Measurements with a Lead Slowing-Down Spectrometer

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Outline

- **Introduction**
  - Why we need nuclear data
  - The RPI nuclear data program
- **LSDS - basic physics and principles of operation**
- **Applications**
  - Fission cross section measurements
  - (n,α) cross section measurements
  - Measurements fission fragment mass and energy distributions
  - Neutron capture reaction rates
  - Assay of used nuclear fuel

Why Should We Care About Nuclear Data?

Reactor Physics Calculations

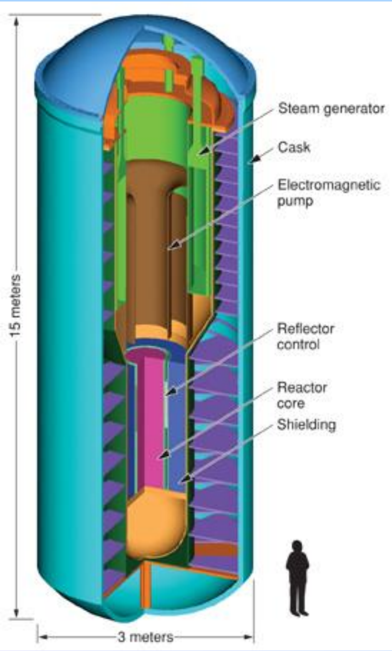
Nuclear Data (Uncertainty)

Geometry Data

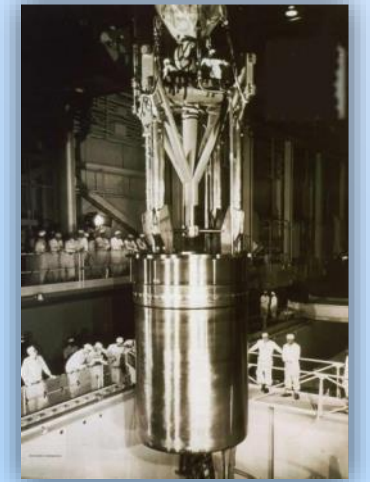
Computational Methods (Physics) (Uncertainty??)

Results Accuracy??

- Effective neutron Multiplication factor
- Neutron flux
- Burnup
- Kinetics

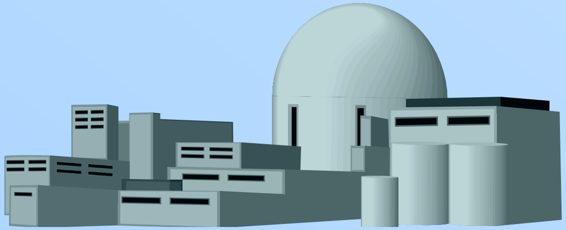


www.llnl.gov



The Shippingport Reactor (Critical in 1957)

http://www.pabook.libraries.psu.edu/palitmap/Shippingport.html



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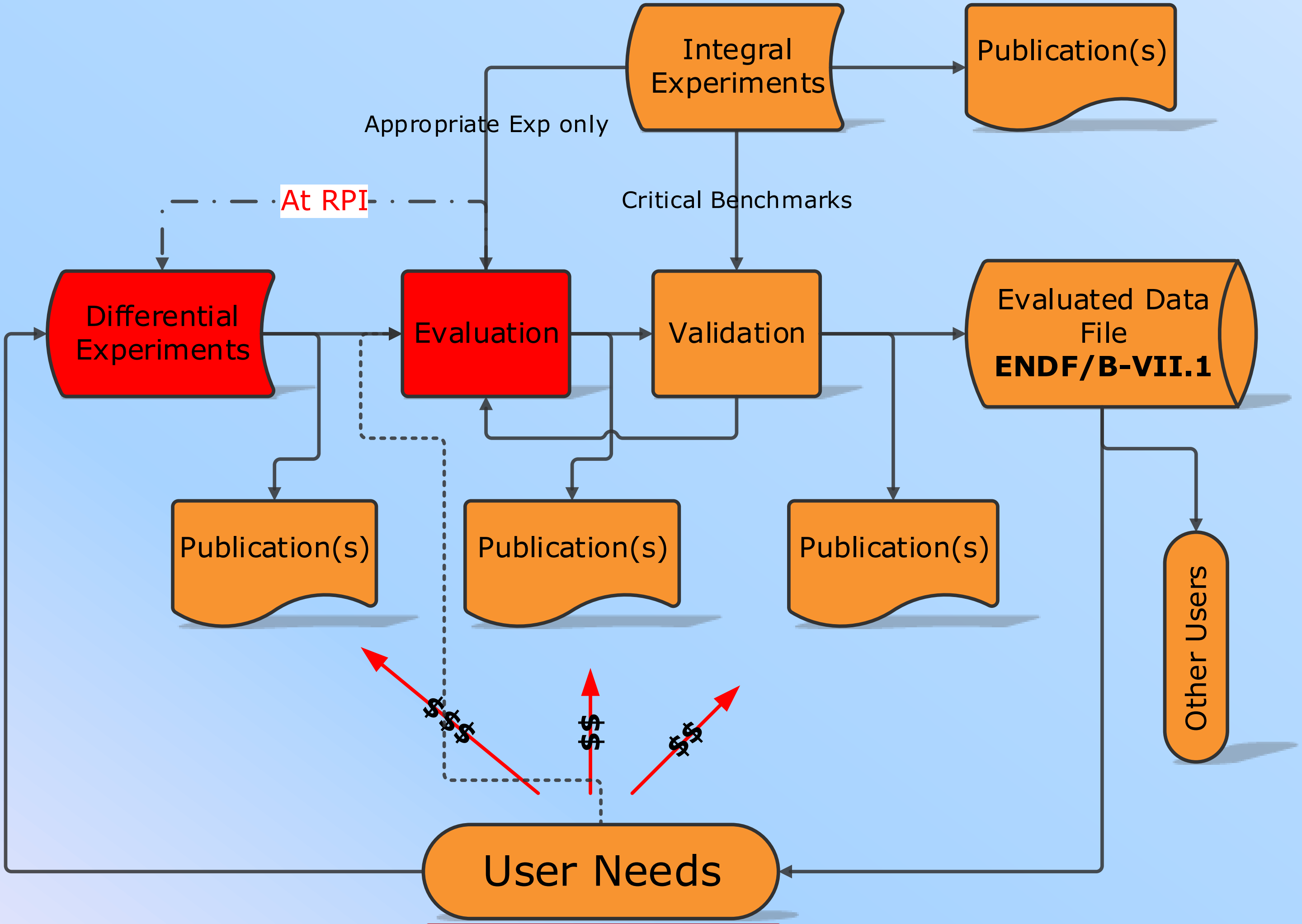
Nuclear Data Lifecycle (Danon's view)

Application Driven



Applications

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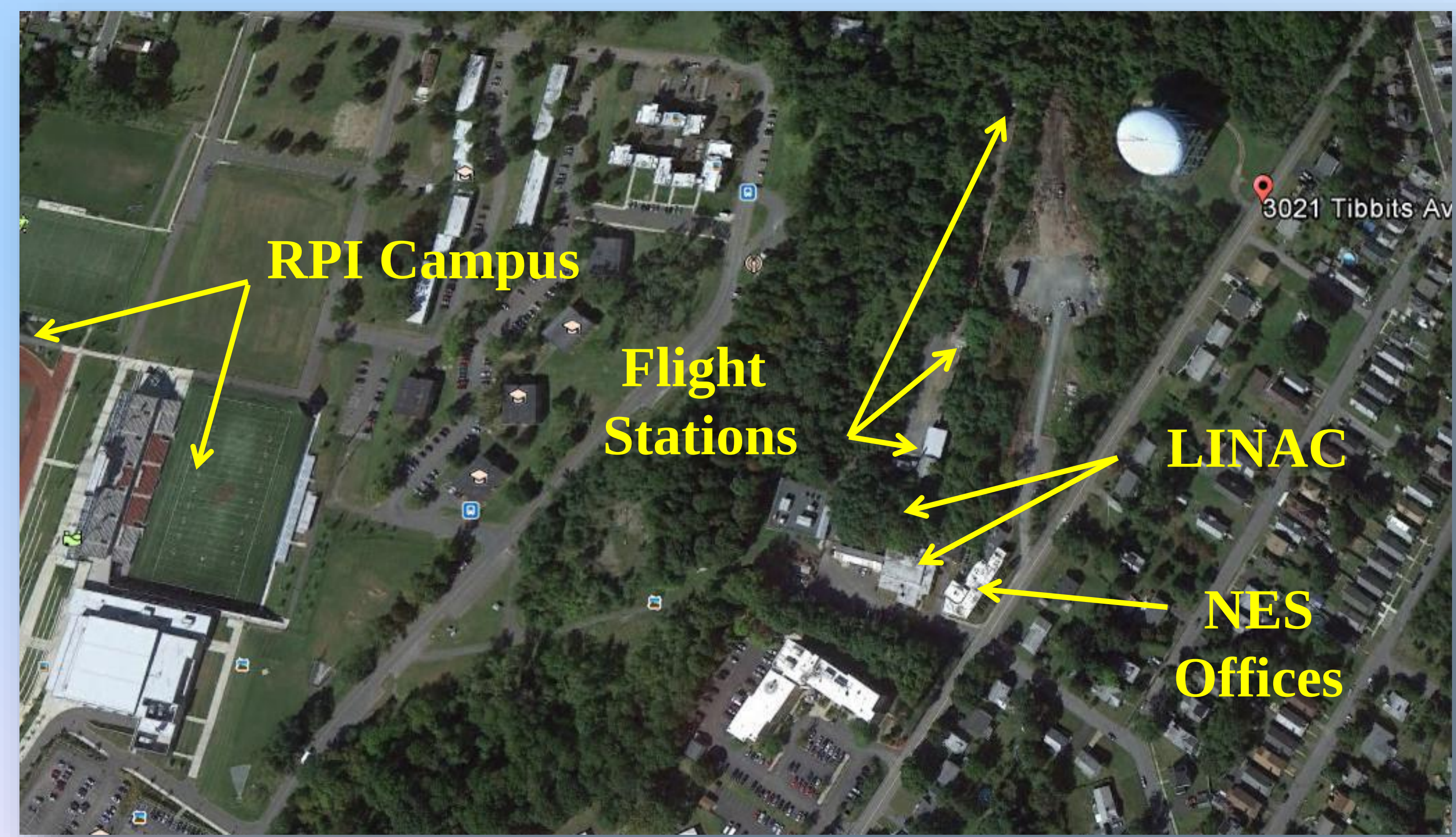
Where is the RPI LINAC ?

- It is on the highest point in Troy, NY



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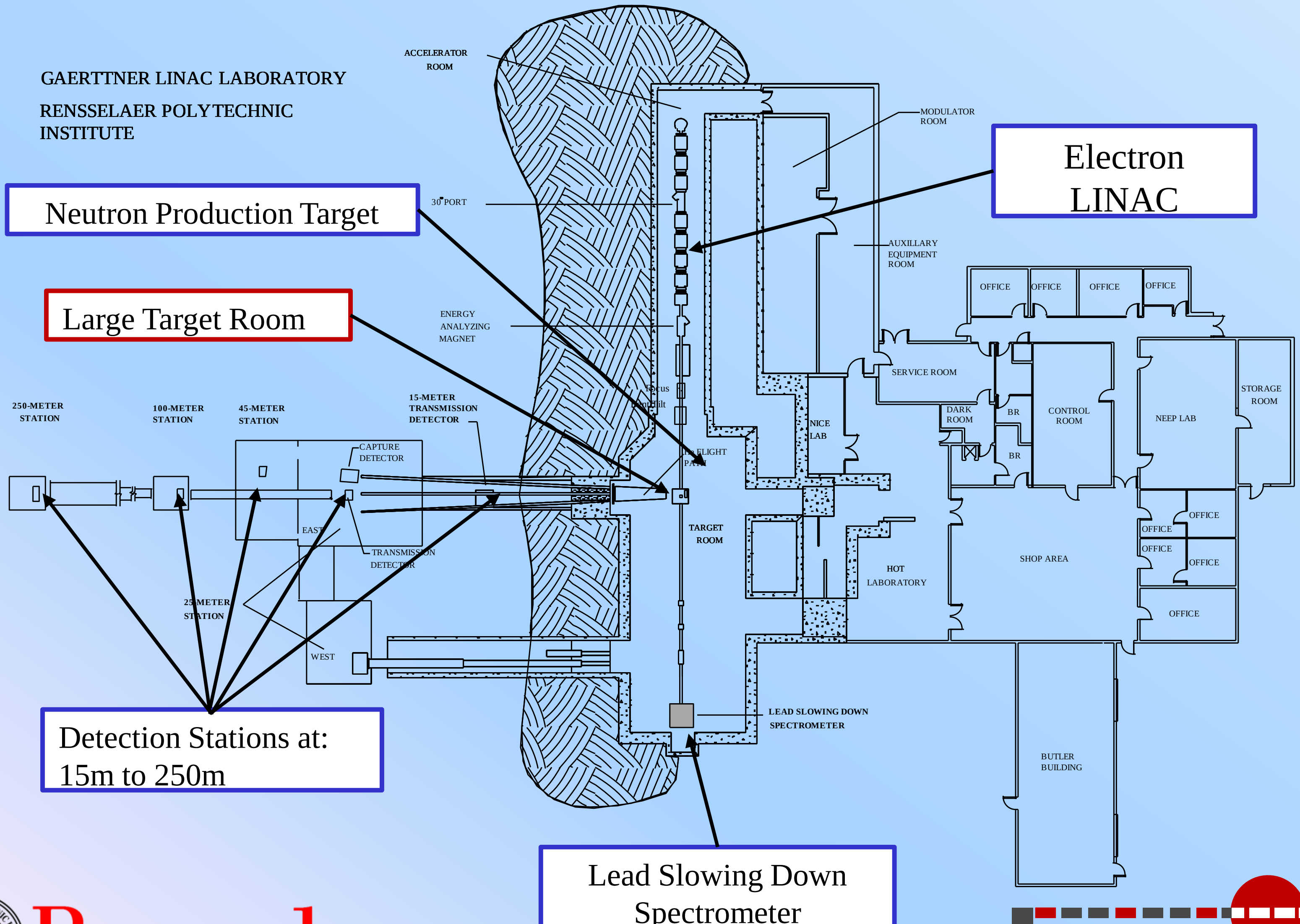
The RPI LINAC Facility

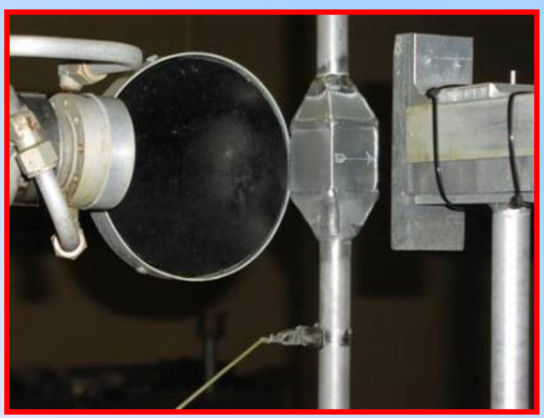


Current Activity

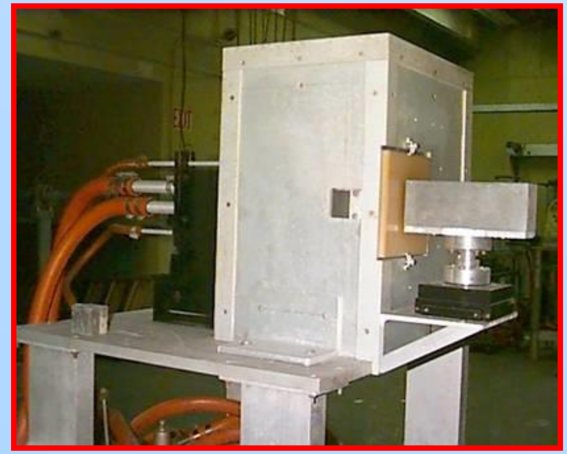
- **Time of flight measurements**
  - Resonance Region
    - Capture (0.01 eV – 2 keV)
    - Transmission (0.001 eV – 100 KeV)
    - Capture to fission ratio (alpha)
    - keV capture detector
    - Neutron scattering (E<0.5 MeV)
  - High energy (0.4-20MeV)
    - Scattering (30 m flight path)
    - Transmission (100m and 250m flight path)
    - Prompt Fission Neutron Spectra
  - High accuracy total cross section measurements using filtered beam
- **Lead Slowing Down Spectrometer**
  - Simultaneous measurement of fission cross sections and fission fragment mass and energy distributions using the RPI lead slowing down spectrometer
  - Measurements of energy dependent (n,p) and (n,α) cross sections of nanogram quantities of short-lived isotopes. (collaboration with LANL).
  - Capture cross section measurements
- **Other**
  - Research on medical isotope production
  - $S(\alpha,\beta)$ measurements (at SNS in ORNL)

Neutron Production Targets

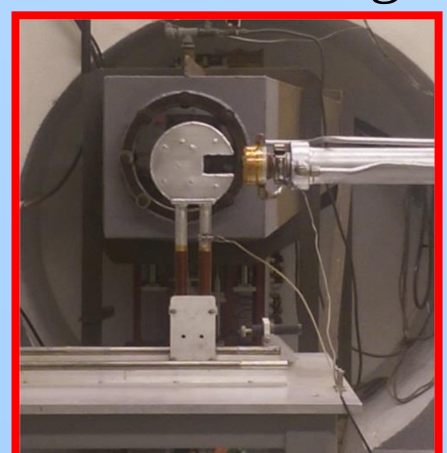
Bare Bounce Target (BBT)



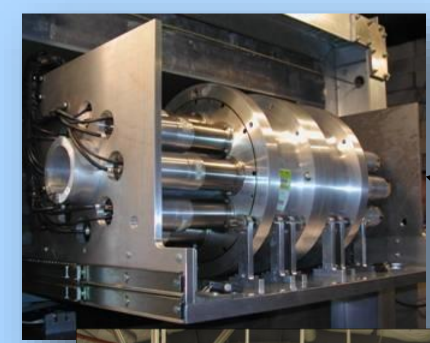
Enhanced Thermal Target (ETT)



PACMAN Target

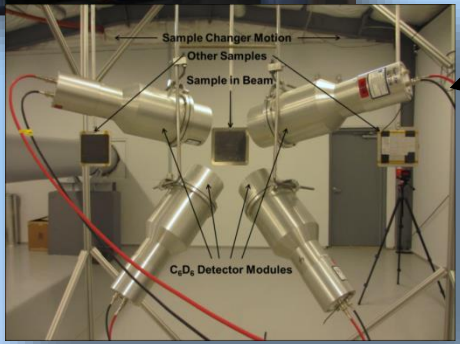


Detectors



25m

Capture/multiplicity

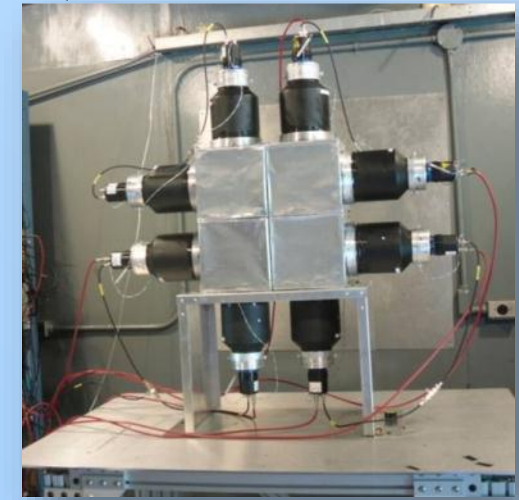


45m

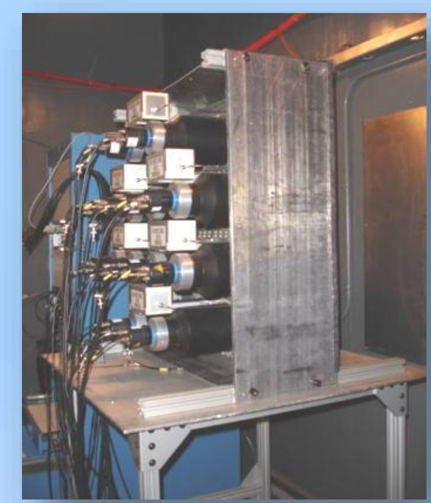
Transmission

15m- 30m

100m

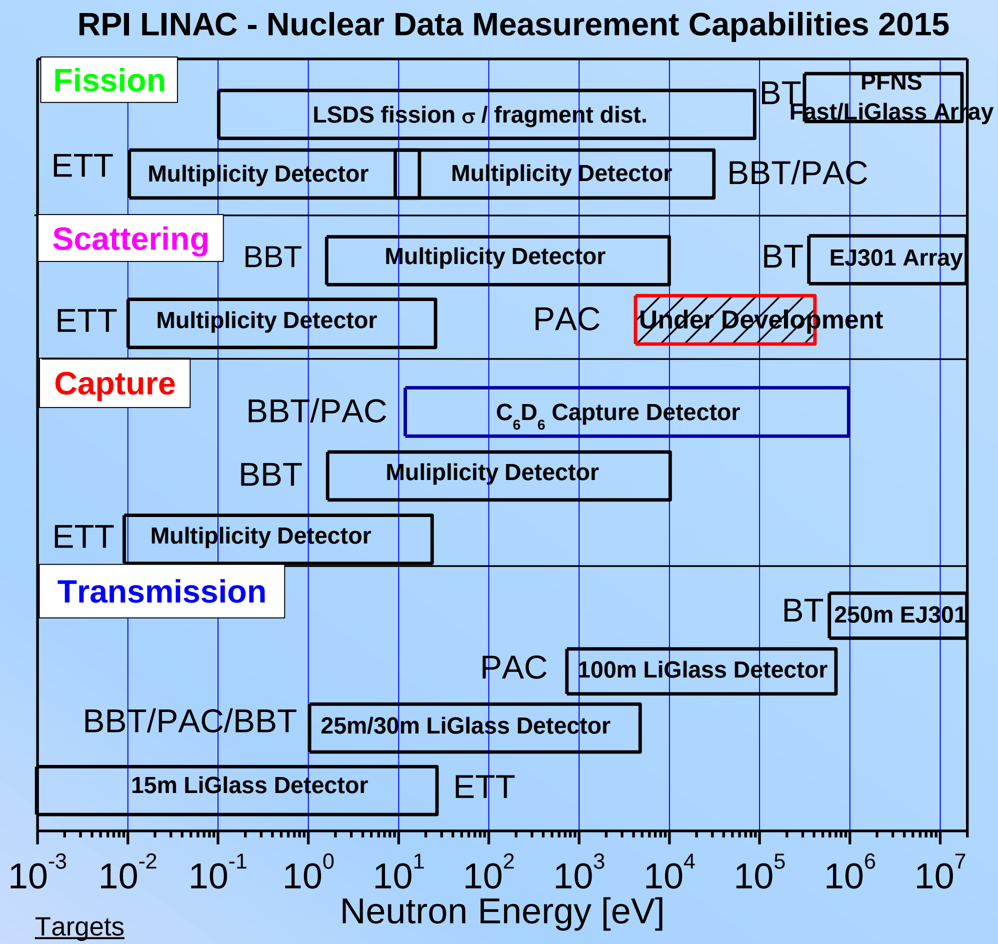


250m



Scattering
PFNS
30m

Capability Matrix and Development



<u>Targets</u>

ETT- Enhanced Thermal Traget

BBT - Bare Bounce Target

BT- Bare Target on Axis

PAC - PacMan Target

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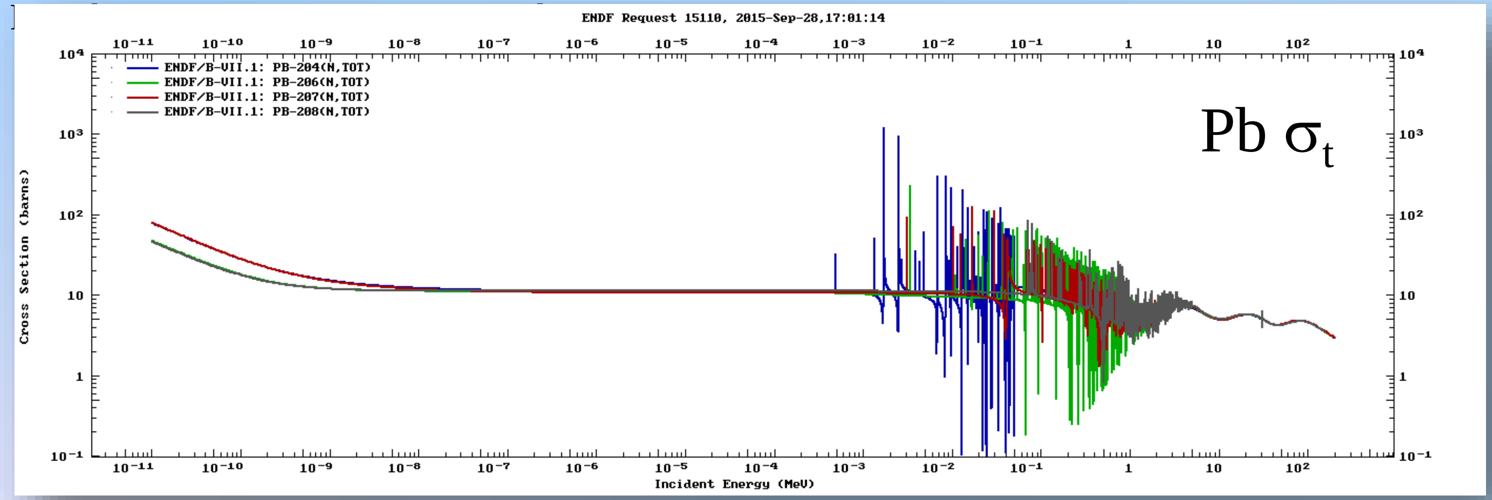
LSDS - Basic Physics

- The idea was developed in 1955 by Bergman et al.
- High purity Pb has low absorption cross section and the primary neutron reaction is scattering.
- Inelastic scattering cause faster (than elastic) neutron energy loss and then the neutron will keep losing energy by elastic collisions
  - The lowest inelastic threshold occurs for Pb-207 is 570 keV
- The energy loss for elastic collision with Pb (A=207) is small (~1% per collision)

$$E_n' - E_n = \frac{1-\alpha}{2} \qquad \text{where} \qquad \alpha = \left(\frac{A-1}{A+1}\right)^2$$

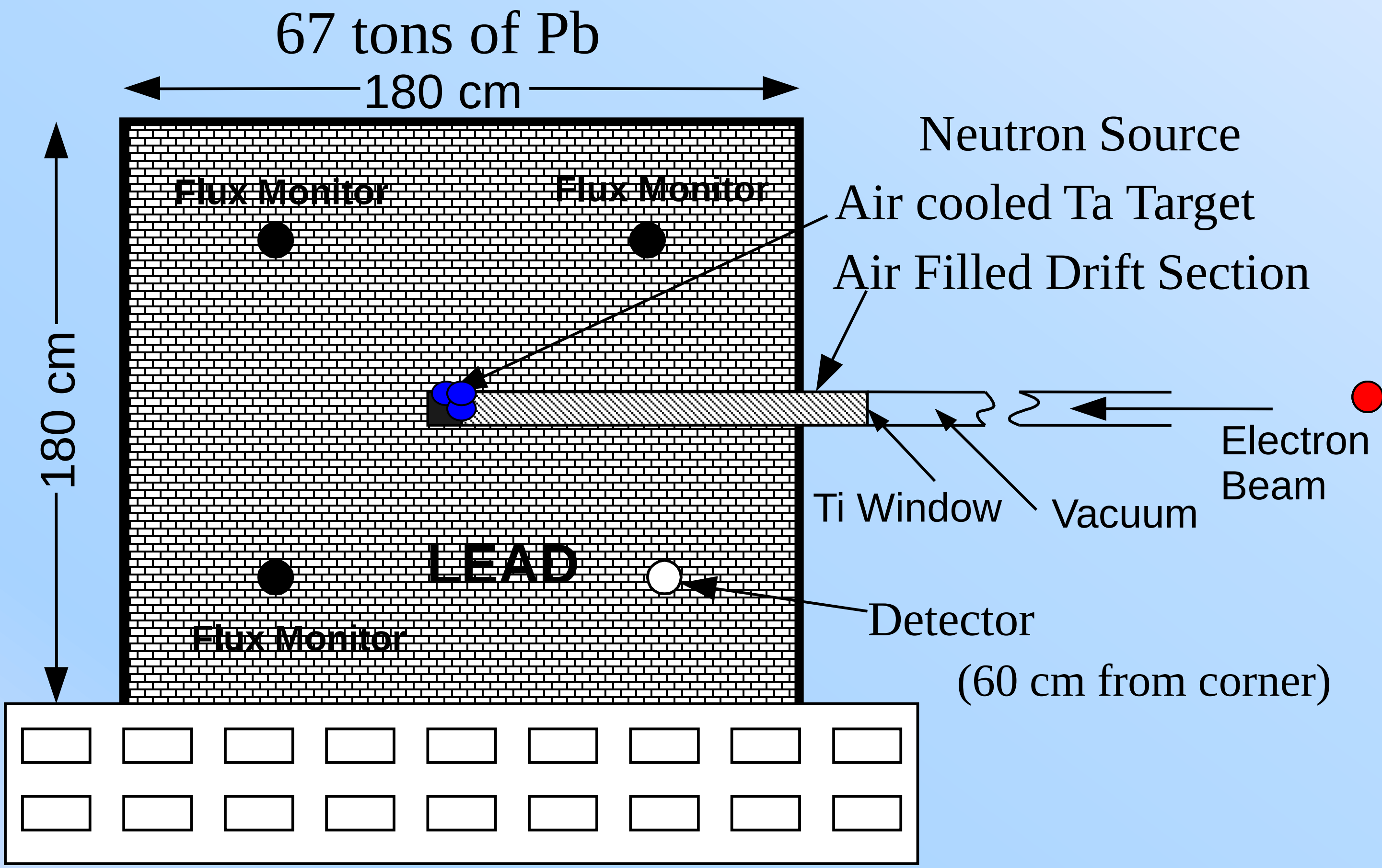


A. A. Bergman, a. I. Isacoff, i. D. Murin, f. L. Shapiro, i. V. Shtranikh, and m. V. Cazar-novsky, "A Neutron Spectrometer Based on Measuring the Slowing-Down Time of Neutrons in Lead", Proc. 1st Int. Conf Peaceful Uses Atomic Energy, Geneva, August 8-20, 1955, United Nations, New York (1955).

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

Number of Collisions in Pb

- Lethargy is defined as $u \equiv \ln\left(\frac{E}{E_0}\right)$. When the neutron loose energy u increases
- The average lethargy gain per collision can be derived to be:

$$\xi = 1 + \frac{\alpha}{1-\alpha}\ln\alpha$$

For Pb, ξ=0.00963

- For multiple collisions (n collision) the lethargy gain is: $\ln\left(\frac{E_0}{E}\right) = n\xi$
- Thus the number of collision when a neutron slows down form E_0 to E:

$$n = \frac{1}{\xi}\ln\left(\frac{E_0}{E}\right)$$

- For example for E_0=1 MeV and E=0.1 eV, ***n*=1674 collisions** (for D n=22)

- **The neutron will have many collision in the Pb which results in a high neutron flux**

Time-Energy Relation

- Given ξ and assuming constant cross section, the time it takes the neutron to slow down from energy E_0 to E can be derived from:

$$du = \xi \Sigma_s v dt$$

- With substitution $du=2dE/E$ the slowing down time can be found:

$$t = \frac{\sqrt{2m_n}}{\xi \Sigma_s}\left(\frac{1}{\sqrt{E}} - \frac{1}{\sqrt{E_0}}\right)$$

- The time energy relation can be obtained:

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

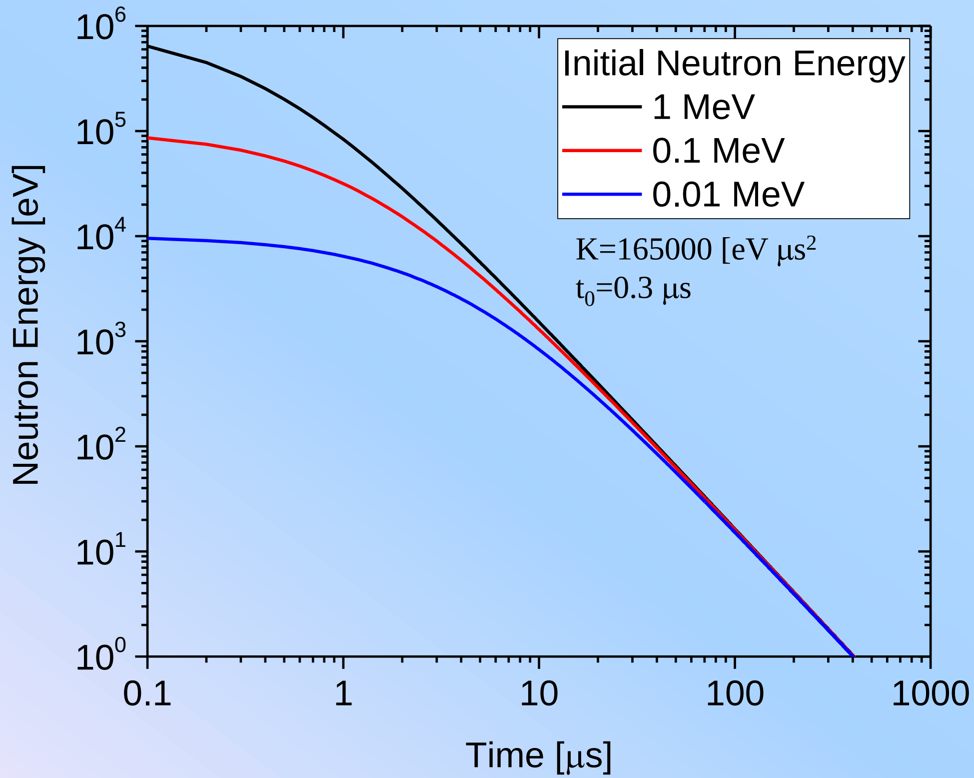
$$K \equiv \frac{2m_n}{\xi^2 \Sigma_s^2}$$

Where t is in [μs] and E in [eV]
t_0=correspond to some effective E_0

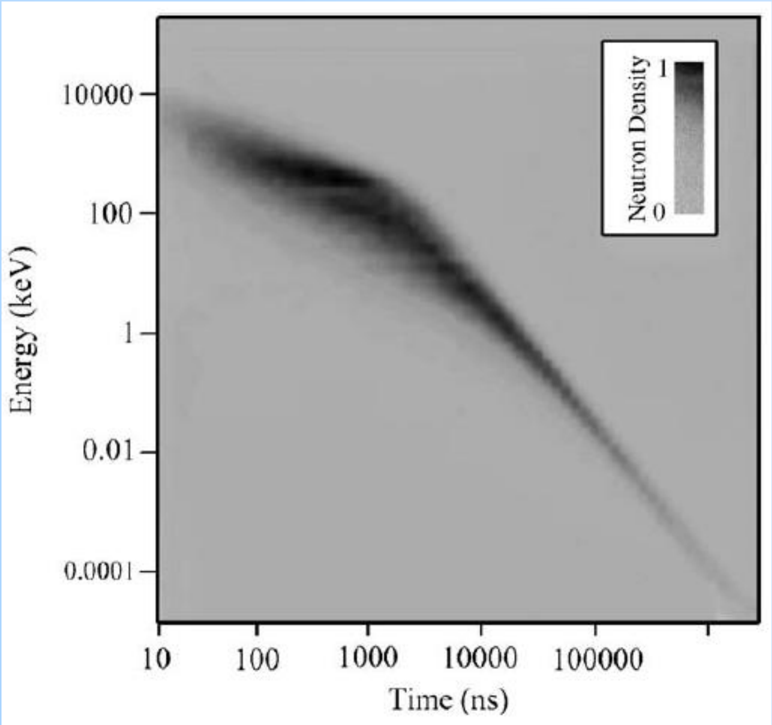
- At RPI we fine tune the constants based on measurements and use
  - K=165000 [eV μs^2] and t_0=0.3 μs
- For example for E=10 eV the slowing down time is: t=128.8 μs
- For E=100 keV the slowing down time is: t=1.5 μs

Energy Focusing

- Different incident energies slowdown at different rates but eventually converge to the same time-energy relation



MCNP simulation for the LANL LSDS



D. Rochman, R. C. Haight, J. M. O'Donnell, A. Michaudon, S. A. Wender, D. J. Vieira, E. M. Bond, T. A. Bredeweg, A. Kronenberg, J. B. Wilhelmy, T. Ethvignot, T. Granier, M. Petit, and Y. Danon, "Characteristics of a Lead Slowing-Down Spectrometer Coupled to the LANSCE Accelerator", *Nuclear Instruments and Methods in Physics Research A*, vol. **550**, Pages 397-413, (2005).

Flux Shape and Energy Resolution

- For infinite medium the neutron flux in the slowing down region is proportional to 1/E
- The energy resolution was derived by Bergmann 1995 as:

$$\left(\frac{\Delta E}{E}\right)_{FWHM} = \left[8\ln(2)\left(\frac{8}{3A} + 4D_0\frac{E}{E_0} + \frac{kT}{E}\right)\right]^{\frac{1}{2}}$$

- Which gives:

$$\left(\frac{\Delta E}{E}\right)_{FWHM} = \left[0.720 + \frac{0.140}{E} + 6.59\times10^{-5}E\right]^{\frac{1}{2}}$$

- For the RPI LSDS the flux shape and energy resolution were obtained from MCNP simulation of the LSDS
- Neutron flux shape:

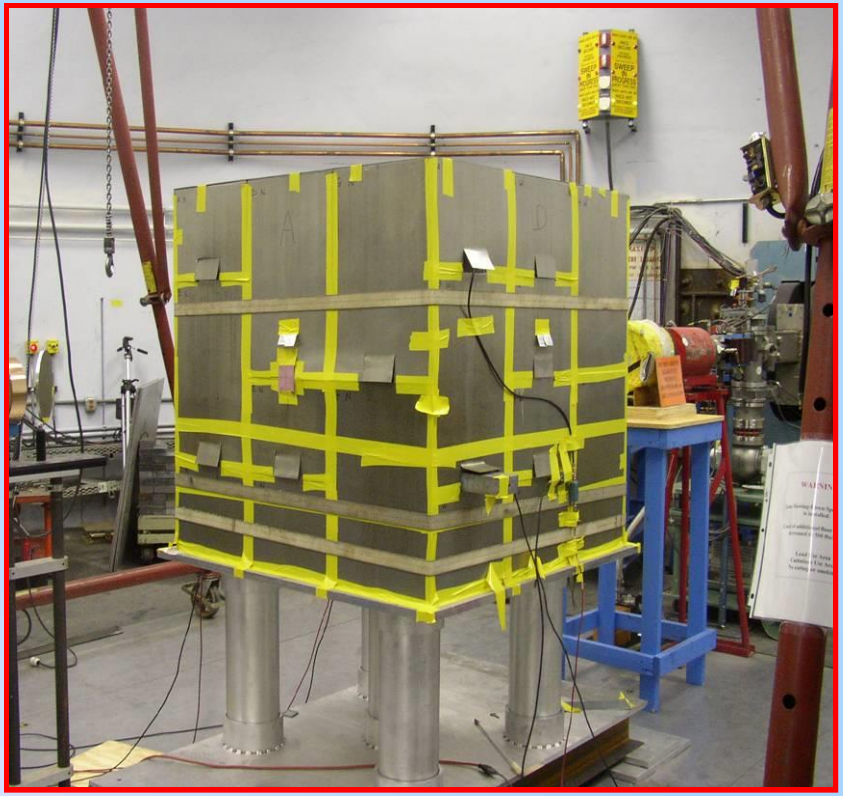
$$\phi(E) = E^{-0.776}\exp\left[-\sqrt{\frac{0.214}{E}}\right]$$

- Energy Resolution:

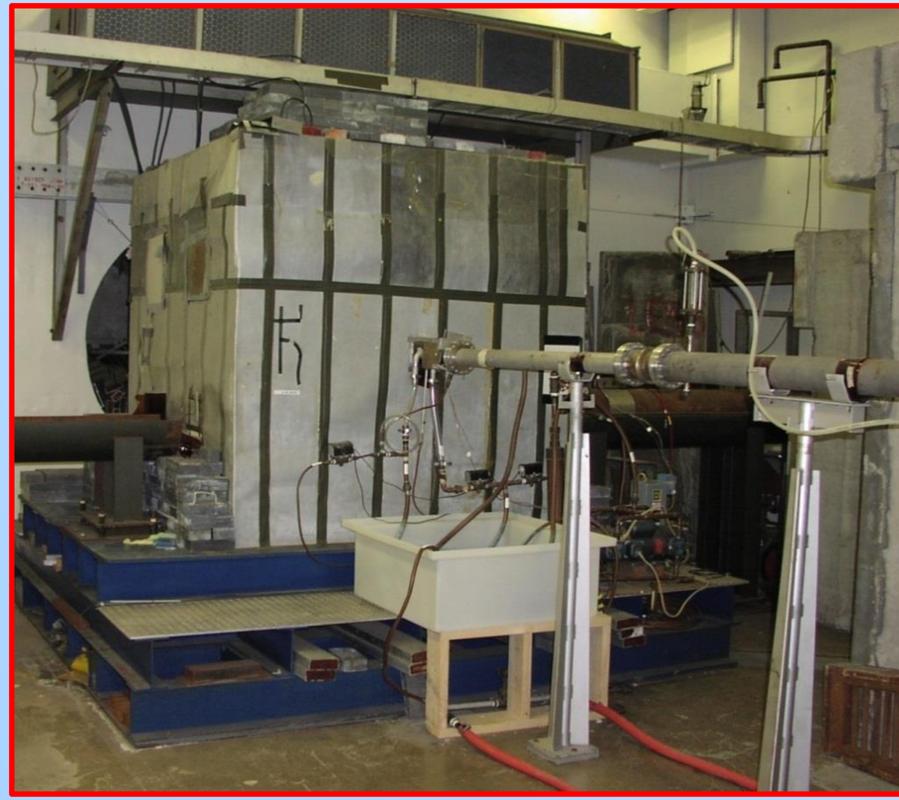
$$\left(\frac{\Delta E}{E}\right)_{FWHM} = \left[0.0835 + \frac{0.128}{E} + 3.05\times10^{-5}E\right]^{\frac{1}{2}}$$

- *The flux and energy resolution can change locally near the detector, if the detector is absorbing or moderating*

Lead Slowing Down Spectrometers



***LANL** – Proton Driven*



***RPI** –Electron Driven*

Two others:

- Russia (Institute for Nuclear Research, Russian Academy of Sciences, Moscow)
- Japan (Kyoto University)

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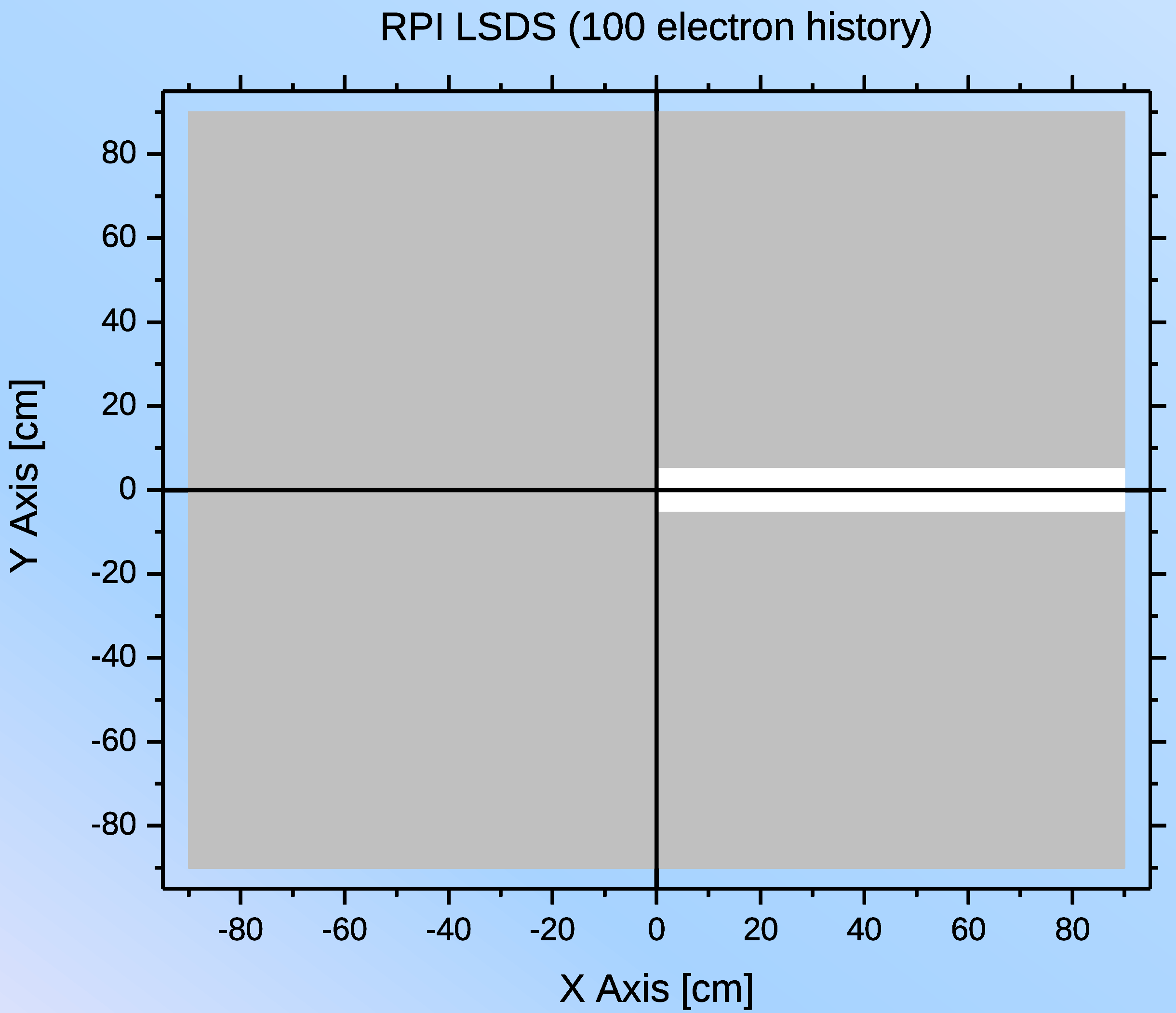
MCNP simulation RPI LSDS - 100 electrons



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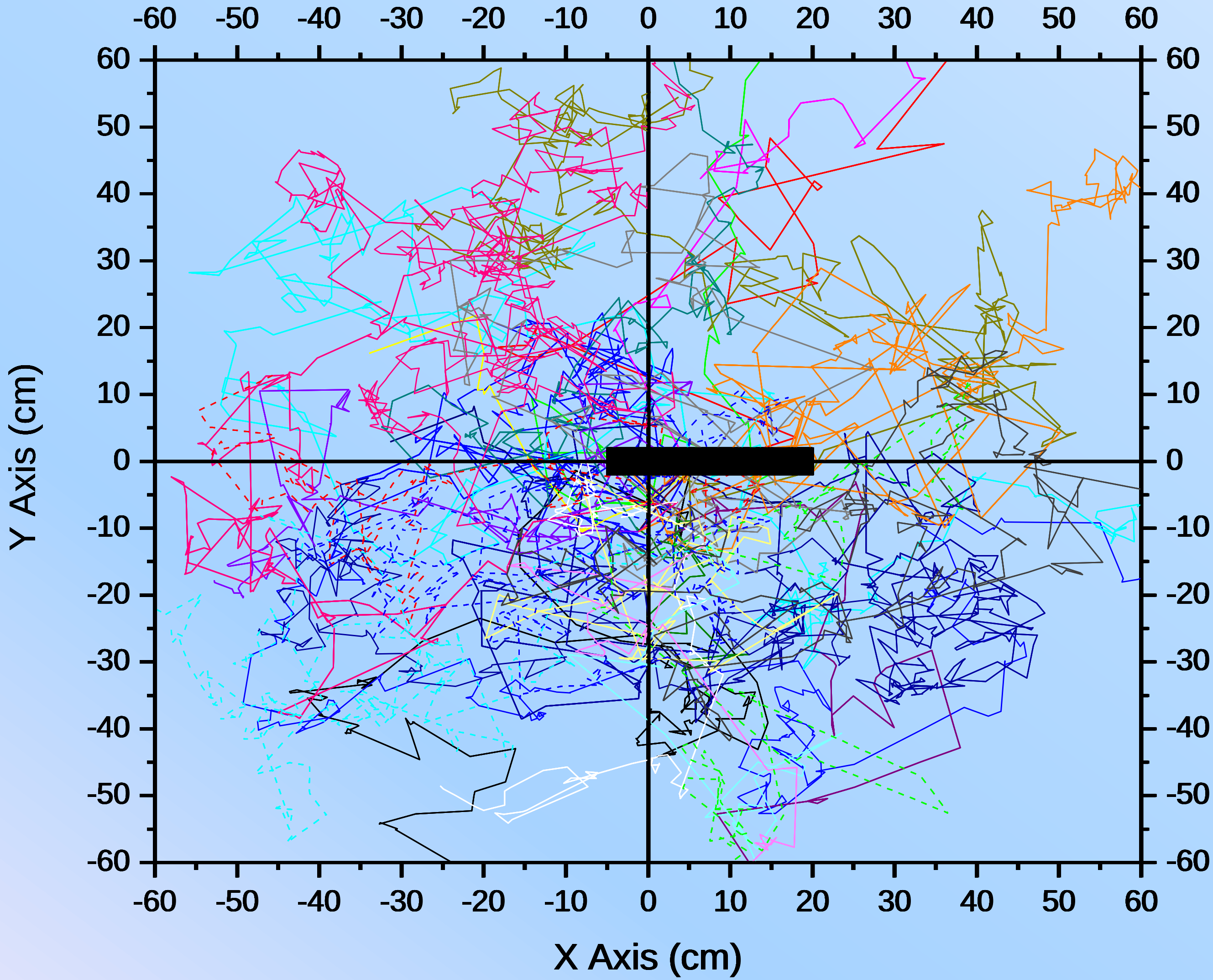
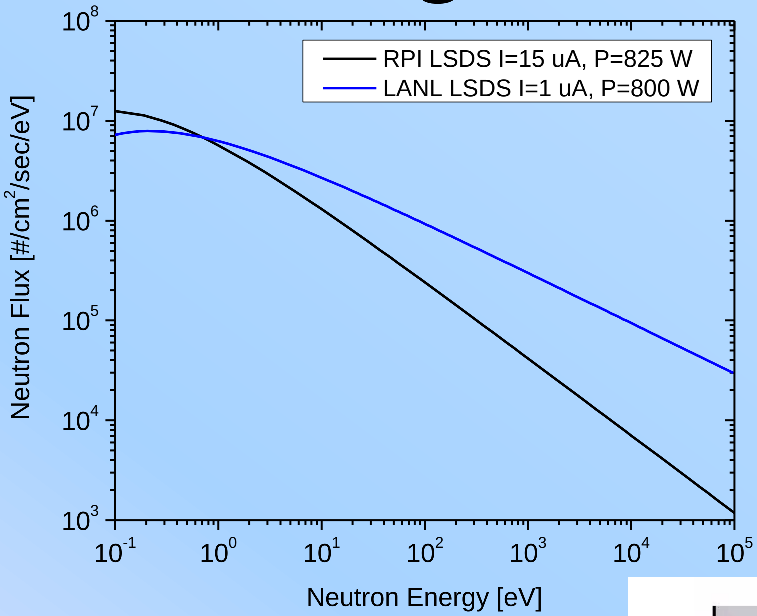
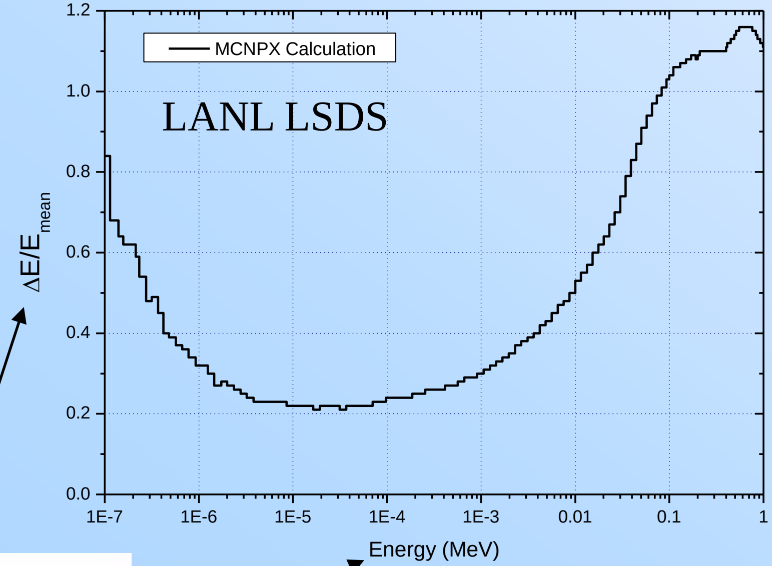
MCNPX Calculations LANL LSDS 1 proton

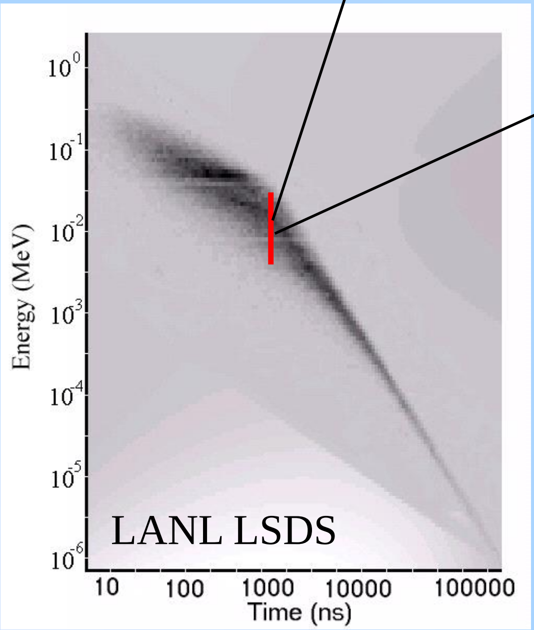


Slowing -Down-Time vs. Energy Relation





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

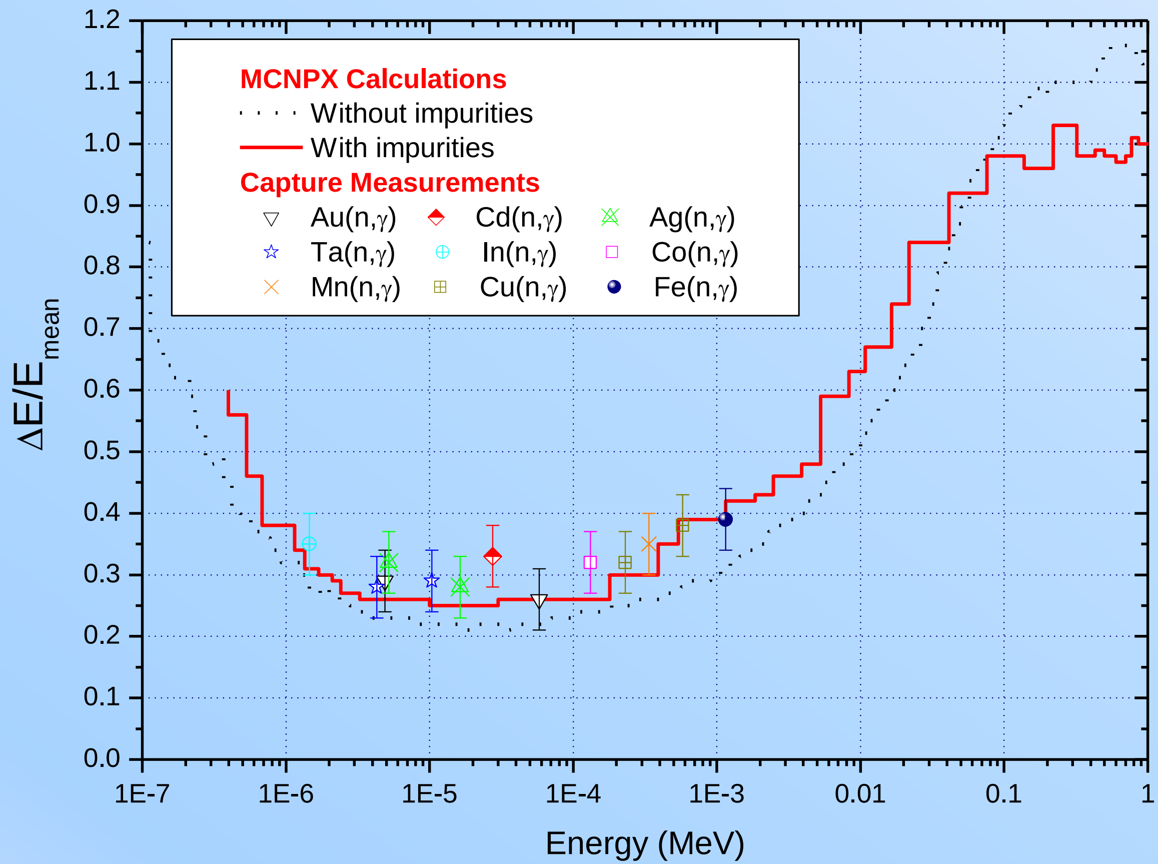


$\overline{E}$

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

Energy Resolution (LANL LSDS)

Foil	Resonance [eV]
$^{115}In(n,\gamma)$	1.5
$^{197}Au(n,\gamma)$	4.9
$^{181}Ta(n,\gamma)$	4.3 10.4
$Ag(n,\gamma)$	5.2 16.3
$Cd(n,\gamma)$	27.5
$^{59}Co(n,\gamma)$	132
$Cu(n,\gamma)$	230 579
$^{55}Mn(n,\gamma)$	336
$^{56}Fe(n,\gamma)$	1150



MCNPX: $K = 161 \pm 5$ keV·μsec^2, $t_0 = 0.4 \pm 0.1$ μsec

Measurements: $K = 161 \pm 1$ keV·μsec^2, $t_0 = 0.4 \pm 0.1$ μsec

RPI LSDS $K = 165$ keV·msec^2, $t_0 = 0.3$ μsec

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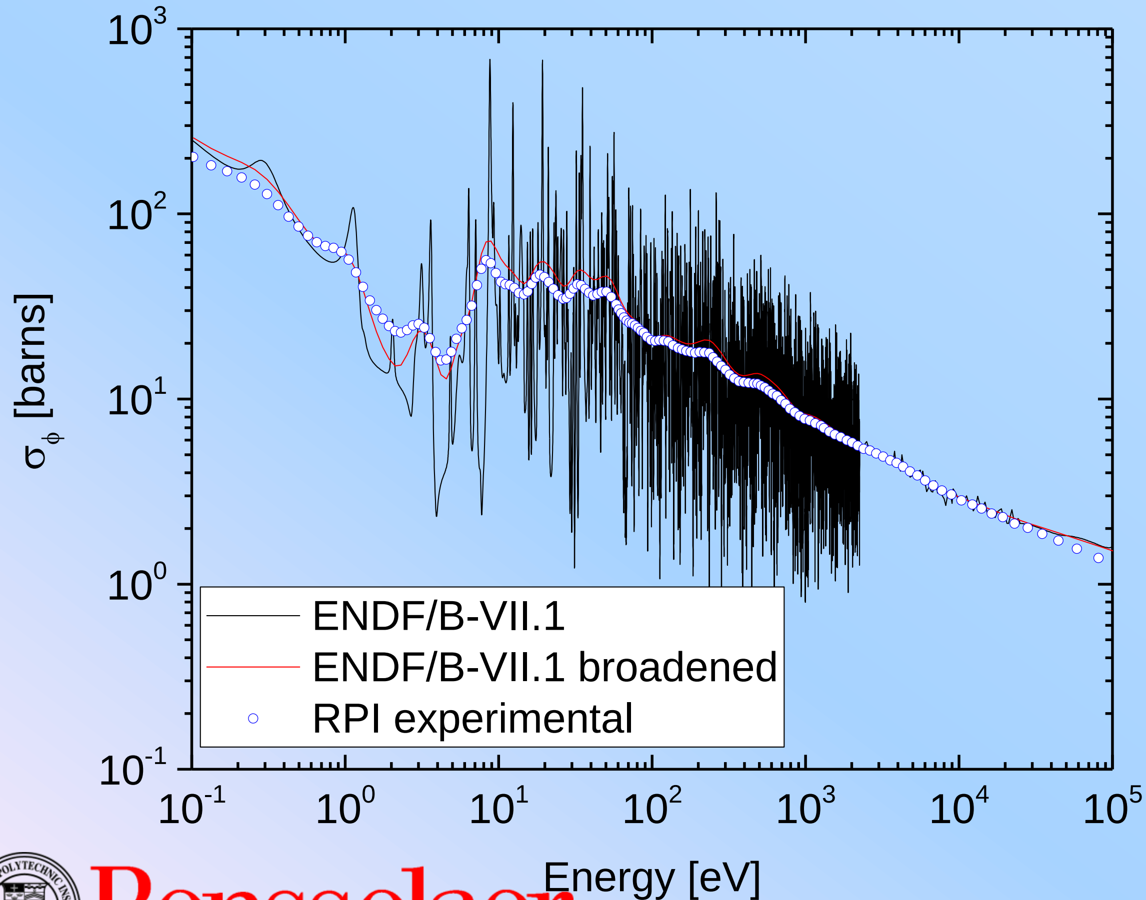
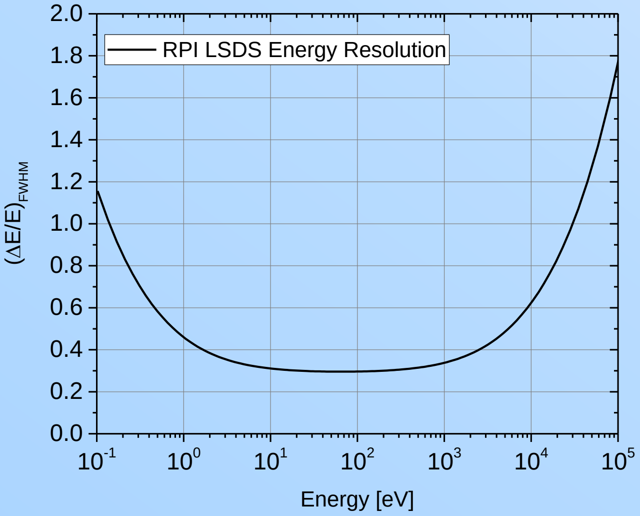
The Effect of Resolution Broadening

- Convoluting the resolution function with the ENDF/B-VII.1 cross section give the expected LSDS measured cross section
- Other effect (neutron return, local perturbations) result is some differences





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What Can You Do With a LSDS?

- Measure neutron interaction cross sections
  - Fission of small samples (ng to μg):
    - ^{239}Pu, ^{242}Cm, ^{245}Cm, ^{247}Cm, ^{250}C, ^{254}Es
    - Sub threshold fission ^{238}U, ^{232}Th, ^{238}Pu
  - Capture reaction rates
  - Fission fragment mass and energy distributions
    - ^{239}Pu, ^{235}U
  - (n,α) cross section for ^{147,149}Sm.
- Assay spent nuclear fuel
  - Demonstrated in Germany and RPI

Other Ideas are Welcome

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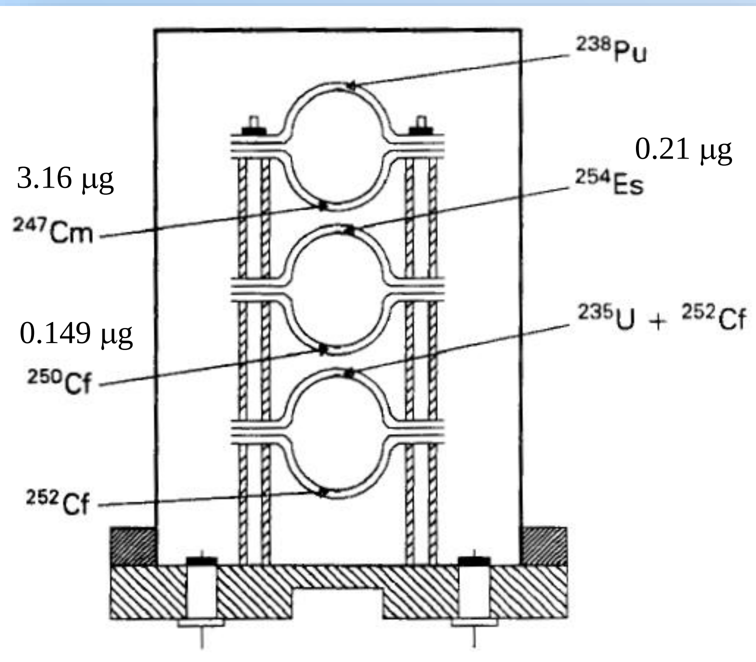
Fission Cross Section Measurements

Fission Cross Section Measurements at RPI

- Use a hemispherical fission chamber to reduce the effect of gamma pile up
- Use fast preamplifier and shaper to reduce the dead time due to gamma flash
- The LINAC is pulsed 90 Hz using a pulse width of 200ns and total power of about 900W
- Counts (C_i) are recorded as a function of the slowing down time (sdt)
- The cross section can be calculated from:



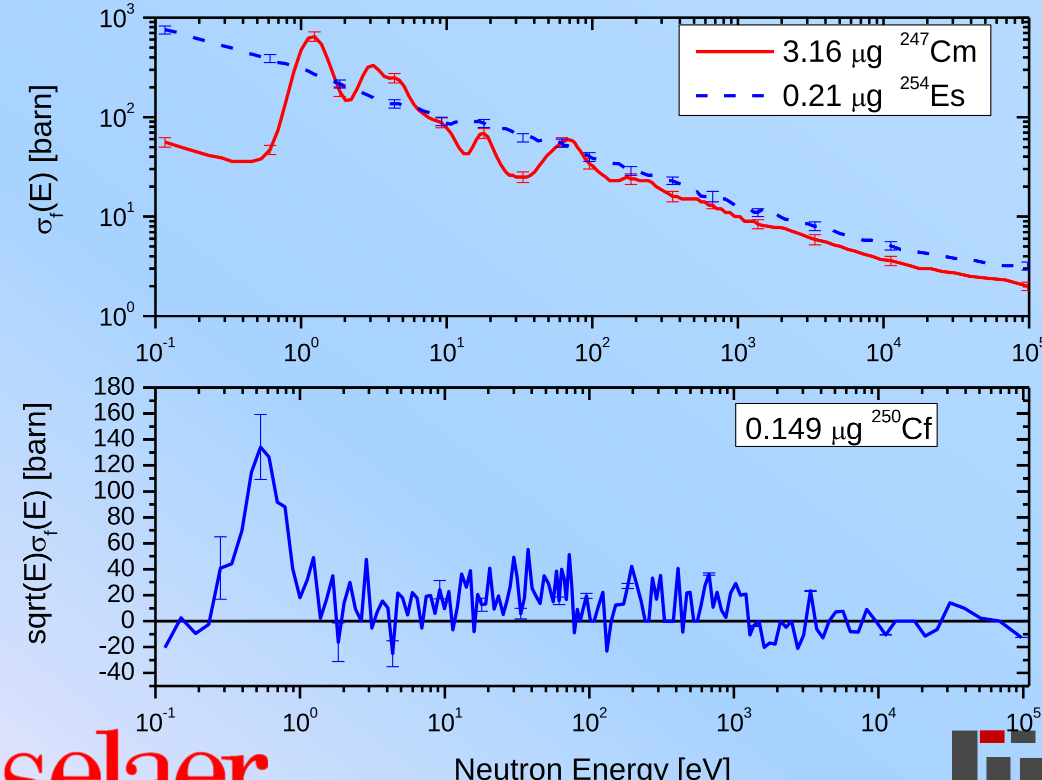
$$\sigma_f(E_i) = \frac{1}{\eta N} \frac{C_i}{p\phi_r(E_i)\Delta E_i}$$

Where:
E_i – neutron energy corresponding to sdt channel I
ΔE_i – energy width of channel i
η - the fission detection efficiency
N – no. of atoms in the sample
$\phi_r(E_i)$ – neutron flux shape
p – normalization factor elative to ^{235}U

Y. Danon, R.E. Slovacek, R.C Block, R. W. Lougheed, R. W. Hoff, M. S. Moore, "Fission Cross-Section Measurements of ^{247}Cm, ^{254}Es, and ^{250}Cf from 0.1 eV to 80 keV", *Nuclear Science and Engineering*, Vol. **109** (4) 341-349 (1991).

Fission Cross Section Measurements at RPI Results

- The ^{254}Es cross section is very interesting since no resonance structure was observed but the cross section is very high.
  - The experiment was repeated at LANCE (LANL) with a fresh sample giving the sample result.
  - The thermal cross section 1749±110 agreed with Halperin et al. 1985.



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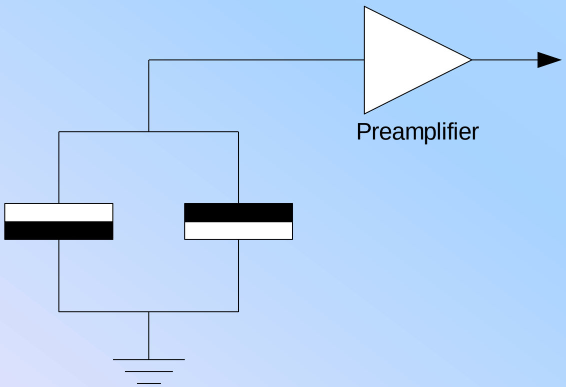
Compensated Solar Cell Detectors

- At LANL compensated detector we required due to large flash effect at t=0
- Use compensated solar cells:
  - Fission Detection
  - Alpha Detection
- Two setups were used

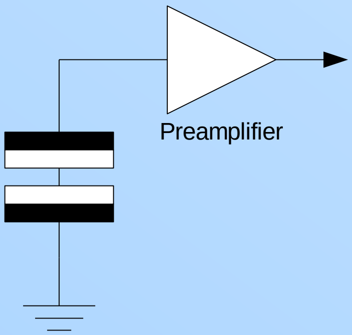
Parallel



Principle of operation

- The sample is on one of the detectors
- During the accelerator pulse both detector respond but because of the inverted polarity they provide zero net current
- Event at the sample is not compensated and will reach the preamplifier

Series (back to back)

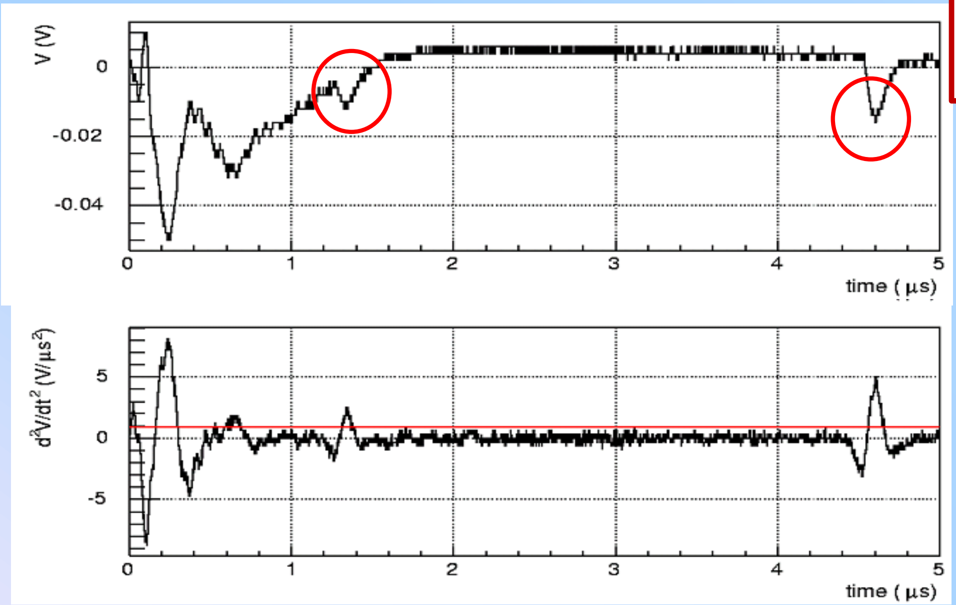


M. Petit, T. Ethvignot, T. Granier , R.C. Haight, J.M. O'Donnell, D. Rochman , S.A. Wender, E.M. Bond, T.A. Bredeweg, D.J. Vieira, J.B. Wilhelmy and Y. Danon "A Compensated Fission Detector Based On Photovoltaic Cells", *Nuclear Instruments and Methods in Physics Research A*, **554**, 340346 (2005).

Digital Data Acquisition

- Use Acqiris DC265
- Sampling rate was set to 50 ns
- In the current runs we digitized a discriminator signal that is wider than the sample interval.
- Advantages - digital filtering

D. Rochman, R. C. Haight, S. A. Wender, J. M. O'Donnell, A. Michaudon, K. Huff, D. J. Vieira, E. Bond, R. S. Rundberg, A. Kronenberg, J. Wilhelmy, T. A. Bredeweg, T. Ethvignot, T. Granier, M. Petit, Y. Danon, “First Measurements with a Lead Slowing-Down Spectrometer at WNR, International conference on nuclear data for science and technology (ND2004)”, Santa Fe, NM, Sep. 26 – Oct. 1, (2004)



Double Derivative
+
Smoothing

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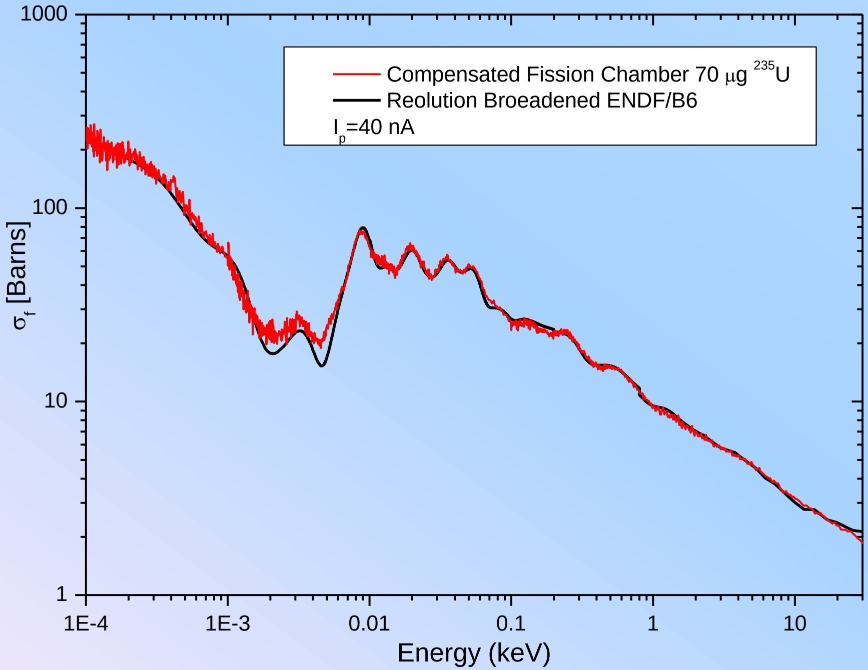
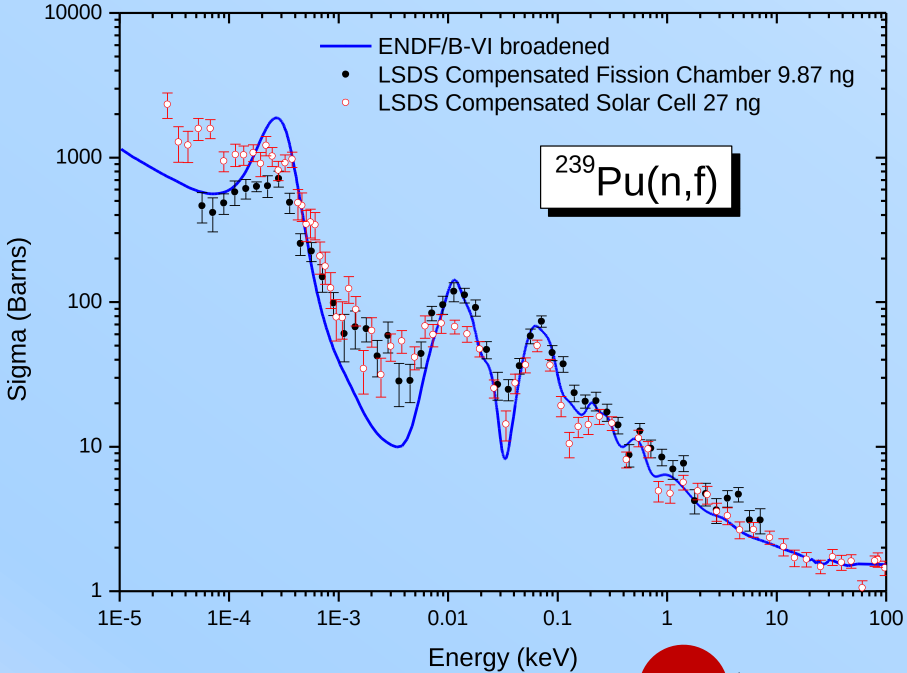
Fission Measurements at LANL - Results

- Develop capabilities
  - Development of compensated detectors
  - Measured ^{235}U, ^{238}U and ^{239}Pu
  - Smallest sample measured- 9.87 ng of ^{239}Pu

^{235}U



^{239}Pu



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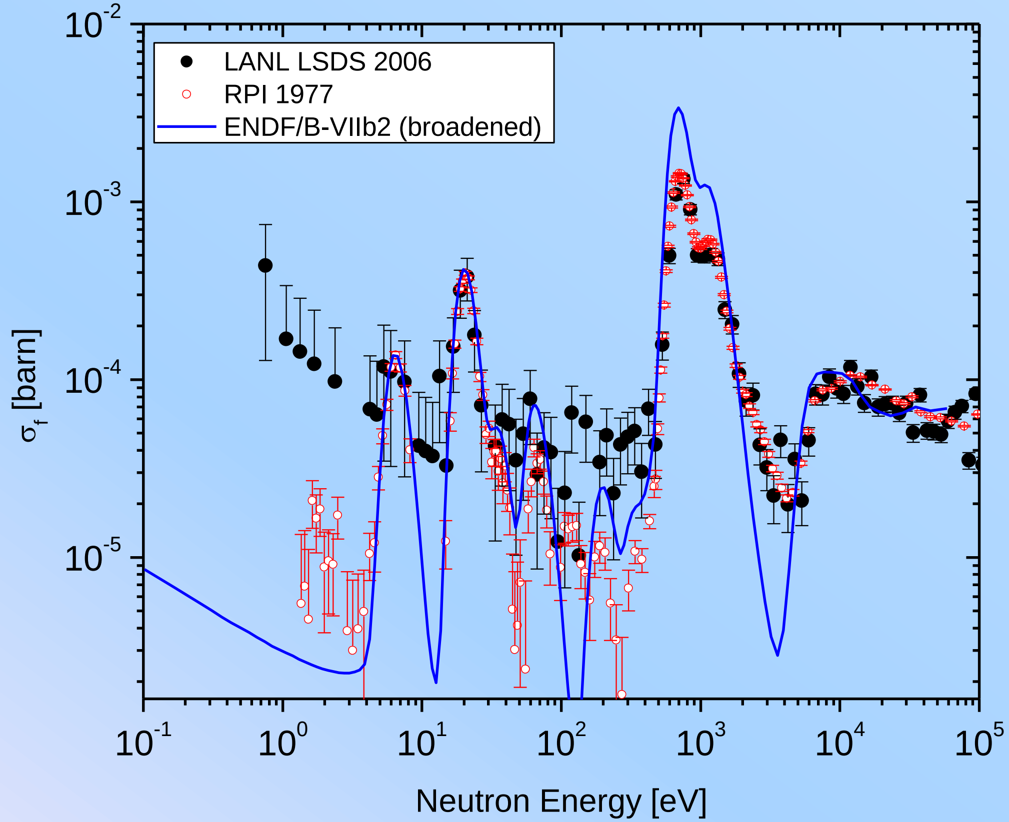
Subthreshold fission in ^{238}U

- At LANL used 3 mg sample with ~1ppm ^{235}U.
- Use a compensated back to back solar cell



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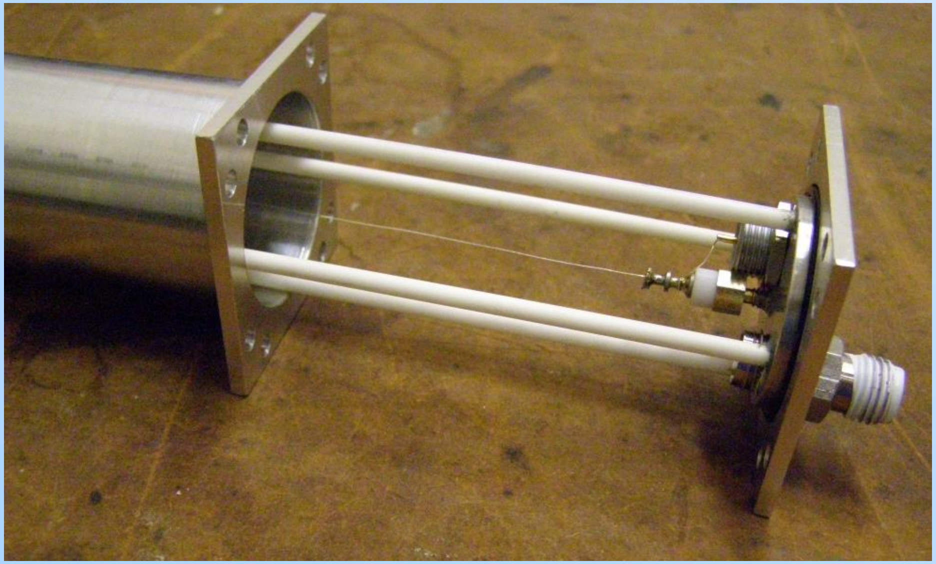
(n,α) Cross Section Measurements

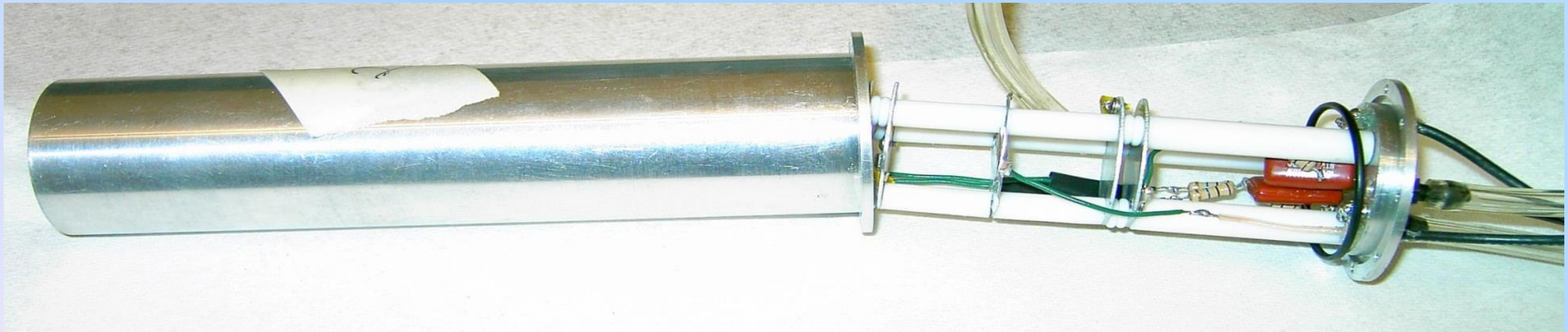
(n,a) Cross Section Measurements

- The problem is small signal relative to fission and the need for fast recovery time after the gamma flash
  - Alphas have energy of a few MeV compared to fission fragment than have energy of tens of MeV
- Examined different detector concepts:
  - Parallel plate ionization chamber
  - Gas Electron Multiplier (GEM) Foil Detector
  - Diamond Detectors
  - Solar Cell Detectors
  - Passivated Implanted Planar Silicon (PIPS) Detectors

Cylindrical and Parallel Plate Ion Chamber

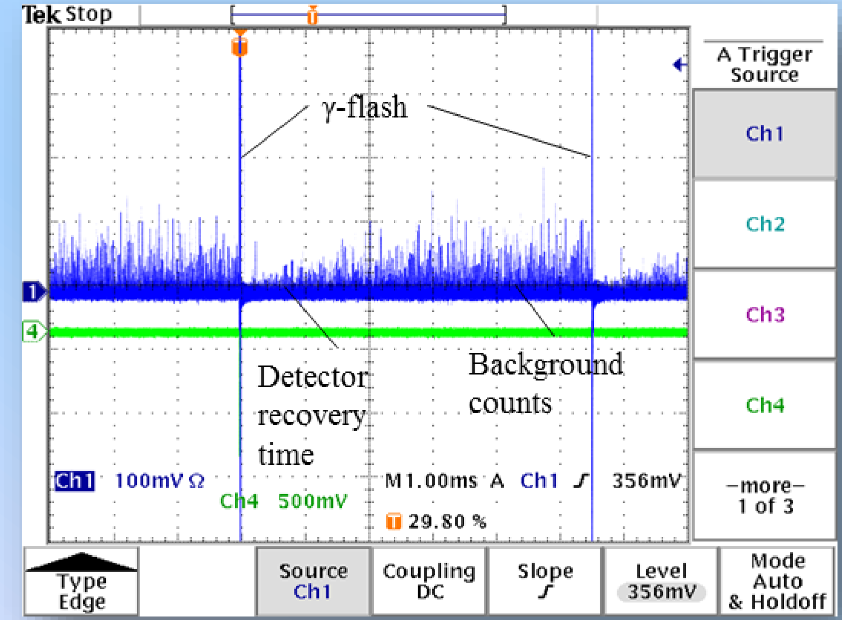
- ☑ Simple design
- ☒ Slow recovery



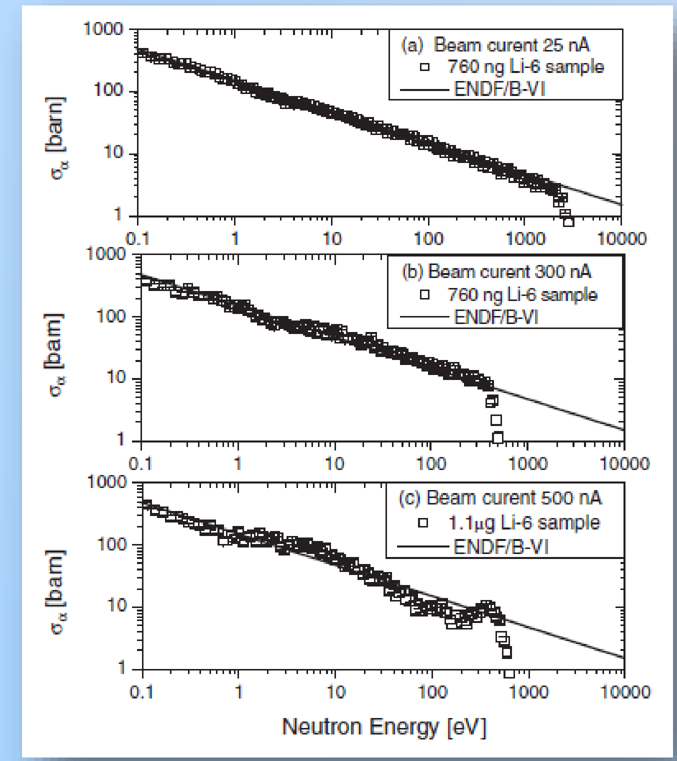


Measurements with Cylindrical Ion Chamber

Cylindrical Ion Chamber



Parallel Plate Ion Chamber



C. Romano, Y. Danon, R. C. Haight, S. A. Wender, D. J. Vieira, E. M. Bond, R. S. Rundberg, J. B. Wilhelmy, J. M. O'Donnell, A. F. Michaudon, T. A. Bredeweg, D. Rochman, T. Granier and T. Ethvignot, "Measurements of (n,α) Cross-Section of Small Samples Using a Lead-Slowing-Down-Spectrometer," Nuclear Instruments and Methods in Physics Research Section A, vol. 562, no. 2, pp. 771-773, 2006.

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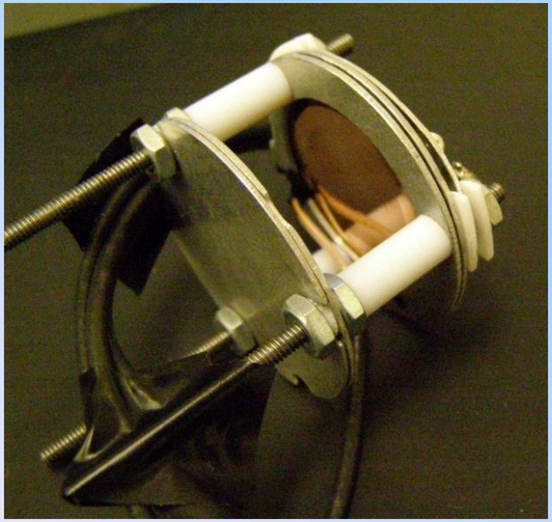
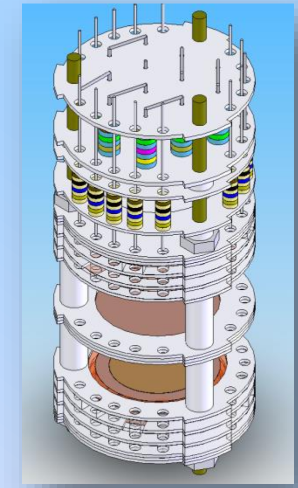
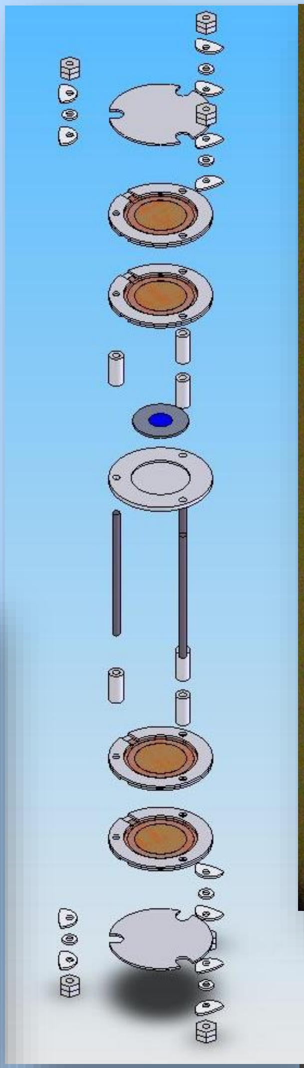
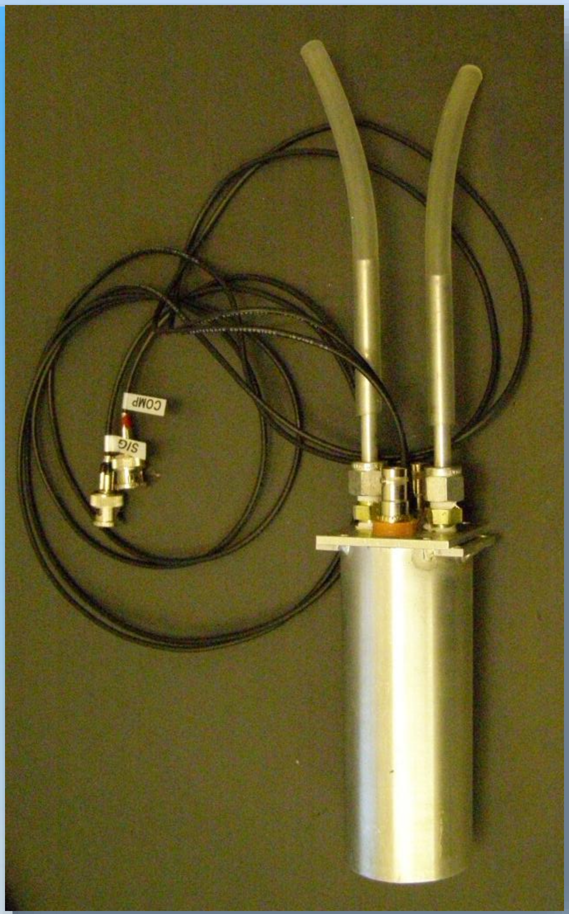
Gas Electron Multiplier (GEM) Foil Detector

- ☑ Fast
- ☑ High internal gain
- ☑ Low noise
- ☒ Complicated design



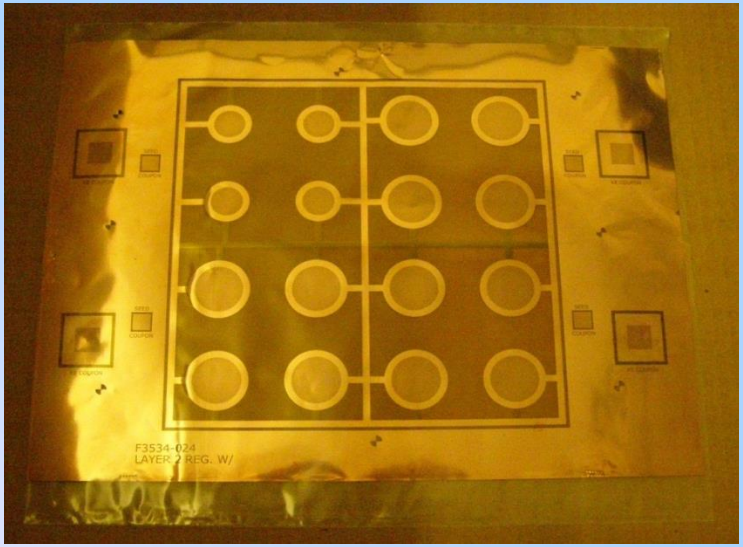


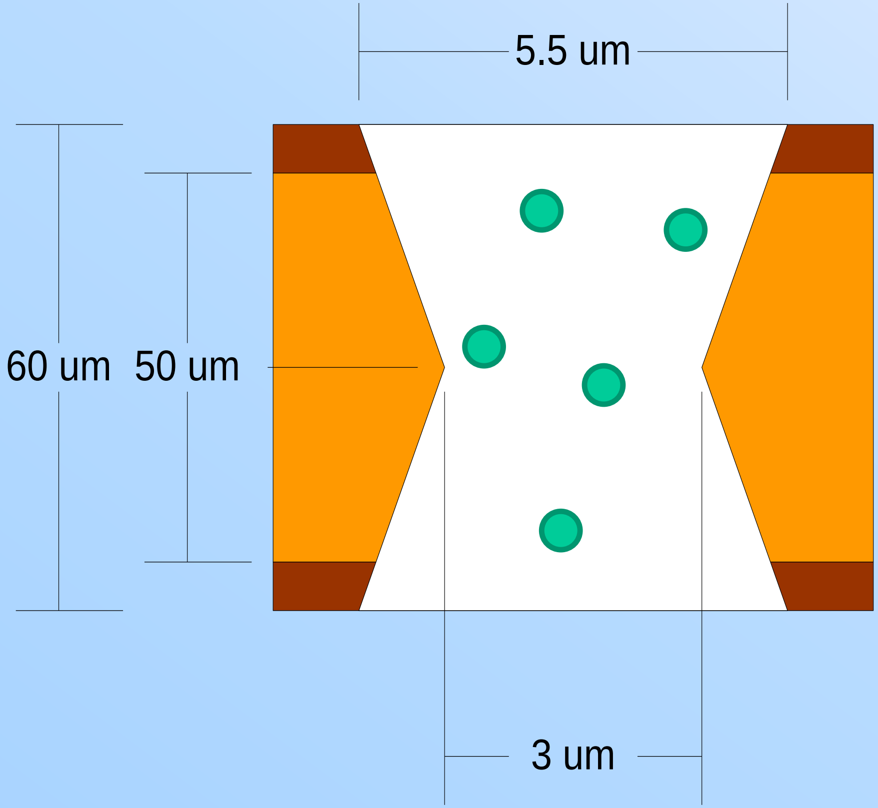




GEM Foil Principles

- 300 – 500 V potential difference across the foil
- Gains of 10^3 - 10^4 [55]





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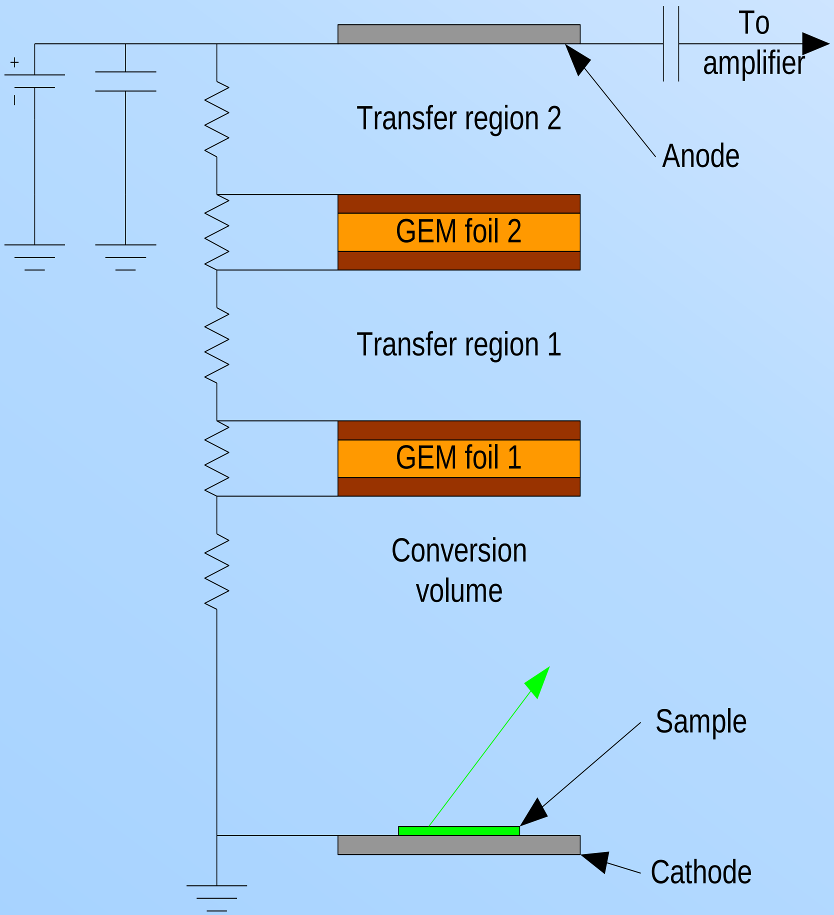
GEM Detector Construction

- α particles or protons generate ion/electron pairs in the conversion volume
- Conversion and transfer regions are operated at a potential that maximizes the electron drift velocity
- Signals are amplified in the GEM foils



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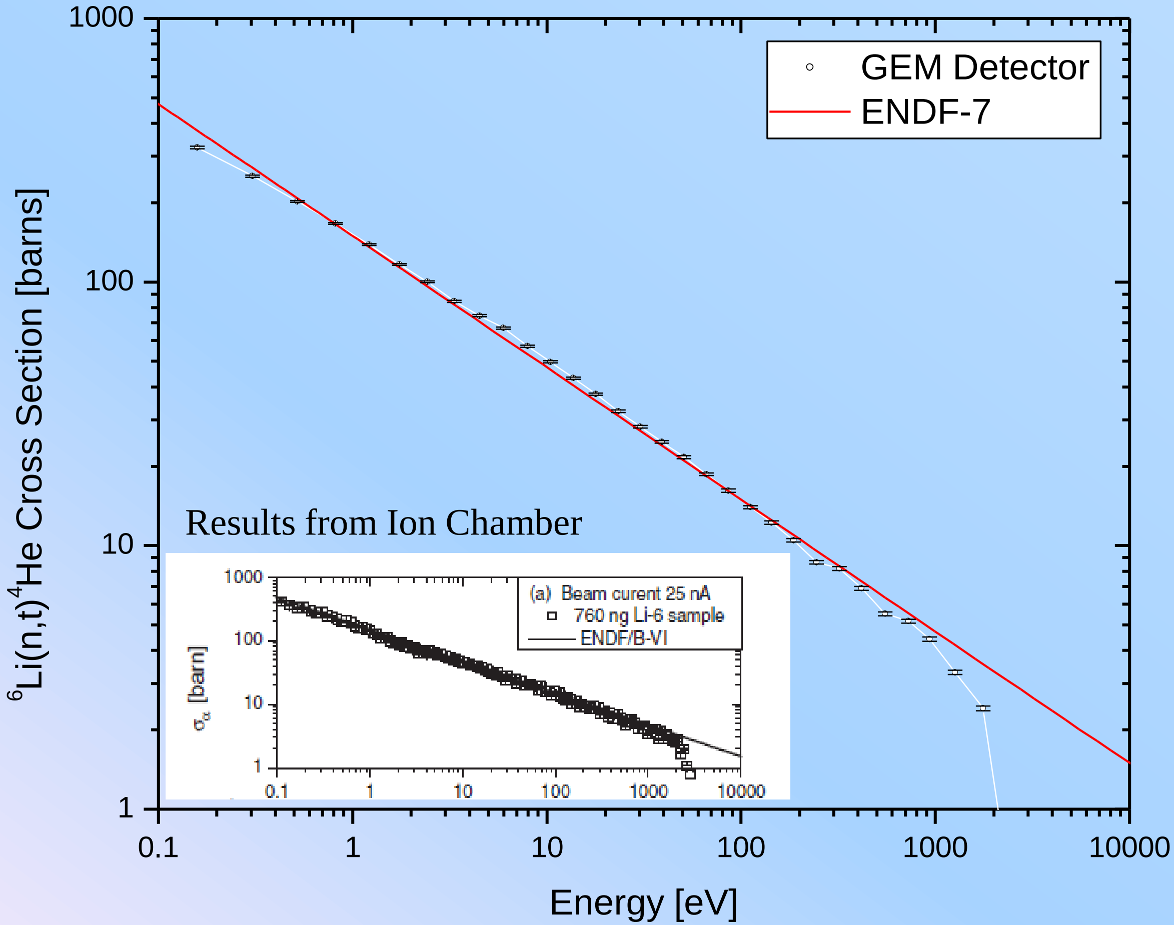
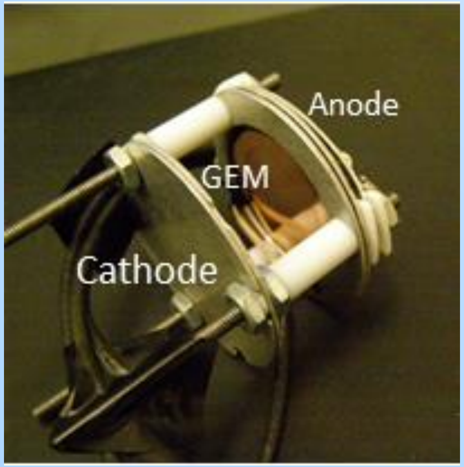
GEM Detector Results





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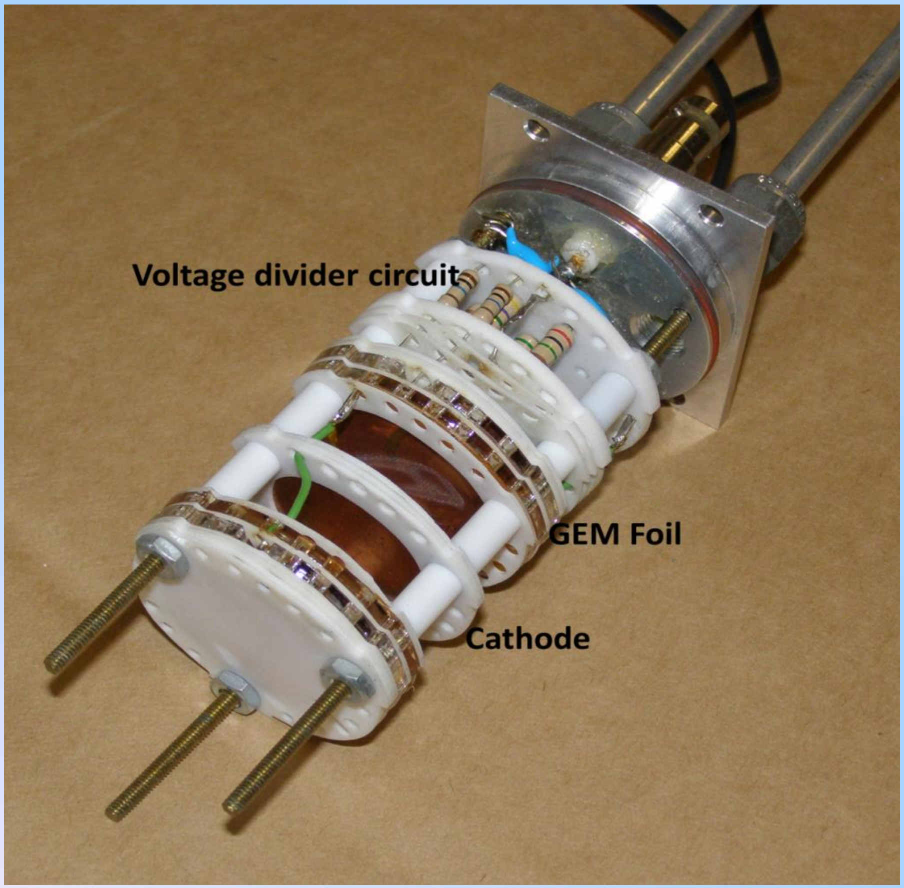
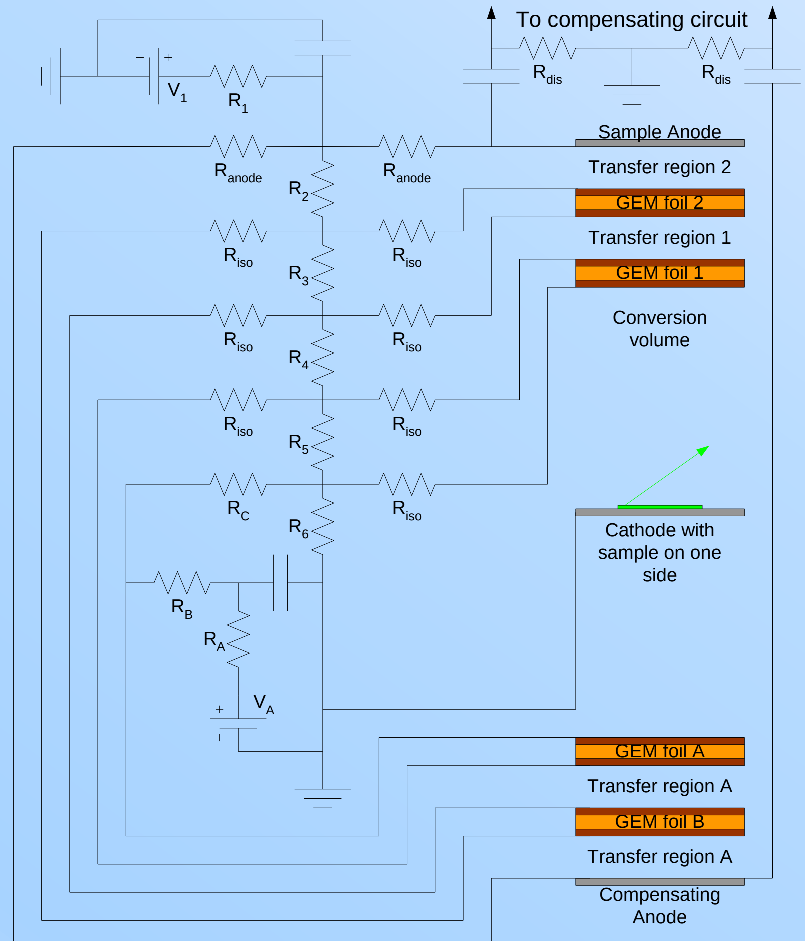
Final Compensated GEM Foil Detector Design





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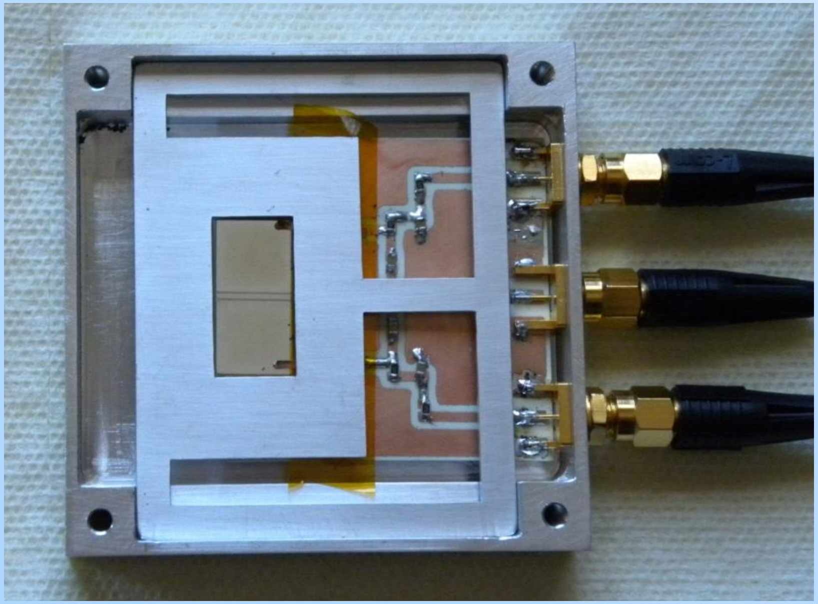
GEM Final Remarks

- GEM detector has a clean signal and a fast recovery time.
- However, if constructed in the wrong environment, it can become contaminated with dust and become prone to sparking.
  - Decrease the internal gain / operate at lower voltages
  - Add a third GEM foil
- Make the conversion volume thinner to make the detector even faster.
- Its limited width and low density would also need to be addressed before (n,p) reactions could be measured.

Compensated Diamond Detector

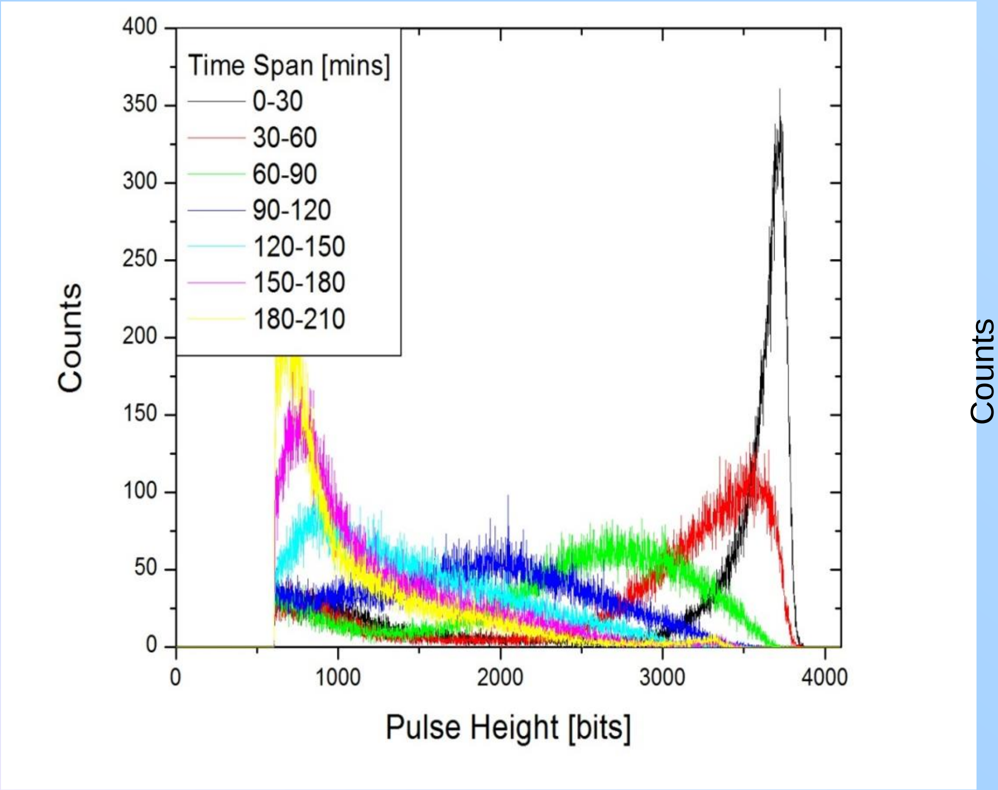
- ☑ Dense/Thin
- ☑ Speed is on par with solid state detectors
- ☑ Good signal-to-noise ratio
- ☒ Commercial – but more problematic than originally thought
- ☒ Charge polarization phenomena

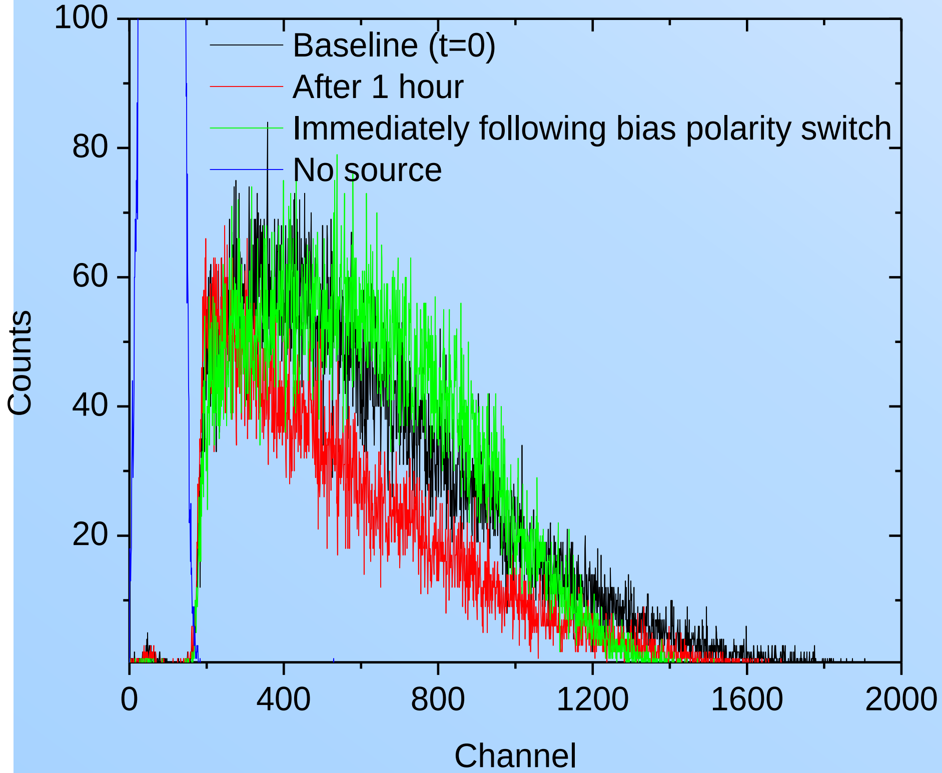


Polycrystalline compensated diamond detector

Charge Polarization Phenomena

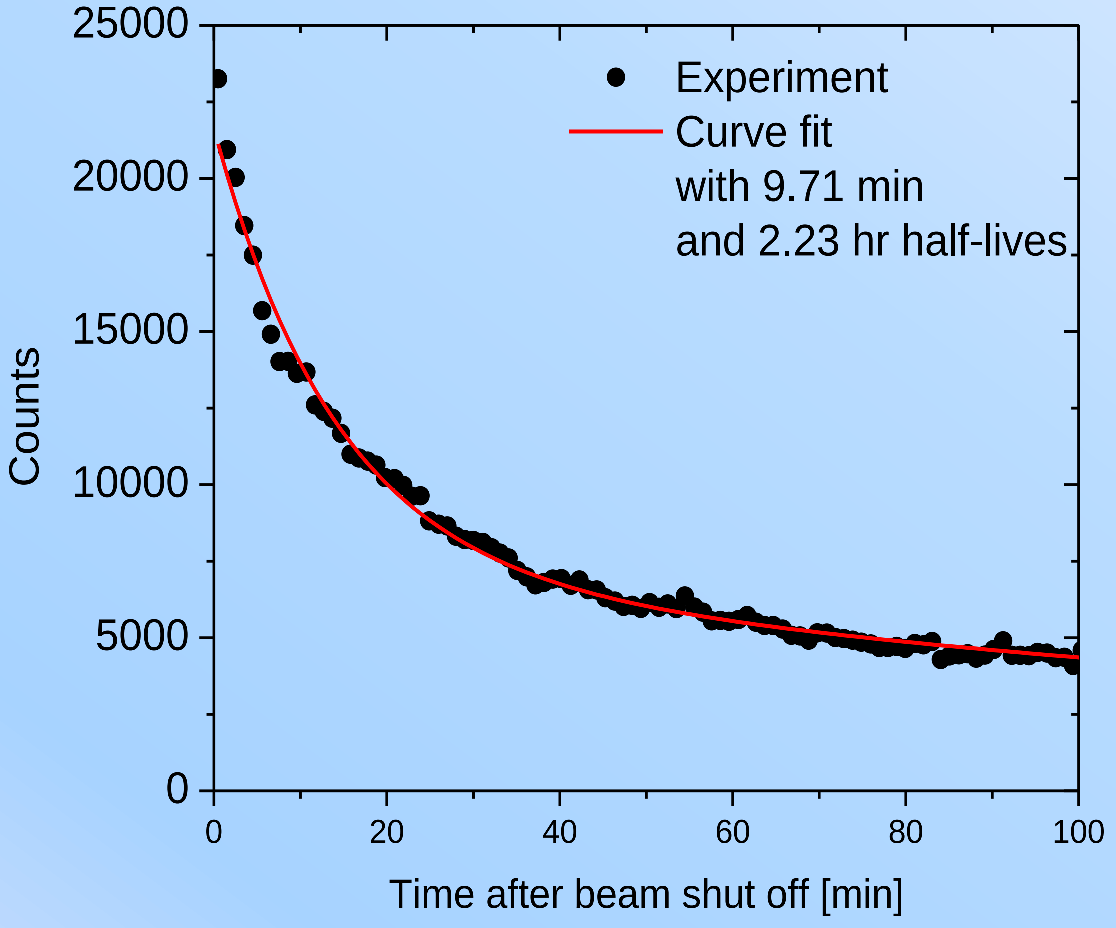
- Due to charge accumulation the detector pulse shape deteriorates
  - Solution reverse bias polarity every 5-10 minutes





Diamond Detector Activation

- 12 µA beam current in the RPI LSDS
- 1 hr irradiation time
- Half-lives possibly correspond to activation of copper



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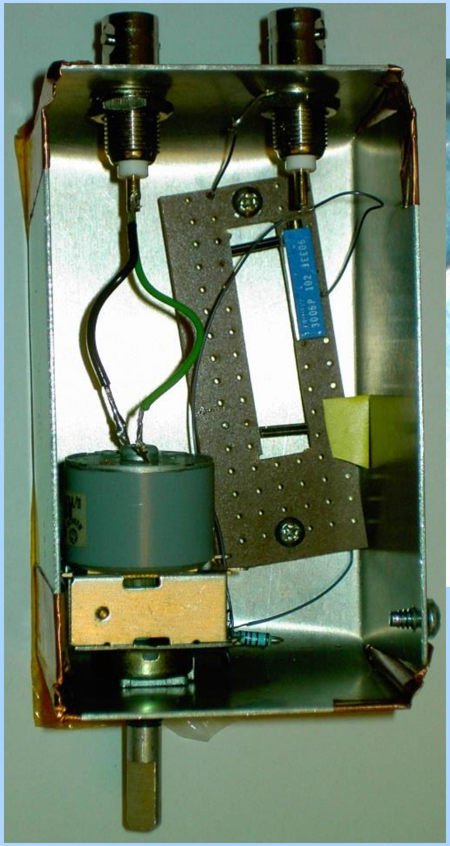
Diamond Detector Final Remarks

- Major breakthroughs 1920 [58], 1955 [57], recently (2000's) [59]
  - Development has been slow
- Single crystalline detectors
  - Have the necessary energy resolution
  - Currently too limited in size to make feasible
- Charge polarization phenomenon still exists

Solar Cell Detectors

- ☑ Cheap
- ☑ Previously used for fission measurements
- ☑ Easy to compensate
- ☒ Succumbs to radiation damage quickly in the LSDS
- ☒ Low signal to noise ratio
- ☒ Low quality control

My students compensated design!!



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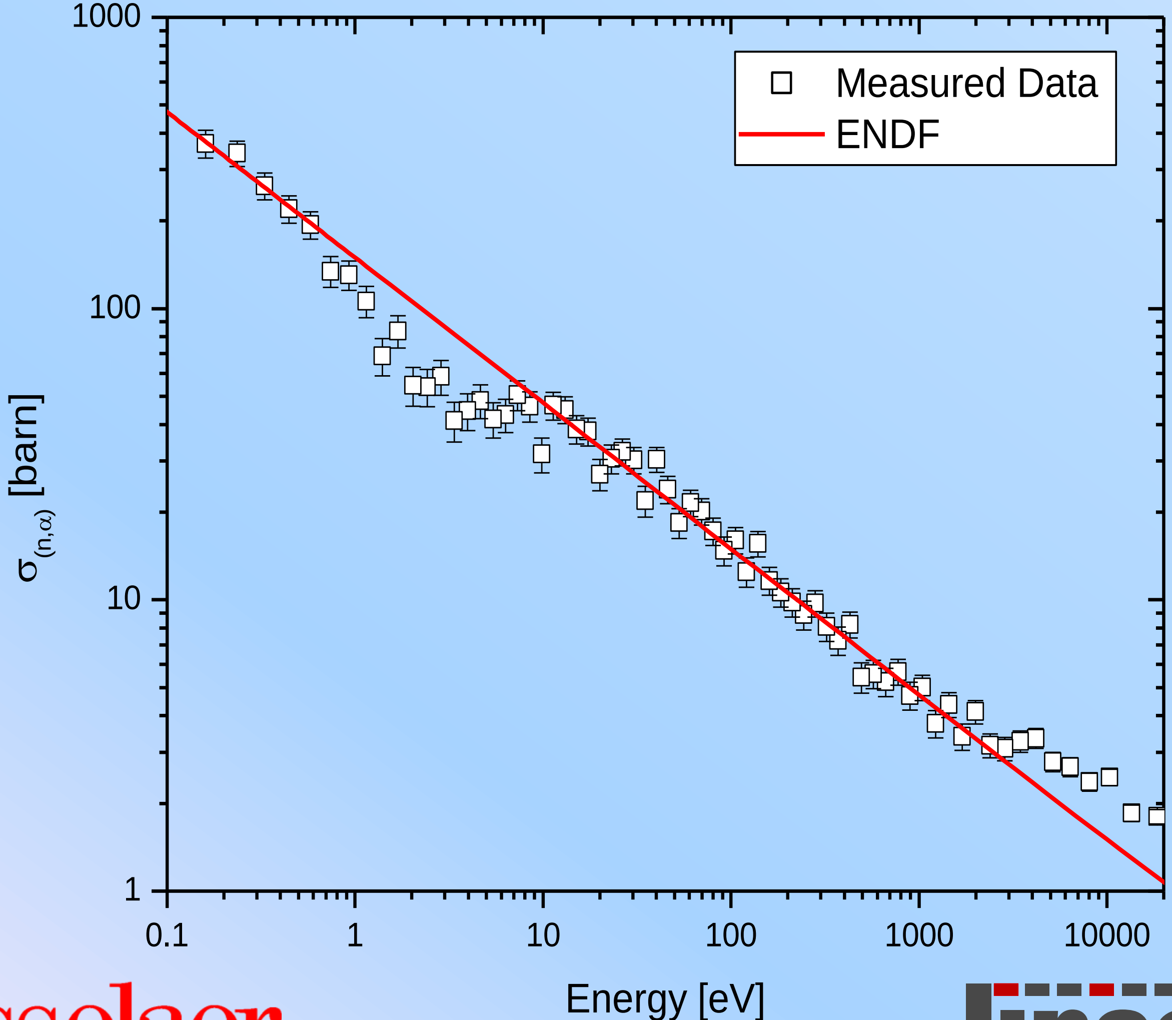
Li(n,α) measurement at LANL



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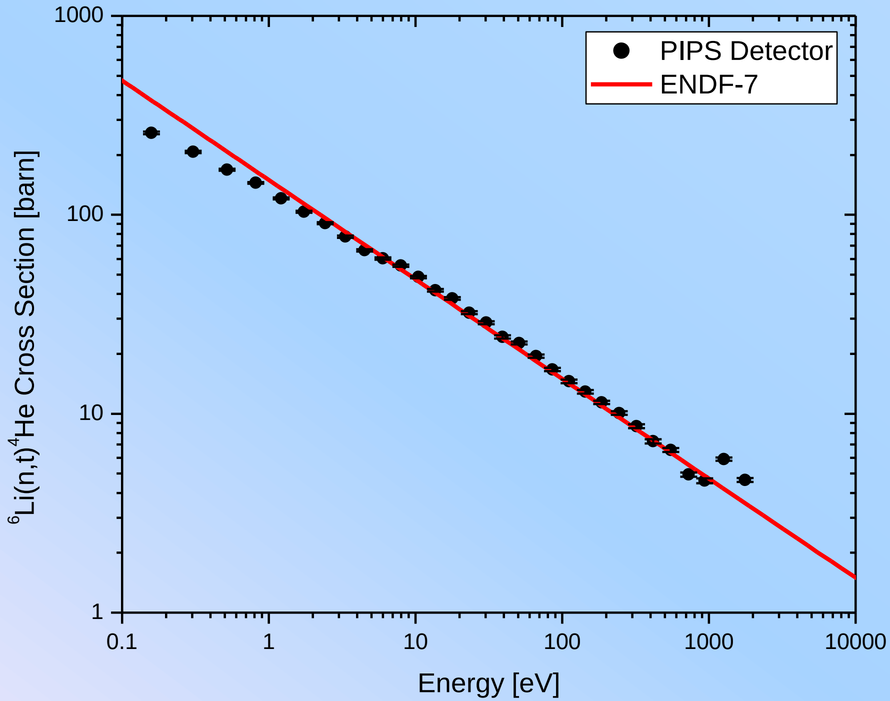
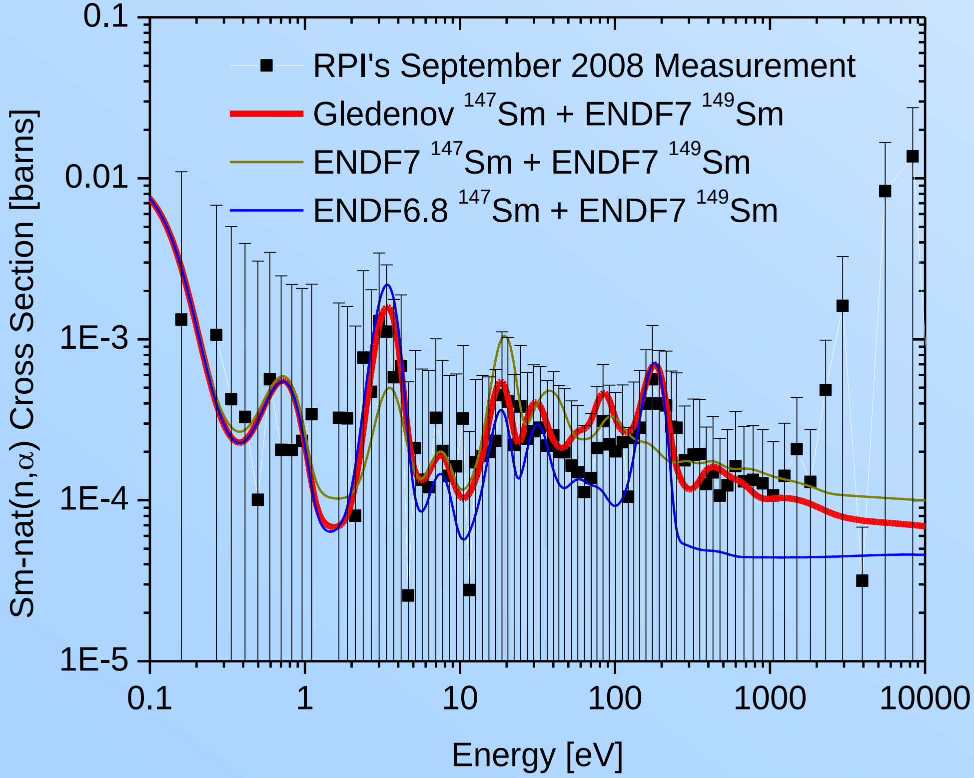
Passivated Implanted Planar Silicon (PIPS) Detectors

- ☑ Commercial
- ☑ Ready made
- ☑ Good signal-to-noise ratio in quiet environment
- ☒ Effects from radiation damage visible in few hours
- ☒ Signals are not easily reversible

Results from PIPS Detectors



First encouraging result at LANCE



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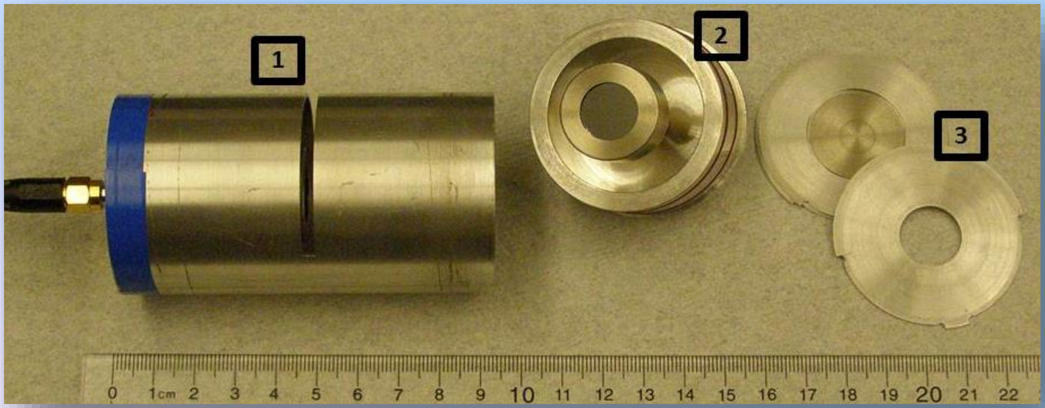
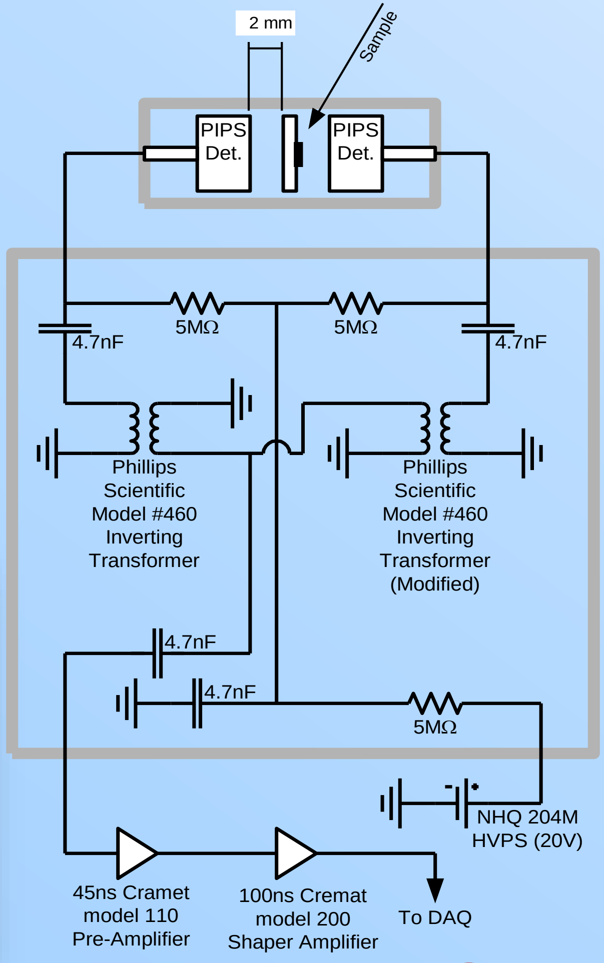
Final PIPS Compensated Detector

- Face-to-face PIPS detector
- PD-150-30-40EPI-40
  - 150 mm^2 active area
  - 40 µm active thicknesses
  - +20 V bias
- Transformer-based compensating circuit





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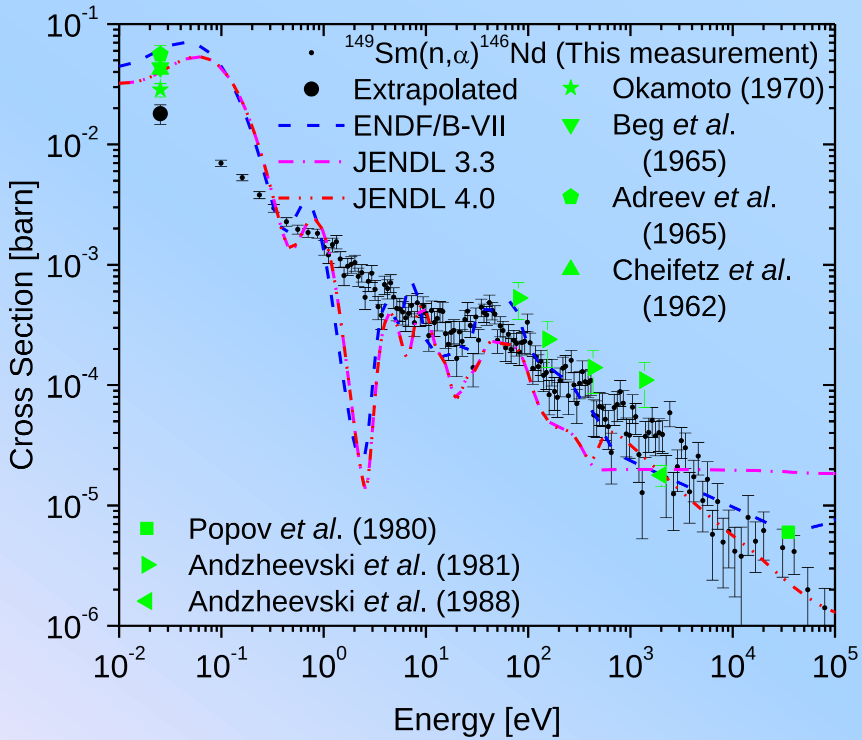
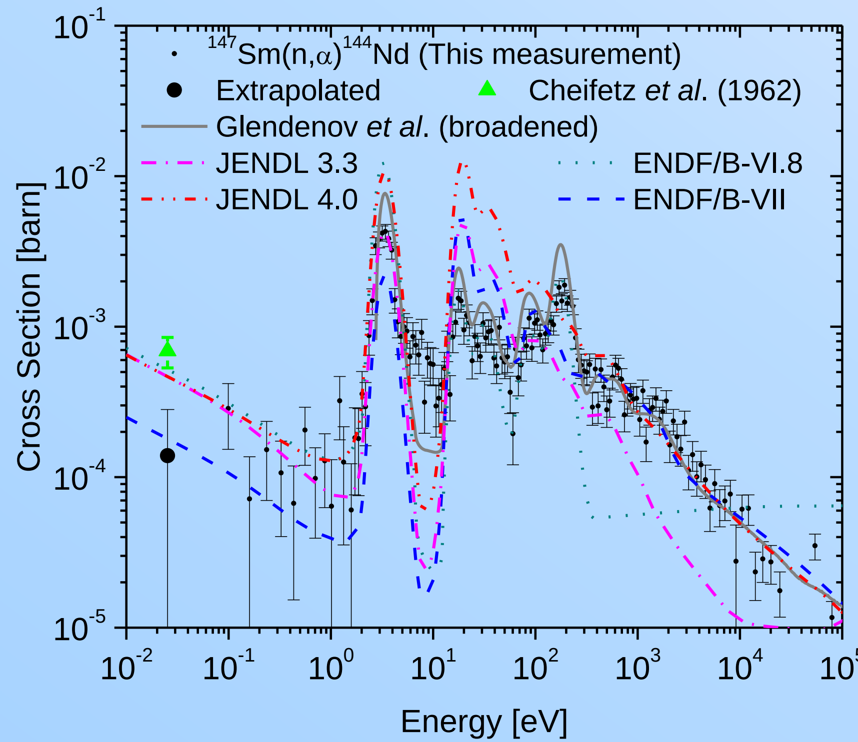
(n,α) Cross Section of ^{147,149}Sm

- Compensated Passivity Implanted Planar Silicon (PIPS) Detector .
  - Commercially available
  - More susceptible to radiation damage.
  - Successfully used at LANL and RPI LSDS

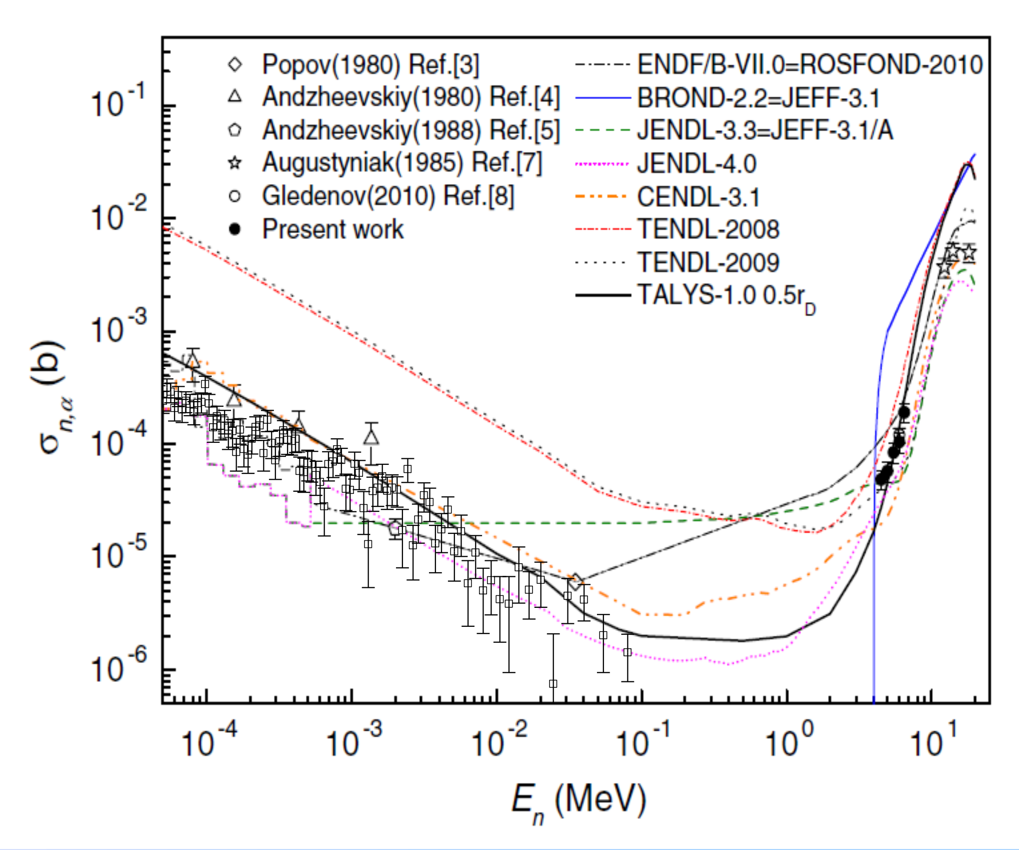




J.T. Thompson, T. Kelley, E. Blain, R.C. Haight, J.M. O'Donnell, Y. Danon, "Measurement of (n,α) reactions on ^{147}Sm and ^{149}Sm using a lead slowing-down spectrometer", Nuc. Inst. and Meth. A, Vol. 673, pp. 16-21, May (2012)

Recent Measurement of ^{149}Sm

- Measured at 4.5- 6 MeV
  - Guohui Zhang et al., "^{149}Sm(n,α)^{146}Nd Cross Sections in the MeV Region", Phys. Rev. Lett. 107, 252502 (2011)



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Measurements Fission Fragment Mass and Energy Distributions using a LSDS

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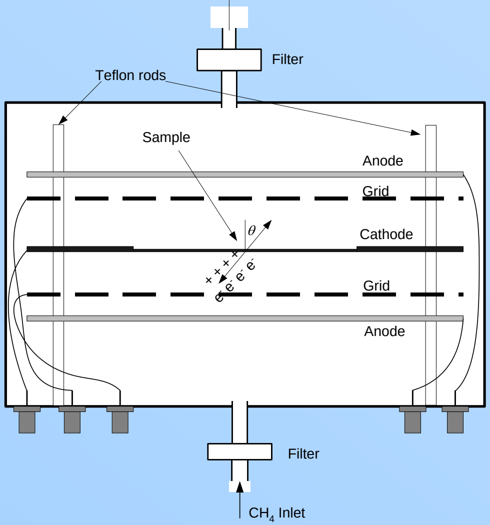
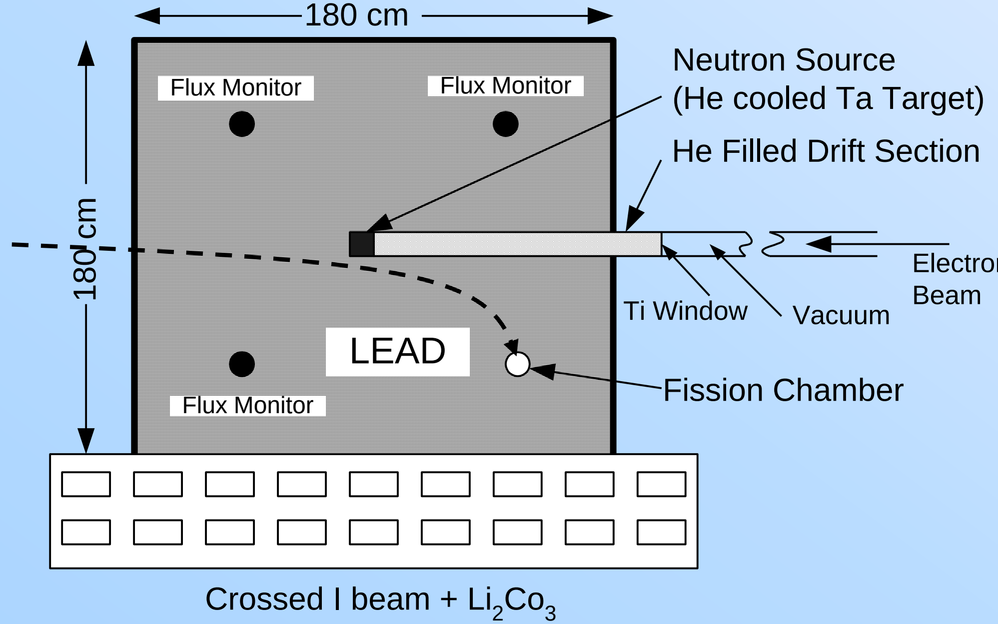
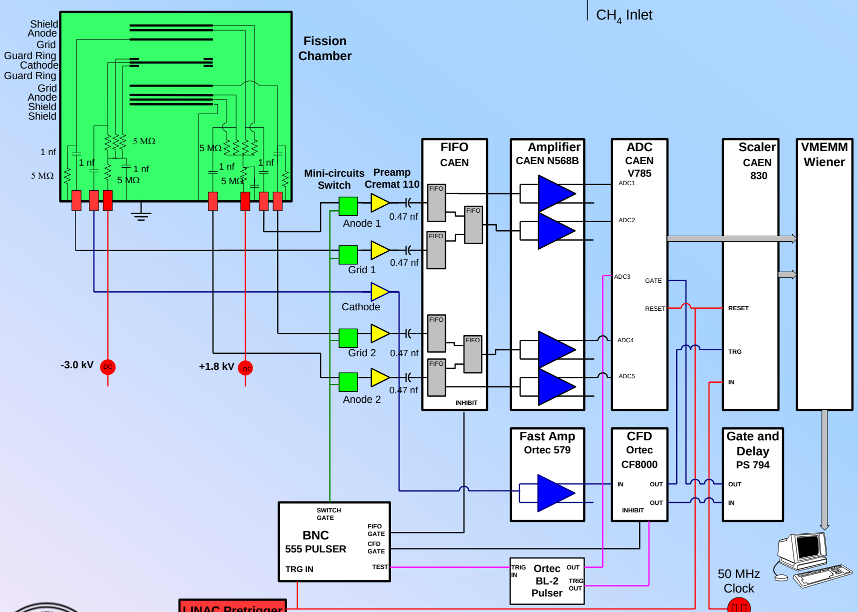
Experimental Setup

Double Gridded Fission Chamber







Multi parameter DAQ

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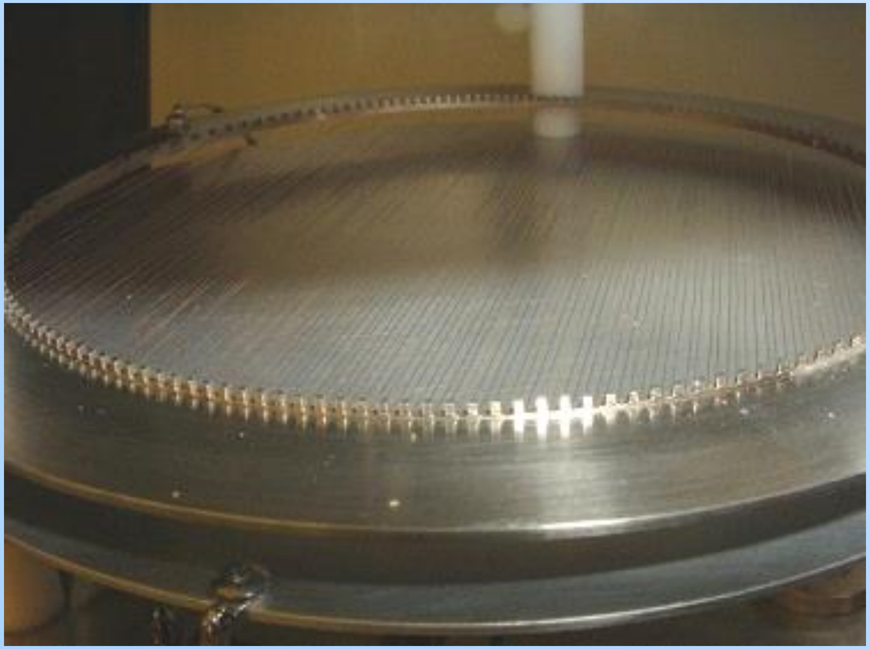
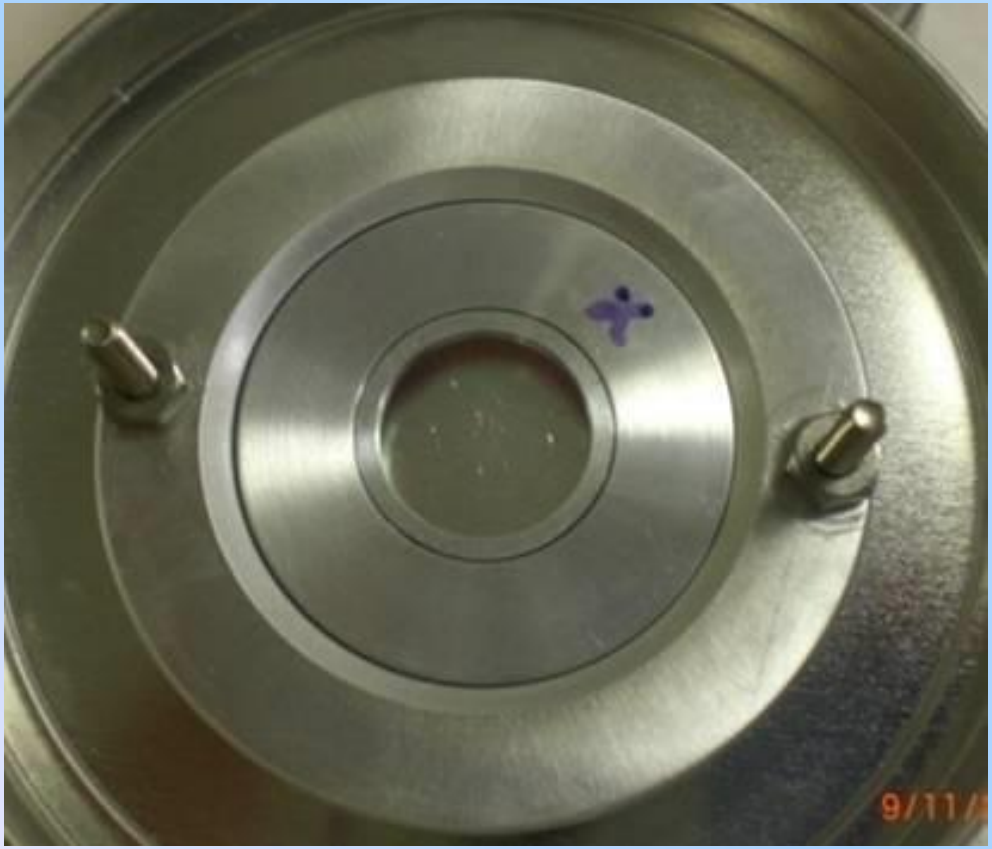
Double-Sided Frisch-Gridded Fission Chamber



Frisch Grid

Thin Samples

34 μg 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

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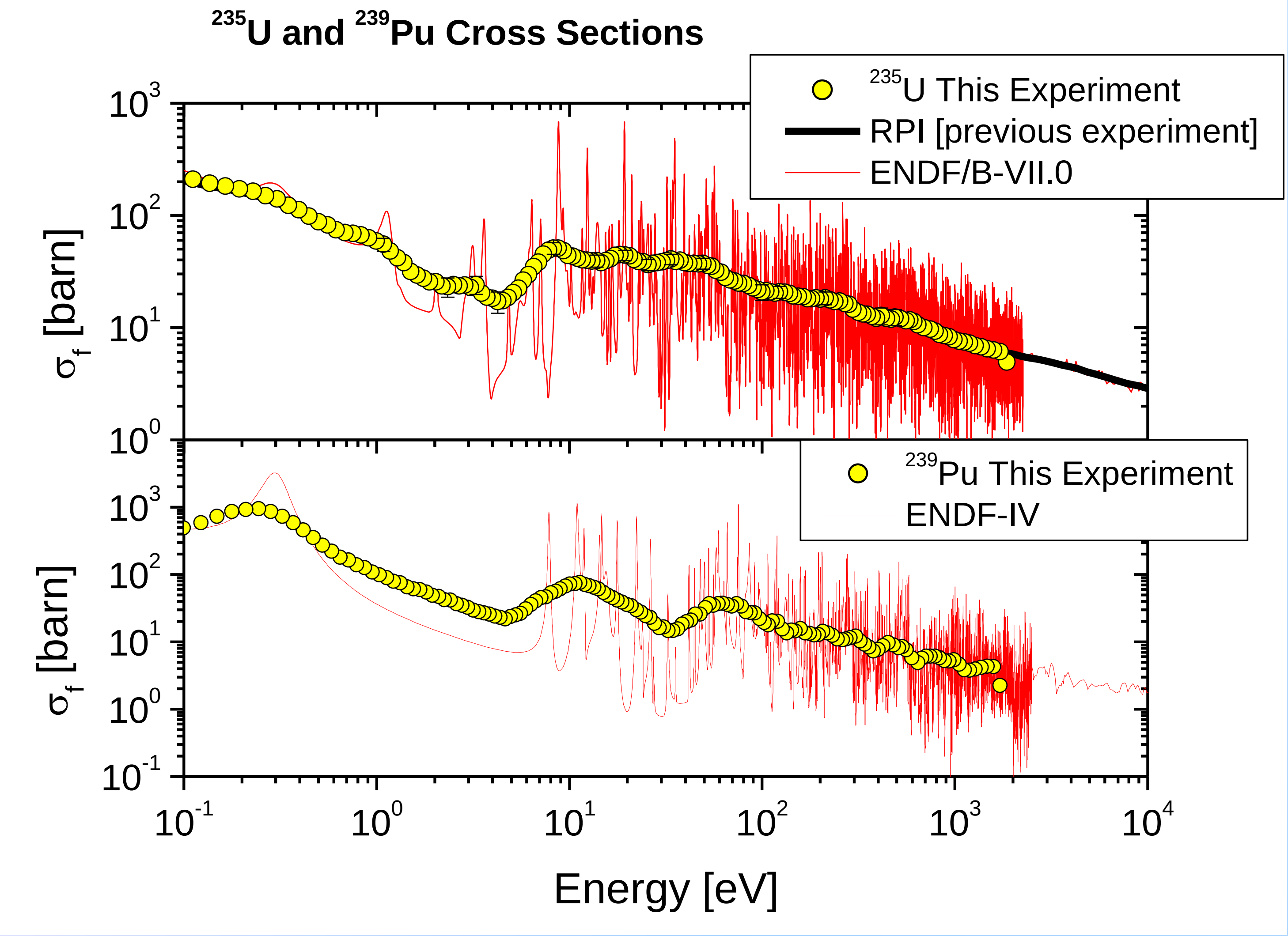
^{235}U and ^{239}Pu Fission Cross Sections



Y. Danon, R.E. Slovacek, R.C. Block, R.W. Lougheed, R.W. Hoff, M.S. Moore, Nuclear Science and Engineering **109**, p. 325 (1991).

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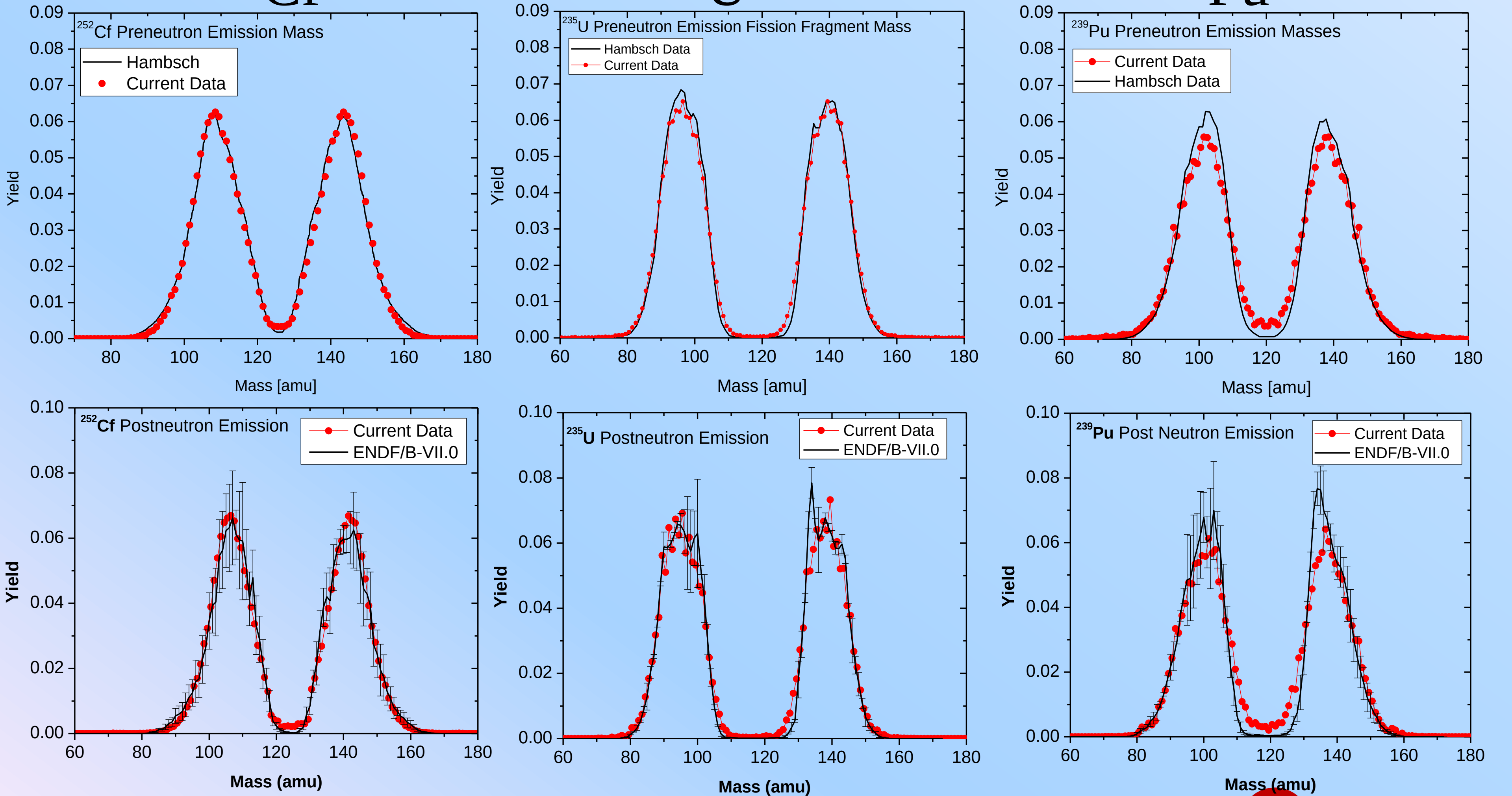
Results – Fission Fragment Mass distribution E_n<0.1 eV

^{252}Cf | ^{235}U | ^{239}Pu



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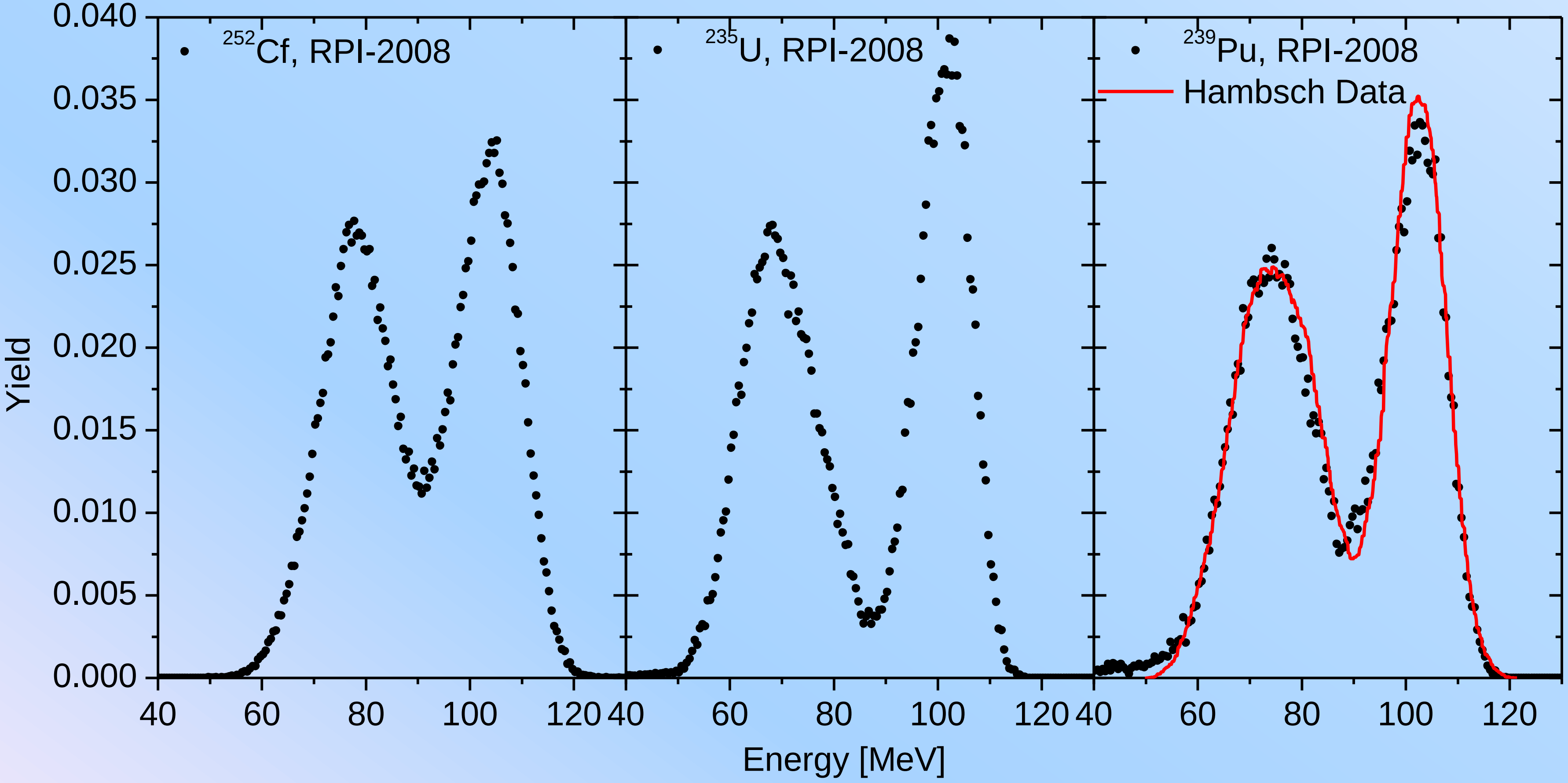
Results – Fission Fragment Energy Distribution E_n<0.1 eV



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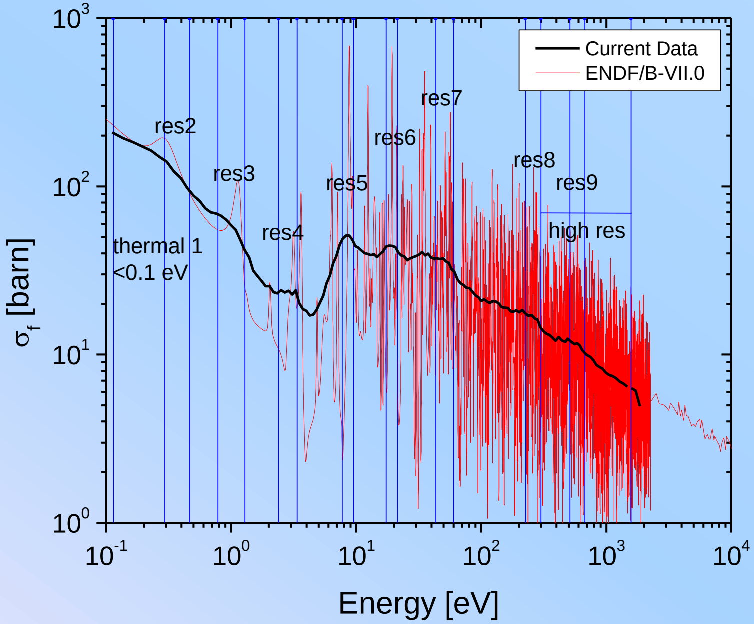
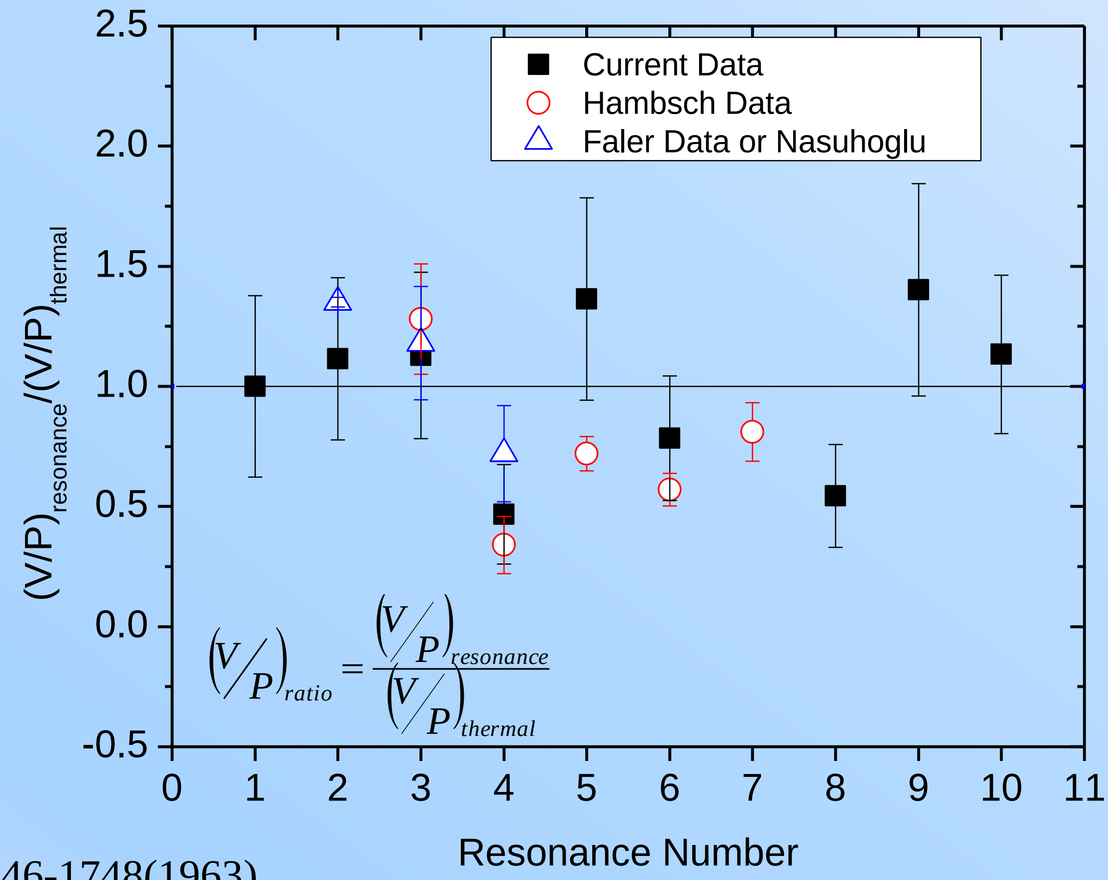
Fission symmetry in resonance clusters





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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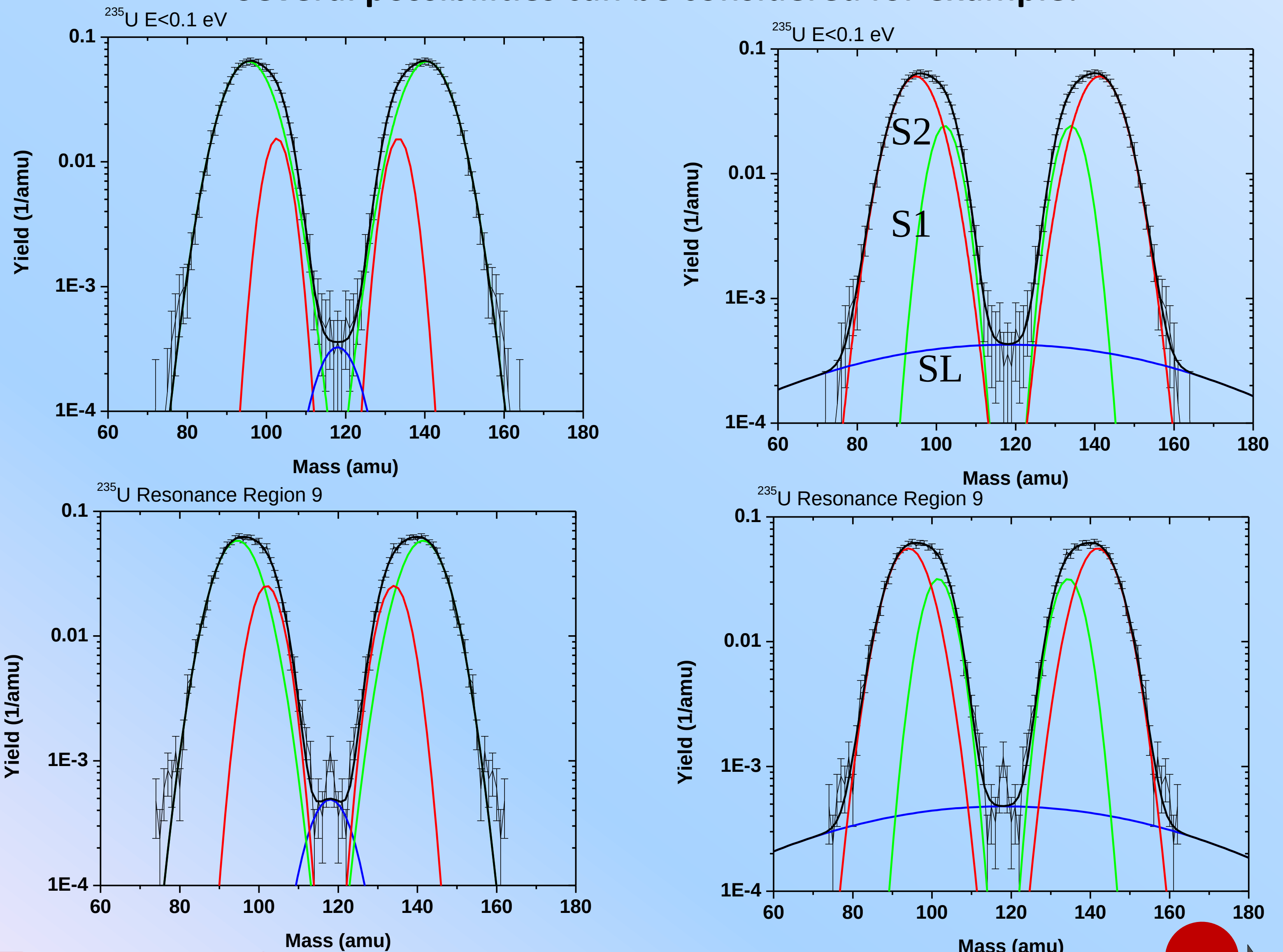
Fitting the fission modes

Several possibilities can be considered for example:



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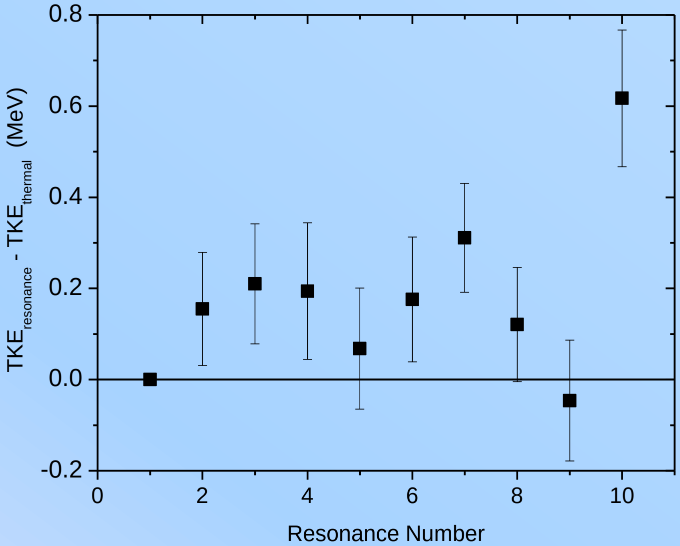
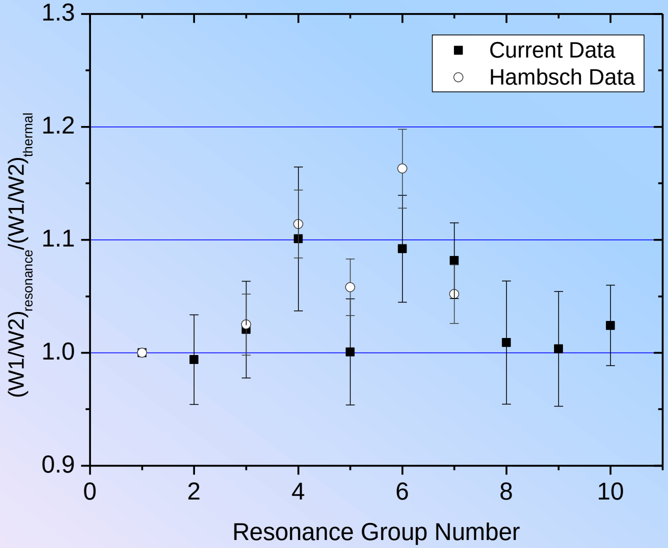
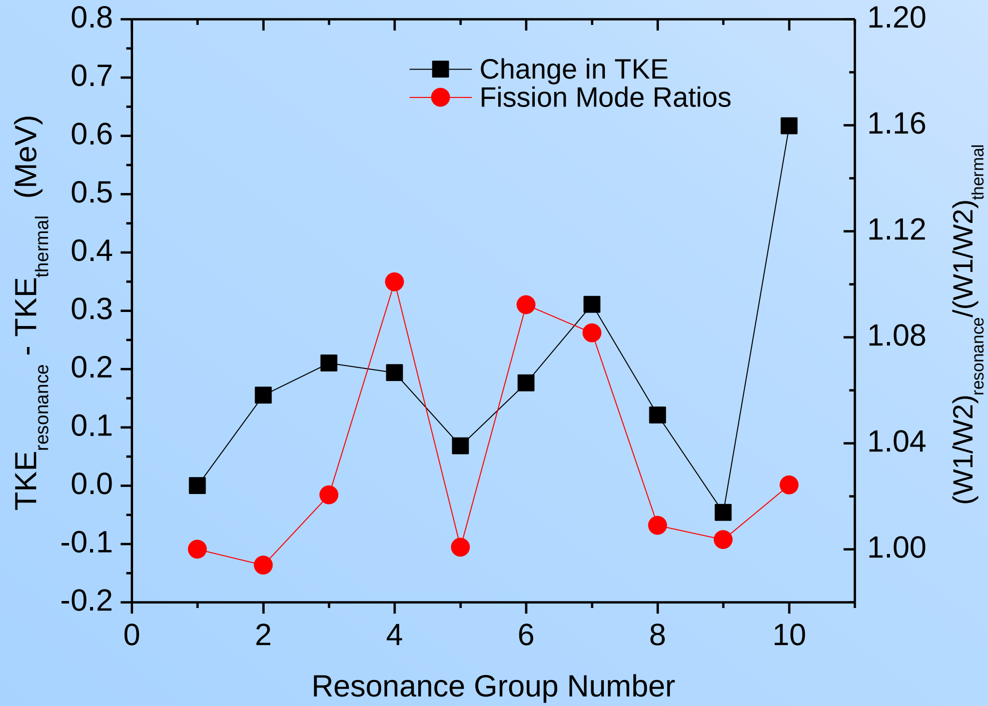
Variations In TKE As A Function of Incident Neutron Energy







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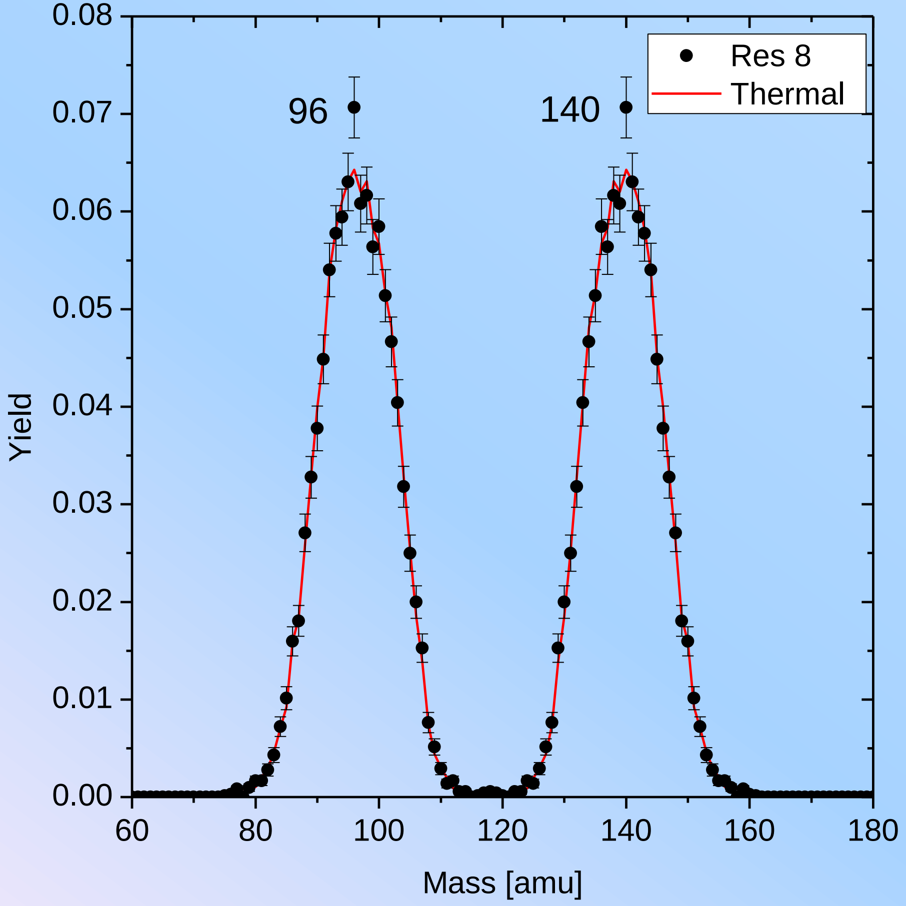
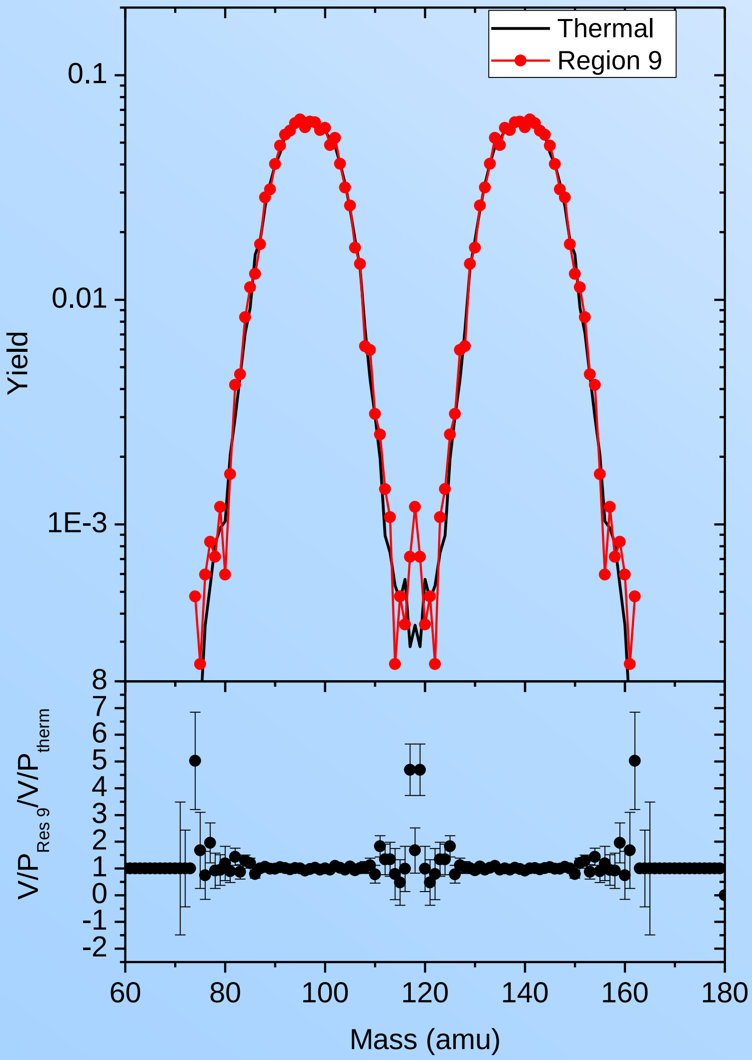
Resonance Regions 8 and 9

- Pre-neutron emission





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Neutron Capture Reaction Rates

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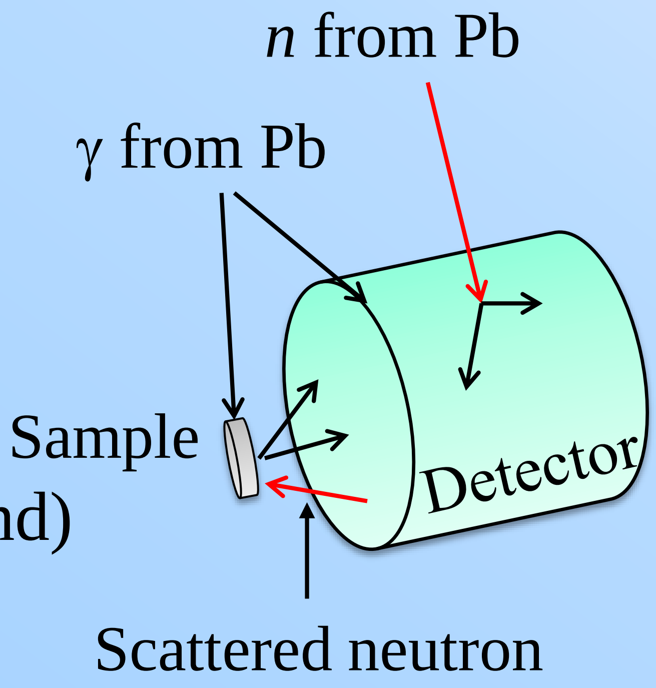
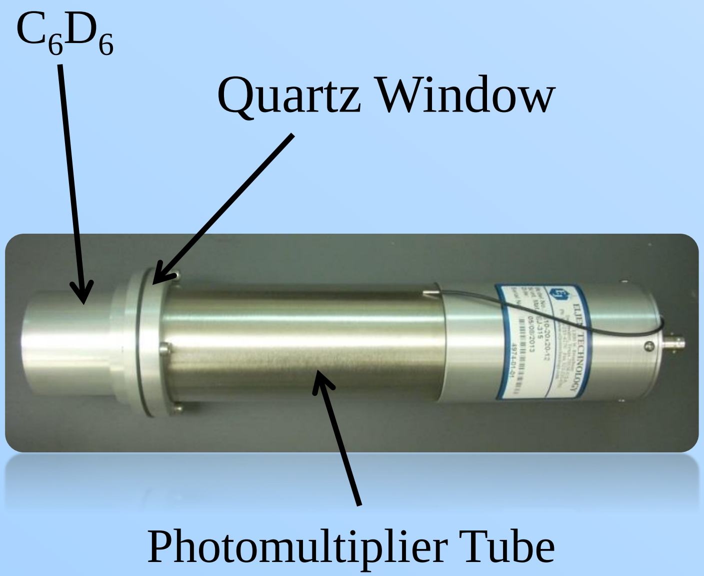
Goal for LSDS measurement

- 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



Detectors Considered

- A number of fast response scintillators were investigated
  - Modeled in MCNP
  - YAP, PWO, LSO, LYSO, C_6D_6 etc.
  - **YAP performed the best**
  - Initial measurements were made



Experimental setup

Detector positions in the LSDS

U-235 fission chamber

- Operate the LINAC at E_e=54 MeV and I<0.1 μA; ~0.8% of max LINAC power when using the RPI-LSDS
- Use two different DAQ systems:
  - Analog – constant fraction discriminator and time-of-flight clock.
  - Digital – Acqiris AP 240 digitizer
- Used two samples:
  - Ta ~ 0.42 g (high cross section)
  - Ni ~ 5 and 15 g (low cross section)
- 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

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Data Acquisition

- Collected data with two systems
  - Analog system
    - Recorded when pulses occurred
    - 204.8 ns bin width was selected
  - Digital system (Acqiris AP240)
    - Recorded when pulses occurred and pulse shape
    - 8 bit digitizer
    - 200 channels, 1 ns channel width

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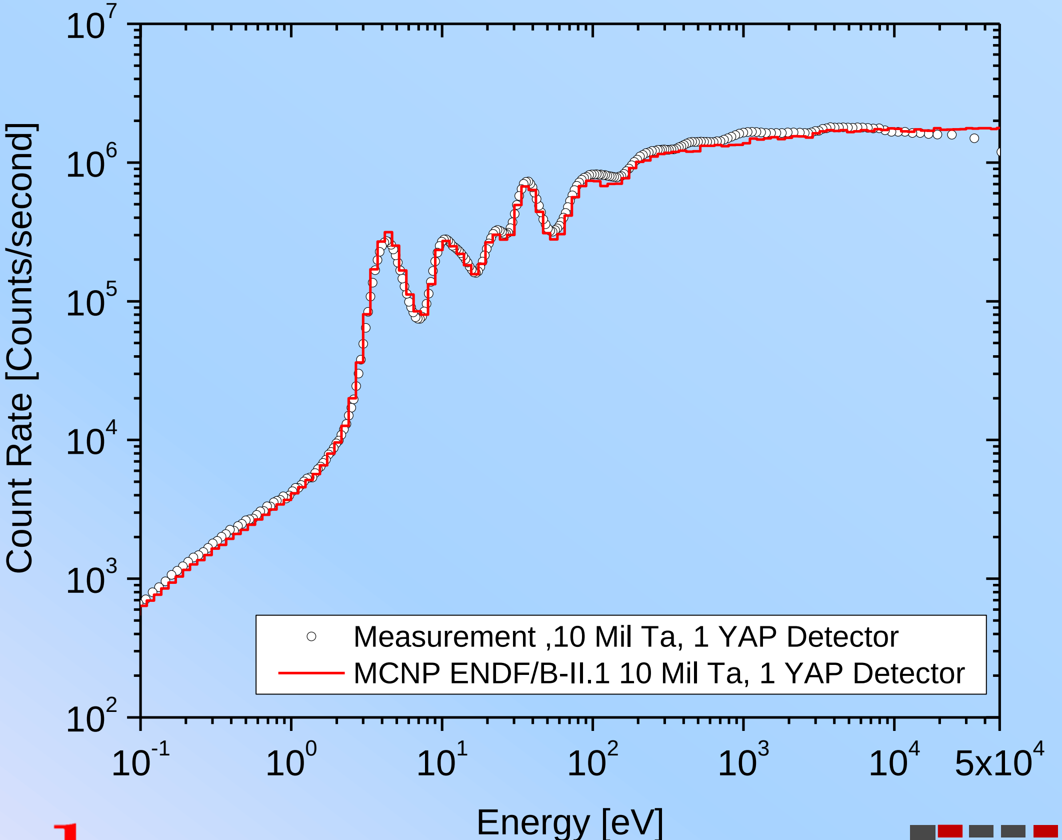
Ta capture rate

- 2mm YAP Detector, 10 Mil Ta Sample
  - Extended high energy range to ~50 keV
  - Good agreement with the simulations



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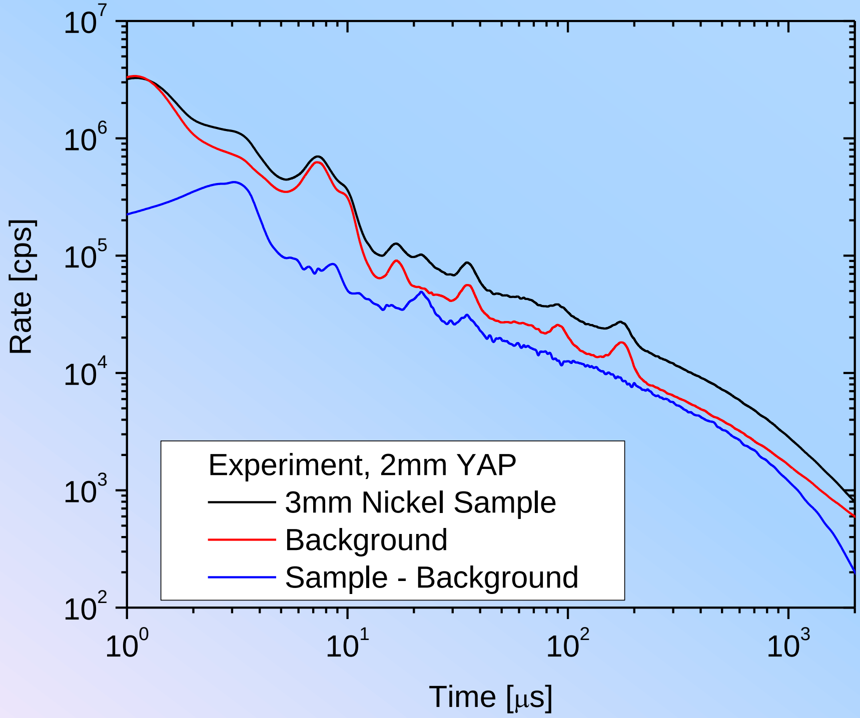
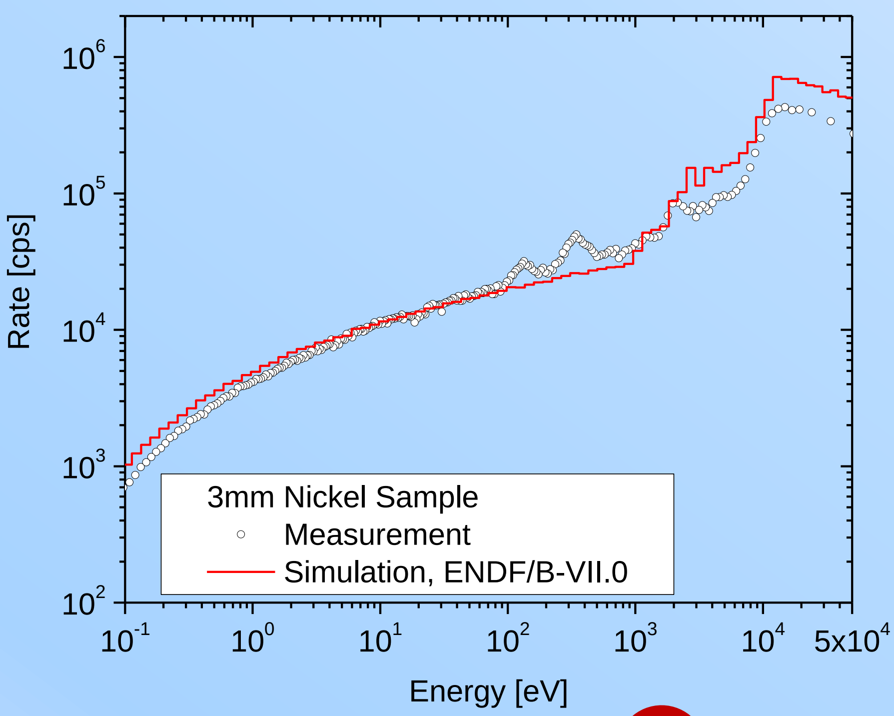
Ni Capture Rate

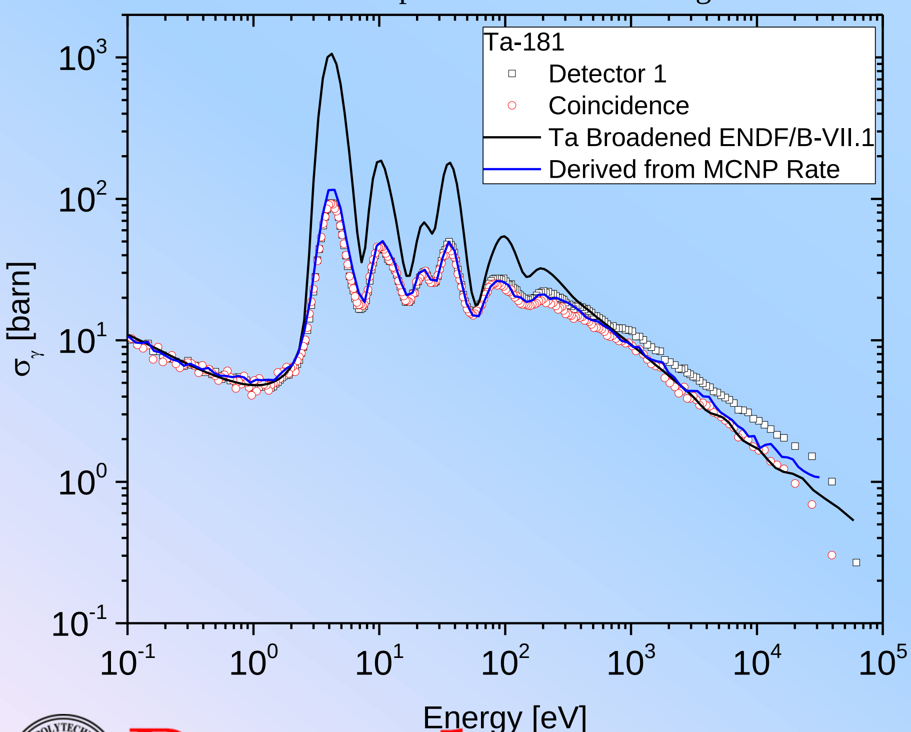
- 2mm YAP Detector, 3mm Nickel Sample
  - Low capture cross section → Low S/B
  - Reasonable agreement with ENDF/B-VII.1 below 100 eV
  - Additional peaks observed near 200-300 eV → possible impurities in sample

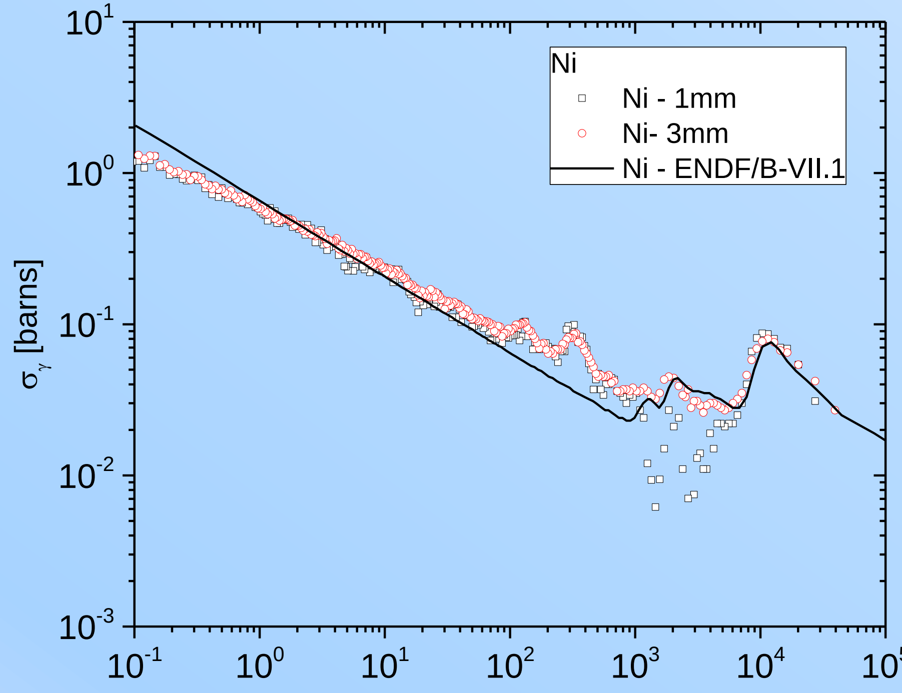




Cross section measurements

- Cross sections can be obtained using the thin sample approximation:
  - $R = \emptyset N\sigma$, where R is the reaction rate, σ is the cross section, and N is the samples atomic density, ϕ is the neutron flux
  - Good assumption when $N\sigma < 0.1$
- Broadened cross section derived from the count rate indicates perturbation of the ideal flux and possibly other contributions
  - Need to improve our understanding





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Assay of Used Nuclear Fuel

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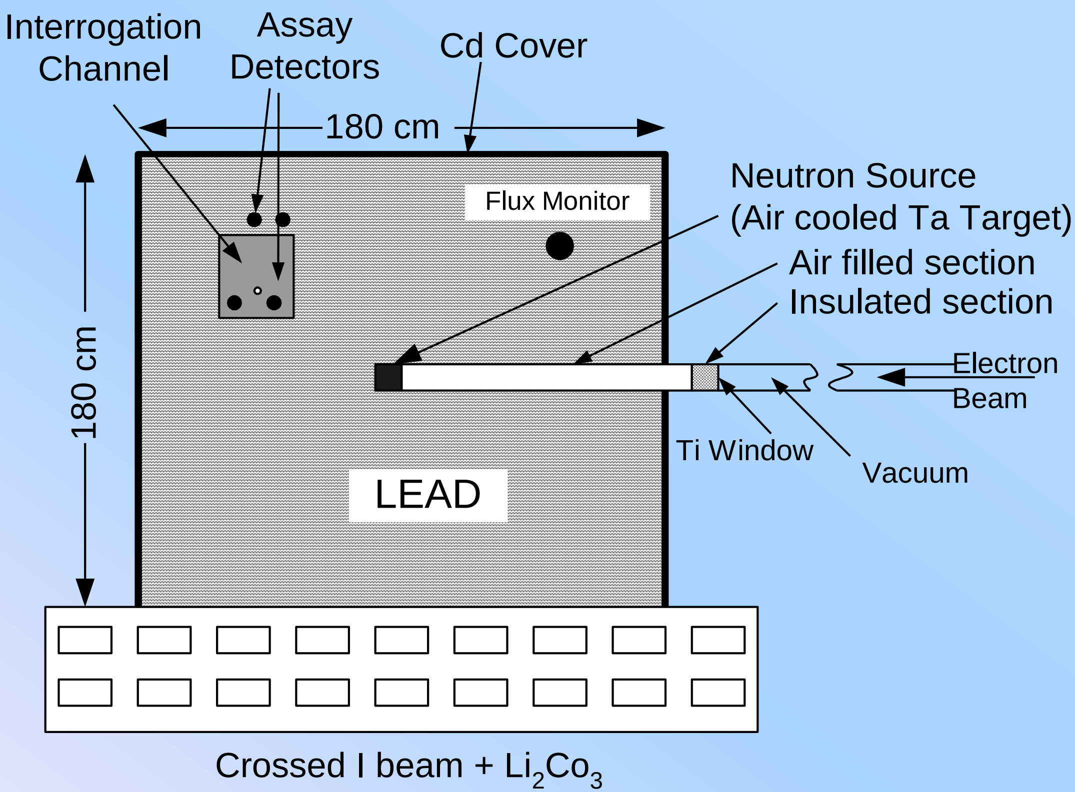
LSDS Used Fuel Assay Basic Principles



- Ta target in the center of the lead cube
  - Neutron production via: (e,γ) followed by (γ,n) reactions
  - Evaporation spectrum with peak energy of ~0.46 MeV
- Neutrons that slowed down interaction with the fuel
- Fission neutrons are detected as a function of slowing down time.

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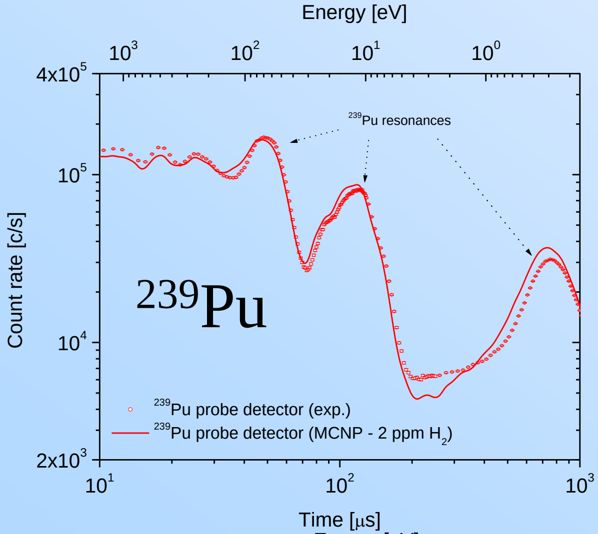
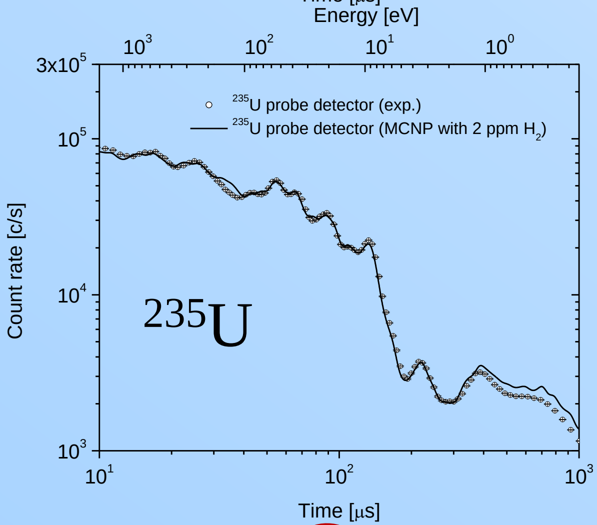
Non-Destructive Assay Using a LSDS

- Each fissile nuclei has a characteristic fission response function (*R*) in the LSDS
  - Resolution broadened fission cross section
  - Amount of material
  - Fast fission neutrons
- Spent nuclear fuel (SNF) gives a combination of different response functions (mainly of ^{239}Pu and ^{235}U)

$$R_{SNF} = A \cdot R_{Pu-239} + B \cdot R_{U-235}$$

- Regression method to find A and B
  - Non-linearity for cases with strong self-shielding





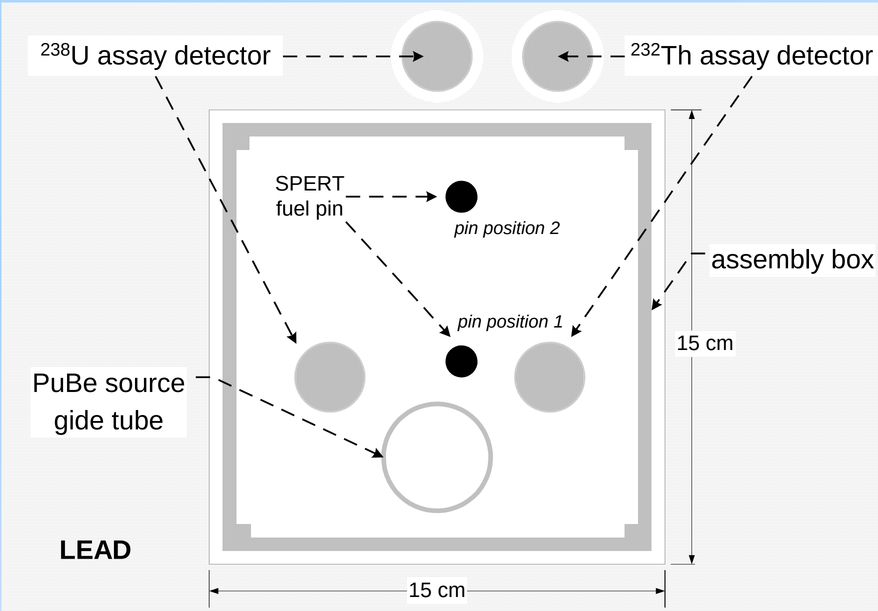
Motivation: Non-Destructive Spent Fuel Assay

- Spent fuel assay:
  - Determination of the fissile material content (mainly ^{235}U, ^{239}Pu)

- Lead Slowing-Down Spectrometer is an attractive option:
  - Active, non-destructive method
  - Does not rely on burn-up information or calculations
  - LSDS have been used for fresh fuel assay (*Karlsruhe; RPI*)
  - Decades of LSDS experience for cross section measurements

- Important information for:
  - Nuclear safeguards
  - Fuel cycle (reprocessing)
  - Fuel storage

Assay Detectors / Measurement System



- ^{238}U and ^{232}Th threshold fission chambers:
  - Fast neutron detector
  - 200 mg content / detector
  - Insensitive to gamma irradiation
  - ^{238}U detector: highly depleted (^{235}U impurity: ~4 ppm)

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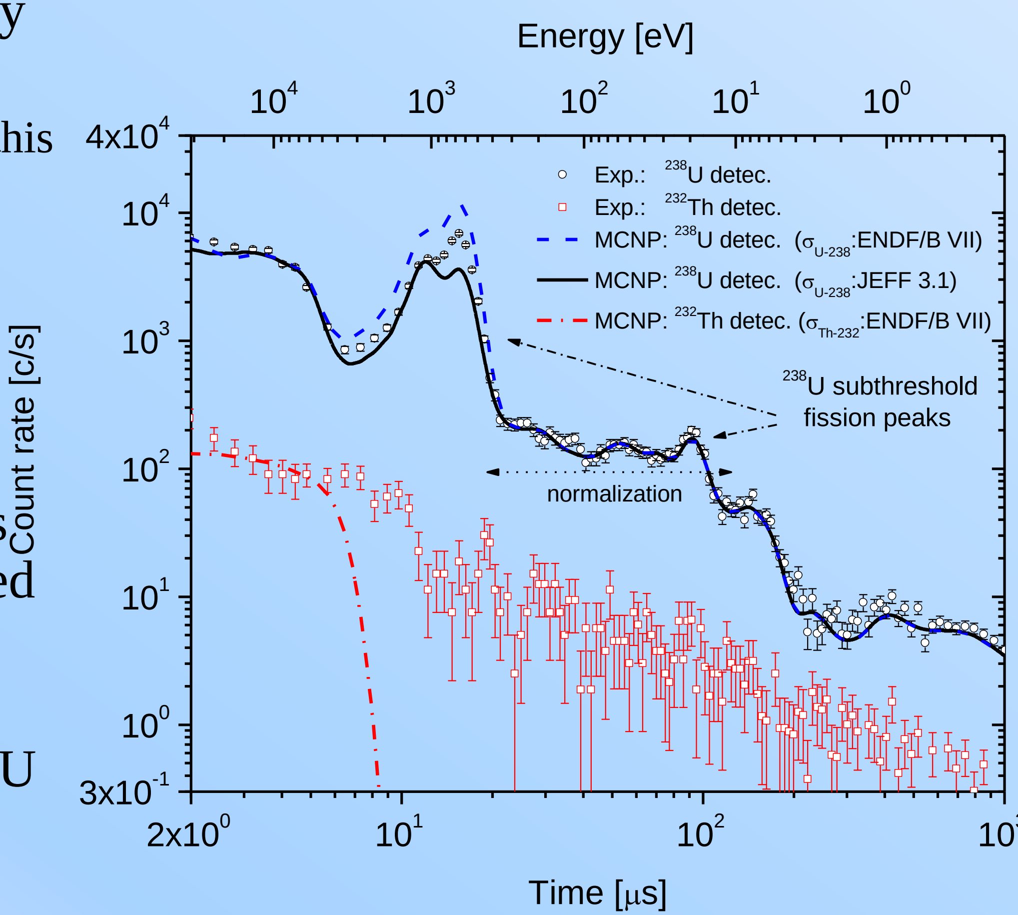
Detector Signal due to Interrogation Flux

- Measurement without any assay specimen
  - Would like to see zero counts (this is a fast neutron detector)
- Background signal of the ^{238}U detector due to:
  - Subthreshold ^{238}U fission
  - ^{235}U impurities (4 ppm)
- ^{238}U subthreshold fission peaks (near 700 eV) are over predicted by ENDF/B VII.0 and under predicted by JEFF3.1
- Overall good agreement for ^{238}U assay detector in both ^{235}U and ^{238}U dominated regions



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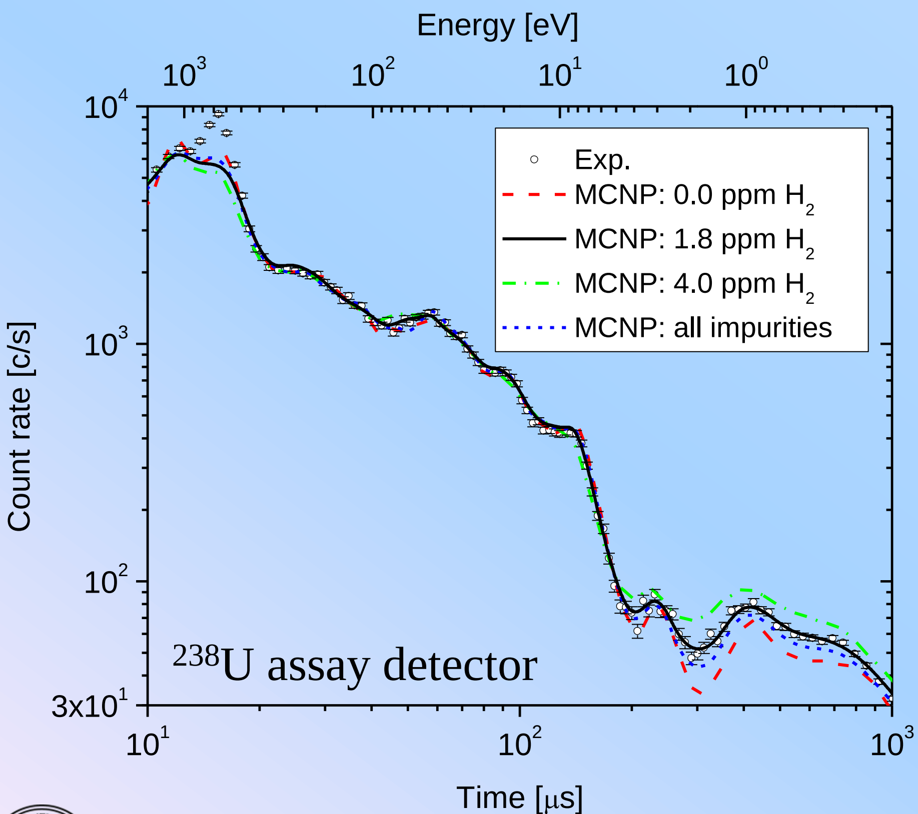
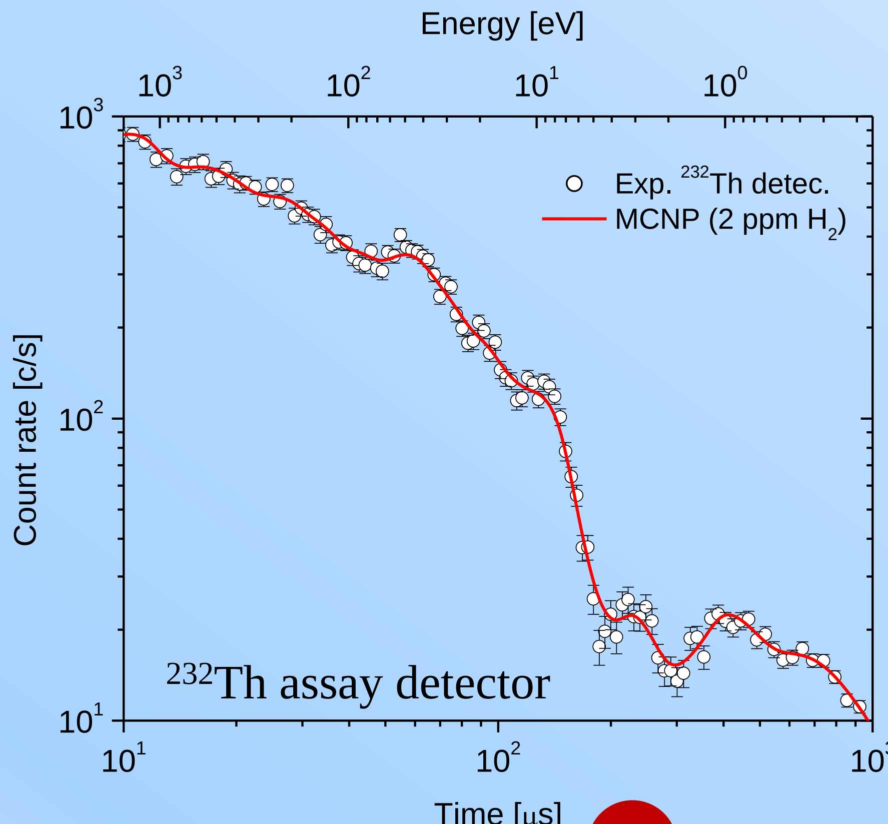
Fresh Fuel Pin Assay

- SPERT fuel pin
  - UOX fuel (35.2 g ^{235}U / pin)
  - ^{235}U enrichment: 4.8 at%

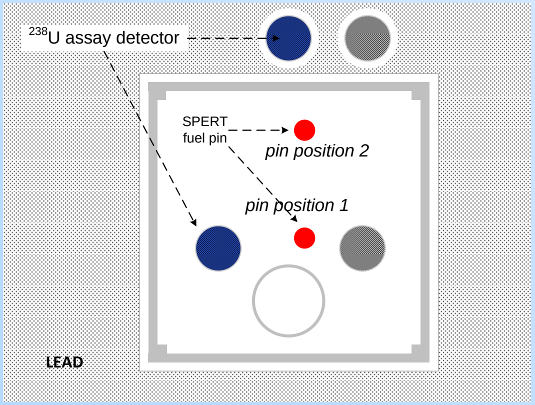
- Impact of H_2 impurities:
  - Decrease of signal structure particularly at low energy

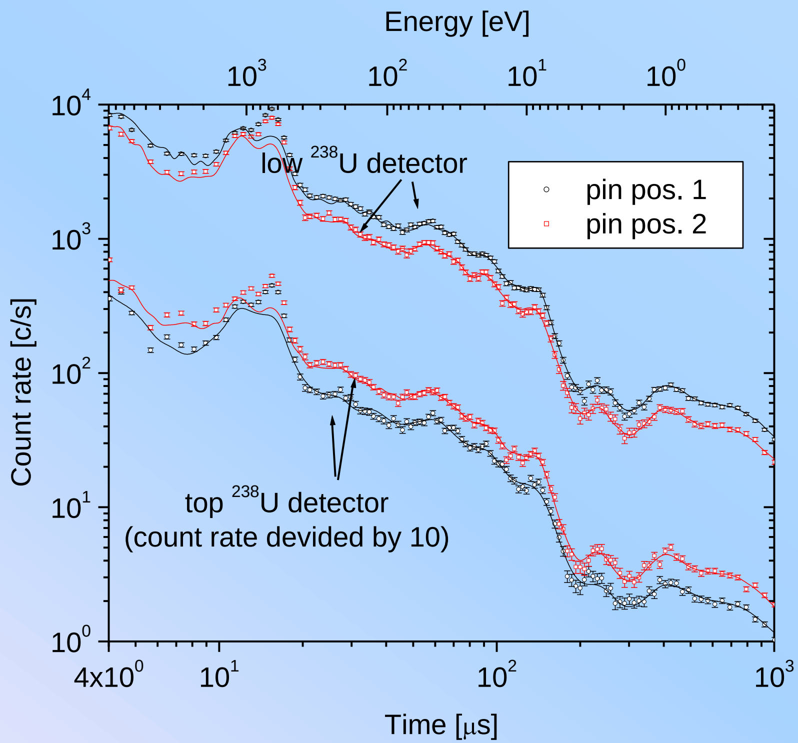


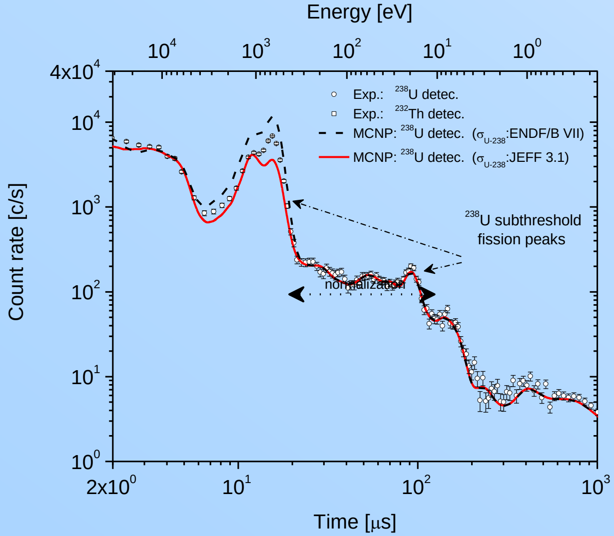


Absolute Count Rate Calculation

- Normalization of the MCNP calculations based on the measurement without a fuel pin
- Prediction of the absolute count rate for the top and low ^{238}U assay detector for two different pin positions







No fuel pin measurement used for normalization

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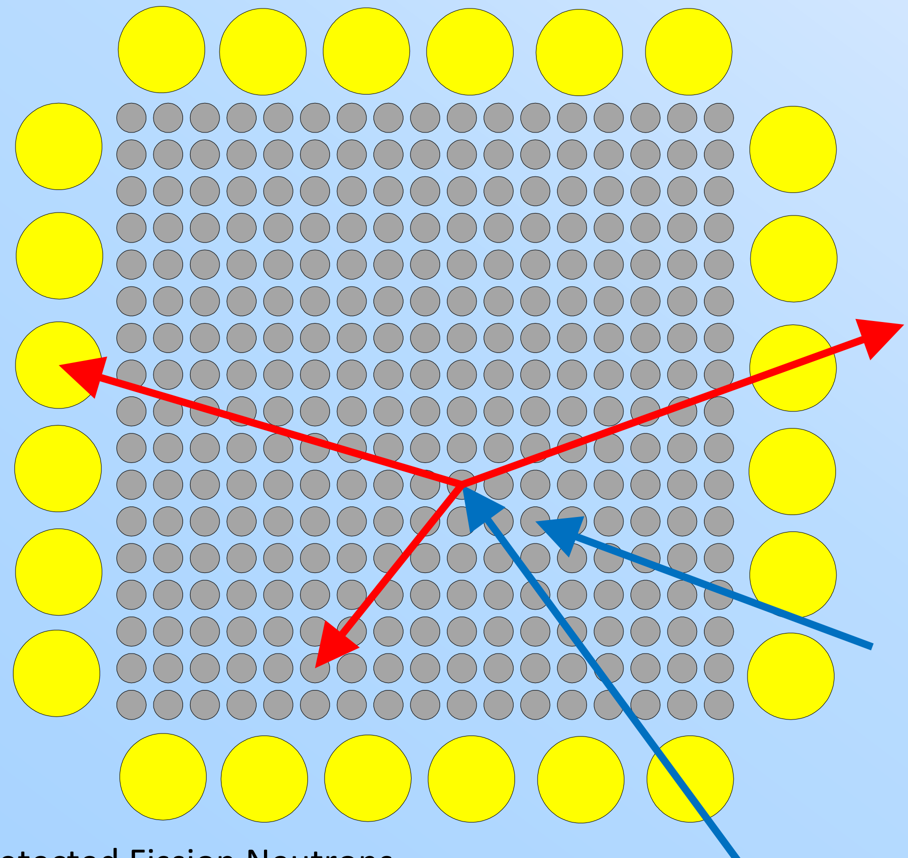
Self Shielding

- Self shielding is a challenging aspect of data interpretation
- Occurs in two energy ranges
  - Interrogation neutrons
  - Fission neutrons
- Occurs in two levels
  - At the single pin level
  - As the assembly level
- Multiplicity helps increase sensitivity to missing fuel pins



C. Romano, Y. Danon and D. Beller, "Fuel Assembly Self Shielding of Interrogation Neutrons In A Lead Slowing-Down Spectrometer", Proc. of the Sixth ANS Intl. Top. Mtg on Nucl. Plant Inst., Control, and Human-Machine Interface Tech. (NPIC&HMIT 2009), American Nuclear Society, LaGrange Park, IL, 2009 (ISBN: 978-0-89448-067-6, on CD-ROM), 2009.

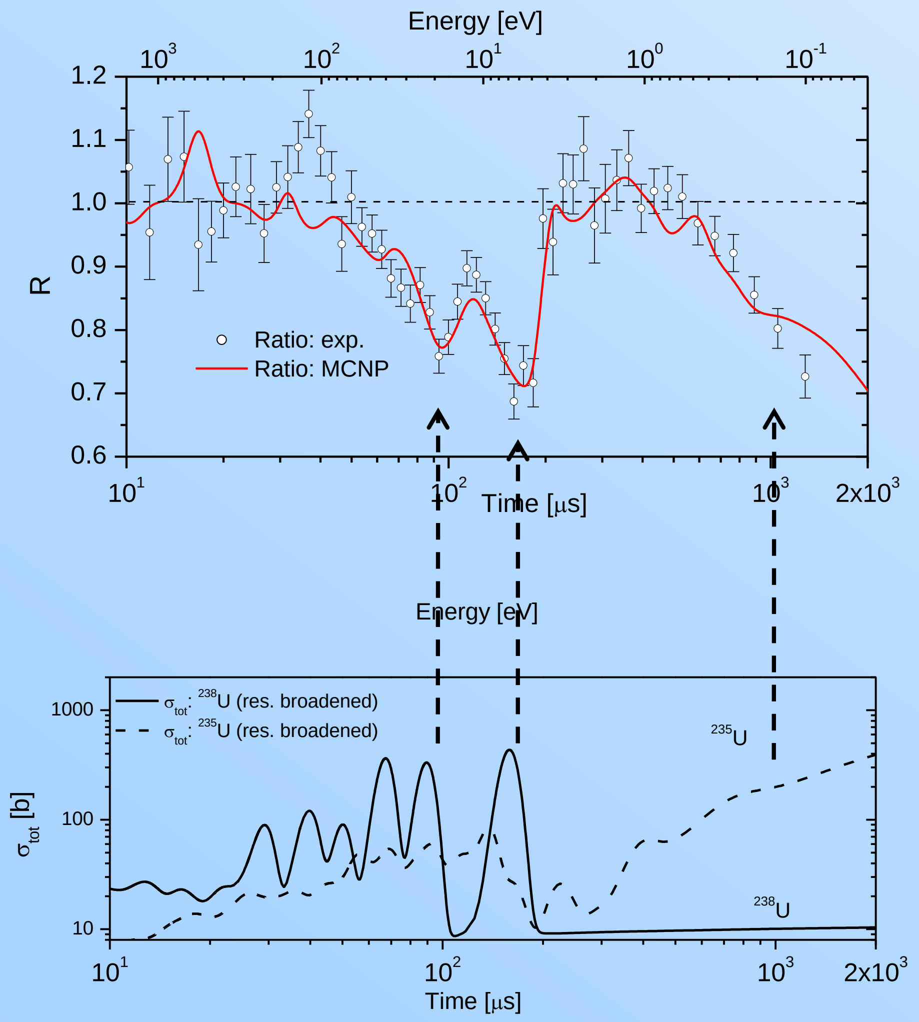
Fuel Pin Self-Shielding

- Estimate of self-shielding: ratio of detector signals

$$R = \frac{\text{Response of assay det. to a fuel pin}}{\text{Response } ^{235}\text{U probe det.}}$$

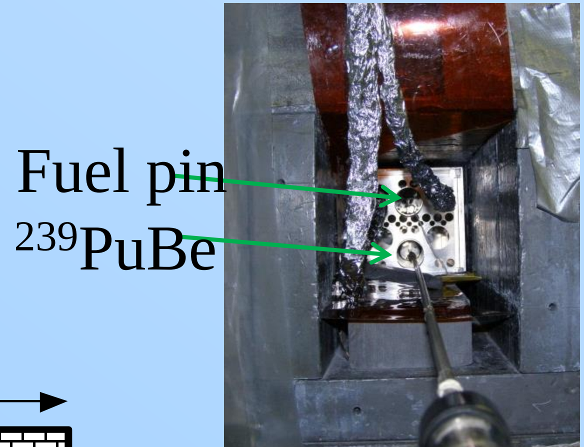
- ^{238}U assay detector response was corrected for sub-threshold fission of ^{238}U

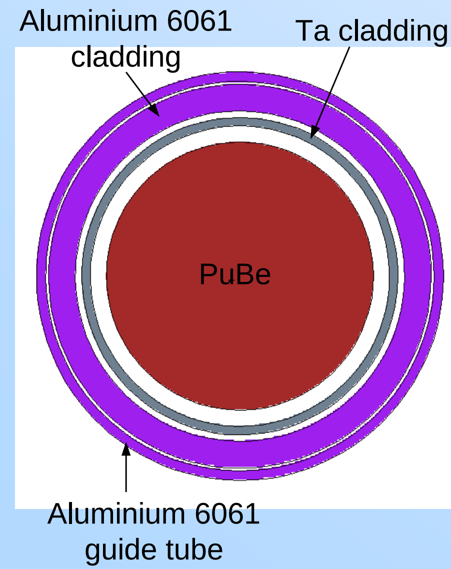
- Self-shielding due to:
  - ^{238}U resonances
  - ^{235}U thermal XS

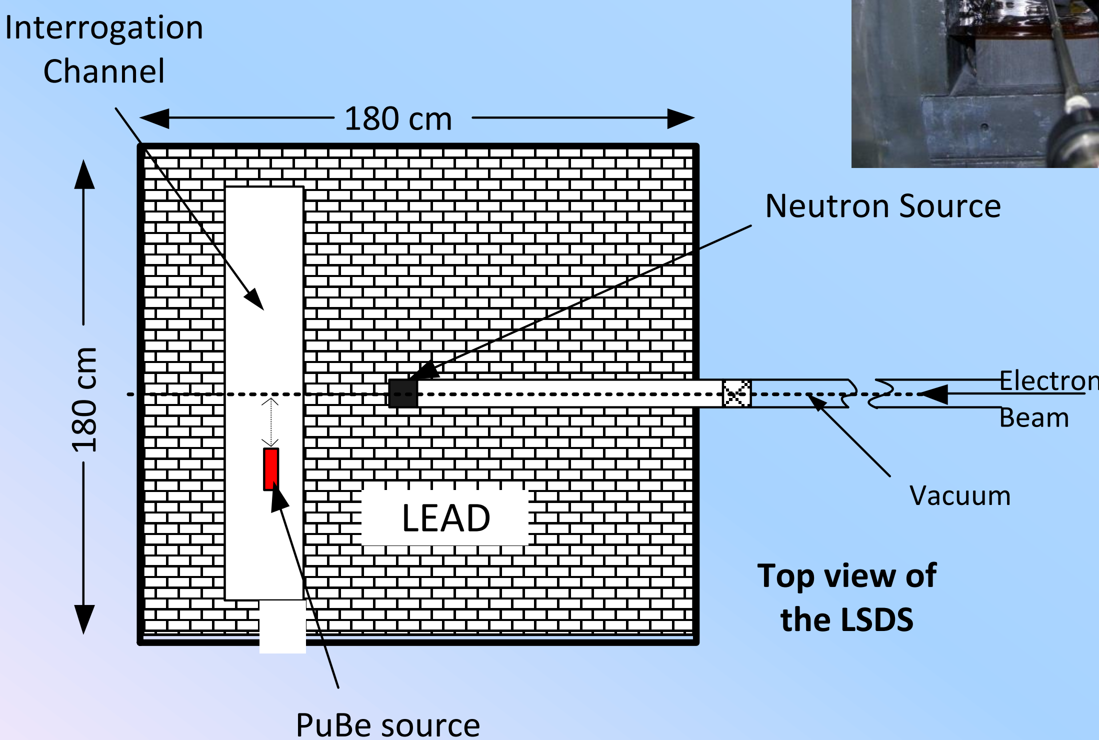


^{239}PuBe Used As a ^{239}Pu Sample

- 96 g ^{239}Pu
- Activity: 1.1×10^{7} n/s







Isotope	w %
^{239}Pu	60.83
^{240}Pu	3.84
^{242}Pu	0.07
^{241}Am	0.26
^{9}Be	35.00

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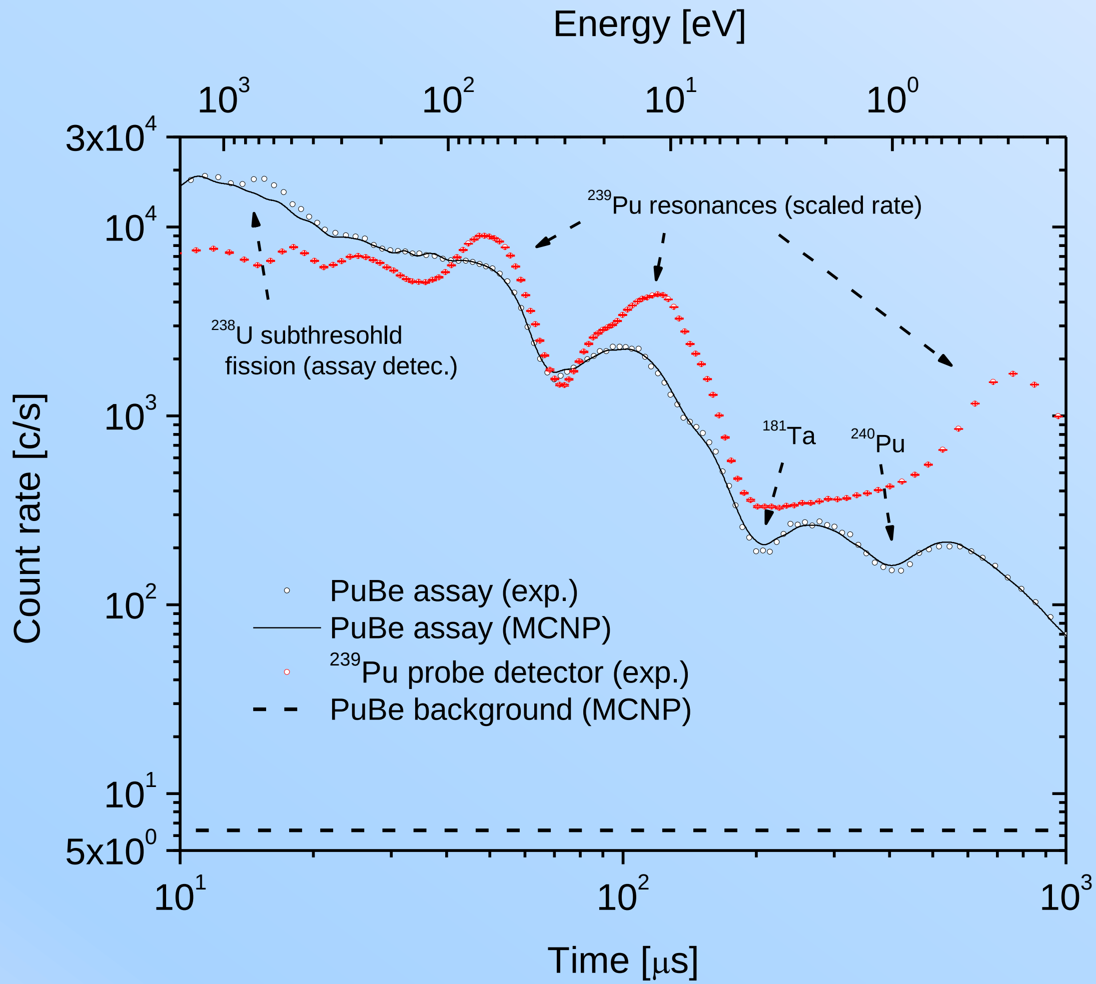
PuBe Source Assay

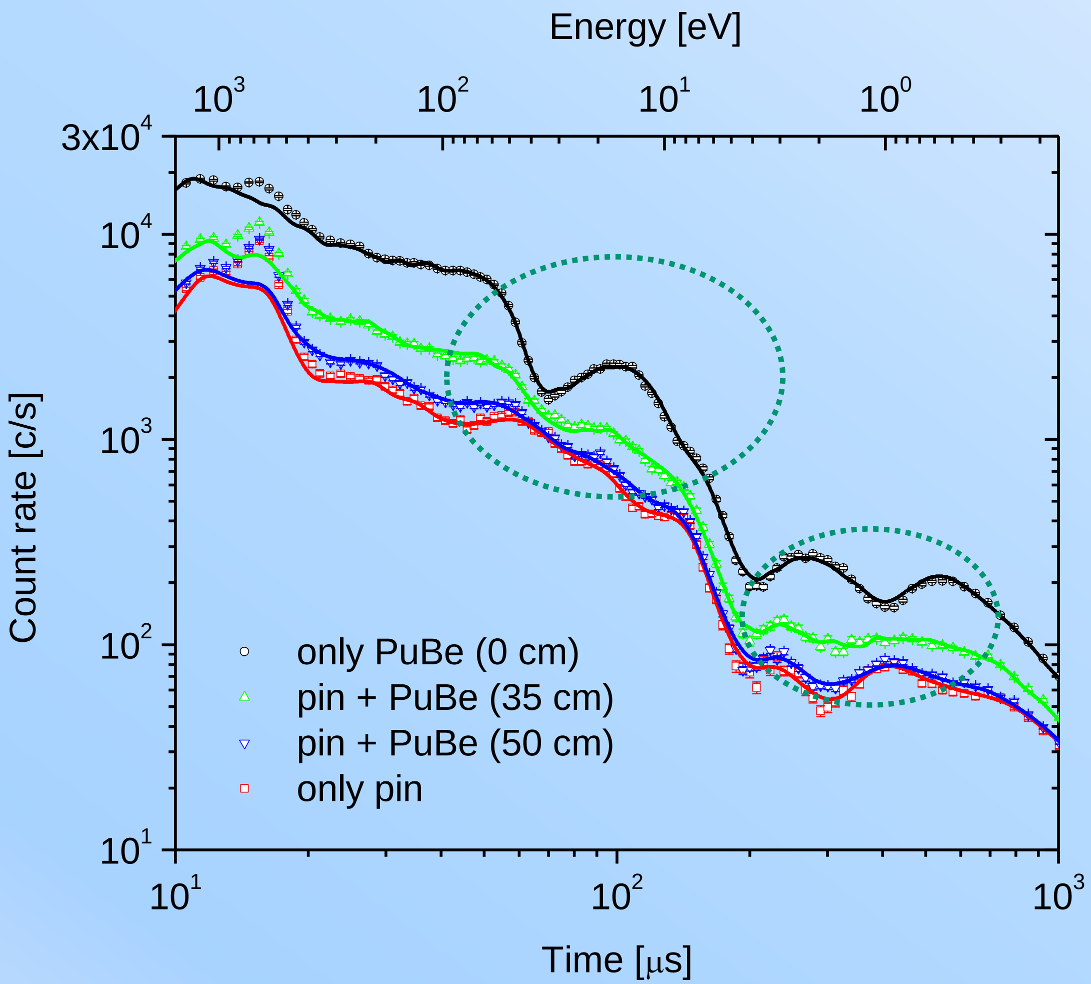
- PuBe source:
  - 96 g ^{239}Pu
  - Activity: $1.1x10^7$ n/s
- Comparison to Probe detector signal:
  - Strong self-shielding
  - ^{240}Pu shielding (mixed with ^{239}Pu)
  - ^{181}Ta shielding (source cladding)
- Background signal:
  - PuBe source: ~ 6.5 c/s



Assay of Fuel Pin and PuBe Source

- The PuBe source was placed at different insertion depths (measured from center)

- Change of the shape of the response function in particular in the 10 – 100 eV region and the 0.3 eV ^{239}Pu resonance



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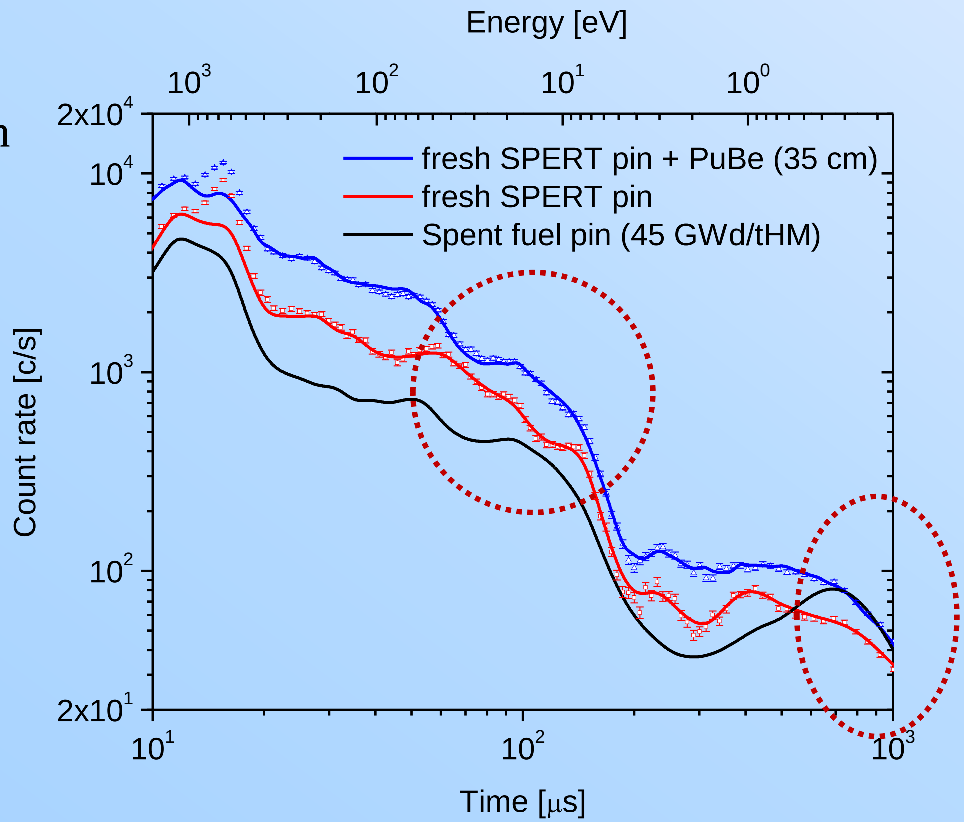
Comparison to a Spent Fuel Pin Assay

- Spent fuel pin model
  - Same geometric dim. as SPERT fuel pin
  - Fuel composition: “OECD Burn-up Credit Criticality Benchmark, Phase II-D; NEA No. 6227 - Case 13b”
  - 5 yrs cooling , 45 GWd/tHM

Nuclide	HM fraction [at%]
^{238}U	97.3
^{235}U	0.86
^{239}Pu	0.60
^{241}Pu	0.13

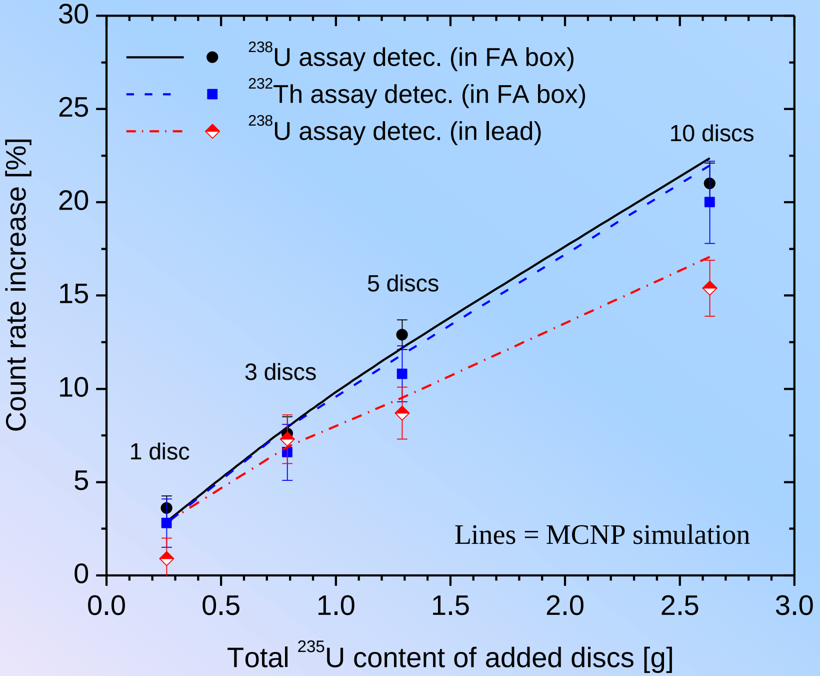


- Lower signal due to lower fissile content (^{235}U and ^{239}Pu)
- ^{239}Pu Peak at ~ 700 μs visible

- Similar structure in 40 – 120 μs frame
- No ^{240}Pu and ^{181}Ta shielding

Sensitivity to ^{235}U mass

- Measure a single fuel pin
  - ^{235}U mass is 35.2 g, 4.85% enrichment
  - Mass contributing to the assay detector ~10.6 g
- Add U (93% ^{235}U disks) ~0.25 g each
- Can detect <2% change is mass in < 10 min measurement

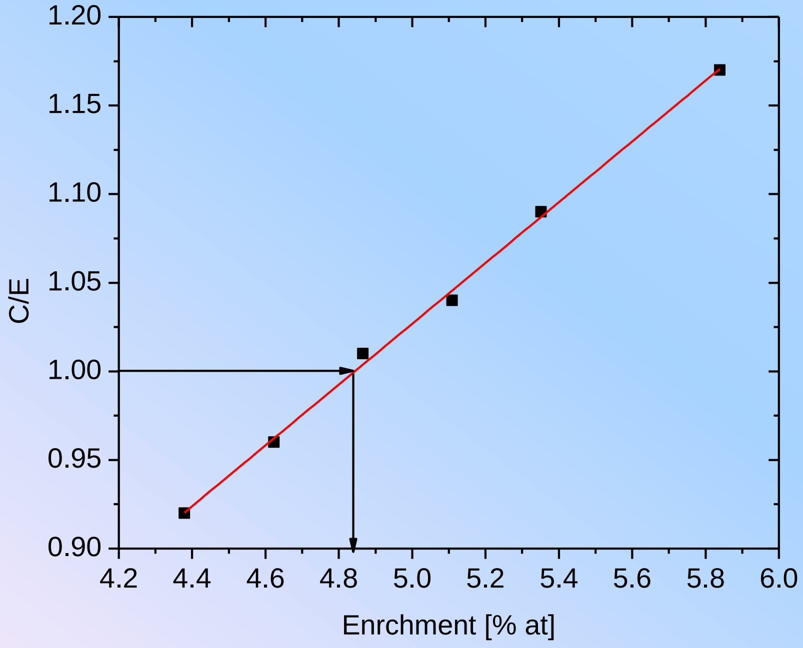


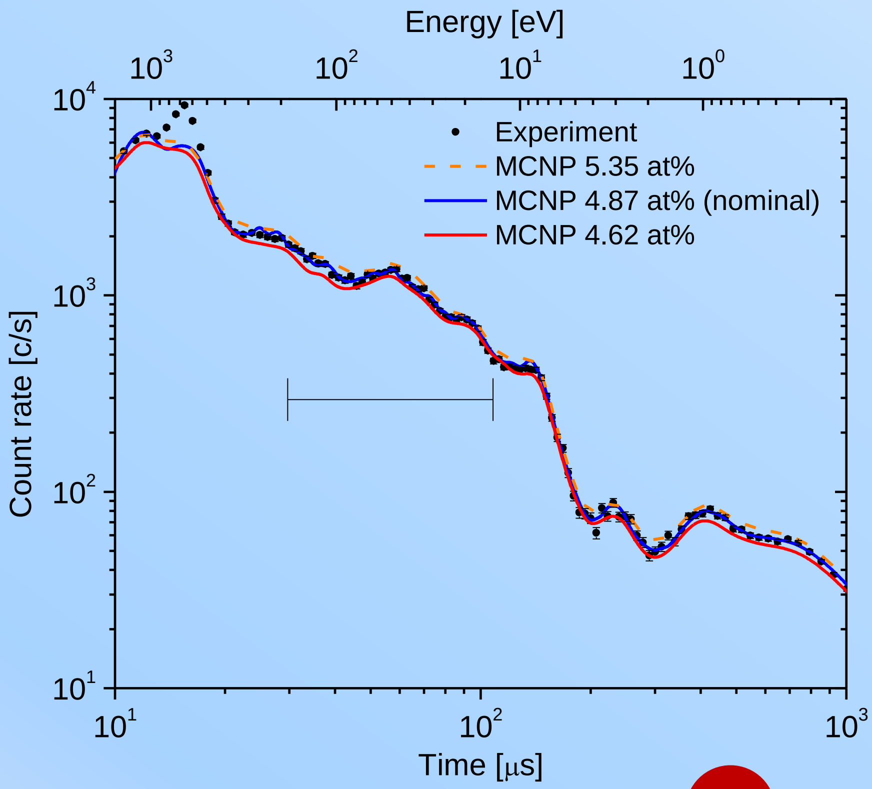
Fuel Pin +
^{235}U disks

Example – Enrichment determination

- Measure a single fuel pin
- Calculate the detector response for different enrichments
  - Plot C/E (30-110 μs) vs. enrichment

ε=4.81±0.10 %(know to be 4.8%)





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Summary

- **Basic theory of the Lead Slowing Down Spectrometer was reviewed:**
  - Time-Energy relation
  - Neutron Focusing
  - Energy resolution
  - Flux shape and intensity
- **The main advantage of the LSDS is a high neutron flux between 0.1 eV to 100 keV**
  - Can also be driven by low power pulsed neutron sources (DT sources)
- **Examples of applications were reviewed**
  - Fission cross section measurements
  - (n,a) cross section measurements
  - Capture reaction measurements
  - Measurements of fission fragment mass and energy distributions.
  - Assay of used spent fuel