Nuclear Data Experiments at LANSCE: Highlights 2008

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Cross Section Evaluation Working Group Meeting
US Nuclear Data Program Meeting
Brookhaven National Laboratory
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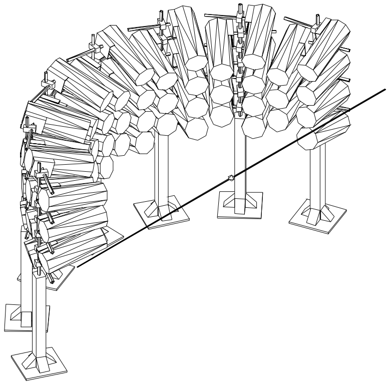
LA-UR-08-06961

Nuclear data measurements at LANSCE are made with several instruments

GEANIE (n,xγ)

FIGARO (n,xn+γ)



DANCE (n,γ)

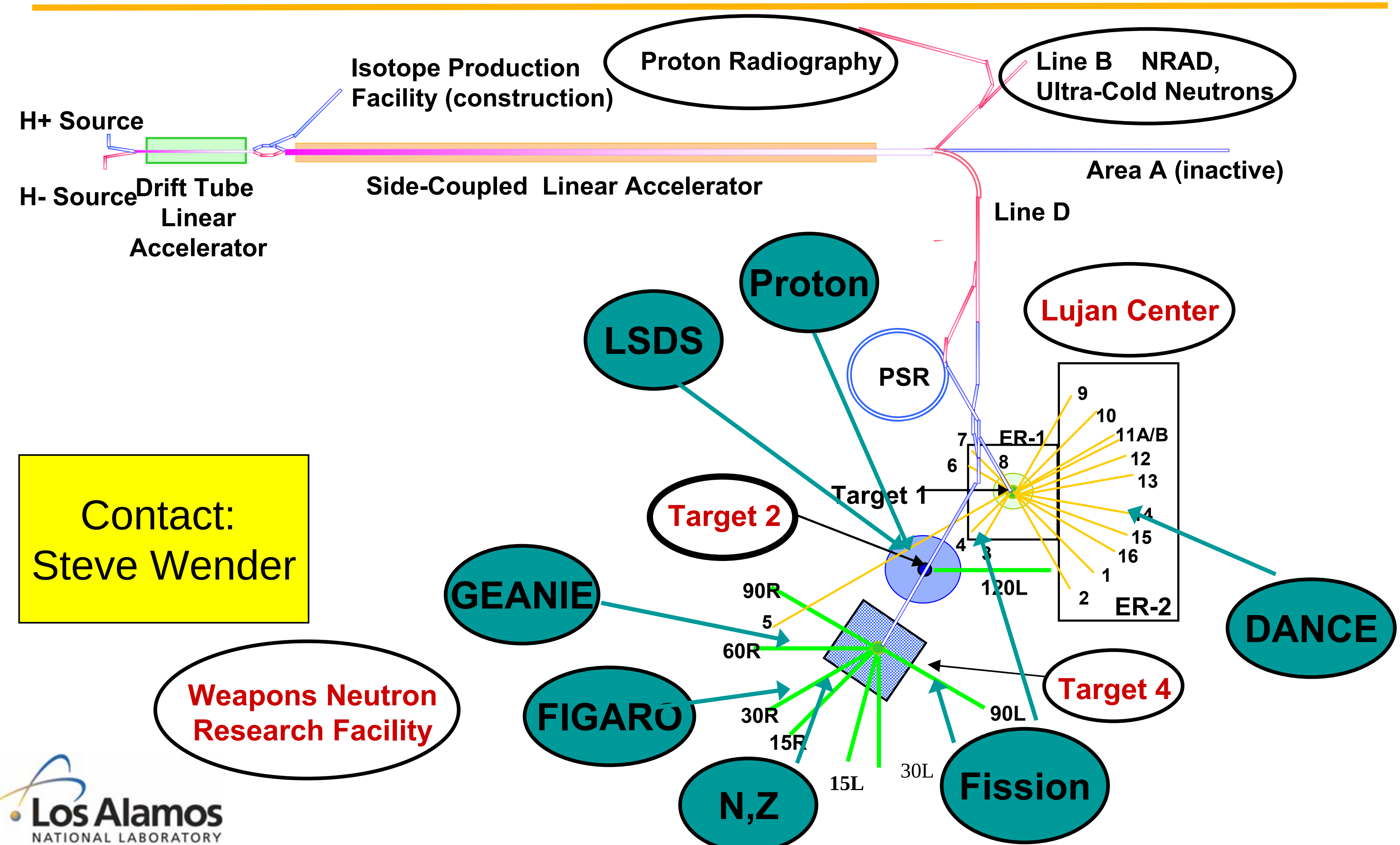
N,Z (n,charged particle)

LSDS

Fission

Double Frisch-grid fission chamber; also standard fission ion chamber; new detector station for fission and (n,alpha)

Nuclear data experiments at LANSCE use neutrons at the Lujan Center, Target 2 and Target 4



Contact:
Steve Wender

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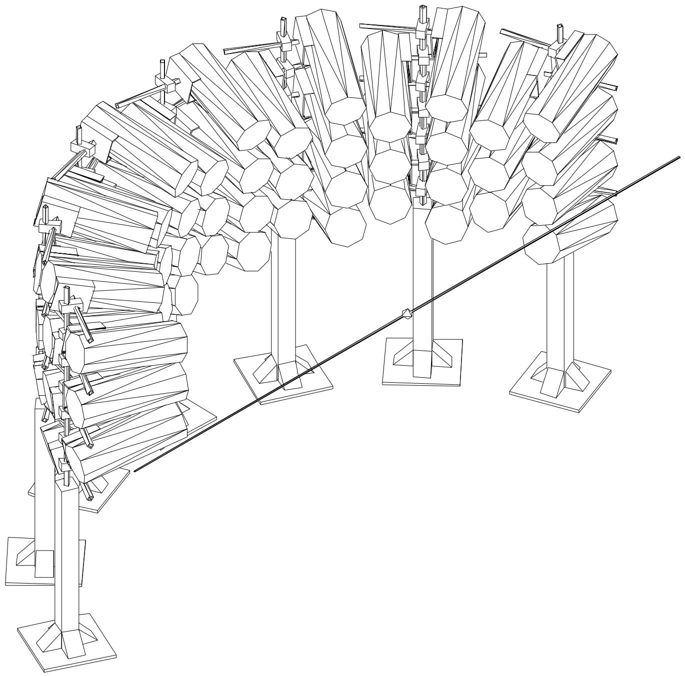
FIGARO (n,xn+γ)



Contact:
Bob Haight
Ron Nelson
Matt Devlin

Present and future experiments at FIGARO/WNR: neutron-emission spectra and ν-bar in fission

Fissionable isotopes: CEA fission chambers in beam

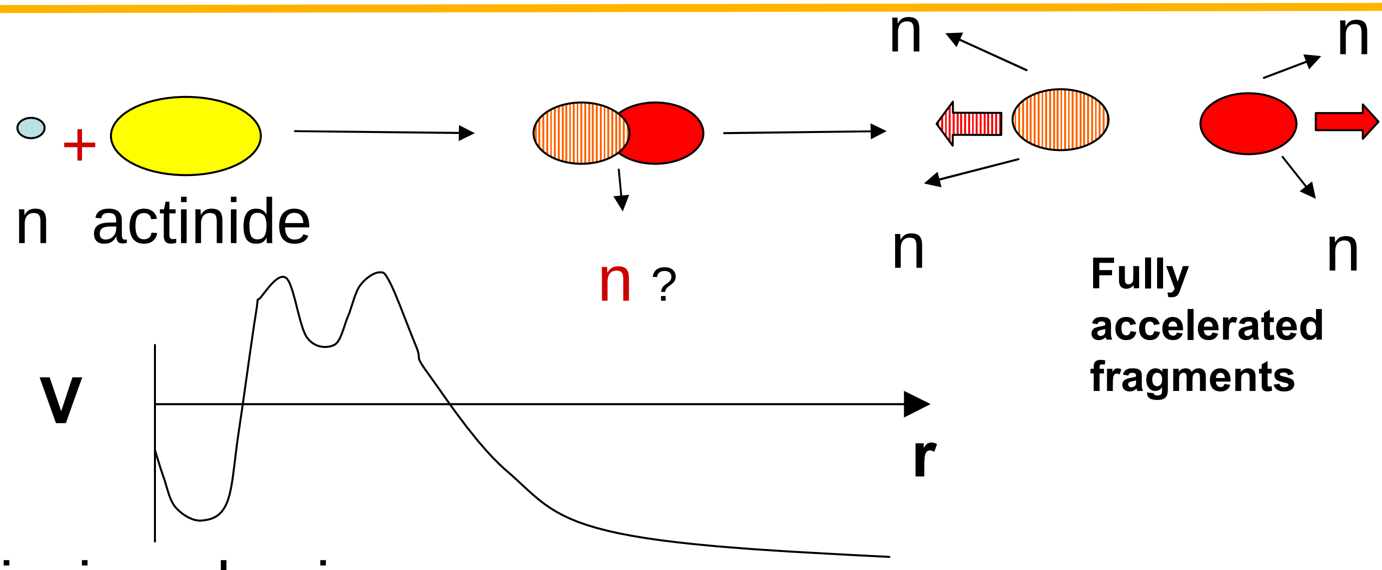
- ^{235}U, ^{239}Pu(n,f) – fission neutron spectra (1-8 MeV)
  - Analyzed for incident neutron energies of 1 – 50 MeV
    LA-UR-08-2585
- New measurement for ^{239}Pu(n,f) – data taken recently
  - Better fission chamber, less background

Other materials: Gamma-ray trigger (HPGe, BaF_2, etc.)

- ^{56}Fe, $^{all\text{-}A}$Mo --In progress

$$1\ \text{MeV} < E_n < 200\ \text{MeV}$$

Fission physics might be illuminated by the measurement of neutron spectrum from fission

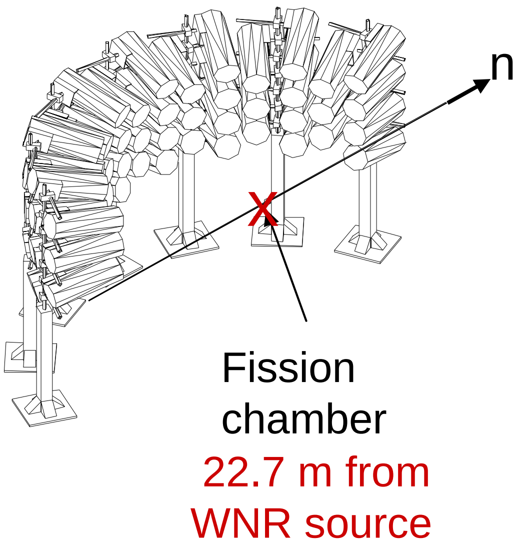


Fission physics

- “Pre-scission” neutrons
- Temperatures (excitation energies) of fragments
- Spectrum changes with energy of incident neutron:
  - Temperature of fragments, pre-equilibrium neutron emission prior to fission, angular distribution effects

FIGARO array of fast neutron detectors

FIGARO (n,xn+γ)

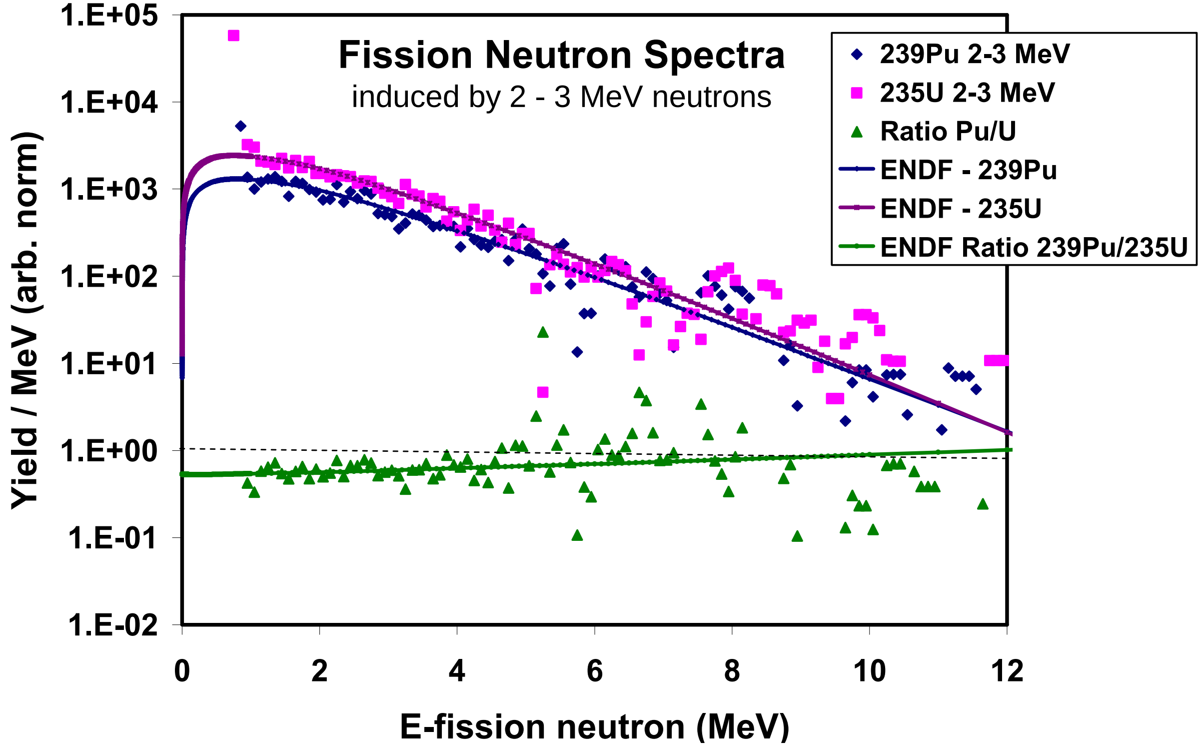


- **20 liquid scintillator neutron detectors**
- **2 gamma-ray detectors**

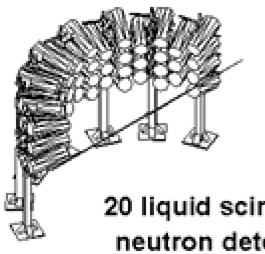
Double time-of-flight experiment

Here are some results -- shapes of fission neutron spectra, arbitrarily normalized

Incident E_n = 2 to 3 MeV



FIGARO (n,xn+γ)

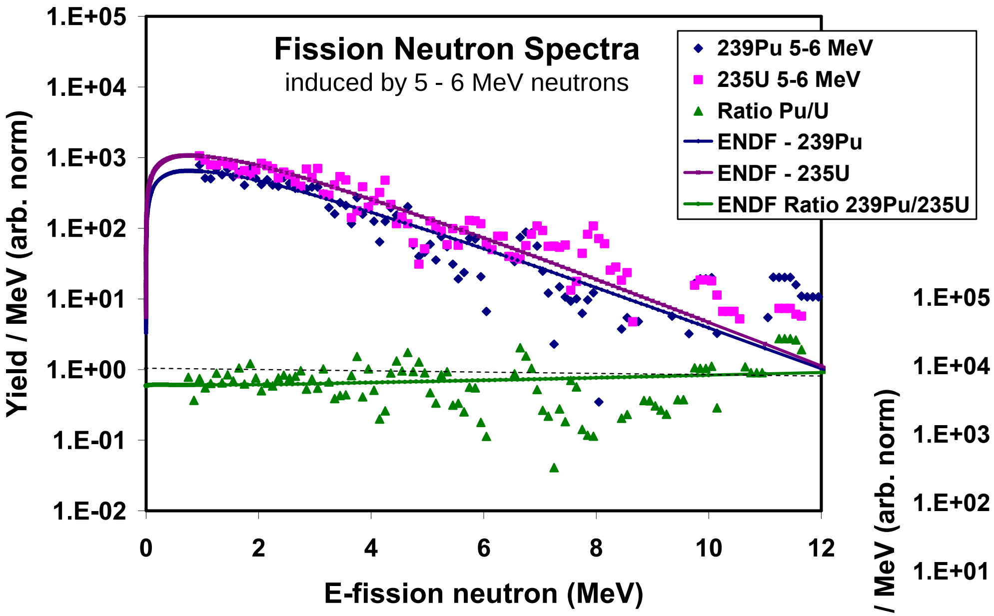


20 liquid scintillator neutron detectors

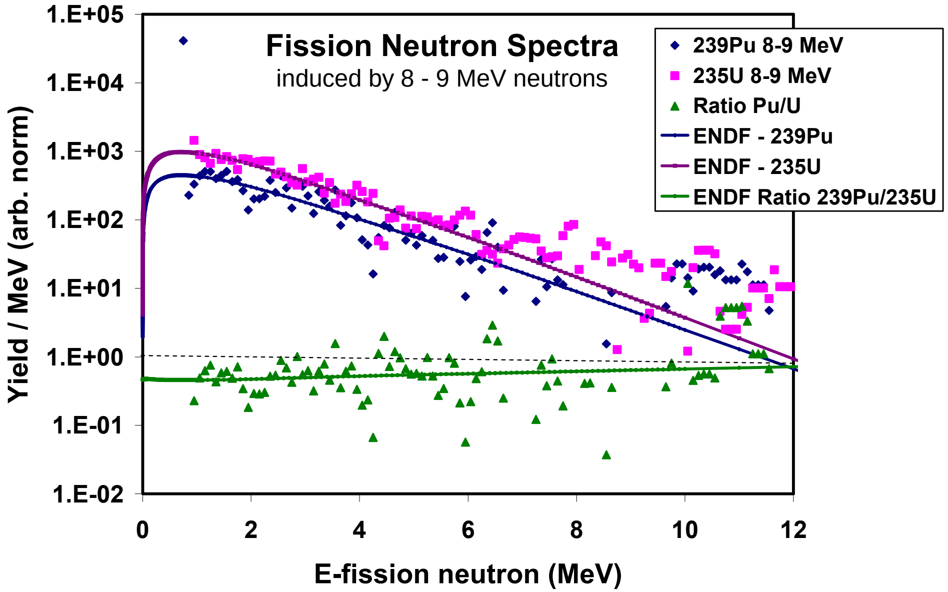
Data are averaged over all neutron detectors – 45 to 135 degrees

Other incident neutron energies show trends

Incident E_n = 5 to 6 MeV

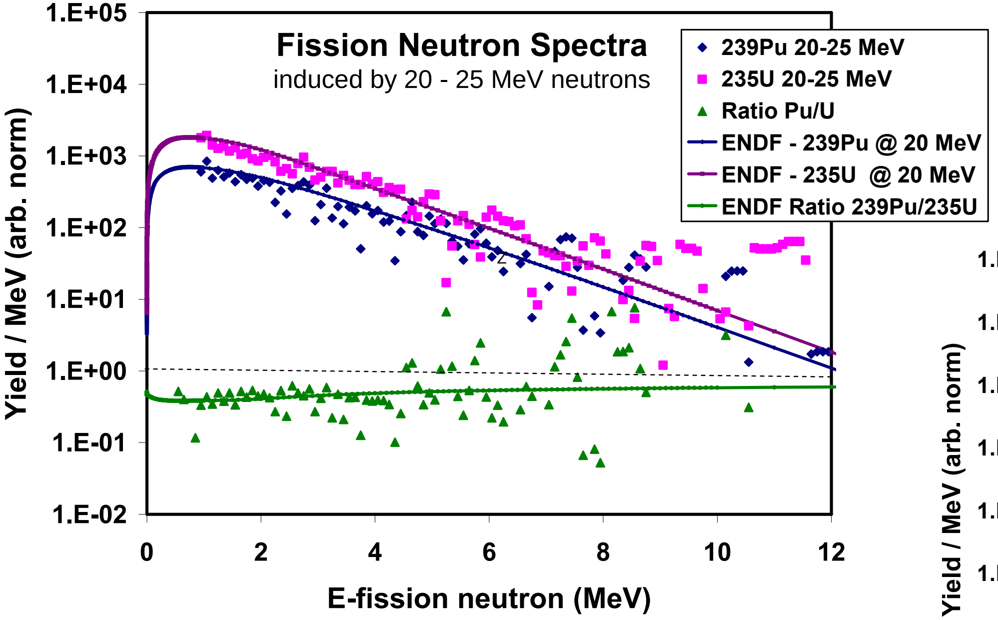


Incident E_n = 8 to 9 MeV

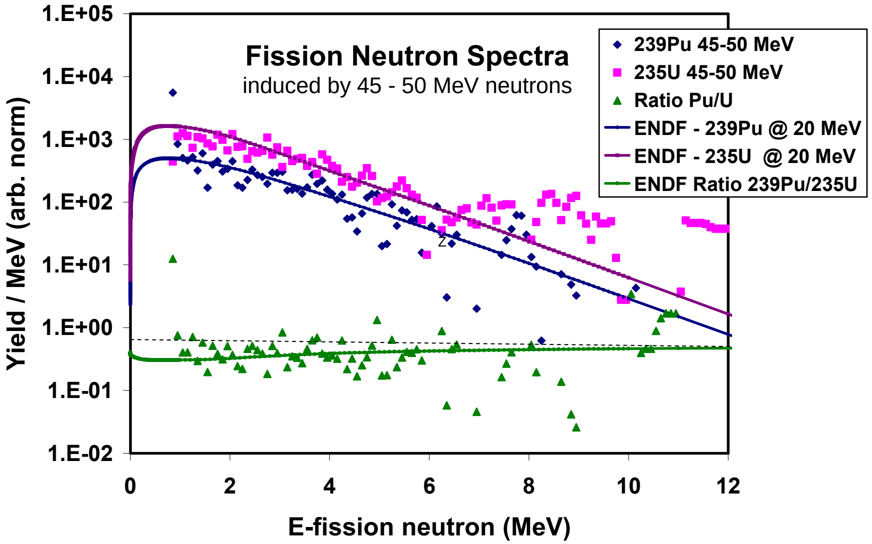


Pre-equilibrium emission becomes obvious at higher incident neutron energies

Incident E_n = 20 to 25 MeV



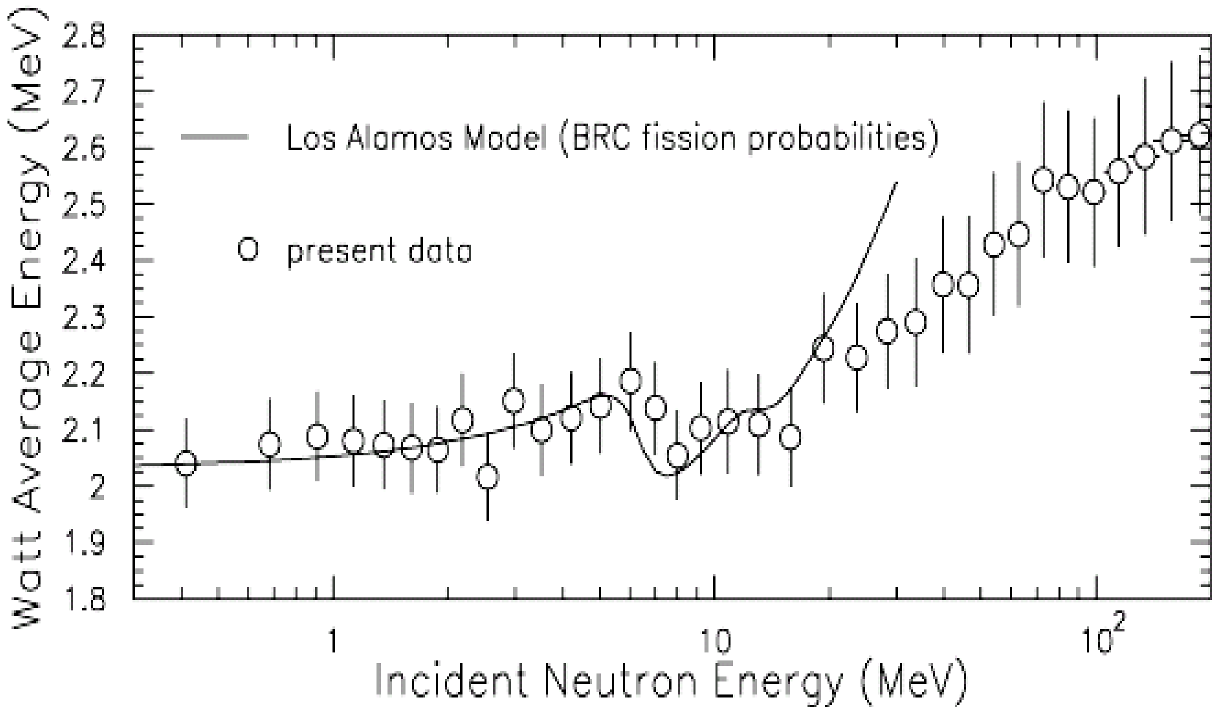
Incident E_n = 45 to 50 MeV



Previously we reported average fission neutron energies for ^{235}U (n,f) and ^{238}U (n,f)

^{235}U

$\langle E_{fn} \rangle$

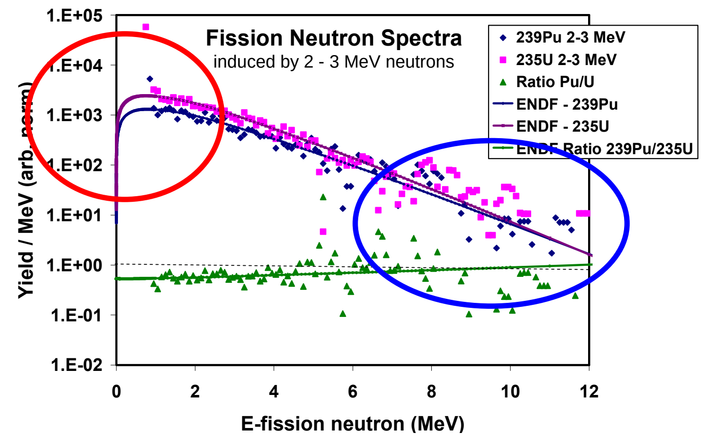


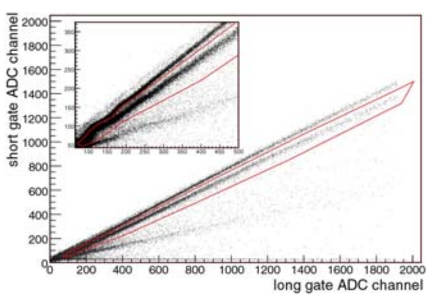
E_{in}

Ref: T. Ethvignot et al., Phys. Rev. Lett. 92, 052701 (2005)

Program of fission neutron output measurements continues

- Reduce background from accidental coincidences
  - Came from neutron scattering on backing foils – 0.12 mm Pt
  - Presently we are using a much better chamber
- Measure fission neutrons below 1 MeV
  - Need better n-gamma discrimination
- Measure fission neutrons better above 8 MeV
  - Better timing on fission chamber (**LLNL-LANL** collaboration)
  - More efficient neutron detectors (larger solid angle for detection)
- Quantify uncertainties better → **covariances**
- More isotopes – ^{235}U, ^{239}Pu, ^{238}U, ^{237}Np, $^{240-244}$Pu, etc.





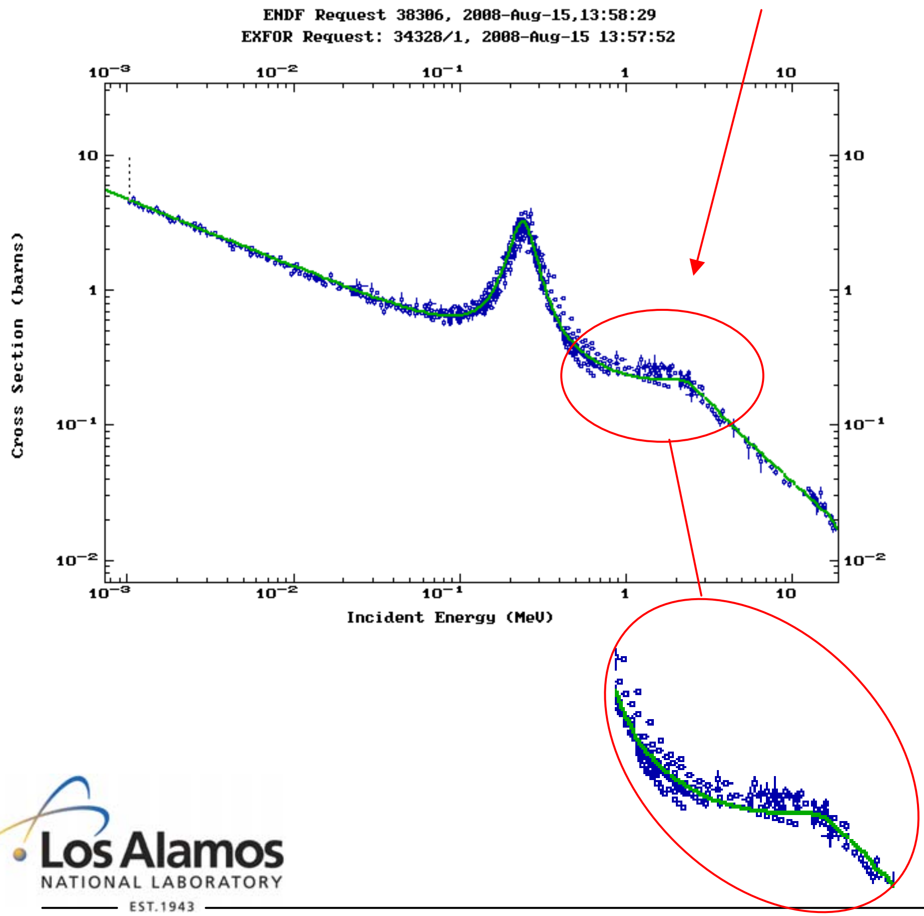
N,Z Reactions

$Z = p, d, t, {}^{3}He, \alpha$

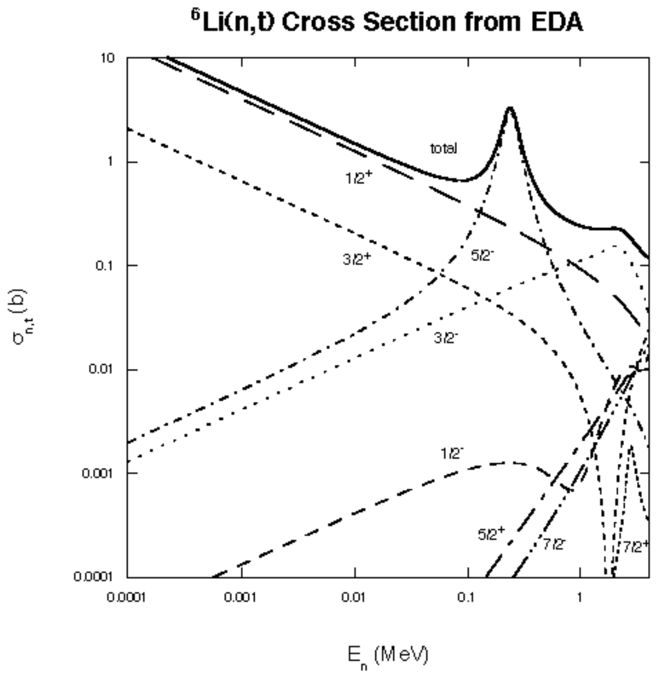
Contacts:
Matt Devlin
Terry Taddeucci
Bob Haight

The ^{6}Li(n,α)t cross section is a standard at low neutron energies (thermal to >10keV), but at MeV energies…

…the experimental data are somewhat discrepant: 20% variation from 1-3 MeV



Interferences between the resonances at 2 MeV leave the R-matrix fit unconstrained



From Gerald M. Hale and Hartmut M. Hofmann, Nuclear Data 2004, AIP Conf. Proc. 769, 2005, 75.

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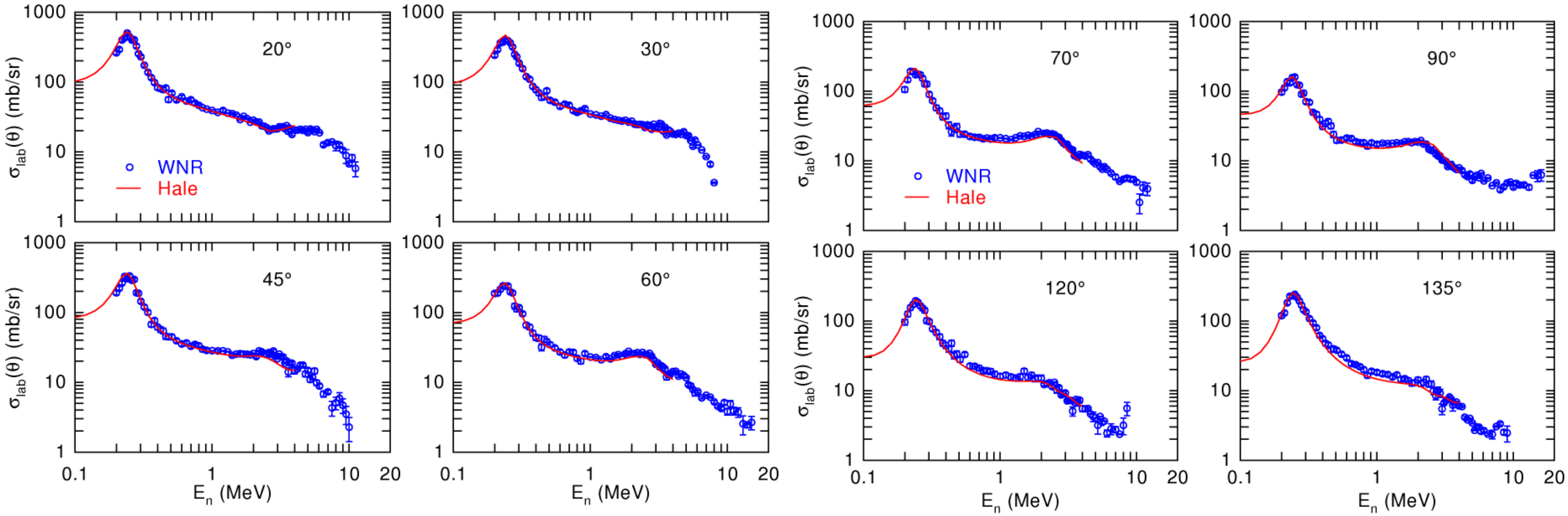
Differential cross section measurements

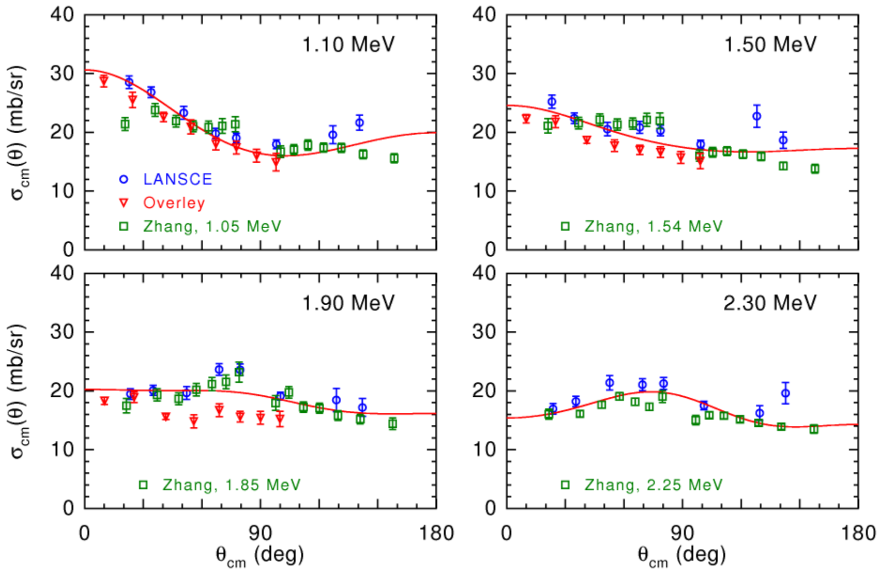
- Uses the N,Z (“Gas production”) apparatus at LANSCE FP30R-A
- Incident neutron energies from Time-of-flight (TOF)
- Beam flux monitored with a ^{235}U fission foil: resulting cross sections are relative to the ^{235}U fission cross section
- Targets were ^{6}LiF (216 μg/cm^2 of Li) on a thin mylar backing (0.84 mg/cm^2), 2” diameter
- Triton data from 2006 and 2007 at eight laboratory angles: 20, 30, 45, 60, 75, 90, 120, and 135 degrees
  - Covering the incident neutron energy for tritons from 0.18 to >10 MeV
- α particle data at five angles (20-75 degrees)
  - Covering neutron energies between 1 and 20 MeV

$^{6}Li(n,t)\alpha$ excitation function measurements

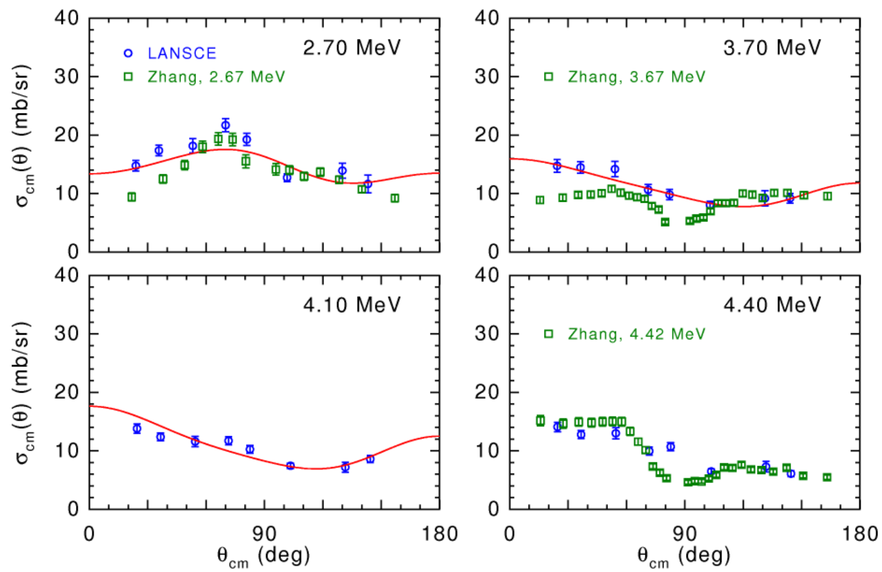
Some examples of results: data and R-matrix analysis for neutron energies from 200 keV – 10 MeV



$^6Li(n,t)\alpha$ triton angular distribution measurements – comparison with other results

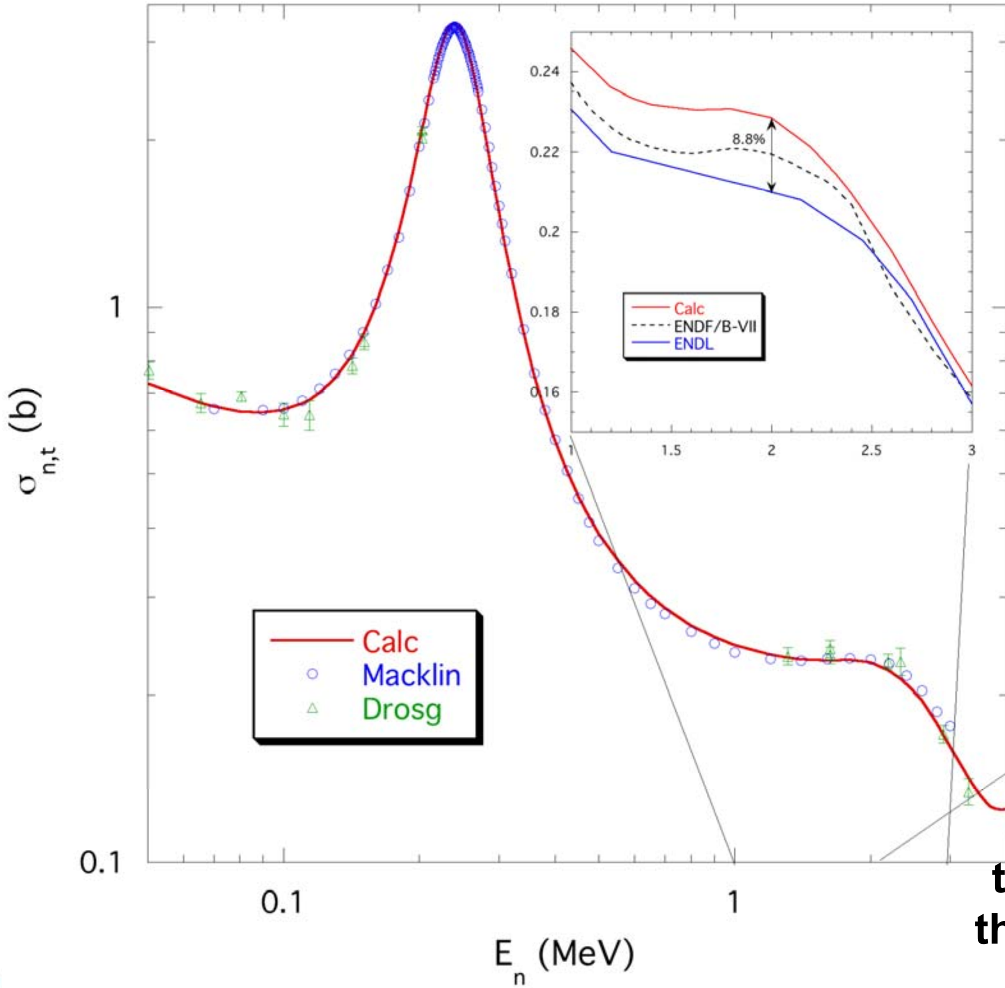


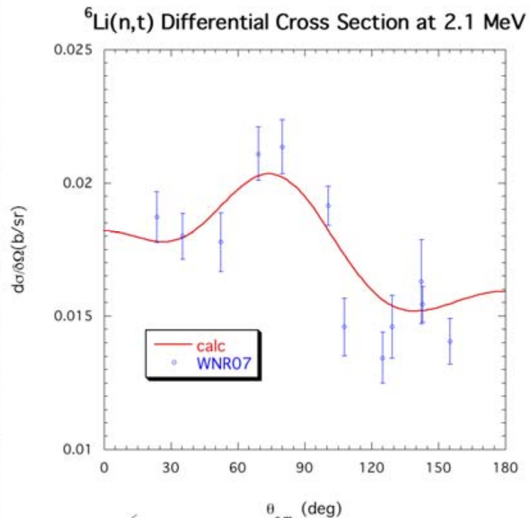
- The Zhang, et al. data appear to have systematic problems at low angles and near 90 degrees – these are probably known issues with the gridded ion chamber technique



Our data indicate that the effects of the resonances near 2 MeV are stronger than the prior evaluation suggested.

^{6}Li(n,α)t reaction cross section: implications of the new LANSCE data





The resulting reaction cross section, after including the new data in the R-matrix fit, is higher than the ENDF/B-VII evaluation in the 2 MeV region, and 8.8% higher than the prior ENDL evaluation.

GEANIE (n,xγ)

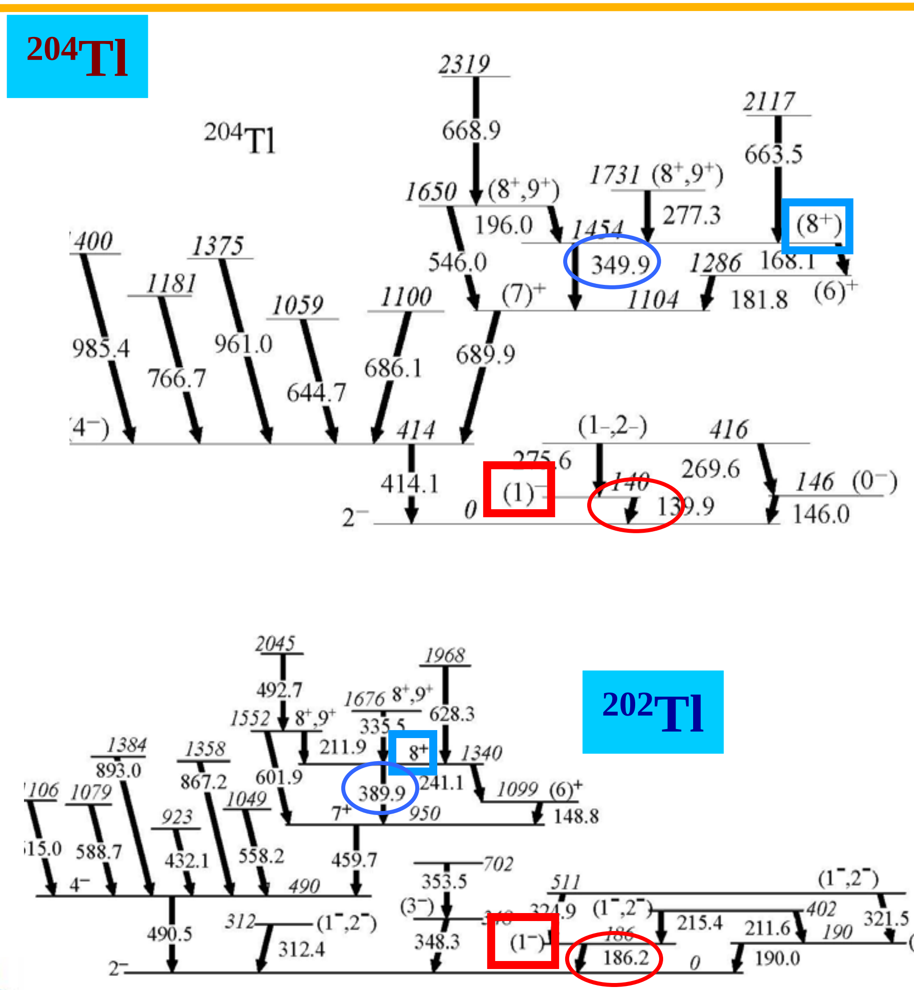
Contact:
Ron Nelson
Nik Fotiades
Matt Devlin

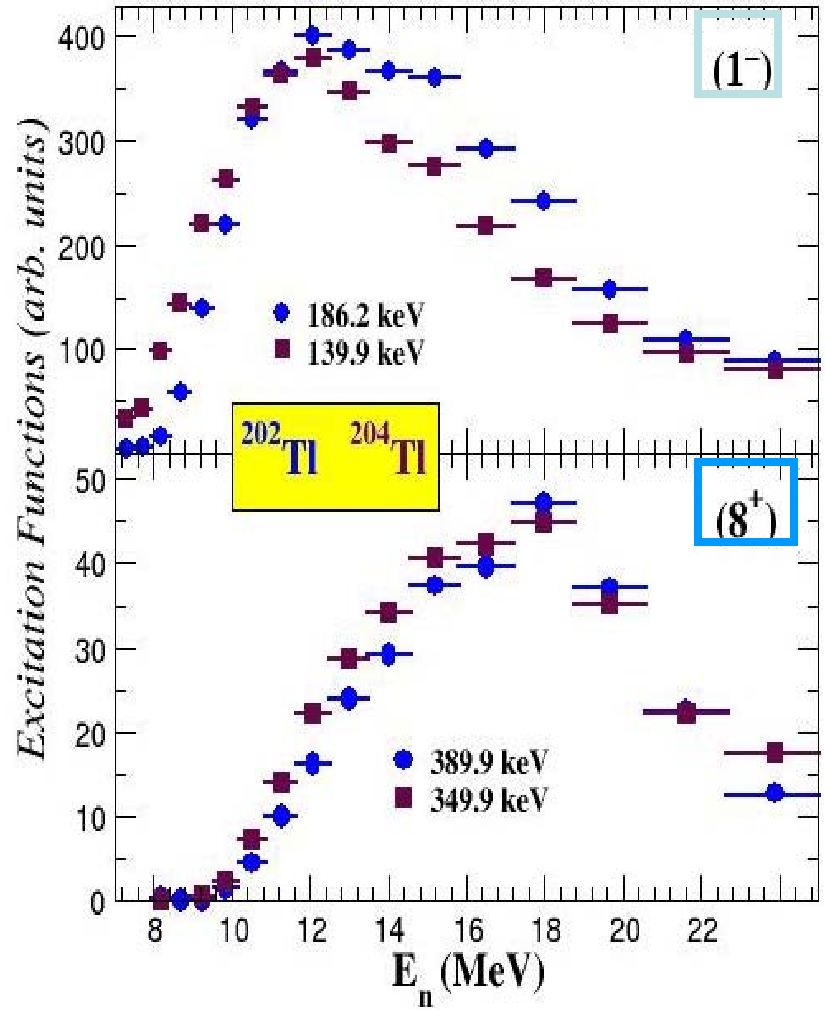
Recent Neutron-Induced Gamma-Ray Measurements with GEANIE at LANSCE/WNR

$1 \text{ MeV} < E_n < 200 \text{ MeV}$

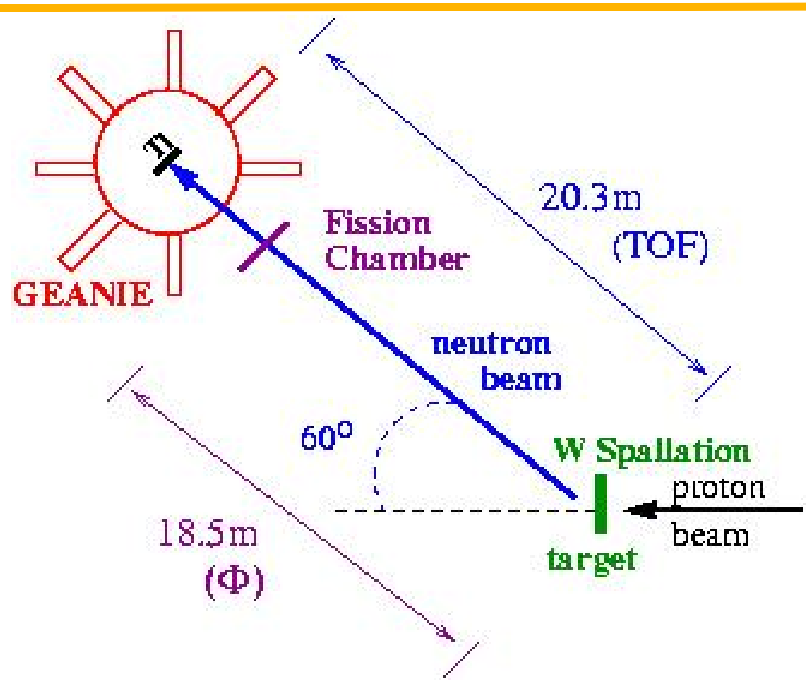
- ^{203,205}Tl(n,2nγ) – N. Fotiades, levels, isomer lifetimes - Phys. Rev. C 76, 0143092 (2007) and submitted to Phys. Rev. C.
- Isomer production in ^{191,193}Ir and ^{197}Au – data analysis completed
- ^{nat}Lu(n,xγ), – levels, isomers – under analysis.
- ^{150}Sm(n,n'γ) – pre-equilibrium analysis continuing – NCState, LLNL
- ^{186}W(n,xγ) – analysis in progress -- NCState, LLNL
- ^{70,72,74}Ge, ^{100}Mo,^{124}Sn, ^{130}Te, ^{138}Ba, data acquired.
- ^{nat}Cu, ^{76}Ge, ^{nat}Pb, (LANL) and ^{nat}Te (UCB) -- (n,xγ) for backgrounds in $0\nu\beta\beta$ decay experiments – analysis in progress
- Fe, Cr (n,xγ) – "Reference cross sections"

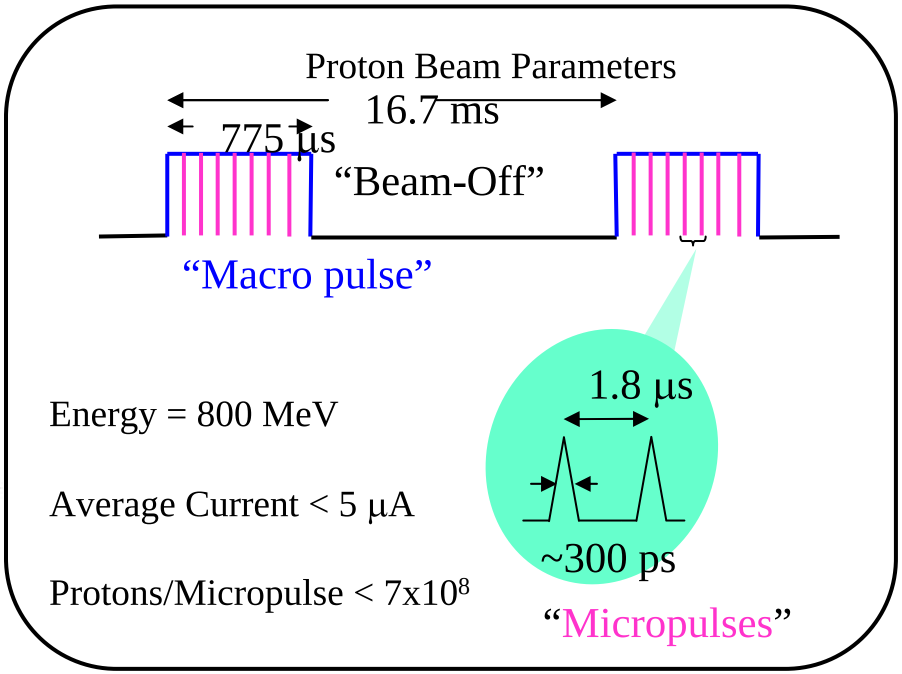
Excitation functions for ^{202,204}Tl from ^{203,205}Tl(n,2n)





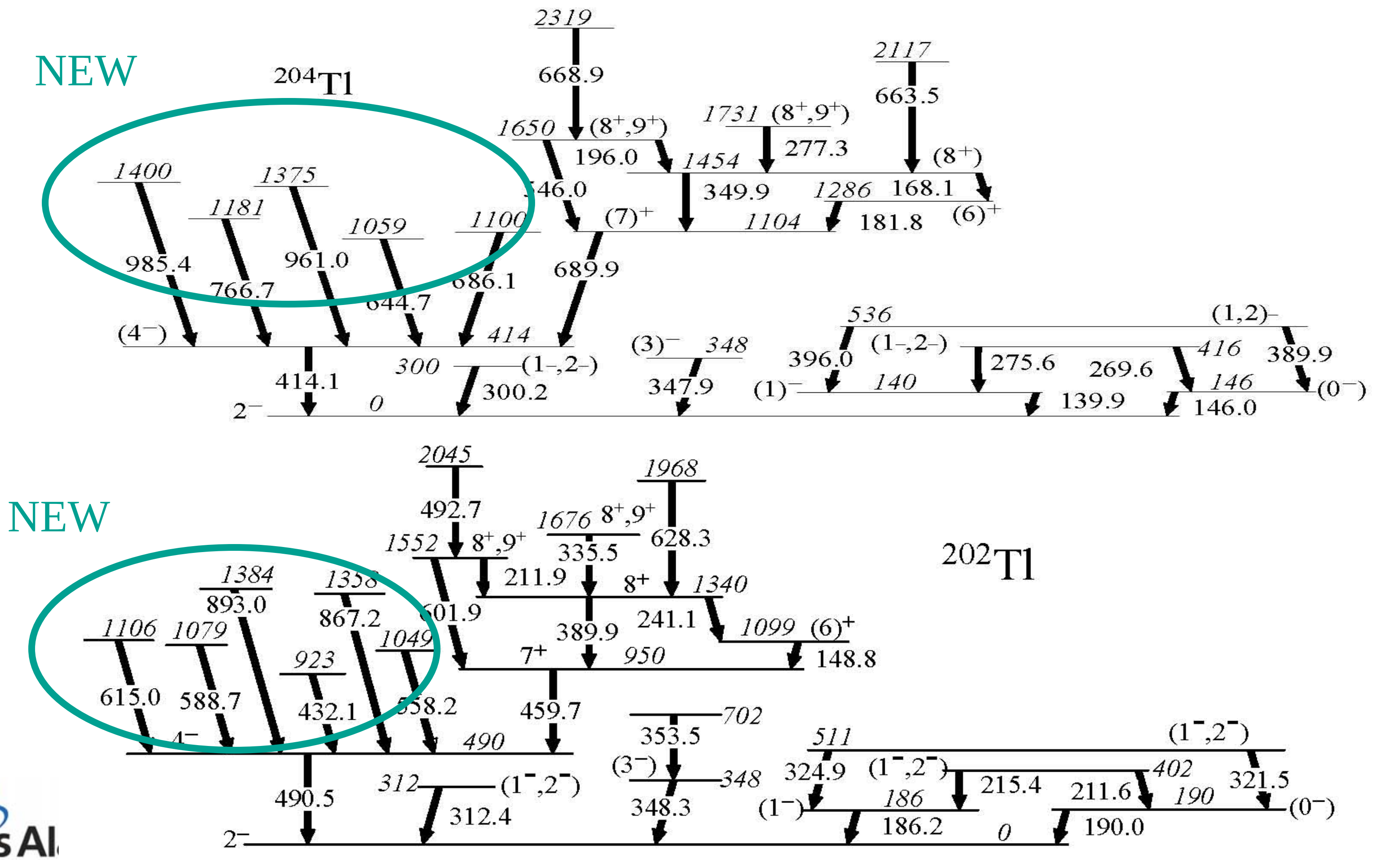
Half-life measurements with GEANIE



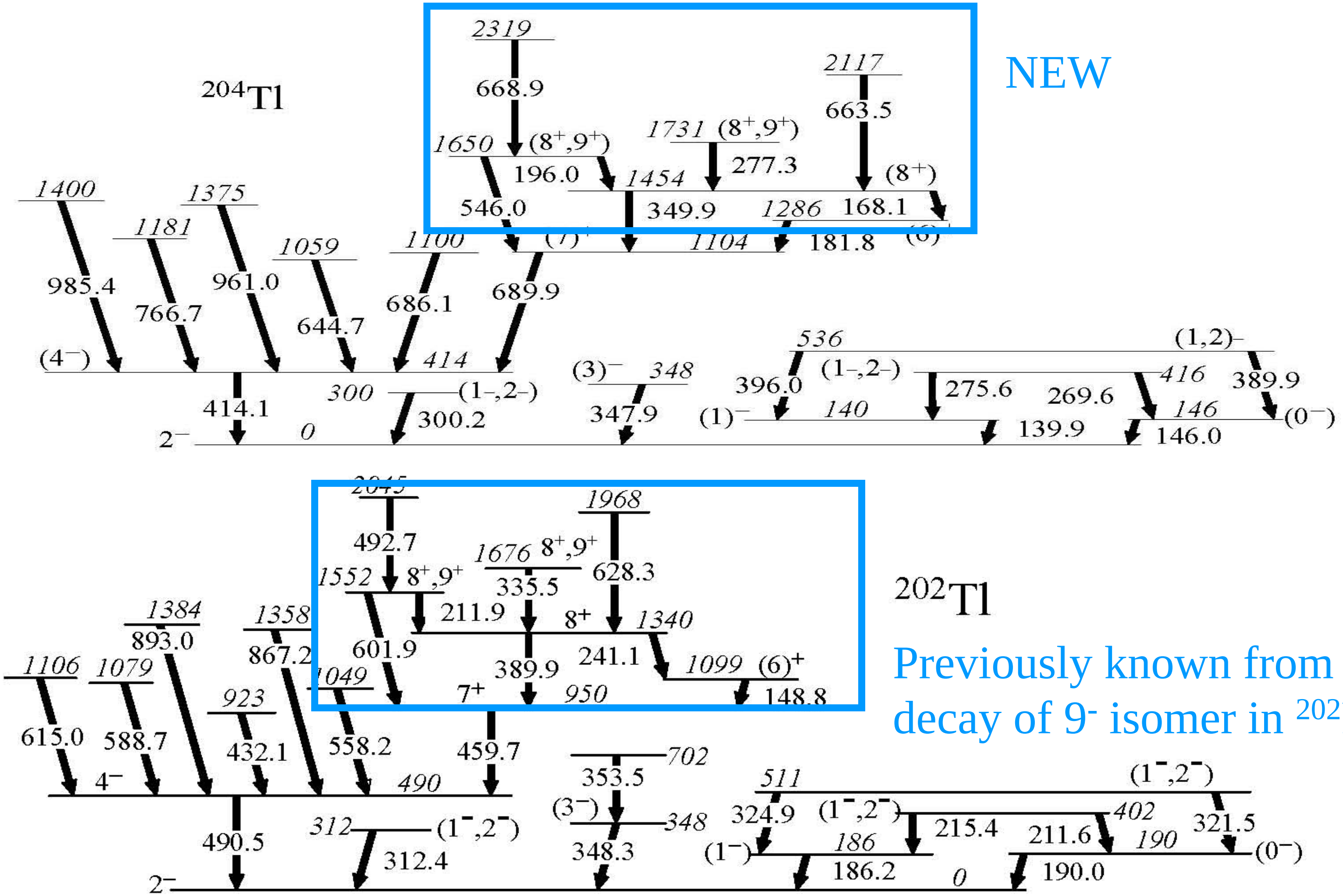


- Measure decay gamma rays during the low background, beam-off period (~16ms)
- Times are established with a very stable 10 MHz clock
- Especially well-suited for µs to ms time range

Level schemes for ^{202,204}Tl



Level schemes for ^{202,204}Tl



NEW

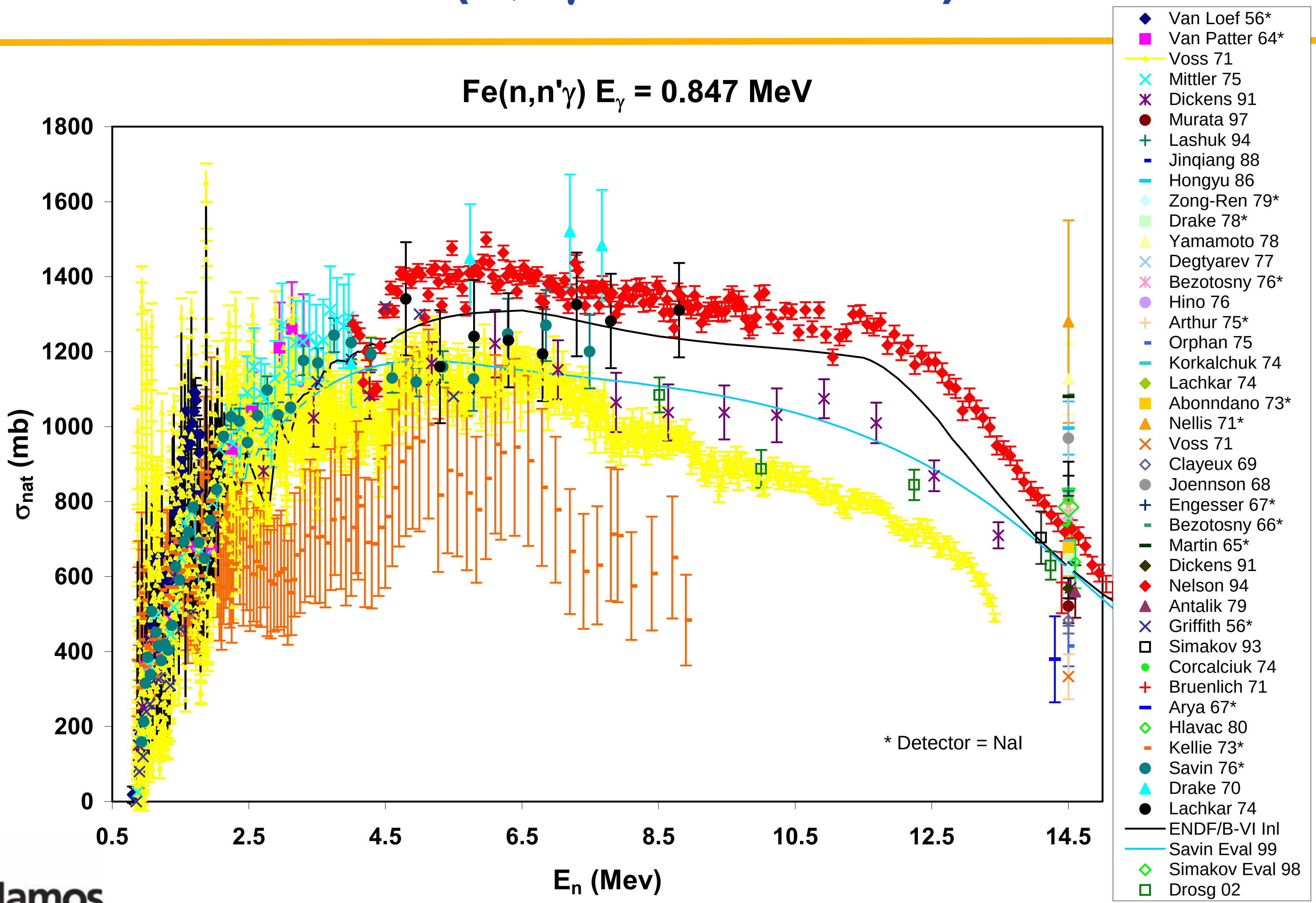
Previously known from β decay of 9- isomer in ^{202}Pb

Reference cross sections provide the metric for other measurements

- Inelastic scattering and reactions at MeV neutron energies
  - A number of secondary standards have been proposed at the 5 to 10% level (C – 4439, Al – 2221 & 3004 keV, Si – 1778 keV, Fe – 847 keV, Cr 1434 keV)
  - Backgrounds tend to be larger due to reactions of neutrons in the detector and surroundings
  - Inelastic scattering on Fe has been used extensively as a secondary standard for neutron-induced gamma-ray production measurements – but there are large differences in measured cross sections and even some variation in evaluations
  - There are drawbacks to all of the above reference cross sections for the most typical incident neutron energy ranges of a few MeV for inelastic scattering and near 14 MeV

Contact: Ron Nelson

Significant differences exist in data sets and evaluations of $^{nat}Fe(n,x\gamma = 0.847\ MeV)$



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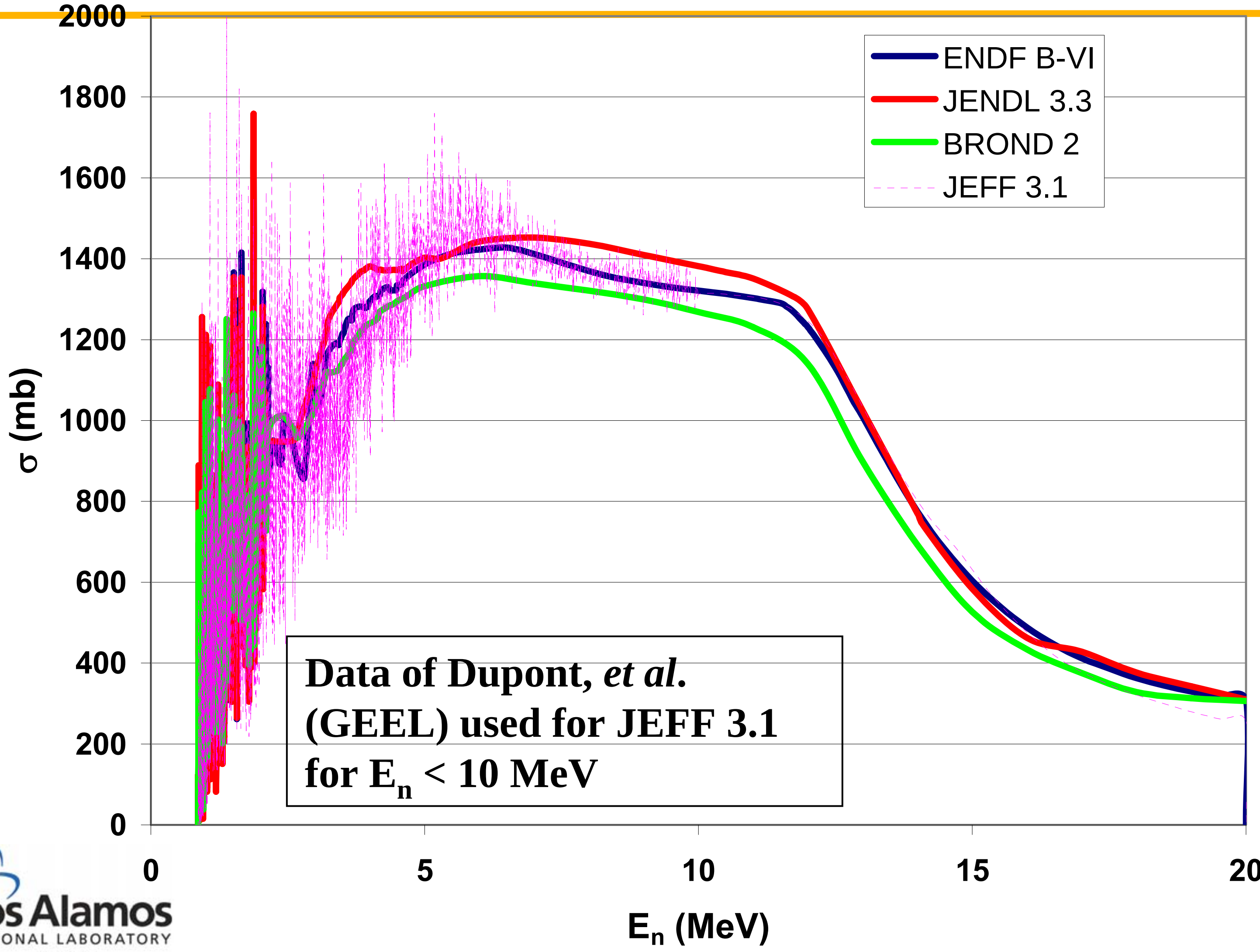
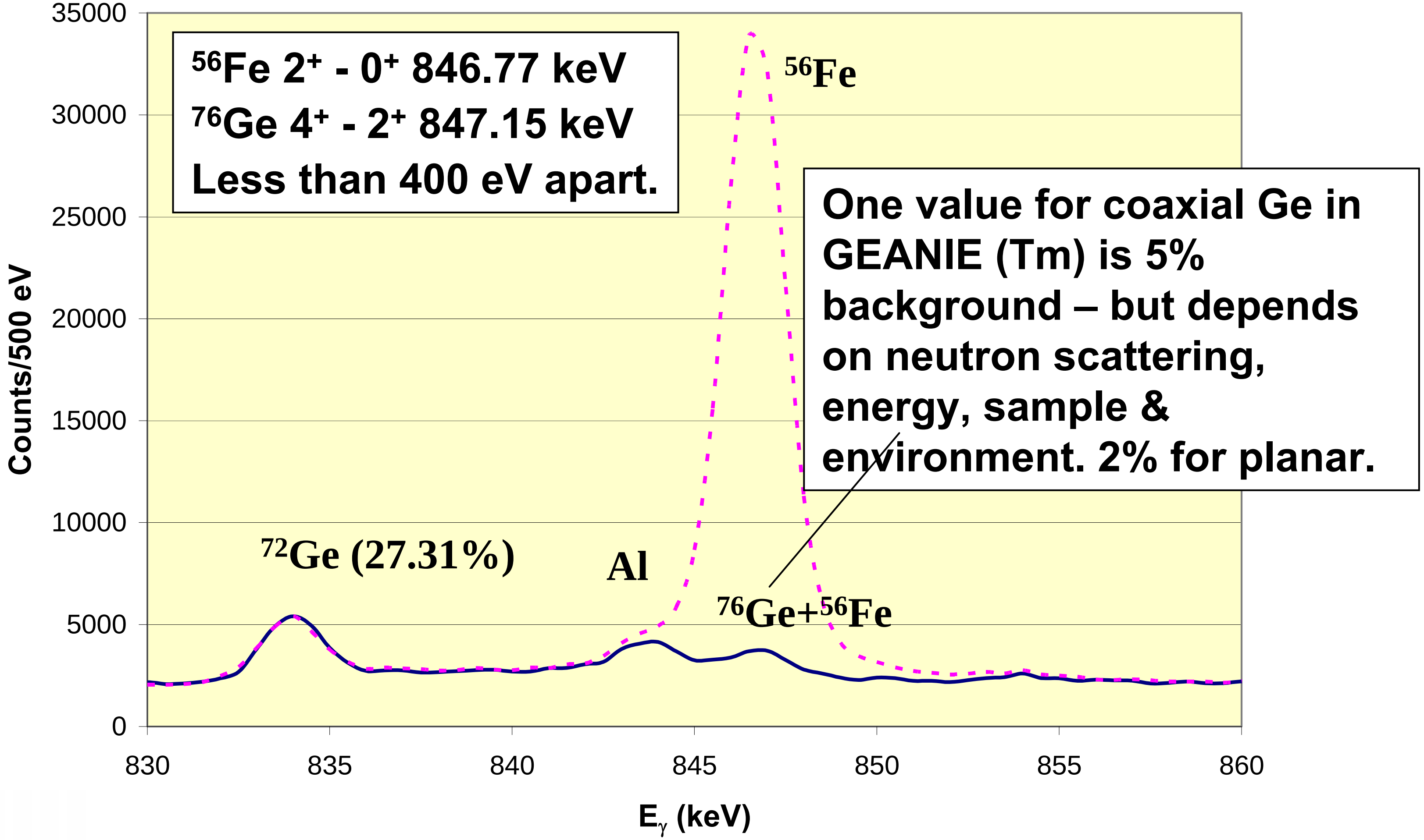
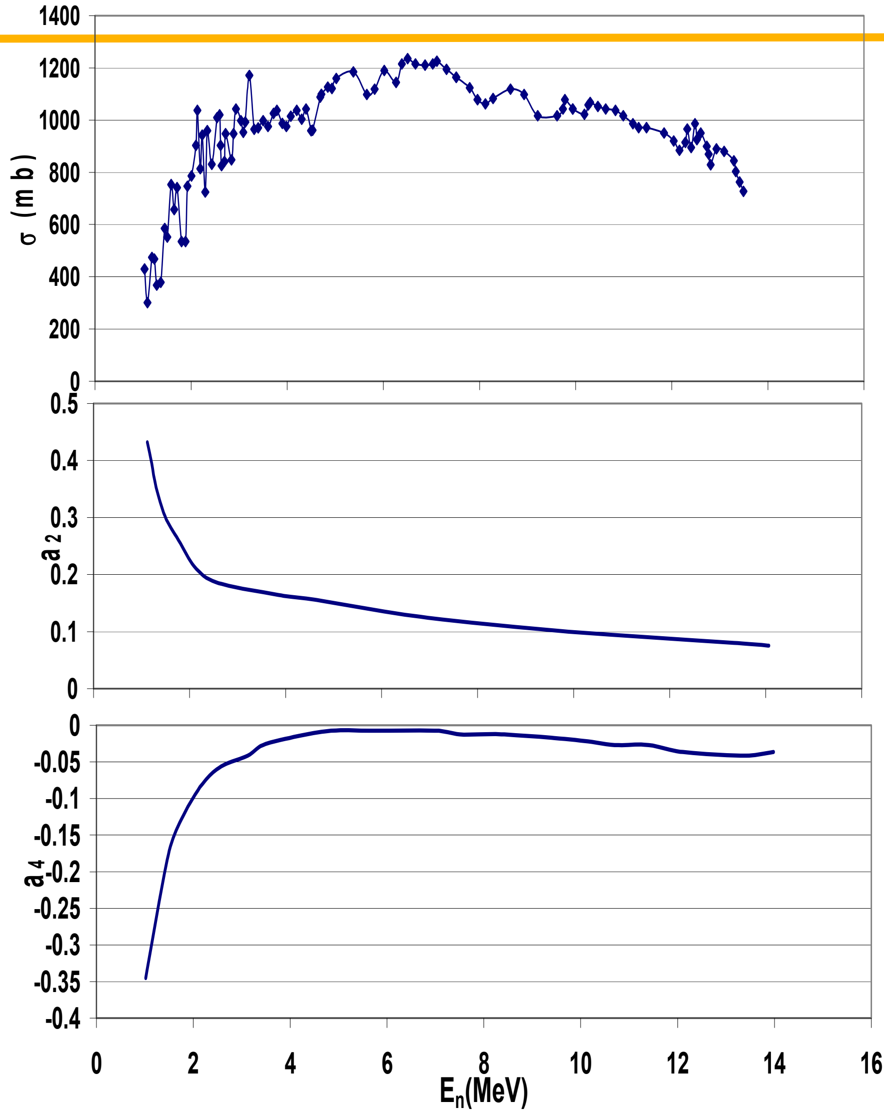
Data evaluations differ on $^{56}Fe(n,x\gamma = 0.847\ MeV)$ cross sections



Iron and aluminum in the detectors & experimental setup, as well as ^{76}Ge (7.83%), may produce backgrounds near 847 keV



Angular distribution coefficients for Fe(n,xγ= 0.847 MeV) show strong effects at low incident neutron energies



Angular Distribution Coefficients

$$\sigma = A_0(1 + a_2P_2 + a_4P_4)$$

From M. Savin, et al., IAEA Report INDC (CCP) **426** p. 95 (2000) translated from Yadernye Konstanty 2 (1999).

Search for a potentially better neutron-induced gamma-ray reference cross section

- Consider all stable elements – prefer them to compounds
  - Eliminate gases and liquids (except as compounds)
  - Eliminate alkali metals
  - Eliminate rare earths that oxidize readily
  - Eliminate expensive (\$/g > Au) elements (Sc & Rh)
  - Require natural abundance > 70 %
- Look first at gamma ray energies $0.1 < E\gamma < 1.5$ MeV
- Want cross sections > 200 mb
- Consider compounds (e.g. BeO, Teflon (CF_2), Li_2CO_3, melamine ($C_3H_6N_6$),) with Be, C, or O that have very few gammas or mainly high energy gammas
- Next slide has results

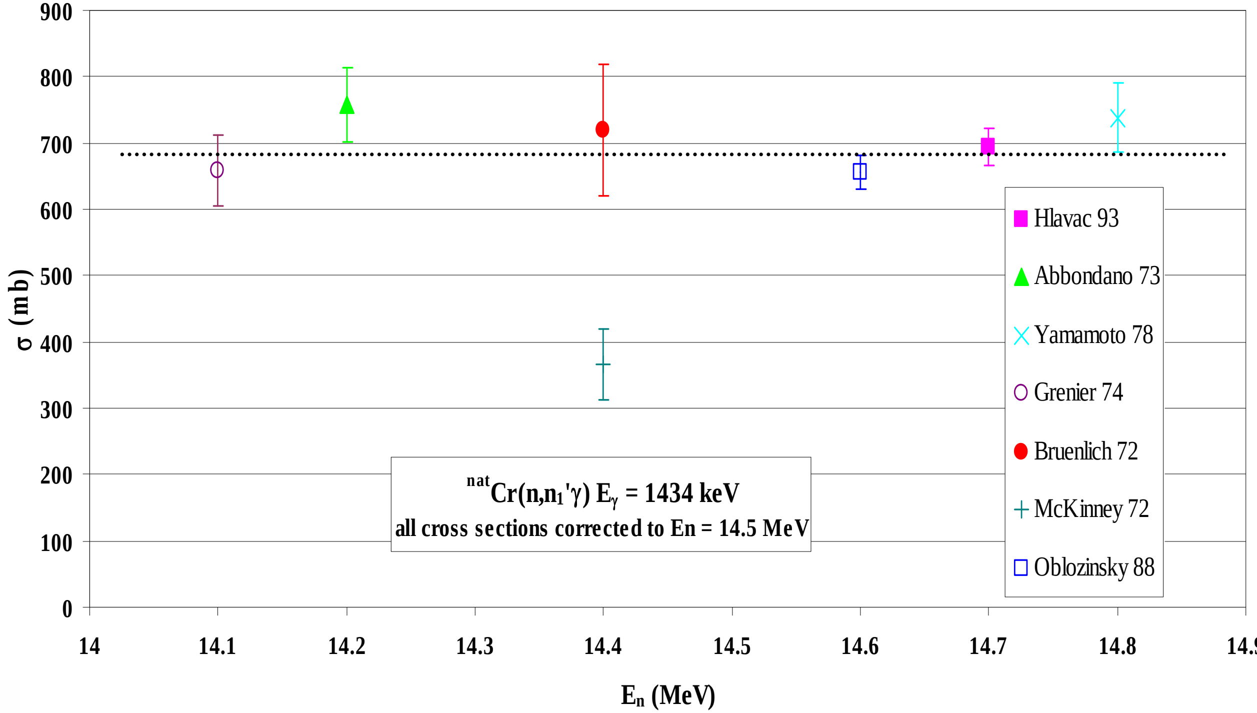
Potential neutron-induced gamma-ray reference samples and their larger cross-section lines

- Chosen "best" cross sections for (n,n') and (n,2n)

Element	Isotope		Eγ	reaction	σ(14 MeV)	Eγ	reaction	σ(14 MeV)	Eγ	reaction	σ(14 MeV)
Niobium	93		**949**	**(n,n')**	**264**	**501**	**(n,2n)**	**263**	**357**	**(n,2n)**	**239**
Gold	197		**147.8**	**(n,2n)**	**490**	**547.5**	**(n,n') 5.0**	**358**			
Titanium	48		**984**	**(n,n')**	**666**	**160**	**(n,2n+n')**	**404**			
Iron	56		847	(n,n')	785	**1238**	**(n,n'+2n)**	**393**			
Chromium	52		1434	(n,n')	695	**935**	**(n,n'+2n)**	**210**			
Manganese	55		156	(n,2n)	542	126	(n,n')	383	212	(n,2n)	299
Magnesium	24		1369	(n,n')	450	472	(n,p) 20ms	105			
Vanadium	51		226	(n,2n)	368	320	(n,n')	313			
Bismuth	209		1006	(n,2n)	210	565.3	(n,2n)	125	650.7	(n,2n)	130

Cr – 1434- keV gamma ray from ^{52}Cr[83.709%](n,xγ) has good consistency for γ-ray cross sections at E_n = 14.5 MeV

- Measurements – 6 of 7 agree within errors
- Evaluation of Simakov, *et al.* at E_n = 14.5 MeV gives a 3.8% error in the evaluated cross section



DANCE (n,γ)

Contact:
John Ullmann
Aaron Couture

Recent DANCE Publications

- Spin measurements for ^{147}Sm + n resonances. P.E. Koehler, et al., Phys. Rev. C 76 025804 (2007)
- Spin and parity assignments for 94,95Mo neutron resonances. S.A. Sheets, et al., Phys. Rev. C 76 064317 (2007)
- Capture cross section of ^{62}Ni. A.M. Alpizar-Vicente, et al., Phys. Rev. C 77 015806 (2008).
- ^{237}Np(n,γ) cross section. E.-I. Esch, et al., Phys. Rev. C 77 034309 (2008).
- ^{241}Am (n,γ) cross section. M. Jandel et al., Phys. Rev. C 78 034609 (2008).

Analysis of DANCE neutron-capture data

^{75}As	(August Keksis, Los Alamos)
^{89}Y	(Andrii Chyzh, NCSU/Los Alamos)
^{143}Nd, ^{149}Sm	(Paul Koehler, Oak Ridge)
^{151,153}Eu	(U. Agvaanluvsan, Stanford; J.A. Becker Livermore. Data available as LLNL Report)
^{152}Eu	(Aaron Couture, Los Alamos. 30 MBq target!)
^{155,156,157,158,160}Gd	(North Carolina State University. B. Baramsai, D. Dashdorj, A. Chyzh, G. Mitchell)
^{175,176}Lu	(Olivier Roig, CEA)
^{203,205}Tl	(Aaron Couture, Los Alamos)
^{240,242}Pu	(Aaron Couture, Los Alamos. Preliminary data reported to GNEP program)
^{242m,243}Am	M. Jandel (Los Alamos)

$^{241}Am(n,\gamma)$ low-energy and thermal results

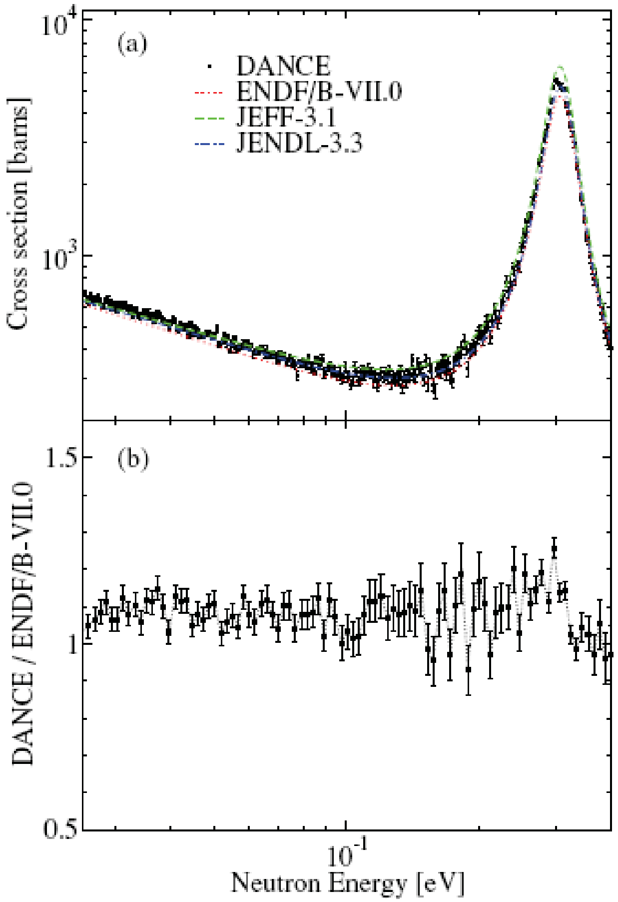


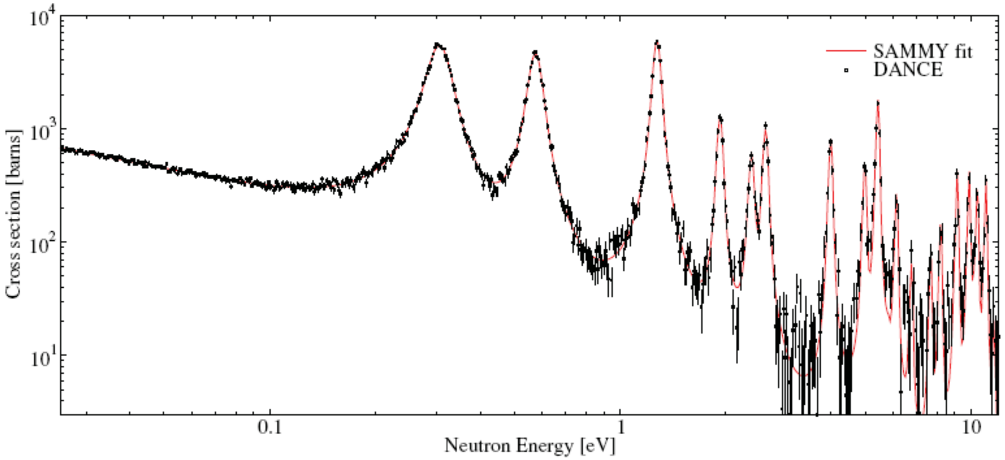
TABLE I. Experimental and evaluated thermal neutron capture cross sections and resonance integrals RI above 0.5 eV for ^{241}Am.

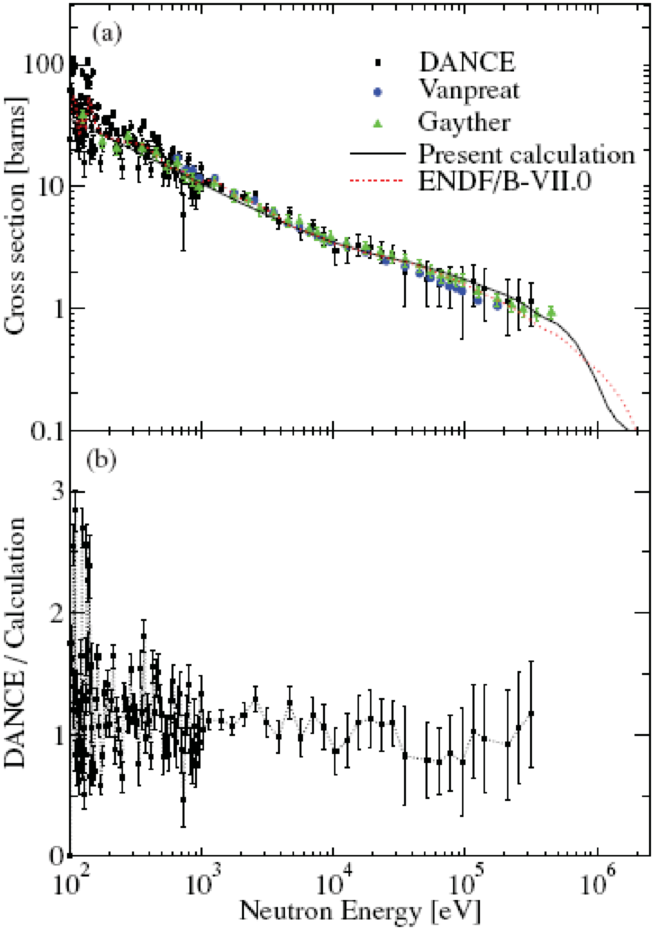
Reference	Year	σ_{th} (b)	RI (b)
Experiment			
This work	2007	665 ± 33	1553 ± 7
Nakamura *et al.*[a] [2]	2007	690	
Fioni *et al.* [3]	2001	696 ± 48	
Maidana *et al.* [4]	2001	672 ± 10	
Shinohara *et al.* [5]	1997	854 ± 58	1808 ± 146
Wisshak *et al.* [12]	1982	625 ± 35	
Belanova *et al.* [6]	1976	622	
Adamchuk *et al.* [7]	1976	600	
Kalebin *et al.* [8]	1976	625 ± 20	
Weston *et al.* [13]	1976	582 ± 50	
Harbour *et al.* [10]	1973	612 ± 25	
Dovbenko *et al.* [9]	1971	654 ± 104	
Pomerance [11]	1955	625 ± 35	
Evaluation			
Mughabghab [33]	2006	585 ± 12	1425 ± 112
JEFF-3.1[b] [30]	2006	647 ± 32	1526.4
ENDF/B-VII.0[c] [29]	2006	620 ± 13	
JENDL-3.3 [31]	2002	639.4	1460

[a]Derived from $\sigma_{0,g} = 628 \pm 22$ b using branching ratio of 0.9 ± 0.09 for the isomer state production.
[b]Based on Refs. [3,4,6,7].
[c]Based on Refs. [8–13].

^{241}Am(n,γ) results in resonance and UR regions

Some examples of data





^{241}Am(n,γ), Low-energy resonances

TABLE II. Resonance parameters for neutron resonances in the energy region between 0.3 and 12 eV. The results obtained from the SAMMY fit to our data are compared with existing values from ENDF/B-VII.0 [29] and Mughabghab [33] evaluations. No error bars are given for our results if that particular parameter was kept fixed during the SAMMY fit.

ENDF/B-VII.0			Mughabghab [33]			This work		
E_0 (eV)	Γ_γ (meV)	$2g\Gamma_n$ (meV)	E_0 (eV)	Γ_γ (meV)	$2g\Gamma_n$ (meV)	E_0 (eV)	Γ_γ (meV)	$2g\Gamma_n$ (meV)
0.308	46.9	0.056935	0.307(2)	46.8(3)	0.0560(5)	0.3051(2)	44.4(3)	0.0622(4)
0.576	47.3	0.0929	0.574(4)	47.2(3)	0.0923(2)	0.5724(3)	43.3(5)	0.1030(7)
1.276	47.9	0.322	1.268(4)	49.6(7)	0.320(8)	1.2718(4)	45.3(7)	0.347(4)
1.928	44.6	0.11133	1.930(5)	44.6(3)	0.113(2)	1.922(1)	41(2)	0.117(2)
2.372	44	0.071666	2.380(8)	42.7(3)	0.07(7)	2.363(2)	50(8)	0.078(3)
2.598	47.6	0.15	2.590(9)	46.6(6)	0.15(2)	2.599(2)	48(3)	0.147(4)
3.973	44.5	0.20966	3.97(1)	44.5(3)	0.22(6)	3.973	44.5	0.208(8)
4.968	43.8	0.17733	4.97(1)	43.8(4)	0.175(4)	4.968	43.8	0.178(7)
5.415	44.2	0.766	5.42(1)	44.2(1)	0.78(2)	5.415	44.2	0.74(2)
5.8	44.2	0.002	–	–	–	5.8	44.2	0.0020(2)
6.117	43.8	0.12366	6.12(1)	43.8(7)	0.127(2)	6.117	43.8	0.127(9)
6.745	44.2	0.032857	6.74(1)	–	0.030(2)	6.745	44.2	0.033(3)
7.659	44.2	0.04557	7.66(1)	–	0.039(2)	7.659	44.2	0.048(5)
8.173	42.7	0.107	8.17(1)	42.7±1.2	0.107(3)	8.173	42.7	0.105(7)
9.113	44.2	0.37733	9.12(2)	44.2(6)	0.379(8)	9.113	44.2	0.364(24)
9.851	43.9	0.39766	9.85(2)	43.9(6)	0.40(1)	9.851	43.9	0.40(2)
10.116	44.2	0.0255	10.12(2)	–	0.026(1)	10.116	44.2	0.027(3)
10.403	42.4	0.148767	10.43(2)	42.4(8)	0.33(1)	10.404(6)	45(4)	0.35(2)
10.997	46.5	0.40333	10.98(2)	46.5(8)	0.40(2)	10.997	46.5	0.41(3)
11.583	44.2	0.021333	11.58(2)	–	0.016(1)	11.583	44.2	0.022(2)

Fission Cross Sections

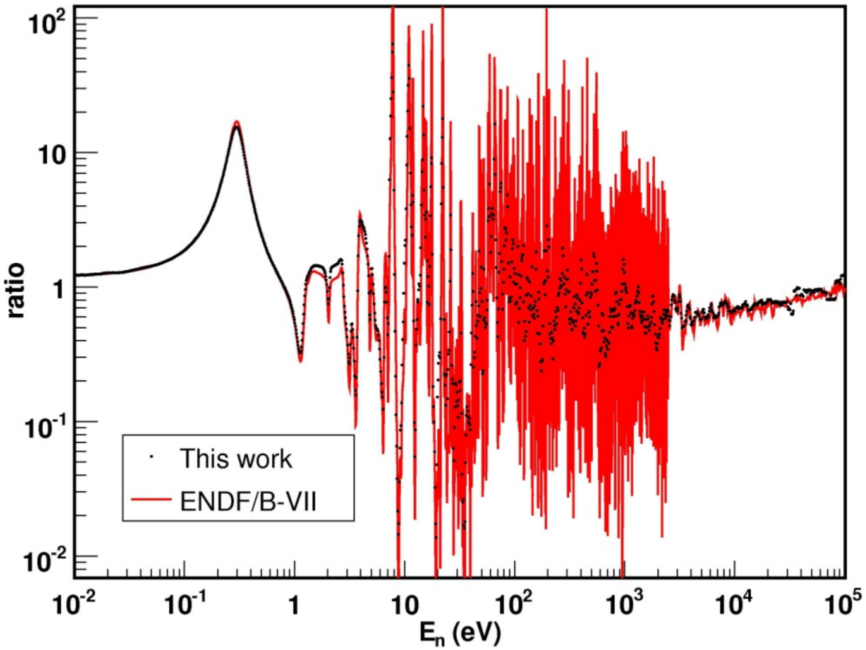
Contact:
Fredrik Tovesson
Tony Hill

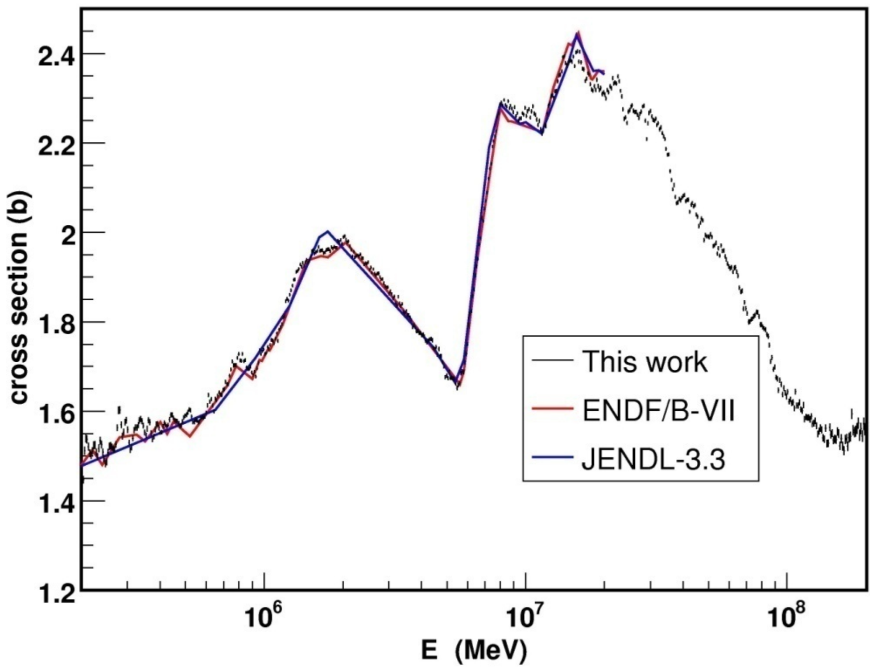
Status of fission cross sections measurements at LANSCE

$$\sim 1 \text{ eV} < E_n < 200 \text{ MeV}$$

- ^{237}Np – published
- ^{240,242}Pu – submitted for publication
- $^{239,\,241}$Pu – data to GNEP evaluators; publication in preparation
- ^{241}Am – milestone for next year; sample needed
- ^{233,238}U – planned
- ^{232}Th – sub-barrier cross section being measured

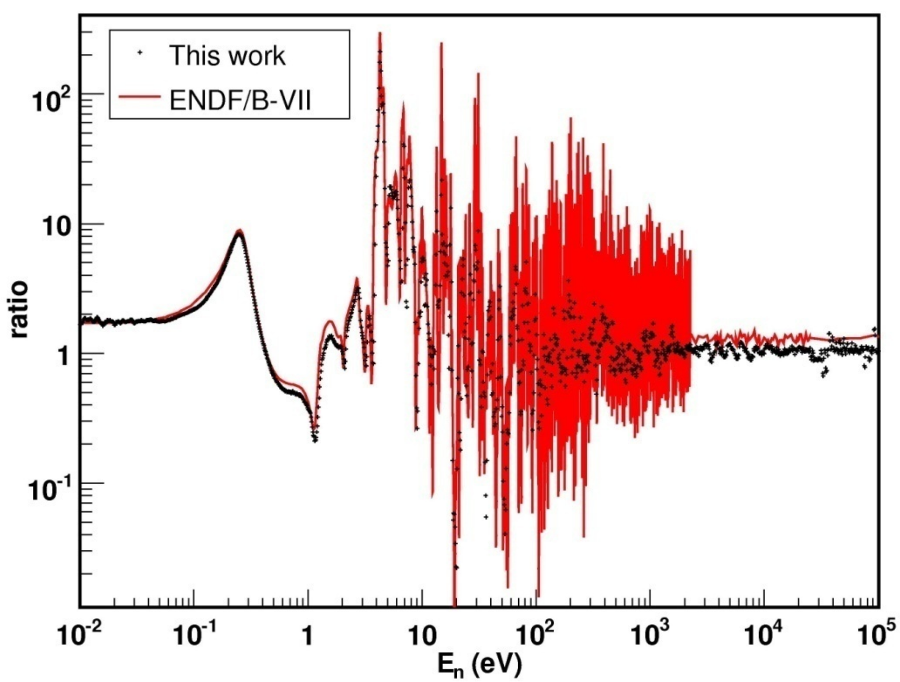
Pu-239 fission

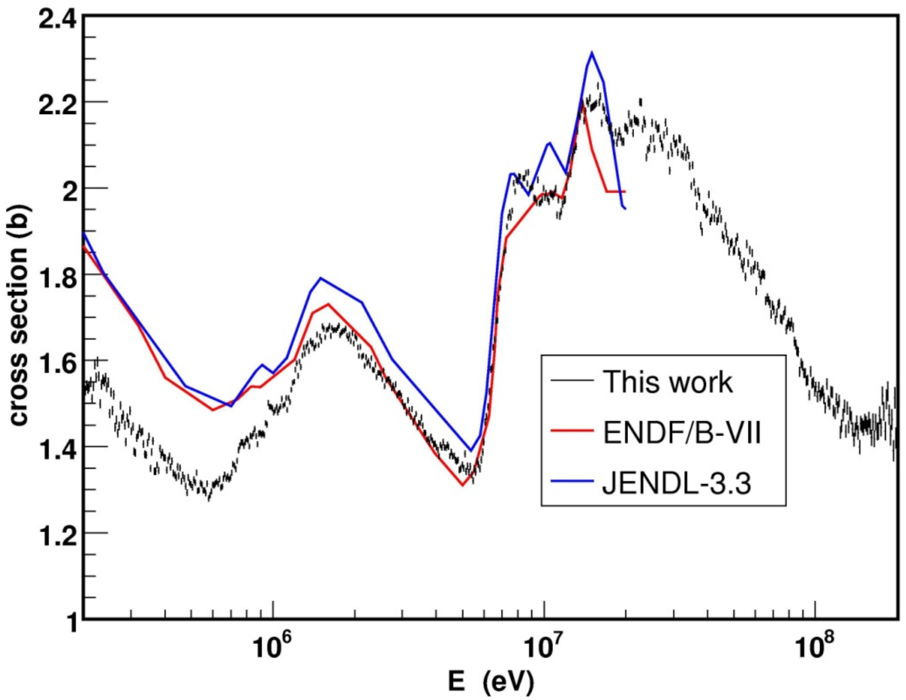




- **Final results delivered to GNEP Sept. 2008**
- **ENDF in better shape than JENDL in the MeV-region**

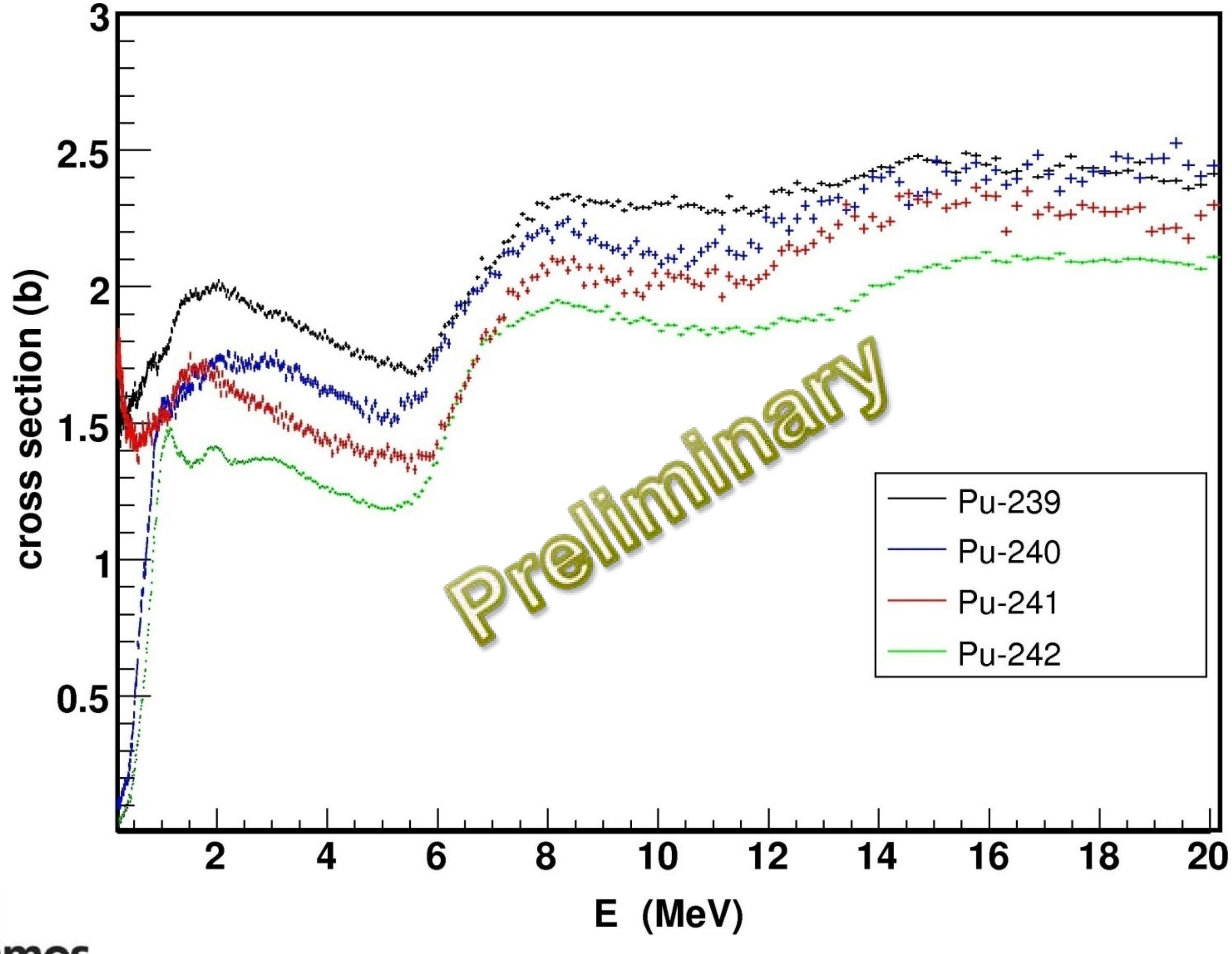
Pu-241 fission





- **20% discrepancies in the 10 keV – 1 MeV region**
- **First measurement above 30 MeV**
- **Only one other measurement in the resonance region in the last 30 years**

Fission cross sections of $^{239\text{-}242}Pu$

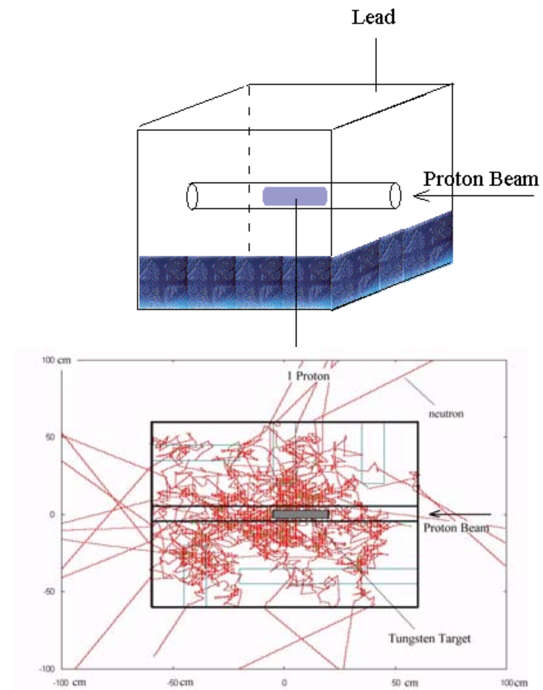


Fission and Other Cross Sections

On Very Small Samples

Contact:
Bob Haight

A Lead Slowing-Down Spectrometer is under development, driven by 800 MeV protons from the PSR



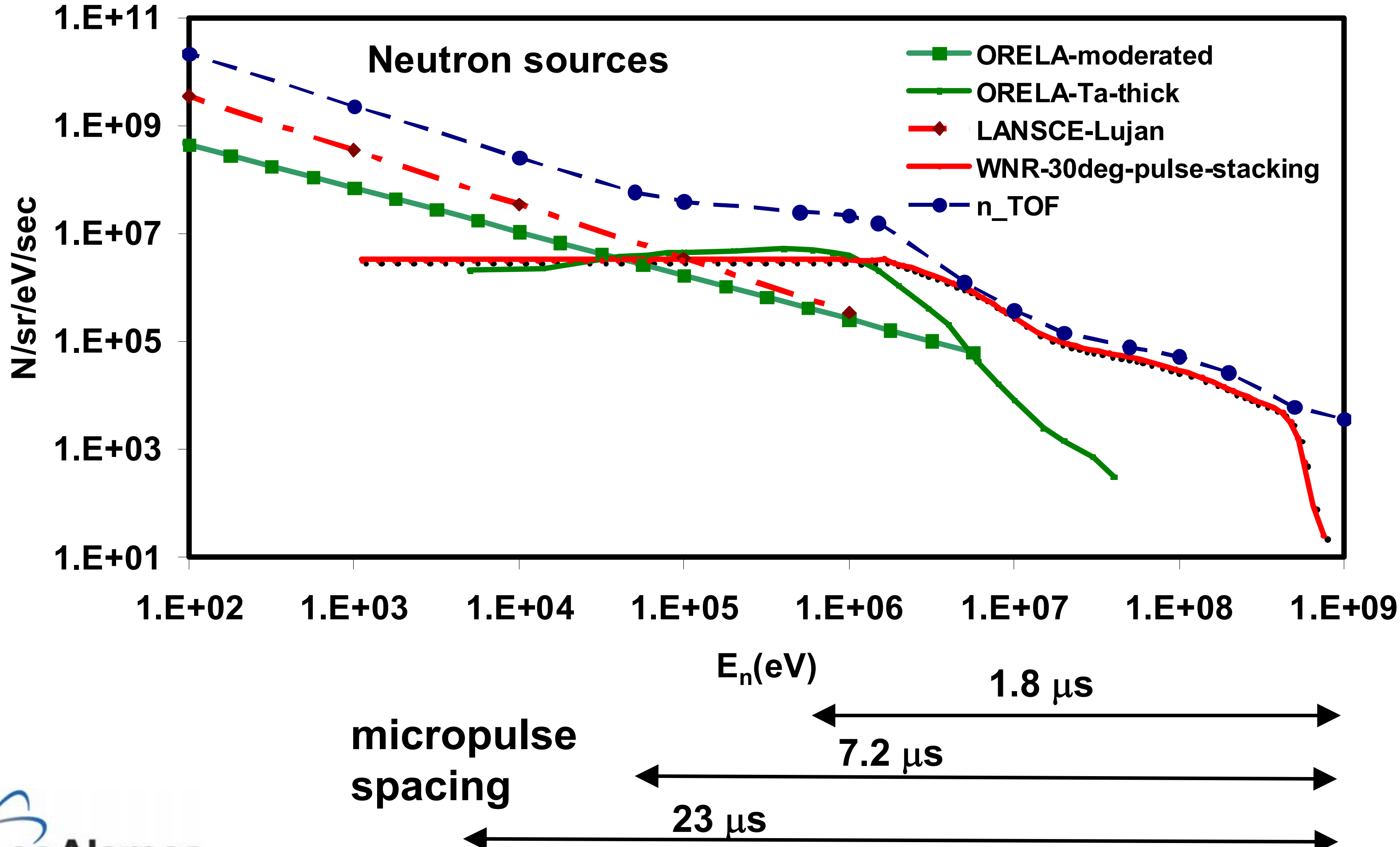
Neutron trajectories following the interaction of 1 proton with the tungsten target in the lead cube

Contact:
Bob Haight

Pulse stacking:

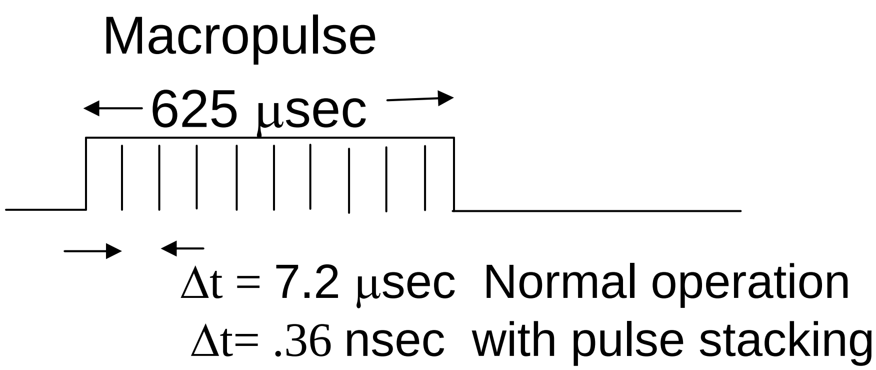
to increase usable flux of neutrons

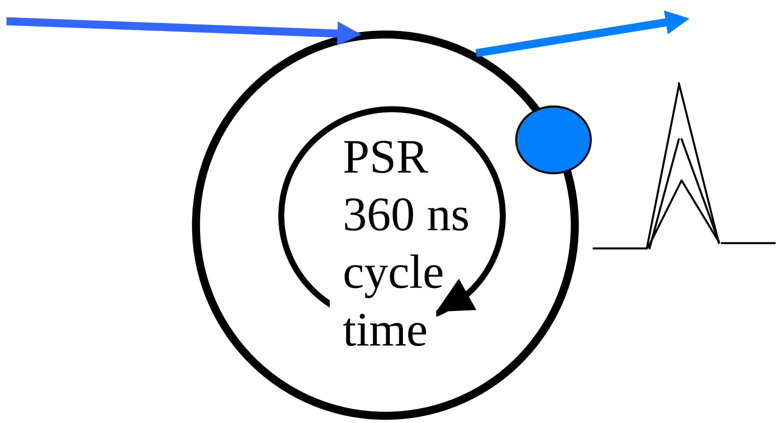
Pulse stacking will extend WNR source to lower usable neutron energies



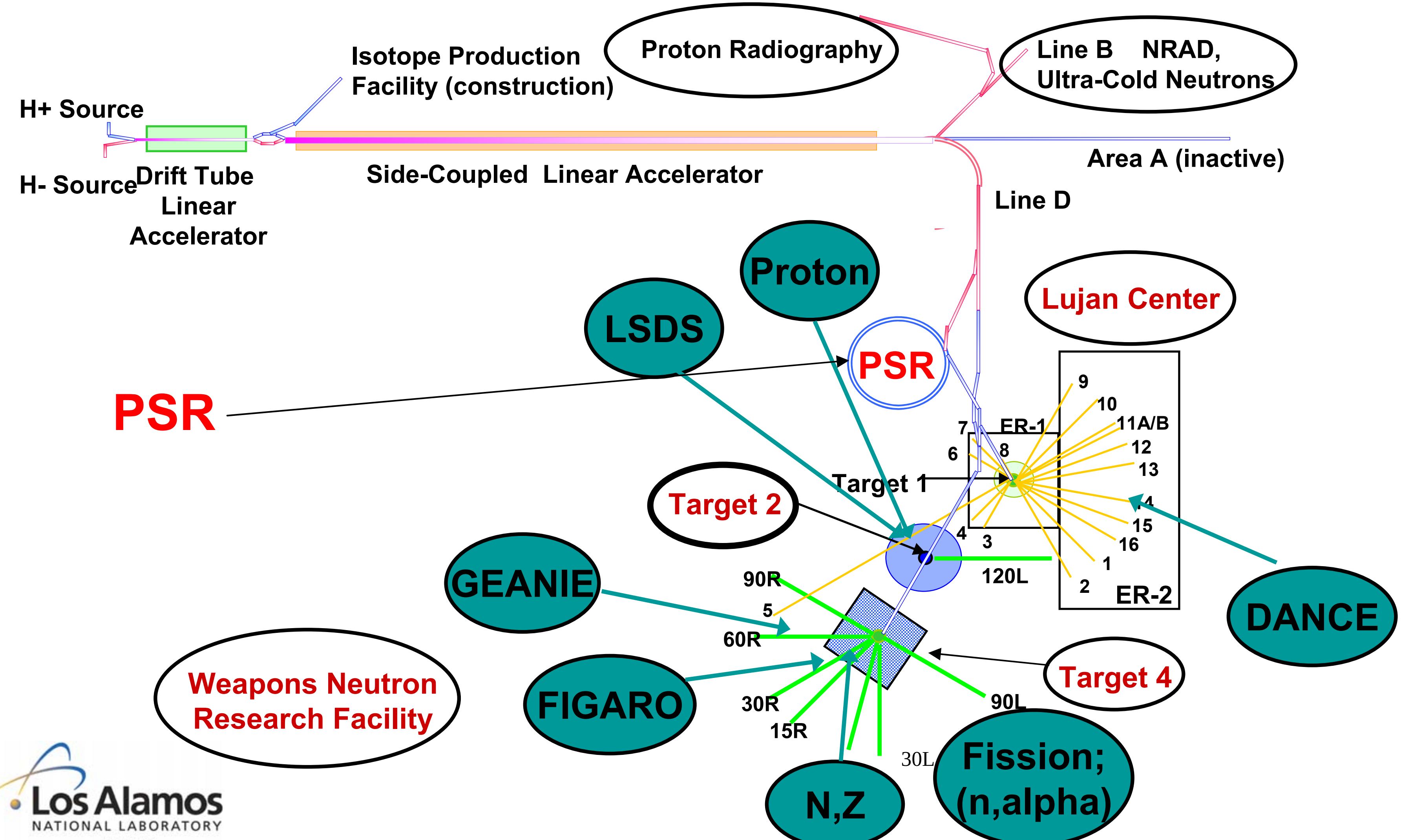
Stack single micropulses in PSR for entire macropulse (Full-fill mode)

- For 10 keV neutrons need pulse separation of 7.2 μsec (10 m)
- Separate micropulses by 360 ns in macropulse. Get x20 { 7.2 /.36 } increase in current / macropulse.
- Stack in pulses in PSR for duration of Macropulse (625 μs)
- Operate PSR at 60 Hz: 20 Hz to Lujan Center 40 Hz to Target-4
- 17 msec between pulses, x8 average beam (at 40 Hz) compared 7.2 μsec at Target-4 (at 100 Hz)
- It may be possible to store several pulses in the PSR



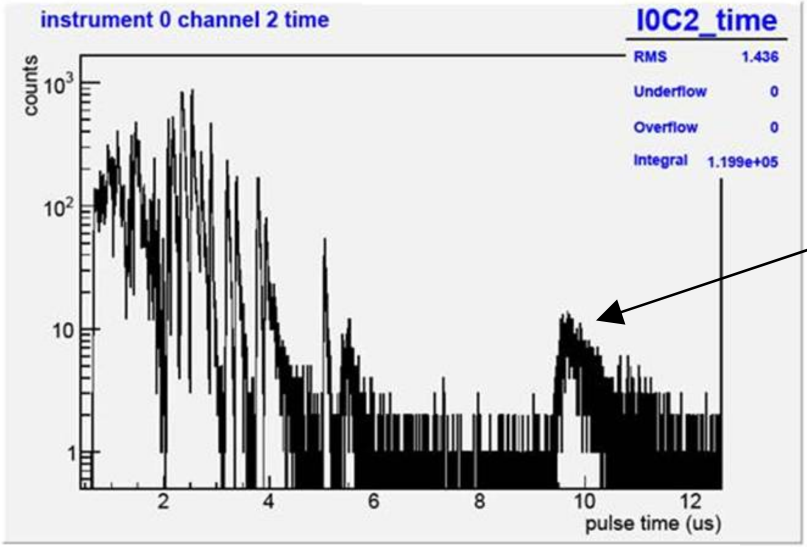


Proton storage ring serves to stack pulses

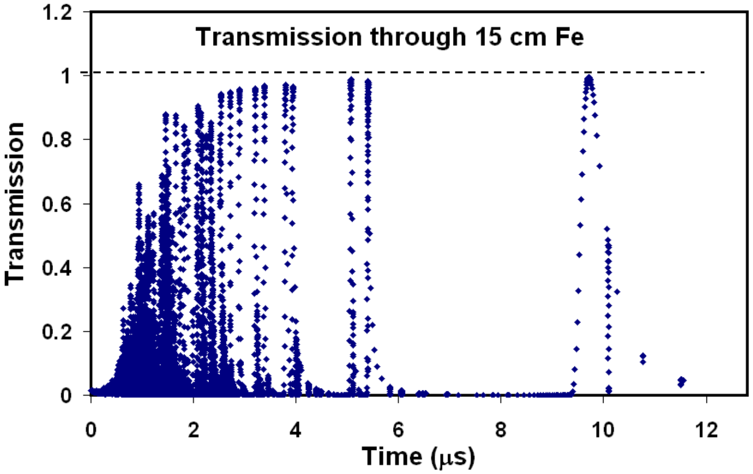


Transmission resonances observed

- 15 cm iron
- ^{6}Li glass detector
- Acqiris digitizer
- 2 minute run

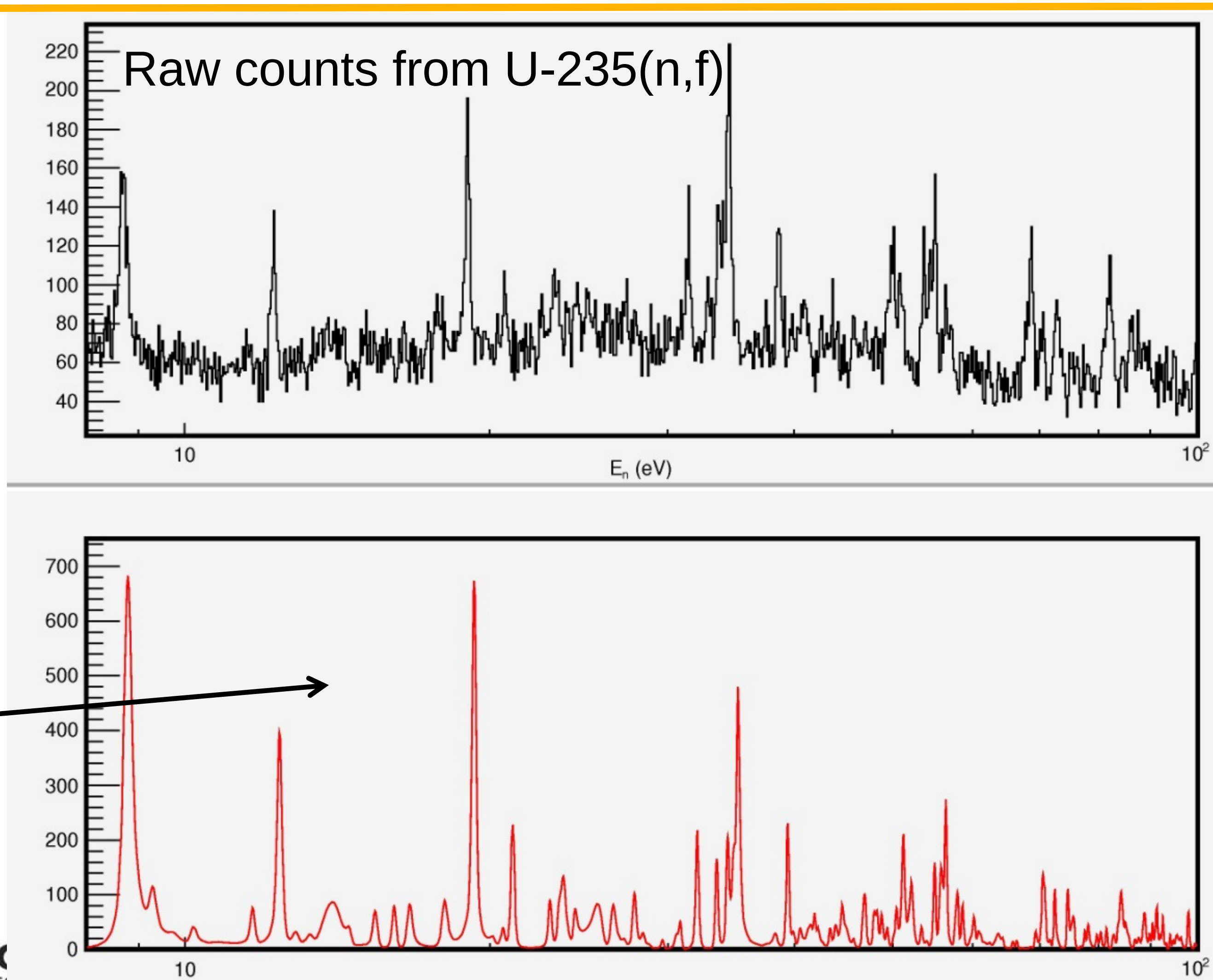


25 keV



→ Filtered beams posssible

Fission cross section resonances observed down in eV region



ENDF

We address the needs of LANSCE sponsors

- National Nuclear Security Administration
  - Program in radchem cross section measurements
    - Neutron capture cross sections on radioactive targets (DANCE)
    - Cross section measurements on high-order (n,2n), (n,xn) reactions (GEANIE)
  - Program in neutron-induced fission measurements
    - Fission product distributions (GEANIE)
    - Energy output in fission: neutron and γ-ray spectra (FIGARO)
    - Nuclear properties of fission products and isomers (GEANIE and FIGARO)
- Office of Nuclear Energy
  - Measurements in support of the AFCI program include:
    - Capture and fission cross section on actinides
    - Gas production: (n,p), (n,α) reactions in structural materials
- Office of Science
  - Support of SNS in understanding pulsed radiation effects on liquid mercury targets
  - Fundamental physics experiments and nuclear data
- National Resource
  - Nuclear science User Facility for defense, basic and applied research
  - Industrial testing of semiconductor devices in neutron beams
  - University research in nuclear science

The LANSCE program in nuclear data involves many laboratories

- GEANIE – LANL, LLNL , INL, ORNL, Bruyères-le-Châtel, NC State
- FIGARO – LANL, Bruyères-le-Châtel, LLNL
- N,Z – LANL, Ohio U
- DANCE – LANL, LLNL, ORNL, NC State, INL, Colorado School of Mines, FZK Karlsruhe
- LSDS – LANL, LLNL, BNL, Bruyères-le-Châtel, RPI
- Fission – LANL, IRMM, LLNL, INL, RPI, NERI universities
- Others – MIT, Kentucky, Kyushu, Harvard,…