Neutron Induced Neutron Emission in ^{235}U and ^{239}Pu

KUMAR SUNIL MOHINDROO
RENSSELAER POLYTECHNIC INSTITUTE

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Motivation - Background

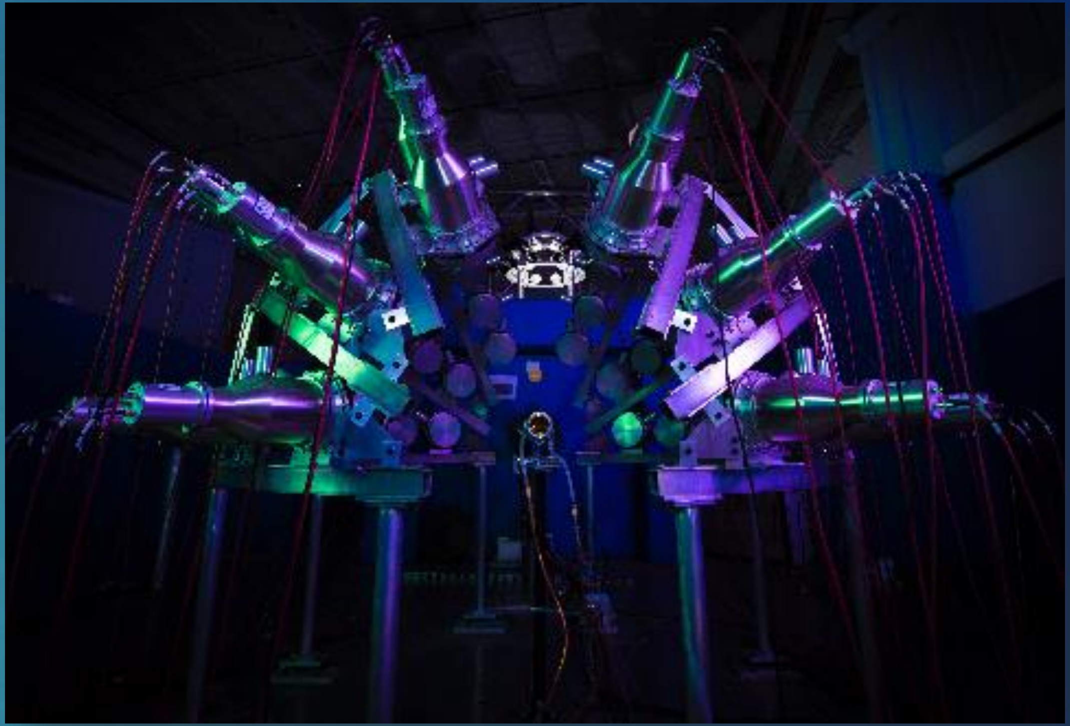
- ^{235}U and ^{239}Pu are important isotopes with significant impact on the nuclear community.
  - supports calculations for nuclear reactor design and operation, criticality safety, and nonproliferation
  - In 2014 CIELO, an international collaboration and respected authority called for the investigation of isotopes including ^{235}U and ^{239}Pu .
- RPI scattering methodology was adapted to supplement this investigation of ^{235}U and ^{239}Pu in the range of 1 to 20 MeV at LANSCE WNR

$$\frac{1}{v}\frac{\delta\varphi}{\delta t} + \widehat{\Omega}\cdot\nabla\varphi + \Sigma_t(r,E)\varphi(r,E,\widehat{\Omega},t)$$

$$= \int_{4\pi} d\widehat{\Omega}' \underbrace{\int_0^{\infty} dE' \Sigma_s(E' \to E, \widehat{\Omega}' \to \widehat{\Omega})}_{Scattering\ Kernel} \varphi(r,E',\widehat{\Omega}',t)$$

$$+ \frac{\chi(E)}{4\pi} \int_{4\pi} d\widehat{\Omega}' \int_0^{\infty} dE' \nu(E')\Sigma_f(E')\varphi(r,E',\widehat{\Omega}',t)$$

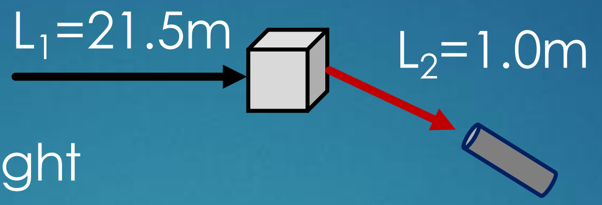
Boltzmann Transport Equation



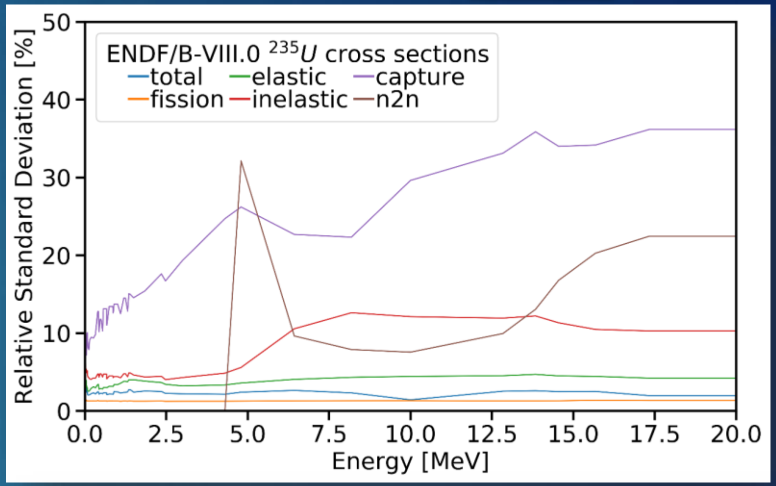
Chi-Nu proton recoil detector assembly with parallel plate avalanche chamber centered in the array

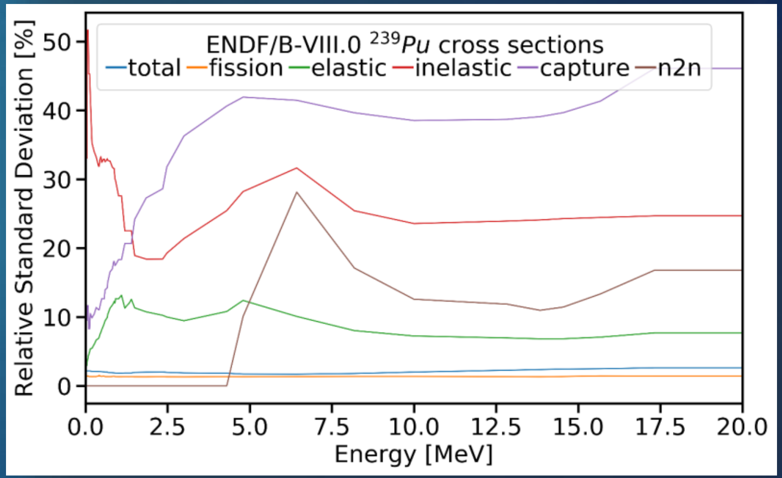
Image source: https://lansce.lanl.gov/facilities/wnr/_assets/image1.jpeg

Theory – Double time of flight experiments



- Energy is inferred through time of flight
- Neutron yield equation shows relative contribution of different reactions to the emission spectrum
  - Low uncertainties on total and fission cross sections serve as a constraint
  - Angular distribution is conserved as a constraint
  - Primarily constraining inelastic, capture, and n2n contributions





Time of flight equation

$$E(t) = m_0 c^2 \left(\sqrt{1 - \frac{(L_1 + L_2)^2}{c^2} \frac{1}{t^2}} - 1 \right)$$

$$\int_{-1}^{1} f(\mu) d\mu = 1$$

Angular distribution conservation

First collision approx. provides insight

$$Y(E) = \underbrace{\phi(E)}_{neutron\,flux} \cdot \underbrace{\left(1 - e^{-\Sigma_t(E)x}\right)}_{probability\ to\ interact} \cdot \underbrace{\frac{\sigma_s(E) + \bar{\nu}\sigma_f(E) + \cdots}{\sigma_t}}_{neutron\ production} \cdot \underbrace{\frac{f(E,\mu)}{2\pi}}_{angular\ distribution} \cdot \underbrace{\eta(E')}_{detection\ efficiency} + \underbrace{Y_{ms}(E)}_{multiple\ scattering}$$

Experimental Setup at WNR LANL

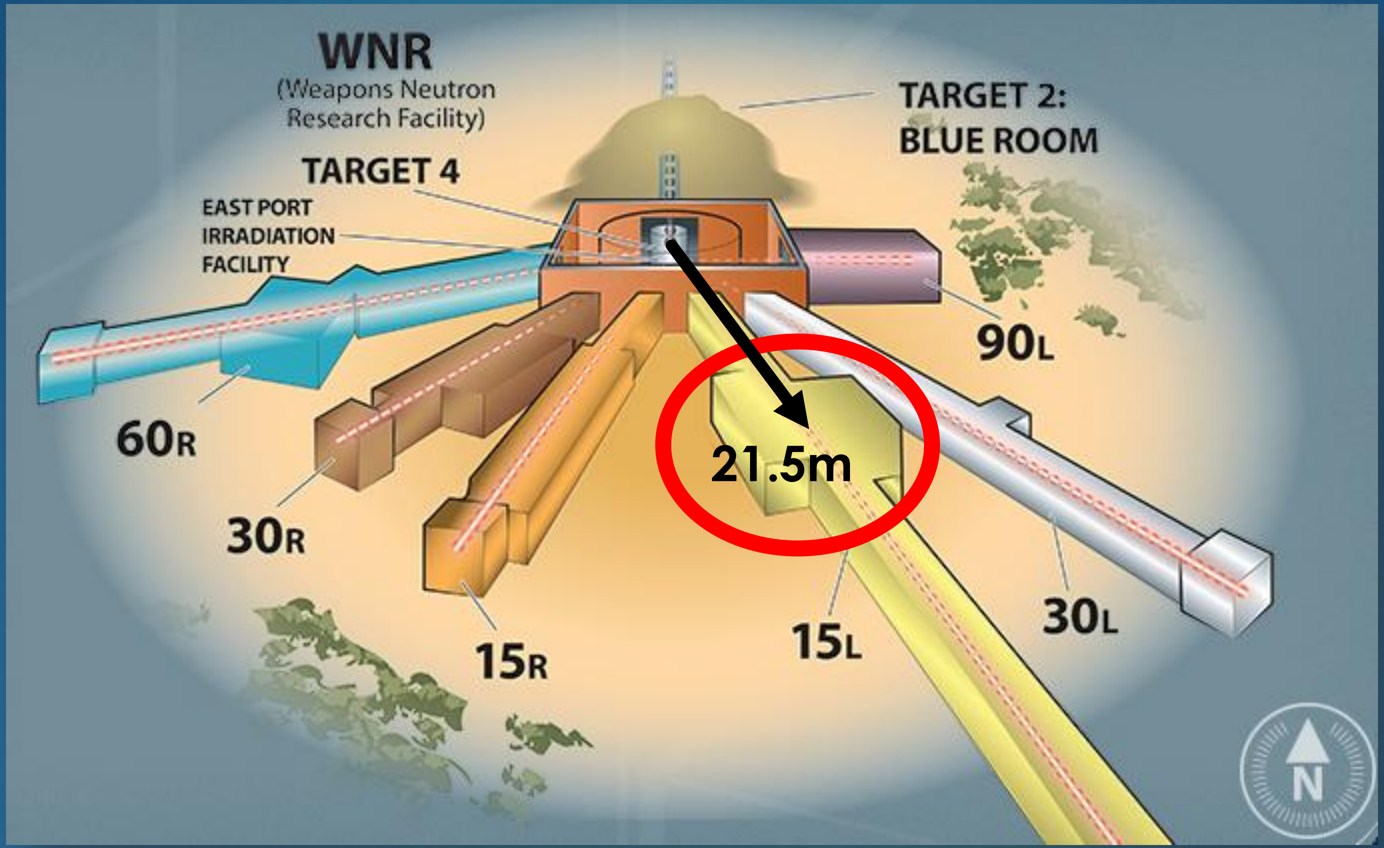
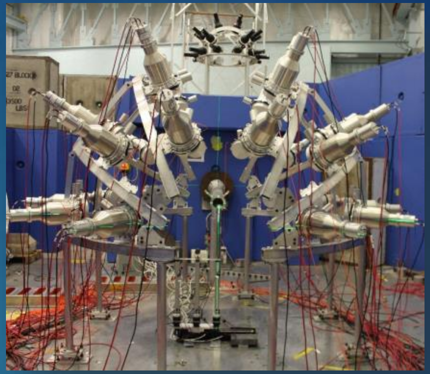


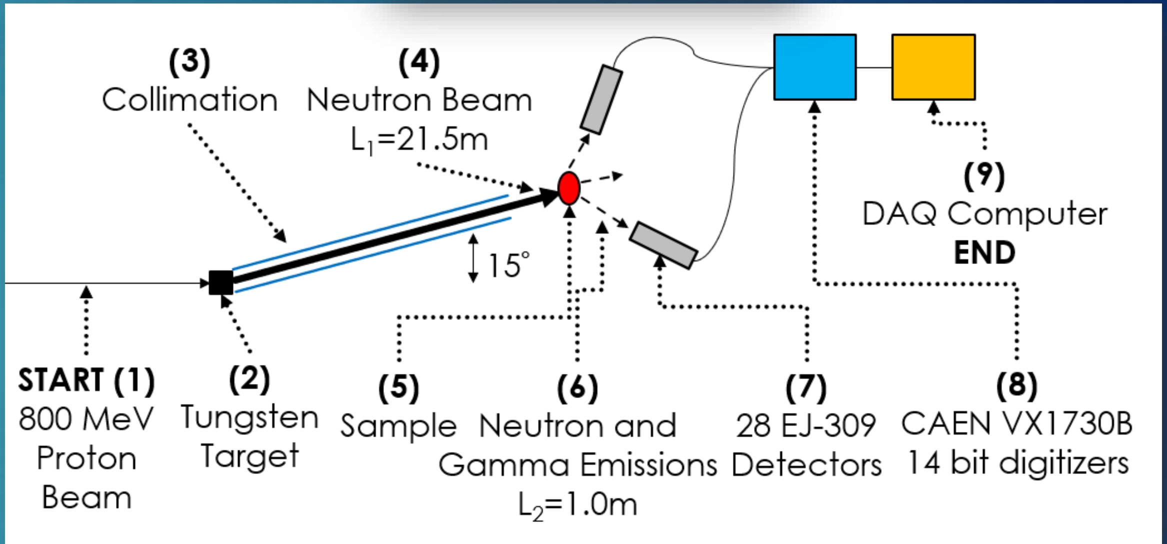
Image source: https://lansce.lanl.gov/facilities/wnr/_assets/images/WNR-flight-paths-Target-4.png

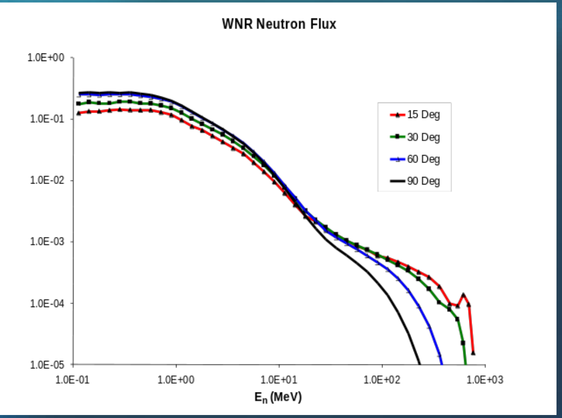
Experiment was performed at Los Alamos National Laboratory using the LANSCE proton accelerator at WNR.
Sample Interrogation was performed on flightpath 15-L at the CHI-NU flight station.

Experimental Setup – Overview



- High energy protons induce spallation on a Tungsten target
- Neutrons up to 800 MeV are emitted isotropically and collimated into a beam
- A sample is placed in the neutron beam and the emissions are recorded by EJ-309 liquid scintillation detectors
- The detection signal is routed through a digitizer and the timestamp, head, tail, and total integral information, and detector number are written to disk.
- Four CAEN VX1730B 14-bit digitizers
  - 500 MHz sampling rate





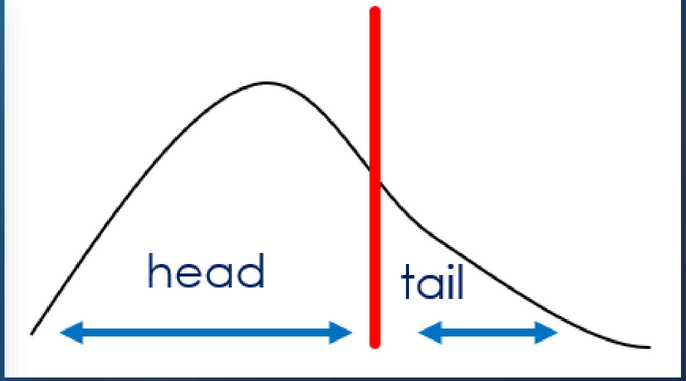


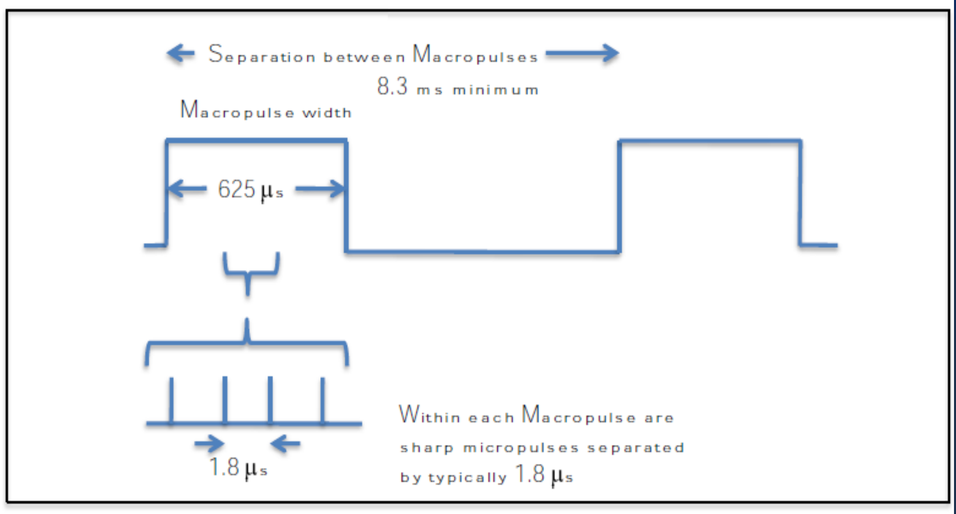
Image source: https://lansce.lanl.gov/facilities/wnr/_assets/images/wnr-neutron-flux.png

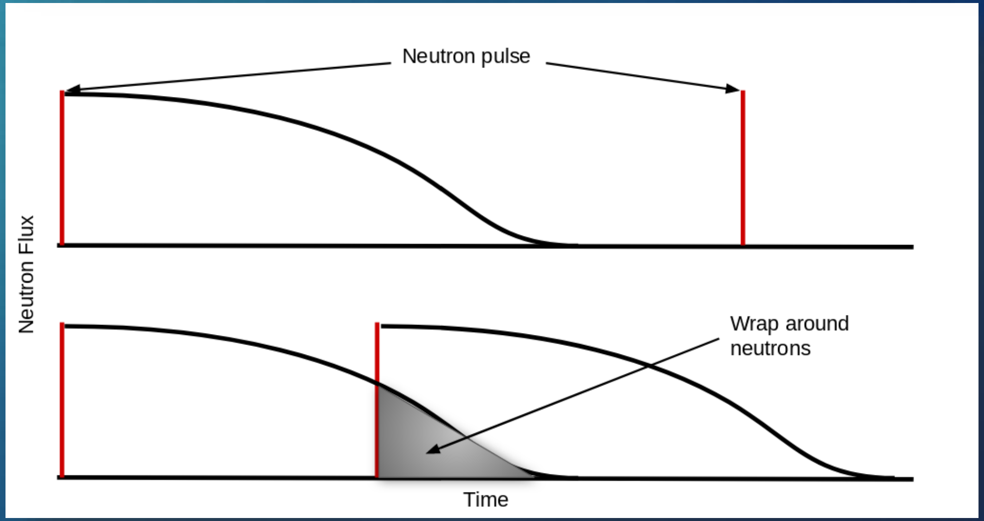
Experimental Setup –Beam Timing

- The accelerator has a firing cycle of up to 120 Hz which lasts for 625 μs. This firing cycle is called a macropulse.

- Within each macropulse there are 347 micro pulses. The micropulses are the actual firing impulse of the accelerator and they are separated by 1.8 μs. The width of the micropulses is dependent on tuning parameters but is typically less than 100 ps at the exit of the LINAC.

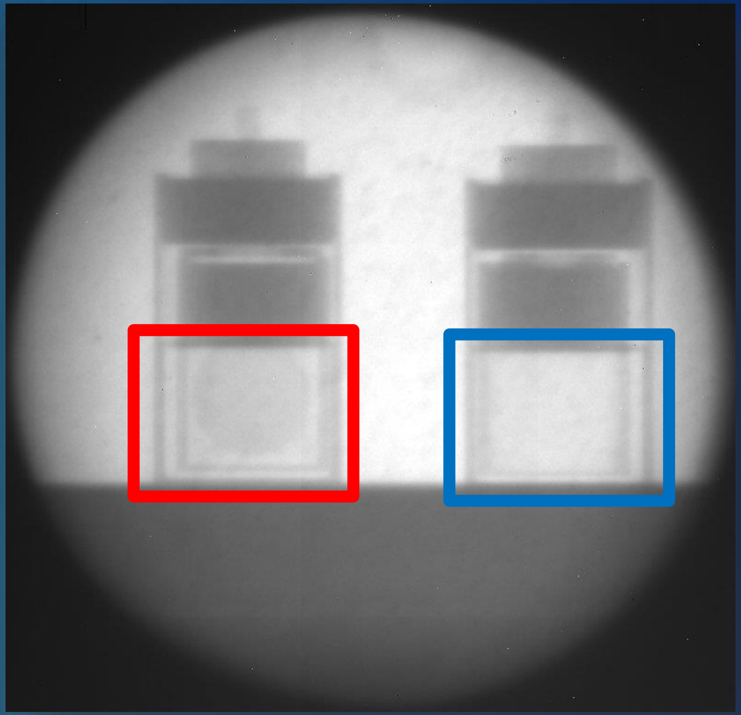
- At our flight path distance of $L=L_1+L_2=22.5m$ the wrap around effect causes contamination in the low energy region and prevents us from measuring below 0.81 MeV.





Pu-Ga alloy sample

- 24.61g sample encapsulated in stainless steel assumed to be 304
- 0.47g sample in similar encapsulation used as blank
- Uncertainties assumed to be
  - ± 0.01g
  - ± 0.001cm

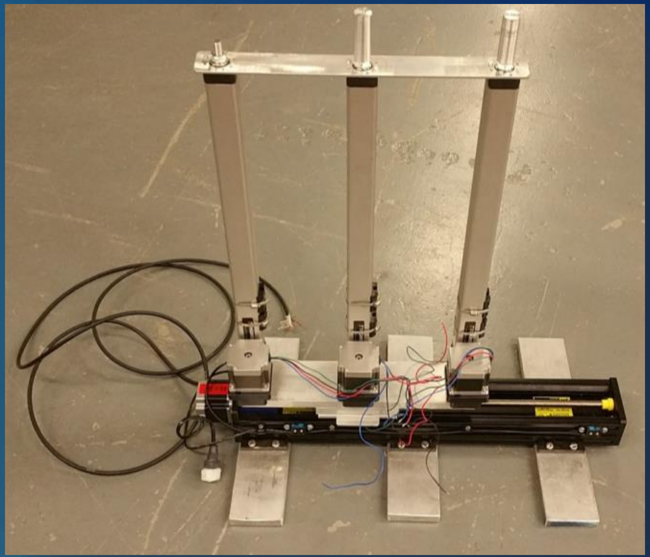


Isotope	Atomic percentage
^{238}Pu	0.0110
^{239}Pu	93.9273
^{240}Pu	5.8847
^{241}Pu	0.1384
^{242}Pu	0.0388
^{244}Pu	0.0000

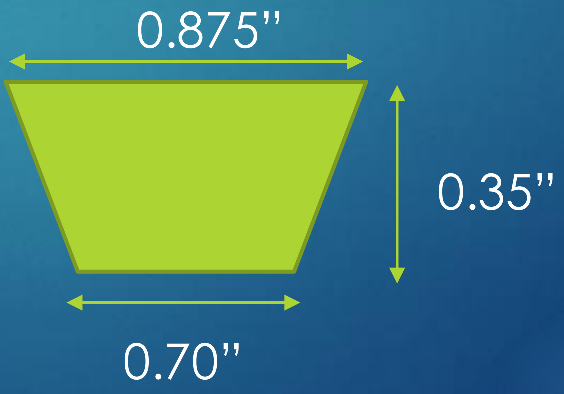
Sample	Net wt. (g)	Diameter (cm)	Nominal thickness (cm)	n(Pu) (atoms/b)	n(Ga) (atoms/b)
Sample	24.61	2.550	0.307	1.195×10^{-2}	4.41×10^{-4}
Blank	0.47	2.543	0.006	2.296×10^{-4}	8.44×10^{-6}

HEU sample

- Encapsulated in ~0.9mil thick aluminum foil (determined through measured mass and area using typical aluminum density 2.71g/cc)
- Sealed with Kapton tape
- 49.5g 93% ^{235}U
- Truncated cone geometry
- 17.647 g/cc calculated based on fixed mass and dimensions
- Uncertainties assumed to be
  - ± 0.01g
  - ± 0.01"



Sample Changer
10-minute run cycles



Methodology - Overview

- Compare detailed MCNP simulations to experiment with different evaluations for U-235 and Pu-239
  - Both sample and "open" were measured and simulated
  - We are comparing the net experimental spectrum to the net simulated spectrum
- Normalize net simulations (ENDF/B-VIII.0) to net measured carbon scattering

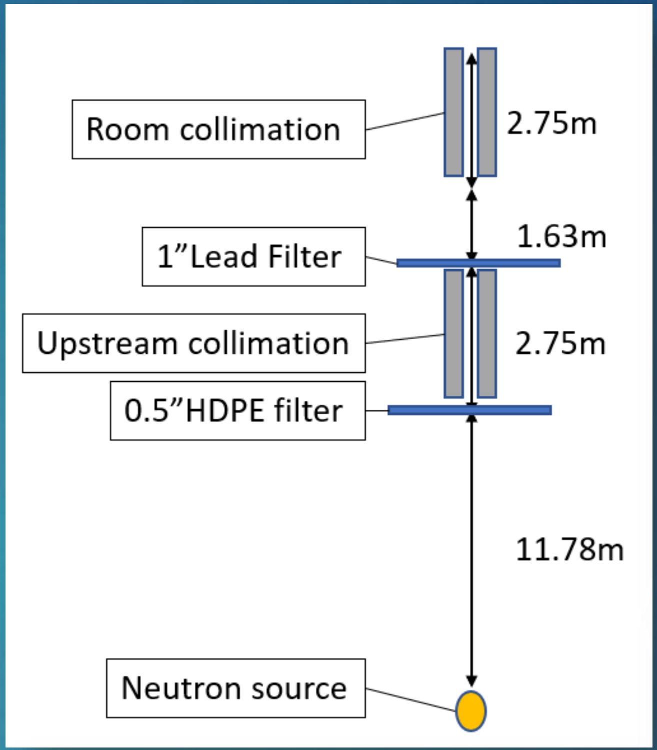
$$C_{net} = \frac{C_{sample}}{N_{Sample}} - \frac{C_{open}}{N_{open}}$$

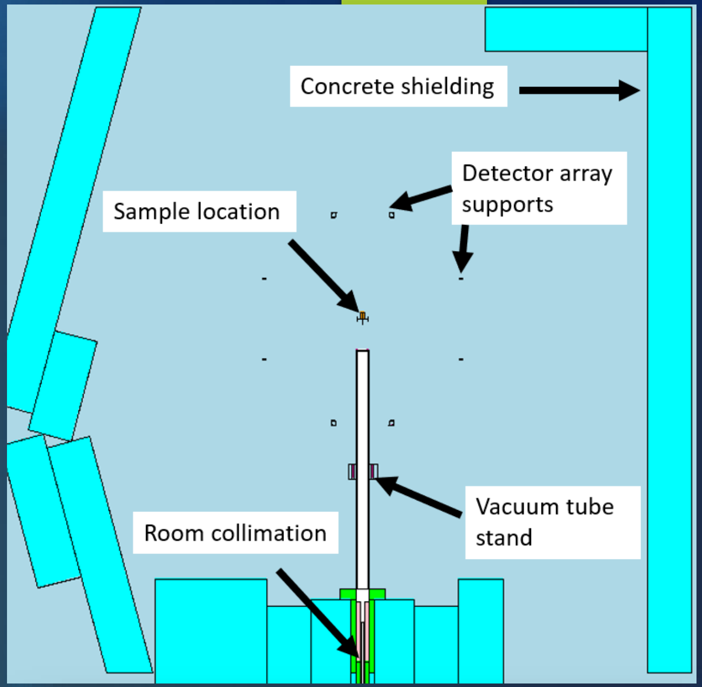
$$\frac{\sigma_{net}}{C_{net}} = \sqrt{\left(\frac{\sigma_{C_{sample}}}{N_{Sample}}\right)^2 + \left(\frac{\sigma_{C_{open}}}{N_{open}}\right)^2}$$

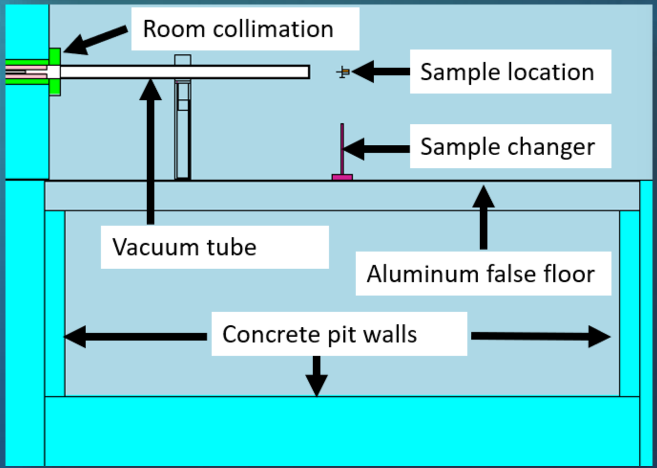
$$\sigma_{total} = \sigma_{stat} + \sigma_{systematic}$$

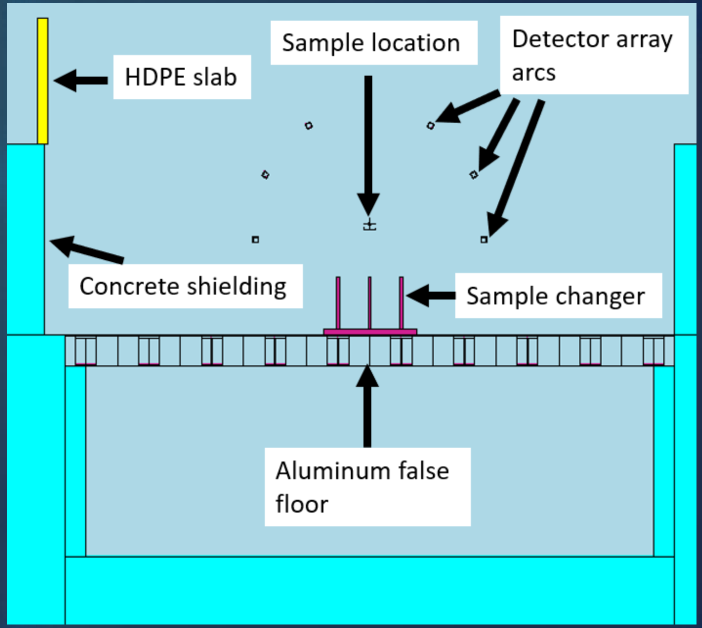
Geometry and room return

- Simulation includes:
  - Room (walls + floor)
  - Other structures near the detectors
  - Beam filters
- The contribution from the room ranges from .59% at 1 MeV to a maximum of 9.93% at 5.67 MeV and reduces to 2.71% at 20 MeV
- Room return is accounted for in the simulations



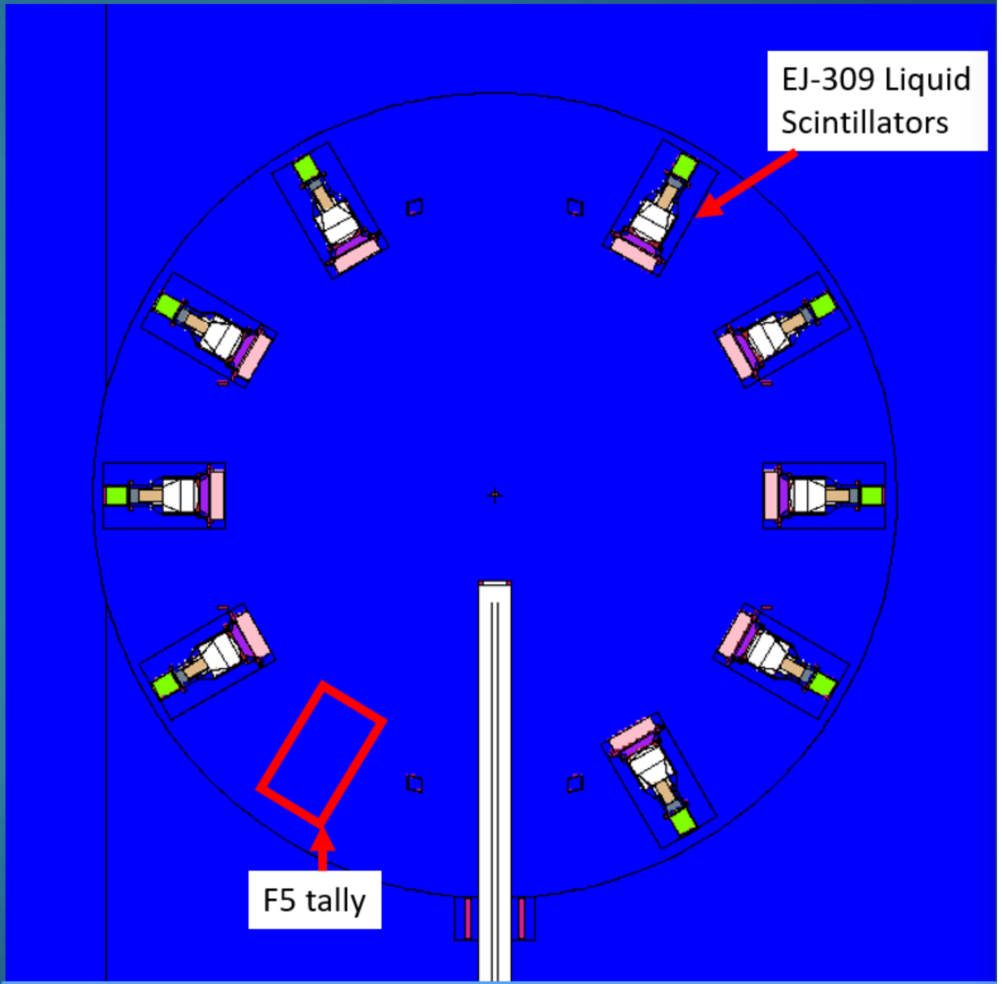






Cross Talk

- Scattering from all detectors to one detector was evaluated
- Overall contribution is about 1.5 %
- Was not corrected for and is included in the systematic uncertainty

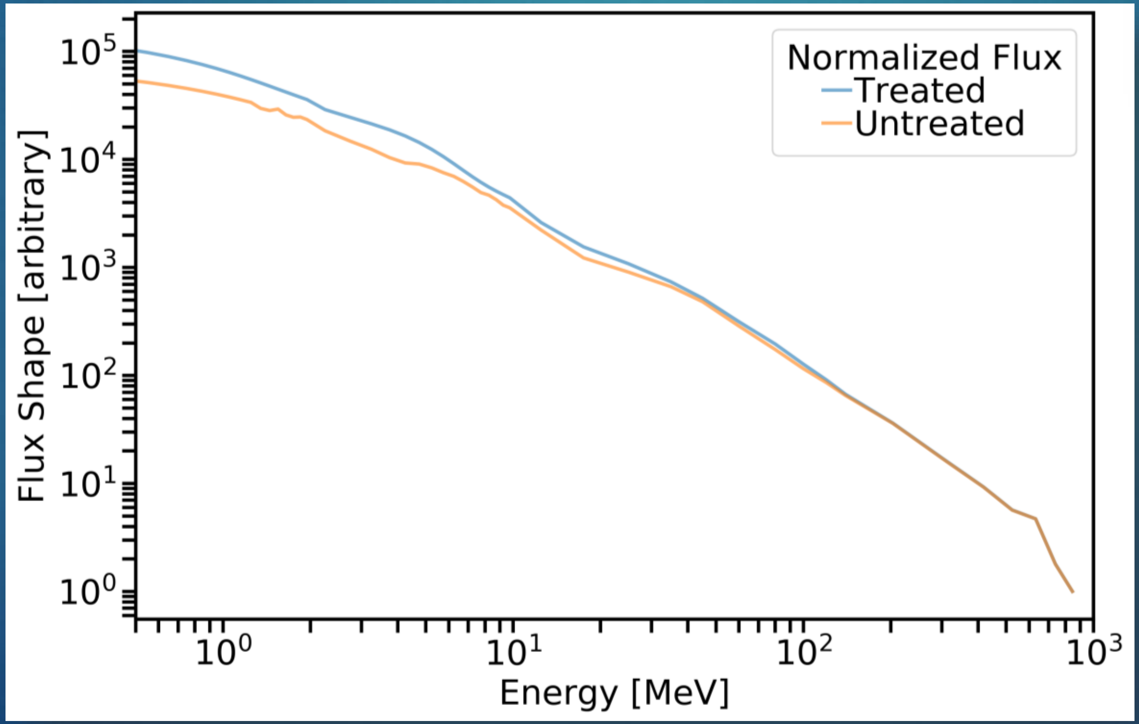


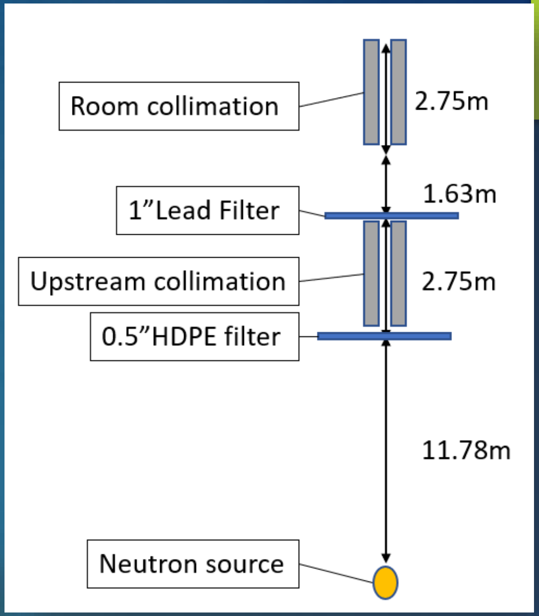
The effect of crosstalk is incorporated into systematic uncertainty

det	angle	% integral increase
0	150	1.35
2	120	1.46
4	90	1.45
6	60	1.41
8	30	0.60
10	135	1.63
12	105	1.35
14	75	1.26
16	45	1.53
18	150	1.31
20	120	1.23
22	90	1.23
24	60	1.58
26	30	0.89
27	150	1.16
29	120	0.98
31	90	1.31
33	60	1.31
35	30	0.68
37	135	1.42
39	105	1.23
41	75	1.21
43	45	1.50
45	150	1.13
47	120	1.25
49	90	1.09
51	60	1.33
53	30	0.85

Neutron Flux shape

- Measured using a U-235 fission chamber
- Dips are due to material in the neutron beam and were removed by analytical calculation to obtain a smooth flux shape.

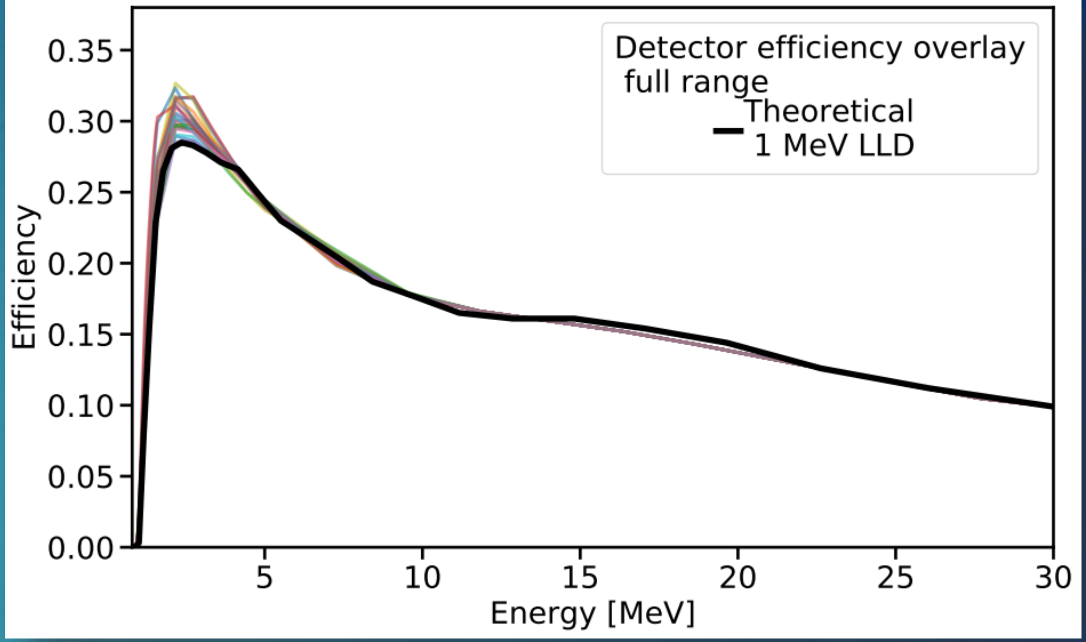


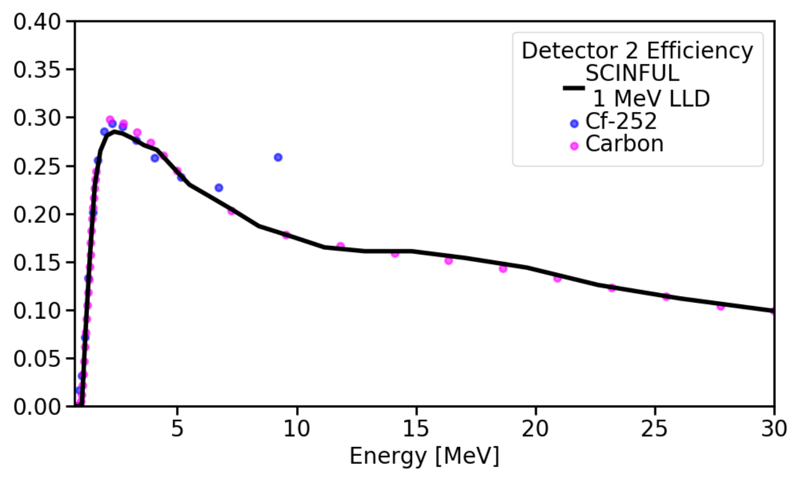


Detector Efficiency

- Measured using Cf-252 as a reference for lower energy range
- Obtained from a 7 cm thick carbon sample by comparing measurement and simulation
- The efficiency was smoothed to remove carbon resonances
- Compared to SCINFUL calculation for verification.

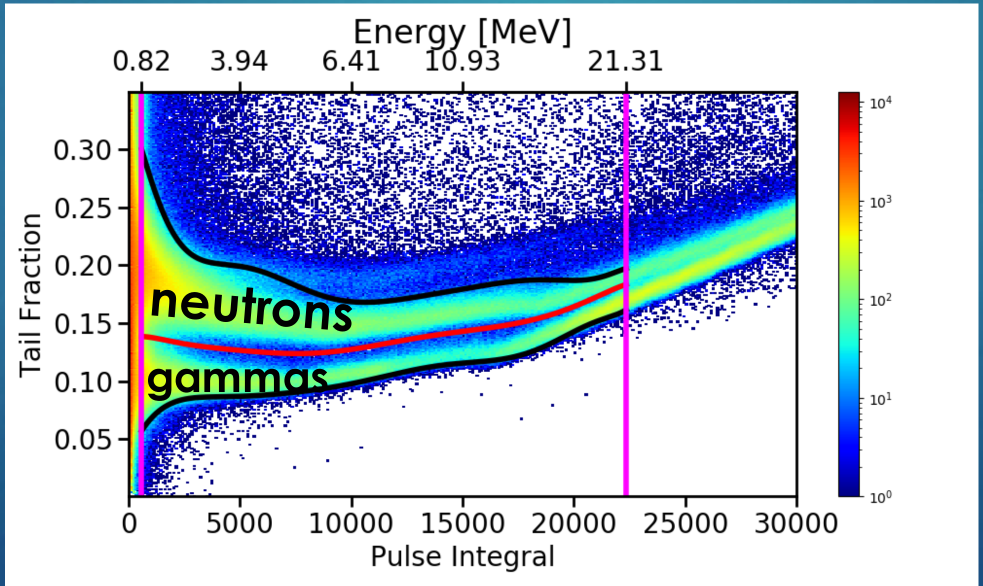
- **Note: different detectors have different efficiencies in energy range of 1-4 MeV**





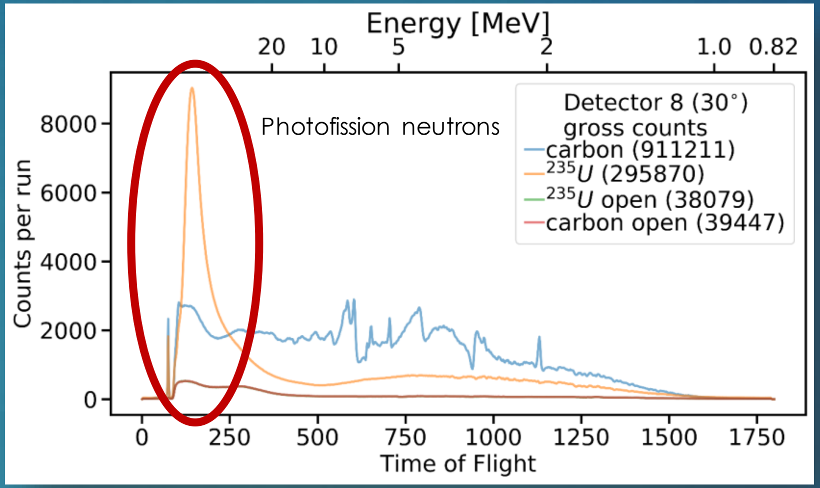
Data Analysis: Pulse Shape Discrimination

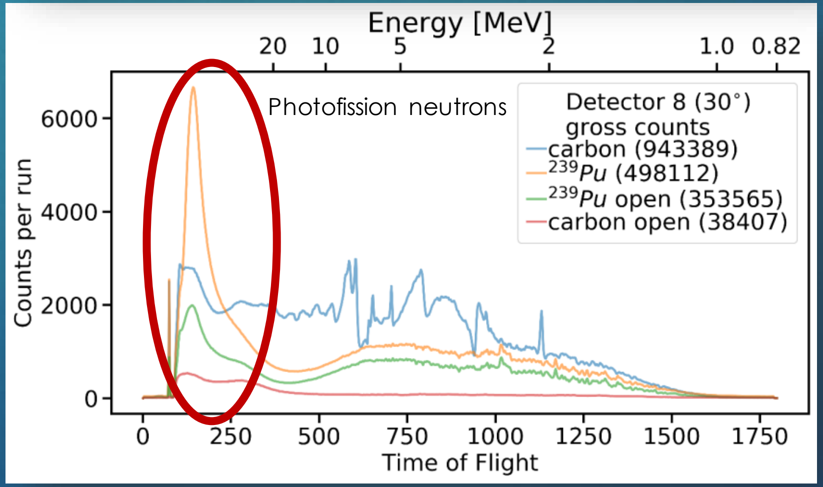
- Pulse shape analysis was used to reject detected gammas
- Good separation between gammas and neutrons was observed from 1-20 MeV

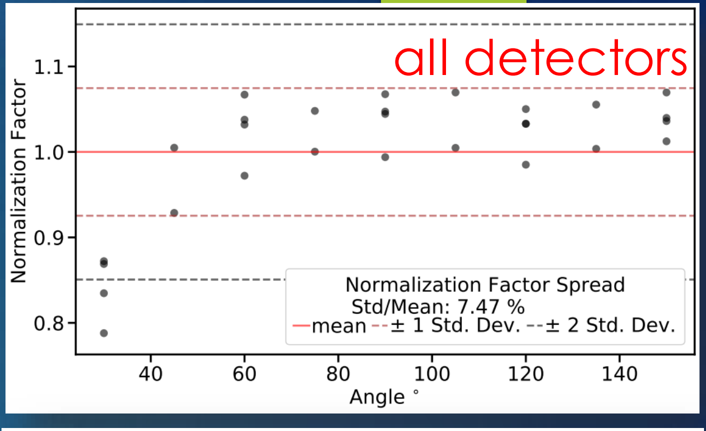


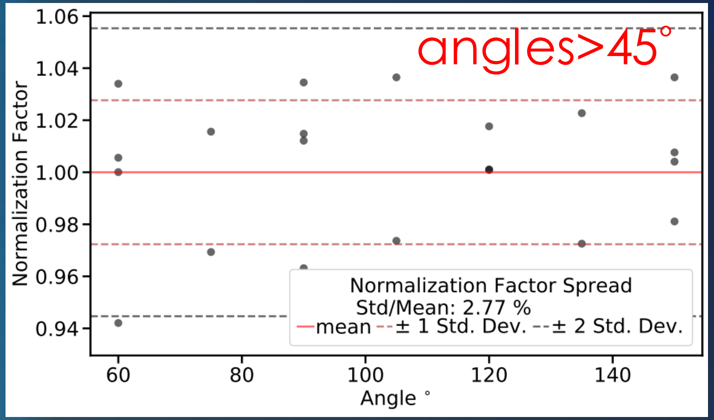
Gross Signal and statistical uncertainty

- ^{235}U Gross Signal
  - ^{235}U encapsulation does not separate from open beam
- ^{239}Pu Gross Signal
  - The ^{239}Pu encapsulation is a large portion of its signal
- Large signal from photofission seen in ^{235}U and ^{239}Pu
- Normalization performed only for carbon reference in range 20 MeV to 1.2 MeV
- Ideally this ratio is constant across all detectors, and deviation represents our systematic uncertainty.

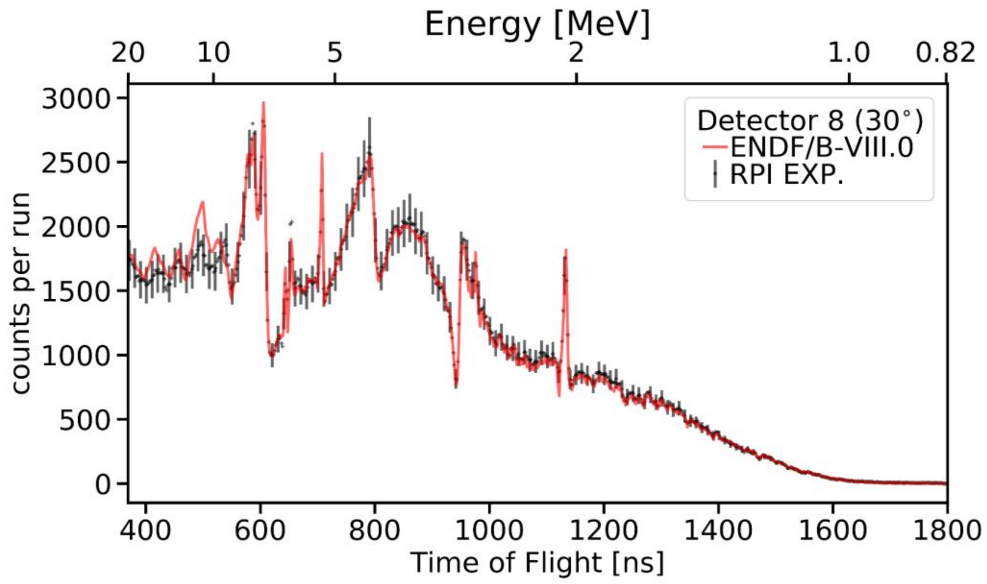


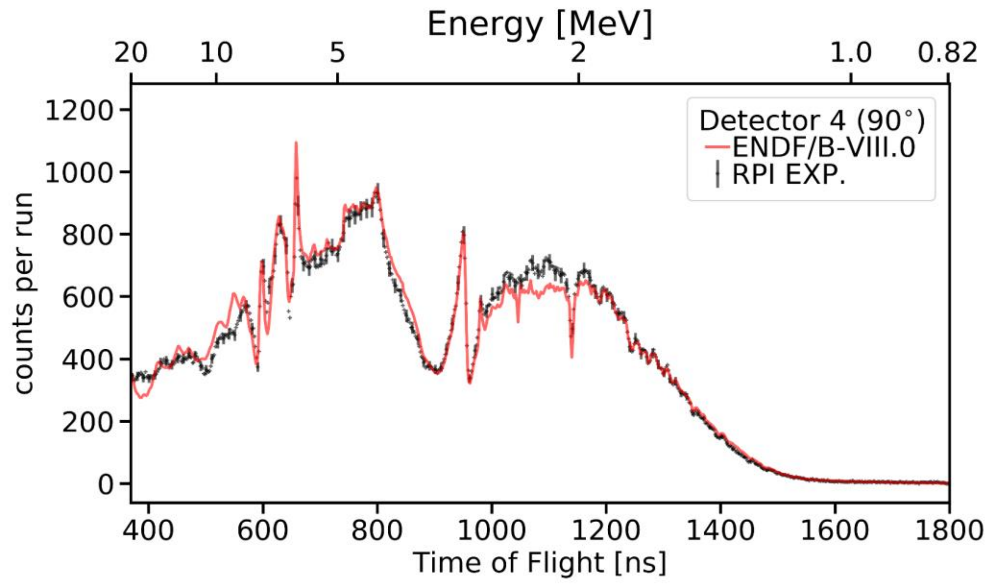




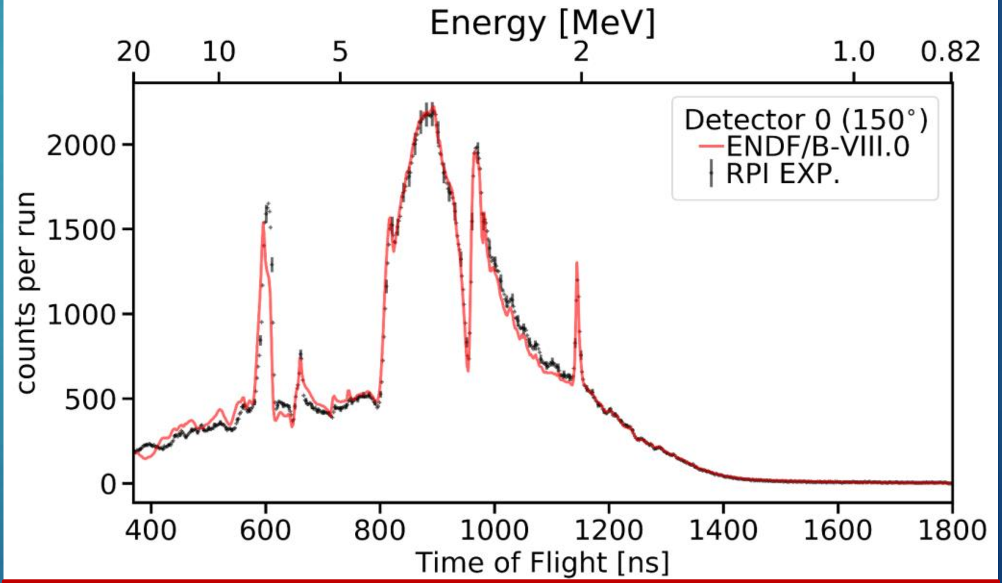


Carbon



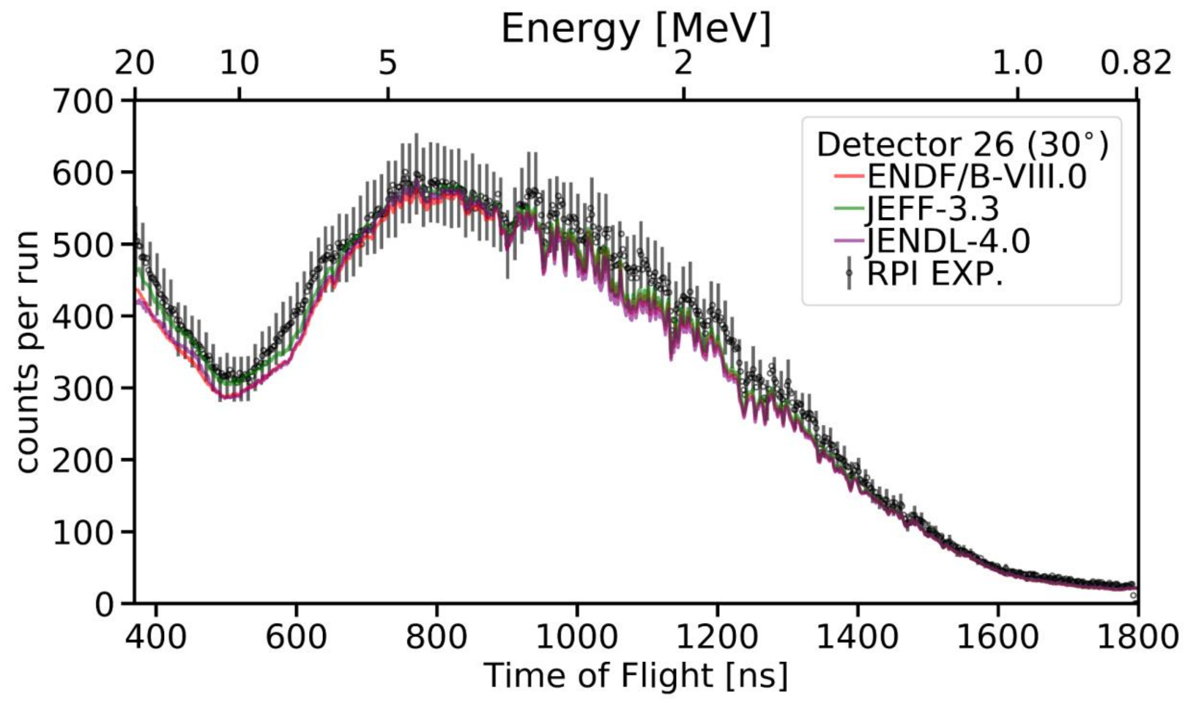


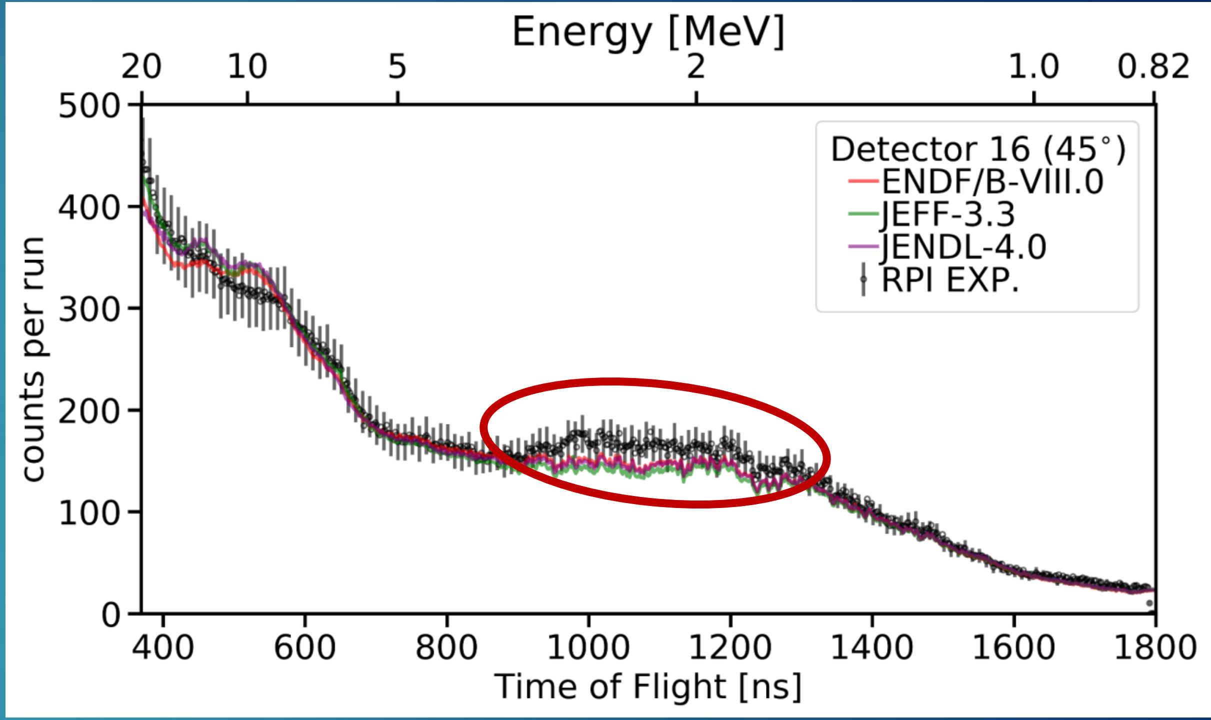
- Overall Carbon measurements and simulation are in good agreement at all angles
- For some angles and incident neutron energies differences are observed
- Further investigation of carbon double differential cross section might be needed

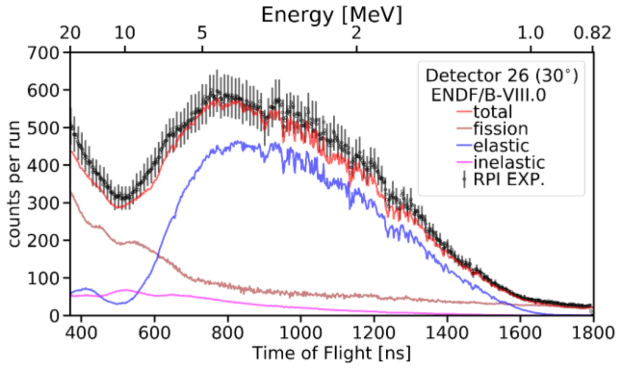


Discrepancies found are not correlated with shapes of the flux or efficiency, or discrepancies found in ^{235}U and ^{239}Pu.

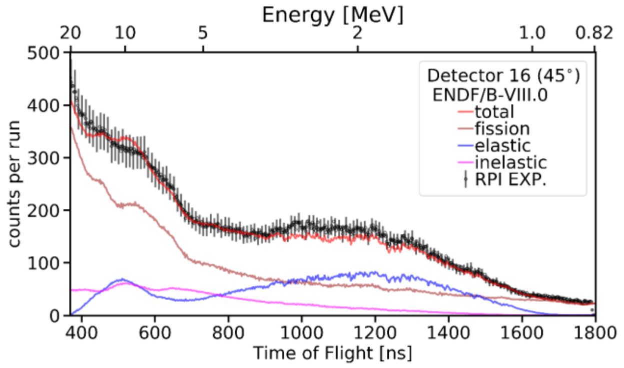
^{235}U forward angles



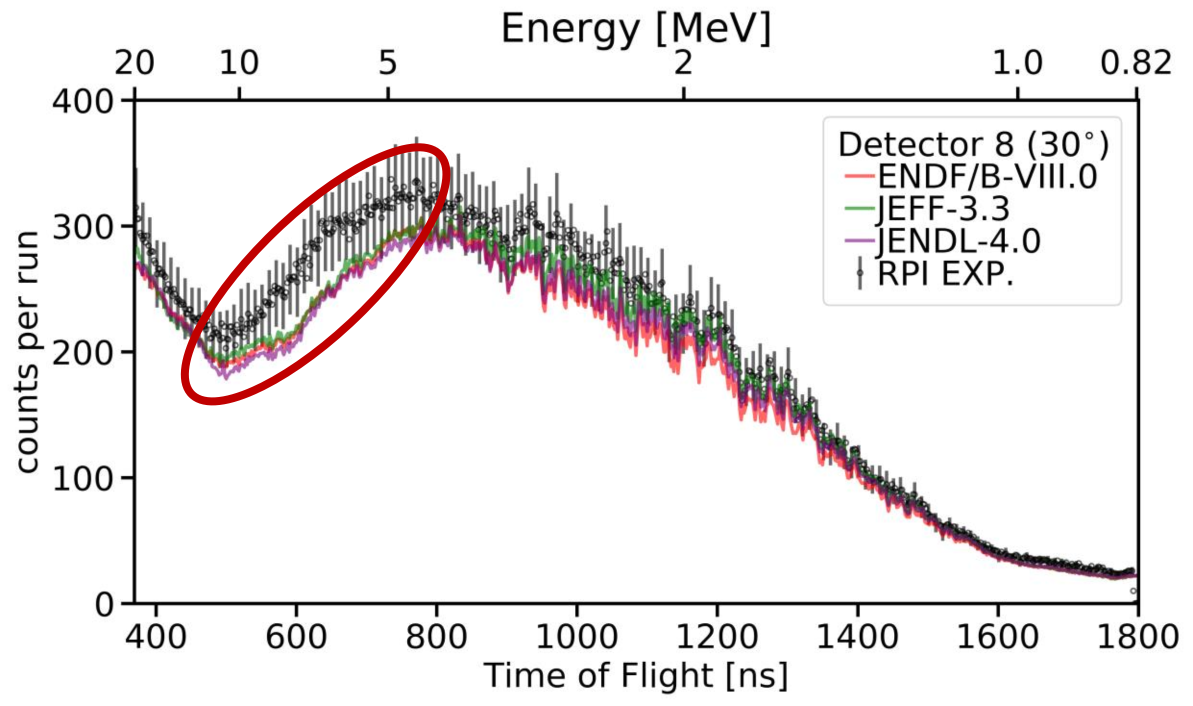


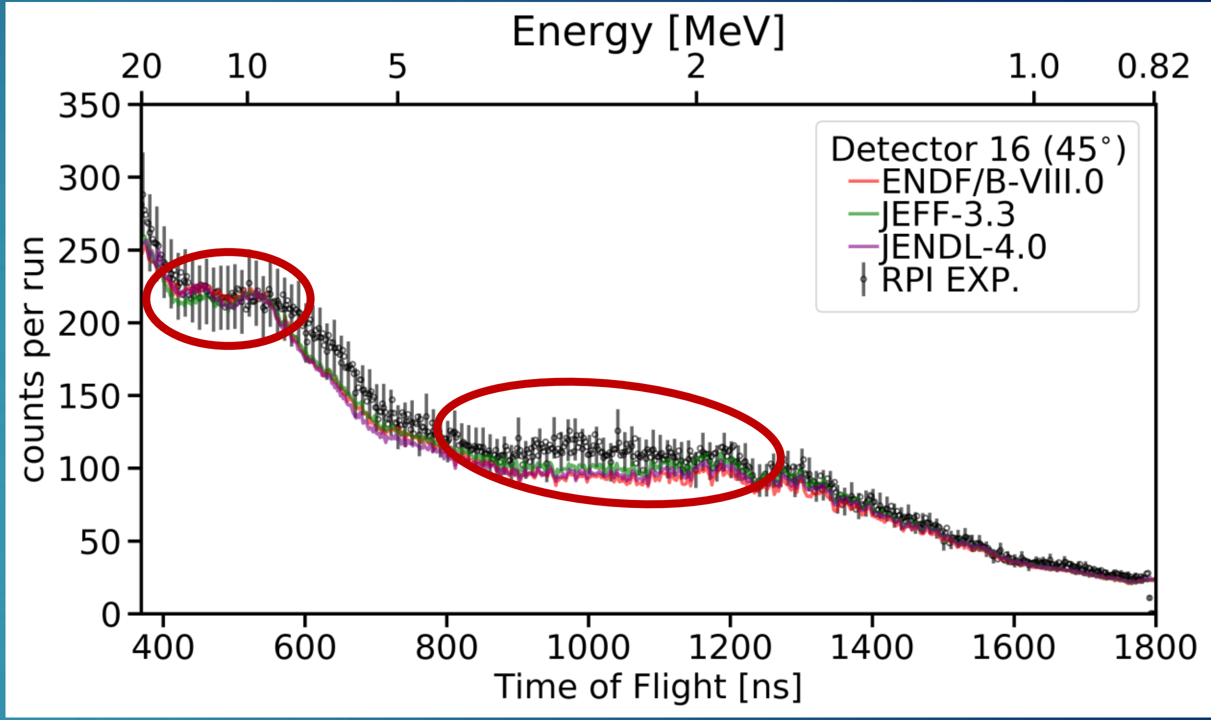


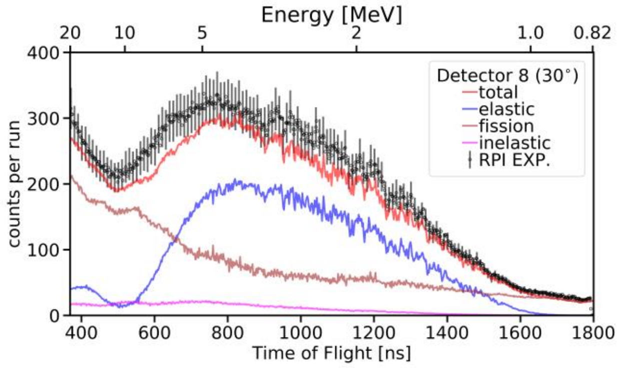
- Plotted uncertainty includes statistical and systematic uncertainty (normalization)
- 30° systematically low at edge of uncertainty
- 45° under-predicts 2-3 MeV
- Scattering dominance drops rapidly



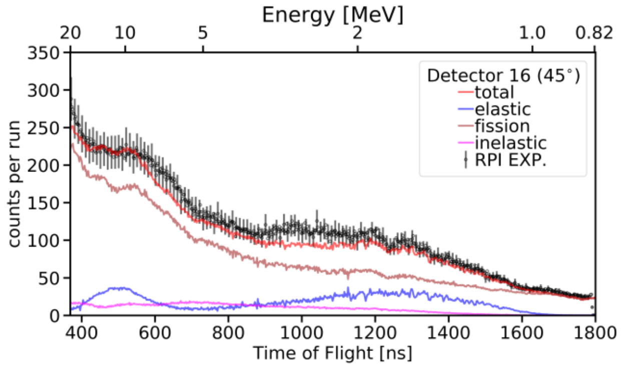
^{239}Pu forward angles





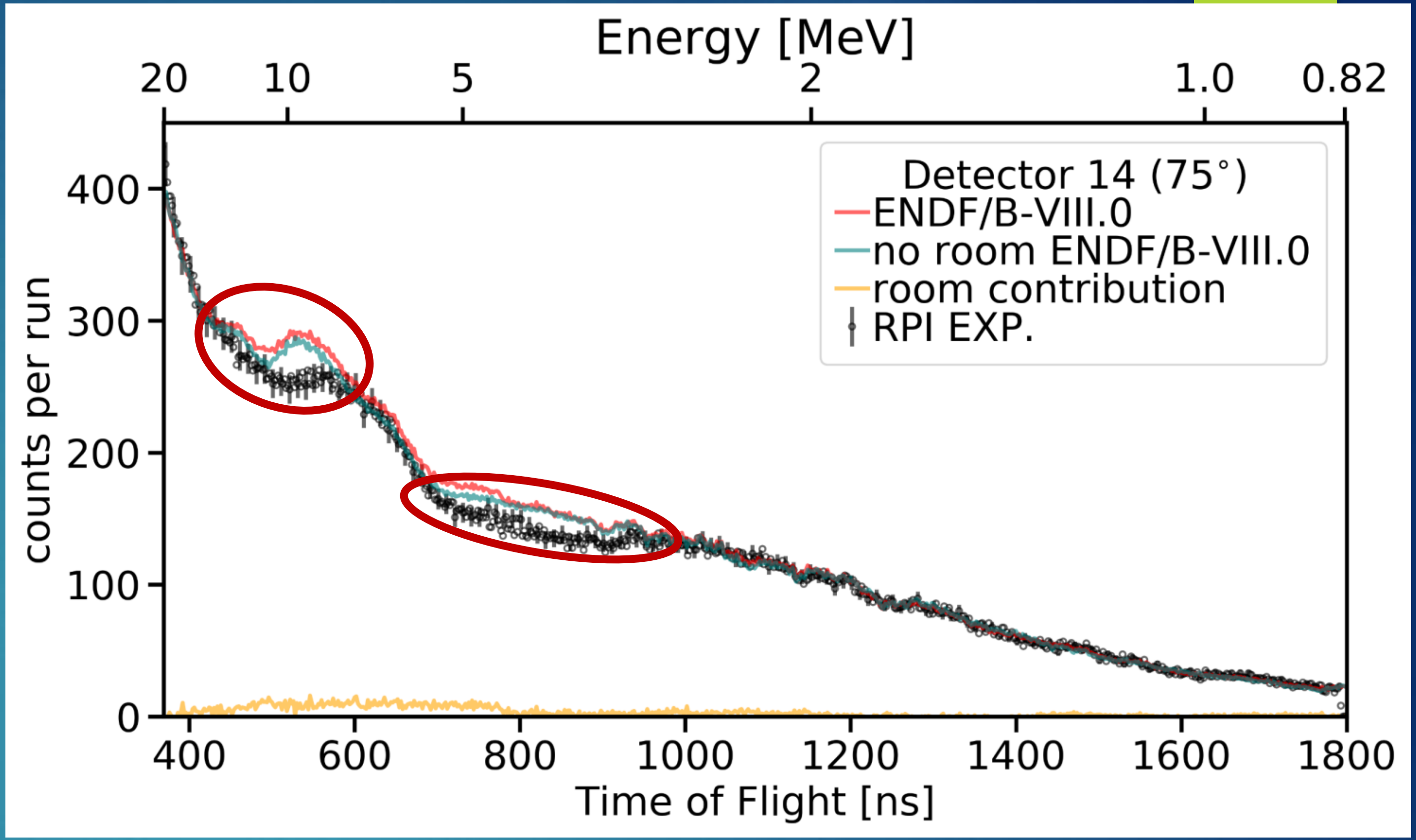


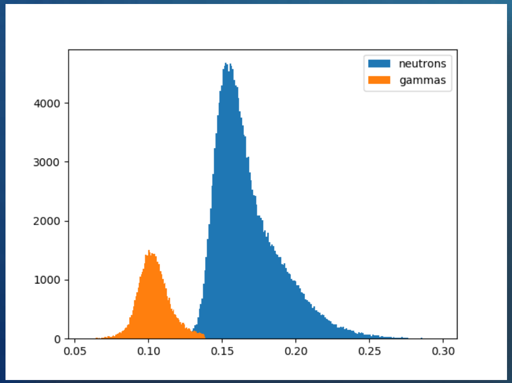
- 30° systematically low at edge of uncertainty and under-predicts 5-10 MeV
- 45° under-predicts 2-3 MeV

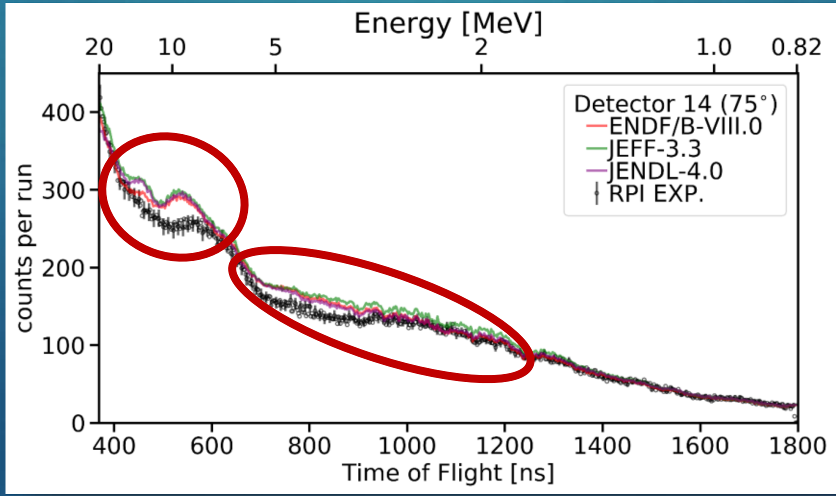


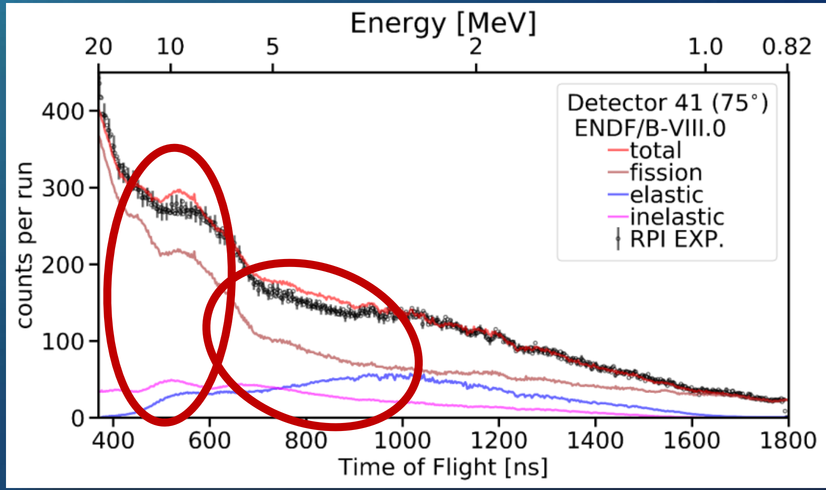
^{235}U

- Room return is not causing the shape discrepancy
- JEFF-3.3 shows over-estimation from 2-3.3 MeV
- All libraries show overestimation from 3.3-5 MeV
- All libraries over-estimate third-chance fission
  - May be related energy spectrum of emitted neutrons from fission/(n,xn)
  - ENDF/B-VIII.0 performs best in the feature at 12 MeV
- PSD holds good separation through 400-600 ns

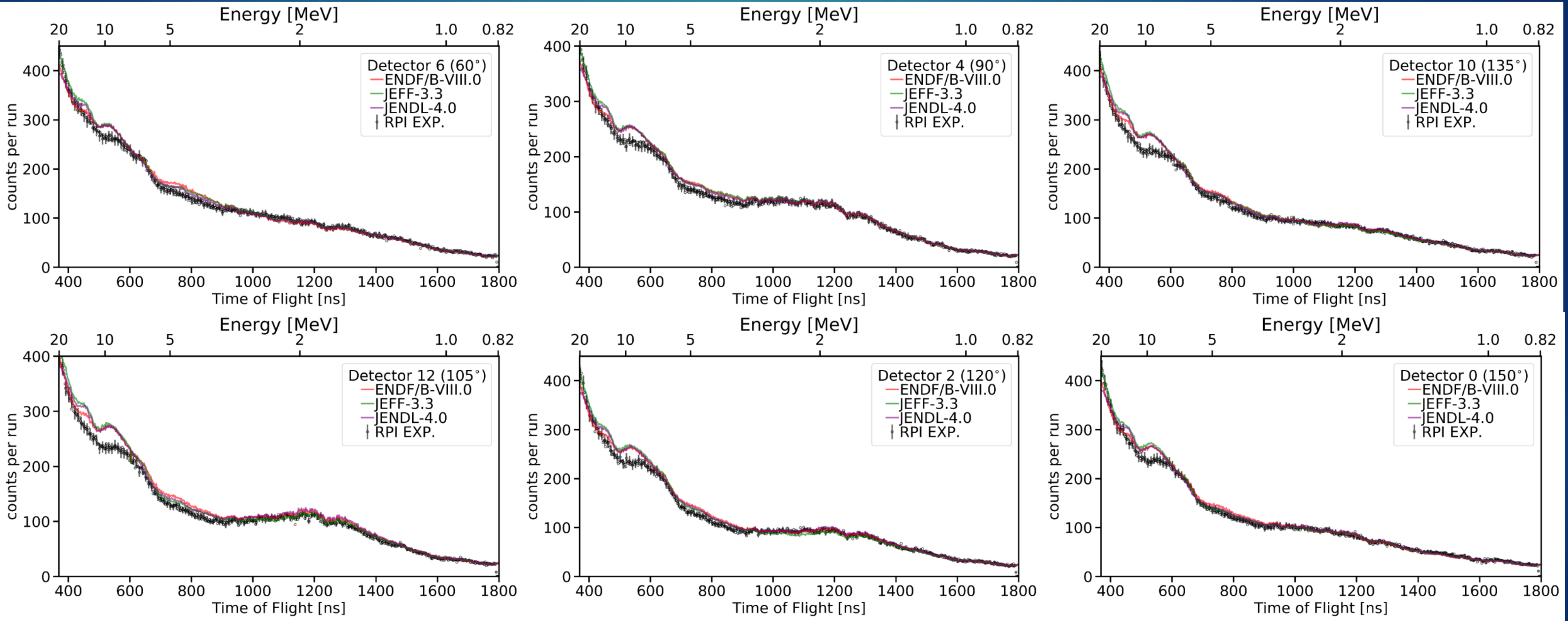




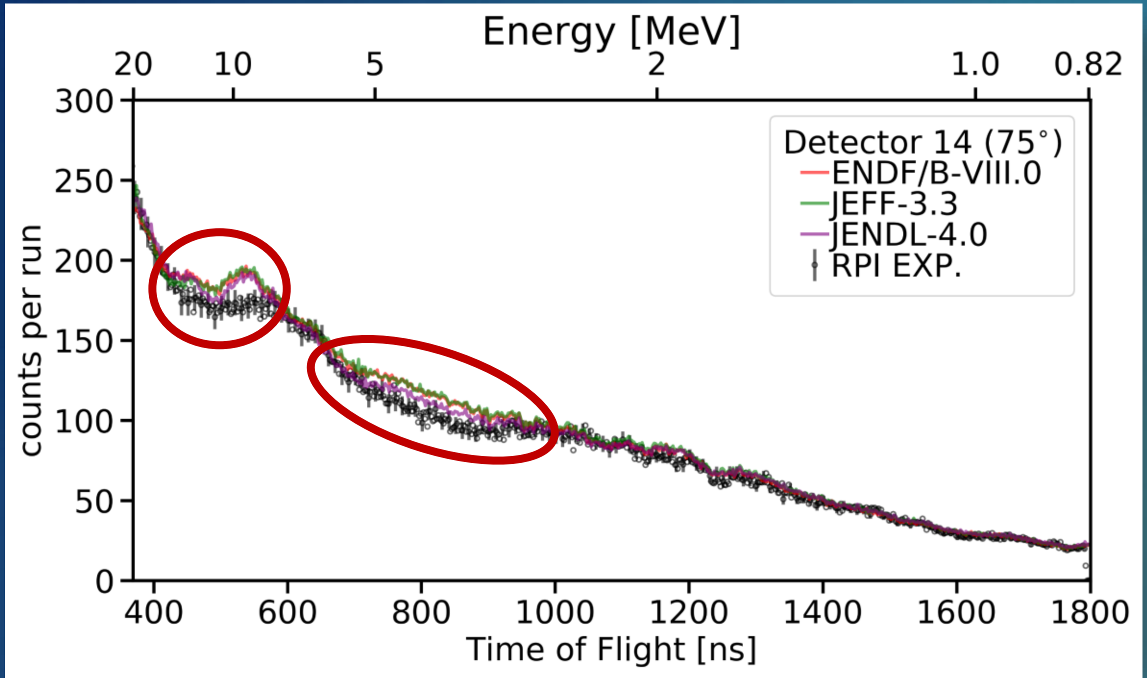


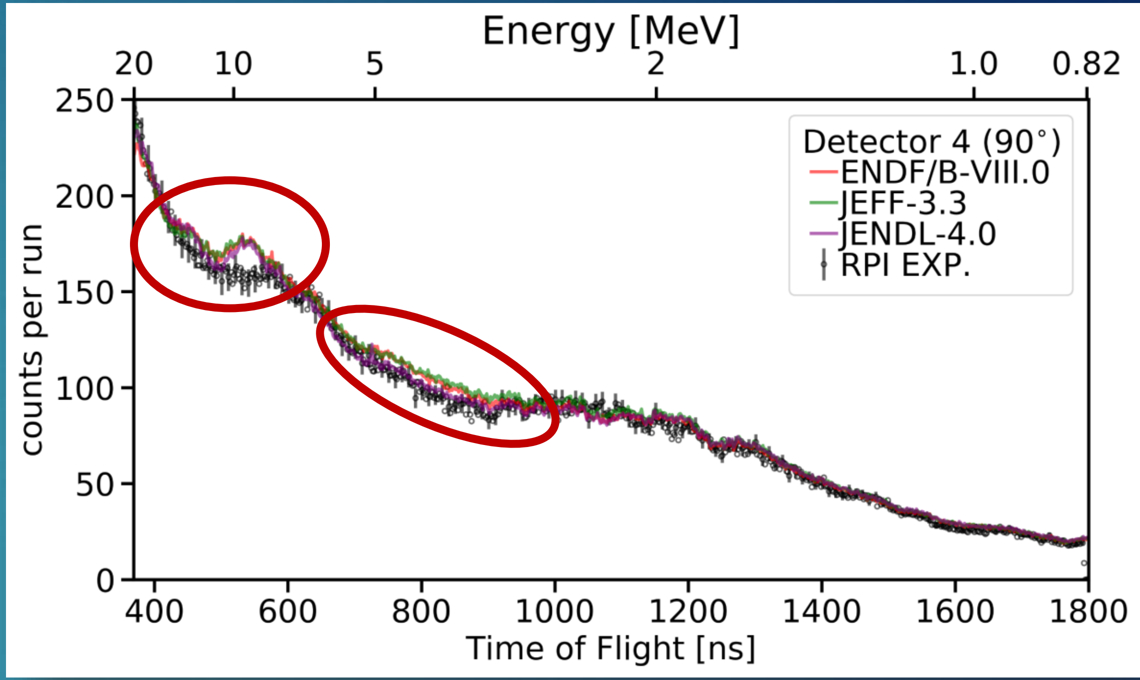


^{235}U – Angles ≥ 60°



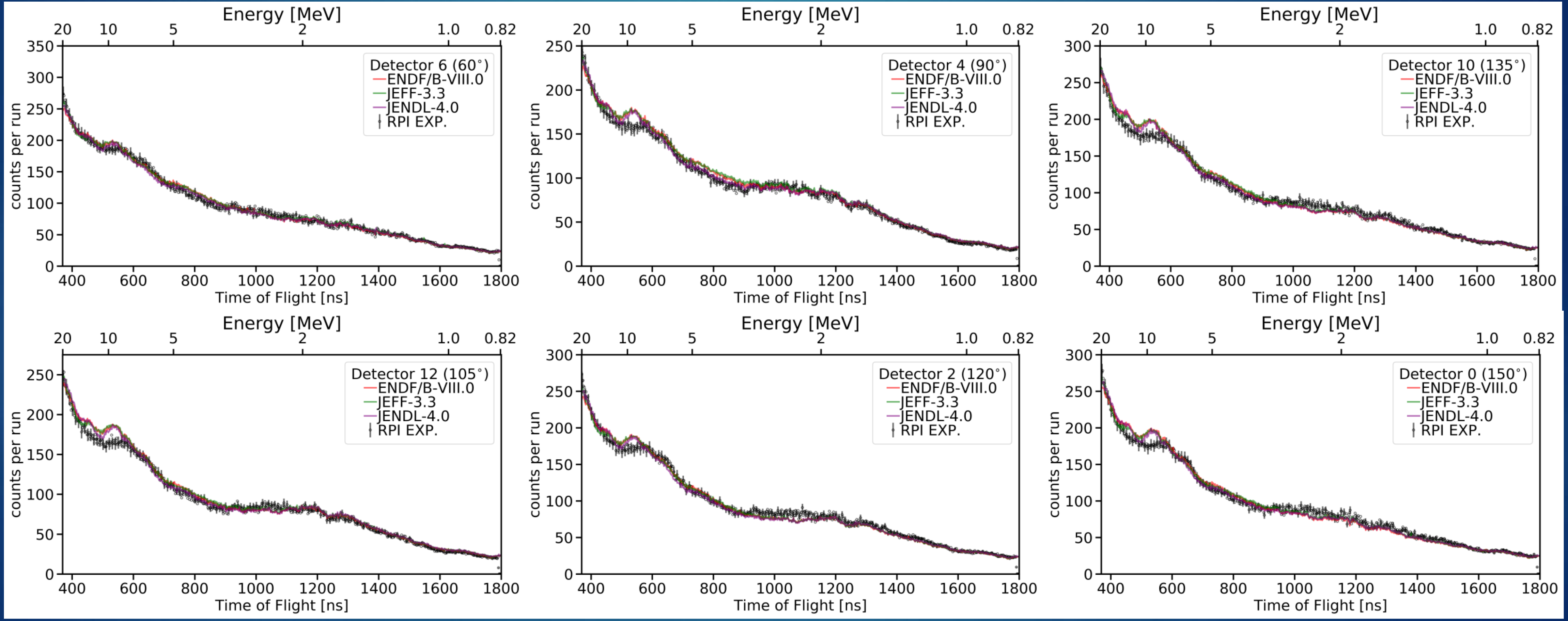
^{239}Pu central angles





- Discrepancy between 3.3-5 MeV with JENDL-4.0 performing closest to the experiment
  - Only at 60, 75 and 90 degrees
- Features at 10 and 12 MeV are still discrepant but not to the extent seen in ^{235}U

^{239}Pu – Angles ≥ 60°



Conclusions

- This work has shown that the RPI scattering methodology is usable for the measurement of neutron emission spectra to identify nuclear data discrepancies at LANSCE and can be used for future nuclear data investigations.

- A re-evaluation of both ^{235}U and ^{239}Pu above 2 MeV utilizing these results is recommended.
  - The third-chance fission discrepancy is seen in both ^{235}U and ^{239}Pu at angles ≥ 60 degrees. This may be related to either cross sections or the energy distribution of emitted neutrons.
  - ^{235}U suffers from what is expected to be a cross-section discrepancy between 3.3-5 MeV (seen at all angles ≥ 60 degrees)
  - ^{239}Pu suffers from localized issues appearing at different angles and energies
- An investigation of carbon nuclear data above 2 MeV is recommended.