Energy Dependence of Fission Product Yields from ^{235}U, ^{238}U and ^{239}Pu for Incident Neutron Energies Between 0.5 and 14.8 MeV

Matthew Gooden

11/4/2014

Nuclear Data Week at BNL

Collaborators

- **TUNL**
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    - B. FALLIN
    - C. HOWELL
    - **W. TORNOW**
  - NC State Univ.
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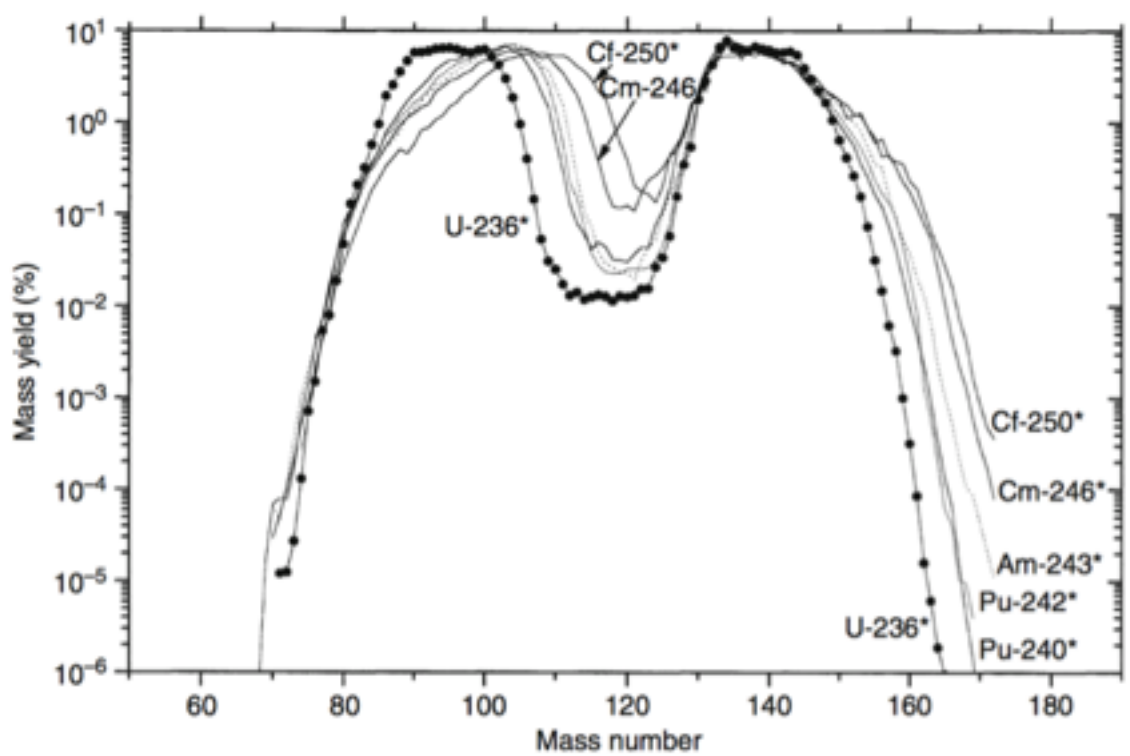
Outline

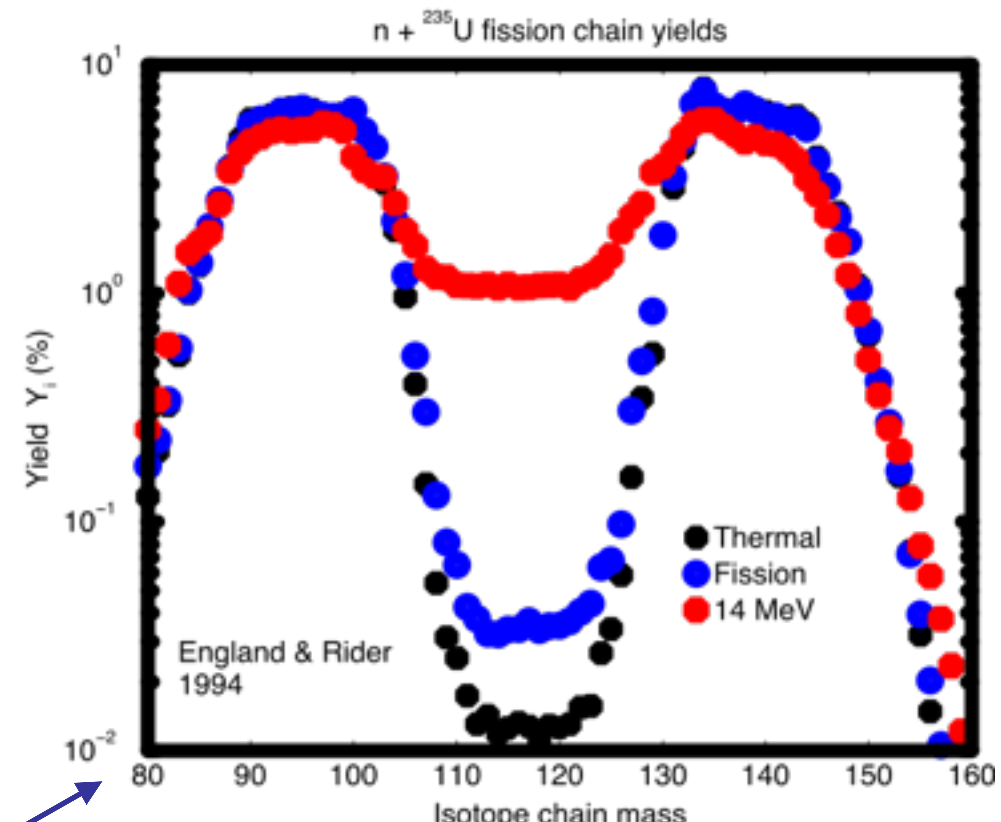
- Motivation
  - Physics
  - Application
- Experimental Design: activation at TUNL, fission chambers and γ-ray counting
- Results
- Future Work

Motivation:

- Rigidity of High Mass peak position from N=82,Z=50 and low-energy side of Low Mass peak due to N=50.



- Washed out shell structure leading to more symmetrical mass splits



- How does this asymmetry evolve with incident neutron energy for ^{235}U, ^{238}U, and ^{239}Pu on a finer scale? Since only 3 energies have been carefully investigated this is currently a challenge.

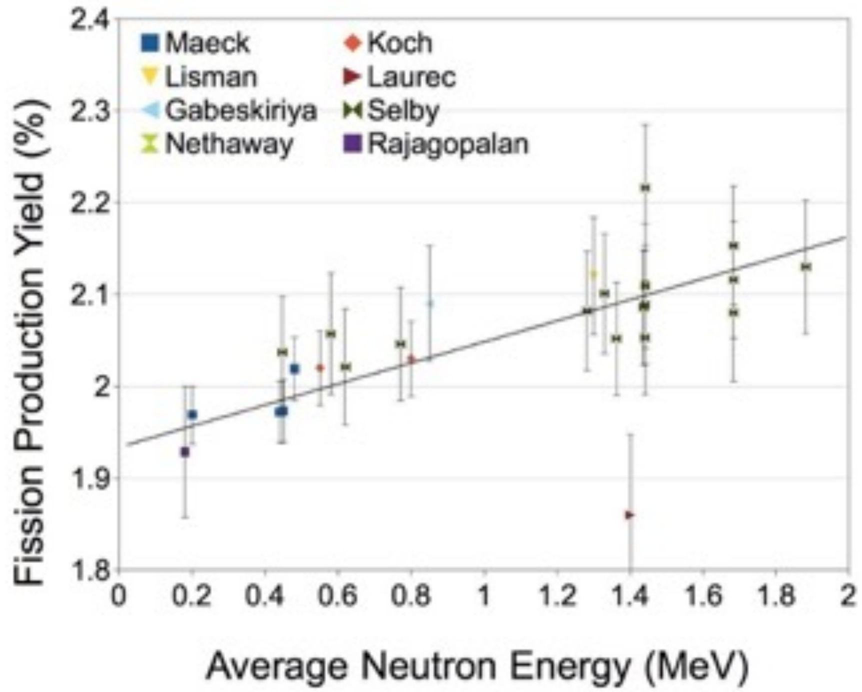
Motivation:

- Joint LANL/LLNL fission product review panel endorsed a possible energy dependence of $^{239}Pu(n,f)^{147}Nd$ fission product yield with fission neutrons:

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

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

- Low-energy data mostly from critical assemblies and fast reactors



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. Tompson et al. Nucl. Sci. Eng. **171**, 85 (2012)

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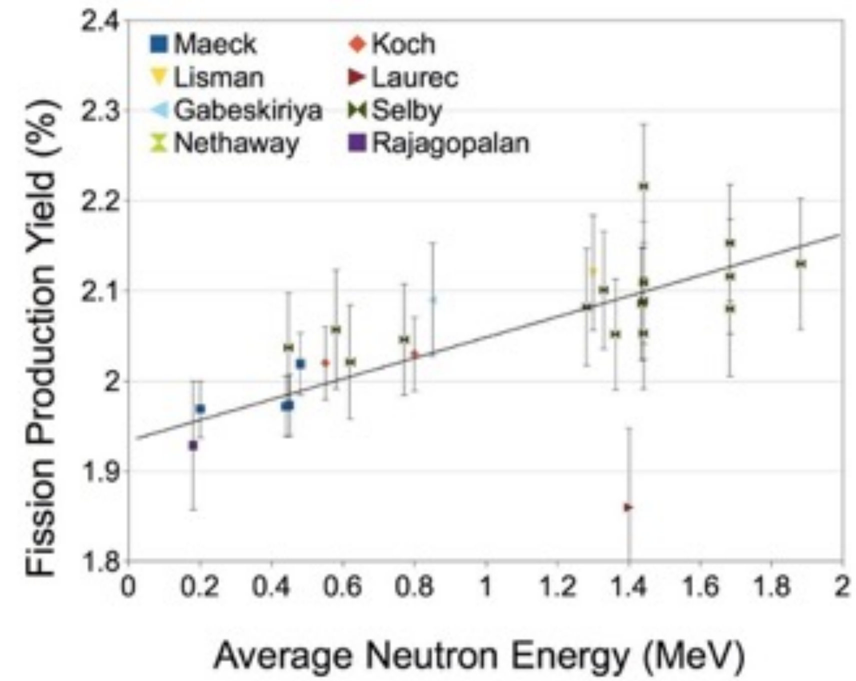
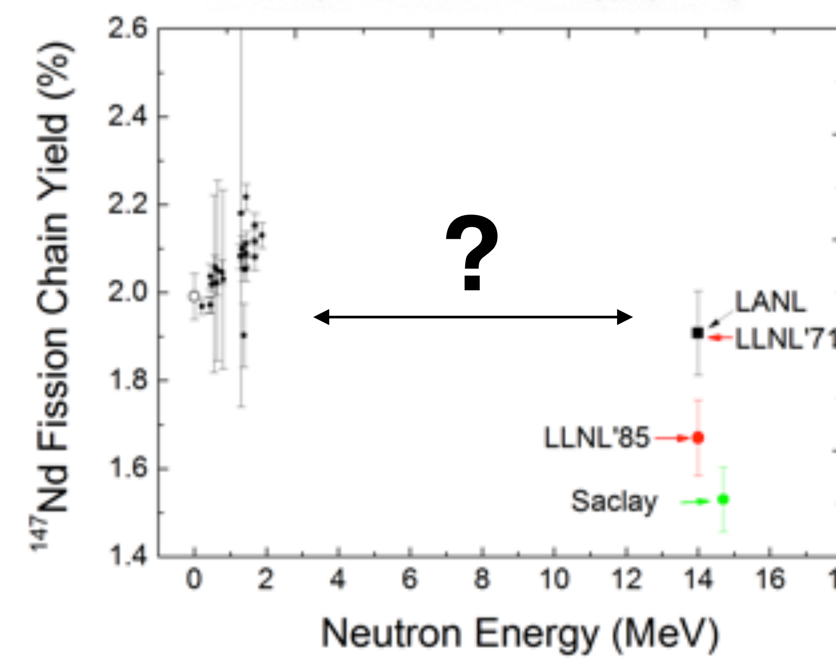
- Low-energy data mostly from critical assemblies and fast reactors

- Very scarce experimental data at the MeV-range

- Large discrepancy (~24%) at 14 MeV





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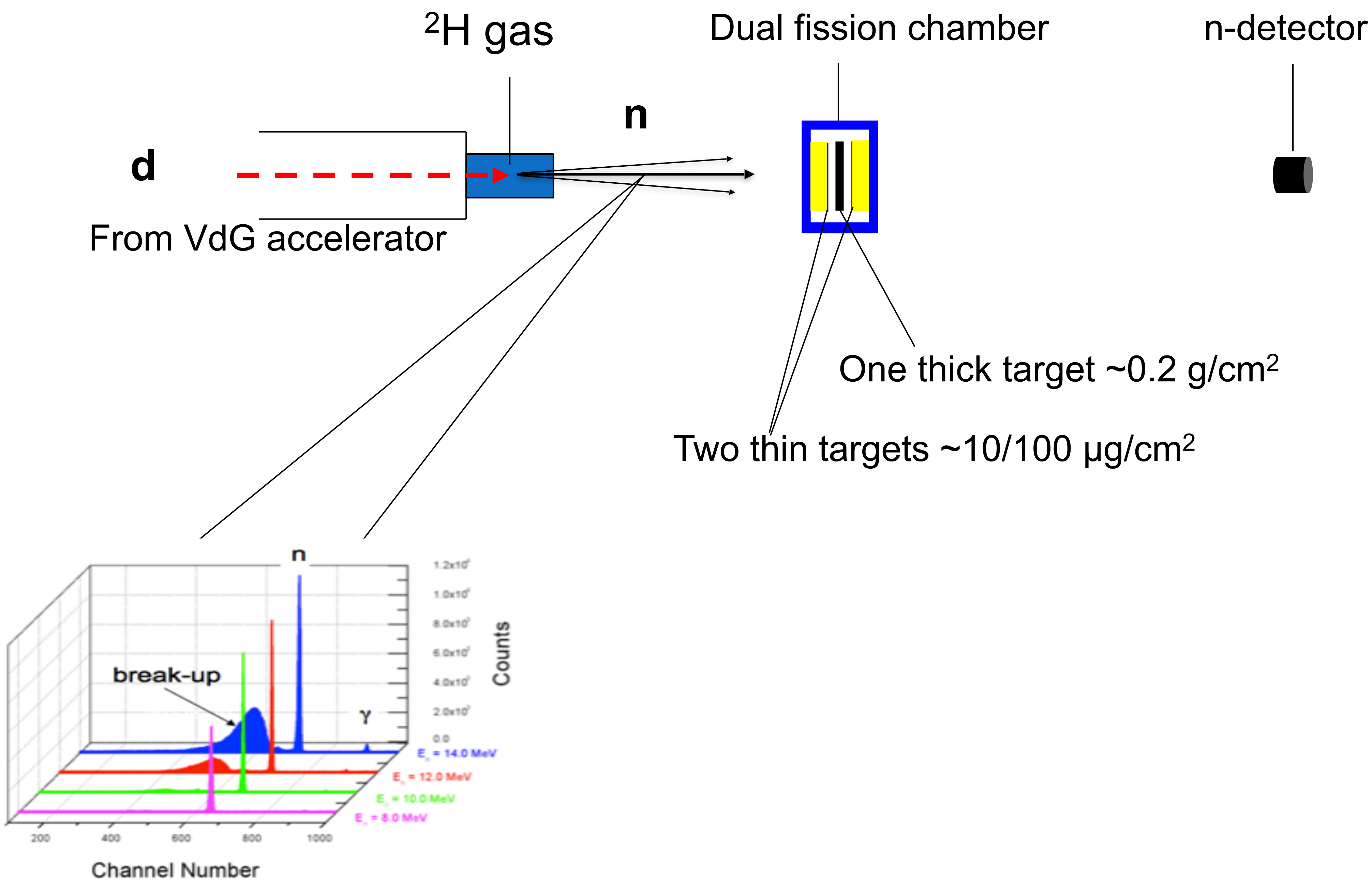
Experiment: Activation

Monoenergetic Neutron Irradiation



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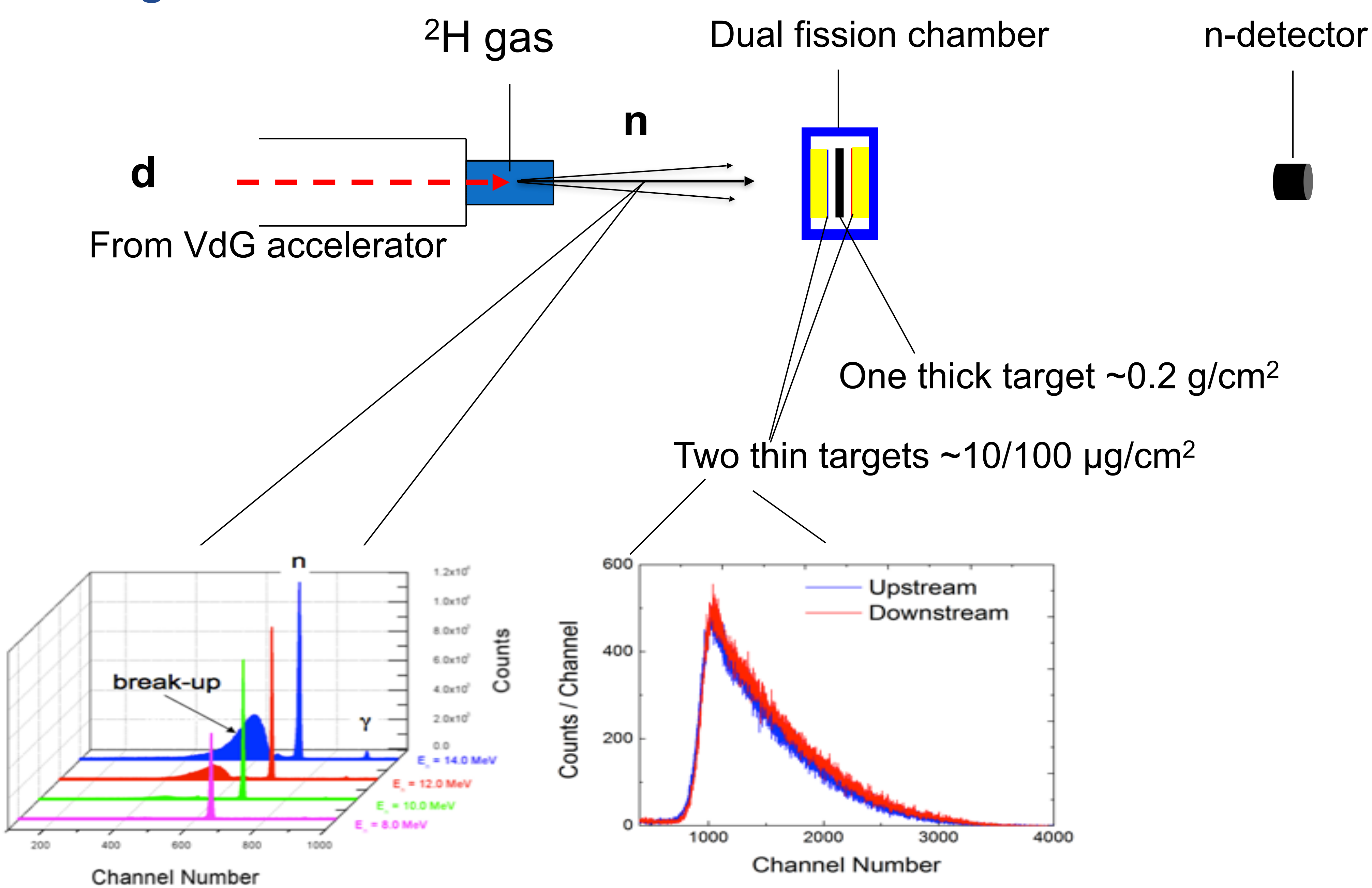

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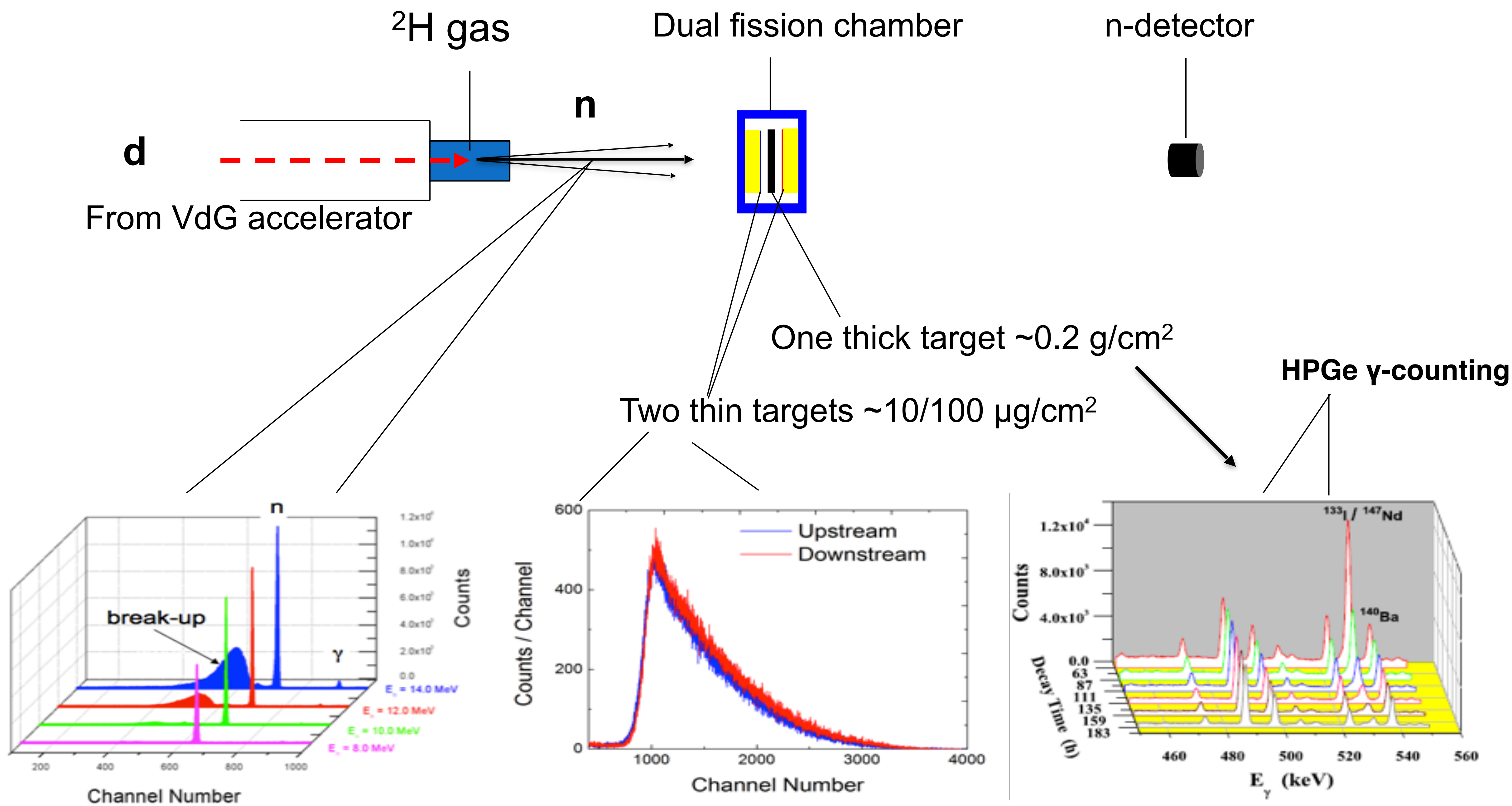
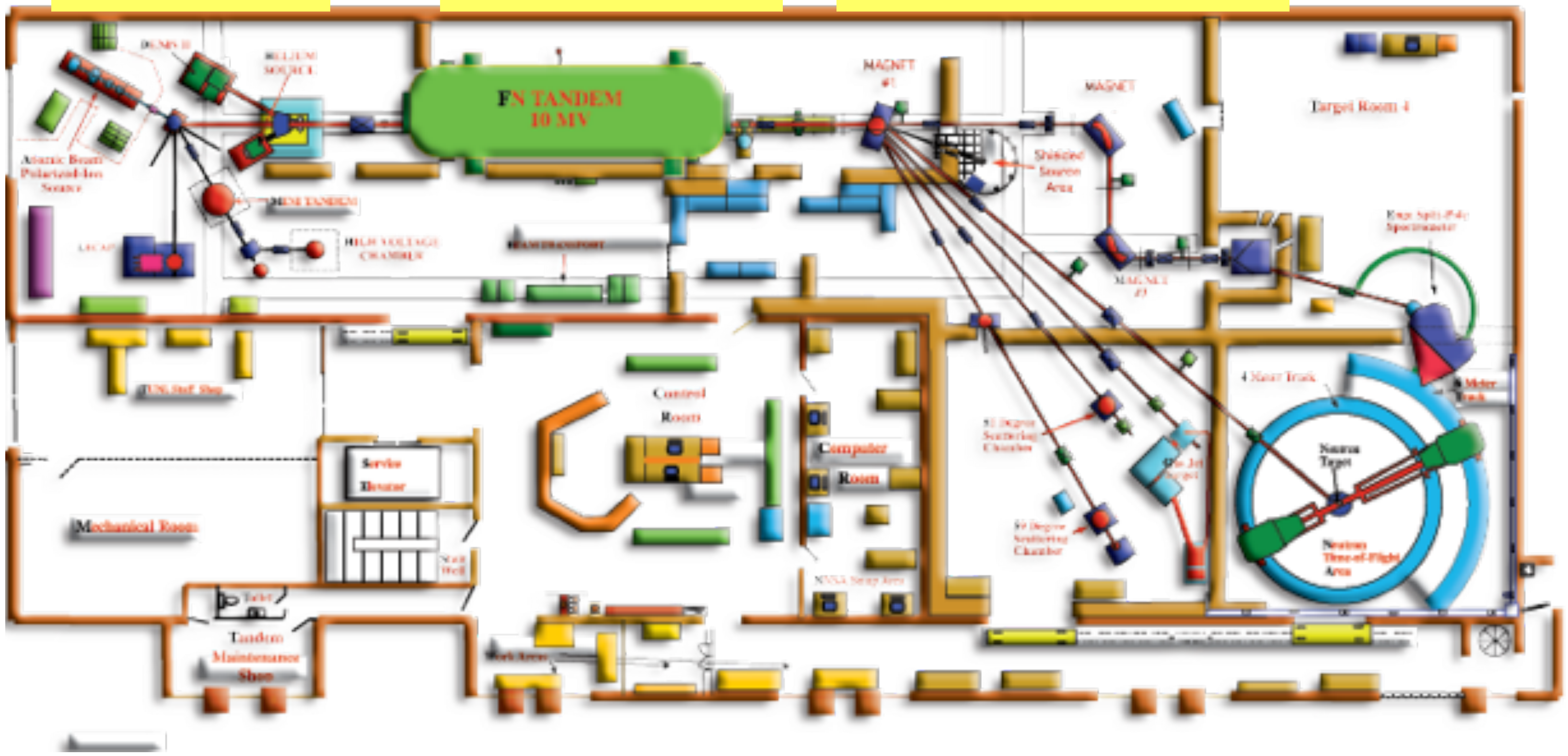


TUNL Facility

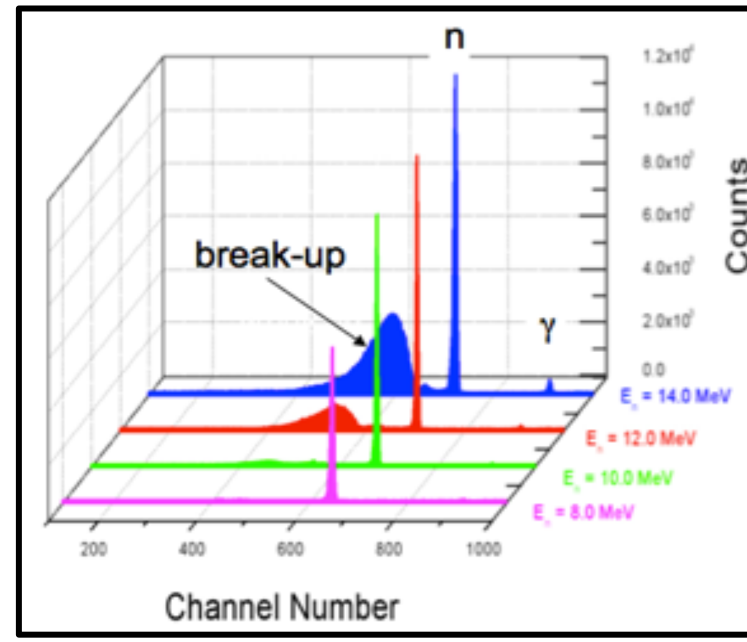
DENIS source

FN TANDEM 10MV

Shielded neutron source area



Quasi-monoenergetic neutrons



Flux on target ($10^7 - 10^8$) cm^{-2} s^{-1}

Energy spread dE/E = 0.1 to 0.40

$^7Li(p,n)^7Be$; Monoenergetic neutrons: 0.1 – 0.65 MeV

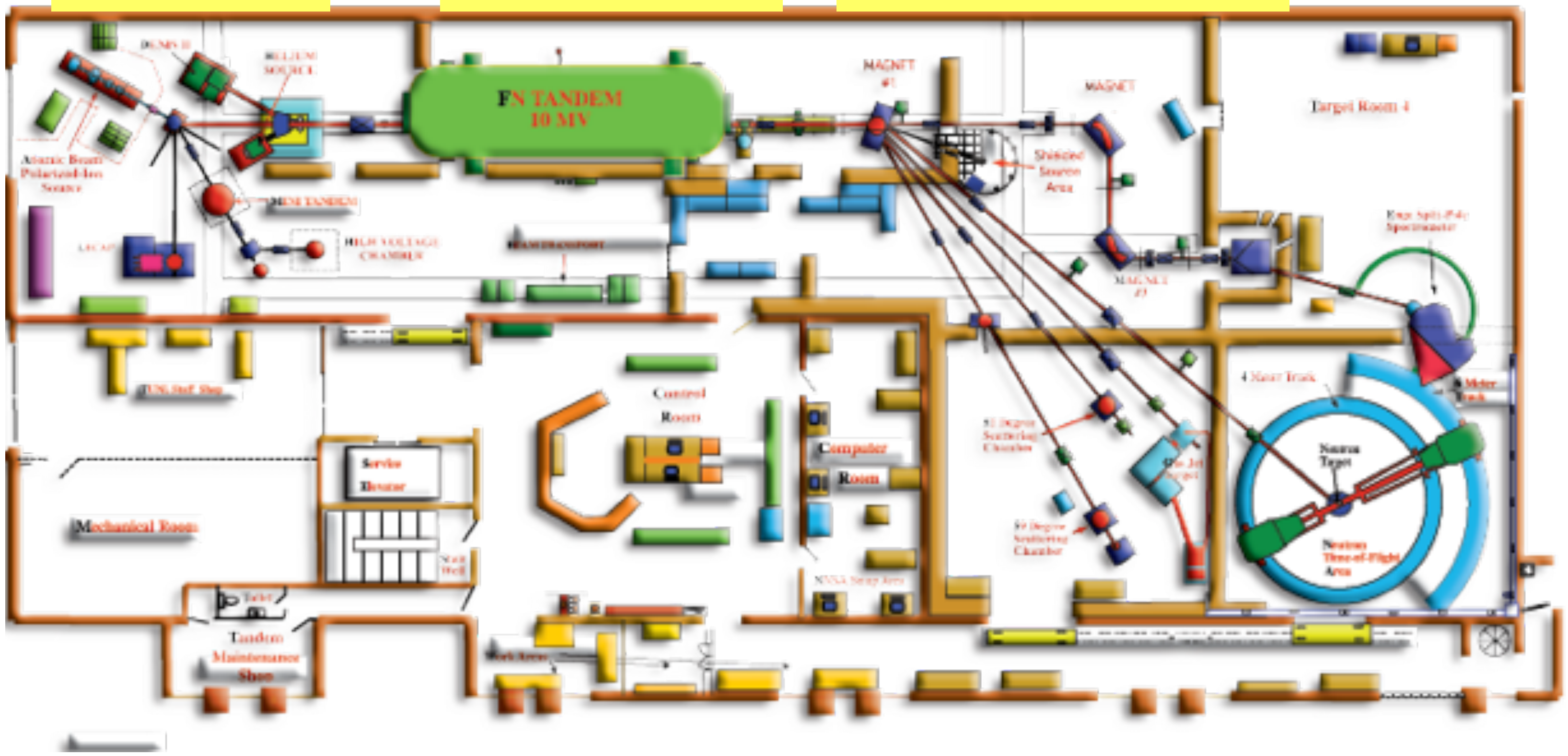
$^3H(p,n)^3He$; Monoenergetic neutrons: 0.5 – 7.7 MeV

$^2H(d,n)^3He$; Monoenergetic neutrons: 4.0 – 7.7 MeV

$^3H(d,n)^4He$; Monoenergetic neutrons: 14.8 – 20.5 MeV

TUNL Facility

DENIS source FN TANDEM 10MV Shielded neutron source area



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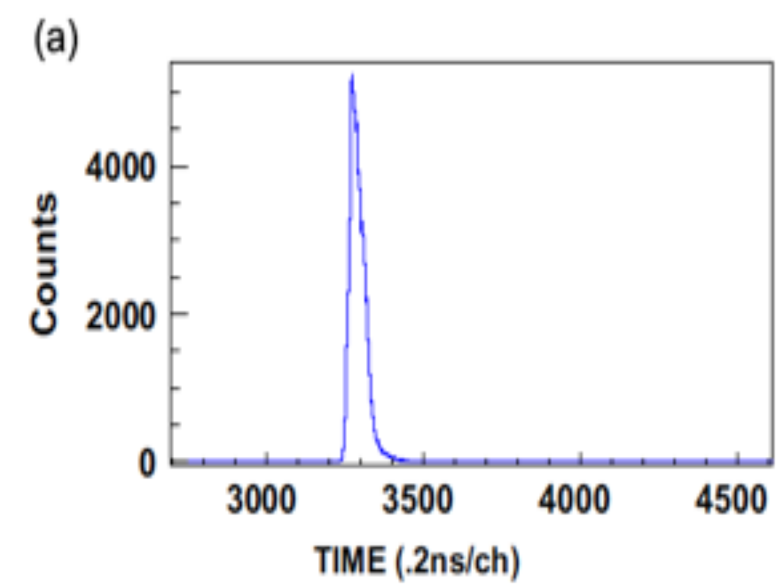

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$^{2}H(d,n)^{3}He$; Monoenergetic neutrons: 4.0 – 7.7 MeV

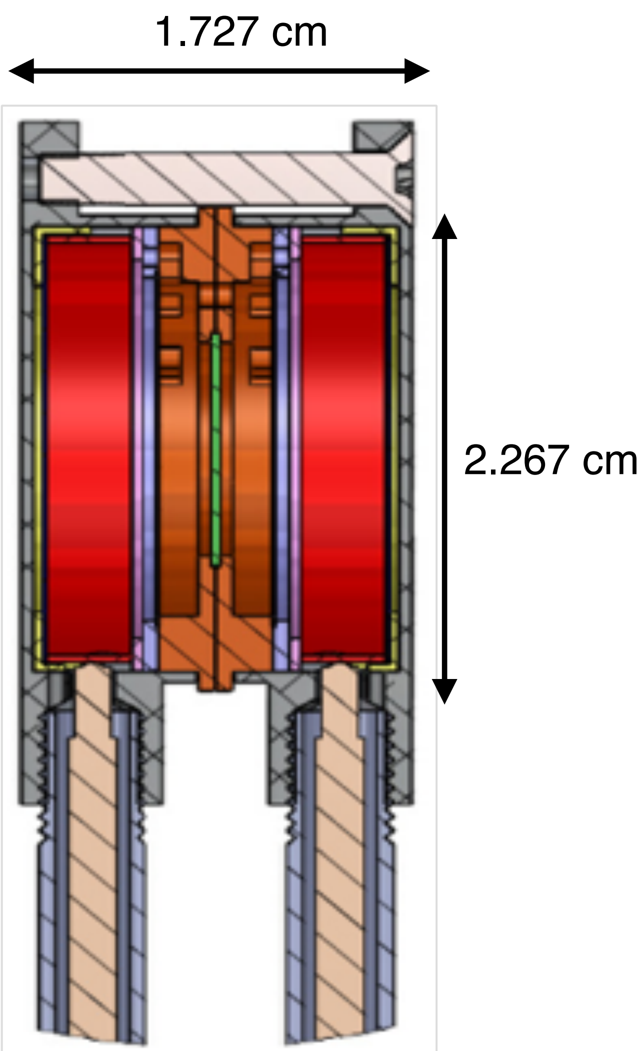
$^{3}H(d,n)^{4}He$; Monoenergetic neutrons: 14.8 – 20.5 MeV

Flux on target $6x10^{6}$ cm^{-2} s^{-1}

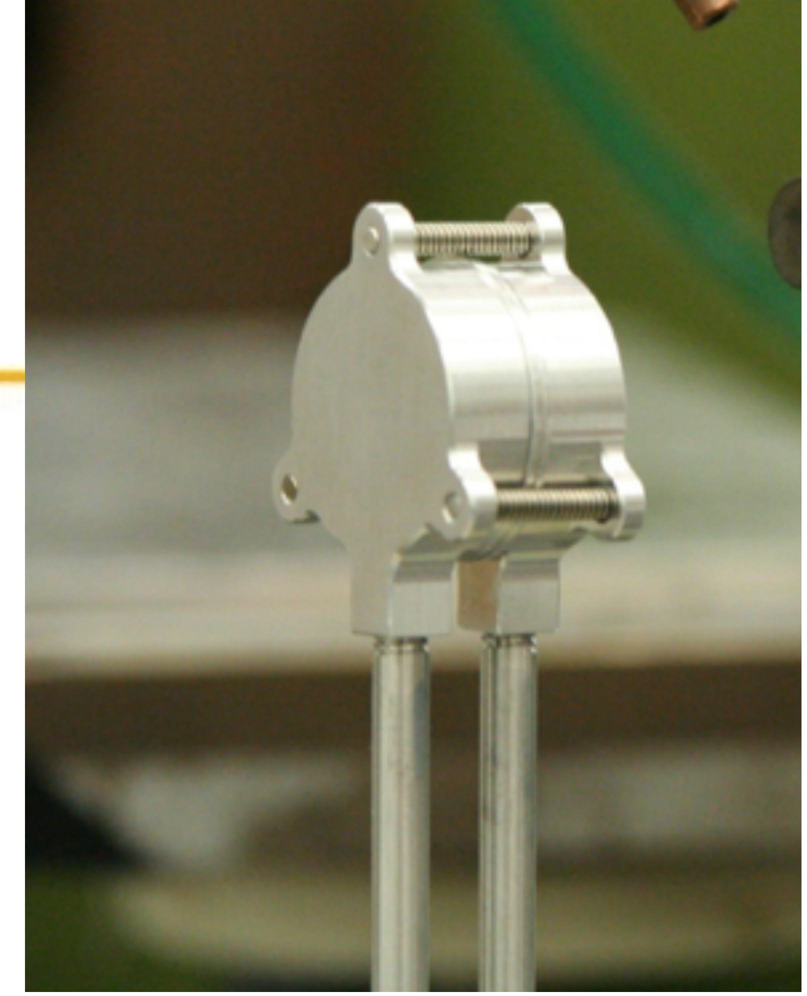
Energy spread = 120 keV

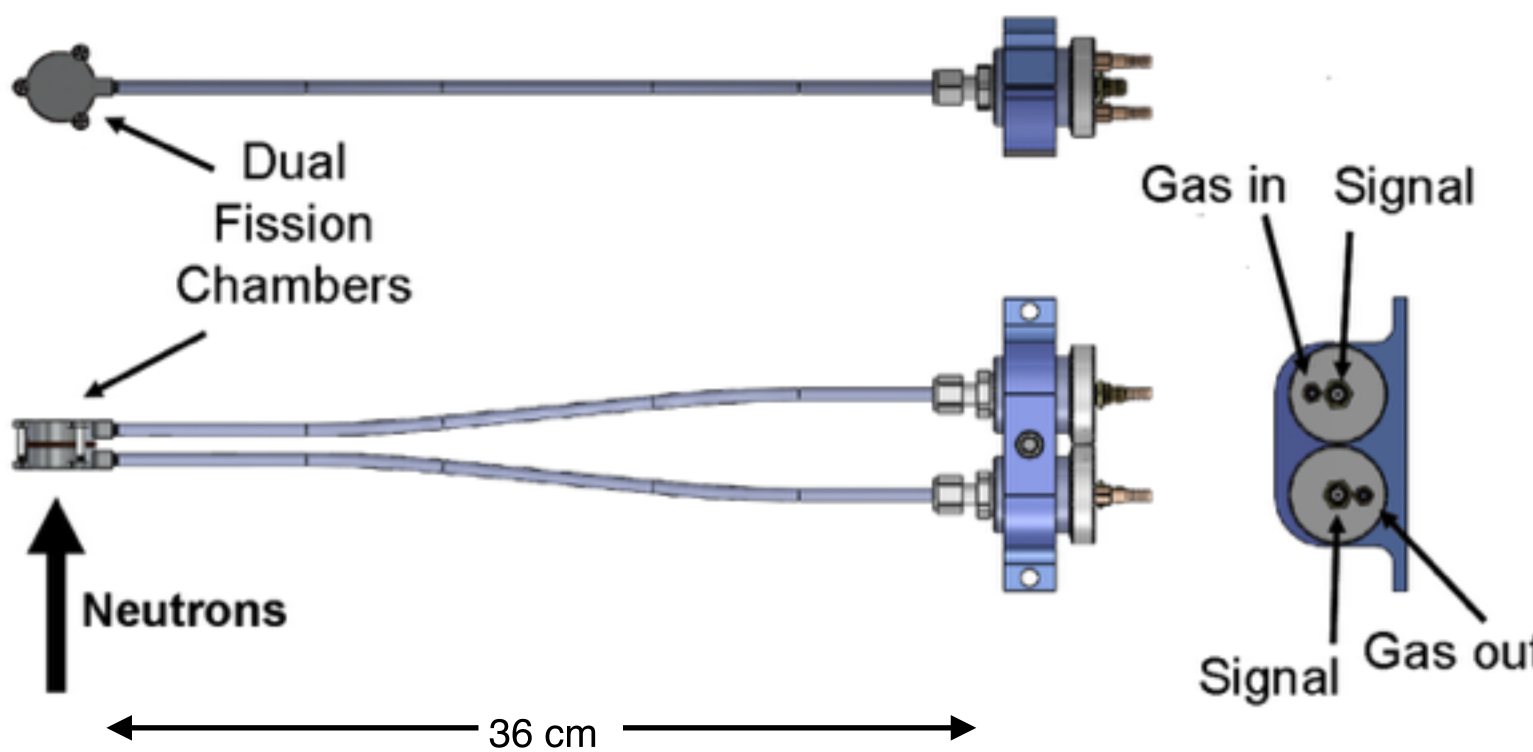
Fission Chambers

- Continuous flow of P-10 as a counting gas
- Signals and gas carried in stainless steel ‘arms’

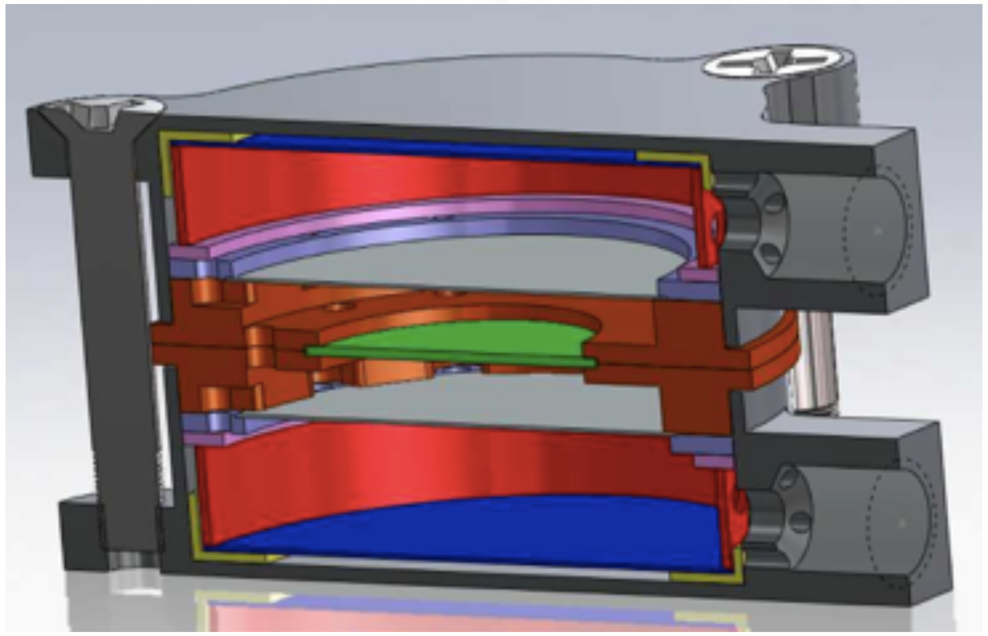


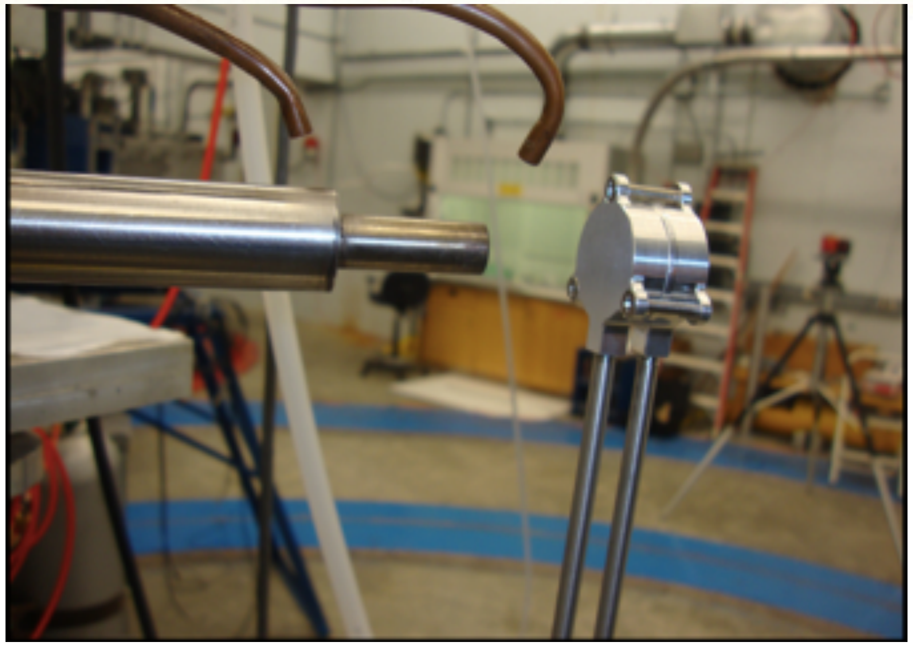
- **Chamber Active Volume**
  4.623 mm depth
  22.098 mm diameter
  = 1.773 cm^3





Fission Chambers



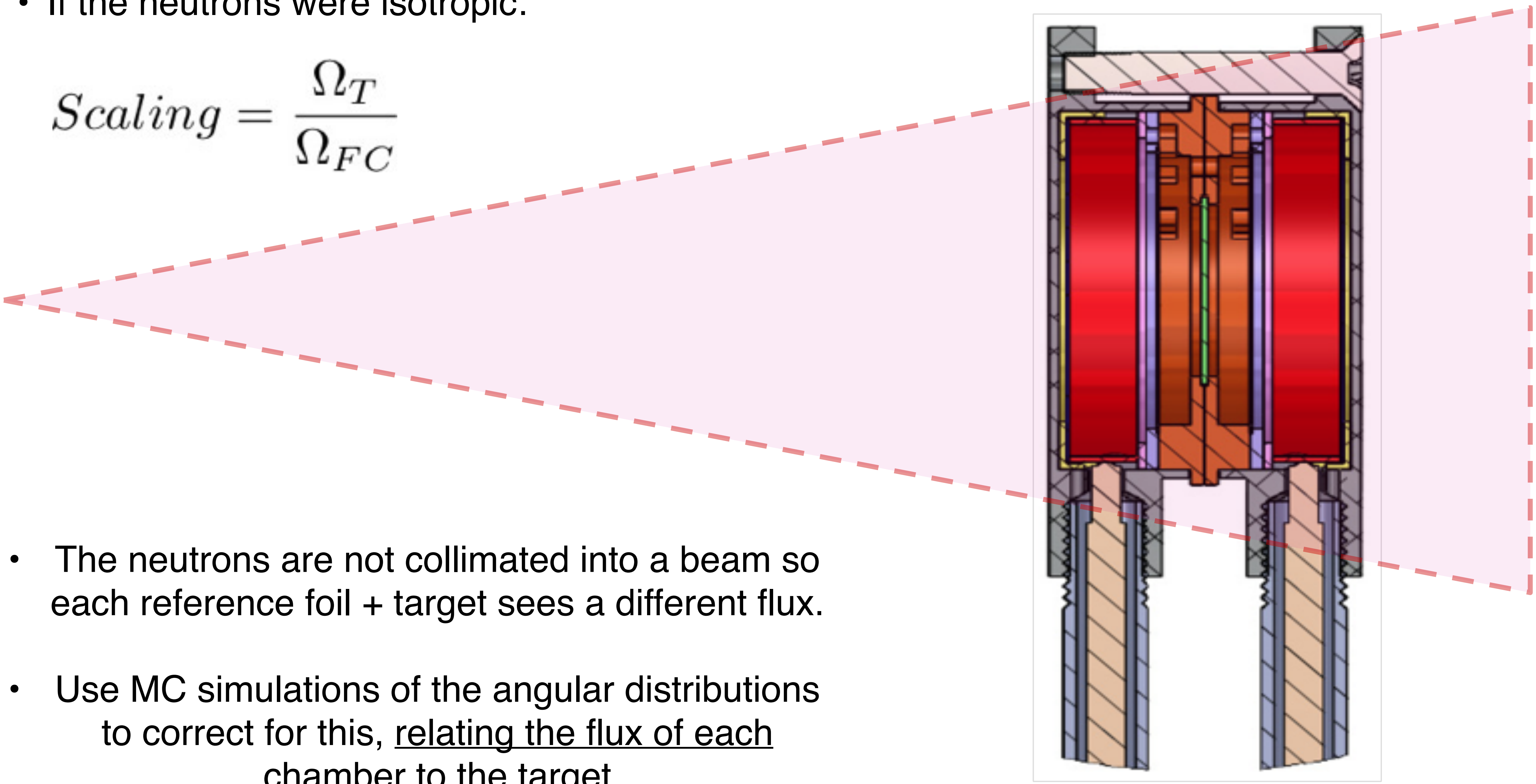


- 3 dual-fission chambers each dedicated to a target isotope: ^{235}U, ^{238}U or ^{239}Pu based on design of Grundl and Gilliam
- Dedicated thin ^{235}U, ^{238}U (~100 μg/cm^2) and ^{239}Pu (~10 μg/cm^2) foils electroplated on 0.5`` titanium backing for each respective chamber
- Dedicated thick (200-400 mg/cm^2) ^{235}U(93.27%), ^{238}U(99.97%) and ^{239}Pu(98.4%) targets

Fission Chambers

- If the neutrons were isotropic:

$$Scaling = \frac{\Omega_T}{\Omega_{FC}}$$



- The neutrons are not collimated into a beam so each reference foil + target sees a different flux.
- Use MC simulations of the angular distributions to correct for this, relating the flux of each chamber to the target

γ-Ray Counting

- γ-ray counting with shielded 60% HPGe detectors using the GENIE daq system with pile-up rejection
- Calibrated using mixed nuclide source to avoid issues with coincidence summing
    - Targets were counted at various distances from 1cm to 5 cm

FPY Calculation

$$Fissions = M_t\ (\phi \cdot \sigma_{n,f})\ \epsilon_f\ t_f$$

$$N_\gamma = M_T\ (\phi \cdot \sigma_{n,f})\ FPY\ I_\gamma\ \epsilon_\gamma\ t_\gamma$$

$M_T(M_t)$ = atoms in the ^{239}Pu thick (thin) target

ϕ = neutron flux ($n.cm^{-2}.s^{-1}$)

$\sigma_{n,f}$ = ^{239}Pu(n,f) fission cross section (cm^2)

FPY = fission product yield of ^{147}Nd per ^{239}Pu fission

I_γ = branching ratio of E_γ

$\epsilon_\gamma(\epsilon_f)$ = counter efficiency of γ-ray (fission) detection

$t_\gamma(t_f)$ = time factor for irradiation and counting periods of γ-ray (fission)

$$\longrightarrow \qquad FPY = \frac{M_t}{M_T}\ \frac{N_\gamma}{Fissions}\ C$$

C = all other factors not explicitly shown (BR, efficiencies, time factors, etc.)

What we have done

Energy	Target	Date	Beam-Time
0.58 MeV	U-235	December 2013	96.5 h
	Pu-239	November 2013	186.5 h
1.37 MeV	U-235	December 2012	89.0 h
	U-238	December 2012	117.8 h
	Pu-239	March 2013	134.1 h
2.37 MeV	U-235	July 2013	54.8 h
	U-238	September 2013	138.1 h
	Pu-239	September 2013	93.5 h
4.52 MeV	U-235	February 2014	91.7 h
	U-238	April 2014	123.6 h
	Pu-239	May 2014	131.5 h
14.8 MeV	U-235	May 2013	142.0 h
	U-238	May 2013	148.6 h
	Pu-239	Feb 2013/Mar 2014	108/228.8 h
		TOTAL =	1885 h

- 5 energies - 15* measurements
- 1885 hours of beam time - almost 80 days!
- Each measurement included ~ 2 months of γ-ray counting
- Just completed another 240 hours of continuous beam time

Fission Products

- ~15 fission fragments have been identified through γ-ray counting

^{91}Sr, ^{92}Sr, ^{95}Zr, ^{97}Zr, ^{99}Mo, ^{103,105}Ru, ^{127}Sb, ^{131,133}I, ^{132}Te, ^{135}Xe, ^{136}Cs, ^{140}Ba, ^{143}Ce and ^{147}Nd

- Half-lives ranging from 2 hrs to >60 days
- Not all FPs have been measured for each target at each energy

Fission Products

- ^{147}Nd is an important reference isotope and there is a major focus
    - A possibly pronounced energy dependence at low-energy

^{91}Sr, ^{92}Sr, ^{95}Zr, ^{97}Zr, ^{99}Mo, ^{103,105}Ru, ^{127}Sb, ^{131,133}I, ^{132}Te, ^{135}Xe, ^{136}Cs, ^{140}Ba, ^{143}Ce and ^{147}Nd

- Determined a single independent yield for the fission product ^{136}Cs

Fission Products

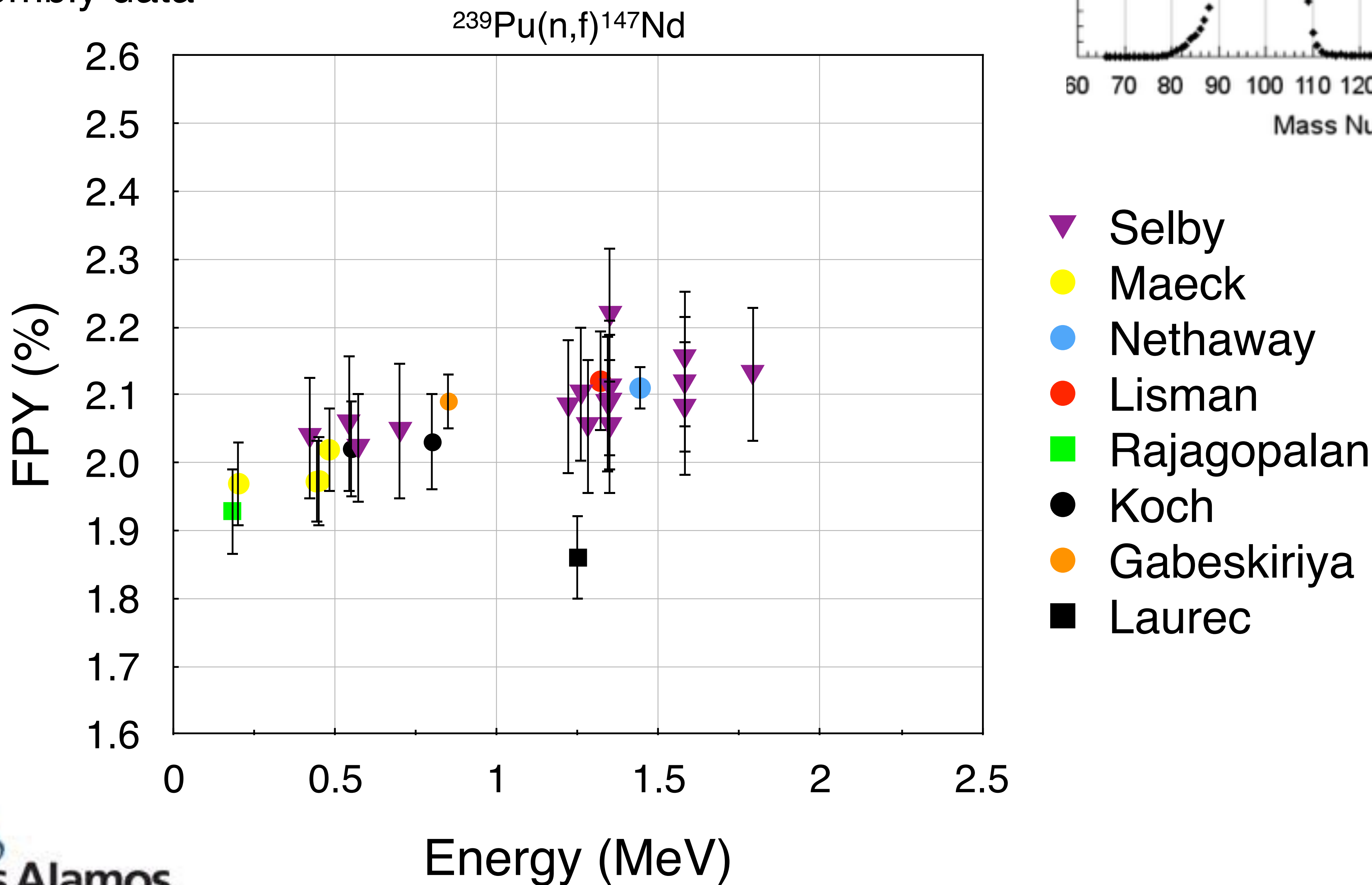
- ^{140}Ba and ^{95}Zr have long histories as important fission calibration references since they showed little-to-no energy dependence
- ^{99}Mo important due to its use as a basis in FPY studies - FPYs built on a '^{99}Mo basis'

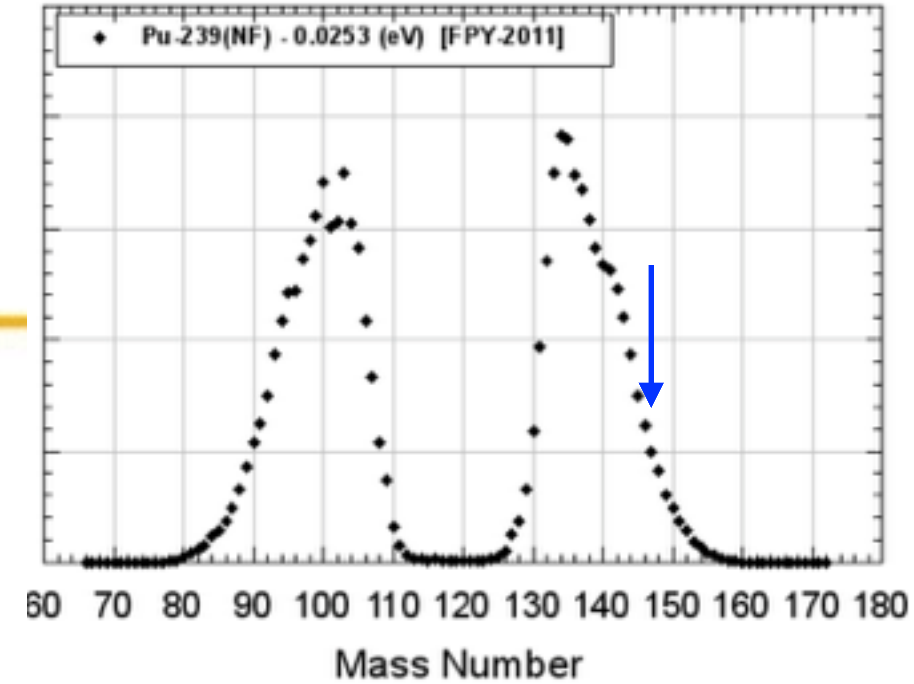
^{91}Sr, ^{92}Sr, ^{95}Zr, ^{97}Zr, ^{99}Mo, ^{103,105}Ru, ^{127}Sb, ^{131,133}I, ^{132}Te, ^{135}Xe, ^{136}Cs, ^{140}Ba, ^{143}Ce and ^{147}Nd

Experimental Results

Results: ^{147}Nd

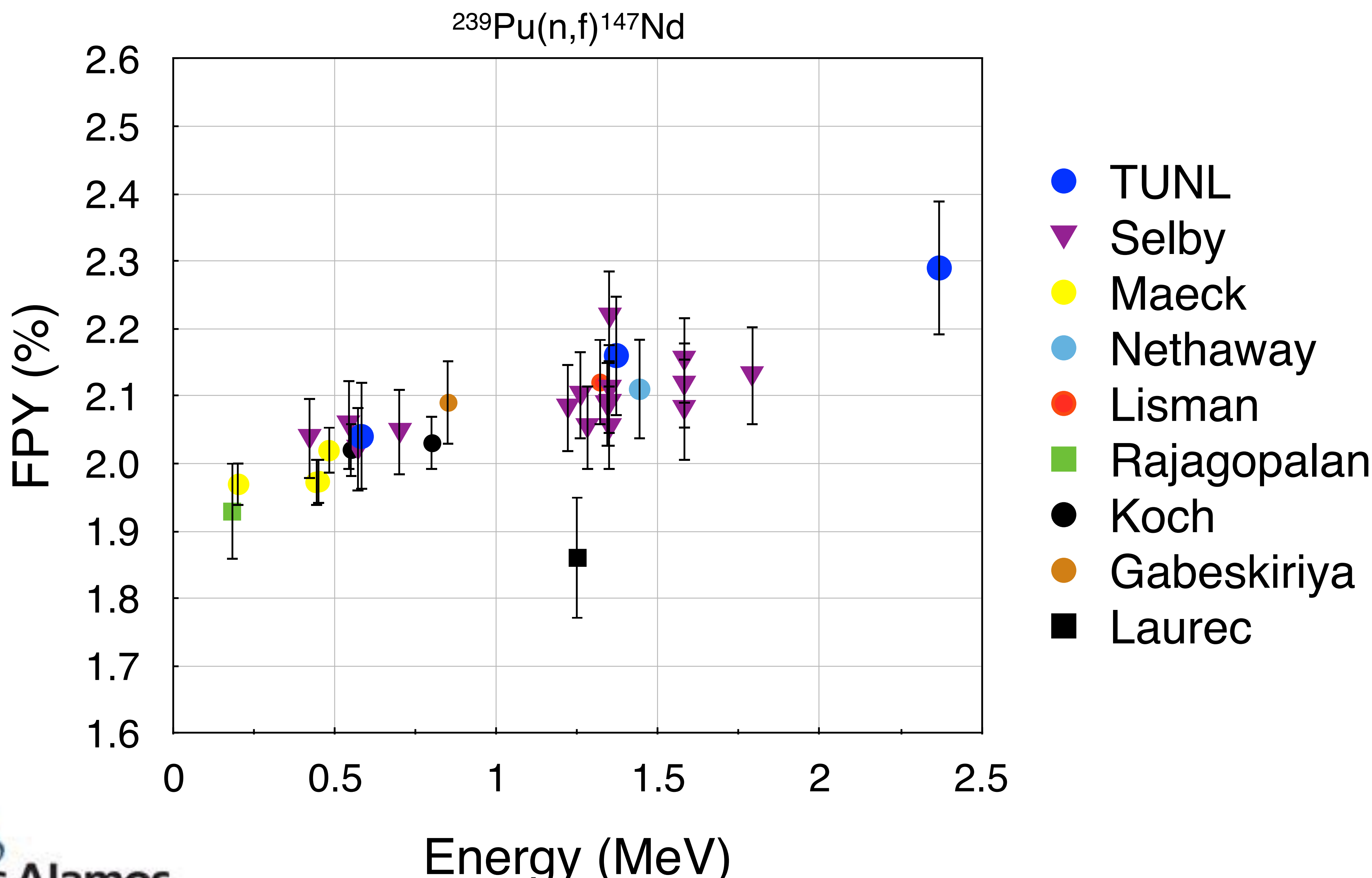
- Existing low-energy critical assembly data





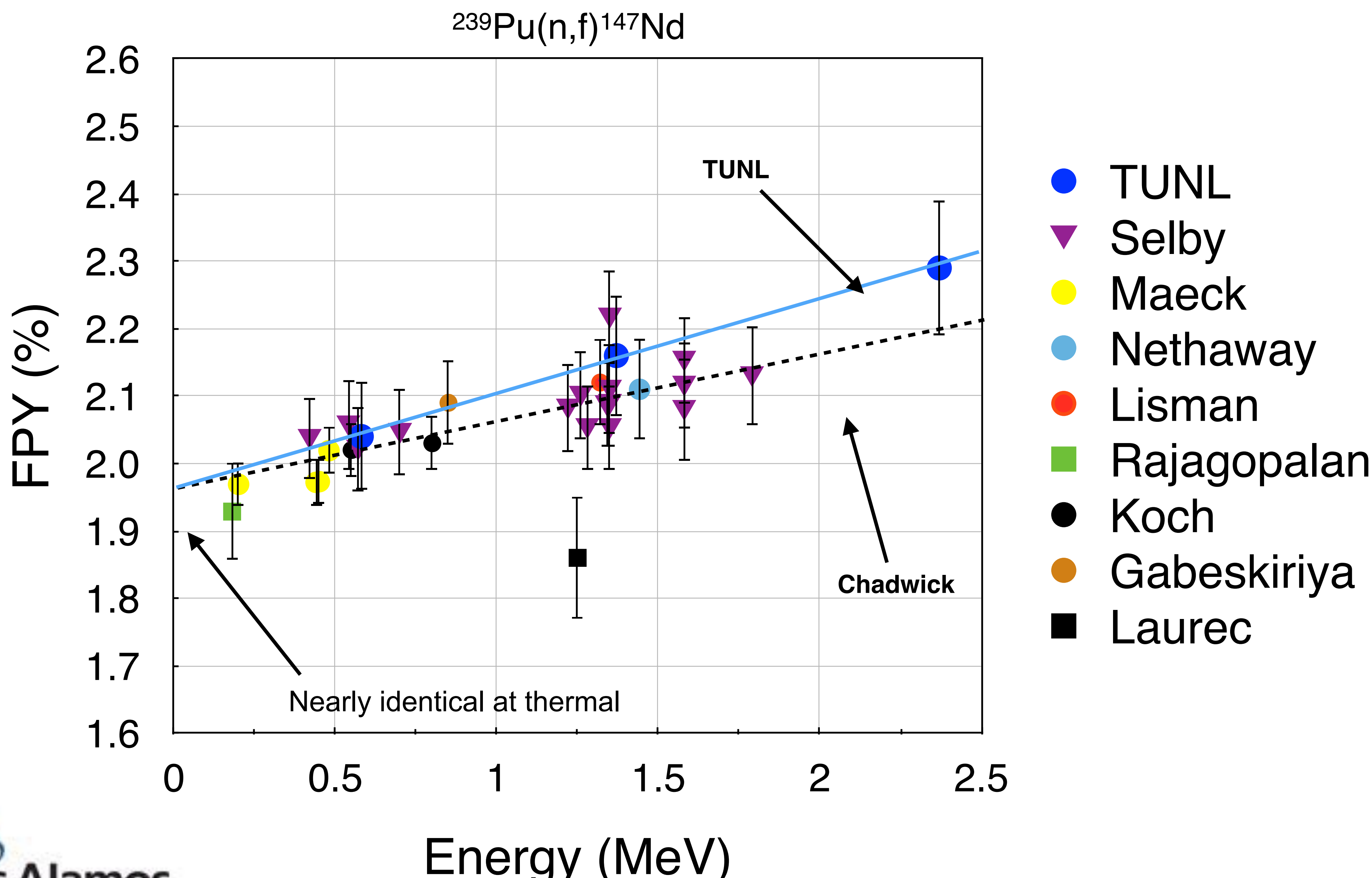
Results: ^{147}Nd

- Nice agreement between 0.58 and 1.37 MeV data with existing measurements
- No data above 2 MeV - How to make a comparison?



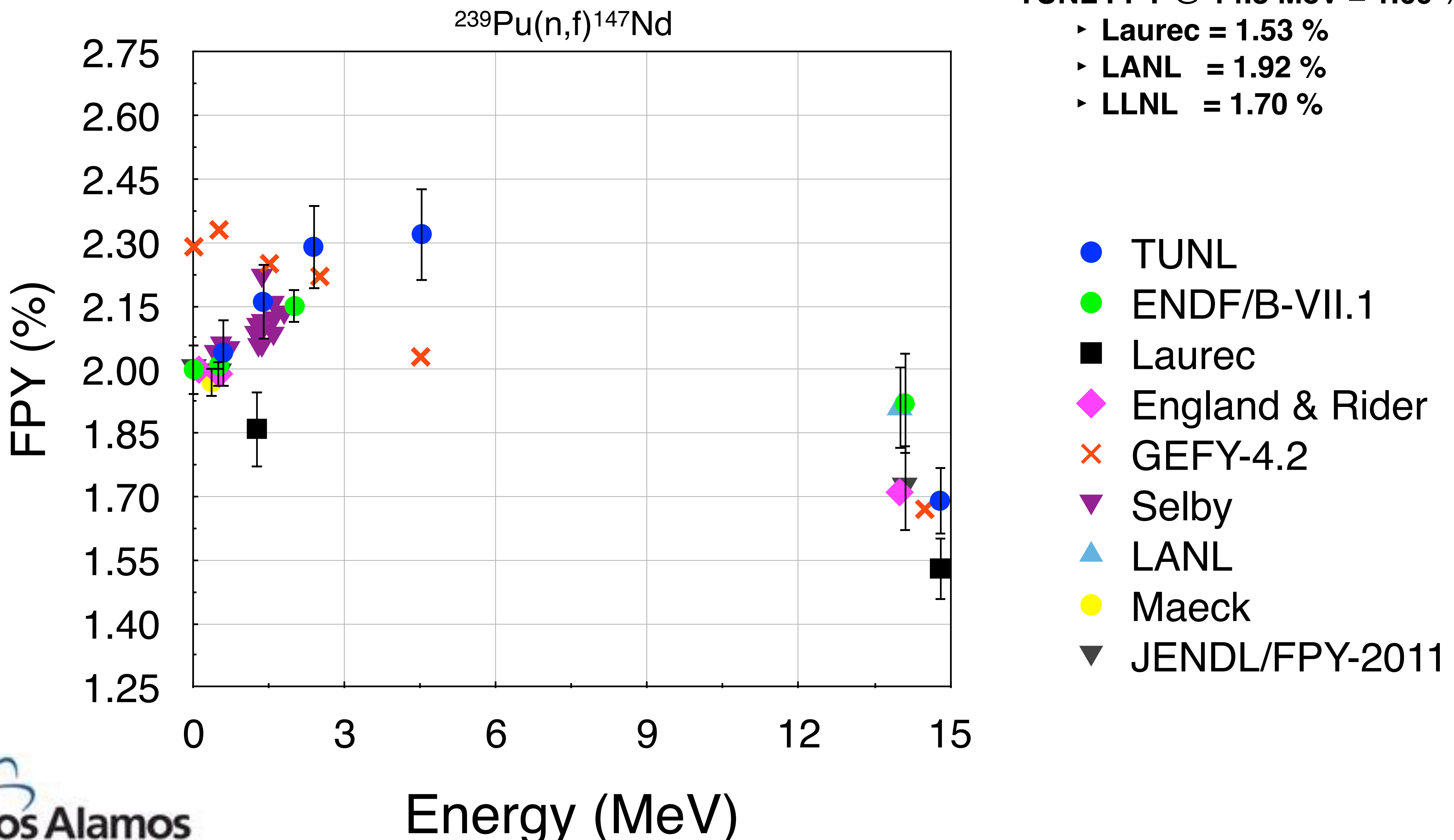
Results: ^{147}Nd

- Chadwick: 4.7 %/MeV   1.961 + 0.091*E
- **TUNL : 7.2 %/MeV**   1.962 + 0.141*E

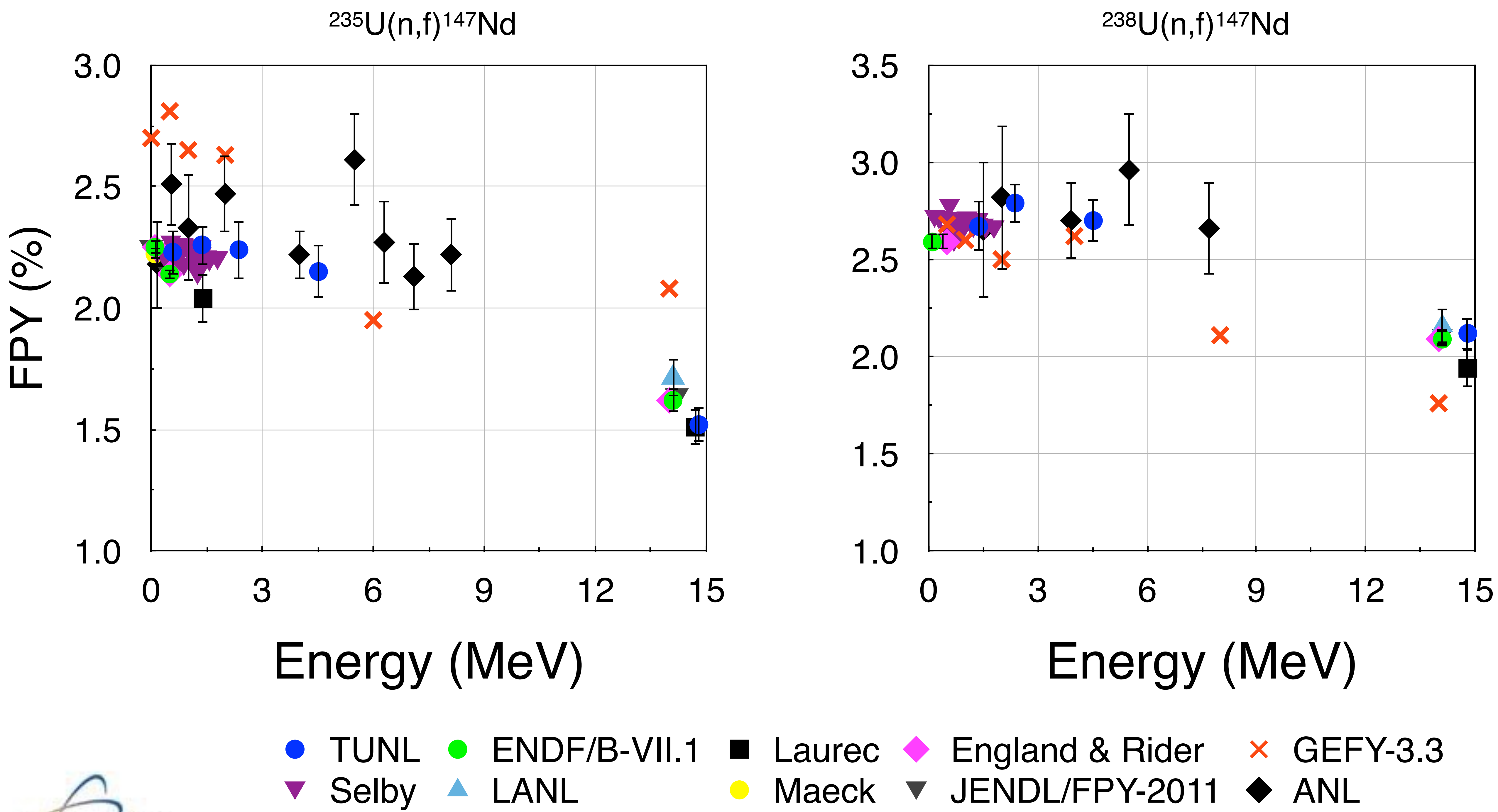


Results: ^{147}Nd

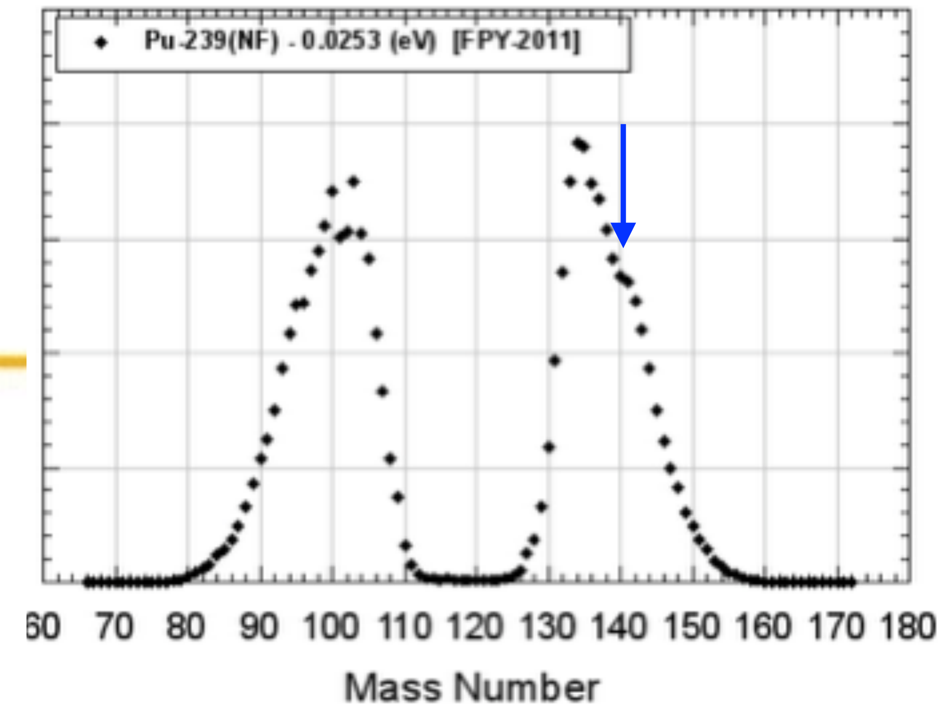
- **Adding data into the 2 - 14 MeV range**
- **TUNL FPY @ 14.8 MeV = 1.69 %**
  - **Laurec = 1.53 %**
  - **LANL = 1.92 %**
  - **LLNL = 1.70 %**

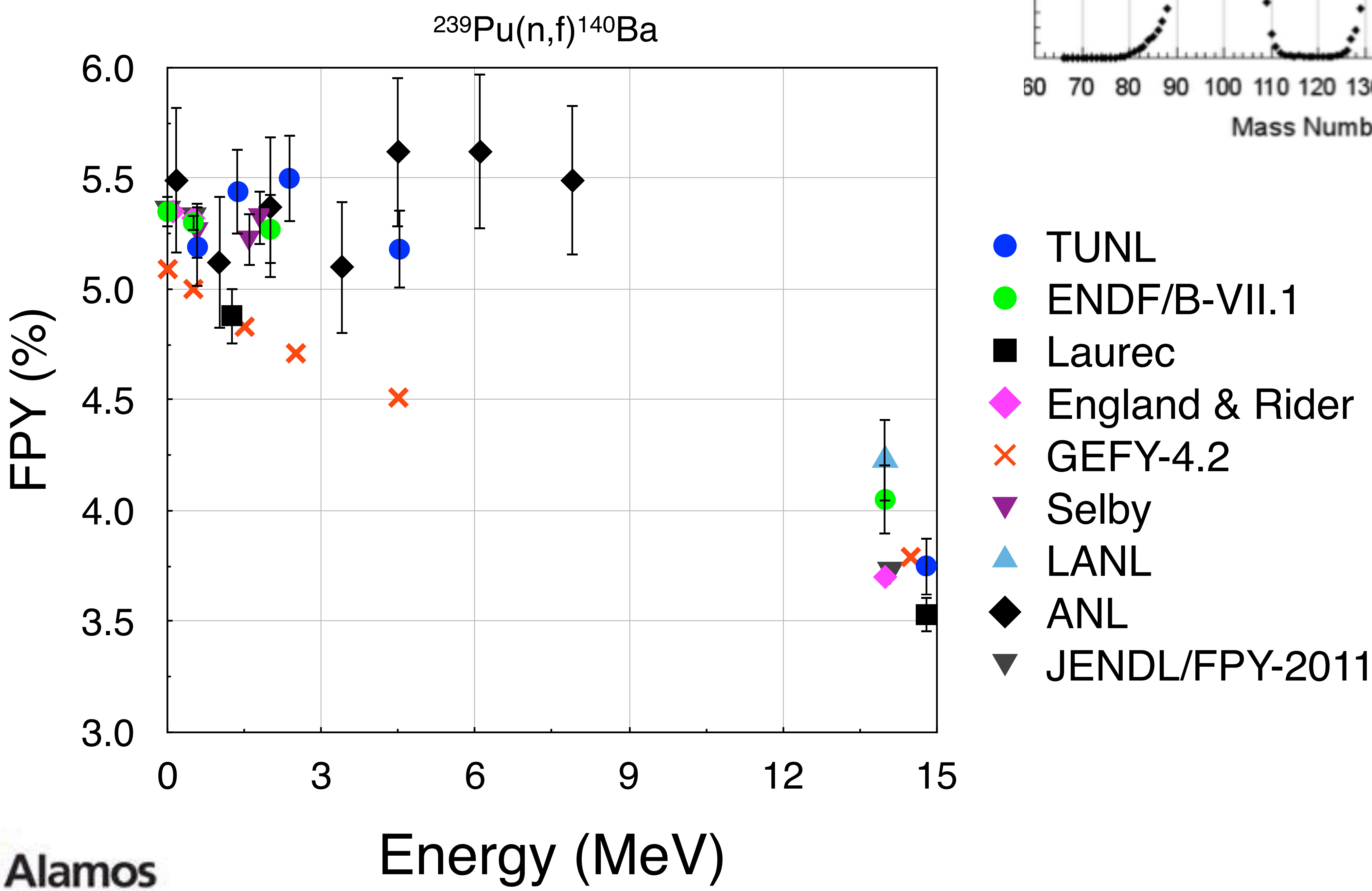


Results: ^{147}Nd



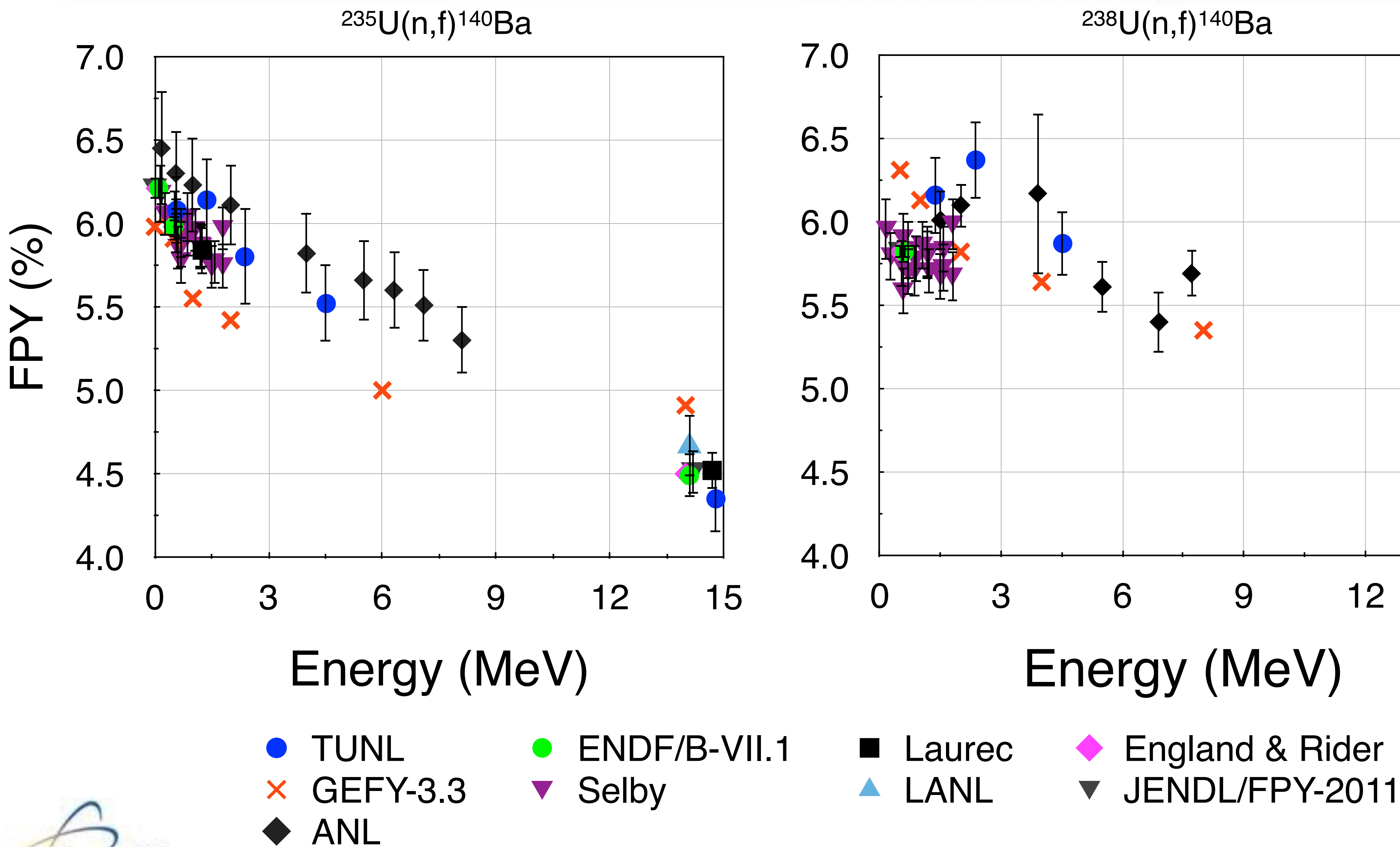
Results: ^{140}Ba





Los Alamos NATIONAL LABORATORY EST. 1943
Operated by Los Alamos National Security, LLC for NNSA

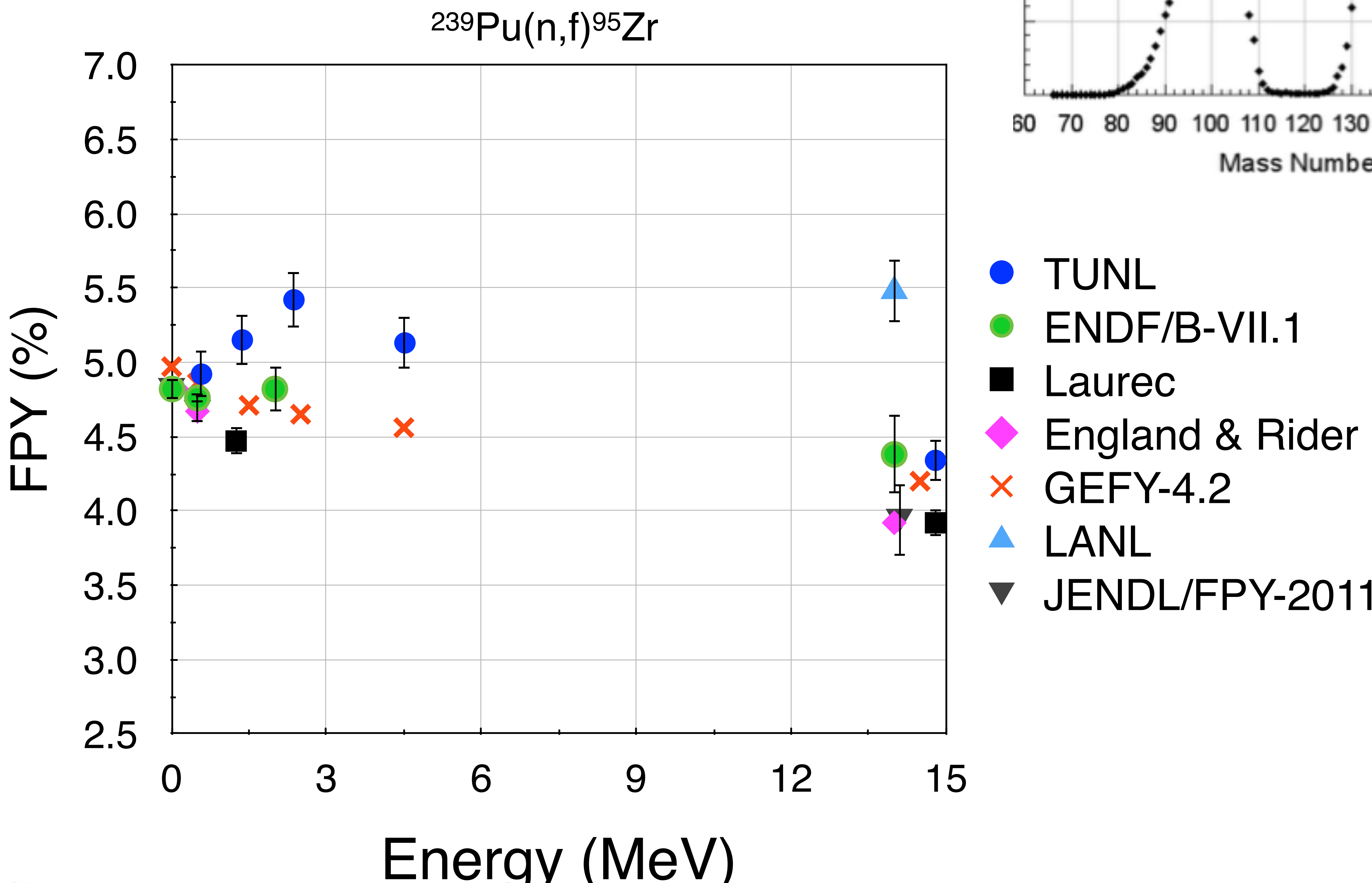
Results: ^{140}Ba

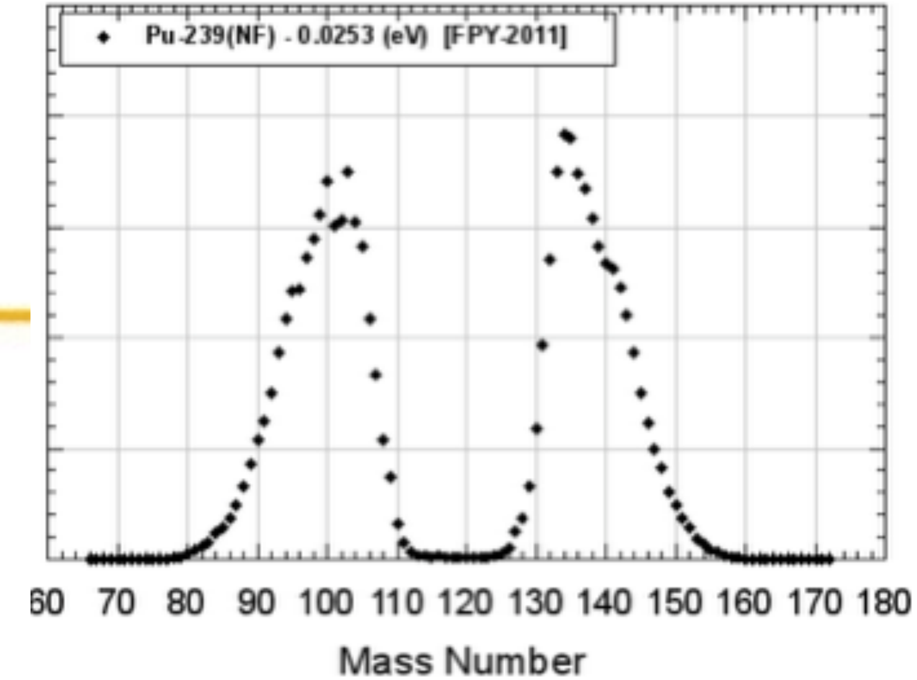


Los Alamos NATIONAL LABORATORY EST. 1943

NNSA

Results: ^{95}Zr





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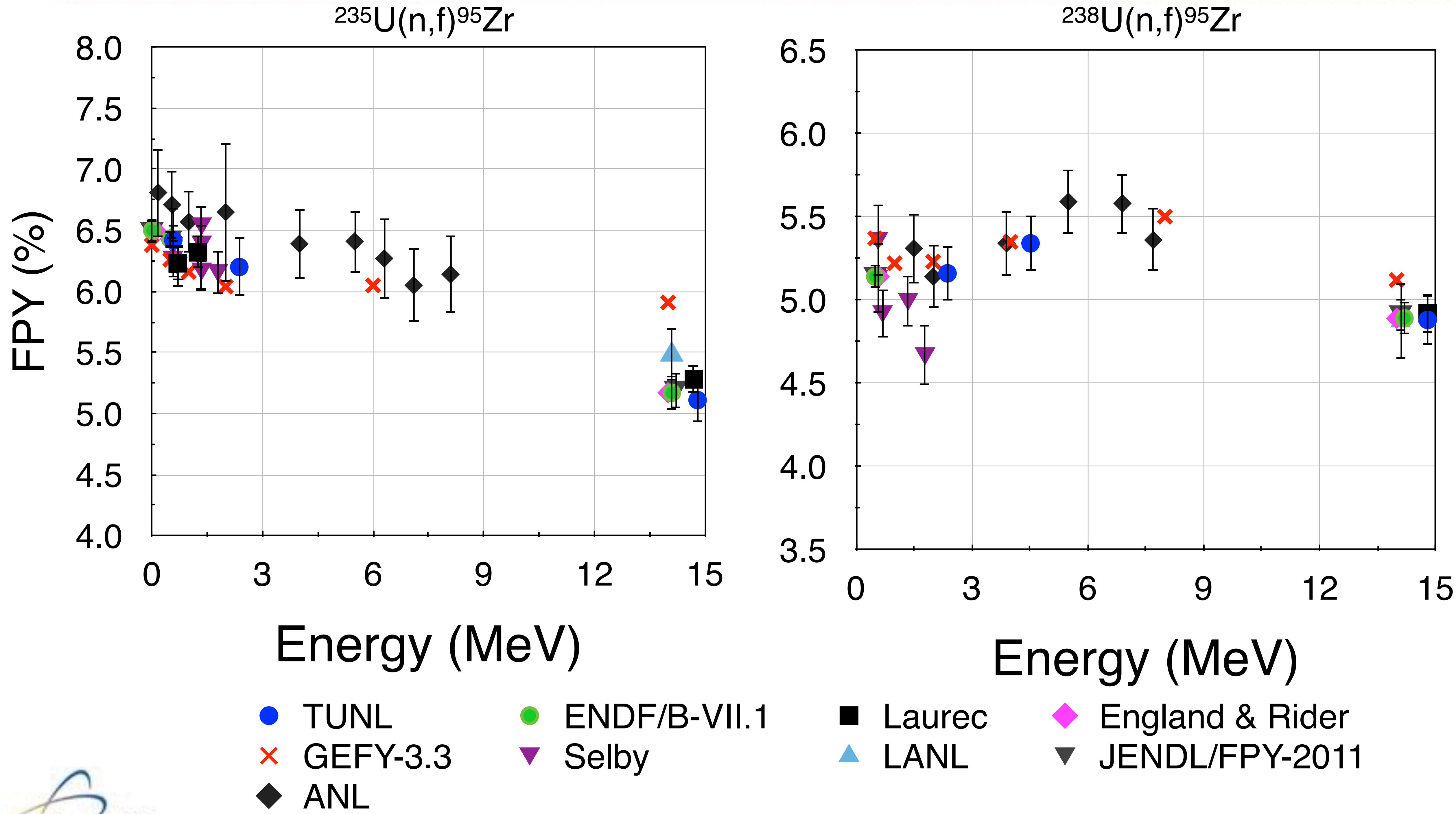


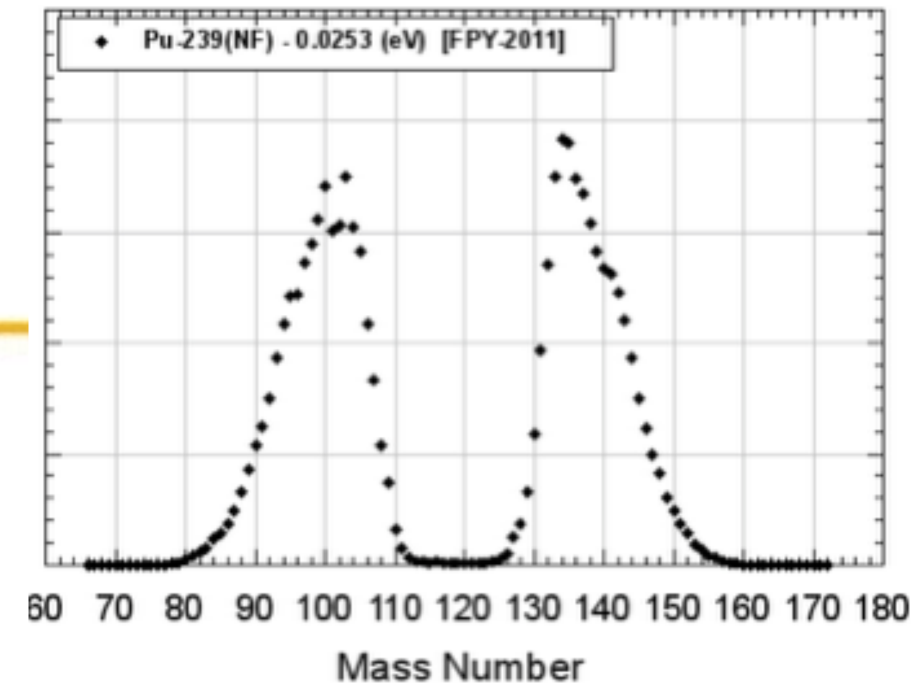
Summary

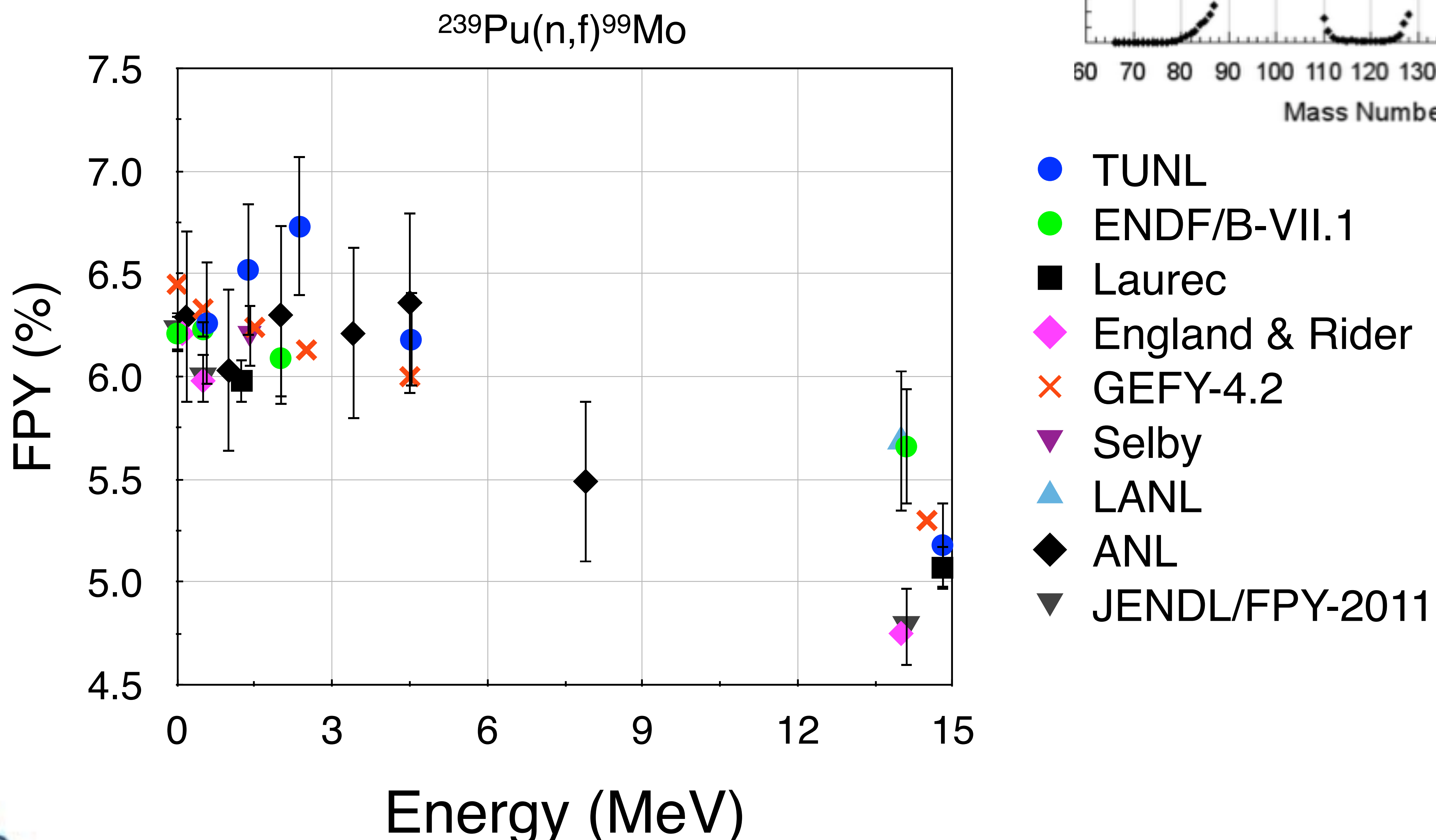
- FPY measurements have been made at neutron energies of **0.56**,**1.37**, **2.37**, **4.52**, 9, **14.8** MeV on $^{235/238}U$ and ^{239}Pu
- Analysis on going for new data at 3.6 MeV
- Data supports a positive slope for ^{147}Nd FPY from $^{239}Pu(n,f)$ as suggested by Chadwick.
- NIM (9 MeV) and PRL (data presented) articles being prepared

THANK YOU

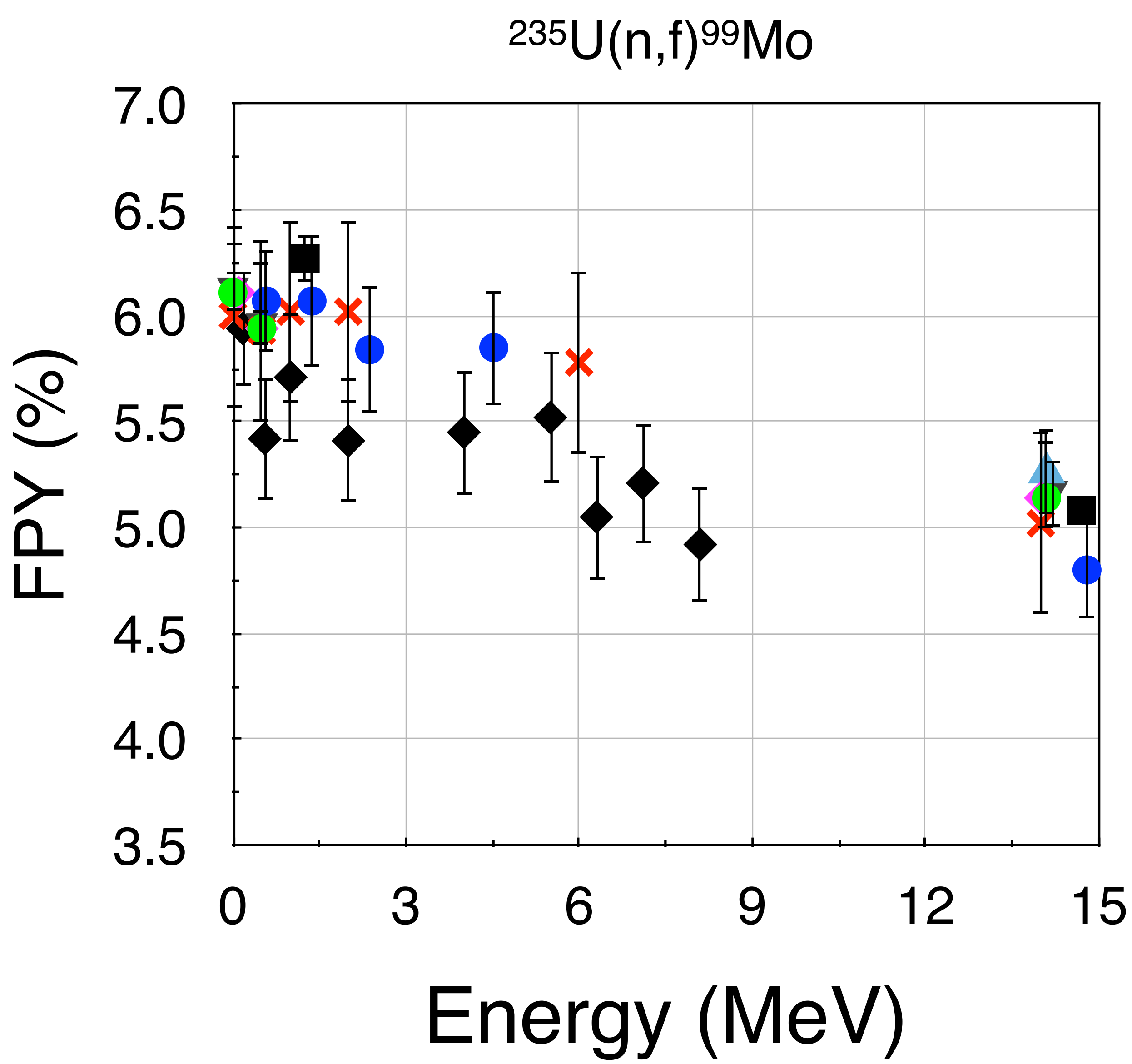
EXTRA SLIDES

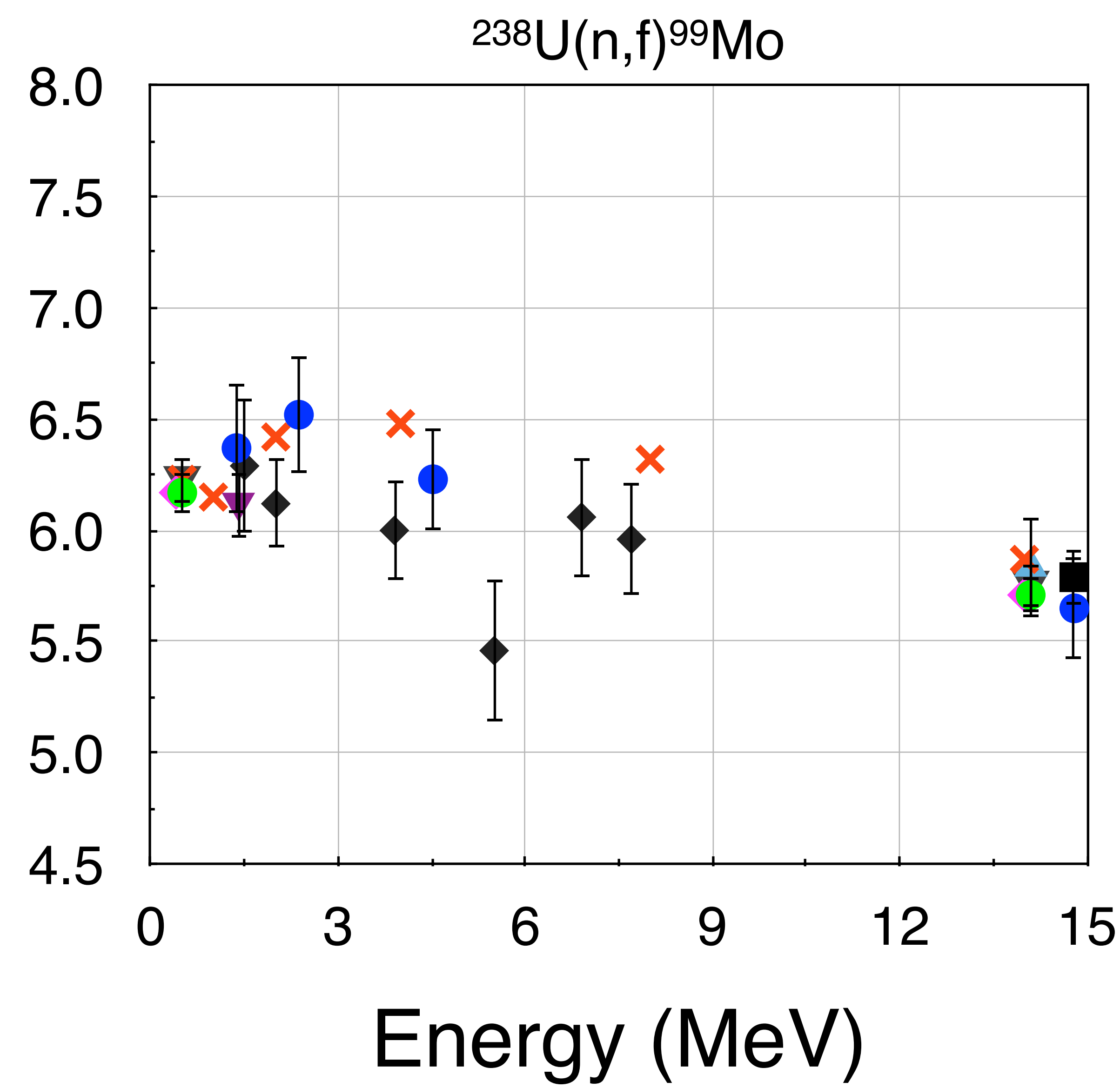
Results: ^{99}Mo





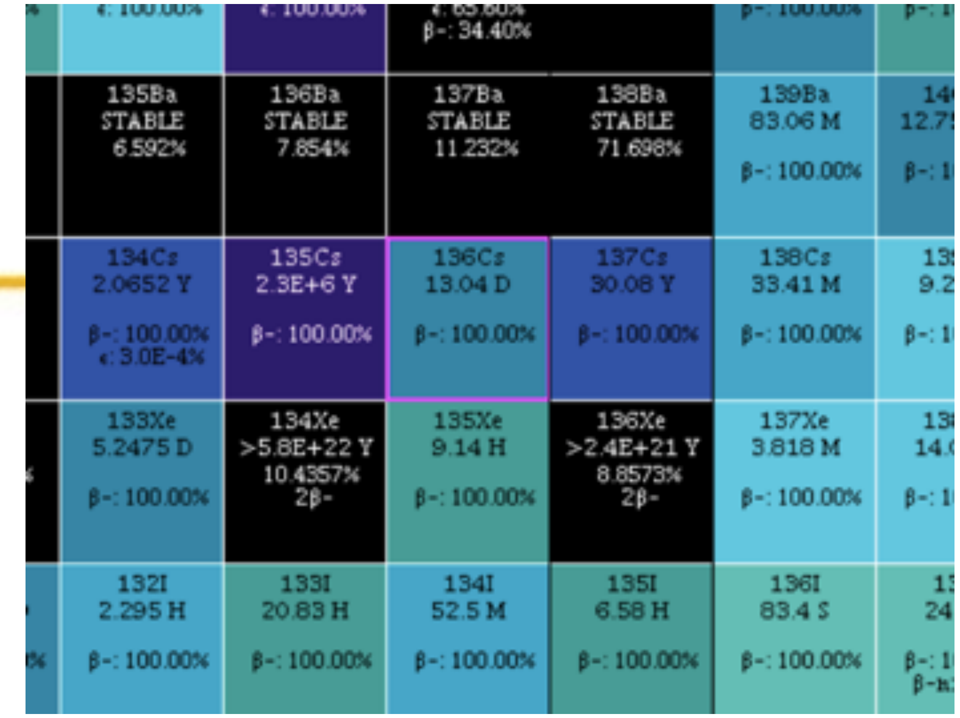
Results: ^{99}Mo





TUNL ENDF/B-VII.1 Laurec England & Rider GEFY-3.3
LANL JENDL/FPY-2011 ANL

Results: ^{136}Cs



***Independent Yield**