Measurements and Simulation of Neutron Capture Gamma Cascades

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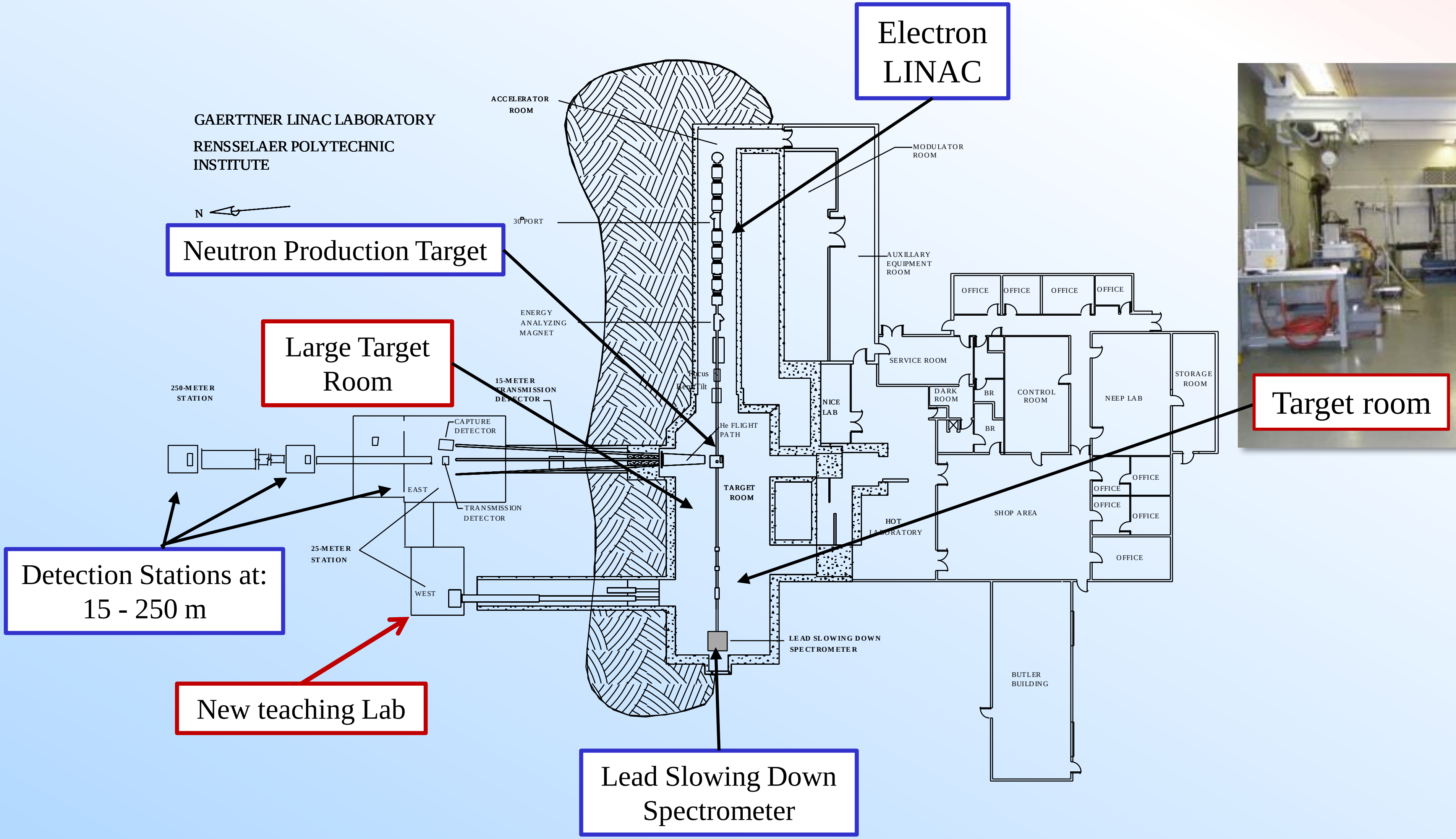
Outline

- The Gaerttner LINAC lab at RPI
- Previous measurements with the multiplicity detector
- Capture gamma cascade project overview
- Example ^{56}Fe ^{55}Mn, ^{59}Co
- Capture cascades in $^{235, 238}$U
- Conclusions and outlook

The RPI Gaerttner LINAC Facility



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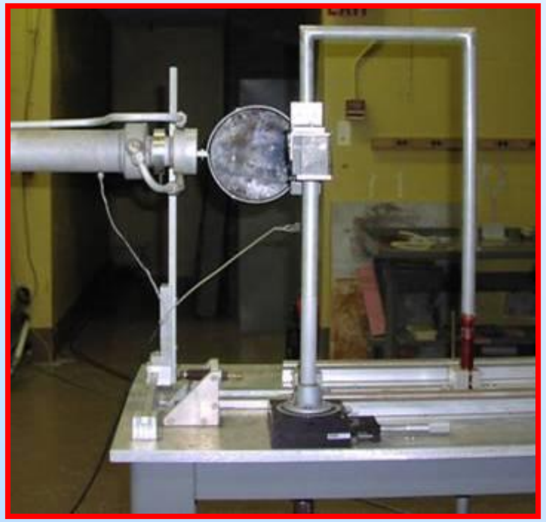
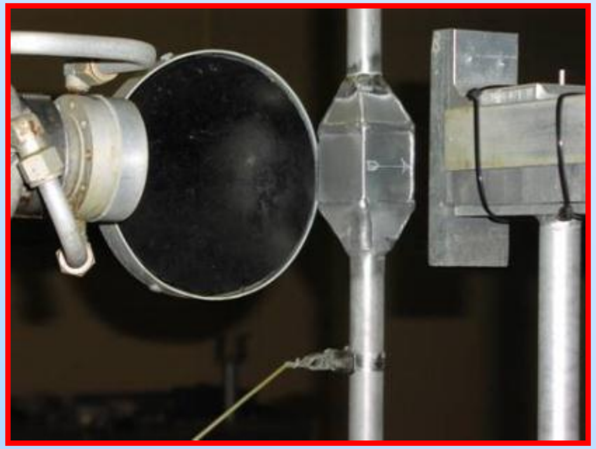
Neutron Production Targets (electrons → neutrons)

Bare Bounce Target (BBT)

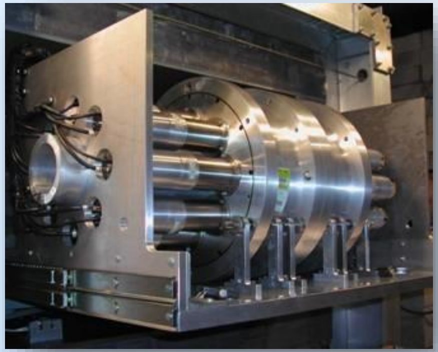




Enhanced Thermal Target (ETT)

PACMAN target

Detector Systems



Capture/Multiplicity

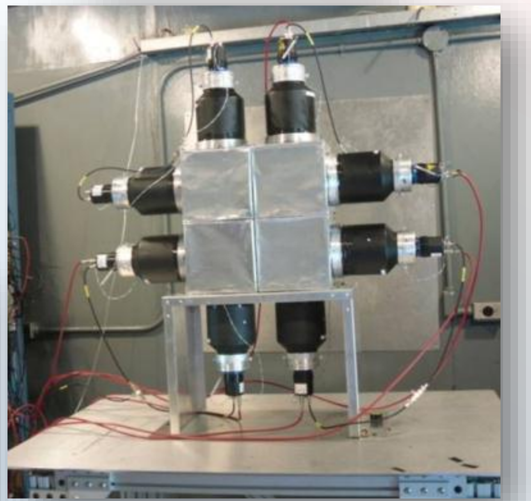
25m

45m

Transmission

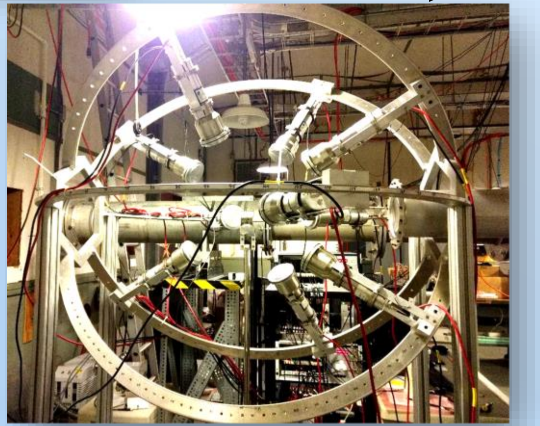
35m

100m



250m

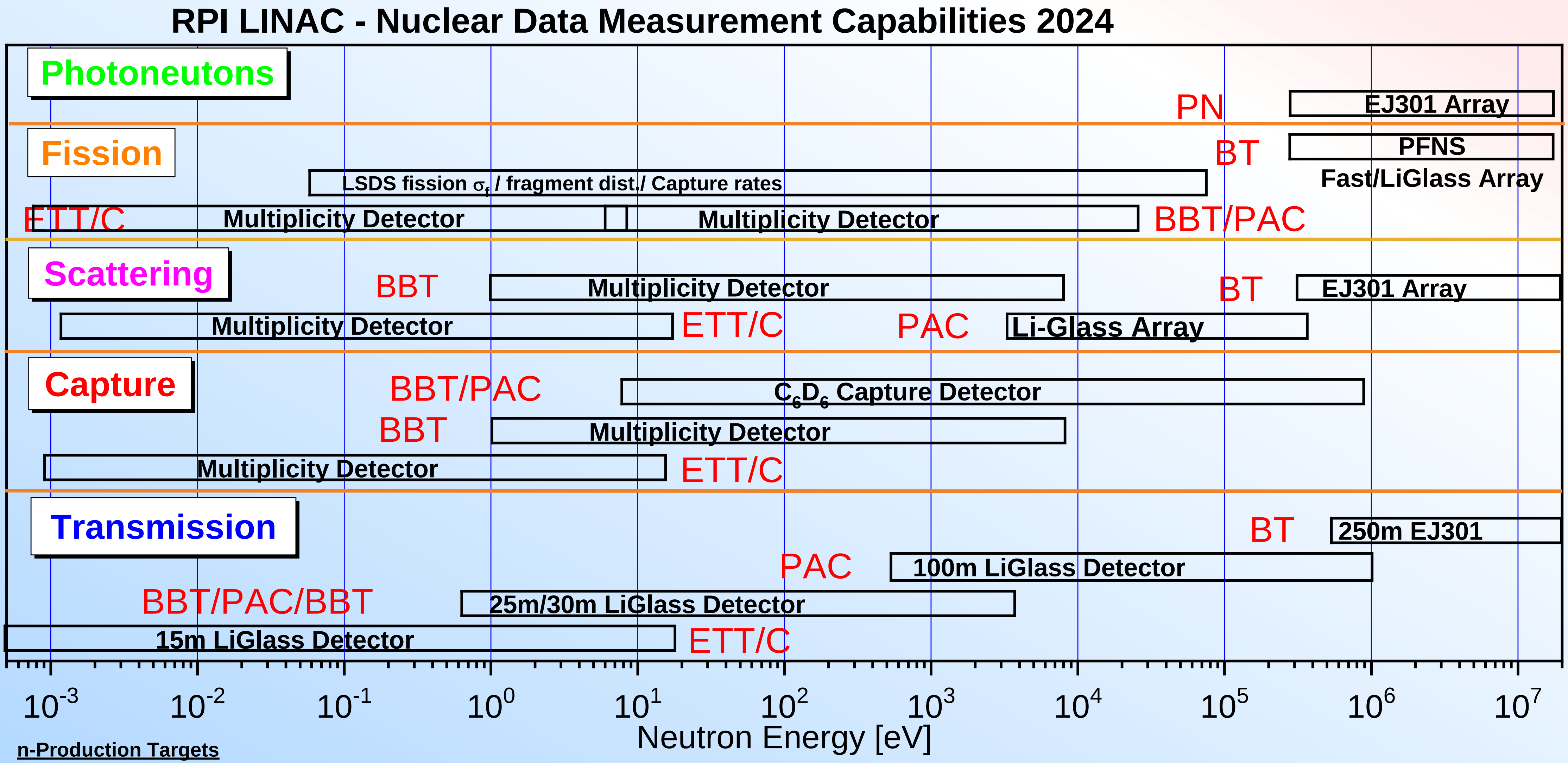
Neutron Scattering



keV Scattering @ 35m

Fast Scattering and PFNS @ 30m

Capability Matrix and Development



n-Production Targets

ETT- Enhanced Thermal Target
ETTC - ETT + cold moderator
BBT - Bare Bounce Target
PAC - PacMan Target
PN - Photoneutron target
BT- Bare Target on Axis

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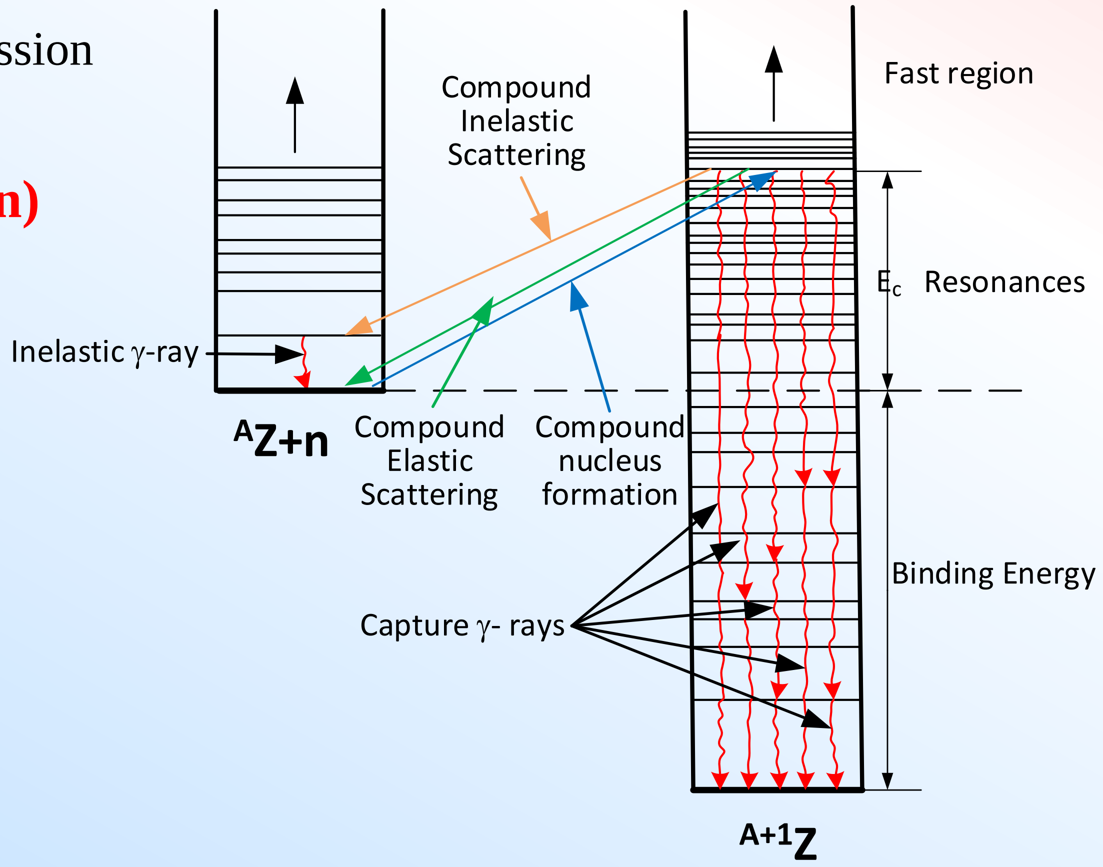
Compound Nucleus Connection

- **At RPI, we mostly measure *neutron induced reactions***
  - Incident neutron energy range sub-thermal thermal to 20 MeV
  - Measurements include transmission, capture, scattering and fission
  - Focus is nuclear data for applications
- **In this talk, the focus will be neutron capture (and fission)**

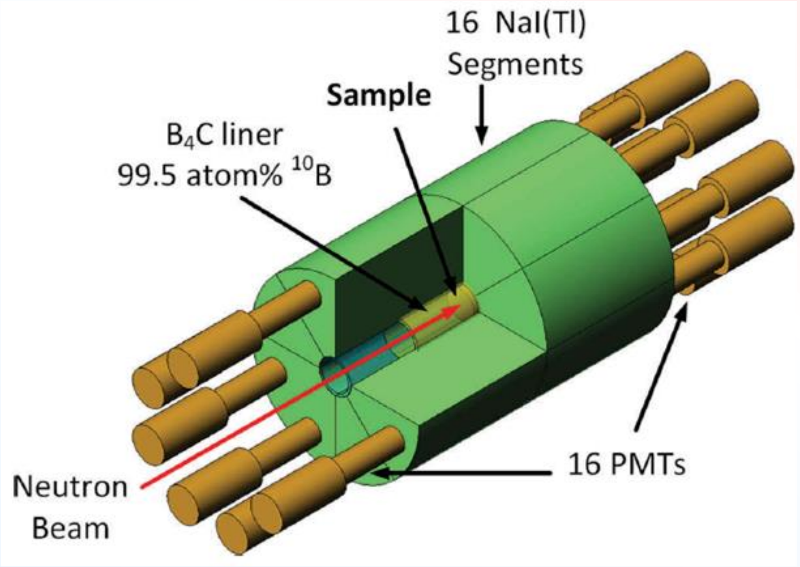
$$n + {}^{A}_{Z}X \rightarrow {}^{A+1}_{\ \ Z}X^{*}$$

$n + {}^{A}_{Z}X$	Elastic Scattering
$n + {}^{A}_{Z}X^{*}$	Inelastic Scattering
${}^{A+1}_{\ \ Z}X + \gamma$	Radiative Capture
${}^{A1}_{z1}A + {}^{A2}_{z2}B + \nu n + \nu' \gamma$	Fission



RPI Capture γ-Ray Multiplicity Detector

- **16 segment NaI(Tl) γ-ray multiplicity detector**
  - 20 L of NaI(Tl) surrounding the sample
  - A 1 cm thick B_4C ceramic sleeve (enriched to 99.5 atom% in ^{10}B) is used inside the detector to absorb neutrons scattering from the sample
  - Up to 96% efficiency for detecting γ-ray cascades
  - Located ~25.5 m from the neutron-producing target
  - Time-of-flight (TOF) method used to determine incident neutron energy
- **Used for neutron capture yield and γ-ray spectra measurements**
  - Incident neutron energies: 0.01 eV – 3 keV
- **16 Channel 250 MHz 14-bit Digitizer (SIS3316-250-14)**
  - Collects digitized pulse data generated during each coincidence event on all 16 detector segments to determine the γ-ray energy deposited in each detected event



Previous work - ^{235}U capture & fission yield data - epithermal measurement

- Principle:
  - If E_{dep}> binding energy (6.55 MeV for ^{235}U) → fission
  - Gamma multiplicity is higher for fission.
- Normalize experimental fission yield to thermal point or a resonance

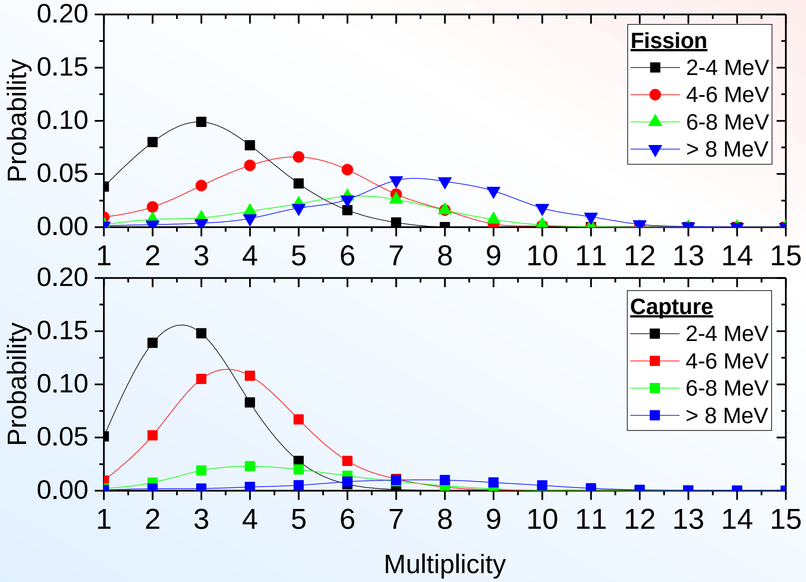
$$Y_f^{ENDF} = k_1 \cdot Y_f \text{ solve for } k_1 \text{ @ 19.3 eV res } \left(\frac{\Gamma_f}{\Gamma} = 0.63\right)$$

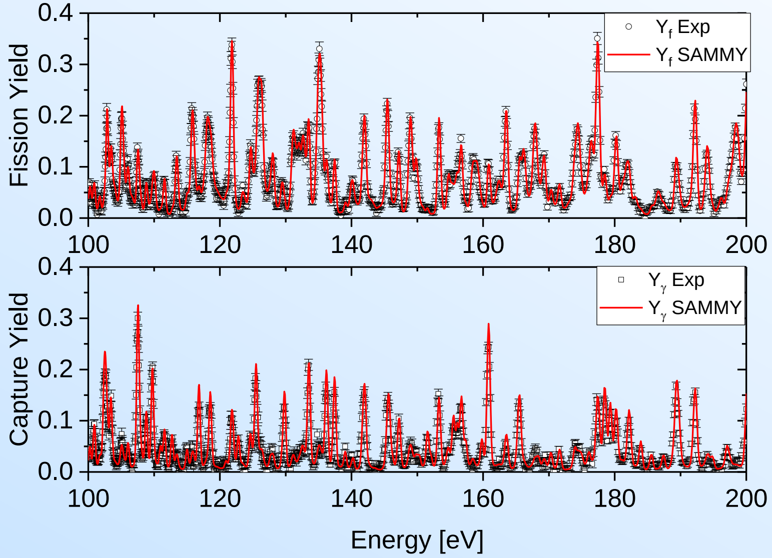
- Use two equations for predominantly capture and fission resonances

@ 11.7 eV res $\left(\frac{\Gamma_\gamma}{\Gamma} = 0.86\right)$ @ 19.3 eV res $\left(\frac{\Gamma_f}{\Gamma} = 0.63\right)$

$$Y1_\gamma^{ENDF} = k_2 \cdot Y1_\gamma - k_3 \cdot k_1 \cdot Y1_f \qquad Y2_\gamma^{ENDF} = k_2 \cdot Y2_\gamma - k_3 \cdot k_1 \cdot Y2_f$$

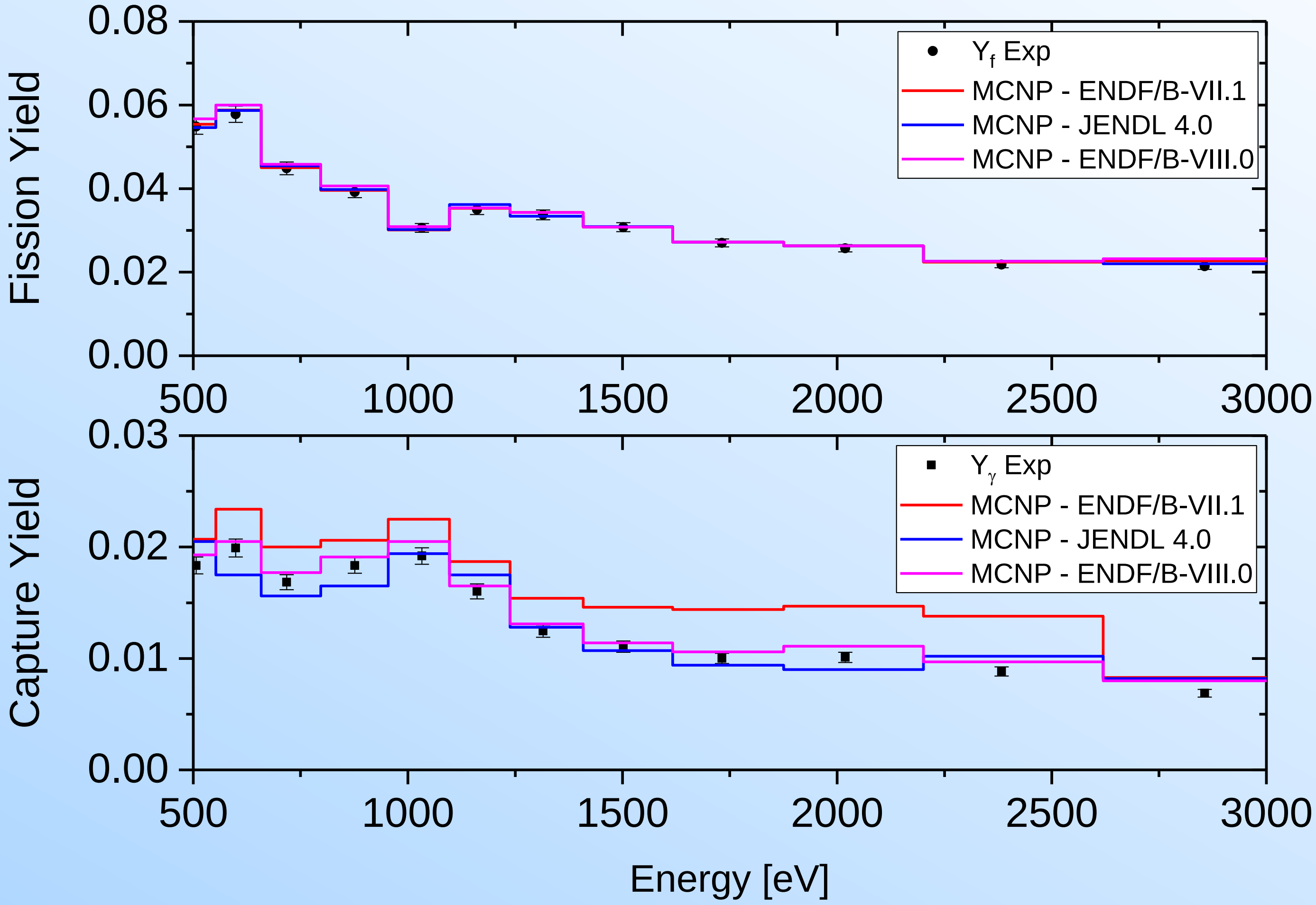
- Solve the two equations for k_2 and k_3





Y. Danon, D. Williams, R. Bahran, E. Blain, B. McDermott, D. Barry, G. Leinweber, R. Block and M. Rapp, "**Simultaneous Measurement of ^{235}U Fission and Capture Cross Sections From 0.01 eV to 3 keV Using a Gamma Multiplicity Detector**", Nuclear Science and Engineering, vol. 187, no. 3, pp. 291-301, 2017, DOI:10.1080/00295639.2017.1312937

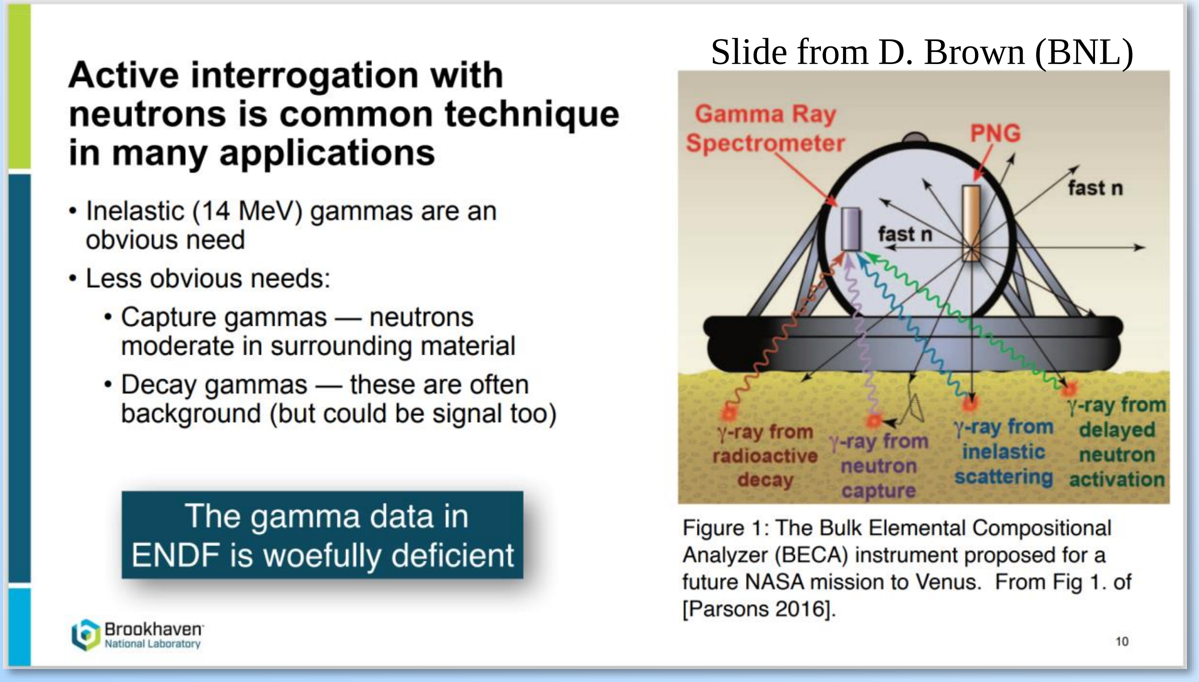
Previous work - Comparing ^{235}U fission and capture yields with evaluations



- Fission is in excellent agreement with evaluations
- Capture uncertainty was 3%
- Helped resolved issues in ENDF/B-VII.1
- Helped validated ENDF/B-VIII.0.

Project Motivation

- **There is a need for accurate capture γ-ray cascade data for applications**
  - Examples include: γ-ray heating in reactors and other critical systems, active neutron interrogation (non-proliferation efforts), detector response
  - Supporting γ-Rays Induced by Neutrons (GRIN) Collaboration
- **Applications can measure capture gammas using a single detector or in coincidence**



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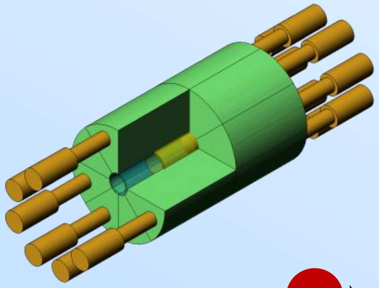
Project Overview

Primary Objective: Develop methodology & provide data to validate the accuracy of neutron capture γ-ray data stored in nuclear data evaluations

1. **Develop experimental methods** to measure γ-ray cascades produced by thermal neutron capture reactions using the RPI Gaerttner Linear Accelerator (LINAC) & Capture γ-Ray Multiplicity Detector
2. **Update simulation methods** to model neutron capture γ-ray transport using Monte-Carlo code(s)
3. **Assess the quality of neutron capture γ-ray cascade evaluations** by comparing measurements to simulations using evaluated γ-ray cascades data inputs
4. **Produce deliverables**: benchmark methodology, experimental data, and computation methods for simulation

Neutron Capture Data Validation Methodology



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Experimental Data

Experimental Parameters

Modified MCNP 6.2

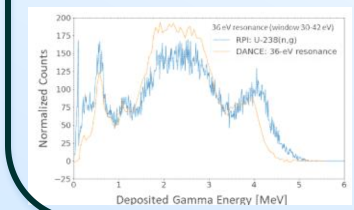
γ–ray cascades, Probability Distributions, etc…

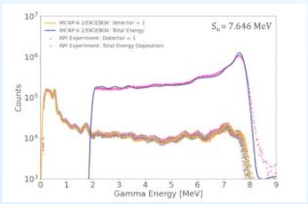
Nuclear Data Evaluation from ENDF/B-VIII.0, DICEBOX (ENSDF + RIPL-3) or other sources

julia Processing Code

Compare various results:

- Neutron capture γ-ray spectra
- γ-ray cascade multiplicity spectra
- Angular distributions of γ-rays
- Total γ-ray energy deposition





Simulation Methods
mod-MCNP-6.2

<u>Step 1</u>: γ-ray cascades are generated using an external code (i.e., DICEBOX) and are written to a file

<u>Step 2</u>: Run mod-MCNP-6.2, for each γ-ray producing event:

1. Read in γ-ray cascade from file
2. Transport γ-ray cascade through the detector geometry
3. Tally γ-ray energy deposition in detector cells

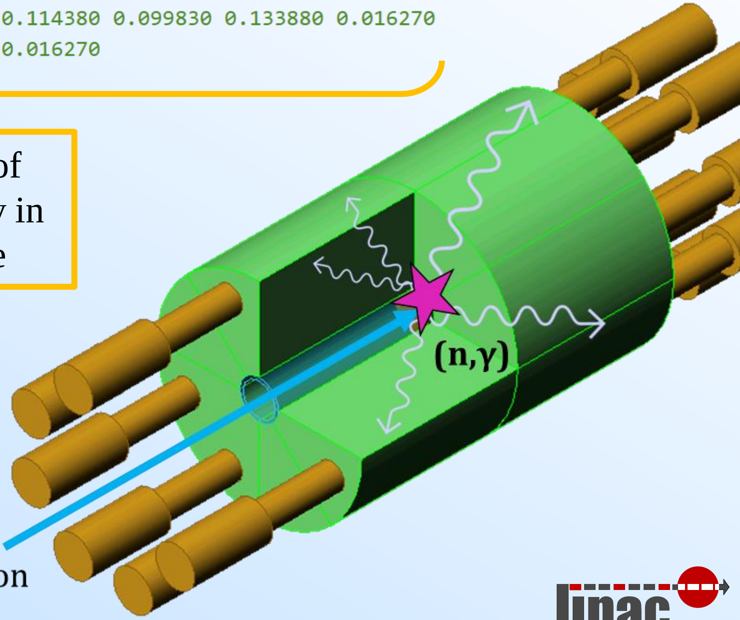
<u>Step 3</u>: Process the output file using event-by-event analysis including coincidence and compare to experimental data

Input cascades

```
6 1.750820 1.641250 2.420890 0.099830 0.133880 0.016270
4 3.739200 1.383080 0.924390 0.016270
5 1.077170 3.693640 0.816570 0.072930 0.402630
4 3.073690 1.329380 1.257240 0.402630
9 3.352750 1.874900 0.095160 0.092480 0.081960 0.074270 0.047810 0.173210 0.270400
5 1.523820 2.843460 1.102700 0.190330 0.402630
7 1.167430 2.036980 1.306060 1.283420 0.118900 0.133880 0.016270
7 2.718790 1.697300 0.732220 0.645580 0.118900 0.133880 0.016270
5 3.656540 2.156420 0.099830 0.133880 0.016270
5 2.228480 2.021640 1.662670 0.133880 0.016270
6 1.127790 1.617990 1.222300 1.115140 0.963450 0.016270
6 3.090780 1.588910 1.133270 0.099830 0.133880 0.016270
6 0.699690 2.839370 2.254830 0.118900 0.133880 0.016270
6 0.788570 1.263900 1.914530 1.346860 0.346450 0.402630
7 0.917810 2.500300 1.769250 0.625600 0.099830 0.133880 0.016270
5 1.480550 2.052000 2.054830 0.072930 0.402630
8 0.917080 2.426180 1.294120 1.061200 0.114380 0.099830 0.133880 0.016270
5 4.761050 1.032840 0.118900 0.133880 0.016270
```

Number of γ-rays in cascade

Energy of each γ-ray in cascade



Simulation Methods

Transport γ-ray cascades through the RPI Capture Detector

MCNP-6.2/ACE

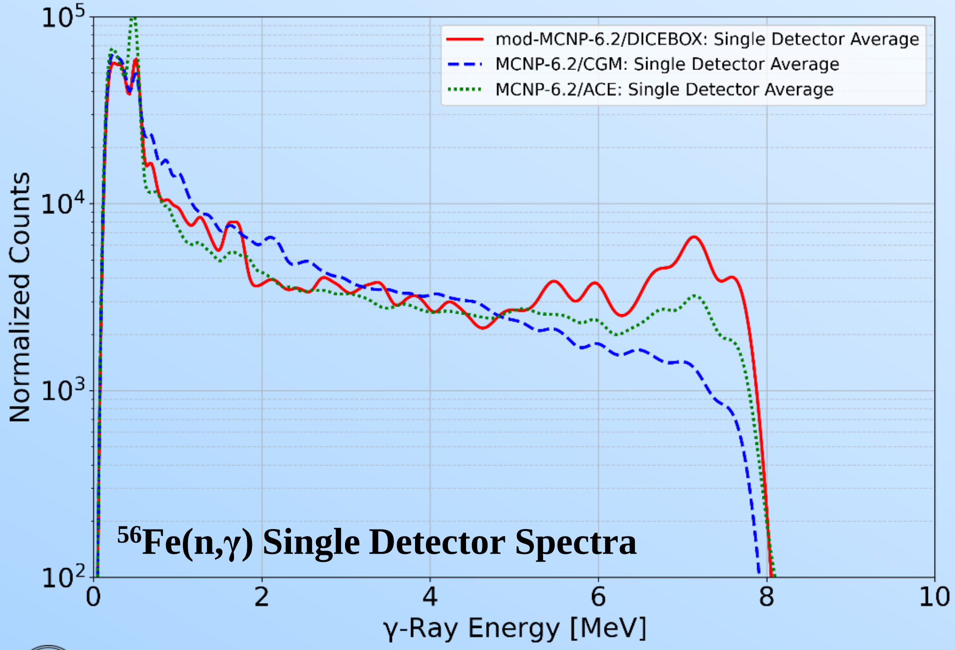
- Extracts γ-ray data from ENDF/B-VIII.0
- Transported through the detector geometry using MCNP-6.2
- Total energy deposition spectra is *expected to disagree* with experimental data because the simulation does not include *coincidence*

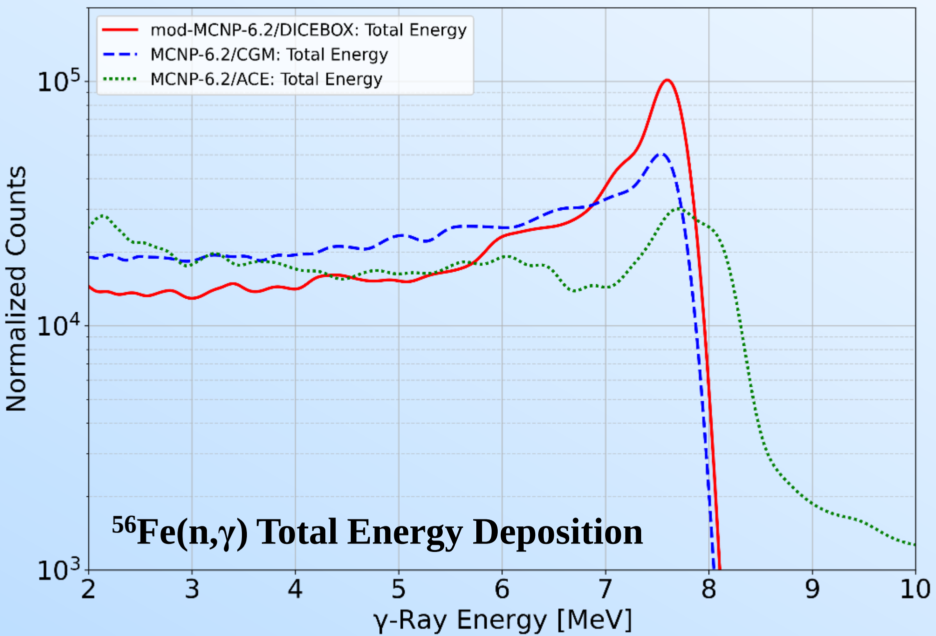
MCNP-6.2/CGM

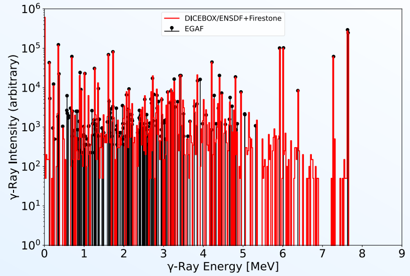
- **C**ascading **γ**-Ray **M**ultiplicity
- Produces correlated secondary γ-ray emissions (cascades)
- Transported through the detector geometry using MCNP-6.2

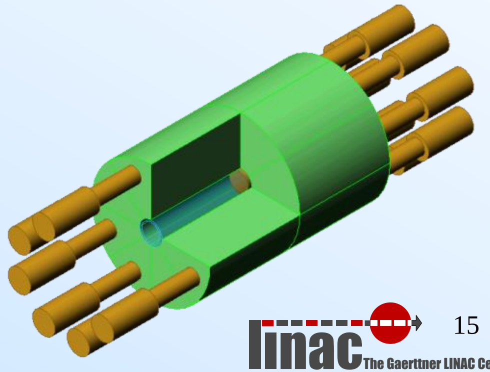
mod-MCNP-6.2/DICEBOX+Firestone

- Cascades generated using DICEBOX + primary intensities from ENSDF/R. B. Firestone et. al., Phys. Rev. C **95**, 014328 (2017).
- mod-MCNP-6.2 banks the cascades to transport through the detector geometry for every γ-ray producing event
- Tally γ-ray energy deposition in detector segments (enables event-by-event analysis including coincidence)









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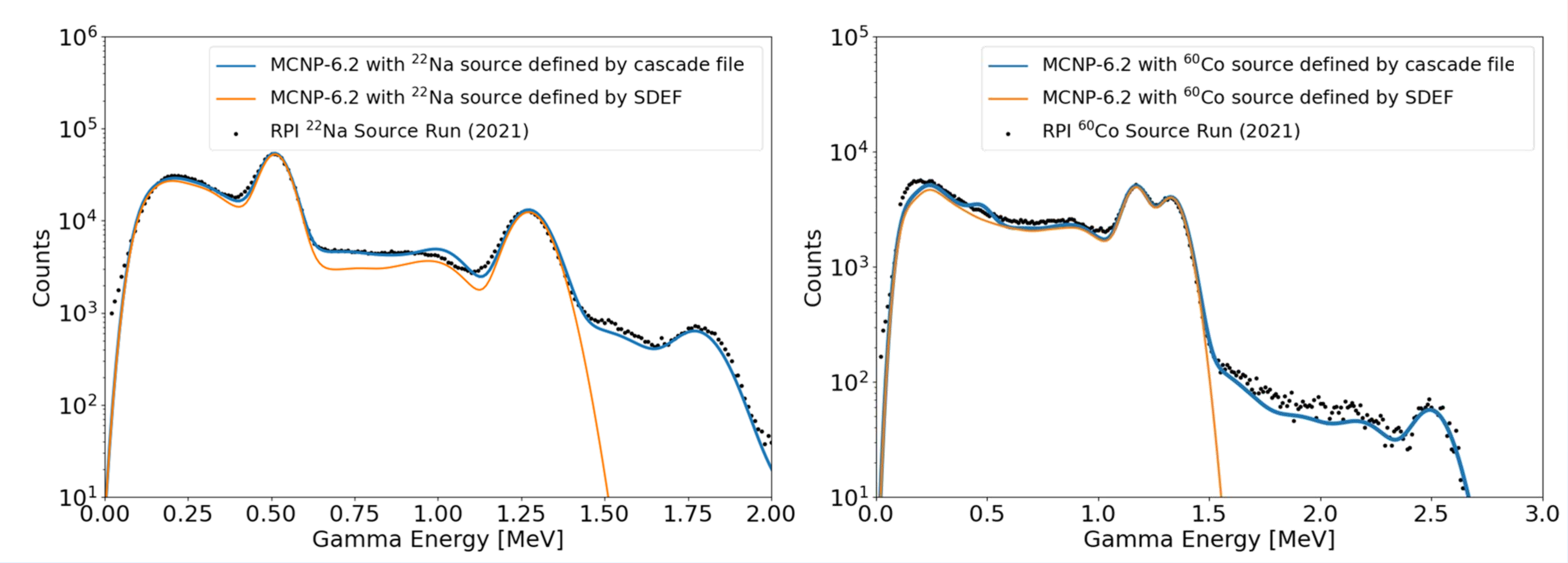
Validation of Coincidence Modeling

Test Cases: ^{22}Na & ^{60}Co coincidence γ-sources



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Simulation Methods

Generate Neutron Capture γ-Ray Cascades using DICEBOX

DICEBOX

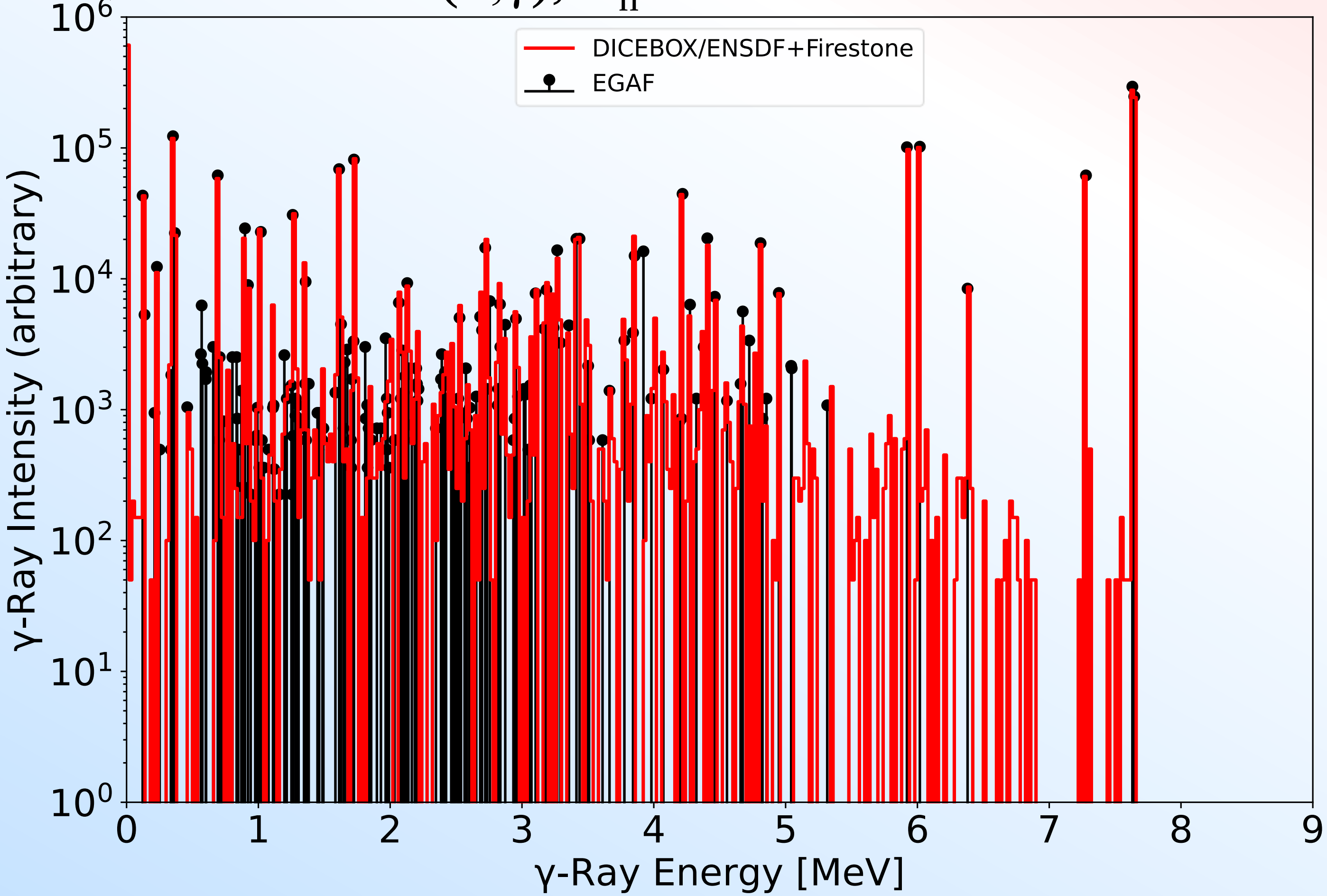
Models full γ-ray cascades using evaluated nuclear data (ENSDF + RIPL-3)

EGAF

Shows experimentally measured primary γ-ray lines

(does not necessarily represent the full cascade)

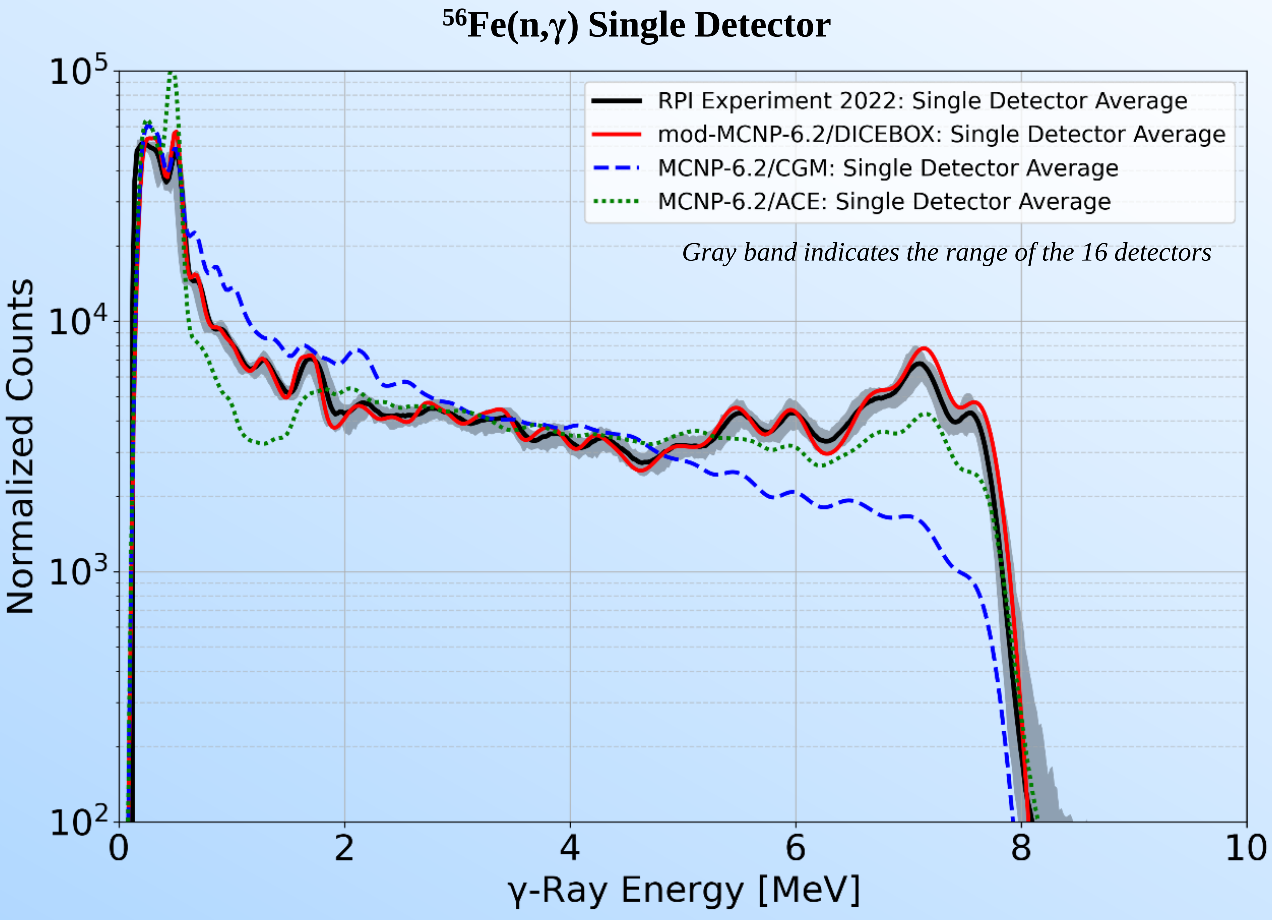
^{56}Fe(n,γ), E_n = thermal

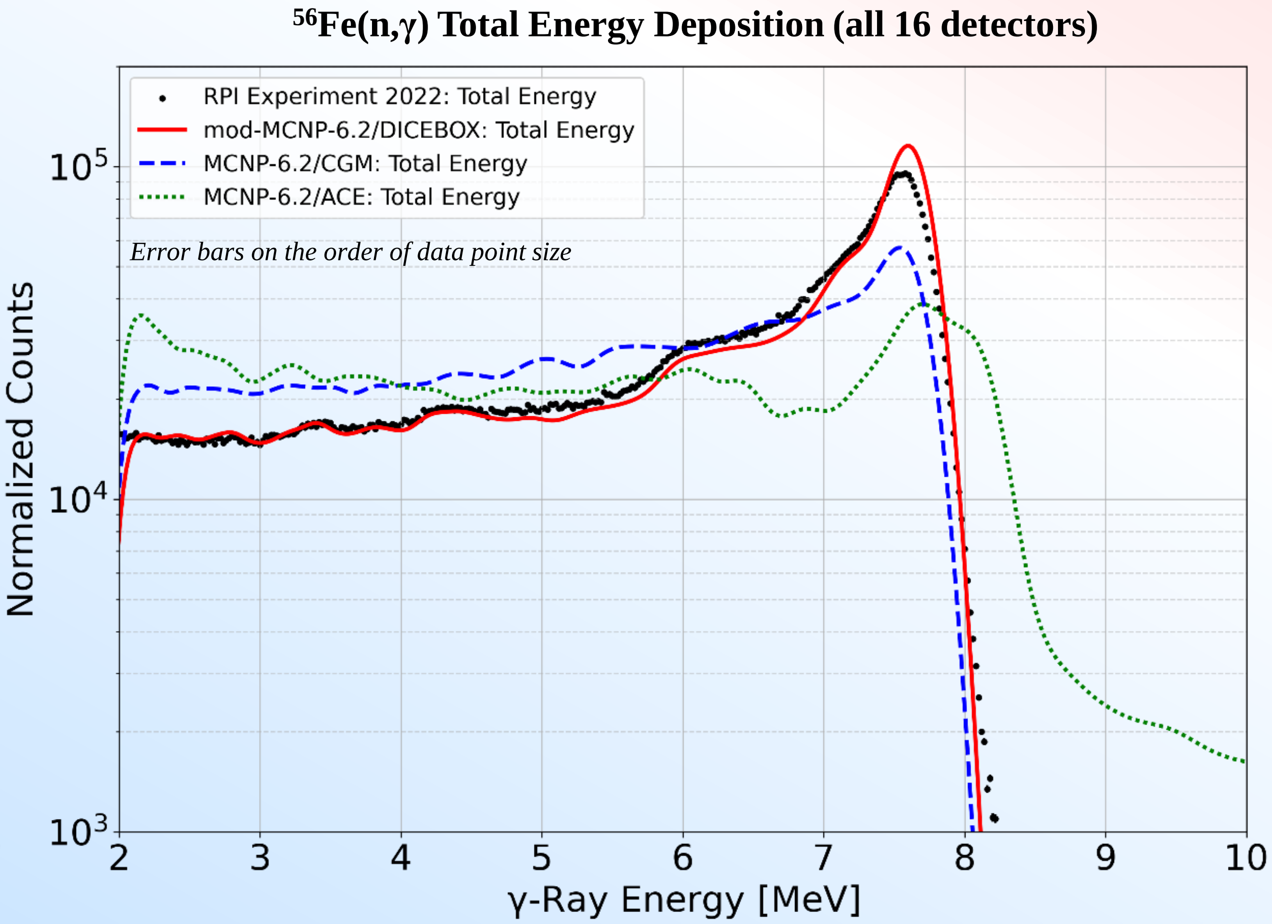


DICEBOX input tuned to R. B. Firestone et. al., Phys. Rev. C **95**, 014328 (2017).

Validating the Experimental & Simulation Methods

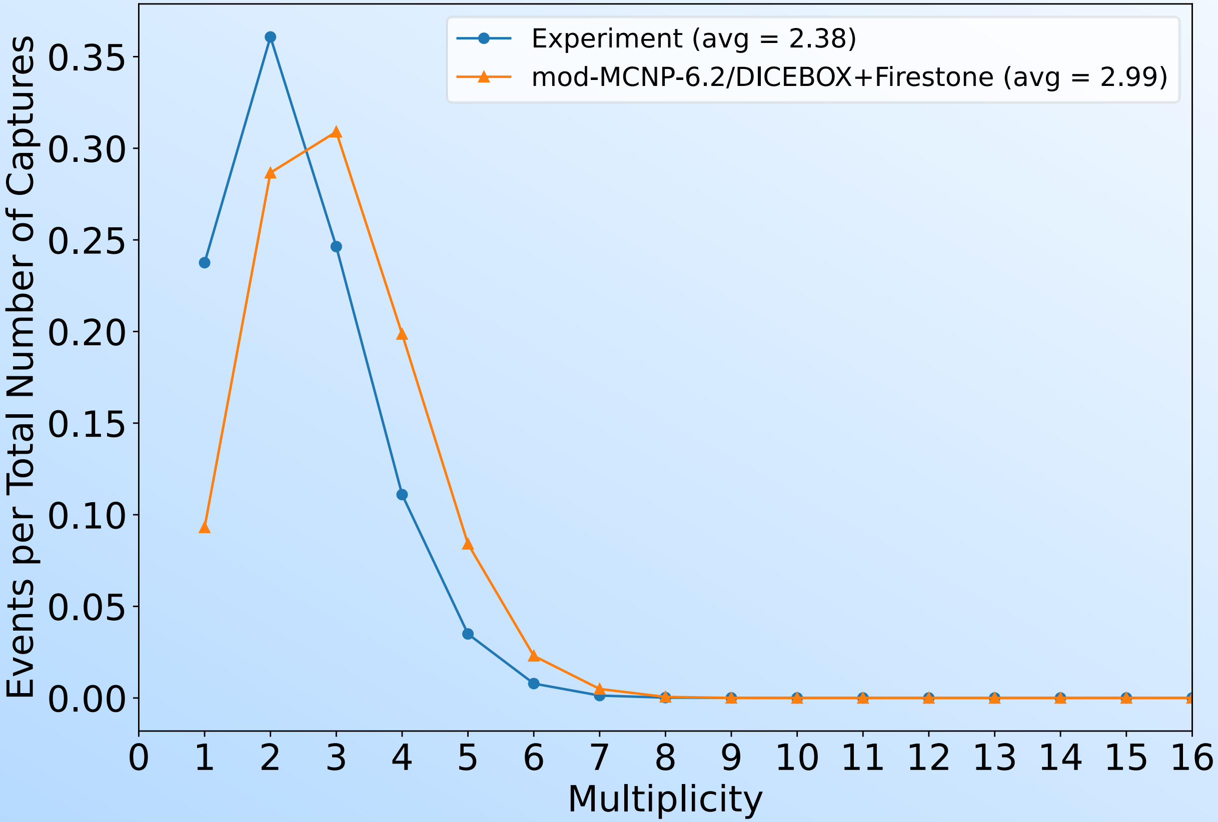
^{56}Fe Thermal Neutron Capture



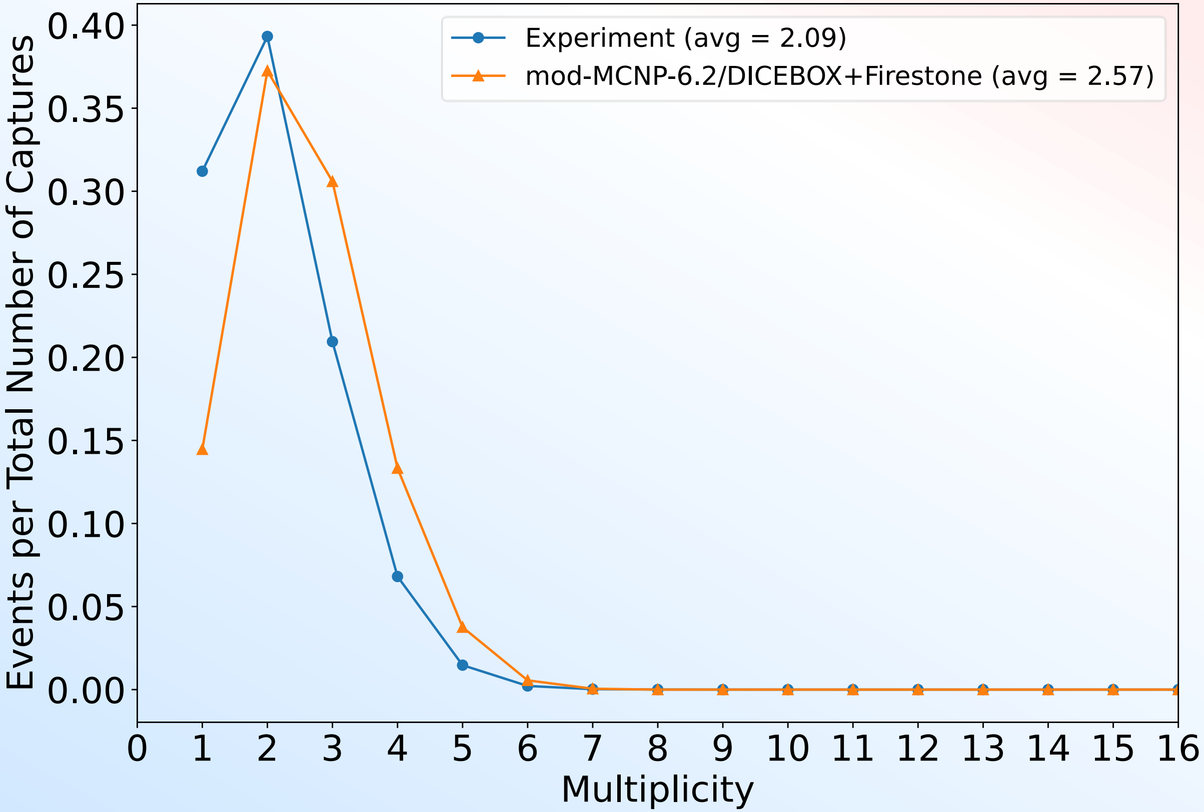


^{56}Fe(n,γ) Multiplicity Distribution

100 keV LLD on Experiment and Simulation



250 keV LLD on Experiment and Simulation



- The experimental multiplicity curve seems to have lower average
- Need to find a way to add a constraint in DICEBOX calculations

Improving DICEBOX Cascades

^{55}Mn Thermal Neutron Capture

DICEBOX/ENSDF

Models full γ-ray cascades using evaluated nuclear data

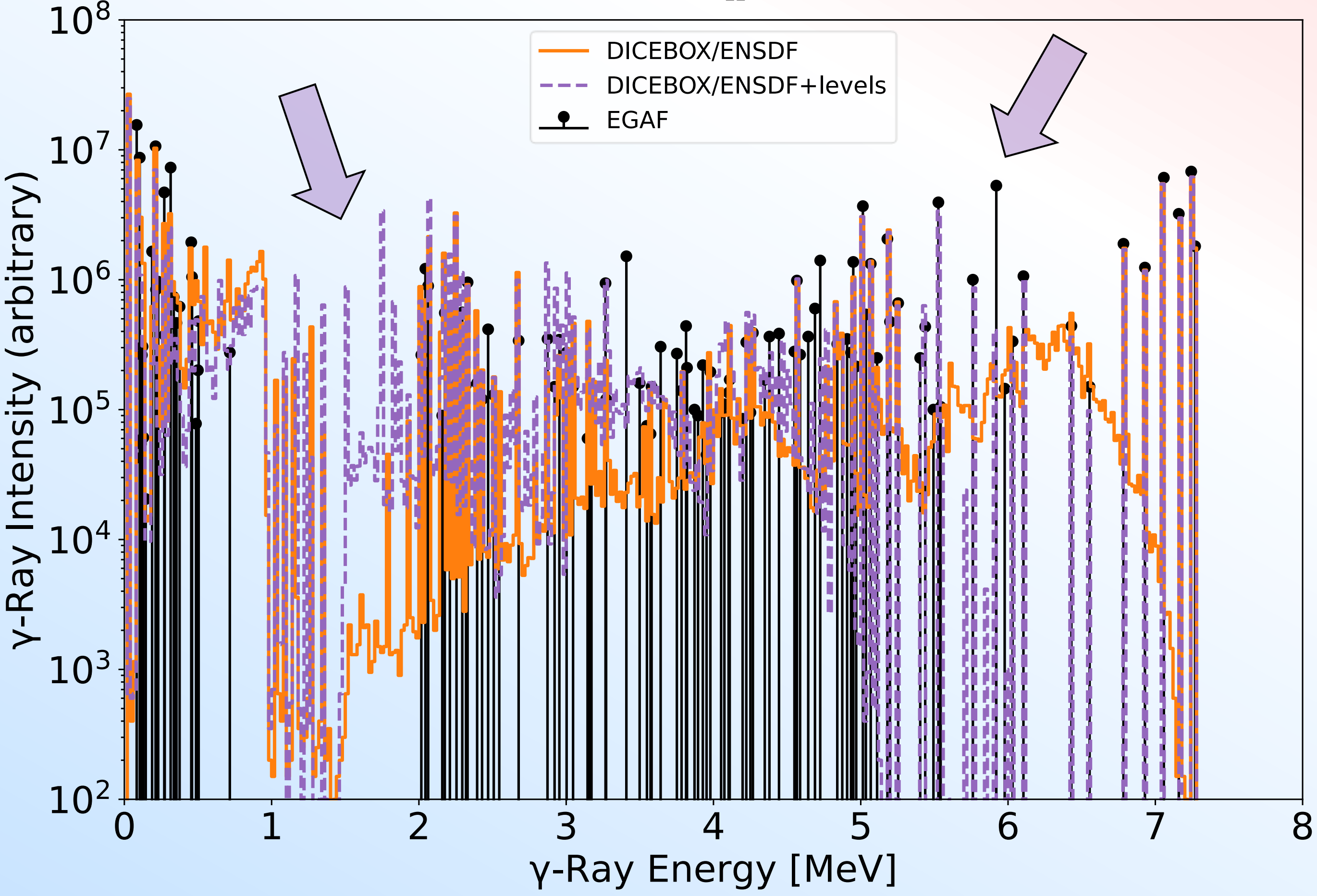
DICEBOX/ENSDF+levels

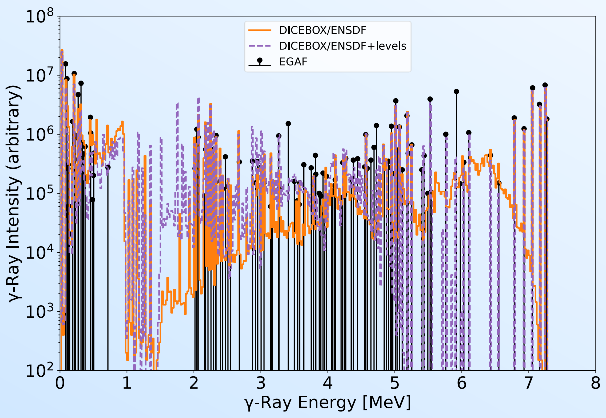
Models full γ-ray cascades using evaluated nuclear data + **additional levels that DICEBOX previously excluded**

EGAF

Experimentally measured γ-ray intensities

^{55}Mn(n,γ), E_n = thermal

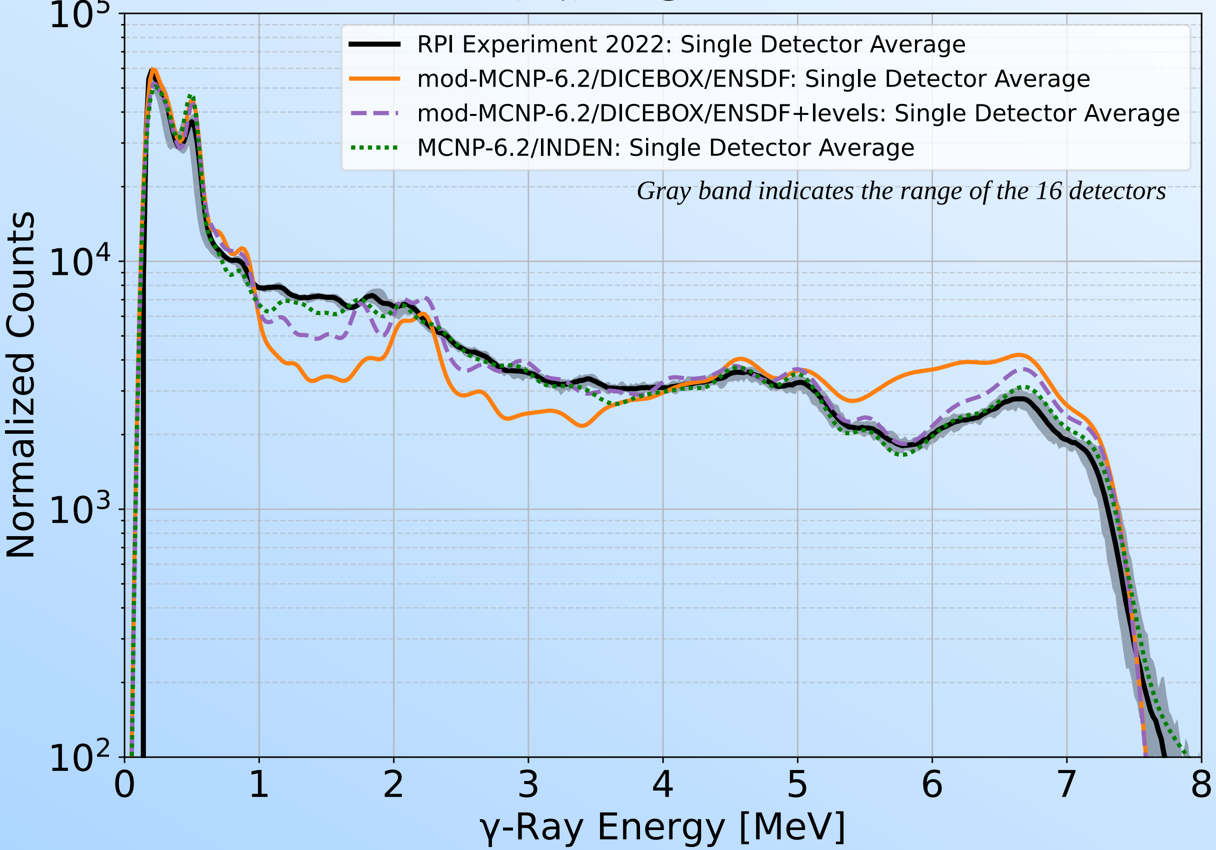




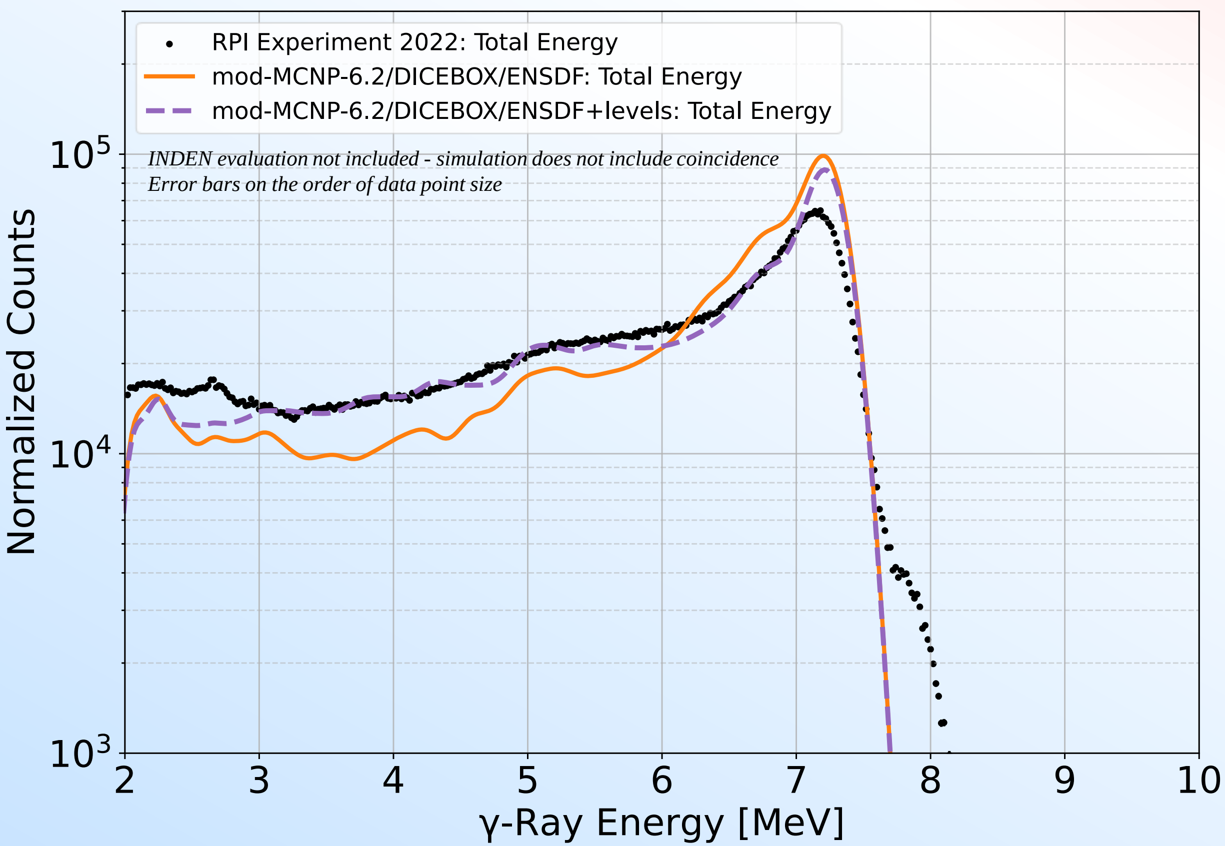
Improving DICEBOX Cascades

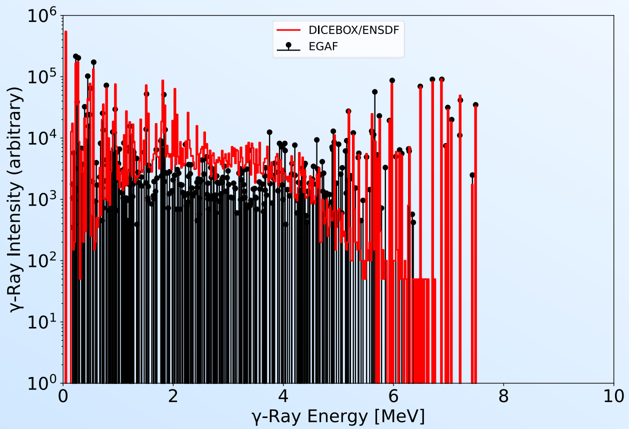
^{55}Mn Thermal Neutron Capture

^{55}Mn(n,γ) Single Detector



^{55}Mn(n,γ) Total Energy Deposition (all 16 detectors)

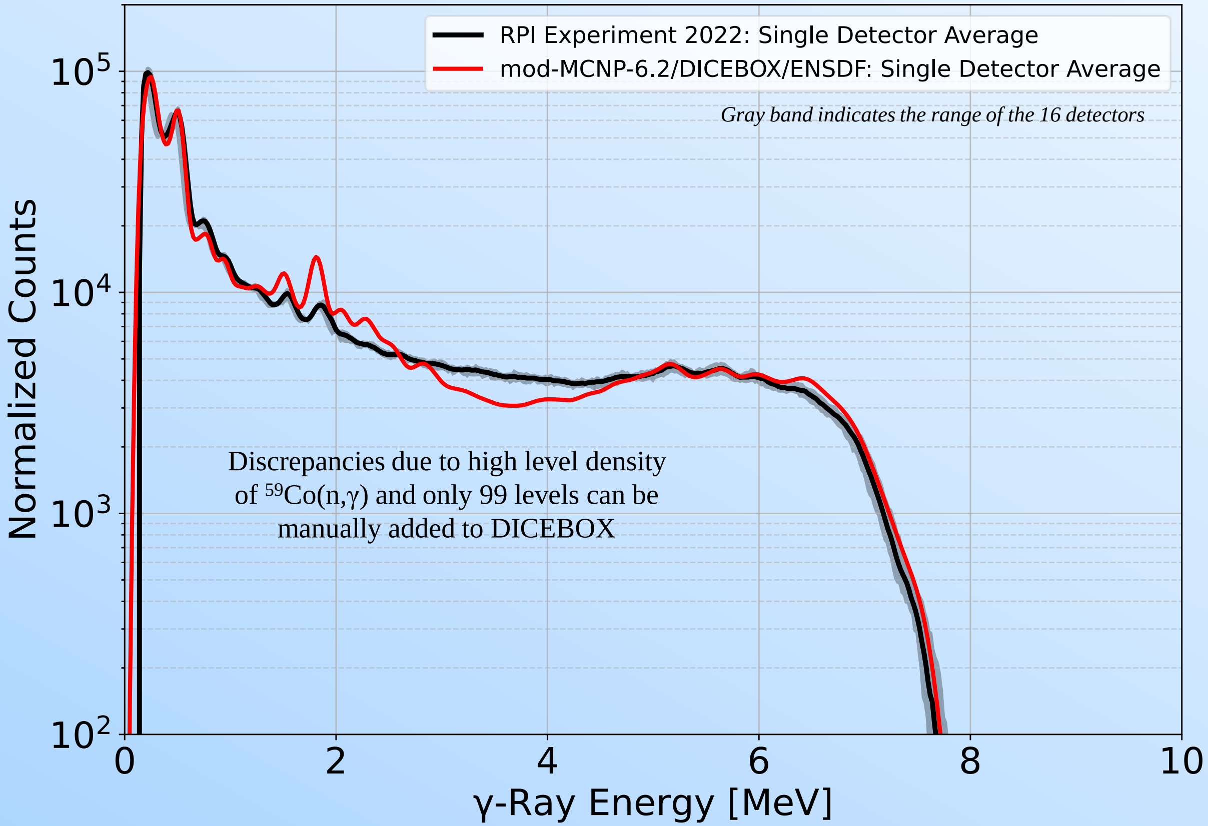




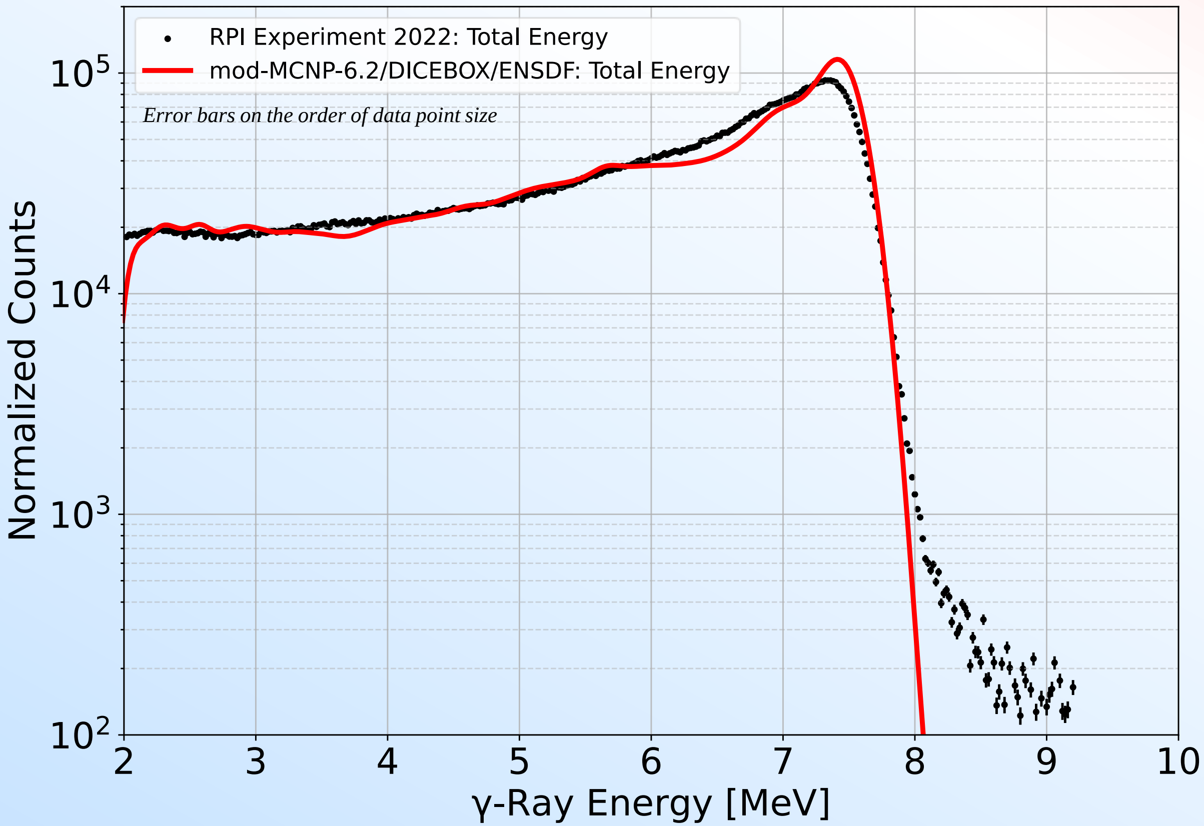
Analysis of Other Isotopes

^{59}Co Thermal Neutron Capture

^{59}Co(n,γ) Single Detector



^{59}Co(n,γ) Total Energy Deposition (all 16 detectors)



Initial work on ^{238}U and ^{235}U Neutron Capture Spectra

^{nat}U Analysis: Thermal Region Capture Measurement

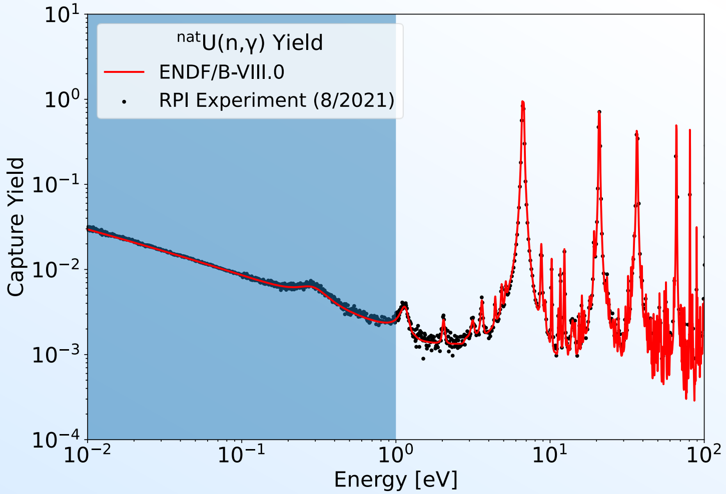
- Sample: 20 mil ^{nat}U
- Neutron Energy: 0.01 – 1 eV
- Total γ-Ray Energy Deposition: 2 – 20 MeV

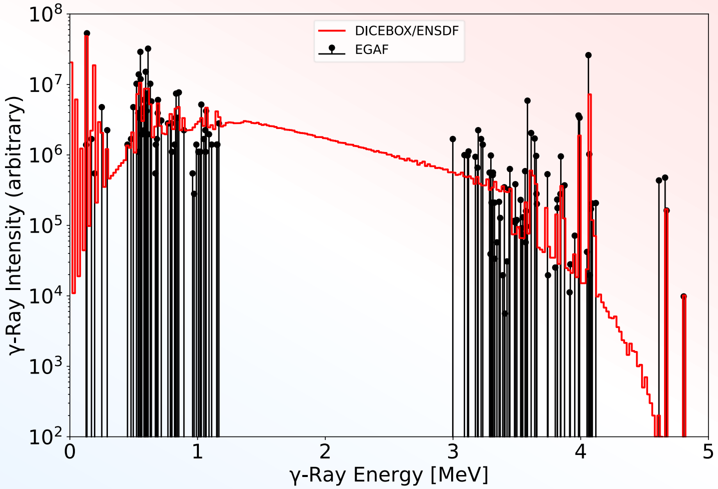
- ^{238}U thermal capture cascades from DICEBOX compared to EGAF

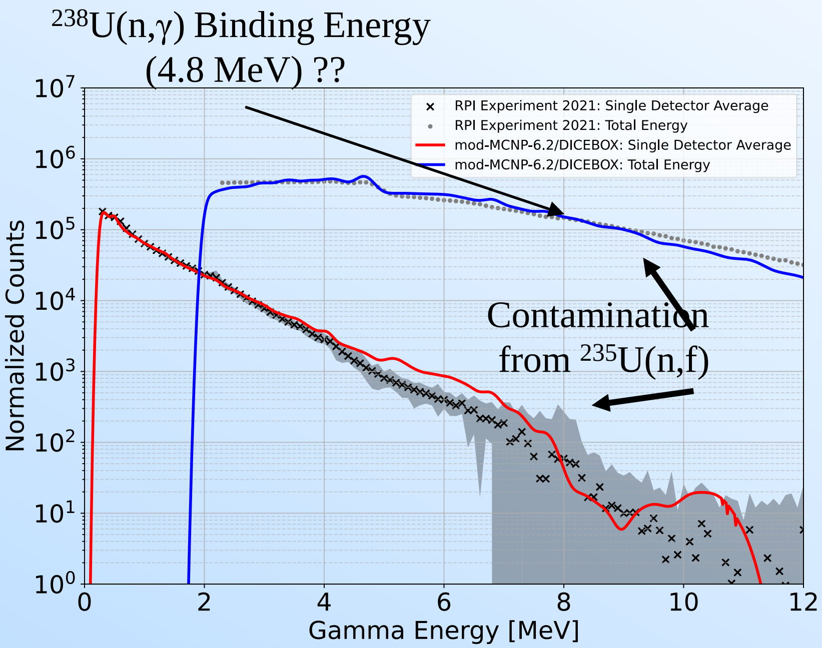
- Other calculated capture fission γ-rays are generated using CGM

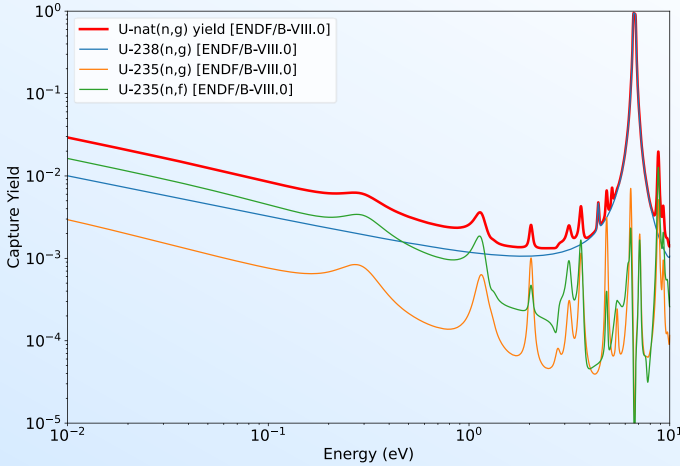
- Thermal γ-rays spectra is dominated by γ-rays produced from ^{235}U(n,f) reaction

- **Use resonance structure to isolate capture and fission spectra**



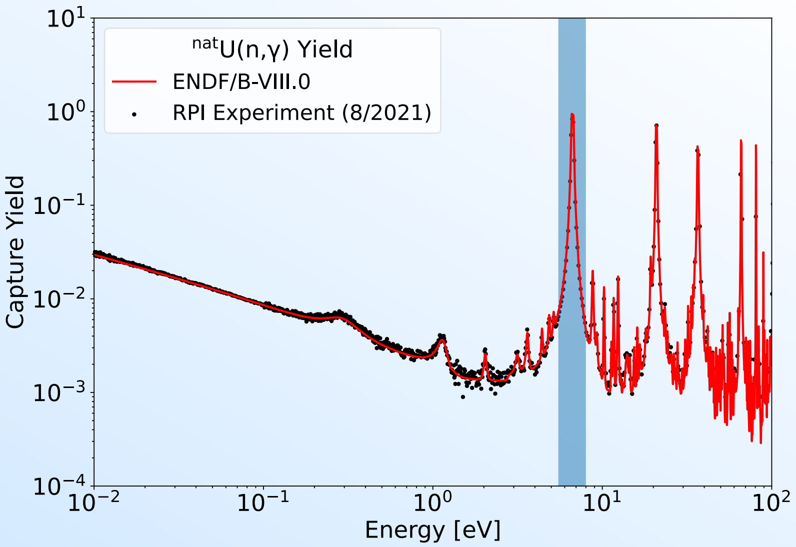


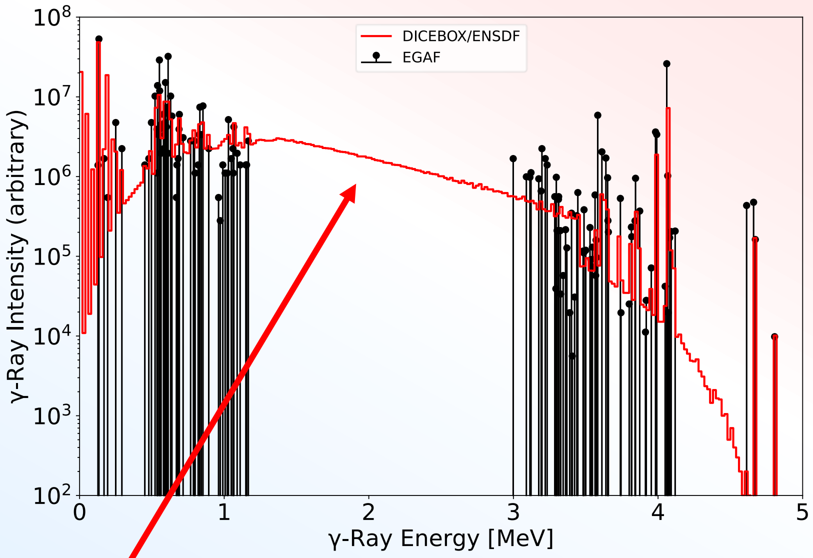


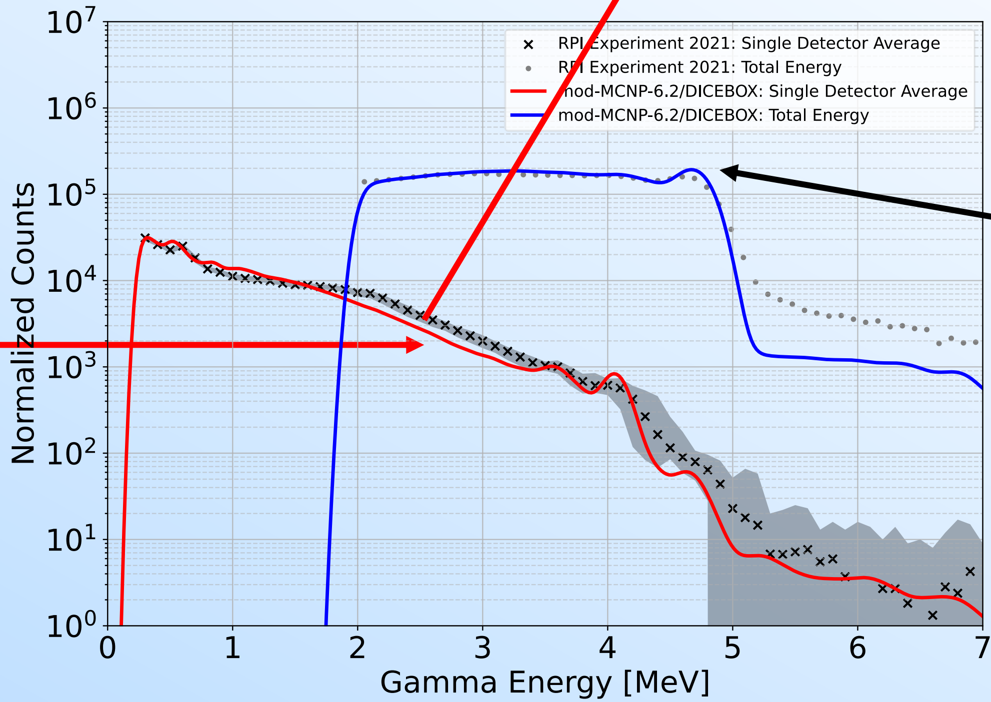


natU Analysis: Capture Resonance γ-Ray Spectra

- Use neutron energies in the first large capture resonance to compare γ-ray spectra
  - Contributions from U-235 fission is very small
  - Neutron Energy: 5.5 – 8.0 eV (6.67 eV resonance)
- Total γ-Ray Energy Deposition: 2 – 20 MeV
- ^{238}U thermal capture cascades from DICEBOX
- Note differences between measurements and simulation when primary gammas seem to be missing

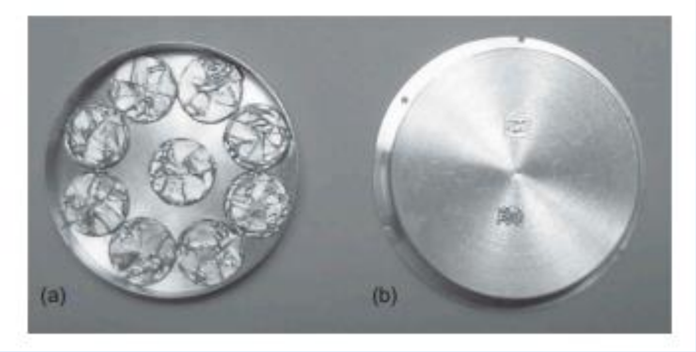






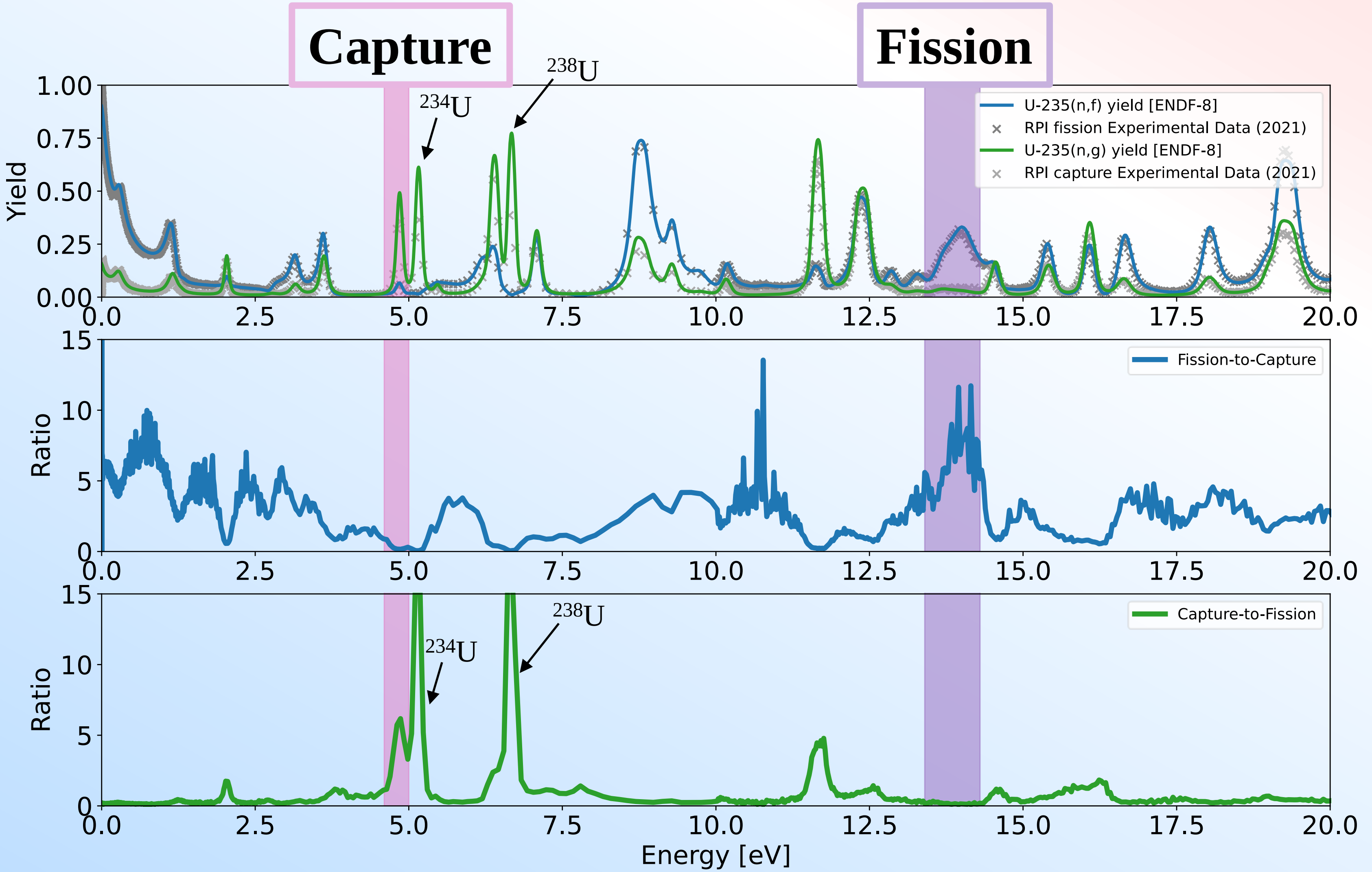
^{238}U(n,γ) Binding Energy (4.8 MeV)

^{235}U Analysis: Compare γ-Ray Spectra from Resonances Dominated by Capture/Fission



Y. Danon, et. al., "**Simultaneous Measurement of ^{235}U Fission and Capture Cross Sections From 0.01 eV to 3 keV Using a Gamma Multiplicity Detector**", *Nuclear Science and Engineering*, vol. 187, no. 3, pp. 291-301, (2017).

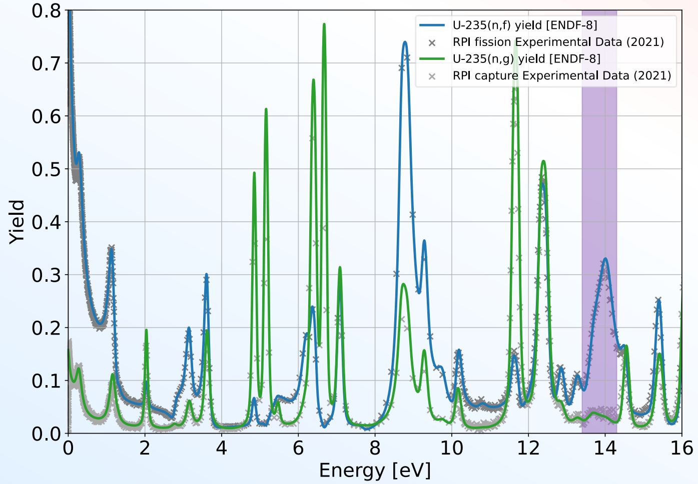
- Sample (above) used in previous ^{235}U neutron capture measurement at RPI
- Use two methods to separate fission and capture γ-ray spectra
    1. Normalization by eye (assume fission does not include capture)
    2. Include the contribution of each reaction (fission in capture and capture in fission)

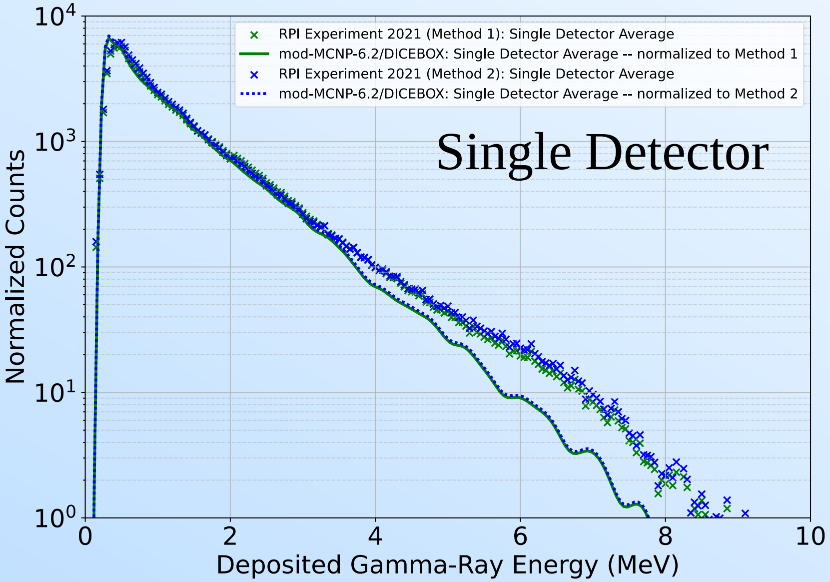


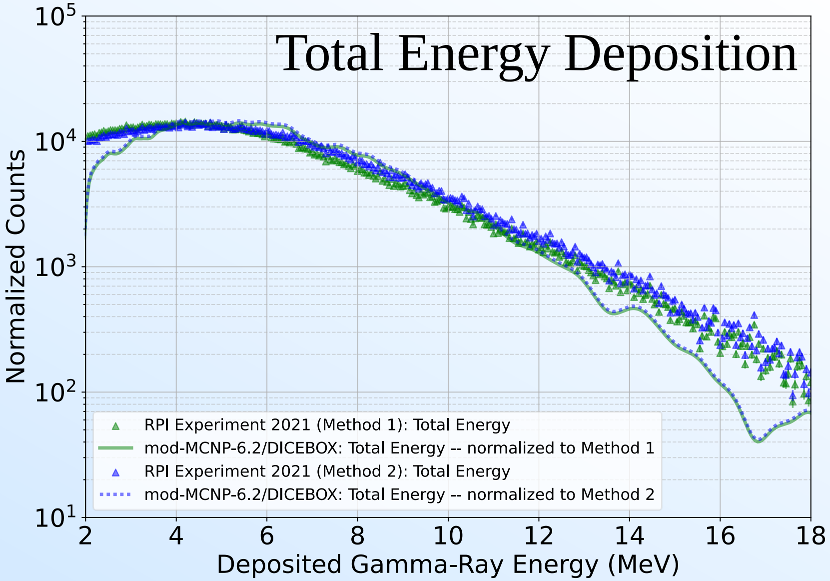
Note: capture resonances from impurities of ^{234,238}U are shown

^{235}U Analysis: Fission Resonance γ-Ray Spectra

- Use resonance structure to isolate ^{235}U fission-dominated spectra
  - Neutron Energy: 13.4 – 14.4 eV
  - Total γ-Ray Energy Deposition: 2 – 20 MeV
- *Corrected* single detector and total γ-ray spectra compared to mod-MCNP-6.2/DICEBOX (fission induced γ rays are generated using CGMF)
  - **No correction - assume fission resonance has no contamination from capture γ rays**
  - **Ratio of the contribution of each reaction (removes small amount of capture in the fission spectrum)**
- **For fission spectra the two methods give about the same result**

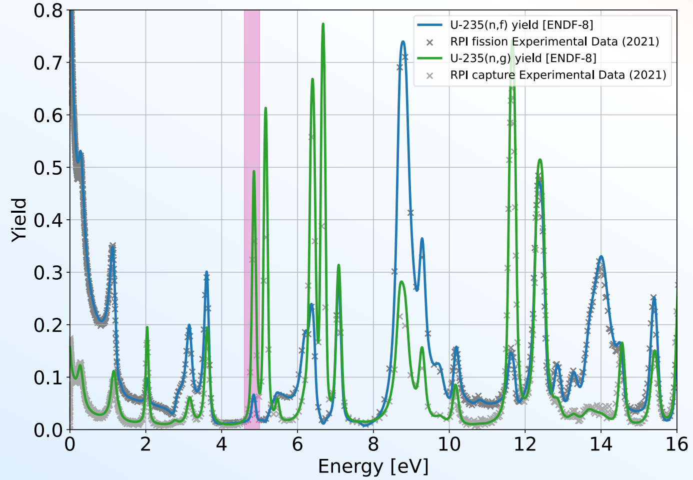


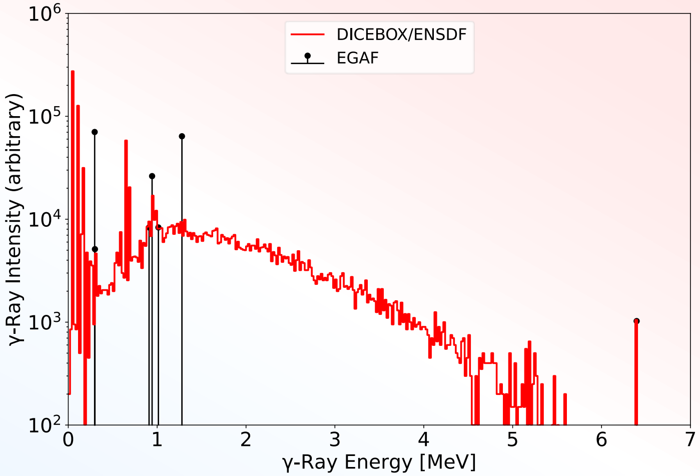


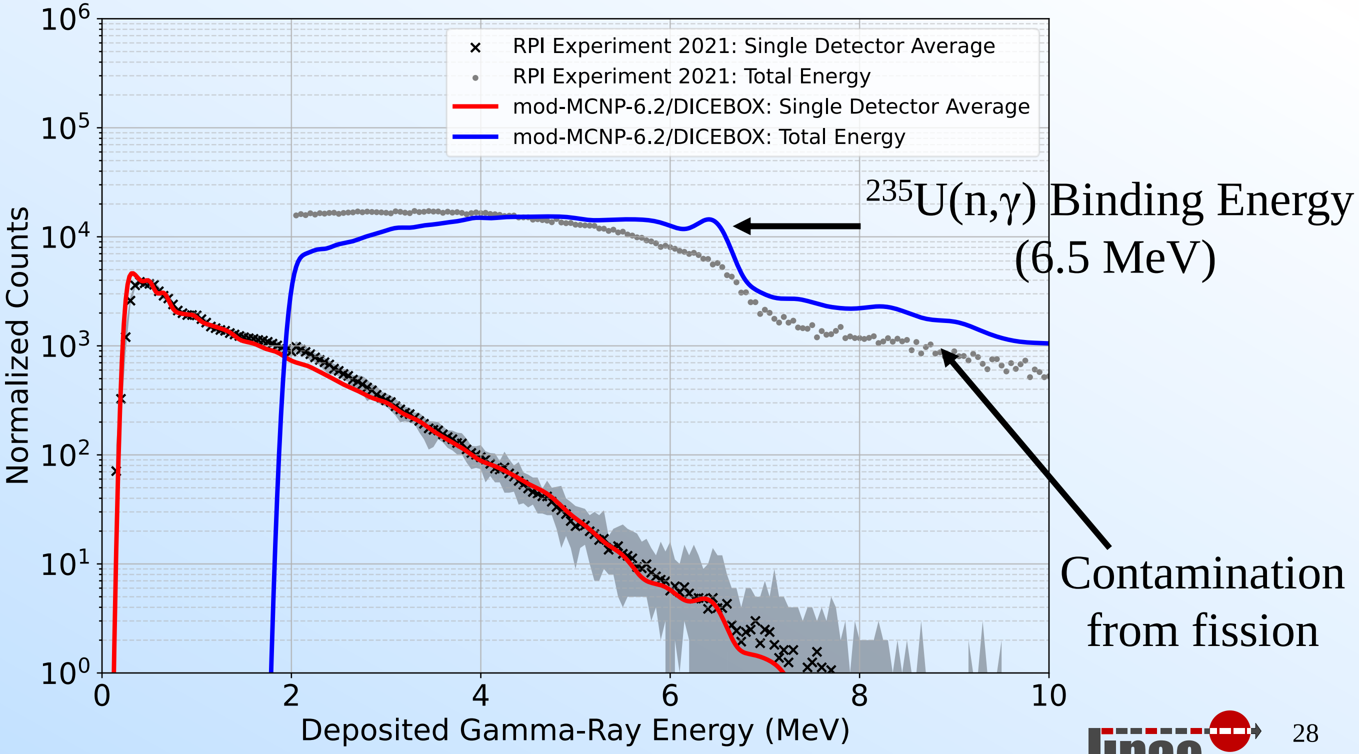


^{235}U Analysis: Capture Resonance γ-Ray Spectra

- Use resonance structure to isolate ^{235}U capture-dominated spectra
  - Neutron Energy: 4.6 – 5.1 eV
  - Total γ-Ray Energy Deposition: 2 – 20 MeV
- Capture gamma data available for ^{235}U is very sparse
- ^{235}U thermal capture cascades from **DICEBOX/ENSDF** compared to **EGAF**
- Coincidence (**total energy spectra**) shows evidence of background fission γ rays that need to be subtracted
- **Use the two methods to correct for this!**



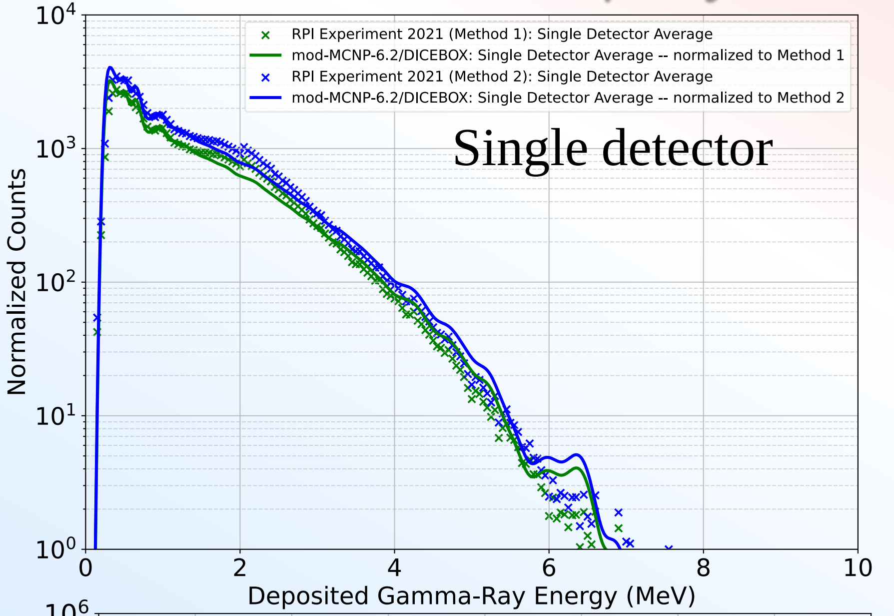


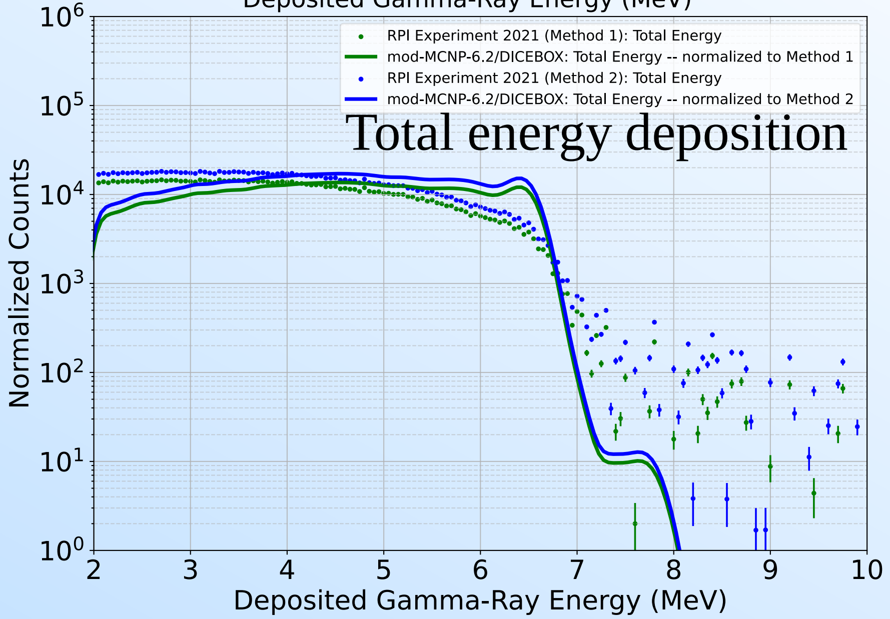


^{235}U Analysis: Capture Resonance γ-Ray Spectra

Corrections to remove contribution from fission γ rays

- Capture mostly spectra
  - Neutron Energy: 4.6 – 5.1 eV resonance
  - Total γ-Ray Energy Deposition: 2 – 20 MeV
- ^{235}U thermal capture cascades from **DICEBOX/ENSDF** compared to **EGAF**
- *Corrected* single detector and total γ-ray spectra compared to mod-MCNP-6.2/DICEBOX (simulation uses NONU card to remove fission γ rays)
  - **Normalization by eye**
  - **Ratio of the contribution of each reaction**
- Single detector response simulated well
- Total energy spectrum missing binding energy peak
  - needs more work

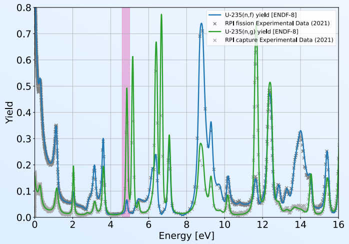


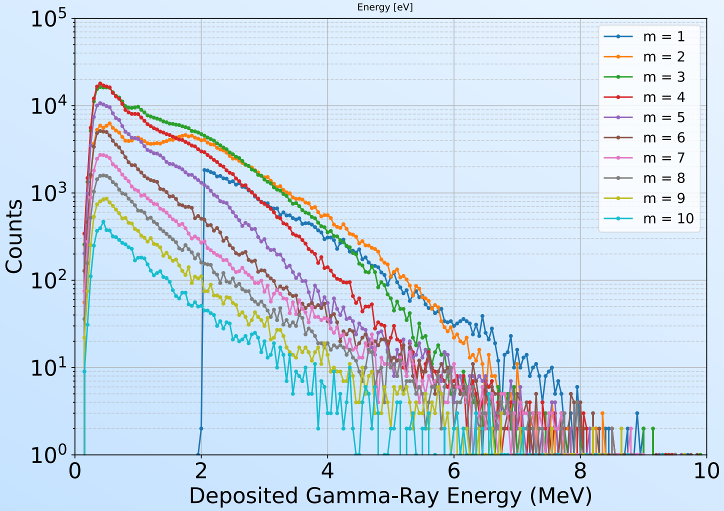


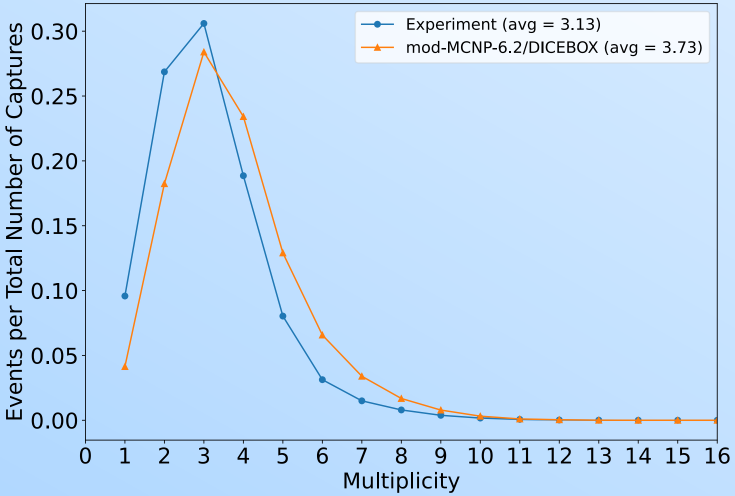
^{235}U(n,γ) Multiplicity Spectra

As expected, measured multiplicity is higher for the fission resonance

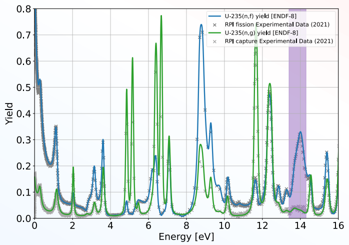
Capture

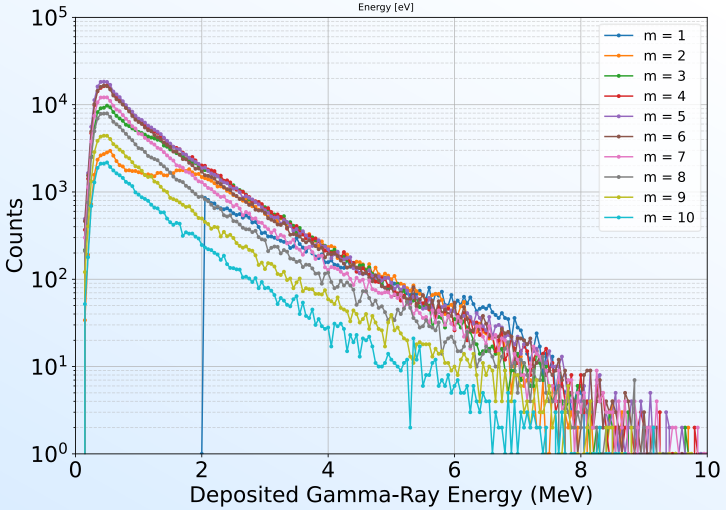


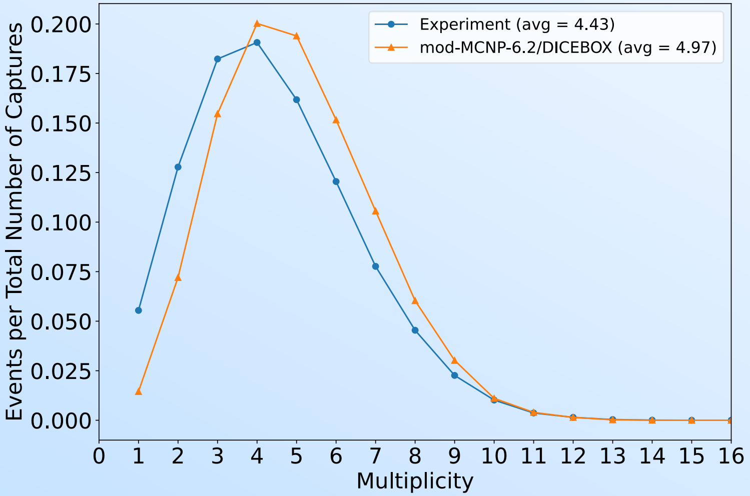




Fission







Conclusions and outlook

- **Demonstrated** that the multiplicity detector can measure event-by-event capture cascade data in coincidence mode in the thermal and resolved resonance region.
- **Developed** a modified version of MCNP that can read capture cascades, transport the gammas, and produce an event-by-event data file.
- Comparison of experiment and simulations showed that when the capture gamma cascade is well known, good agreement is obtained.
- Differences between experiment and simulation provides information on where the evaluated data can be improved
- **The project provides experimental validation and thus helps produce better capture gamma evaluations**

- **Next step** is to create a useable validation or benchmark suite
  - Quantify the uncertainties in both experiment and simulation
  - Create a definition of a benchmark for this type of data
  - Create benchmark suites that includes the experimental data and computation tool to compare with simulations
  - Find the best path to disseminate the data and codes

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