History of Pu-239 fission cross section measurements

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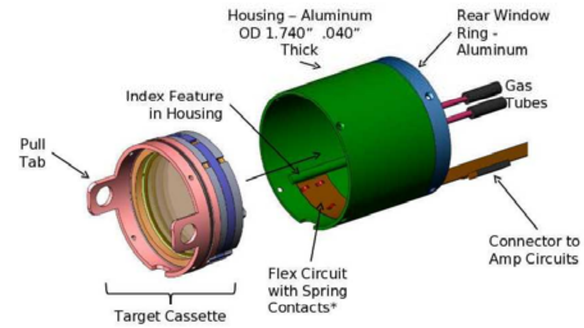
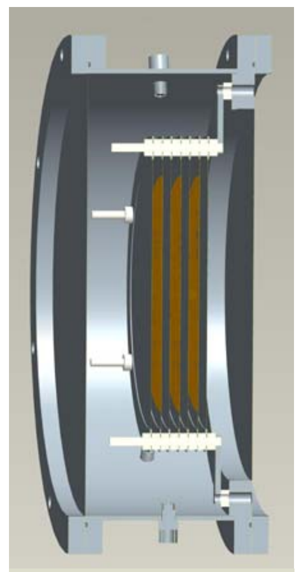
Outline

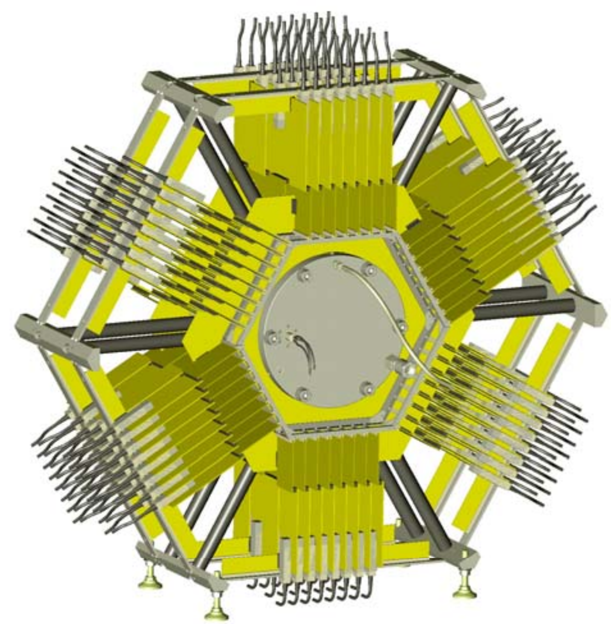
- **Experimental approaches**
  - Detectors
  - Neutron sources
  - Flux monitoring
  - Data analysis
  - Uncertainties
- **Experimental data in EXFOR**
  - Pre-1970
  - Absolute measurements
  - Relative measurements up to 20 MeV
  - Spallation source measurements

Detection methods

- **Ionization chambers**
  - Fritsch-gridded
  - Parallel plate
  - PPAC
- **Surface barrier**
- **Scintillators**
- **Track-etch detectors**







Neutron production

- **Quasi mono-energetic neutrons**
  - Accelerators: Van de Graaff (Geel, Ohio State U.), cyclotron (UU)
  - Li(p,n): 0 – 3.8 MeV (above 3.8 the Li(p,n’) reaction contributes)
  - T(p,n): 1.0 - 5.0
  - D(d,n): 3.5 – 10.0 MeV (deuterium break-up above 7 MeV)
  - T(d,n): 14.1-22.0 MeV
- **White neutron sources**
  - Photonuclear neutron production (ORELA, GELINA).
    - Electron beam (100 MeV) on uranium target (Geel). Energy range from sub-thermal to 20 MeV.
  - Spallation (LANSCE, PNPI, nTOF)
    - Proton beam on heavy target, such as lead or tungsten
    - Neutron range sub-thermal to hundreds of MeV’s

Flux measurements

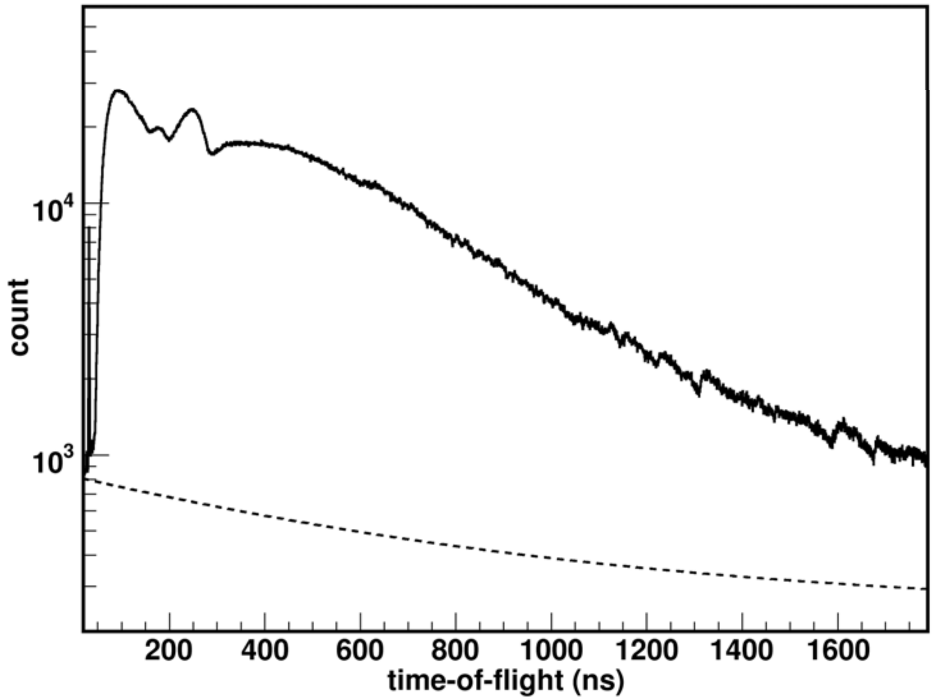
- **Relative cross section measurements**
  - U-235(n,f).
    - Standard evaluation
    - ~1% uncertainty in fast region
    - Many systematic uncertainties in the fission counting are reduced
  - H(n,n)H
    - The proton recoil in neutron scattering on hydrogen is detected
    - Uncertainty is fraction of percent
    - Detected with different system than the fission event = larger systematic uncertainties
  - B(n,α), Li(n, α)
    - Standards in the low-energy range (below 100 keV)
- **Absolute cross section measurements**
  - Associated particle method
    - In the neutron producing reaction the associated particle is detected, guaranteeing that a neutron was produced

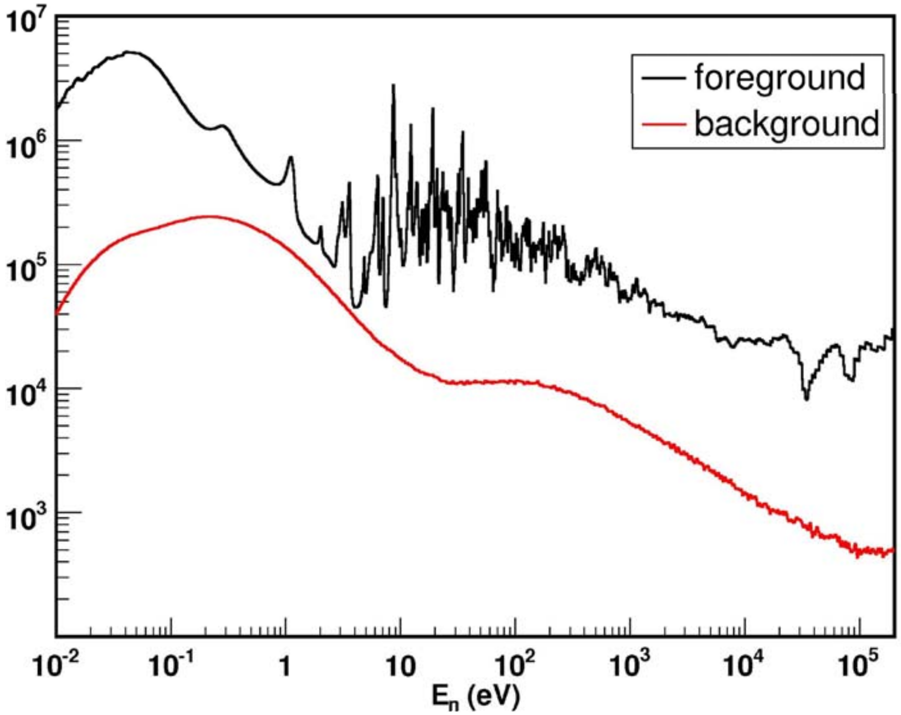
Cross section analysis

$$\frac{\sigma_{Np}}{\sigma_U} = \frac{N_U}{N_{Np}} \cdot \frac{\varepsilon_2(E)}{\varepsilon_1(E)} \cdot \frac{\Phi_2(E)}{\Phi_1(E)} \cdot \frac{w_1^{-1}(E) \cdot C_1(E)}{w_2^{-1}(E) \cdot C_2(E) - C_2^b(E)} - \frac{N_{Pu}}{N_{Np}} \cdot \frac{\sigma_{Pu}(E)}{\sigma_U(E)} - \frac{N_{U3}}{N_{Np}} \cdot \frac{\sigma_{U3}(E)}{\sigma_U(E)}$$

- **Statistical uncertainties**
- **Energy independent systematic uncertainties**
- **Energy dependent systematic uncertainties**

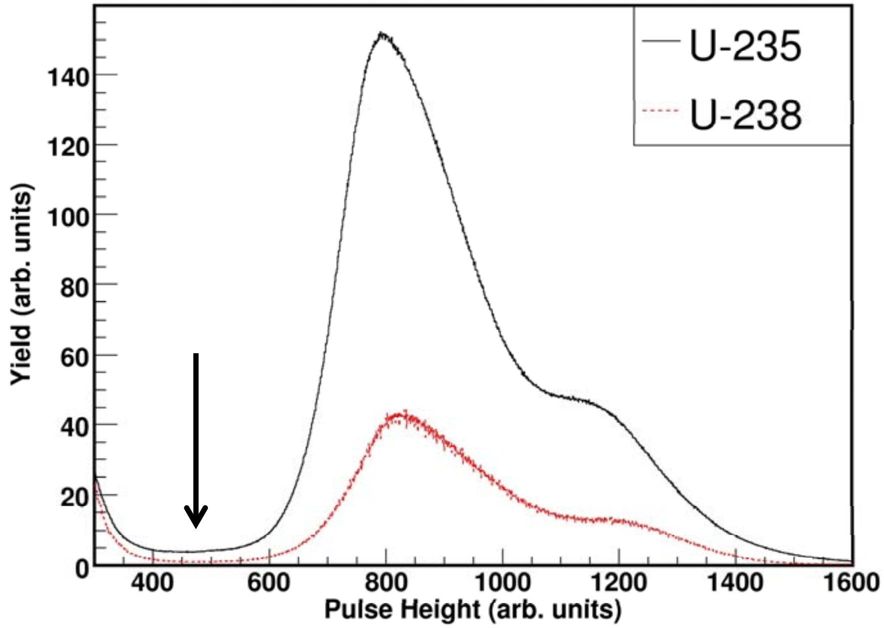
Neutron background

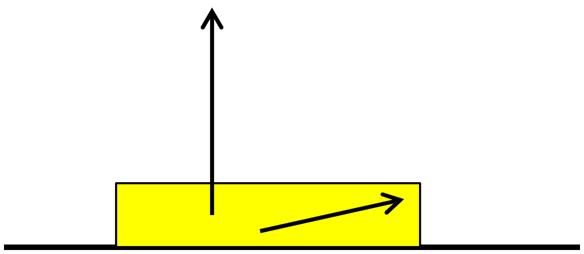




- **Neutron background causes fission events that needs to be subtracted**
- **Room scattering – neutrons scatter on collimation, air, detector resulting in loss of correlation between TOF and energy**
- **Frame overlap – Short pulse structure in TOF measurements results in "wrap-around" neutrons**

Detection efficiency





- **Fission fragment absorption in sample**
  - Thin (<200 ug/cm2) samples are typically used. Still, some 2-3% of events are absorbed
- **Energy straggling**
  - Straggling causes a tail of the energy distribution, and some events fall below detection threshold

Dead time corrections

$$p_i = \frac{N_i}{N_p - \sum_{j=i-D-1}^{i-1} N_j} = w^{-1} \cdot N_i$$

P. B. Coates, Rev. Sci. Instrum. 63, 1992

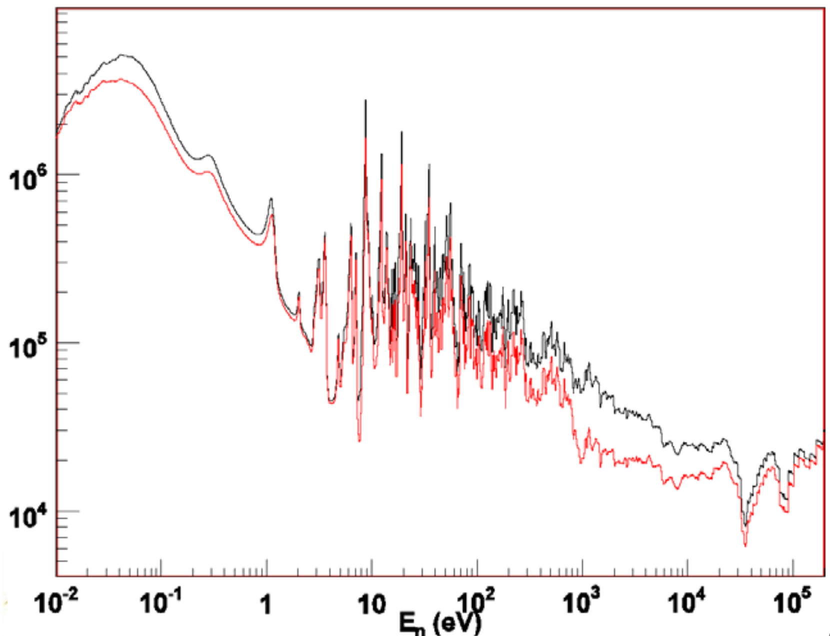
p_i = probability (per T0) to have event in bin i

N_i = measured events in bin i

N_p = number of T0's

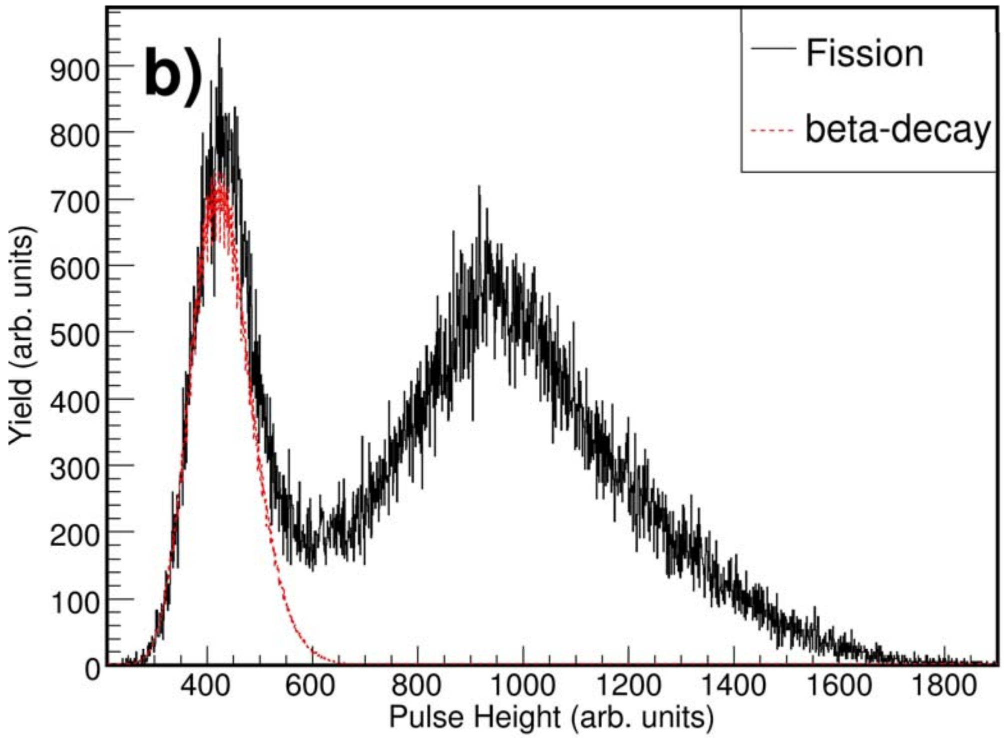
D = deadtime in bins

^{235}U events



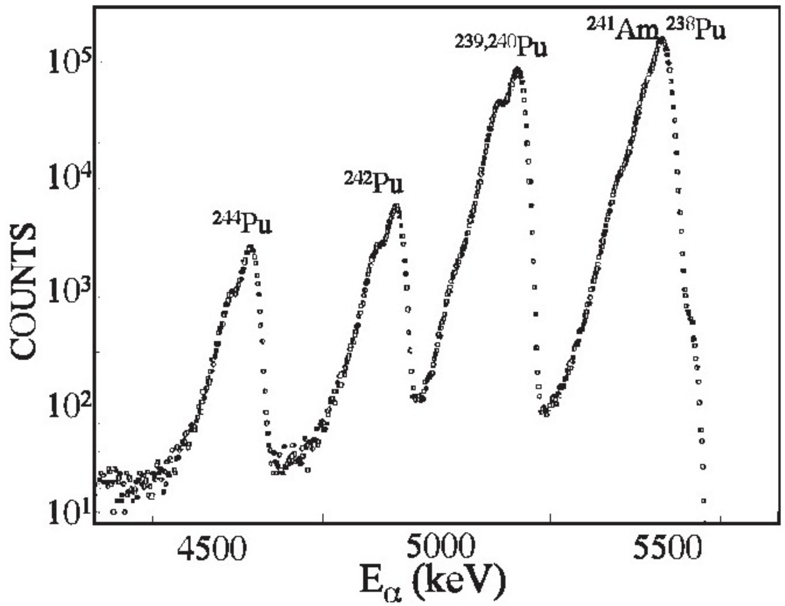
- The dead-time correction is exact at constant event rates. We correct every 10 seconds worth of data, assuming stable beam on this time scale.
- Hardware scalers measures the integral live-time. The only input parameter D is fine tuned until there is perfect agreement with scalers.

Fission identification



- **Background events**
  - Radioactive decay
  - Neutron-induced charged-particles
  - Gammas
- **Rejection criteria**
  - Total energy deposited
  - TOF

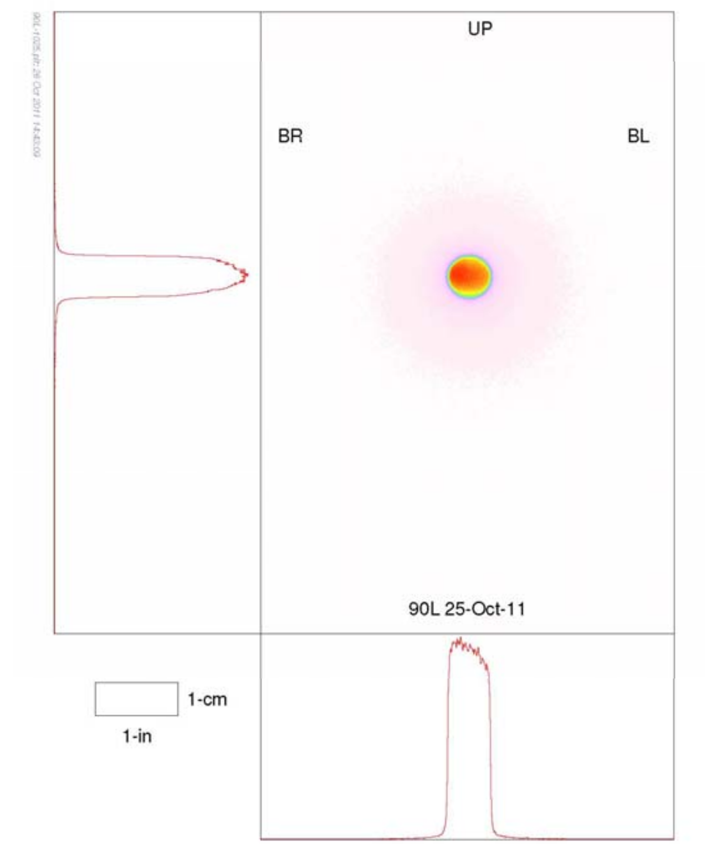
Mass determination



Spectrum from alpha counting of Pu-244 samples used for fission cross section measurement by Staples and Morley

- **Samples are typically counted relative to standards. Pu-239 samples are alpha- or gamma counted.**
- **Enriched samples are needed for fission cross sections. Pu-239 is available with higher than 99% enrichment. Contamination levels can be determined using alpha spectroscopy.**

Sample and beam profile non-uniformities



- **Sample uniformity**
  - Highly dependent on method of preparation
  - Evaporated samples can be uniform to within 1-2%
  - Can be measured using counting with mask
- **Beam profile**
  - Imaging techniques used to determine profile
  - Typically not energy differential

Covariance of experimental uncertainties

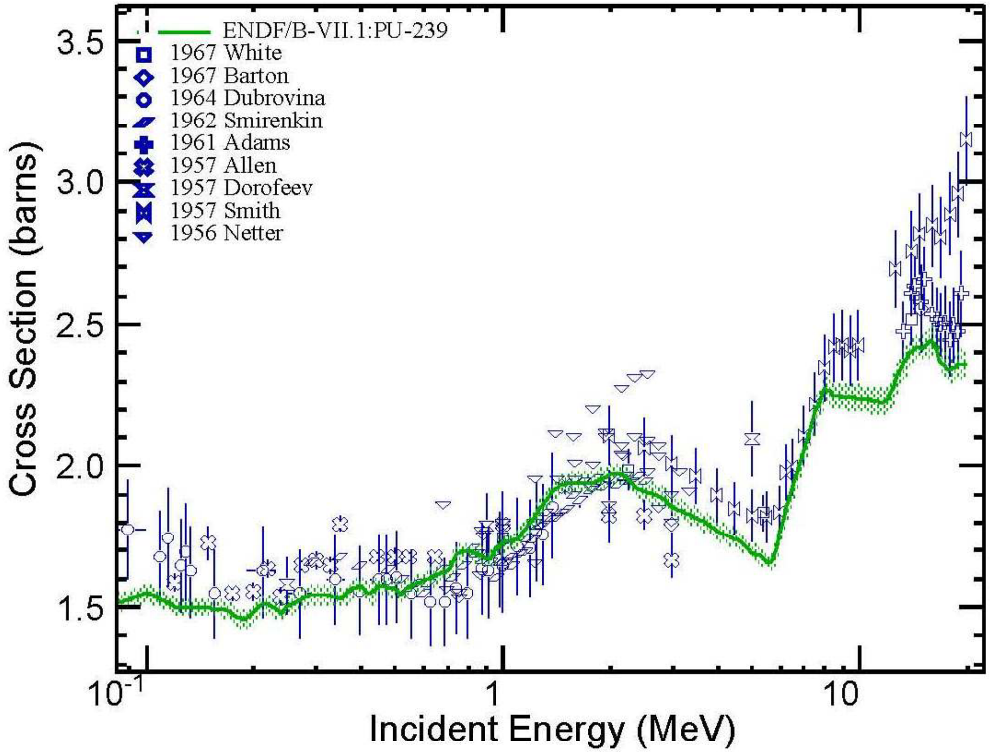
Elements in the covariance matrix are given by

$$C_{ij} = R_i R_j \left[\sigma_s^2 \delta_{ij} + d_i d_j + b_i b_j + u_i u_j + p_i p_j + \sigma_n^2\right]$$

Where

$R_{i,j}$ = measured ratio value
σ_s = relative statistical uncertainty
$d_{i,j}$ = relative uncertainty due to dead-time corrections
$b_{i,j}$ = relative uncertainty due to background corrections
$u_{i,j}$ = relative uncertainty due to U-233 contamination corrections
$p_{i,j}$ = relative uncertainty due to Pu-239 contamination corrections
σ_s = normalization uncertainty

Experimental work pre-1970

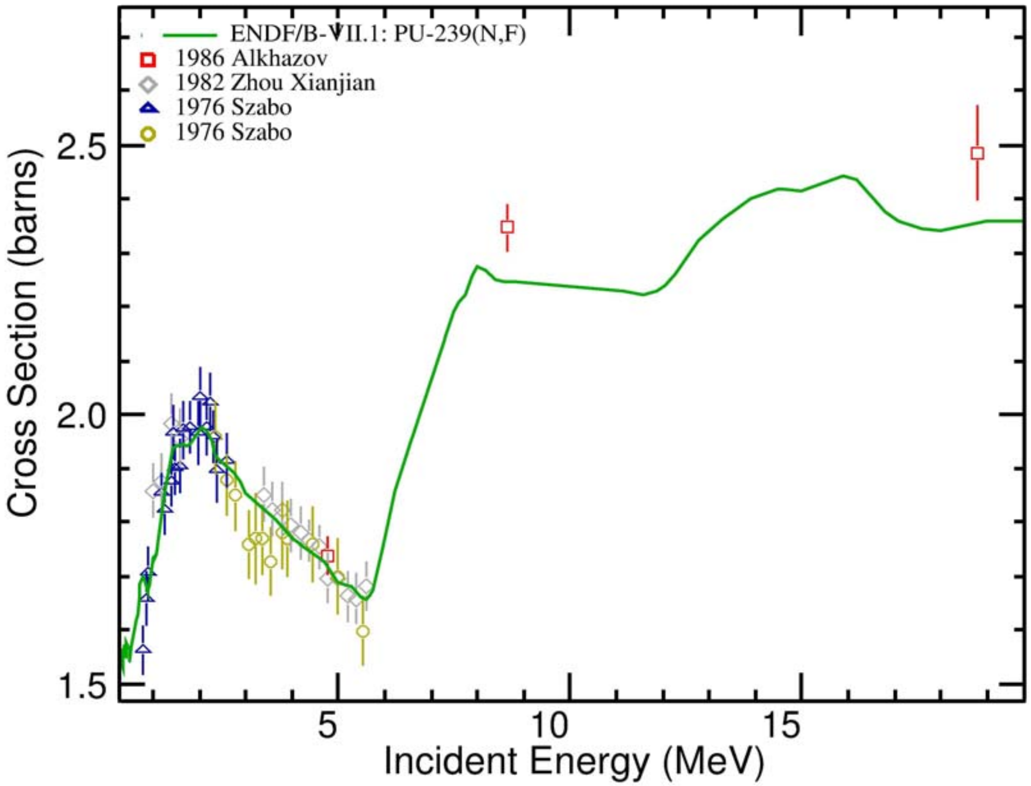


- Generally higher cross sections than current evaluation
- Large discrepancies above 10 MeV
- Fairly large experimental uncertainties, or non at all

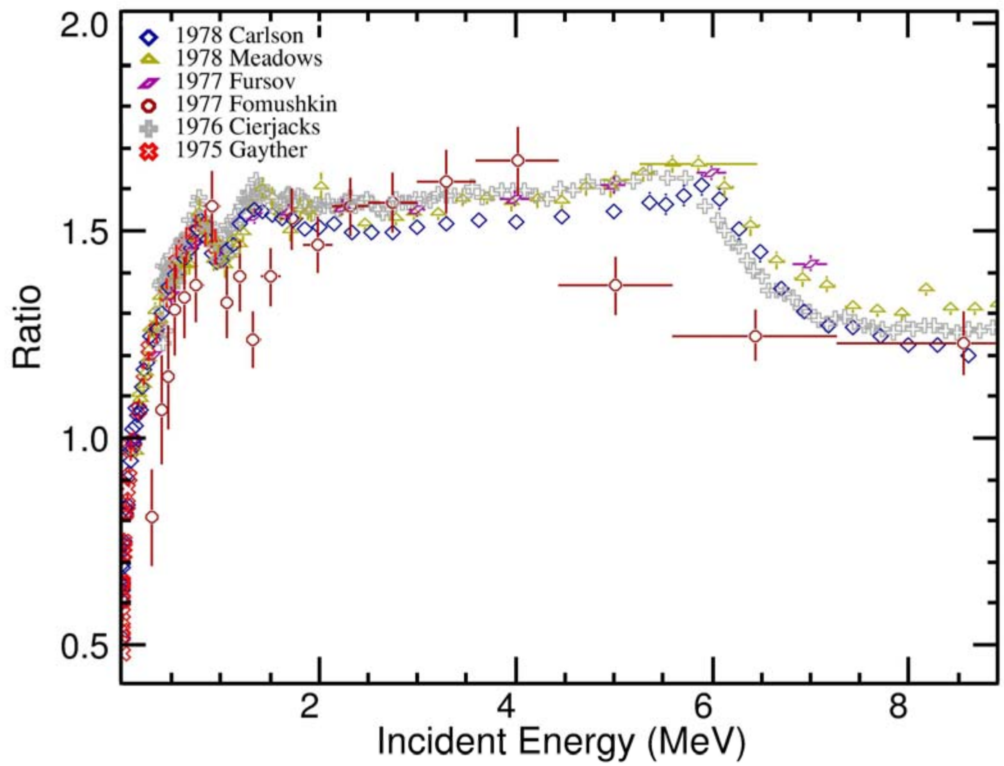
Absolute measurements

Author	Year	Energy (MeV), points	Neut. Prod.	Detector	Flux mon.
I. D. Alkhazov et al.	1986	4.8-18.8, 3	D-D, D-T	Ion. Chamber	Assoc. part, α
Xianjian et al.	1982	1.0-5.8,16	P-T, D-D	Ion. Chamber	H(n,n)H, p
M.C.Davis et al.	1978	0.14-0.96, 4	Na-Be, La-Be, Na-D, and Ga-D	track-etch detectors	Calibrated N-source
K. Kari	1978	0.99-20.9, 168	?, cyclotron, TOF	Gas scint.	H(n,n)H, p
I. Szabo et al.	1976	0.035-5.53, 54	P-Li, P-T,D-D	Frisch	Assoc. part, H(n,n)H

Absolute measurements

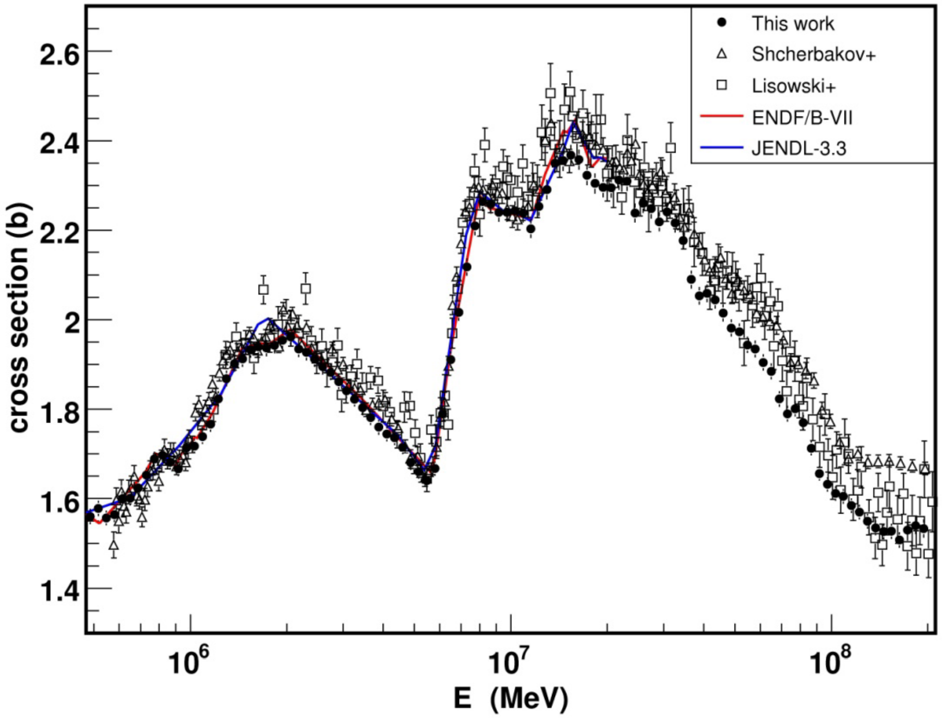


Relative measurements in the fast region



- All relative to U-235
- Fairly consistent, except for Fomushkin
- Discrepancies are generally around 5% above 1 MeV

Measurements at spallation sources



- **Lisowski et al. performed measurements in late 1980's at LANSCE**
- **Shcherbakov et al. made similar measurements at PNPI in 1990's. Difference: flight path length, pulse spacing.**
- **New measurements at LANSCE performed in recent years**

Summary

- Large data base of Pu-239 energy differential fission cross sections.
- Most data set were collected with the same type of detectors, neutron sources, samples.
- The uncertainties in all individual measurements are >1%. More realistically >2%.
- Fairly large discrepancies exist above 10 MeV.
- There is plenty of measurements relative to U-235. An absolute, or relative to H(n,n), would be of significantly more value.