

Exceptional service in the national interest



Nuclear Material Characterization and Imaging using fast neutrons

Peter Marleau

UC Davis NSSC Summer School, 2016



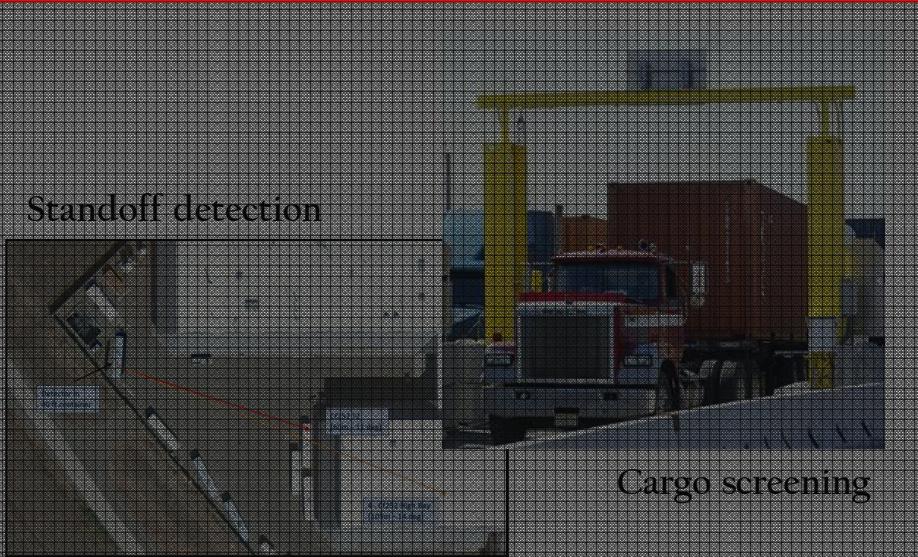
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Sandia National Laboratories



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

SNM characterization/imaging

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

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
- Warhead confirmation for verification of treaty compliance
 - Challenge problem
 - Authentication vs. Certification
 - Comparison measurements
 - CONfirmation using a Fast-neutron Imaging Detector with Anti-image NULL-positive Time Encoding (CONFIDANTE)

Nuclear Material?

As defined by the IAEA:

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

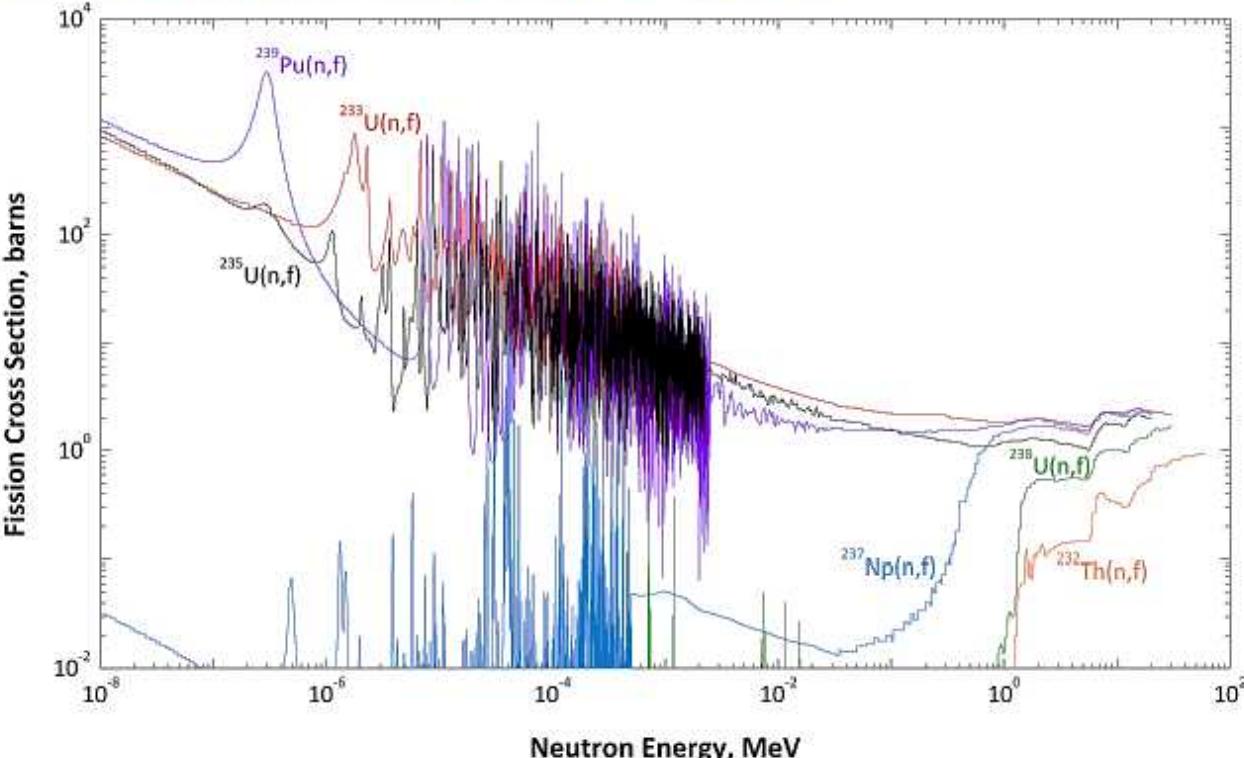
Special Nuclear (fissile) Material – U-233, U-235, Pu-239

Source Material – everything that is not special

What's so special?

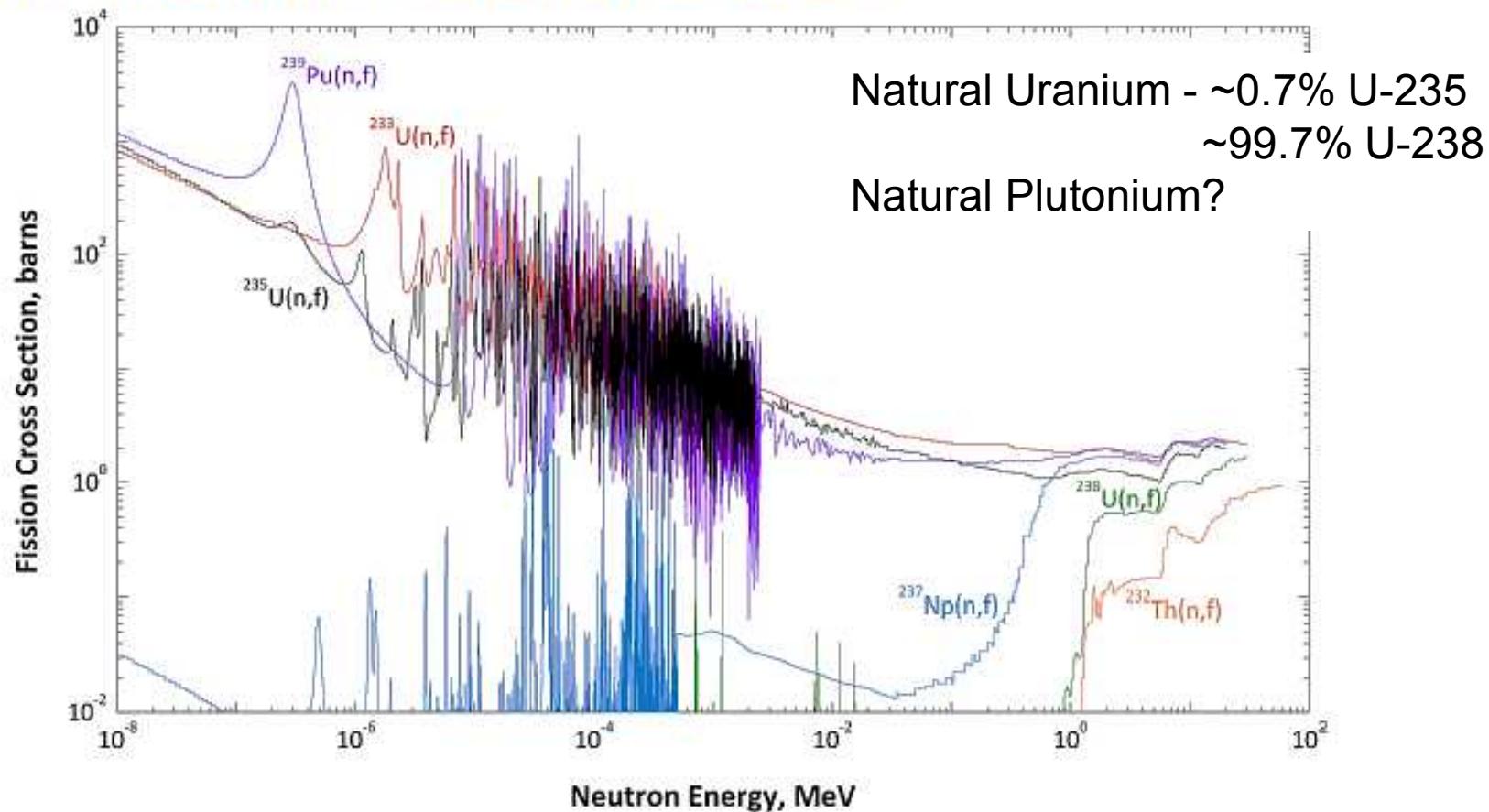
Isotope	Induced Thermal Fission Multiplicity ν
^{232}U	3.13
^{233}U	2.4
^{234}U	2.4
^{235}U	2.41
^{236}U	2.2
^{238}U	2.3
^{237}Np	2.70
^{238}Pu	2.9
^{239}Pu	2.88
^{240}Pu	2.8
^{241}Pu	2.8
^{242}Pu	2.81
^{244}Cm	3.46
^{252}Cf	4.06

The Neutron Fission Cross Sections



Where can I get some?

The Neutron Fission Cross Sections

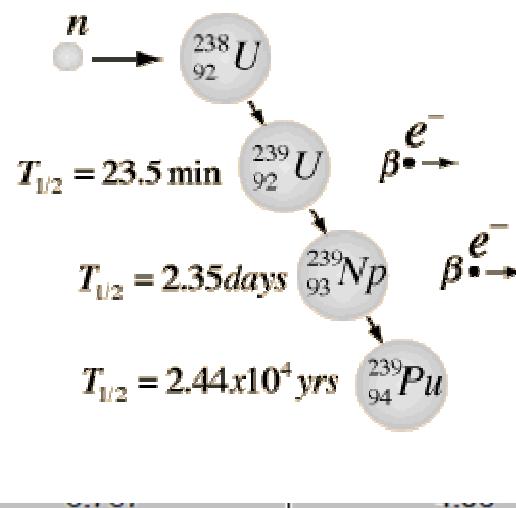


Can I make it?

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}		

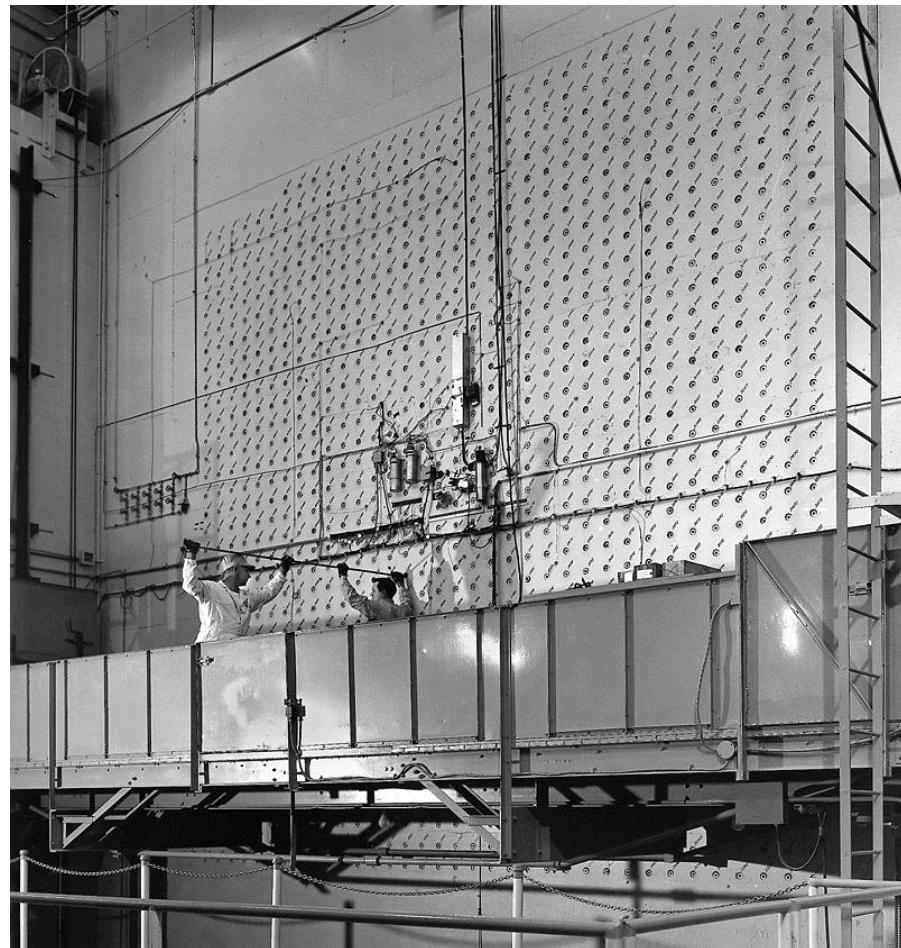
There isn't natural Plutonium to be found, but ...



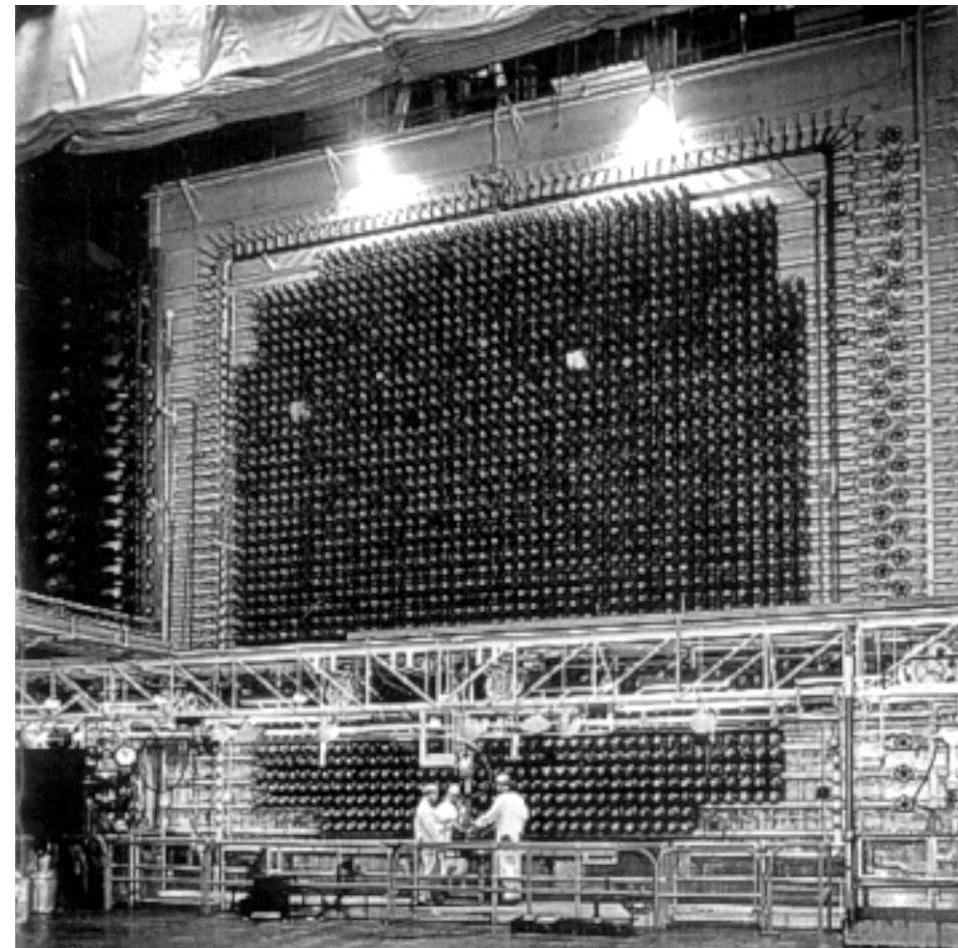
Ref: "Panda Book", values with \pm have significant uncertainty

First Nuclear Reactors

ORNL X-10 Graphite Reactor
graphite moderated, air cooled

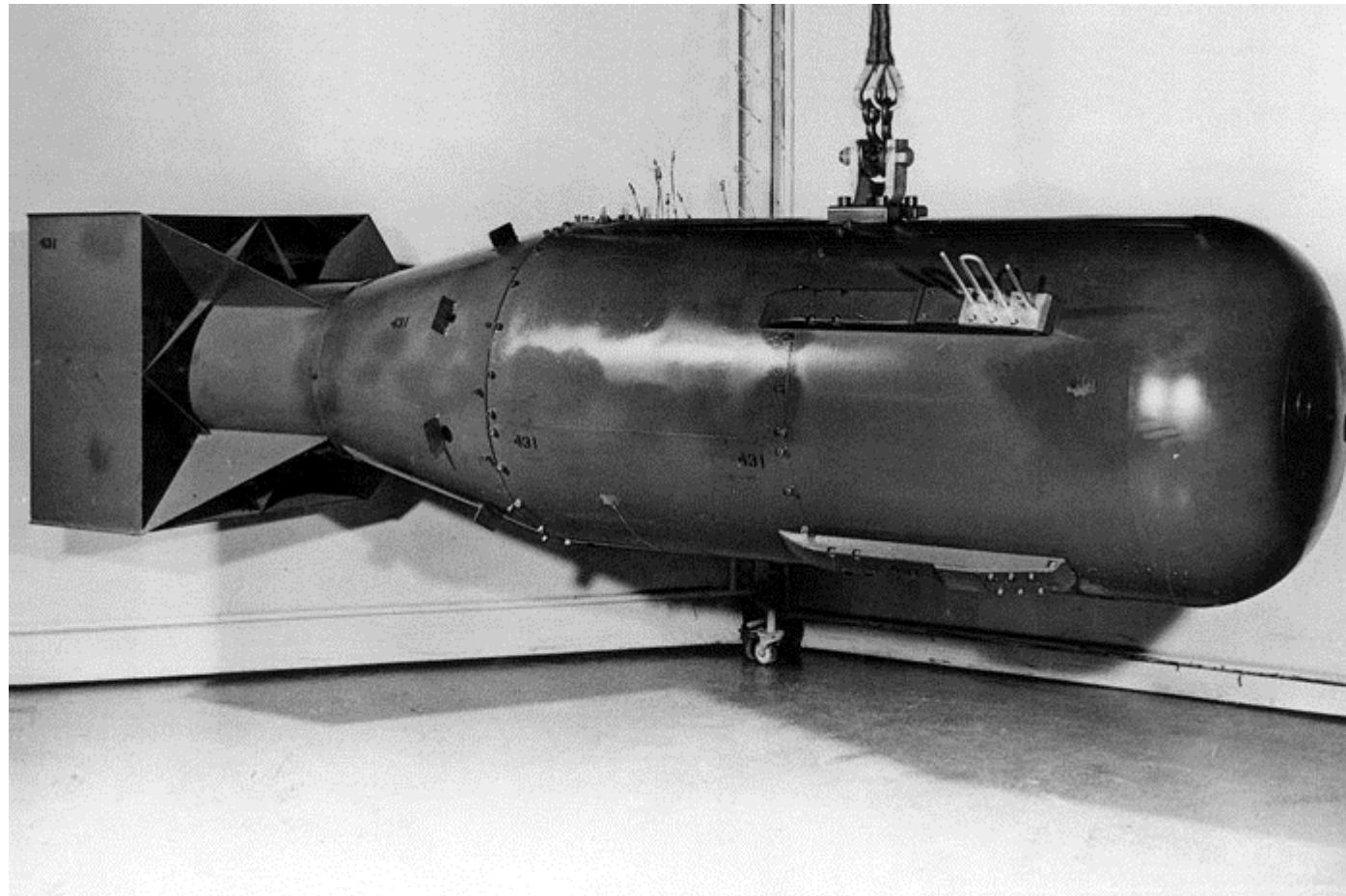


Hanford B Reactor
graphite moderated, water cooled



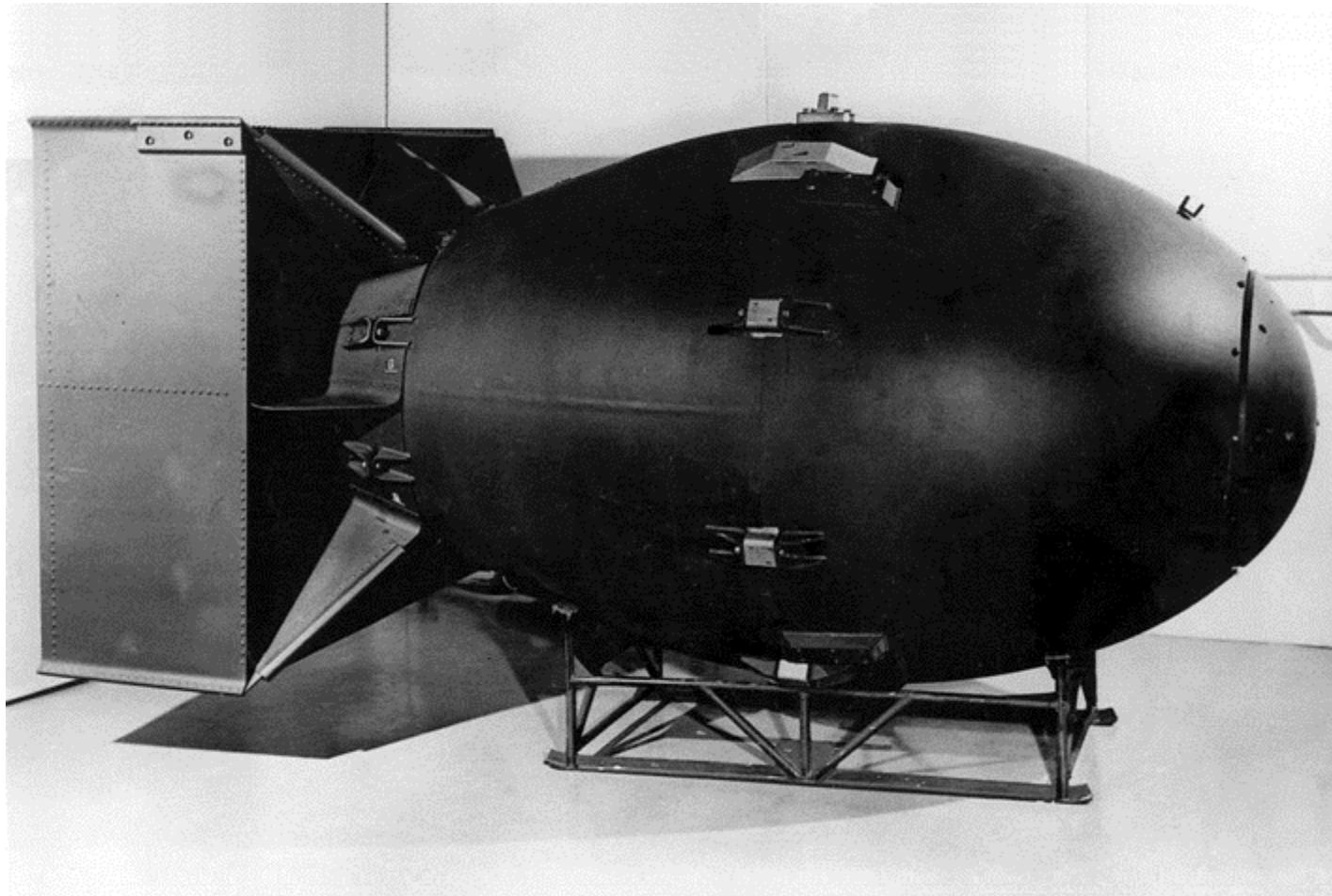
Little Boy

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

3. Total neutron rate

- Assay of the contents of specific materials

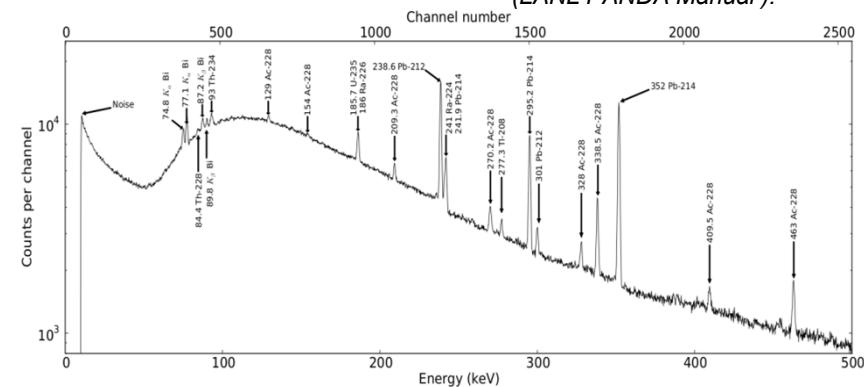
4. Correlated counts

- 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

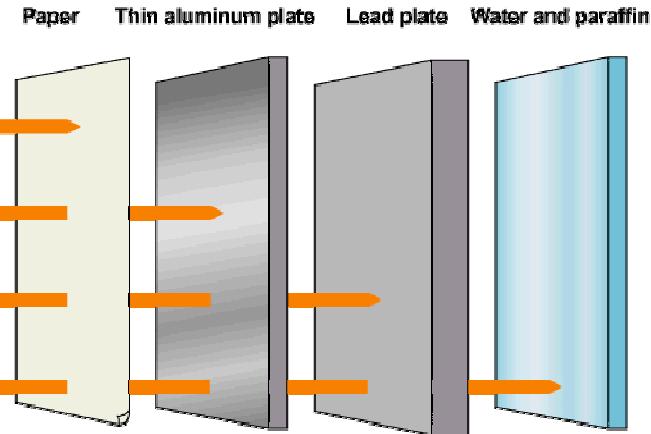
Gamma-rays

Isotope	Energy (keV)	Activity ($\gamma/\text{g-s}$)
^{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 ($\gamma/\text{g-s}$)	Mean Free Path (mm) (High-Z, ρ)	Mean Free Path (mm) (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"

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

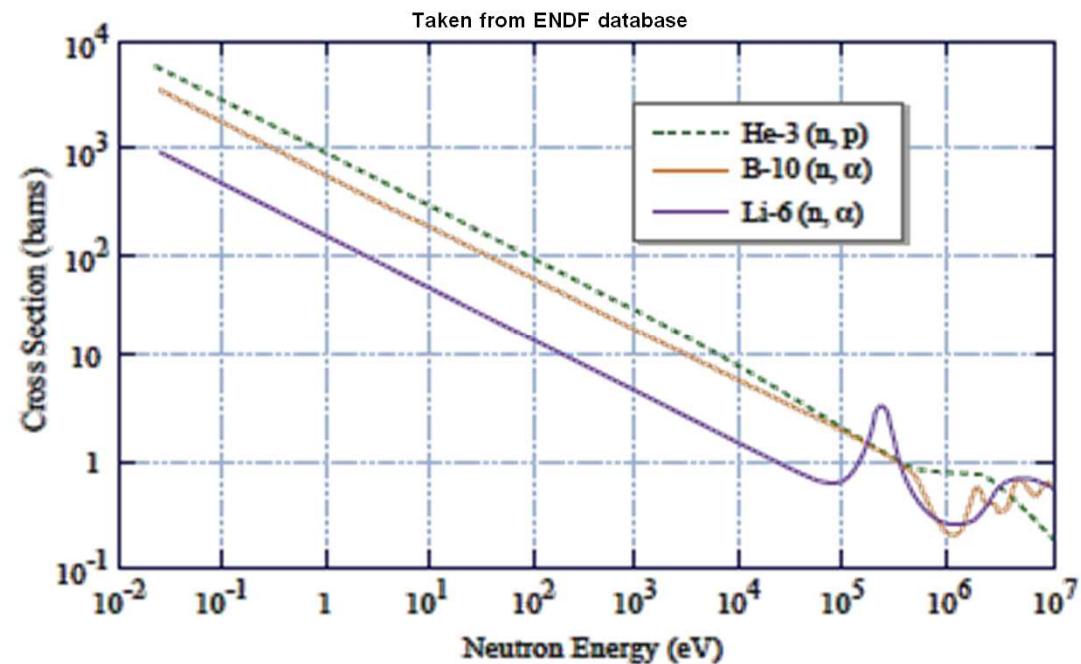
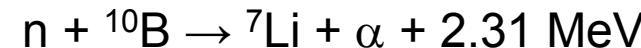
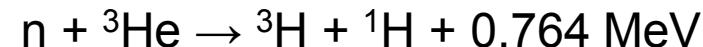
4. Correlated counts

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

 He-3 based technologies

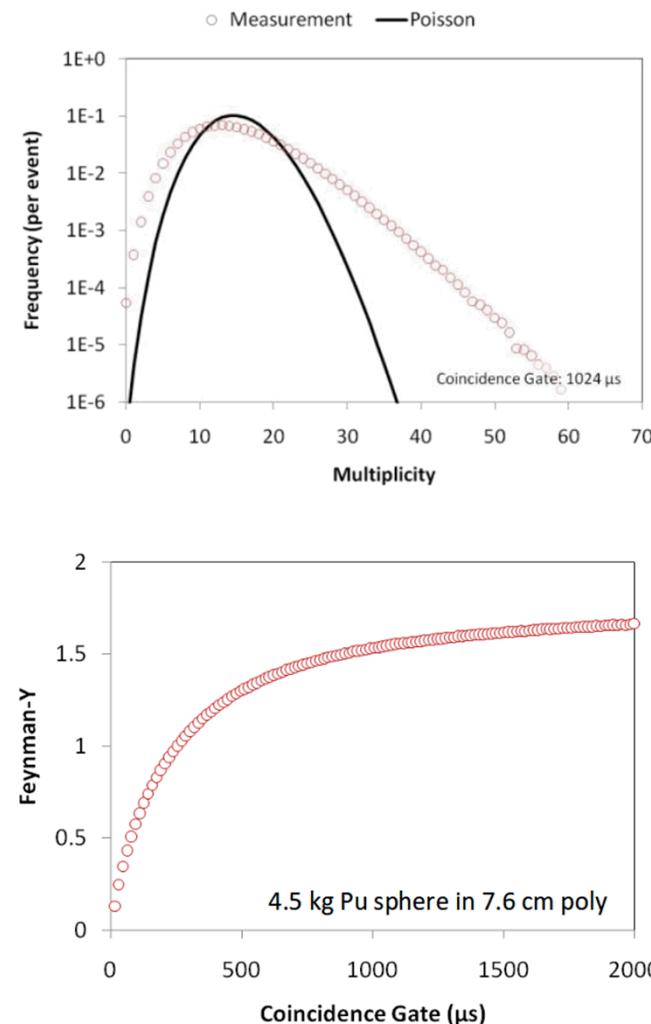
Thermal Neutron Detection

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



Feynman-Y Approach

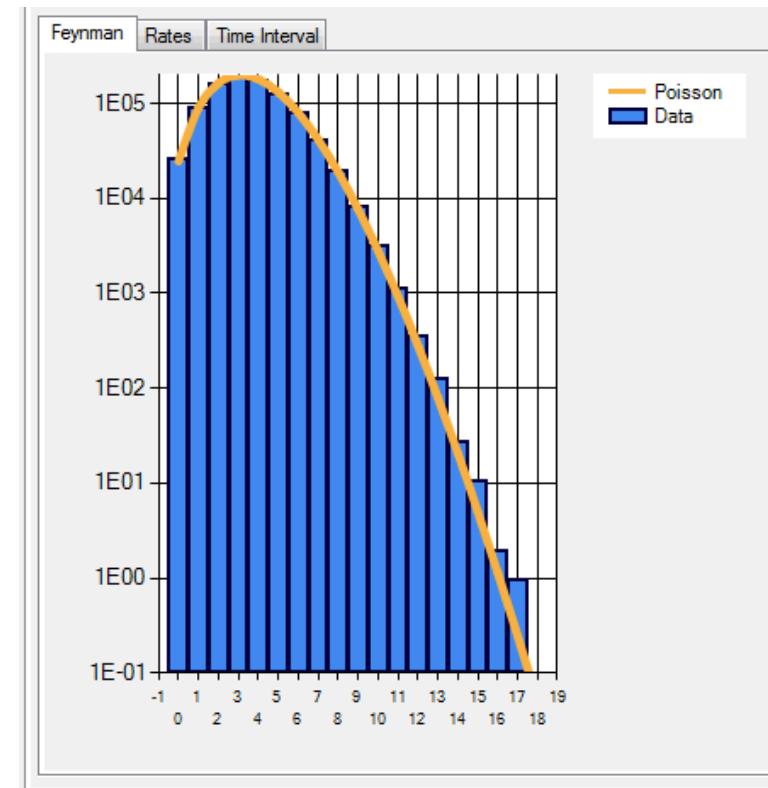
- Measures correlated counts in a fixed gate
- Fission chains create variance in excess of Poisson distribution
- $$\frac{\sigma^2}{\mu} = 1 + Y$$
 - σ^2 : variance
 - μ : mean



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

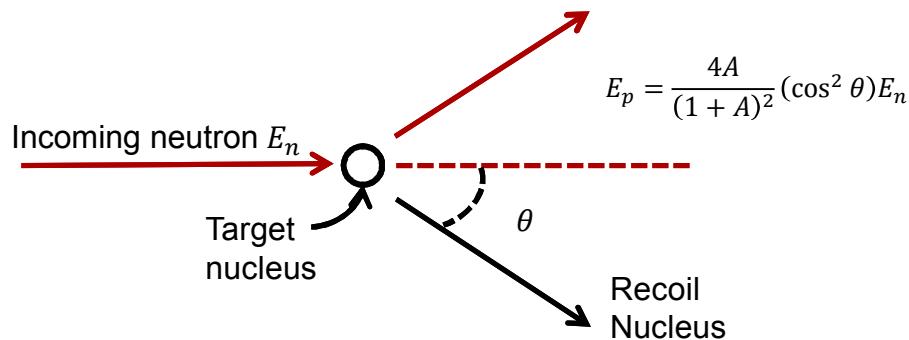
Motivation for using correlated timing

- Multiplicity analysis requires:
 1. High efficiency
 2. Strong knowledge of the efficiency
 - a) Distance to the source.
 - b) Room return.
 - c) Neutron energy spectrum.
 3. Properties of neutron emitting material (multiplicity, cross sections, etc.)
 - a) Isotopes – $\bar{\nu}$
 - b) (α, n) – uncorrelated singles
- We focus on fast fission chain timing:
 1. Can be low efficiency
 2. Does not require knowledge of that efficiency
 3. Can statistically identify the contributions due to spontaneous fission/ (α, n) and fission chains.

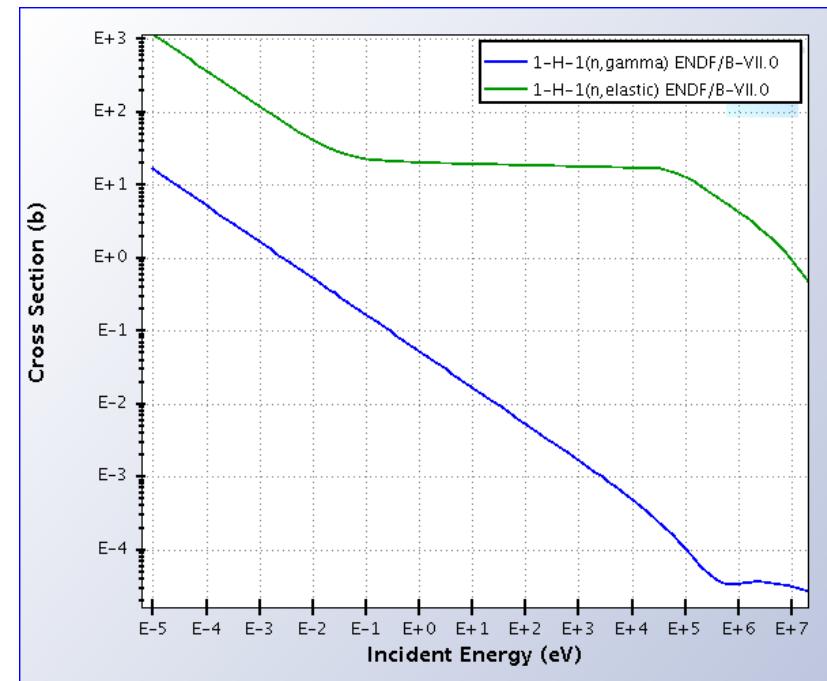


Fast Neutron Detection - Elastic

Deposited Neutron Energy



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



Pulse Shape Discrimination

Pulse Shape Dependence on Interacting Particle

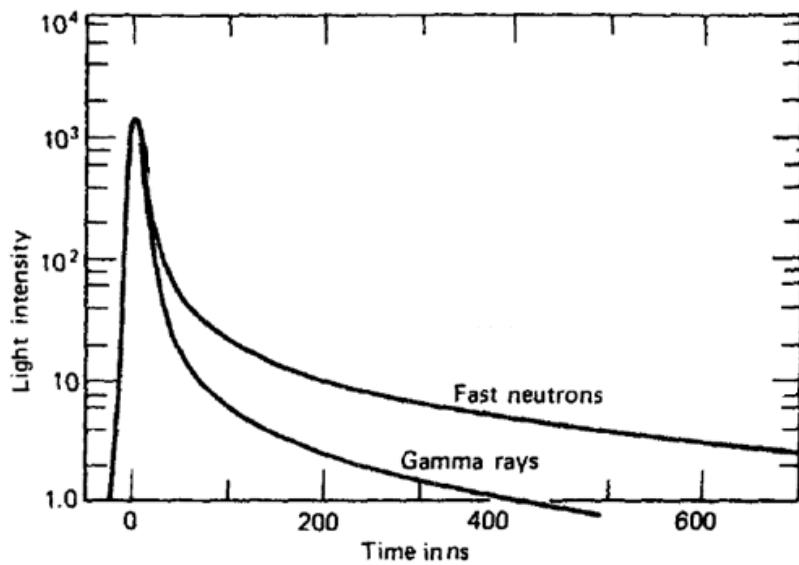
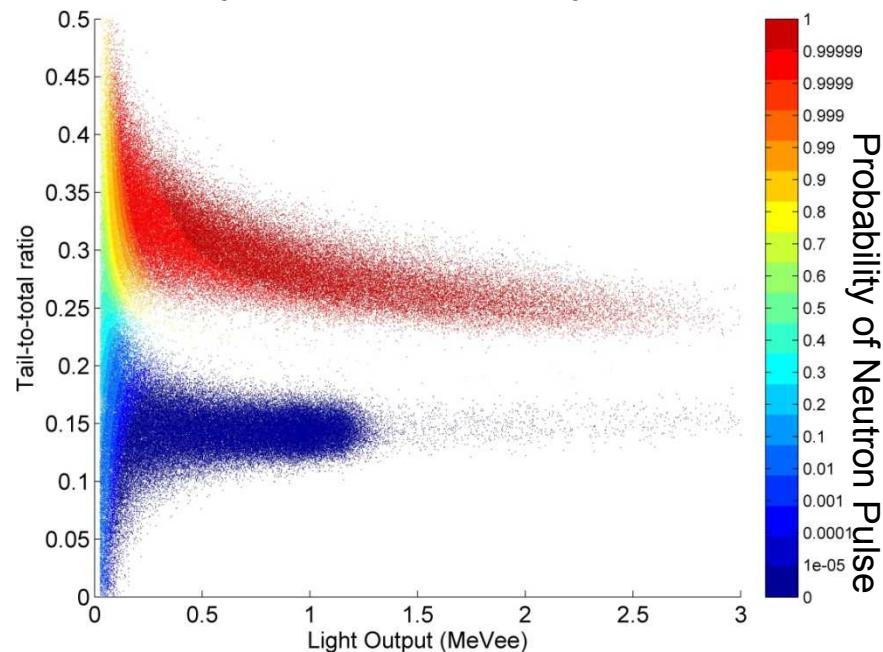
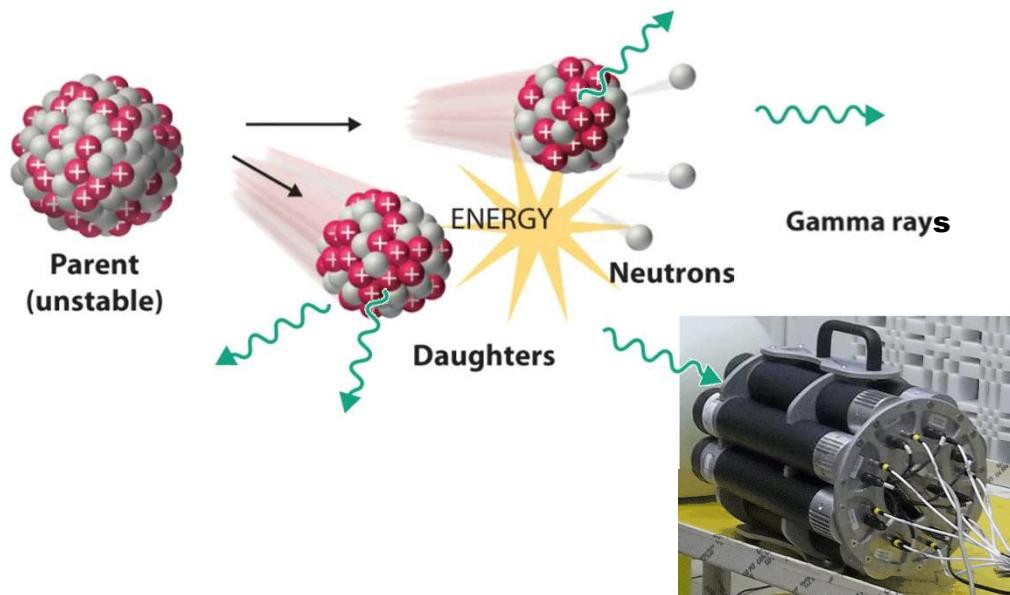


Figure from Glenn Knoll *Radiation Detection & Measurement 3rd Edition*

Bayesian Probability Map

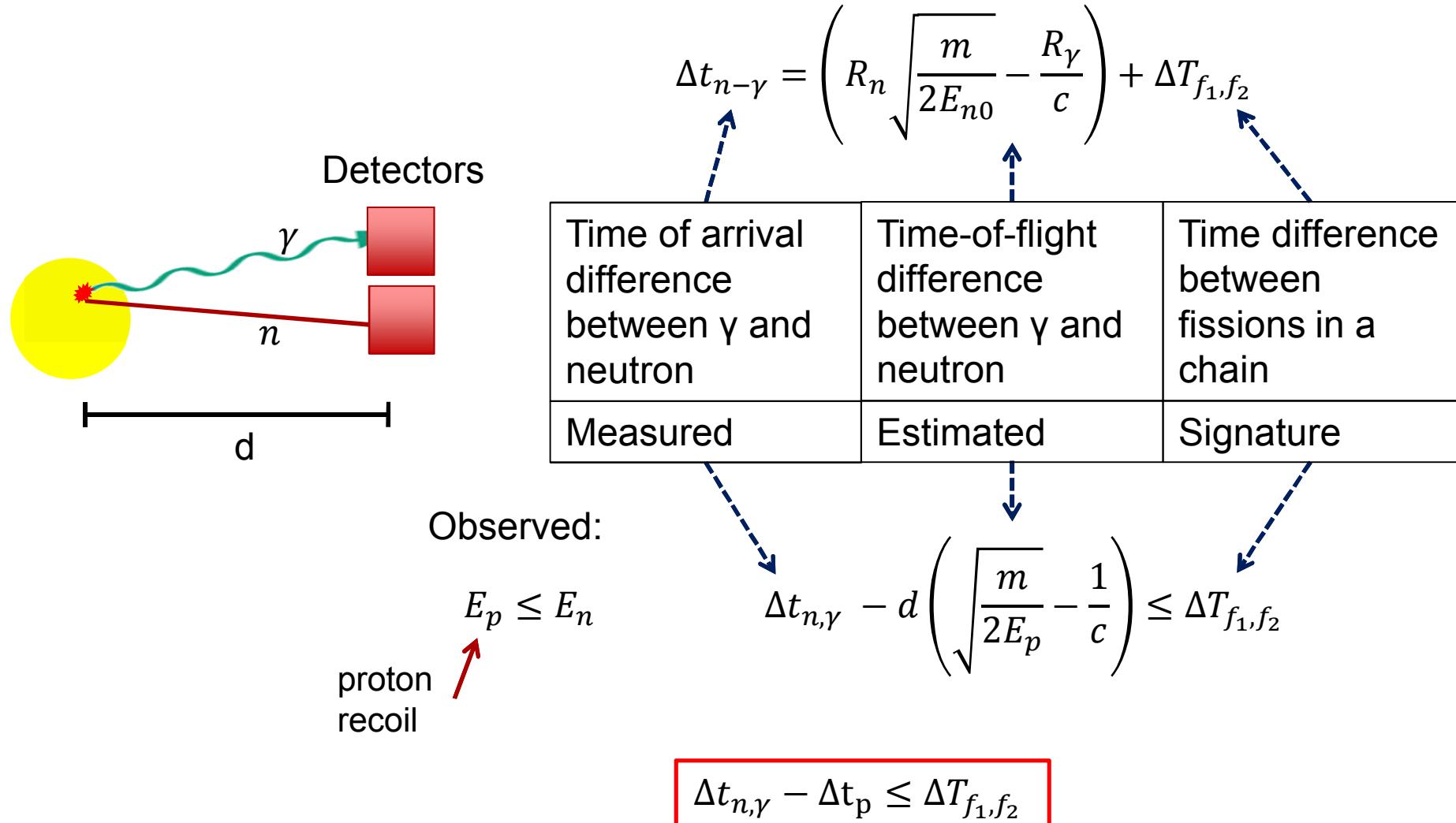


Multiplicity 2 correlation

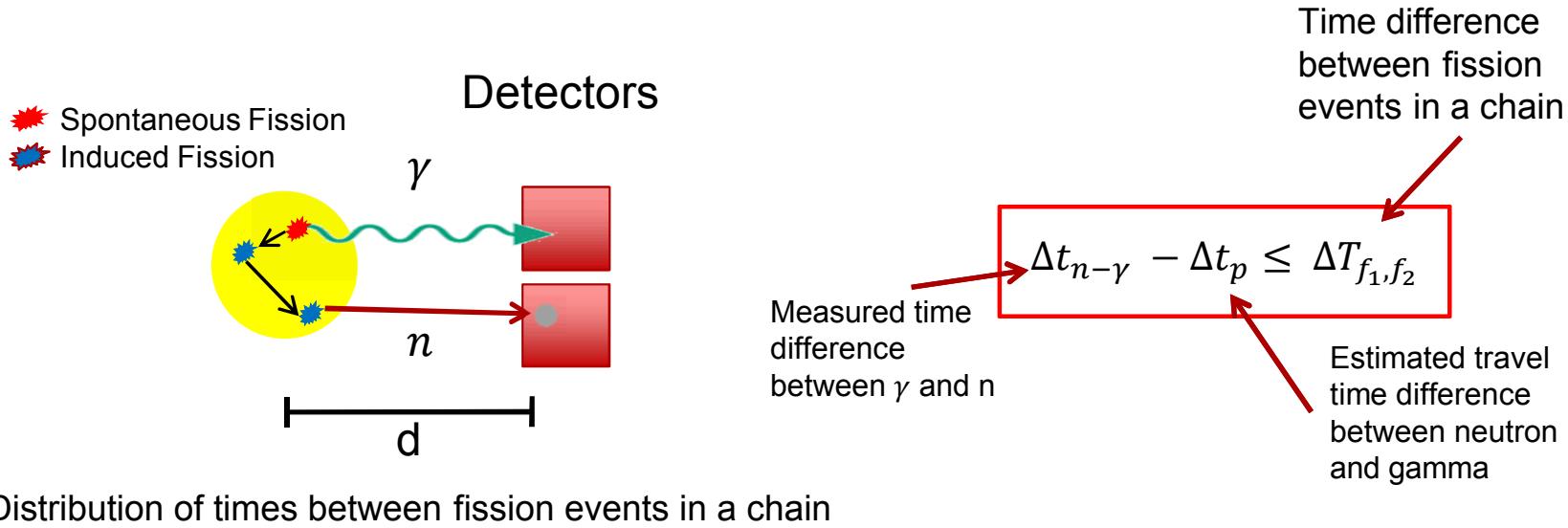


- Neutrons and gamma-rays are emitted nearly simultaneously during the fission process.
- Minimum multiplicity equals two:
 1. **Gamma-gamma:** great for timing, but a lot of detector cross talk and uncorrelated background.
 2. **Neutron-neutron:** without event by event energy, expected correlated timing 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).

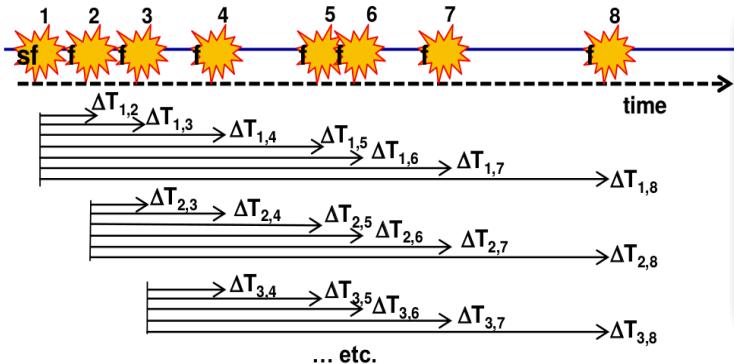
Neutron-Gamma Time Correlation



Neutron-Gamma Correlation – Fission Chain



Distribution of times between fission events in a chain



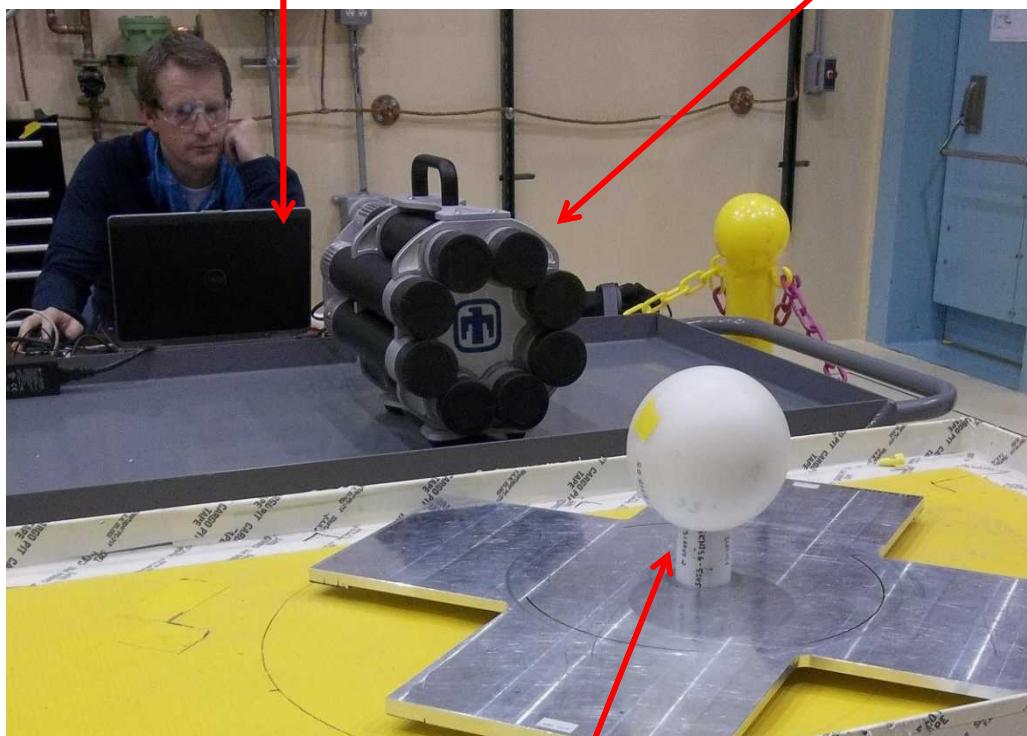
$$\Delta T_{f_1,f_2} > 0 \text{ if } f_1 = \gamma \text{ & } f_2 = n$$

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

Correlated Radiation Signatures - Stilbene Array

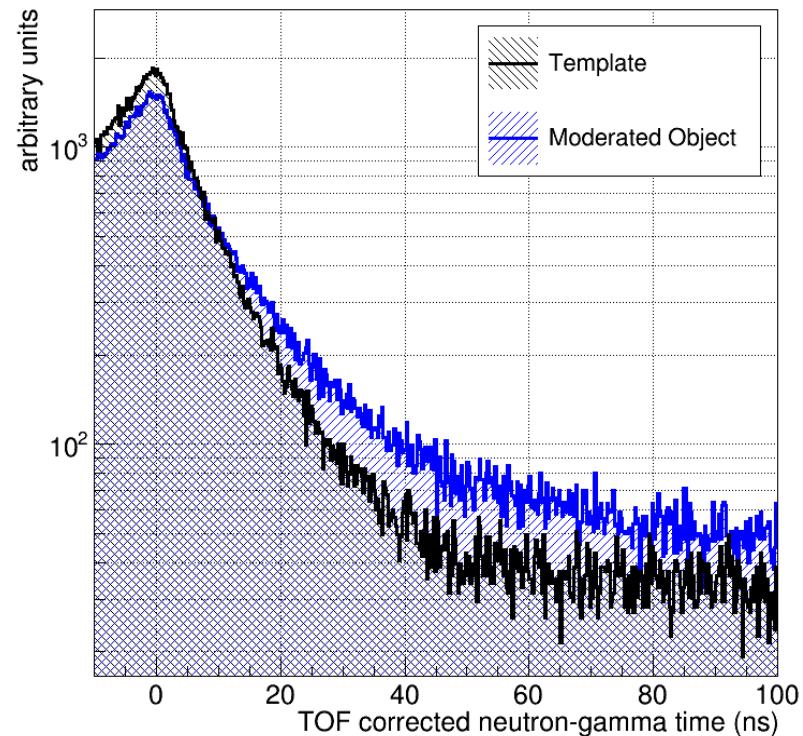
Computer/digitizer

Stilbene array



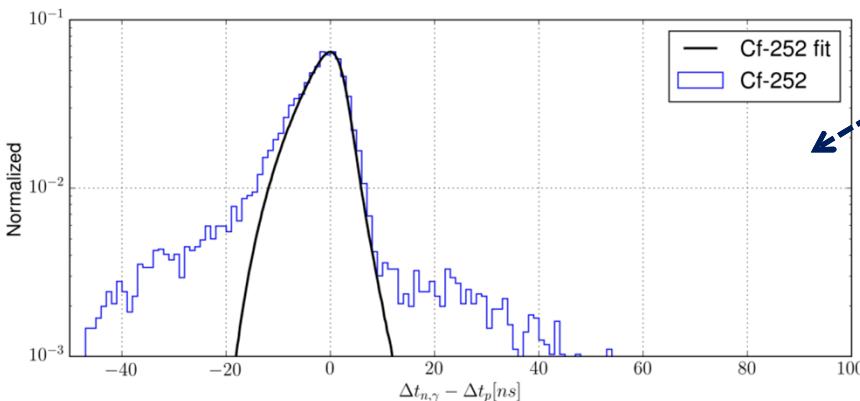
Beryllium Reflected Plutonium (BeRP) ball
in a 1" shell of High Density Polyethylene

Timing distributions for the bare
and moderated BeRP ball

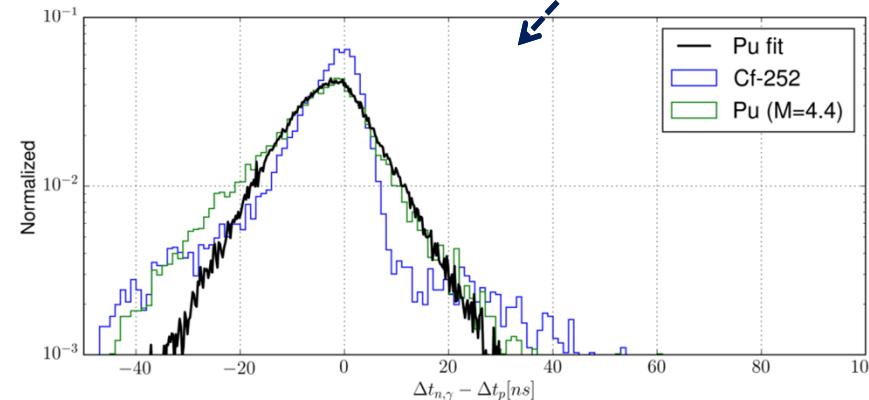


99% True Positive,
1% False Positive:
8 seconds

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



Non-fissile fission source



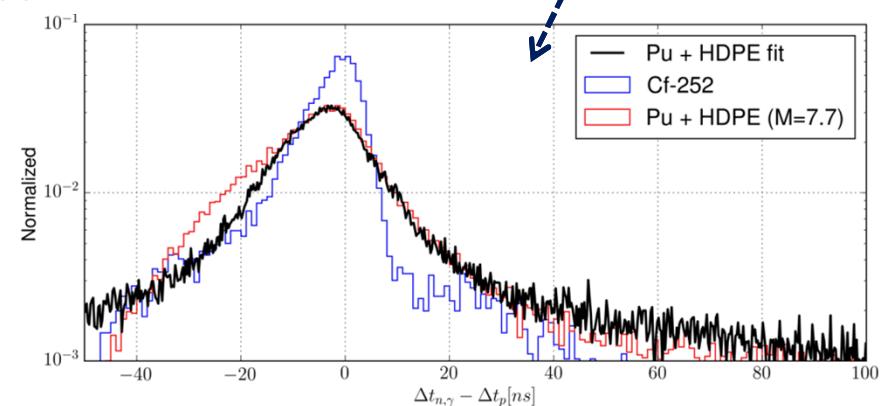
Fissile material
(BeRP ball, $M = 4.4$)

Moderated Fissile
material
(BeRP ball, $M = 7.7$)

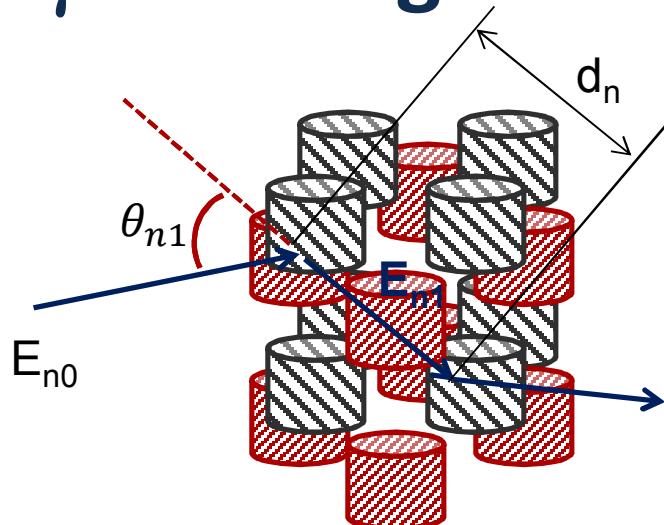
$\Delta T_{f_1, f_2}$ can discriminate:

1. Fissile vs. fissionable
2. Fissile, different multiplications
3. Moderated vs. reflected or bare

Simple fission chain models fit the distribution well!



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
---	--	---

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

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

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

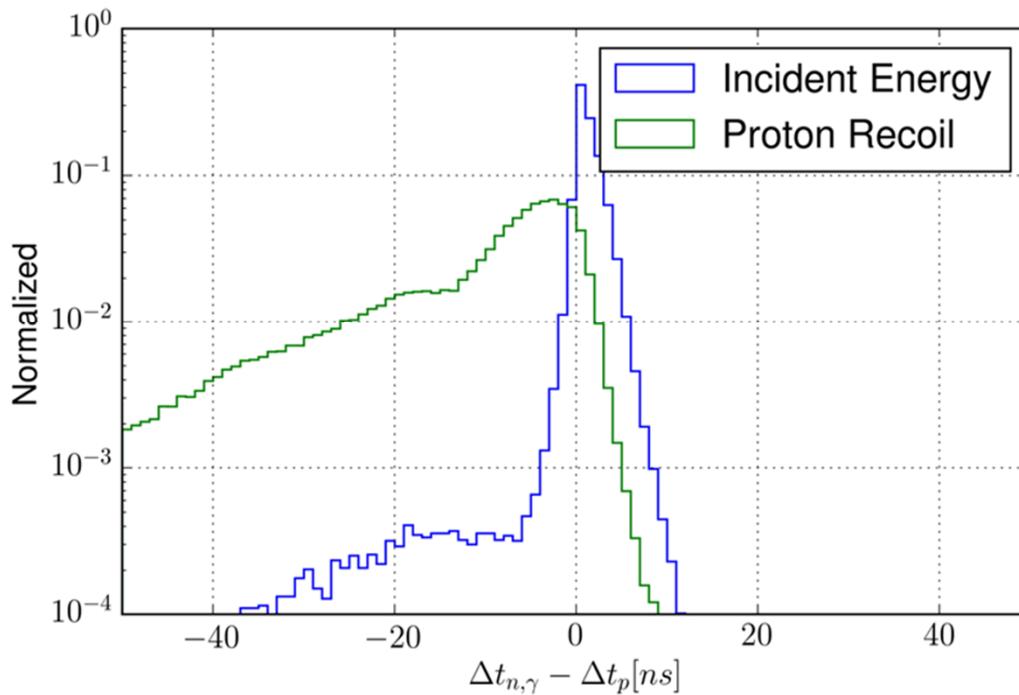


MINER: the Mobile Imager of Neutrons for Emergency Response

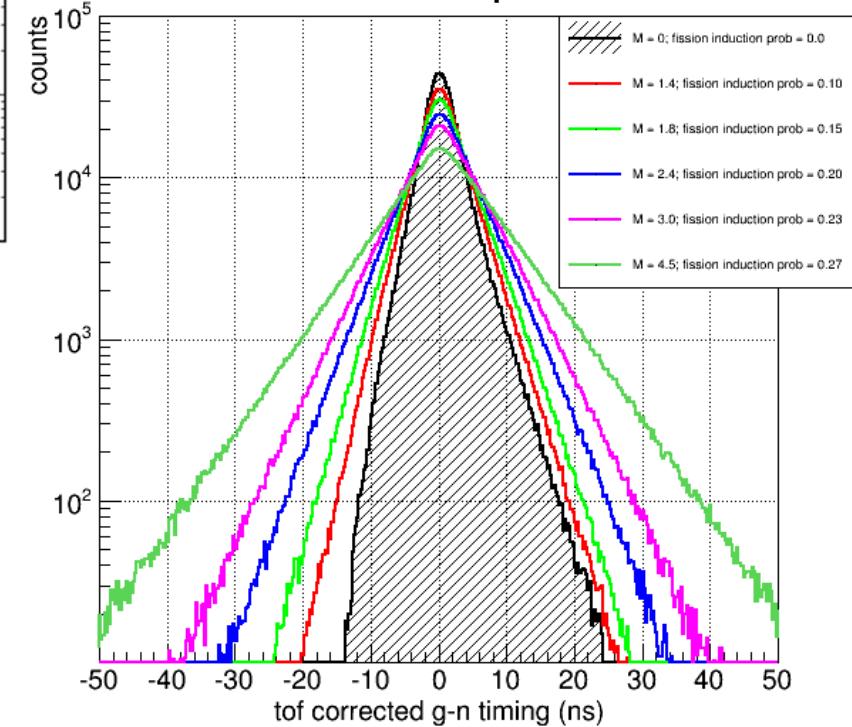
16 independent
3" x 3" EJ-309
liquid scintillator
cells

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

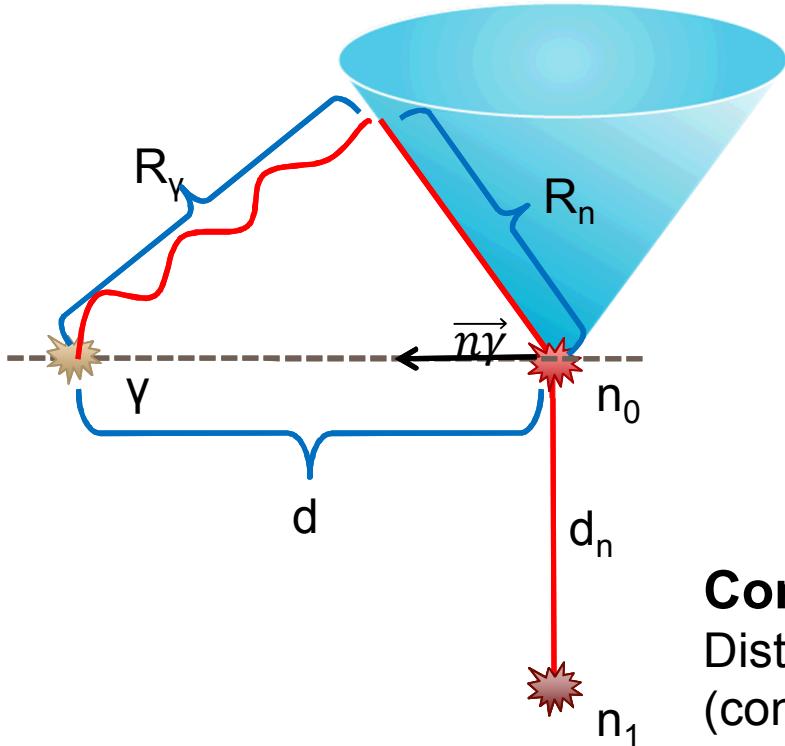
Cf-252 Simulation



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)

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

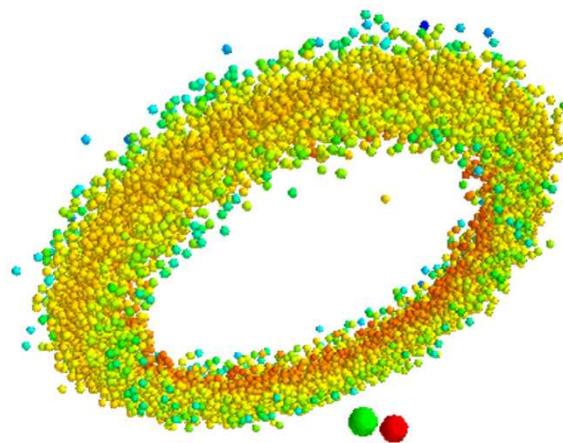
$$R_n = \frac{c^2 \Delta t v_n - d v_n^2 \mu + \sqrt{v_n^2 (c^2 (\Delta t^2 v_n^2 - 2 d v_n \mu \Delta t + d^2) + v_n^2 d^2 (\mu^2 - 1))}}{c^2 - v_n^2}$$

where

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

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

Single correlated event back-projection



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

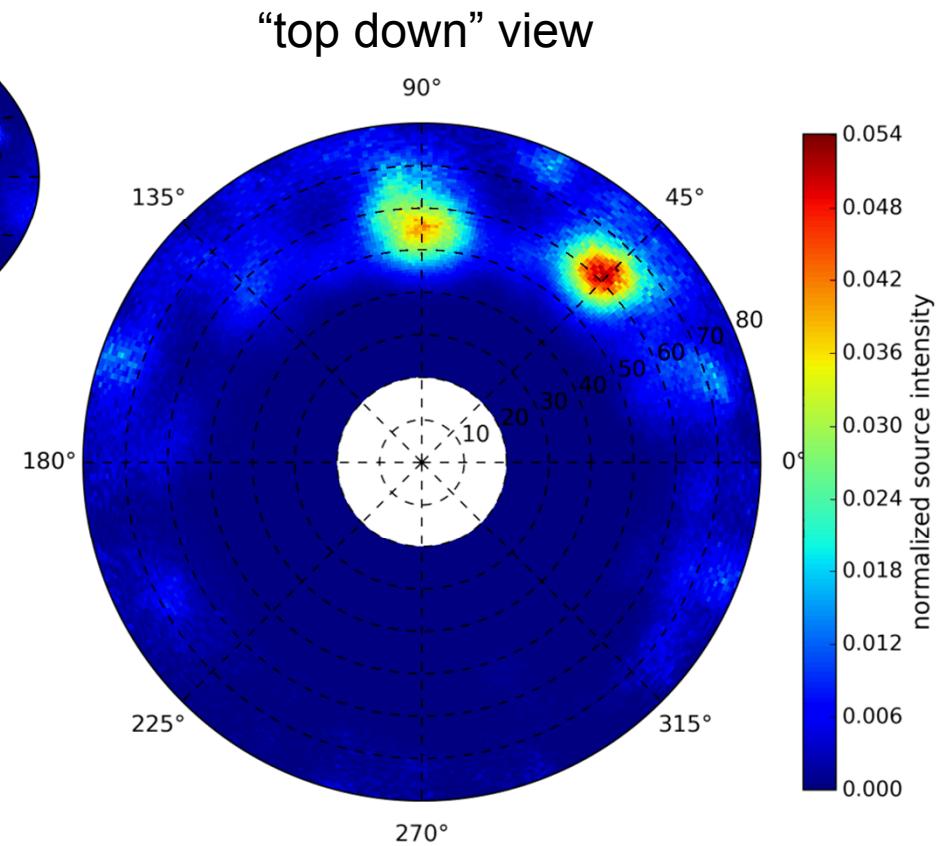
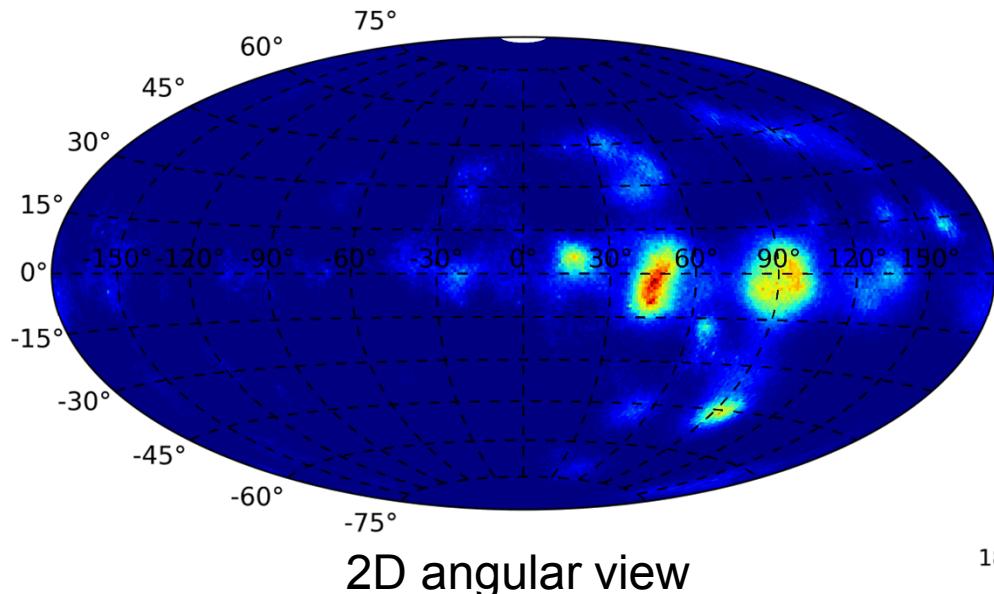
+

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

γ -n-n 3-D reconstruction

Very preliminary results

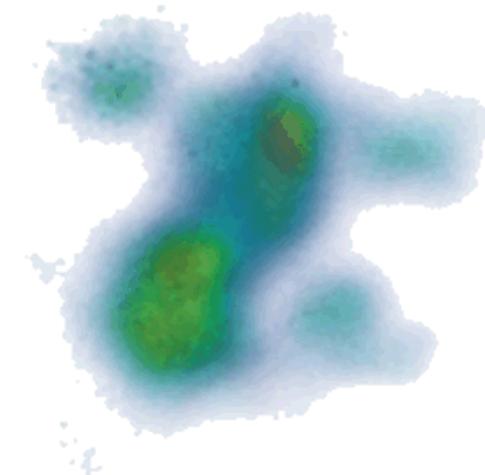
(Stochastic Origins Ensemble Reconstruction (SOE)),
Two Cf-252 fission sources, 50 & 60 cm from MINER



γ -n-n 3-D reconstruction



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



Preliminary

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

+

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

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

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.

Exceptional service in the national interest



CONFIRMATION using a Fast-neutron Imaging Detector with Anti-image NULL-positive Time Encoding (CONFIDANTE)

Peter Marleau



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Certification vs. Authentication: It's not just for hardware

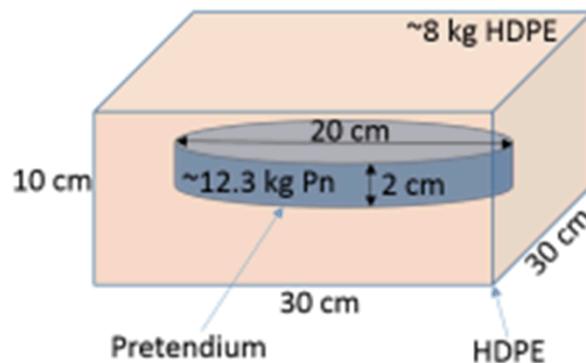


Certification – the process by which a host party gains confidence that sensitive information regarding an entity or facility remains secure.

Authentication - the process by which a monitoring party gains confidence that reported characteristics of an entity reflect the true state of that entity

Challenge problem

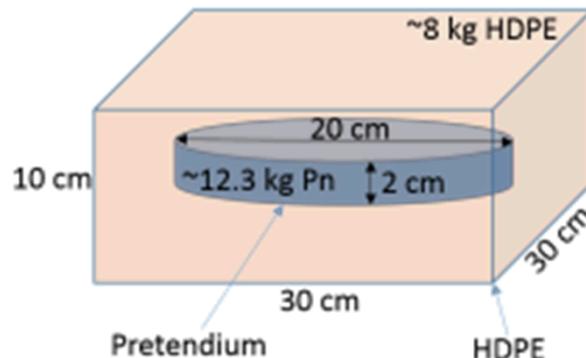
- The inspecting party has or had access to measure item T, which is known to be a valid type 1 treaty accountable item (TAI) through some other mechanism.



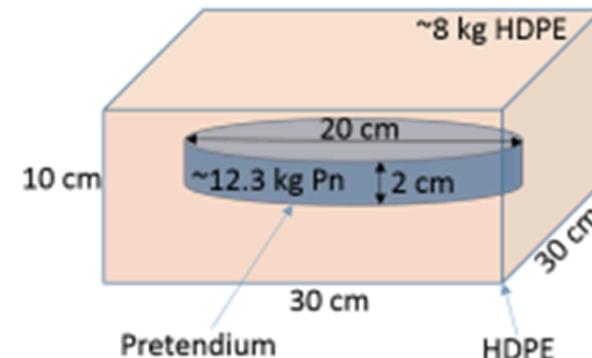
**Object T = valid
type 1 TAI**

Challenge problem

- The inspecting party has or had access to measure item T, which is known to be a valid type 1 TAI through some other mechanism.
- In the course of an inspection, the host presents item X and declares it as a type 1 TAI
- Item X should pass the verification measurement if it is a type 1 TAI, and fail if it is significantly different.



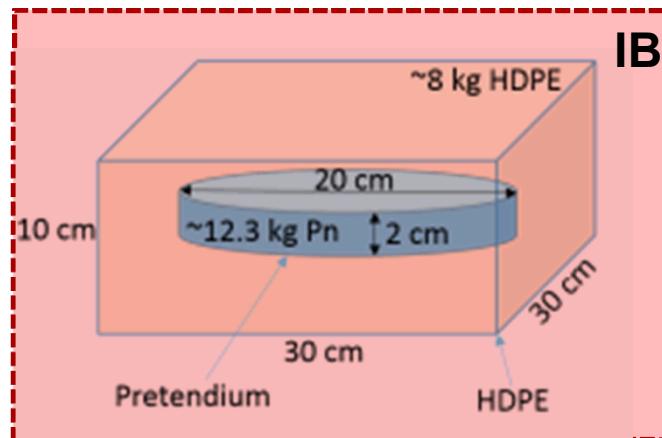
**Object T = valid
type 1 TAI**



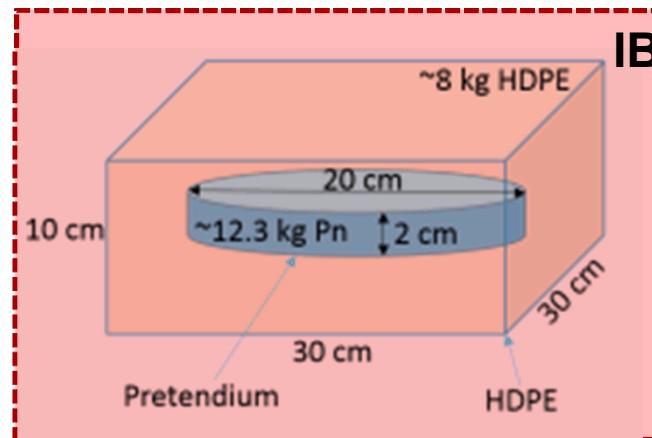
Object X = ?

Challenge problem

- The inspecting party has or had access to measure item T, which is known to be a valid type 1 TAI through some other mechanism.
- In the course of an inspection, the host presents item X and declares it as a type 1 TAI
- Item X should pass the verification measurement if it is a type 1 TAI, and fail if it is significantly different.
- The host must be confident that the inspector has not learned the diameter d of the pretendium in item X, or any type 1 TAI



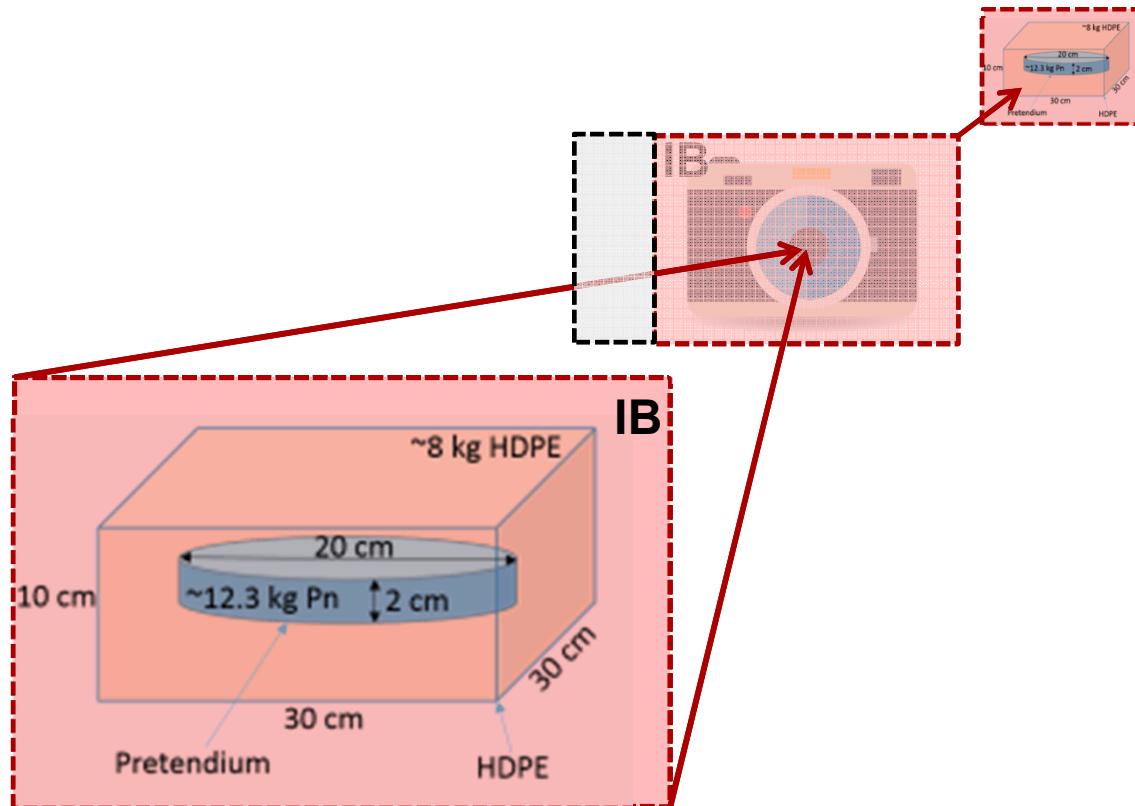
**Object T = valid
type 1 TAI**



Object X = ?

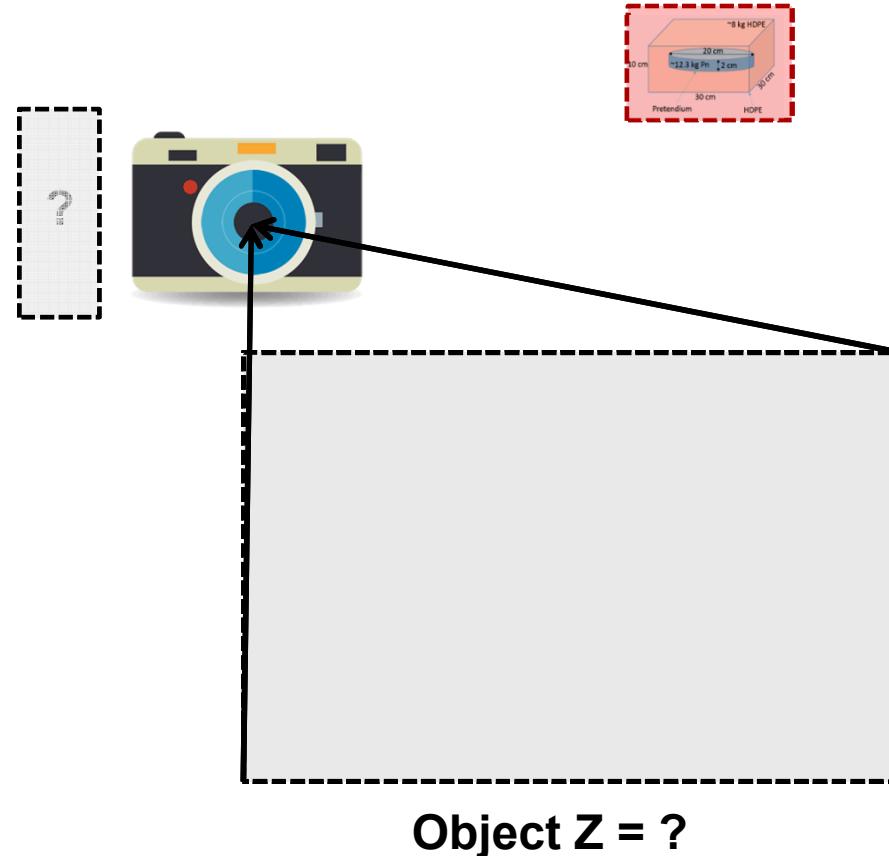
Templates - generation

- We could generate a template behind an information barrier (IB) ...



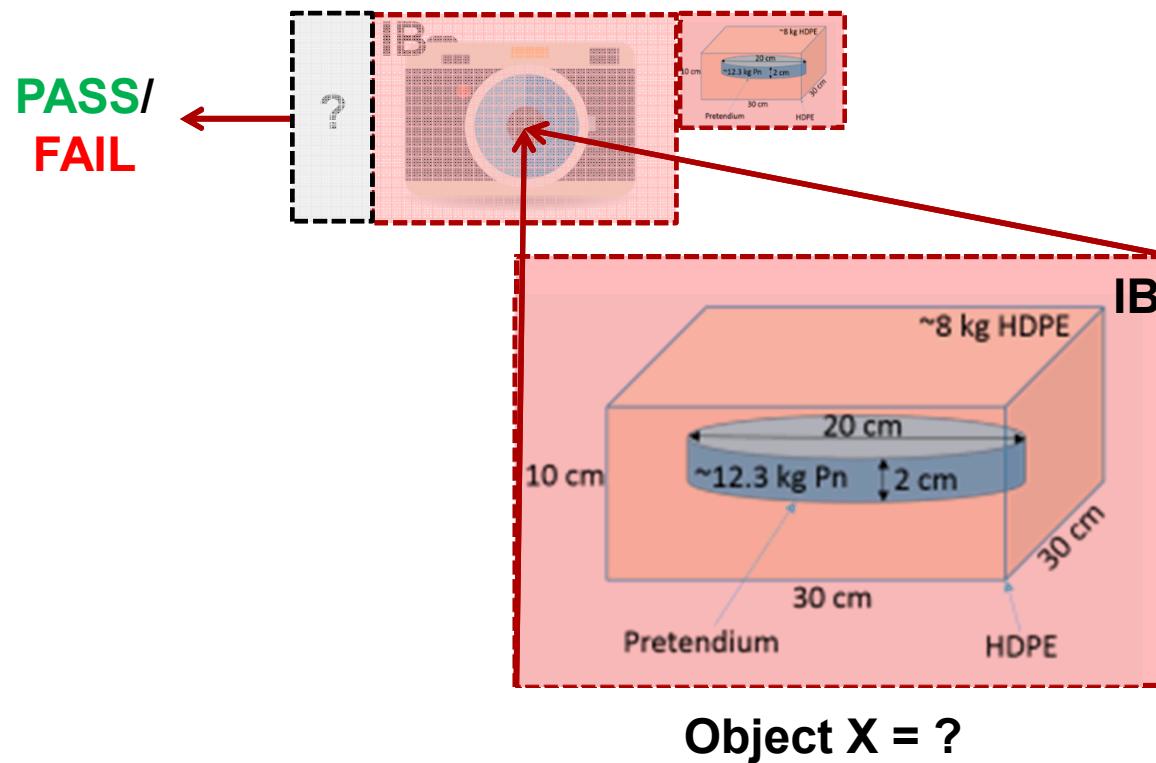
Templates - authentication

- Authenticate equipment ...



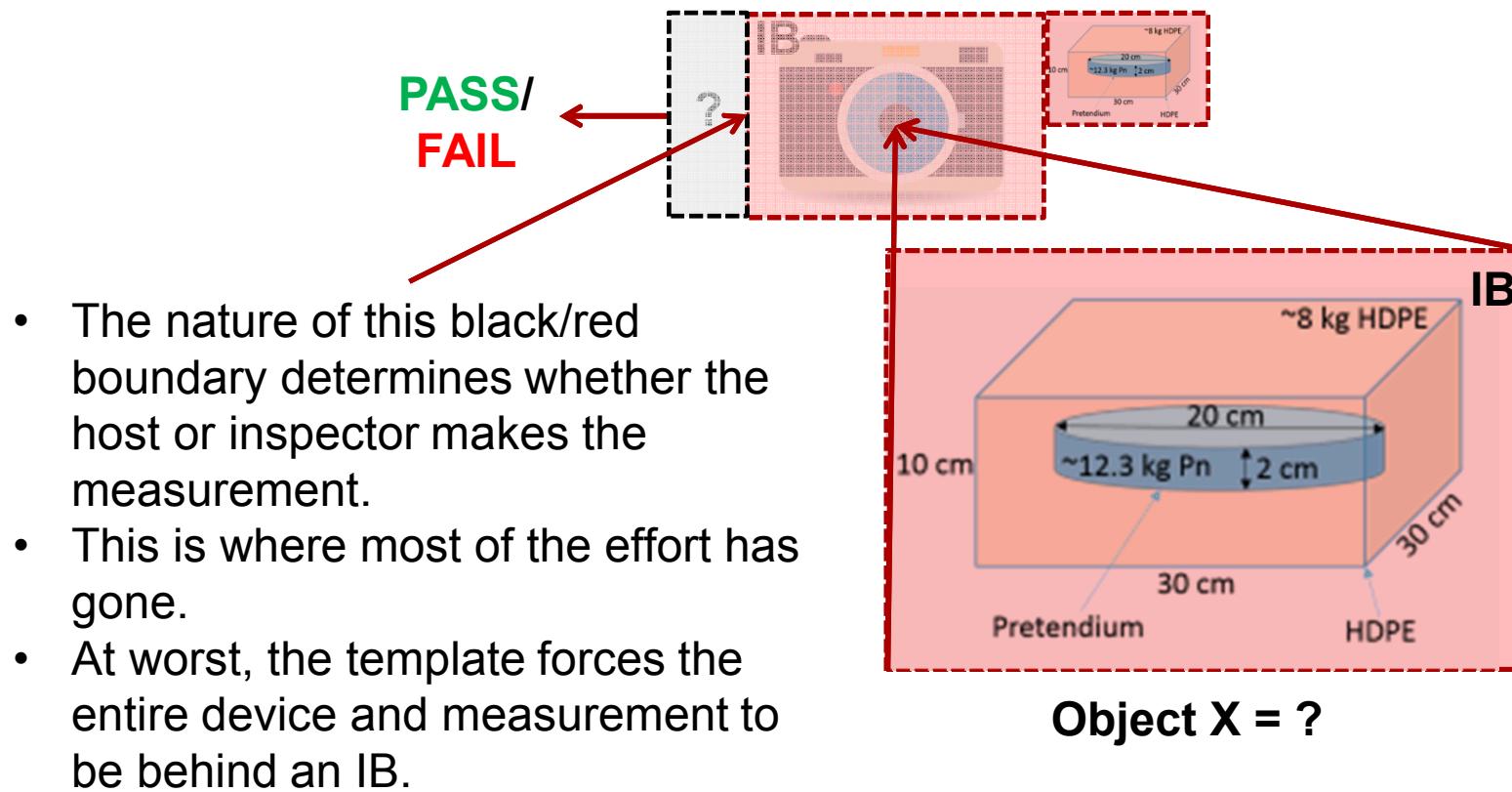
Templates - comparison

- Make comparison measurement...



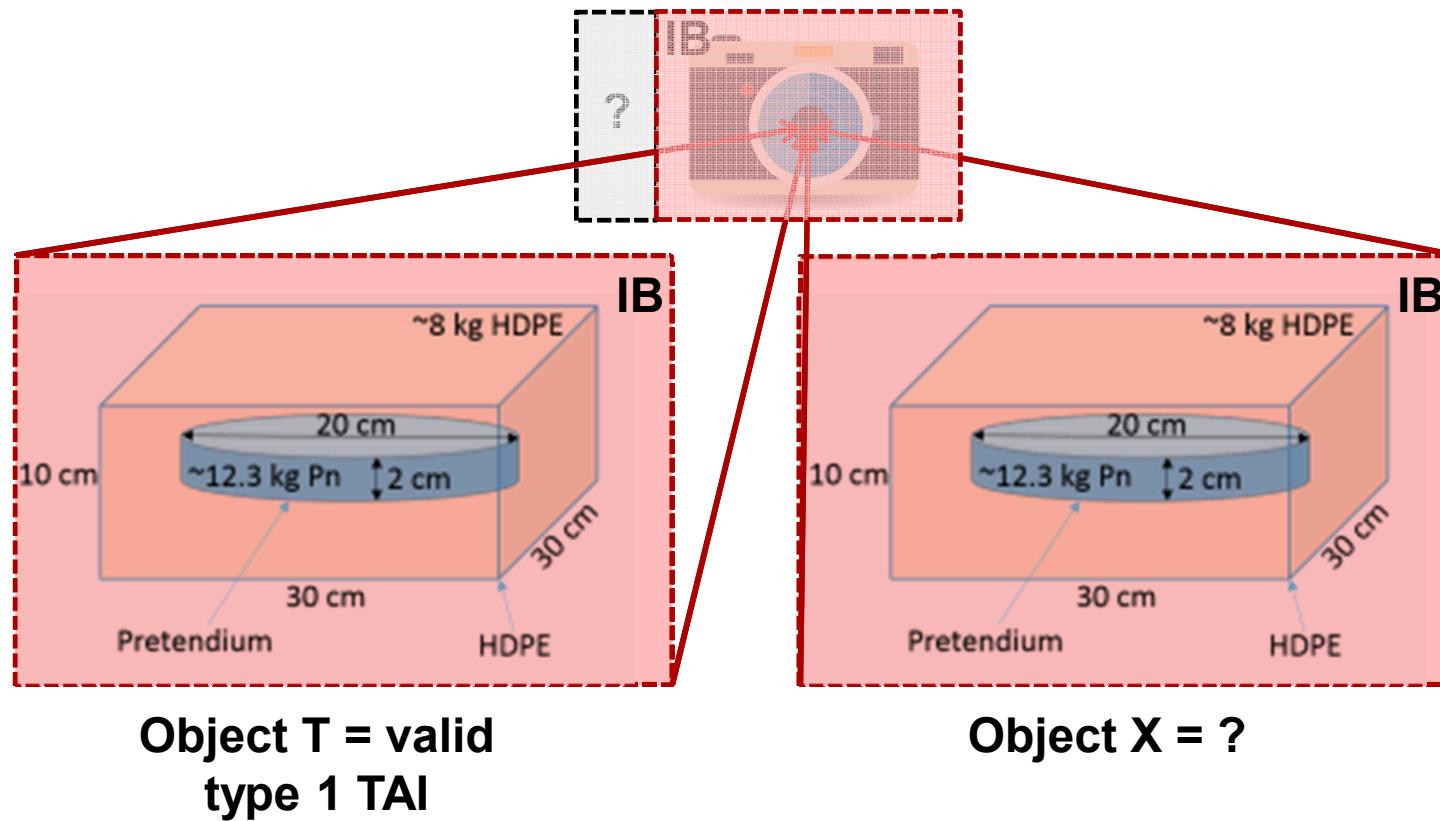
Templates – who measures?

- Who makes the measurement? Is the measurement itself authenticatable?



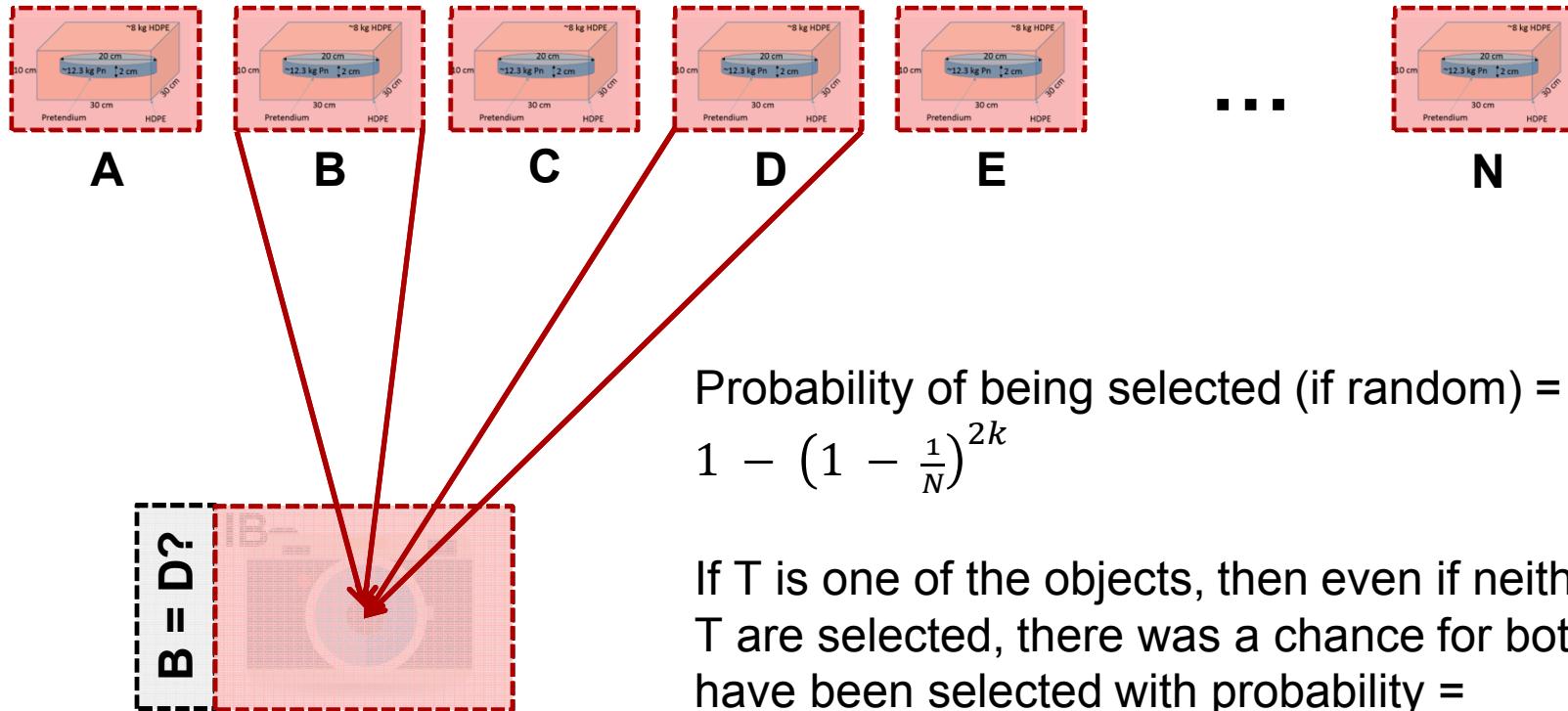
Proposal – comparison measurements

- Can we compare two objects directly without generating a template?
- If one object is T, then X is confirmed as a type 1 TAI.
- If neither object is T, then they are confirmed to be identical, but not T.
- If multiple object comparisons are confirmed and even one is T, then all objects are confirmed as type 1 TAIs.



Proposal – CONOPS and Inspector choice

- Presented with N objects and k comparison measurements will be made.



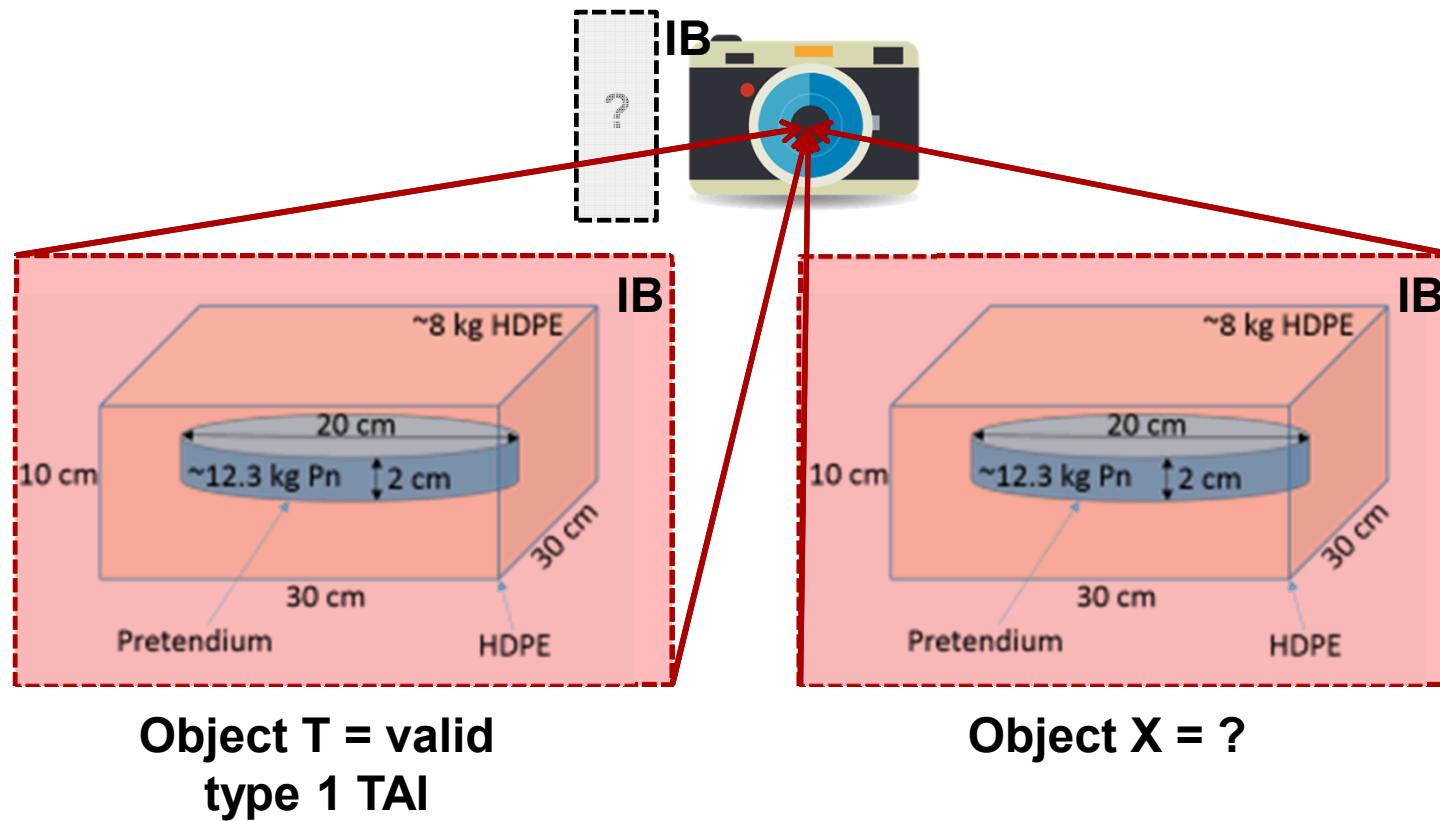
If T is one of the objects, then even if neither X nor T are selected, there was a chance for both to have been selected with probability =

$$\left(1 - (1 - \frac{1}{N})^{2k}\right)^2$$

providing some degree of confidence

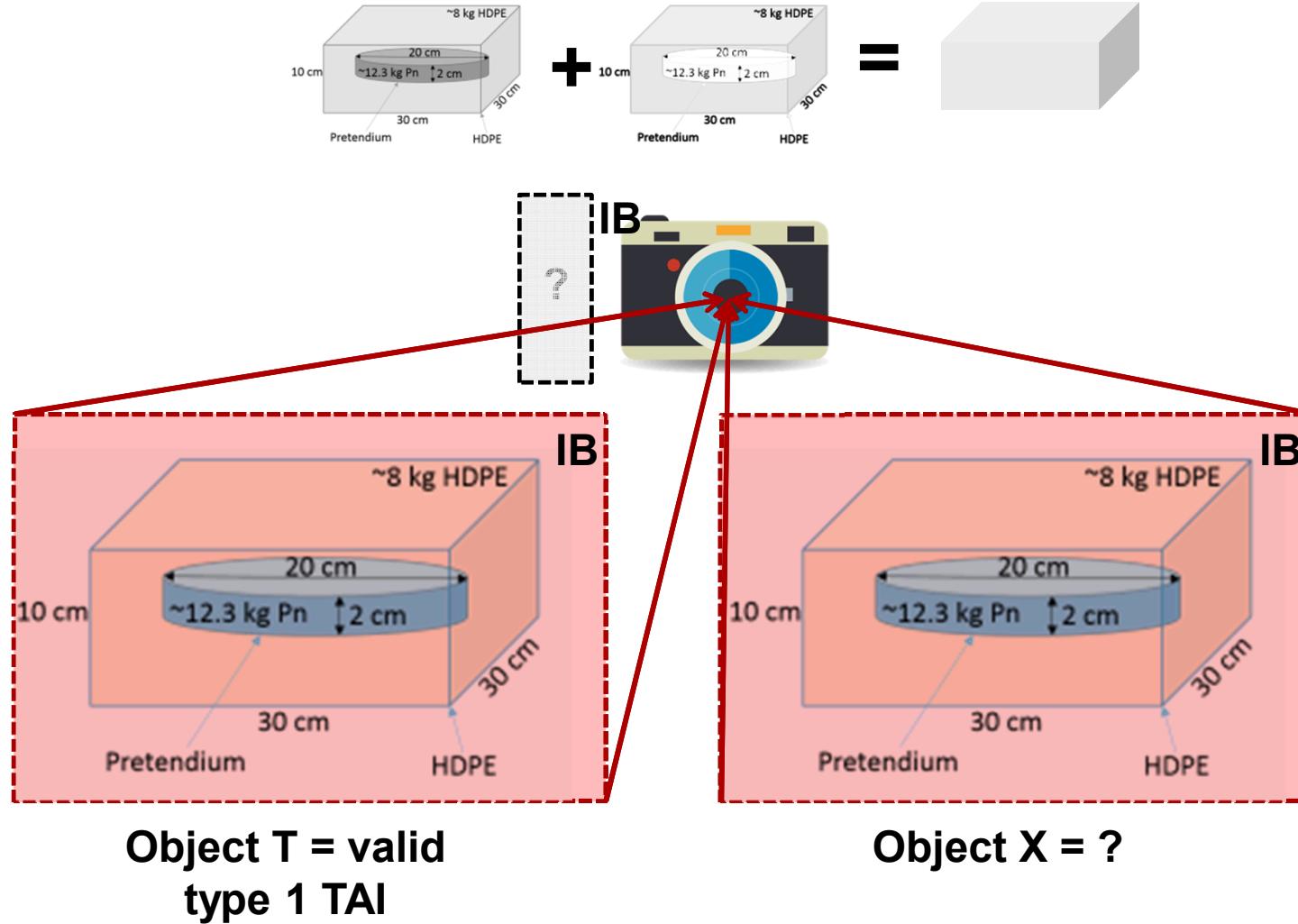
Proposal – comparison measurements

- Is there a physical implementation of the confirmation measurement that the inspector can watch and authenticate?
- **It would be great if we could get a physical NULL as an indication of positive confirmation at all times, even during the measurement.**



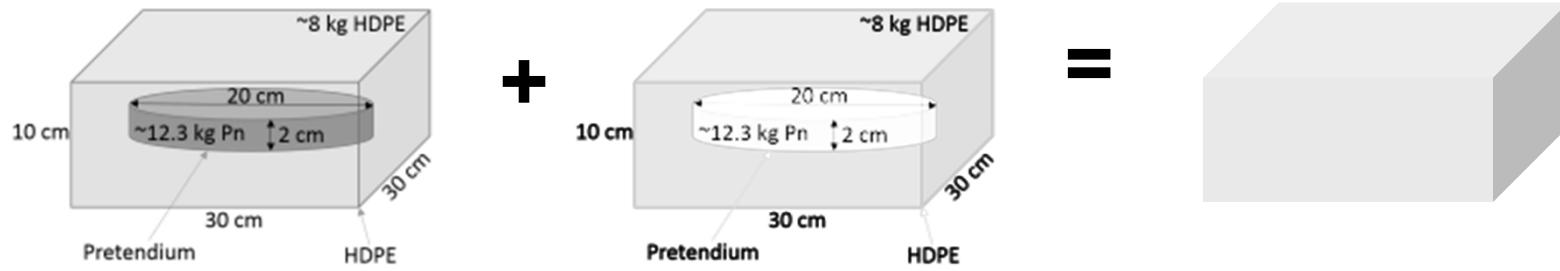
Proposal – complementary comparison

- What we need is to turn one image into the complement of the other ***at all times***.

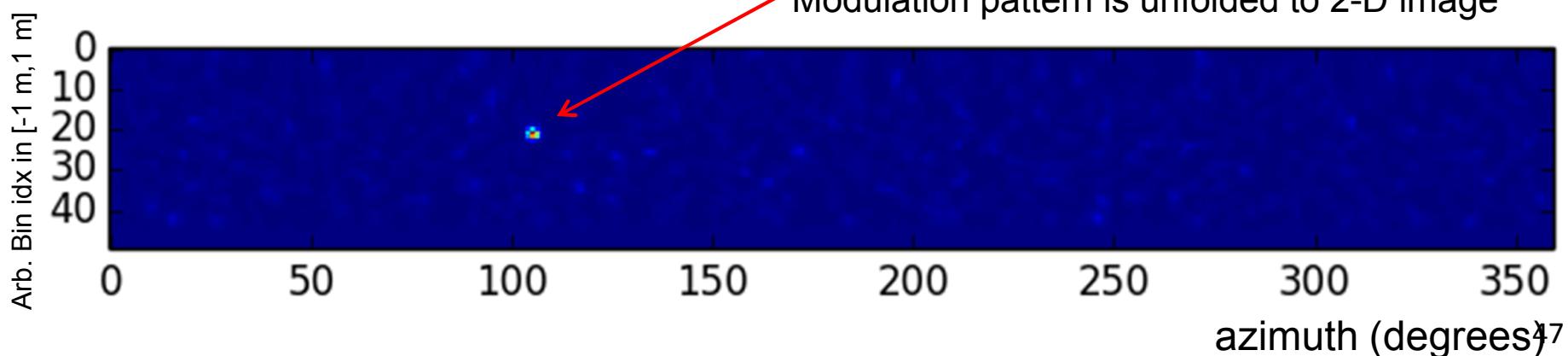
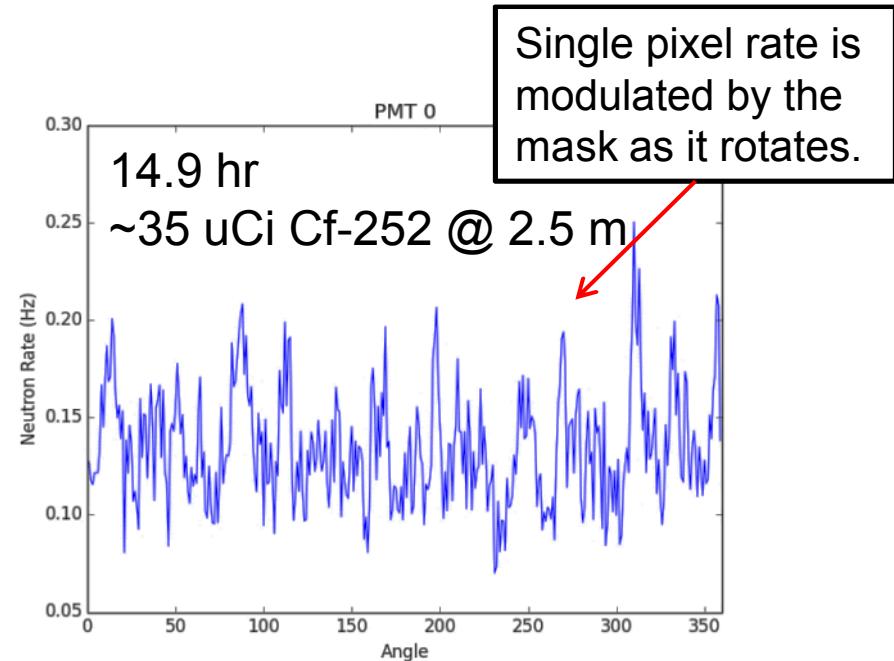
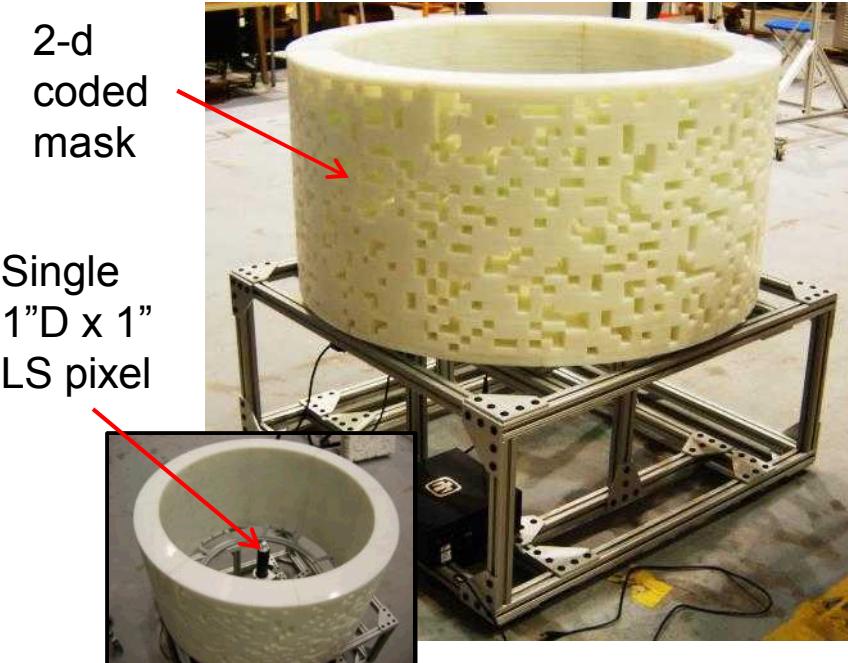


Ok, but how? ...

- One image is the complement of the other *at all times*.



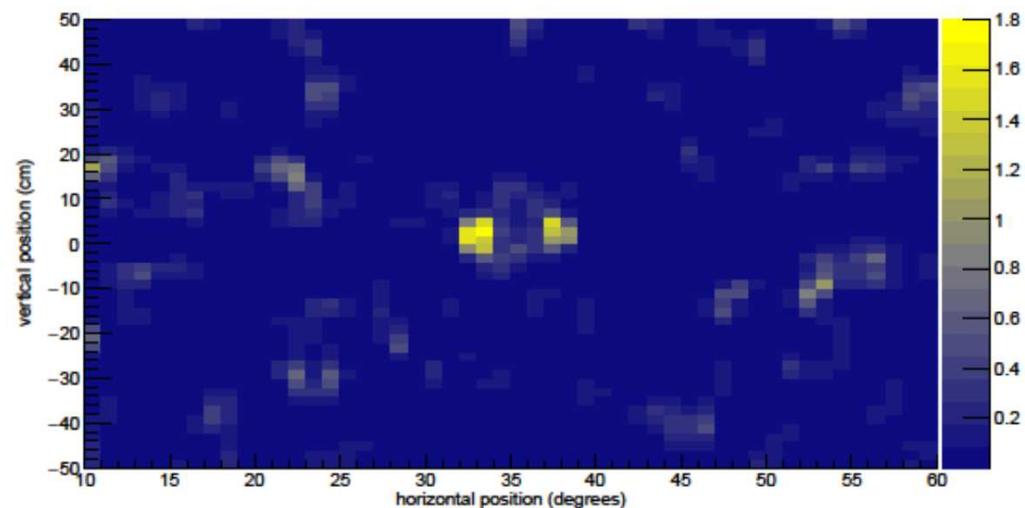
2D Time-encoded Imaging (TEI)



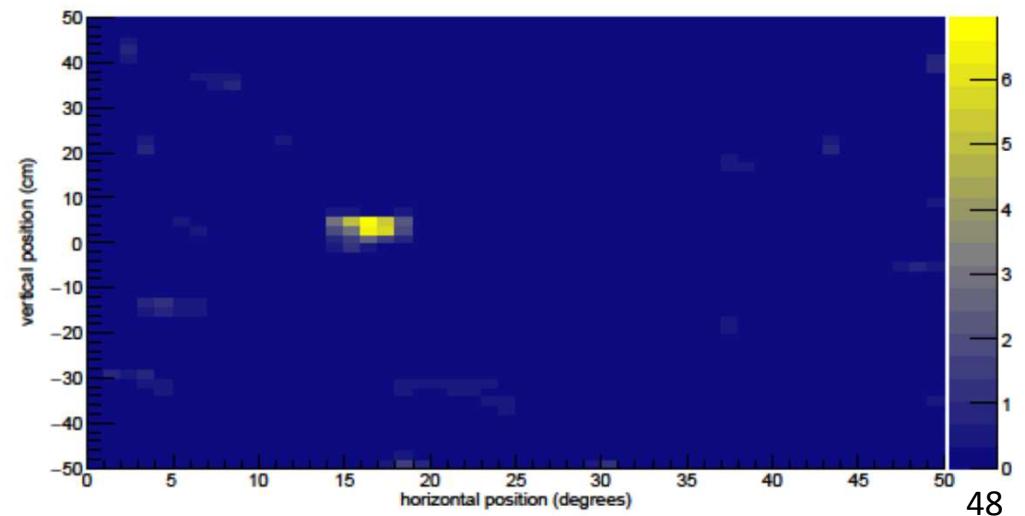
TEI-2D imaging – two point sources

Two 1.4×10^5 n/s ^{252}Cf point sources at 2.0 meters stand-off.

5 degree separation in 1 hour
(50 mlem iterations)



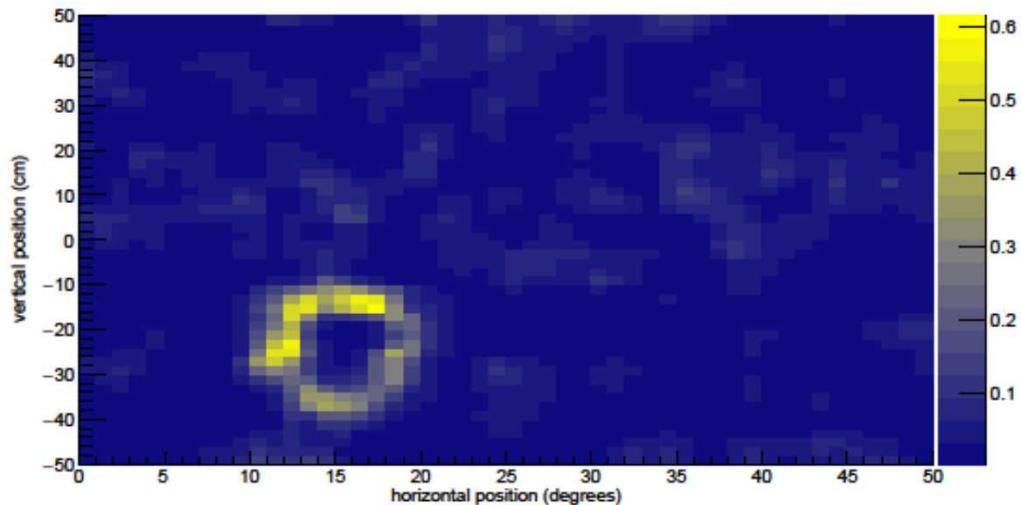
2 degree separation in 24 hours
(250 mlem iterations)



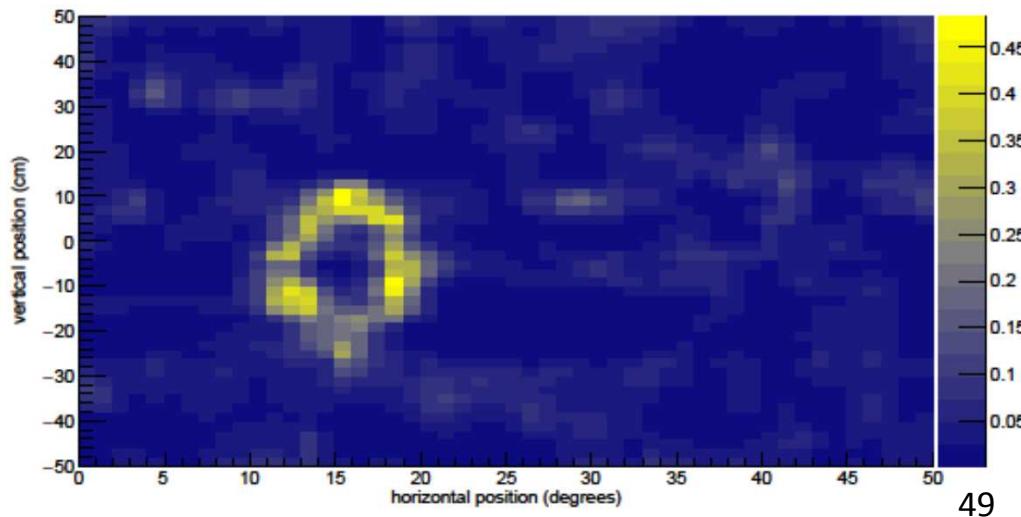
TEI-2D imaging – extended sources

A single 1.4×10^5 n/s ^{252}Cf source move through an extended pattern at 2 m.

72 hours
(100 mlem iterations)

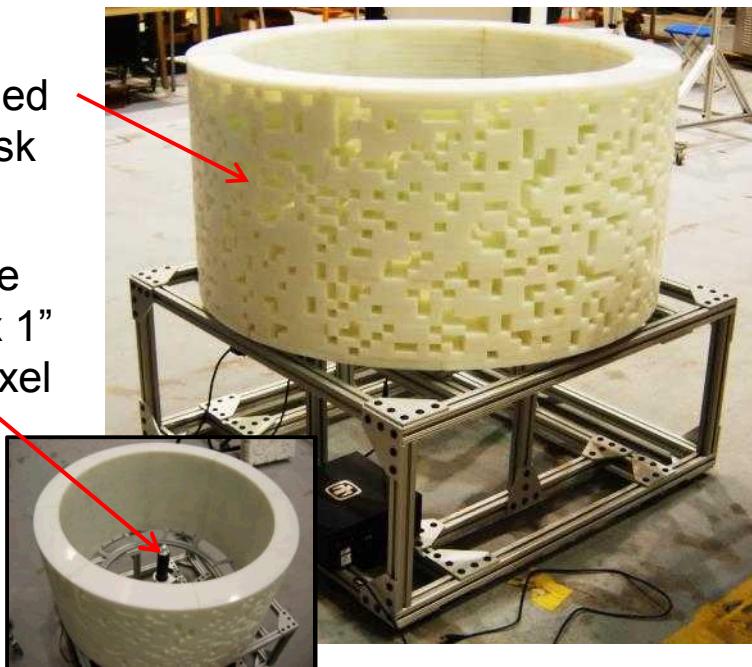


94 hours
(100 mlem iterations)



2D TEI – confirmation measurements?

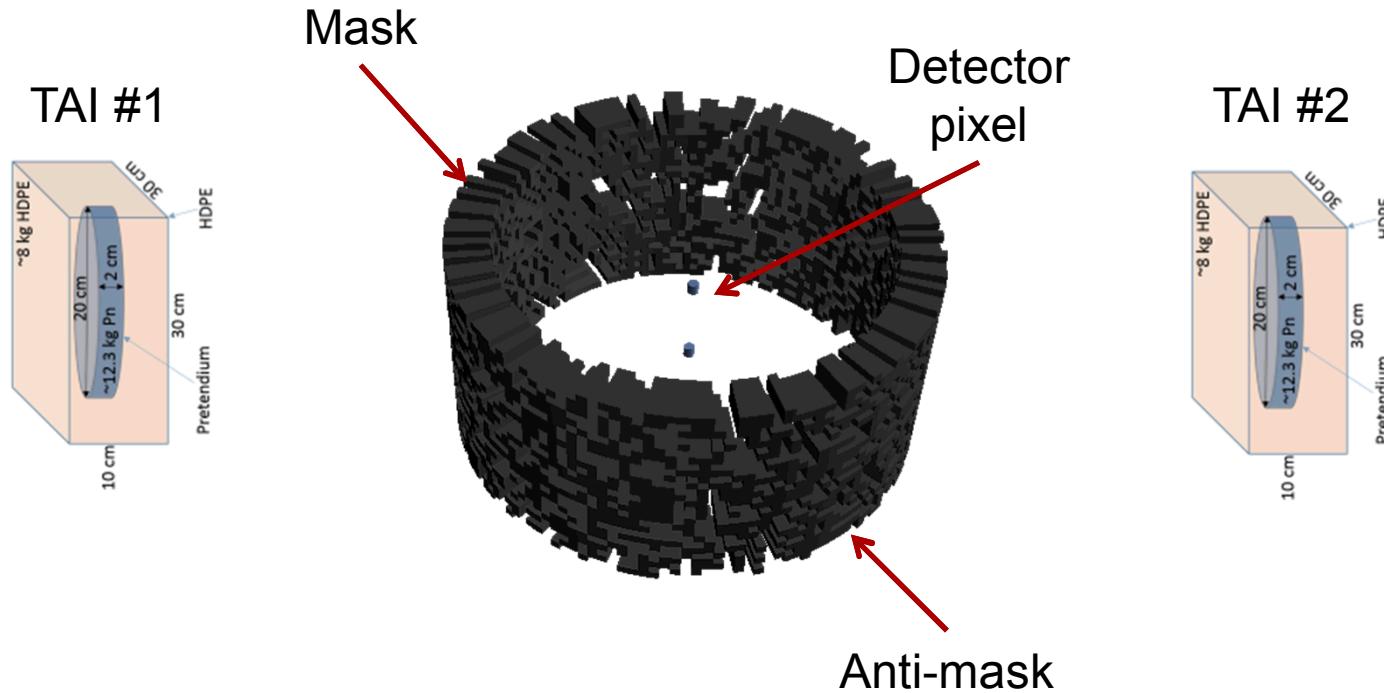
2-d
coded
mask



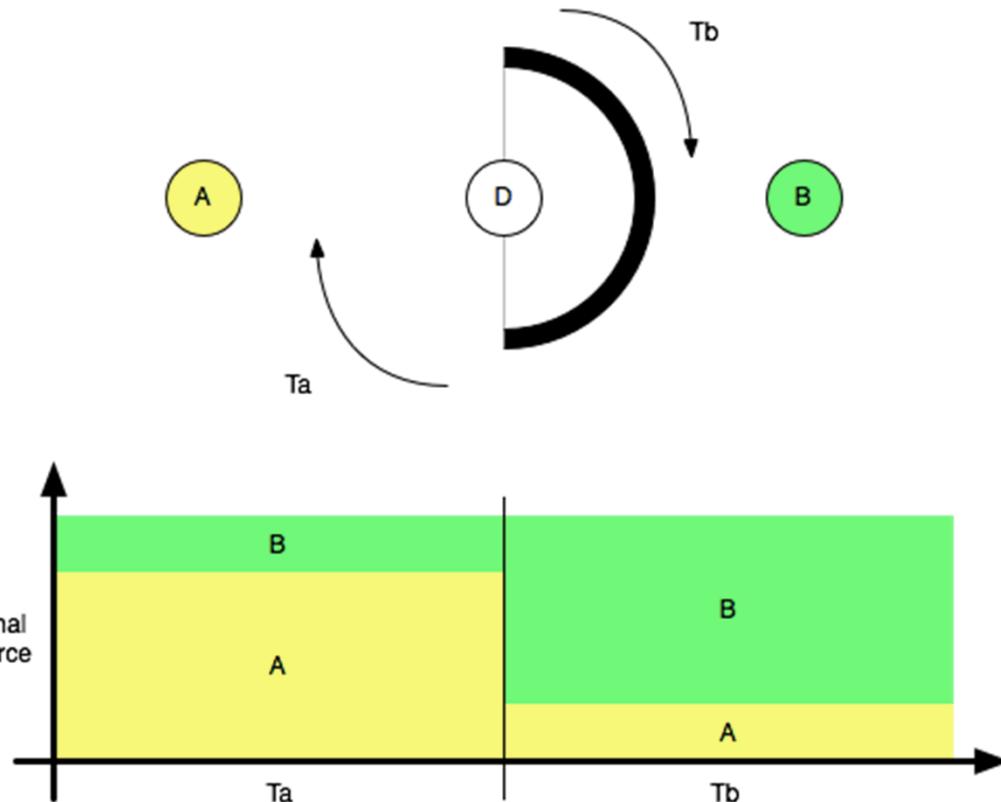
- **TEI is simple**
 1. Only one instrumented channel.
 2. Minimal calibration issues
 - a) Information encoded in the relative rate of a single detector.
 - b) Absolute gain doesn't matter.
 - c) Gain can drift over time.
 3. Potential real-time analysis
 - a) Single data stream.
 - b) Events can be processed one at a time and update a test statistic.
- **Can we design a TEI confirmation system such that the detection rates can be monitored by an inspector without putting sensitive information at risk?**

Here's where the magic happens ...

If the mask is designed such that one side is the anti-mask of the other,
then **TAI #2 projects the anti-image of TAI #1 at all times**
if and only if they are identical!

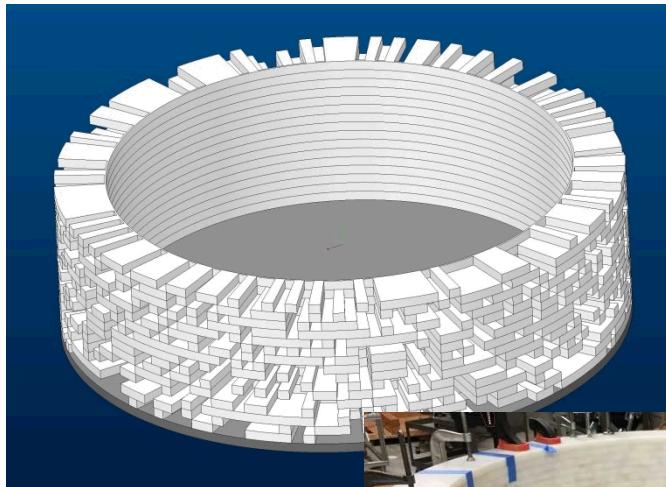


A very simple example



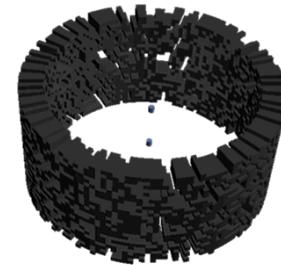
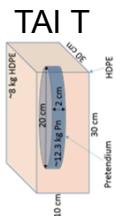
- For example, take a very simple mask: half mask, half aperture.
- The fraction of total count rate coming from A and B is unknown at any given angle.
- In this example, the location (and shape) of the boundary between regions is not revealed.

We're working on it

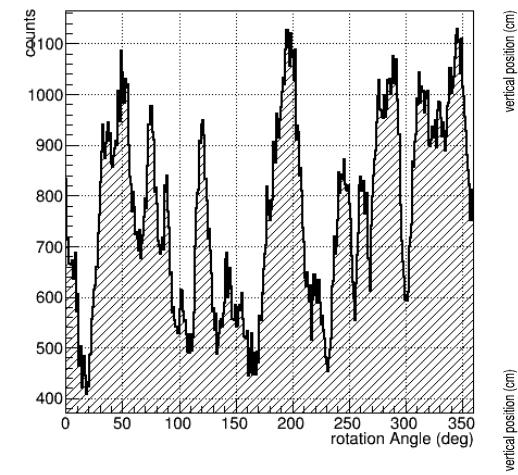


Modeling results - Single type 1 TAI (5e5 counts)

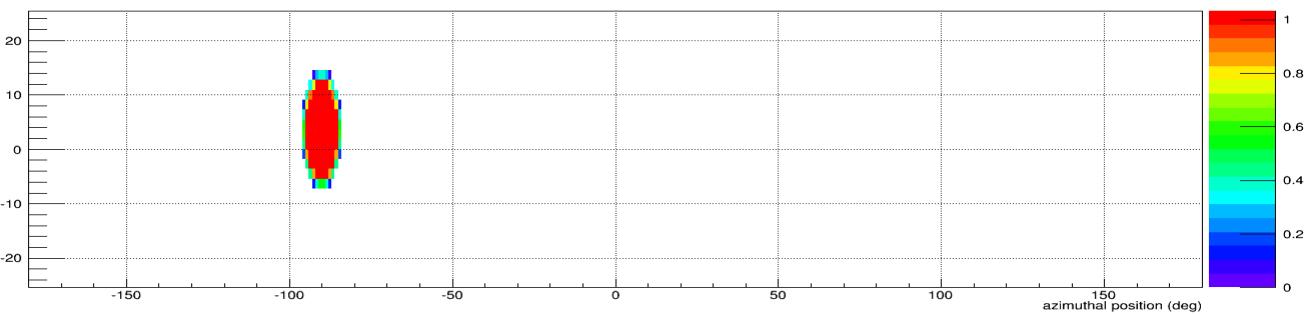
The system still functions as a standard TEI imager for any single object.



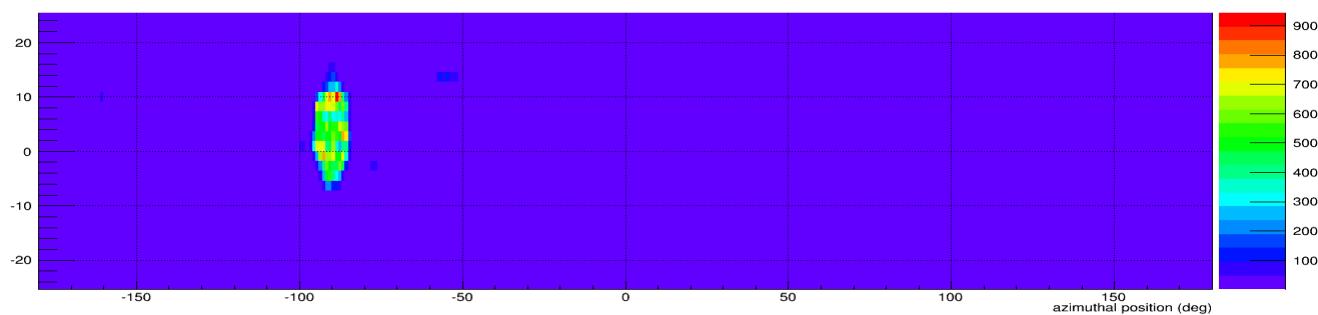
Pixel Counts



Iso-Background plus Source

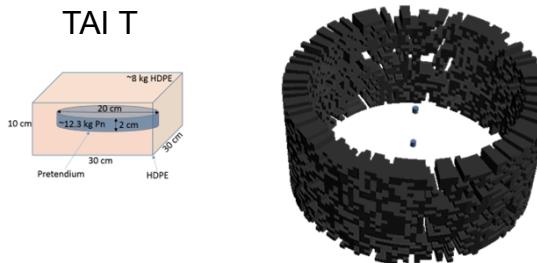


Reconstructed Image (MLEM)

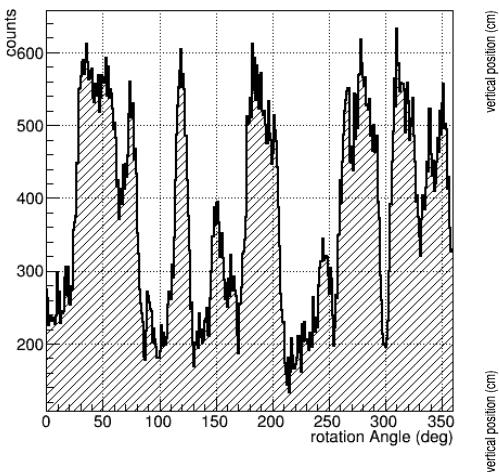


Modeling results - Single type 1 TAI (2.5e5 counts)

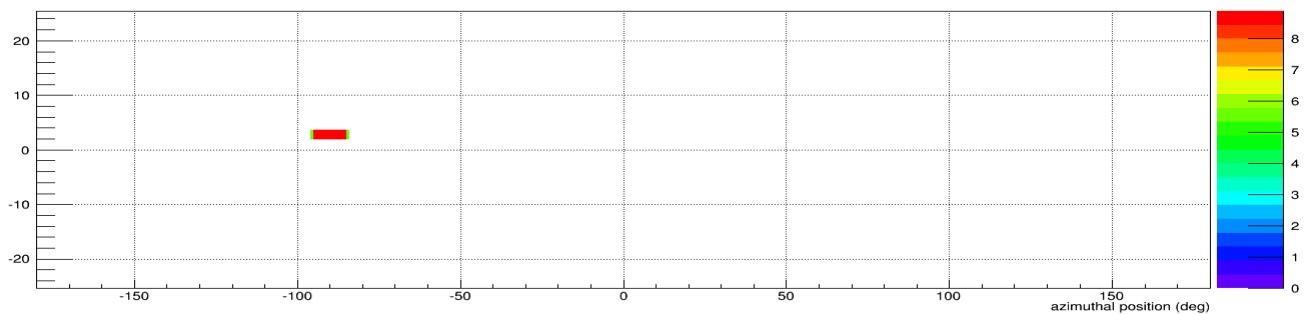
This orientation provides better discrimination between objects T and F.



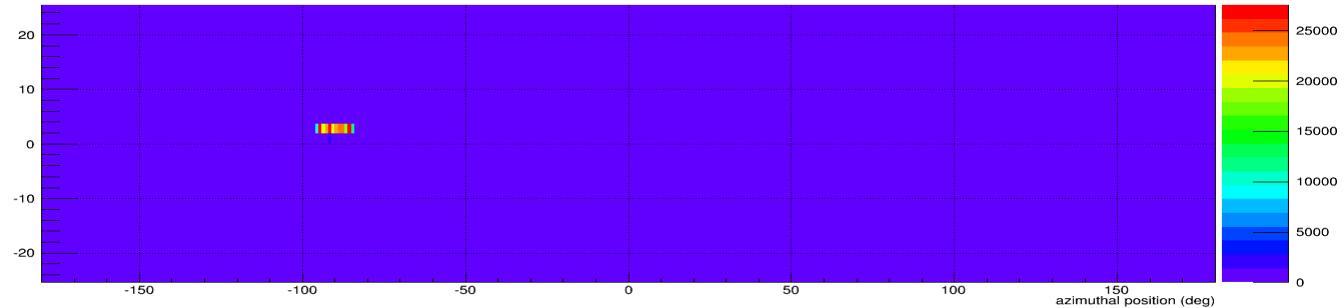
Pixel Counts



Iso-Background plus Source



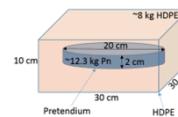
Reconstructed Image (MLEM)



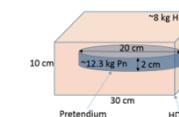
Modeling results – T vs. X (5e5 counts)

True “null”-positive
confirmation comparison
measurement between
two type 1 TAIs.

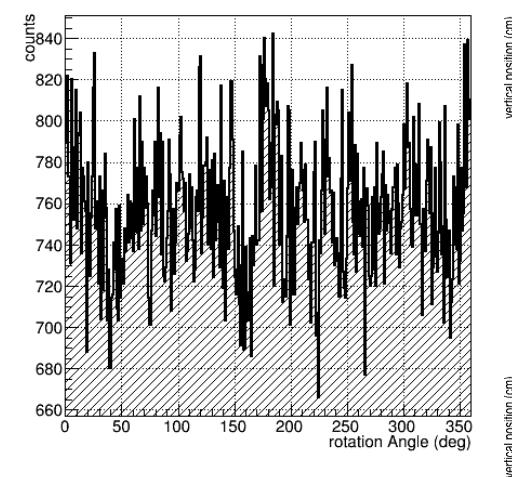
TAI T



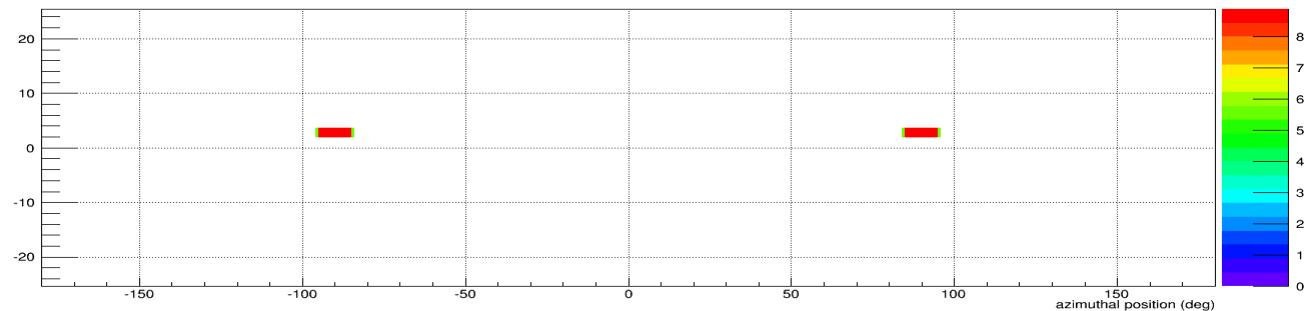
TAI X



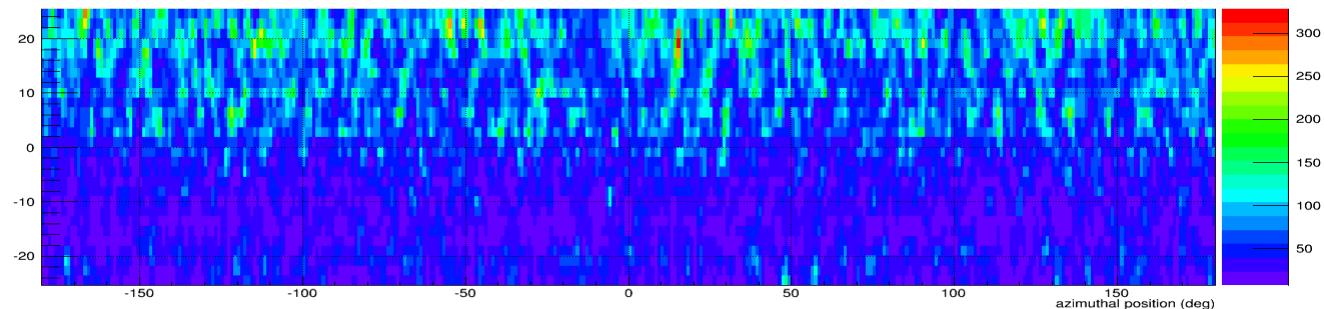
Pixel Counts



Iso-Background plus Source



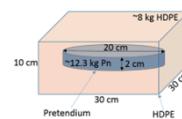
Reconstructed Image (MLEM)



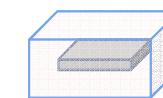
Modeling results – T vs. F (5e5 counts)

True non-null-negative
confirmation comparison
measurement between
objects T and F.

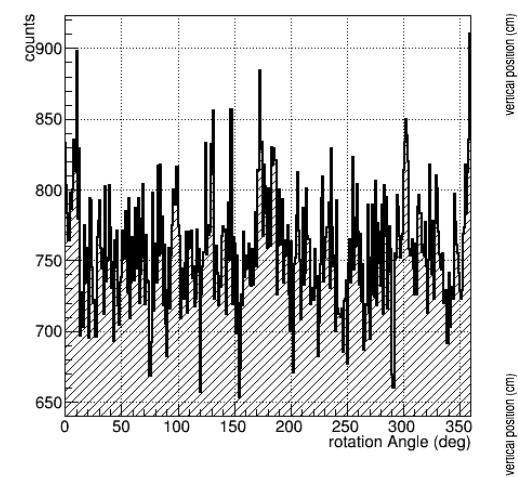
TAI T



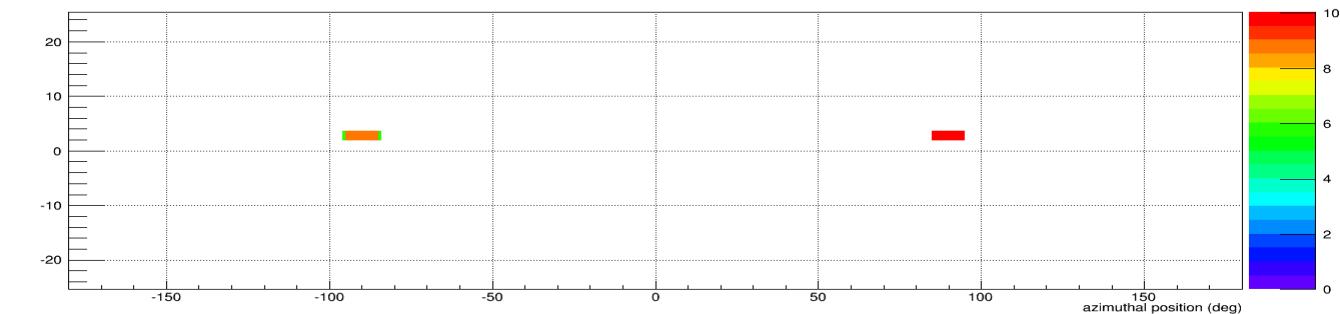
TAI F



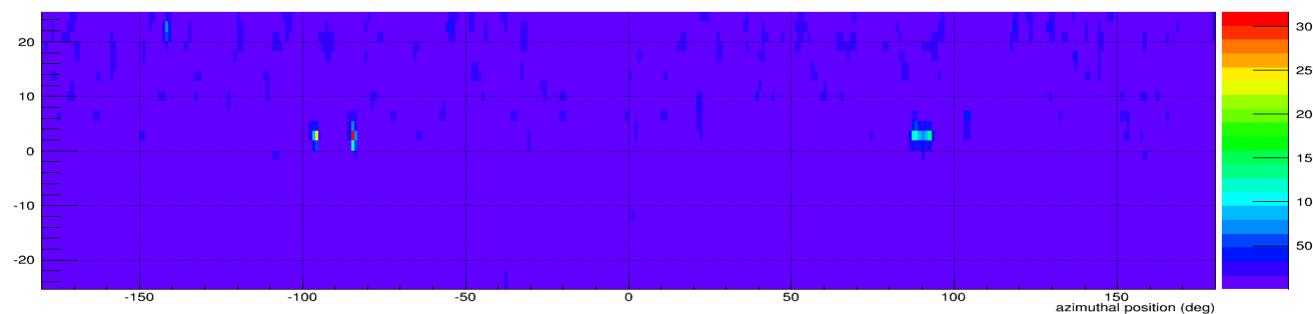
Pixel Counts



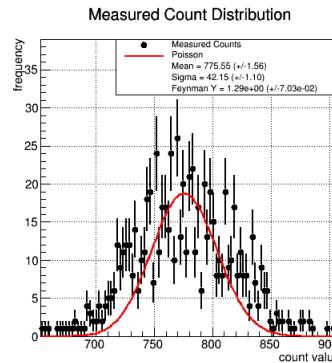
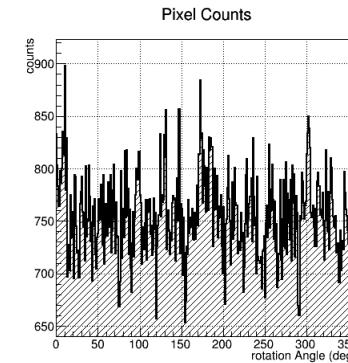
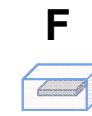
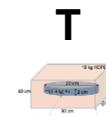
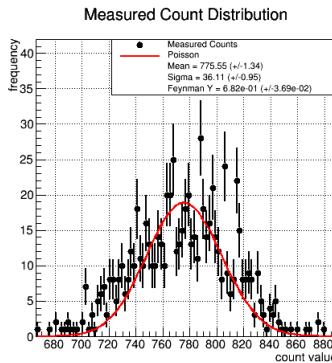
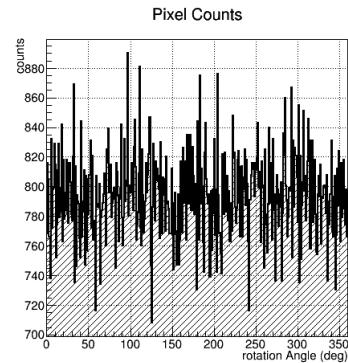
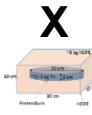
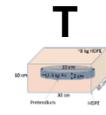
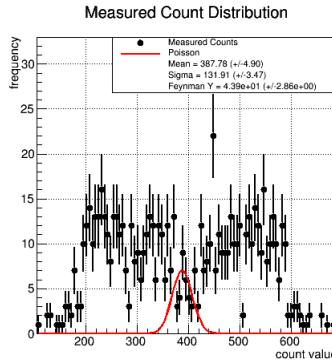
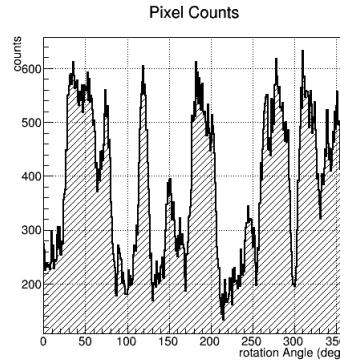
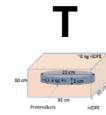
Iso-Background plus Source



Reconstructed Image (MLEM)



Single Test Statistic – Feynman Y



$$\text{Feynman Y} = \left(\frac{\text{variance}}{\text{mean}} - 1 \right)$$

= 86.8 (+/- 5.7) →
Far from Poisson

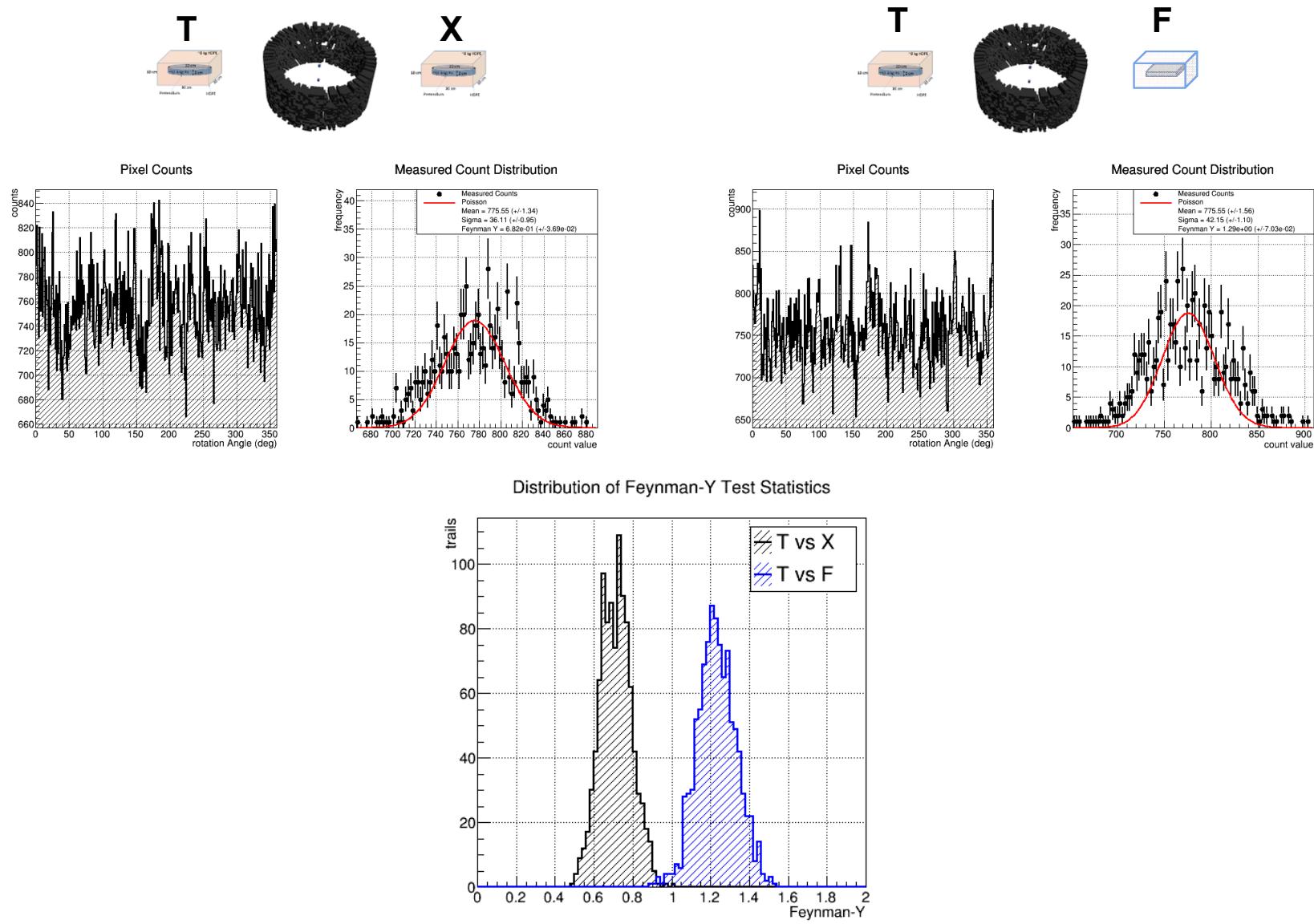
$$\text{Feynman Y} = \left(\frac{\text{variance}}{\text{mean}} - 1 \right)$$

= 0.68 (+/- 0.04) →
Fairly Poisson

$$\text{Feynman Y} = \left(\frac{\text{variance}}{\text{mean}} - 1 \right)$$

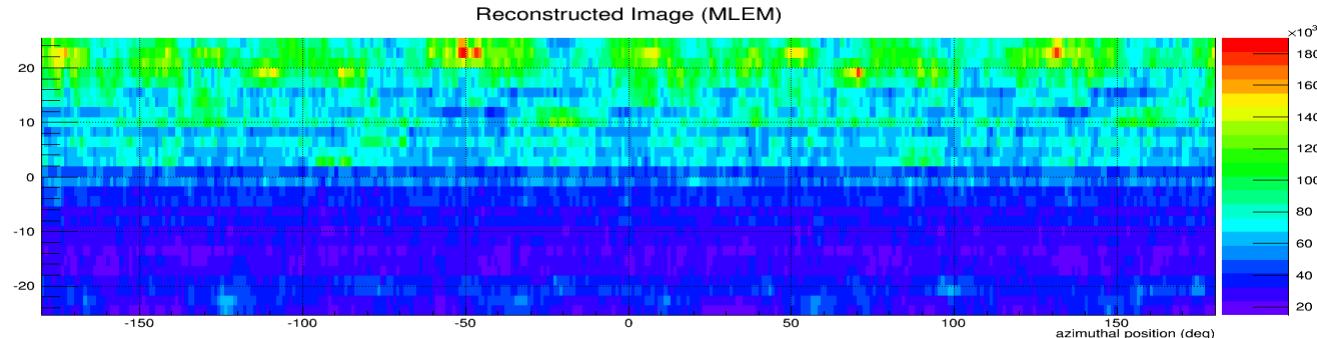
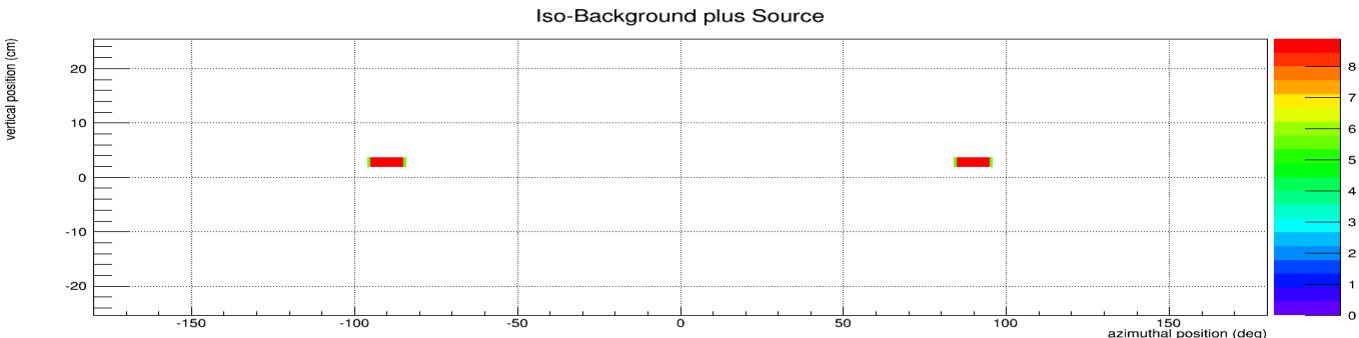
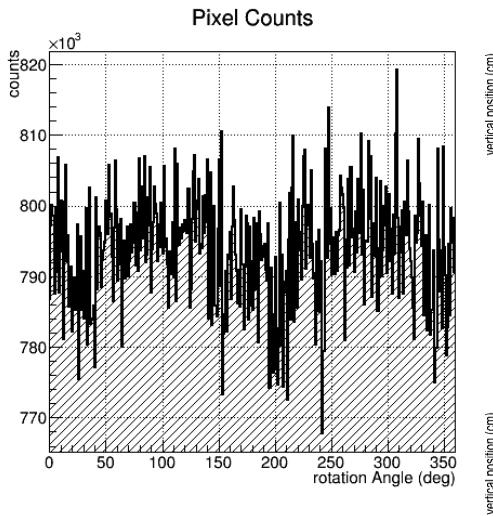
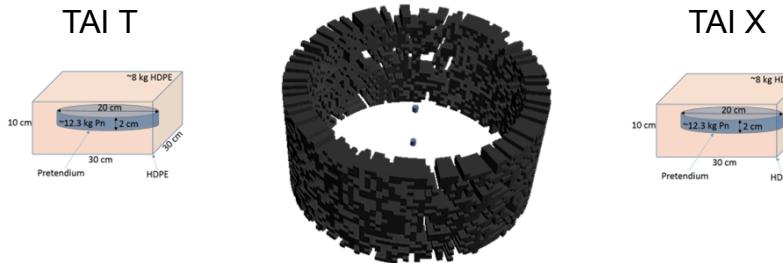
= 1.3 (+/- 0.07)
→ Less Poisson

Feynman Y Test Statistic – 1000 trials of 5e5 counts



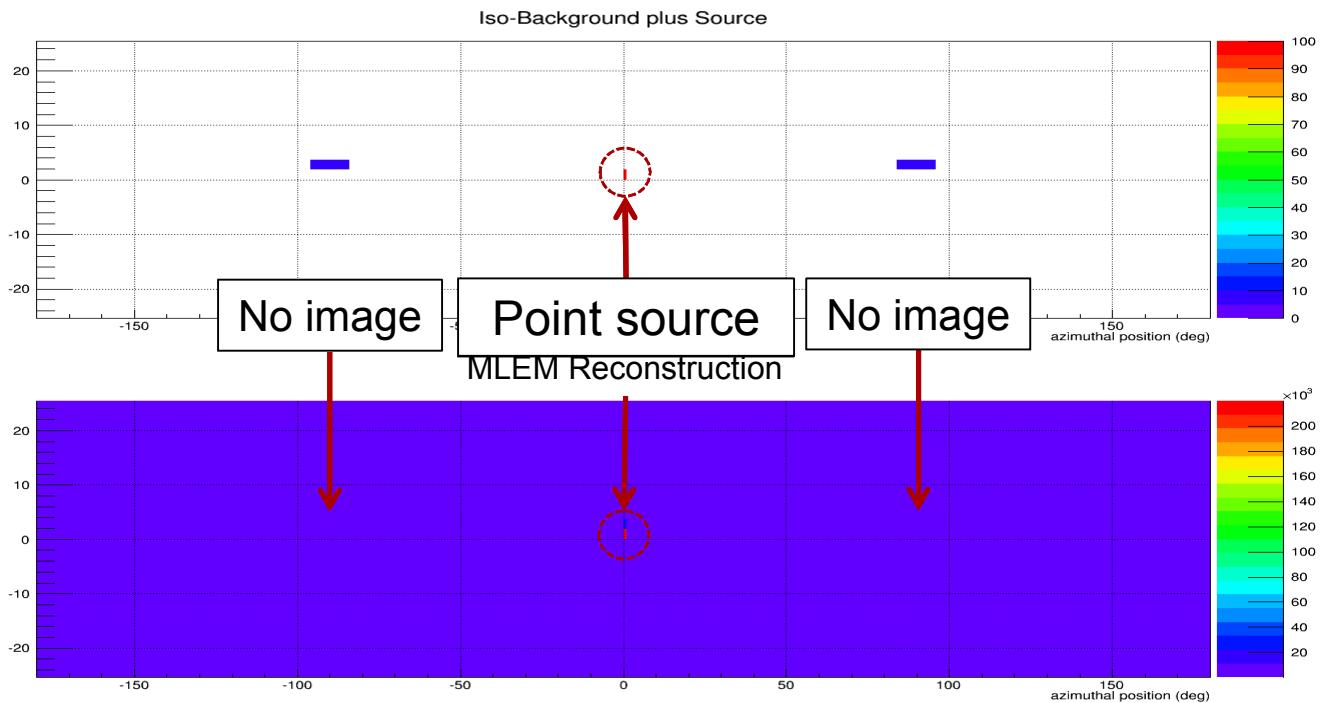
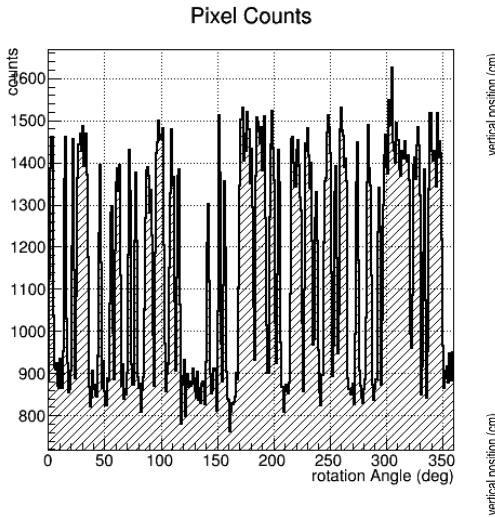
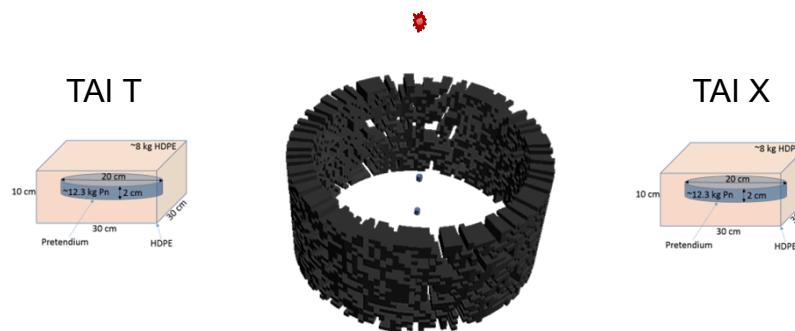
Modeling results – T vs. X (1000 trials of 5e5 counts)

Even after summing 1000 trials worth of data, there isn't much evidence that sensitive information is present. **This must be made more rigorous.**



Modeling results – T vs. X plus point source (8e5 counts)

If (and only if) the TAIs are identical, only the third source is visible!



Conclusions

A properly designed two-dimensional time-encoded imager can:

1. Confirm that two objects are identical in a single measurement with NULL (constant rate) indicating a positive result.
2. Because a NULL (constant rate) is present at all times, the inspecting party might be allowed full access to the measurement and data.
3. The Feynman-Y test statistic can be updated to further protect against sensitive information loss.
4. Can image any third inspector provided object during the confirmation measurement without revealing the first two objects as an authentication measure.

Extra Slides

Neutron Coincidence Counting Equations

$$1. \quad S = F\epsilon M v_{s1} (1 + \alpha)$$

$$2. \quad D = \frac{F\epsilon^2 f_d M^2}{2} \left[v_{s2} + \left(\frac{M-1}{v_{i1}} \right) v_{s1} (1 + \alpha) v_{i2} \right]$$

$$3. \quad T =$$

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

- 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