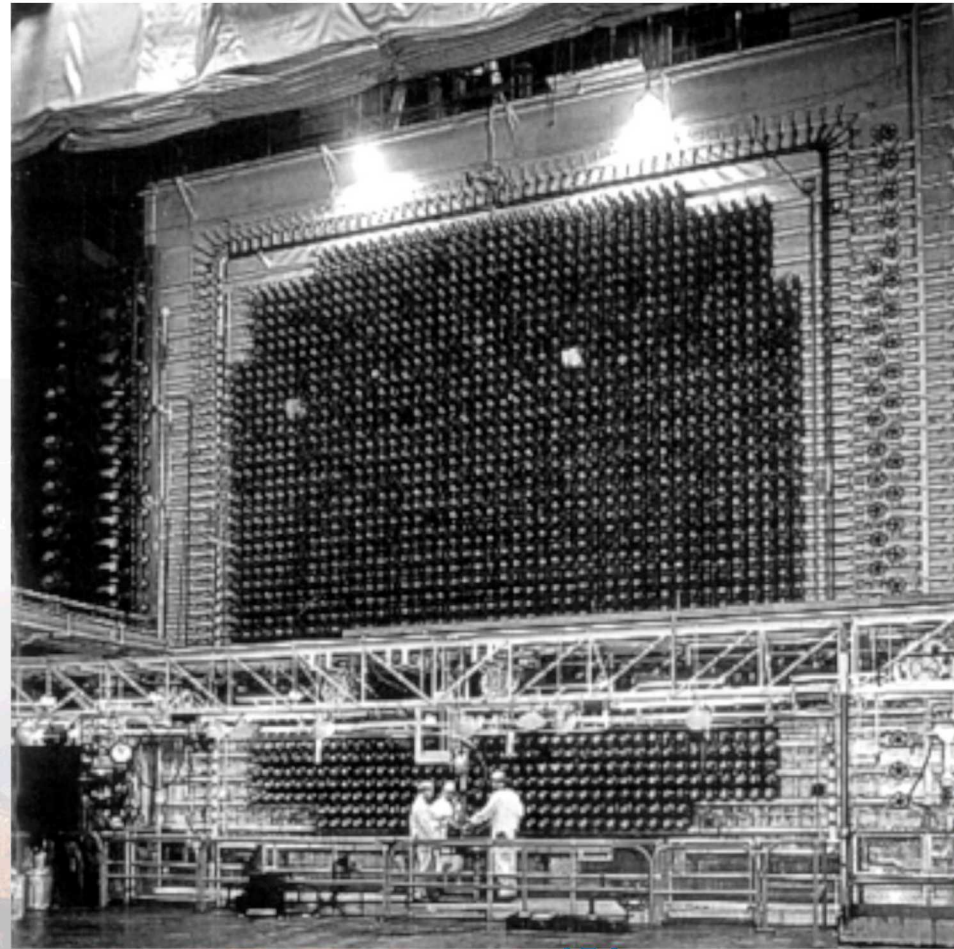
Ref: "Panda Book", values with $\pm$ have significant uncertainty

First Nuclear Reactors

1943 - ORNL X-10 Graphite Reactor
graphite moderated, air cooled

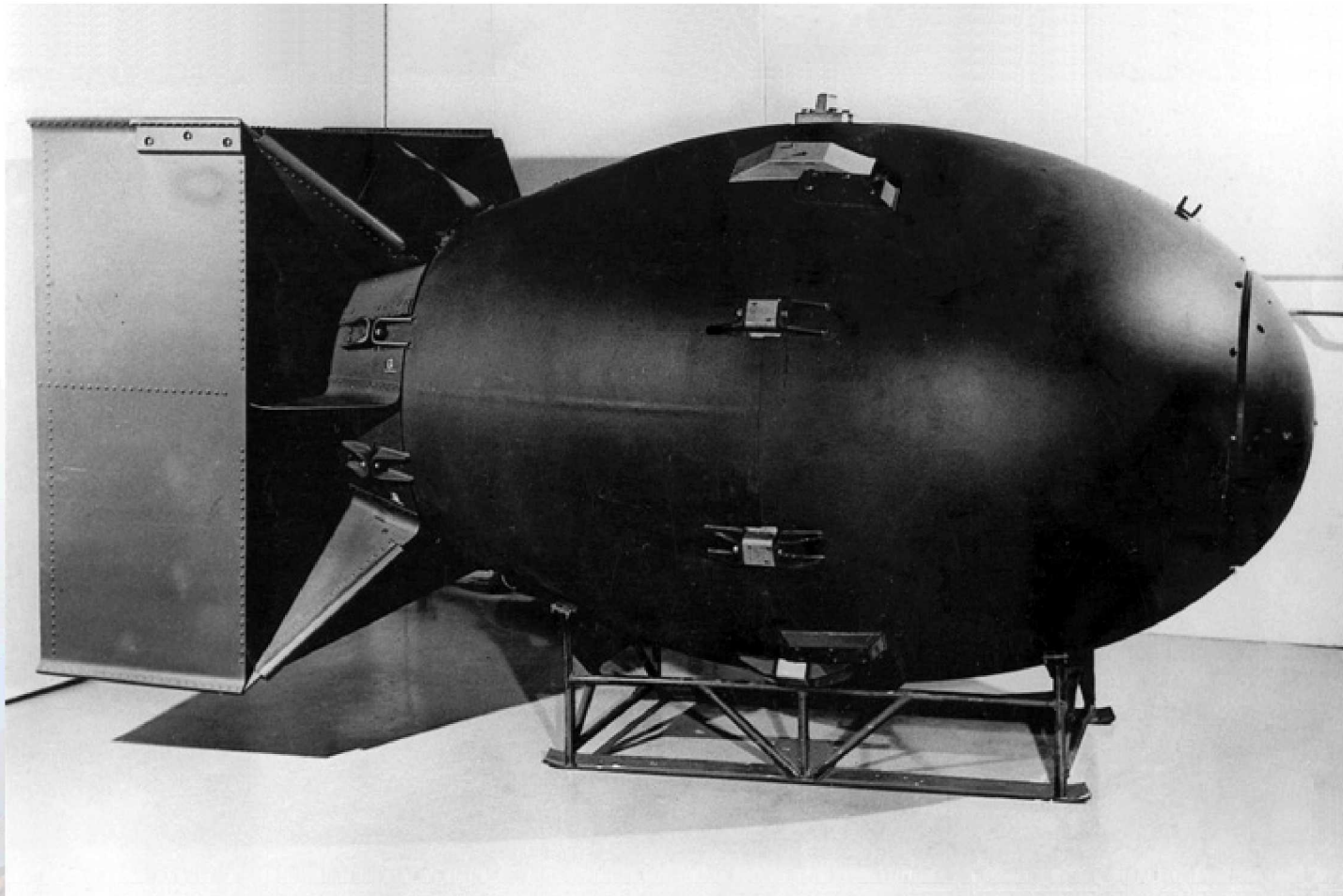


1944 - Hanford B Reactor
graphite moderated, water cooled



Fat Man - 1945

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



Measurable Unique Signatures of Fissile Material

1. Radiography

- Geometry

2. Gamma spectrum

- Isotopic content + surface area/shielding

3. Total neutron rate

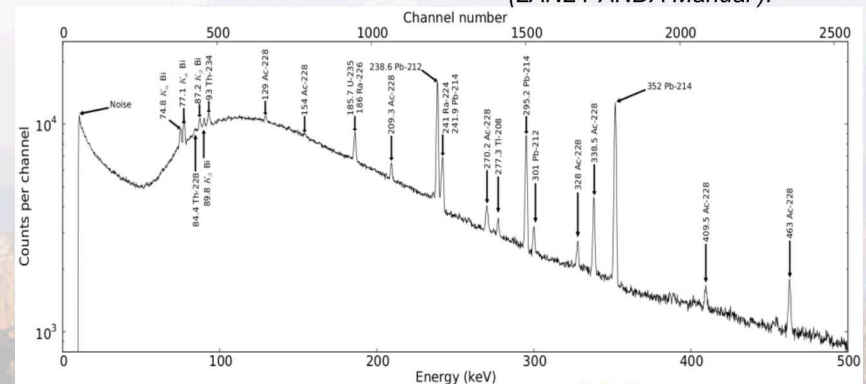
- Assay of the contents of specific materials

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).





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

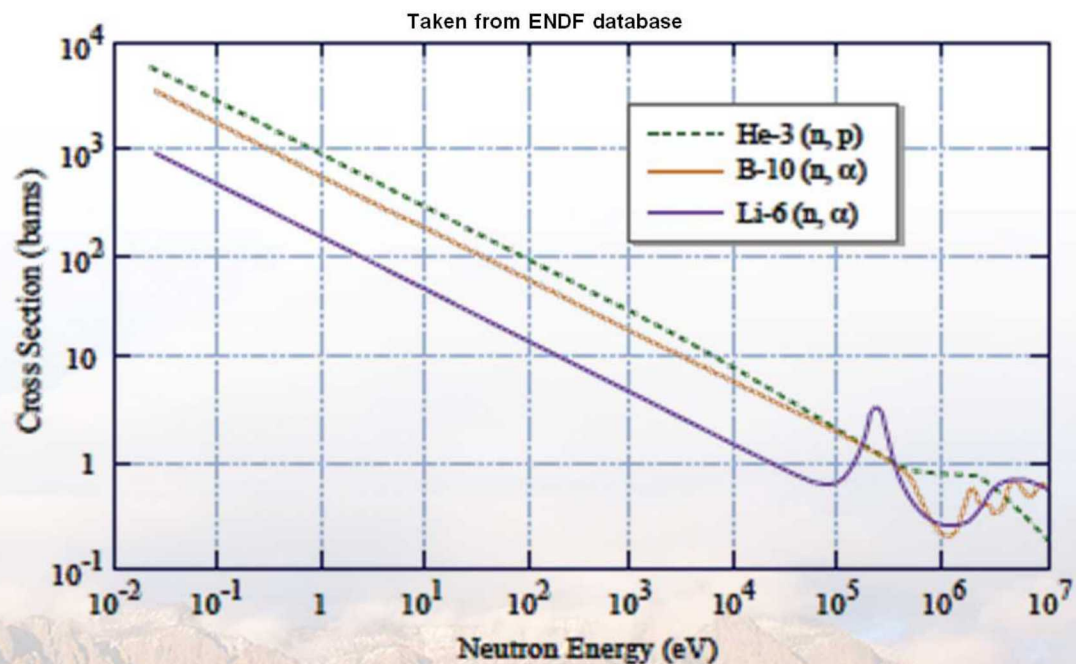
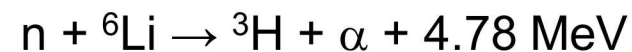
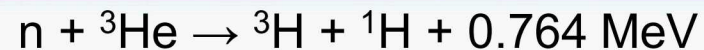
4. Correlated counts

- Requires high efficiency, necessitates large detection system
- Efficiency has to be well known
- Detector lifetime of 10-30 μ s is long compared to fission chain processes
- Neutron energy information is lost due to moderation

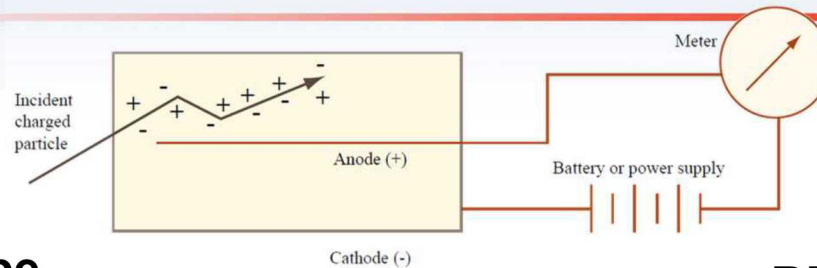
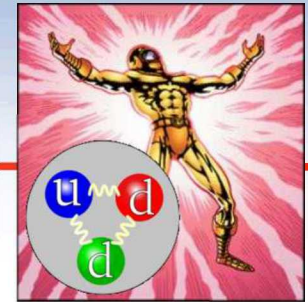
→ He-3 based technologies

Thermal Neutron Detection

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

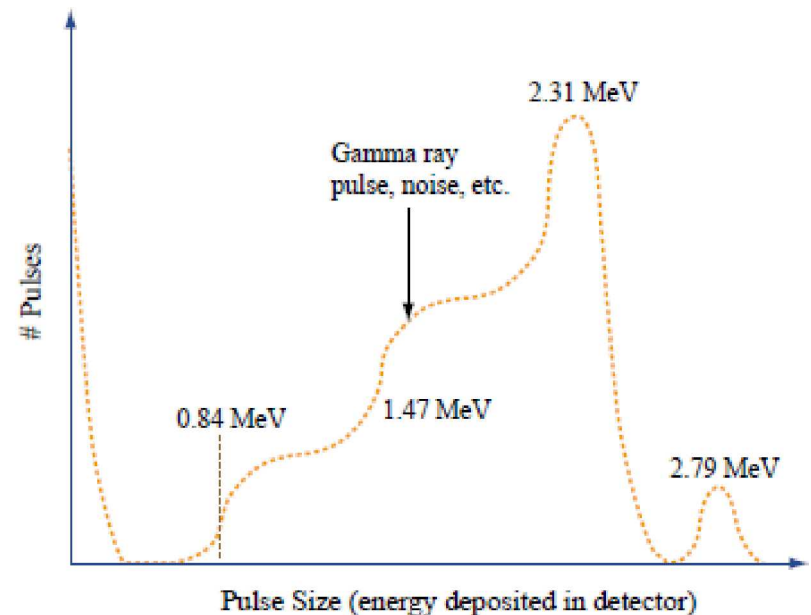
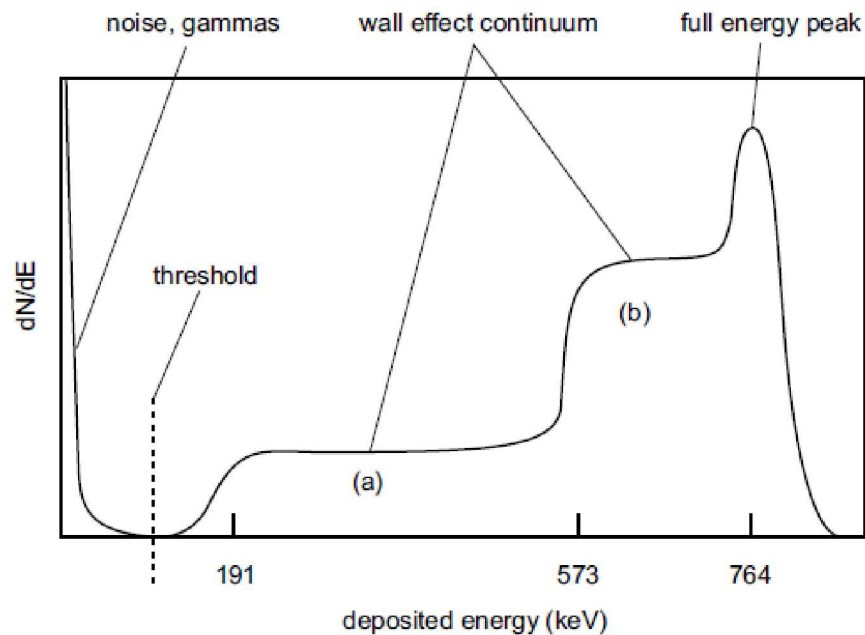
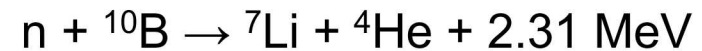
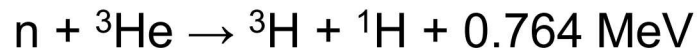


Gas Filled Neutron Counters

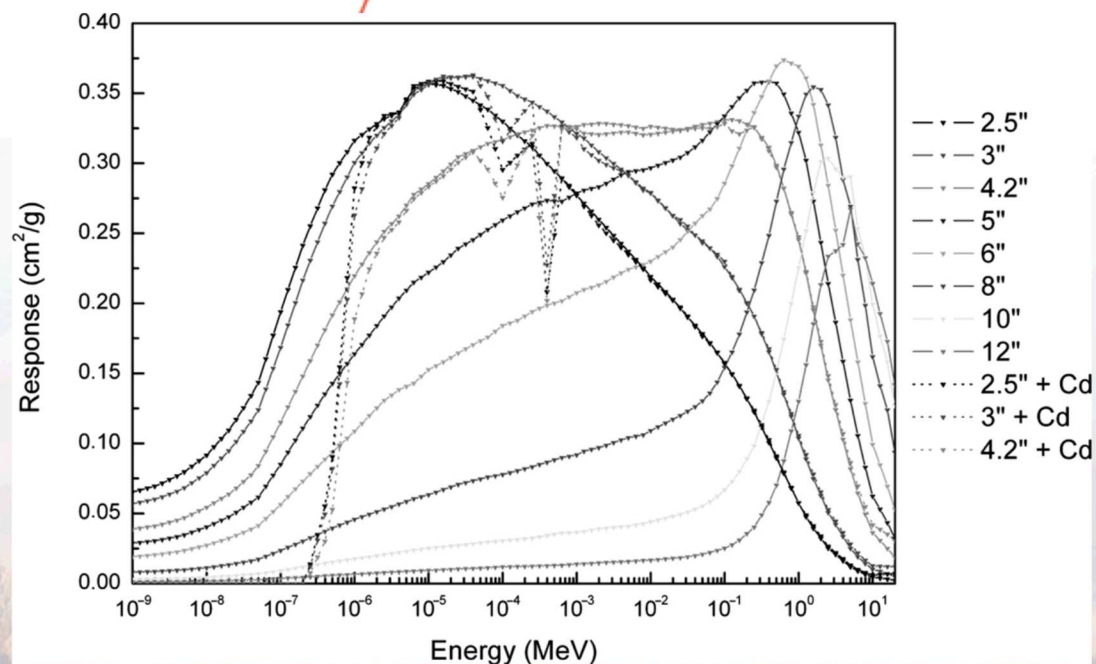
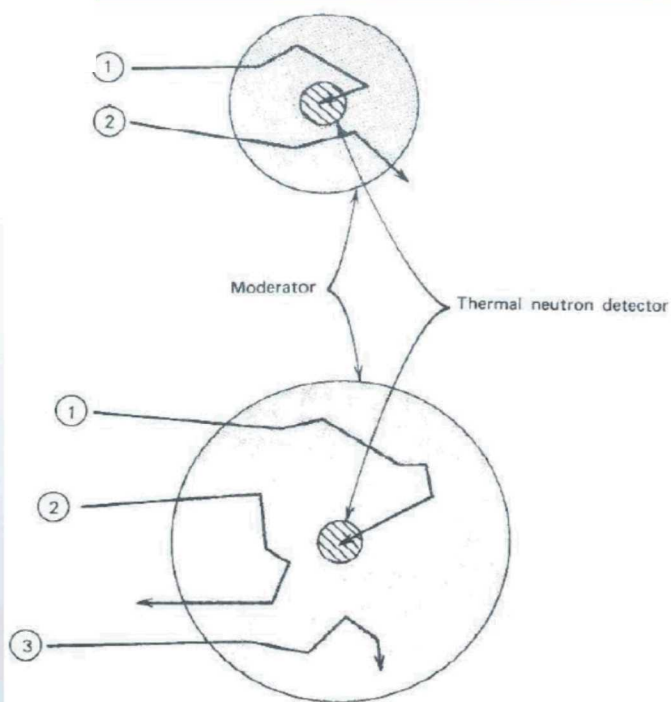
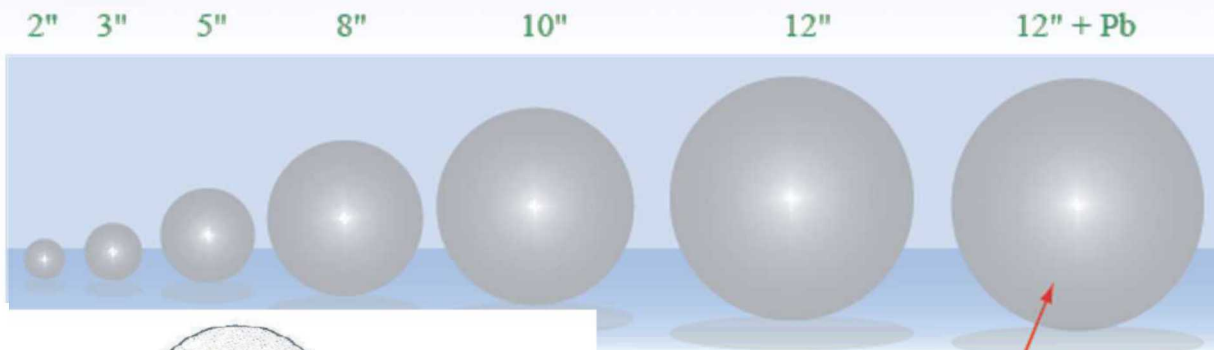
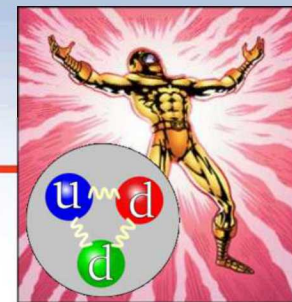


He3 Tube

BF3 Tube



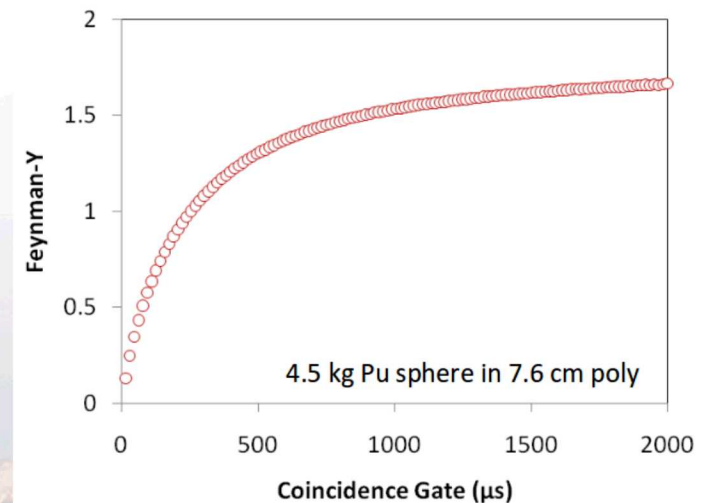
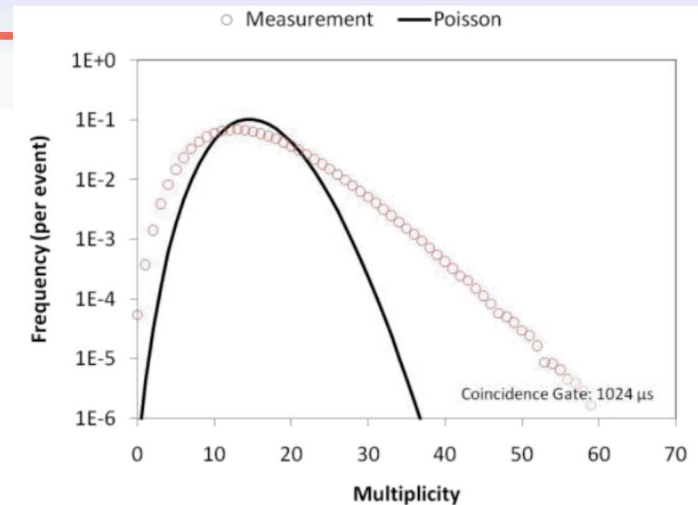
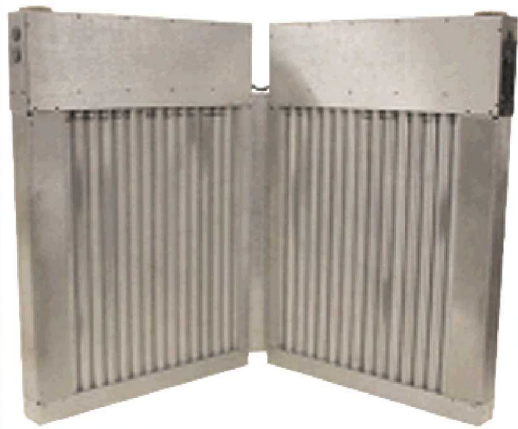
Fast Thermal Neutron Counters



Ref.: G. Knoll, Radiation detection and measurement, 3rd ed., p. 539

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>



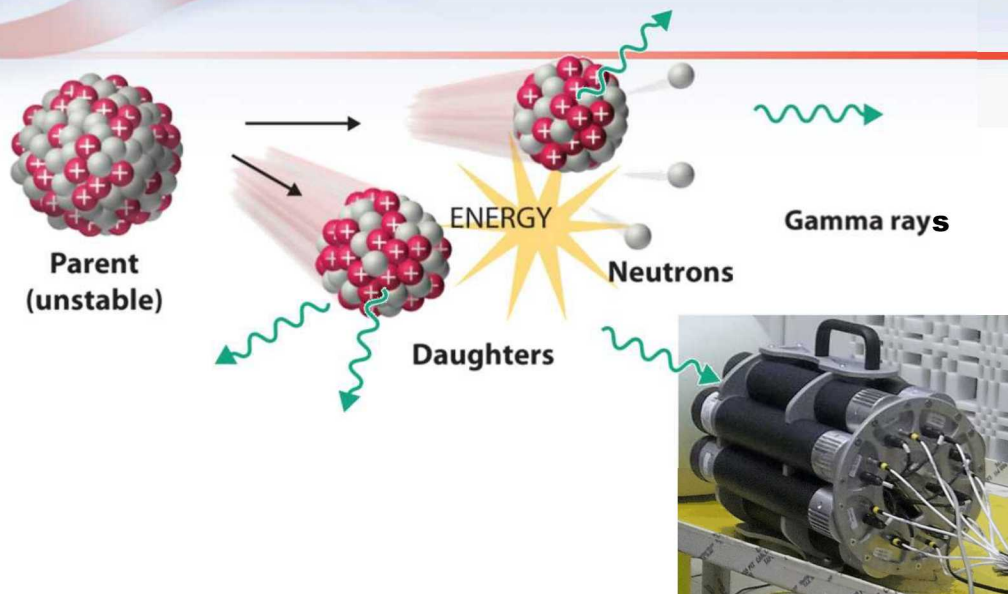
Sandia National Laboratories

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

- **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

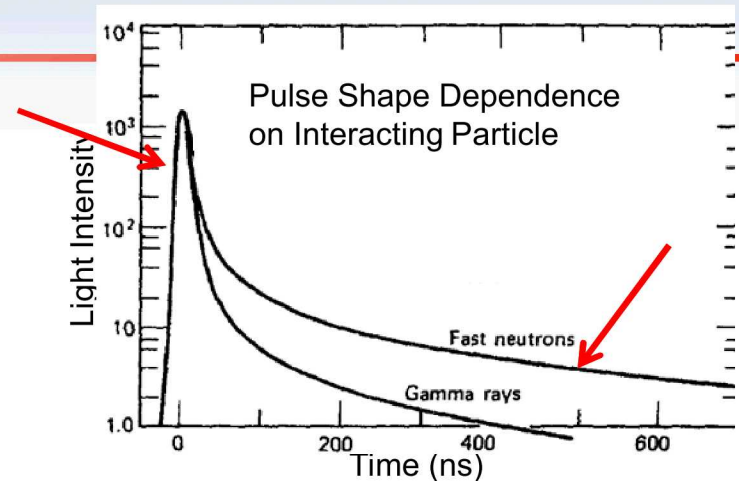
- Sub-nanosecond timing resolution

2. Incident neutron energy

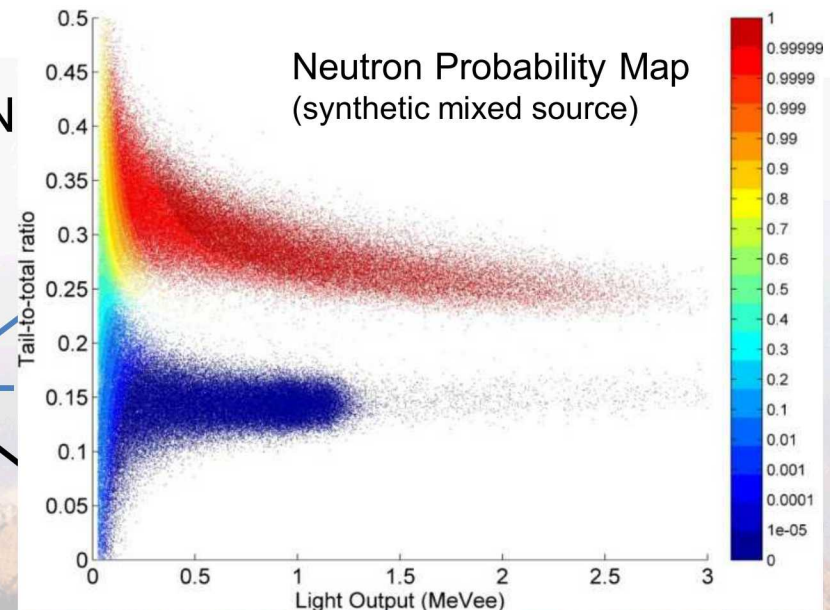
- Light output proportional to proton recoil

3. PSD Capable

- Discriminate neutrons and gamma rays
- Gamma rays provide clean timing signature of correlated event



Glenn Knoll Radiation Detection & Measurement 3rd Edition



Detection Efficiency

Table 13-1. Neutron and gamma-ray interaction probabilities in typical gas proportional counters and scintillators

Thermal Detectors	Interaction Probability	
	Thermal Neutron	1-MeV Gamma Ray
^3He (2.5 cm diam, 4 atm)	0.77	0.0001
Ar (2.5 cm diam, 2 atm)	0.0	0.0005
BF_3 (5.0 cm diam, 0.66 atm)	0.29	0.0006
Al tube wall (0.8 mm)	0.0	0.014
Fast Detectors	Interaction Probability	
	1-MeV Neutron	1-MeV Gamma Ray
^4He (5.0 cm diam, 18 atm)	0.01	0.001
Al tube wall (0.8 mm)	0.0	0.014
Scintillator (5.0 cm thick)	0.78	0.26

From T.W. Crane and M.P. Baker, Chapter 13, Neutron Detectors



Energy Deposition

Table 13-2. Neutron and gamma-ray energy deposition in typical gas proportional counters and scintillators

	Alpha or Proton Range (cm)	dE/dx for 400-keV Electron (keV/cm)	Average Neutron Reaction Energy Deposited (keV)	Electron Energy Deposited (keV) ^a	Ratio of Neutron to Electron Energy Deposition
Thermal Detectors					
³ He (2.5 cm diam, 4 atm)	2.1	1.1	~500	4.0	125
³ He (2.5 cm diam, 4 atm) + Ar (2 atm)	0.5	6.7	~750	24.0	30
BF ₃ (5.0 cm diam, 0.66 atm)	0.7	3.6	~2300	25.7	90
Fast Detectors					
⁴ He (5.0 cm diam, 18 atm)	0.1	6.7	1000	48	20
Scintillator (5.0 cm thick)	0.001	2000	1000	400	2.5

^aThis calculation assumes a path length of $\sqrt{2} \times$ tube diameter.

From T.W. Crane and M.P. Baker, Chapter 13, Neutron Detectors



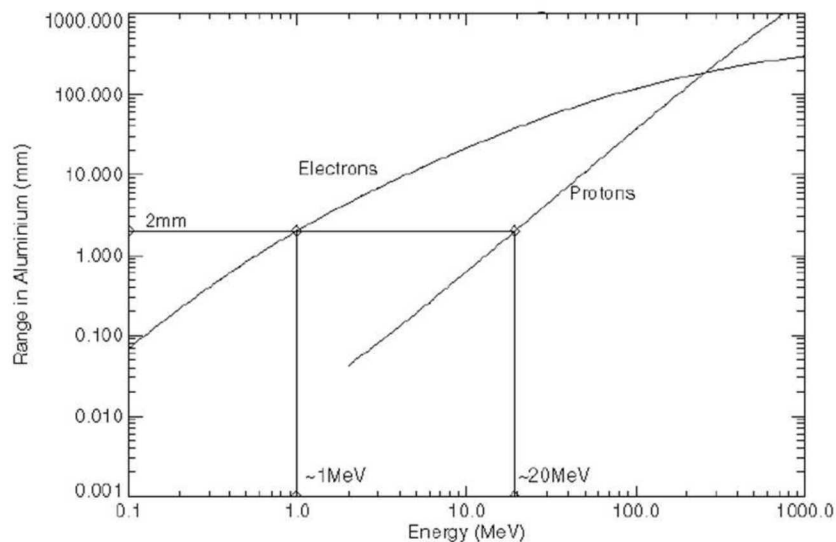
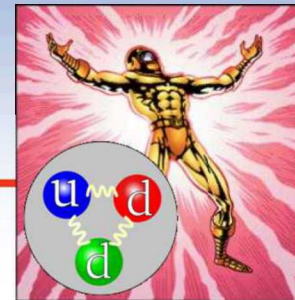
Discrimination – It's all about dE/dx



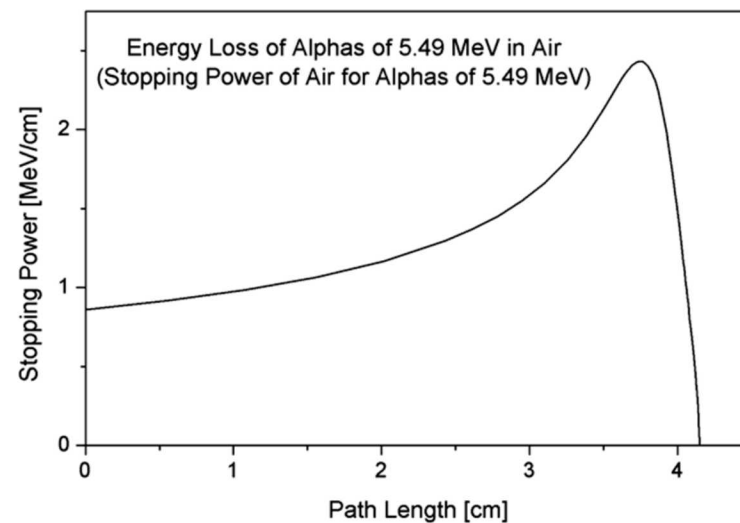
$$-dE/dx \sim M Z^2/E$$

$$M_e = 511 \text{ keV}$$

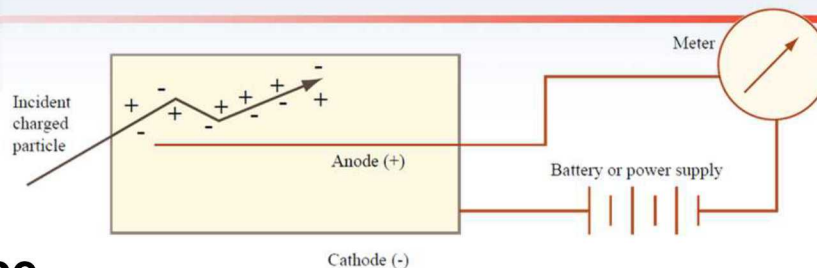
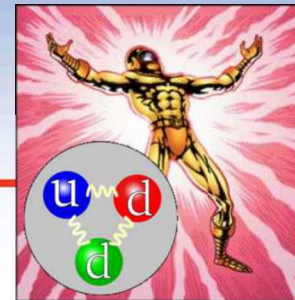
$$M_p = 938 \text{ MeV}$$



Mean ranges of protons and electrons in aluminum [Source: E.J. Daly, A.Hilgers, G. Drolshagen, and H.D.R. Evans, "Space Environment Analysis: Experience and Trends," ESA 1996 Symposium on Environment Modelling for Space-based Applications, Sept. 18-20, 1996, ESTEC, Noordwijk, The Netherlands.]

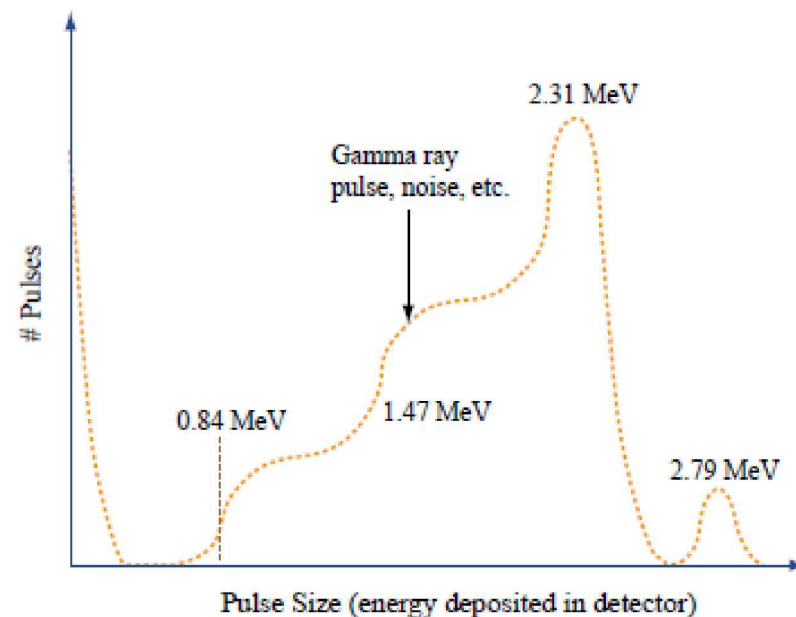
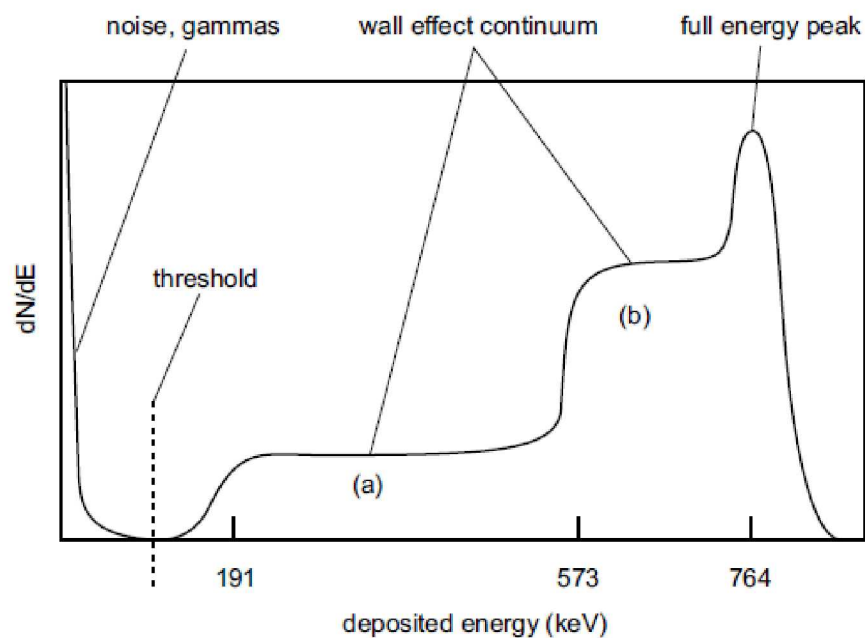
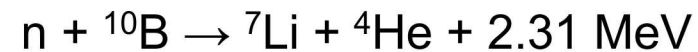
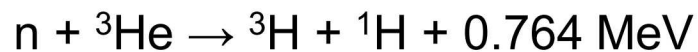


Discrimination – It's all about dE/dx



He3 Tube

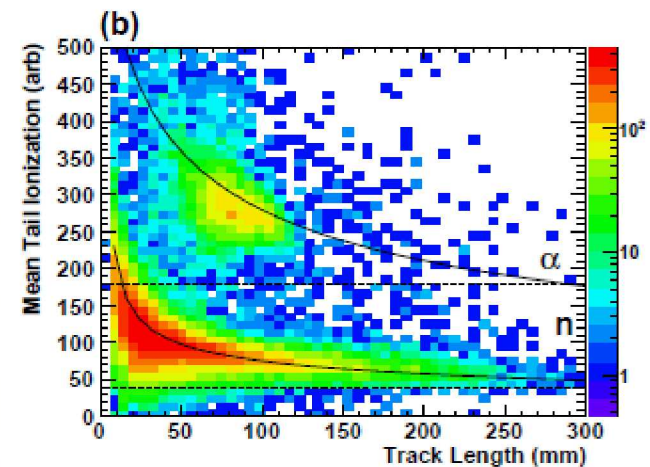
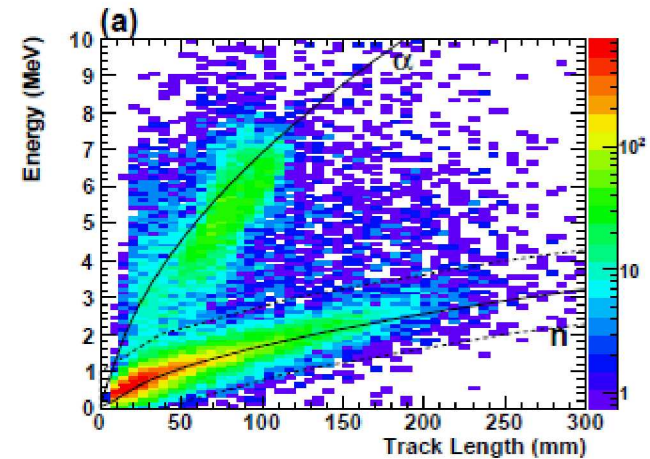
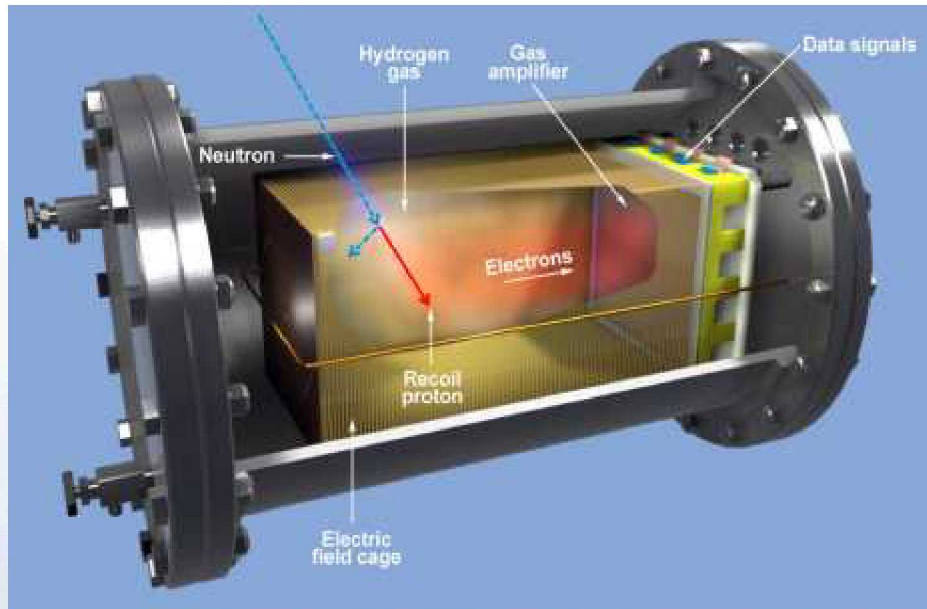
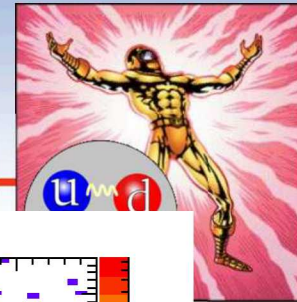
BF3 Tube



Discrimination – It's all about dE/dx



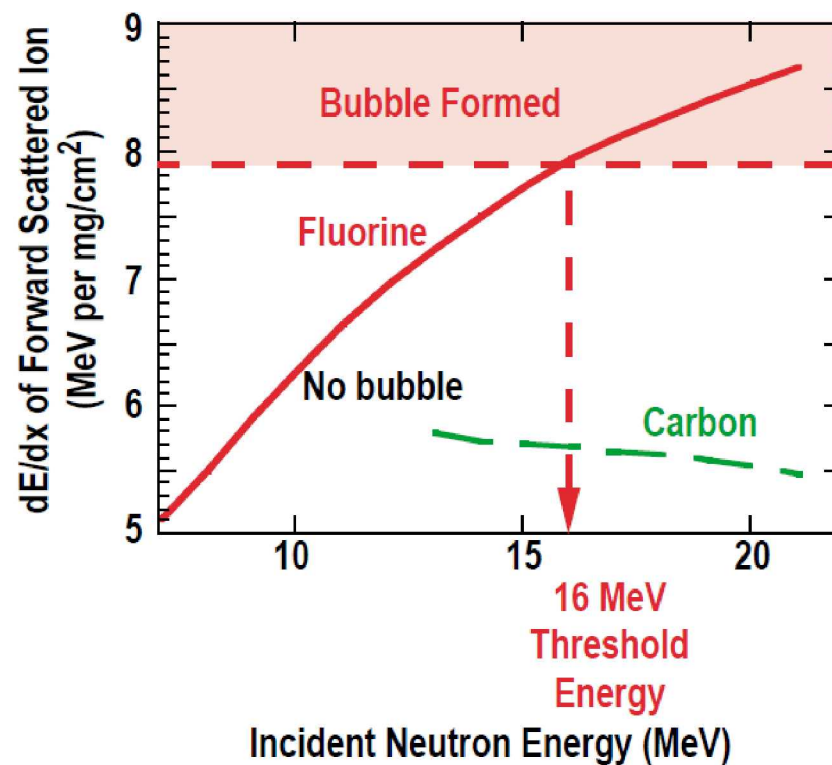
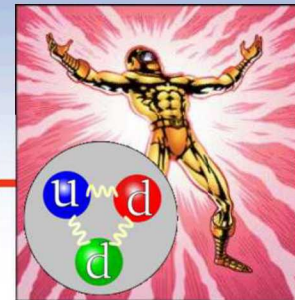
$$-dE/dx \sim M Z^2/E$$



Discrimination – It's all about dE/dx



$$-dE/dx \sim M Z^2/E$$



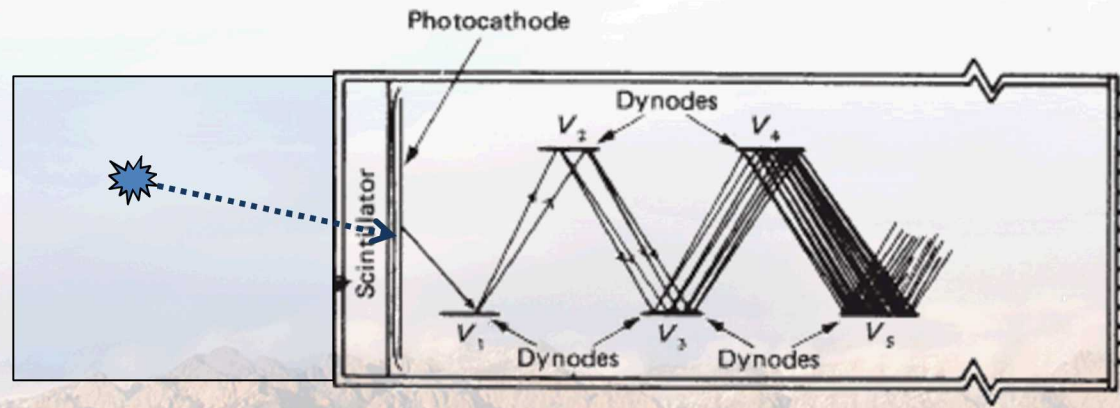


Neutron Detection – Scintillator

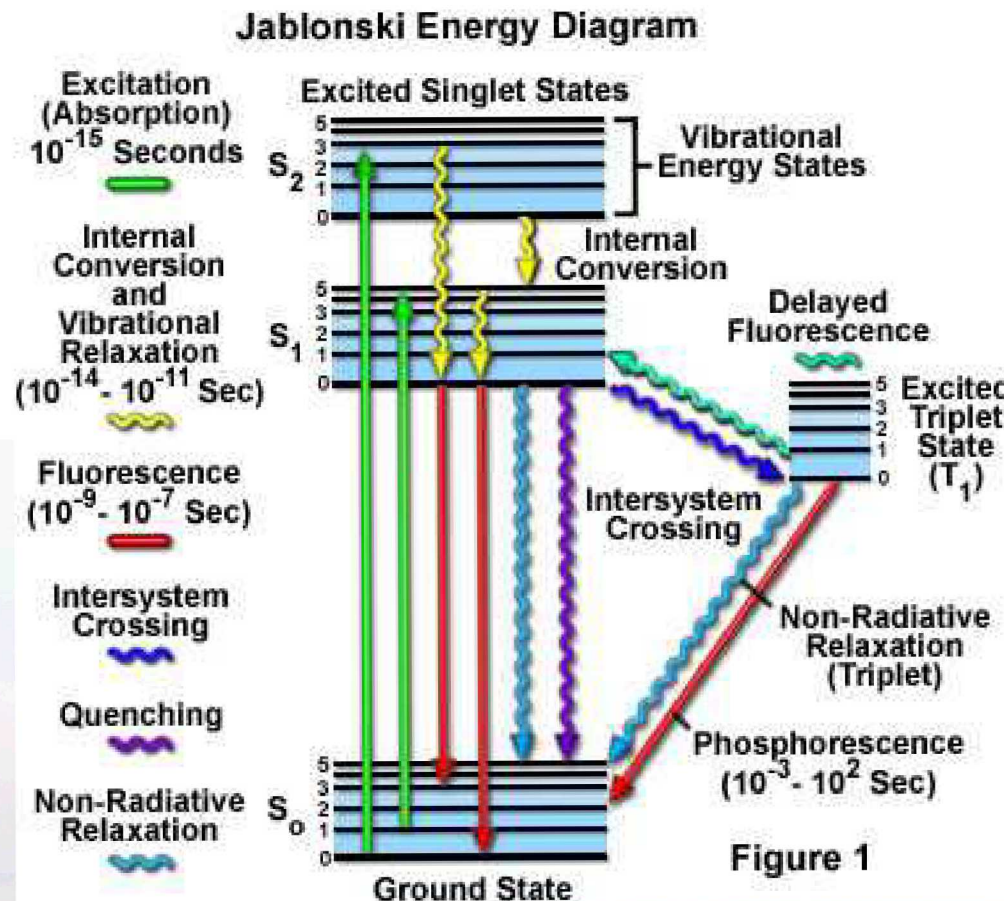
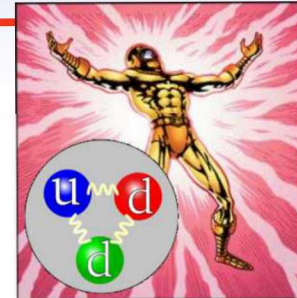
- Luminescence - When a material is excited and it subsequently gives off light.
- How it is excited determines the type of luminescence.
- Scintillation – luminescence produced by ionizing radiation excitation.
- Fluorescence – photoluminescence or scintillation that has a fast decay time (ns to μ s).
- Phosphorescence – same as fluorescence, but with much slower decay time (ms to seconds)

Neutron Detection – Scintillator

- Desirable Properties:
 - High conversion efficiency
 - Short decay times
 - Transparent to its own emission
 - Wavelength matched to photodetector
 - Inorganic – NaI(Tl), CsI(Tl), LaBr, BGO
 - Organic – Anthracene, Stilbene, Plastic, Liquid



Neutron Detection – Organic Scintillator



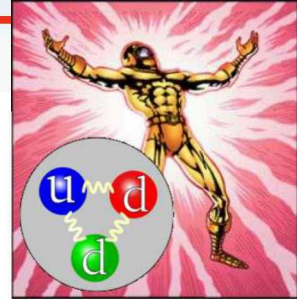
$$T_1 + T_1 = S_0 + S_1$$

<http://micro.magnet.fsu.edu/primer/techniques/fluorescence/fluorescenceintro.html>

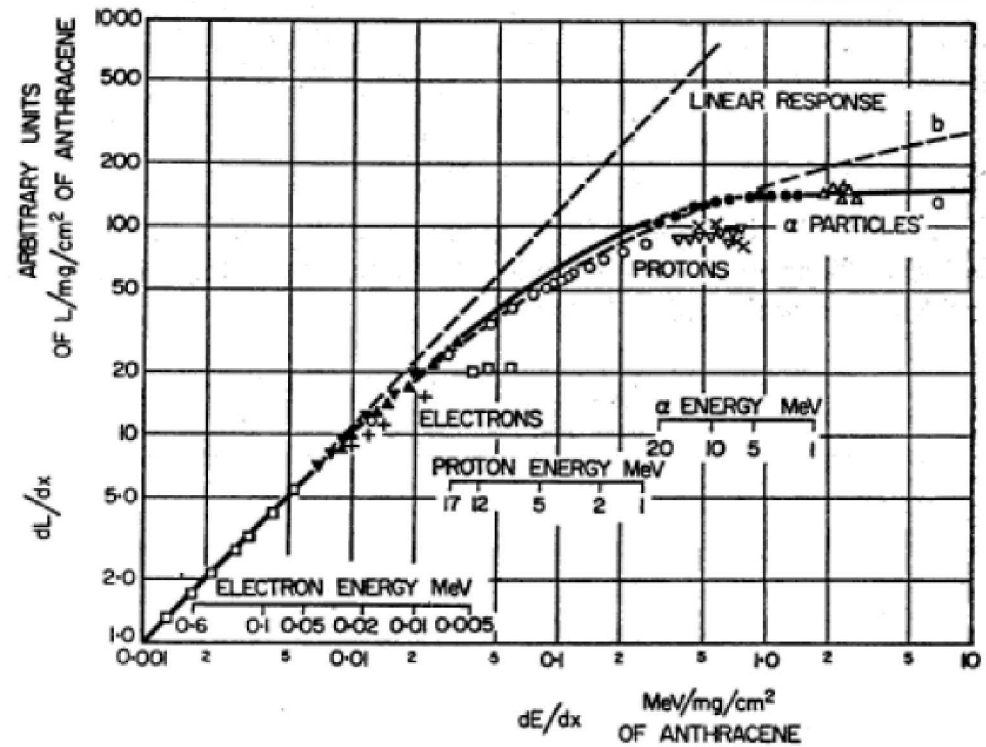


Sandia National Laboratories

Neutron Detection – Quenching, Birk's Law



- start with linearity at low ionization density $\frac{dL}{dx} = A \frac{dE}{dx}$
- let the density of excited molecules be proportional to the ionization density $B \frac{dE}{dx}$
- let k be the fraction that is quenched $\frac{dL}{dx} = \frac{A \frac{dE}{dx}}{1 + kB \frac{dE}{dx}}$
- for small dE/dx , approaches linearity
- for large dE/dx , approaches saturation $\frac{dL}{dx} = \frac{A}{kB}$



from Birks

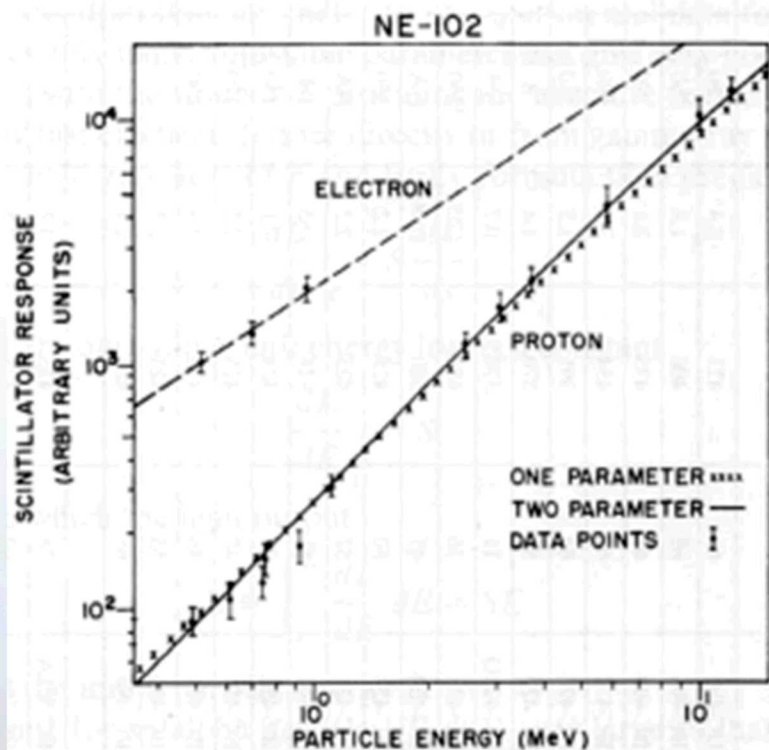


Sandia National Laboratories

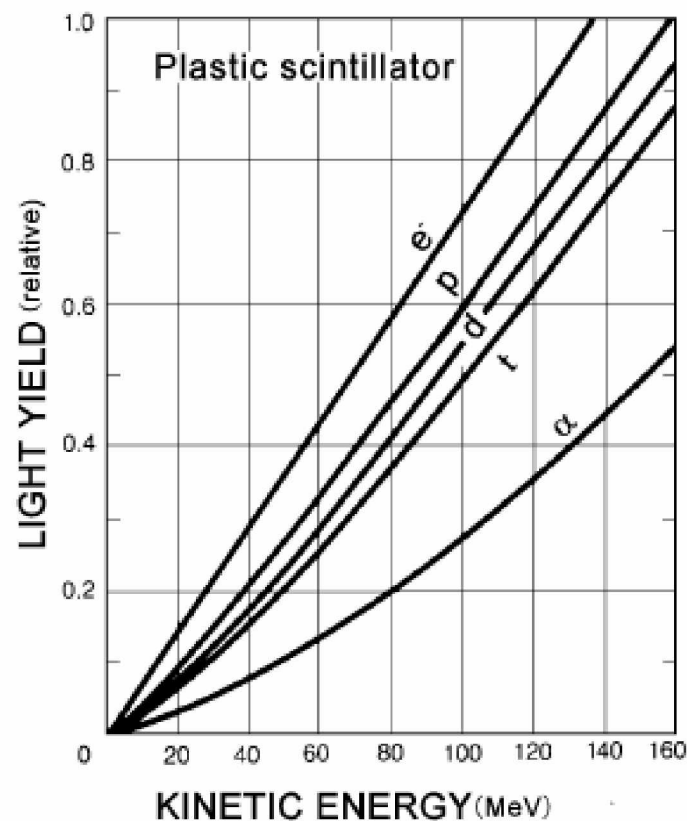
Neutron Detection – Quenching, dE/dx



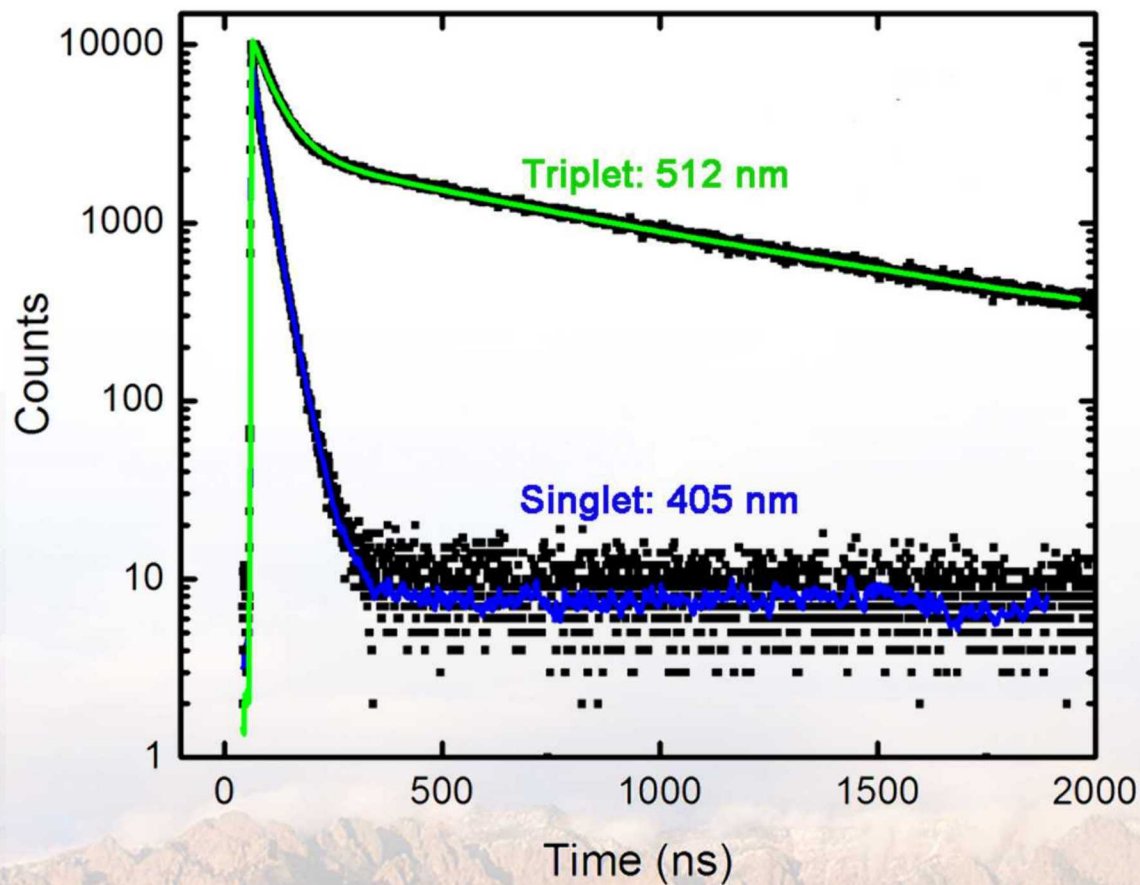
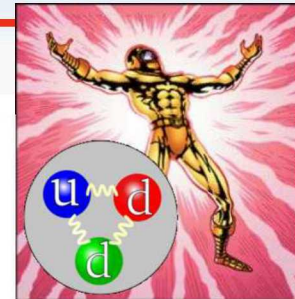
$$L(E) = \int_0^E \frac{S \cdot (dE/dx)}{1 + kB \cdot (dE/dx)} \left(\frac{dE}{dx} \right)^{-1} dE$$



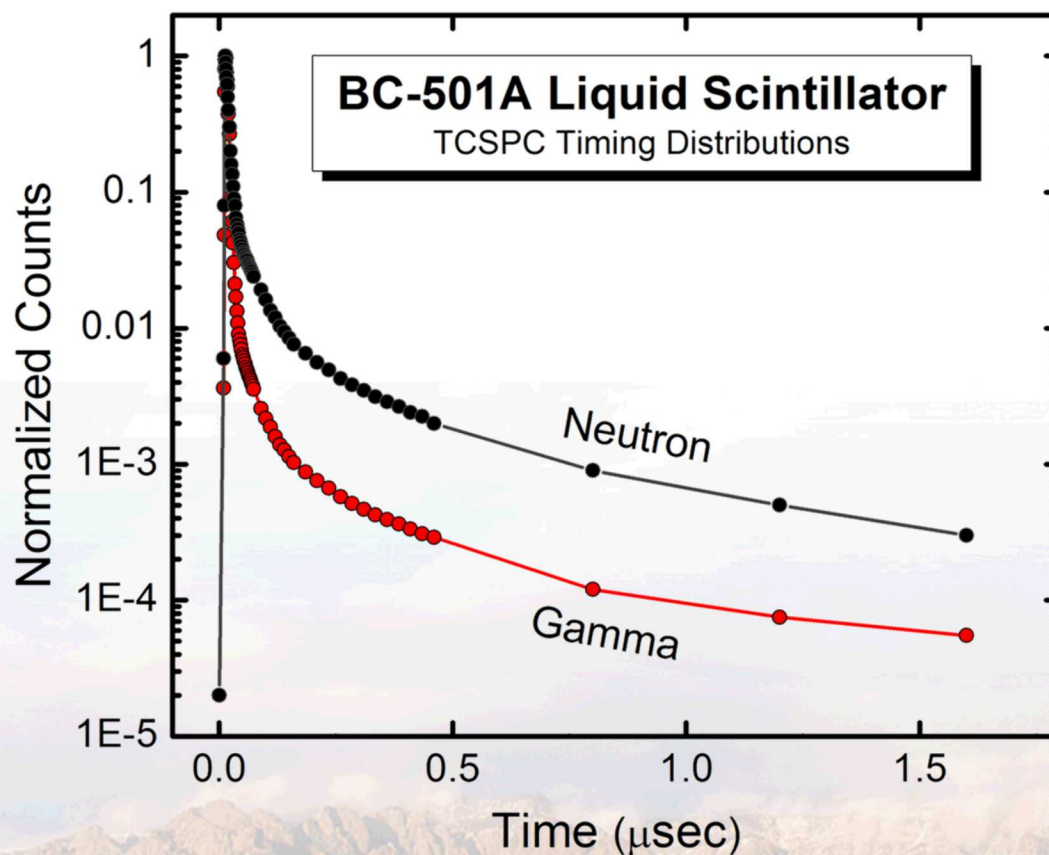
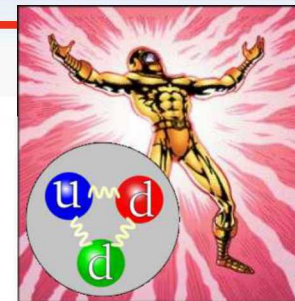
from Gooding and Pugh



Neutron Detection – Slow Component

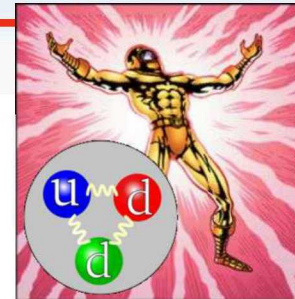


Pulse Shape Discrimination

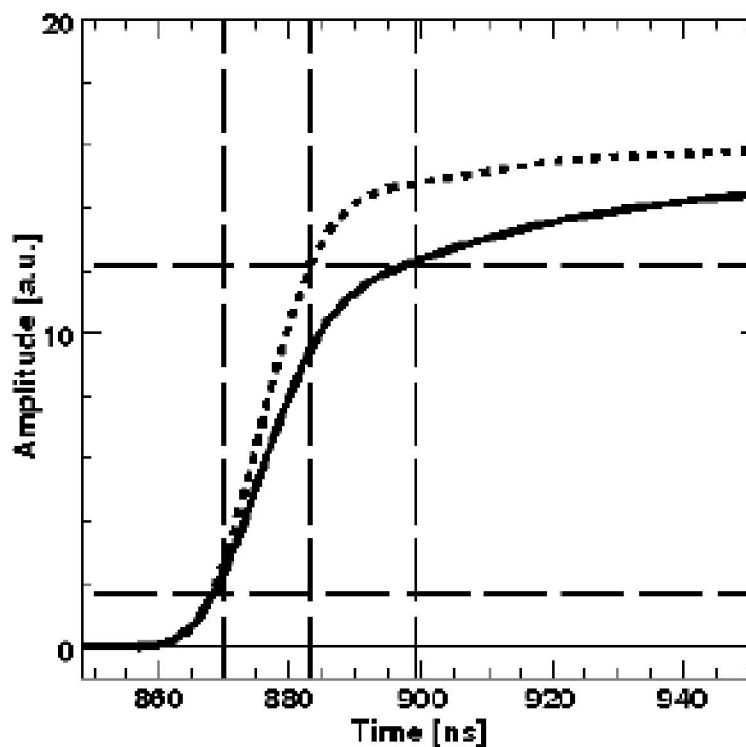


*Szczesniak, T. et al *IEEE Trans. Nucl. Sci.* **2010**, 57, 3846.

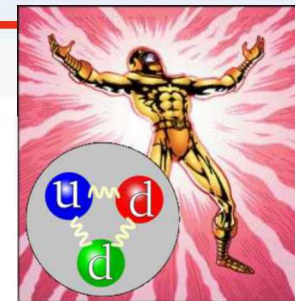
PSD - Integrated Rise Time



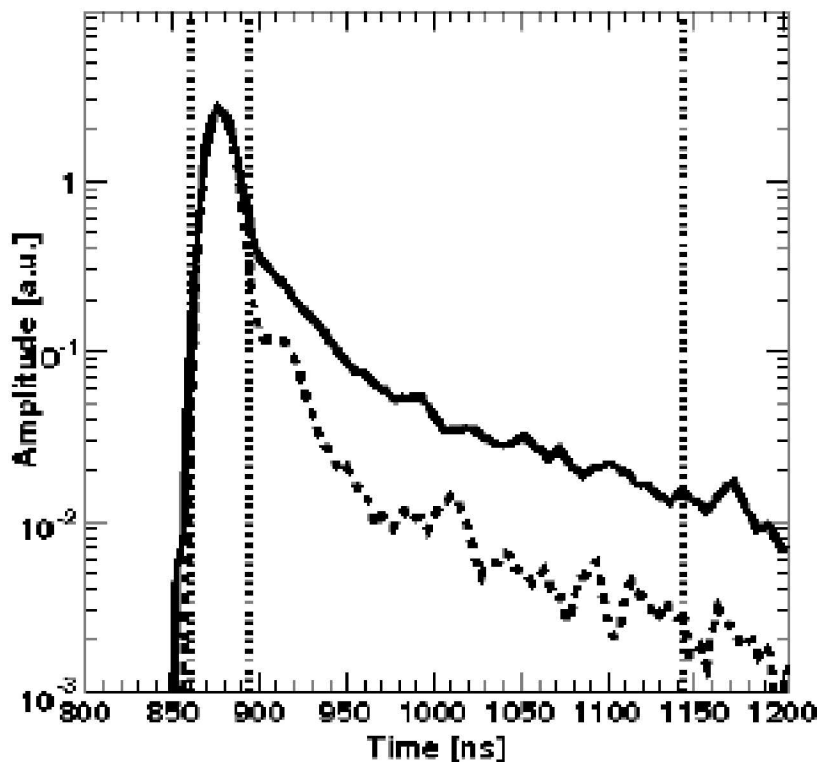
- Digitally integrate the pulse and take the rise time.
- PSD is taken as time between 10% and some fraction of total integral.



PSD - Slow component



- Locate the start of the slow decay component and compare integrals in prompt and slow.
- In this case:
Fast = 0-33 ns
Slow = 33-283 ns



PSD – Gatti and De Martini integral



Starts by leading edge

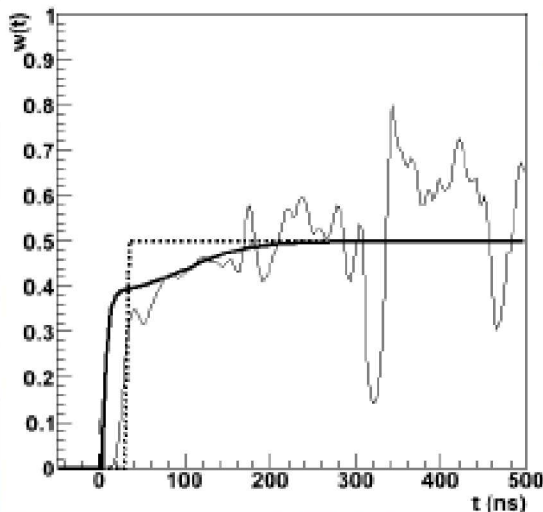
Integrate the entire pulse with a suitable weight

$$S = \int_0^T p(t) w(t) dt$$

The optimal weight can be shown to be

$$W(t) = \frac{\bar{n}(t) - \bar{y}(t)}{\sigma_n^2(t) + \sigma_y^2(t)}$$

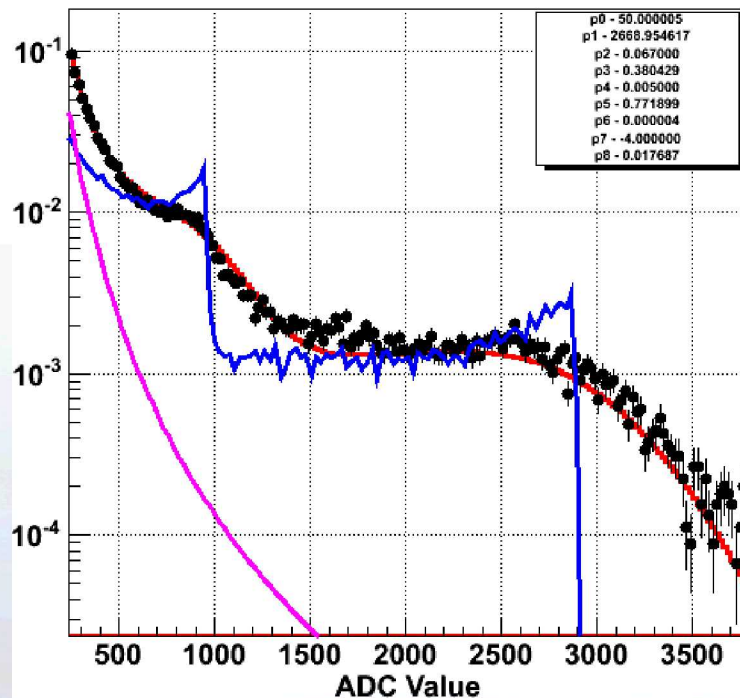
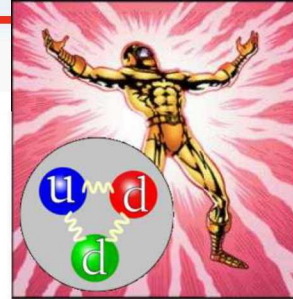
E. Gatti, F. de Martini, in: Nuclear Electronics, Proceedings of International Conference at Belgrade, Vol. II, IAEA, Vienna, 1962, p. 265.



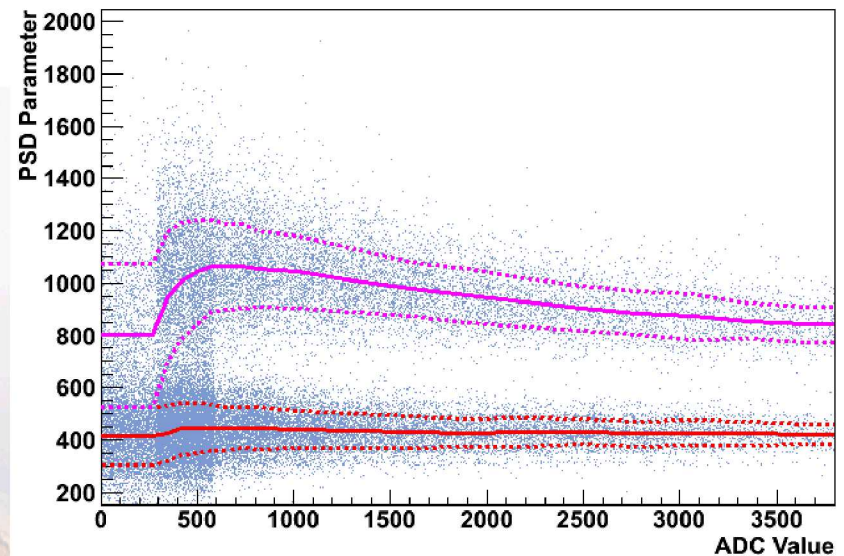
PSD – Calibration



$$FOM = \frac{|X_y - X_n|}{(W_y + W_n)}$$



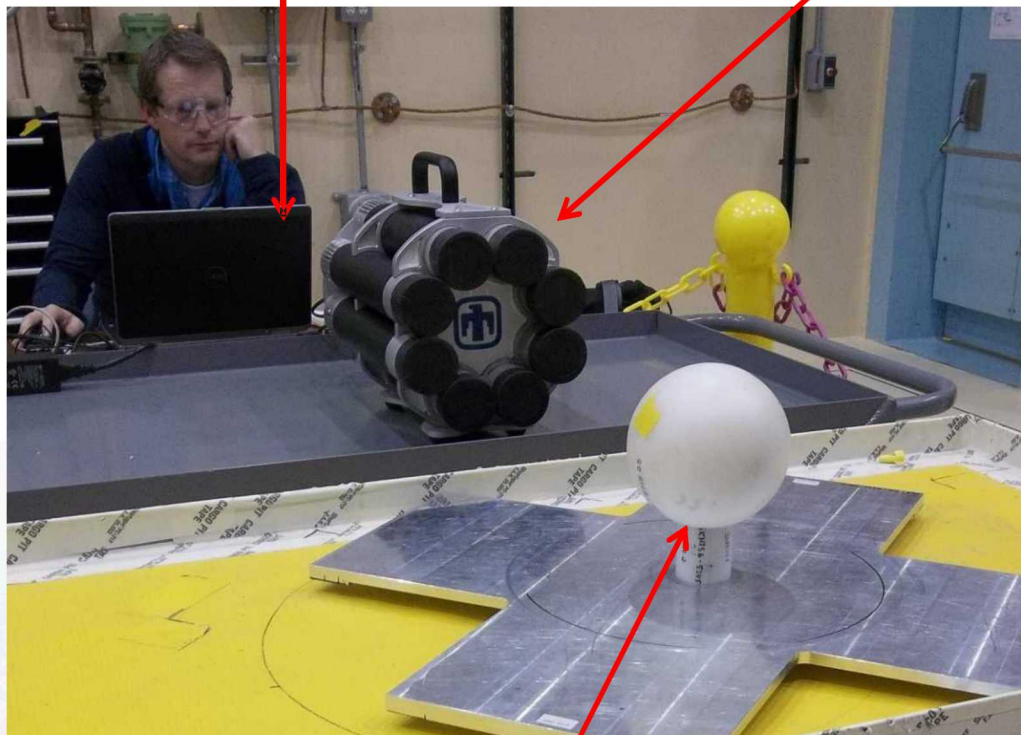
PSD vs. ADC - det1_ch0_z0



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



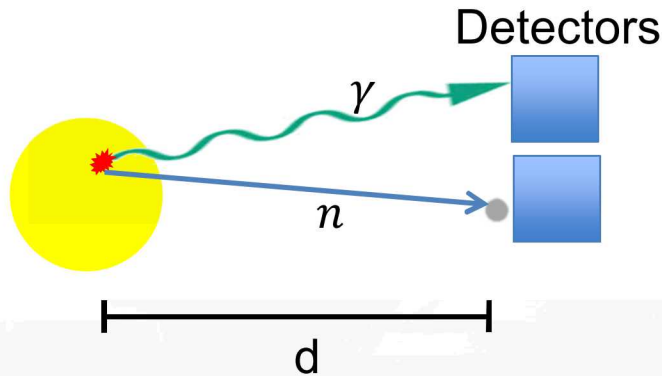
Same Fission Gamma-Neutron Correlations

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



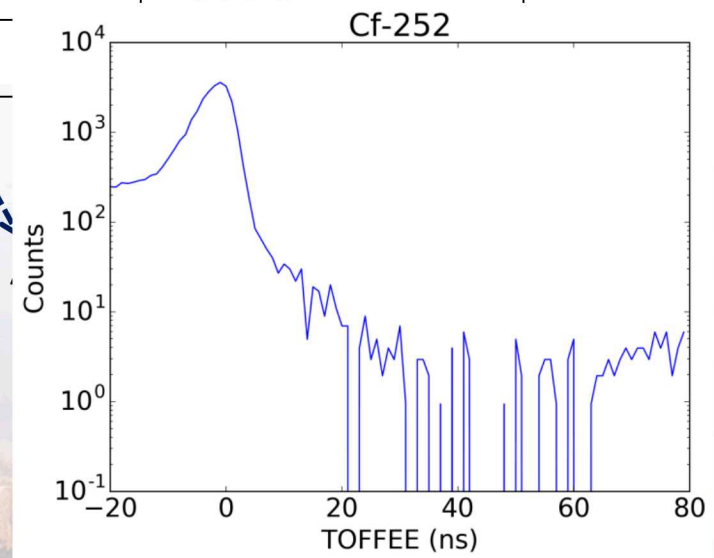
$$\Delta T_{n-\gamma} - \left(R_n \sqrt{\frac{m}{2E_n}} - \frac{R_\gamma}{c} \right) = 0$$

Time of arrival difference between γ and neutron	Time of flight difference between γ and neutron
$\Delta t_{n,\gamma}$	Δt_p
$\Delta t_{n,\gamma} - \Delta t_p \leq 0$	

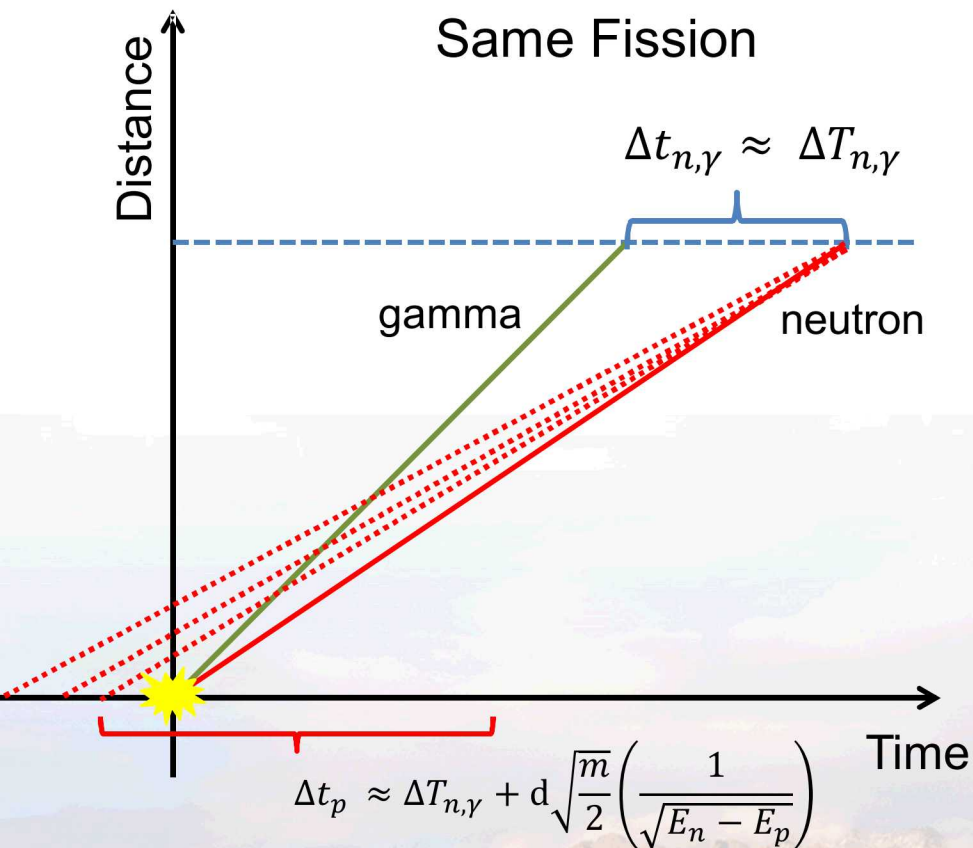
Measured

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

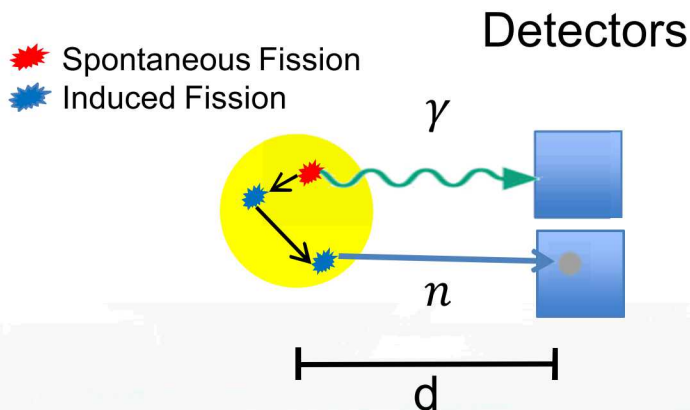


1. Neutron and gamma are born from the same fission event
2. They travel the same distance to the detectors
3. The measured and actual difference in time-of-flight is equivalent
4. When subtracting estimated time of flight of the neutron, the result is less than zero

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

Neutron-Gamma Correlation – Fission Chain

Time Of Flight Fixed by Estimated Energy (TOFFEE)



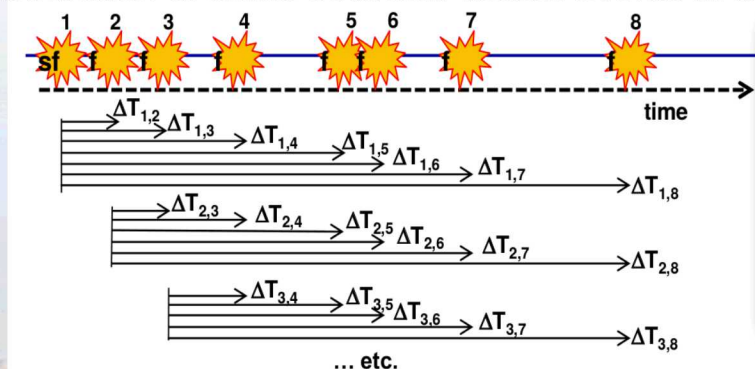
Time difference between fission events in a chain

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

Measured time difference between neutron and nearest γ

Estimated travel time difference between neutron and gamma

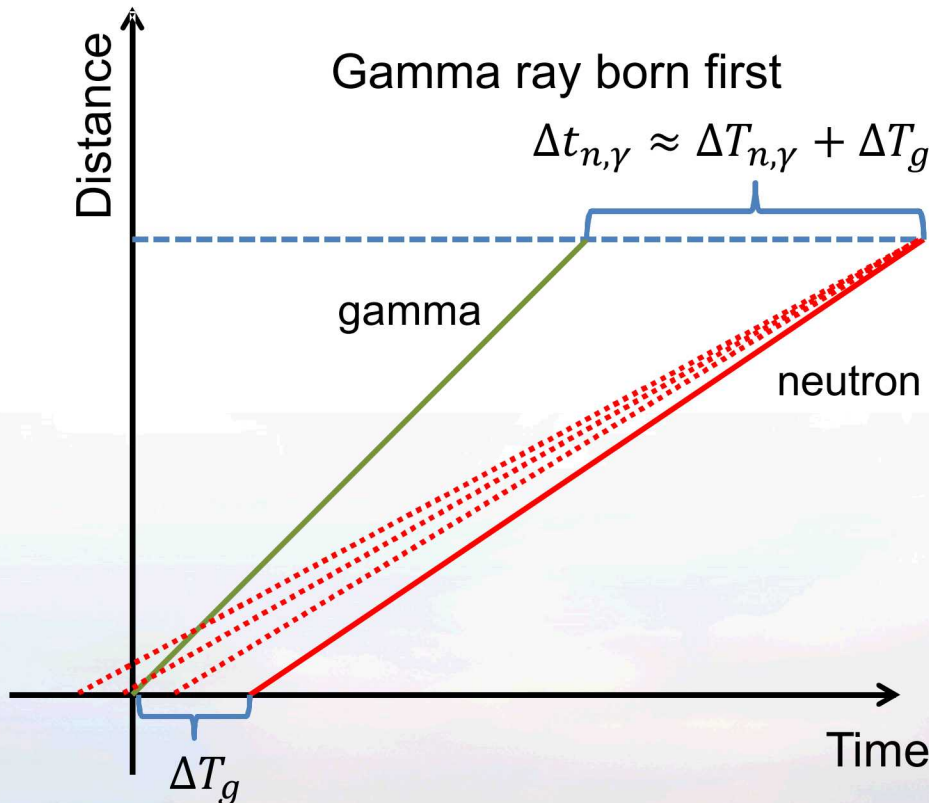
Distribution of times between fission events in a chain



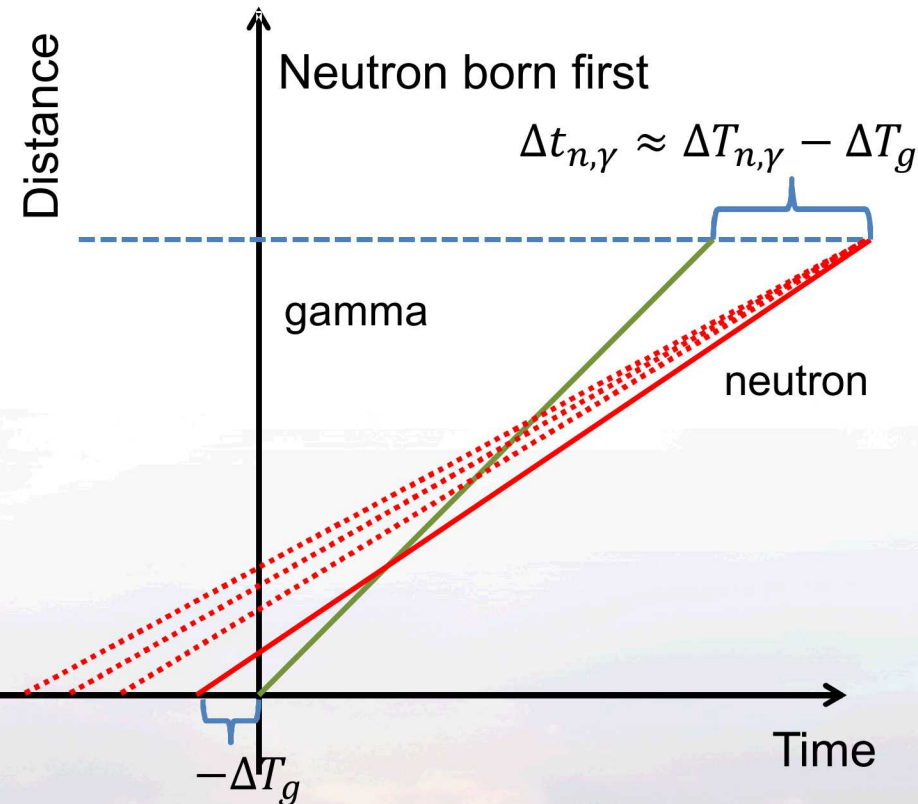
$$\Delta T_g > 0 \text{ if } f_1 = \gamma \text{ \& } f_2 = n$$

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

Fission Chains Gamma-Neutron Space-Time



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



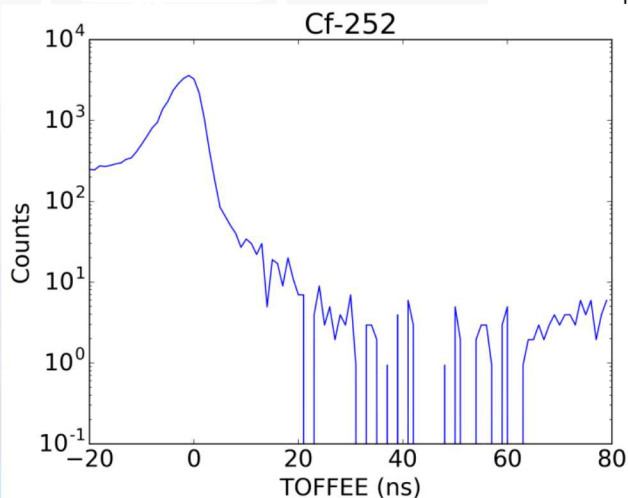
$$\Delta t_{n,\gamma} - \Delta t_p \leq -\Delta T_g$$

TOFFEE Sensitivity to Fissile Material and Configuration

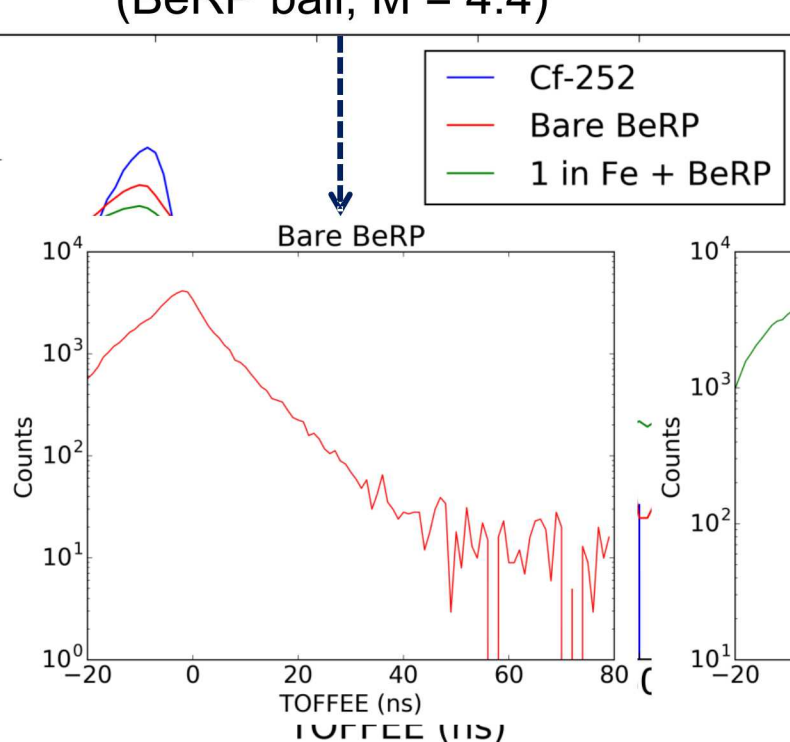
TOFFEE is sensitive to:

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

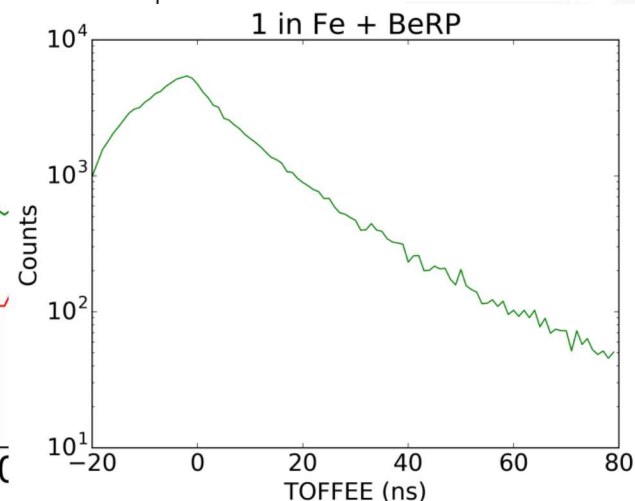
Non-fissile fission source
(Cf-252, $M=0$)



Fissile material
(BeRP ball, $M = 4.4$)



Reflected Fissile
material
(BeRP ball, $M = 6.48$)

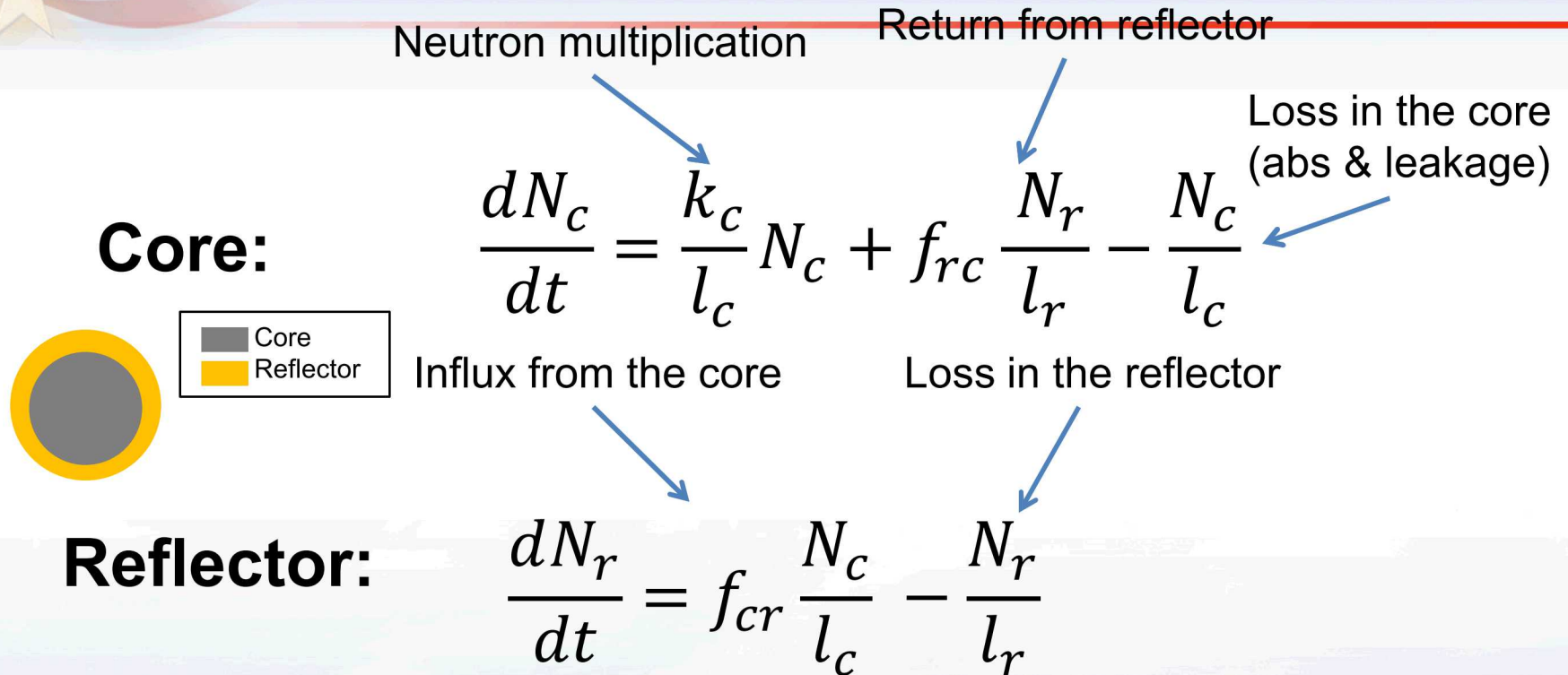


Extracting Physical Parameters from TOFFEE

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



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

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

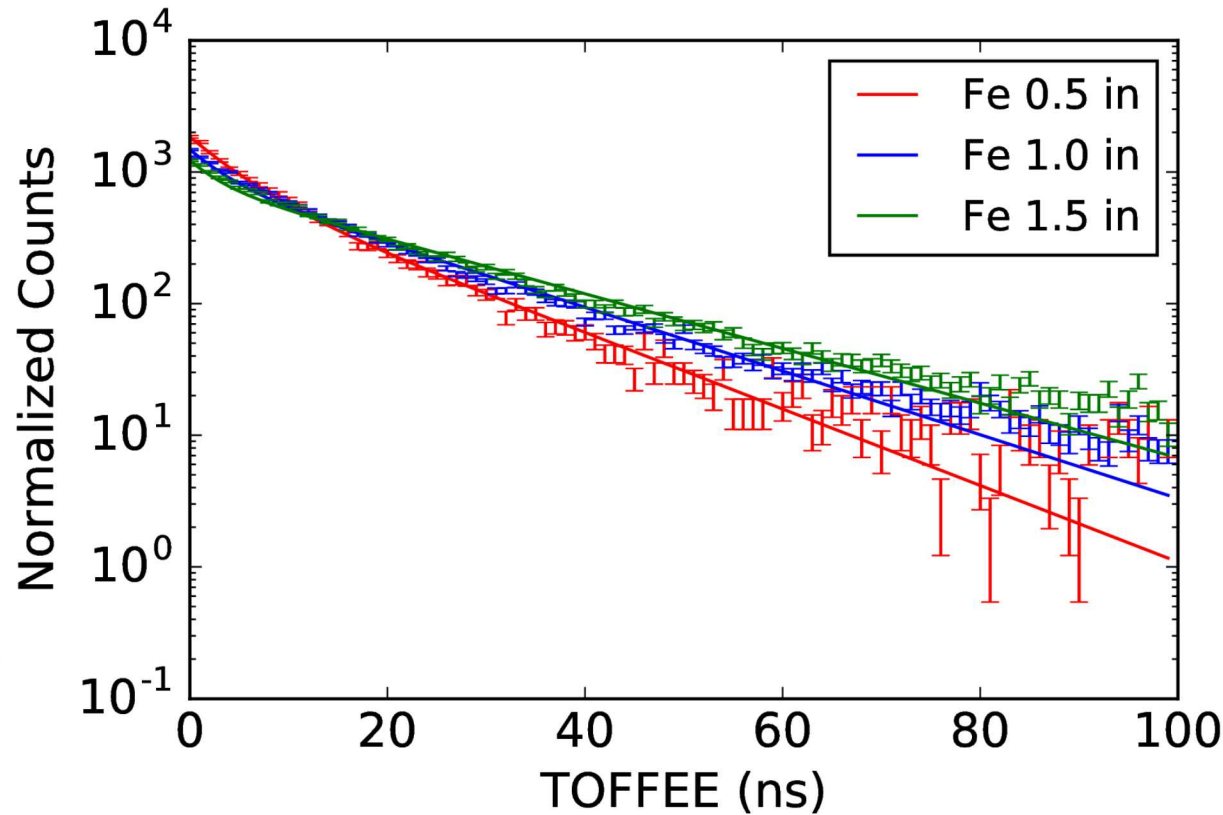
$$r_{1,2} = \frac{\pm \sqrt{4l_cl_r(f + k_c - 1) + (l_c - l_r(k_c - 1))^2} - l_c + l_r(k_c - 1)}{2l_cl_r}$$

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

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

Fitting Strategy for Reflected Configurations

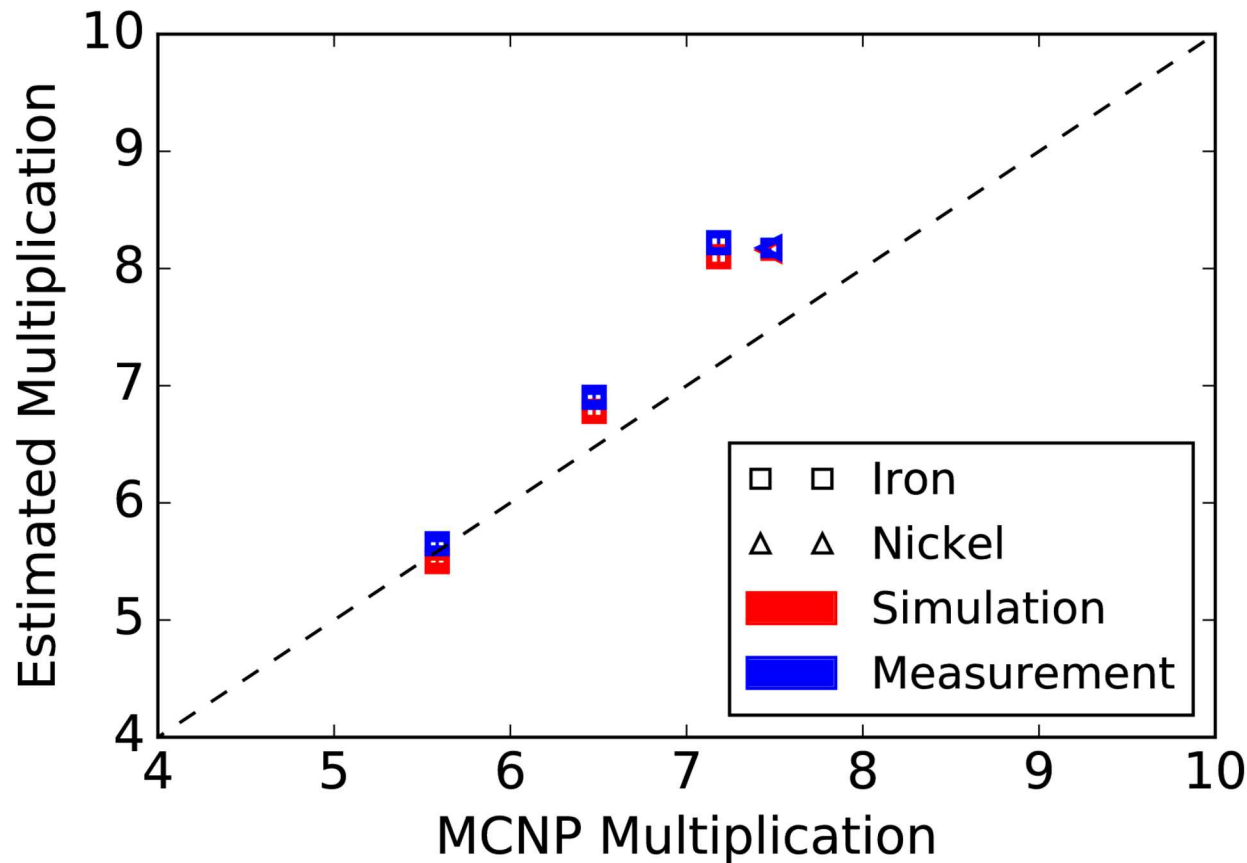
$$N_c(t) = N_o[(1 - R)e^{tr_1} + Re^{tr_2}]$$



- 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

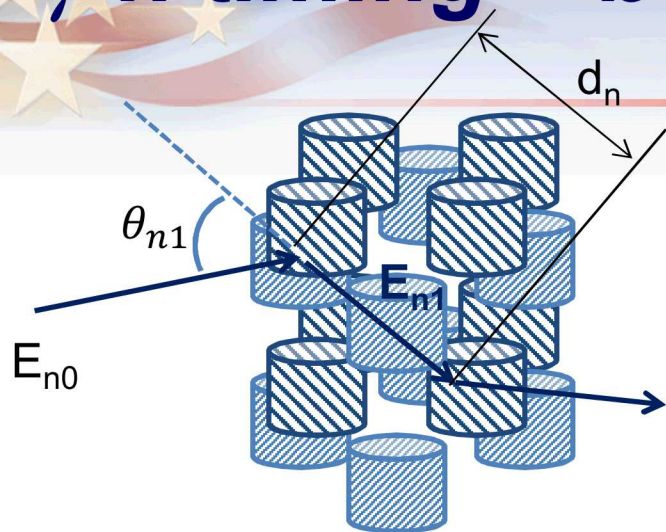
Estimating Multiplication of Reflected Assemblies

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

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

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

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

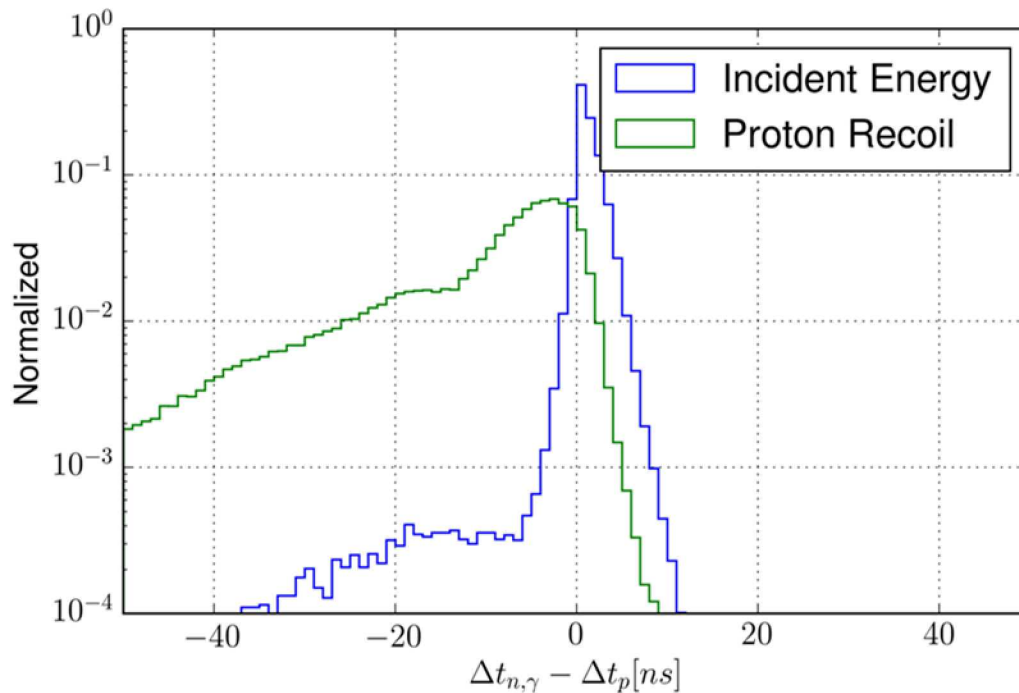


MINER: the Mobile Imager of Neutrons for Emergency Response

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

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

Cf-252 Simulation



Different Multiplications

