Experimental Fission Research at the Gaerttner LINAC Center at Rensselaer Polytechnic Institute

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LANL FIESTA Fission Workshop, Sep. 10 - 12, 2014

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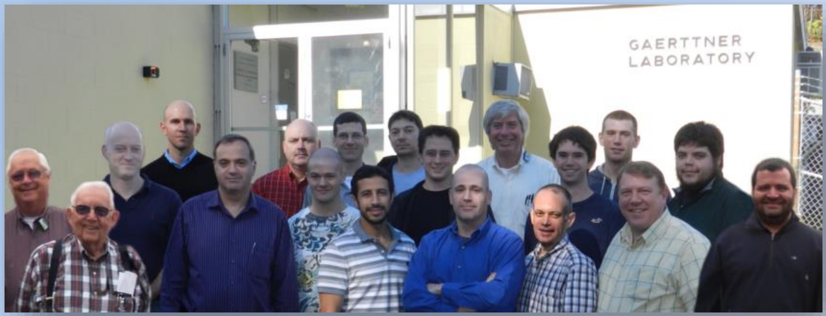
Collaboration (fission related)

- **RPI**
  - PI: Y. Danon, Professor, Director Gaerttner LINAC Center, Nuclear Engineering Program Director
  - Graduate Students: Z. Blain, N. Thompson, D. Williams, A. Daskalakis, B. McDermott, A. Youmans
  - Undergraduate students: K. Mohindroo, Amanda Lewis
- **KAPL**
  - Dr. T. Donovan, Dr. G. Leinweber, Dr. D. Barry, Dr. M. Rapp, Dr. R. Block, B. Epping
- **LANL (fission / LSDS)**
  - Dr. R. Haight, Dr. P. Talou
- **ORNL (sample for LSDS measurements)**
  - Dr. Romano



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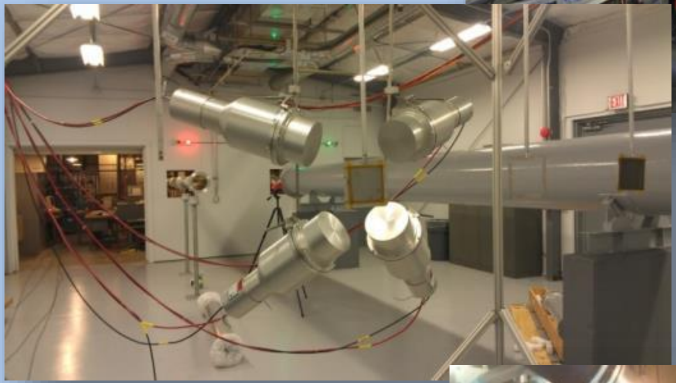
The Nuclear Data Program at the Gaerttner LINAC Center at RPI

- Driven by a 60 MeV pulsed electron LINAC ~ $\mathbf{10^{13}}$ **n/s**
- **Neutron transmission**
  - Resonance region: 0.001 eV- 1000 keV,
  - High energy region: 0.4- 20 MeV
- **Neutron Capture**
  - Resonance region: 0.01-1000 eV
- **Neutron Scattering**
  - High energy region: 0.4 MeV- 20 MeV
- **LSDS**
  - Assay of used nuclear fuel
- 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
- **Started a major refurbishment project (~$10M)**



Focus on

- **Historical Perspective**
- **Lead Slowing Down Spectrometer (LSDS)**
  - Simultaneous measurements of fission cross section and fission fragment mass and energy distributions of small samples.
- **Time-of-Flight and gamma tagging**
  - Measurements of fission neutron spectra (also < 1 MeV) as a function of the incident neutron energy using a gamma tag.
  - Fission cross section using a gamma tag.

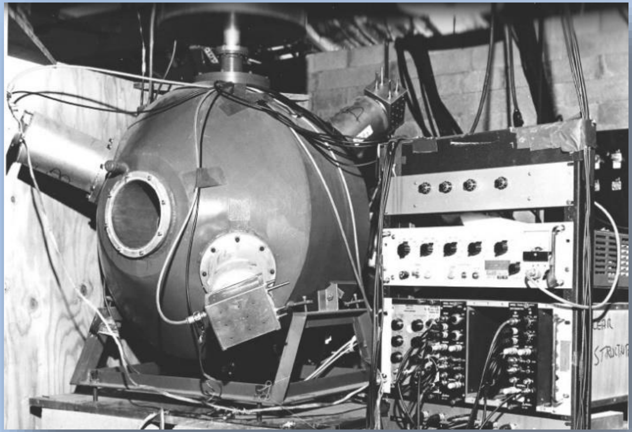
Early Fission Related Work at RPI
Cross Section Measurements and PFN multiplicity

- ^{232}Th
  - Nakagome, Y., Block, R.C., Slovacek, R.E. and Bean, E.B., "Neutron-Induced Fission Cross Section of **^{232}Th** from 1 eV to 20 keV," Physical Review C, Vol. 43, No. 4, April, 1991.
- ^{233}U
  - Weston, L.W., Gwin, R., de Saussure, G., Ingle, R.W., Todd, J.H., Craven, C.W., Hockenbury, R.W. and Block, R.C., "Neutron Fission and Capture Cross-section Measurements for **Uranium-233** in the Energy Region 0.02 to 1 eV," Nuclear Science and Engineering, 42, 143-149, 1970.
- ^{235}U
  - Kaushal, N.N., Malaviay, B.K., Becker, M., Burns, E.T. and Gaerttner, E.R., "Measurement and Analysis of Fast Neutron Spectra in Uranium Depleted in the Uranium-235 Isotope," Nuclear Science and Engineering, Vol. 49, n 3, p. 330-48, November, 1972.
- ^{238}U
  - Slovacek, R.E., Cramer, D.S., Bean, E.B., Valentine, J.R., Hockenbury, R.W. and Block, R.C., "^{238}U(n,f) Measurements Below 10 keV," Nuclear Science and Engineering, 62, 455-462, 1977.
- ^{239,240,241}Pu
  - Weinstein, S. and Block, R.C., "Neutron Multiplicity-Spin State Correlations for ^{239}Pu Resonances," Physical Review Letters, Vol. 22, No. 5, 195-198, February, 1969.
  - Gwin, R., Weston, L.W., Saussure de, G., Ingle, R.W., Todd, J.H., Gillespie, F.E., Hockenbury, R.W. and Block, R.C., "Simultaneous Measurement of the Neutron Fission and Absorption Cross Sections of Plutonium-239 Over the Energy Region 0.02 eV to 30 keV," Nuclear Science and Engineering, 45, 25-36, 1971.
  - Hockenbury, R.W., Moyer, W.R. and Block, R.C., "Neutron Capture, Fission, and Total Cross Sections of **Plutonium-240** from 20 eV to 30 keV," Nuclear Science and Engineering, 49, 153-161, 1972.
  - M. S. Moore, O. D. Simpson, and T. Watanabe, J. E. Russell and R. W. Hockenbury, Fission Cross Section of **Pu-241**, Phys. Rev. 135, B945–B952 (1964).
- ^{244,246,247,248}Cm, ^{254}Es
  - Maguire, Jr., H.T., Stopa, C.R.S., Block, R.C., Harris, D.R., Slovacek, R.E., Dabbs, J.W.T., Dougan, R.J., Hoff, R.W. and Lougheed, R.W., "Neutron-Induced Fission Cross-Section Measurements of ^{244}Cm, ^{246}Cm and ^{248}Cm," Nuclear Science and Engineering, 89, 293-304, 1985.
  - Danon, Y., Slovacek, R.E., Block, R.C., Lougheed, R.W., Hoff, R.W. Moore, M.S., "Fission Cross-Section Measurements of **^{247}Cm, ^{254}Es and ^{250}Cf** from 0.1 eV to 80 keV," Nuclear Science and Engineering, 109, 341-349, 1991.

RPI Old Gd Loaded Scintillator -1969

- S. Weinstein, R. Reed, R.C. Block ,Neutron Multiplicity Measurements for ^{233}U, ^{235}U and ^{239}Pu Resonance Fission, Second IAEA Symposium on Physics and Chemistry of fission,IAEA-SM-122/113, 1969.
- Measured neutron multiplicity in a short slowing down time window following a fission event.

Gd loaded Scintillator tank



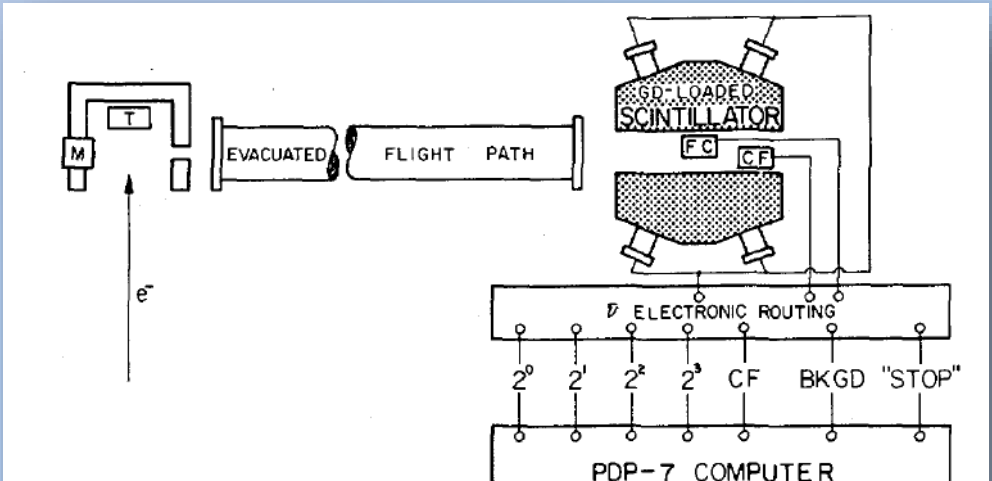
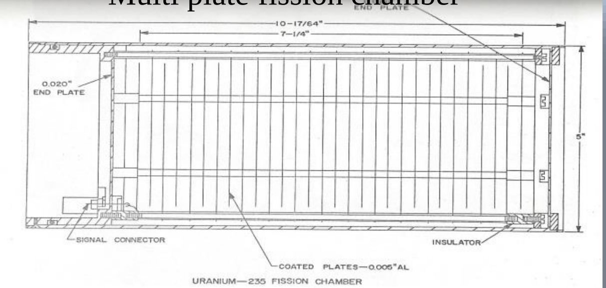


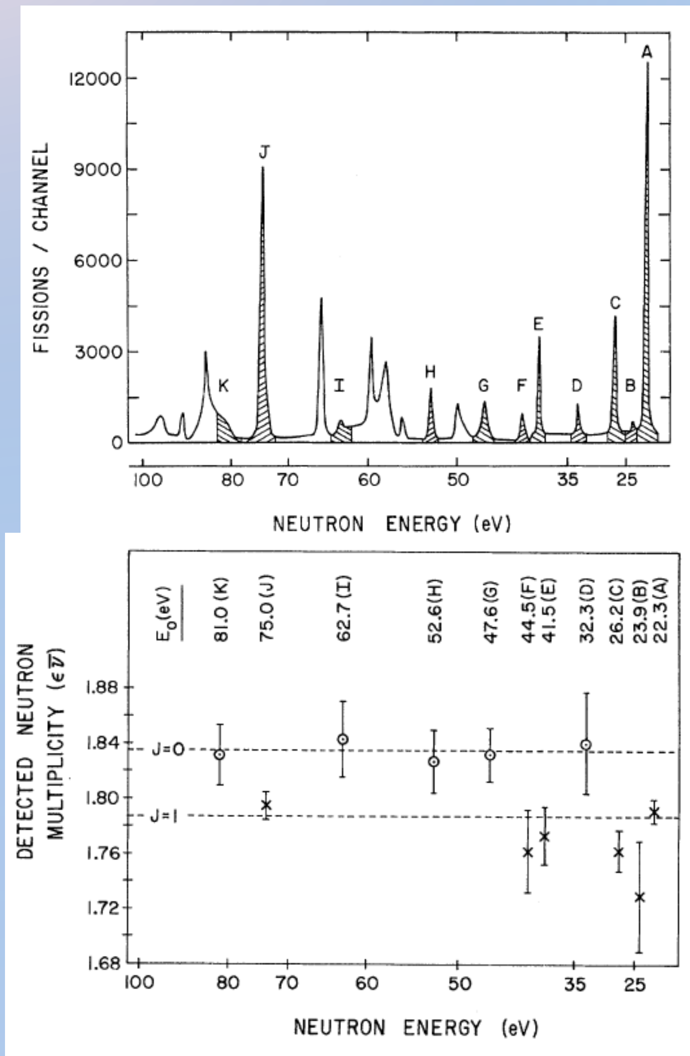
FIG. 1. Major components of experiment.

Multi plate fission chamber



Schematic Diagram - ^{235}U Fission Chamber
Figure 2.5

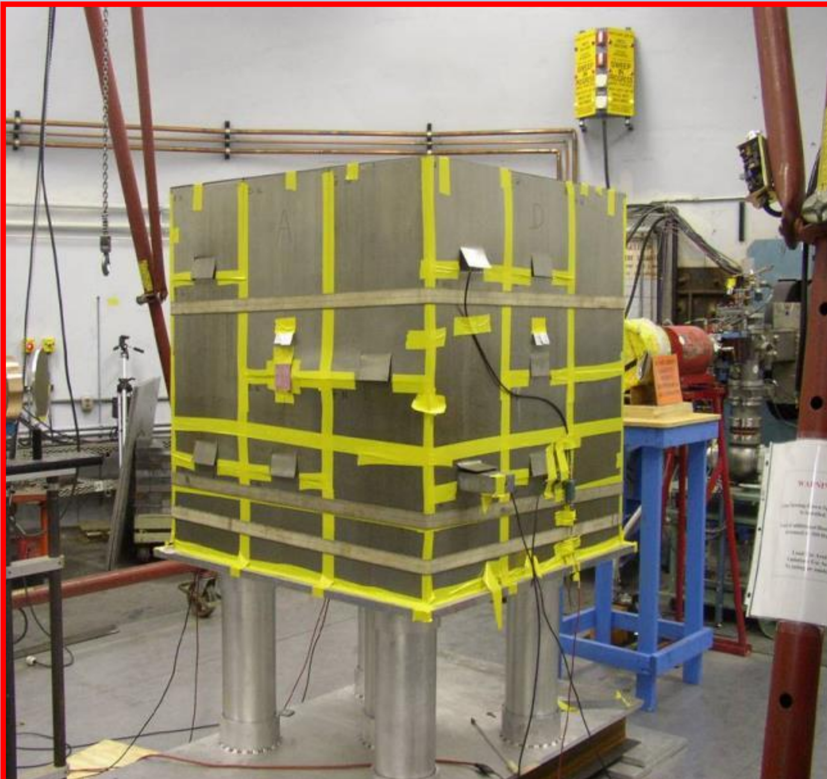
Results - 1969



- Observed fluctuations in ^{239}Pu neutron multiplicity were used to infer resonance spin states.

Weinstein, S. and Block, R.C., "Neutron Multiplicity-Spin State Correlations for ^{239}Pu Resonances," Physical Review Letters, Vol. 22, No. 5, 195-198, February, 1969.

Lead Slowing Down Spectrometers in the US



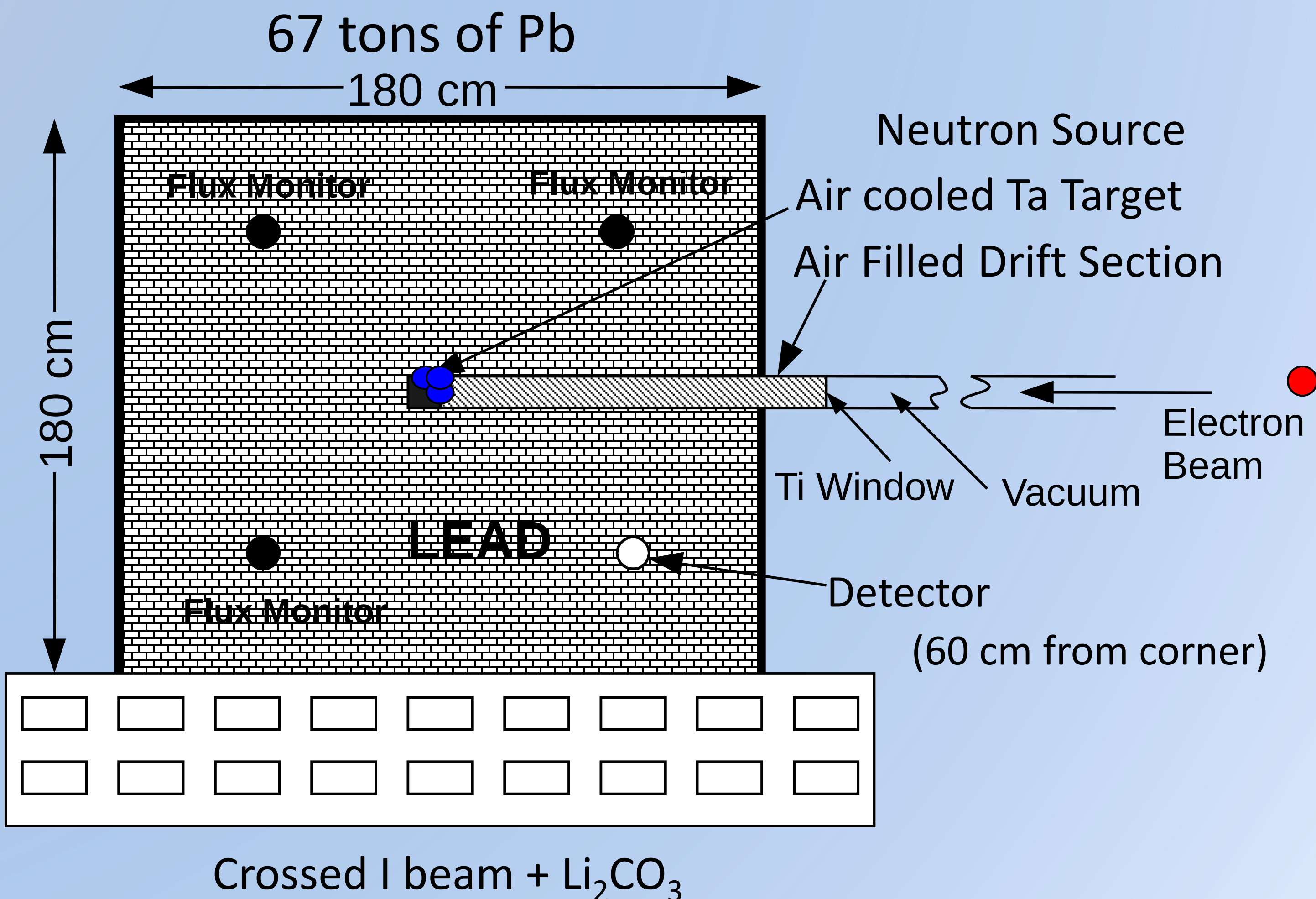
***LANL** – Proton Driven*

***RPI** –Electron Driven*

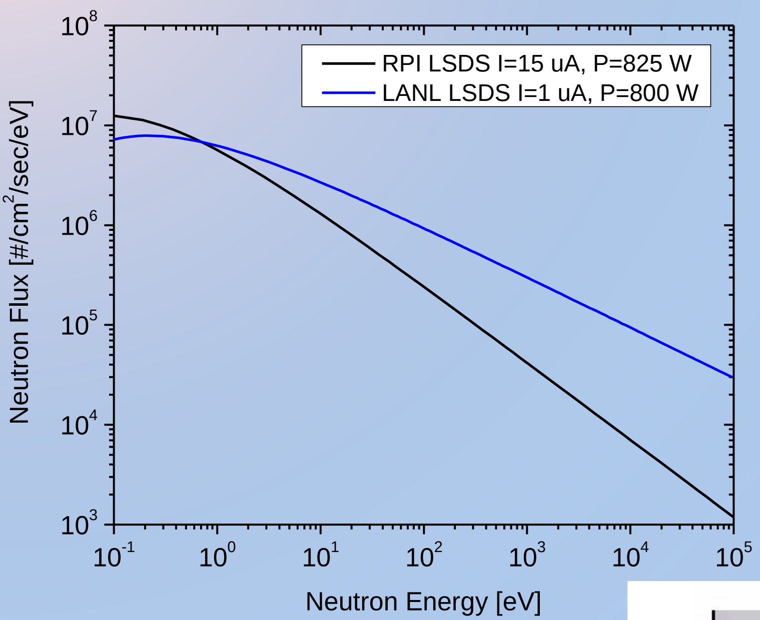
Why Use a Lead Slowing-down Spectrometer

Showing - Lead Slowing-down Spectrometer at RPI

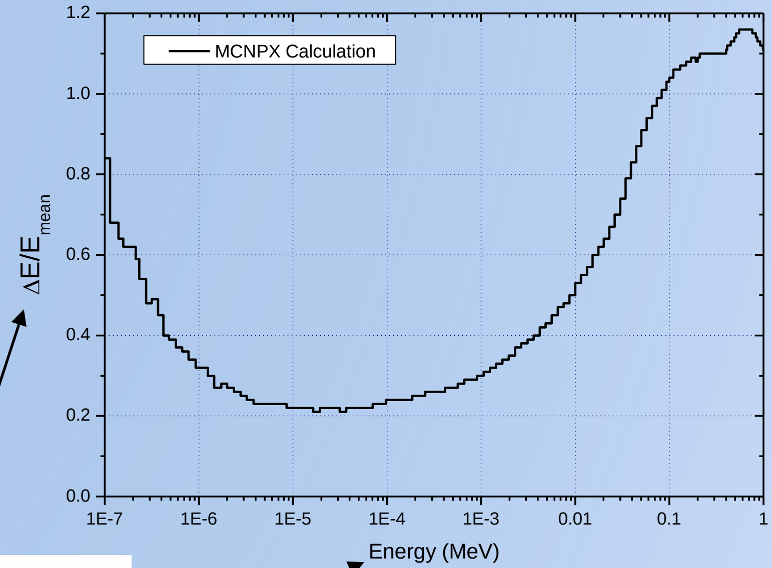
- Tantalum target in the center produces neutrons.
- Neutrons scatter elastically with the Pb.
- Neutrons can pass through the same position several times.
- **About 10^3-10^4 times higher flux than an equivalent neutron TOF experiment.**



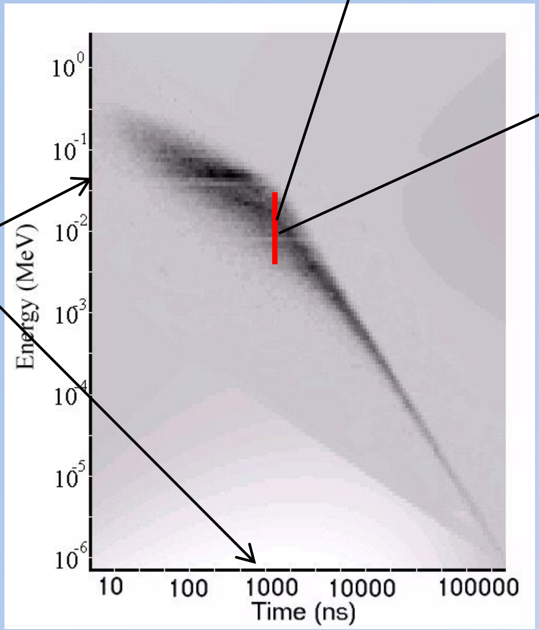
Slowing -Down-Time vs. Energy Relation



LANL LSDS



$$\frac{\Delta E}{\overline{E}}$$



$$\overline{E}$$

For E_n>50 KeV detector recovery of <1 μs is required

$$\overline{E} = \frac{K}{(t+t_0)^2}$$

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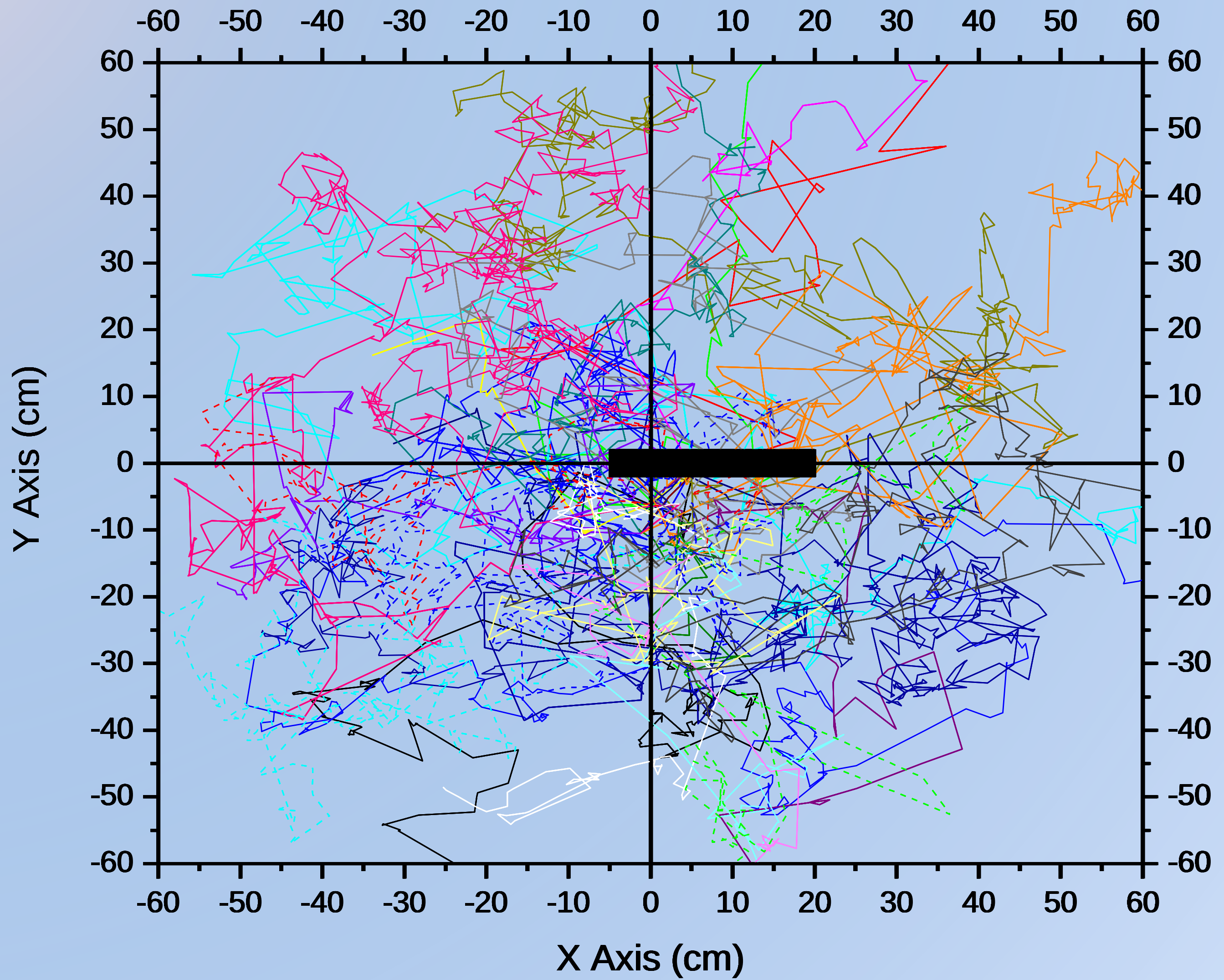
MCNPX Calculations LANL LSDS 1 proton



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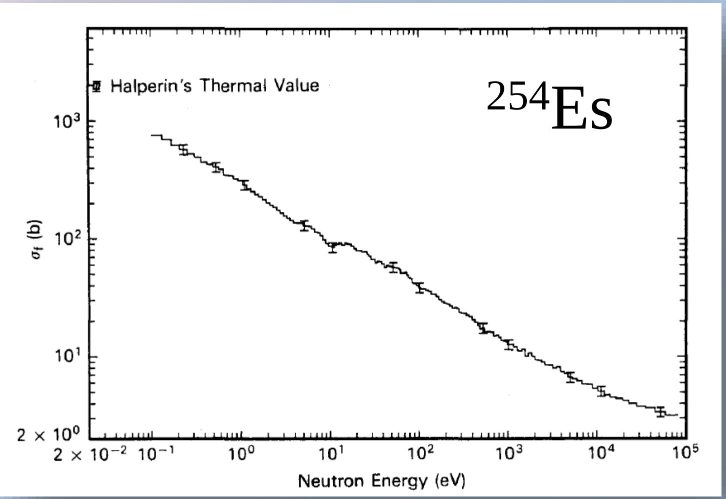
LSDS Applications

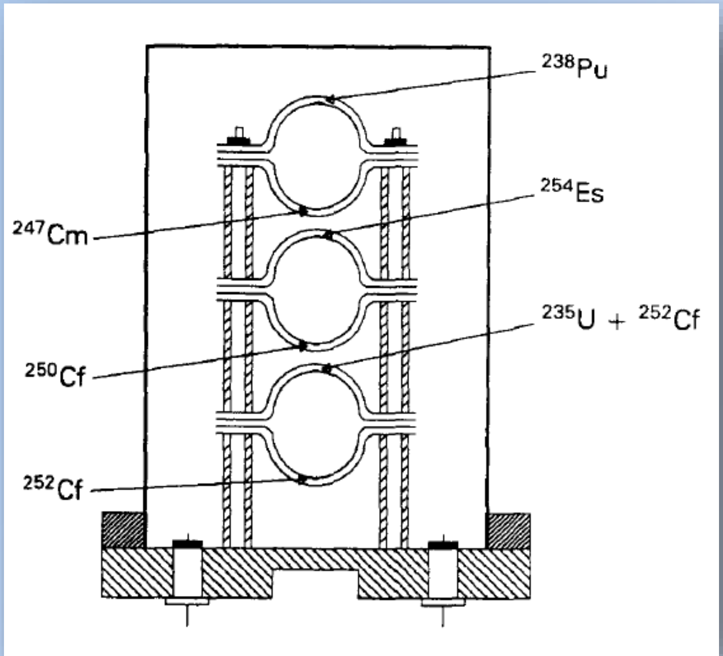
- Assay of use nuclear fuel
  - The slowing down spectrum induces fission in the fissile material such as ^{235}U and ^{239}Pu
  - To first order the reaction rate as a function of time is a linear combination of the of U and Pu content.
  - Use threshold fission detector with ^{238}U (indicated some problems in the sub threshold fission of ^{238}U).
- Fission cross section measurements
- Fission fragment spectroscopy
- (n,α) and (n,p) cross section measurements
- Capture cross section measurements

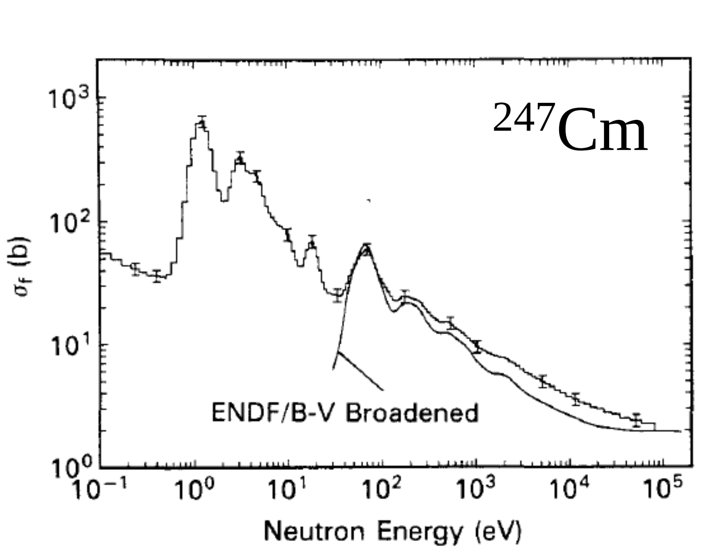
The LSDS is a high flux environment which requires appropriate detector development

LSDS - Fission cross section measurement example



Hemispherical fission ion chamber reduces alpha particle pileup

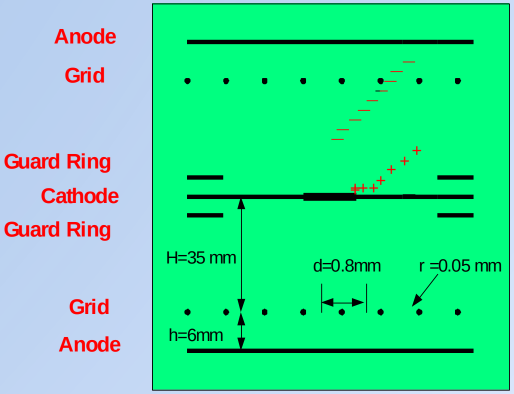




Danon, Y., Slovacek, R.E., Block, R.C., Lougheed, R.W., Hoff, R.W. Moore, M.S., "Fission Cross-Section Measurements of ^{247}Cm, ^{254}Es and ^{250}Cf from 0.1 eV to 80 keV," Nuclear Science and Engineering, 109, 341-349, 1991.

Simultaneous Measurements of Fission Cross Section and Fission Fragment Mass and Energy Distributions of Small Samples

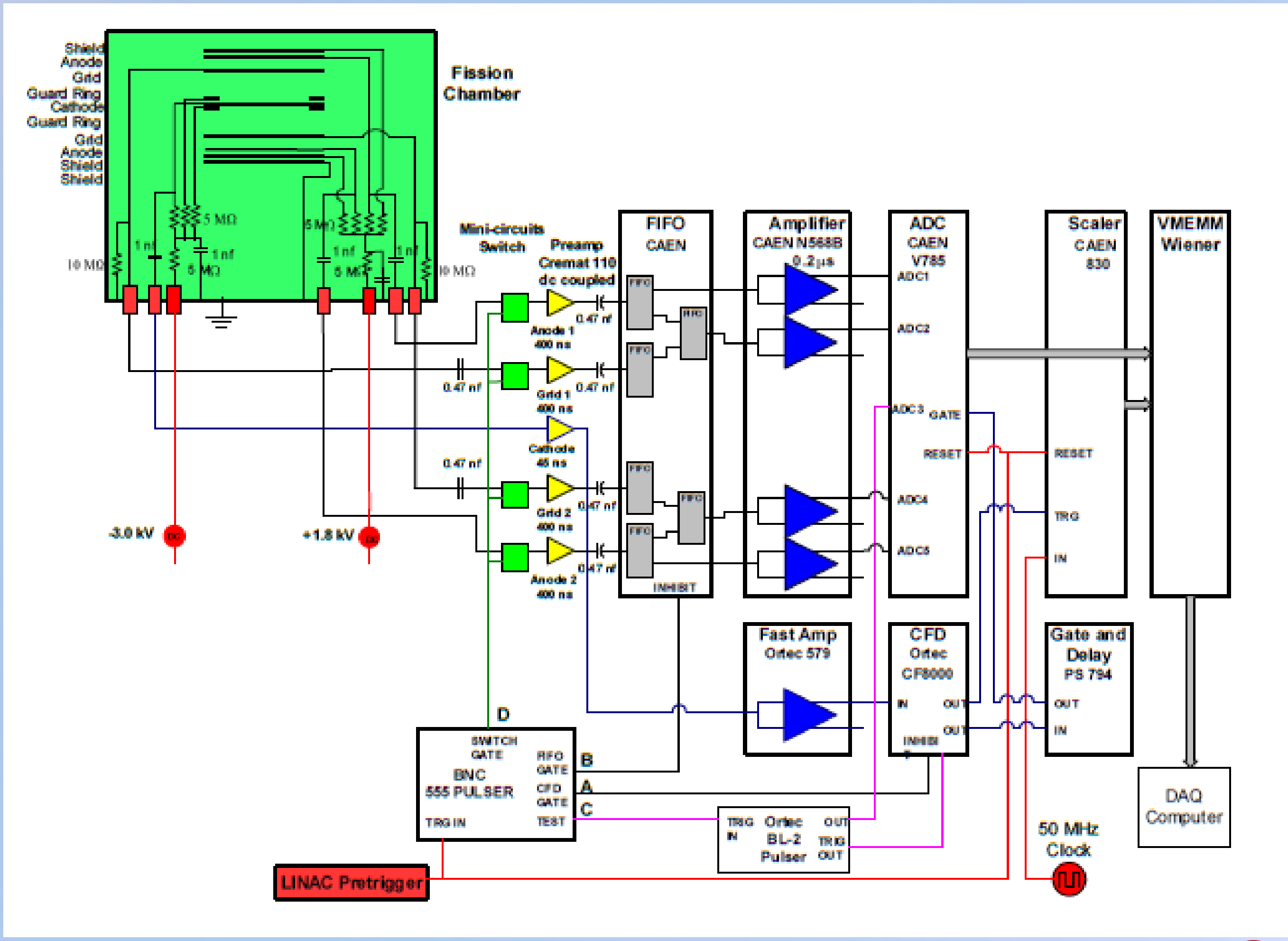
- Use a double gridded fission chamber with the RPI LSDS
- Use kinematics to compute
  - Fragment angle relative to the normal to the sample
  - Fragment energy distribution
  - Fragment mass distribution
- Future improvements
  - Improve by converting to digital electronics (next slide)
    - Enables more flexibility in the data analysis
- Advantages
  - Low mass samples <40μg)
  - High data throughput (<8h for above mass)



C. Romano, Y. Danon, R. Block, J. Thompson, E. Blain, E. Bond, "Fission Fragment Mass And Energy Distributions As A Function of Neutron Energy Measured In A Lead Slowing Down Spectrometer", Phys. Rev. C, 81, 014607 (2010).

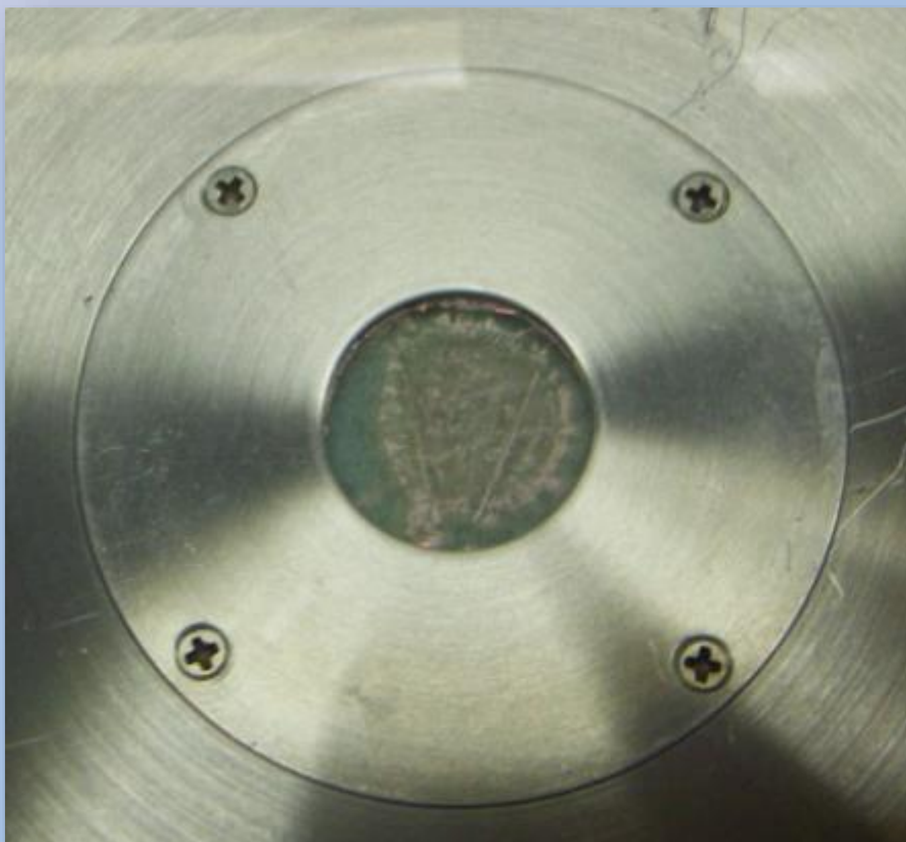
Electronics and DAQ

- VME based system with preamplifiers near the LSDS



Thin Samples

34 mg sample of ^{239}Pu made at INL



Sample Frame

- 2500 A thick polyimide
- 1.5 cm aperture
- Made by Luxel Corporation

Actinide Sample is made by dissolving actinide in acid and dropping measured amounts on the film

- **25 µg** ^{235}U sample made at LANL
- 0.41 ng ^{252}Cf made at RPI

Fragment Mass and Energy

Iterative procedure to find the preneutron and postneutron mass and energy

Initial guess:

$$m_{T_i}^* = m_{B_i}^* = A/2$$

$$m_{T_i} = m_{T_i}^* - \nu(m_{T_i}^*)$$
$$m_{B_i} = m_{B_i}^* - \nu(m_{B_i}^*)$$

$$E_{T_i} = E_{T_i} + PHD(m_{T_i})$$
$$E_{B_i} = E_{B_i} + PHD(m_{B_i})$$

$$m_{T_i}^* = \frac{E_{B_i}}{E_{T_i}\left[\frac{m_{T_i}^* m_{B_i}}{m_{B_i} m_{T_i}^*}\right] + E_{B_i}} A$$

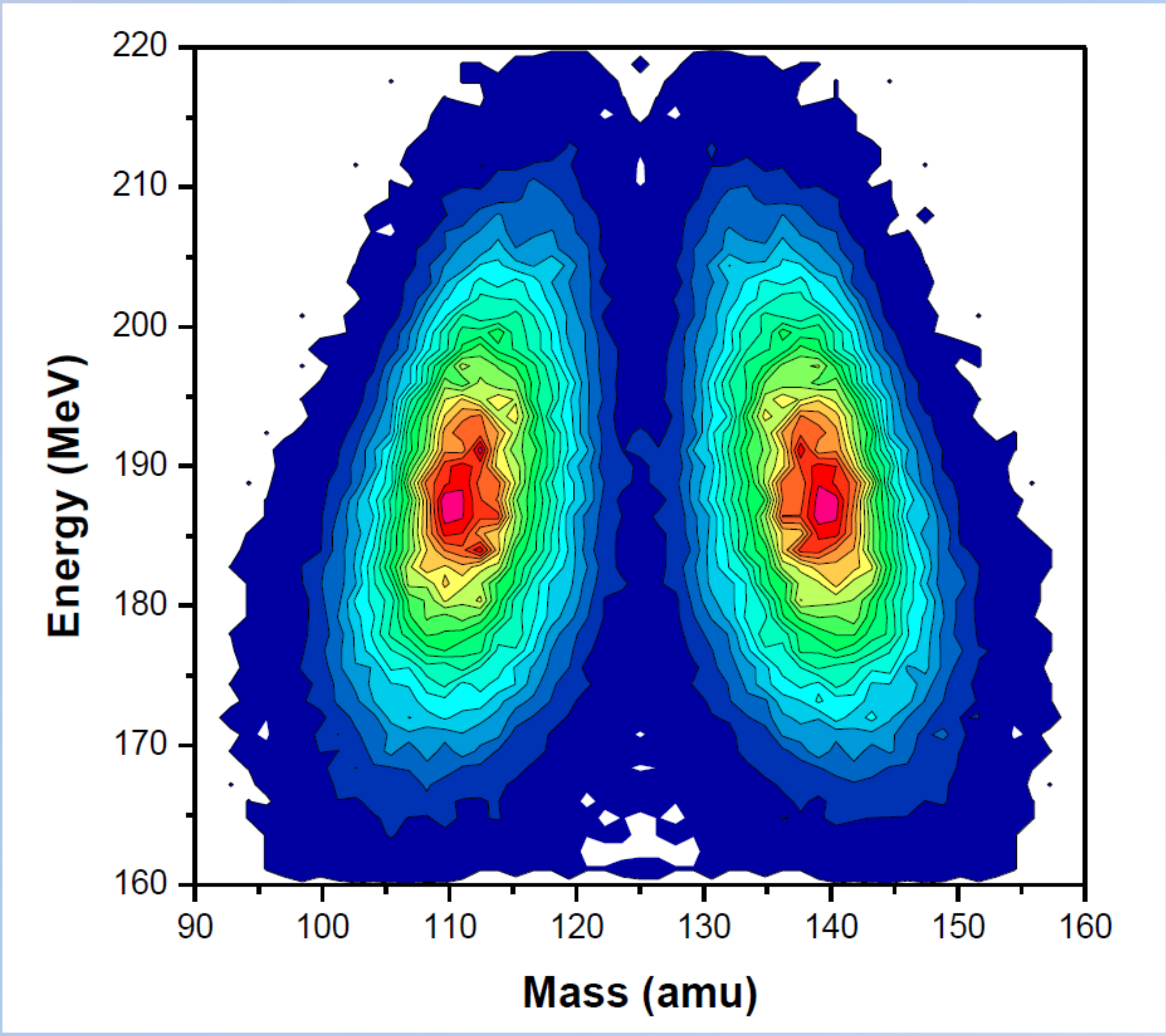
$$m_{B_i}^* = \frac{E_{T_i}}{E_{B_i}\left[\frac{m_{B_i}^* m_{T_i}}{m_{T_i} m_{B_i}^*}\right] + E_{T_i}} A$$

$$E_{T_i}^* = \frac{m_{T_i}^*}{m_{T_i}} E_{T_i} \qquad E_{B_i}^* = \frac{m_{B_i}^*}{m_{B_i}} E_{B_i}$$

$$if \quad m_{T_i}^* = m_{T_{i-1}}^* \pm 0.25 \quad and \quad m_{B_i} = m_{B_{i-1}} \pm 0.25 \quad then\ done$$

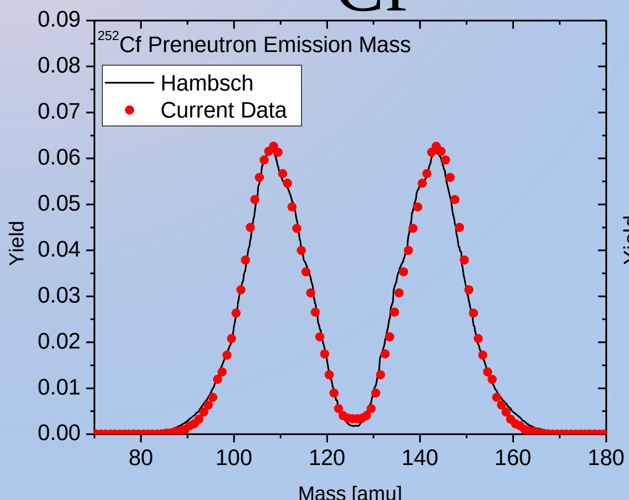
F.J. Hambsch, H.H. Knitter and C. Budtz-Jorgensen, Fission Mode Fluctuations in the Resonances of 235U(n,f), Nuclear Physics A491 (1989) 56-90

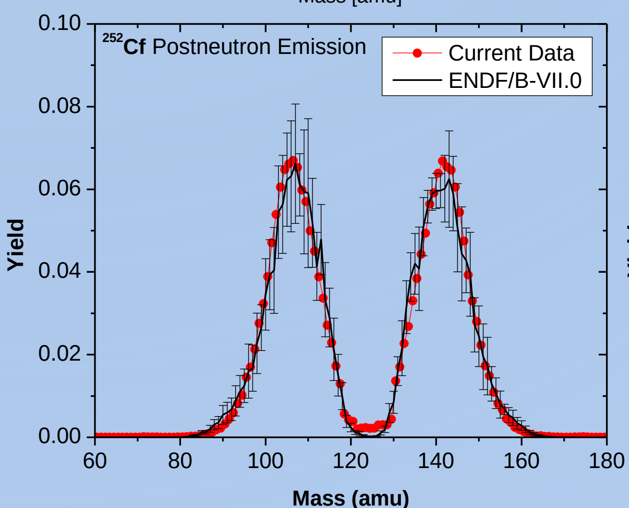
^{252}Cf Contour Plot of TKE vs Fragment Mass



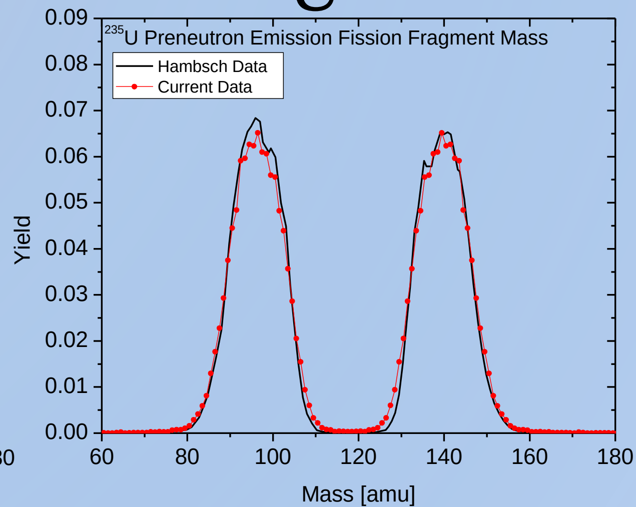
Results – Fission Fragment Mass distribution E_n<0.1 eV

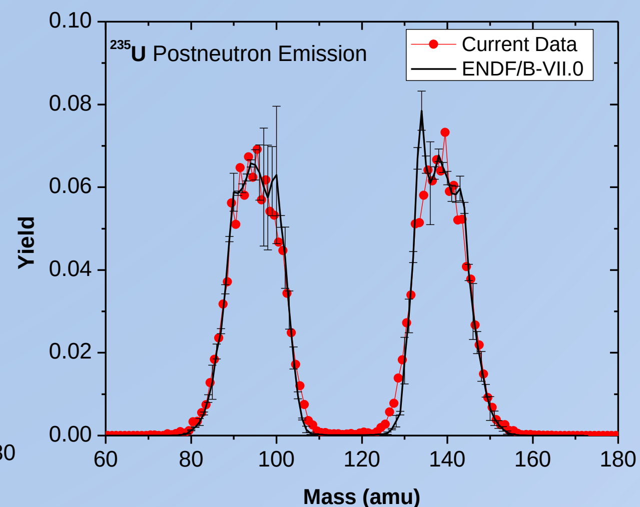
^{252}Cf



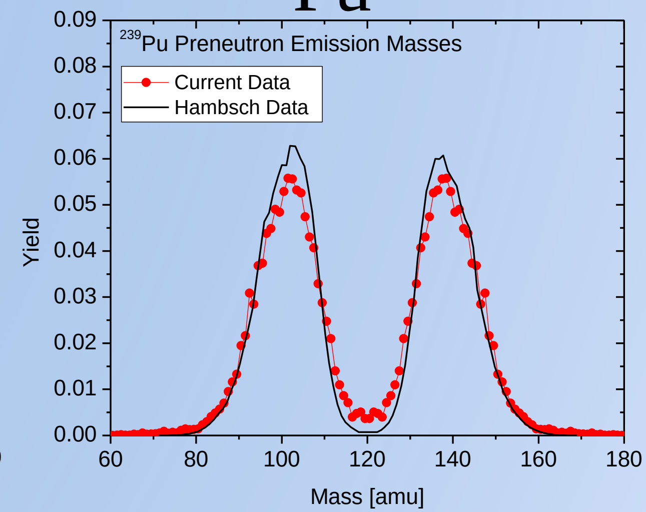


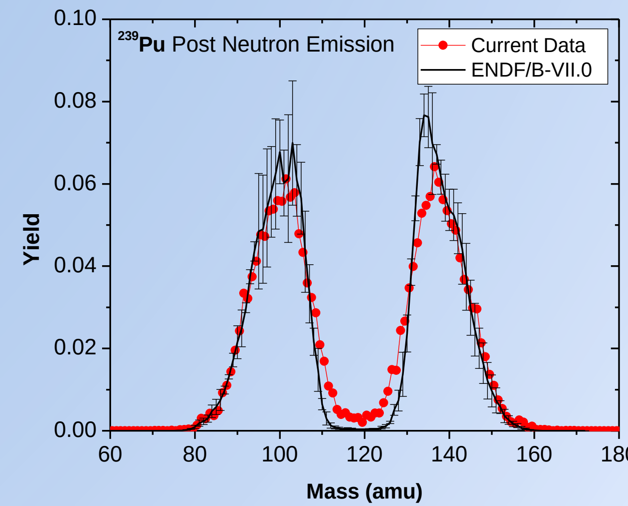
^{235}U





^{239}Pu





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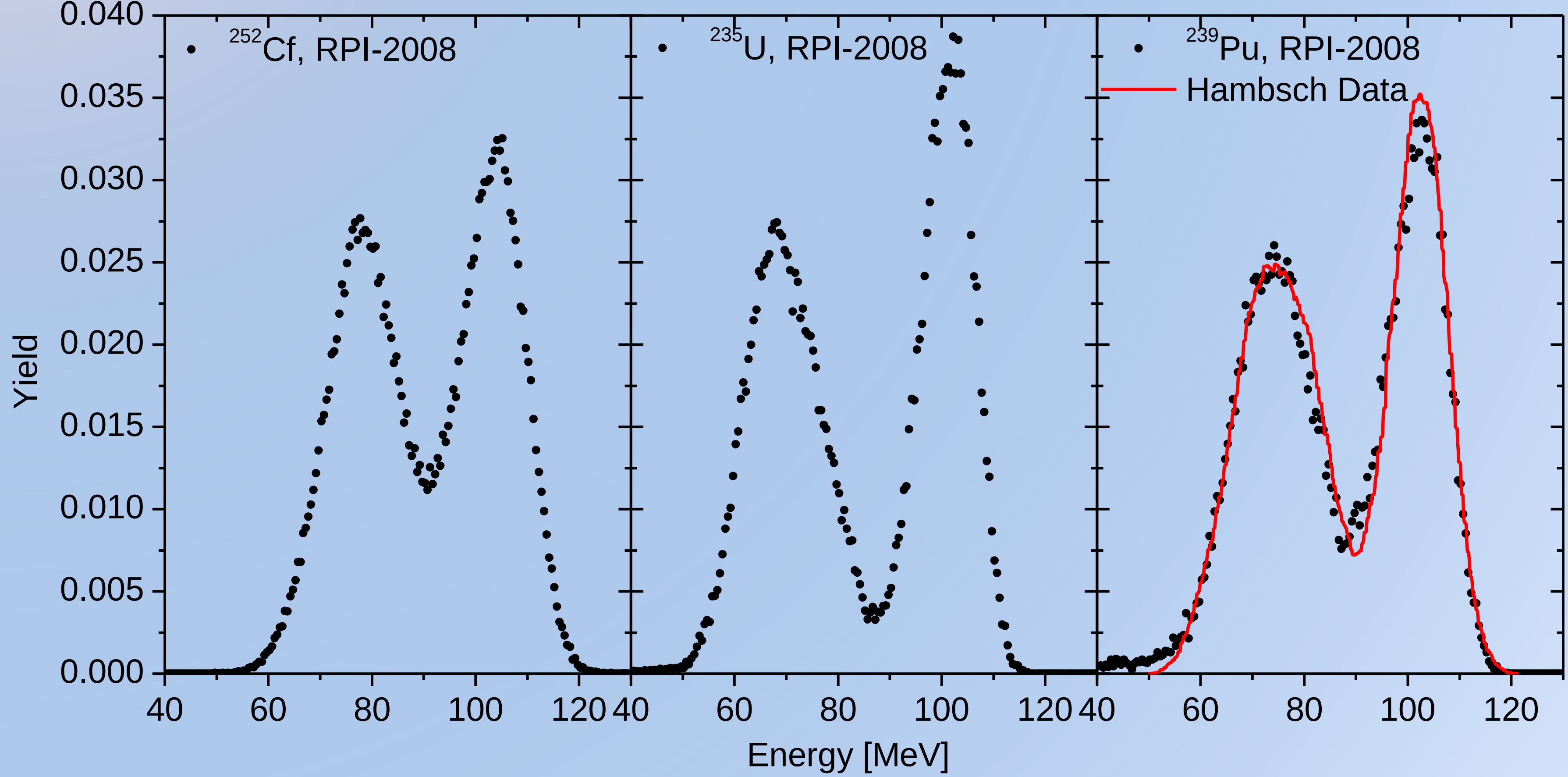
Results – Fission Fragment Energy Distribution E_n<0.1 eV



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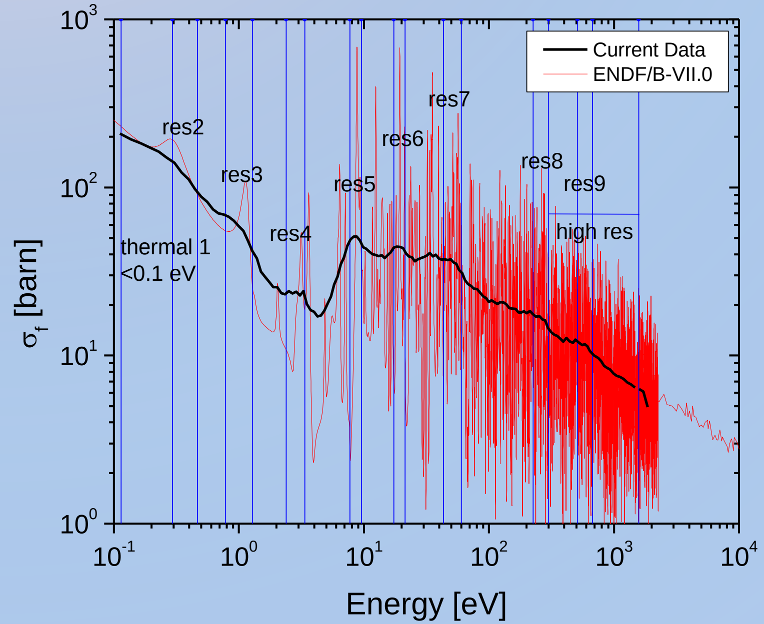
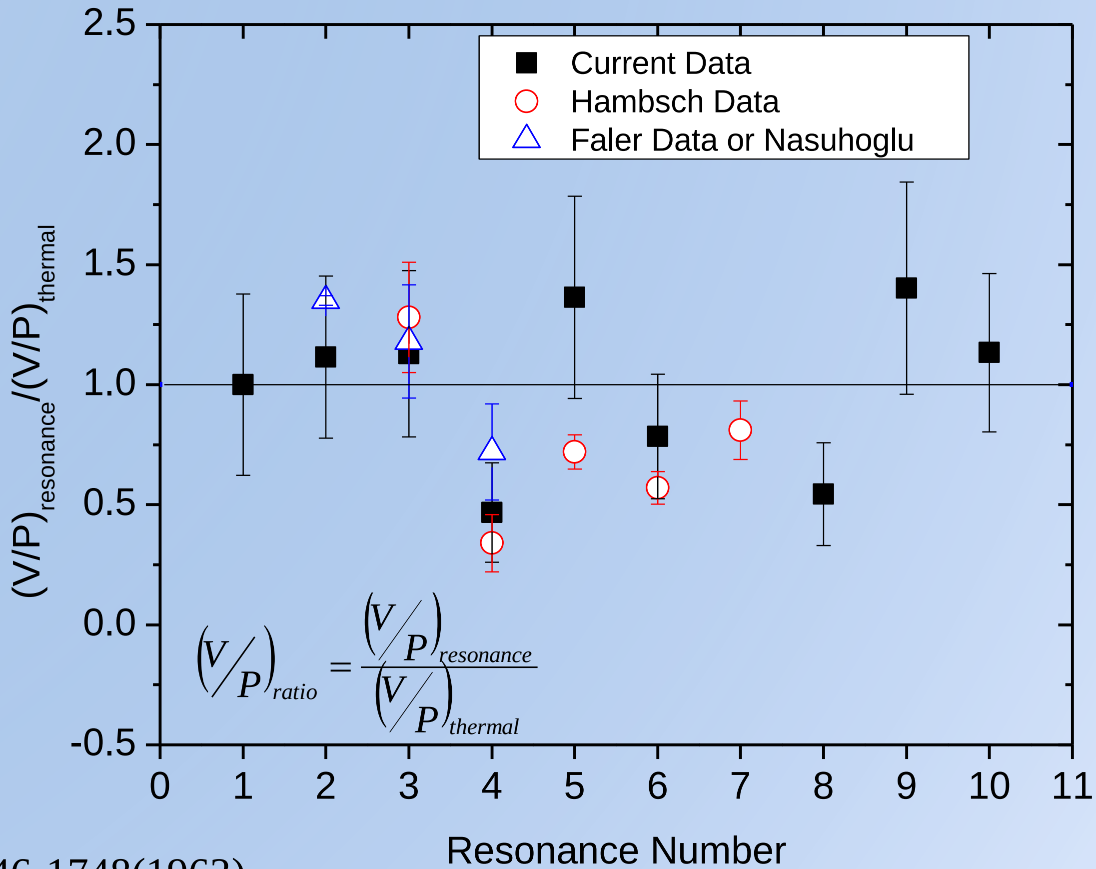
Fission symmetry in resonance clusters





K. T. Faler, R. L. Tromp, Phys. Rev. **131,** 1746-1748(1963).
R. Nasuhoglu, et al., Phys. Rev. **108**, 1522 (1957).

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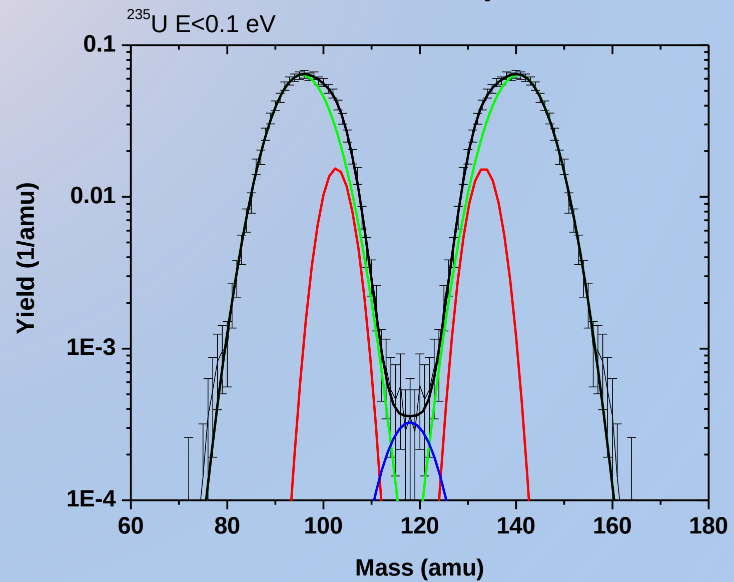
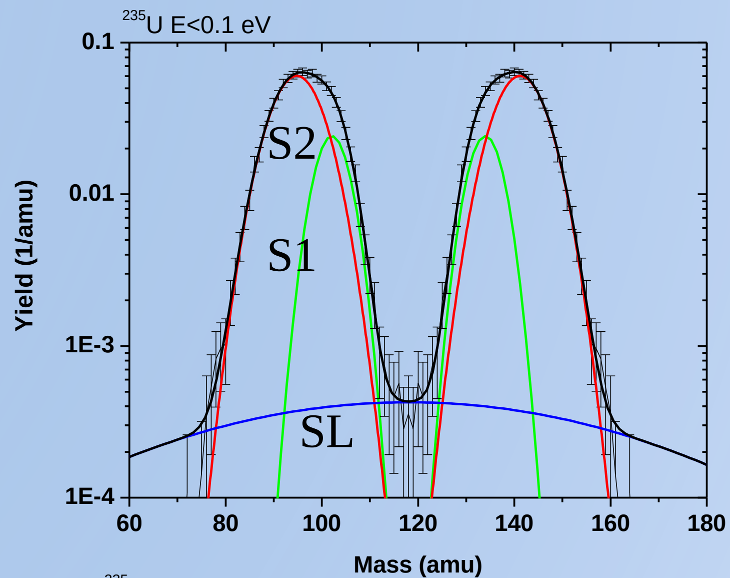
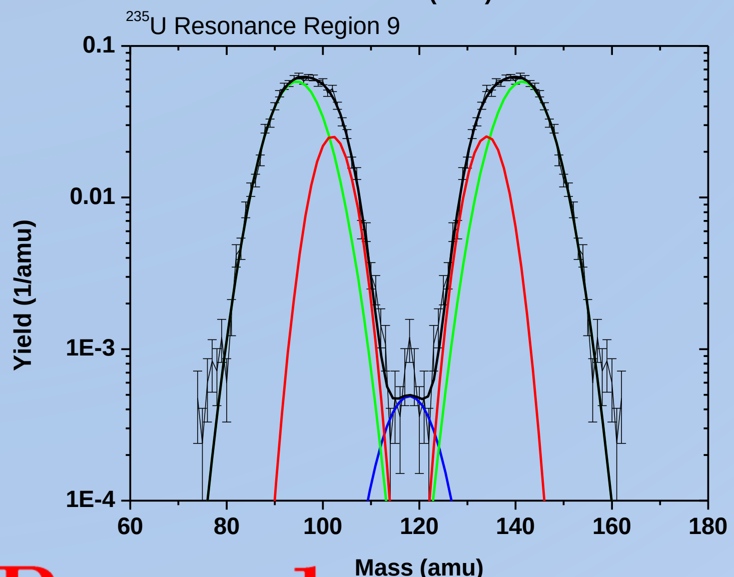
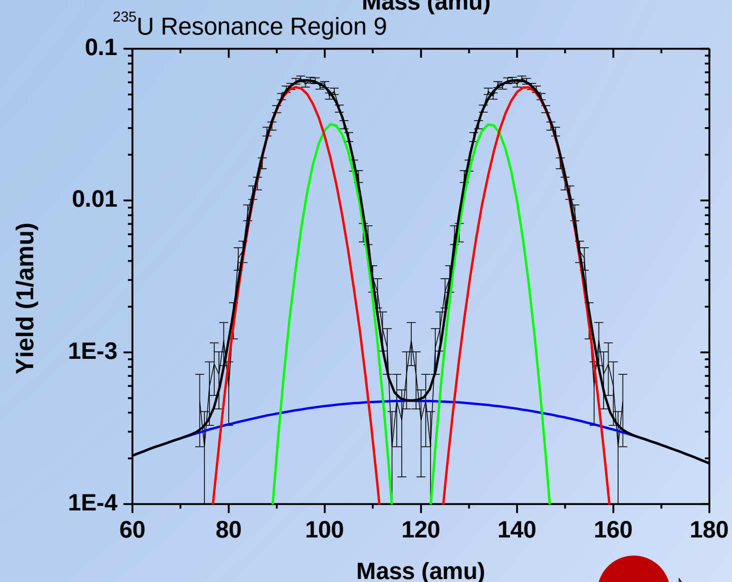
Fitting the fission modes

Several possibilities can be considered for example:

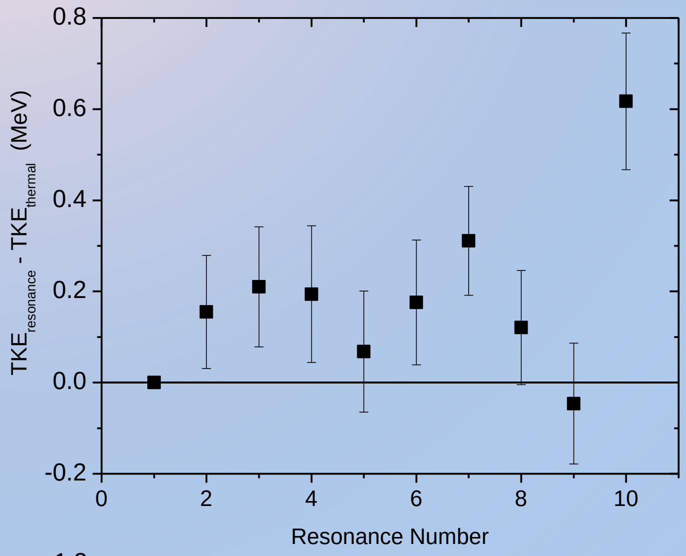


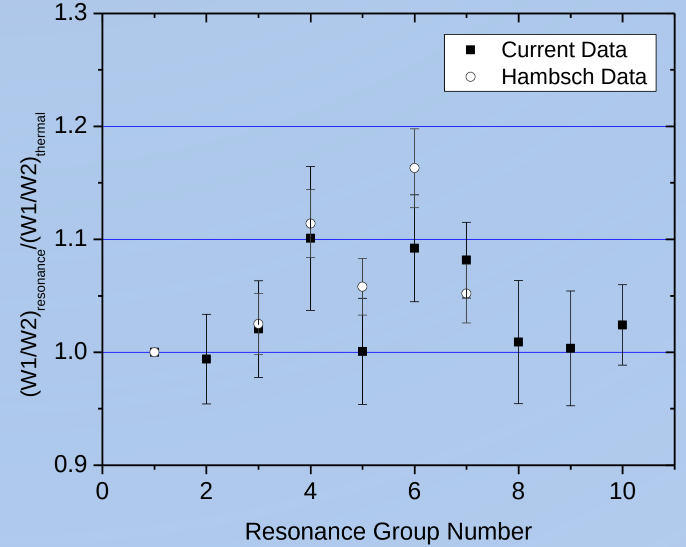


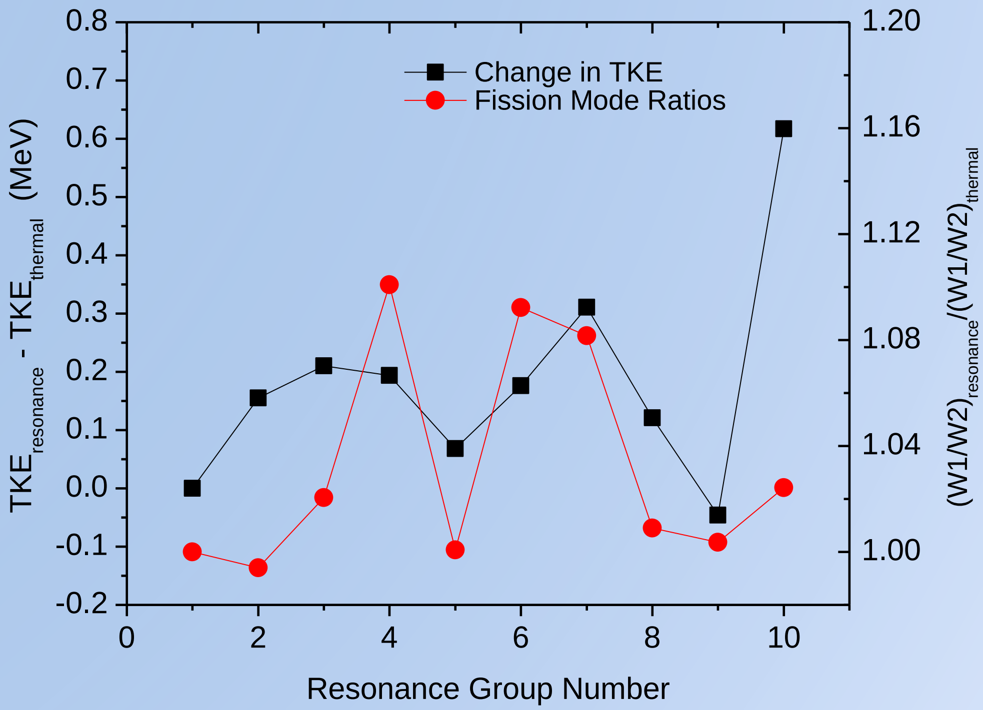




Variations In TKE As A Function of Incident Neutron Energy ^{235}U







$W_{1,2}$ weight of $S_{1,2}$ respectively

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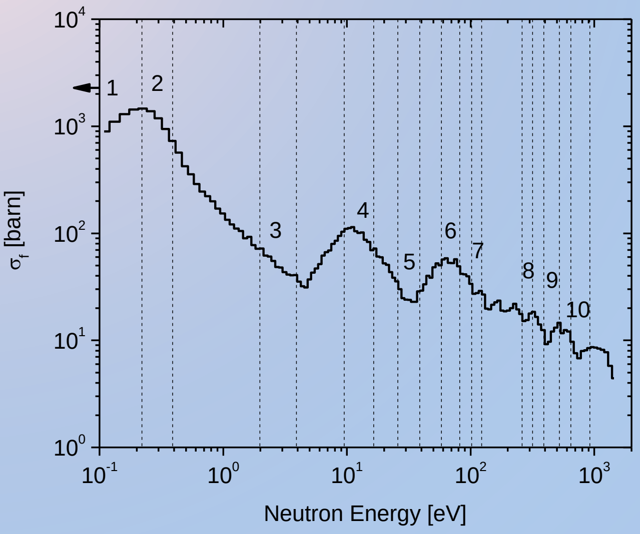
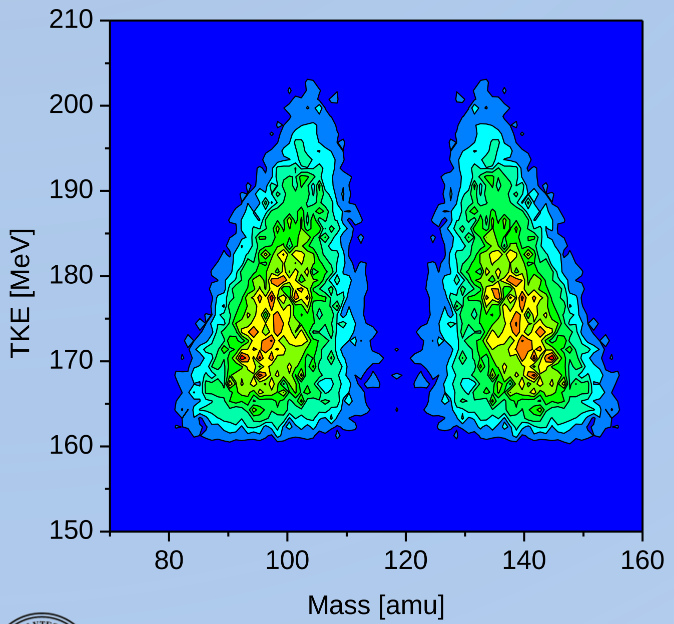
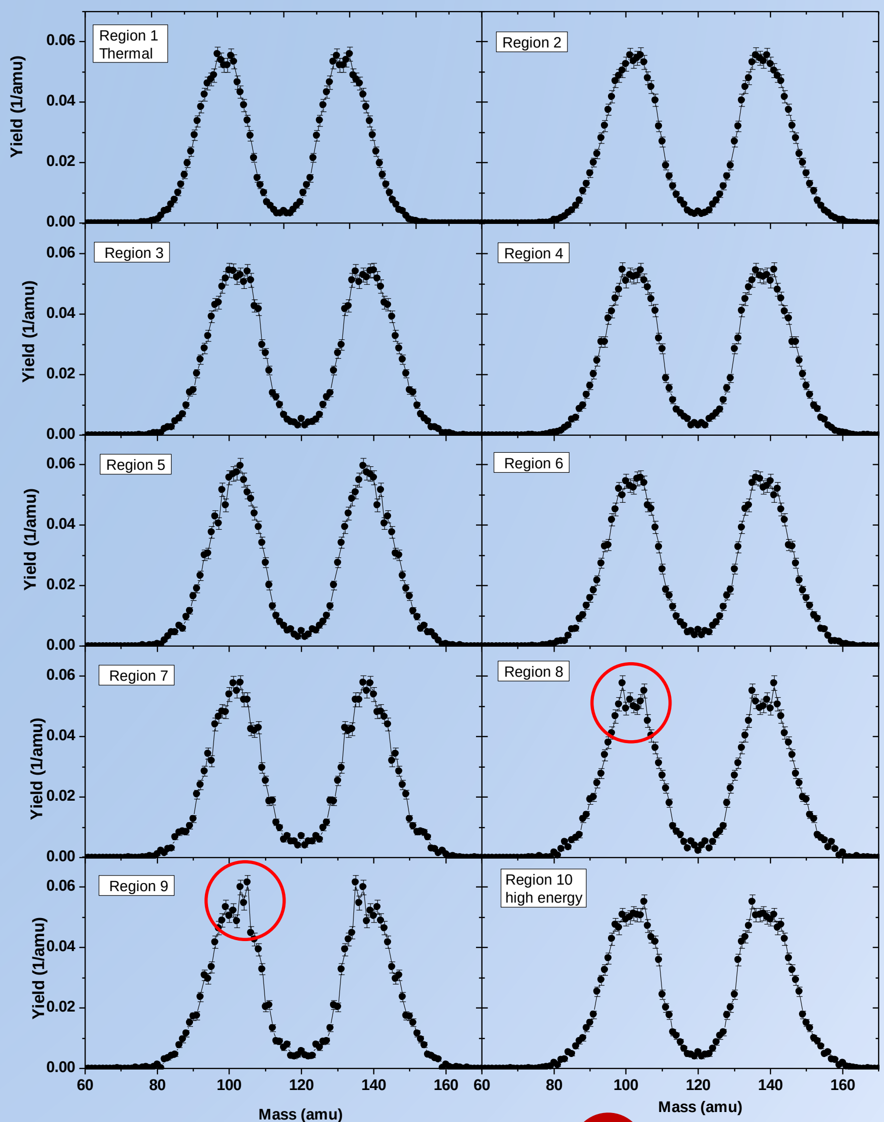
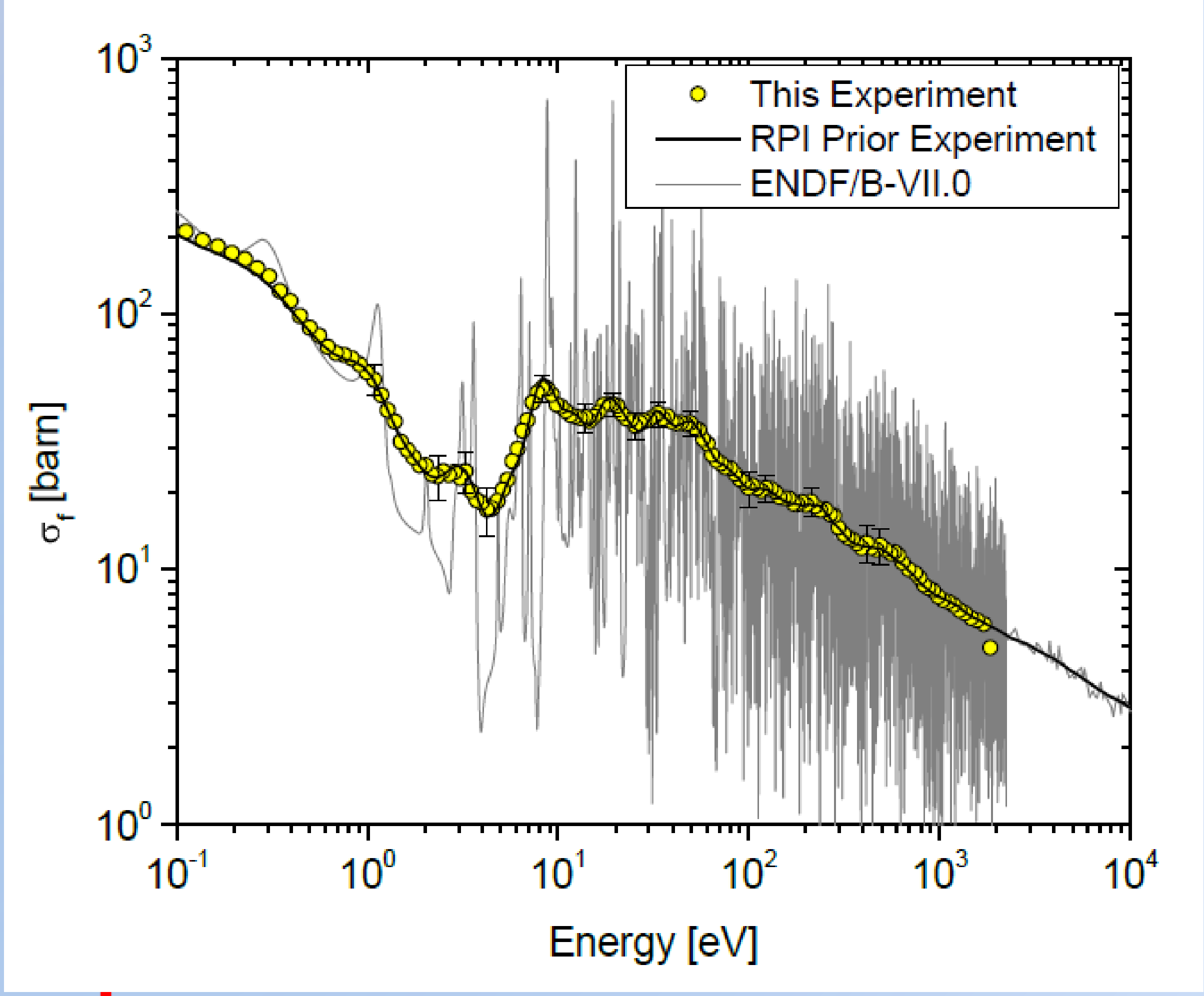
^{239}Pu - Results







^{235}U Fission Cross Section Measured at the Same experiment



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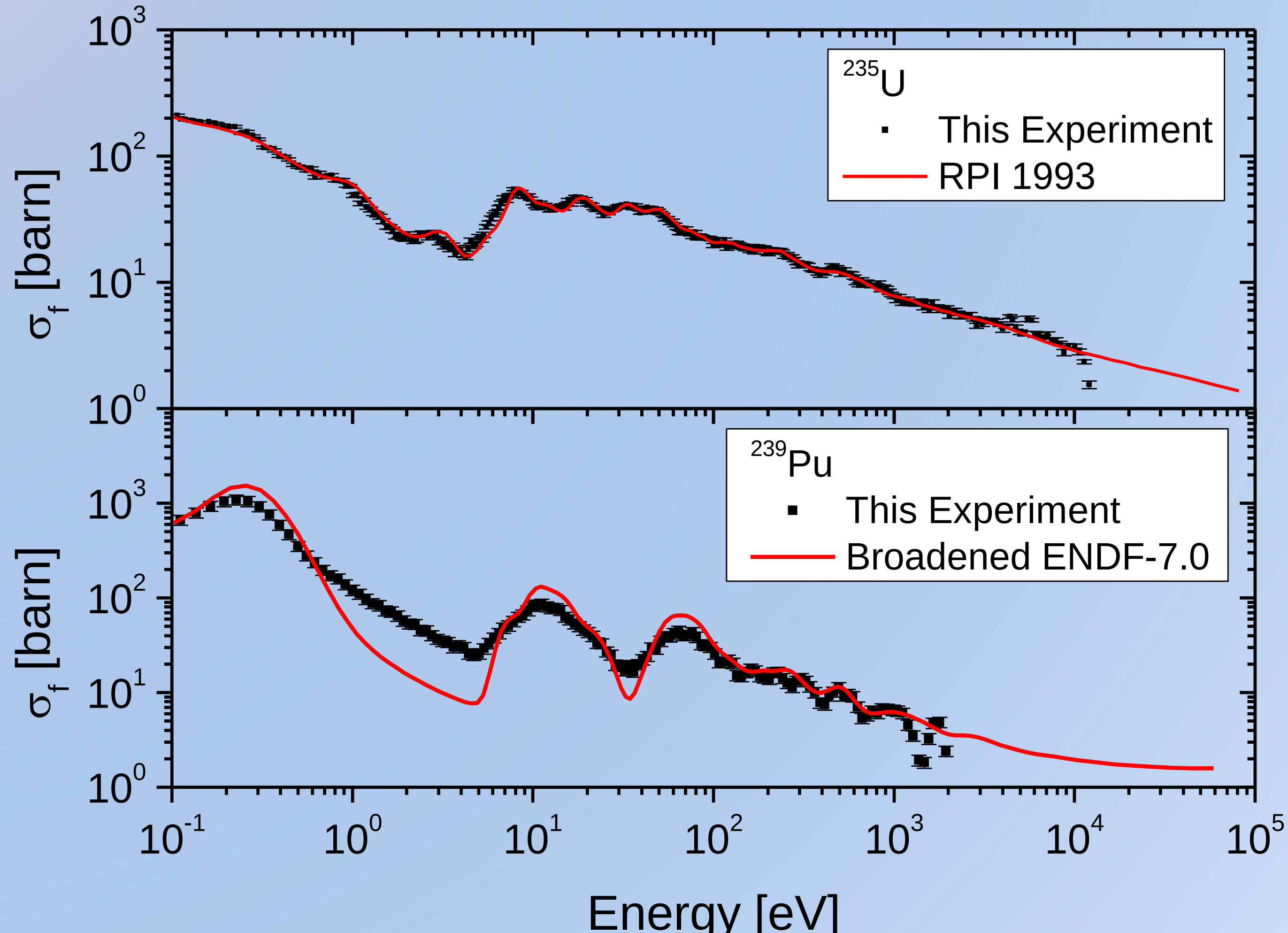
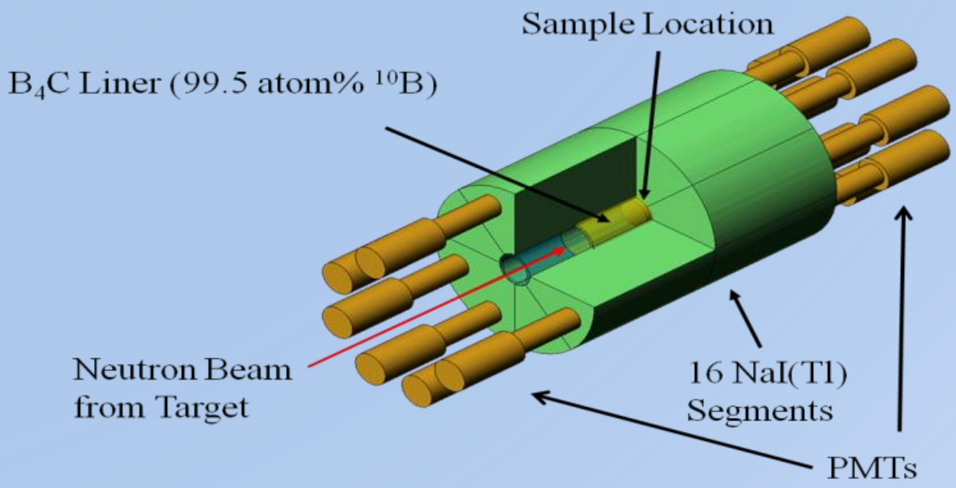
Results – Measured Fission Cross Section



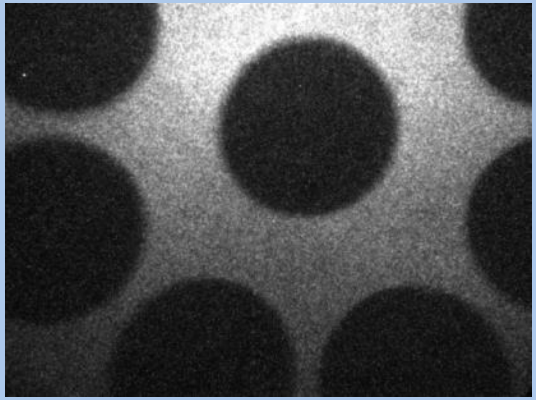
Fission and Capure Cross Section Measurement Using a Gamma Tag

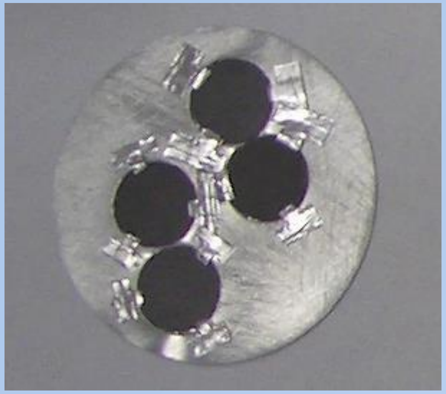
- 16 optically isolated segments NaI(Tl)
- Located at 25 m flight station
- 1.0 cm thick B_4C liner enriched in ^{10}B (captures scattered neutrons)
- Each event categorized by TOF channel, multiplicity, & total γ energy group (up to four groups)



Samples

- ^{235}U: Multiple ½” discs 93% enriched in ^{235}U
- Additional samples: Depleted U & Au (~ 30mil each)
- Empty sample holder: background subtraction





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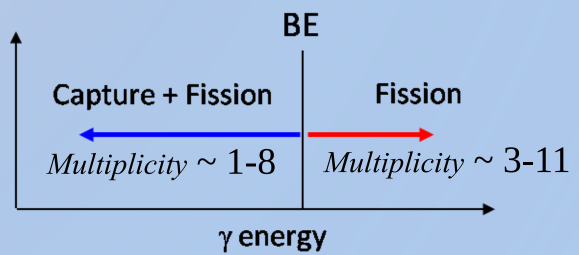
Measurements of ^{235}U Capture & Fission Yields

- **Thermal** measurement with enriched ^{235}U sample
- 16 Segment Multiplicity Detector with 4 E_γ groups
- Good agreement with SAMMY calculations
- Extracting Capture Yield from data with mixture of capture and fission events



- Challenges: **Normalization**

➢ Normalize experimental fission yield to thermal point

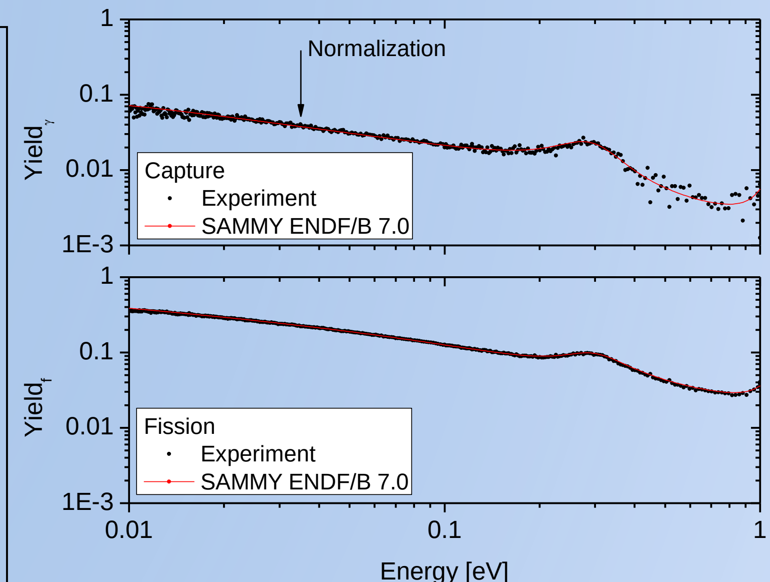
$$Y_f^{ENDF} = k_1 \cdot Y_f$$ Solve for k_1 @ 0.0253 eV

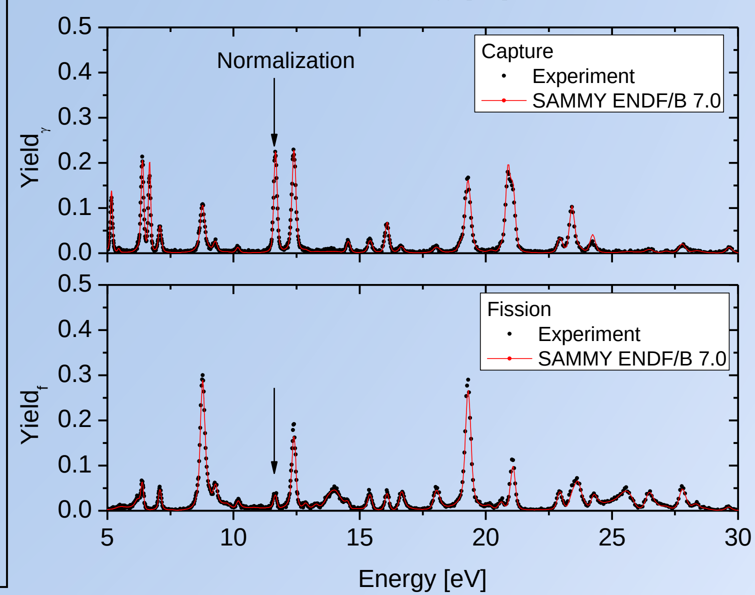
➢ Use two equations for a predominantly capture resonance and predominantly fission region (thermal)

@ 11.7 eV res $\left(\frac{\Gamma_\gamma}{\Gamma} = 0.86\right)$ @ 0.0253 eV

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

➢ Solve the two equations for k_2 and k_3





^{235}U Capture & Fission Yield Data - Epithermal Measurement

- Challenges:
  - Normalization
  - False capture due to neutron scattering

➢ Normalize experimental fission yield to resonance

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

➢ Use two equations for predominantly capture and fission resonances

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

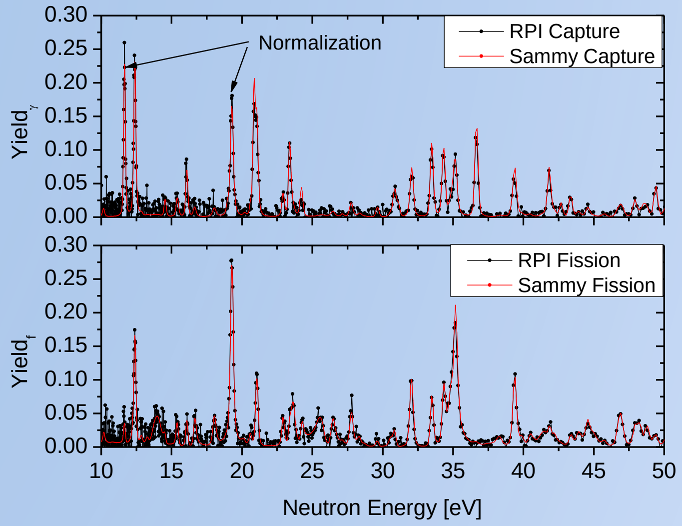
$$Y1_\gamma^{ENDF} = k_2 \cdot Y1_\gamma - k_3 \cdot k_1 \cdot Y1_f \qquad Y2_\gamma^{ENDF} = k_2 \cdot Y2_\gamma - k_3 \cdot k_1 \cdot Y2_f$$

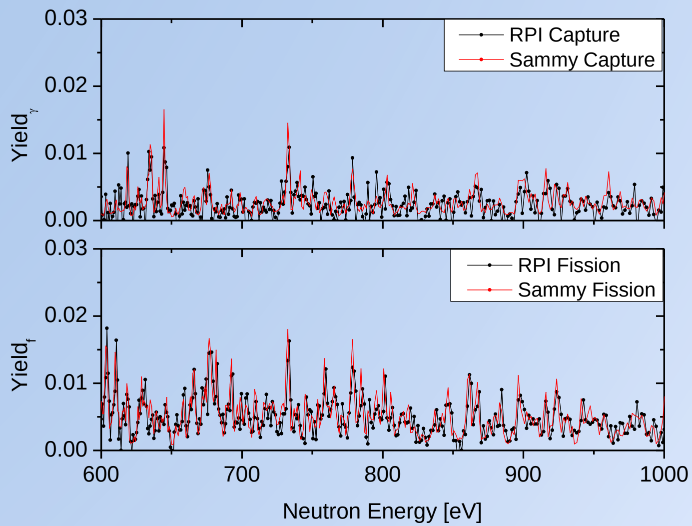
➢ Solve the two equations for k_2 and k_3

► **Need 2 resonances with known parameters** ◄

Provides data to address WPEC subgroup 29 report

"Uranium-235 Capture Cross-section in the keV to MeV Energy Region"





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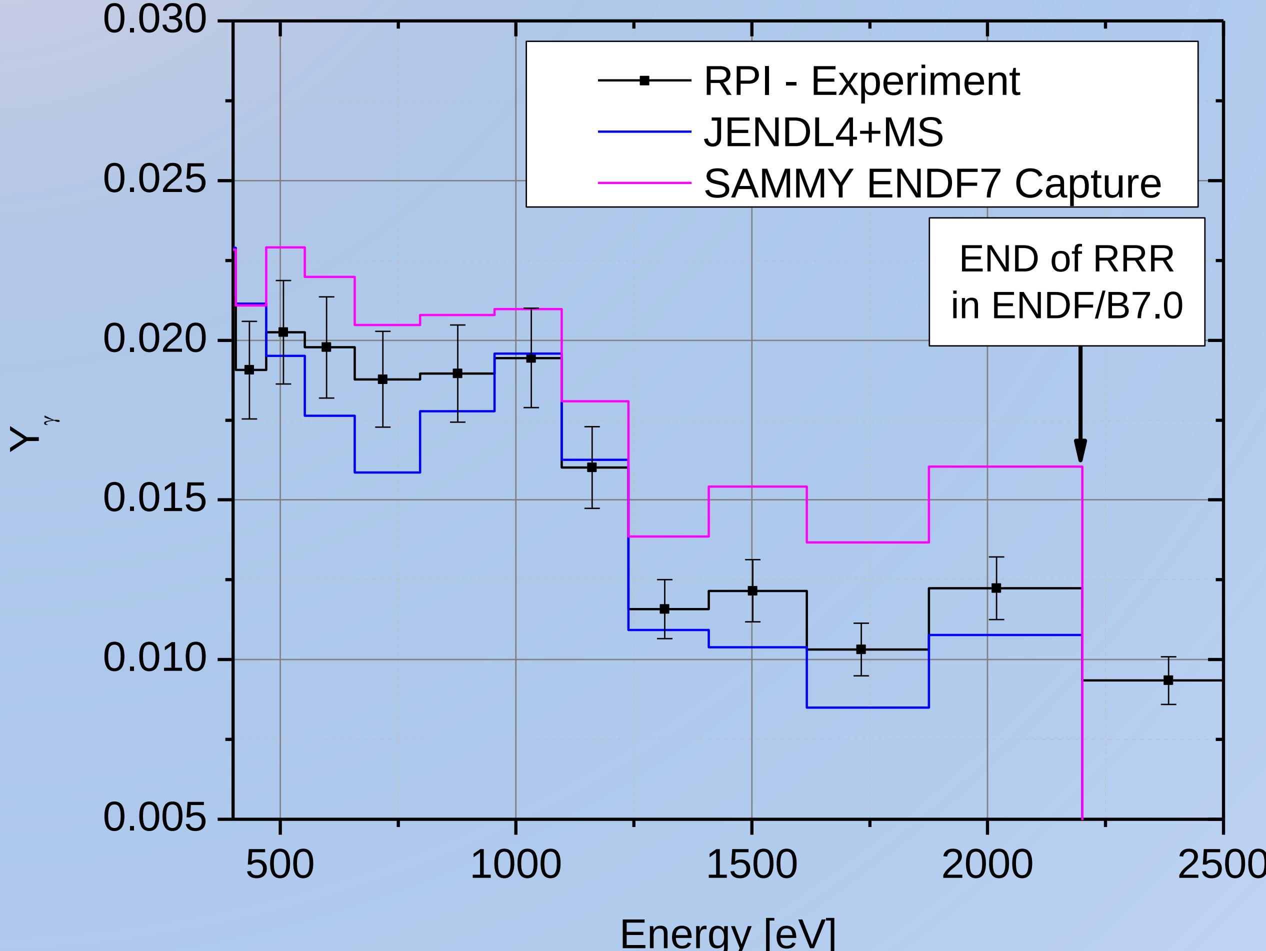
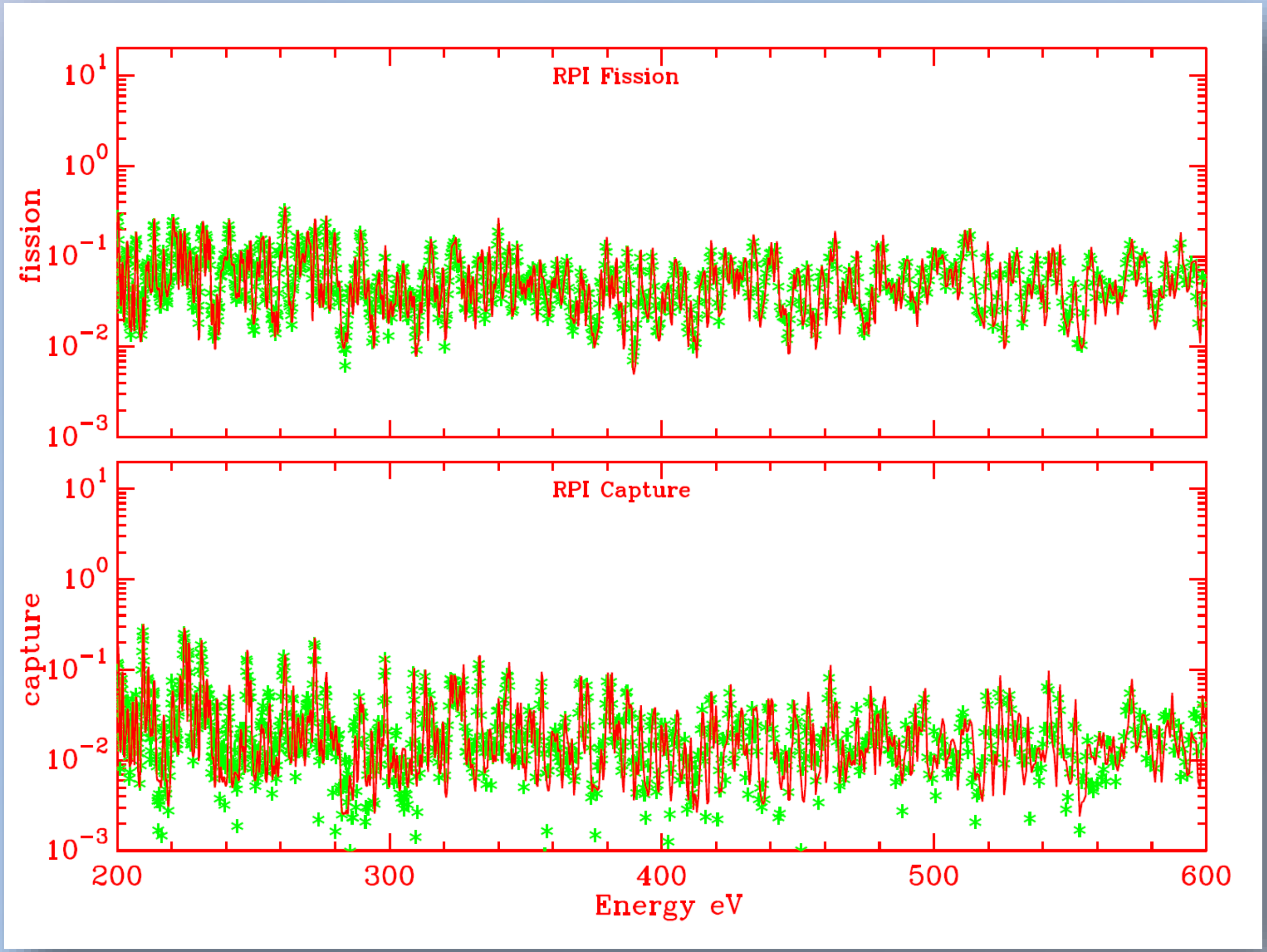
Comparing ^{235}U Fission and Capture with Evaluations

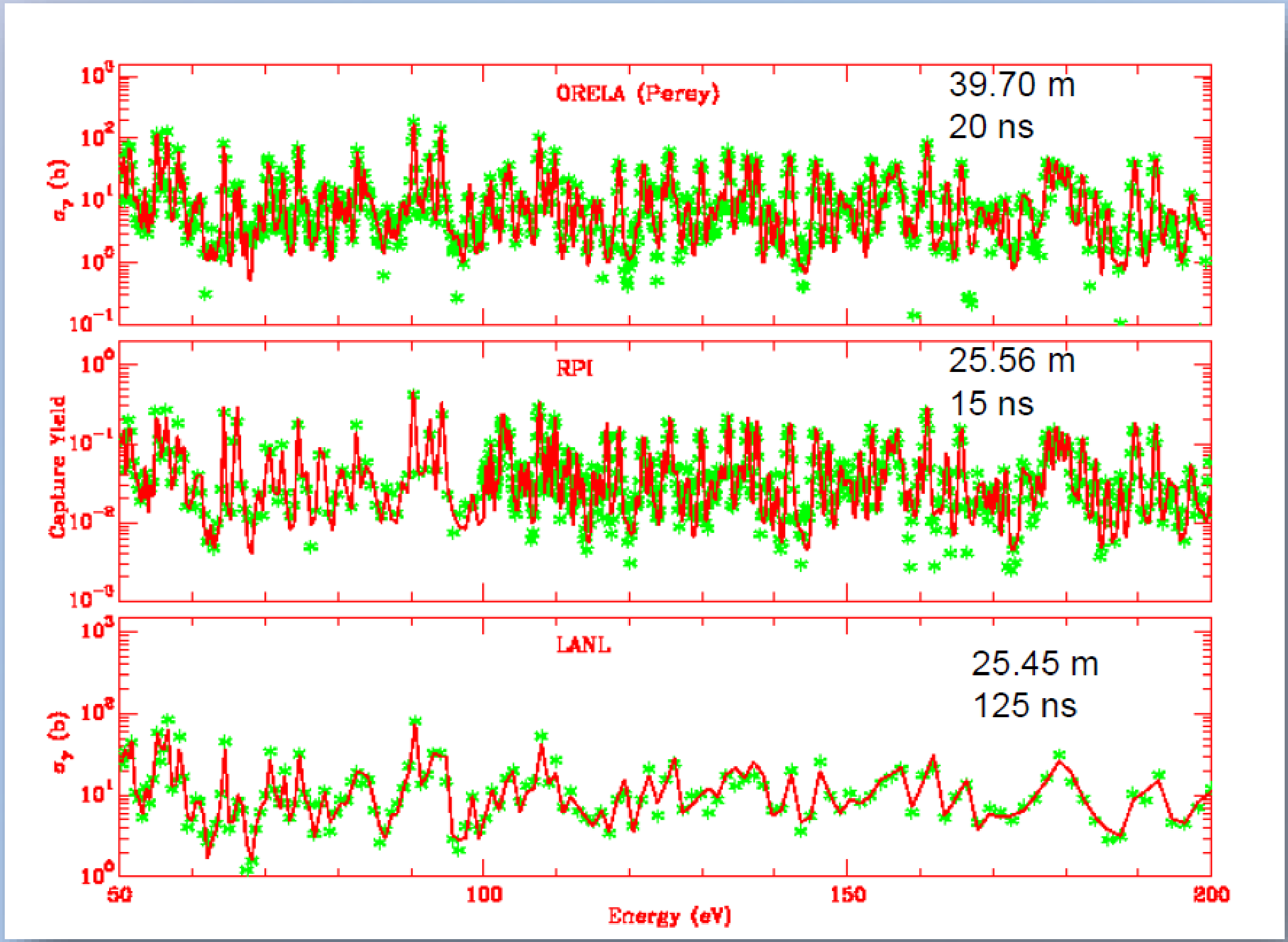


- Fission is in excellent agreement with evaluations
- Capture data has up to 8% multiple scattering that must be taken into account during the analysis
- Capture yield uncertainty is about 8%
- **0.4-1 keV capture data is closer to ENDF/B-7.0**
- **1-2 keV ENDF/B7.0 too high JENDL 4.0 too low.**
- **E>1 keV data is slightly higher than evaluations but within uncertainties.**

ORNL SAMMY fit to ^{235}U to RPI fission and Capture Yields

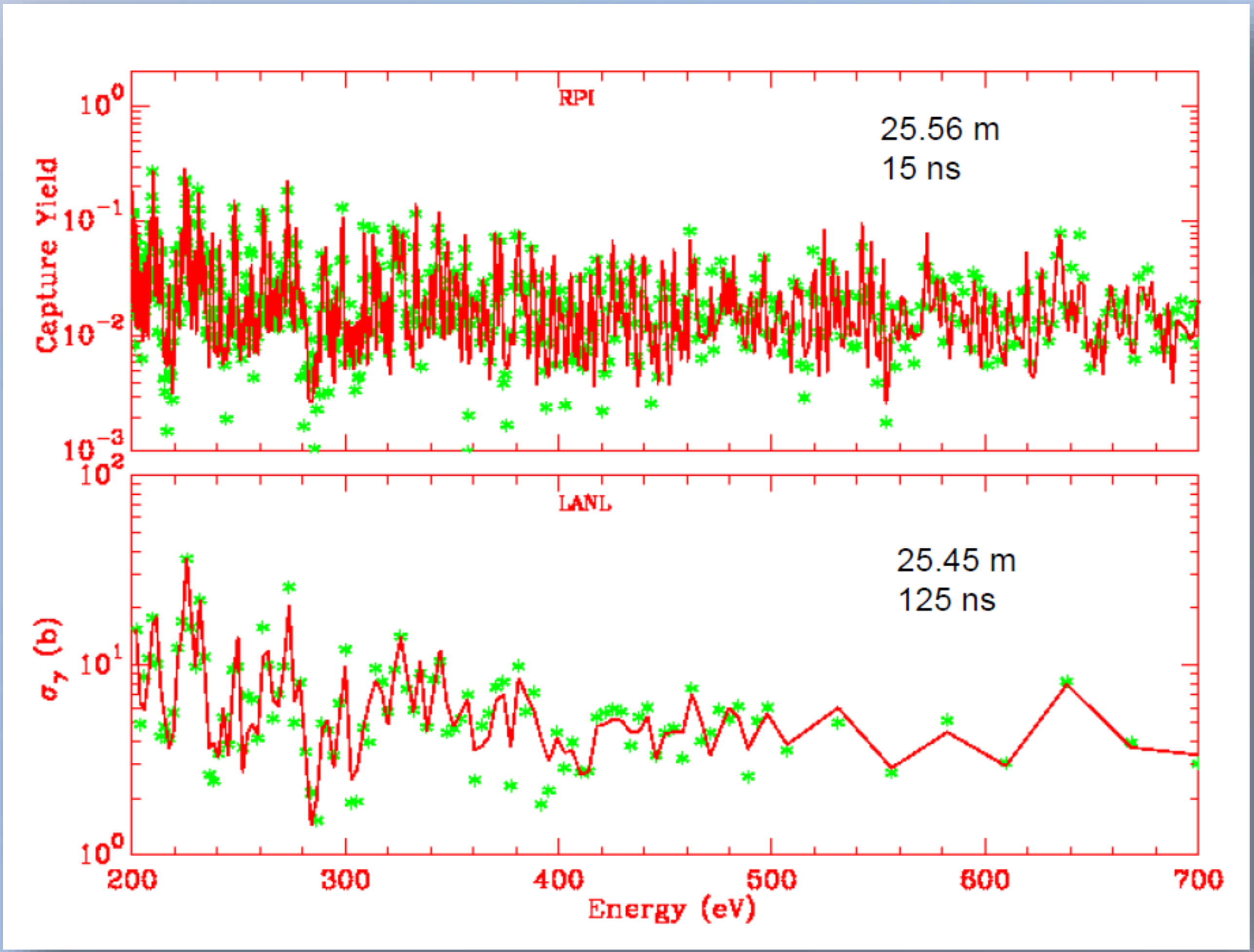


^{235}U (n,γ) Low Energy Region



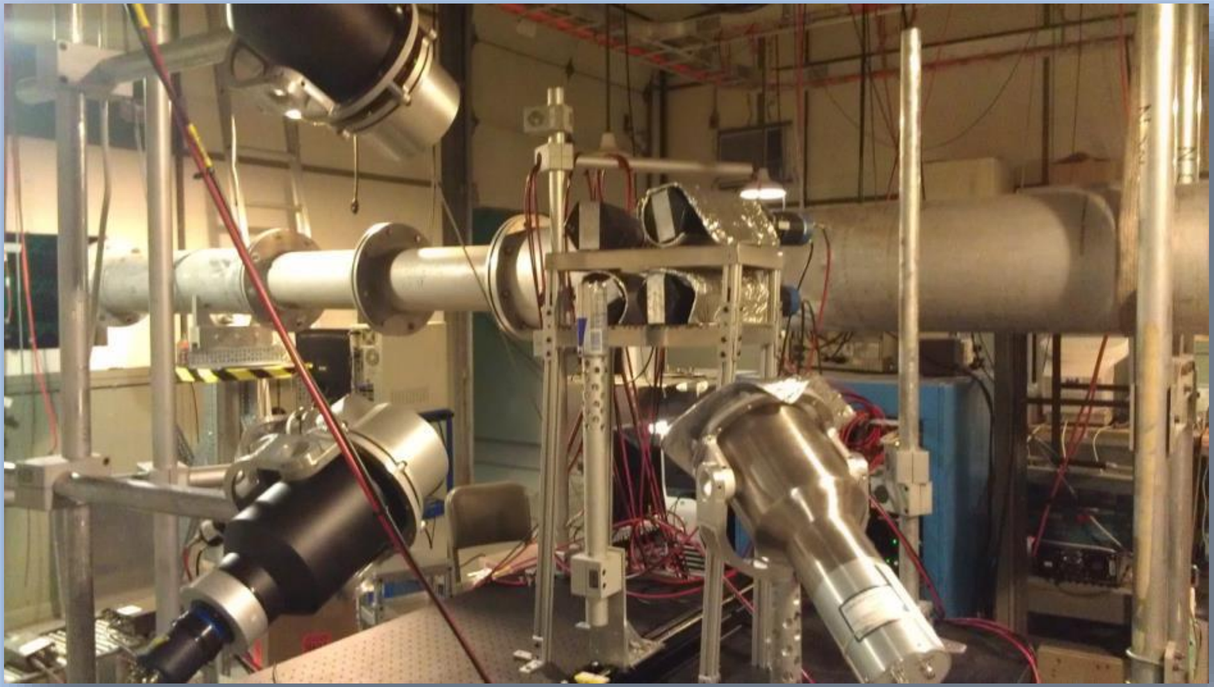
From ORNL 2013 CSEWG presentation

^{235}U (n,γ) High Energy Region



From ORNL 2013 CSEWG presentation

Prompt Fission Neutron Spectra

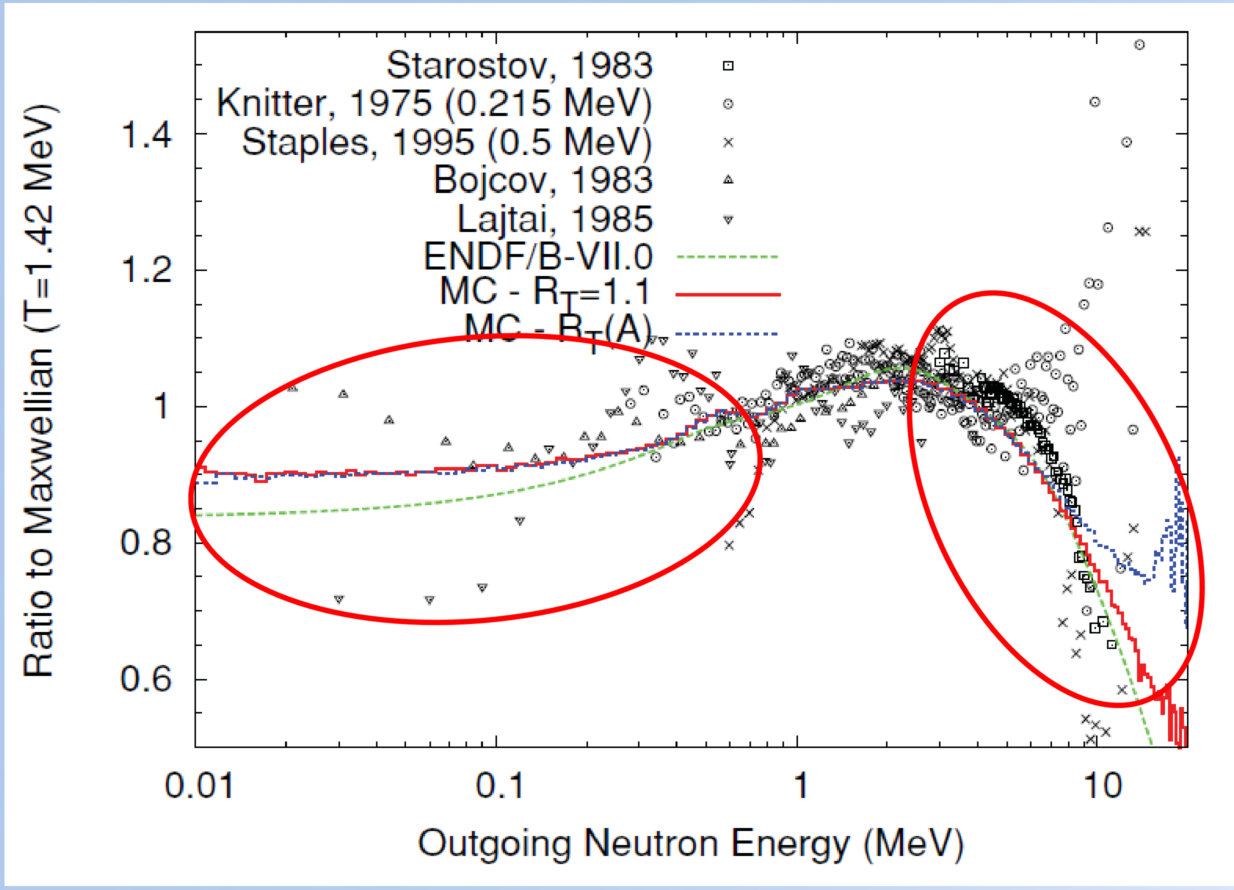


Motivation/Fission Spectra

- $E_{emission}$<0.5 MeV and $E_{emission}$> 6 MeV
  - Very little or no experimental data
  - High uncertainty in current models

P. Talou, B. Becker, T. Kawano, M. B. Chadwick, and Y. Danon, "Advanced Monte Carlo modeling of prompt fission neutrons for thermal and fast neutron-induced fission reactions on ^{239}Pu", Phys. Rev. C 83, 064612 (2011).

^{239}Pu, E_n=Thermal



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Fission Spectrum Measurement

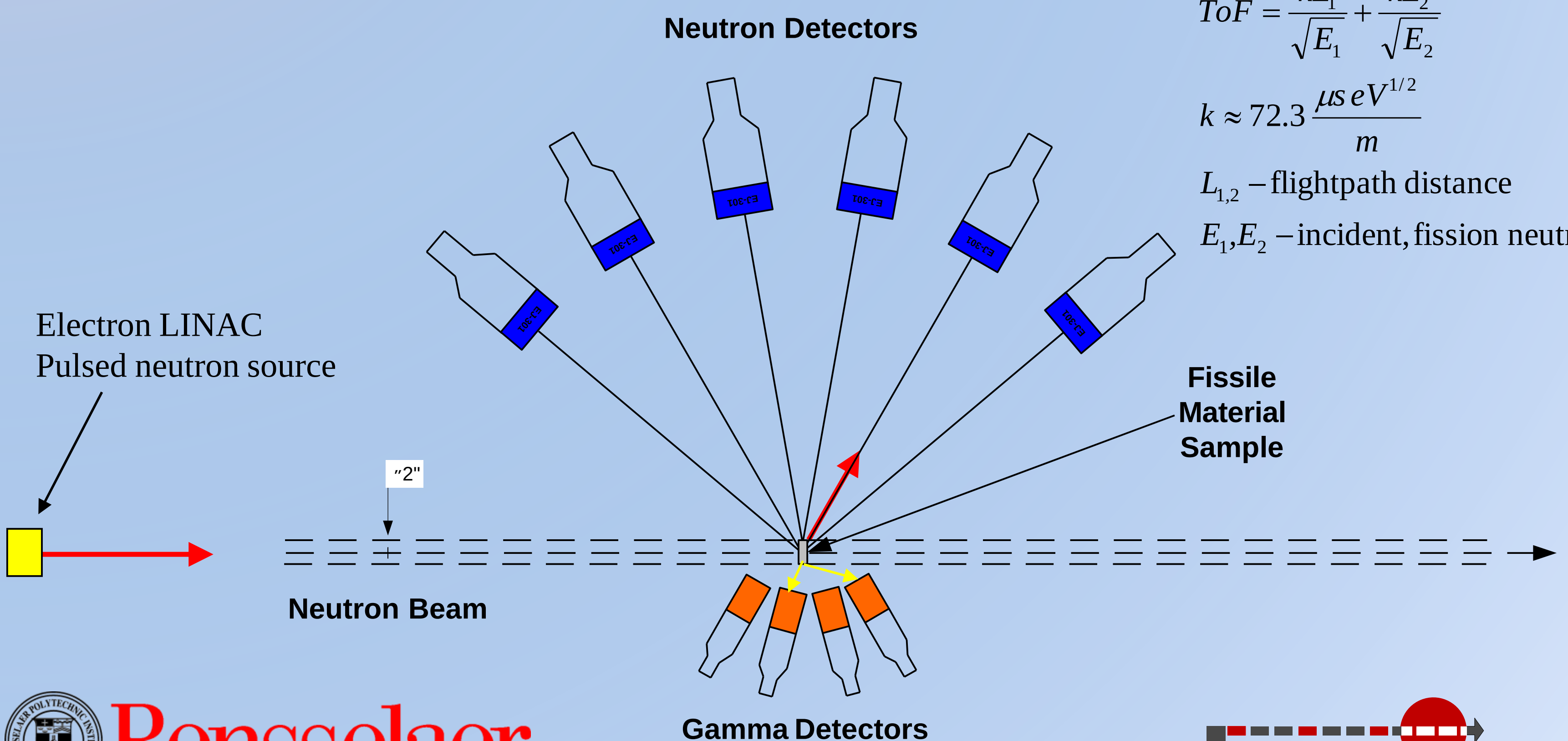
- Use the double TOF method
- Use a gamma tag for fission (instead of traditional fission chamber)
- Use a combination of Liquid Scintillators and Li-Glass neutron detectors

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

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

$L_{1,2}$ – flightpath distance

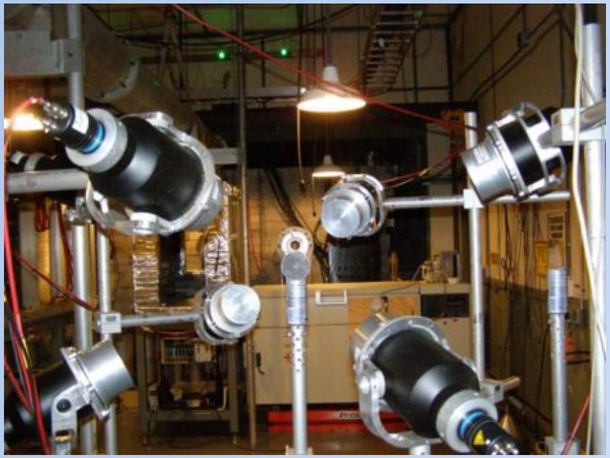
E_1, E_2 – incident, fission neutron energy



Gamma Tagging

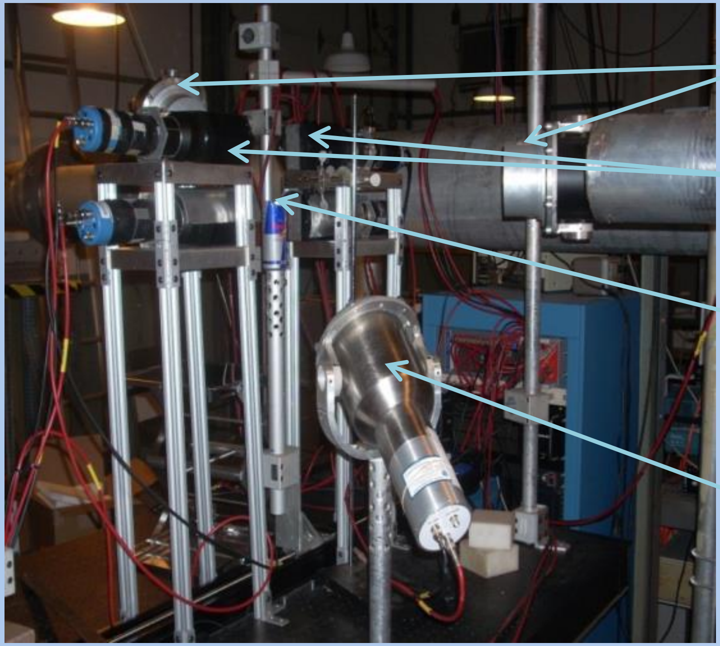
- Advantages
  - Eliminated the need to construct a complicated multiplate fission chamber
  - Simpler sample preparation
  - Can use relatively large samples
  - Can increase the detected fission rate
- Disadvantages
  - False fission detection due to:
    - Random coincidence for radioactive decay
    - Neutron interactions with the gamma detection
    - Beam related:
      - Gamma capture (no neutron emission)
      - Inelastic Scattering (single gamma emitted)
      - Increased background (the biggest problem, use simulations to characterize)

Fast neutrons scattering detector array



Experimental Setup

- Neutron Detectors
  - EJ-204 Plastic Scintillator
    - 0.5” x 5”
    - 47 cm away from center of sample
  - 2 EJ-301 Liquid Scintillators
    - 3” x 5”
    - 50 cm away from center of sample



- Gamma Detectors
  - 4 BaF_2 detectors on loan from ORNL
  - Hexagonal detectors 2” x 5”
  - 10 cm from center of sample
  - ¼” lead shield between detectors
    - Reducing scattering between detectors

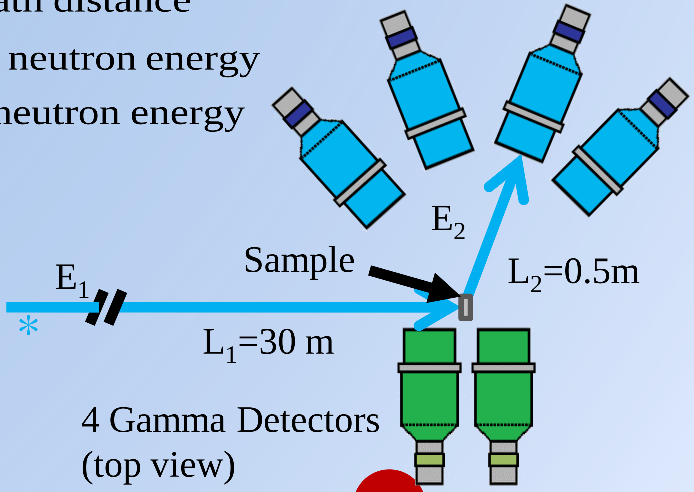
$$ToF = \frac{kL_1}{\sqrt{E_1}} + \frac{kL_2}{\sqrt{E_2}}$$

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

$L_{1,2}$ – flightpath distance

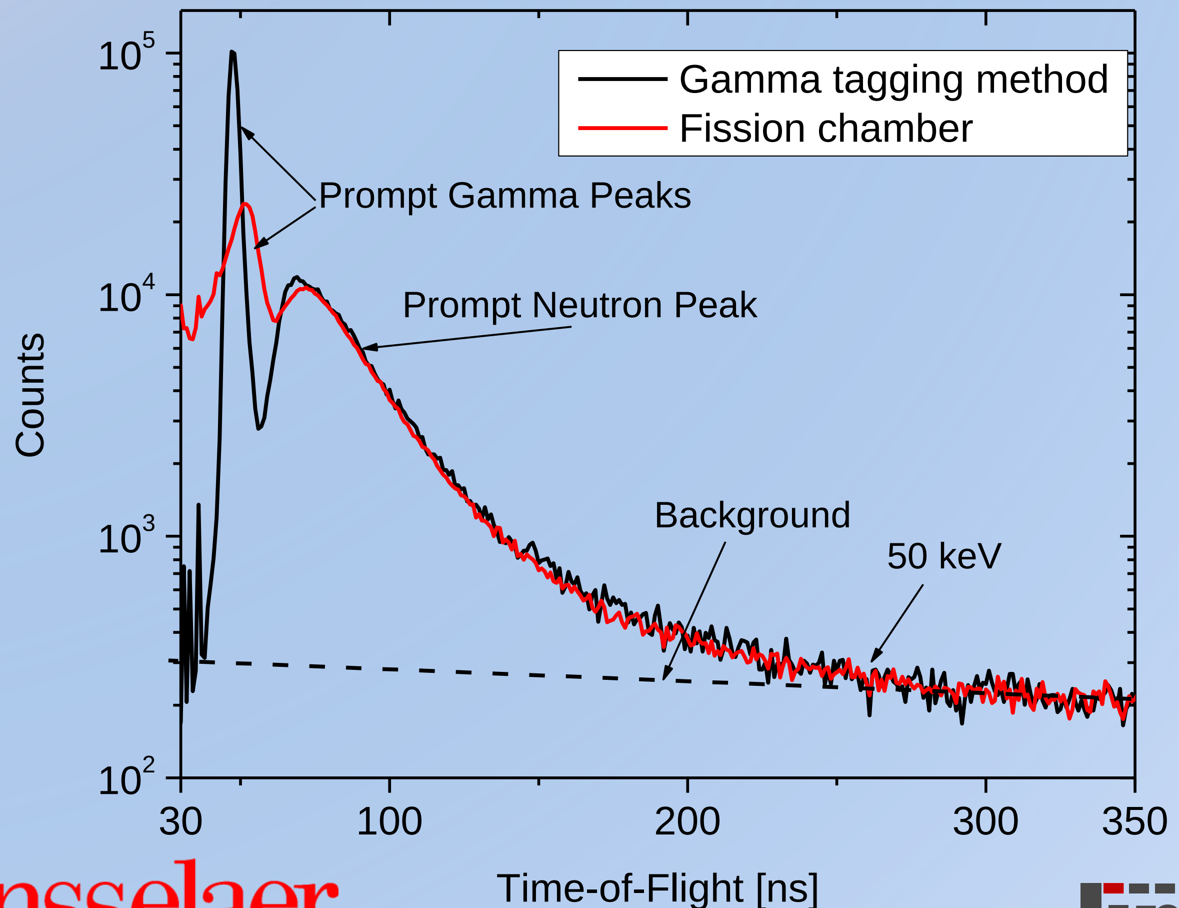
E_1 – incident neutron energy

E_2 – fission neutron energy



Gamma Tagging - EJ-204

- Gamma tagging method corrected for 30% detection efficiency compared to 83% detection efficiency with fission chamber

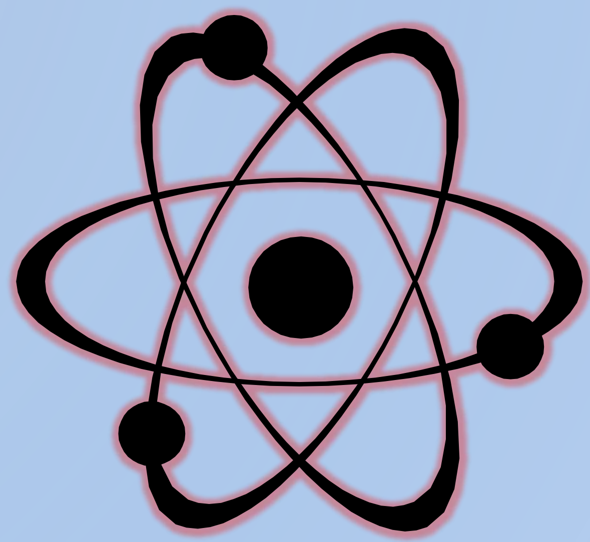


Results

Will be shown in Zeke Blain's talk

Summary

- Simultaneous Measurements of Fission Cross Section and Fission Fragment Mass and Energy Distributions of Small Samples.
  - Demonstrated the feasibility of this method
  - Works with small mass sample ~20-40 ug.
  - Data collection with small sample is very quick (<10 hours)
  - Fission cross section can also be measured at the same time
  - Data was obtained for ^{252}Cf, ^{235}U and ^{239}Pu and compares well to other measurements
  - Room for improvement in the DAQ system
- Fission and capture measurements from prompt fission gamma
  - Eliminates the fission detector in the conventional approach
  - Utilize gamma multiplicity and total energy
  - Demonstrated on ^{235}Ufrom thermal to 2.5 keV
- Fission spectra
  - Demonstrated and characterized the concept of gamma tagging with a ^{252}Cf sample.
  - Studied the effect of a discriminator on measurements with a fission chamber (applicable to detector calibration with ^{252}Cf).
  - Performed PFNS measurement ^{238}U



Thank You