Experiments with Neutron Induced Reactions

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Rensselaer
Mechanical, Aerospace and Nuclear Engineering

linac The Gaerttner LINAC Center

Collaboration

- RPI
  - PI
    - Y. Danon - Professor, Director Gaerttner LINAC Center, Nuclear Engineering Program Director
  - Research Associate
    - E. Blain – Measurement of prompt fission neutron spectra
  - Graduate Students:
    - N. Thompson – Measurement of capture rates with the Lead Slowing Down Spectrometer
    - K. Mohindroo – Measurement of fast neutron scattering
  - Undergraduate students
    - A. Lewis – Low energy neutron detector response
- LANL (Scattering/ LSDS)
  - R. Haight, M. Devlin, S. Mosby

Graduated 3 PhDs supported by SSAP
Supported 6 undergraduate research projects

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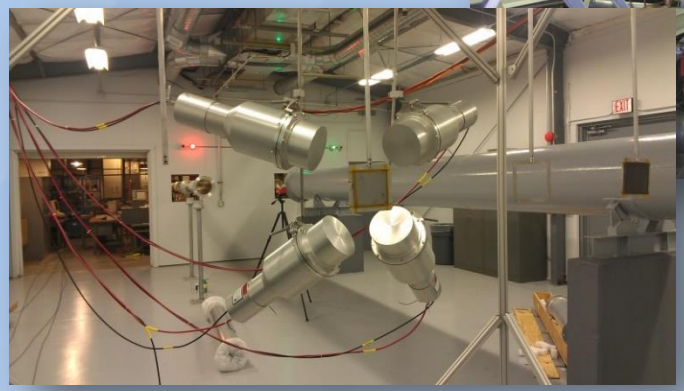
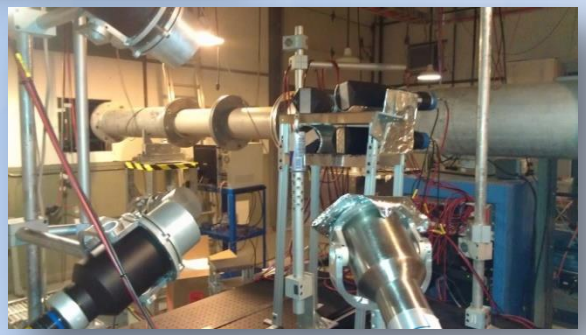
Additional information in the poster session

The Nuclear Data Program at the Gaerttner LINAC Center at RPI

- Driven by a 60 MeV pulsed electron LINAC ~**10^{13} n/s**
- **Neutron transmission**
  - Resonance region + : 0.001 eV - 5000 keV,
  - High energy region: 0.4- 20 MeV
- **Neutron Capture**
  - Resonance region + : 0.01 - 2000 keV
- **Neutron Scattering**
  - High energy region: 0.4 MeV- 20 MeV
- **LSDS**
  - Assay of used nuclear fuel
- **Novel Neutron Detectors**
- **Prompt Fission Neutron Spectrum**
- **LSDS**
  - Fission cross section and fission fragment spectroscopy.
  - (n,α), (n,p) and (n,γ) cross sections on small (radioactive) samples.
- Support from various DOE offices





LINAC 2020 - upgrade and refurbishment project

Research Topics

- **LSDS**
  - Measure capture reaction rates of samples with small mass or small cross sections (LSDS)
- **Time-of-Flight**
  - Measurement of fast neutron scattering and angular distributions for U-235 and Pu-235 at LANL
  - Measurement of prompt fission neutron spectrum of Cf-252 for E>5 MeV

Motivation

- Capture Cross Sections
  - Neutron induced cross sections are important for stockpile stewardship.
    - Radiochemistry for example.
  - Resonance and unresolved capture cross section of samples with low mass or low cross section are important in some cases.
- NEW Neutron Scattering data
  - Elastic and inelastic scattering are hard to measure and result in large uncertainties in the evacuations.
  - New experimental data helps to constrain the models used by evaluators
- Prompt Fission Neutron Spectrum
  - Reduction of PFNS uncertainties is required due to high sensitivity in criticality calculations.
  - Little information for neutron emission below ~1 MeV and above 6 MeV where it is hard to measure.

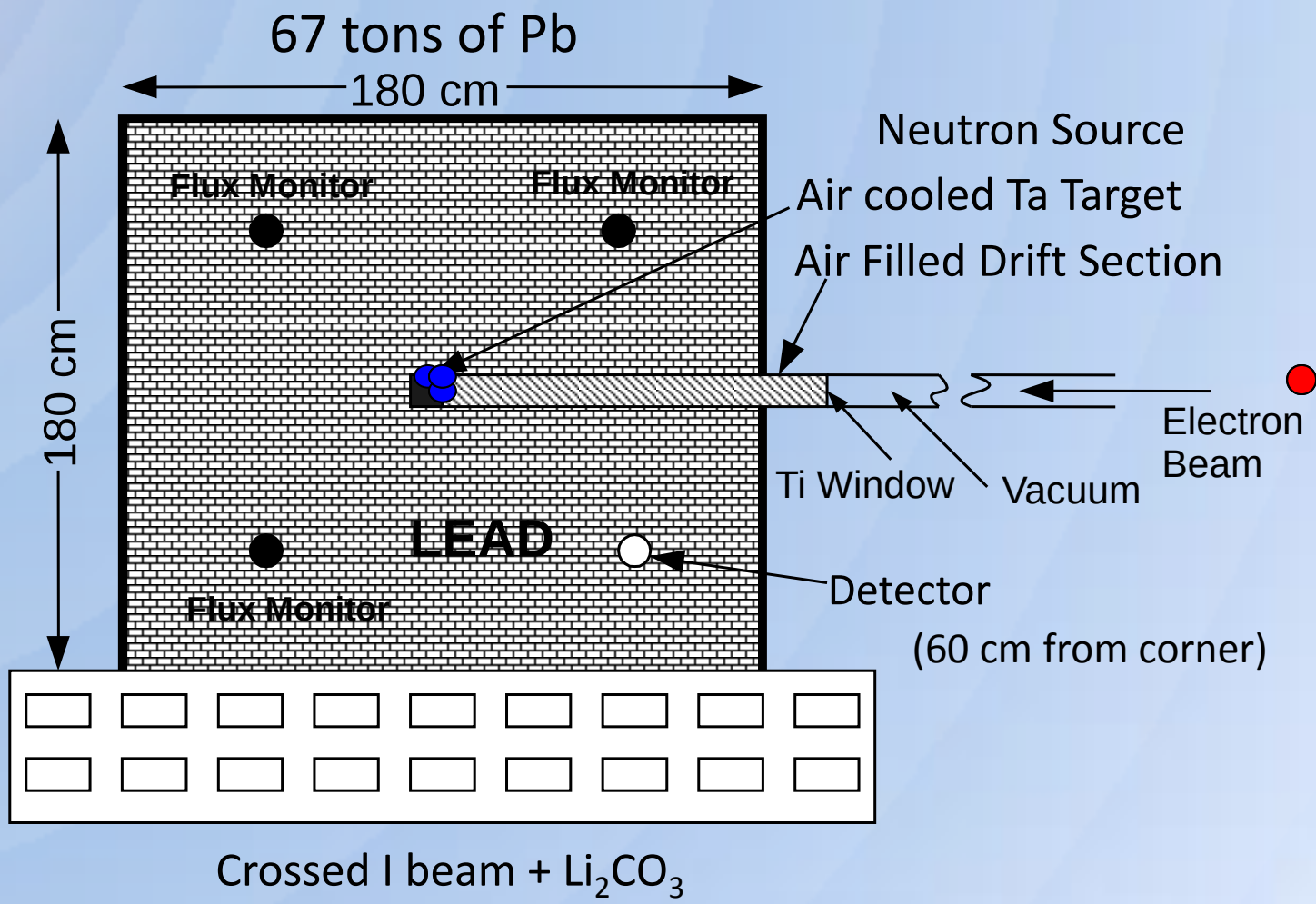
Improvements in nuclear data ➔ more accurate neutron transport calculations

Neutron Capture with the LSDS

Why Use a Lead Slowing-down Spectrometer

Lead Slowing-down Spectrometer at RPI

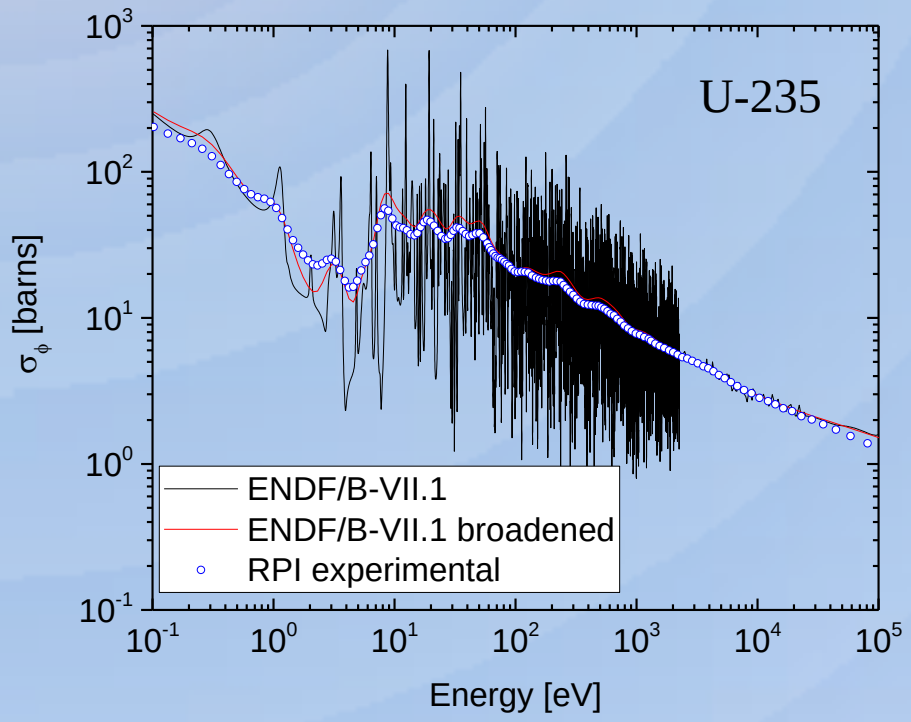
- Tantalum target in the center produces neutrons.
- Neutrons scatter elastically with the Pb.
- Neutrons can pass through the same position several times.
- **About 10^3-10^4 times higher flux than an equivalent neutron TOF experiment.**

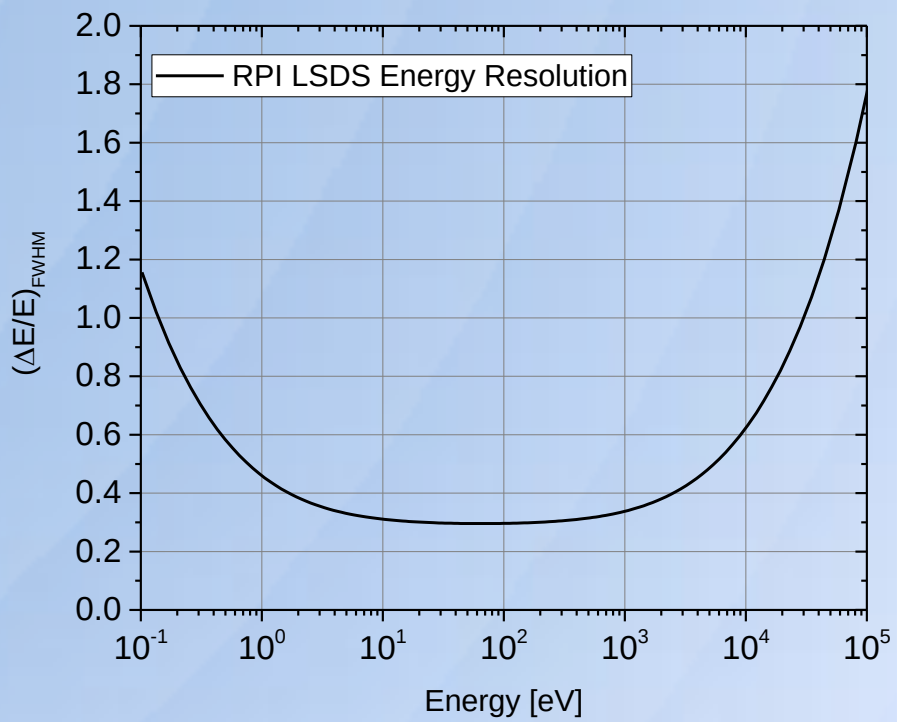


Crossed I beam + Li_2CO_3

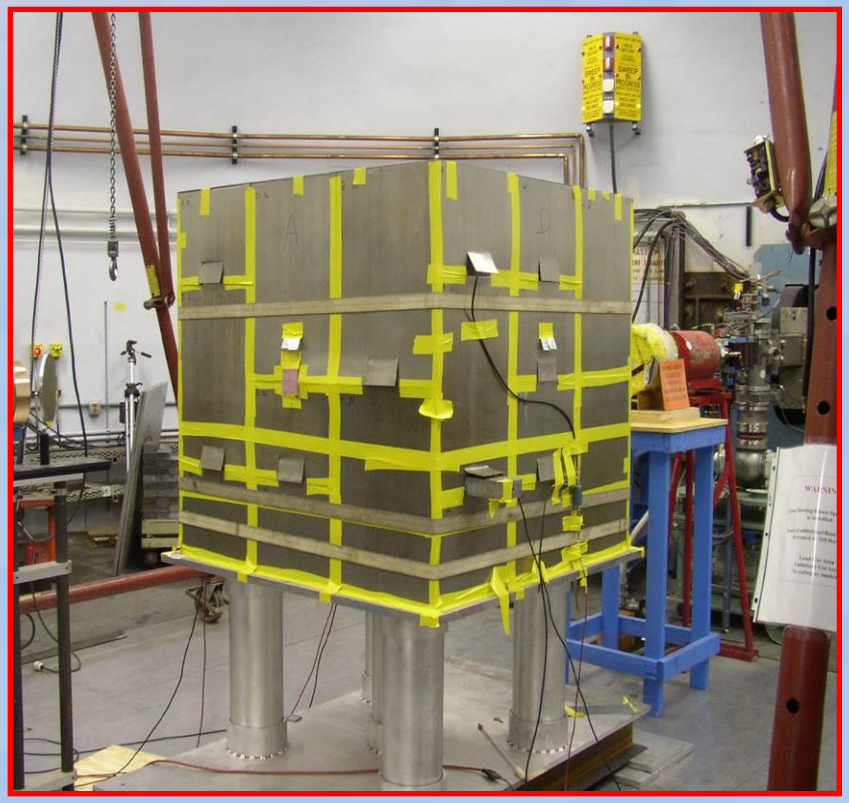
LSDS Energy Resolution

- Best energy resolution is about 30%
- Compromise of high intensity and low resolution
- Provides broad energy resolution cross sections

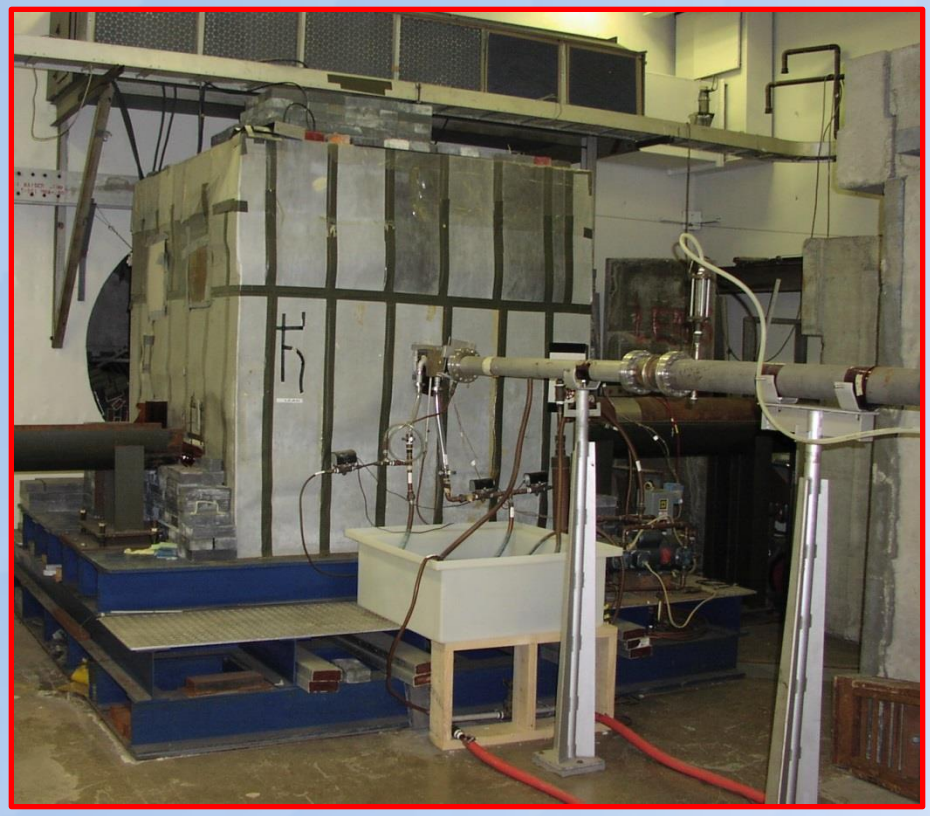




Lead Slowing Down Spectrometers in the US



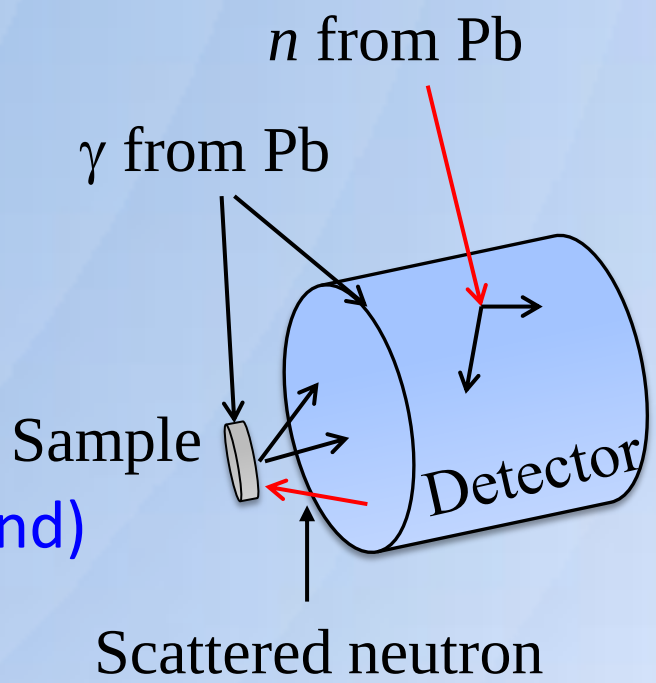
LANL *– Proton Driven*



RPI *–Electron Driven*

Goal for the LSDS measurement

- Use a Lead Slowing Down Spectrometer to measure the capture rate of samples of small mass or small cross section
  - Detect γ from capture
- Criteria for a γ detector
  - Low n-capture cross section
  - Low n-scattering cross section
  - High γ detection efficiency
  - Small detector (reduce background)
  - Fast recovery from gamma flash

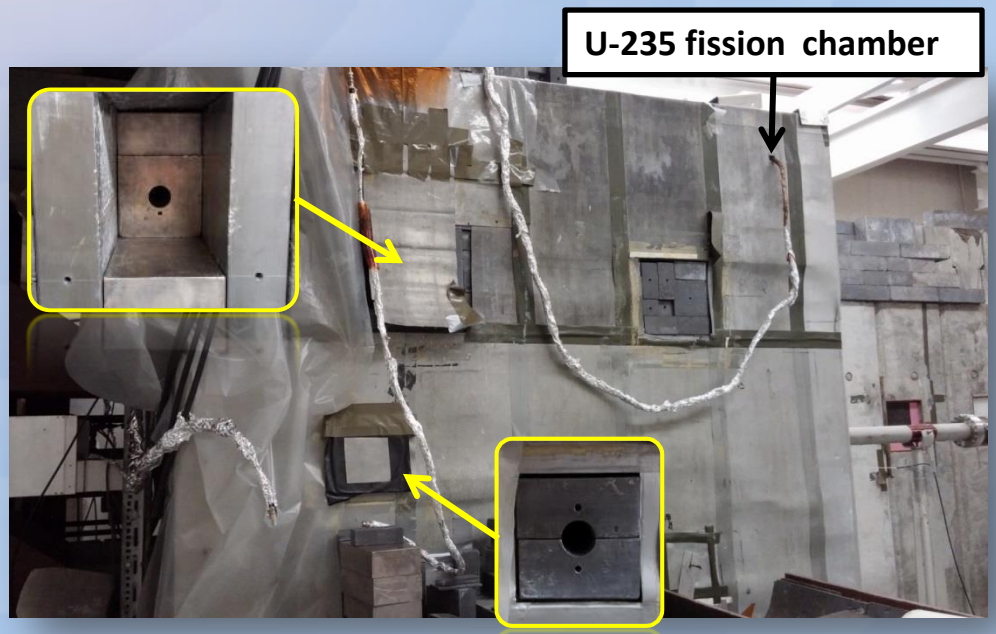


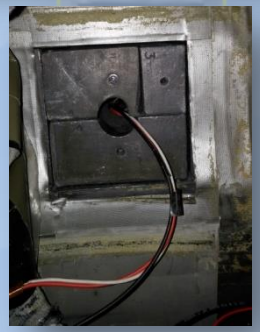
Detector

- After experiments and simulation of different detectors YAP scintillator was selected
- Thin YAP ($YAlO_3$:Ce) scintillator used
  - Good mix of light output, decay time, and low neutron capture and scattering cross sections
- Small sample (0.1 – 2 mm thickness) affixed to the detector

Experimental setup

Detector positions in the LSDS



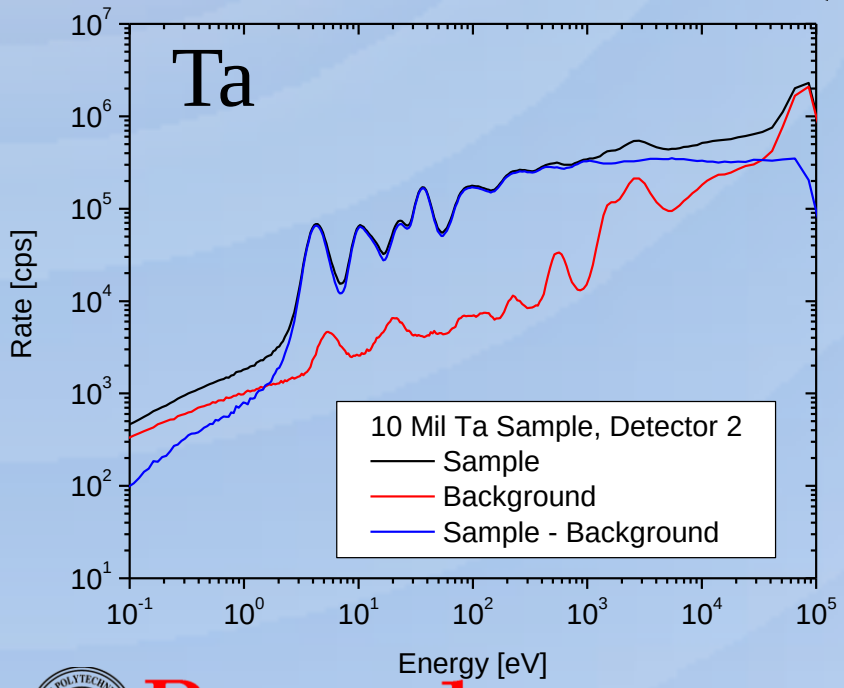


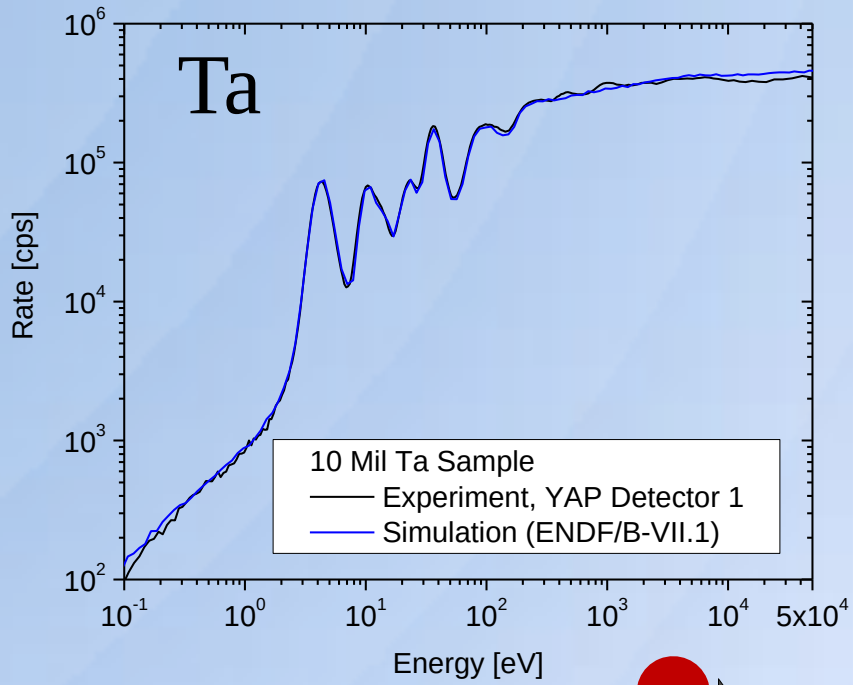
- Operate the LINAC at E_e=54 MeV and I<0.1 μA
  - about 0.8% of max power possible with the RPI-LSDS
- Use Digital DAQ – Acqiris AP 240
- Made two type of measurement:
  - Sample in
  - Sample out
- Used a ~1 mg U-235 fission chamber to monitor the neutron flux in the LSDS

Results, Ta Sample

- Data analysis:
  - Correct for deadtime background and
  - Compare the count rate to detailed MCNP time dependent simulation of the geometry, use tally F6 card to get the energy deposition in the detector.
  - Convert the slowing down time of both the experiment and simulation to energy:

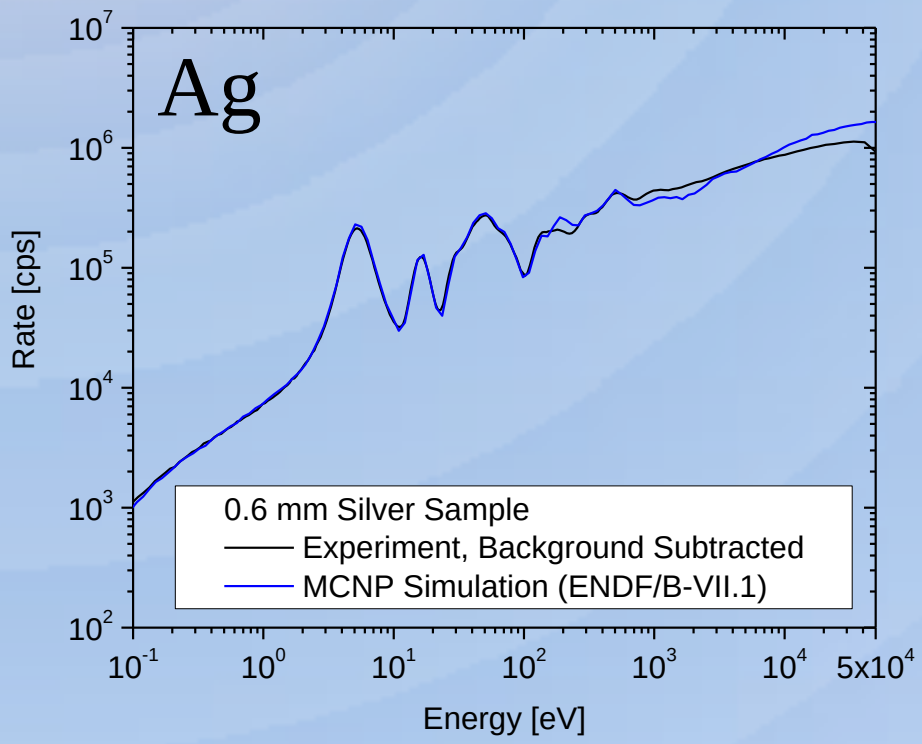
$$E = \frac{165000}{(t + 0.3)^2}$$

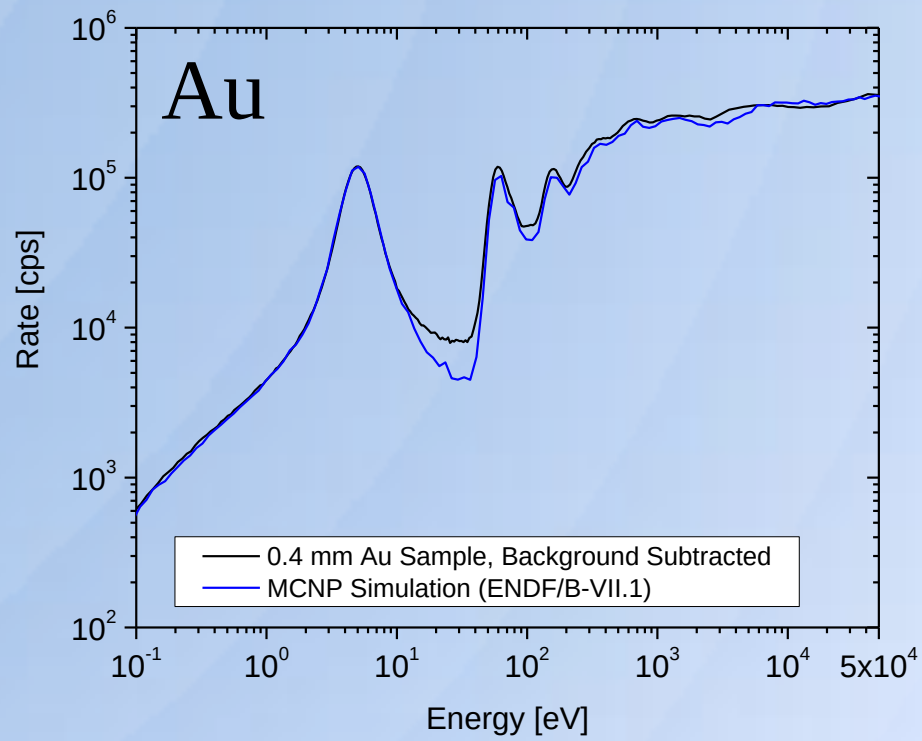




Results Ag and Au

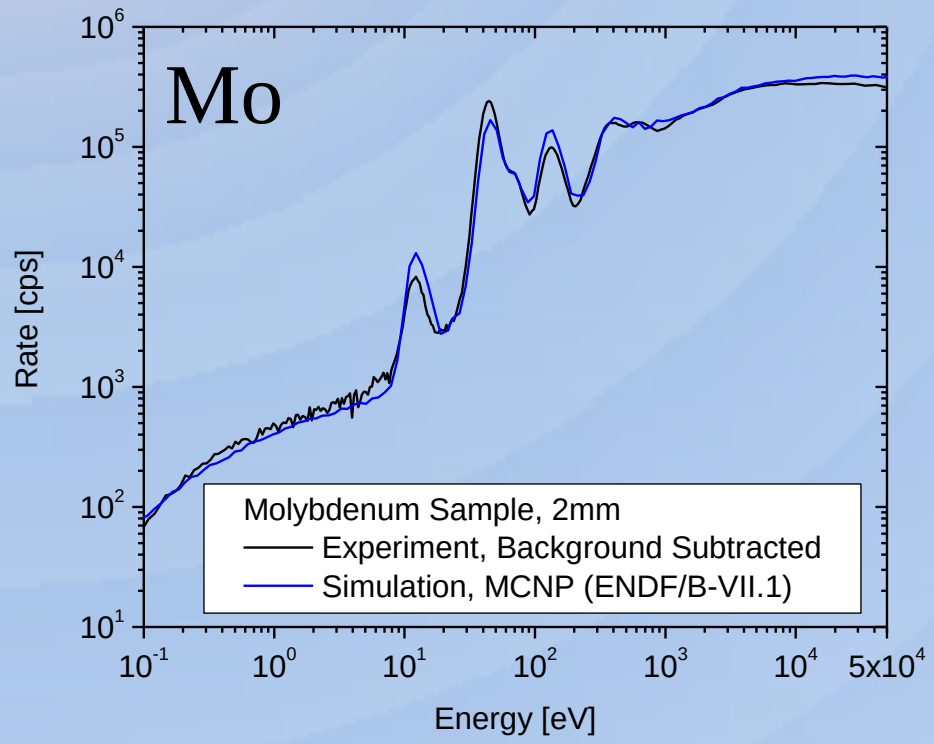
- The simulation has a slightly narrower energy resolution
  - Need to investigate the materials included in the vicinity of the detector
- Possible room return background in Au sample

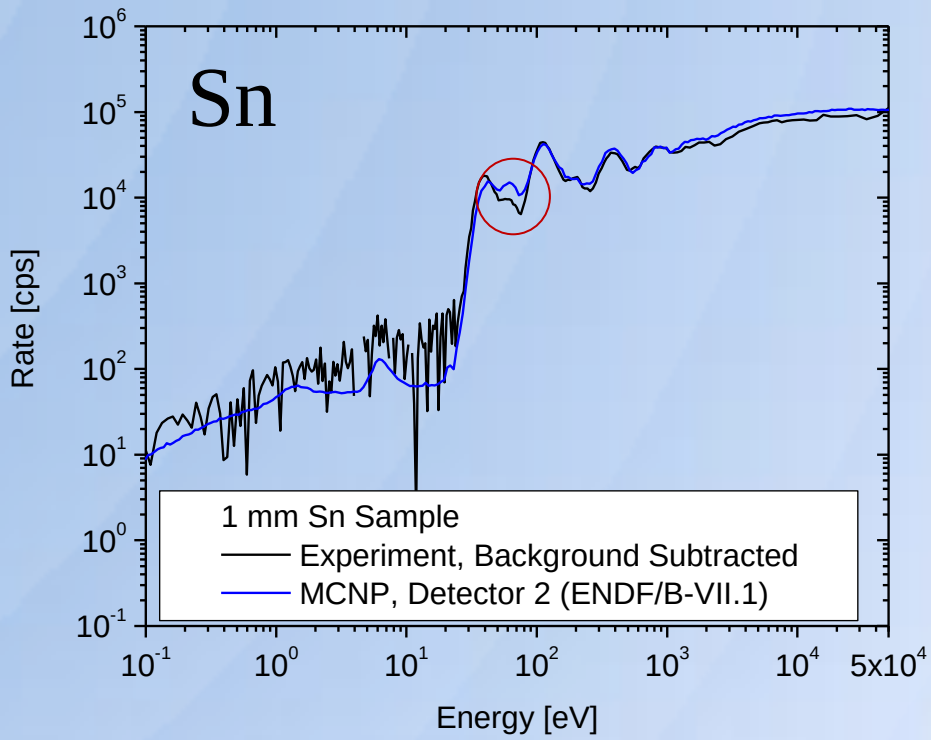




Results Mo and Sn

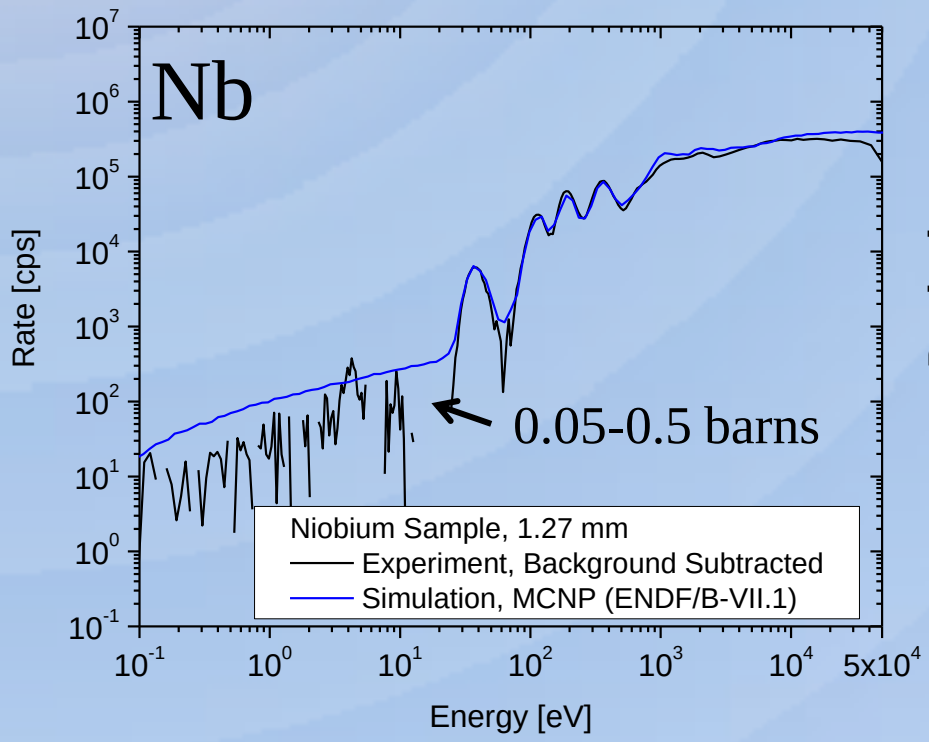
- Possible problems with Mo cross section (Mo has 7 stable isotopes).
- Two different positions and detector give similar results for Sn.
  - Differences near 50 eV

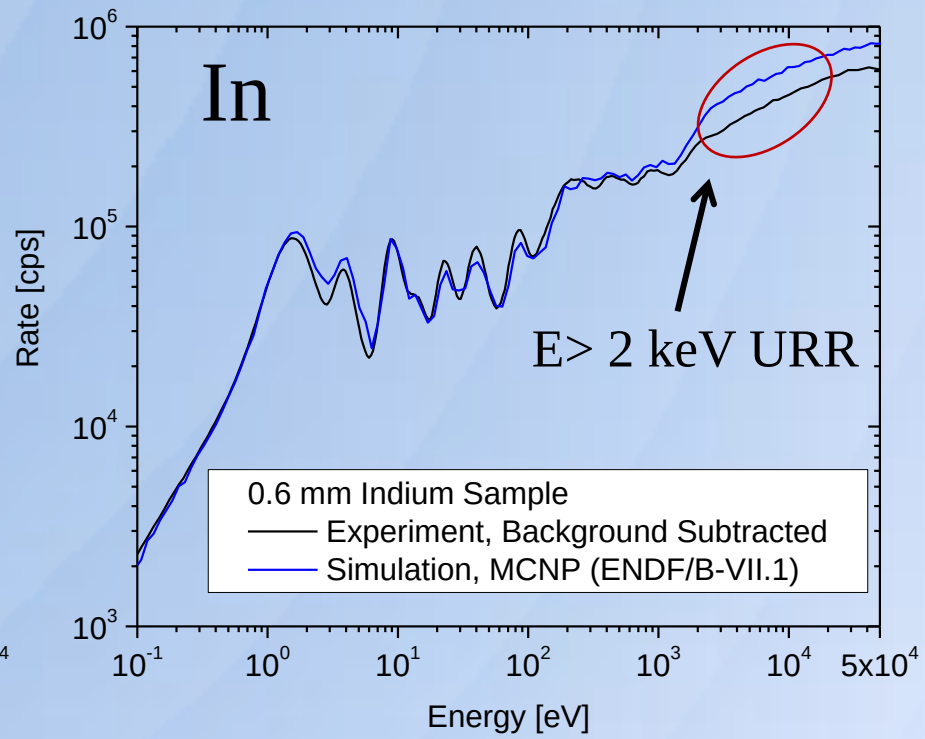




Results Nb and In

- Background is too high for low σ <0.5 barn
- Possibly a problem in the URR region of In

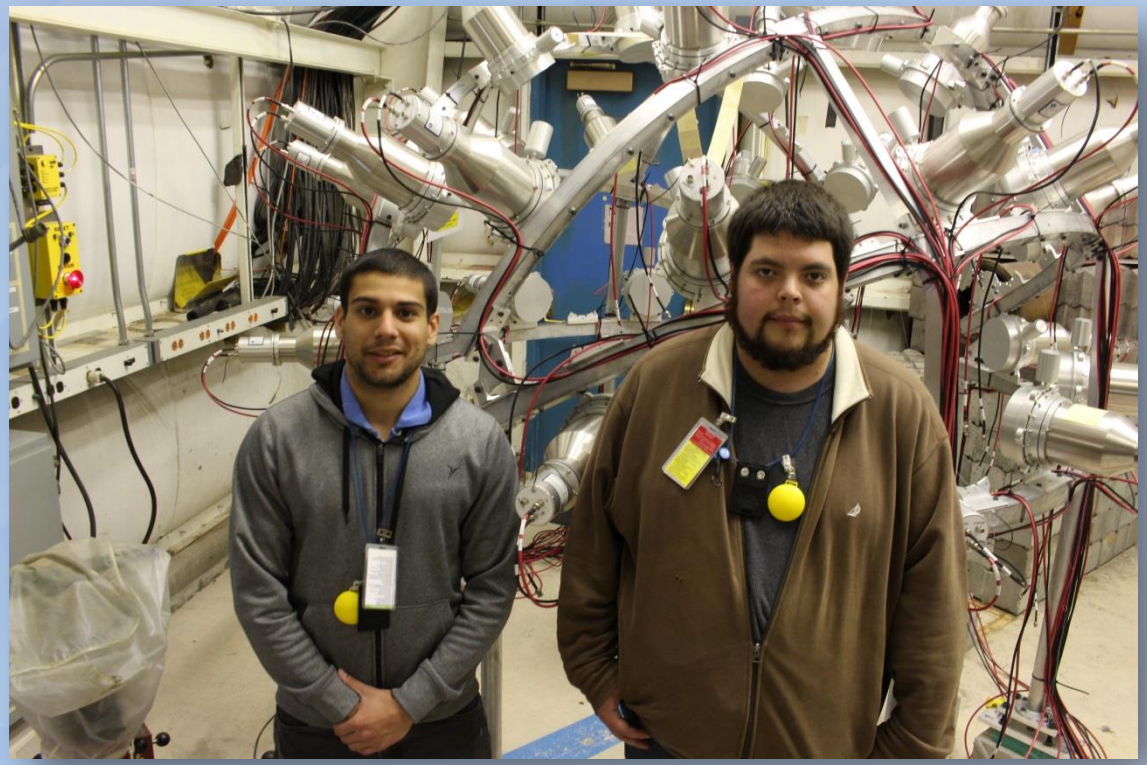


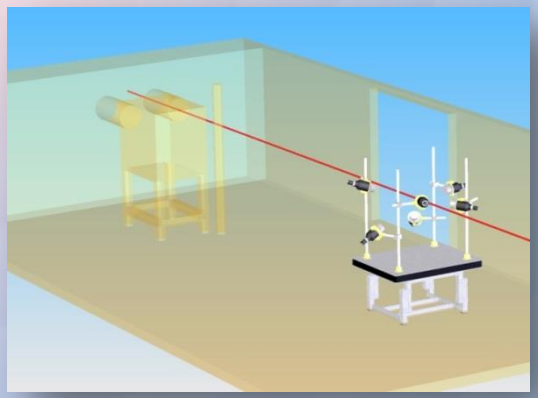


Conclusions – LSDS Capture

- Demonstrated measurements of several materials:
  - In some cases there are significant differences between the simulation and experiment.
- Improve the simulation accuracy
  - Account for H content in LSDS
  - Accurate description of martials near the detector
- Investigate other evaluations.

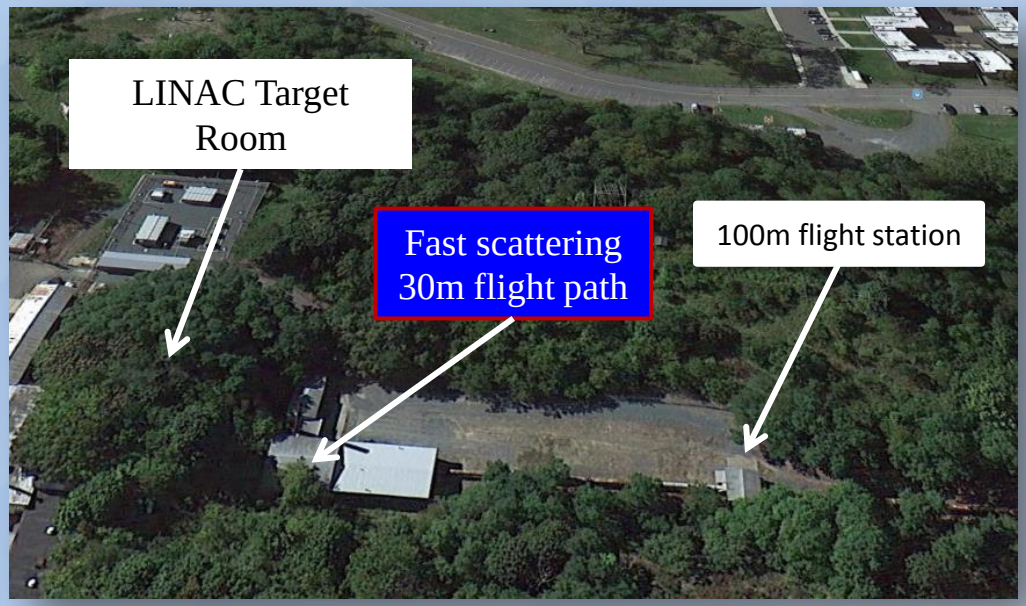
Fast Neutron Scattering





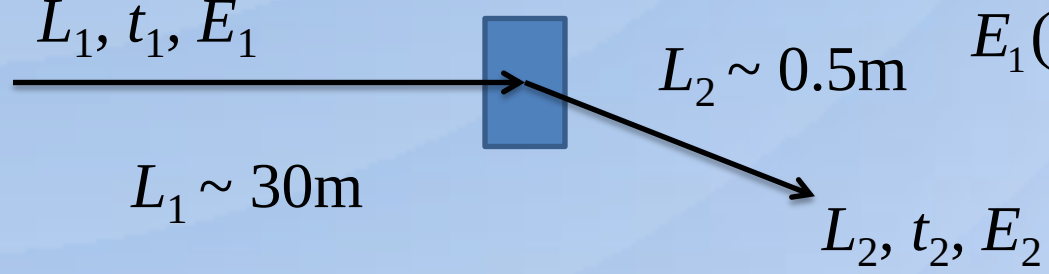
Fast Neutron Scattering at RPI

Quasi-differential neutron scattering and angular distributions.



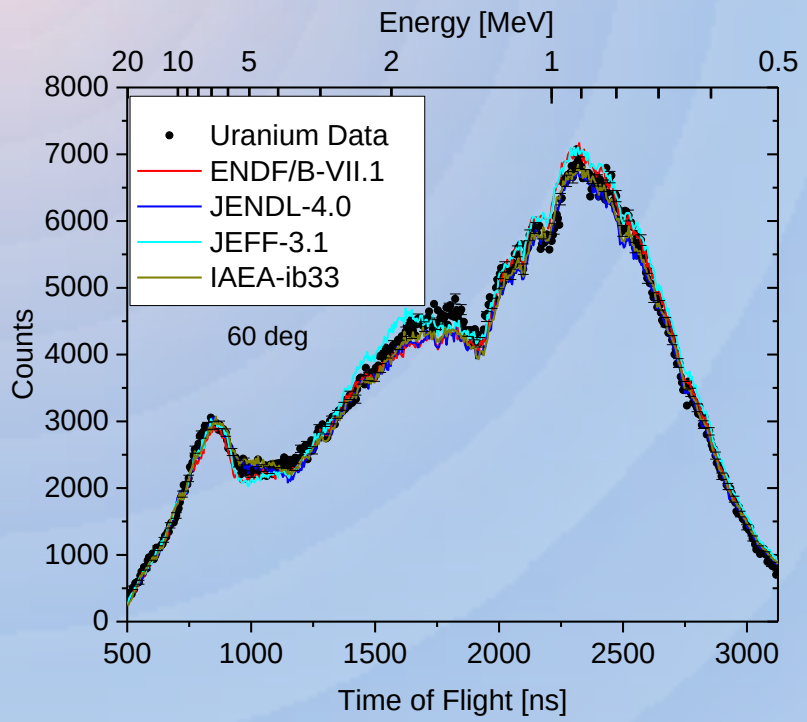
TOF Scattering Measurement

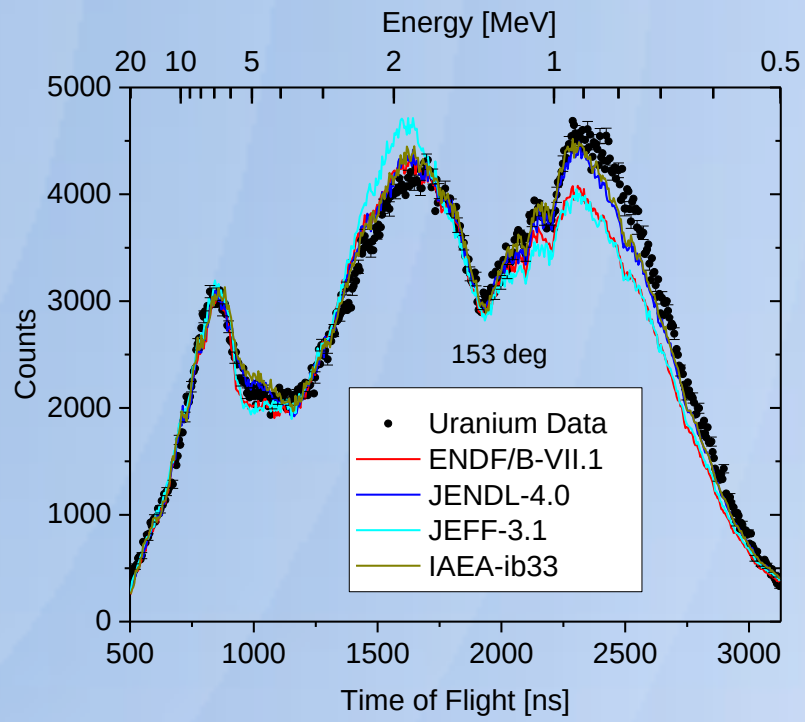
- Measure TOF: $t = t_1 + t_2$; where $t_1 \gg t_2$
- All scattering events: $E_2 < E_1$
- For elastic scattering with A » 1: $E_1 \sim E_2$
- Assuming $L = L_1 + L_2$ than total TOF, t, can be used to calculate the incident neutron energy, $E_1(t)$



$$E_1(t) \approx m_n c^2 \cdot \left(\frac{1}{\sqrt{1 - \left(\frac{L}{c \cdot t} \right)^2}} - 1 \right)$$

^{238}U Scattering Measurement





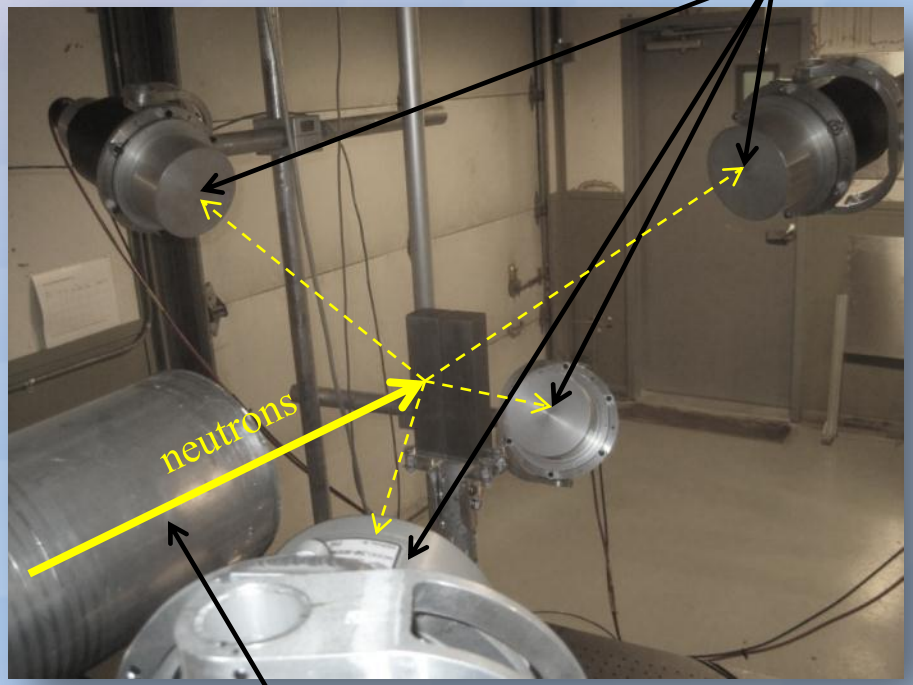
Library	FOM
ENDF/B-VII.1	3.78
JEFF-3.1	5.00
JENDL-4.0	3.34
IAEA-ib33	2.86

Library	FOM
ENDF/B-VII.1	12.60
JEFF-3.1	14.65
JENDL-4.0	4.66
IAEA-ib33	4.37

A.M. Daskalakis, R.M. Bahran, E.J. Blain, B.J. McDermott, S. Piela, Y. Danon, D.P. Barry, G. Leinweber, R.C. Block, M.J. Rapp, R. Capote, A. Trkov, "Quasi-differential neutron scattering from U-238 from 0.5 to 20 MeV", Annals of Nuclear Energy, Volume **73**, Pages 455-464, November 2014.

Fe Scattering Measurement - Setup

EJ-301 Liquid Scintillator Neutron Detectors



Evacuated Flight Tube

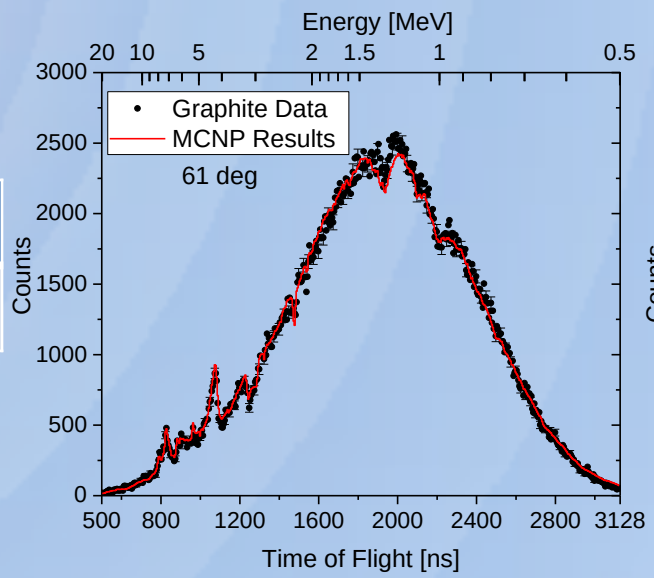
- Fe Sample
- Dimensions 77.0 x 152.6 x 32.2 mm

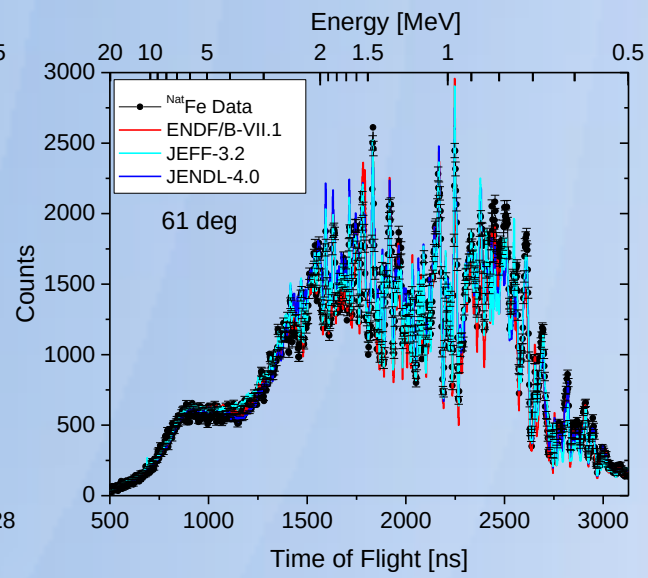
The neutron beam size is smaller than the sample.

A. M. Daskalakis, E. J. Blain, B. J. McDermott, R. M. Bahran, Y. Danon, D. P. Barry, G. Leinweber, M. J. Rapp, R. C. Block, "Separation of Neutron Inelastic and Elastic Scattering Contribution from Natural Iron using Detector Response Functions", 12th International Topical Meeting on Nuclear Applications of Accelerators (AccApp '15), Washington D.C., November 2015.

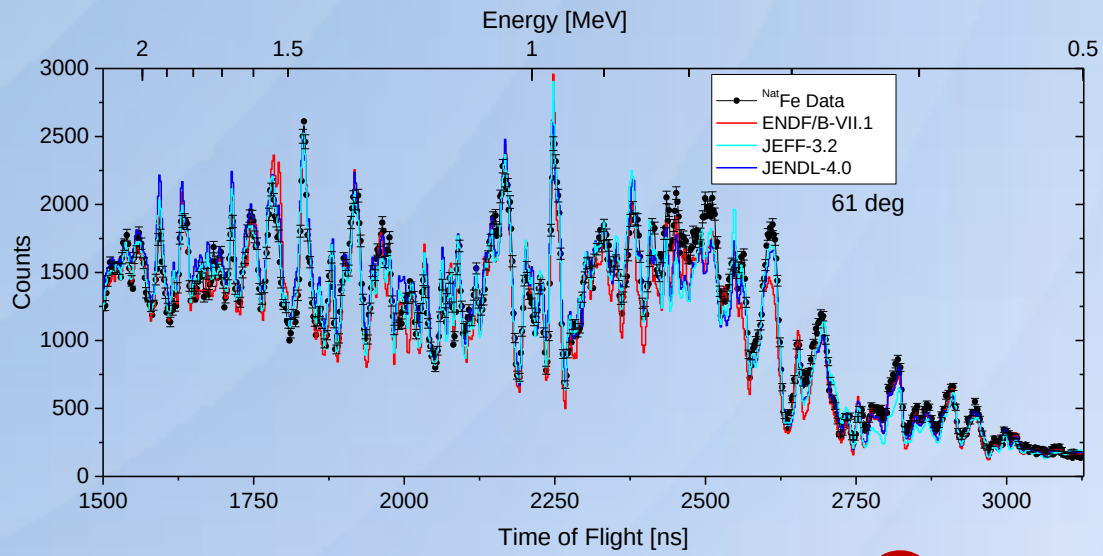
^{nat}Fe Scattering - 61°

Reference	FOM
Graphite	1.3



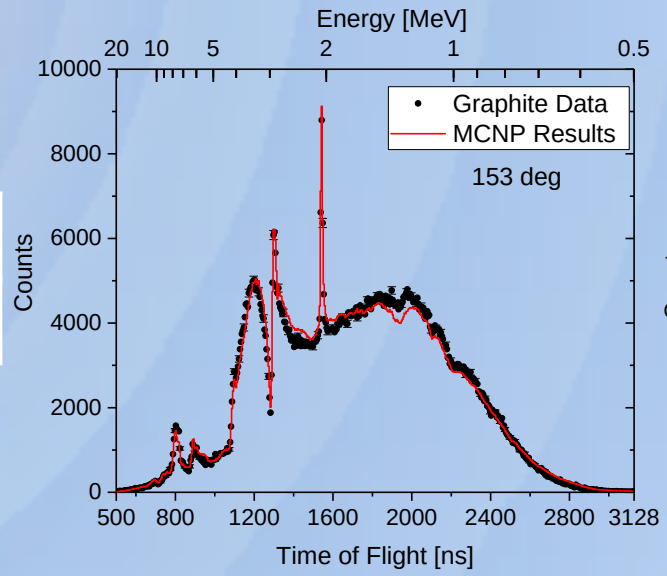


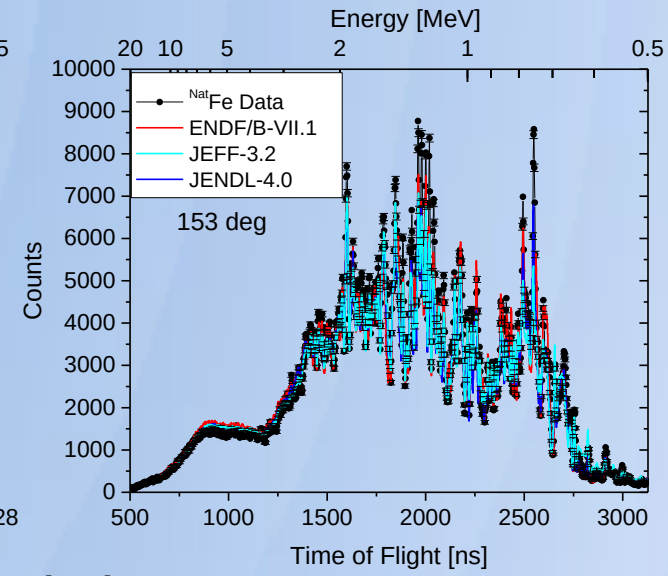
Library	FOM
ENDF/B-VII.1	9.5
JEFF-3.2	9.5
JENDL-4.0	8.4



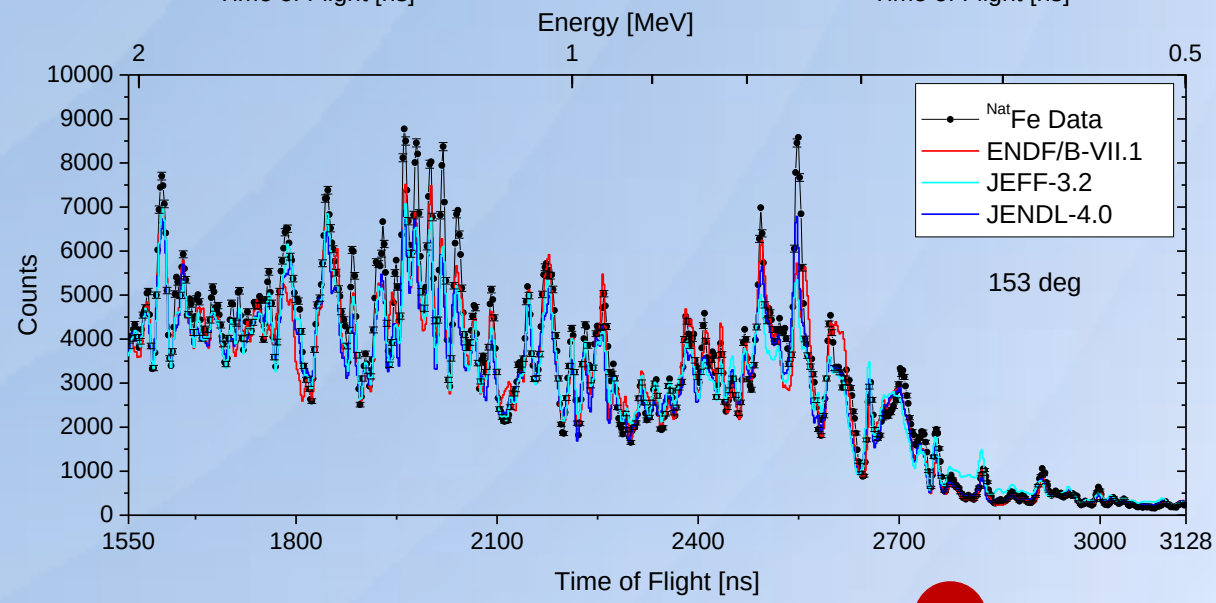
^{nat}Fe Scattering - 153°

Reference	FOM
Graphite	4.9

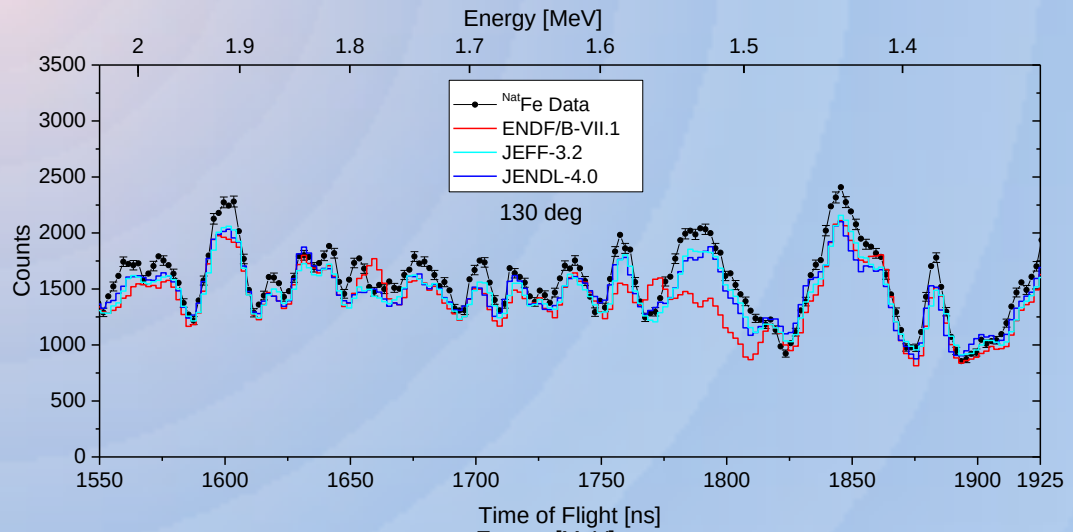


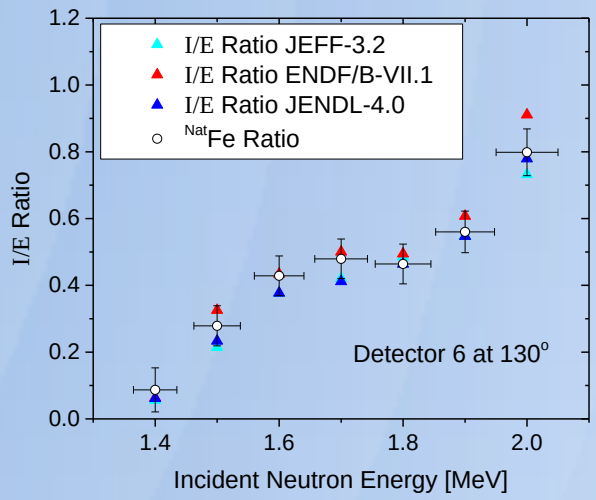


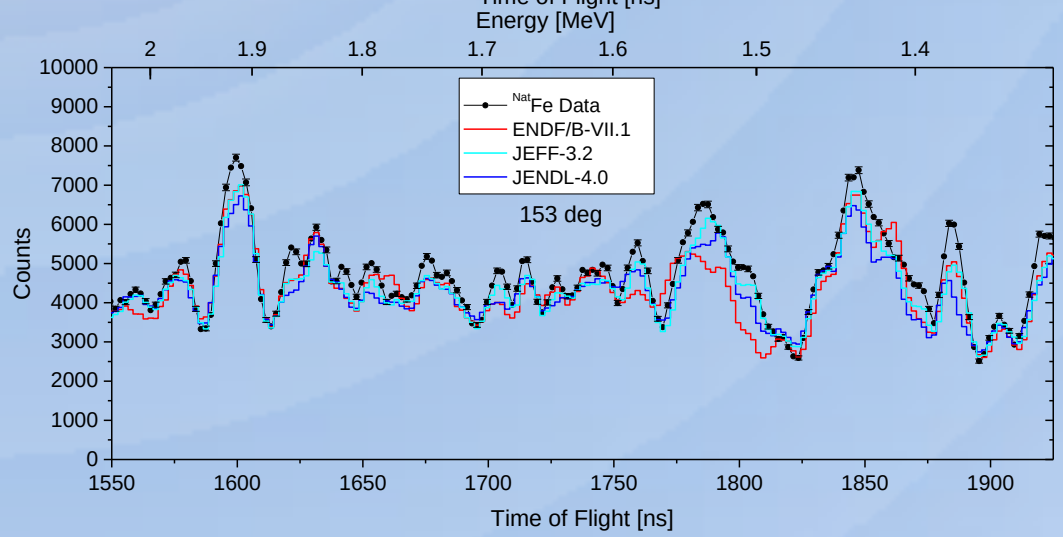
Library	FOM
ENDF/B-VII.1	25.9
JEFF-3.2	39.8
JENDL-4.0	23.4

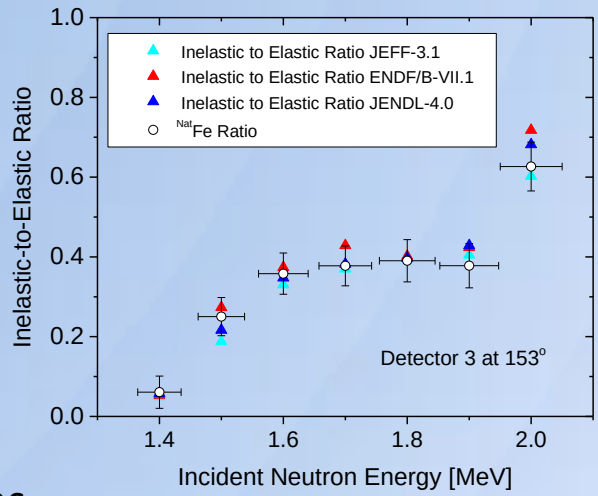


Inelastic to Elastic Ratio



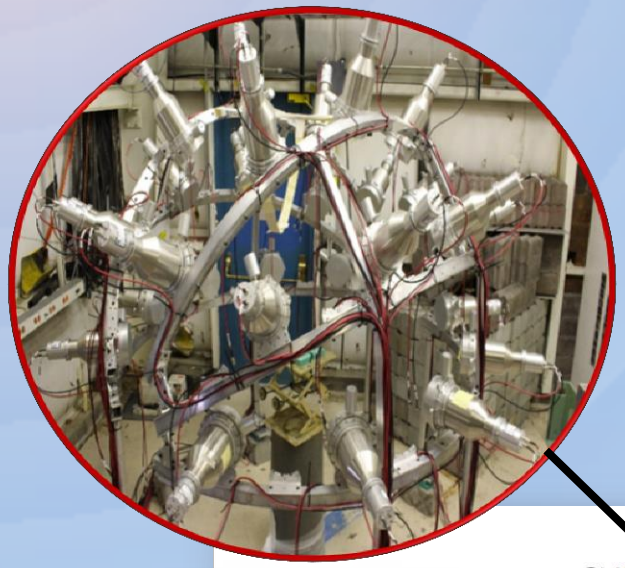




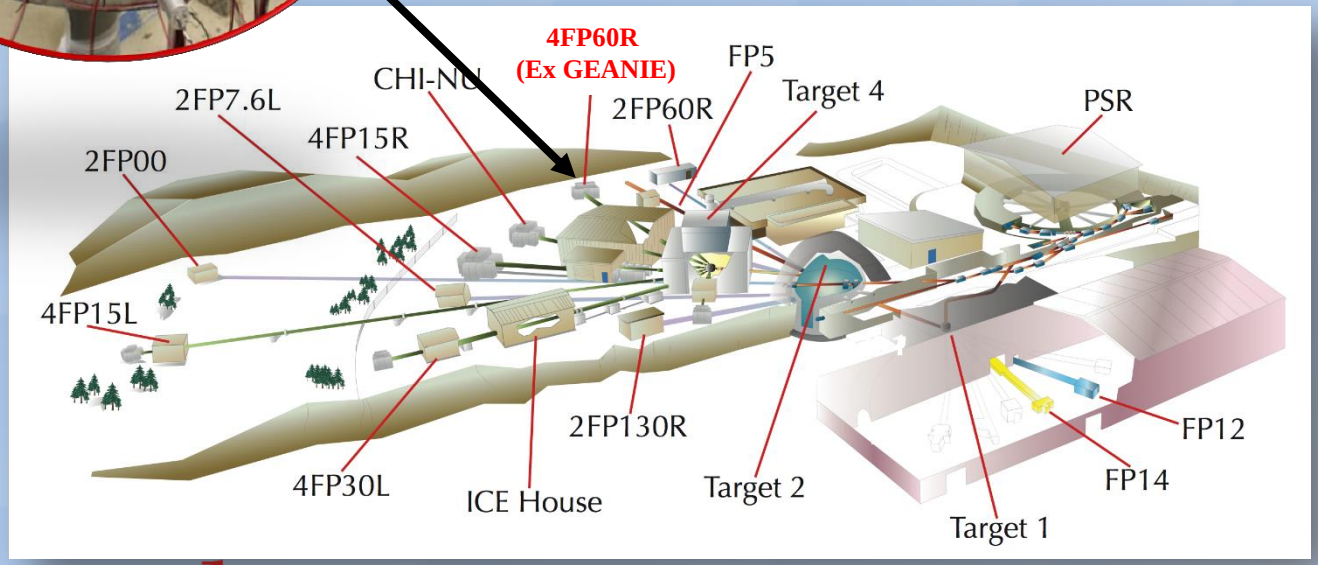


- Multiple scattering effects included in MCNP simulations
- Statistical and systematic uncertainties included in analysis

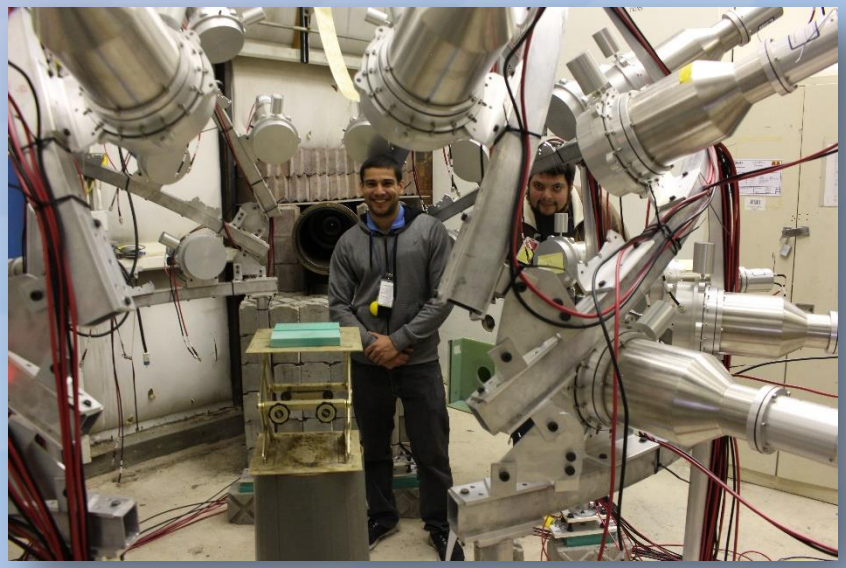
Experimental Setup at LANL



- Used the Chi Nu EJ-309 detector array
  - 56 detectors, arranged in 2 “quarter-spheres”
  - 9 detector slots per row starting at 30 degrees spaced 15 degrees apart spanning 150 degrees encompassing the sample
  - Rows 1 and 3 detectors at 30, 60, 90, 120, 150 degrees
  - Row 2 detectors at 45, 75, 105, 135 degrees
- Detectors were connected to digitizers
  - Pulse shape analysis using long and short gate
  - Full event pulse was also saved



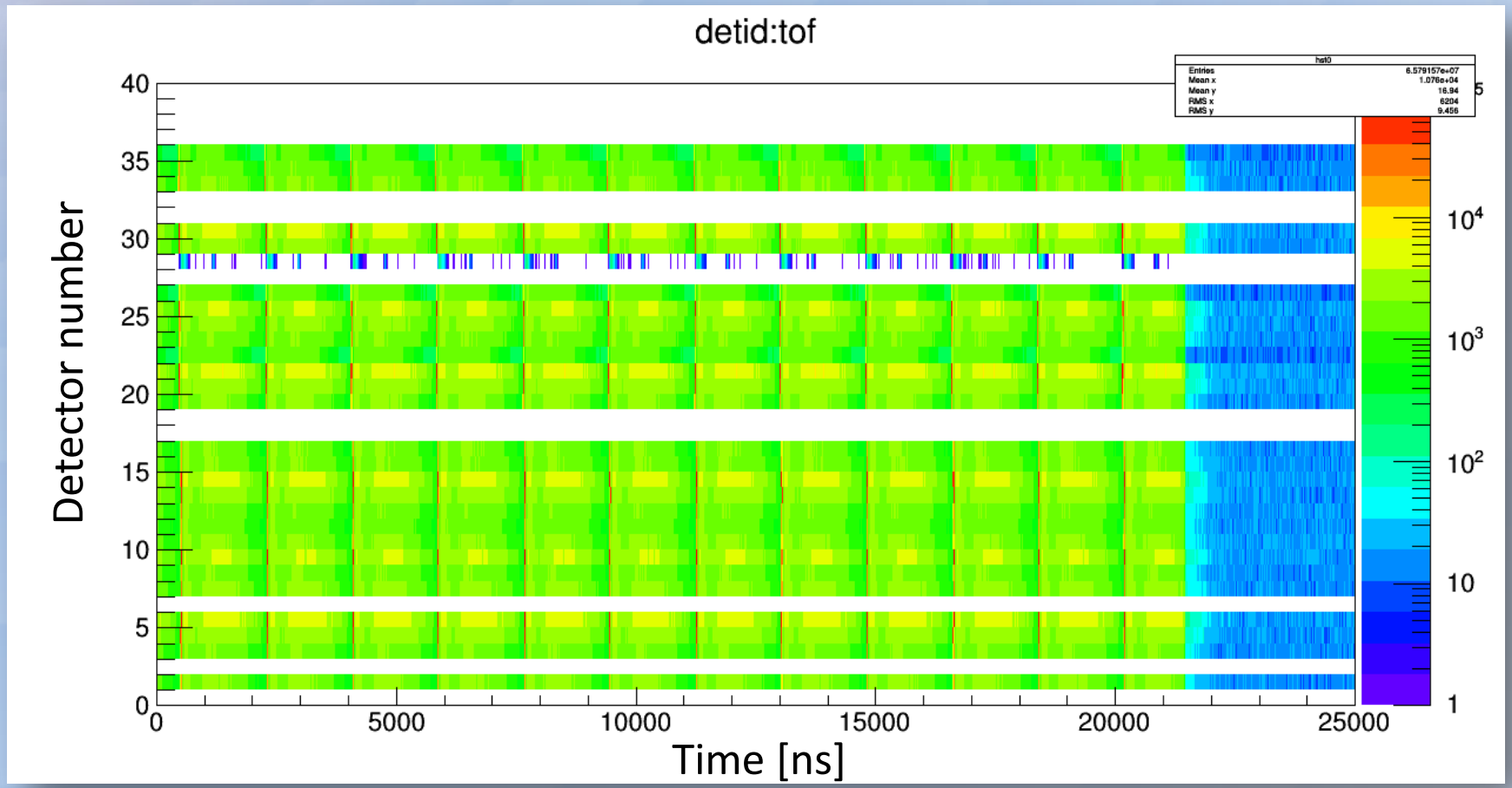
Carbon Scattering



- 56 EJ-301 17.8 cm in diam x 5 cm thick detectors, used only 28
- Used 3 cm and 7 cm graphite samples
- LANSCE operated at 100 Hz
  - 625 us micro-pulse
  - Micro-pulses spaced 1.8 us apart
  - Proton pulse width < 0.5 ns
- U-235 and U-238 fission chambers for flux monitoring
  - Malfunctioned some of the time
  - Run times fluctuate and intermittent beam conditions will require a greater depth of processing before the true quantity of data can be determined
- Used CAEN VX1730B Digitizers
  - 500 MHz sampling rate

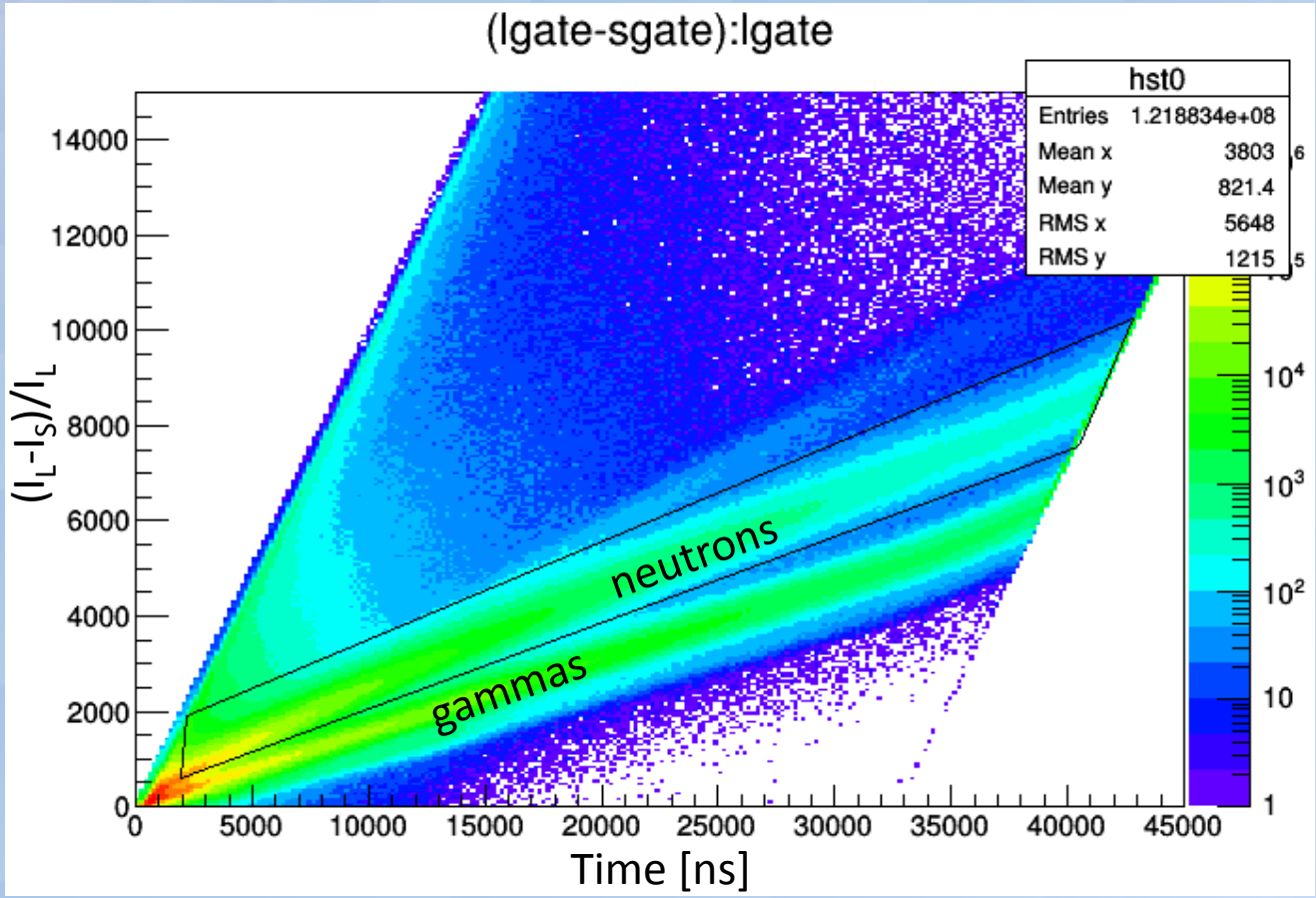
Preliminary Analysis Results

- A cut of the data: time-of-flight for each detector



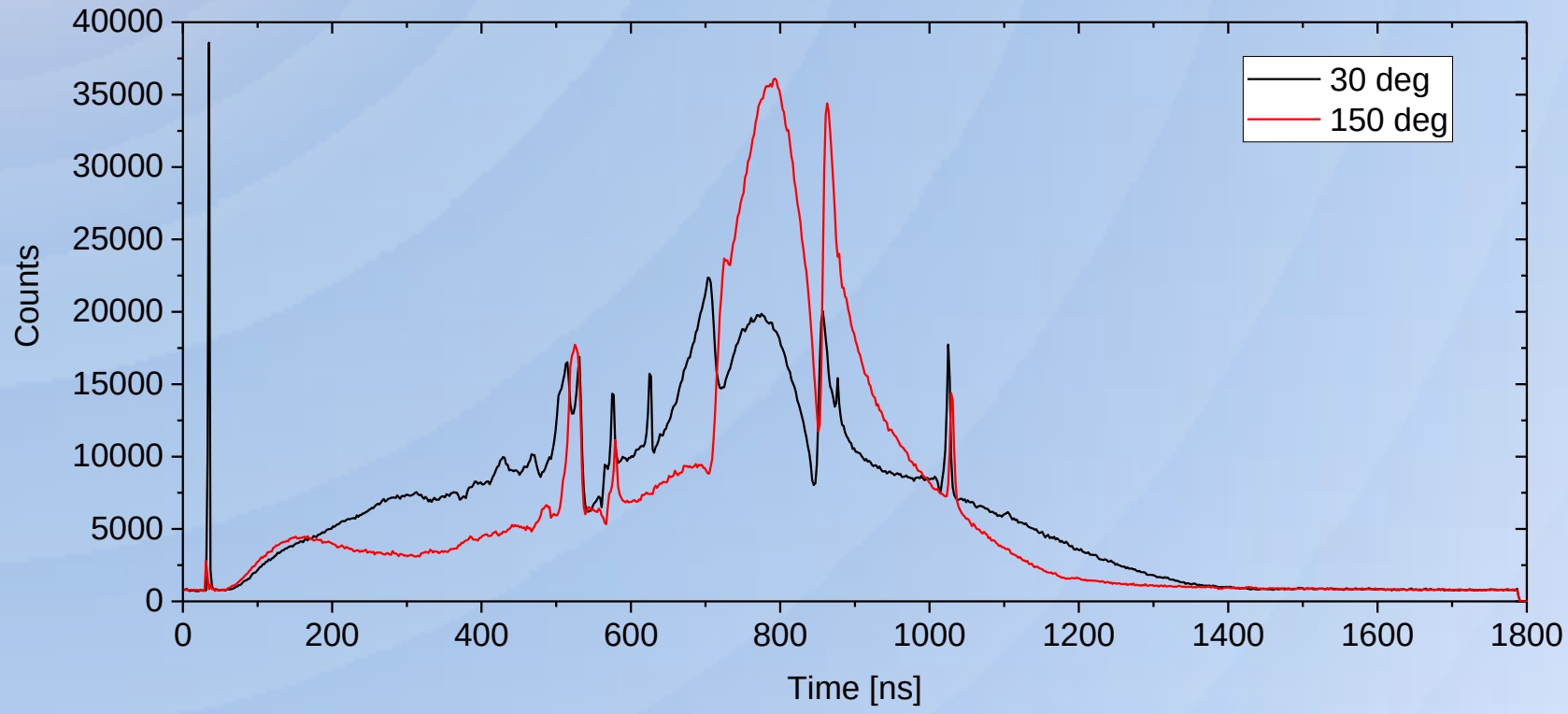
Pulse Shape Discrimination

- Use a long (I_L) and short (I_S) pulse integrals

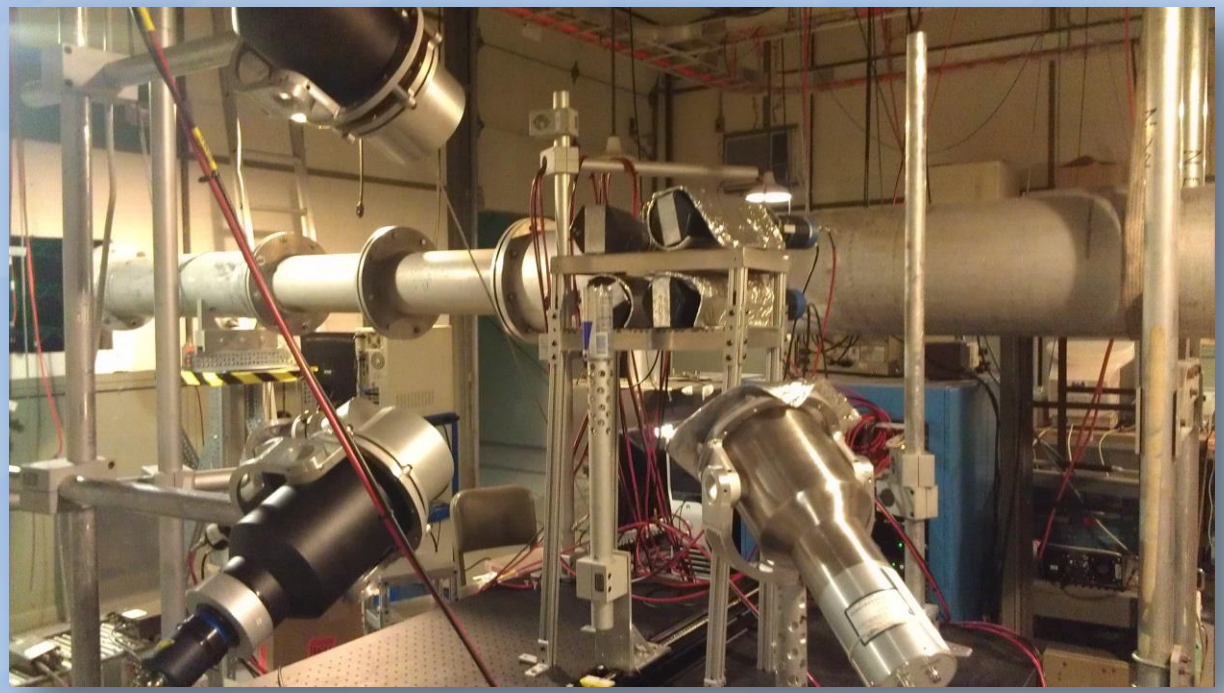


TOF for 7 cm Thick Graphite Sample

- TOF spectra show correct resonace structure of carbon
  - Good time resolution due to short pulse width (<0.5ns), 1 m sample-detector distance and thin (2”) detector.
- As expected the 150 deg data is delayed relative to 30 deg
- Working on MCNP simulations



Prompt Fission Neutron Spectra



Fission Spectrum Measurement Gamma Tagging

- Use the double TOF method
- Use a gamma tag for fission (instead of traditional fission chamber)
- Use a combination of Liquid Scintillators and Li-Glass neutron detectors

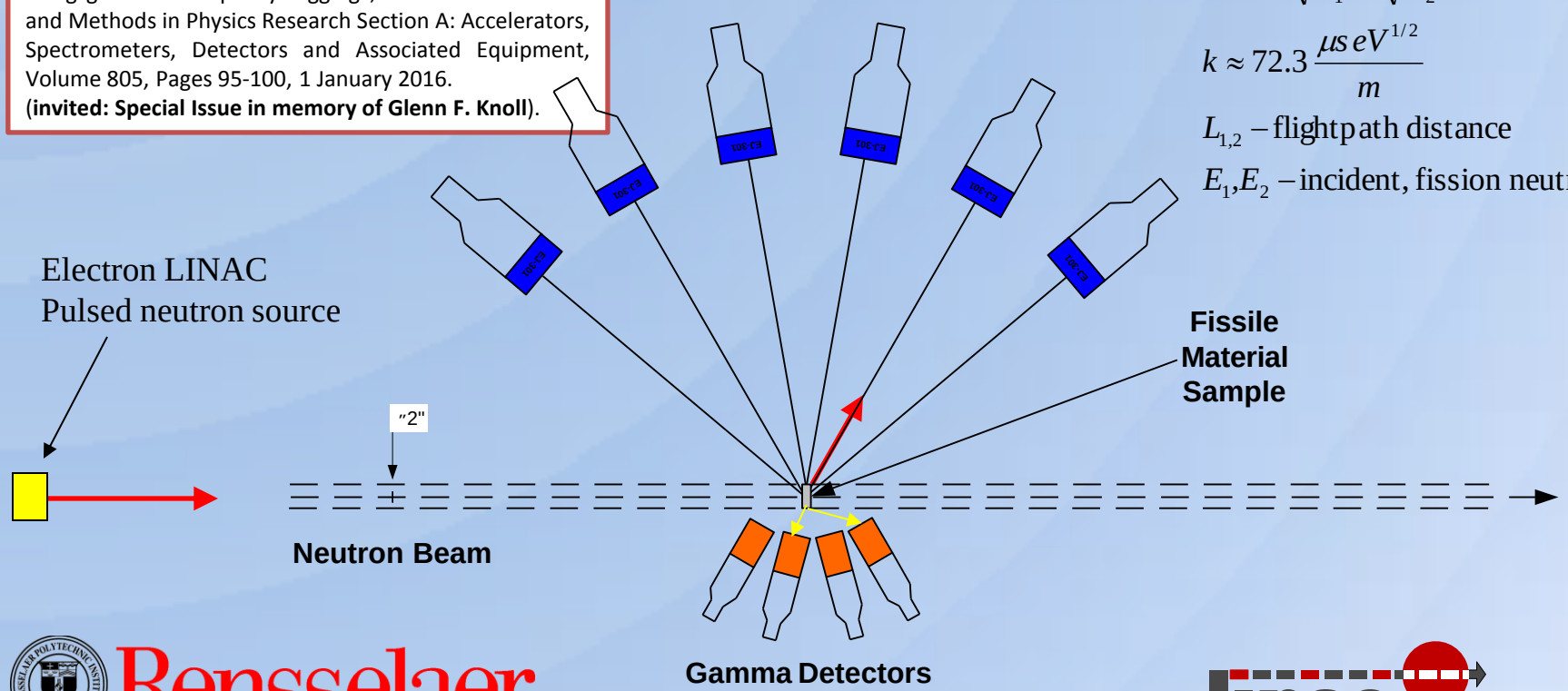
E. Blain, A. Daskalakis, R.C. Block, D. Barry, Y. Danon, "A method to measure prompt fission neutron spectrum using gamma multiplicity tagging", Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, Volume 805, Pages 95-100, 1 January 2016.
(**invited: Special Issue in memory of Glenn F. Knoll**).

$$ToF = \frac{kL_1}{\sqrt{E_1}} + \frac{kL_2}{\sqrt{E_2}}$$

$$k \approx 72.3 \frac{\mu s\, eV^{1/2}}{m}$$

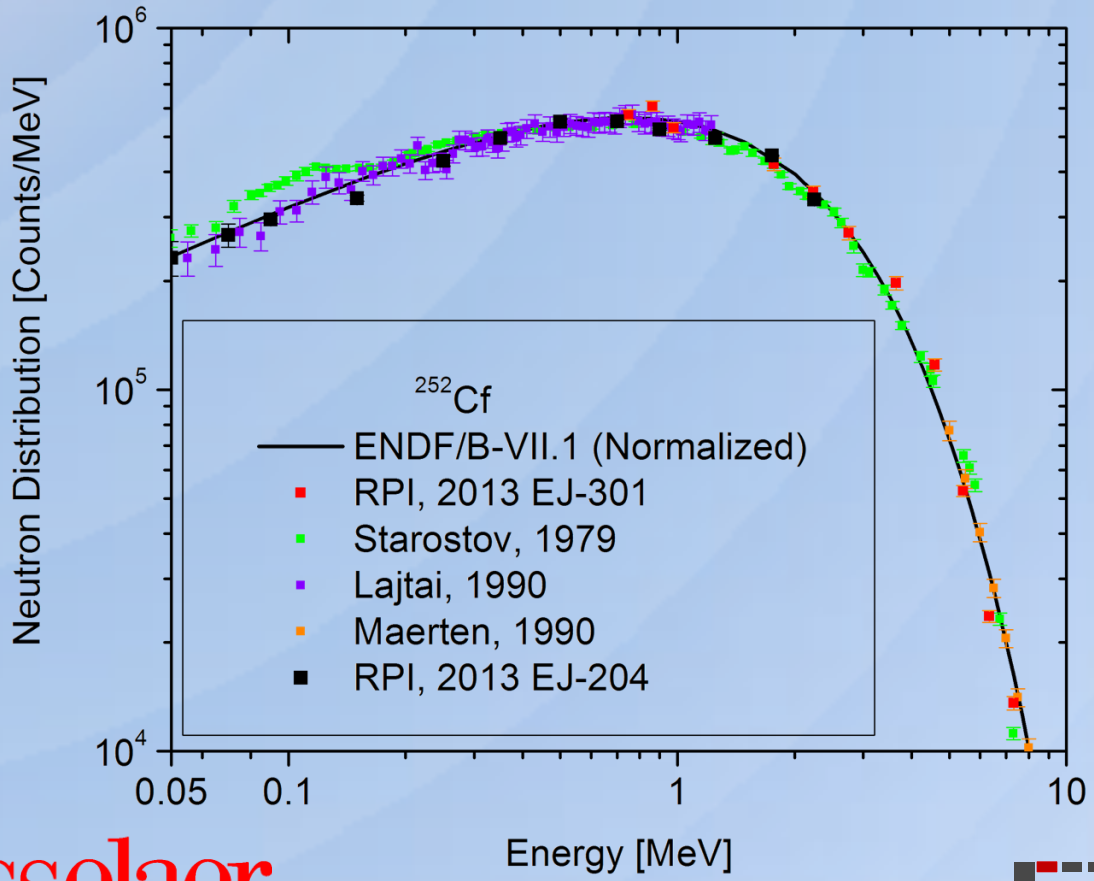
$L_{1,2}$ – flightpath distance

E_1, E_2 – incident, fission neutron energy



^{252}Cf Prompt Fission Neutron Spectrum

- Good agreement seen between the RPI datasets in the overlap region from 0.7 MeV to 2 MeV
- Combined datasets provide measurement from 0.05 Mev to 7 MeV



New Cf-252 Measurement for E>7 MeV

- Used a stronger Cf-252 source.
  - Encountered problems with high count rate and false gamma tagging from random coincidence.
- The experiment will be repeated:
  - Larger distance between the source and gamma detectors.
  - Additional shielding in front of the gamma detectors

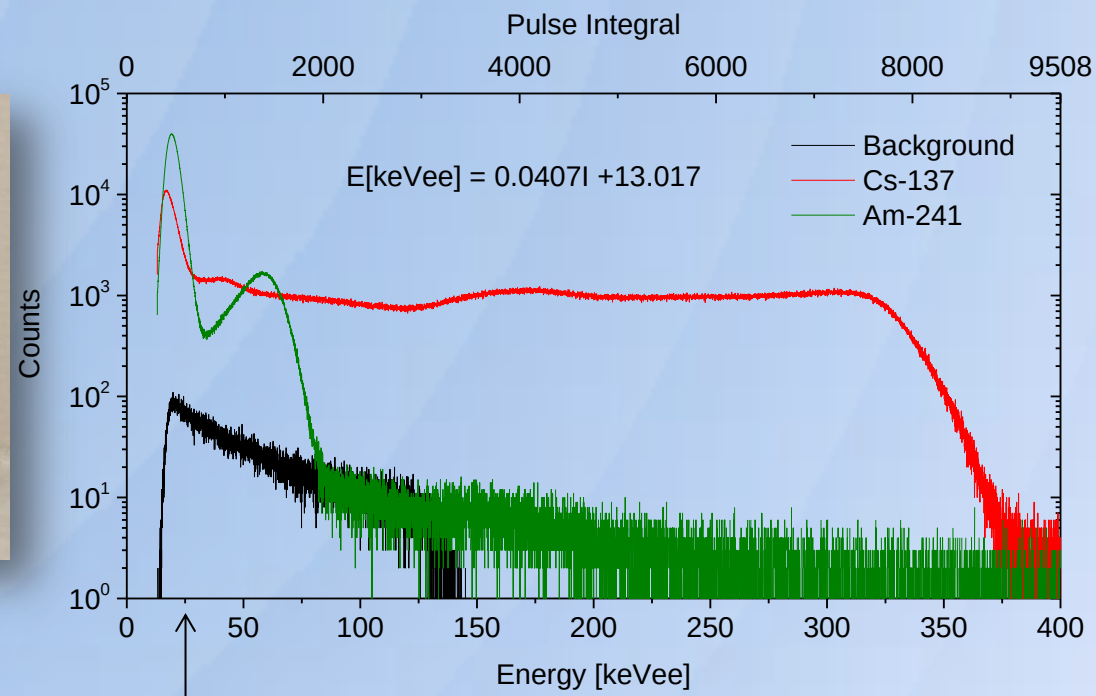
Neutron Detection Below 0.5 MeV

Need for a keV neutron detector

- Required for PFNS and neutron scattering.
- Liquid scintillators are usually used above 0.5 MeV where pulse shape discrimination effectively discriminates gammas.
- Thin plastic scintillator were used for PFNS measurents to 50 keV
  - It is gamma sensitivity and can be used when gamma background is low
- Need a detector which has low sensitivity to gammas, options considered:
  - Use Li-Glass – has low efficiency compared to liquid or plastic scintillators
  - CLYC with Li-6
    - Excellent gamma discrimination
    - Currently under testing CLYC has lower Li-6 concentration and thus the neutron detection efficiency is about 4-5 time lower than Li-Glass
  - **Thin (0.5”) liquid scintillator (EJ-301)**
    - **Need pulse shape analysis at lower neutron energies**

Pulse Shape Discrimination Below 500 keV

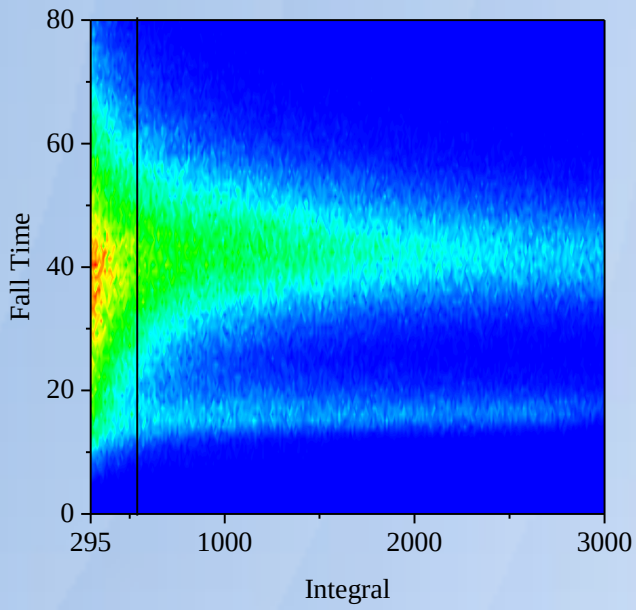
- There is very little work on PSD using liquid scintillation detectors below 500 keV. A thin (0.5”) EJ301 detector is being used here to extend PSD.

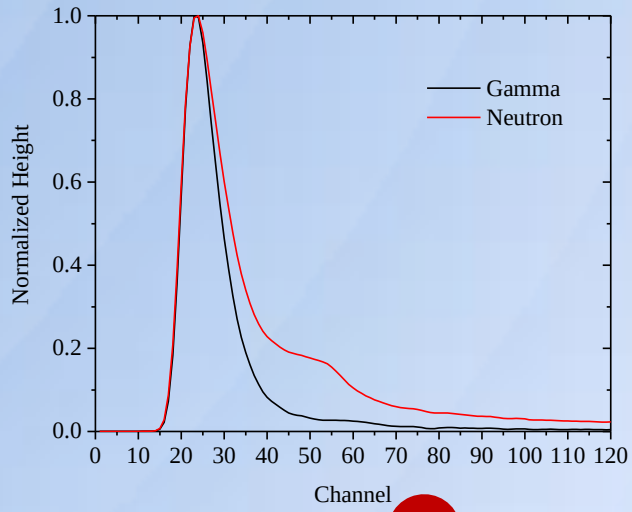


Targeting PSD down to ~25keVee
Corresponds to about 100 keV neutrons

Pulse Shape Discrimination Below 500 keV

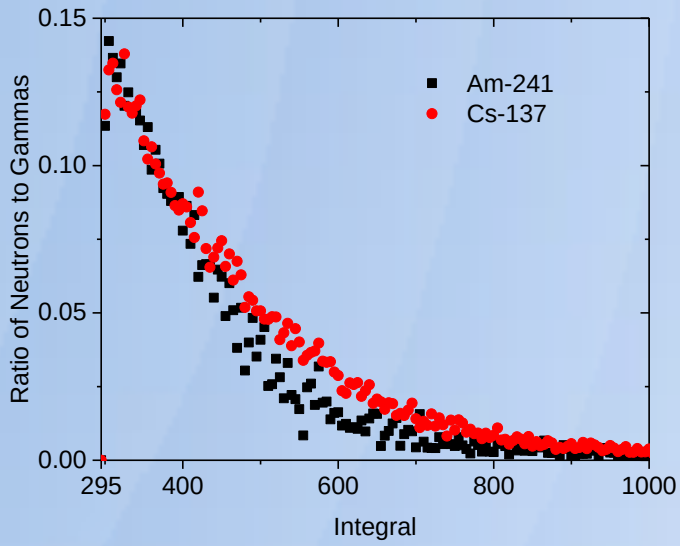
- The fall time and integral were used as a first cut between gammas and neutrons.
- For a ^{252}Cf source, there is visible separation down to an integral of 540 (35 keVee)
- Developed a classifier
  - Using the fall time cut, average pulse shapes were found for gammas and neutrons
  - Detector pulses are peak aligned and normalized. Chi-squared comparison is done between channels 40 and 60 to classify each pulse as either a neutron or gamma

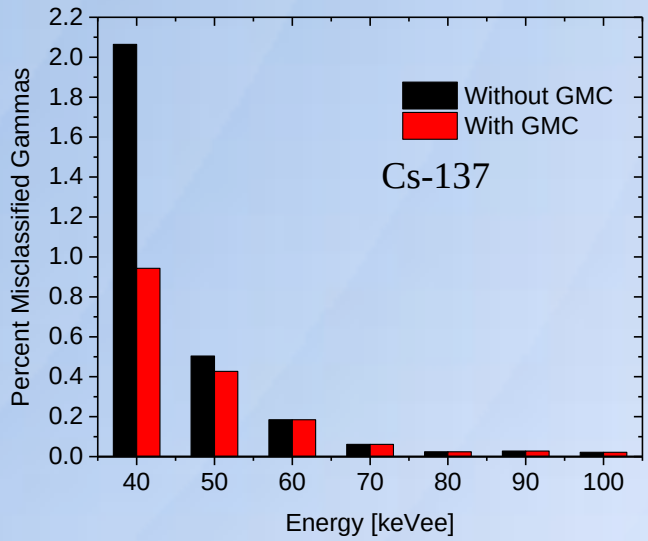




Pulse Shape Discrimination Below 500 keV

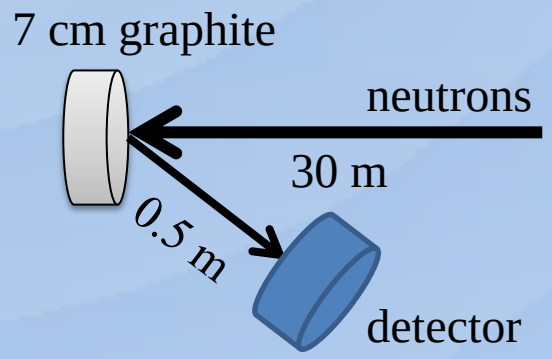
- At low energies, the pulses are small and are more likely to be misclassified
- For low amplitude pulses gammas can be misclassified as neutrons
  - Also neutrons can be classified as gammas, this will reduce the neutron detection efficiency
- Developed a gamma misclassification correction
  - Using an ^{241}Am gamma source, the misclassification was determined as a function of pulse integral.
  - As verification, the classification of a ^{137}Cs source was corrected between 35 keVee and 55 keVee

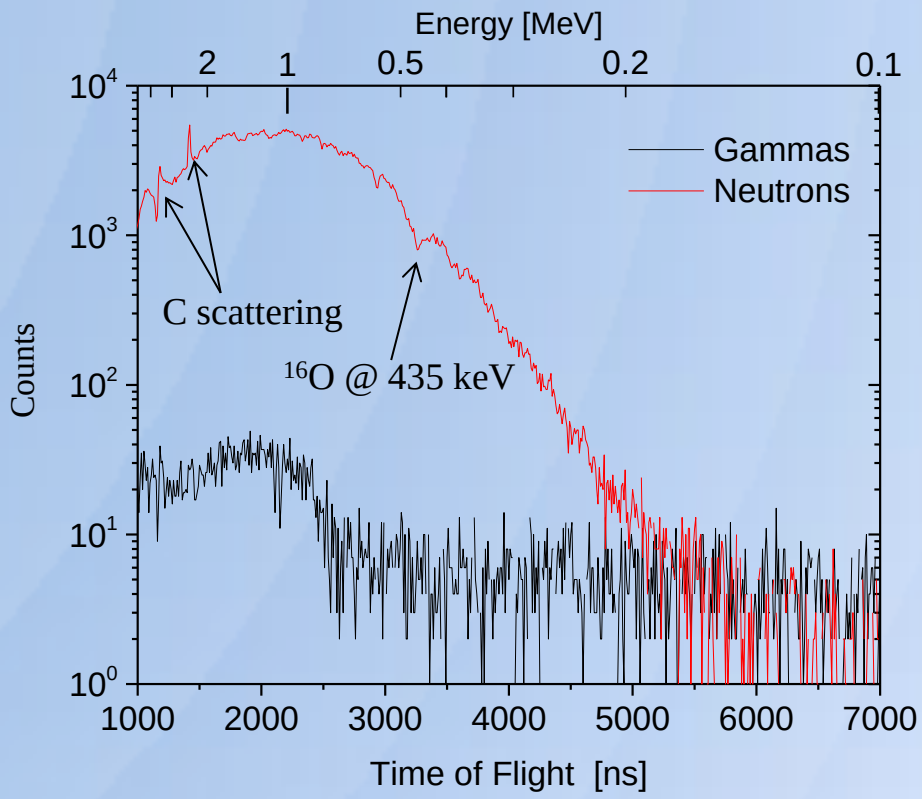




Pulse Shape Discrimination Below 500 keV

- A TOF back scattering from carbon demonstrate the Pulse classification. Preliminary results show neutron detection down to 150 keV





Summary

- Development of a capture detectors for the LSDS is in progress at RPI.
  - Demonstrated Capture measurements of several samples
  - Comparison of experiment and simulation indicates where improvement is needed
- Fast neutron scattering
  - Performed graphite scattering experiment at LANL using Chi-nu array
  - Preliminary results show good energy resolution
  - Need to complete characterization of the system
    - Neutron flux shape (use fission chamber)
    - Detector efficiency (use flux shape, in-beam measurements and MC simulations)
- Prompt fission neutron spectra
  - Gamma tagging was developed and used for Cf-252 and U-238
  - Additional measurement of Cf-252 for E>7 MeV are in progress.

Thank You