Recent Nuclear Data Activity at the RPI Gaerttner LINAC Center

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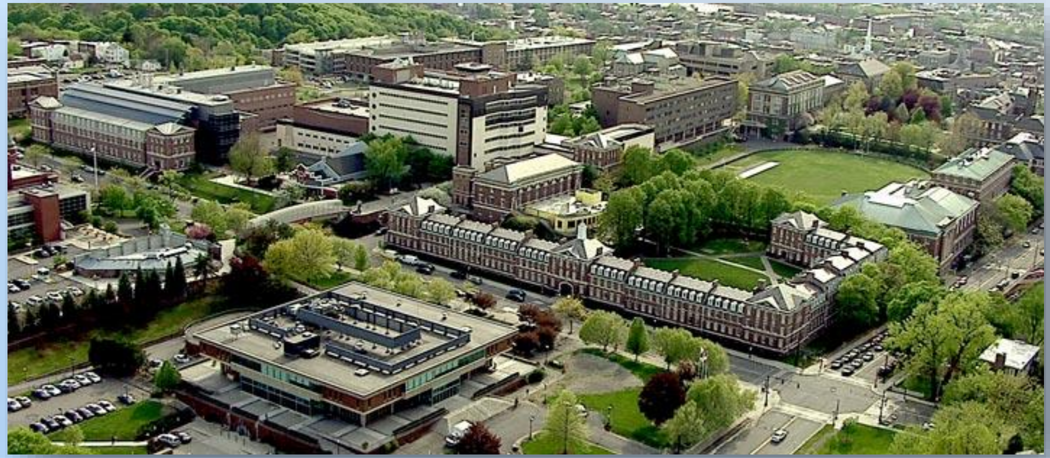

WONDER 2023, 6th International Workshop on Nuclear Data Evaluation for Reactor Applications, June 5-9, 2023
Aix-en-Provence, France

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Outline

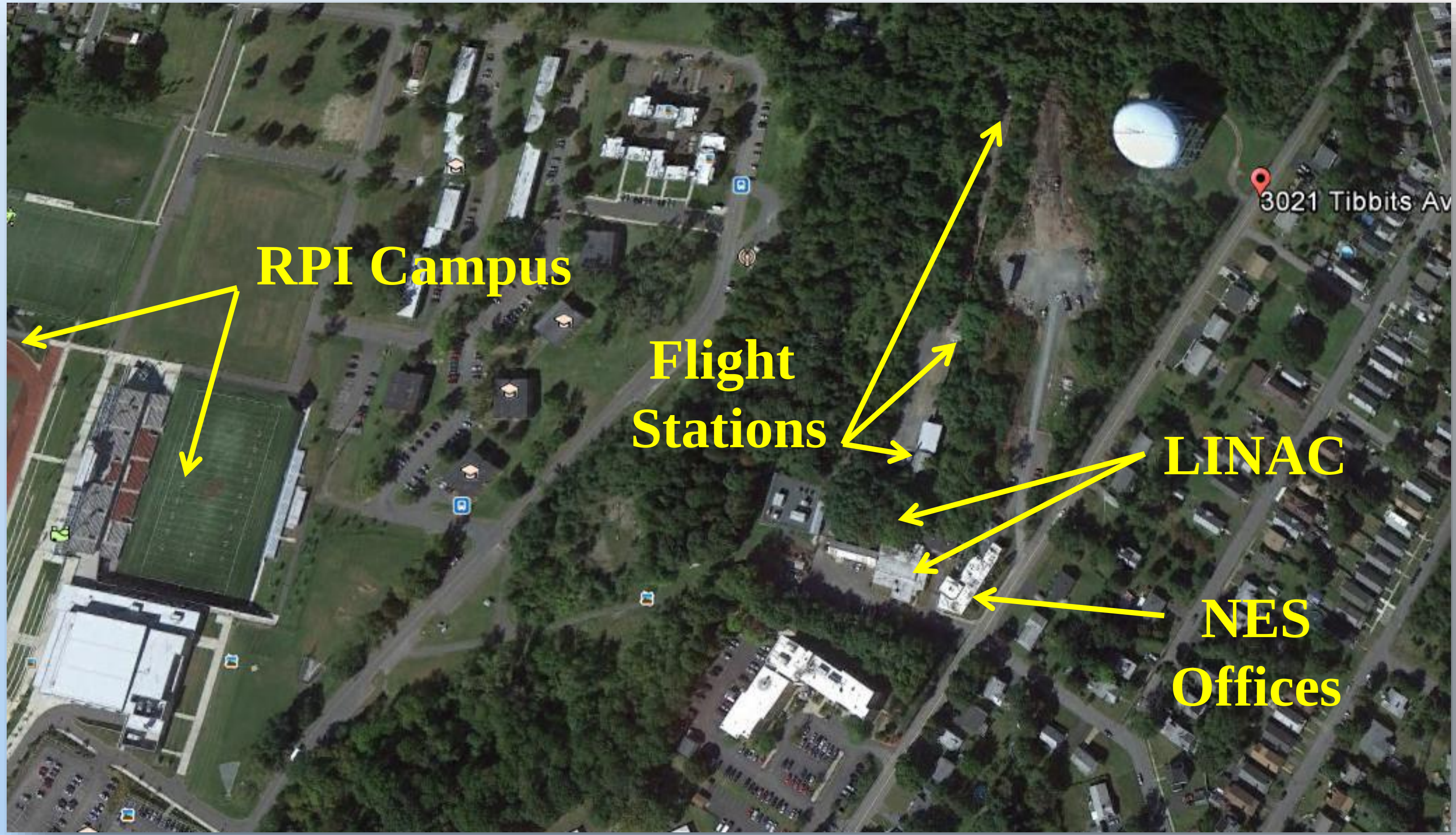
- Gaerttner LINAC center overview
- Thermal cross section measurements (**Dominik Frits**)
- ^{54}Fe Resonance region measurements and evaluation (**Sukhjinder Singh**)
- Capture γ-spectra measurements (**Katelyn Cook**)

Motivation

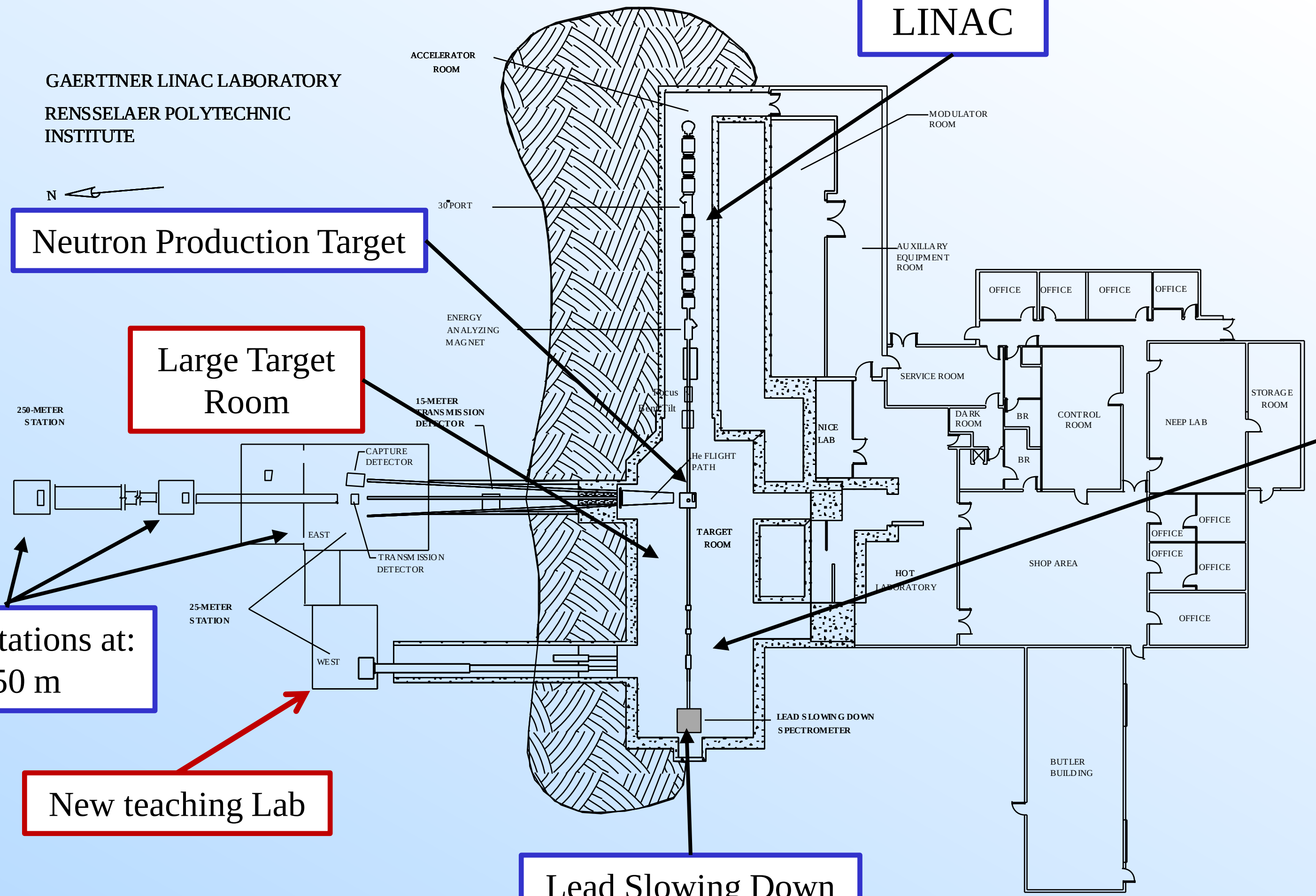
- Driven by 60 MeV electron LINAC
- The RPI Nuclear Data research group is dedicated to development and execution of novel and accurate nuclear data measurements for the improvement of data used in applications
  - Mostly related to nuclear reactor design and analysis, and criticality safety
  - Mostly neutron induced reactions
- There are other research areas that utilize the same accelerator

Where is the RPI Gaerttner LINAC Center?

It is on the highest point in Troy, NY



The RPI Gaerttner LINAC Facility



Electron LINAC

Neutron Production Target

Large Target Room

Detection Stations at: 15 - 250 m

New teaching Lab

Lead Slowing Down Spectrometer

Target room

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Current LINAC Specifications

	Three Sections (Low Energy Port)	Nine Sections (High Energy Port)
Electron Energy	5 to 25 MeV	25 to over 60 MeV
Pulse Width	6 to 5000 ns	6 to 5000 ns
Peak Current	3A (short pulse: 6 to 50 ns) 400 mA (long pulse: 50 to 5000 ns)	3A (short pulse: 6 to 50 ns) 400 mA (long pulse: 50 to 5000 ns)
Average Power	10 kw@ 17 MeV, 5000 ns	>10 kw@ 60 MeV, 5000 ns
Peak Dose Rate	$>10^{11}$ Rads/sec (in Silicon)	n/a
Neutron Production	n/a	**~4 X 10^{13} neutrons/sec**
Pulse Repetition Rate	Single pulse to 500 pps (short pulse) Single pulse to 300 pps (long pulse)	Single pulse to 500 pps (short pulse) Single pulse to 300 pps (long pulse)

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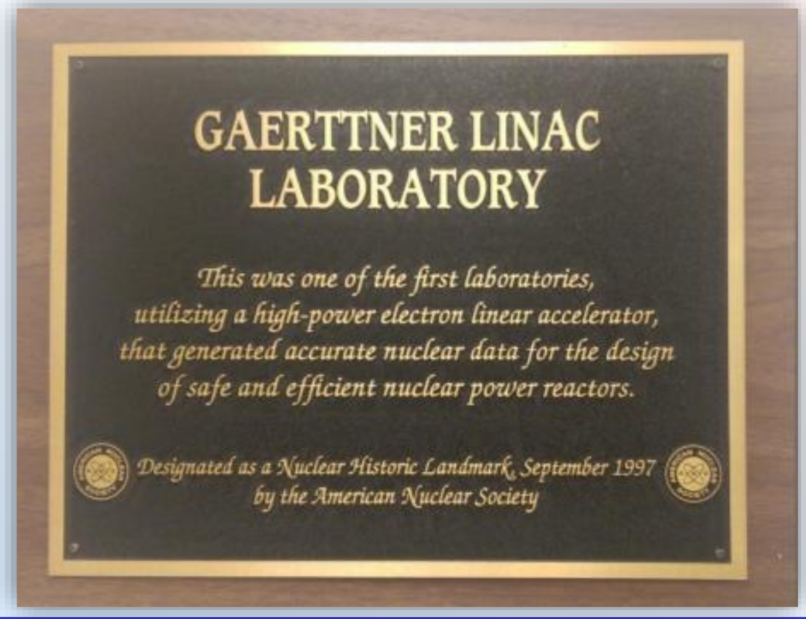
RPI LINAC History

December 1961 - The RPI LINAC started operation Working "continuously" since.

September 1997- LINAC was designated as Nuclear Historic Landmark by the American Nuclear Society



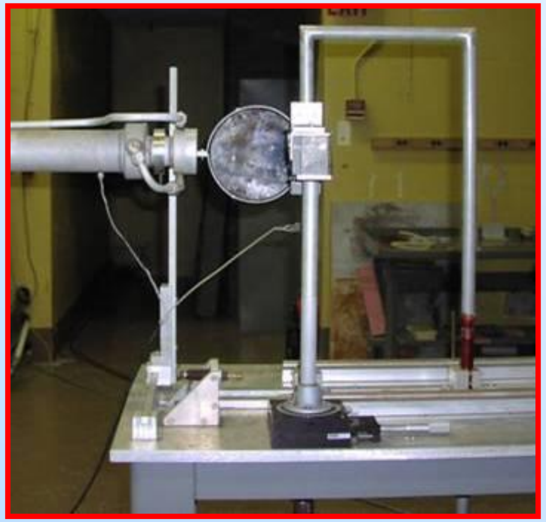
Graduated over 190 students who utilized the LINAC as part of their graduate thesis research

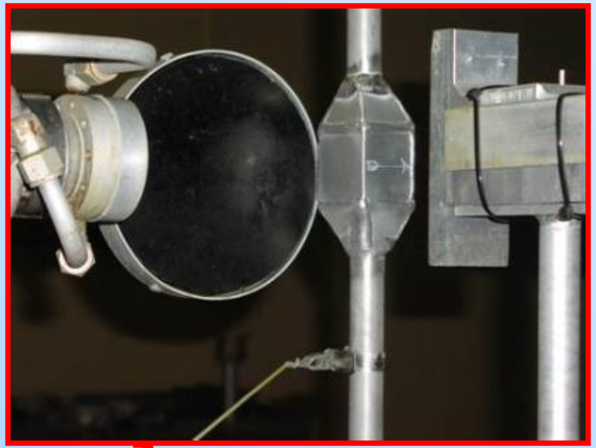
Many years of accumulated experience

2014 - Started a major refurbishment and upgrade project

Neutron Production Targets (electrons → neutrons)

Bare Bounce Target (BBT)





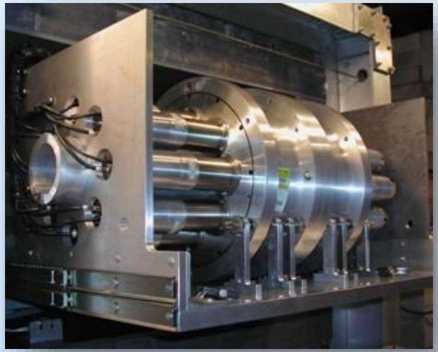
Enhanced Thermal Target (ETT)

PACMAN target

Detector Systems

Capture/Multiplicity

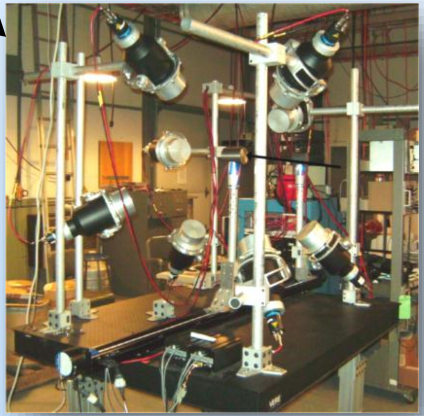
25m



45m

Neutron Scattering

keV Scattering @ 35m

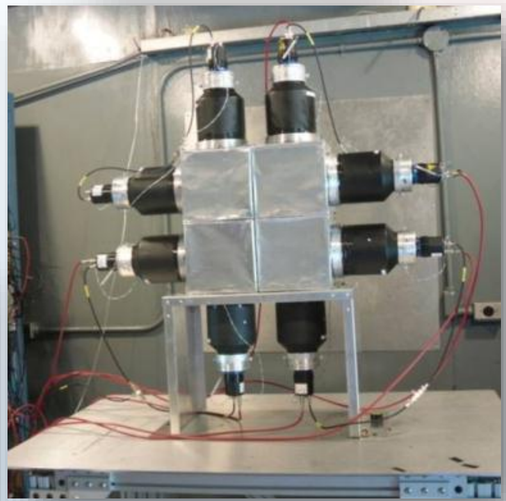


Fast Scattering and PFNS @ 30m

Transmission

35m

100m



250m

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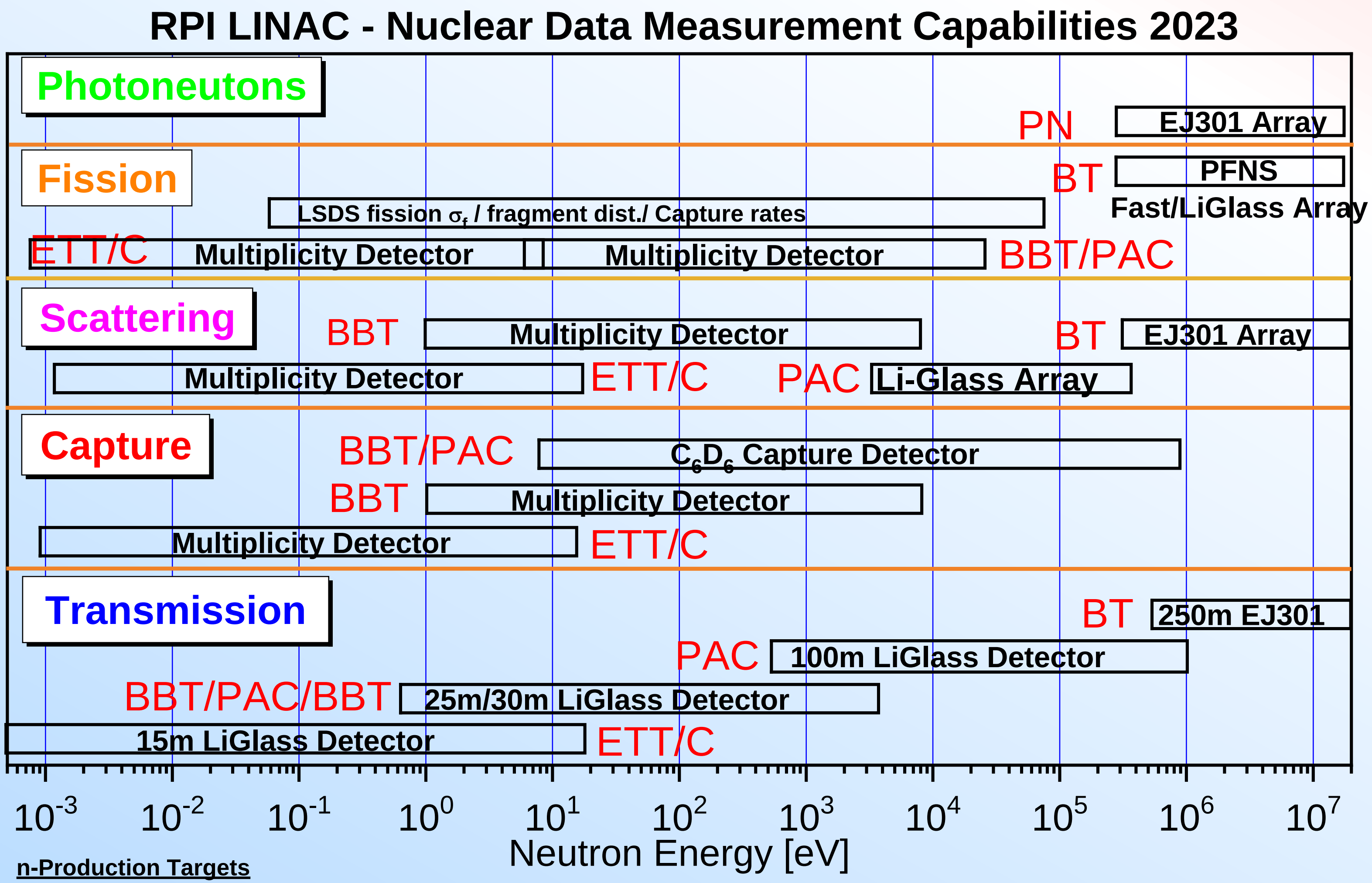
Capability Matrix and Development



n-Production Targets

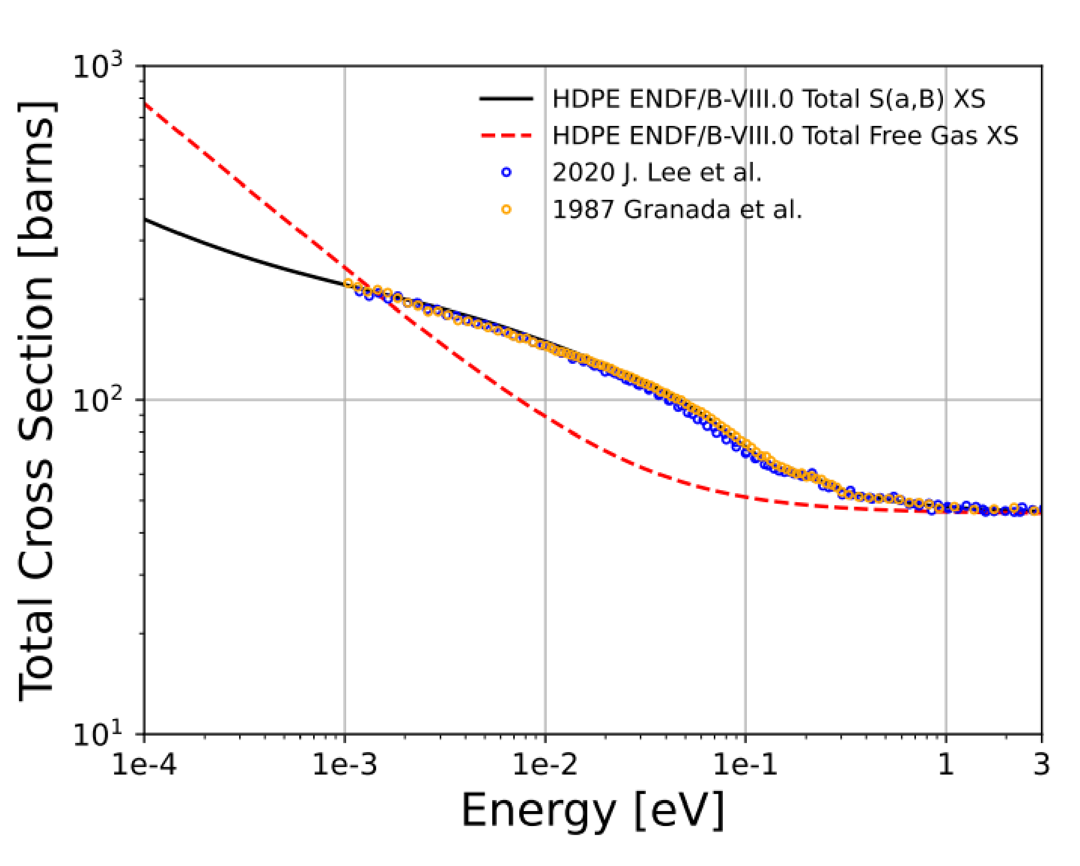
ETT- Enhanced Thermal Target
ETTC - ETT + cold moderator
BBT - Bare Bounce Target
PAC - PacMan Target
PN - Photoneutron target
BT- Bare Target on Axis

THERMAL CROSS SECTION MEASURMENTS

Primary Project Objectives

- Many materials lack high quality total cross section measurements in the 0.5 meV – 1 eV region required for validation of thermal scattering library/law (TSL) evaluations.
  - Examples: polystyrene, Lucite, and yttrium hydride
- Evaluations in this region need validation as thermal scattering can dramatically alter the neutron multiplication factor of a system.

Polyethylene Total Cross Section



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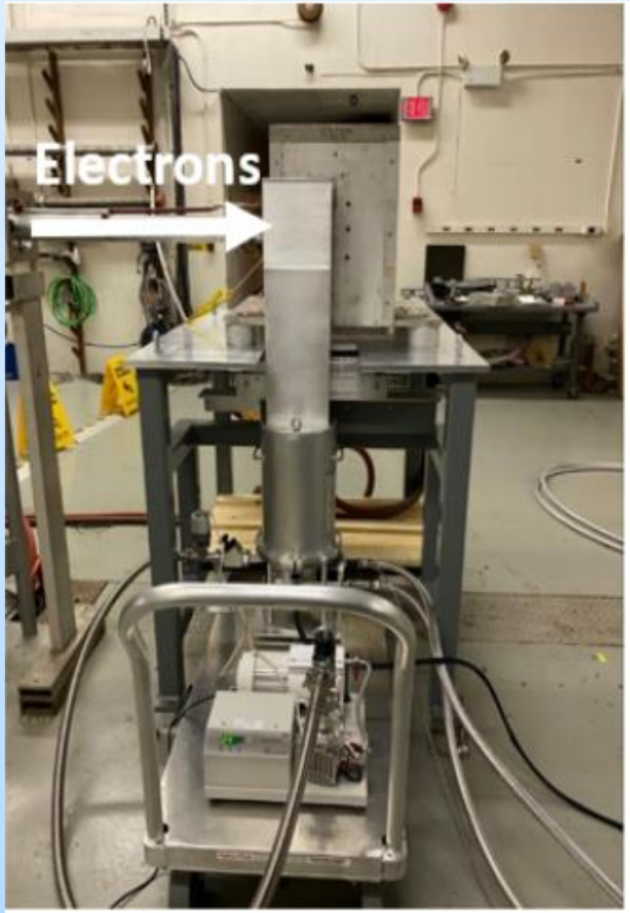
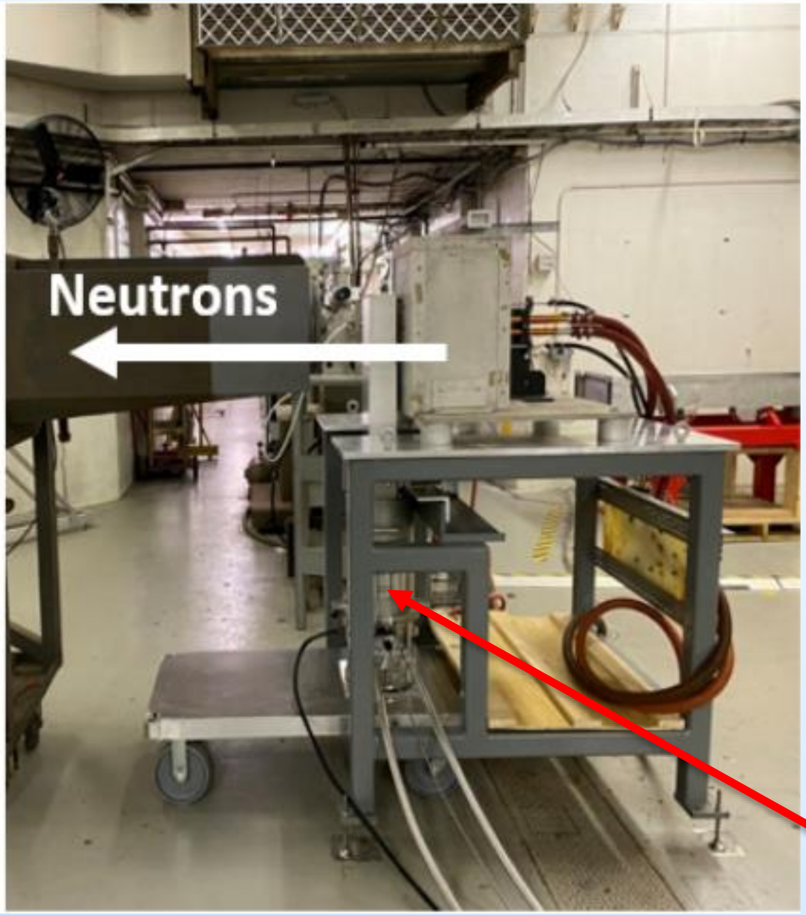
New Cold Neutron Add On



Front View
(detector view)



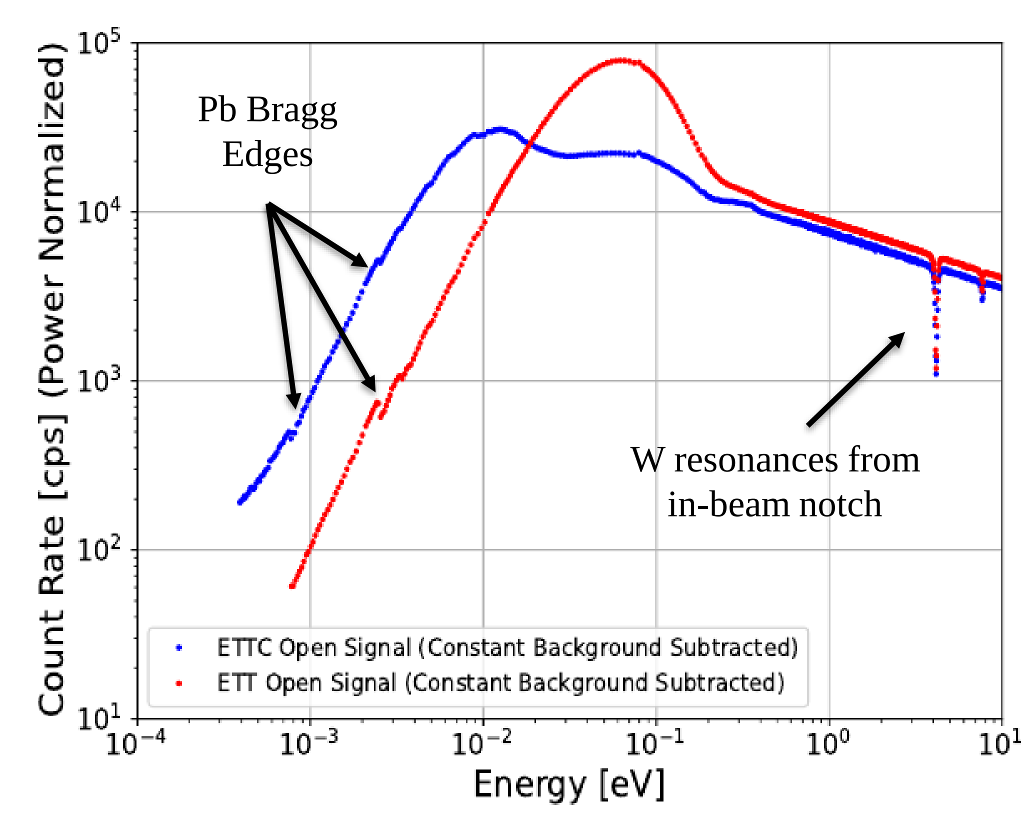
Side View

- Cold moderator is attached to cart that easily rolls in front of ETT target. This is the ETTC configuration.
- Once stable temperature, the vacuum pump is removed, and a helium flight tube is rolled into place.

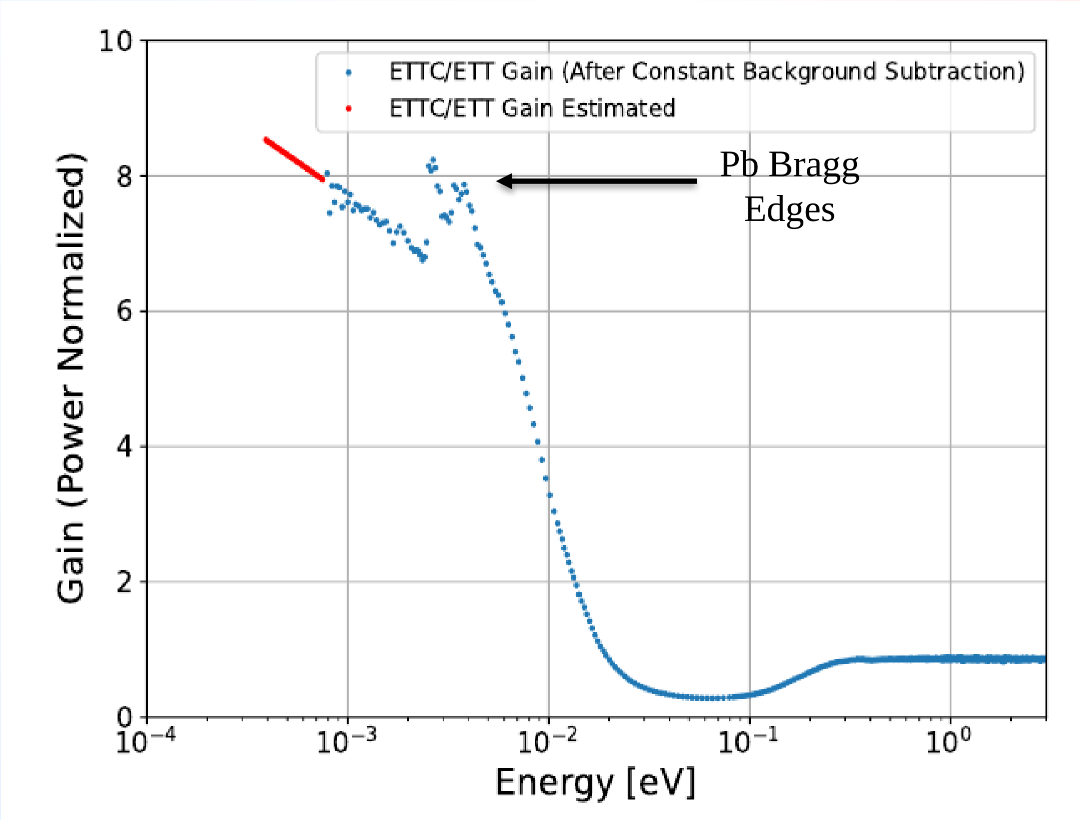
Cryostat rolls
into place

Neutron Intensity Improvement

Power Normalized Measured Flux Spectra For ETT & ETTC



ETTC/ETT Neutron Flux Gain



- Below 0.02 eV, **ETTC produces gain up to 8 over ETT after background subtraction.**

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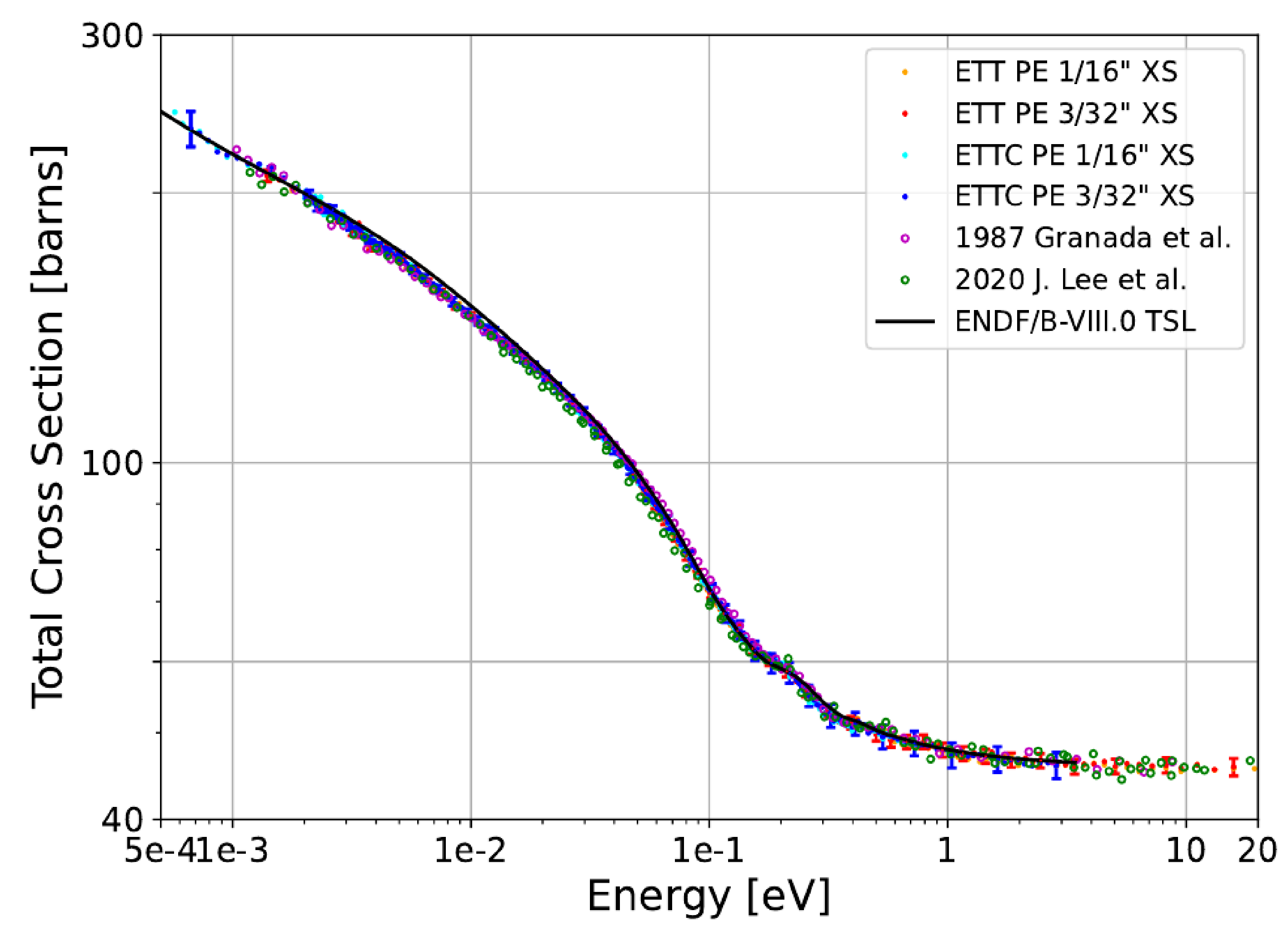
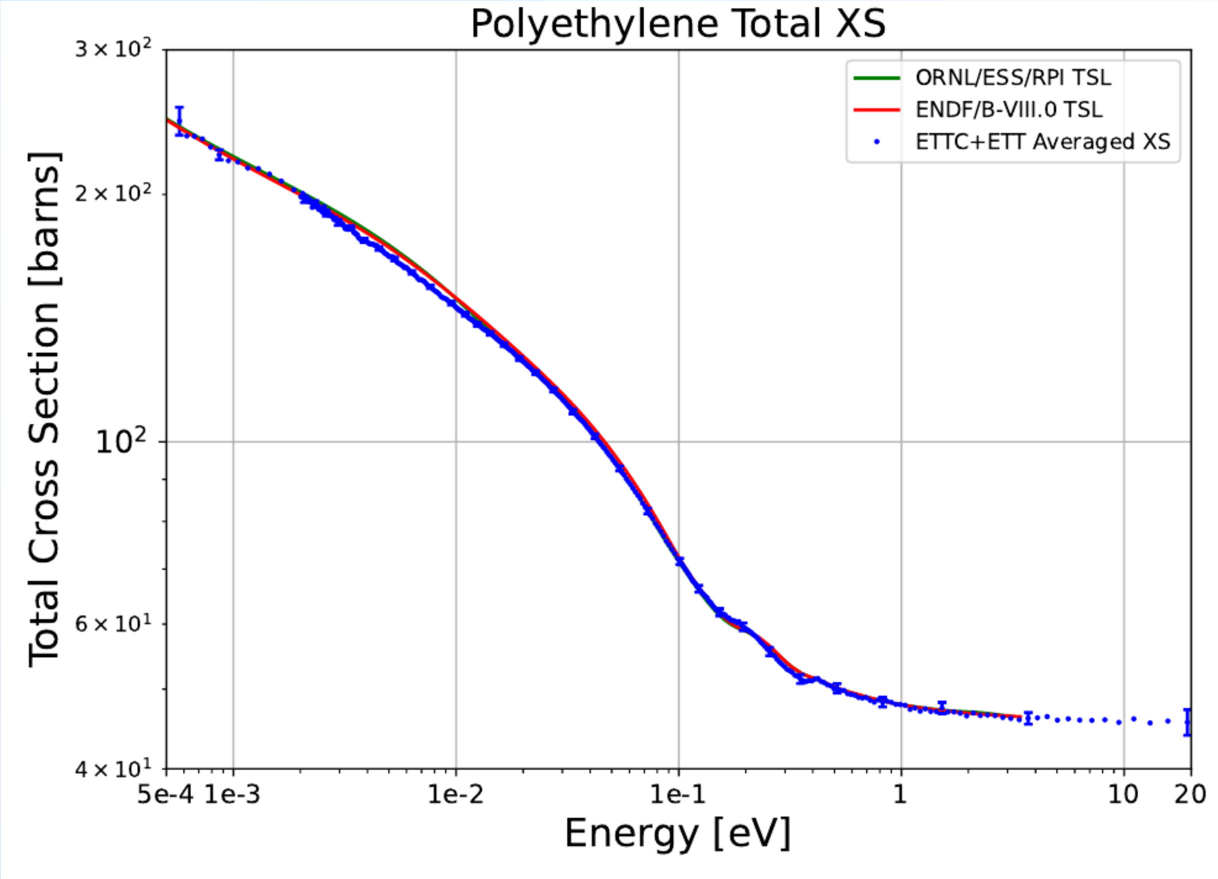
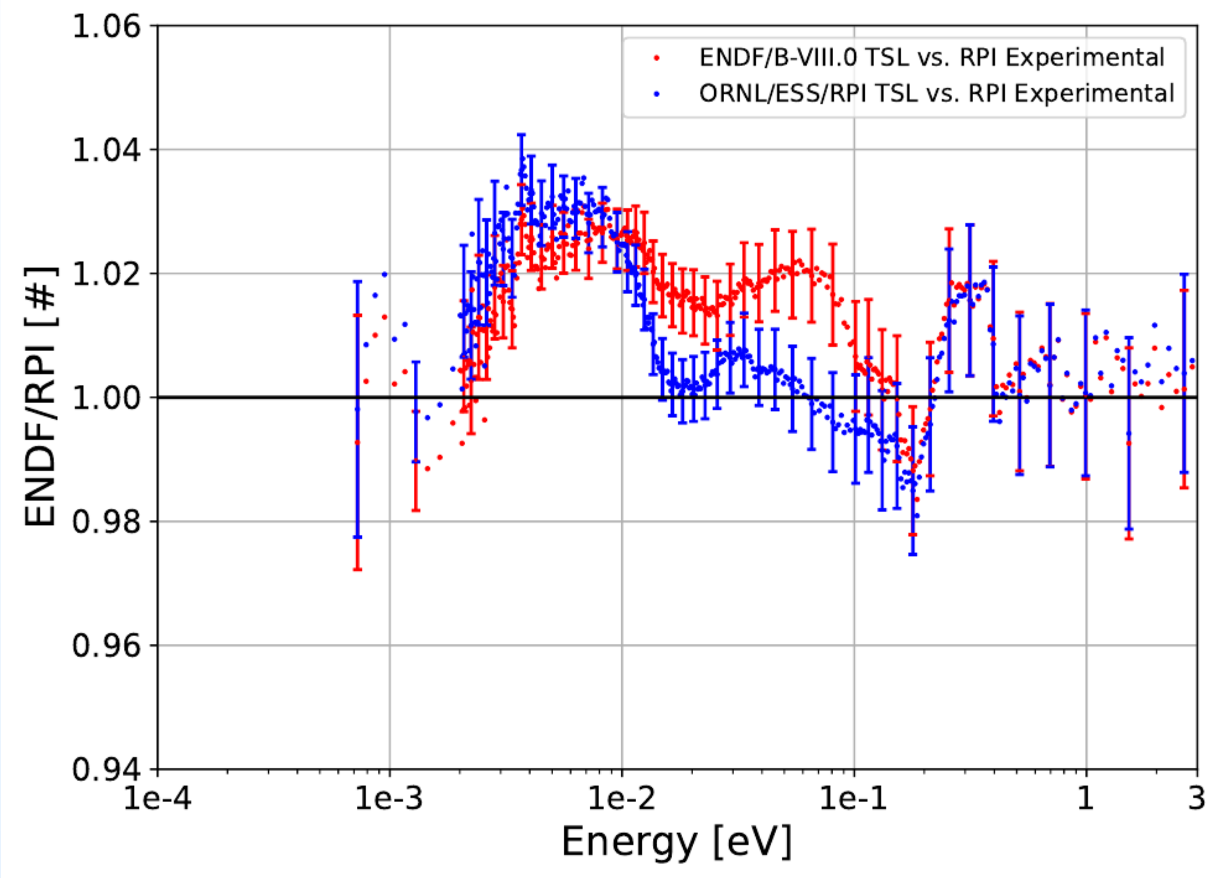
Polyethylene total cross section

- Excellent agreement between ETT and ETTC cross section for polyethylene (PE).
- Serves to validate ETTC system and measurement methodology.
- Good agreement between all RPI measured cross sections, other measured cross sections, and TSL evaluation.
- Experimental error bars account for all experimental sources of uncertainty and were calculated via a covariance matrix.



Polyethylene: averaged total cross section





- Weighted average of all polyethylene measurements performed
- In general, the ORNL/ESS/RPI TSL evaluation agrees with the measured cross section better than the ENDF/B-VIII.0 TSL evaluation.

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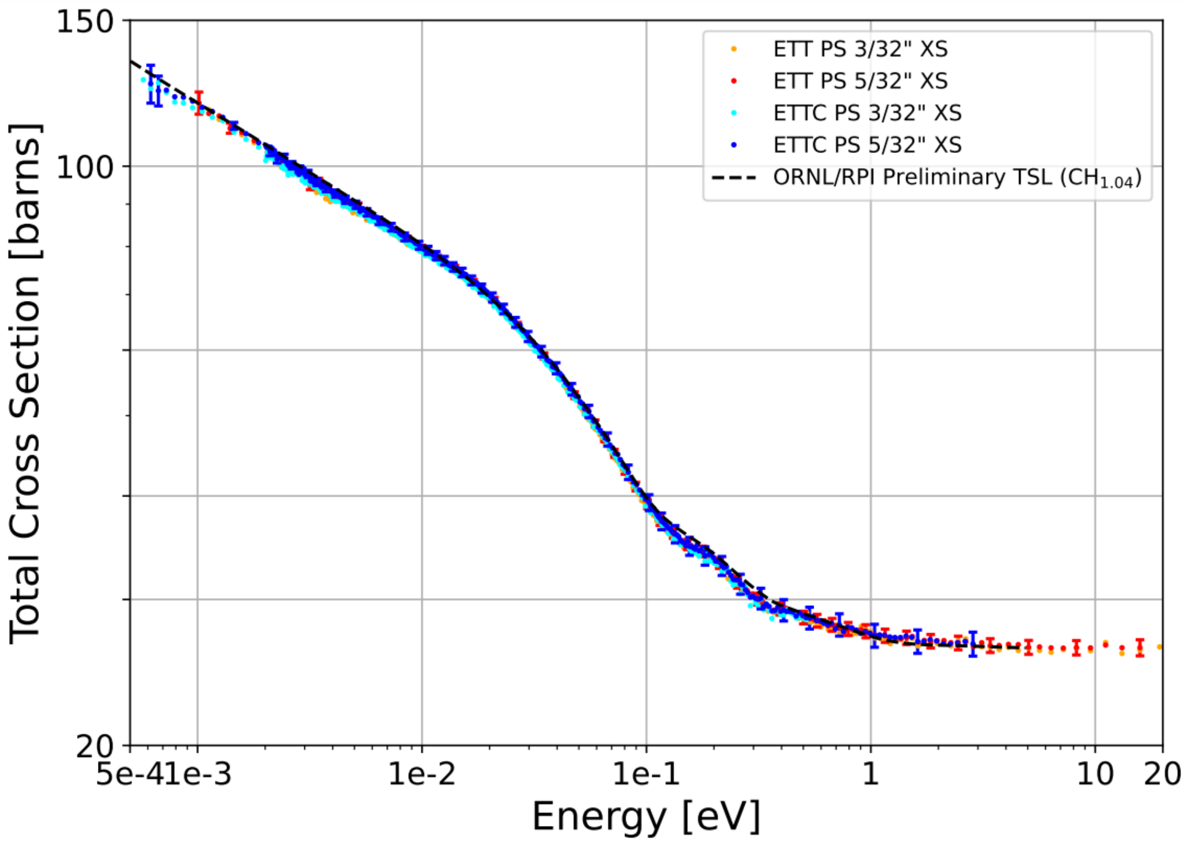
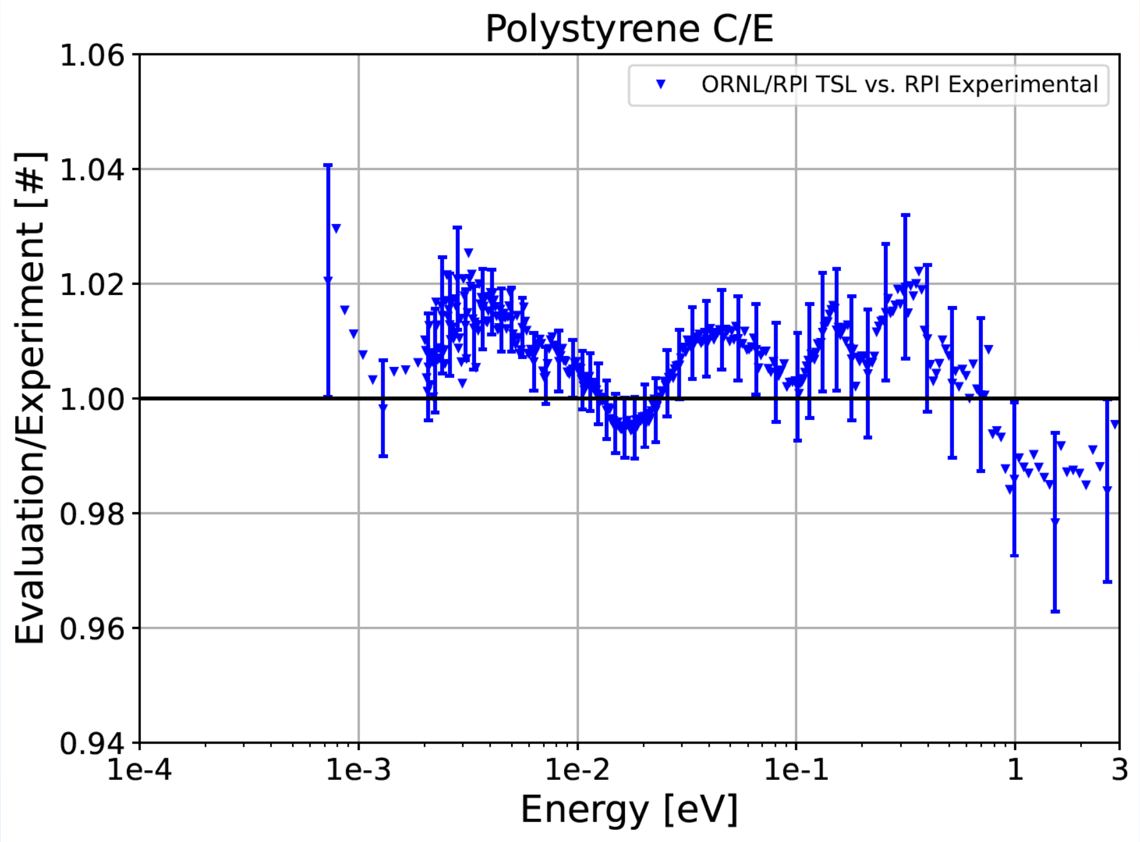
Polystyrene total cross section





- Good agreement between measured cross section and preliminary ORNL/RPI evaluation.

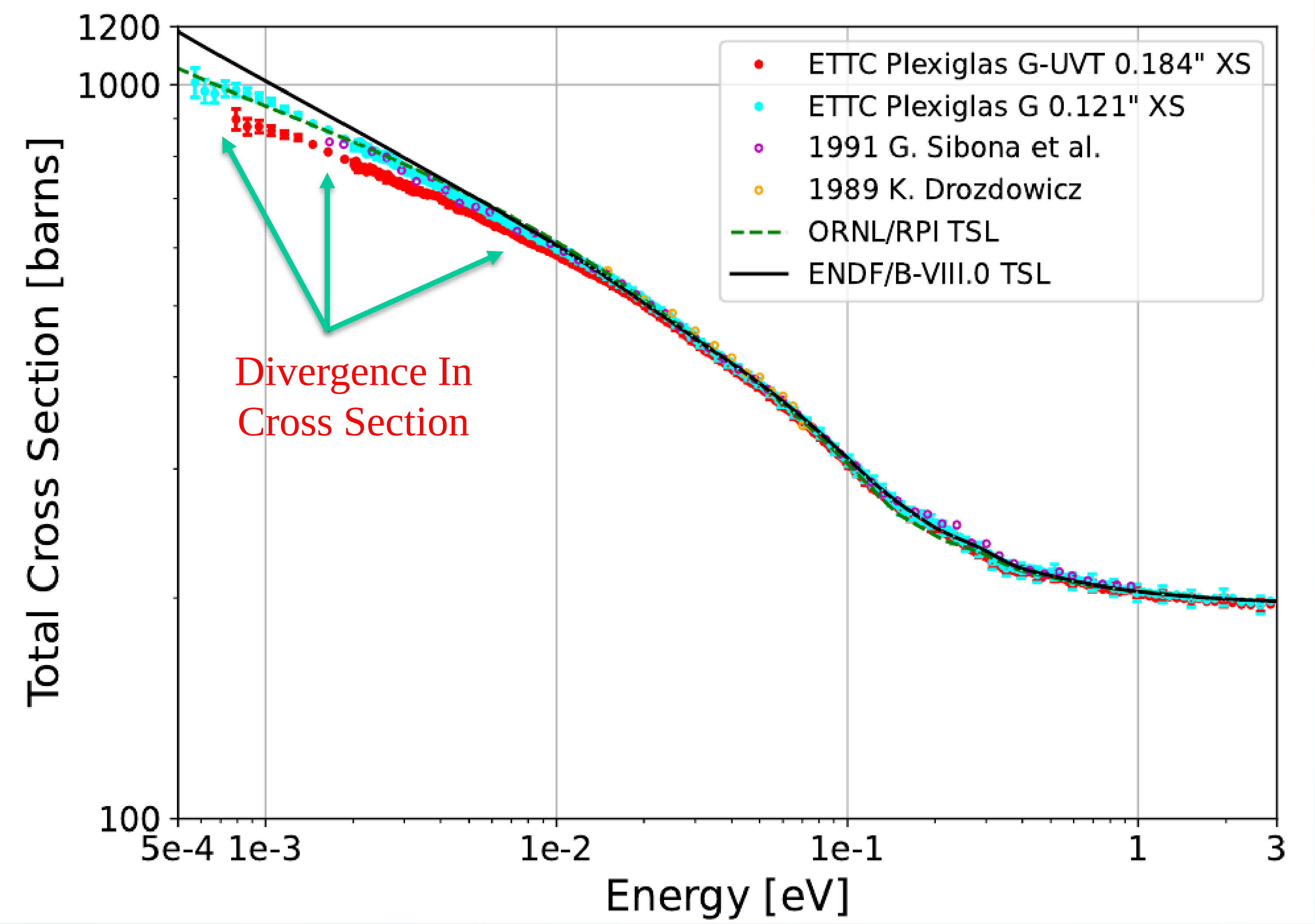
D. Fritz, Y. Danon, K. Ramic, C. W. Chapman, J. M. Brown, G. Arbanas, M. Rapp, T. H. Trumbull, M. Zerkle, J. Holmes, P. Brain, A. Ney, S. Singh, K. Cook and B. Wang, "**Total thermal neutron cross section measurements of hydrogen dense polymers from 0.0005–20 eV**", *Annals of Nuclear Energy*, vol. 183, pp. 109651, 2023, DOI:10.1016/j.anucene.2022.109651

Plexiglas total cross section

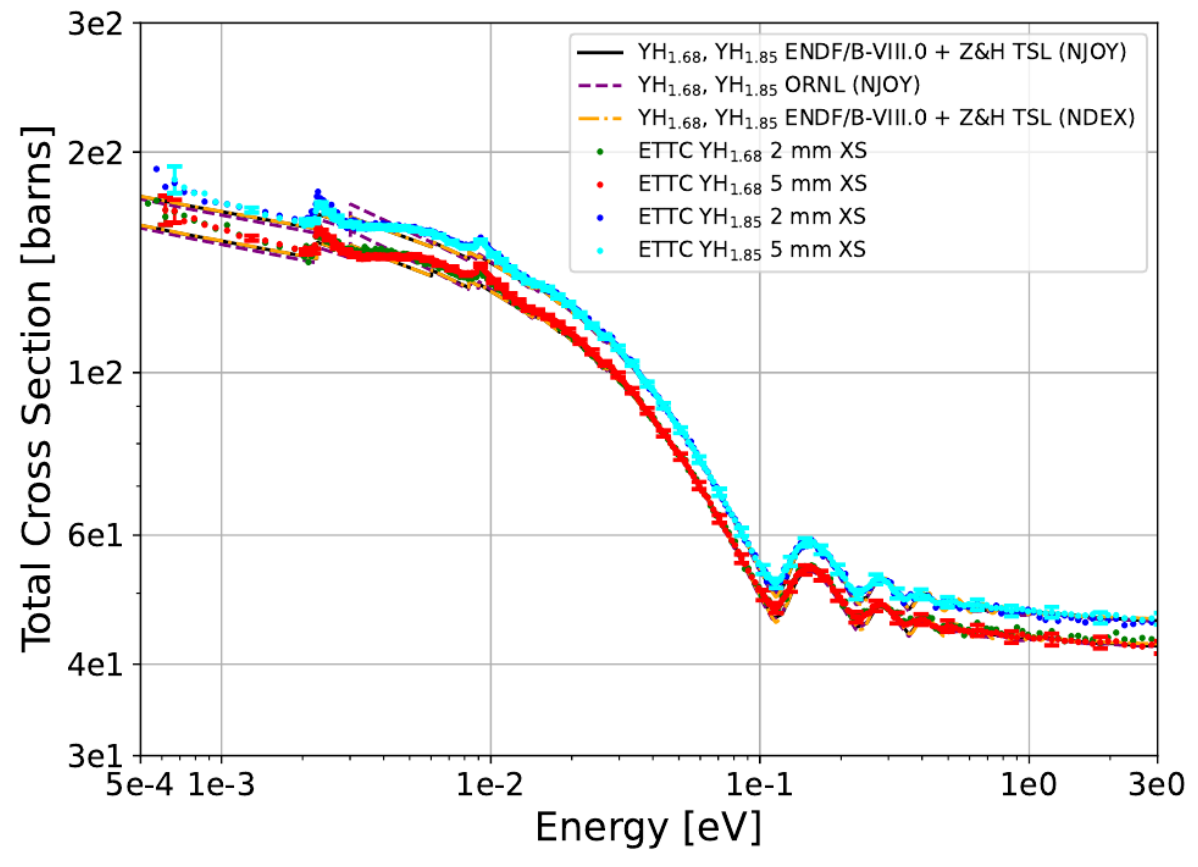
- Divergence in cross section between Plexiglas G and Plexiglas G-UVT occurs below 0.02 eV due to presence of Octadecanoic acid in Plexiglas G.

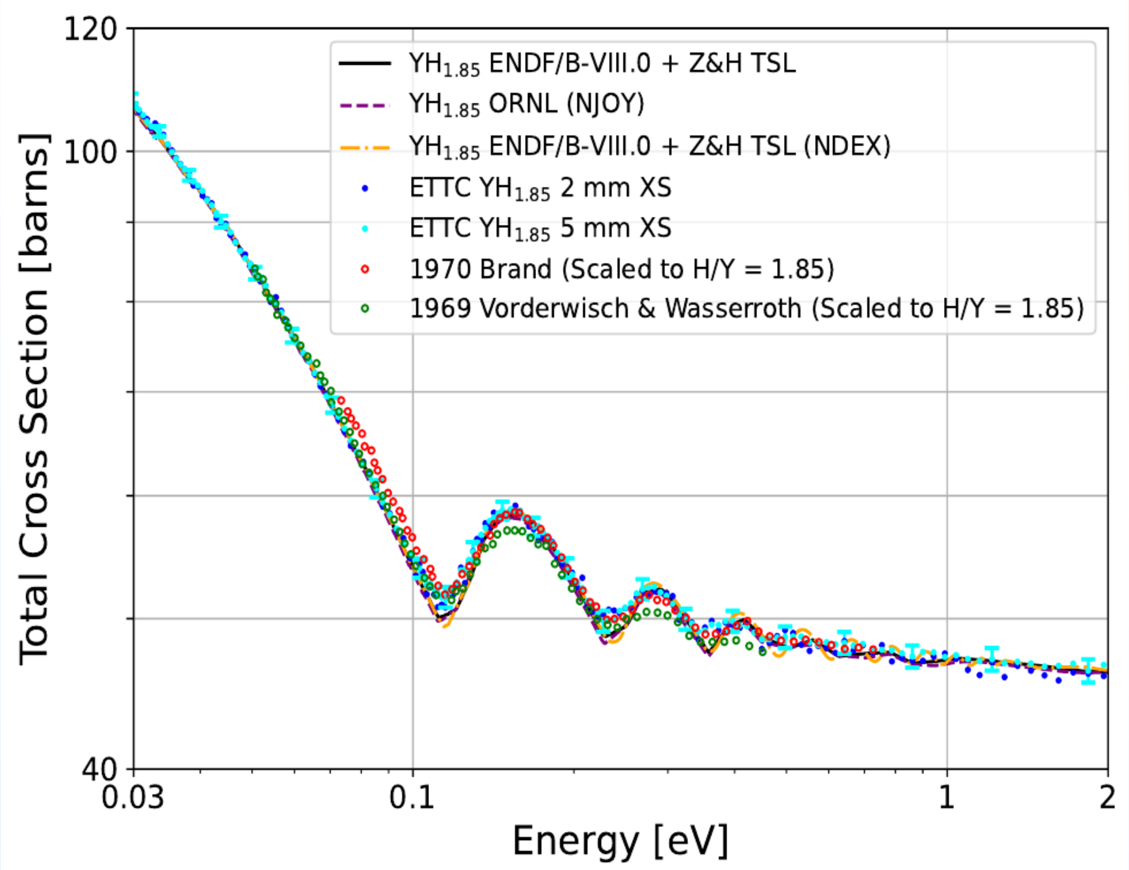
- Existing experimental data and ORNL/RPI TSL evaluation agrees with Plexiglas G

- ENDF/B-VIII.0 TSL evaluation begins to diverge from Plexiglas G cross section below 0.02 eV.



Yttrium Hydride total cross section

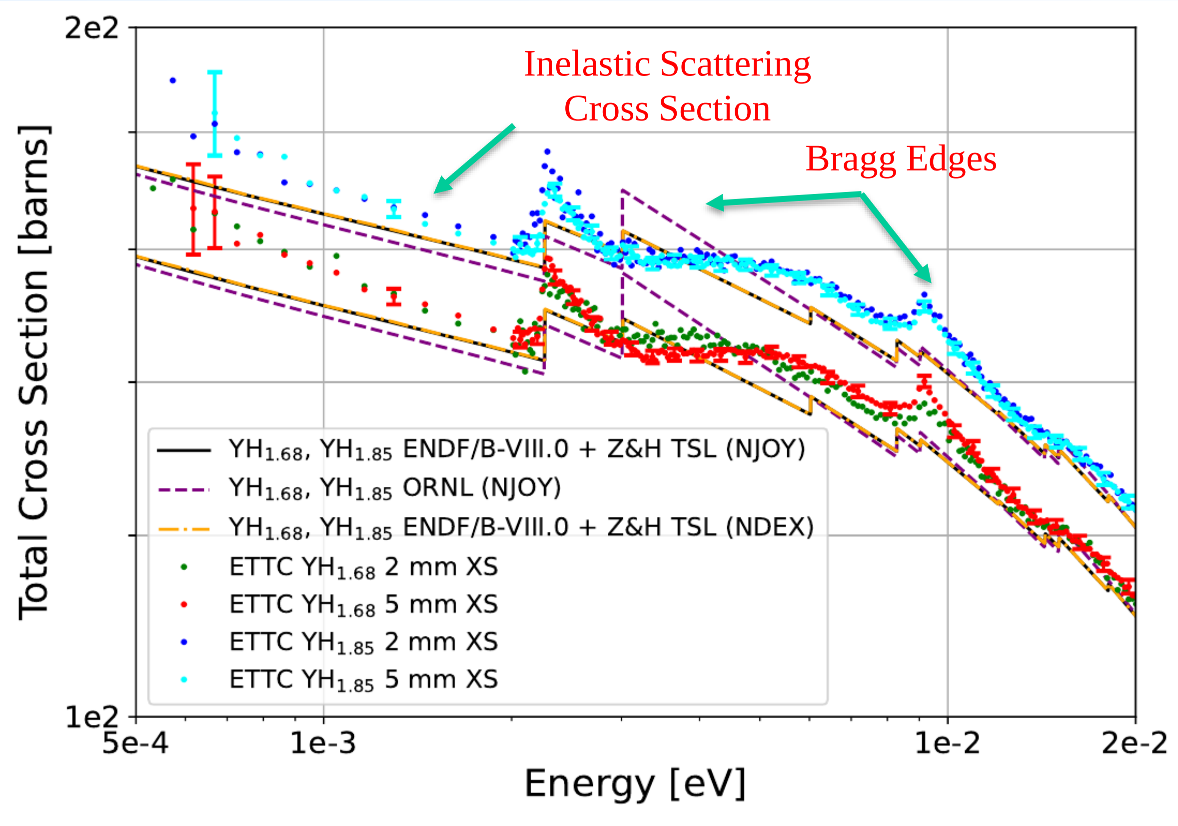


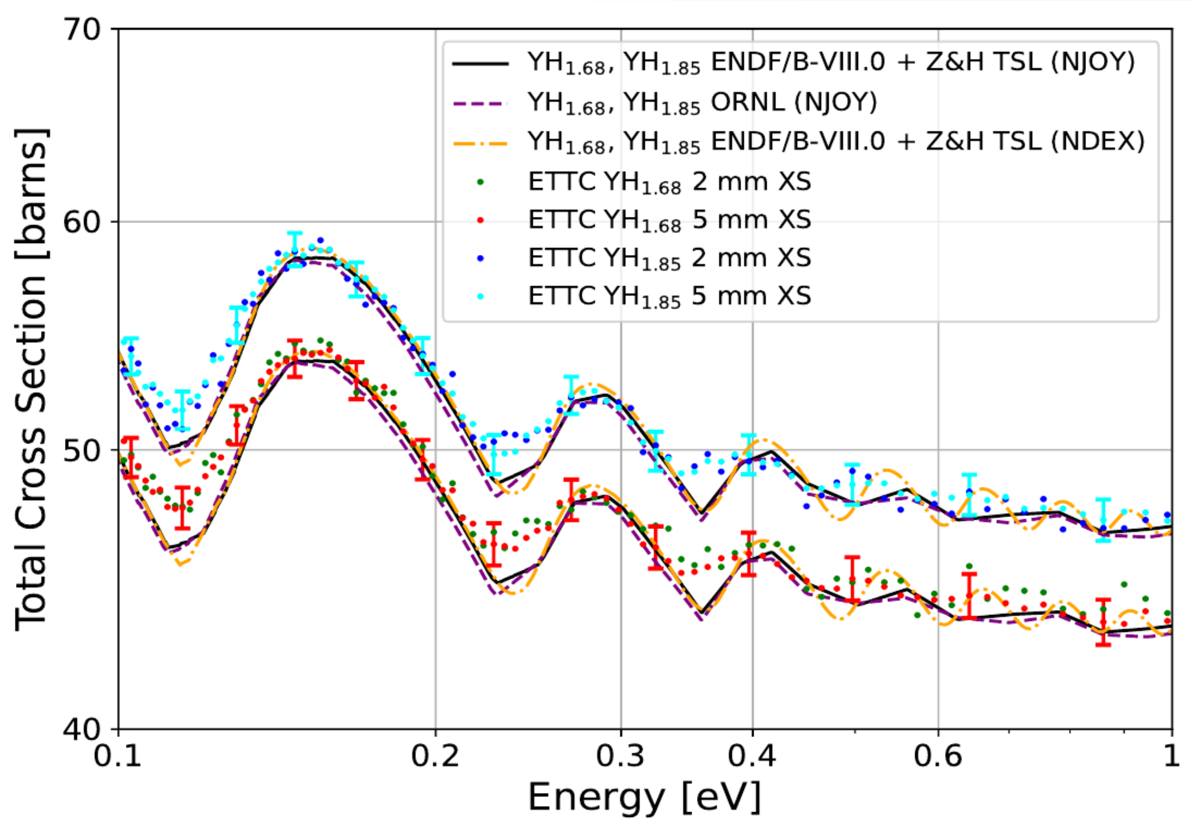


- Generally good agreement with existing experimental data and TSL evaluations.
- Measured cross section extended above 0.8 eV and below 0.05 eV.

D. Fritz, Y. Danon, M. Rapp, T. H. Trumbull, M. Zerkle, J. Holmes, C. W. Chapman, G. Arbanas, J. M. Brown, K. Ramic, X. Hu, S. Singh, A. Ney, P. Brain, K. Cook and B. Wang, "**Total thermal neutron cross section measurements of yttrium hydride from 0.0005 - 3 eV**", *Annals of Nuclear Energy*, vol. 181, pp. 109475, 2023, DOI:10.1016/j.anucene.2022.109475

Yttrium Hydride – Closer Look





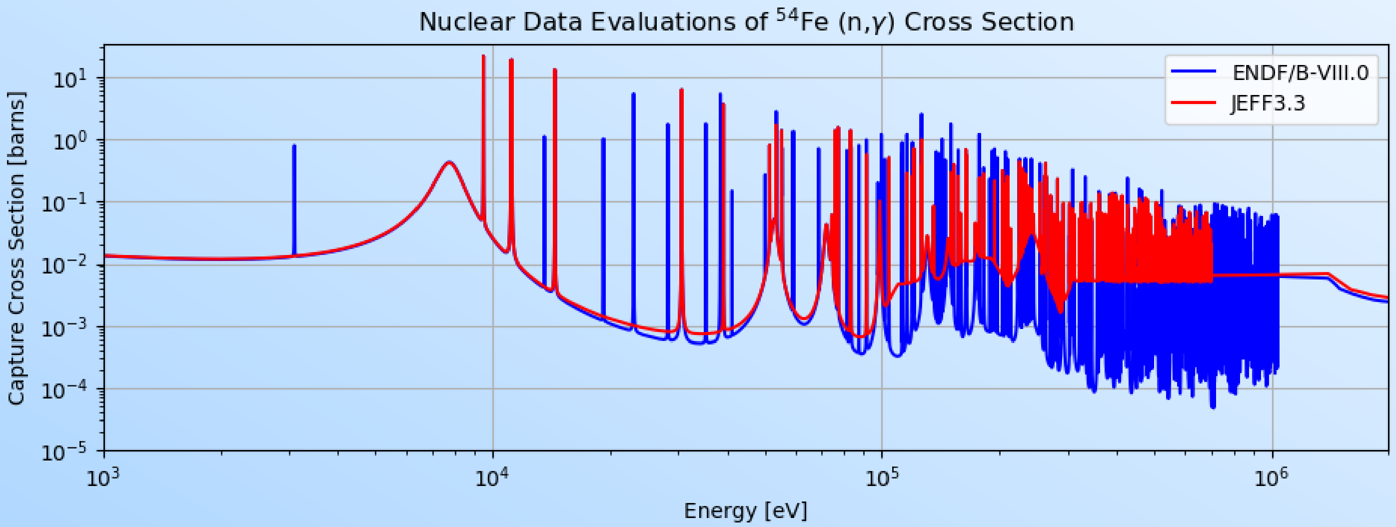
- Misalignment of Bragg edges between experiment and yttrium TSL evaluations.
- Discrepancy in inelastic scattering cross section below last Bragg edge.

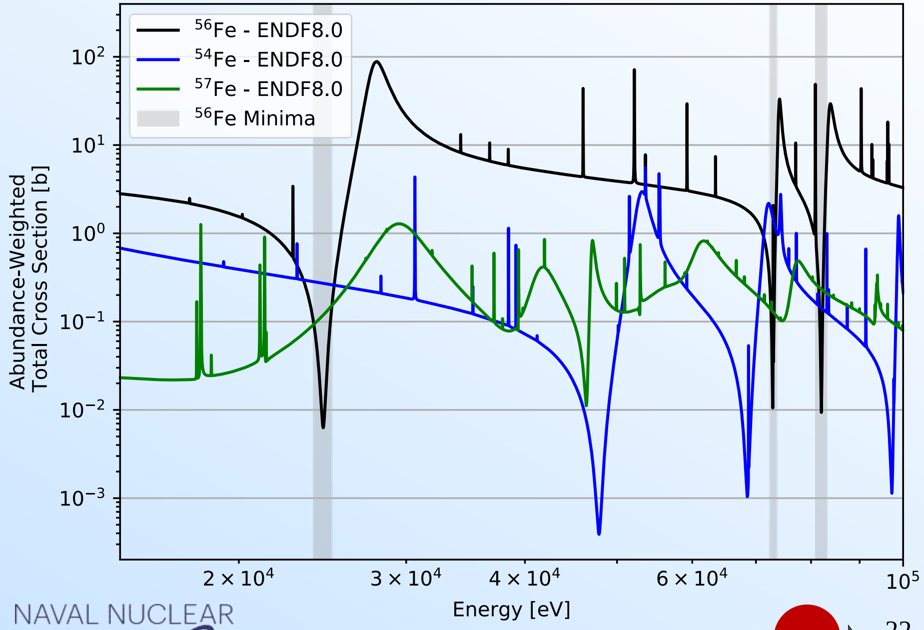
- Misalignment of oscillations between experiment and hydrogen TSL evaluations.
  - Impact of anharmonic behavior and impurities in YHx on neutron scattering not fully understood.

^{54}Fe CROSS SECTION MEASUREMENTS

^{54}Fe (n, γ) Measurement - Motivation

- Fe is important in many nuclear systems and has applications in reactors, shielding, and stellar nucleosynthesis
- Natural Fe and ^{56}Fe cross sections have been studied extensively, but there is a lack of data available in EXFOR of the ^{54}Fe(n, γ) cross section
- There are **various discrepancies between different evaluated data libraries**, where some resonances are present in one evaluation and not the other.
- Accurate knowledge of the cross sections of minor isotopes becomes very important in the ^{56}Fe cross section minima.



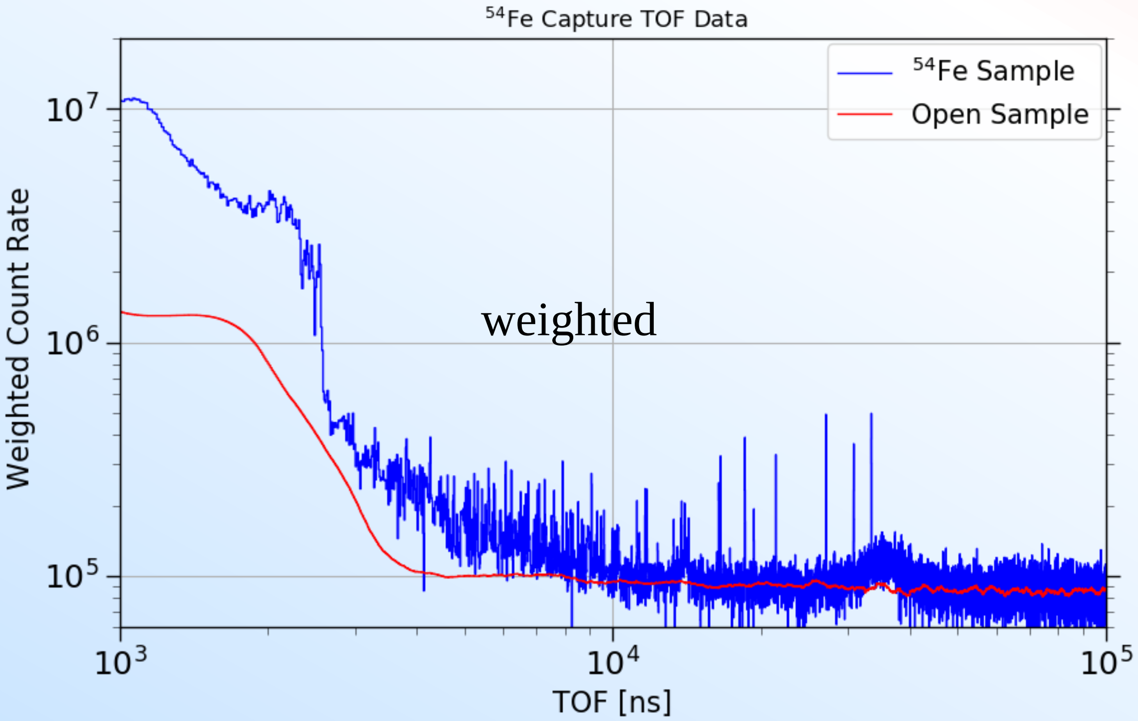


Overview of C_6D_6 Capture Array

- An array of seven C_6D_6 liquid scintillators surrounding the sample of interest at a flight path of 45m
- The system is designed to perform **radiative capture in the keV – low MeV energy range**
- All the detector structural materials have a low capture cross section to minimize neutron sensitivity
  - Materials are mostly thin Al
- System is based on the principle of the **total energy method**
  - Pulse weighting is required

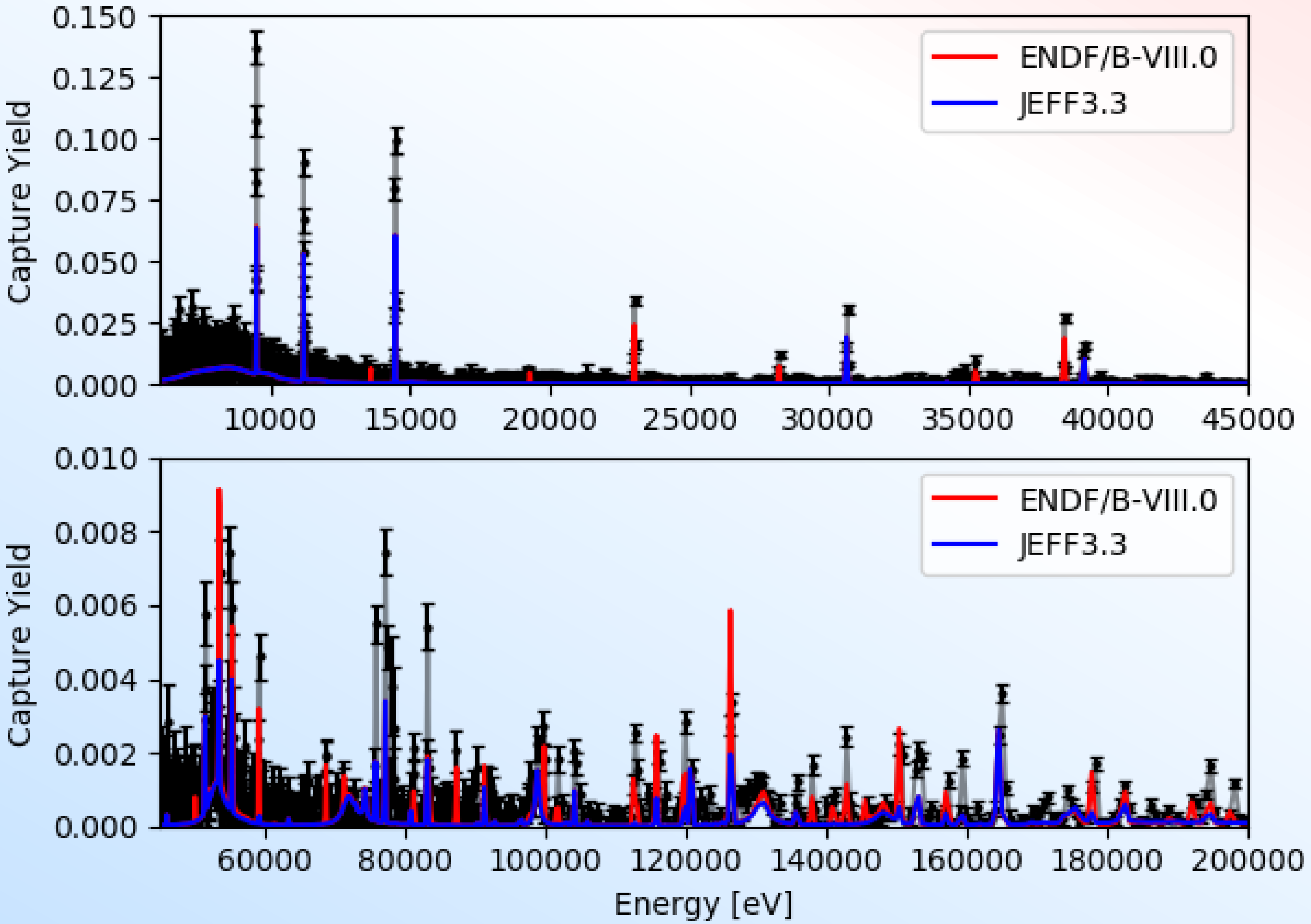
^{54}Fe Capture Measurement Overview

- Capture data were taken of a ~0.021 [a/b] 96% enriched ^{54}Fe sample.
  - 2.5 mm thick, 2" diameter.
- Separate experiment conducted for normalization of capture yield
  - Au, Ta samples used to calculate absolute normalization factors using saturated resonance method
- Resonance structure is clearly observed in the raw ^{54}Fe data.
- Pulse height weighting technique (PHWT) is utilized when processing raw data.
  - Higher energy photon events are weighted more heavily to achieve proportionality of photon energy and detection efficiency.



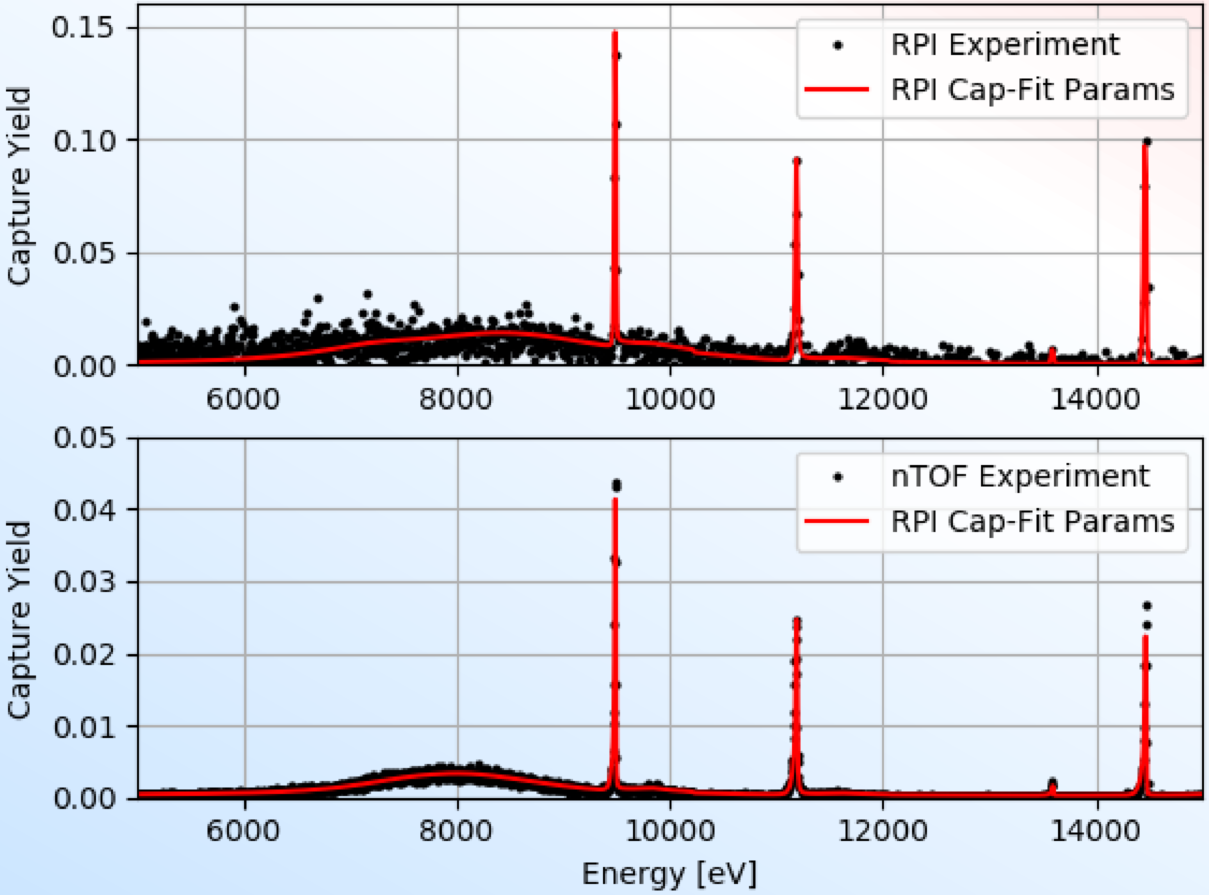
^{54}Fe Radiative Capture – Results

- RPI capture yield disagrees with evaluations at prominent capture resonances.
- Some missing resonances from JEFF3.3 can be seen in RPI experiment.
- Capture data were normalized to black resonance of Au.
- Energy resolution is limited at higher neutron energies.
- Covariance matrix generation is planned before data is released (currently underway).



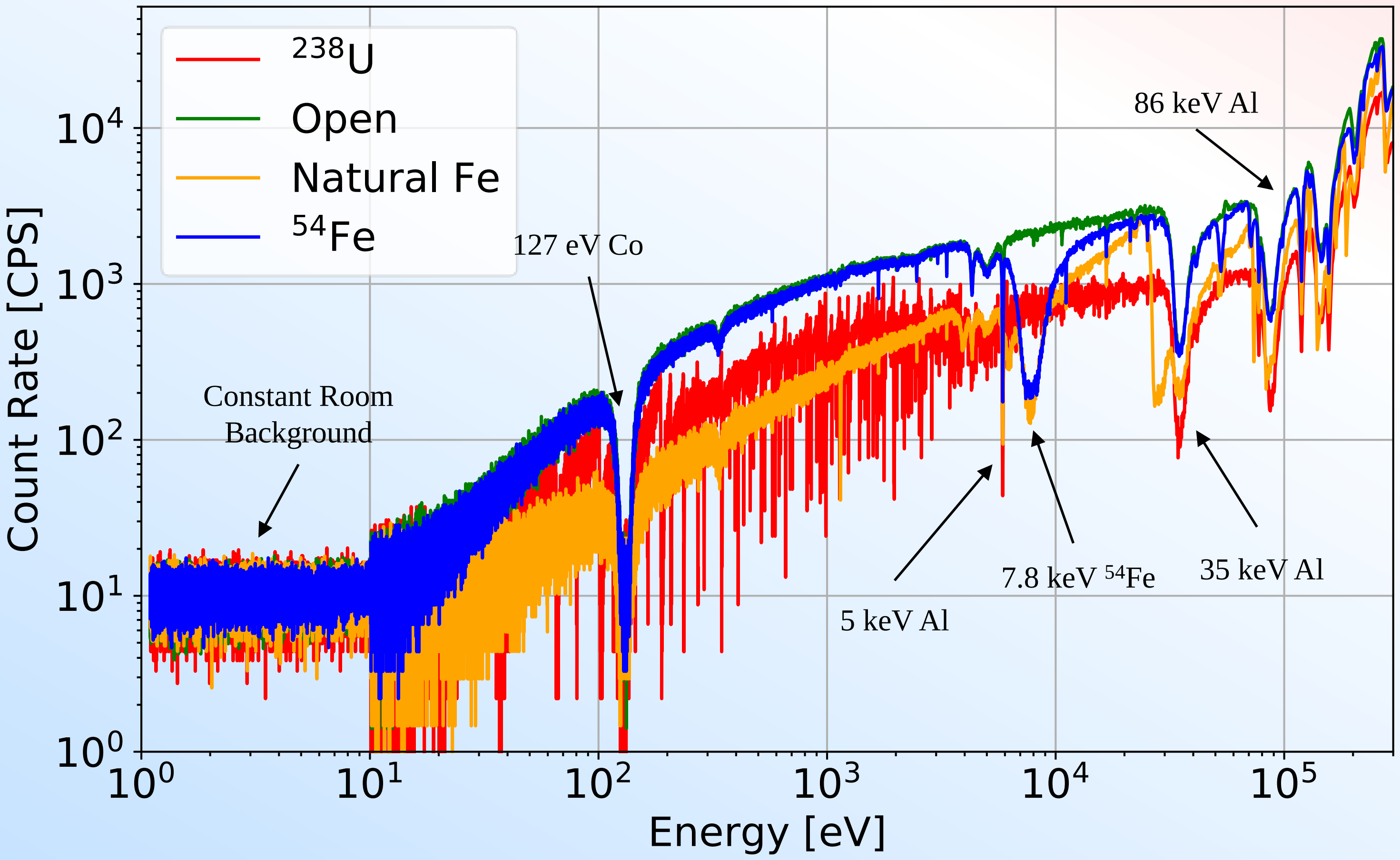
^{54}Fe Capture Data Evaluation

- n_TOF and RPI experiments can be fit simultaneously and agree with one another.
- n_TOF data available in EXFOR needs additional normalization to obtain absolute capture yield
- **Observation:** Some p,d-wave resonances will require large changes in Γ_γ
- **Work Needed:** Extend evaluation up to 1 MeV w/ consideration of data covariances when available.



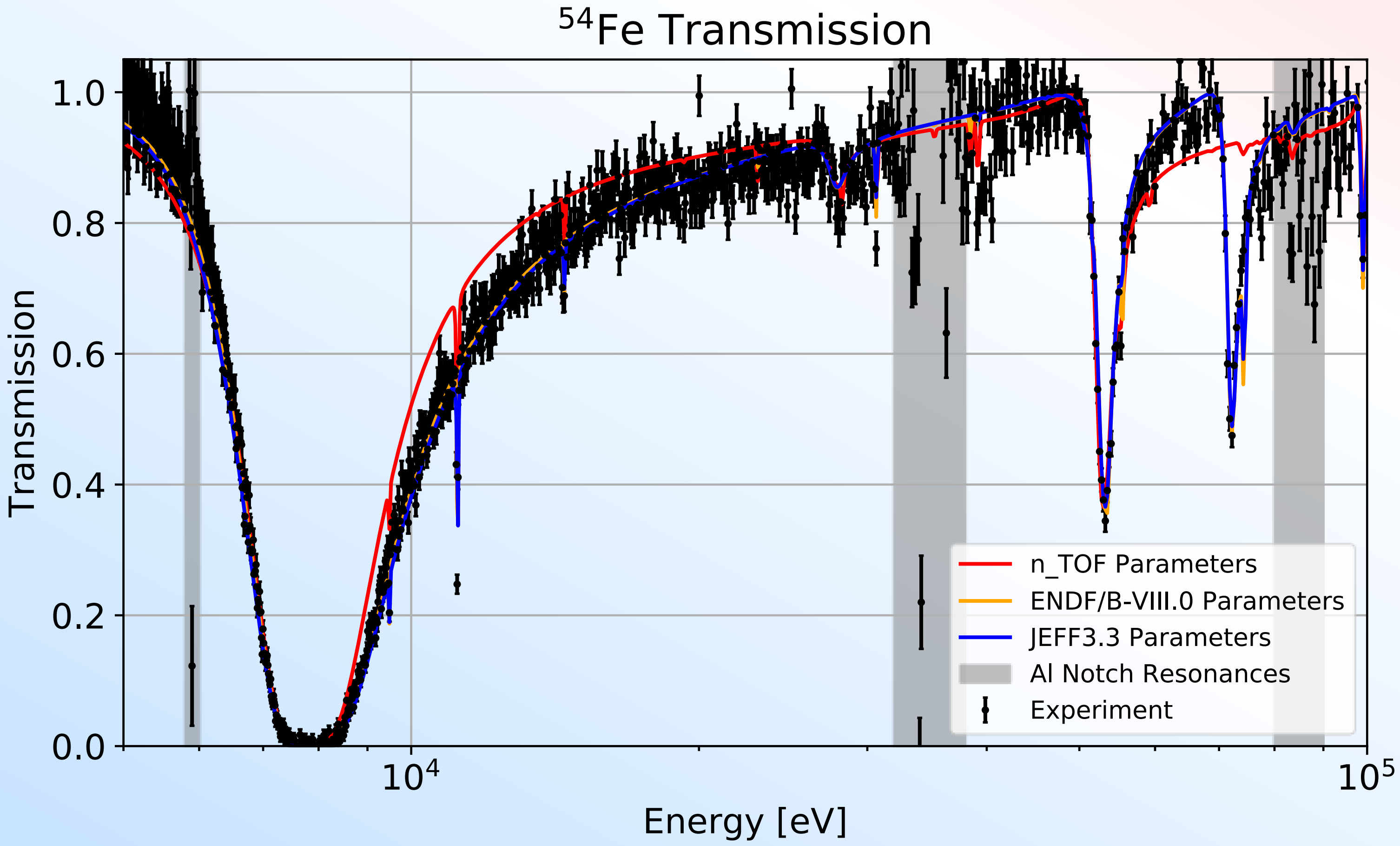
Transmission Experiment Overview

- To improve the fitting of the ^{54}Fe capture data, a transmission measurement to obtain total cross section data was conducted
- A Li-Glass detector at 35m was used to collect data, sample placed at 15m flight path
- 2cm of natural Fe, 625 mil of depleted Uranium, 0.25 cm of ^{54}Fe all measured during experiment
- Black resonances used to determine background shape for each sample



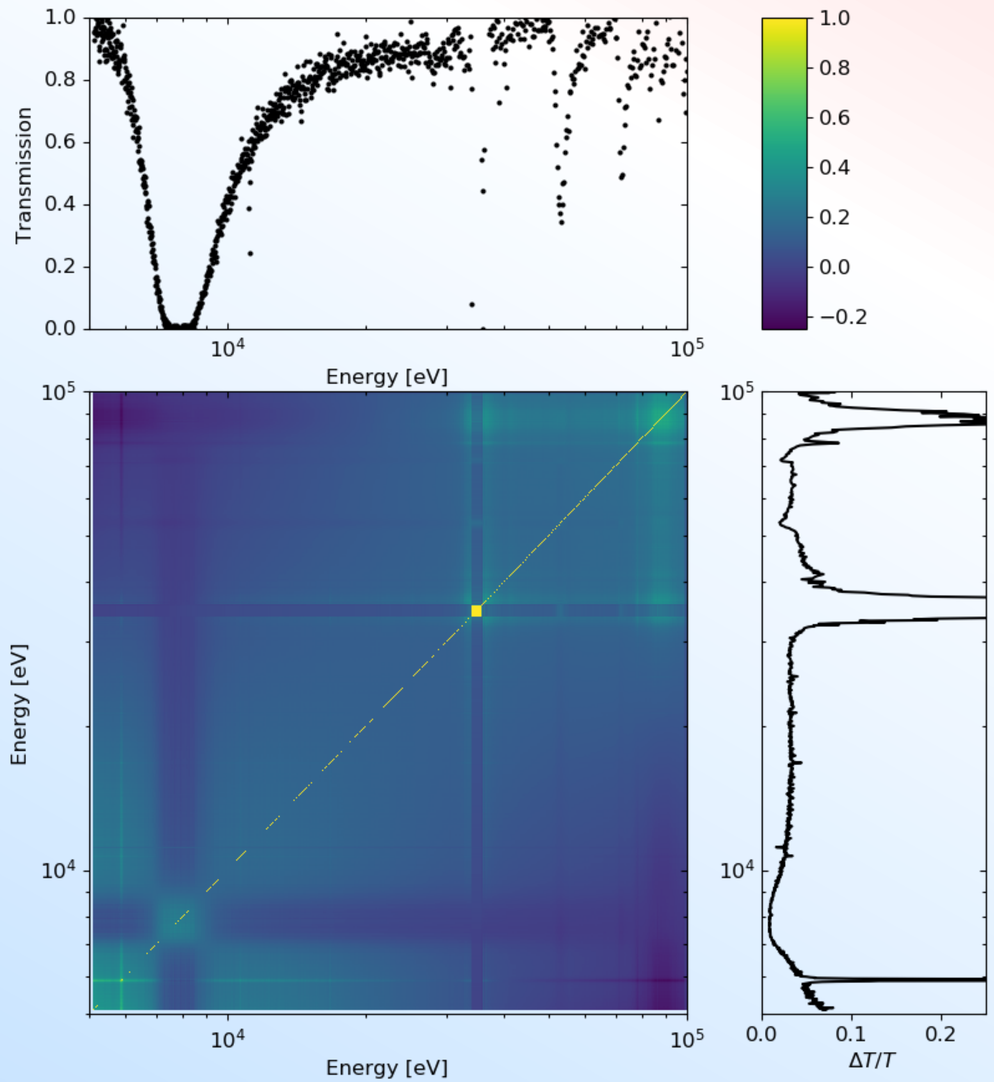
Transmission Experiment Results

- Experiment is most useful below 30 keV, where there is very limited previous data available
- Data between 30-150 keV can be used to support higher energy resolution measurements
- Limited energy resolution above 150 keV
- RPI's approach of combining capture and transmission data will improve resonance evaluation effort for ^{54}Fe.
- Covariance matrix has been generated for this experiment.



Transmission Experiment Covariance

- Covariance matrix has been generated for transmission experiment.
- Data are not heavily correlated.
- Covariance passes mathematical checks
  - Positive definite, eigenvalues all positive, covariance matrix is invertible
- Covariance has been converted into implicit format for use in evaluation w/ SAMMY.



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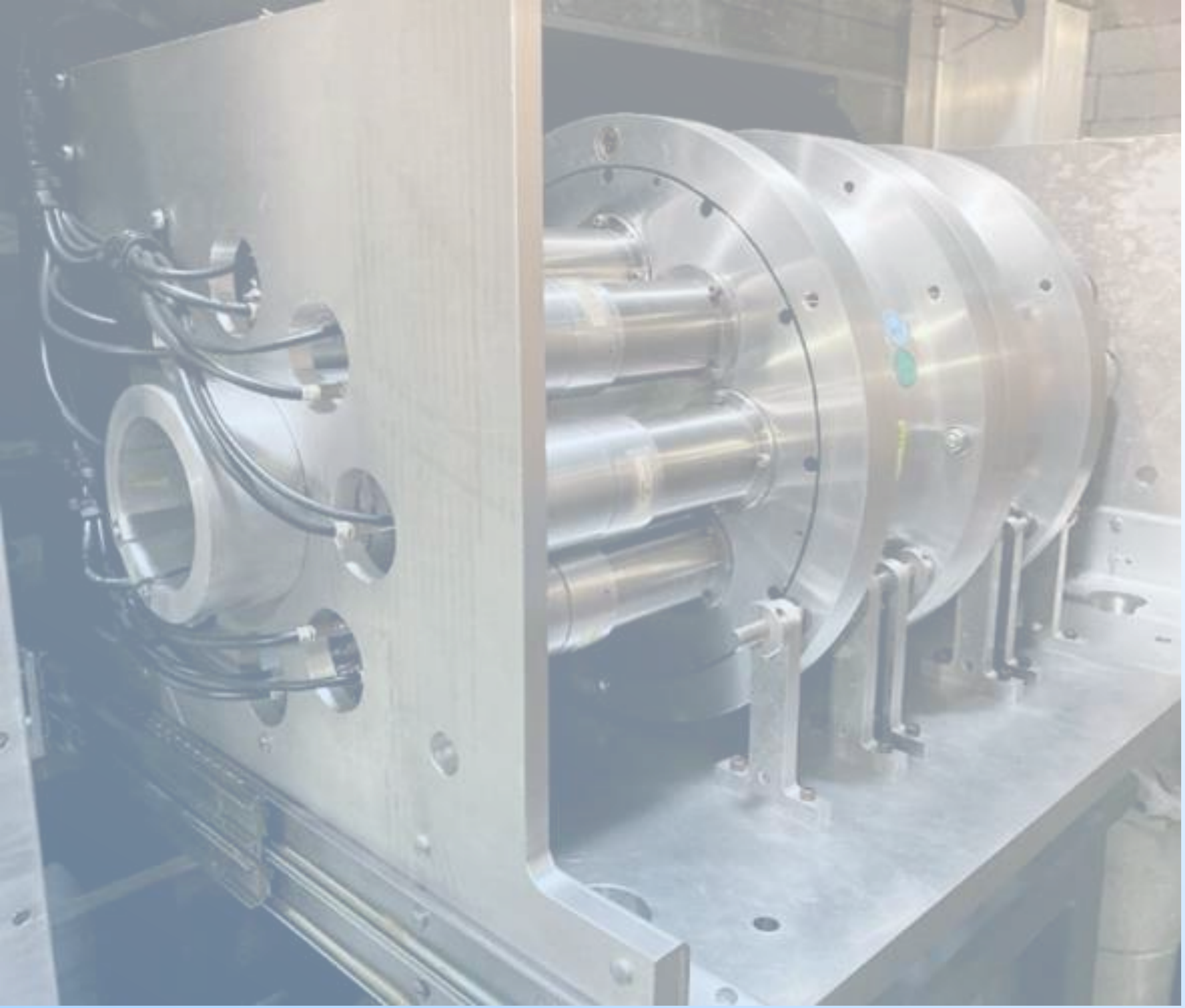
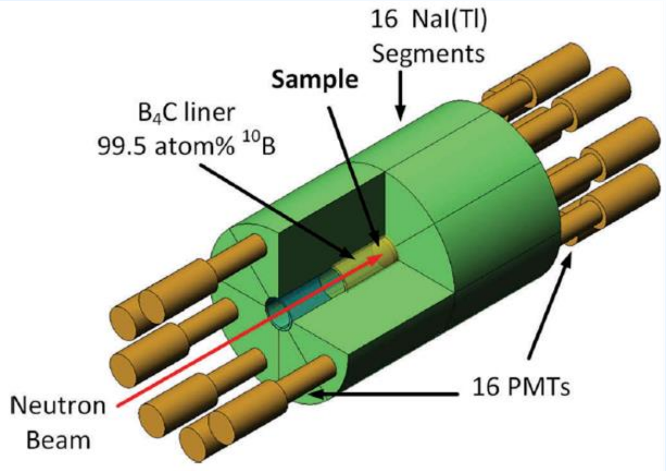


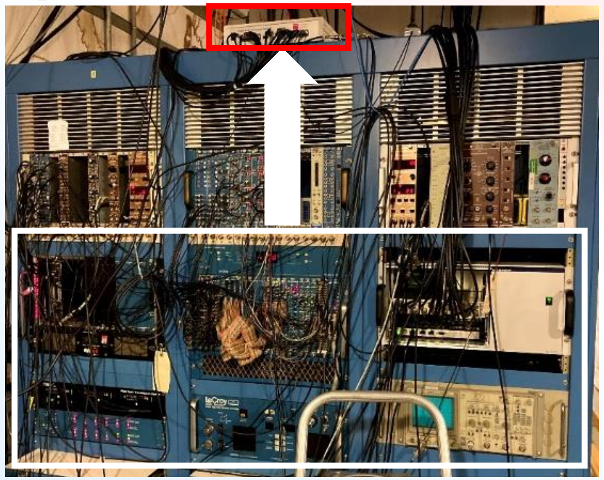
THERMAL NEUTRON CAPTURE γ-SPECTRA MEASUREMENTS

Capture γ-Spectra Measurements: Motivation & Goals

- **Test accuracy of capture data evaluations**
- **Measure capture (and fission) γ-emission spectra** as a function of energy and multiplicity of important nuclear materials in the RRR
  - Interest includes ^{235}U and ^{238}U
- **Generate detailed capture (and fission) γ-cascade data** from experimental results
  - Compare with current simulation tools
  - Constrain models used for reaction and cascade calculations
- **Improve the current models used to simulate γ-emission spectra following neutron capture**

RPI γ-Multiplicity Detector



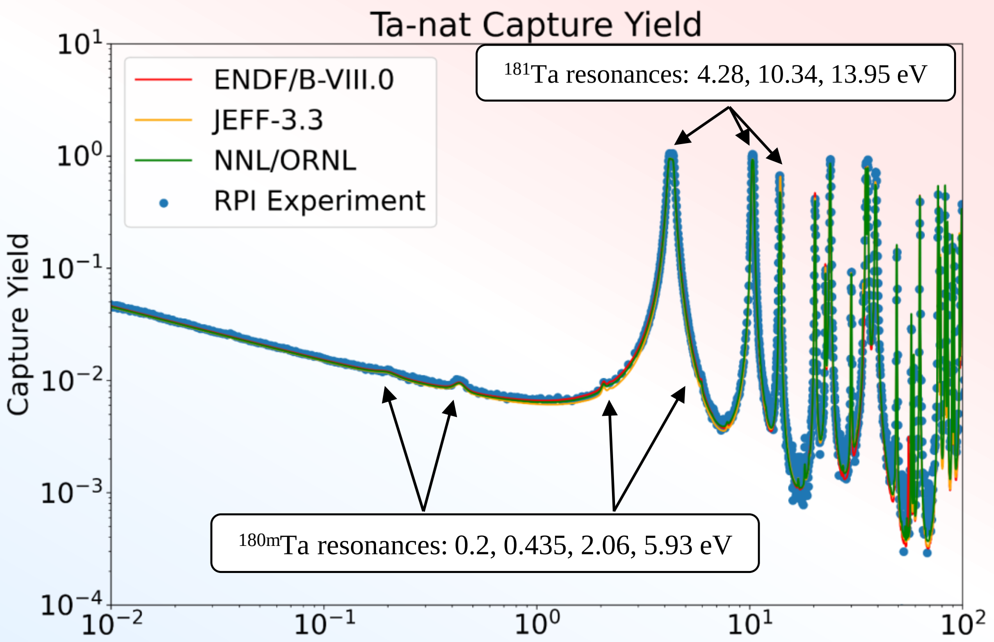


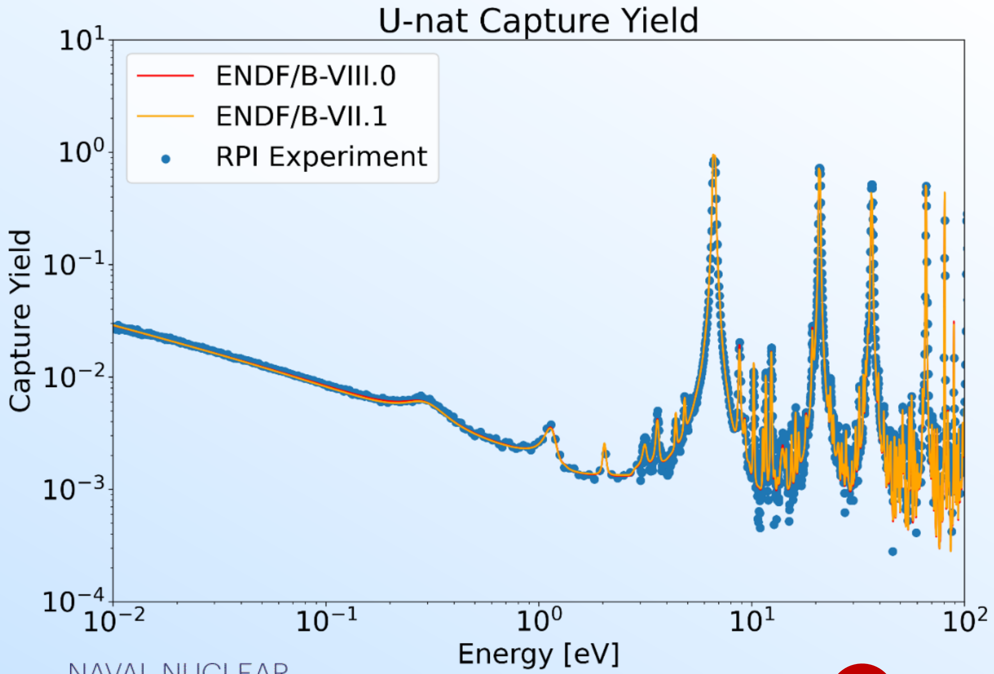
- **16 segment NaI(Tl) γ-multiplicity detector**
  - Total volume: 20 L of NaI(Tl) surrounding the sample
  - Inside the detector is lined (~1 cm) with a B_4C ceramic sleeve which is enriched 99.5 atom% in ^{10}B to absorb scattered neutrons from the sample
  - Up to 96% efficiency for detecting γ-cascades
- **Detector is used for capture yield and γ-spectra measurements**
  - Useful neutron energy range: 0.01 eV – 3 keV
- **New SIS3316-DT 16 Channel Digitizer**
  - Digitize pulse wave for all events on all detectors & obtain the energy of each detected event

Experimental Validation

Low Energy Capture Yield of ^{nat}Ta and ^{nat}U

- Sample thicknesses:
  - 10 mil ^{nat}Ta (0.012% ^{180m}Ta)
  - 20 mil ^{nat}U (0.7% ^{235}U)
- Useful energy range: 0.01 – 100 eV
- Validation of the new DAQ system and processing codes (julia based)
- First-order calculated capture yield is shown for evaluations





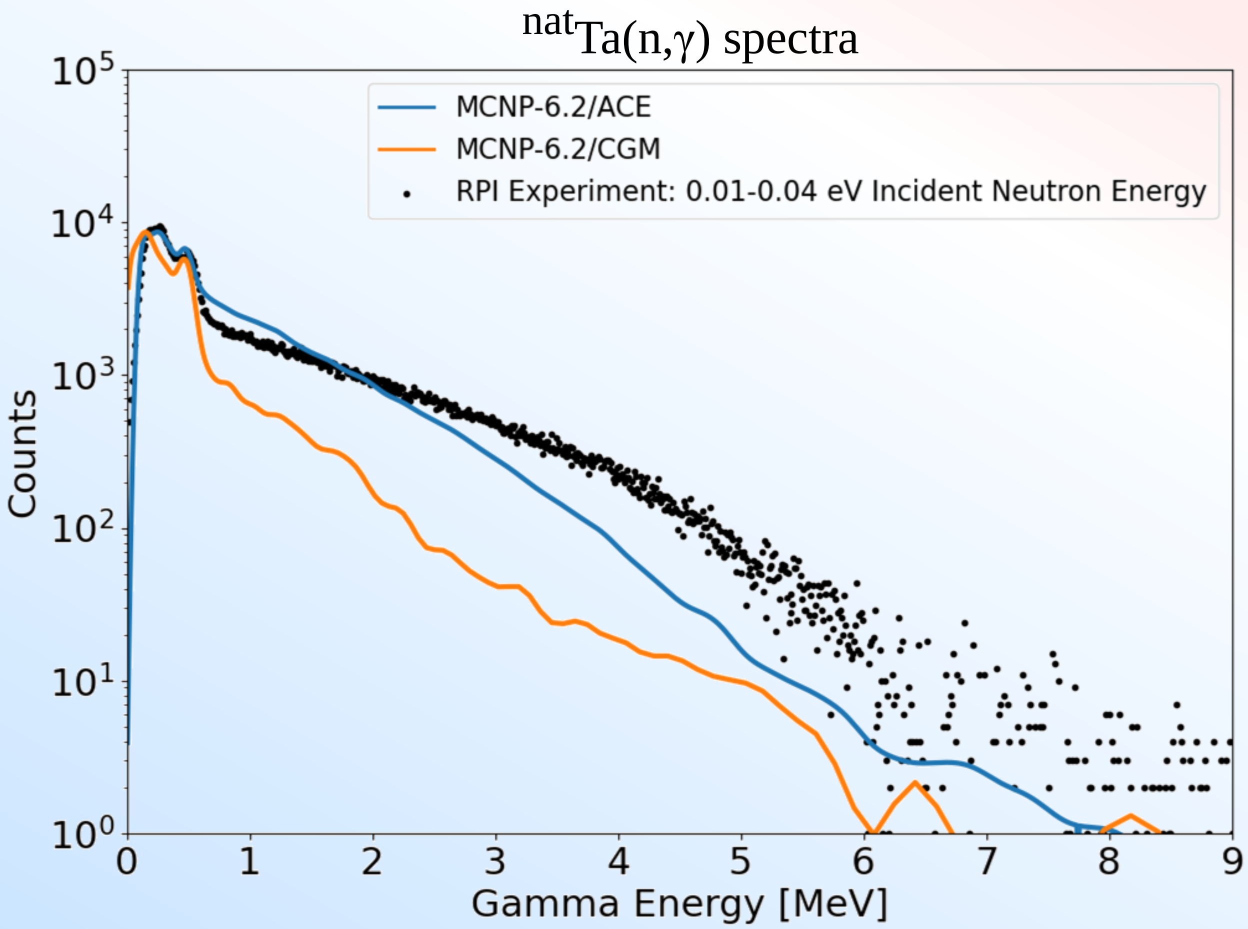
Comparing Experimental γ-Spectra to MCNP Simulations

- Current tools for modeling show a large discrepancy between experimental and simulated γ-spectra for ^{181}Ta(n,γ)

- **MCNP-6.2**
  - Extracts γ-ray data from ACE files (ENDF/B-VIII.0)

- **MCNP-6.2 with CGM**
  - **C**ascading **G**amma-Ray **M**ultiplicity
  - Produces correlated secondary γ-emissions
  - Has some issues

- Need a better way to feed γ-cascades to MCNP



Comparing Experimental γ-Spectra to `MCNP-6.2` Simulations

Step 1: γ-cascades are generated using an external code (i.e., **`DICEBOX`**) and are written to a file

```
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
```

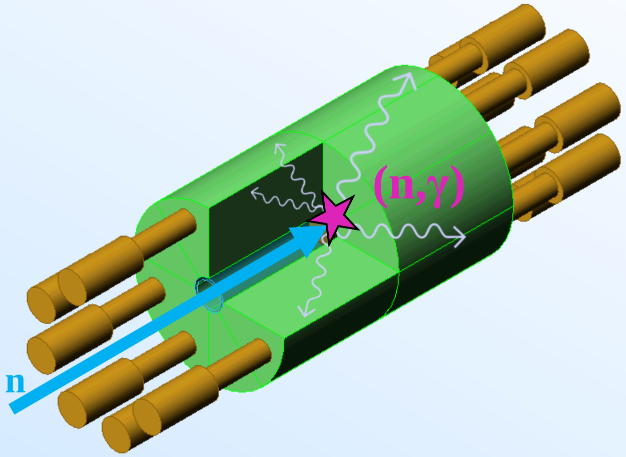
Number of γ-rays in cascade

Energy of each γ-ray in cascade

Step 2: Run modified **`MCNP-6.2`**, for each capture event:

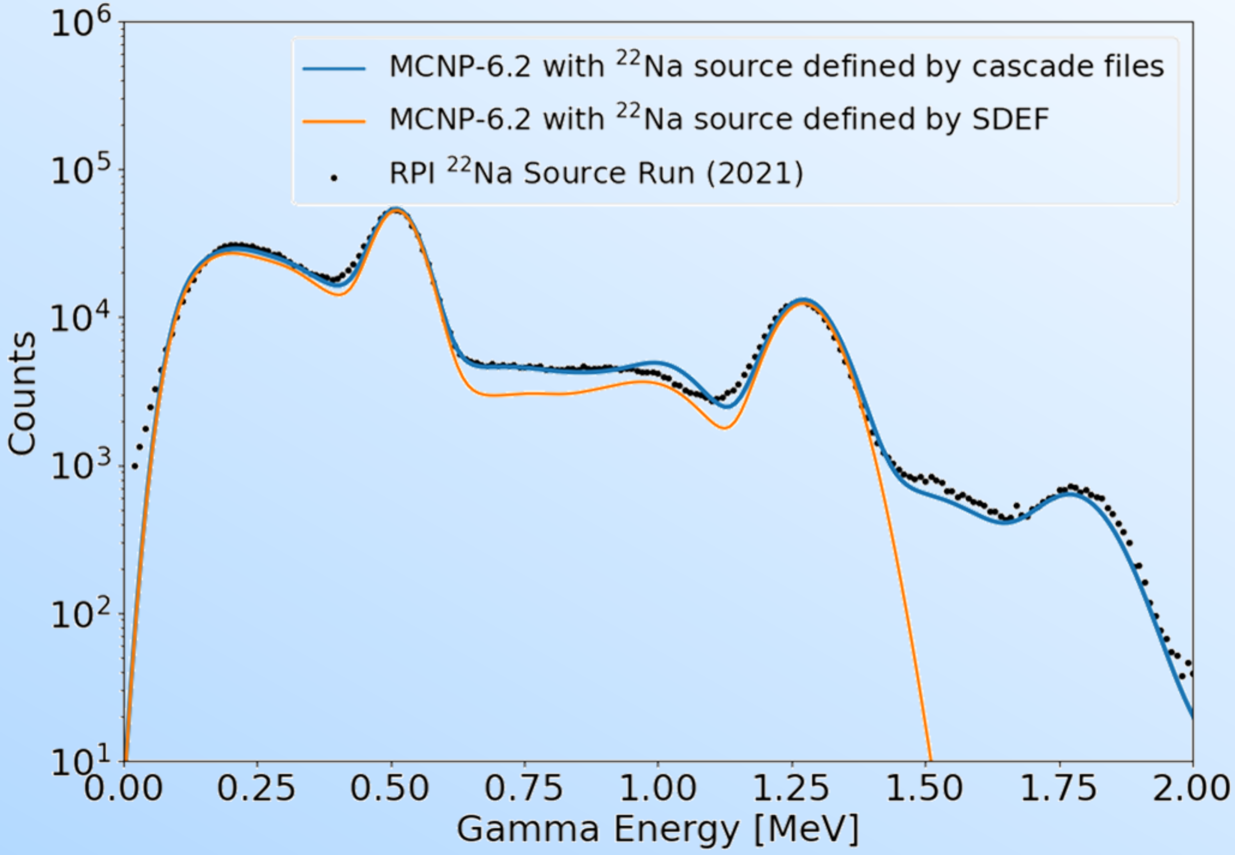
1. Read in γ-cascade from file
2. Transport γ-cascade through the detector geometry
3. Output an event file to tally γ-energy deposition in detector cells

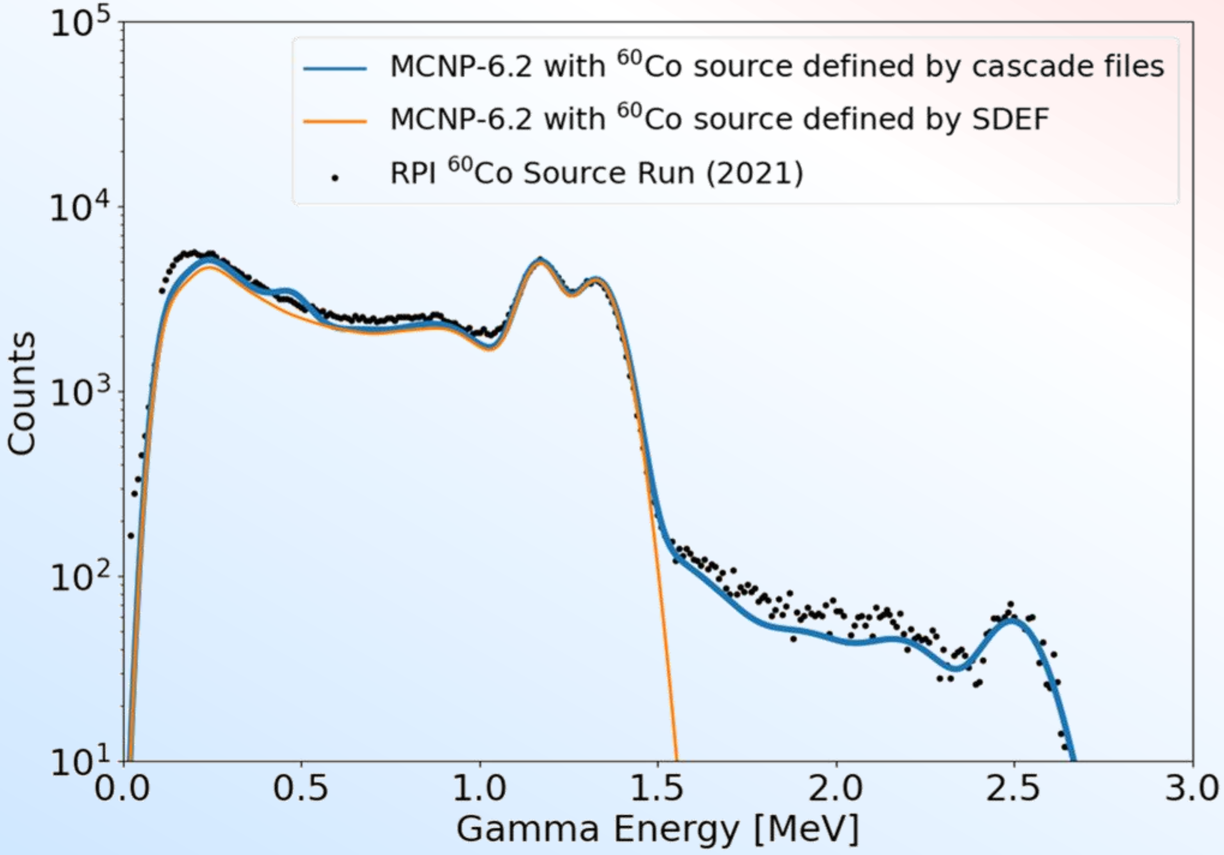
Step 3: Process the output file using event-by-event analysis including coincidence and compare to experimental data



MCNP-6.2/DICEBOX Simulation Validation

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





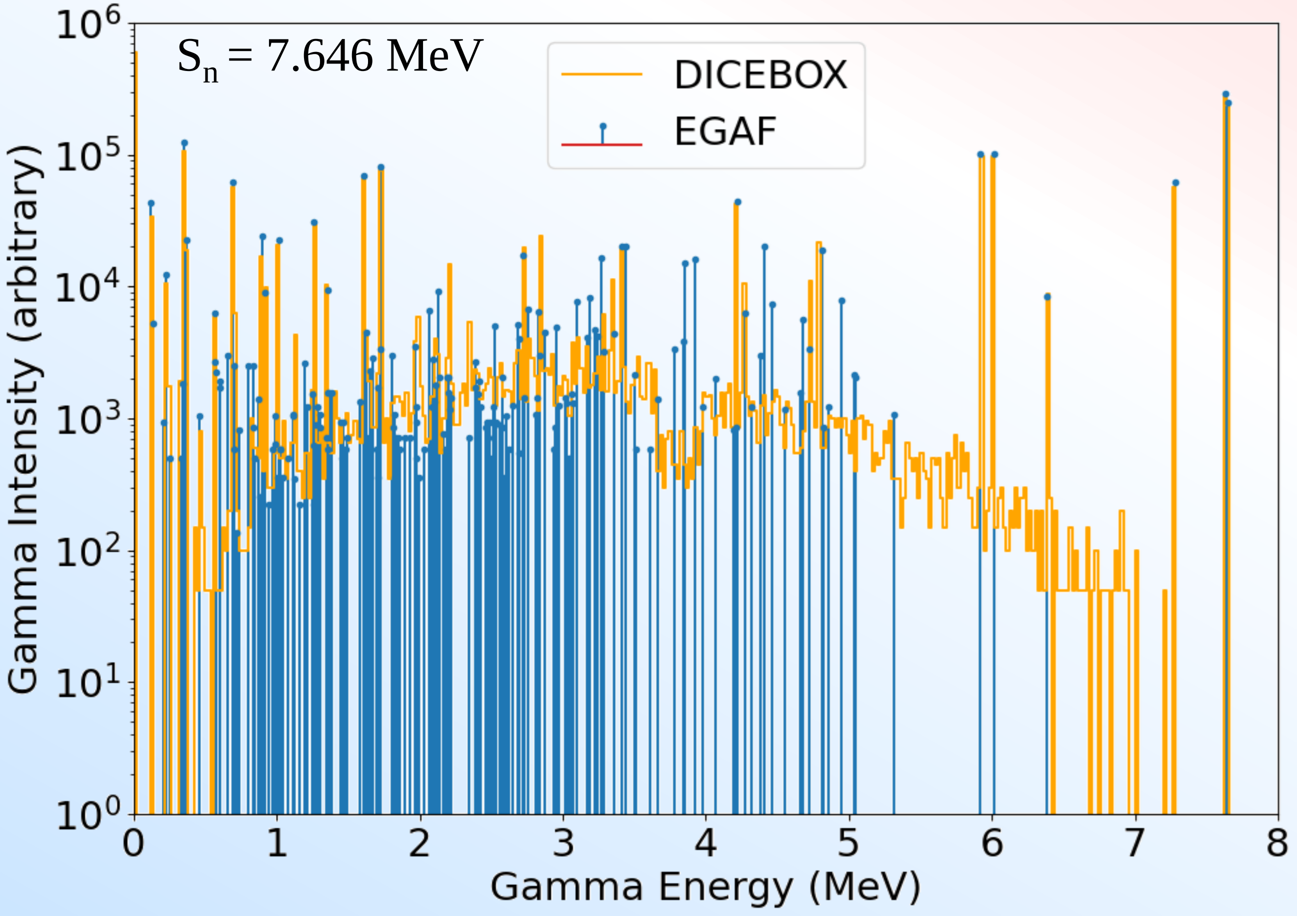
MCNP-6.2/DICEBOX accounts for the high energy sum peak in single detectors resulting from coincidence

Comparing ^{56}Fe DICEBOX Capture γ-Cascades to EGAF

- To measure capture γ-spectra accurately, **the detector system needs to be benchmarked by isotopes with well-known γ-ray data (like ^{56}Fe)**

- **DICEBOX**
  - Models full γ-cascades using evaluated nuclear data

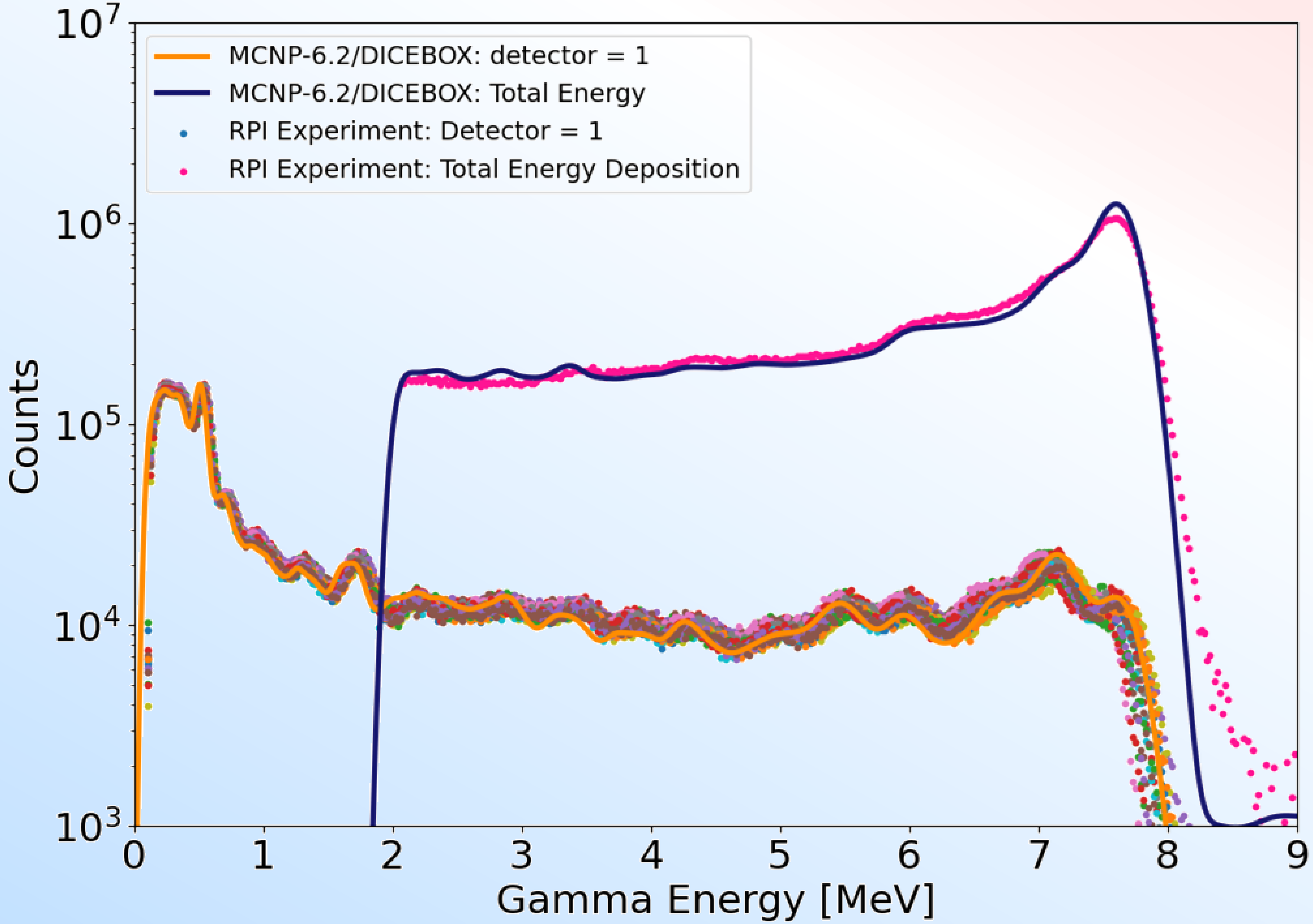
- **EGAF**
  - Shows experimentally measured γ-ray lines (does not represent the full spectrum)



^{56}Fe(n,γ) spectra compared to `MCNP-6.2/DICEBOX` Simulation

- Minor discrepancies between experimental and simulated γ-spectra for ^{56}Fe(n,γ)

- **Conclusion**: experimental γ-spectra agree with **`DICEBOX + MCNP-6.2`** calculations for isotopes with well-known γ-ray data



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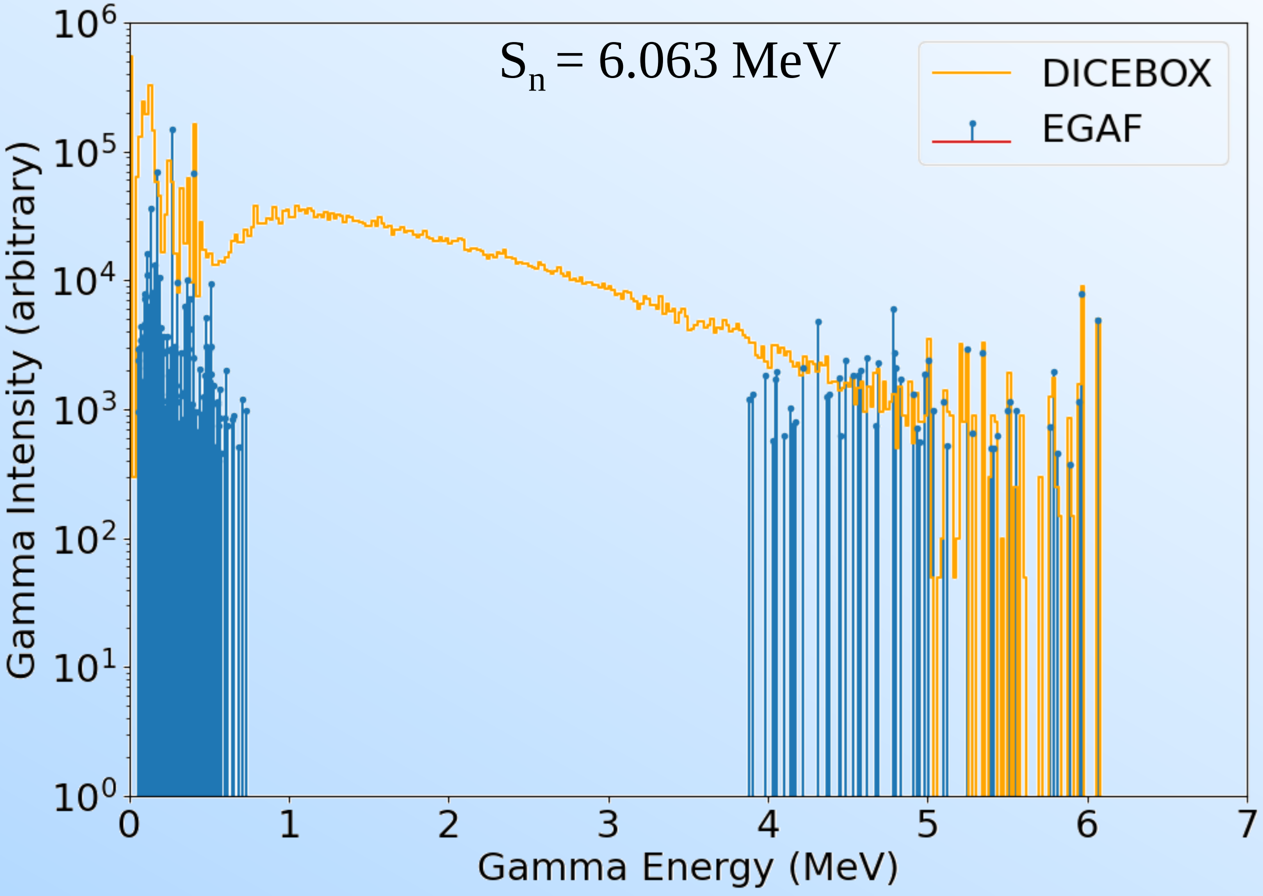
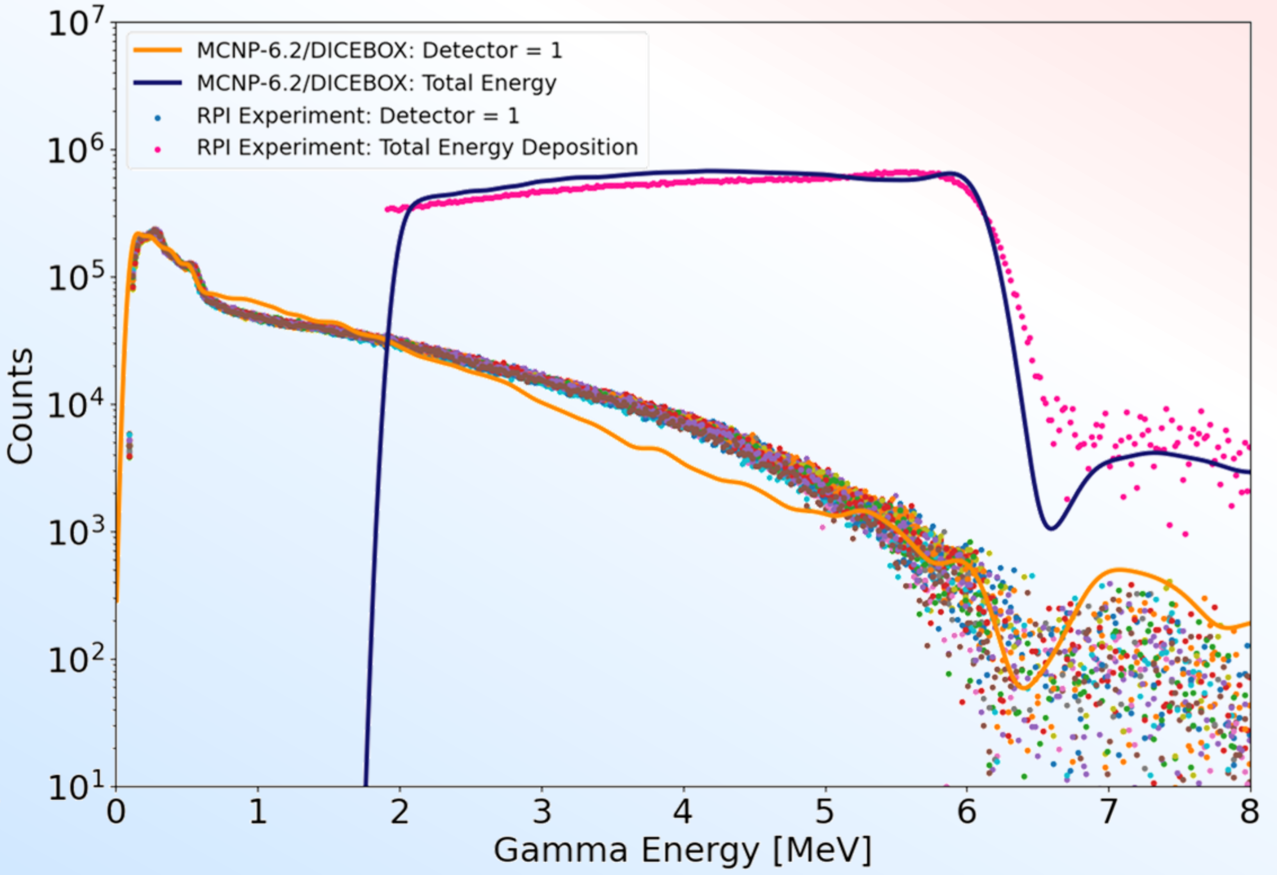
Challenges: Deficiencies in evaluated γ-ray data

^{181}Ta(n,γ) spectra compared to **MCNP-6.2/DICEBOX** Simulation



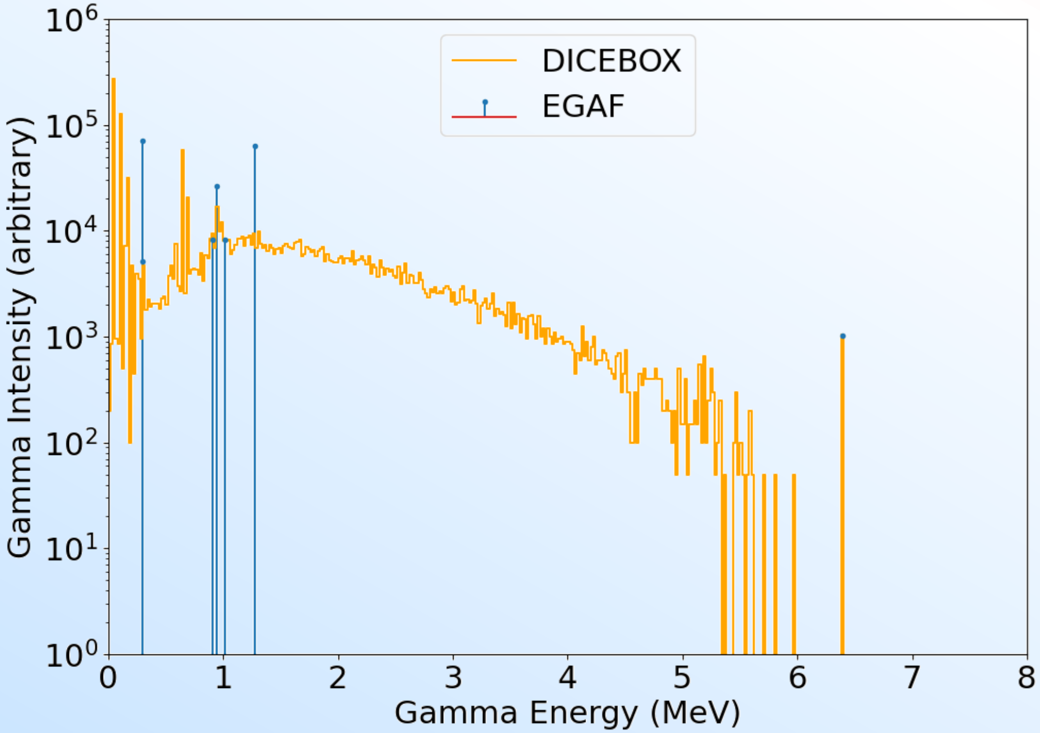


Discrepancy between experimental and simulated γ-spectra

Capture γ-Spectra Measurements: Conclusions

- Experimental, simulation and nuclear data methods were validated for the RPI γ-Multiplicity Detector
- **When neutron capture γ-cascade data is well-known, the γ-emission spectra can be accurately calculated using the `MCNP-6.2/DICEBOX` simulation method.**
- RPI γ-Multiplicity Detector system is now ready for analysis and recommendations for isotopes with deficiencies in γ-ray data (like ^{181}Ta)

Comparing ^{235}U **DICEBOX** Capture γ-Cascades to EGAF



Future Work

- Develop a method for analyzing and adjusting nuclear data for ^{59}Co, ^{55}Mn and other measured isotopes including **^{181}Ta**
- Compare experimental γ-emission spectra with **`MCNP-6.2/DICEBOX`** simulations for **^{238}U and ^{235}U**
  - Most interesting for reactor applications, most difficult to measure and simulate (due to the fission contribution)

Conclusions

- **Thermal measurements**
  - Constructed the Enhanced Thermal Target + Cold Moderator (ETTC).
    - Net neutron flux improved by up to a factor of 8 below 0.02 eV.
  - 14 new total cross section measurements and covariances submitted to EXFOR for various moderator materials over the entire thermal energy region (polyethylene, polystyrene, Plexiglas, yttrium hydride).
  - Published 3 journal articles, 2 conference papers, and a PhD thesis.
- **Fe-54 capture**
  - Completed capture measurements for Fe-54 at 45m flight path
  - Completed transmission measurements of Fe-54 sample at 35m flight path
  - Evaluation of Fe-54 resonance parameters in progress
- **Capture γ-cascade measurements**
  - Upgraded data acquisition system to Digitizers
  - Perfumed measurements on several samples including Ta, Fe-56, U-235, U-238
  - Developed methodology to simulate capture gamma cascade and transport them with MCNP
  - Comparison of experiments with simulation validates capture gamma evaluations and identifies deficiencies.

THE 6TH BIANNUAL WORKSHOP FOR INELASTIC AND ELASTIC NEUTRON SCATTERING (WINS 2023)

October 10 - 12, 2023
Rensselaer Polytechnic Institute

In-person workshop at:

Gaerttner LINAC Center
3021 Tibbits Avenue, Troy NY, USA 12180

The topics of interest include:

- Recent experimental results
- New experimental setups and techniques
- Theoretical developments and advance-ments
- Nuclear data evaluations
- Covariance and uncertainty analysis

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