On the Renaissance of Nuclear Fission

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Outline

1. Fission Product Yield Measurements for $^{235,238}U(n,f)$ and $^{239}Pu(n,f)$ between E_n=0.0 and 15 MeV

2. Nuclear Forensics I: Prompt Fission Neutrons
3. Nuclear Forensics II: Fission Isomers ^{134m}Te and ^{136m}Xe

4. Reactor Antineutrino Anomaly

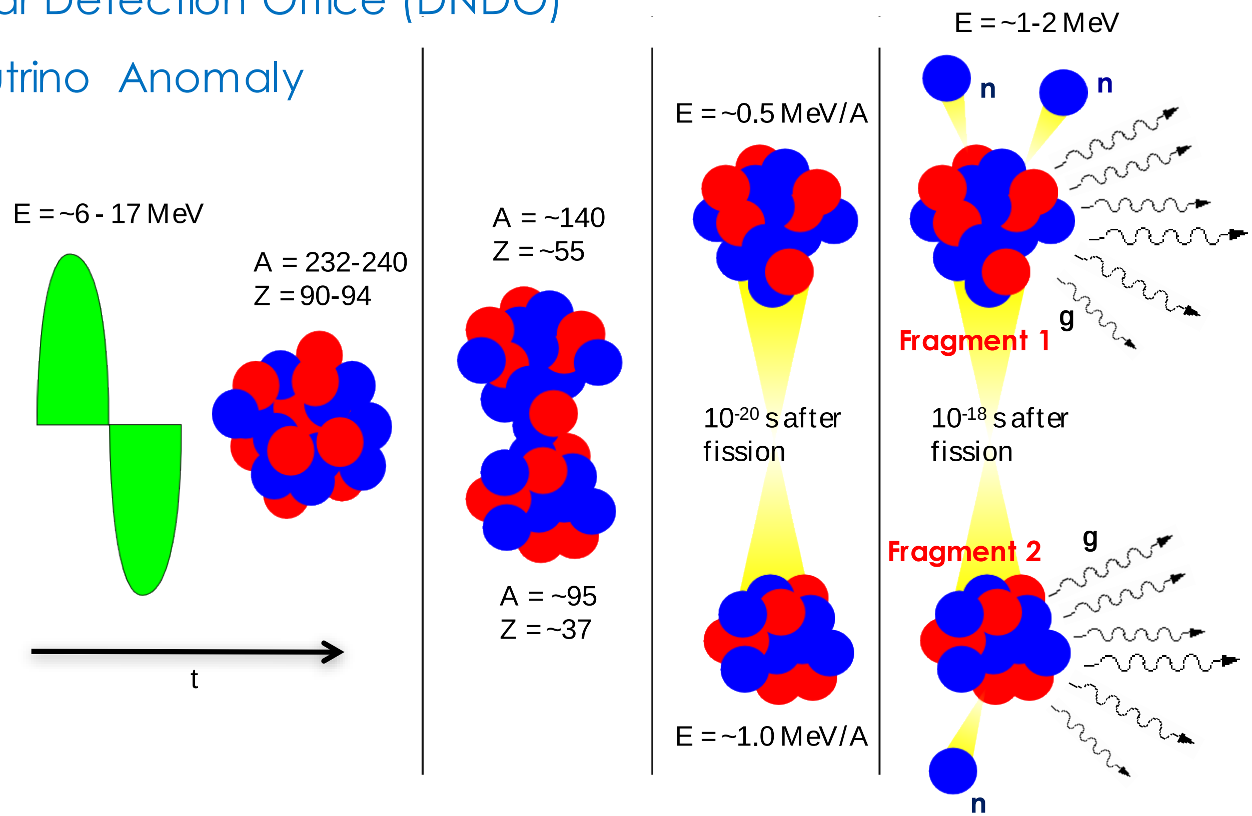
Motivations:

- Advance fundamental understanding of the fission process

- Stockpile stewardship science applications
- National Nuclear Security Administration (NNSA)

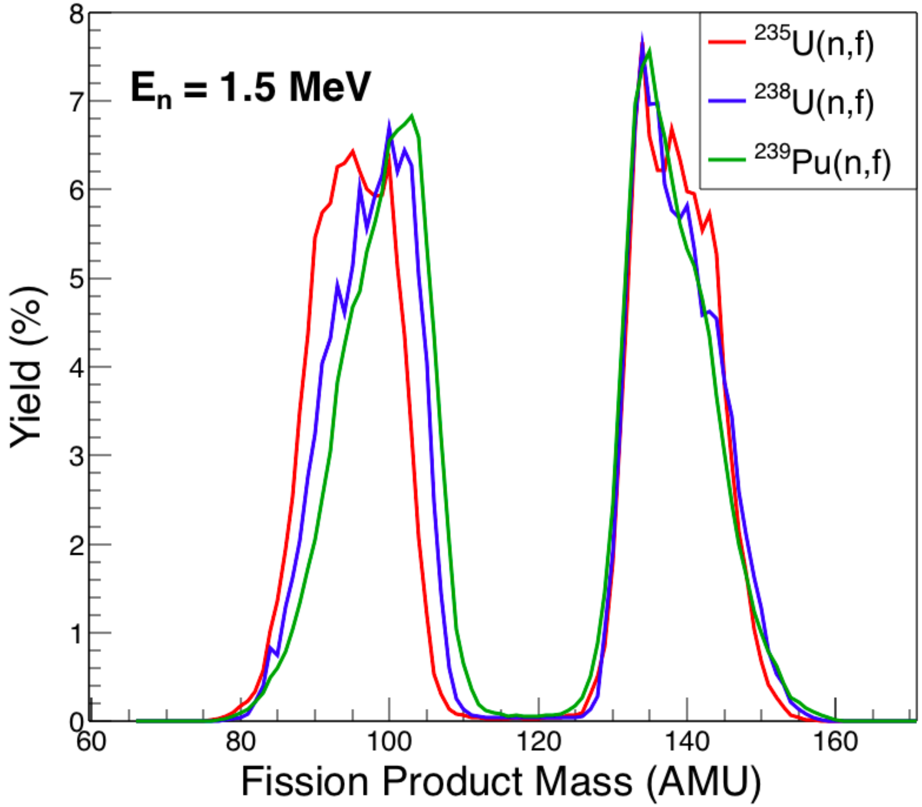
    - Remote detection of Special Nuclear Materials and Nuclear Forensics
    - Domestic Nuclear Detection Office (DNDO)
    - Reactor Antineutrino Anomaly

December 1938: Otto Hahn and Fritz Strassmann
January 1939: Lise Meitner and Otto Frisch
Before then: Enrico Fermi and his experimental group made important contributions



Fission Product Yields (FPYs)

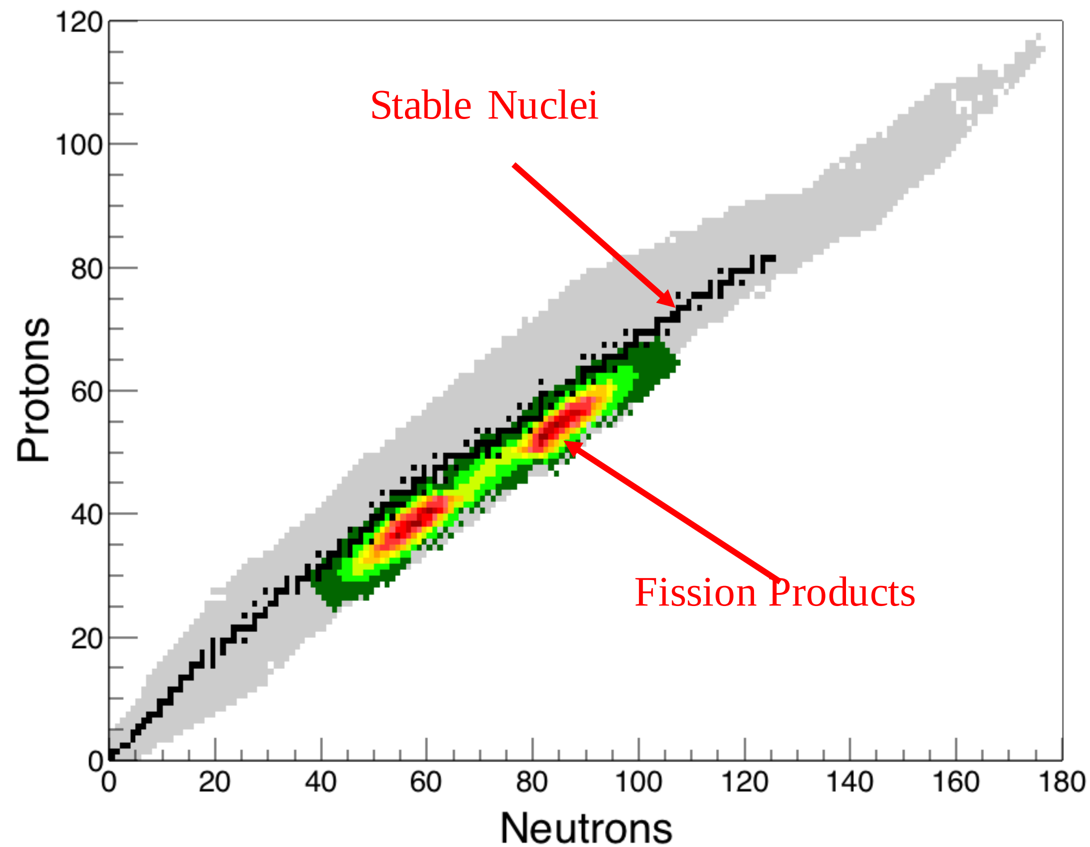
- Shape and energy evolution of FPY mass distribution is a sensitive probe of the fission process
- Need for high-precision FPY data
  - Stockpile Science
  - Nuclear Energy
  - Nuclear Forensics
  - Antineutrino Anomaly
  - Benchmark for Microscopic Fission Models

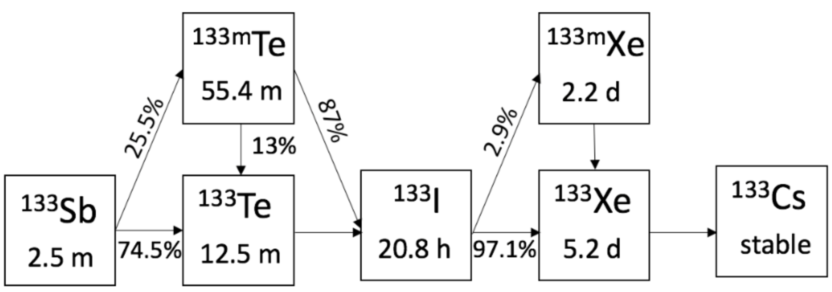


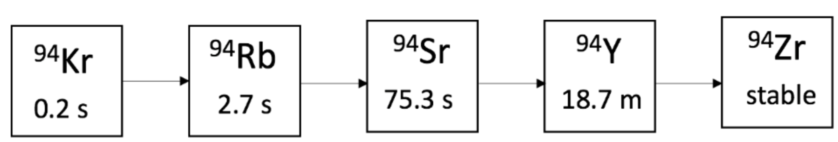
Data from T.R. England and B.F. Rider, LA-UR-94-3106 (1994)

Fission Product Yields Landscape

- Neutron-rich fission products β-decay to stability
- Identify with E_γ and $t_{1/2}$
- Determine cumulative & independent yields







Motivation for FPY measurements

- **Resolve the long-standing difference between LLNL and LANL with respect to selected fission product data**

- Joint LANL/LLNL fission product review panel suggested the possible energy dependence $^{239}Pu(n,f)^{147}Nd$ fission product yield:

  4.7%/MeV from 0.2 to 1.9 MeV (M. Chadwick)

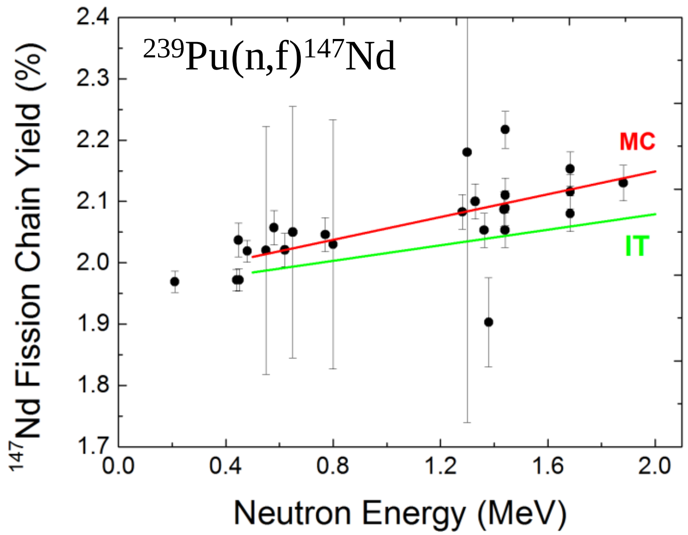
  3.2%/MeV from 0.2 to 1.9 MeV (I. Thompson)

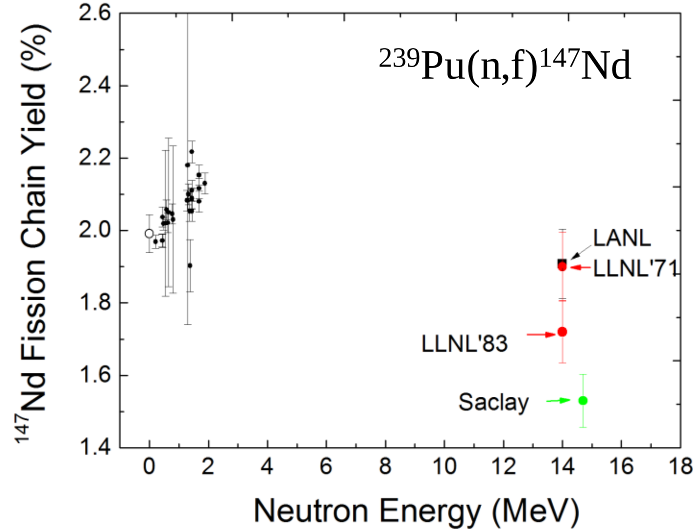
  Relative slope (%) = 100* [(FPY(E2)-FPY(E1)) / (E2-E1)] / FPY(E1)

- Mostly low energy data from critical assemblies or fast reactors

- Not many data exist in the MeV range

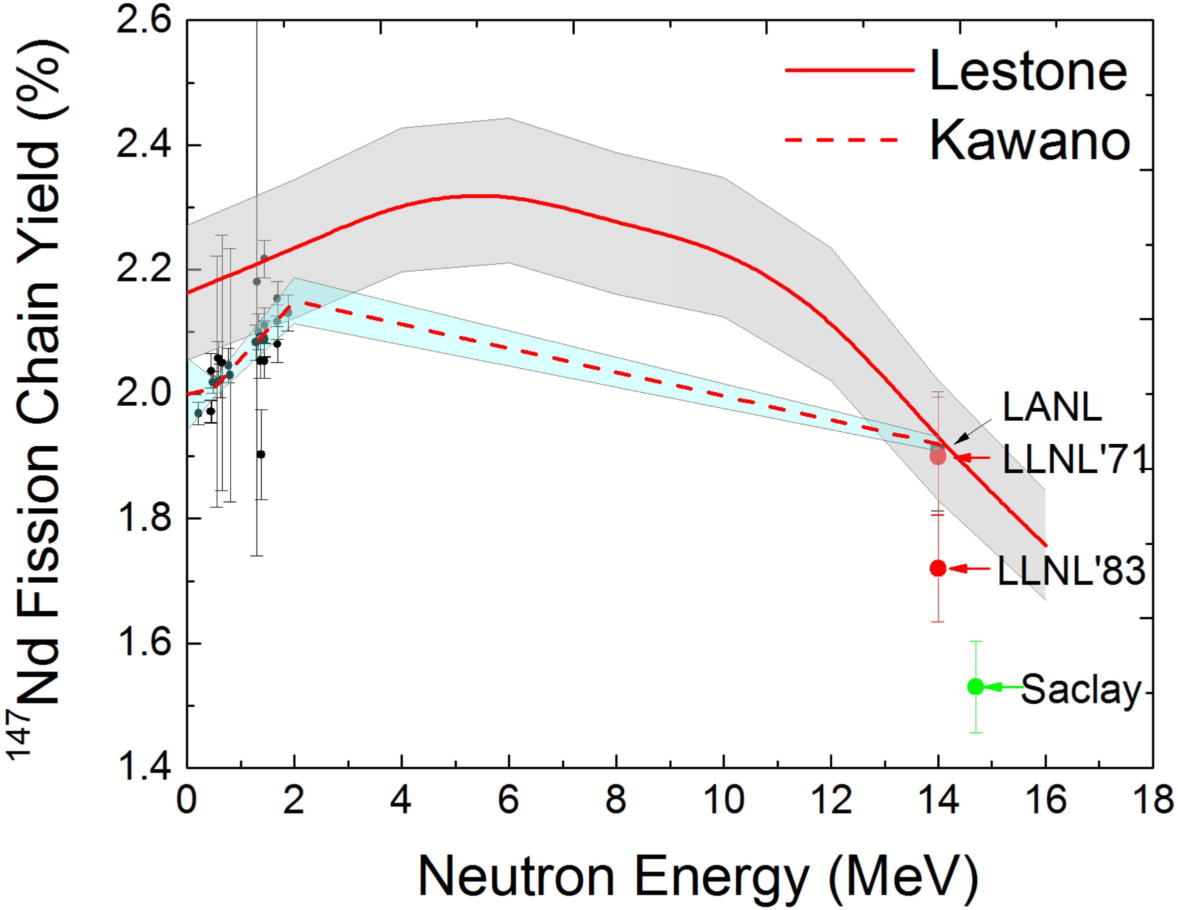
- Large discrepancy (~20%) at 14 MeV





M.B. Chadwick et al. Nuclear Data Sheets 111 (2010) 2923; H.D Selby et al. Nuclear Data Sheets 111 (2010) 2891.
P. Baisden et al, LLNL-TR-426165, 2010; R. Henderson et al. LLNL-TR-418425-DRAFT; I. Thompson et al. Nucl. Sci. Eng. **171**, 85 (2012)

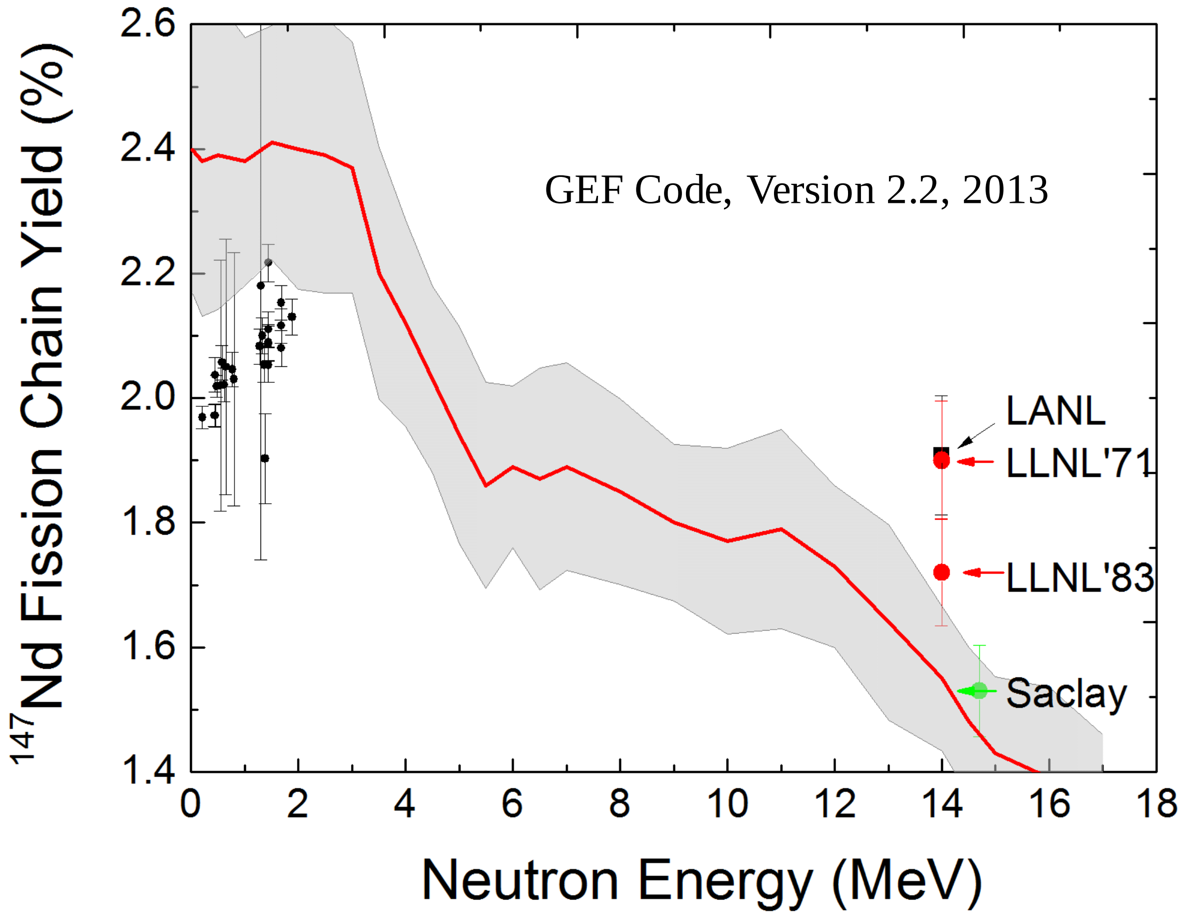
Theory Prediction



J. Lestone. Nuclear Data Sheets 112 (2011) 3120

T. Kawano *et al*. J. Nucl. Sci. Technol. 50 (2013) 1034

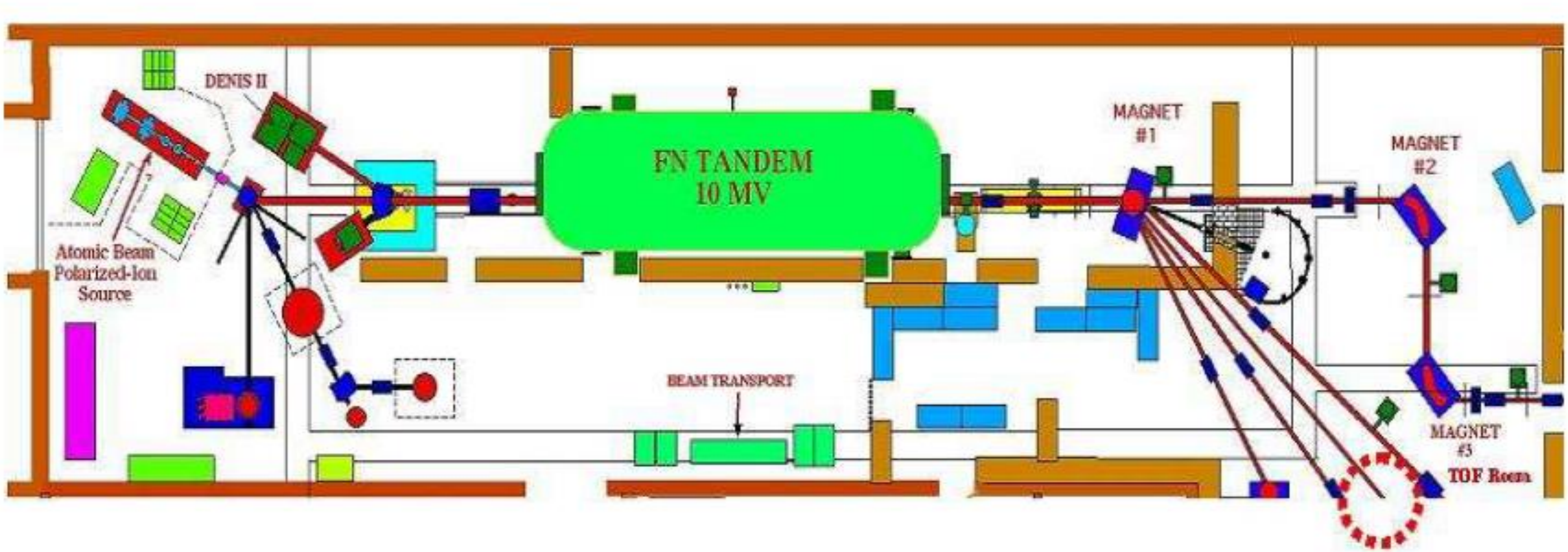
Theory Prediction



Approach

- Mono-energetic neutron beams
- Dual fission chambers → to get absolute yields
- High-resolution γ-ray spectroscopy

Floor Plan of TUNL (Triangle Universities Nuclear Laboratory)



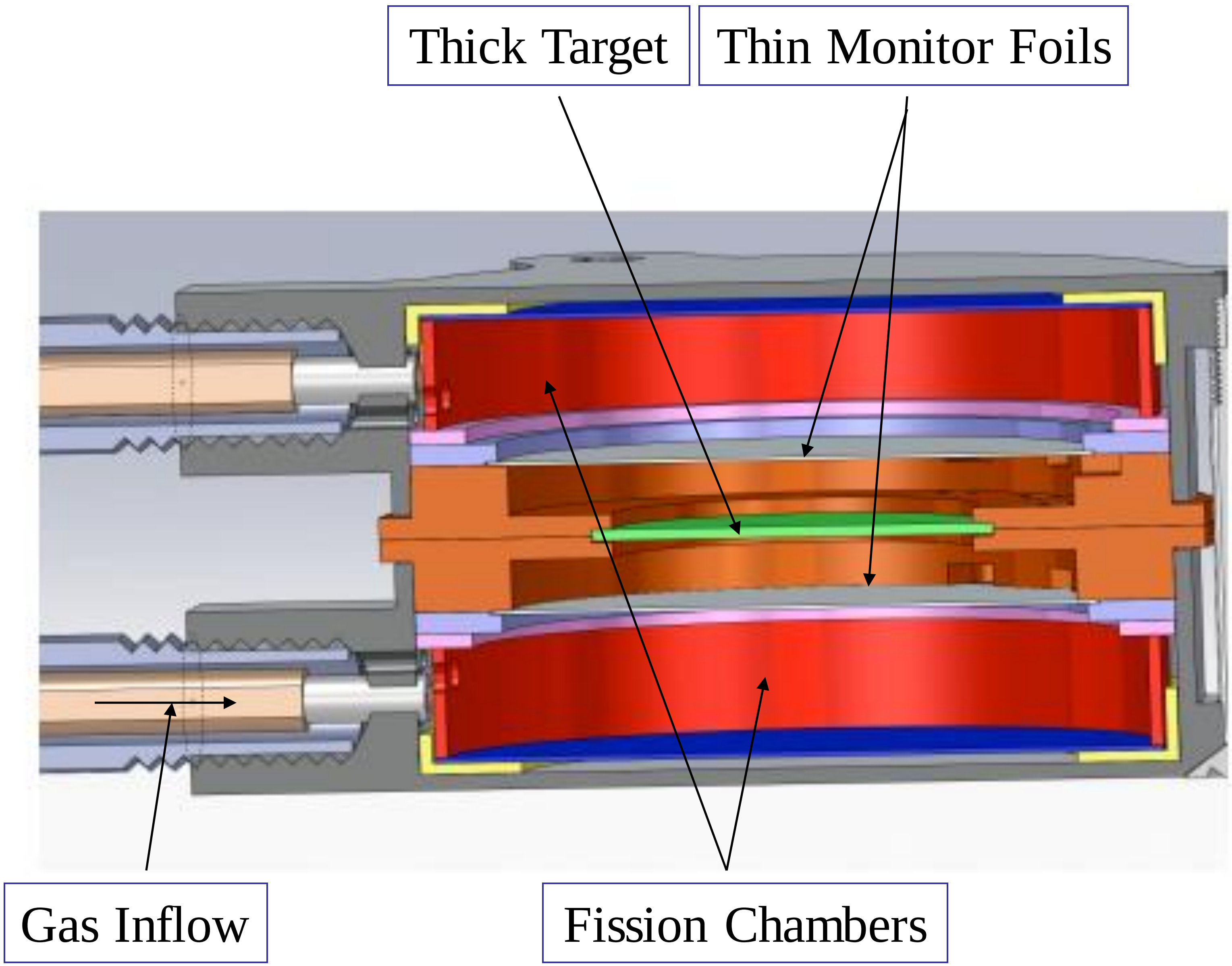
For neutron energies below E_n=0.6 MeV: $^7Li(p,n)^7Be$

For neutron energies below E_n=4 MeV: $^3H(p,n)^3He$

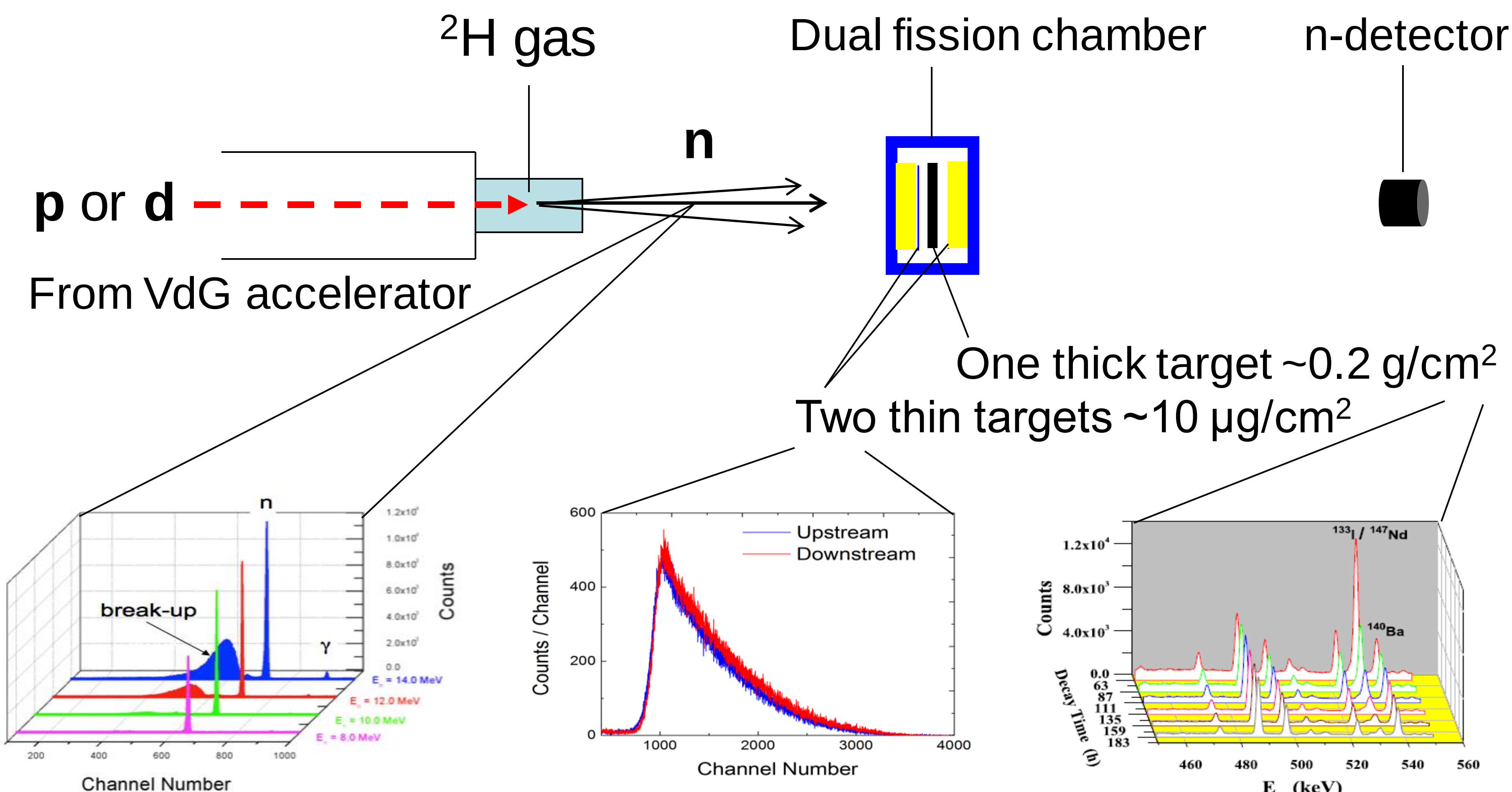
For neutron energies above E_n=4 MeV: $^2H(d,n)^3He$

For neutron energies above E_n=14.5 MeV and below 30 MeV: $^3H(d,n)^4He$

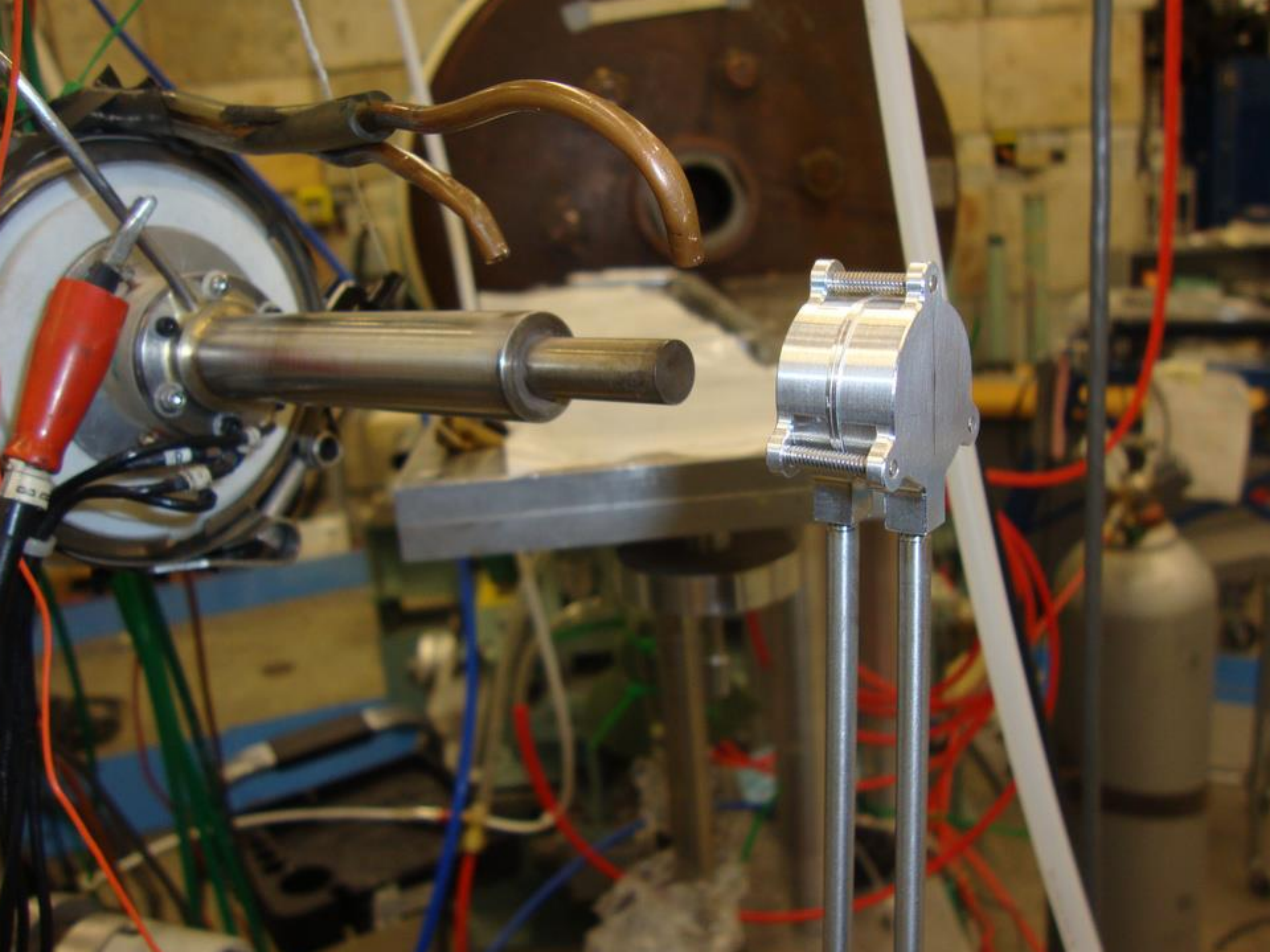
Dual Fission Chamber

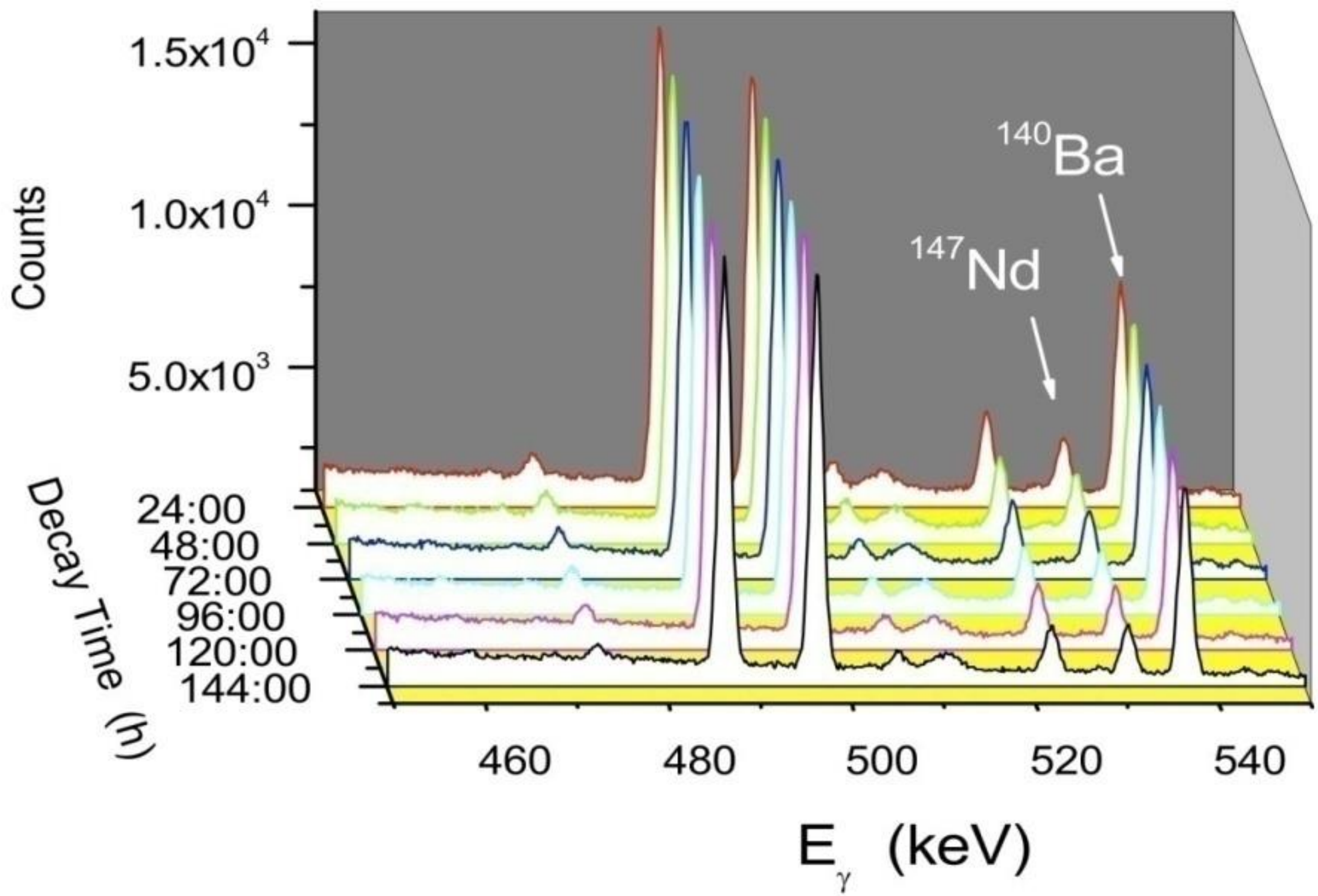


Monoenergetic Neutron Irradiation



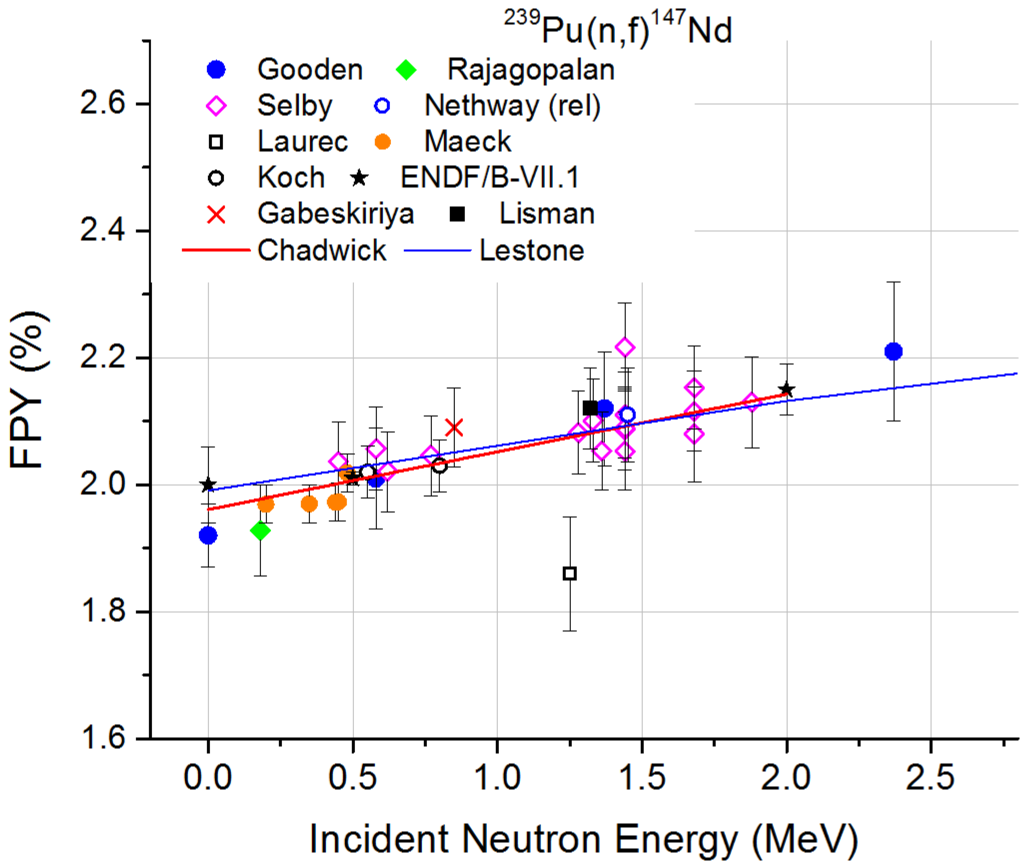
$$(\text{Gamma\_count} / \text{Fission\_count}) = (m_{thick} / m_{thin}) * \text{FPY} * C$$





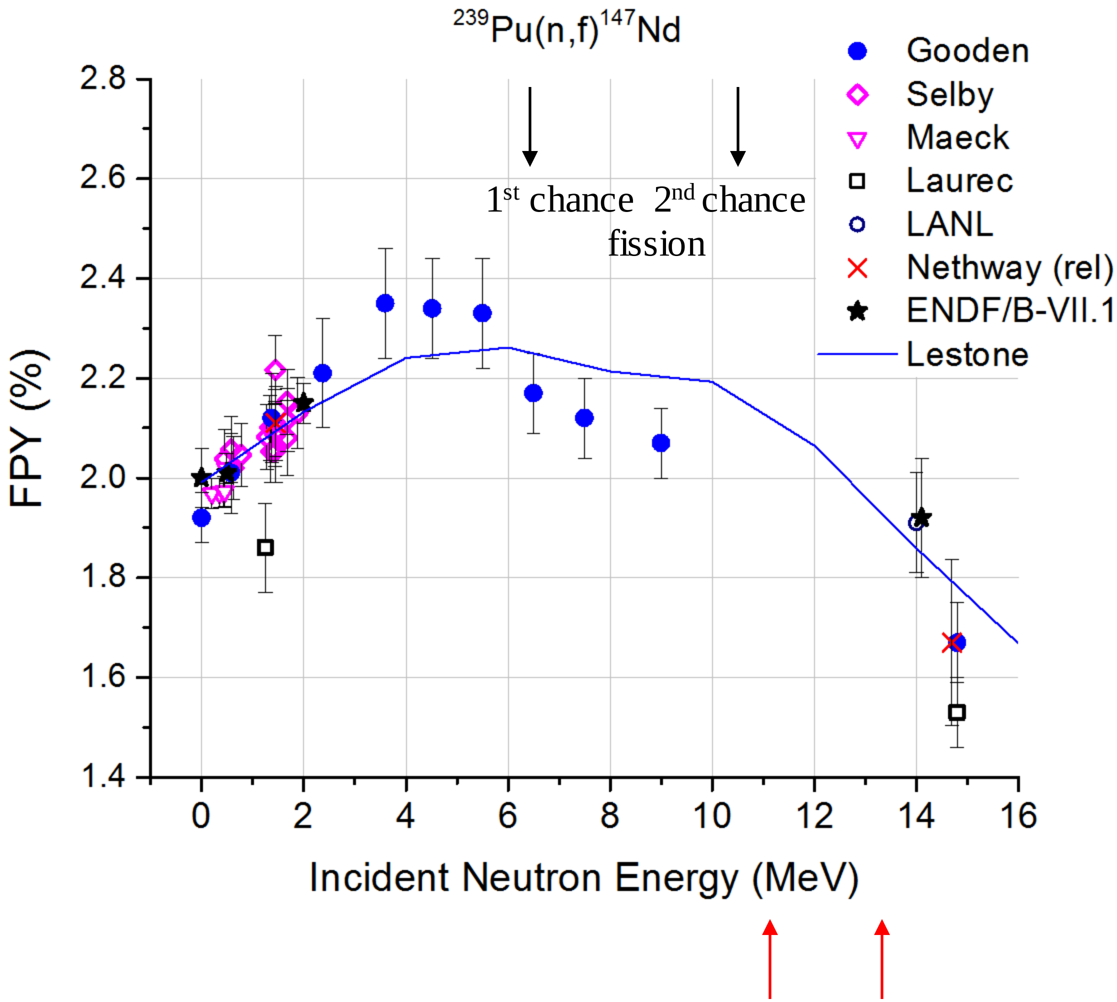
Counts
1.5x10^4
1.0x10^4
5.0x10^3
^140Ba
^147Nd
Decay Time (h)
24:00
48:00
72:00
96:00
120:00
144:00
460
480
500
520
540
Eγ (keV)

Fragment	E_γ (keV)	$T_{1/2}$	I_γ %
^{91}Sr	1024.3	9.63(5) h	33.50(1.10)
^{92}Sr	1383.93	2.611(10) h	90(4)
^{95}Zr	724.192	64.032(6) d	44.27(22)
^{97}Zr	743.36	16.749(8) h	93.09(16)
^{99}Mo	739.5	65.976(24) h	12.26(22)
^{103}Ru	497.09	39.247(13) d	91.0(1.2)
^{105}Ru	724.3	4.44(2) h	47.3(5)
^{127}Sb	685.7	3.85(5) d	36.8(2.0)
^{131}I	364.489	8.0252(6) d	81.5(8)
^{132}Te	228.16	3.204(13) d	88(3)
^{133}I	529.872	20.83(8) h	87.0(3)
^{135}Xe	249.794	9.14(2) h	90(3)
^{136}Cs	1048.07	13.16(3) d	80(3)
^{140}Ba	537.261	12.7527(23) d	24.39(23)
^{143}Ce	293.266	33.039(6) h	42.8(4)
^{147}Nd	531.016	10.98(1) d	13.37(27)



$^{239}Pu(n,f)^{147}Nd$
Gooden
Rajagopalan
Selby
Nethway (rel)
Laurec
Maeck
Koch
ENDF/B-VII.1
Gabeskiriya
Lisman
Chadwick
Lestone
2.6
2.4
2.2
2.0
1.8
1.6
FPY (%)
0.0
0.5
1.0
1.5
2.0
2.5
Incident Neutron Energy (MeV)

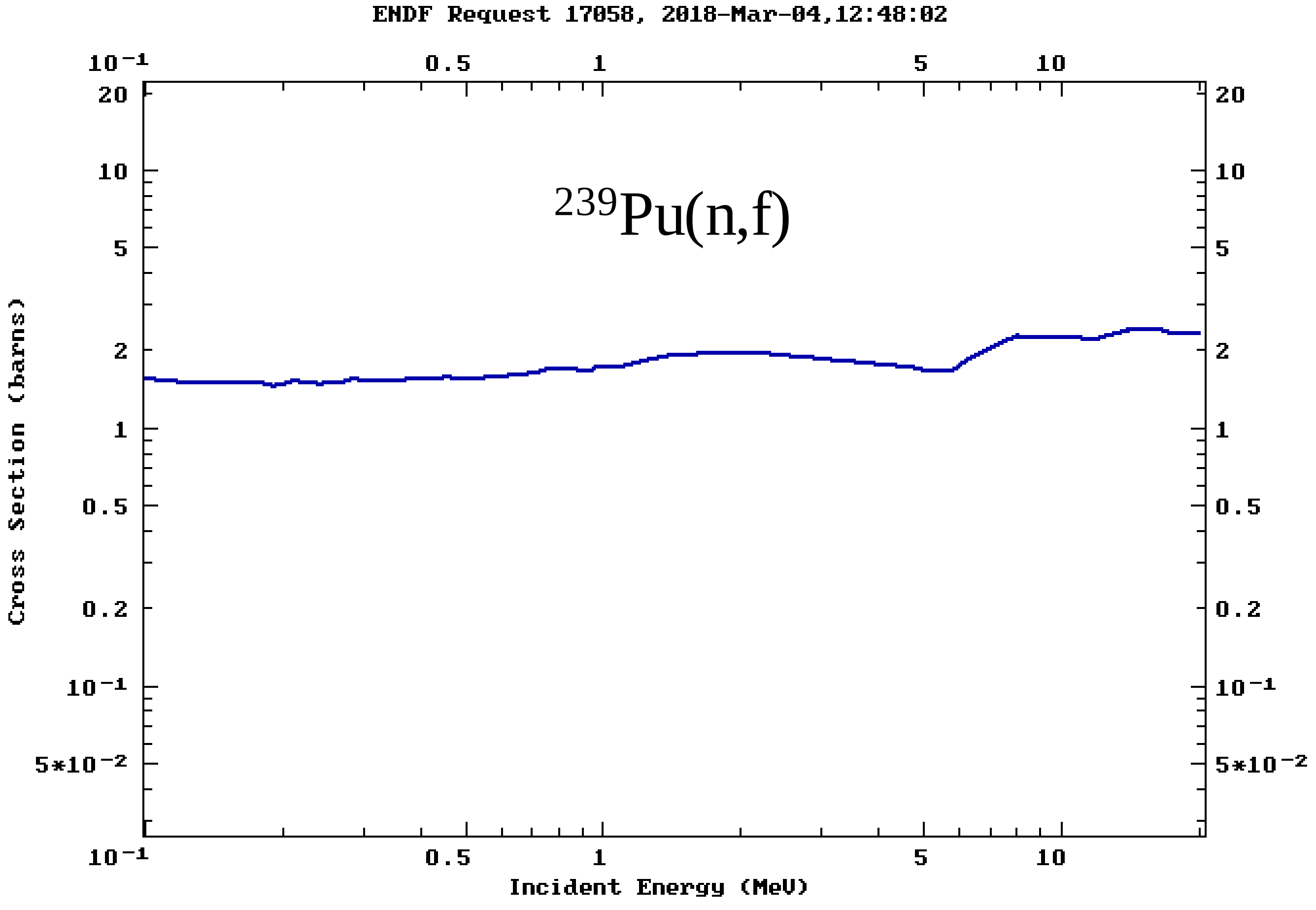
Preliminary



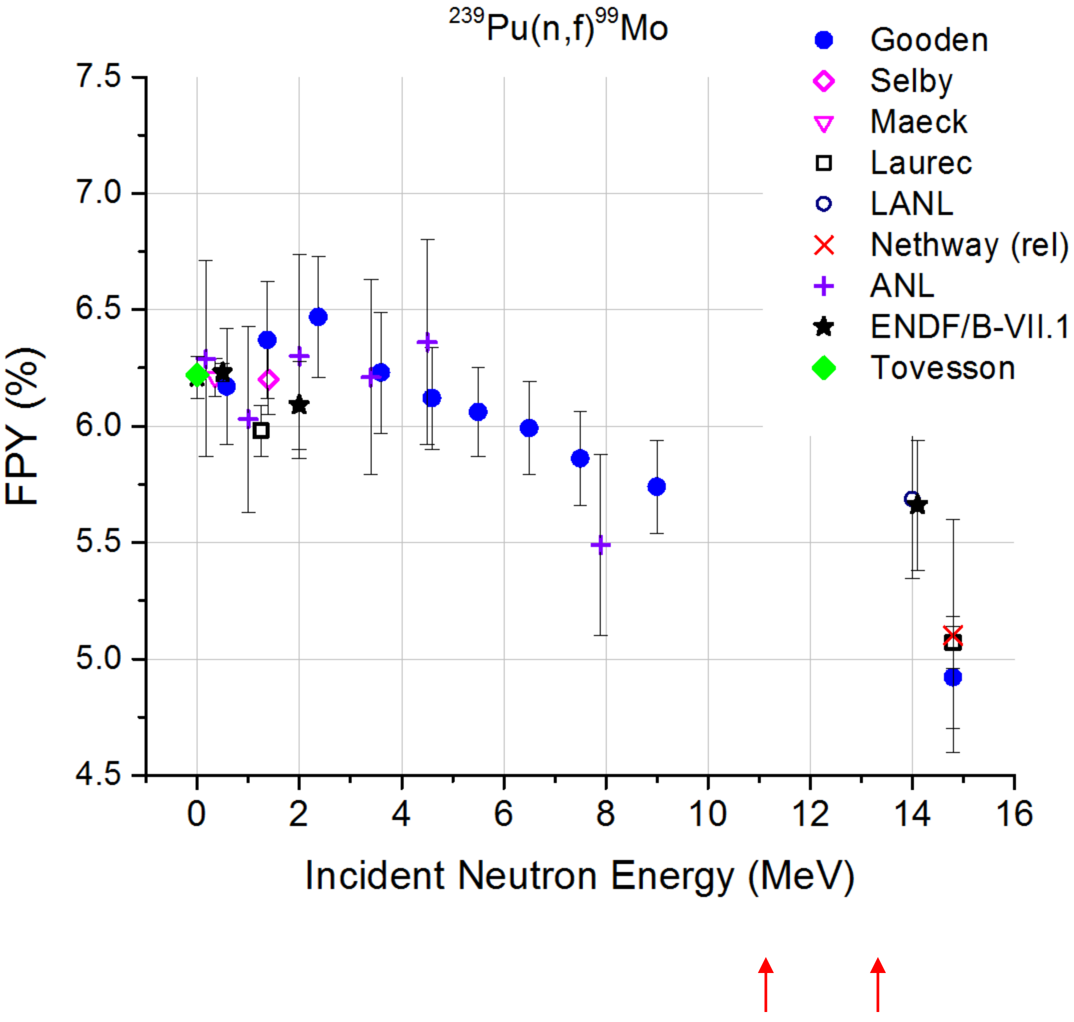
^{239}Pu(n,nf)

^{239}Pu(n,2nf)

9 MeV datum not corrected for off-energy neutrons from the $^{2}H(d,n)^{3}He$ reaction

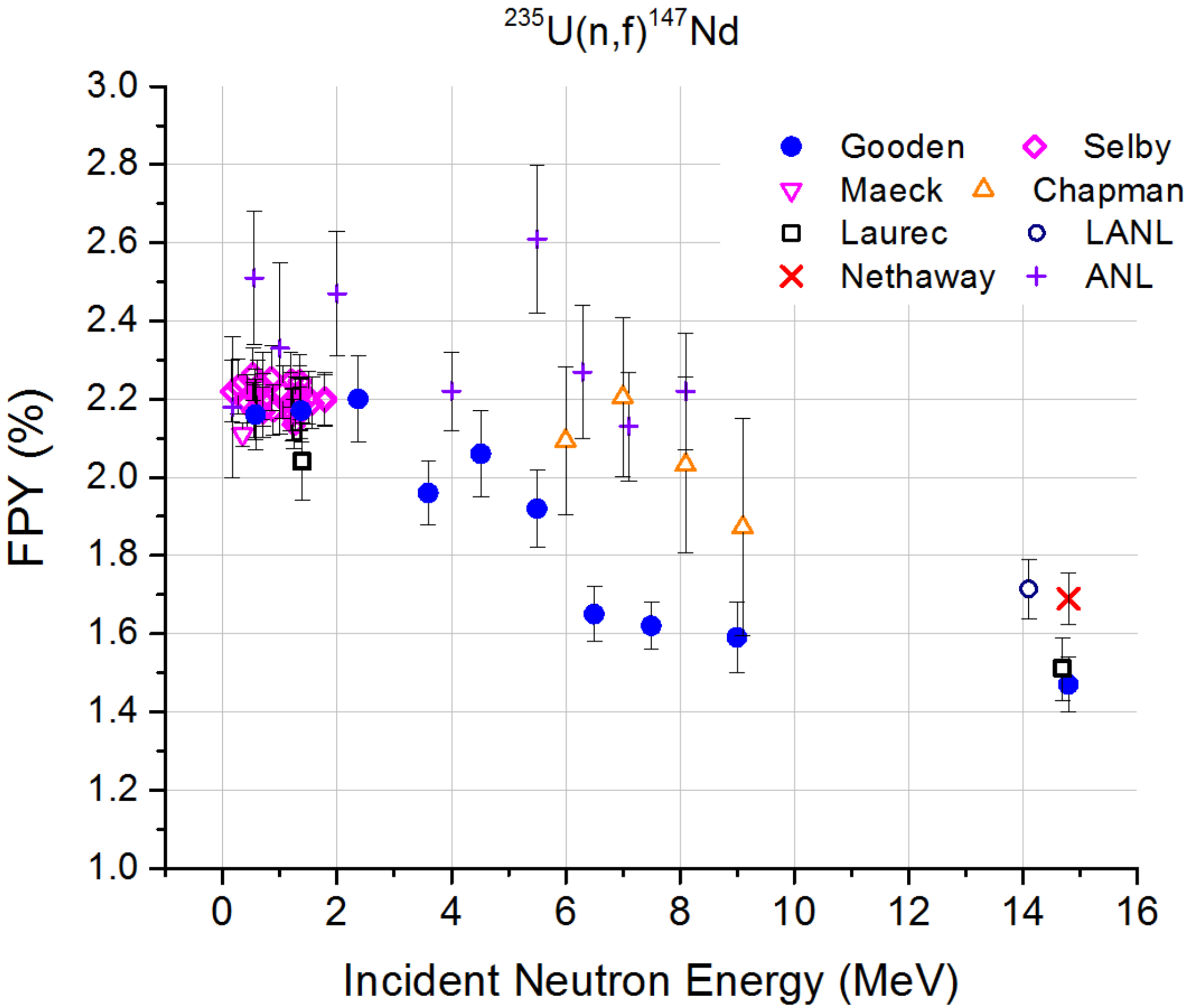

ENDF Request 17058, 2018-Mar-04,12:48:02
$^{239}Pu(n,f)$
Cross Section (barns)
Incident Energy (MeV)
10^{-1}
0.5
1
5
10
20
2
0.2
$5*10^{-2}$

Preliminary

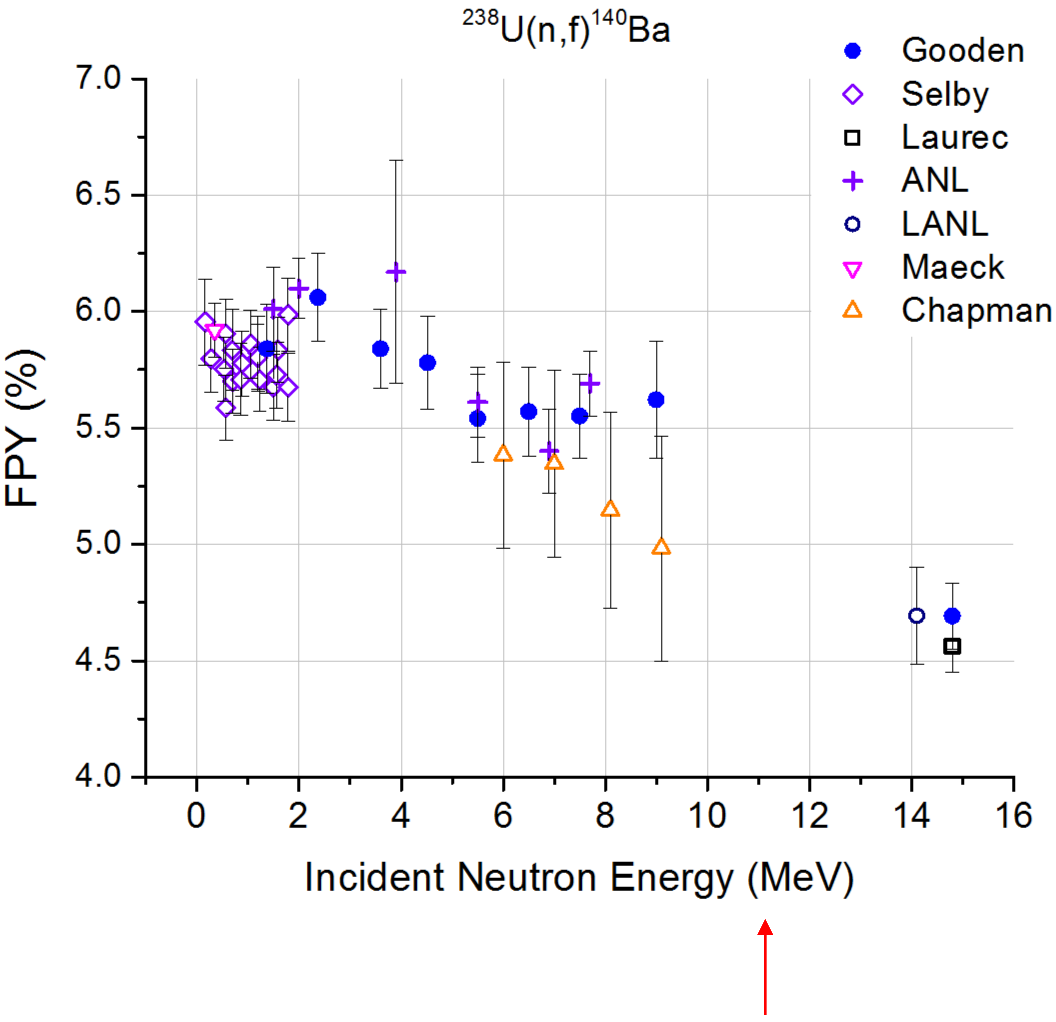


Datum at 9 MeV not corrected for off-energy neutrons from the ^{2}H(d,n)^{3}He reaction

Preliminary

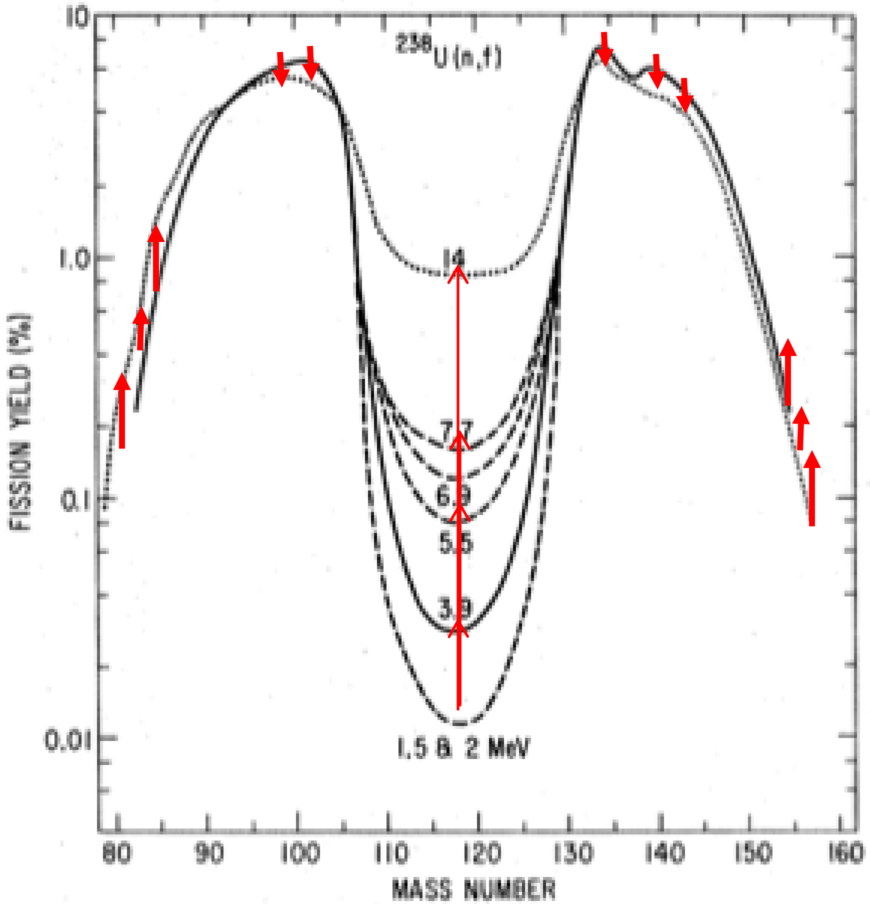


Preliminary



9 MeV datum not corrected for off-energy neutrons from $^{2}H(d,n)^{3}He$ reaction

Fission Product Mass Distribution: What have we learned so far?

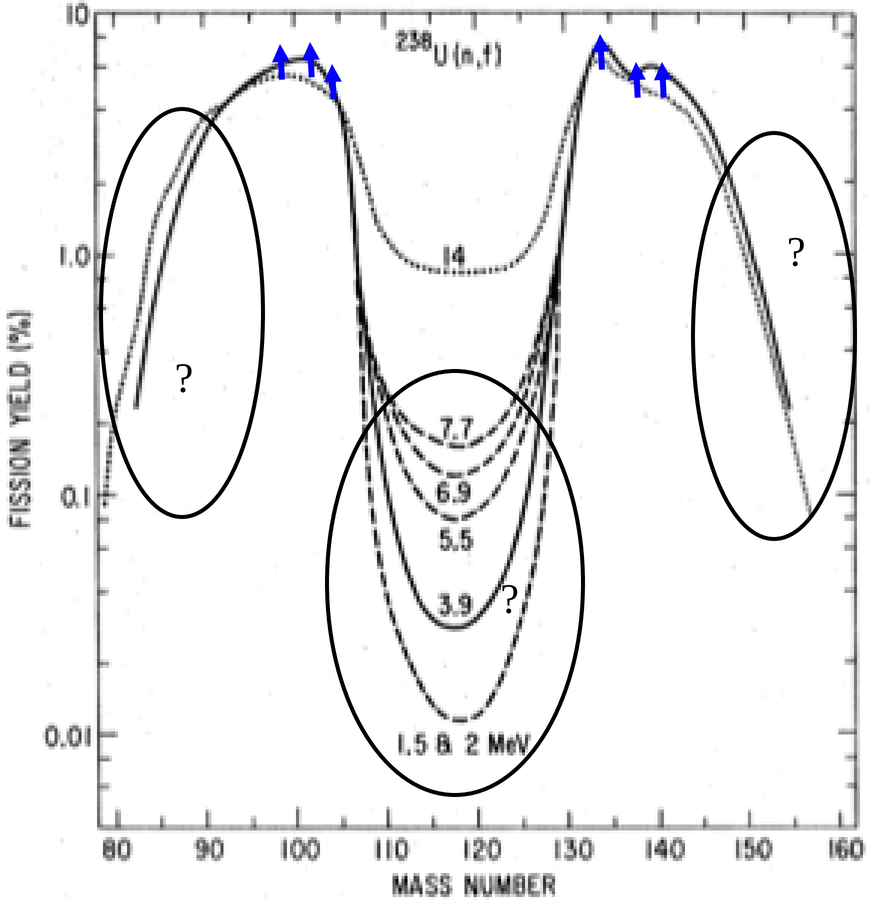


At higher energies, 5 – 15 MeV, all is consistent with what we know

- The symmetric FPYs increase
- The two asymmetric FPY's slightly decrease
- The very asymmetric FPY's (the wings) slightly increase

AT

Fission Product Mass Distribution: What have we learned so far?



At higher energies, 5 – 15 MeV, all is consistent with what we know

- The symmetric FPYs increase
- The two asymmetric FPY's slightly decrease
- The very asymmetric FPY's (the wings) slightly increase

- At lower energies, 0.5 –5 MeV, a new phenomenon

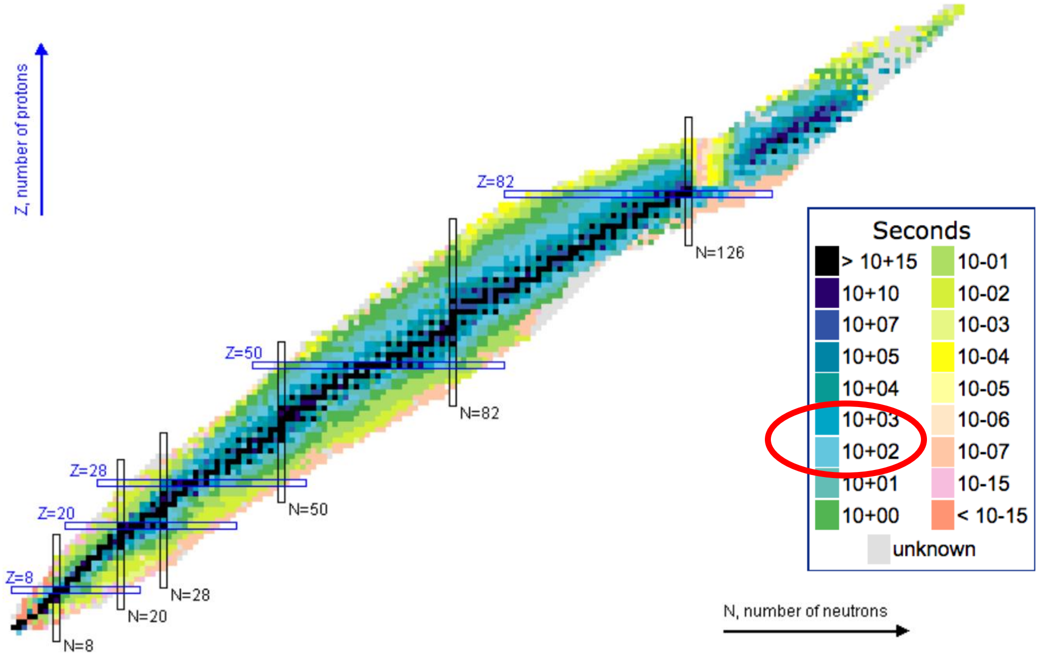
Expand effort on basic theory of fission to understand evolution of yields at low neutron energies

Conjecture: Interplay between pairing and shell effects

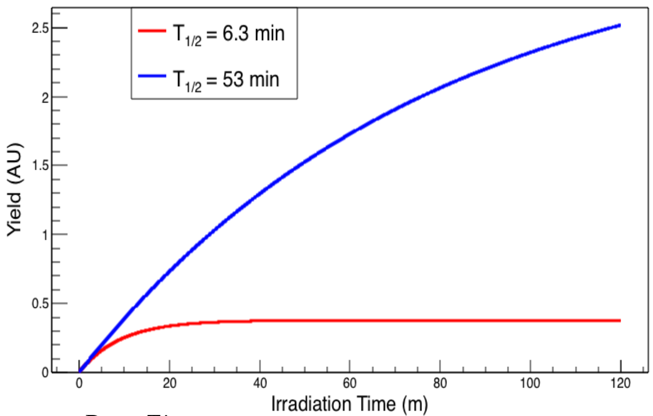
AT

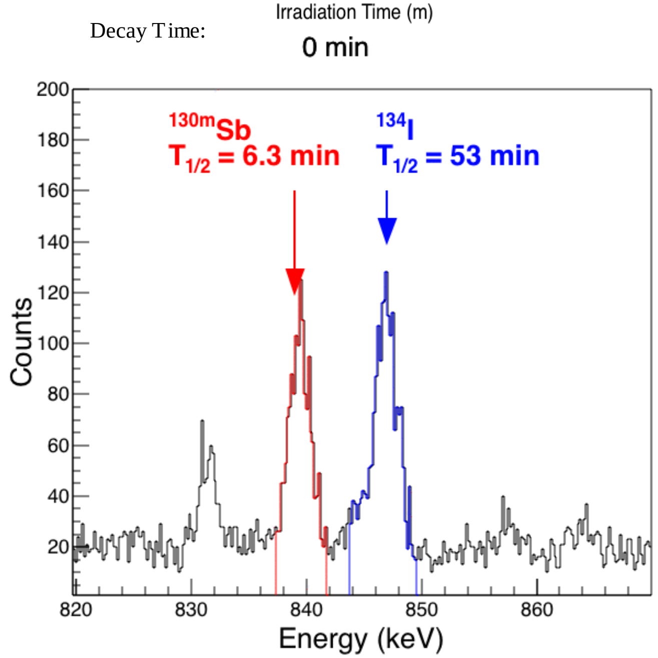
Short Lived FPY Measurements

- Expose to neutron beam for 1 hr, begin counting immediately after (<5 min) and count continuously for 3-4 days
- Reduce activity from long lived FPs, halving background



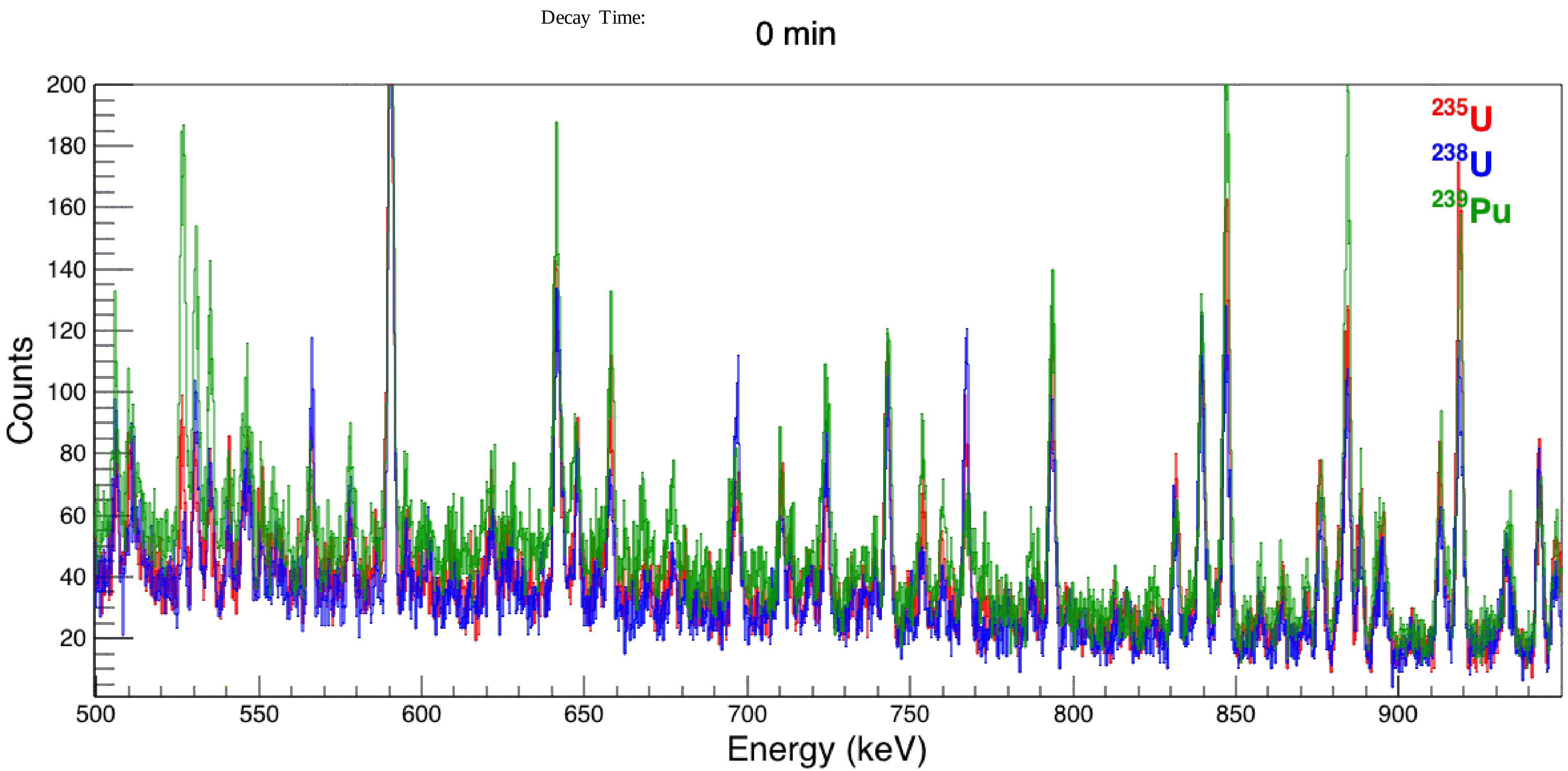
https://www.nndc.bnl.gov/chart/





JS

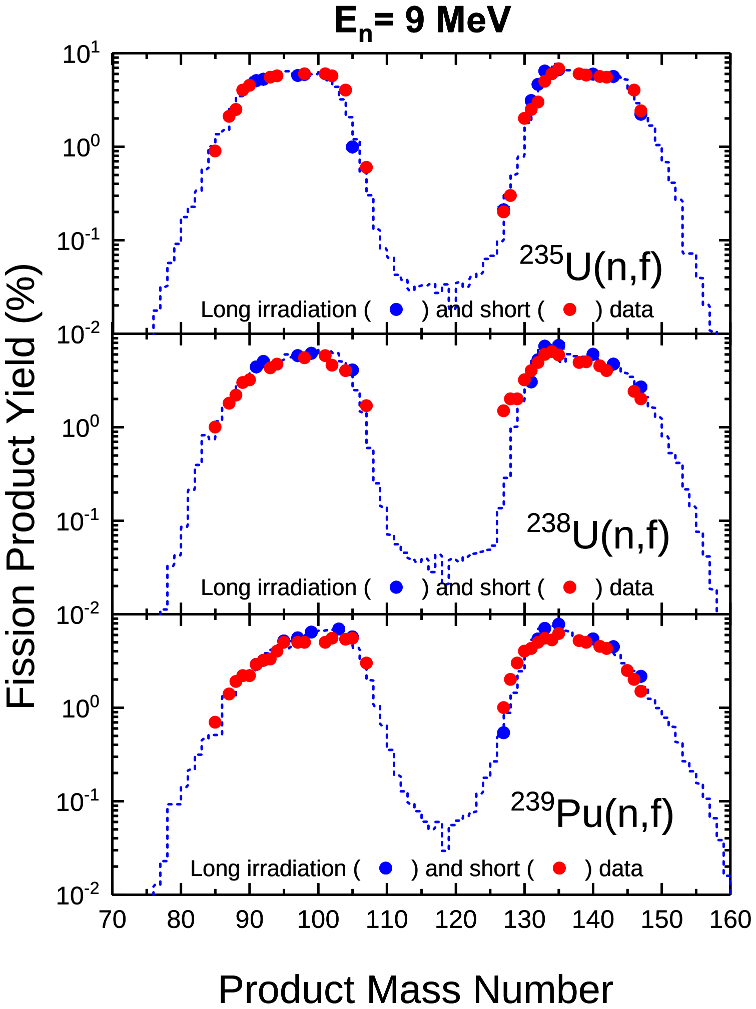
Fission Product γ-Ray Spectra at E_n = 9 MeV



JS

FPY Mass Distribution: Long & Short Irradiation

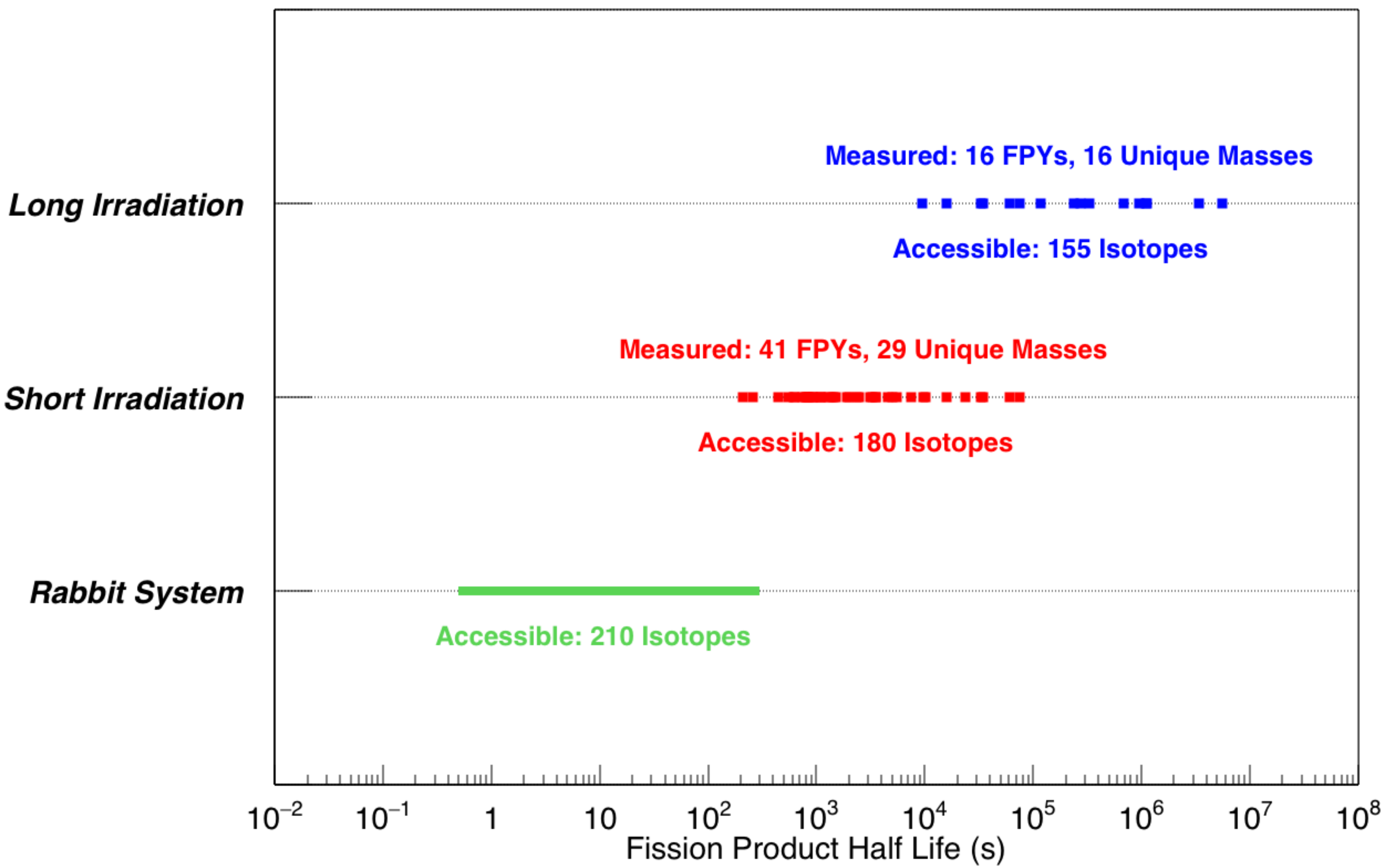
- Self-consistent, systematic approach to measuring FPYs
- Long-lived (days or weeks)
- Short-lived (minutes or hours)
- Constrain the mass distribution



Preliminary

JS

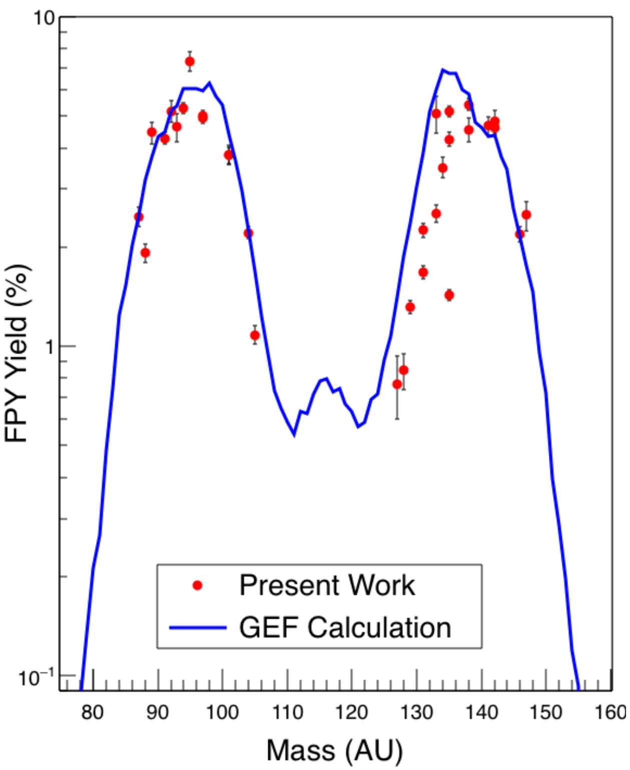
Measured Fission Product Yields



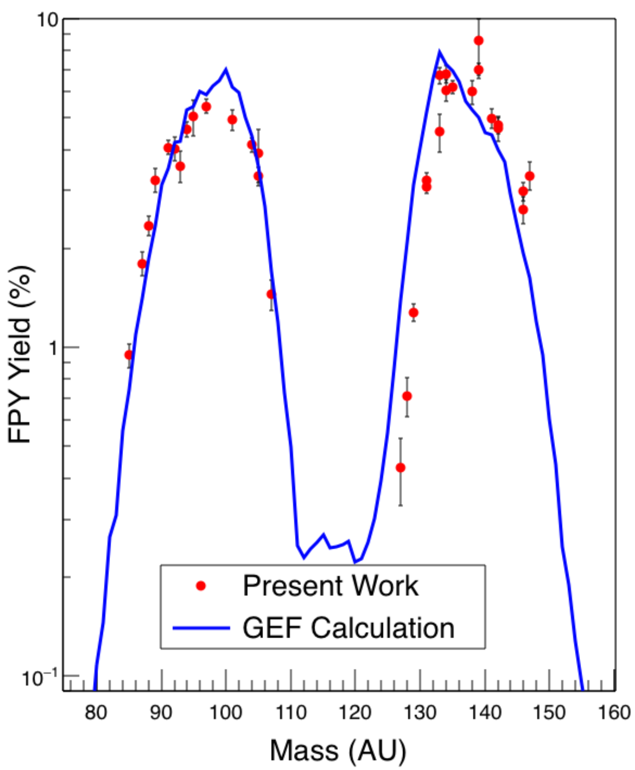
Independent ← Cumulative Fission Product Yields

Preliminary Results from E_n = 9 MeV

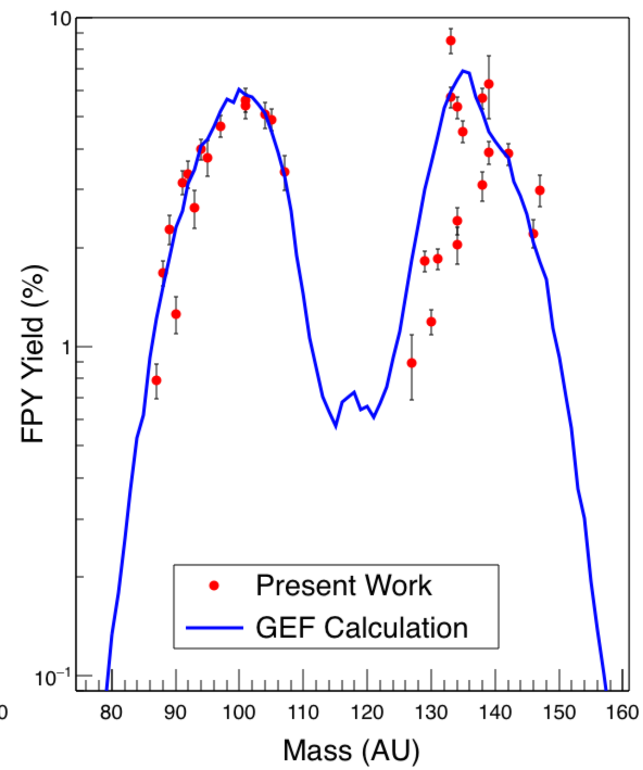
235U



238U

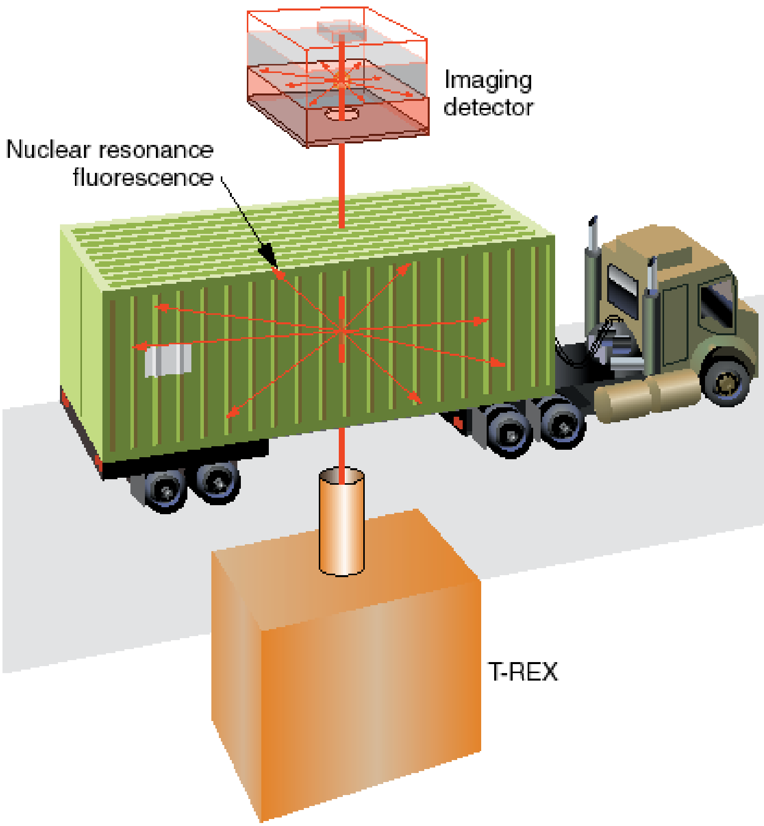


239Pu



JS

Fission and Nuclear Forensics I



Detection of special nuclear materials

- Non-destructive interrogation
- If positive signal: identification
- 10 seconds per cargo container

U.S. Department of Homeland Security, Domestic Nuclear Detection Office:

NO neutrons anymore!

Use γ rays (*i.e.*, high energy photons) below 10 MeV

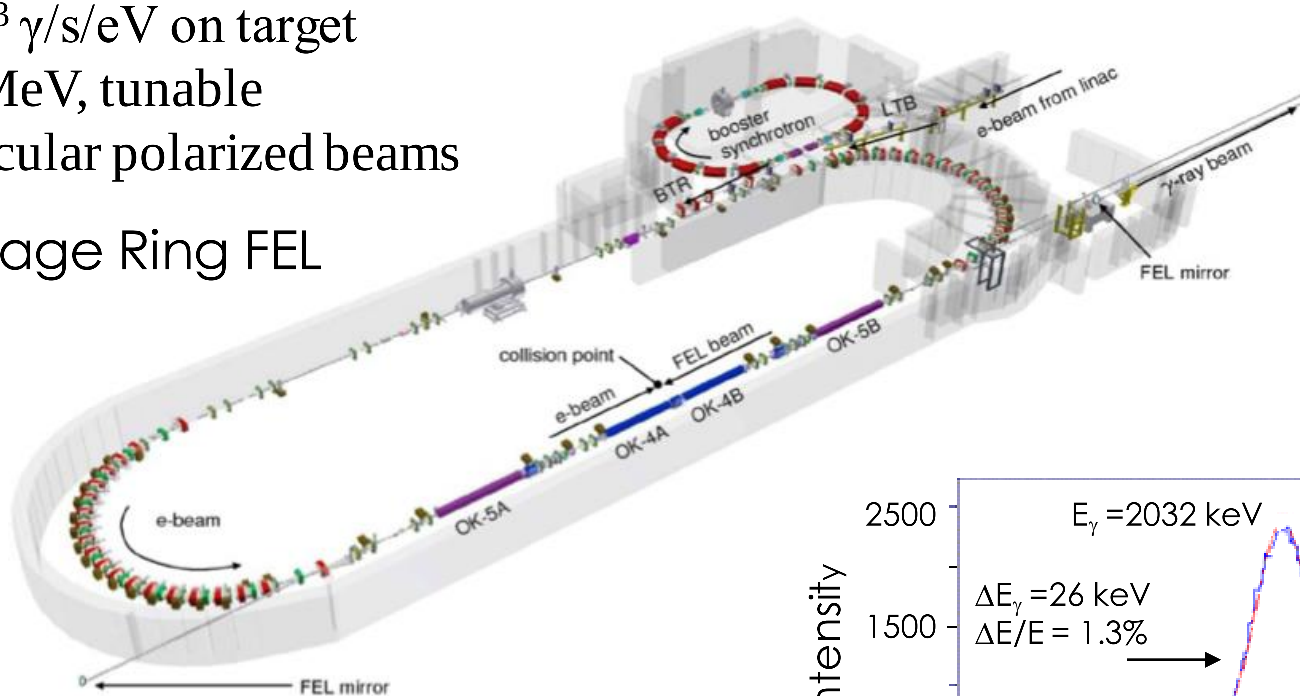
(γ,f) ~5 times smaller than (n,f)

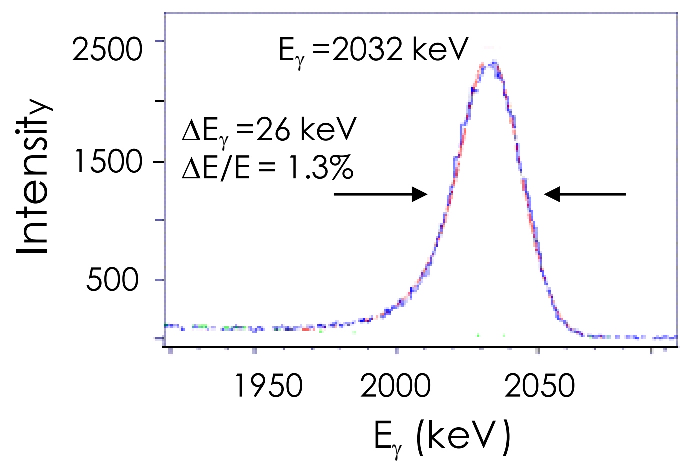
High-Intensity γ-Ray Source @ TUNL

HIγS is the most intense Compton γ-ray source in the world

Produces γ-rays by Compton backscattering
Intensity = 10^3 γ/s/eV on target
E_γ = 1 – 100 MeV, tunable
Linear and circular polarized beams

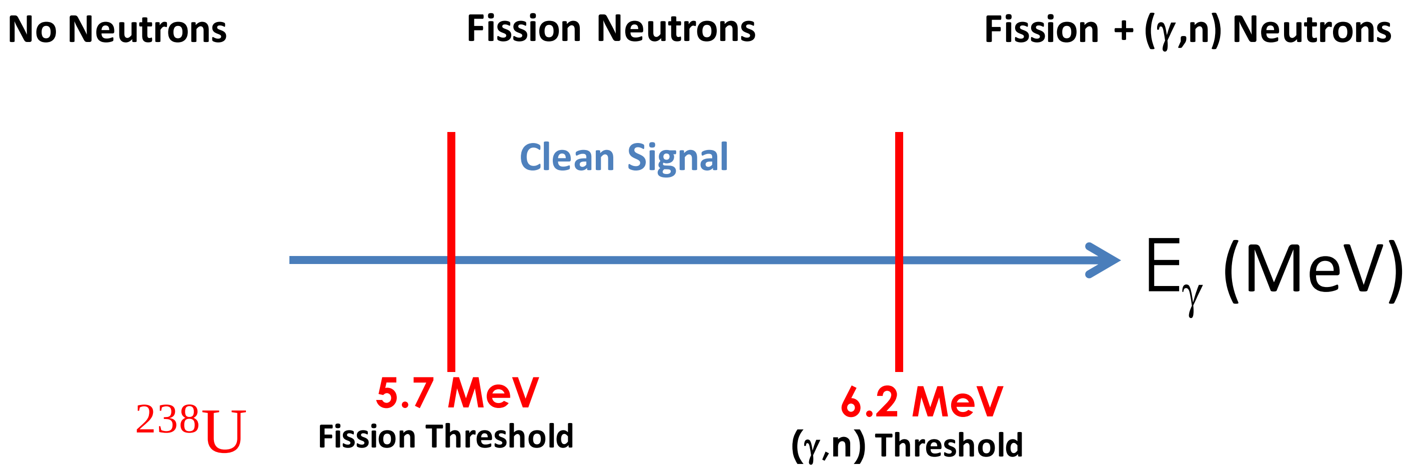
1.2 GeV Storage Ring FEL





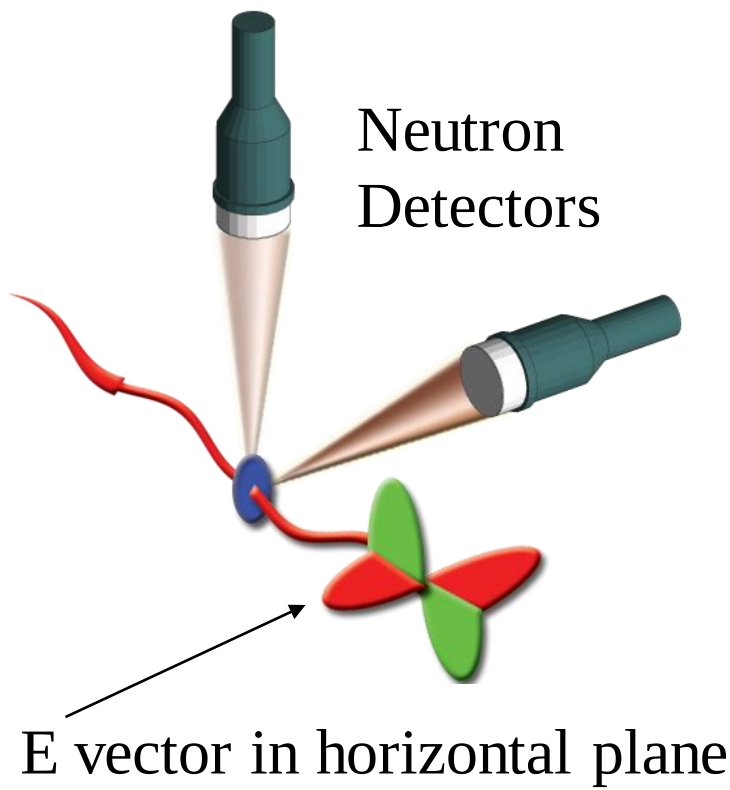
Beam time structure: Rep rate = 5.58 MHz (T = 180 ns) and Δt = 100 ps

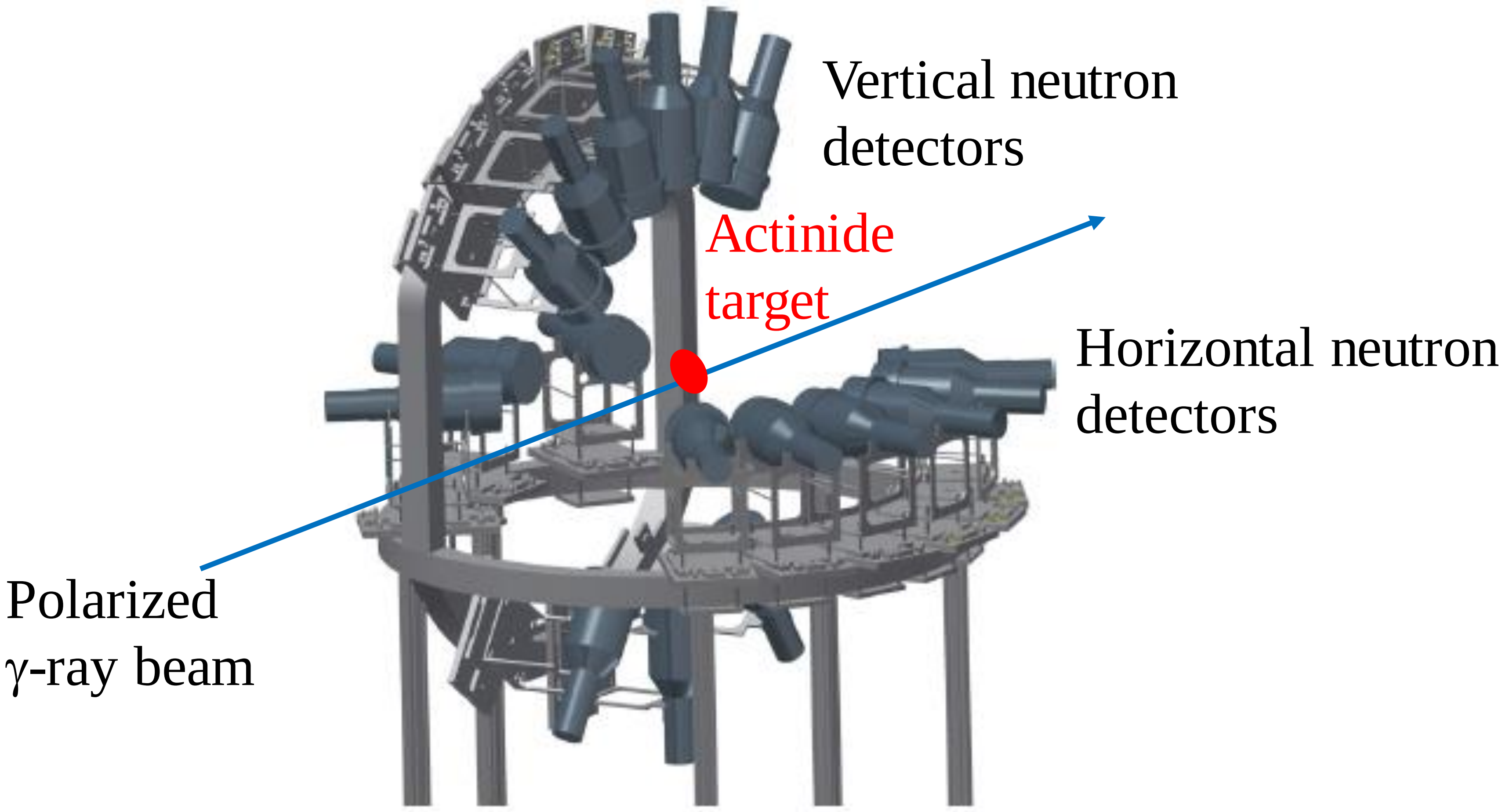
Prompt fission neutrons



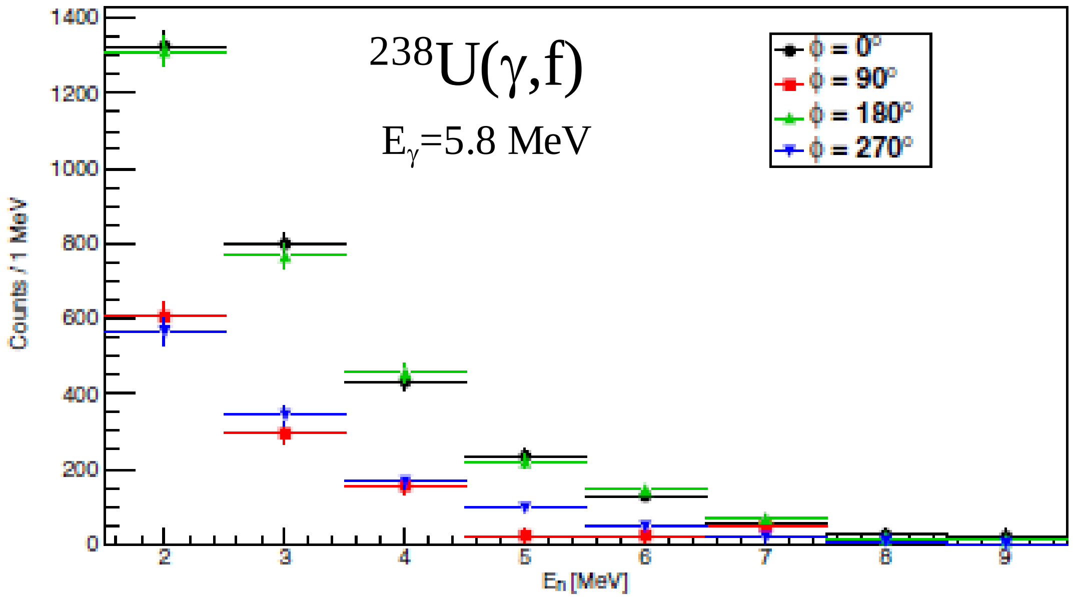
- Typical energy range E_γ = 5.8 - 7.0 MeV
- Only other stable isotopes which can produce neutrons at these energies are ^{2}H and ^{9}Be
- The neutron energy detection threshold is 1.5 MeV
- All neutrons below (γ,n) threshold are fission neutrons

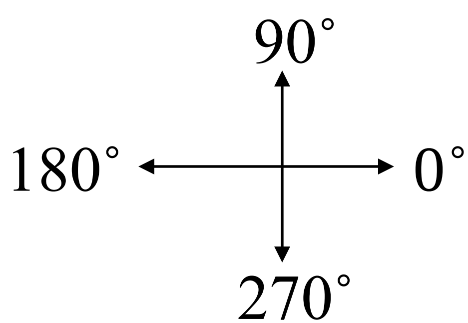
- Polarized γ-ray induces fission of target nuclei
- Prompt neutrons are detected both parallel and perpendicular to the plane of polarization of the incident γ-ray




Vertical neutron detectors
Actinide target
Horizontal neutron detectors
Polarized γ-ray beam

Mueller et al.

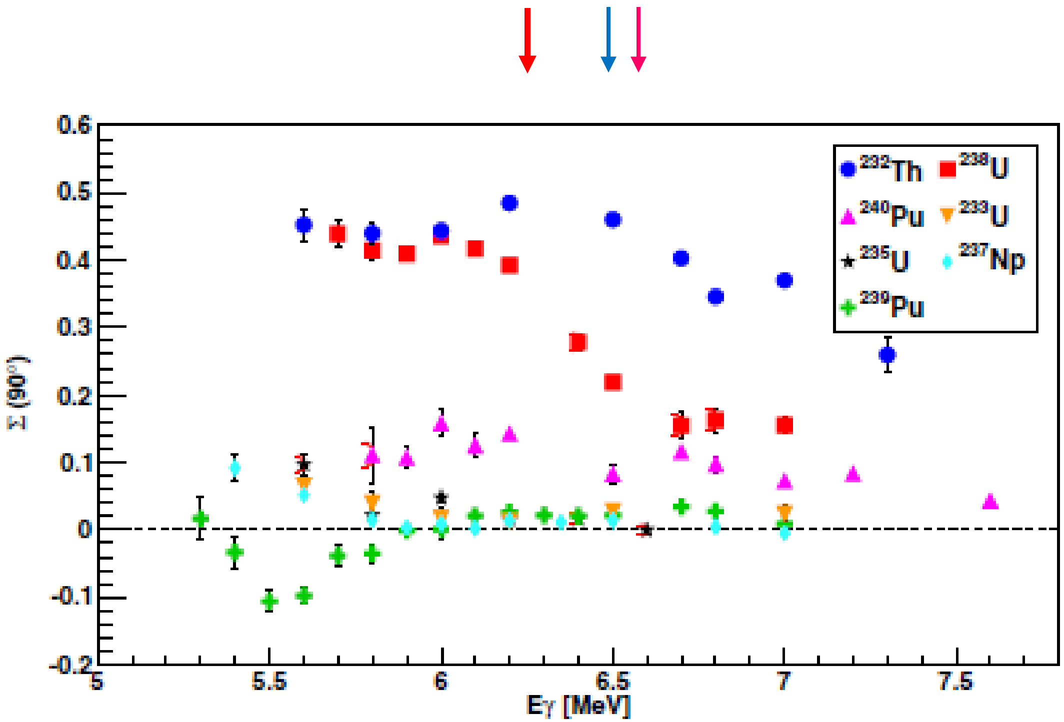




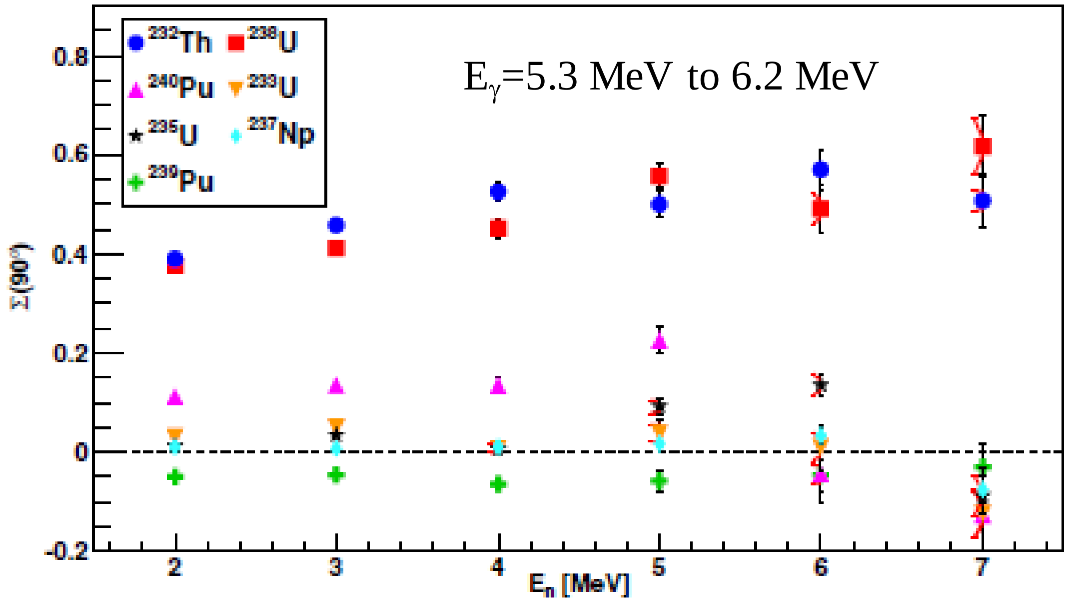
E vector : Polarization Plane

Mueller et al

(γ,n)



$$\Sigma(\theta) = \frac{W(\theta,0^\circ) + W(\theta,180^\circ) - W(\theta,90^\circ) - W(\theta,270^\circ)}{W(\theta,0^\circ) + W(\theta,180^\circ) + W(\theta,90^\circ) + W(\theta,270^\circ)}.$$



Mueller *et al*.

$$\Sigma(\theta) = \frac{W(\theta,0^\circ) + W(\theta,180^\circ) - W(\theta,90^\circ) - W(\theta,270^\circ)}{W(\theta,0^\circ) + W(\theta,180^\circ) + W(\theta,90^\circ) + W(\theta,270^\circ)}.$$

Even-even actinides ^{232}Th and ^{238}U show large asymmetries, but smaller for ^{240}Pu

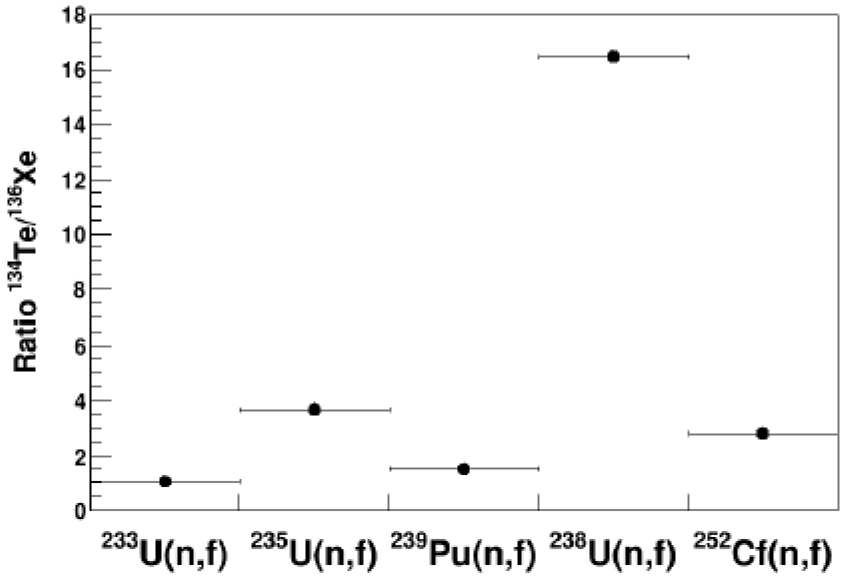
Odd-A actinides show little to no asymmetries

Possible reason: Different spin and level densities

Nuclear Forensics II

Applications

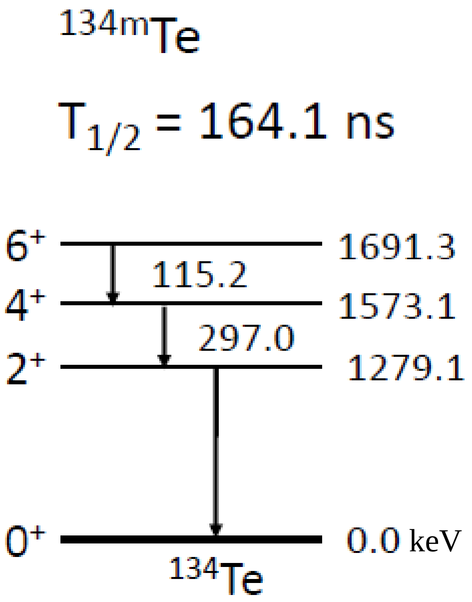
- Active interrogation of special nuclear materials
  - Non-destructive
  - Prompt and delayed radiation
- Production of ^{134m}Te and ^{136m}Xe is unique to fission
- Large fission yield: 2-6%
- Ratio dependent on identity of the fissile material

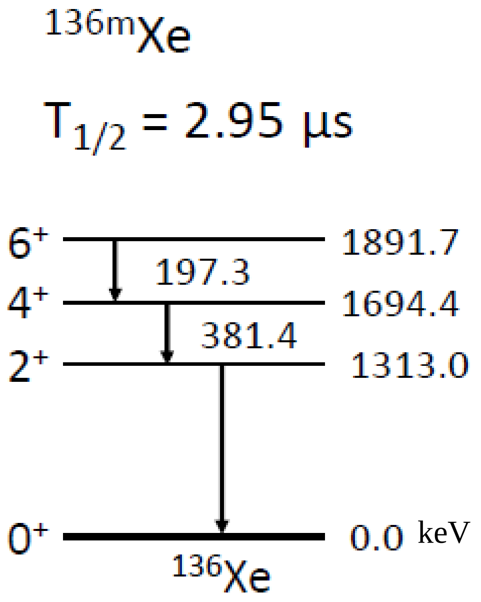


ENDF for prompt fission products

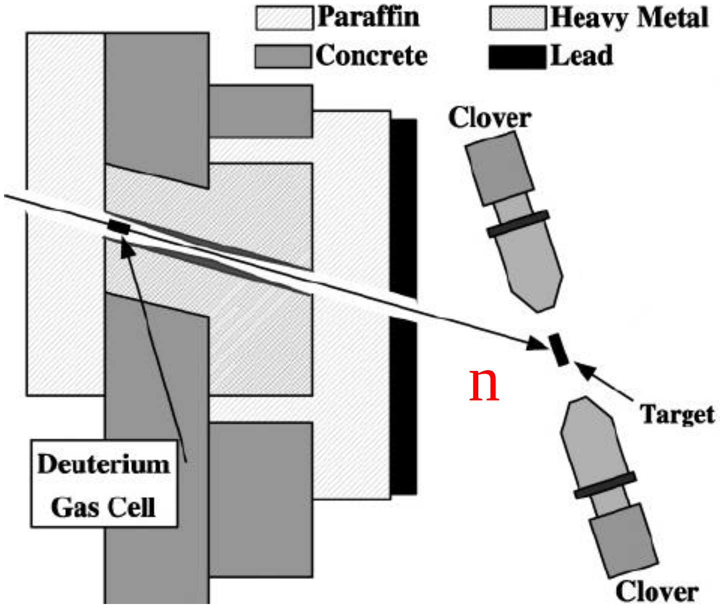
Fission Isomers

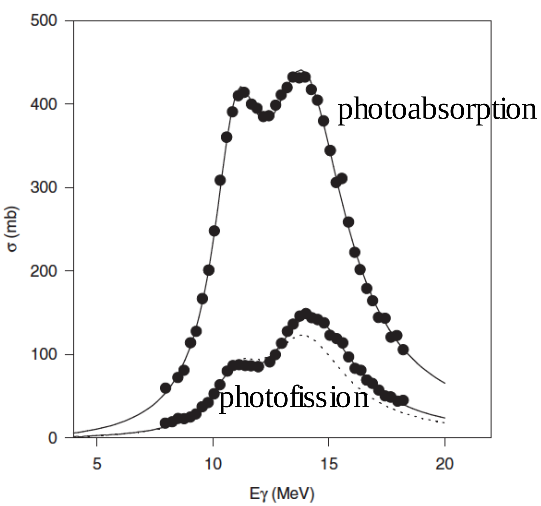
- On average 7±2 units of angular momentum are transferred to fission products
- Isomeric states are likely heavily populated
  - Fission product yield data only exists for spontaneous fission of ^{252}Cf



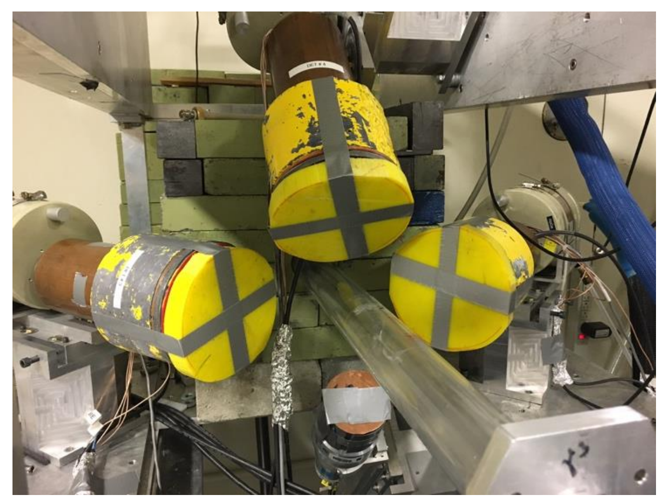


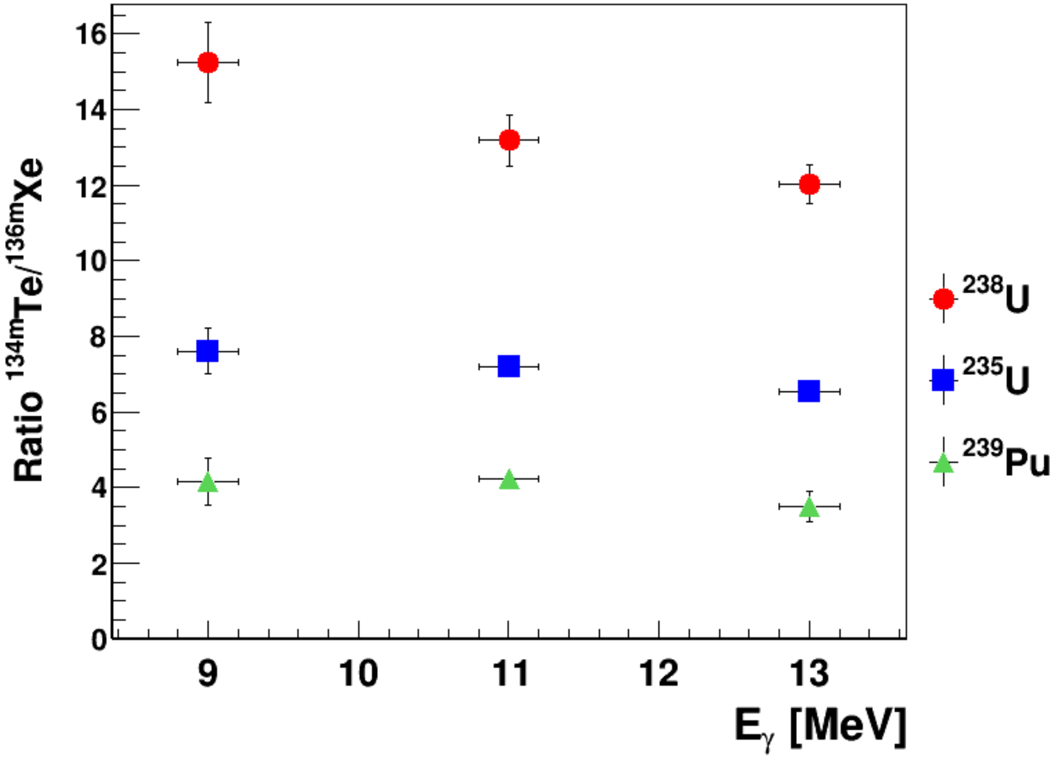
N=82 even-even closed shell nuclei

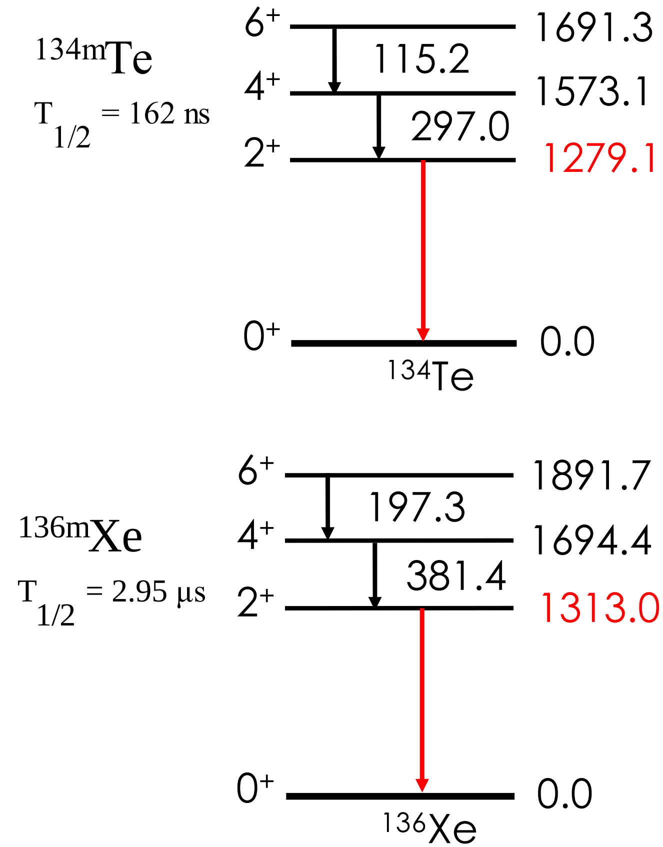


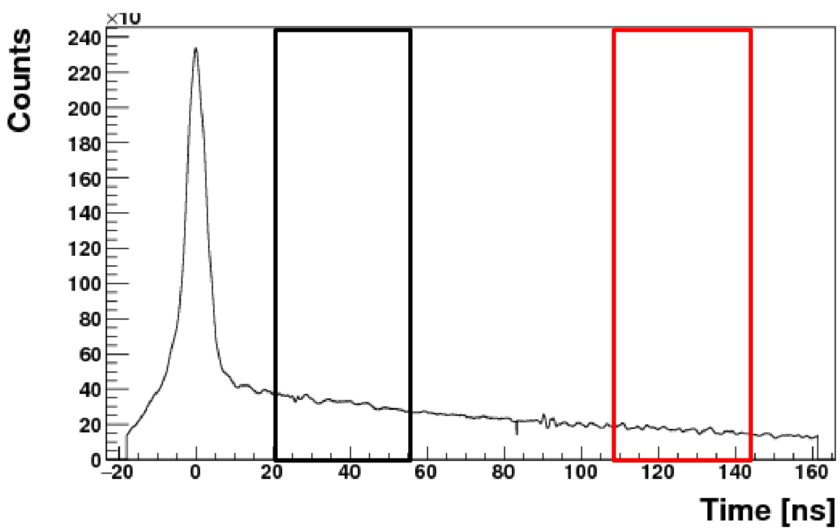


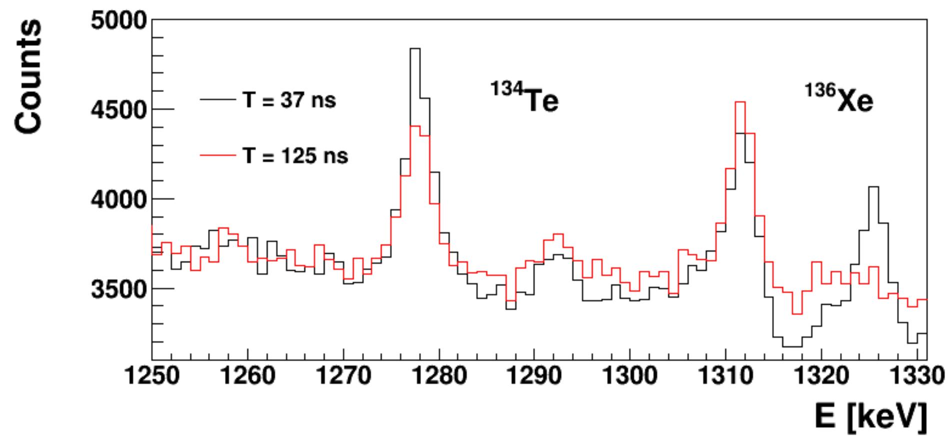
With γ rays as well
More easy!







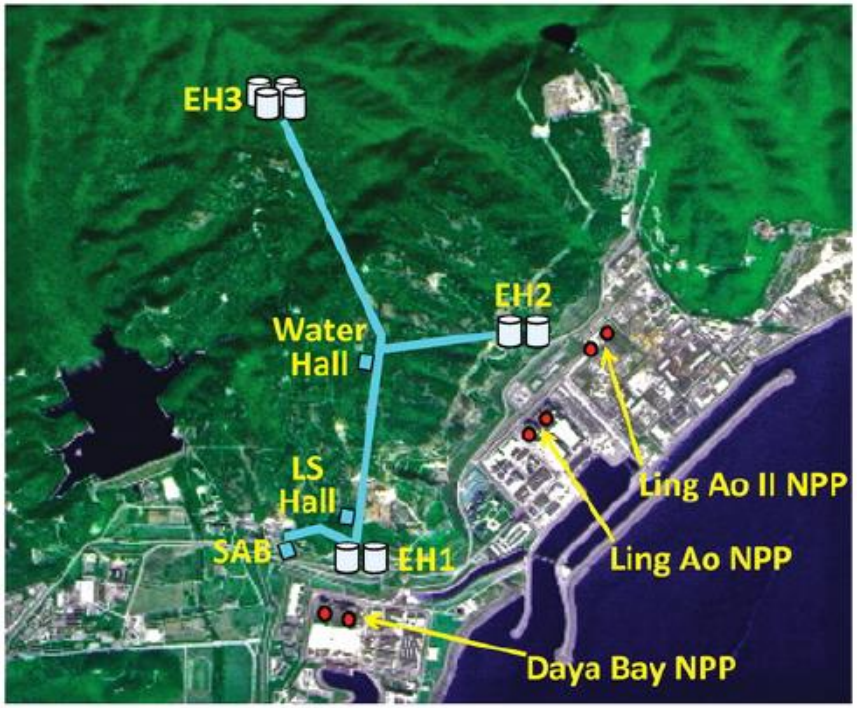




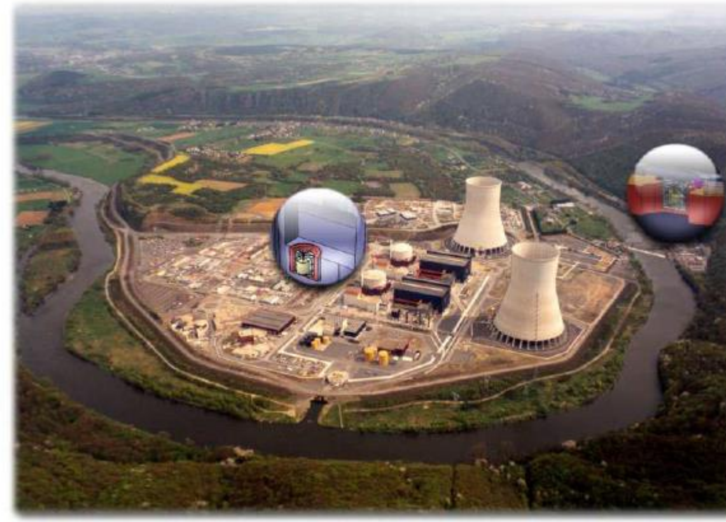
Conclusion: The isotope dependence in the ratio of the FPY of ^{134m}Te/^{136m}Xe is substantial, supporting the feasibility of using this observable to identify fissile materials with neutron beam or γ-ray beam interrogation methods

Reactor antineutrino anomaly

Daya Bay/China

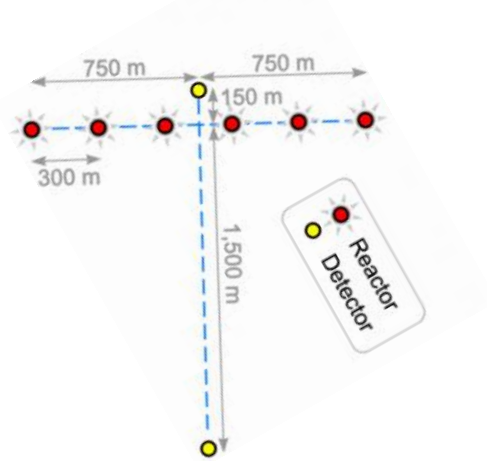


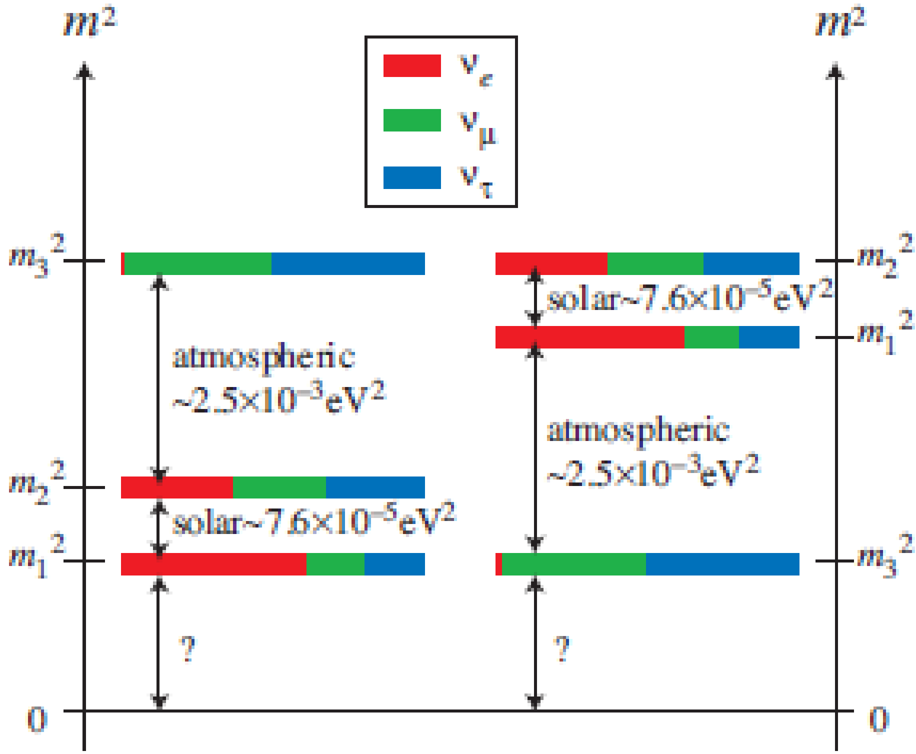
Double Chooz/France



Goal: θ_{13} mixing angle

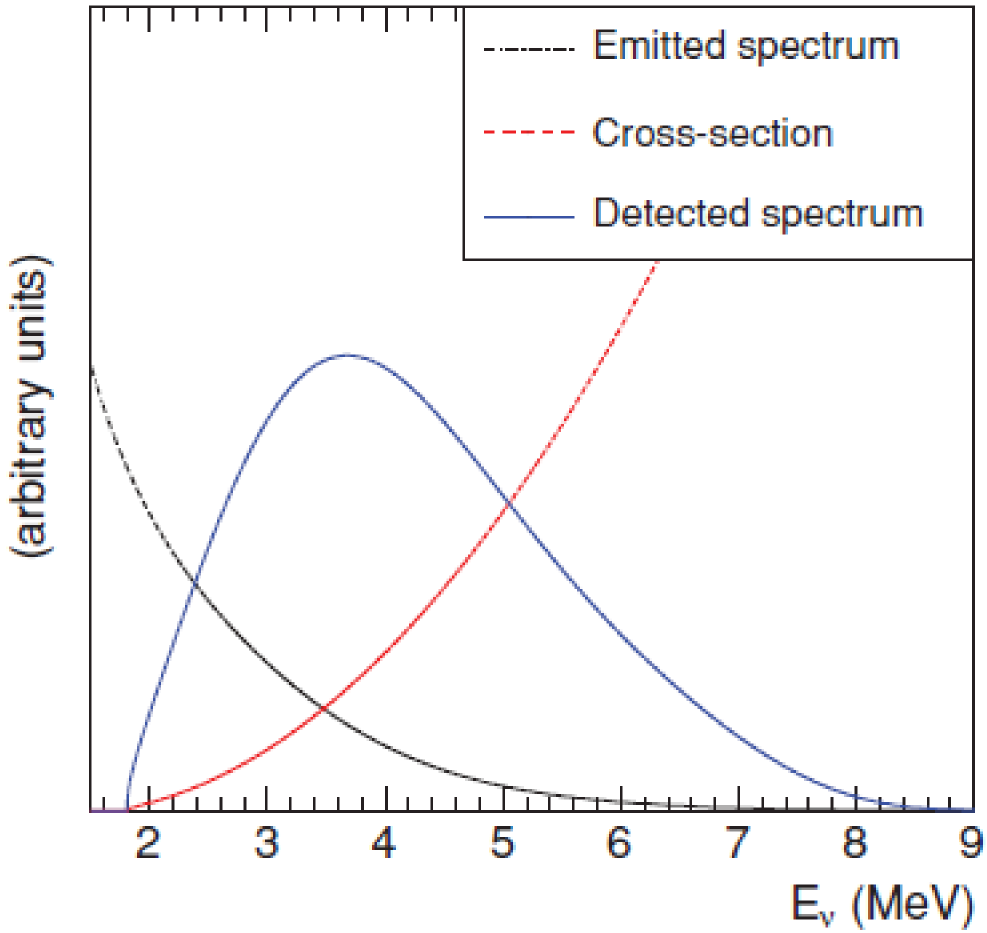
RENO/South Korea

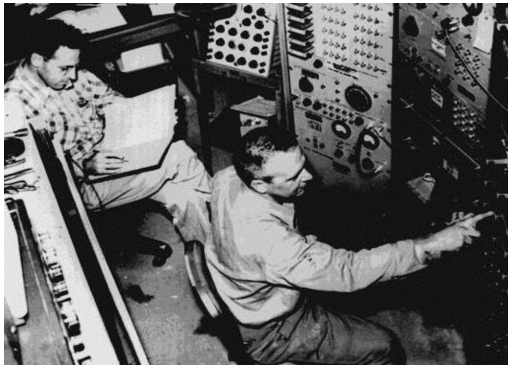




m²
m²
ν_e
ν_μ
ν_τ
m₃²
m₂²
m₁²
atmospheric
~2.5×10⁻³eV²
solar~7.6×10⁻⁵eV²
?
0
m₂²
m₁²
m₃²
solar~7.6×10⁻⁵eV²
atmospheric
~2.5×10⁻³eV²
?
0

Inverse β decay: $\bar{\nu}_e + p = e^+ + n$

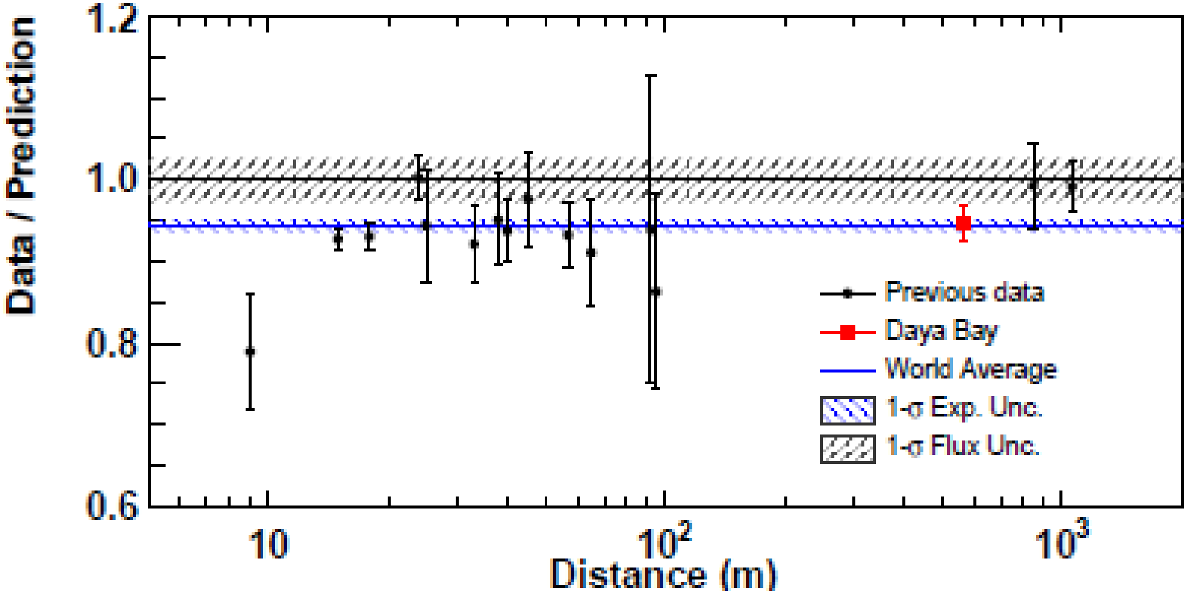




1956

First observation of neutrinos by Fred Reines and Clyde Cowan at the Savannah river reactor

Antineutrino flux and model predictions (Huber @ Mueller)

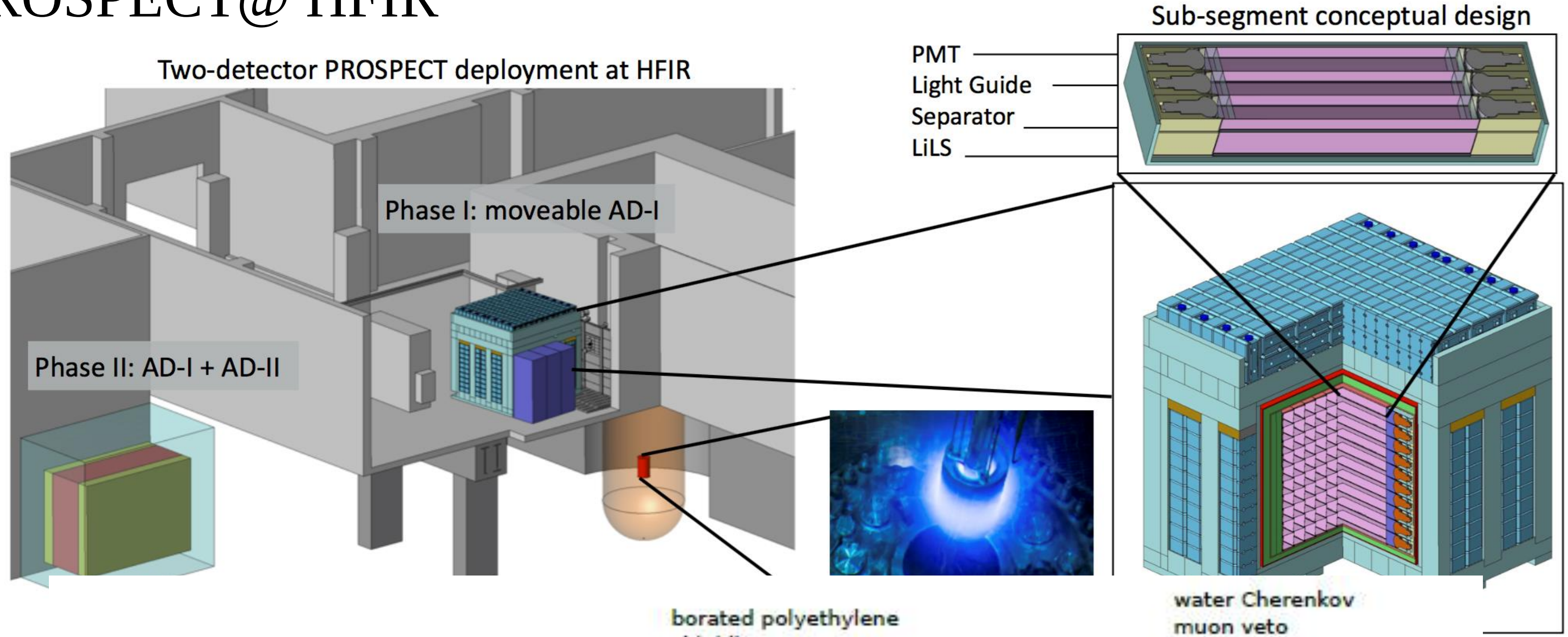


Antineutrino anomaly #1: Measured Flux ~6% too low

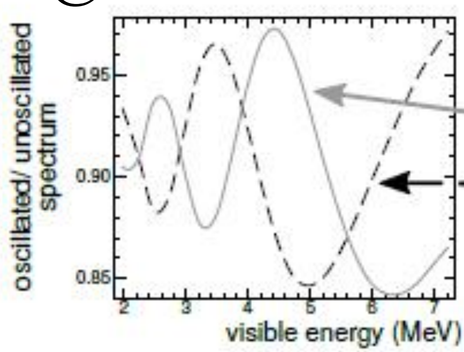
Conjecture: sterile neutrinos

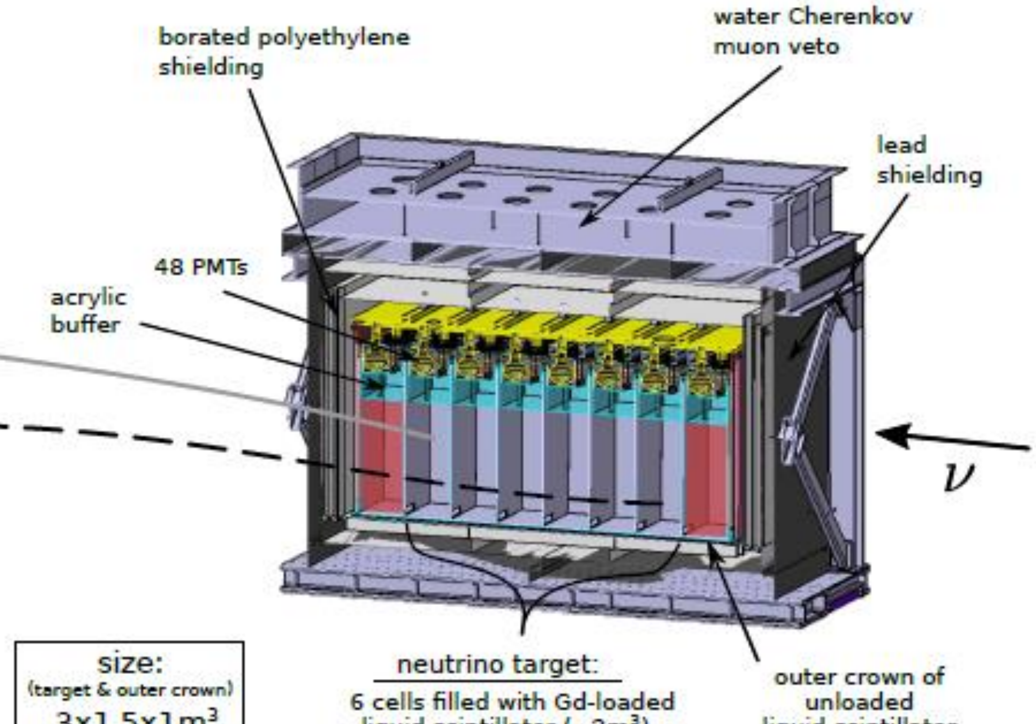
Fission product yields ????

PROSPECT@ HFIR



Stereo @ILL





Antineutrino anomaly #2:

"Bump" or "Shoulder" at 5 – 6 MeV antineutrino energy

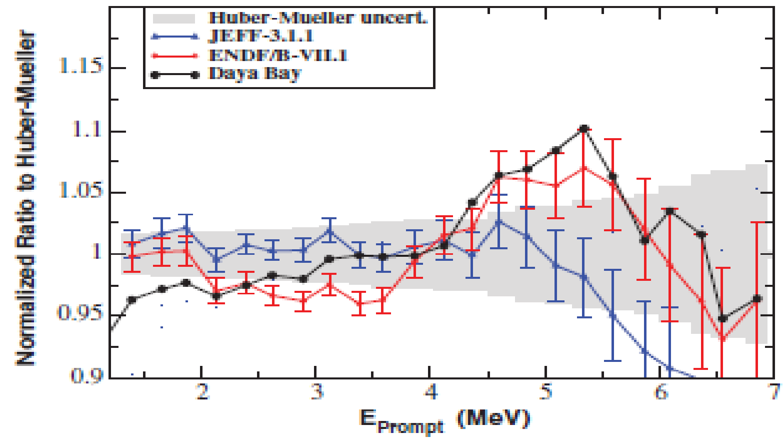
Conjecture: Fission product yield problem

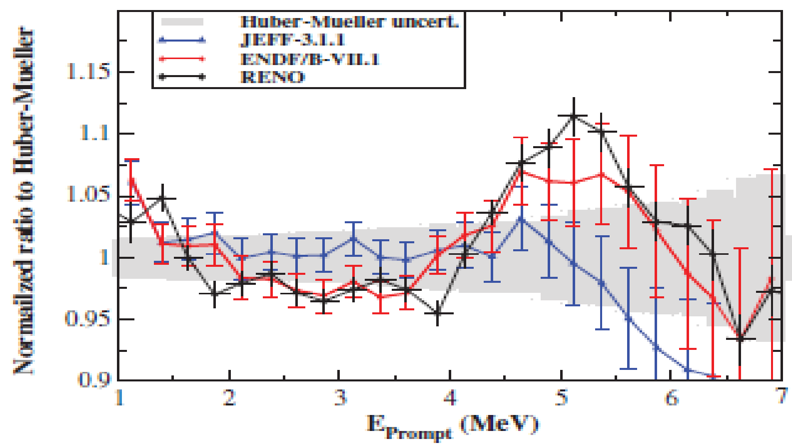
Only a few short-lived fission products contribute to shoulder (Hayes *et al*. 2015)

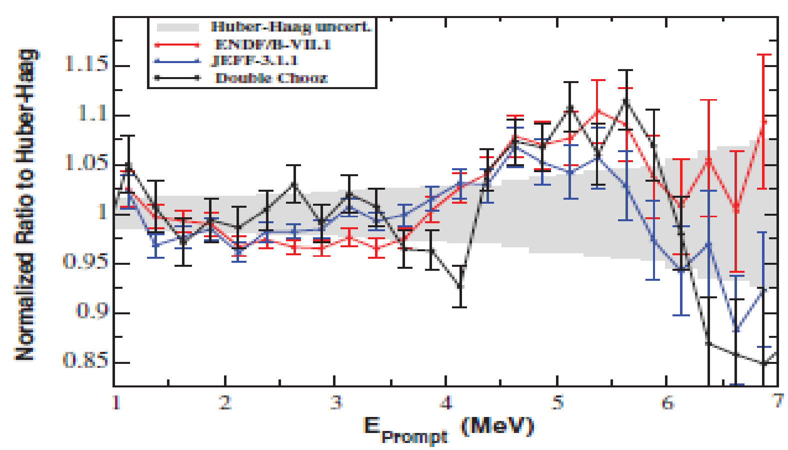
^{89,90}Br, ^{95}Rb

^{96,97,98,98m,100}Y, ^{134}Sb

Nucleus	JEFF Y_{F_i} (%)		ENDF Y_{F_i} (%)	
	^{235}U	^{239}Pu	^{235}U	^{239}Pu
^{89}Br	1.36	0.50	1.08	0.35
^{90}Br	0.49	0.10	0.56	0.25
^{95}Rb	0.66	0.26	0.77	0.44
^{96}Y	4.72	2.88	6.0	4.35
^{97}Y	2.08	1.22	4.89	3.75
^{98}Y	1.07	0.68	1.92	1.52
^{98m}Y	1.97	1.87	1.11	1.19
^{100}Y	0.30	0.21	0.61	0.35
^{134m}Sb	0.52	0.19	0.36	0.20







Is ^{238}U the culprit?
Why ^{238}U?

^{238}U fission product distribution is shifted more to neutron-rich nuclei than those for ^{235}U and ^{239}Pu:

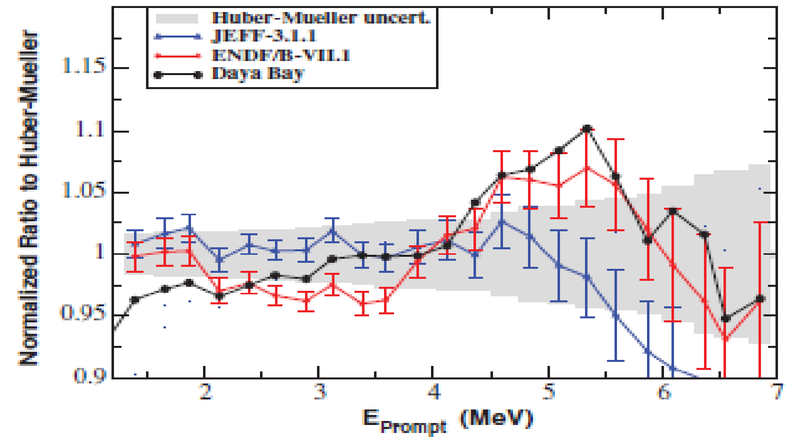
→ more antineutrinos emitted

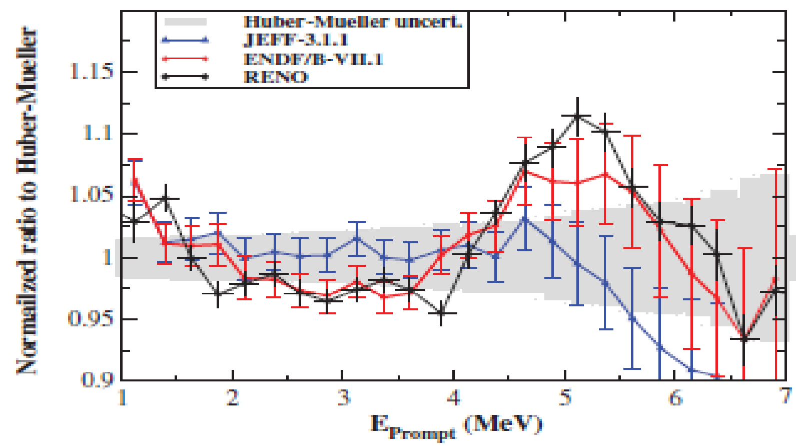
β-decay Q-values increases with neutron number:

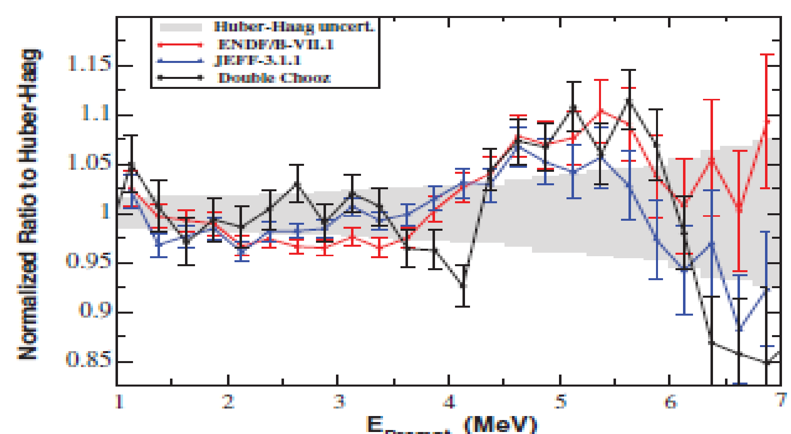
→ antineutrinos have higher energies

^{238}U β-decay energy spectra were not measured in the original work of Schreckenbach *et al*. at ILL

Huber & Mueller converted measured β-decay energy of Schreckenbach *et al*. spectra into antineutrino energy spectra





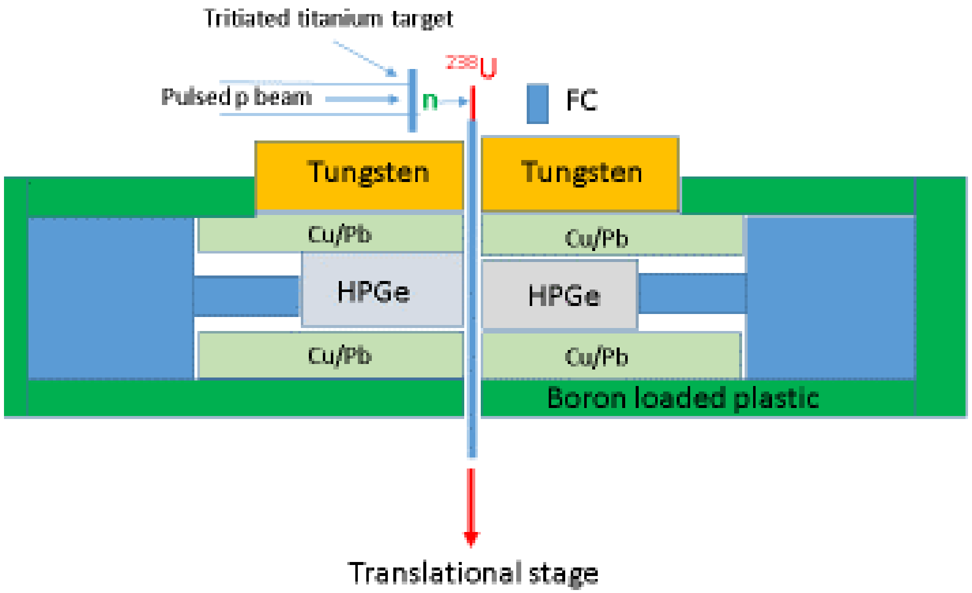


Will try to measure the short-lived fission product yields of

$^{89,90}Br$, ^{95}Rb, $^{96,97,98,98m,100}Y$, ^{134}Sb

in $^{238}U(n,f)$

Half-live times between 0.4 s and 10 s



Summary

There is a revival of Nuclear Fission research

driven by

Special Applications & Fundamental Science

Thank you!