Measurements and evaluation of structural materials at RPI

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International Nuclear Data Evaluation Network (INDEN) Evaluated Nuclear Data of the Structural Materials, December 16-20, 2024, IAEA, Vienna

Recent relevant work

- Iron (Sukhjinder Singh)
  - Fe-54 capture and transmission in the keV region.
  - Evaluation of RRR in progress

- Zr evaluation (Greg Siemers)
  - New evaluation in progress for ^{90}Zr

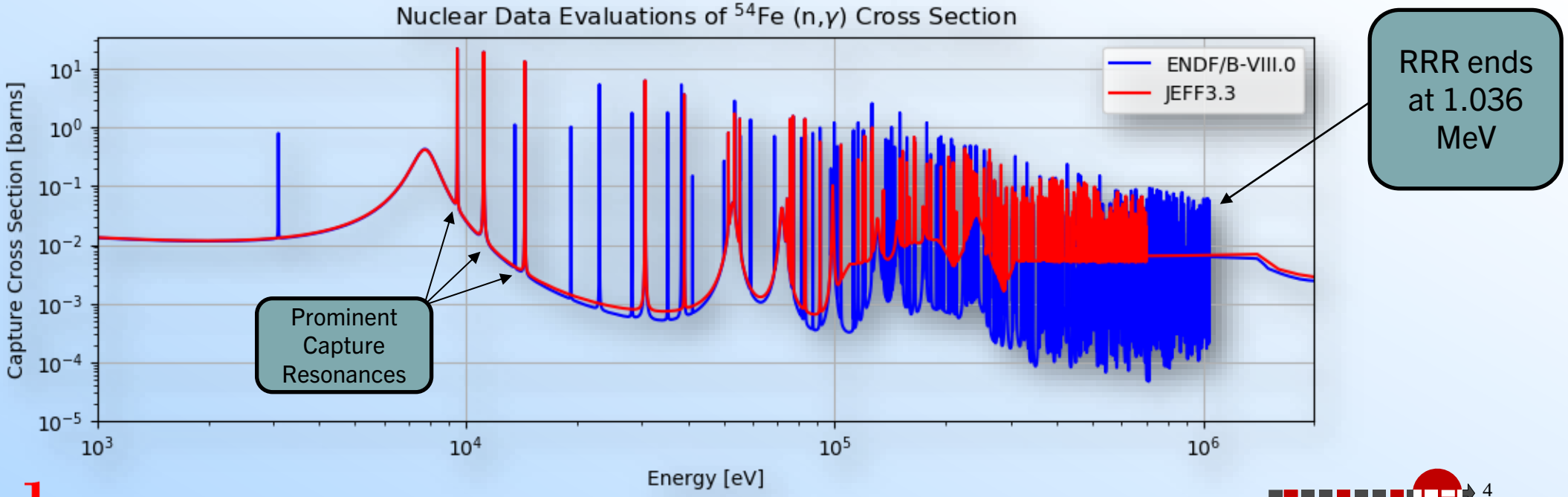
- Quasi differential scattering for F and Ta (Greg Siemers)

- Neutron capture gamma cascade spectra validation (Katelyn Keparutis, Ian Parker)

Fe-54 measurements and evaluation

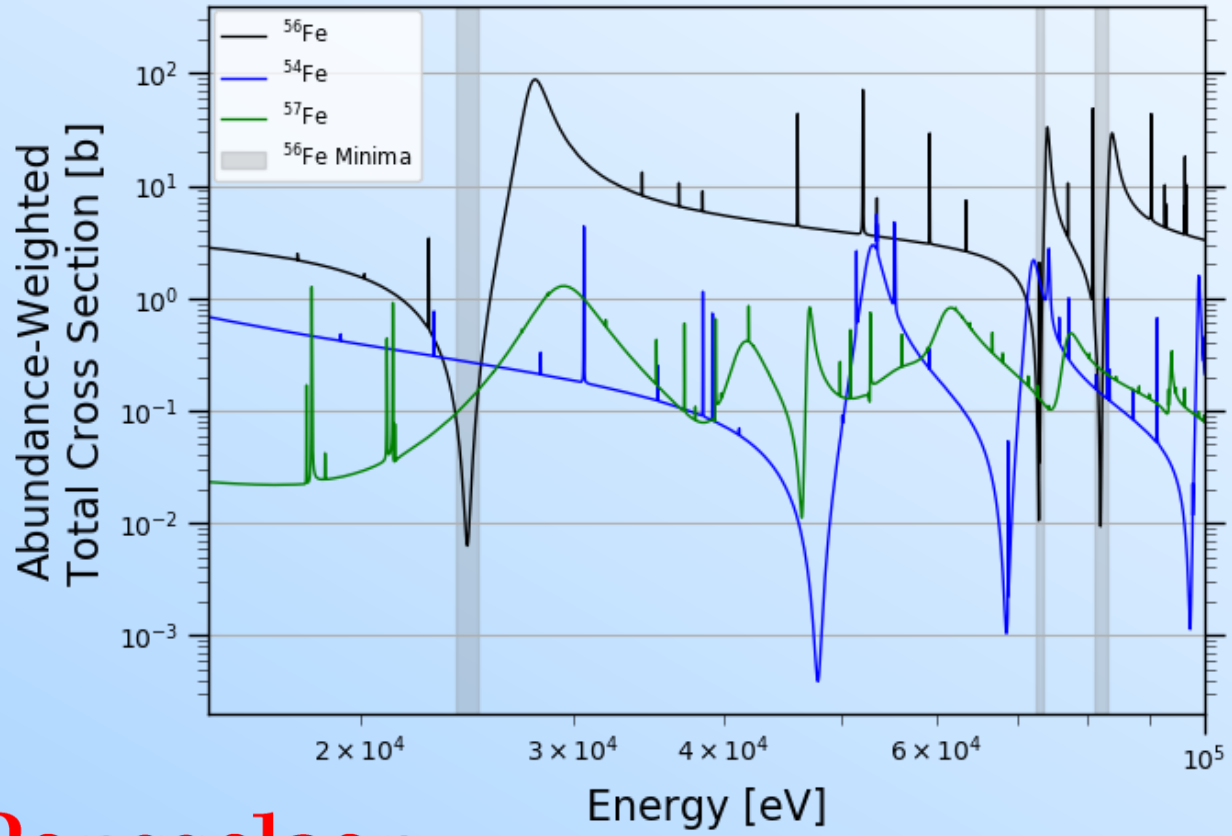
^{54}Fe Project Overview

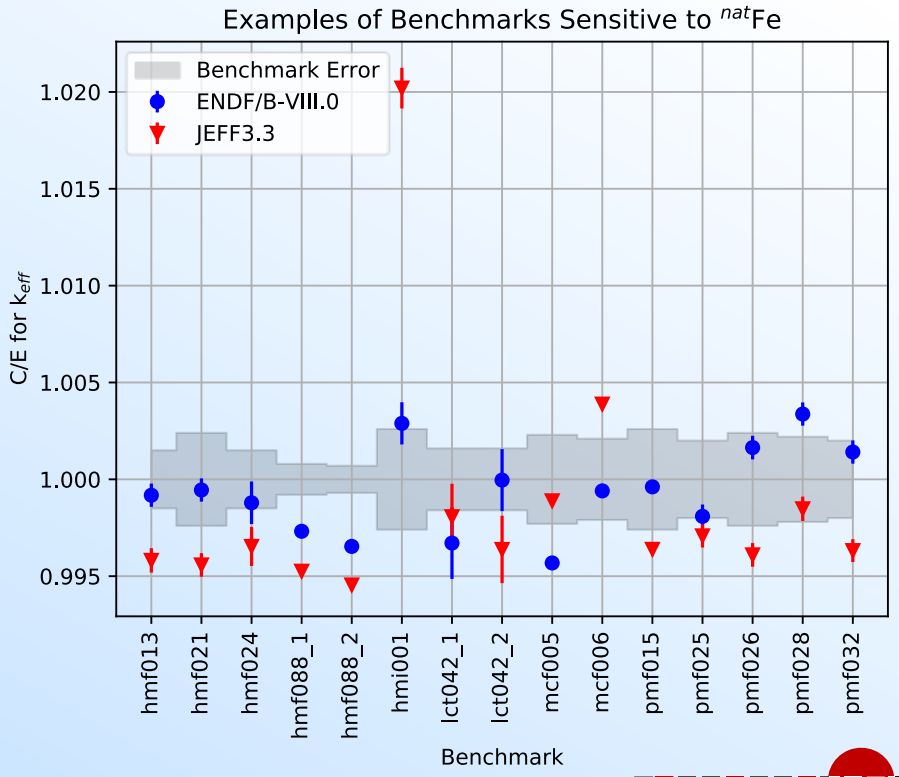
- **Motivation:**
  - Criticality safety calculations require high accuracy nuclear data to reduce uncertainties
  - ^{54}Fe neutron cross sections have not been well-studied relative to ^{56}Fe.
    - Iron is important in shielding, criticality safety, and stellar nucleosynthesis
- **Project Goals:**
  - Perform new RRR evaluation for ^{54}Fe cross section in the keV region using RPI and EXFOR nuclear data.
    - Evaluation will include covariances along w/ consideration of measurement covariances from RPI experiments.
  - Perform radiative capture and transmission measurements of ^{54}Fe in the keV energy region



Importance of ^{54}Fe Evaluations

- Iron is very important to study with respect to radiation shielding, criticality safety, and stellar nucleosynthesis.
- Since Fe is an important structural material in nuclear reactors, benchmarks can be shown to have sensitivity to ^{nat}Fe cross sections.
- In the minima of ^{56}Fe cross sections, neutrons can stream through a shielding wall and pose a health risk.
  - **The cross sections of minor isotopes is important in the minima of ^{56}Fe.**





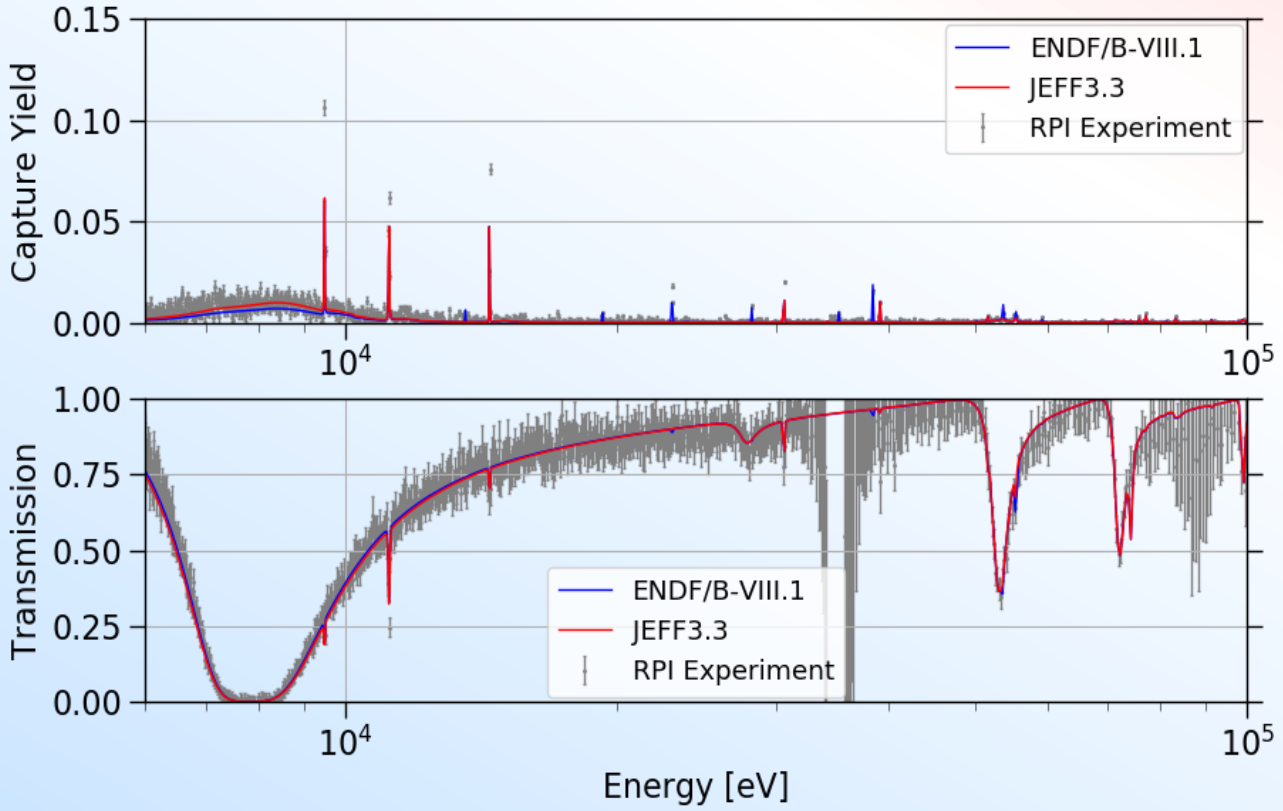
Overview of RPI Experiment Campaign

- Radiative Capture:
  - An array of seven C_6D_6 liquid scintillators surrounding the sample of interest at a flight path of 45m.
  - Enriched Fe-54 sample (~0.021 at/b) measured along with Open, Au, B_4C, Pb samples
  - Pulse height weighting used to reduce data down to capture yield
  - Useful to ~150 keV
- Transmission:
  - Li-6 glass detector at 35m used to collect sample-in, sample-out data using analog electronics.
  - Enriched Fe54 sample (~0.021 at/b) measured along with Open, Depleted Uranium, and Natural Fe.
  - Fixed notch materials kept in beam during the duration of the experiment.
  - Useful to about ~150 keV, will be used to fit resonance in the low energy region.

Overview of RPI Measurement Campaign

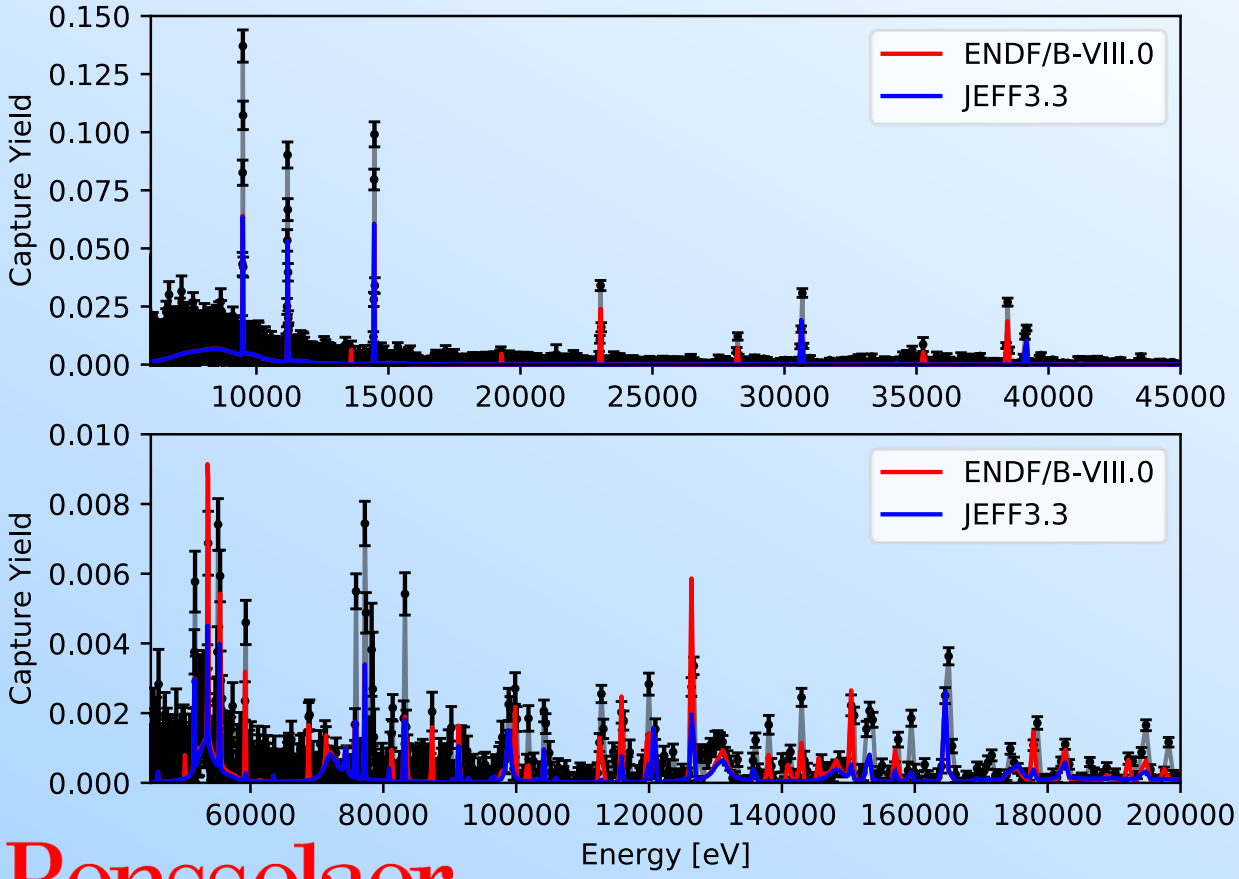
- RPI previously conducted transmission and radiative capture yield measurements as a part of this work
- 0.021 a/b enriched metallic ^{54}Fe sample was used for both experiments.
- Full energy-energy correlations were generated.
  - Transmission can capture data include **experimental covariances**.

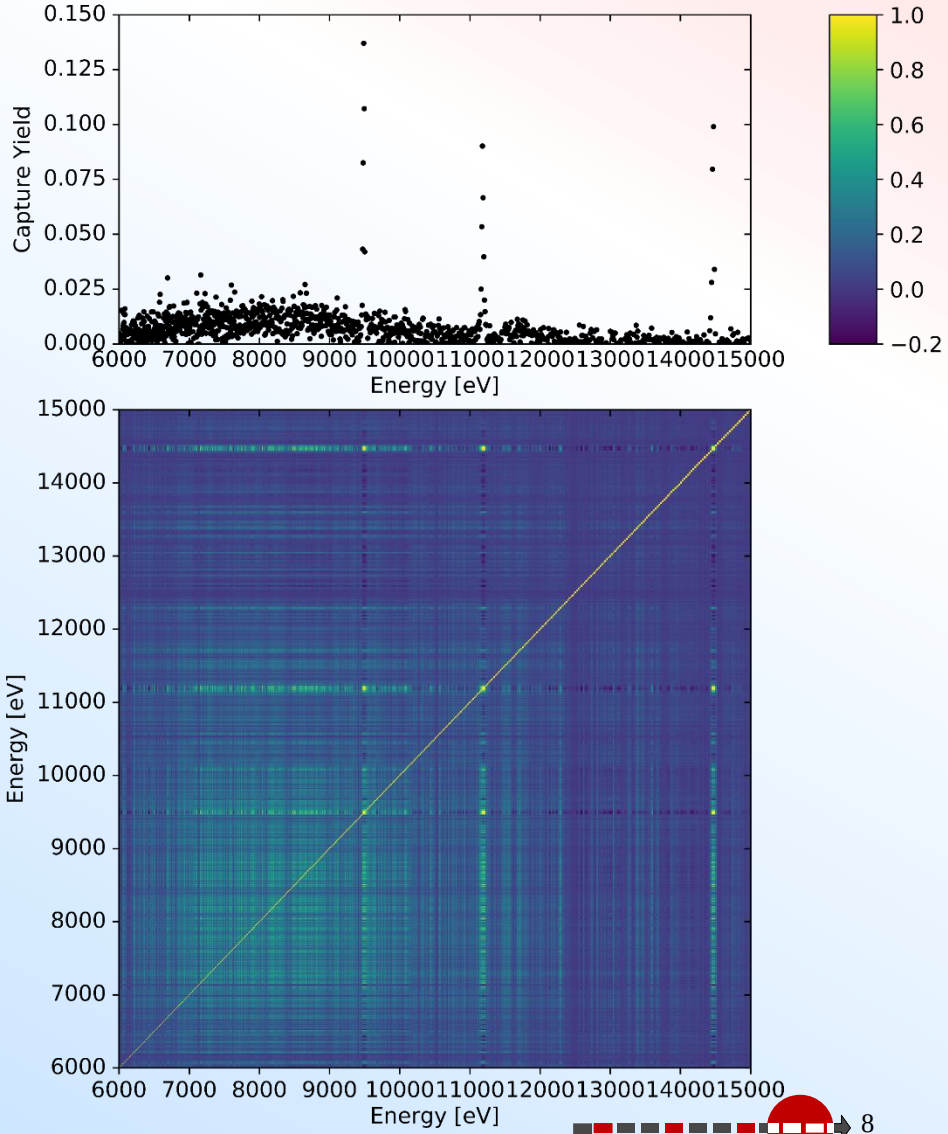
Parameter	Capture	Transmission
Flight Path [m]	45	35
Energy Region [keV]	1-150	0.01 - 150
Detector	C_6D_6	^{6}Li Glass



^{54}Fe Capture Experiment

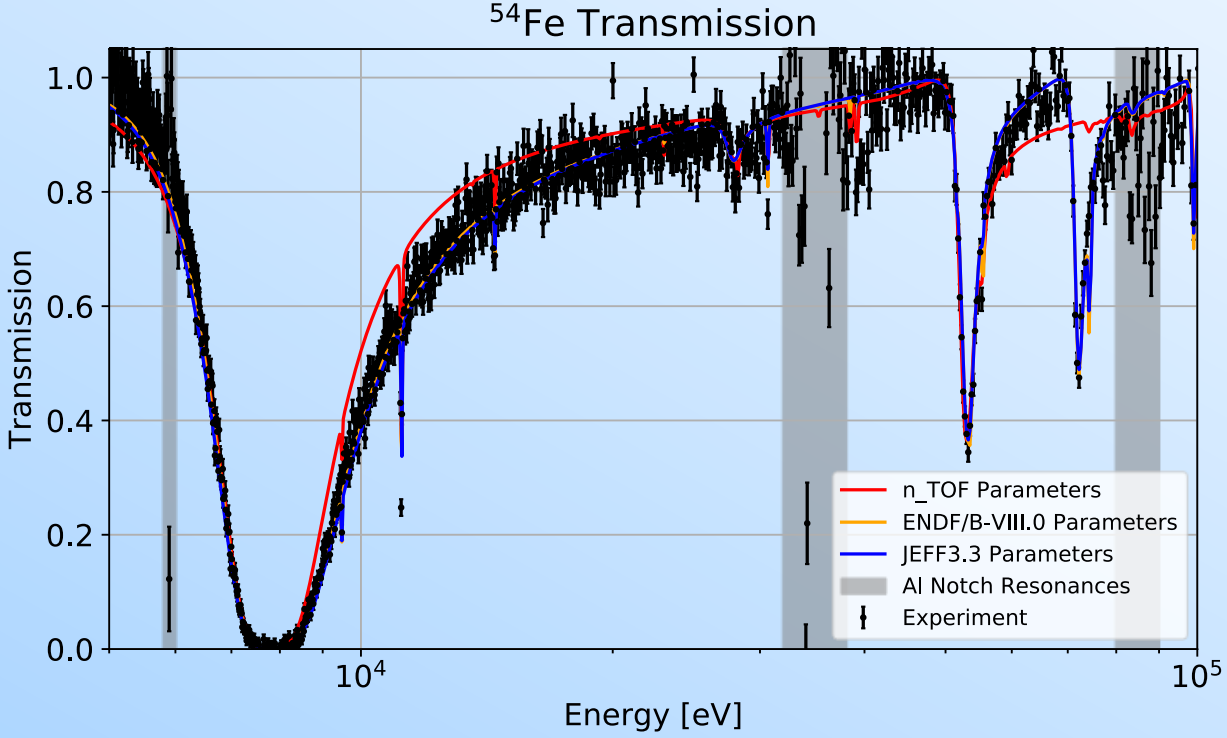
- Capture yield shows large discrepancies.
- Stronger correlations between resonances are present in the experiment.

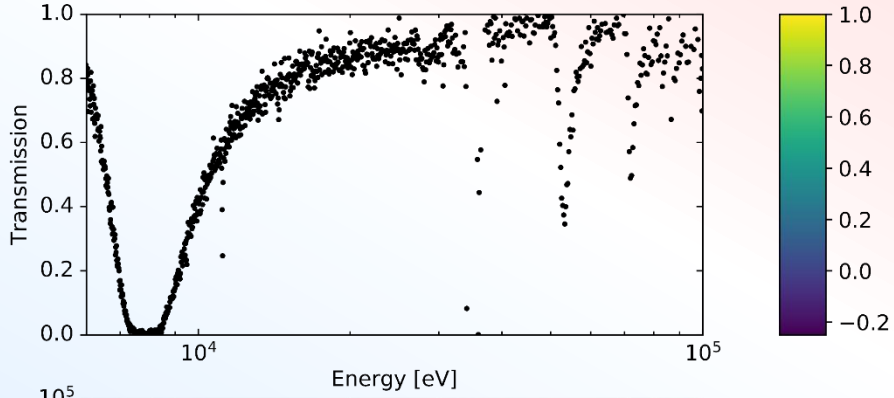


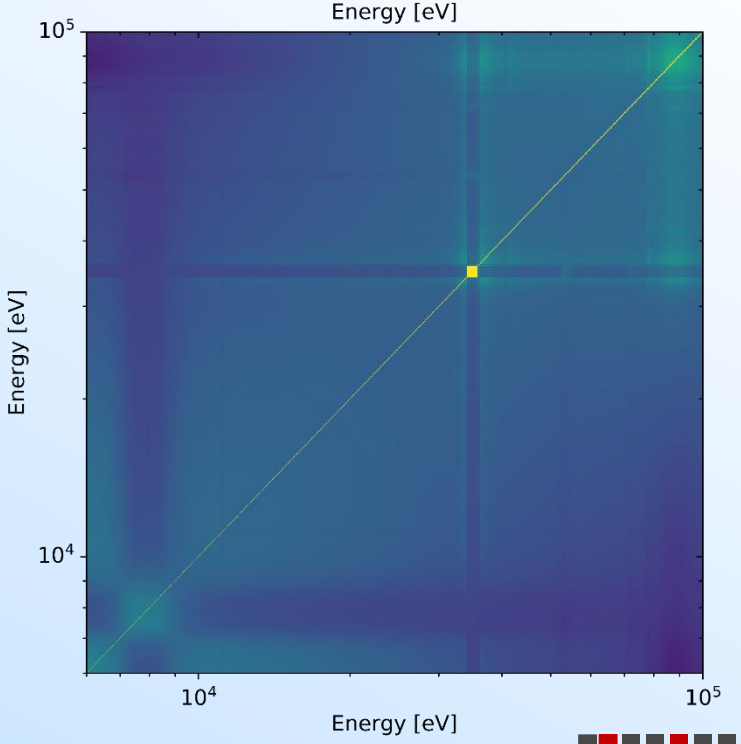


^{54}Fe Transmission Experiment

- Transmission is less sensitive to changes in evaluations.
- Covariance passes all mathematical checks.
- Small correlations are present in the transmission experiment.







RPI Evaluation Results

Experiment or Evaluation	Value [mb] @ 30 keV
ENDF/B-VIII.0	27.13
RPI Eval	29.8 ± 0.9
KADoNiS-0.3	29.6 ± 1.3
n_TOF Exp	30.8 ± 1.6
Allen Exp	33.6 ± 2.7

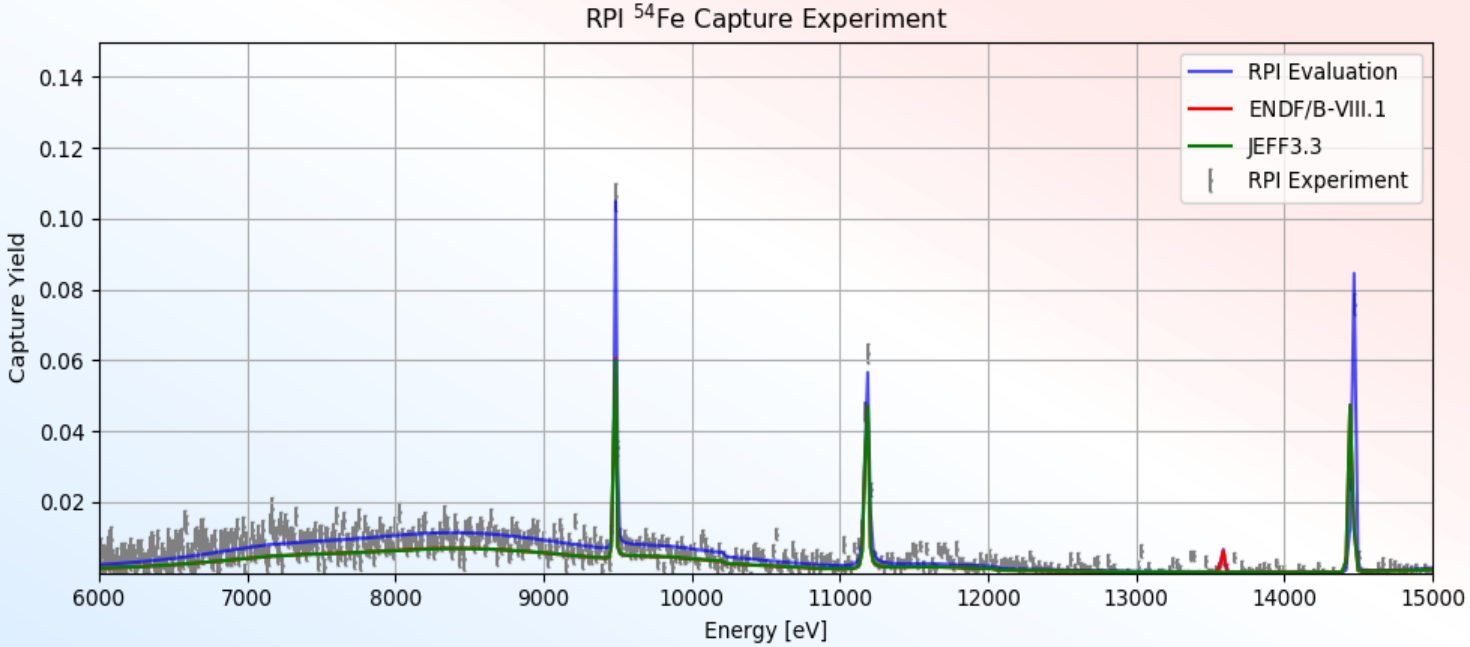
Source	σ_t [barns] at 0.0253 eV
RPI Evaluation	4.46 ± 0.05
ENDF/B-VIII.0	4.43
JEFF-3.3	4.45
NIST	4.45
Atlas	4.47 ± 0.12

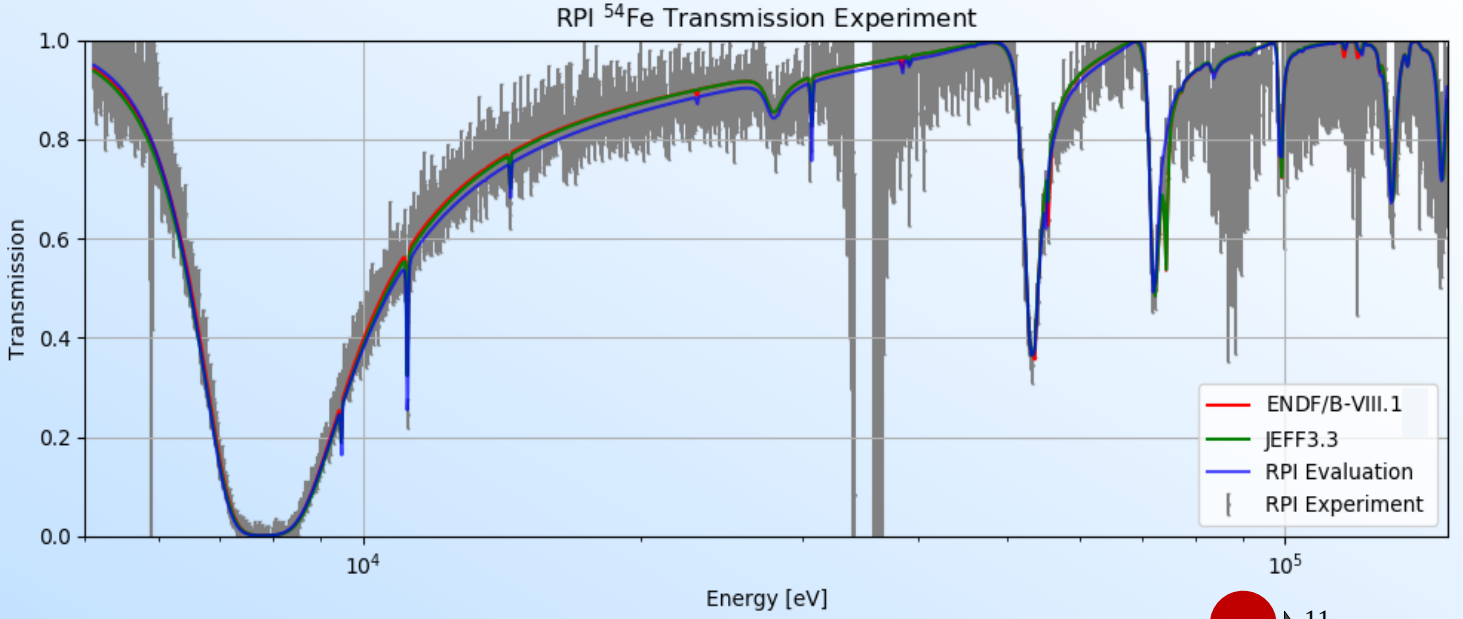
Source	σ_γ [barns] at 0.0253 eV
RPI Evaluation	2.27 ± 0.02
ENDF/B-VIII.0	2.25
JEFF-3.3	2.25
Litvinskij Capture	2.28
Wallner Capture	2.26 ± 0.15
NIST	2.25
Atlas (2018)	2.30 ± 0.07

Experiment	Value @ 481 keV
Wallner	6.01 ± 0.28 [mb]
n_TOF Exp	6.04 [mb]
RPI Eval	6.10 ± 0.14 [mb]

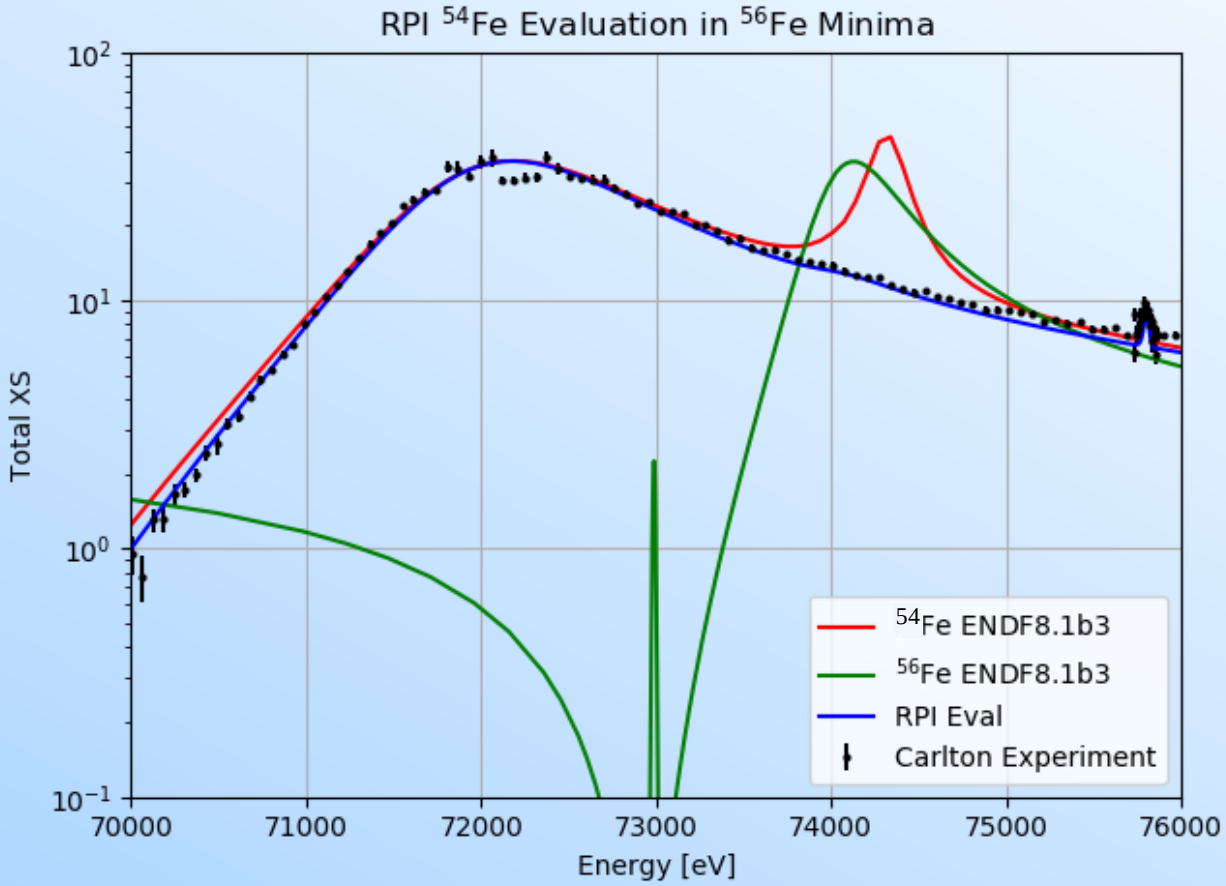
RPI Evaluation

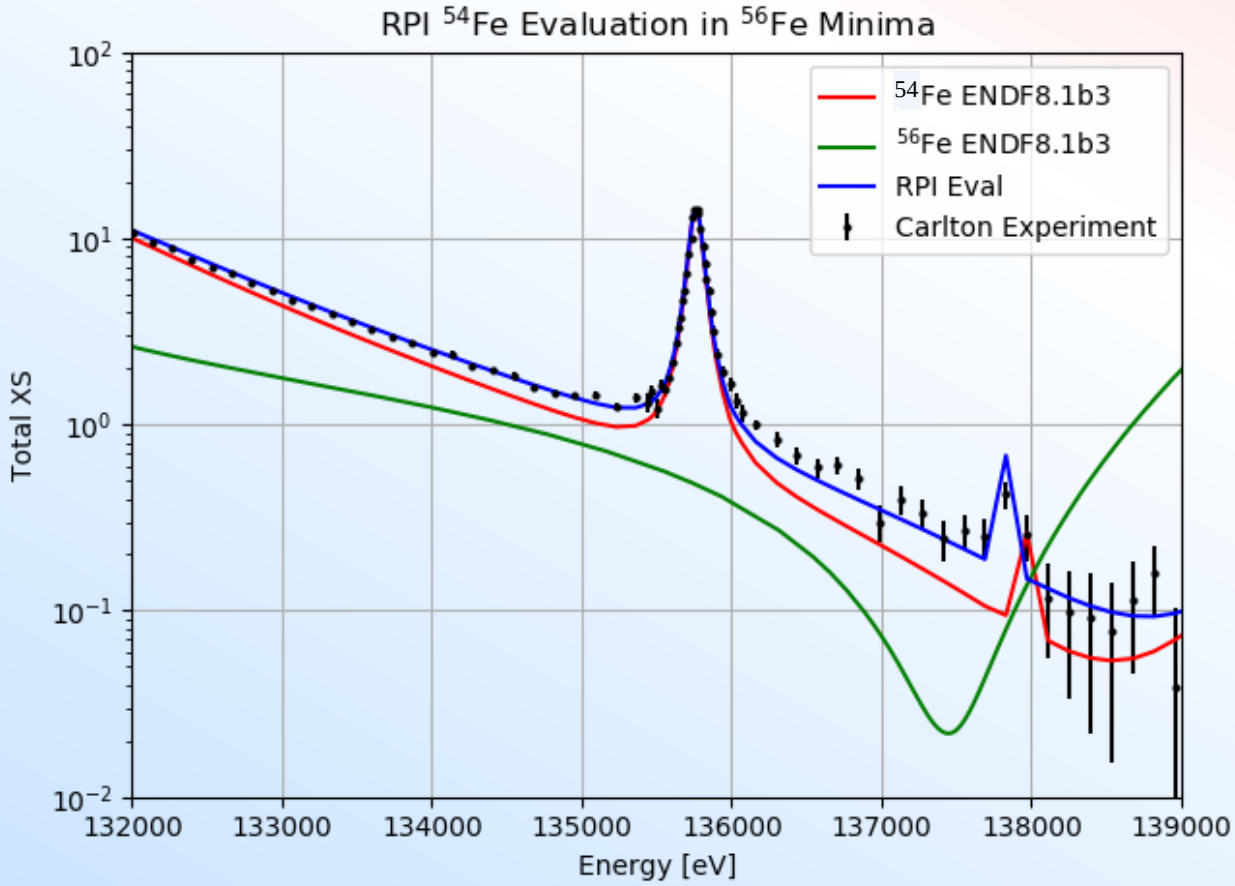
- Capture widths were increased for prominent capture resonances
  - Both experiments suggested an increase was needed in the capture cross section for these d-wave resonances
- The direct capture process was considered in the evaluation, a direct capture background was discarded in favor of bound levels.
  - Collaboration w/ Luiz Leal (ORNL) and Goran Arbanas (ORNL) to determine magnitude of direct capture cross section.
- **Resonance parameter fitting considered experimental data covariances.**
  - This has not been done in the past.





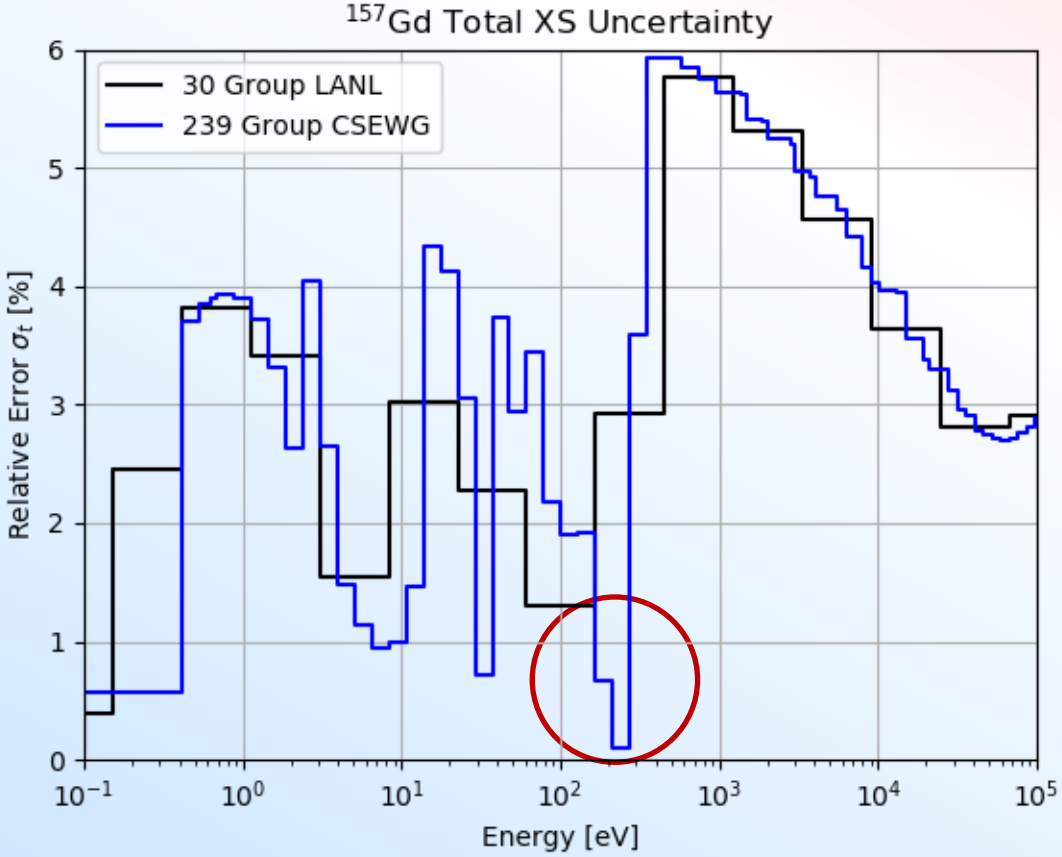
^{54}Fe Changes Near ^{56}Fe XS Minima





The Issue with Nuclear Data Covariances

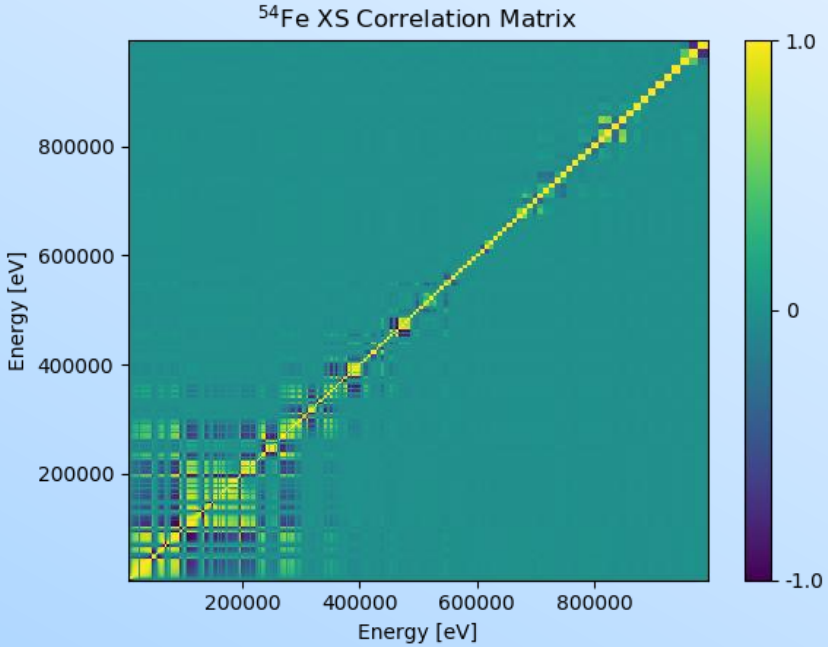
- Covariance information is required when submitting a new evaluation to ENDF
- Two issues arise with this approach:
  - **Resonance parameter covariances when converted to cross section covariances lose systematic uncertainties**
  - As a result, grouped cross section uncertainties are sensitive to the grouping structure chosen.
- Several examples of accepted evaluations in ENDF/B-VIII.1 show issues when inspecting covariance data

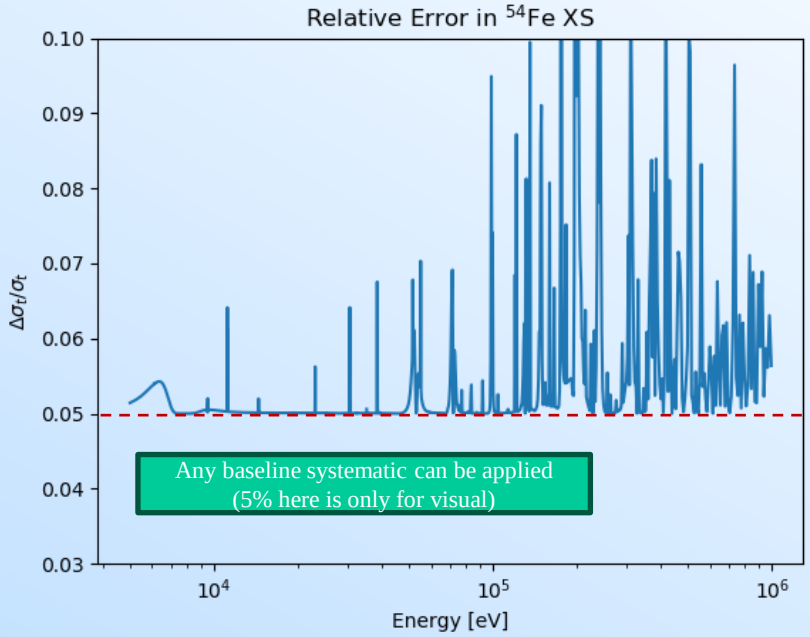


A Proposed Solution for Future Evaluations

- A different and novel approach for including uncertainties into the ENDF format is currently being investigated.
  - Compatible with ENDF format, preserves correlations and constrains total XS.
  - ENDF file contains MF=32 and systematics are propagated separately into MF=33.

$$V = g^{\times} m^{\times} g'$$



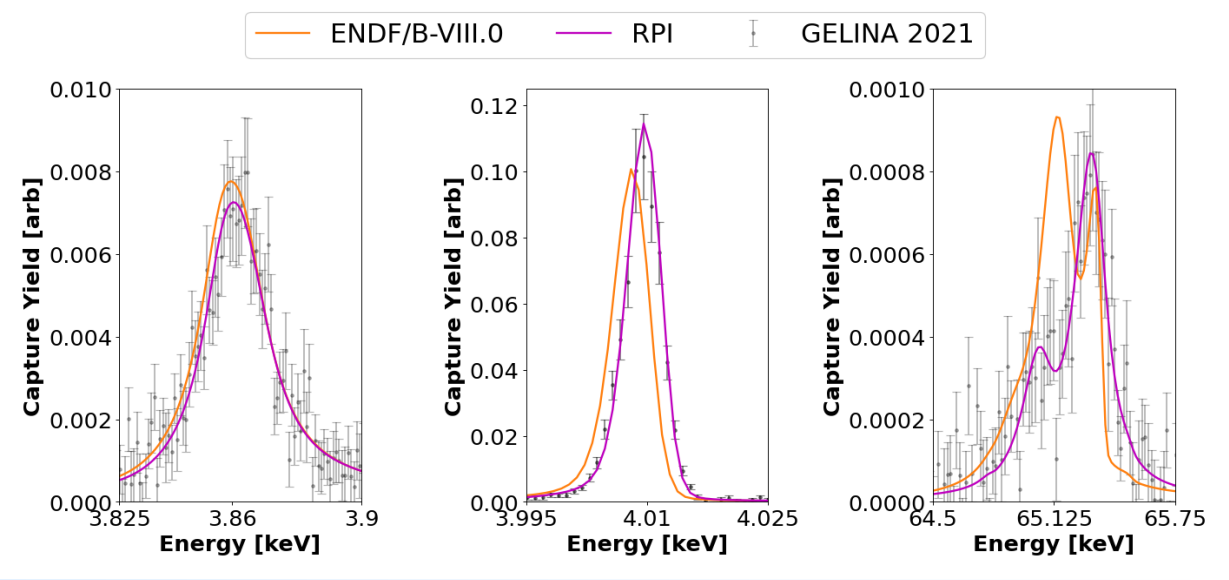


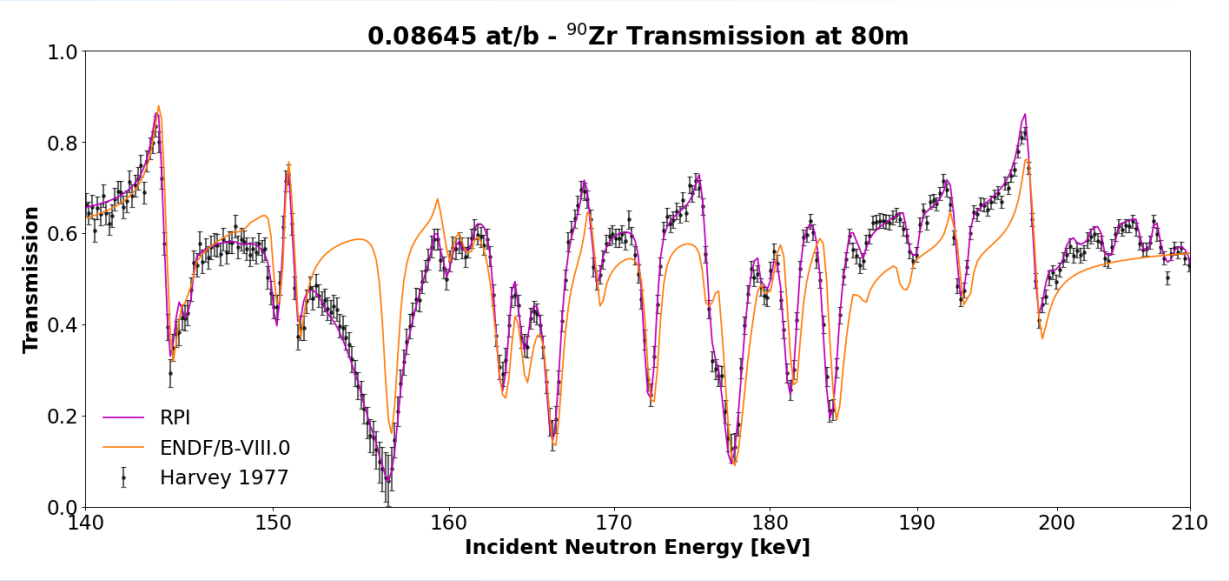
$\boldsymbol{V}$ = Full xs covariance matrix
$\boldsymbol{g}$ = Partial derivatives of resonance parameters + nuisance variables w.r.t. cross section
$\boldsymbol{m}$ = Covariance matrix of resonance parameters + nuisance variables

^{90}Zr Evaluation

^{90}Zr Resonance Evaluation

- Datasets fitted in evaluation:
  - **Transmission:**
    1. de L. Musgrove, et al. 1977[1] - 0.08645 at/b metallic enriched ^{90}Zr @ 80m (^{6}Li)
    2. de L. Musgrove, et al. 1977[1] - 0.08645 at/b metallic enriched ^{90}Zr @ 200m (NE110)
  - **Capture:**
    1. GELINA 2021 - 0.00558 at/b metallic enriched ^{90}Zr @ 48m (C_6D_6)
- Channel radius of 6.31 *fm* adopted from Fröhner recommendation[2]
  - Distant levels to represent R_∞

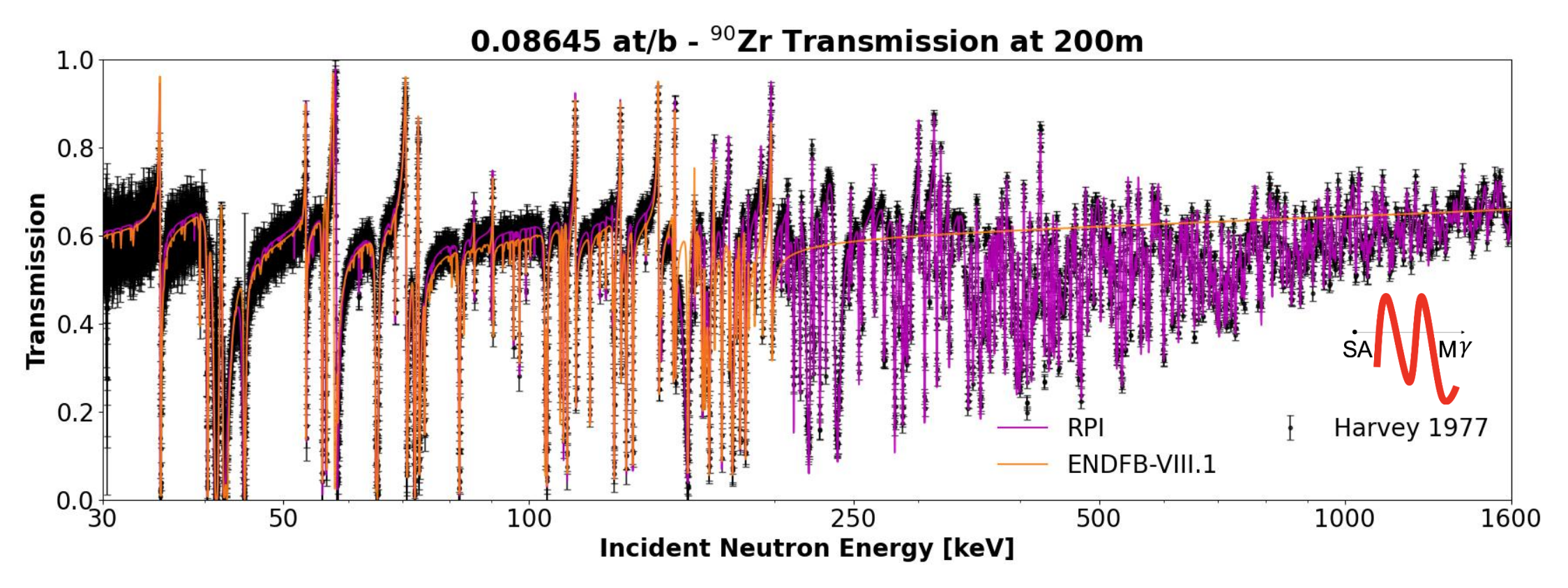




1. de L Musgrove, Harvey, and Good *Neutron Resonance Parameters of 90Zr Below 300 keV*, Aust. J. Phys, 1977, **30,** 379-89
2. Frohner and Bouland *Treatment of External Levels iun Neutron Resonance Fitting: Application to the Nonfissile Nuclide 52Cr* Nucl. Sci. Eng. 2001, **137,** 70-88

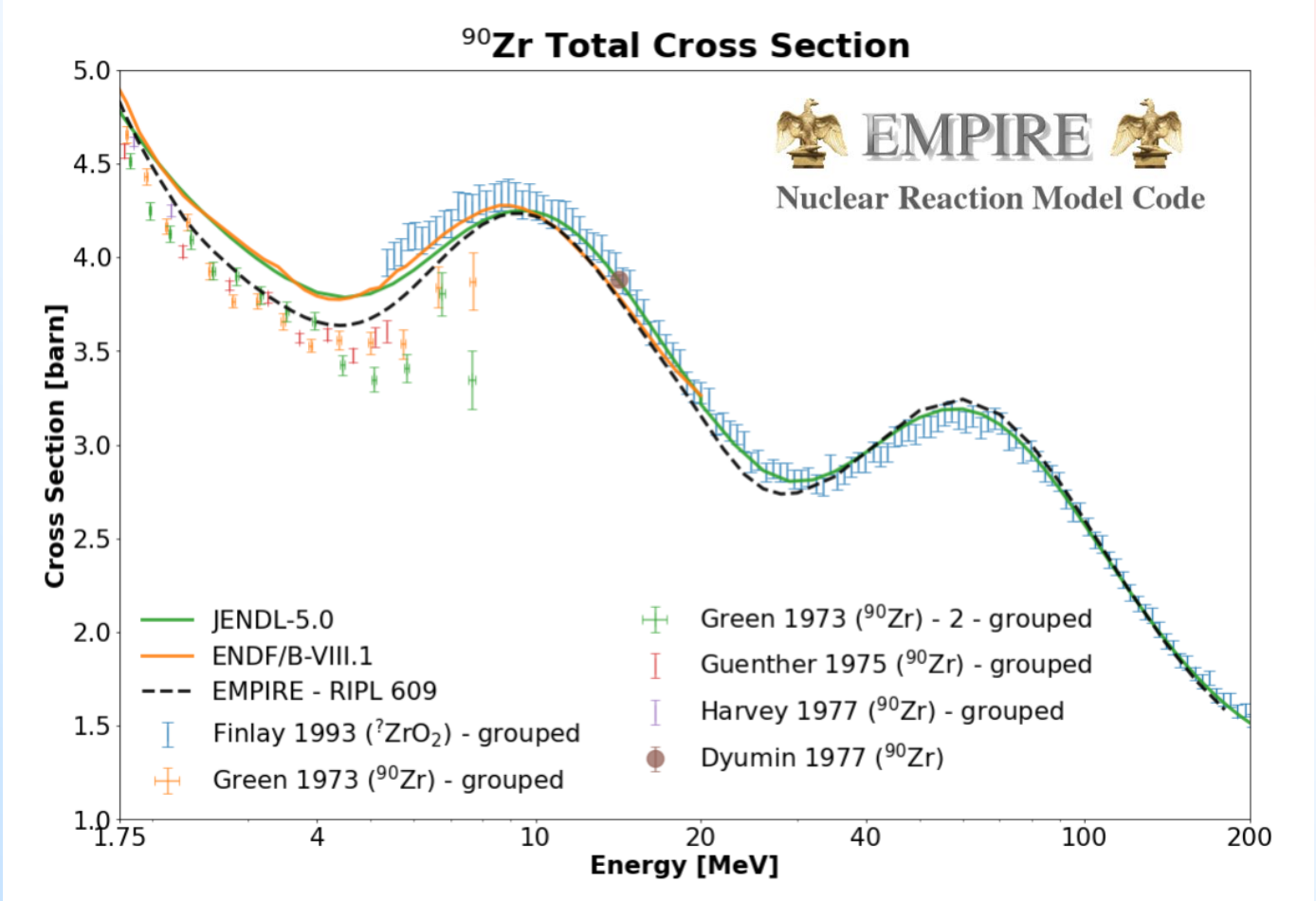
^{90}Zr Resolved Resonance Region Extension

Resonance parameters fit to ^{90}Zr transmission data up to first excited state, 1764 keV.
Limit of current ENDF/B-VIII.1 RRR is 200 keV.



Beginning ^{90}Zr Fast Region Evaluation

- New soft rotational coupled-channels optical model potential (OMP) developed by Capote and Soukhovitskii for Zr isotopes (RIPL 609)
- RIPL 609 shows good agreement to experiment, particularly below 10 MeV
- Finlay 1993 sample measured suspected to be elemental Zr abundance rather than enriched $^{90}ZrO_2$

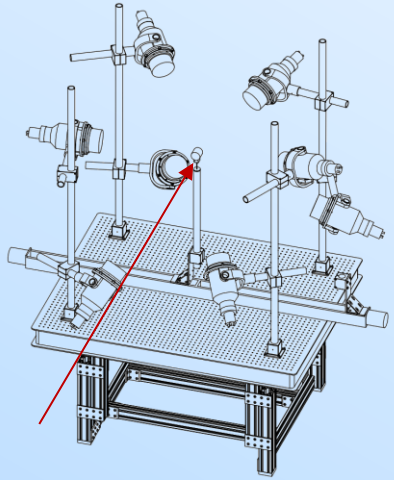


High Energy Quasi-Differential Neutron Emission Measurements from ^{181}Ta and ^{19}F

Quasi-Differential Measurement Methodology

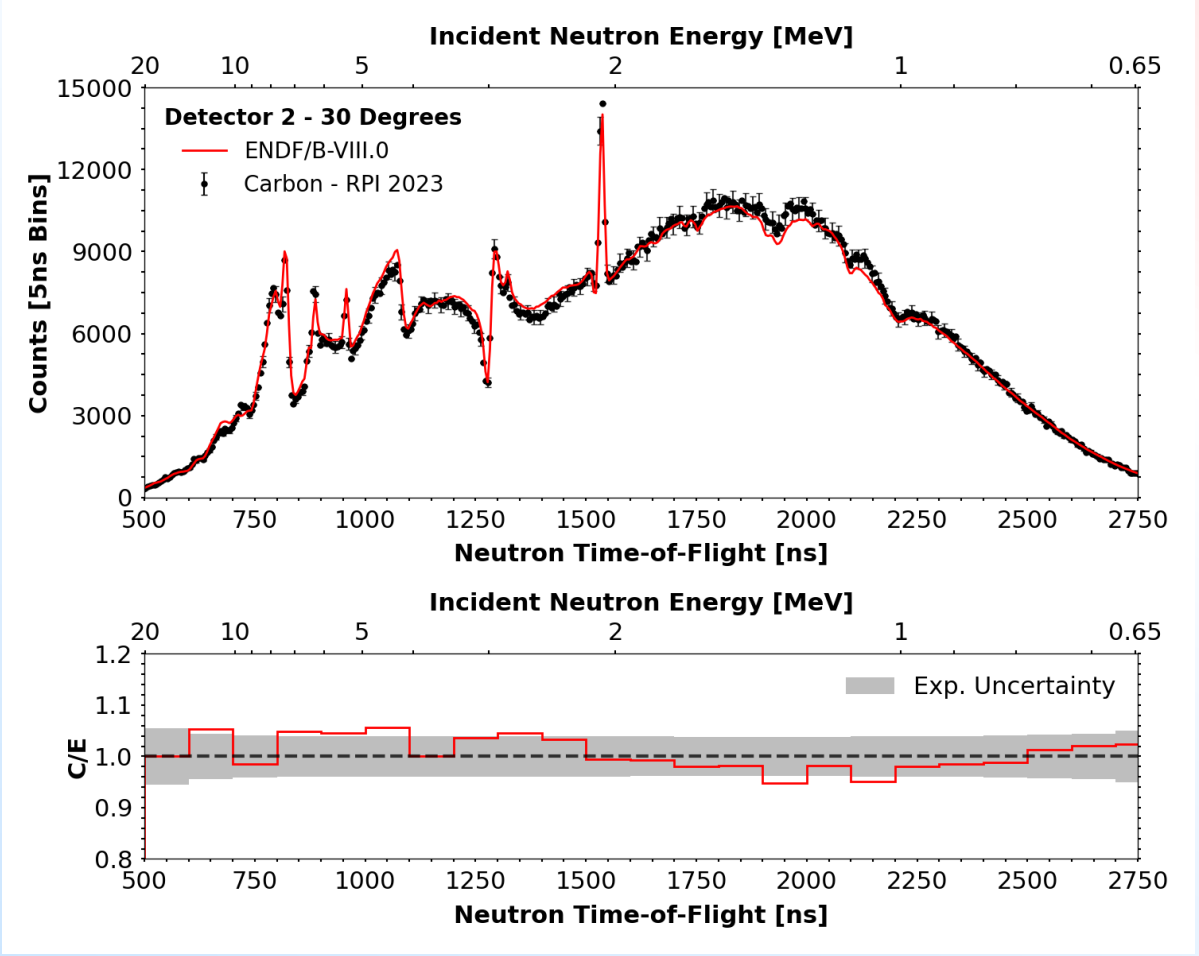
1. Conduct differential neutron time-of-flight experiment on sample of interest, validation sample, and open beam
   - Due to sample size, the experiment is dominated by multiple scattering interactions
2. Perform MCNP transport calculation of validation (Carbon) measurement using measured neutron flux and detector efficiencies
   - This validates experimental geometry and reproduction of known validations sample
3. Perform MCNP transport calculation of sample of interest measurement using measured neutron flux and detector efficiencies
   - Differences present in nuclear data evaluations of the sample of interest are compared to the experimental data to validate performance or show needs for improvement

Measured Ta-181 and TEFLON $(C_2F_4)_n$ with the objective of getting data on F.



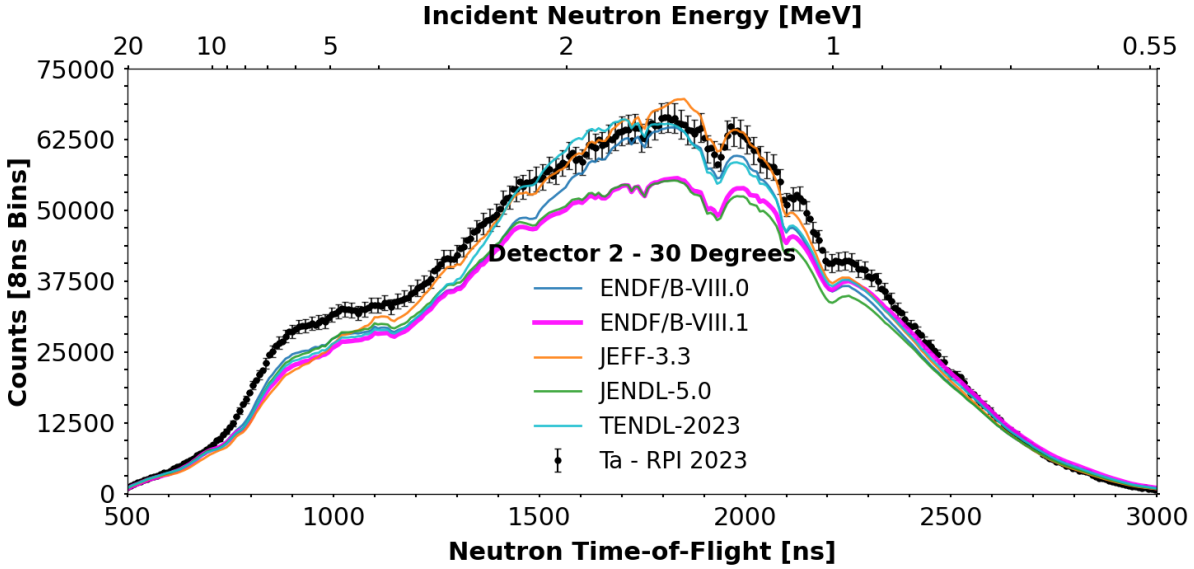
Validation Measurements of a Carbon Sample

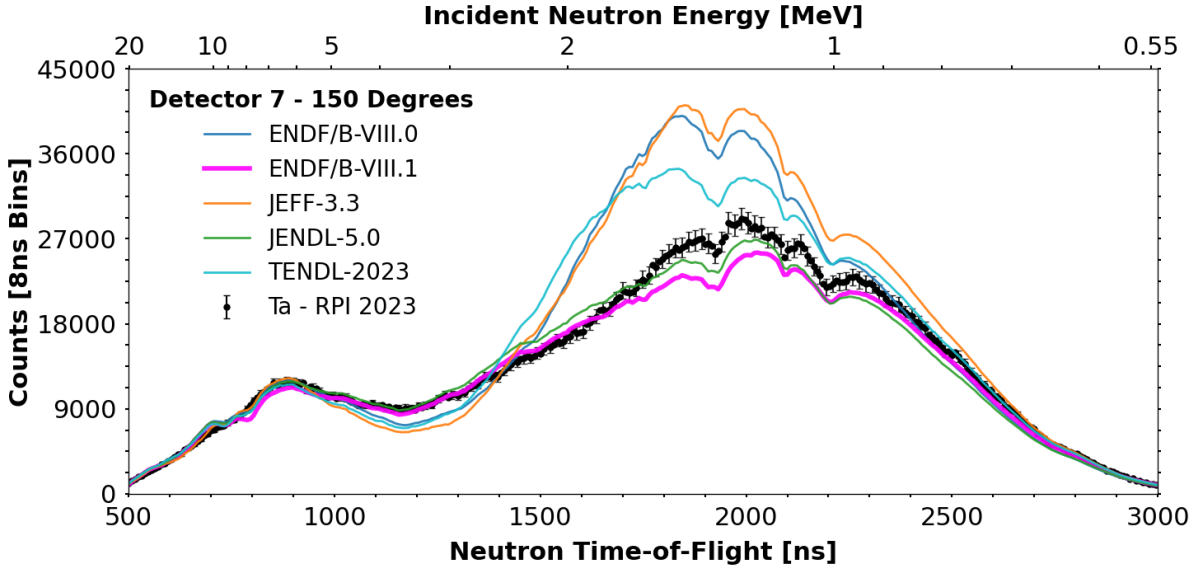
- Deviations of measured carbon data from evaluation adopted as experiment systematic uncertainty
  - 3.9% and 3.4% for Ta and Teflon experiments respectively
  - Improved from previous measurements[1,2] where systematics were ~6%
- Detector efficiencies determined from in-beam measurements
- Deadtime correction utilized legacy RPI algorithms recently validated for SIS-3305 digitizer[3]



1. E. Blain et. al, *Nucl. Sci. and Eng.*, **196**, 2, 121-132 (2022)
2. Daskalakis A. et. al, *Ann. Nucl. Eng.* **110**, 603-12 (2017)
3. B. J. McDermott, *Ph.D. Thesis, Rensselaer Polytechnic Institute (2016)*

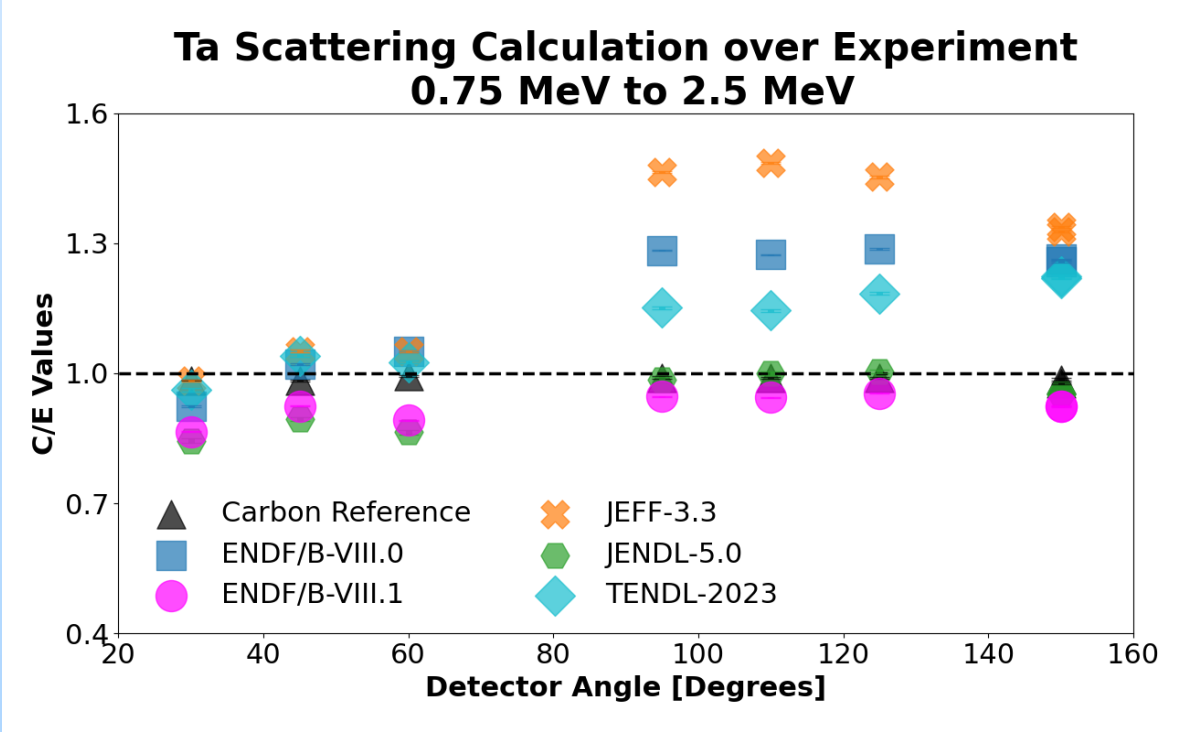
Ta - Some Issues Remain at Very Forward Angles, but Large Improvements in Backward Angles

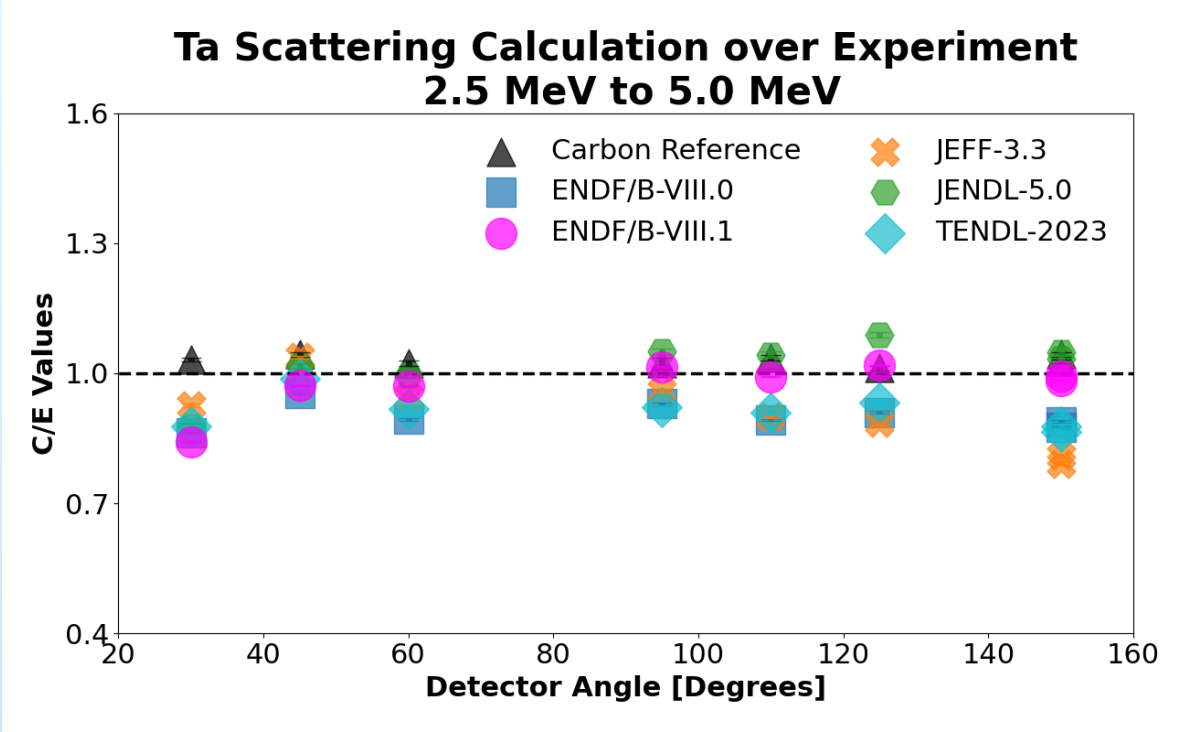




Tantalum Scattering Kernal Performance

Improvements made over previous ENDF/B-VIII.0 ^{181}Ta evaluation to resolve the significant overprediction of neutron scattering at backward angles in the fission neutron energy regime.

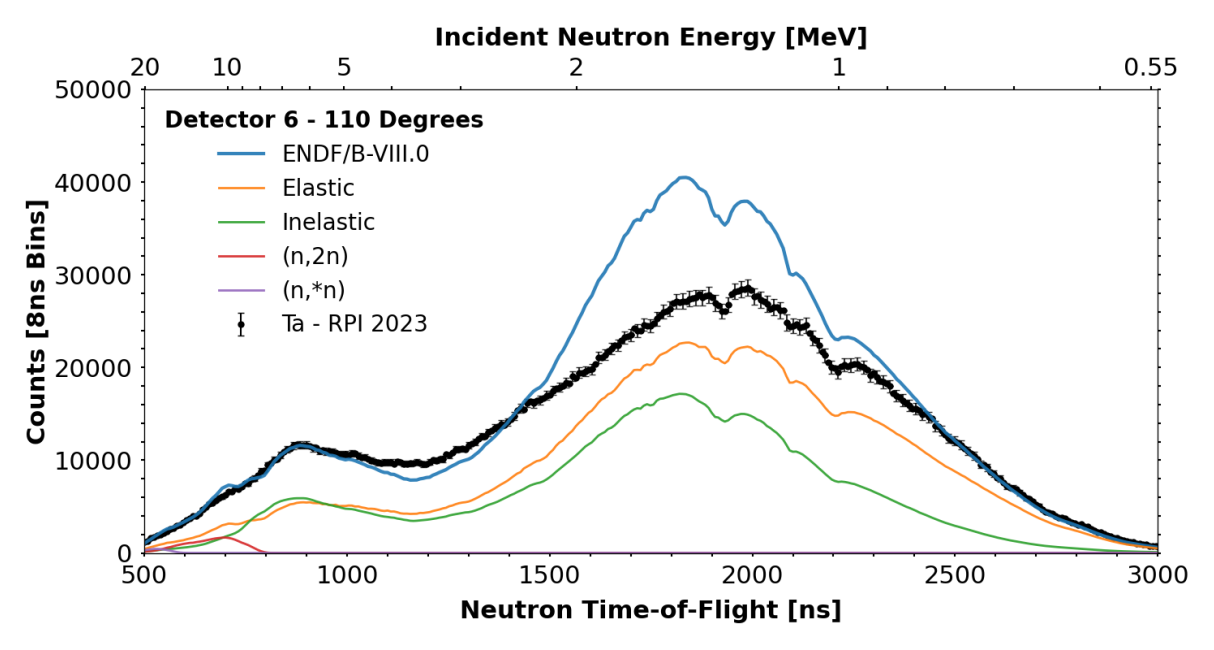


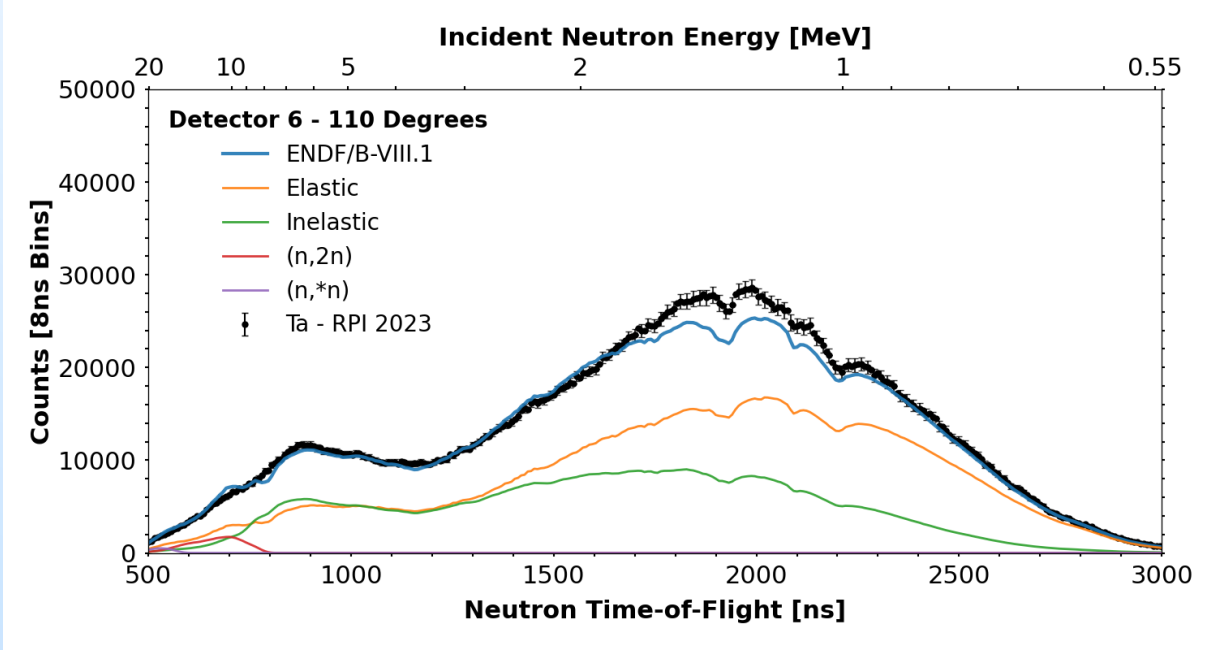


Changes Observed in Scattering Channels

Moving from ENDF/B-VIII.0 to ENDF/B-VIII.1 a large reduction of both elastic and inelastic neutron scattering is observed below 3 MeV at backward scattering angles to correct the large imbalance in the tantalum scattering kernel.

ENDF/B-VIII.0 → **ENDF/B-VIII.1**

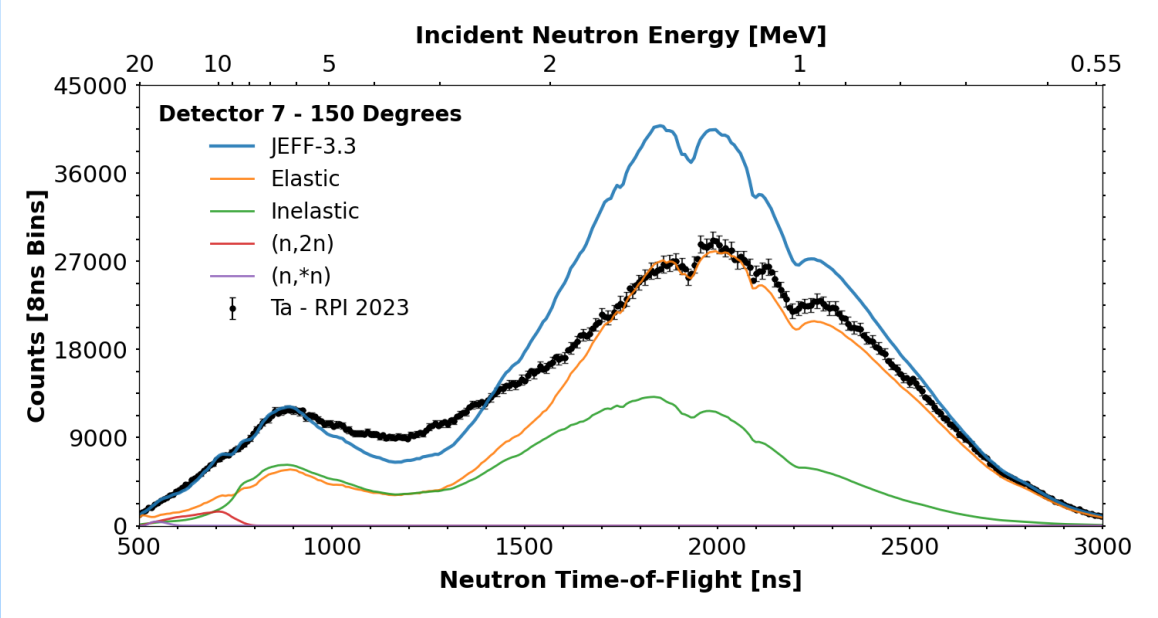


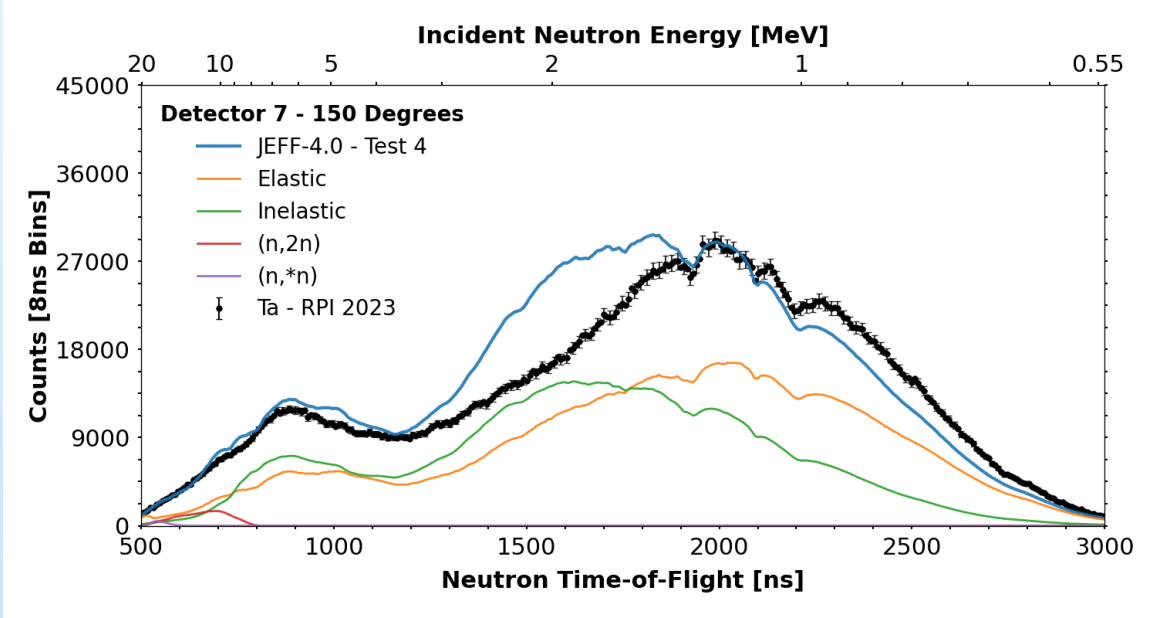


Large Changes Observed in Inelastic Scattering

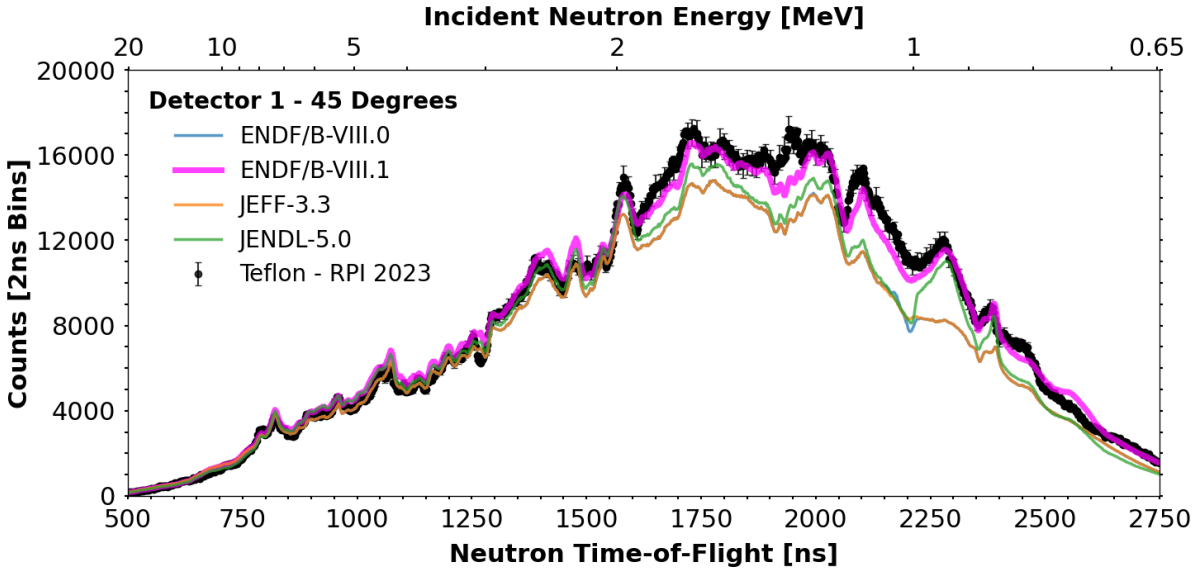
Moving from JEFF-3.3 to JEFF-4 (T4) a large reduction of elastic neutron scattering is observed below 2 MeV along with an increase in inelastic neutron scattering above 1.5 MeV.

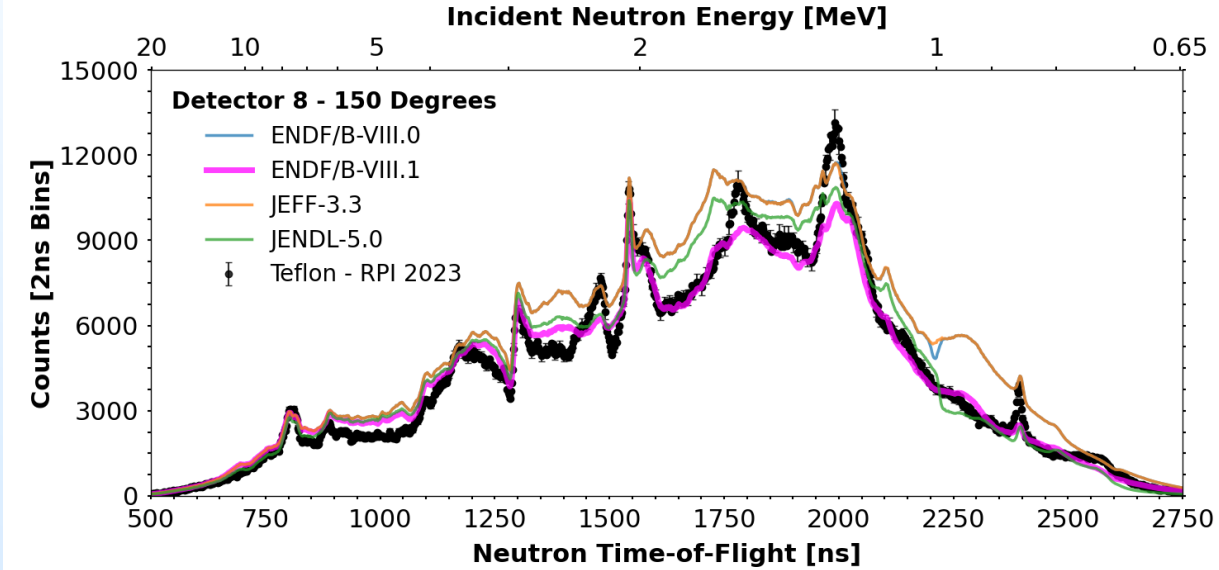
JEFF-3.3 → **JEFF-4 (T4)**





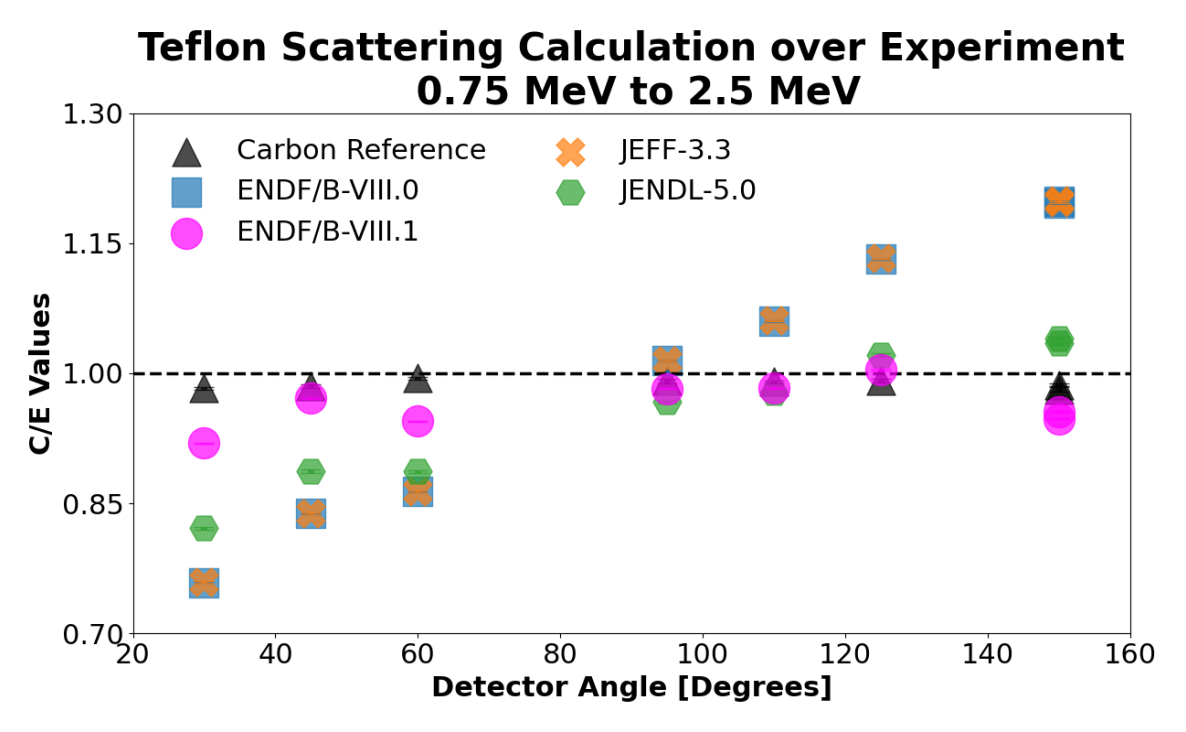
Teflon (^{19}F) – Good Performance at Forward Angles, But Issues With Resonance Anisotropy

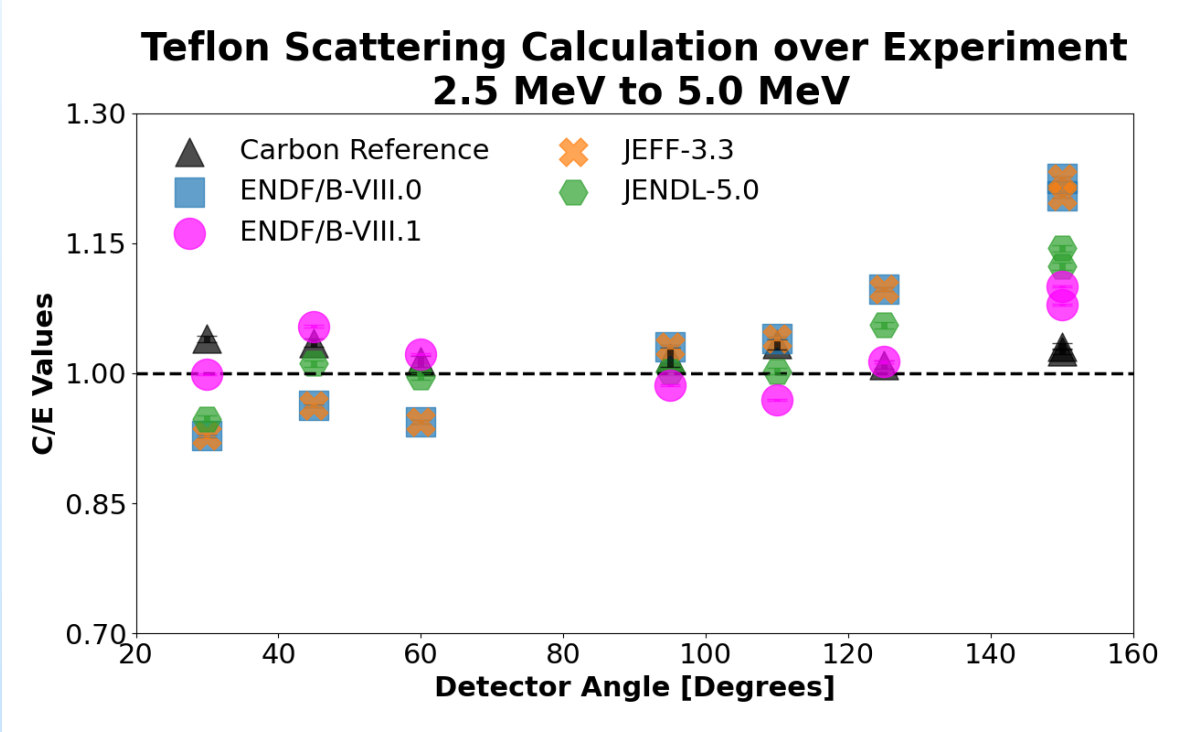




^{19}Fluorine Scattering Kernal Performance

Adoption of INDEN ^{19}F evaluation yields improvement made over previous ENDF/B-VIII.0 ^{19}F evaluation with respect to the experimental data by addressing large scattering kernel imbalance in the fission neutron energy regime.





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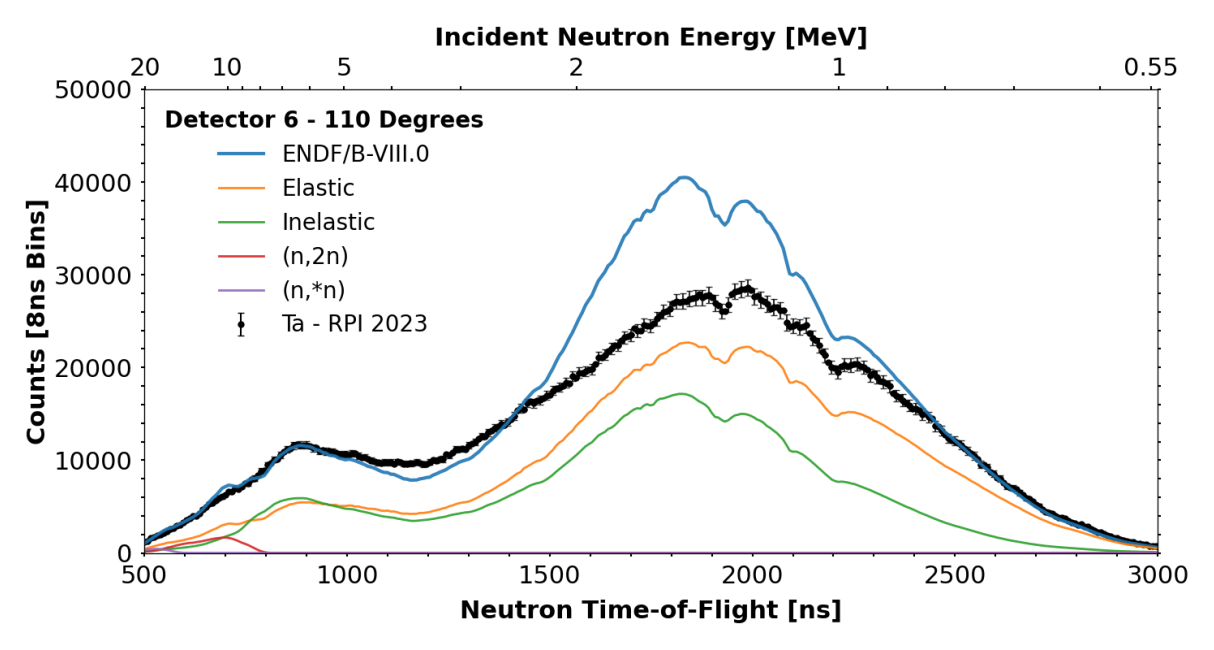
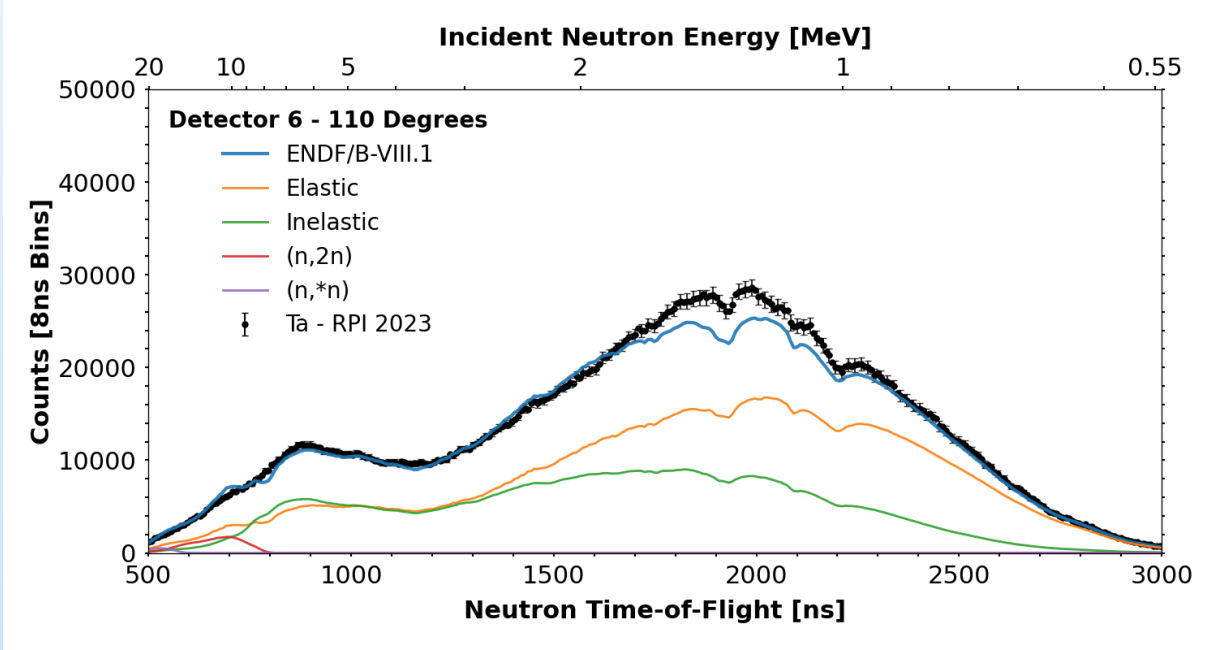
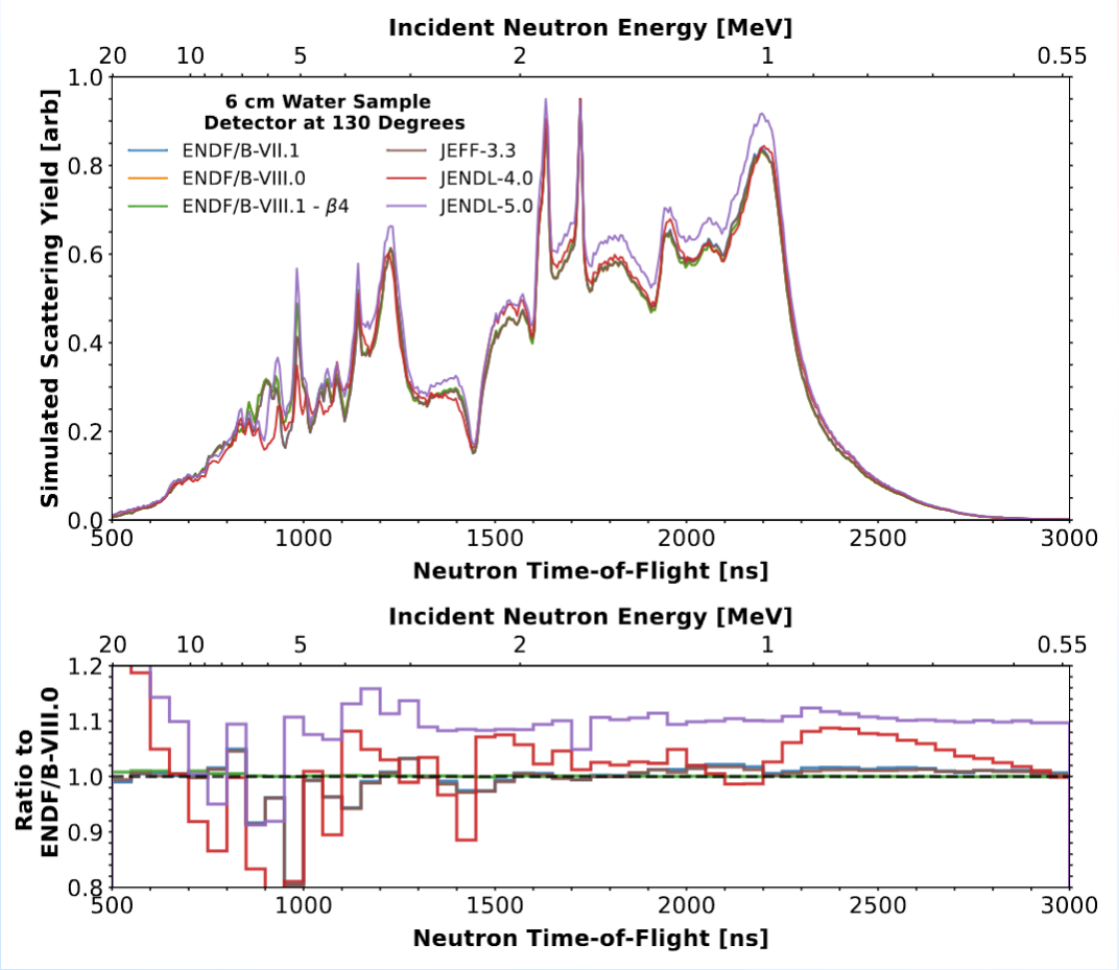


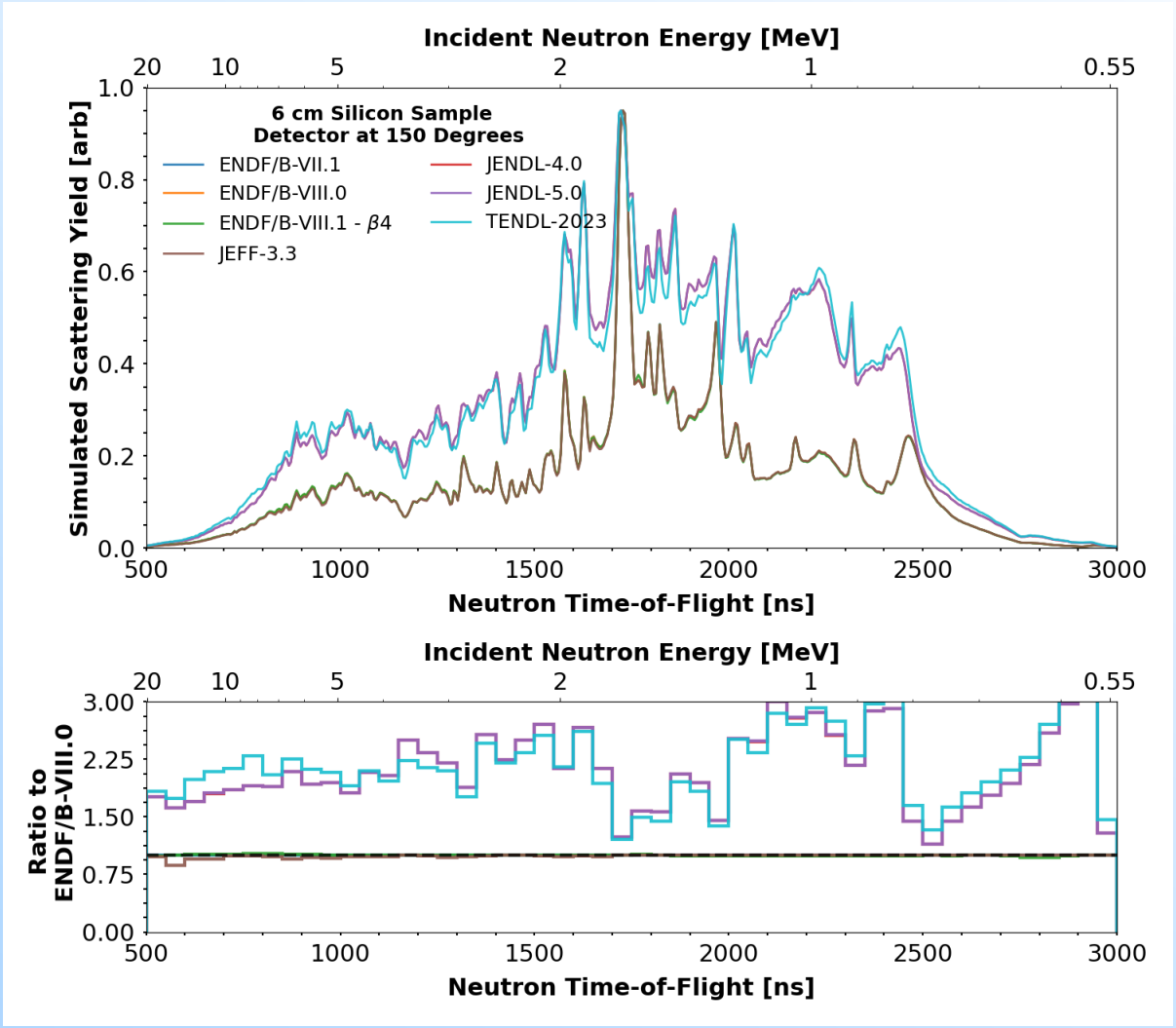


Conclusions and Future Experiments at RPI

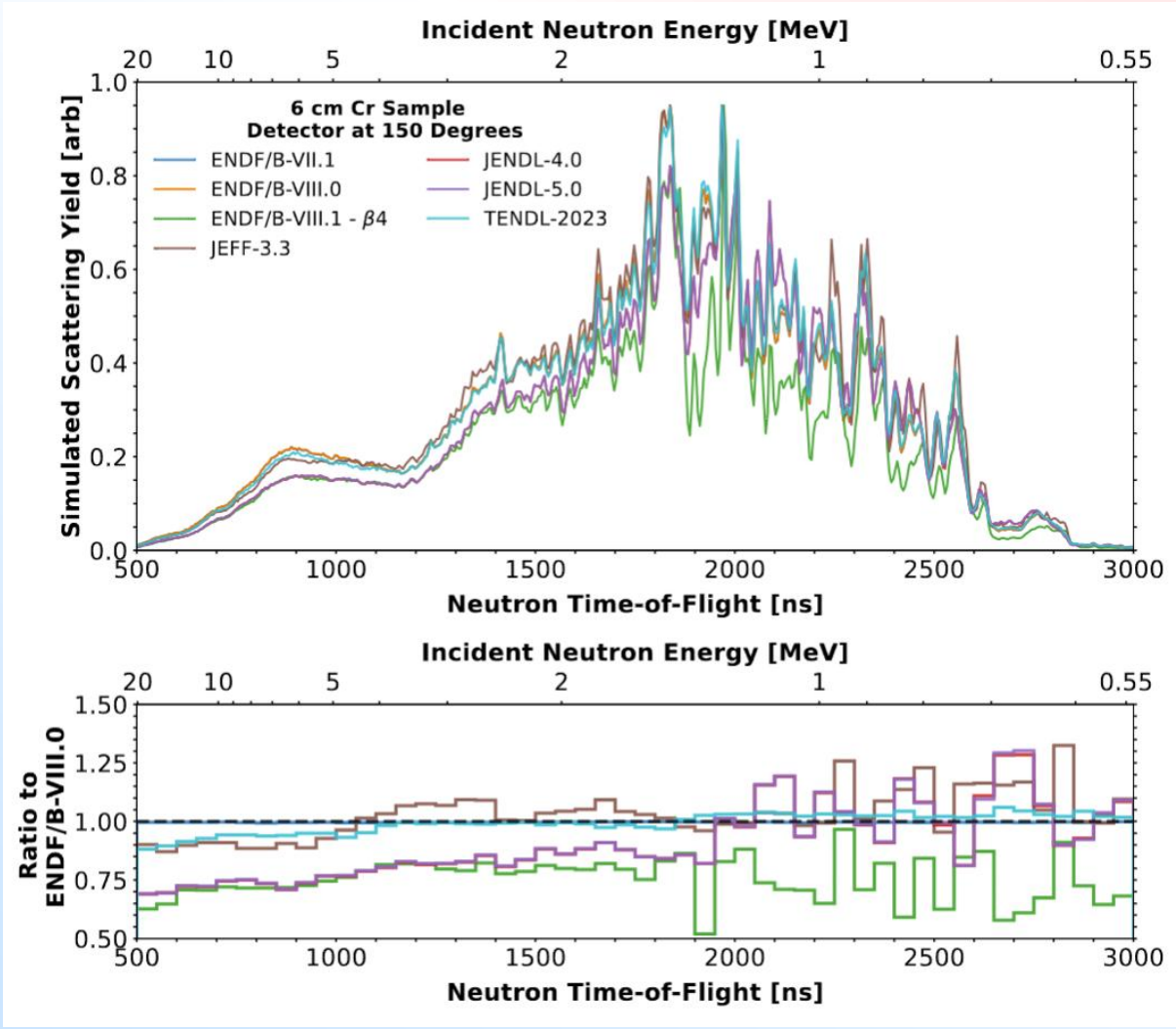
- RPI neutron scattering measurements of ^{nat}Ta, ^{nat}Cu, and Teflon were used as validation platforms for ENDF/B-VIII.1
  - Be experiment also used for validation in earlier beta versions
- RPI scattering experiments can be further improved by performing benchmark analysis
- Conflicts in evaluated nuclear datasets for the following elements can be resolved with RPI neutron scattering measurements:
  - ^{6,7}Li, B, N, O, Na, Mg, Al, Si, Cl, Ti, V, Ni, Mn, Nb, Cr, Y, Ba, Sn, Hf, W



Silicon



Chromium



Neutron-Induced γ-ray Spectra Measurements at the RPI LINAC

K. Keparutis and I. Parker

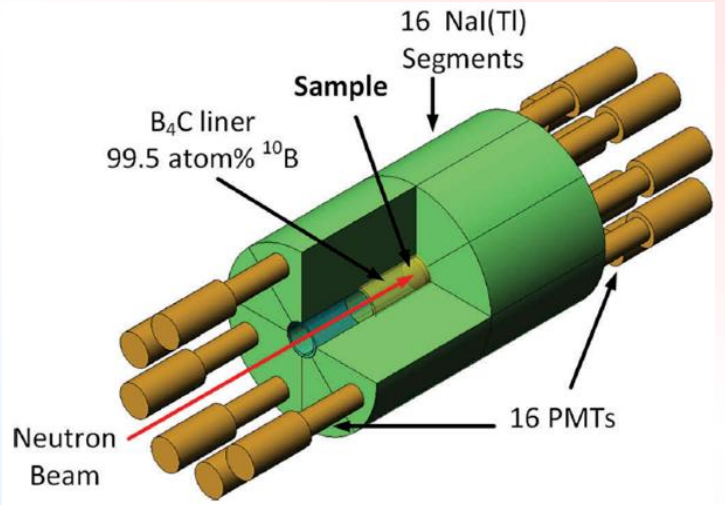
This work was partially performed under appointment to the Rickover Fellowship Program in Nuclear Engineering sponsored by Naval Reactors (NR) Division of the National Nuclear Security Administration (NNSA).

This material is partially based upon work supported by the U.S. Department of Energy, Office of Science, Office of Nuclear Physics, under Award Number DE-SC0024679.

RPI Neutron-Induced γ-ray Spectra Measurements

Measurements coupled with updated simulation methods provide a tool that can be used to **assess the accuracy of γ-ray production data** stored in nuclear data libraries

- Samples measured at RPI with **incident neutron energies of 0.01-100 eV**: ^{56}Fe, ^{55}Mn, ^{59}Co, ^{nat}Ta, ^{nat}U, ^{235}U, ^{nat}Cd, ^{nat}Au, ^{nat}In, and NaCl
- Updated simulation method: **mod-MCNP6.2/DICEBOX**
  - γ-ray cascades generated using DICEBOX and transported through the detector geometry
  - Writes an output file that saves γ-ray energy deposition in detector segments (enables event-by-event analysis including coincidence)



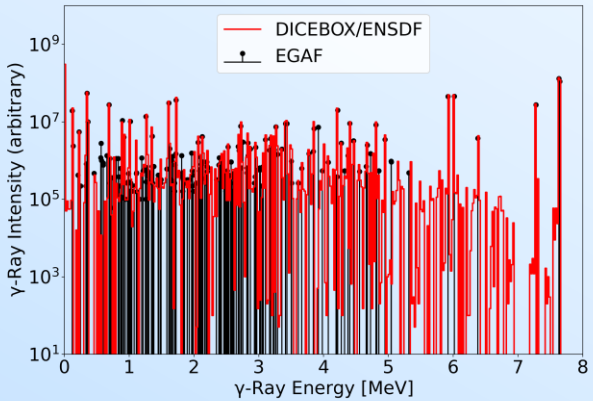
Motivation: improving γ-ray production data

- Increase the accuracy of **reactor and shielding calculations**
- Understand the effects of **γ-ray heating** in nuclear reactors
- Improve **isotope identification** for active neutron interrogation

Awarded DOE Grant (FY23-25) as a follow-on project with the GRIN collaboration:
Development of Benchmark Measurements for Capture Gamma Cascades (DE-SC0024679)

- Benchmark deliverables: measurement data, simulation tools, and benchmark template

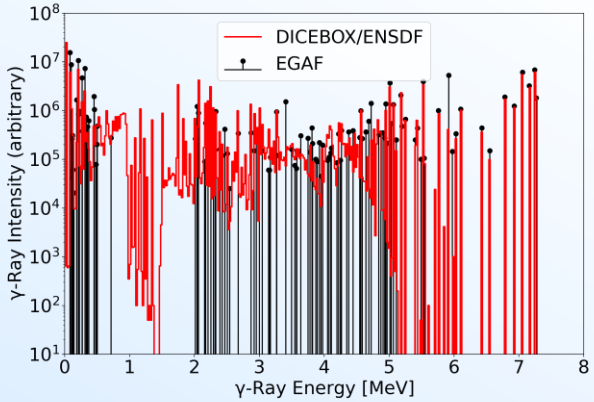
^{56}Fe(n,γ), E_n=TH



^{56}Fe discrete levels from R. B. Firestone et. al., Phys. Rev. C 95, 014328 (2017)

No statistical models

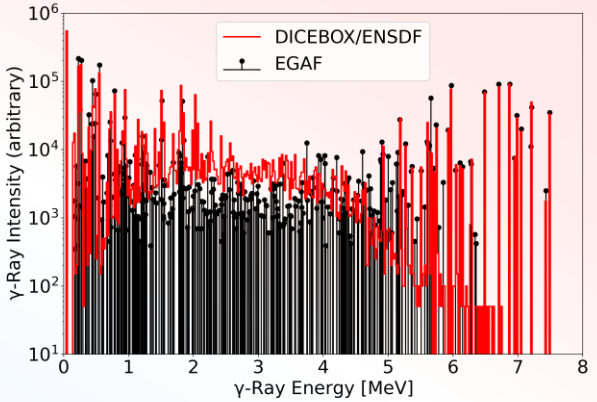
^{55}Mn(n,γ), E_n=TH

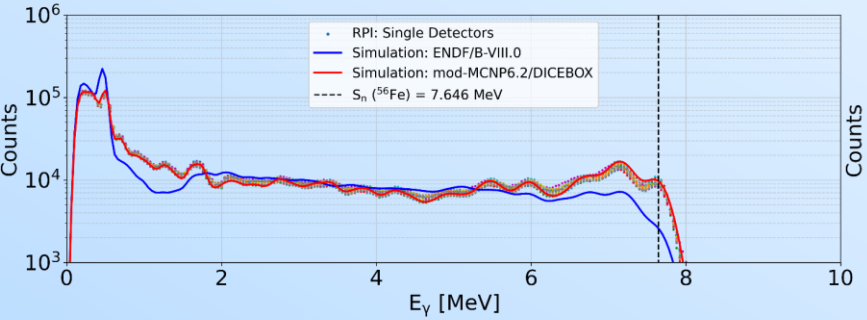


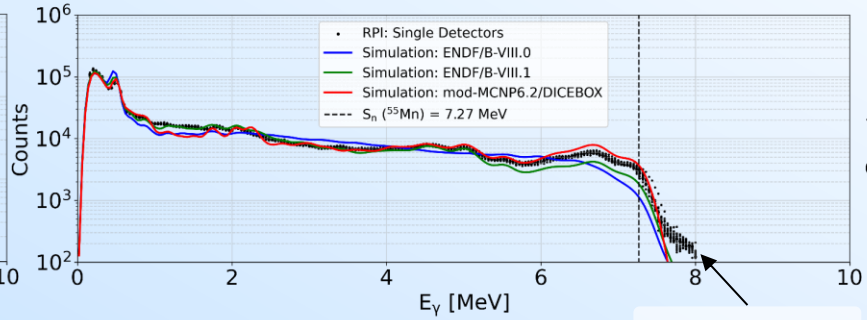
^{55}Mn & ^{59}Co discrete levels from ENSDF (n,γ), E=TH data

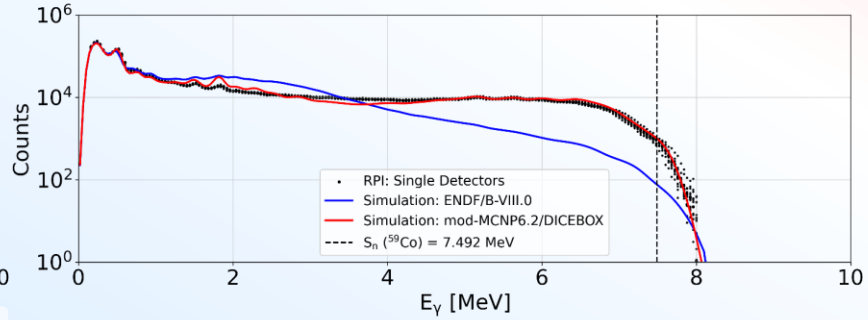
LD model: Back-Shifted Fermi Gas (BSFG)
E1 PSF: Modified General Lorentzian (MGLO)
M1 PSF: Single-Particle (SP)
E2 PSF: Standard Lorentzian (SLO)
Parameters from RIPL-3

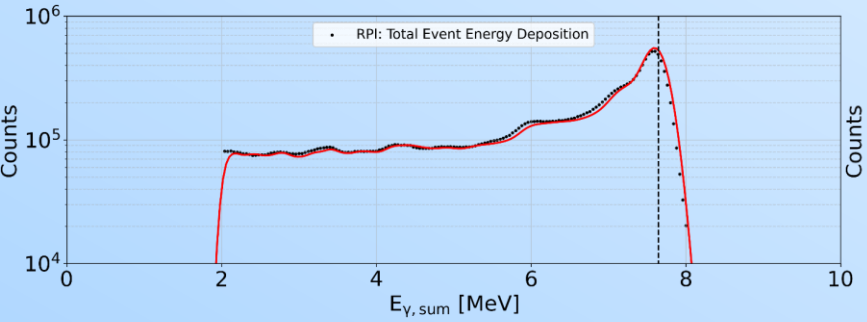
^{59}Co(n,γ), E_n=TH

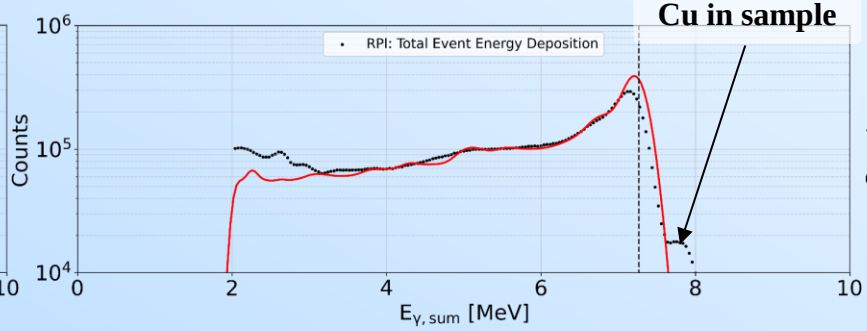


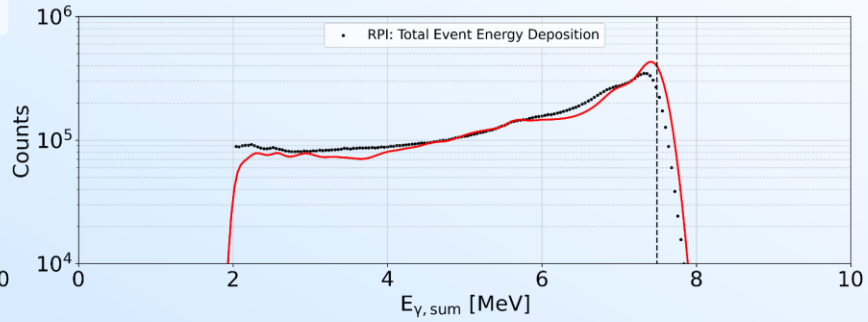








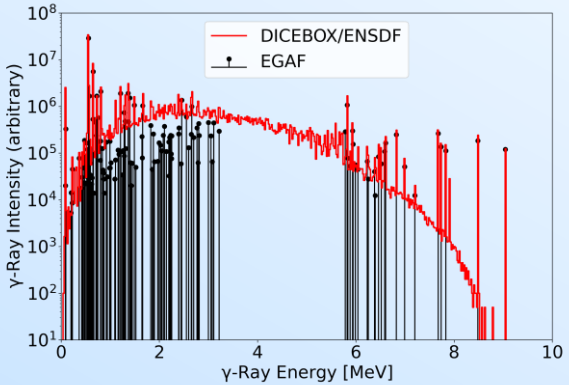




Cu in sample

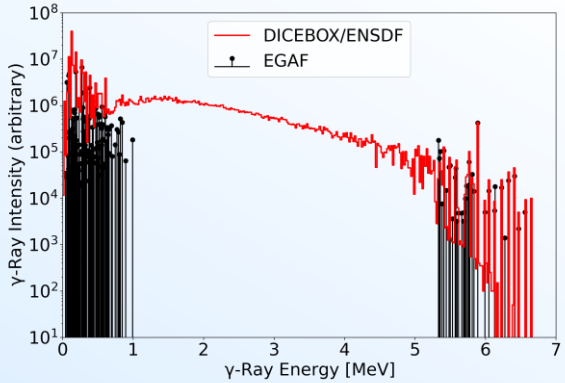
RPI experimental data (black points) are results for $0.01 \leq E_n(eV) \leq 1.0$ and $2.0 \leq E_{\gamma,sum}(MeV) \leq 8.0$

^{113}Cd(n,γ), E_n=TH



^{113}Cd, ^{115}In, & ^{197}Au discrete levels are from ENSDF (n,γ), E=TH data

^{115}In(n,γ), E_n=TH



LD model: Back-Shifted Fermi Gas (BSFG)

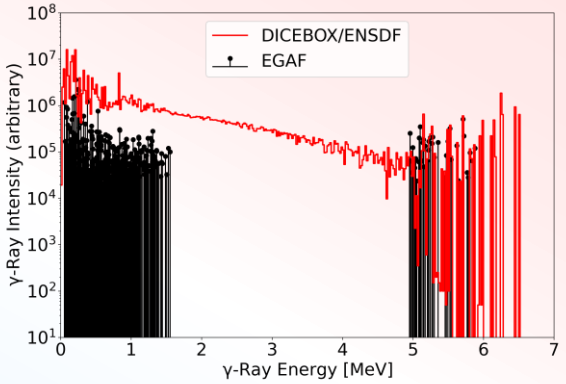
E1 PSF: Standard Lorentzian (SLO)

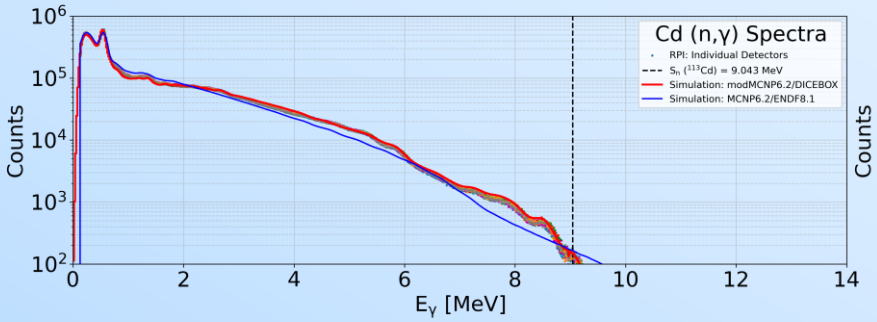
M1 PSF: Single-Particle (SP)

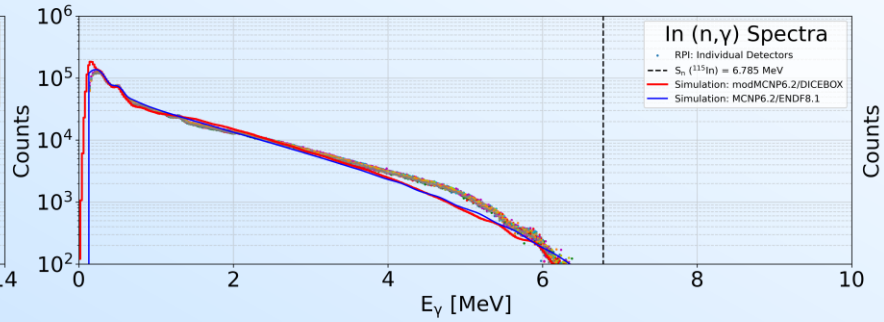
E2 PSF: Standard Lorentzian (SLO)

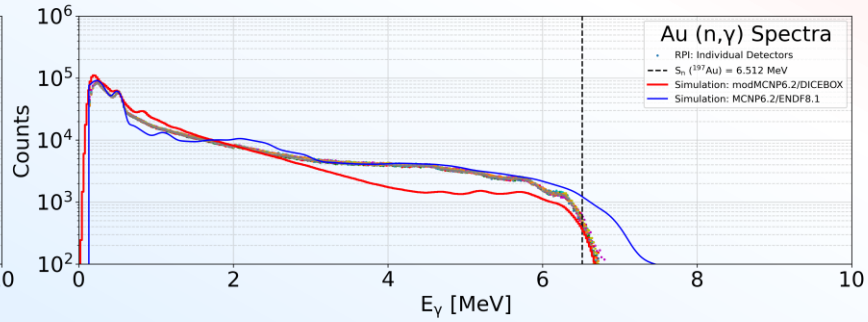
Parameters from RIPL-3

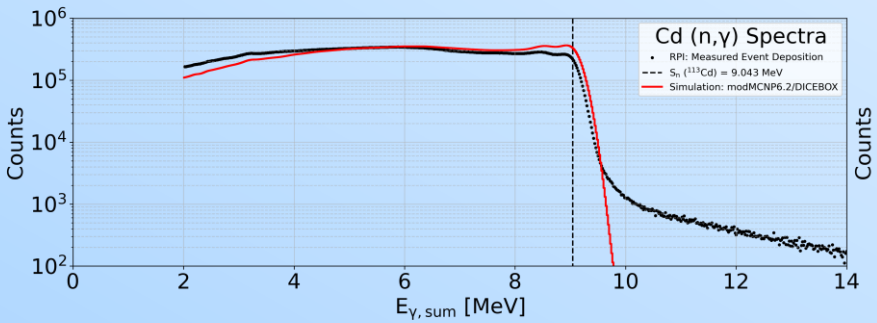
^{197}Au(n,γ), E_n=TH

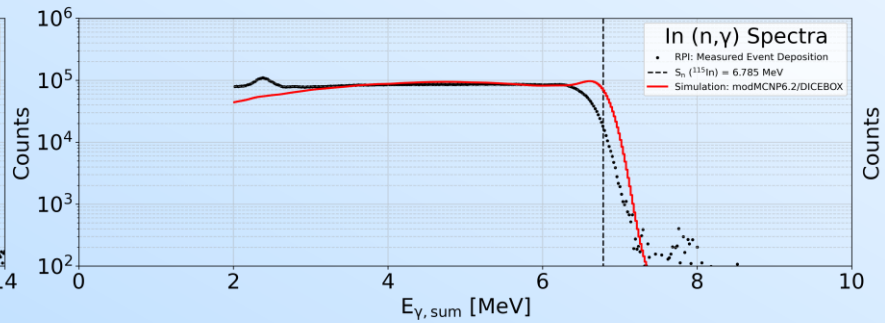


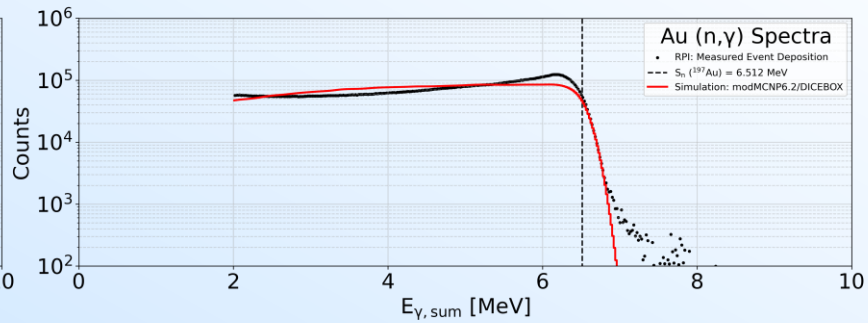








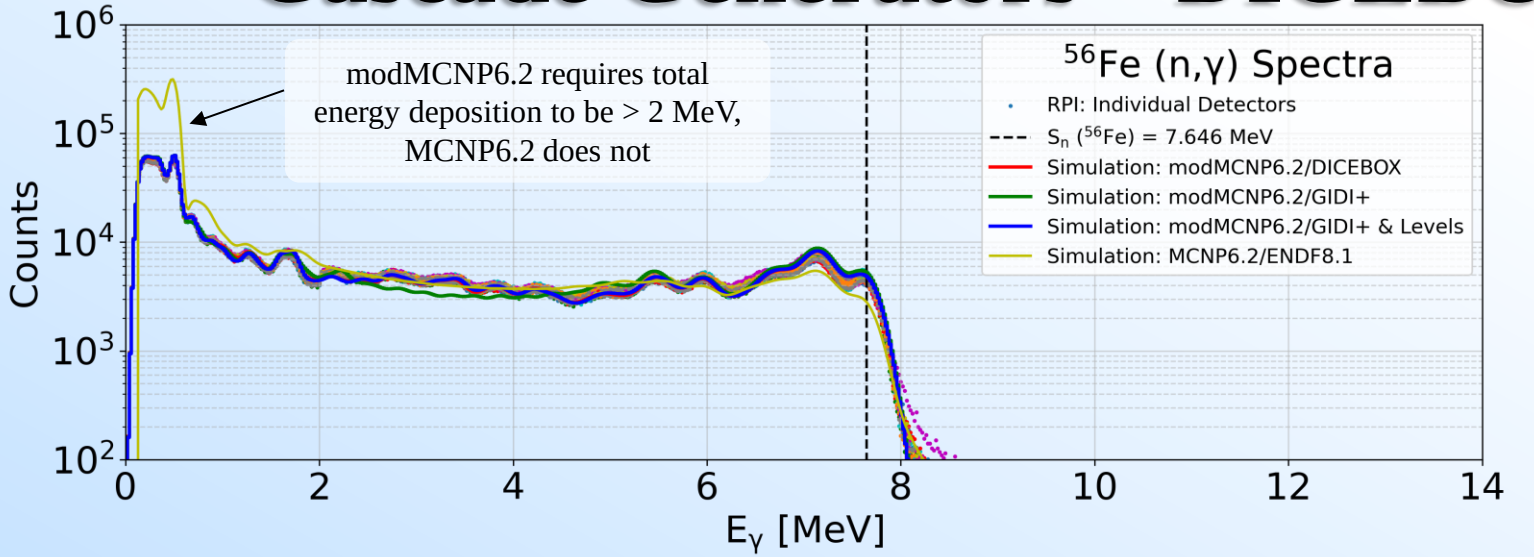


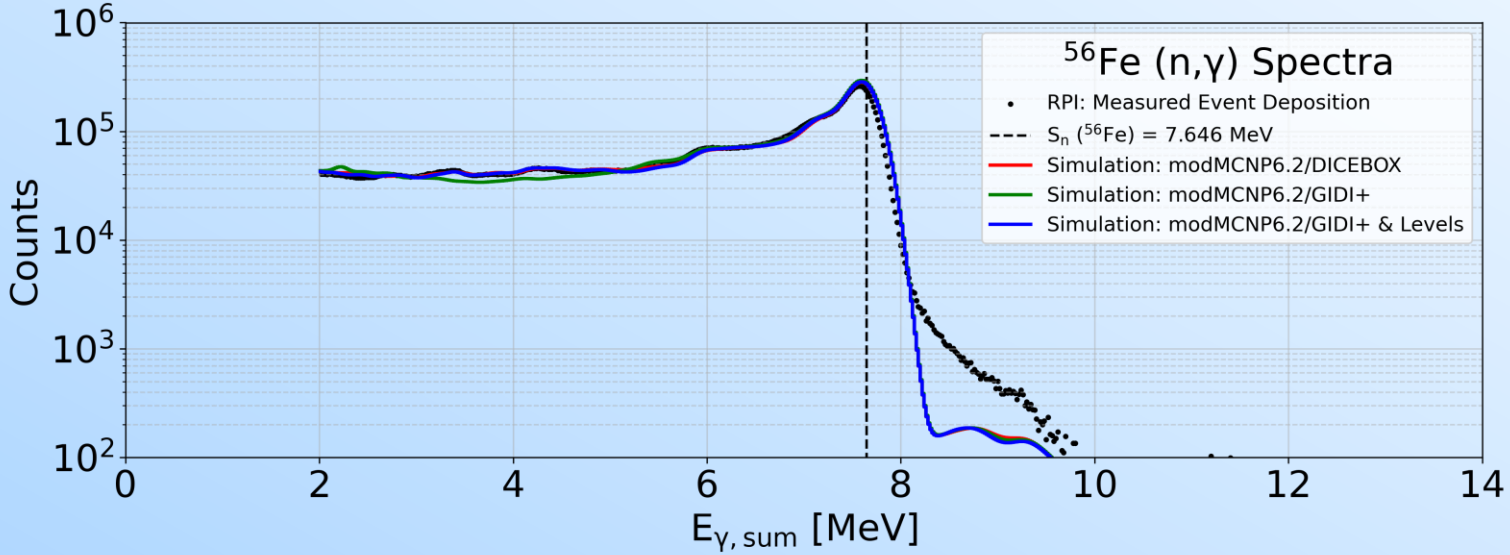


RPI experimental data (black points) are results for $0.01 \leq E_n(eV) \leq 1.0$ and $2.0 \leq E_{\gamma,sum}(MeV) \leq 20.0$

Work of Ian Parker & Emanuel Chimanski

Cascade Generators – DICEBOX/GIDI+





1 week visit in September to work with Emanuel/GRIN to compare DICEBOX and GIDI+

GIDI+ uses ENSDF evaluated discrete levels
Primary E_γ = [7.646 – 5.921 MeV]

GIDI+ & Levels uses similar discrete levels to DICEBOX
(R. B. Firestone et. al., Phys. Rev. C 95, 014328 (2017))

Validation complete of cascade generators; both perform equally well when inputs have accurate discrete levels

Summary

- RPI is working on several measurements and evaluations of structural materials
- ^{54}Fe measurements and evaluation.
  - RRR measurements completed, evaluation nearly complete.
- ^{90}Zr evaluation is progress
  - RRR energy was extended to first excited state at1764 keV.
  - Working on incorporating capture measurements.
  - Preliminary fast calculation.
- Quasi differential scattering
  - Ta and TEFLON measurements and simulation completed
  - Ta - comparison with stimulation shows that ENDF/B-VIII.1 is an improvement over ENDF/B-VIII.0
  - F - comparison with simulation shows that ENDF/B-VIII.1 is an improvement but there is room for additional improvements that might require fitting resonances.
  - Looking at other materials including fusion related.
- RPI neutron-induced γ-ray spectra measurements and simulations
  - Several measurements performed.
  - Methodology to compare the experiments to nuclear structure data was developed.
  - Observed differences between experiments and simulations indicate where capture gamma evaluations can be improved.