How we can improve nuclear fission data for applications and fundamental physics

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Physics with Secondary Hadron Beams in the 21st Century

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Why neutrons?

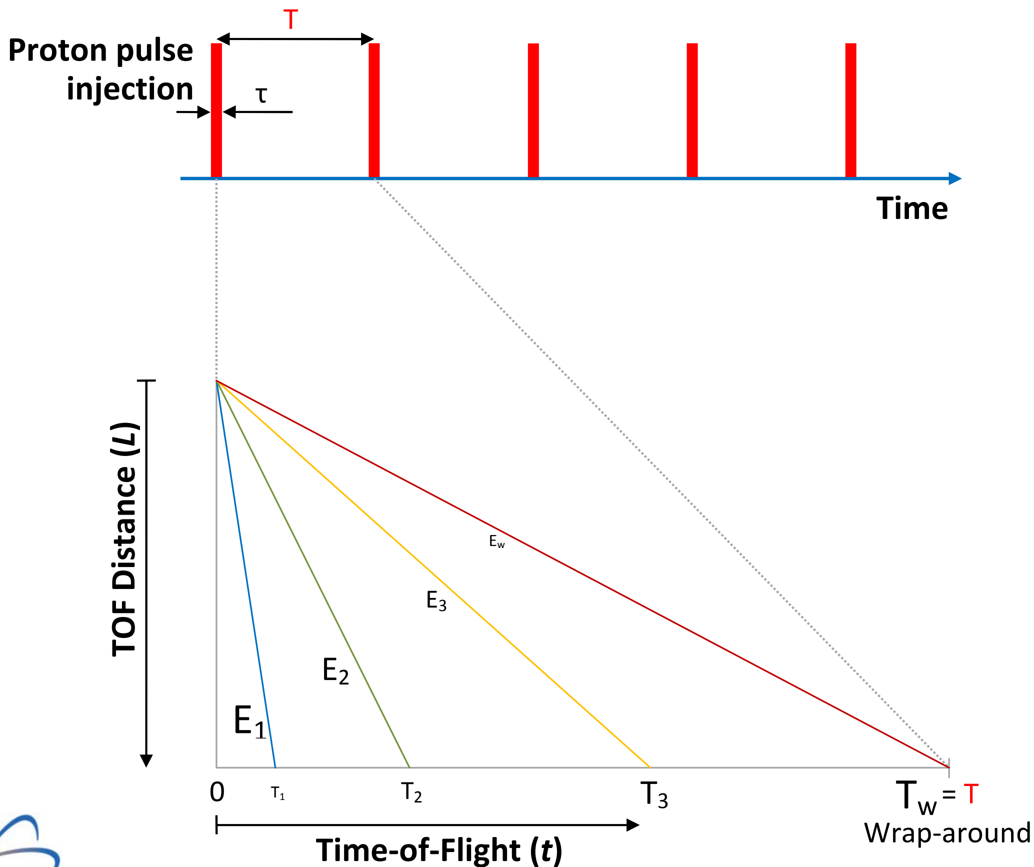
- **High demand from nuclear industry for good quality data, first of all neutron-induced reaction cross sections for actinides and sub-actinides**
  - **Development of new advanced nuclear reactors**
  - **Decreasing nuclear data uncertainties will decrease nuclear reactor construction safety margins incorporated into reactor's design**
  - **Industry keeps developing: in Dec. 2011 NRC approved first nuclear power plant license since 1978 for the power plant expansion in Georgia**
  - **Public pressure is as high as ever**

Why neutrons (cntd)?

- Defense physics needs
- Accelerator-based conversion of weapon's grade plutonium
- Transmutation of nuclear waste, especially actinides
  - Yucca Mountain nuclear waste repository doesn't accept new waste effective in 2011
- Basic understanding of fission process, fundamental physics, etc.

Why a pulsed neutron source?

- **The most accurate and effective method to measure neutron energy is the time-of-flight method**



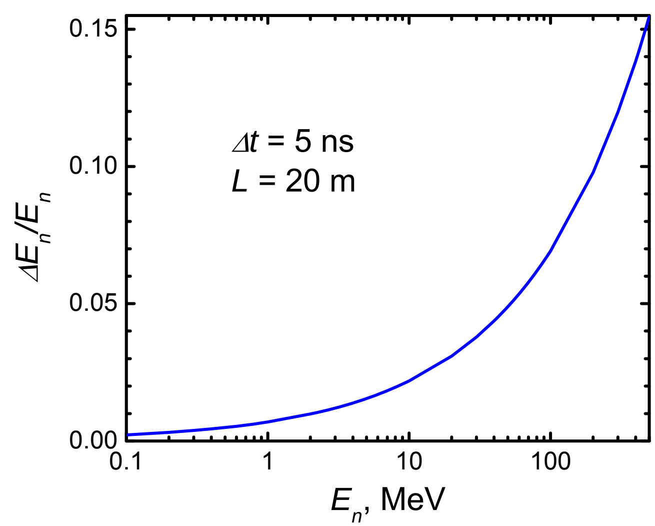
Handbook of Spallation Research. Wiley-VCH, Berlin, 2009

$$E_n = \left(\frac{72.3 \cdot L}{t}\right)^2$$

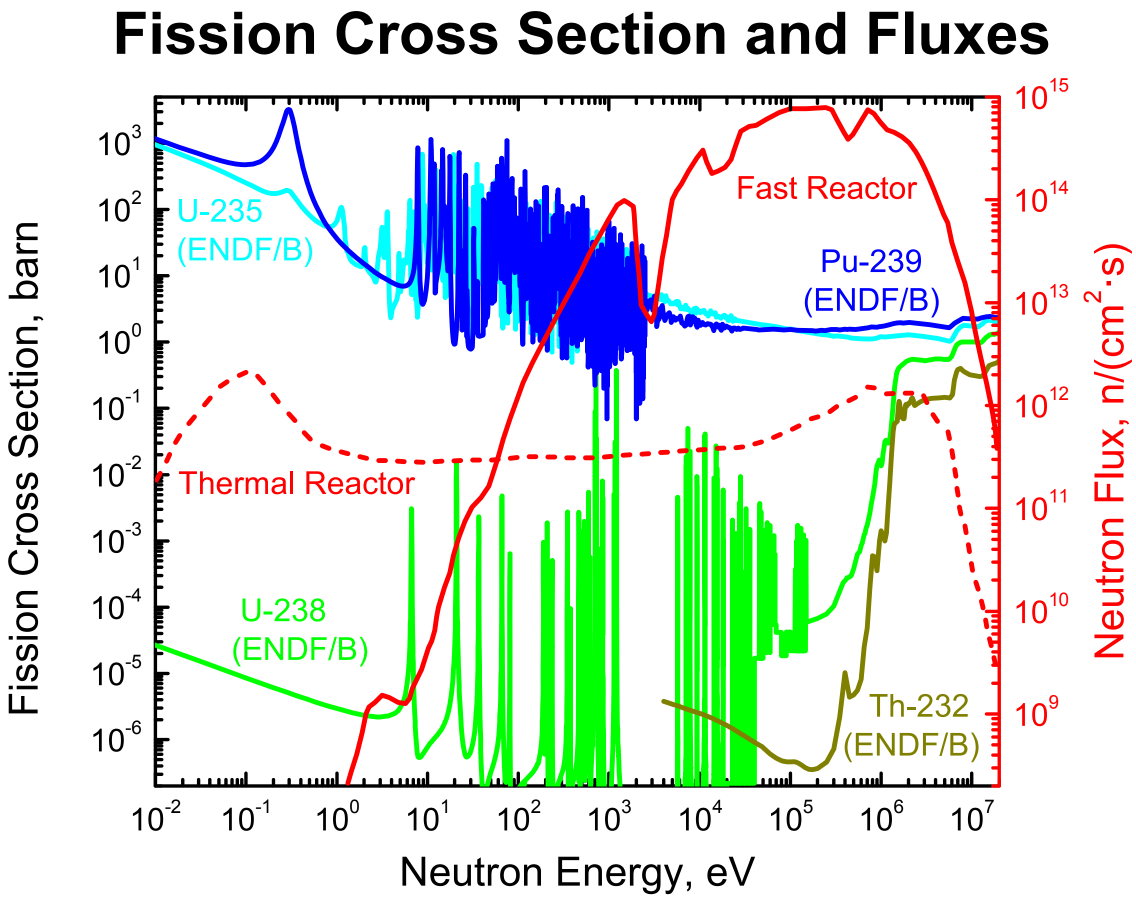
$$\frac{\Delta E_n}{E_n} = 2\frac{\sqrt{E_n} \cdot \Delta t}{72.3 \cdot L}$$

Nonrelativistic approximation for simplicity
E_n is in eV
t is in μs
L is in meters

$$\Delta t = \tau + \cdots$$

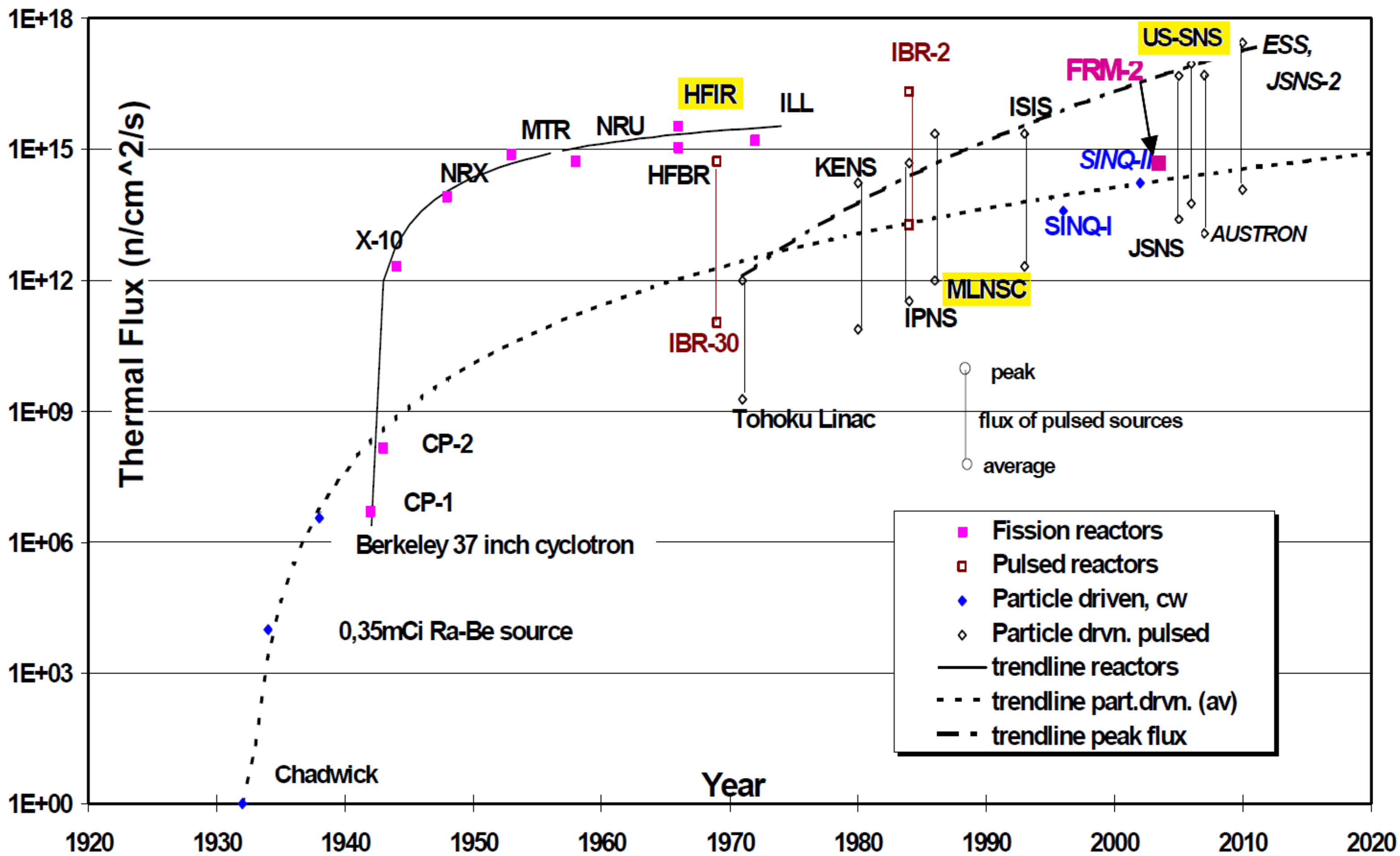


What is the energy range of interest?



- Neutron flux shapes folded with cross sections determine the region of interest (both energy and isotopes)
- The magnitude of the neutron flux drives cross section uncertainty requirements

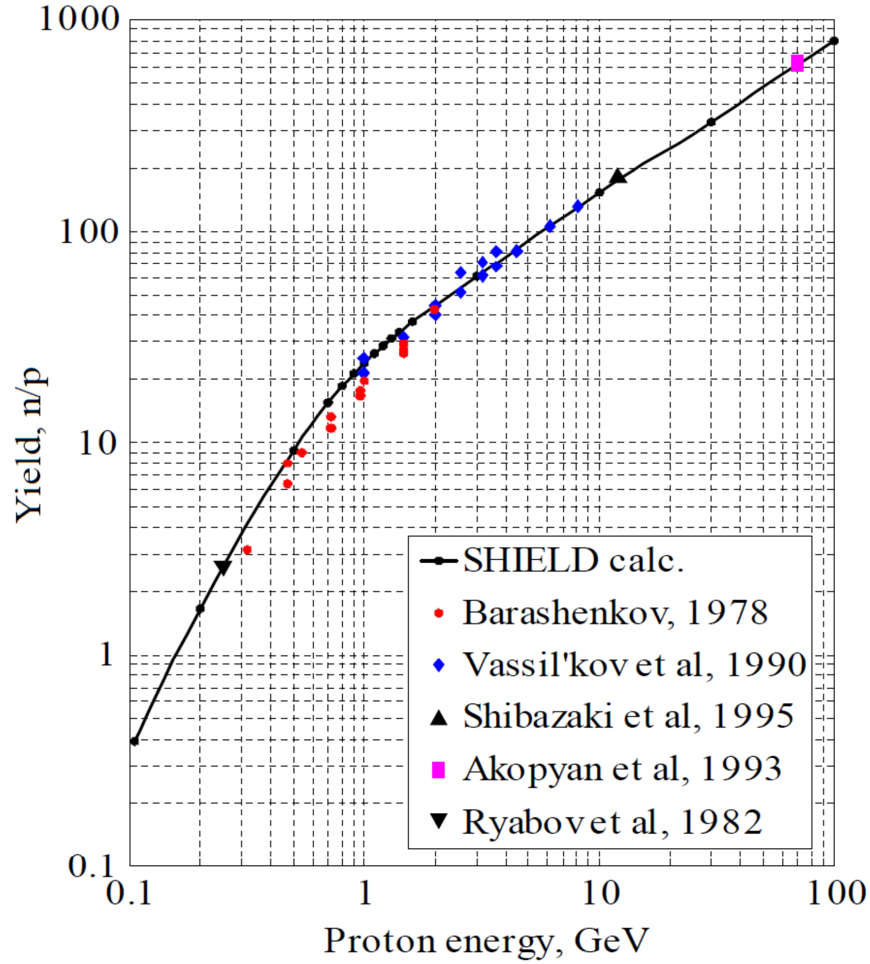
History of neutron source development



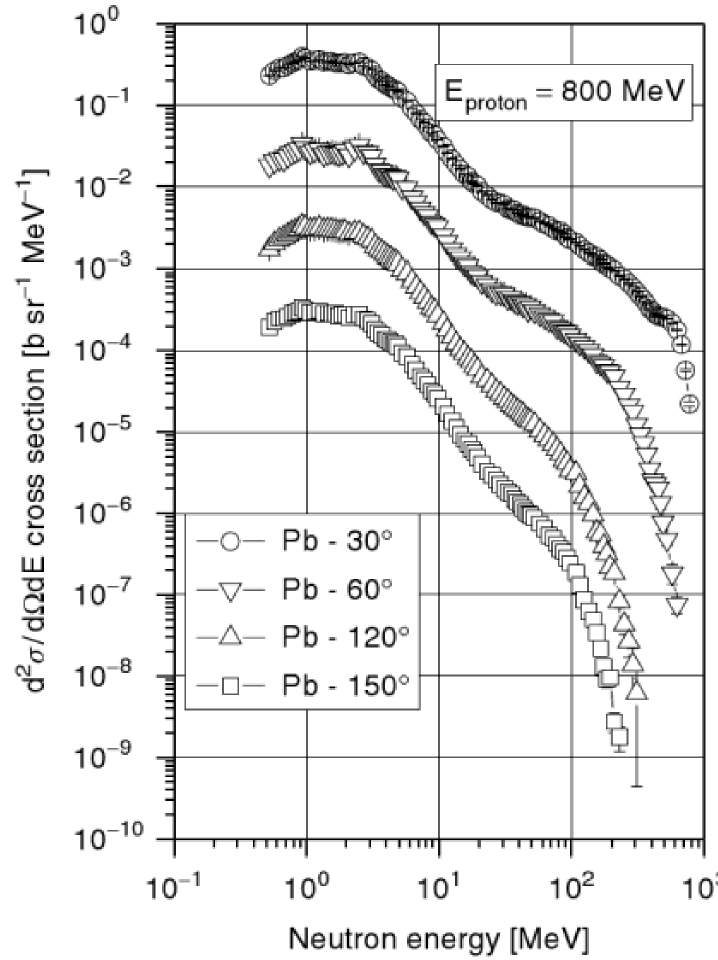
Highlighted are active facilities in the US

IAEA-TECDOC-1439, 2005

Spallation sources: n/p yield and neutron energies (thick Pb target)



NIM A 374 (1996) 70
Preprint INR RAS 1280, 2011



Nucl.Sci.Eng. 112 (1992) 78

Two components:

- The cascade component up to the energy of the incident protons

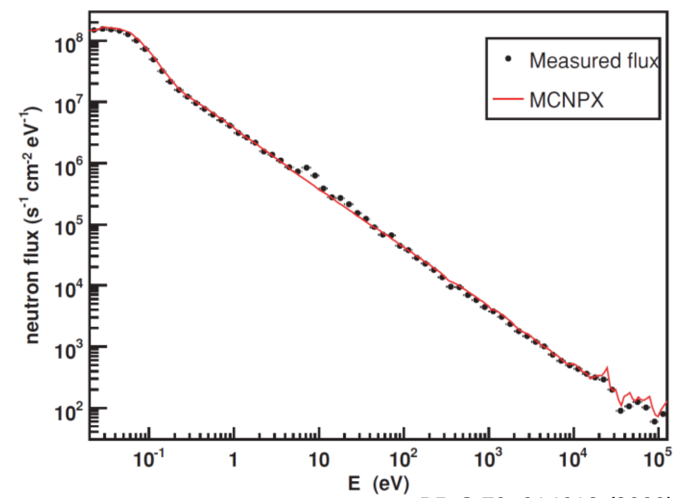
- The evaporation component which is dominant up to ~20 MeV

Moderated neutron sources

- Enrich neutron flux with epithermal neutrons
  - High content hydrogen material is used for effective neutron moderation, e.g. polyethylene, water, etc.
  - Varying of moderator thickness one can change the ratio of faster/slower neutrons
- Very specific moderator – liquid hydrogen – provides cold and ultra cold neutrons. This is a very unique field of neutron physics

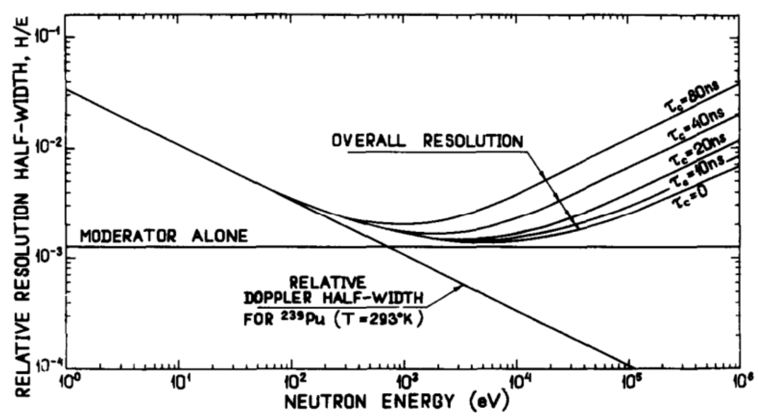
Details about this field are in the following report of S.Dewey

Neutron flux of moderated source, LANSCE



PR C 79, 014613 (2009)

Time resolution of moderated source, GNEIS



NIM A 242 (1985) 121

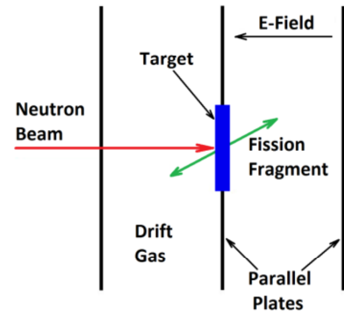
What facilities are available for fast neutrons?

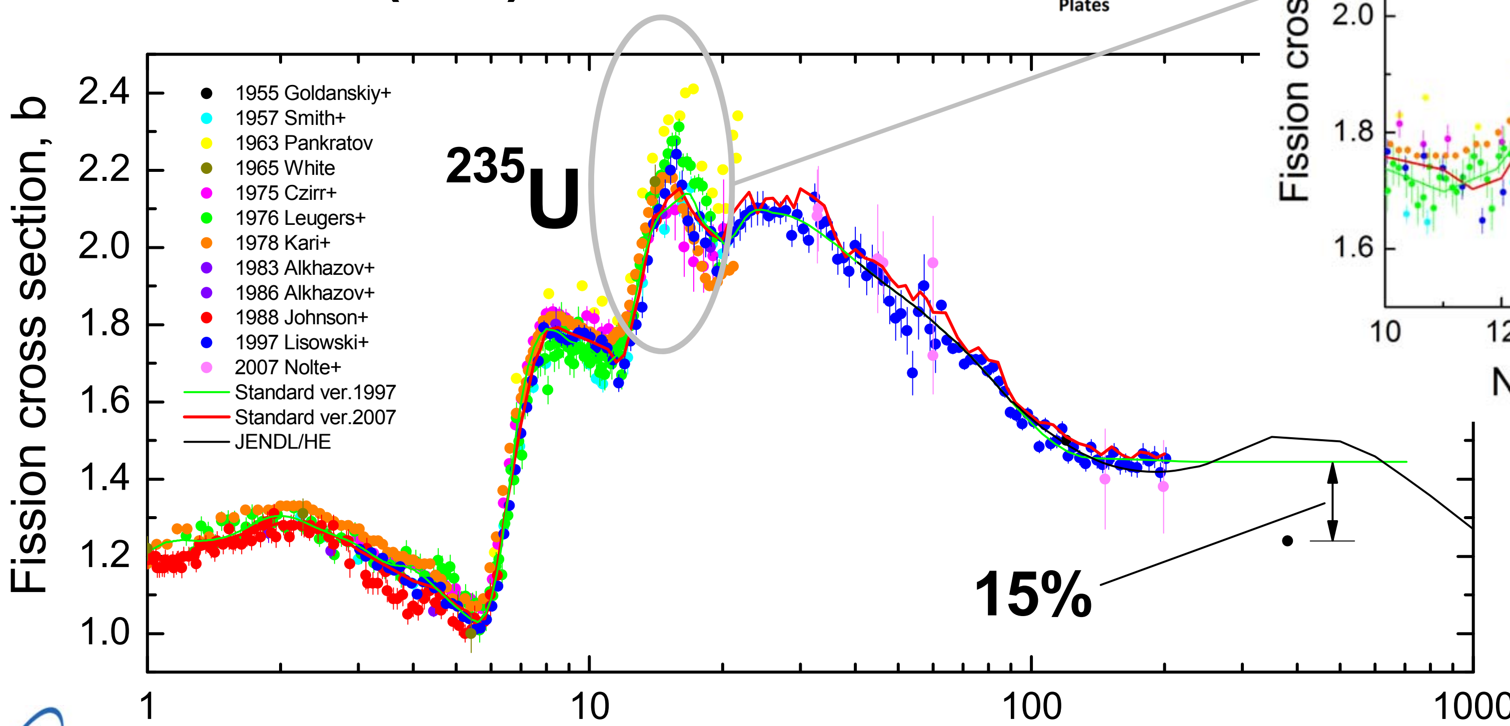
	WNR	GNEIS	n_TOF	SNS	J-PARC
Proton energy, MeV	800	1,000	20,000	1,000	3,000
Proton current, μA	1.8	2.3	0.5	1,400	300
Target	W	Pb	Pb	Hg	Hg
Number of produced neutrons per proton	10	20	250	25	75
Total neutron yield per second	$1\cdot10^{14}$	$3\cdot10^{14}$	$8\cdot10^{14}$	$2\cdot10^{17}$	$1.5\cdot10^{17}$
Proton pulse length on target, ns	0.2	10	6	700	1,000
Pulse frequency, Hz	13,900	50	0.4	60	25

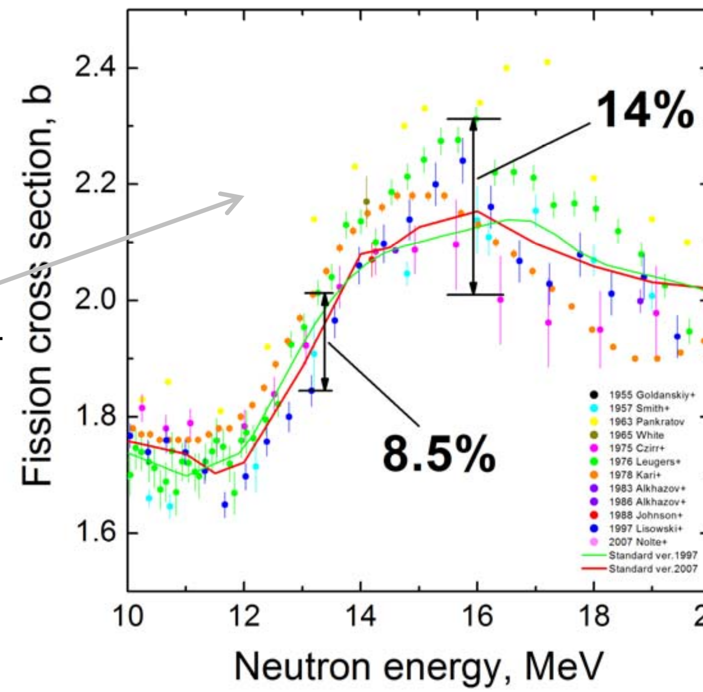
Handbook of Nuclear Engineering, Springer, 2010
NIM A 242(1985)121

Existing ^{235}U(*n,f*) XS data for fast neutrons

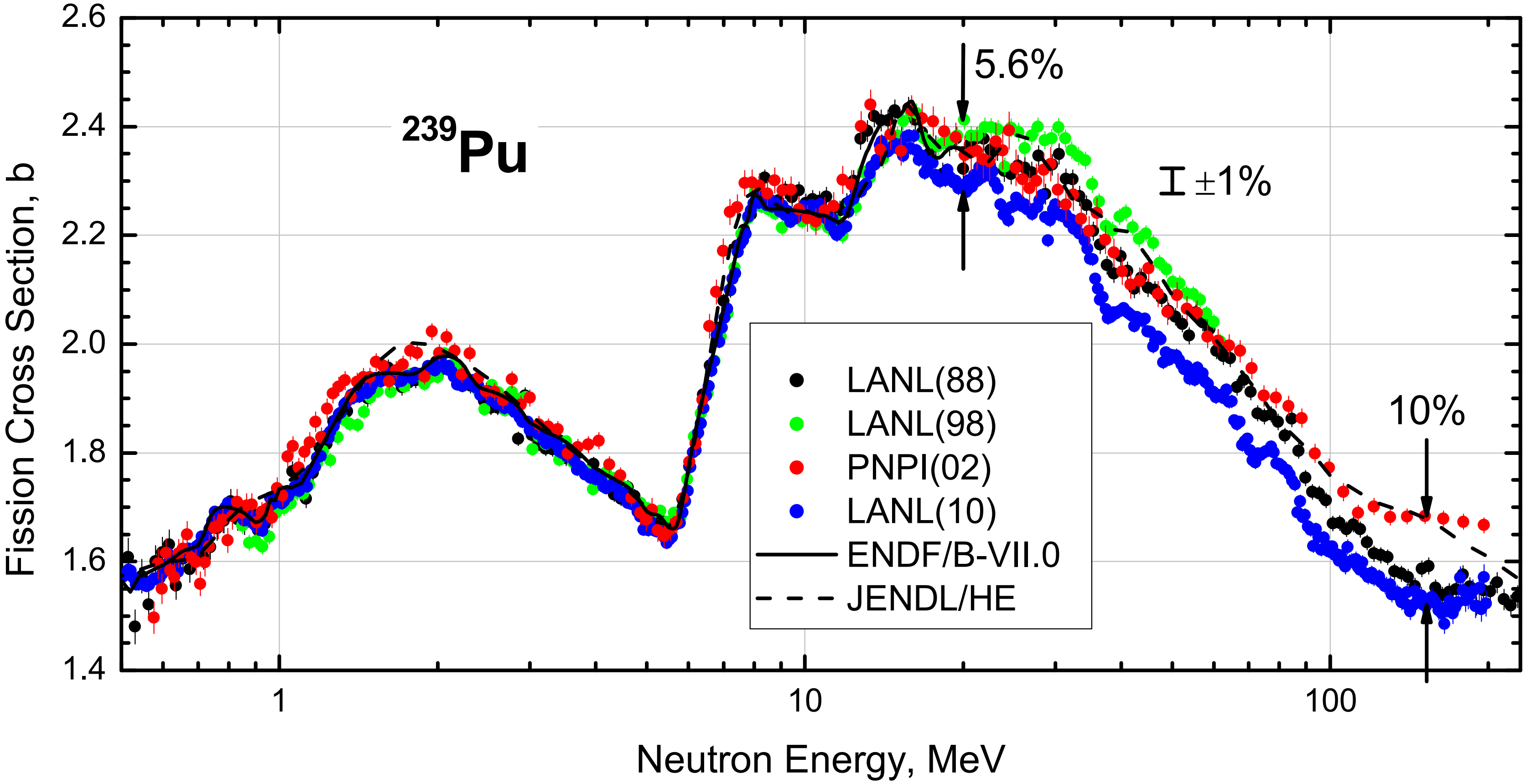
- **The most popular detector is a parallel-plate fission ionization chamber (FIC)**





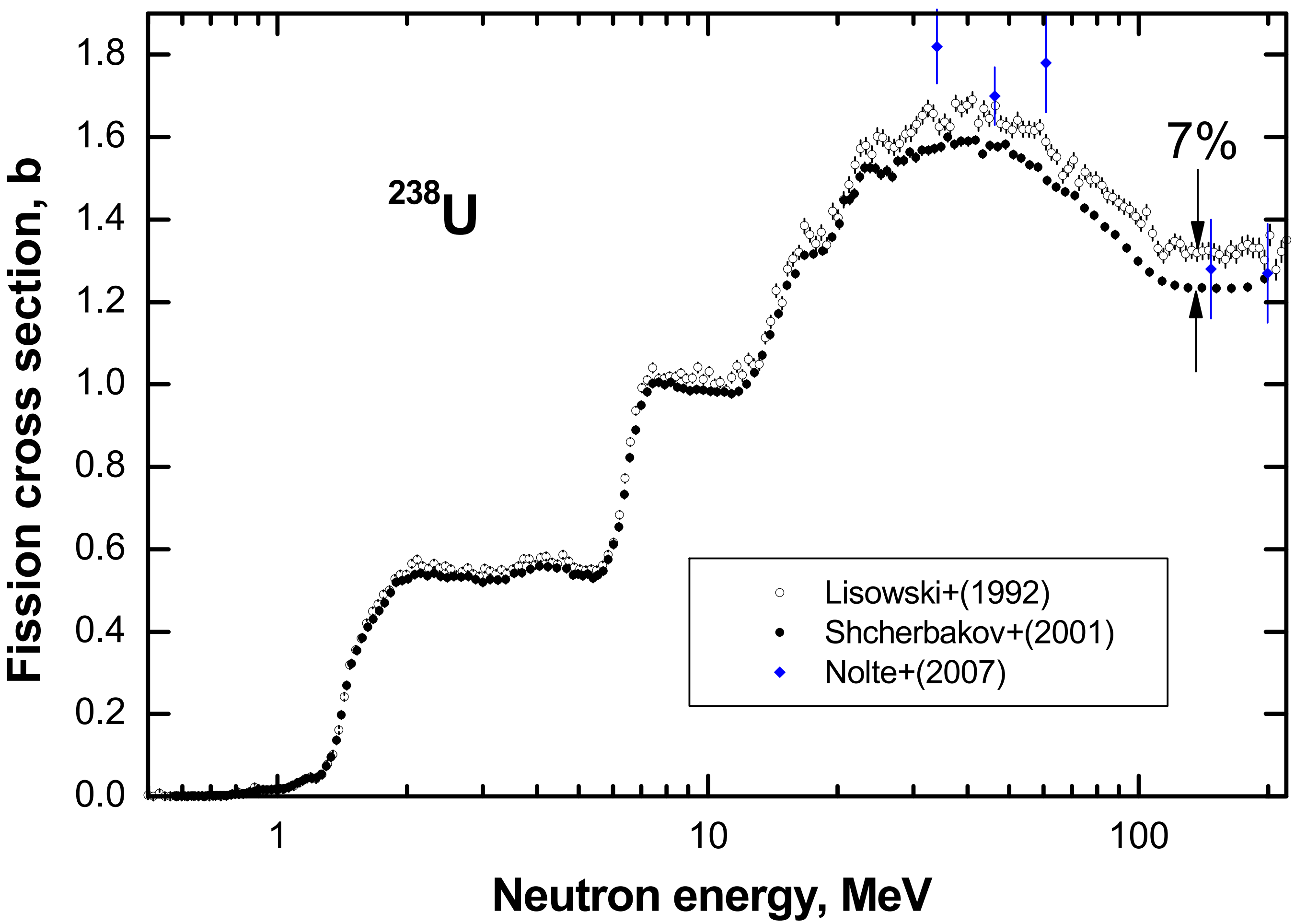


Existing $^{239}Pu(n,f)$ XS data for fast neutrons



***Ref.**:*
LANL(88): Lisowski et al., ND-1988(1988)97
LANL(98): Staples et al., Nucl.Sci.Eng. 129(1998)149
PNPI(02): Shcherbakov et al., J.Nucl.Sci.Tech. S2(2002)230
LANL(10): Tovesson et al., Nucl.Sci.Eng. 165(2010)224

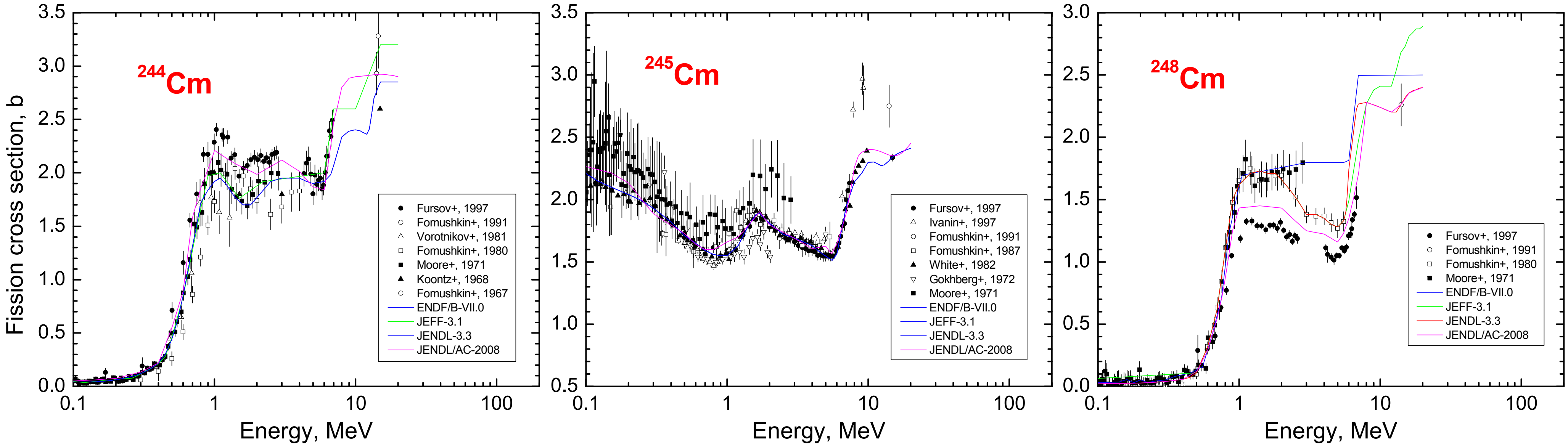
Existing $^{238}U(n,f)$ XS data for fast neutrons



- Data were taken using FIC as a ration to ^{235}U (LANL and PNPI)
- Data of Nolte were obtained relative to *np*-scattering

Ref.:
Lisowski: Lisowski et al., ND-1991(1992)732
Shcherbakov: Shcherbakov et al., J.Nucl.Sci.Tech. S2(2002)230
Nolte: Nolte et al., Nucl.Sci.Eng. 156(2007)197

Existing data for fast neutron-induced fission of some minor actinides



From the NEA Nuclear Data High Priority Request List for fission cross section of minor actinides

^{244}Cm	Initial *vs* target uncertainties (%)	
Energy Range	**Initial**	**ADMAB**
6.07 - 2.23 MeV	31	3
2.23 - 1.35 MeV	44	3
1.35 - 0.498 MeV	50	2

^{245}Cm	Initial *vs* target uncertainties (%)	
Energy Range	Initial	ADMAB
2.23 - 6.07 MeV	31	7
1.35 - 2.23 MeV	44	6
0.498 - 1.35 MeV	49	3

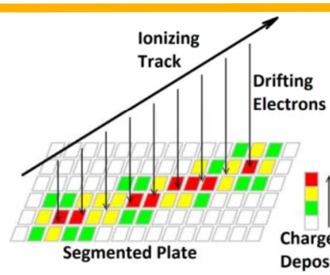
Requested accuracy is defined by the Accelerator-Driven Minor Actinides Burner (ADMAB)

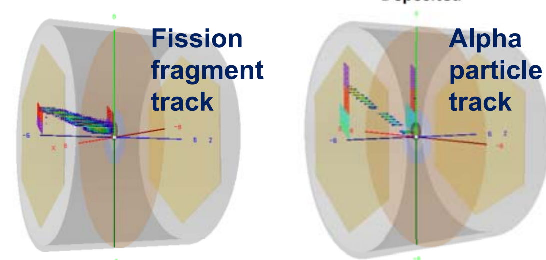
Systematic errors associated with conventional FICs

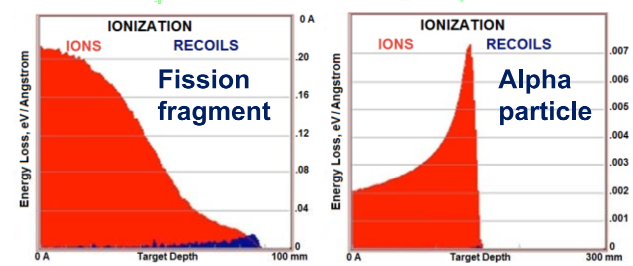
1. **Separation of fission and background events in pulse-height spectra:**
   - **Spallation and fragmentation inputs**
   - **Radioactivity of used targets**
   - **Electronic noise**
2. **Anisotropy of emitted fission fragments**
3. **Neutron flux normalization: mostly data were obtained from ^{235}U ratio, very few were normalized to (*n,p*)-scattering**
4. **Beam profile and sample uniformity**
5. **Charged particle contamination of neutron beam**
6. **…**

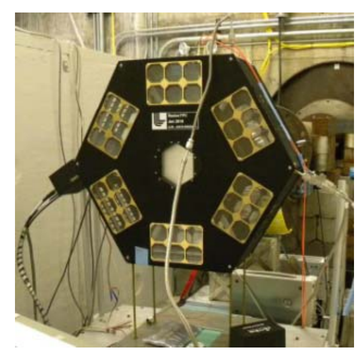
TPC can address most of the systematic uncertainties associated with conventional FICs

- **Time Projection Chamber (TPC) technology based on highly pixelated readout anodes**
- **Full 3D event reconstruction gives a "snapshot" of ionization tracks in a fill gas**
- **Particle identification based on specific ionization loss**
- **The TPC experiment, involving a collaboration of 4 national labs and 6 universities, is currently running at the fast neutron beam facility WNR at LANL**





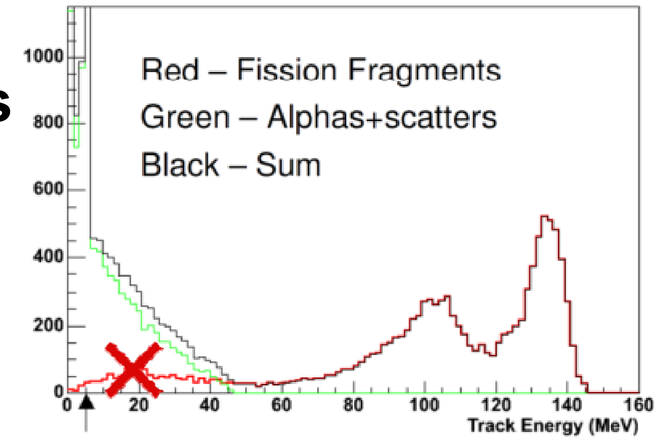




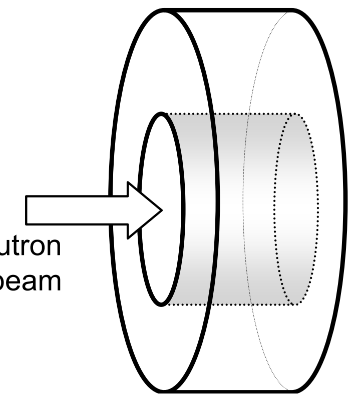
There are more details in following report of F.Tovesson

Other ways to avoid systematic uncertainties associated with conventional FICs

- **An ionization fission chamber with gaseous actinide target doesn't lose fission fragments emitted at steep angles**



- **Uranium hexafluoride UF_6 can be a suitable compound for investigating U isotopes as it has a boiling point at 56.5°C**
  - **There is no "edge-effect" due to two fragment emission**
  - **It is expected much better resolution between fission fragments and small PH signals in a pulse height spectrum**
  - **Separate active volume outside the main one should be used for the background neutron input estimation**



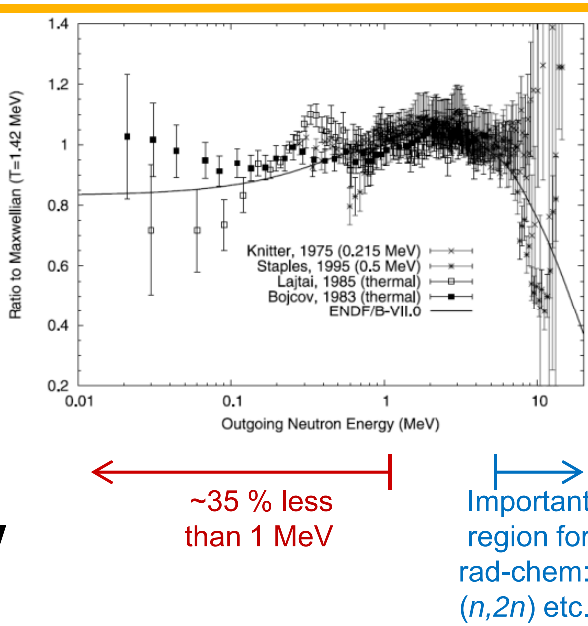
***Ref.**: Laptev, Strakovsky, Briscoe, Afanasev, Proposal NSF-ARI-MA (DNDO) Grant ECCS-1139985, 2011*

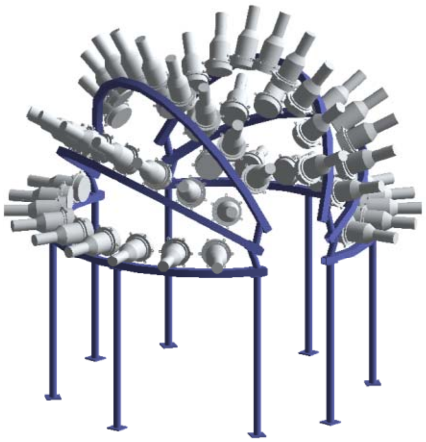
- **For the Pu isotopes, plutonium hexacarbonyl $Pu(CO)_6$ can be considered.**

***Ref.**: Thanks to W.Loveland, Oregon State University, for pointing out this opportunity*

Prompt Fission Neutron Study

- Existing data establish the prompt fission neutron spectrum quite well in the energy range from 1 to about 5 MeV
- The Chi-Nu experiment to study the prompt fission neutron spectrum will also run at the WNR
  - Parallel plate avalanche counter as a fission trigger
  - 20 ^{6}Li-glass detectors to measure neutron output below 1 MeV
  - 50 liquid scintillator detectors to measure neutron output from 0.5 to 12 MeV
- Chi-Nu goals are
  - Below 1 MeV, to reduce the neutron output uncertainties from ~10% to ~5%
  - Above 6 MeV, to reduce uncertainties from 20-50% to 10-20%

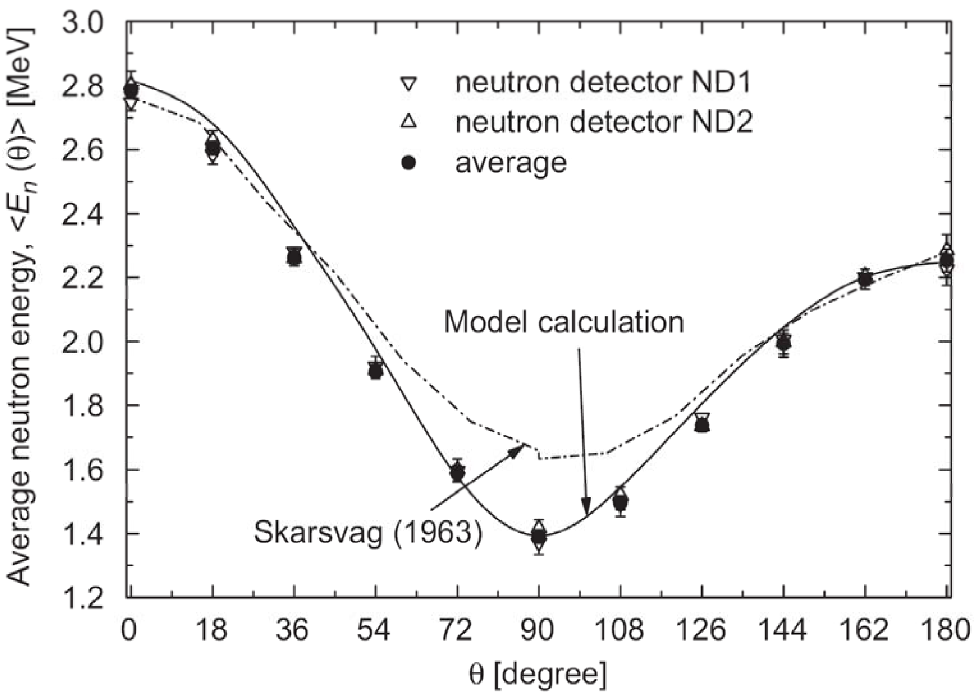




Ref.: *Noda, Haight et al., Phys.Rev.C 83, 034604 (2011)*

Angular distribution of PFN relative to FF

Recent measurement for thermal neutrons



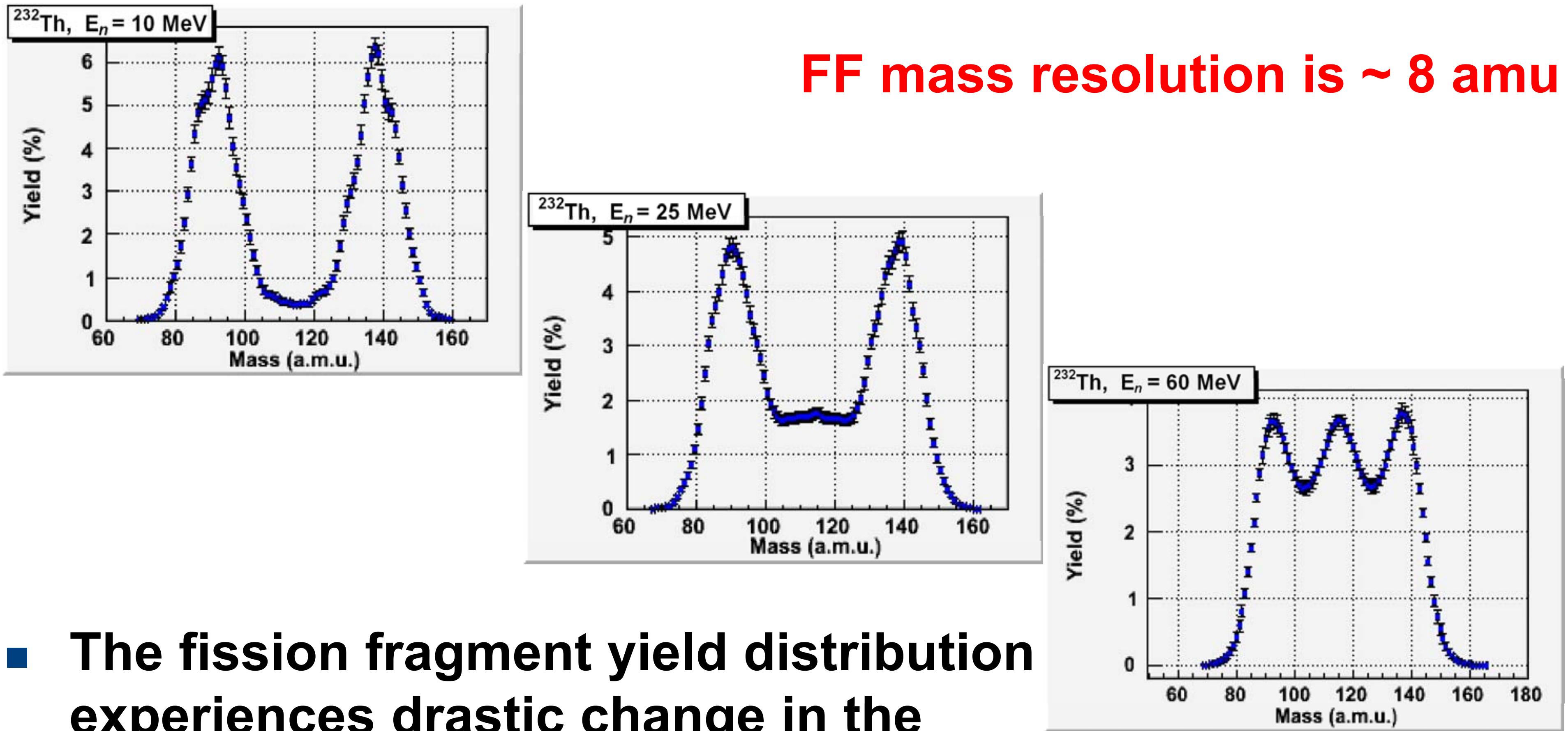
Ref.: *A.Vorobyev et al., NIM A 598 (2009) 795*

- Fission neutrons from the $^{235}U(n_{th},f)$ reaction were detected at several angles
- Angular resolution is 18°
- Model calculation was done on the basis of the assumption that neutrons are emitted only from fully accelerated fragments
- Obtained an estimate of upper limit for “scission” neutrons less than 5% of the total neutron output
- From all existing works: the contribution of scission neutrons to the total yield of PFN ranges from 1% to 20%, so even existence of “scission” neutrons can hardly be considered proven

Fission fragment yield distribution

- The fission fragment yield distribution changes drastically with neutron energy above ~10 MeV, from asymmetric to symmetric
- There are no data above neutron energy of about 20 MeV except for ^{238}U and ^{232}Th. No data for the most important nuclei ^{235}U and ^{239}Pu !
- These data are vitally important for both fission theory and many applications including fast reactors, ADS systems, and special nuclear devices
- This gives strong motivation for the new LANL experiment SPIDER, designed to obtain fission fragment yields with mass resolution up to 1 amu following fast neutron-induced fission

Existing data for fission fragment yield distribution as a function of neutron energy for ^{232}Th



FF mass resolution is ~ 8 amu

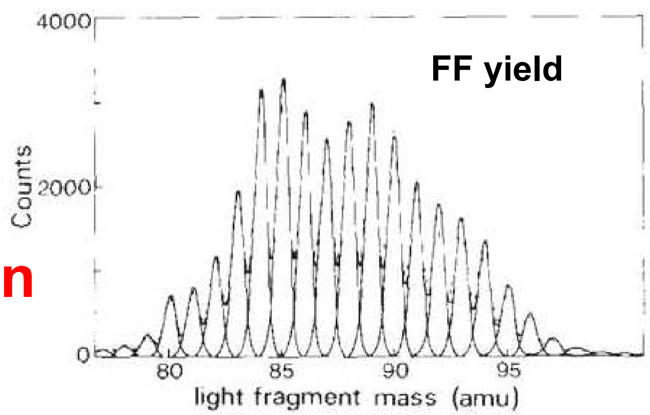
- **The fission fragment yield distribution experiences drastic change in the energy range from 10 to 60 MeV**

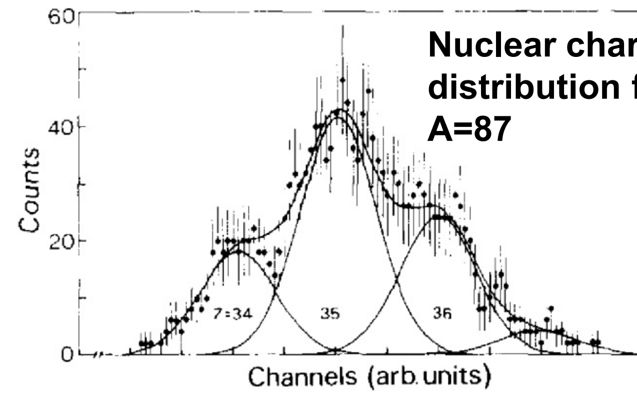
Ref.: *Simutkin et al., AIP Conf.Proc. 1175(2009)393*

High resolution fission fragment yield measurements

- **Measured at ILL, France for thermal neutron**

^{229}Th(n_{th},*f*) with COSI-FAN-TUTTE spectrometer



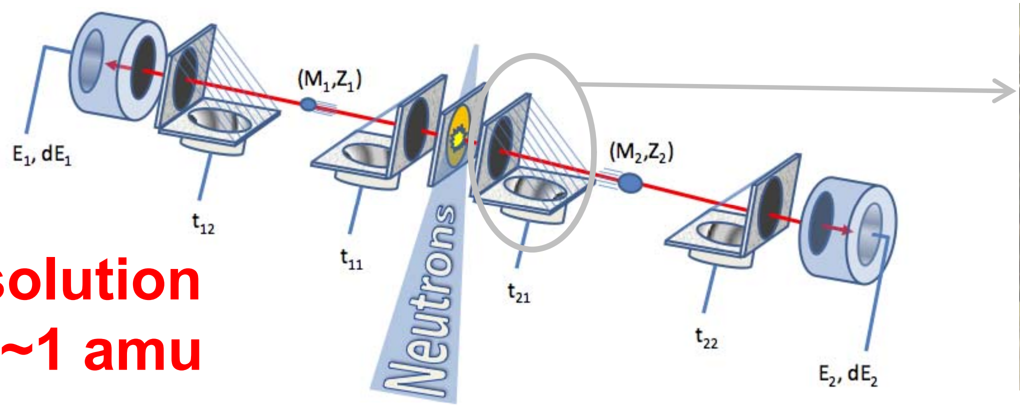


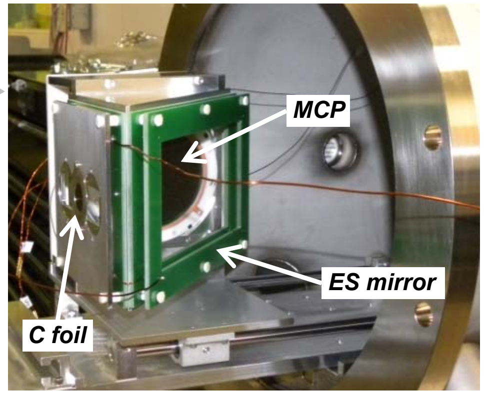
FF mass resolution is ~ 0.64 amu

***Ref.**: Boucheneb et al., Nuc.Phys.A 502(1989)261c*

- **SPIDER experiment to measure energy-dependent neutron-induced FF yields with high resolution in progress at LANL**

***Ref.**: White, Tovesson et al., Report LA-UR-11-01125*



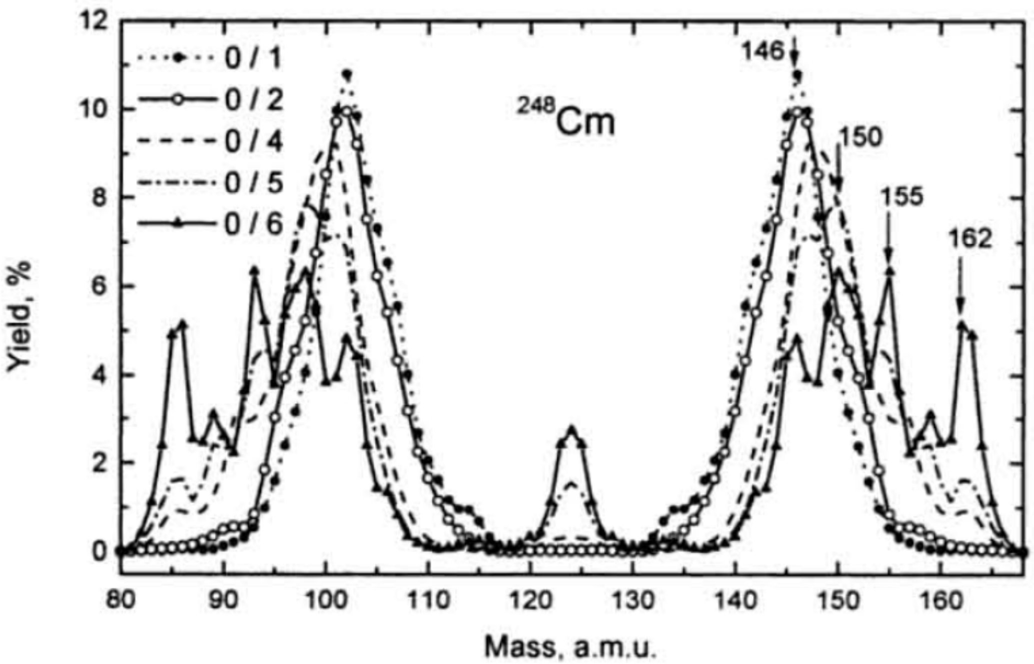


FF mass resolution is expected ~1 amu

There are more details about SPIDER in following report of F.Tovesson

Correlation between PFN multiplicity & FF properties

Search for correlation between PFN multiplicity & FF properties in SF of ^{252}Cf, ^{244}Cm, and ^{248}Cm was done using a 4π neutron detector and twin Frisch gridded FIC



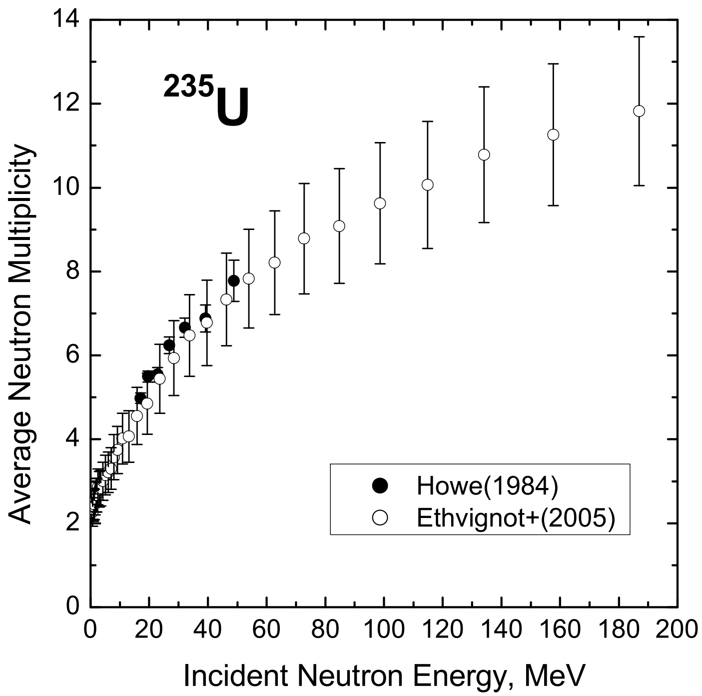
***Ref.**: V.Kalinin et al., Fission: Pont d'Oye V (2003)73*

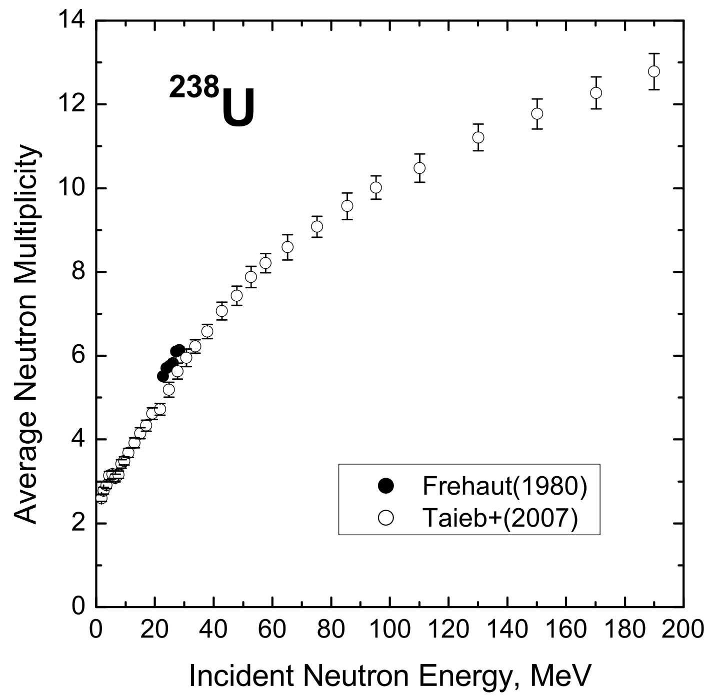
- Fission fragment mass distribution for fixed numbers of emitted neutrons ν_L/ν_H for spontaneous fission of ^{248}Cm
- There are no such data for fast neutron-induced fission. The very first attempt of such measurement was done for thermal and 0.3 eV neutron induced fission of ^{235}U and ^{239}Pu [*Batenkov et al., AIP Conf. Proc. 769 (2005)1003*]
- Fission theories are most sensitive for correlated data, source: LANL-LLNL fission workshop, Feb 3-6, 2009

FF mass resolution is ~ 3.5 amu (FWHM for ^{252}Cf)

Average fission neutron multiplicity

- Average fission neutron multiplicity as a function of incident neutron energy is known quite well for nuclei ^{235}U, ^{238}U, and ^{237}Np, up to 200 MeV





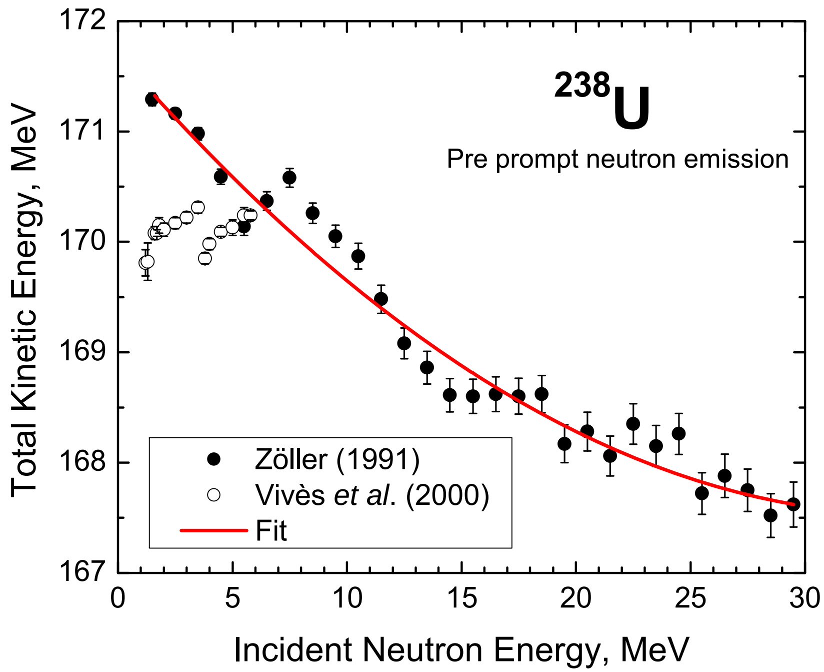
Ref. *(^{235}U):*
R.Howe,
Nucl.Sci.Eng. 86(1984)157
T.Ethvignot et al.,
PRL 94(2005)052701

Ref. *(^{238}U):*
J.Frehaut ,
EXFOR data 21685.003
J.Taieb et al.,
ND-2007 (2007)429

- For other nuclei important to the nuclear cycle the data are limited: for ^{232}Th < 50 MeV, ^{239}Pu < 30 MeV, ^{233}U < 15 MeV, $^{243,241}Am$ < 11 MeV, etc.

Total kinetic energy of fission fragments as a function of neutron energy

- Data on total kinetic energy of fission fragment are required for both theory and applications to calculate total energy release in fission (along with that for prompt and delay fission neutron, prompt and delay fission gamma-ray energy, etc.)

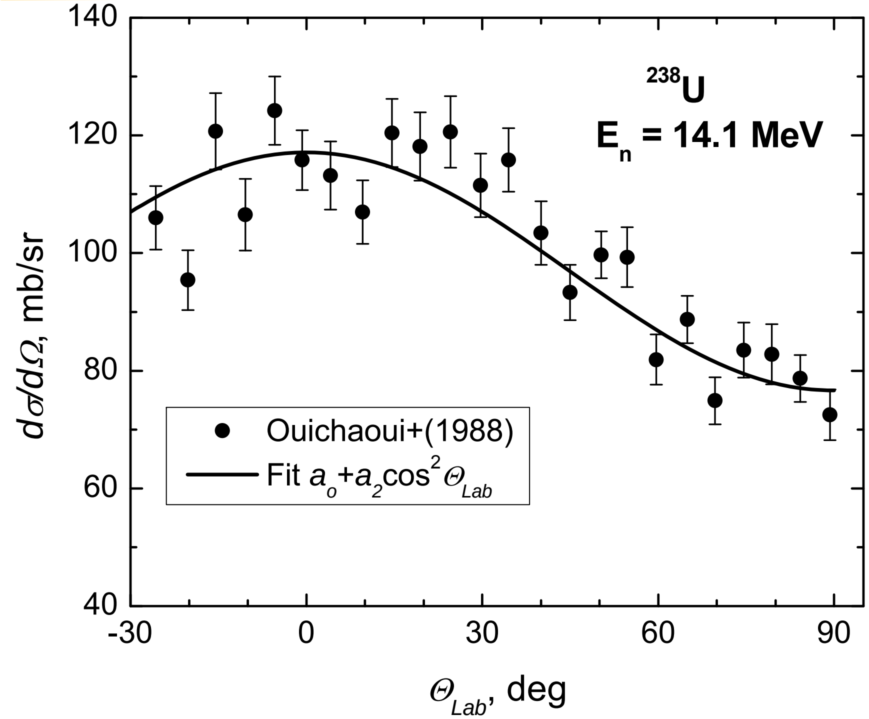


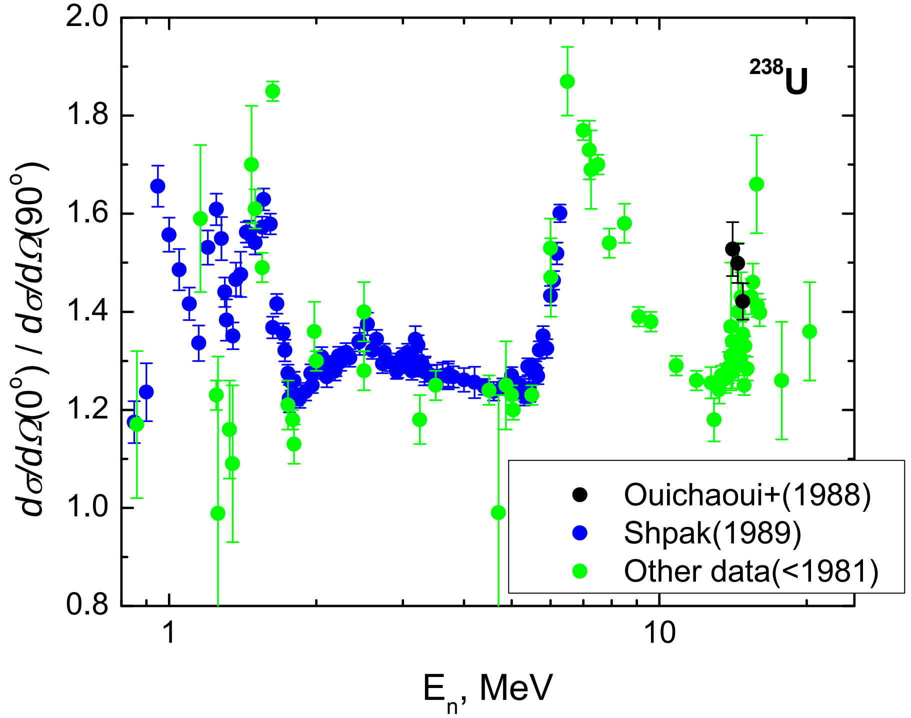
Ref.:
Data: C.Zöller, PhD Theses (1991)
F. Vivès et al., NP A 662(2000)63
Fit: D.Madland, NP A 772(2006)113

- There is noticeable disagreement among existing data below 5 MeV

- The quadratic fit (to Zöller data) doesn't describe structure near different chance fission thresholds because the corresponding experimental data for ^{235}U and ^{239}Pu are much lower in quality and the energy range is limited (there are no data above 15 MeV)

Angular anisotropy of fission fragments





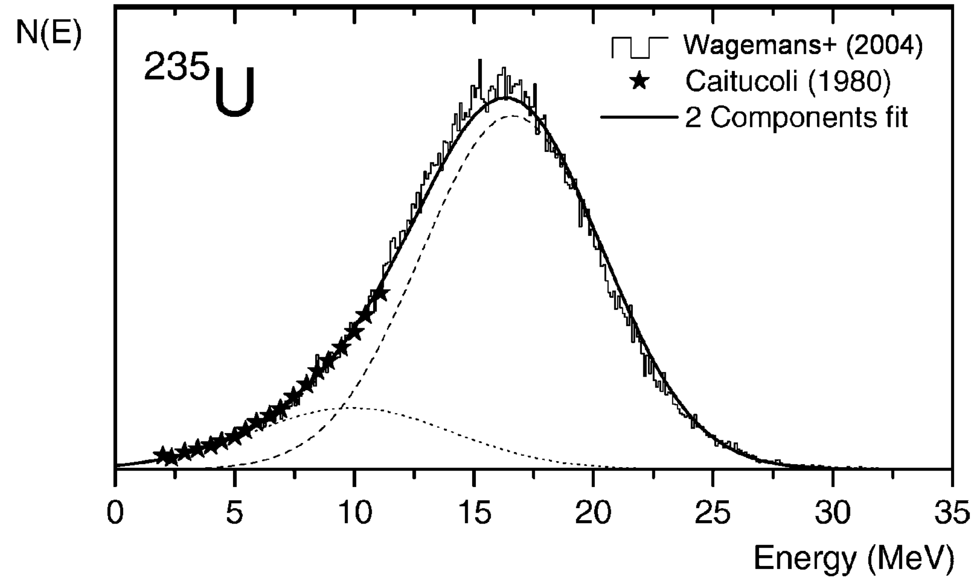
- It is used in fission cross section measurements to calculate a correction for FF absorption in a target
- Provide information about the projection of the total angular momentum J of the fissioning nucleus along the nuclear symmetry axis at saddle point deformation

Ref.:
Ouichaoui et al., Acta Phys.Hung. 64(1988)209
Shpak, Yadernaya Fizika 50(1989)922

Ternary fission yields

- **Important for both fission theory and applications (gas emission characteristics)**
- **Studied quite well at thermal energy:**
  - **^{235}U ratio binary/ternary fission was measured quite well: 536±10** [*C.Wagemans et al., PR C 33(1986)943*];
  - **energy distribution of ternary α's measured** [*C.Wagemans et al., NP A 742(2004)291*];
  - **etc.**
- **Confident separation of all ternary particles in SF ^{252}Cf** [*M.Mutterer et al., PR C 78(2008)064616*]

E-distribution for the ^{235}U(n_{th},*f*) ternary α's



Prospective investigations of ternary fission

- **Very little data for fast neutron-induced fission. Better understanding needed of:**
  - **Variation of ternary-to-binary fission ratios in the resonance energy region. E.g., a correlation of ternary fission yields and the relative contribution of standards I and II fission modes was found in ^{235}U(*n,f*) resonances at energy from thermal to 2500 eV**

    [*S.Pomme et al., NP A 587(1995)1*]
  - **Angular anisotropy in ternary nuclear fission and its dependence on neutron energy. Most fission theories predict ternary particles to be formed at scission. A way to confirm this assumption is to analyze the angular distribution of fission fragments**

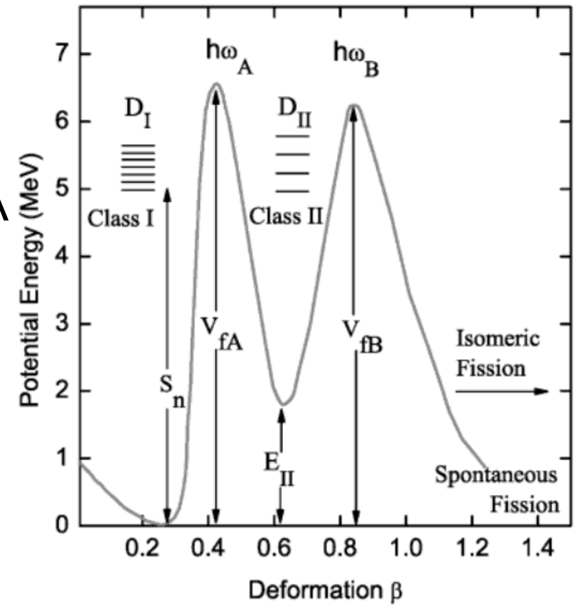
    [*S.Dilger, PhD Theses(2004)*]
  - **"Quaternary" fission with an apparently independent emission of two charged particles?**

    [*M.Mutterer, Pont d'Oye V (2003)135*]

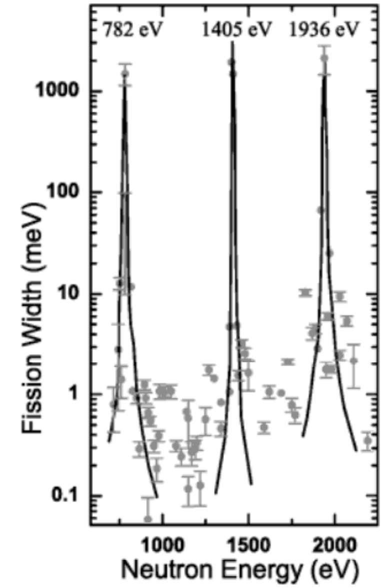
Subthreshold fission as a search for class-II states

- Gives unique information about fission barriers
- Direct demonstration of double-humped structure of potential energy of heavy nuclei and existence of class-II states
- First systematic study of class-II clusters with high energy resolution was done at the ORELA facility, ORNL for ^{240}Pu

The double-humped fission barrier

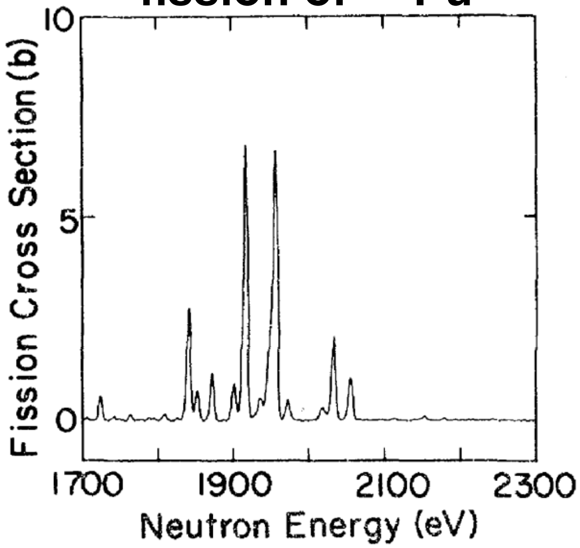


Clustering of fission strengths



***Ref.**: Mughabghab, Atlas of Neutron Resonances, 2006*

1936 eV cluster in subthreshold fission of ^{240}Pu

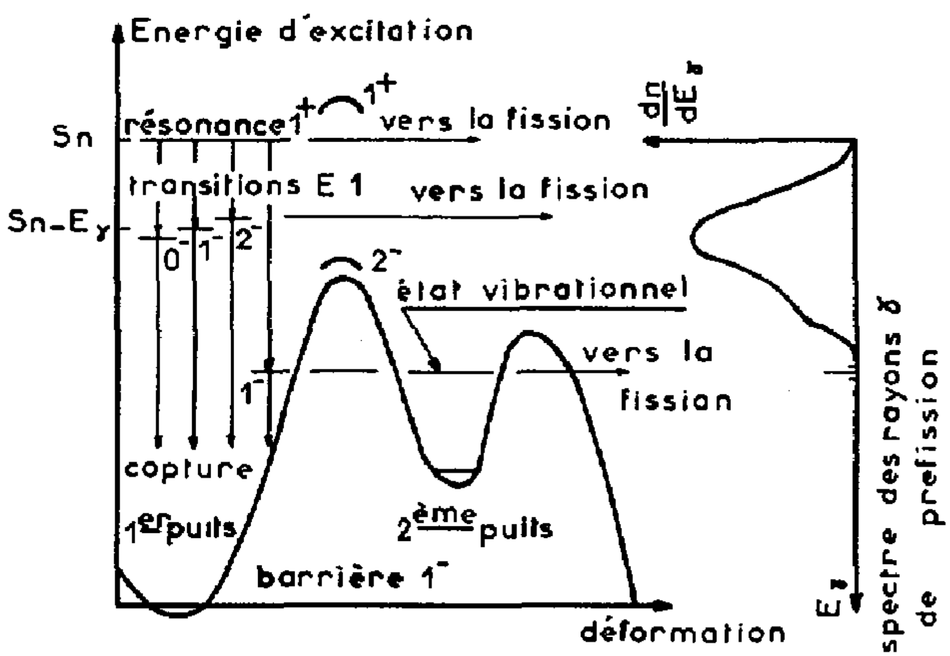


***Ref.**: Auchampaugh, Weston, PRC 12, 1850 (1975)*

- Clustering of subthreshold fission strengths observed in ^{240}Pu was explained in terms of the double-humped fission theory
- Average level spacing of class-II states estimated of D_{II} = 450±50 eV. Based on that and known D_I, a value of E_{II} = 1.52±.08 MeV was derived

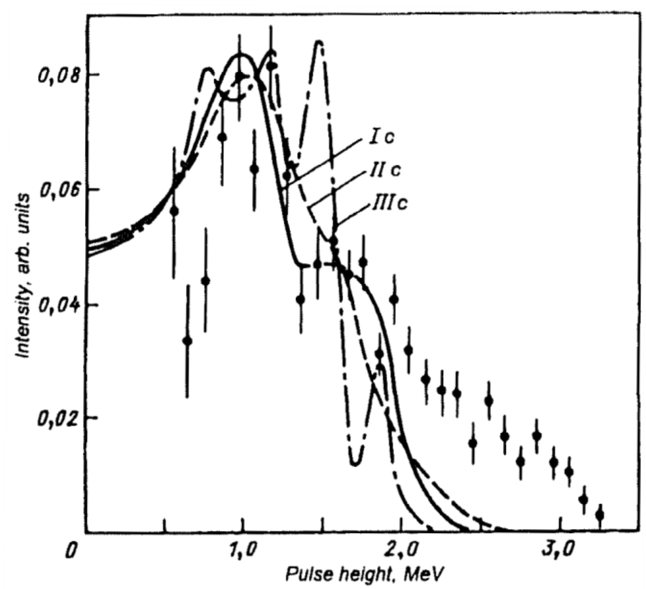
Search for γ-transitions to class-II states

Descriptive schematic of the (n, γf) reaction mechanism



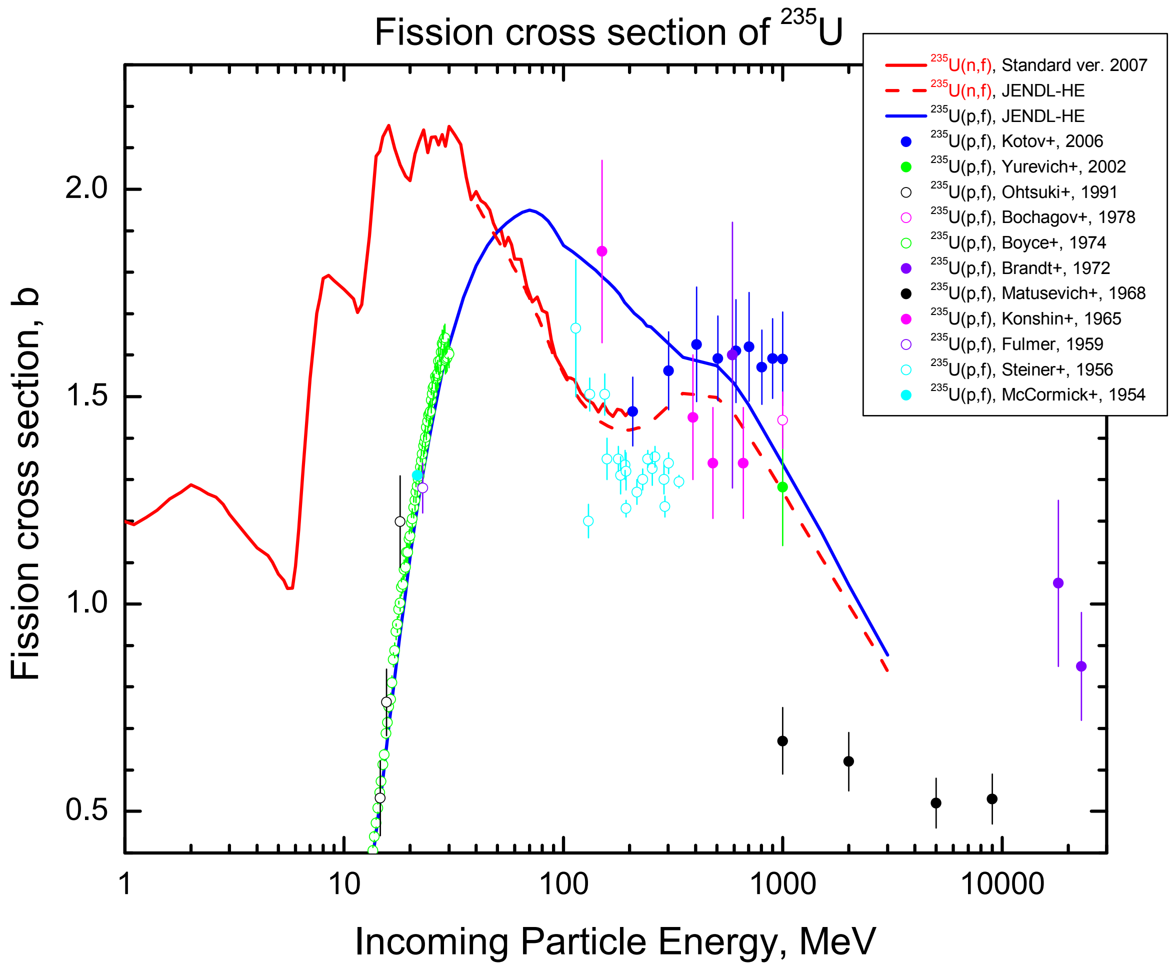
Ref.: J.Trochon, Physics & Chemistry of Fission, 1979, Vol. I, p.87

- Differences in pulse-height γ-ray spectra for weak (Γ_f < 10 meV) and strong (Γ_f > 10 meV) fission 1^+-resonances of ^{239}Pu in epithermal neutron energies
- Few possible structures could be interpreted as a γ-transitions between class-II states
- Confident discovery of class-II γ-transitions will give unique information about the structure of the fission barrier



Ref.: O.Shcherbakov et al., ASAP Proc. (2002) 123

Proton-induced fission of ^{235}U



- **This energy range is important for future ADC systems, e.g., for transmutation of nuclear waste**
- **Existing data are very scarce and are in contradictory**
- **The ALICE code calculation for the JENDL-HE data file shows a peak near 300 MeV, which comes from a broad reaction cross section shape from the optical model [***Private communication from T.Kawano, 2009***]. New experimental data are needed to confirm that result**
- **Relevant proton energy range should be available for the JLab EIC booster**

Single-event effects (SEEs) in electronics investigations

- Increasing complexity and density of an electronic chips increases their susceptibility to cosmic radiation. Most SEEs in avionic electronics at aircraft cruising altitudes are caused by neutrons
- Electronics industry is very interested in checking the stability of their electronics against neutron irradiation
- Spallation neutron sources can imitate cosmic neutron flux but with intensities millions of times higher
- Many facilities all around the world built irradiation facilities, which are in high demand

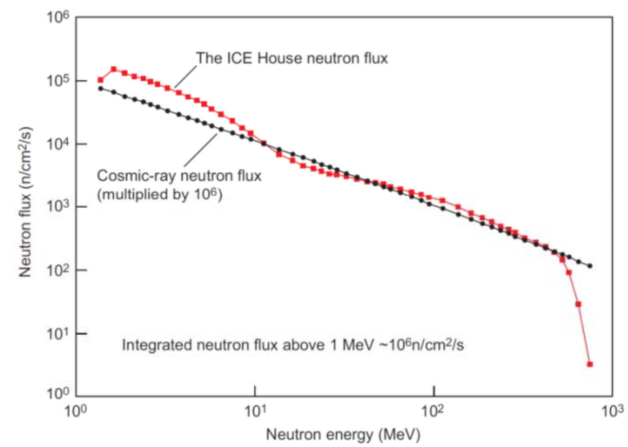
Active facilities in SEEs studies

Facility	Affiliation	Energy, MeV*)	Neutron source	Ref.
ICE House	LANL, USA	800(p)	White spectrum	J.Korean Phys.Soc. 59 (2011) 1558
TRIUMF Neutron Facility	TRIUMF, Canada	70-500(p)	White spectrum	IEEE Radiation Effects Data Workshop, 2003, p.149
ANITA	The Svedberg Lab., Uppsala Univ., Sweden	180(p)	White spectrum	IEEE Radiation Effects Data Workshop, 2009, p.166
VESUVIO	Rutherford Appleton Laboratory, UK	800(p)	White spectrum	Appl. Phys. Lett. 92, 114101 (2008)
GNEIS	PNPI, Russia	1,000(p)	White spectrum	Instrum. Exper. Tech. 53 (2010) 469
RCNP	Research Center for Nuclear Physics, Osaka Univ., Japan	14-198(n)	Quasi-monoenergetic	42nd Annual IEEE Int. Reliability Physics Symp., 2004, p.305
…				

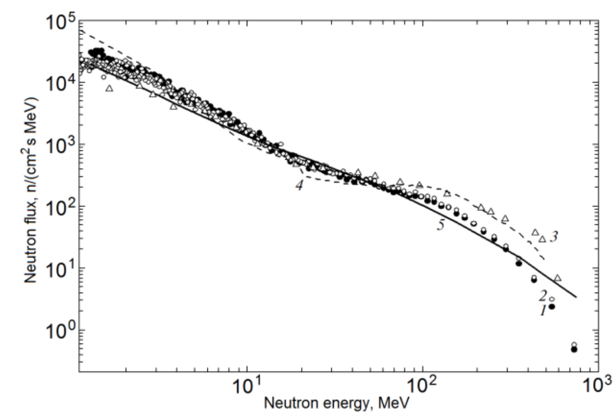
*) (p) indicates proton beam energy on spallation target; (n) energy of neutron beam is shown.

- **With the EIC booster proton energy of 3,000 MeV it may be possible to obtain better imitation of cosmic-ray neutron spectrum to higher energies**

Neutron flux of the ICE House irradiation facility at LANL



Neutron flux of the irradiation facility at GNEIS



Summary

- **In many years of fission research, a large volume of experimental data was accumulated**
- **There is still a significant lack of fission data for fast neutrons. Improving the quality of existing data will make a crucial impact on important applications and nuclear theory**
- **The proposed EIC complex at JLab promises to be a facility for acquiring high-quality experimental fission data and to carry out applied research**