

LA-UR-06-7894

Nuclear Data Experiments at LANSCE: Highlights 2006

**Robert C. Haight
Los Alamos National Laboratory**

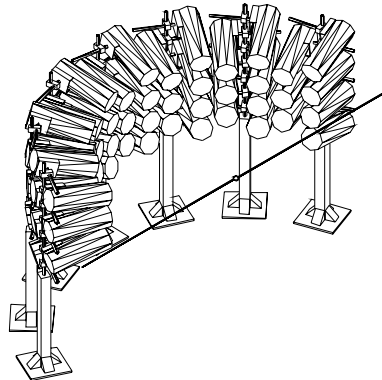
**Cross Section Evaluation Working Group Meeting
US Nuclear Data Program Meeting
Brookhaven National Laboratory
November 6-9, 2006**

Nuclear data measurements at LANSCE are made with several instruments

GEANIE (n,x γ)



FIGARO (n,xn+ γ)



DANCE (n, γ)



N,Z (n,charged particle)



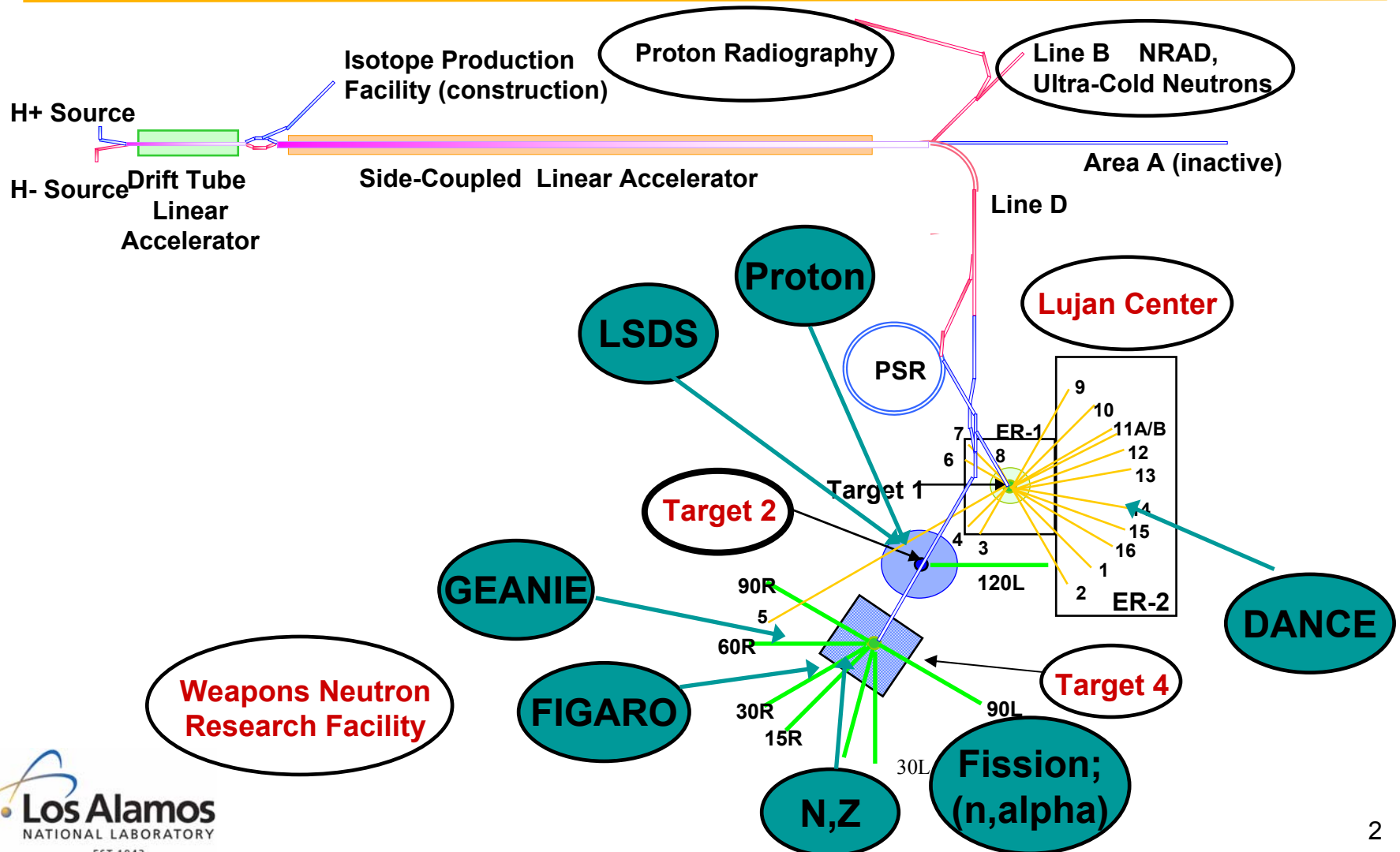
Fission

LSDS



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



GEANIE (n,x γ)



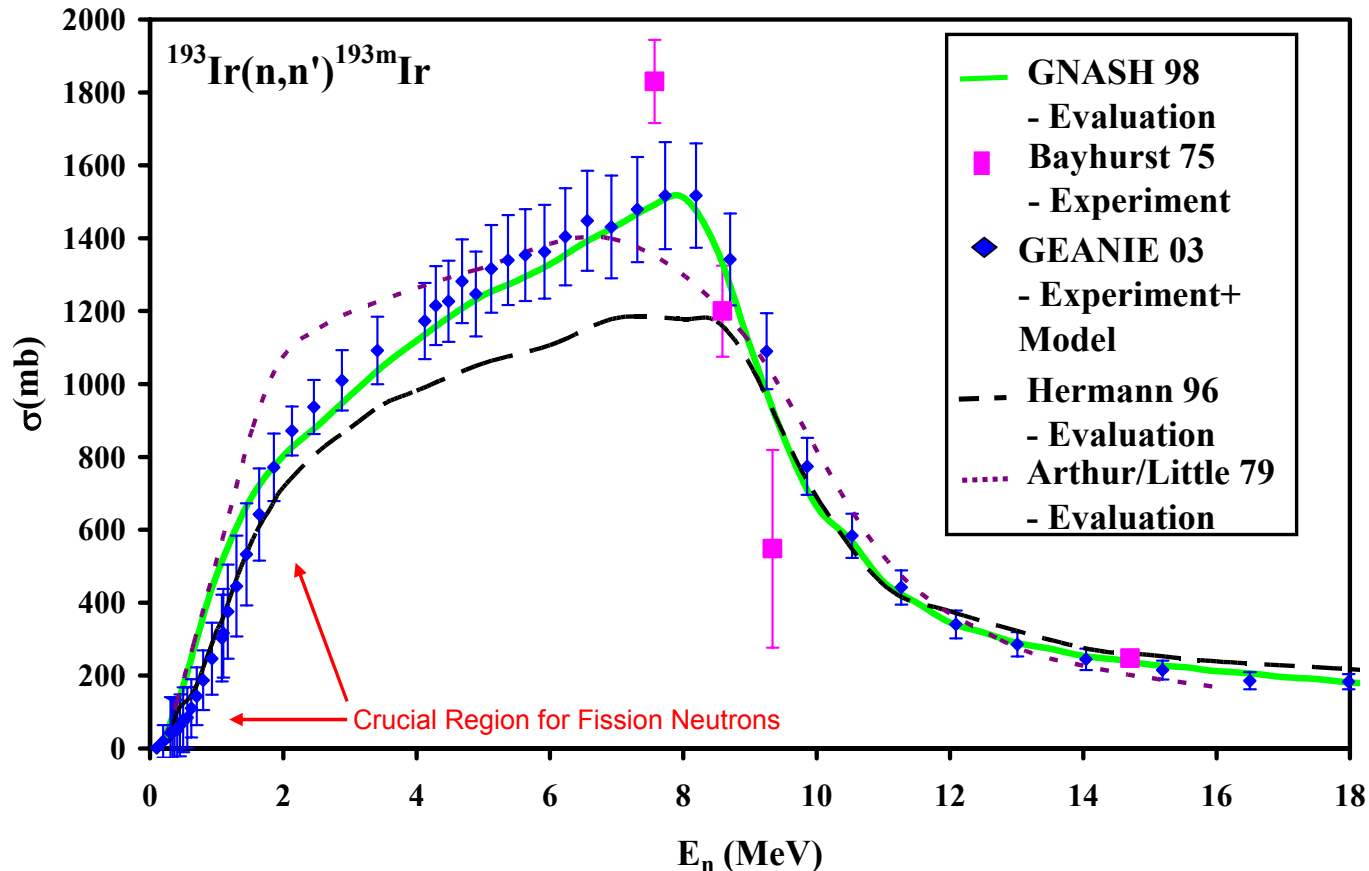
Recent & Planned GEANIE Neutron-Induced Gamma-Ray Cross Section Measurements at LANSCE/WNR

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

- $^{103}\text{Rh}(n,x\gamma)$, ^{169}Tm , $^{203,205}\text{Tl}$ – levels, isomers – under analysis
- $^{48}\text{Ti}(n,x\gamma)$ – dissertation 2005 - D. Dashdorj (NCSU/LLNL)
- $^{150}\text{Sm}(n,n'\gamma)$ – pre-equilibrium analysis continuing
- ^{186}W , $^{233}\text{U}(n,2n)$ data acquired
- $^{100}\text{Mo}(n,x\gamma)$, $^{124}\text{Sn}(n,x\gamma)$, $^{130}\text{Te}(n,x\gamma)$, $^{138}\text{Ba}(n,x\gamma)$ data acquired
- $^{70,72,74}\text{Ge}(n,x\gamma)$ data acquired
- $^{\text{nat}}\text{Pb}$ and $^{\text{nat}}\text{Te}$ for $0\nu\beta\beta$ decay experiment backgrounds – measurements in progress
- Planned Samples: ^{136}Xe , other Xe and Kr isotopes

Contact:
Ron Nelson

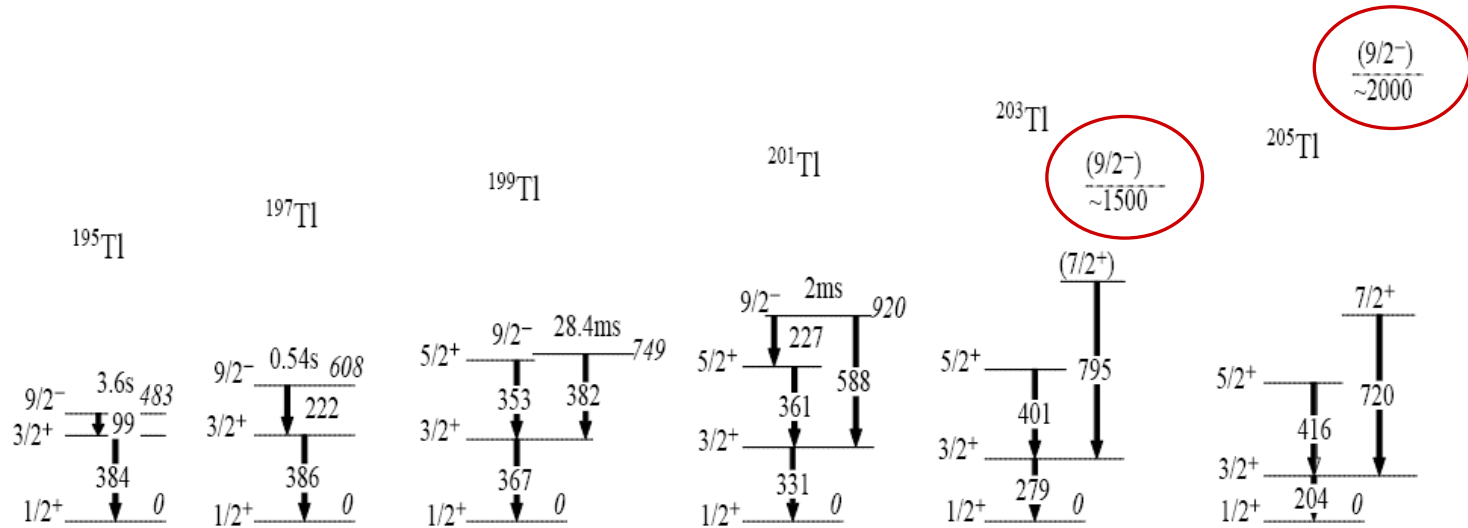
New GEANIE data significantly improve the $^{193}\text{Ir}(n,n')^{193\text{m}}\text{Ir}$ cross section database



GEANIE
LLNL/LANL

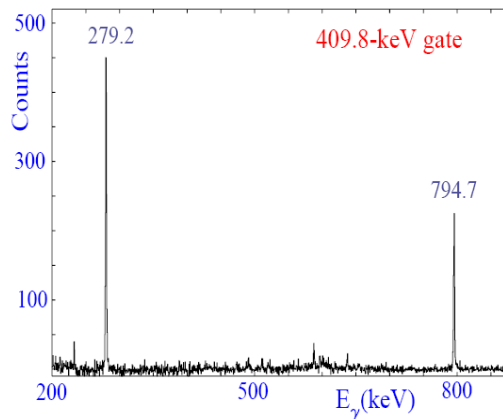
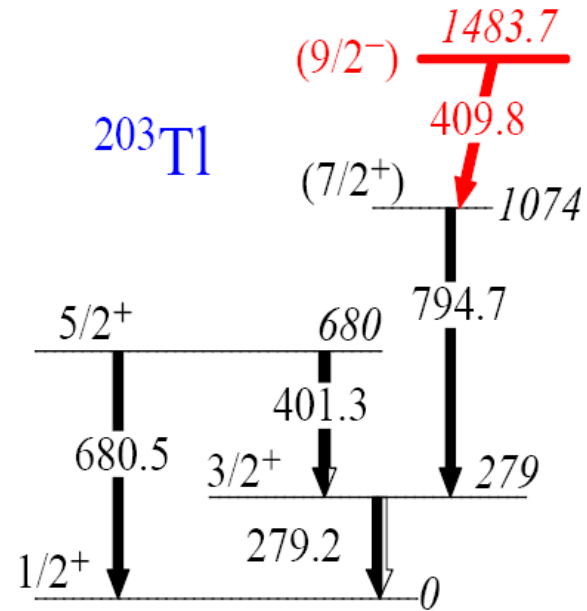
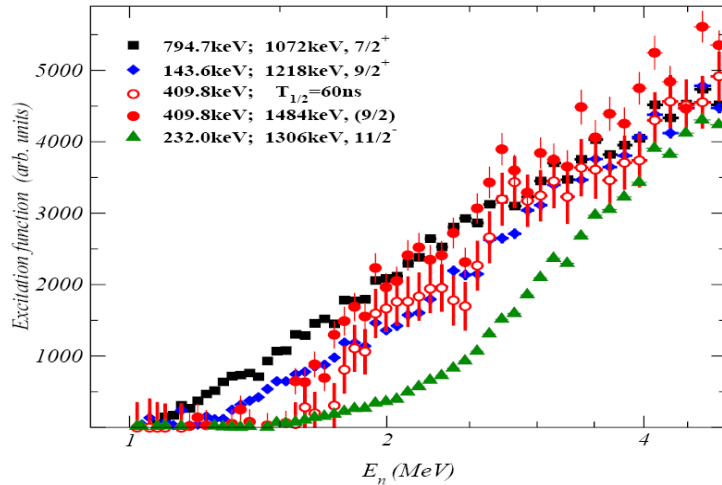
Structure in odd Thallium isotopes

$9/2^-$ isomers from the odd proton in the $h_{9/2}$ orbital



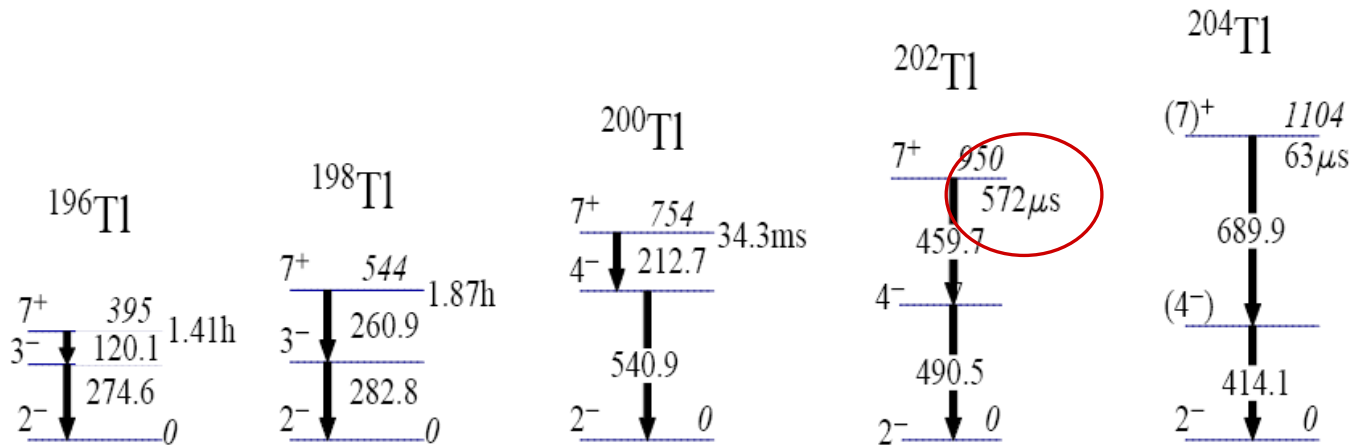
----- G. E. Arenas Peris and P. Federman, Phys. Rev. C 38, 493 (1988)

Half-life corrected excitation function of 408.9 keV

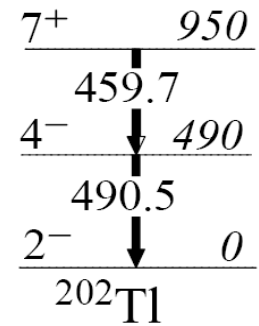
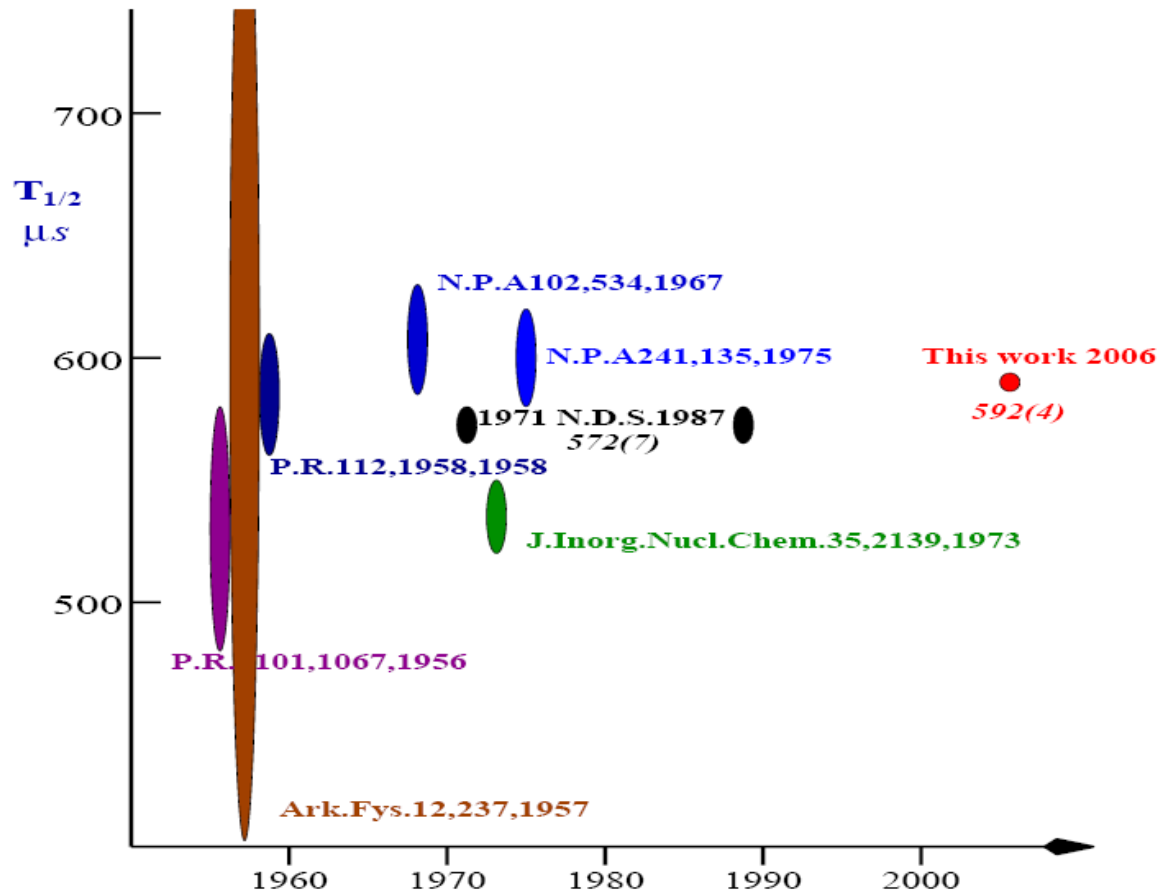


Structure in even Thallium isotopes

7^+ isomers from the **odd proton** in the $s_{1/2}$ orbital
and the **odd neutron** in the $i_{13/2}$ orbital



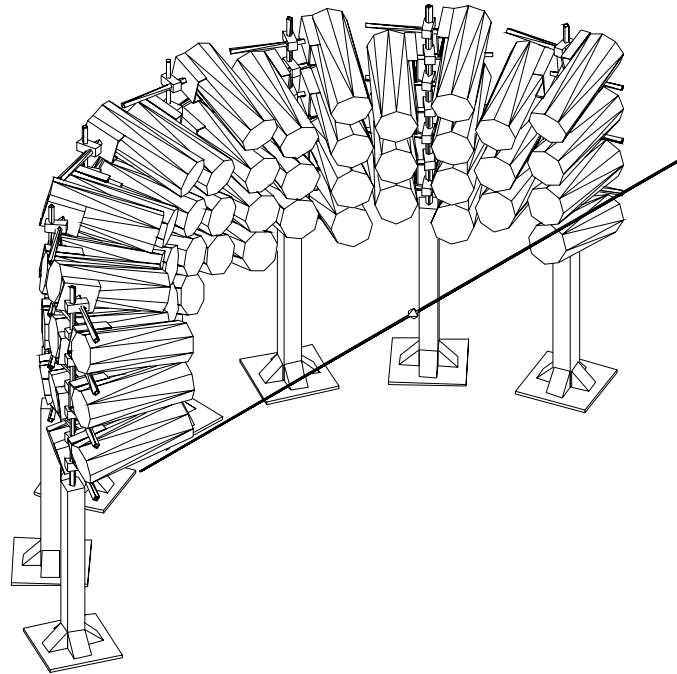
History of the ^{202}Tl 7⁺ isomer half-life



Some conclusions on states in Tl-isotopes

- Excited states in $^{199-203}\text{Tl}$ are studied with GEANIE using the $^{203}\text{Tl}(n,x\gamma)$ reaction
- Gamma excitation functions are measured from beam-on data
- Half-lives of isomers are determined between beam macropulses
- 1484-keV state is a candidate for the $9/2^-$ isomer in ^{203}Tl ; half-life probably in the nanosecond range
- Half-life of the 7^+ isomer of ^{202}Tl [592(4) μs in this work vs. 572(7) in the 1989 evaluation]
- Results of life-time measurements in $^{199-201}\text{Tl}$ in agreement with previous values

FIGARO ($n, xn+\gamma$)



Present and future experiments at FIGARO/WNR: neutron-emission spectra and $\bar{\nu}$ in fission

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

Fission Chamber in beam

- $^{239}\text{Pu}(n,f)$: E_{fn} , $\bar{\nu}$ In progress
- $^{235}\text{U}(n,f)$: E_{fgamma} R. Nelson, in progress
- $^{237}\text{Np}(n,f)$: E_{fn} , $\bar{\nu}$ Data being analyzed by CEA

Gamma-ray trigger (HPGe or BaF₂)

- ^{nat}Ba , ^{nat}Sr , ^{56}Fe In progress

Contact:
Bob Haight

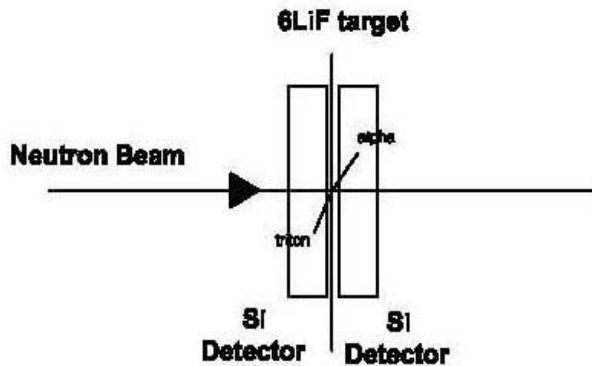
N,Z = (n,charged particle) cross sections
-- studied in two ways

${}^6\text{Li}(n,t)\alpha$ measurements at WNR: cross section and angular distribution

- The ${}^6\text{Li}(n,t)\alpha$ reaction cross section in the few MeV region has relatively large experimental uncertainties
- Specifically, we are measuring
 - 1) The reaction cross section for $0.1 < E_n < 10$ MeV using a Si detector “sandwich” technique
 - 2) The angular distribution of tritons, to improve the theoretical modeling and to identify the spins and parities of the (unbound) states in the MeV region
- Results will be ready in early 2007

Contacts:
Matt Devlin
Terry Taddeucci

${}^6\text{Li}(n,t)\alpha$ cross section measurement with Si detectors: the method

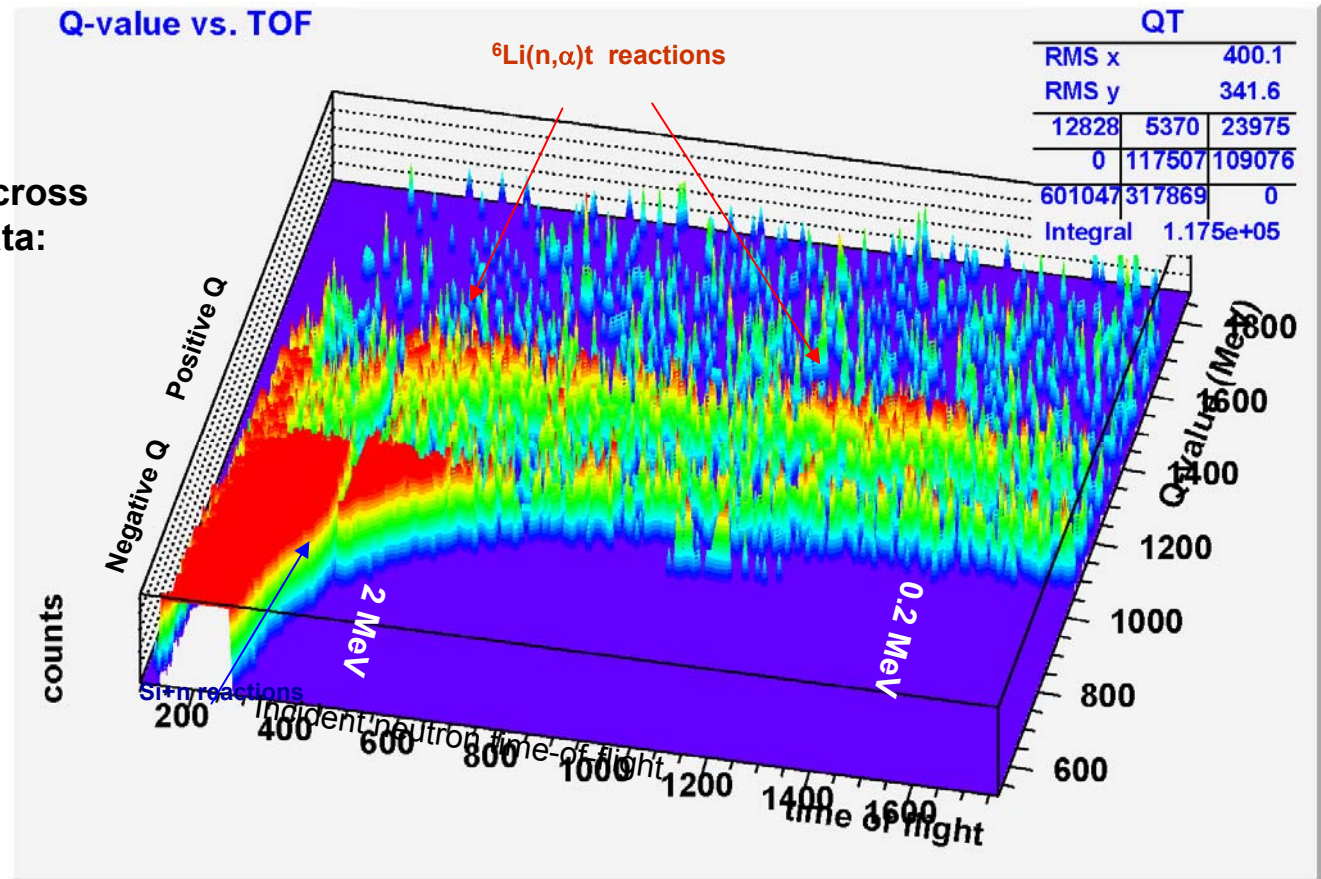


- Si detectors are selected to be thick enough to stop tritons and alphas up to $E_n = 25$ MeV
- For higher neutron energies both products go forward
- Si detectors obtained from ORTEC, Canberra
- Systematic errors will include angular distribution effects (some of the solid angle is not covered) and downscattering of neutrons in the Si upstream

${}^6\text{Li}(n,\alpha){}^3\text{H}$ data at 40m and 7.2 μs show good separation of reaction from background

Preliminary cross section data:

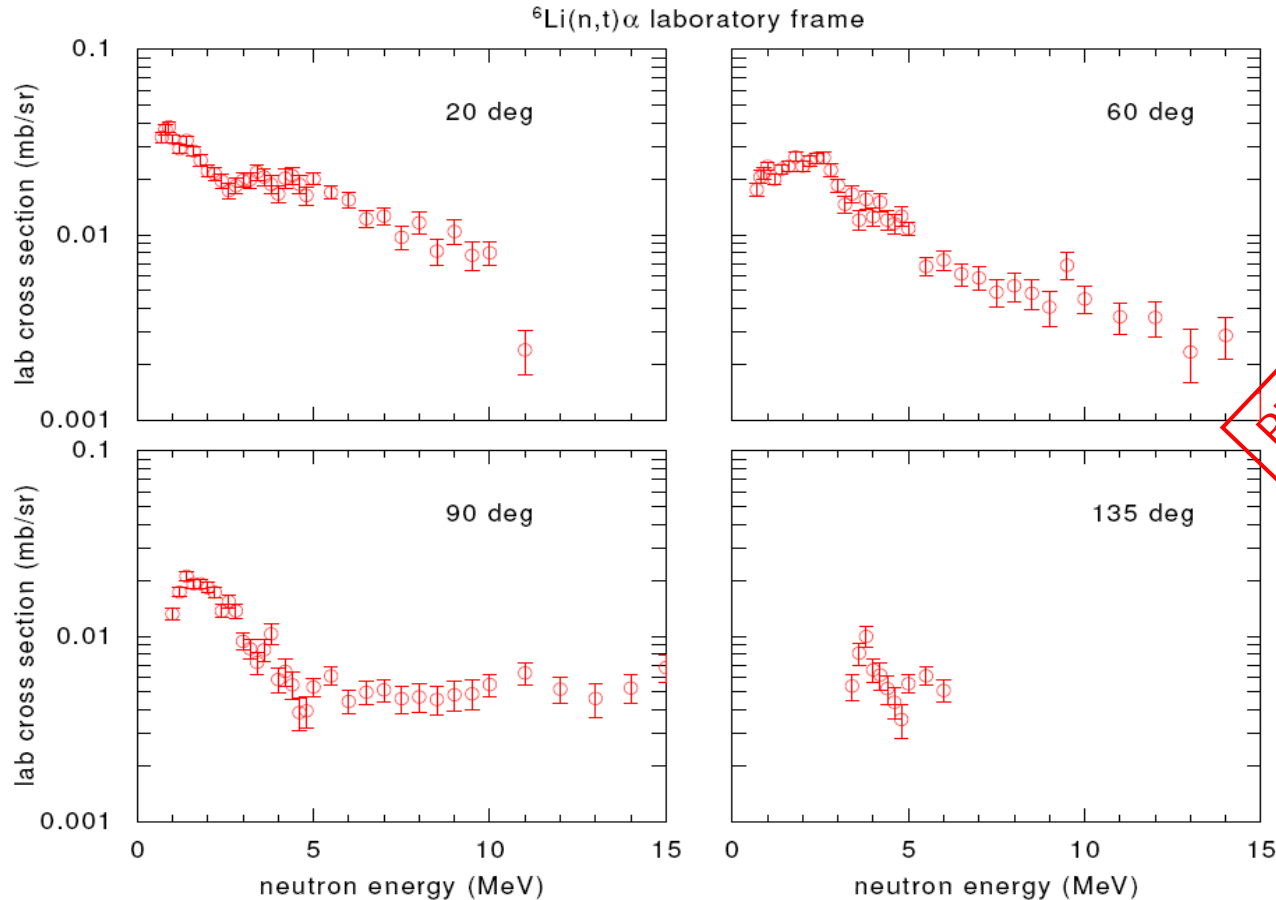
${}^6\text{Li}(n,\alpha)t$ reactions are the band with positive Q-values



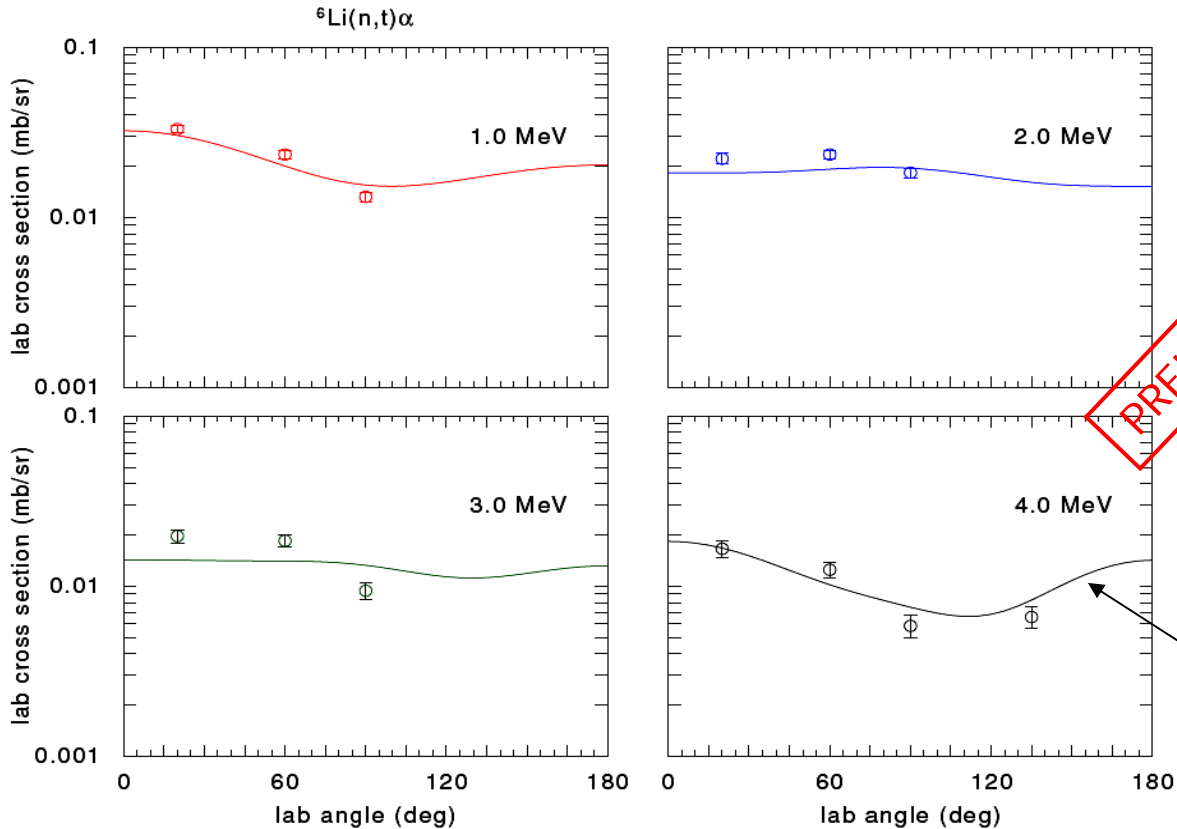
N,Z double-differential cross sections are studied with “standard” detector telescopes



${}^6\text{Li}(n,\alpha)$ preliminary results for differential cross section are very promising

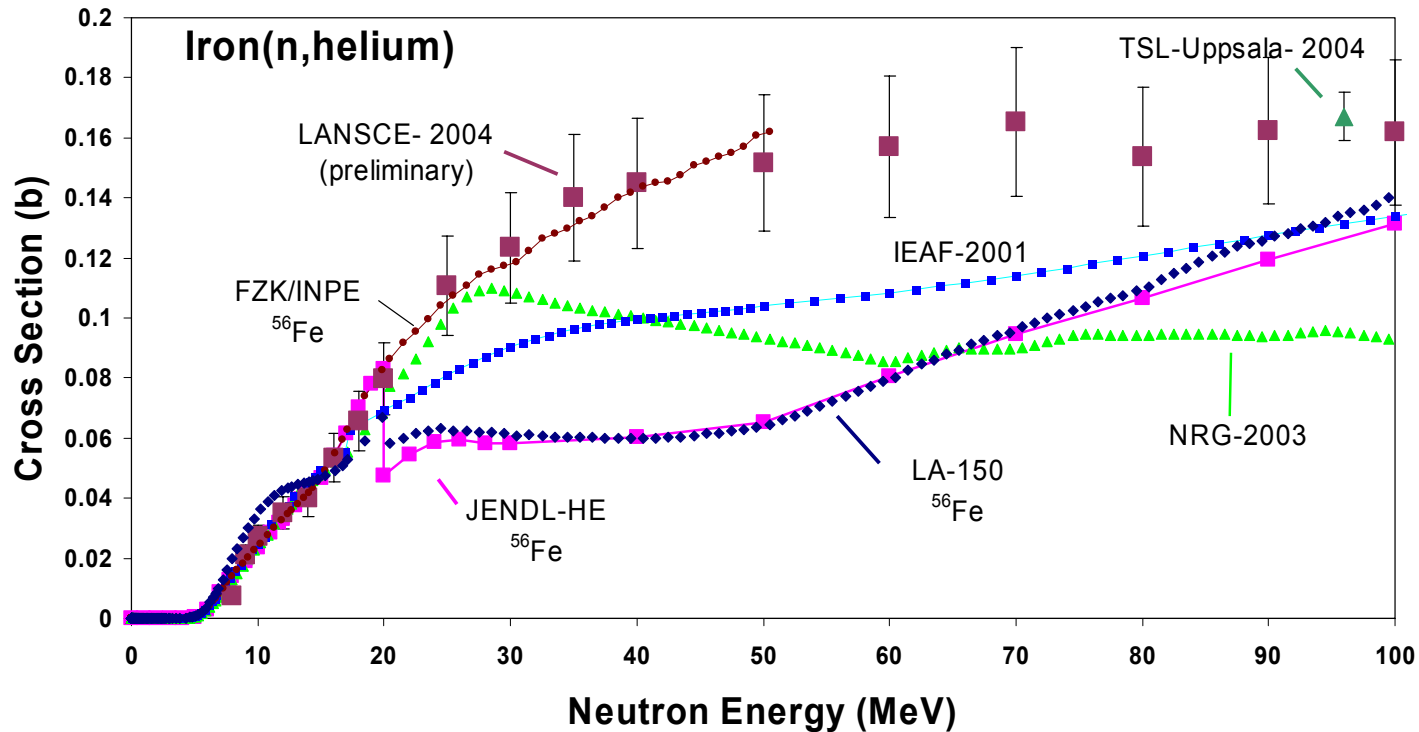


Angular distribution data will aid R-Matrix analysis



R-Matrix
calculation
(previous
parameters)

We measure hydrogen and helium production cross sections for the Advanced Fuel Cycle Initiative



These data differentiate among evaluations

Contact:
Bob Haight

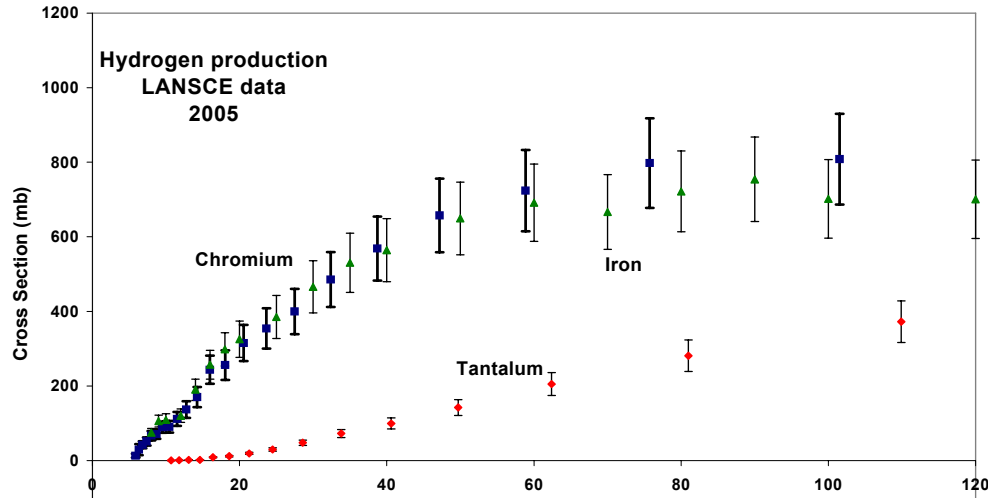
Plans for future hydrogen and helium production cross sections for the Advanced Fuel Cycle Initiative

$$1 \text{ MeV} < E_n < 100 \text{ MeV}$$

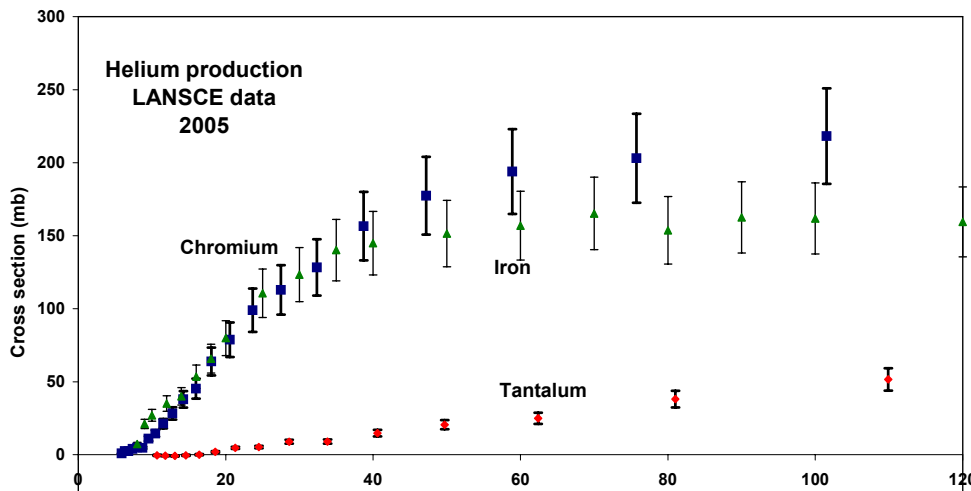
- Zr(n,xp) and (n,x α) -- nearly completed
- Mo(n,xp) and (n,x α) -- planned

Goal is to determine, e.g. helium production / dpa for accelerated radiation damage analysis

Previous data are for Iron, Chromium, and Tantalum

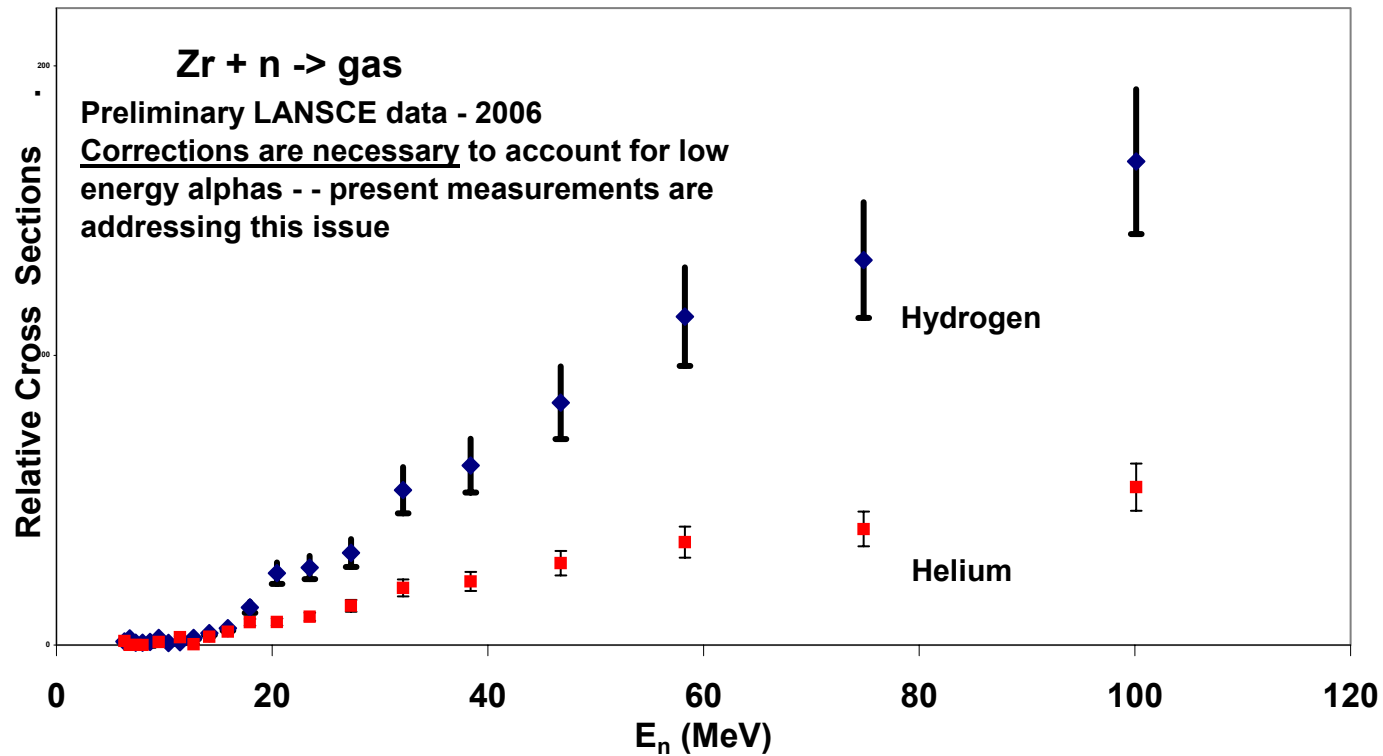


Hydrogen



Helium

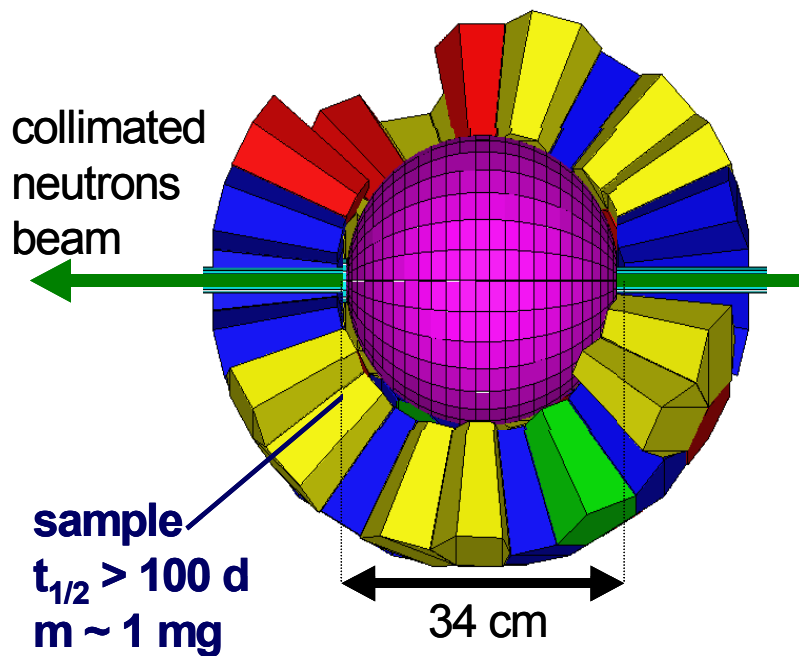
Partial data have been taken for Zirconium



DANCE (n, γ)



Detector for Advanced Neutron Capture Experiments - DANCE



neutrons:

- spallation source
- thermal .. 500 keV
- 20 m flight path
- $3 \cdot 10^5 \text{ n/s/cm}^2/\text{decade}$

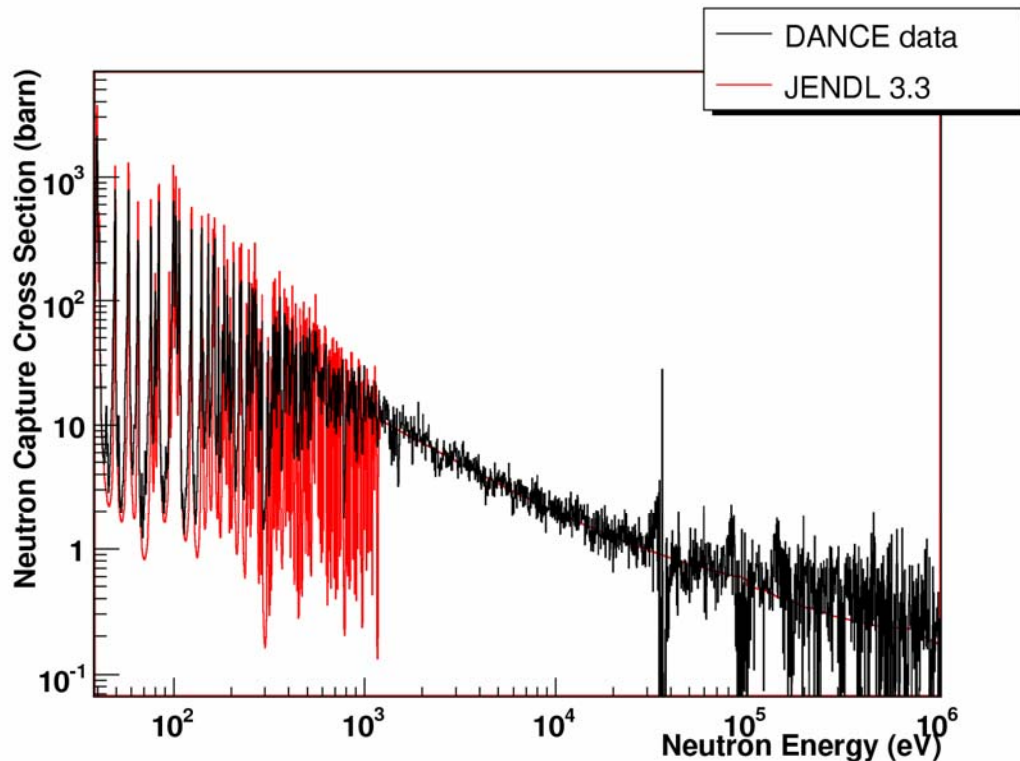
γ -Detector:

- 160 BaF_2 crystals
- 4 different shapes
- $R_i=17 \text{ cm}$, $R_a=32 \text{ cm}$
- 7 cm ${}^6\text{LiH}$ inside
- $\epsilon_\gamma \approx 90 \%$
- $\epsilon_{\text{casc}} \approx 98 \%$

Contacts:

John Ullmann
Rene Reifarth
Bob Rundberg

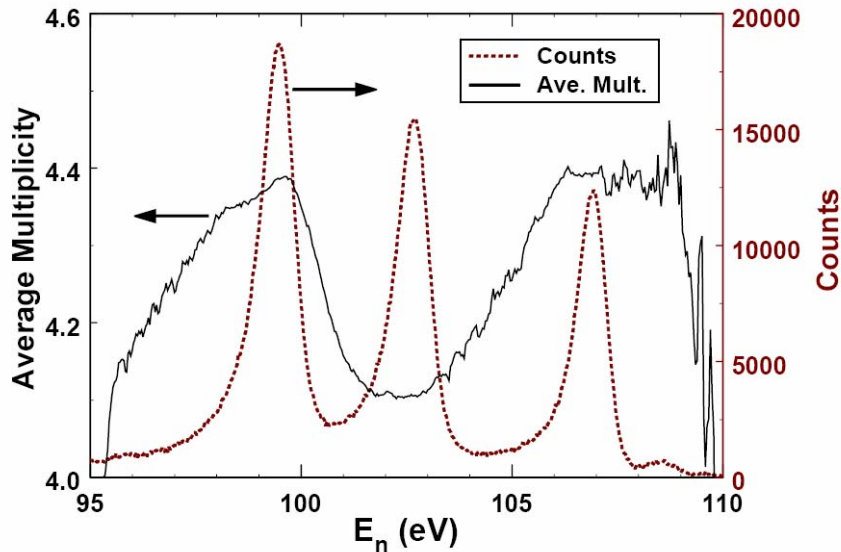
$^{147}\text{Sm}(n,\gamma)$, 10 mg



Comparison of preliminary data from a 10 mg ^{147}Sm sample at DANCE (**black**) and the JENDL-3.3 evaluation (**red**) around the low keV region. The evaluated data could be confirmed over a broad energy range.

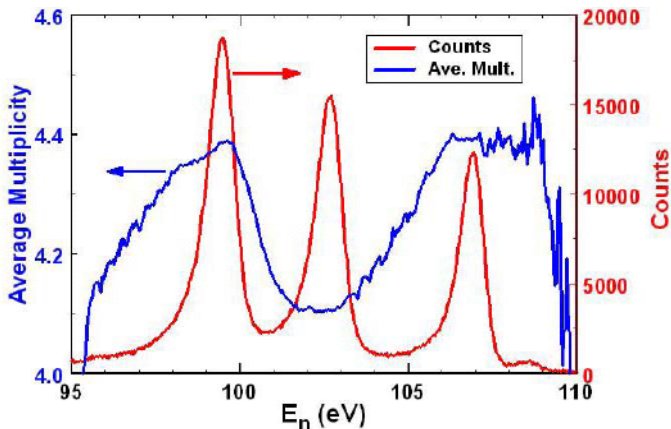
Contact:
Paul Koehler
(ORNL)

$^{147}\text{Sm}(n,\gamma)$, 10 mg

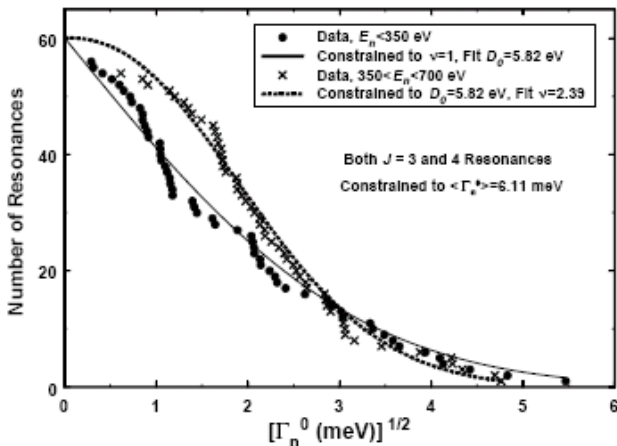


Spin-assignment using the “average multiplicity” technique. Provided the nuclear structure is favorable, DANCE is a very powerful tool for this technique – thanks to the high segmentation.

Spin Assignments for $^{147}\text{Sm} + n$



$\langle M \rangle$ and Counts for several ^{147}Sm resonances



Spin assignments made for 140 resonances < 1 keV

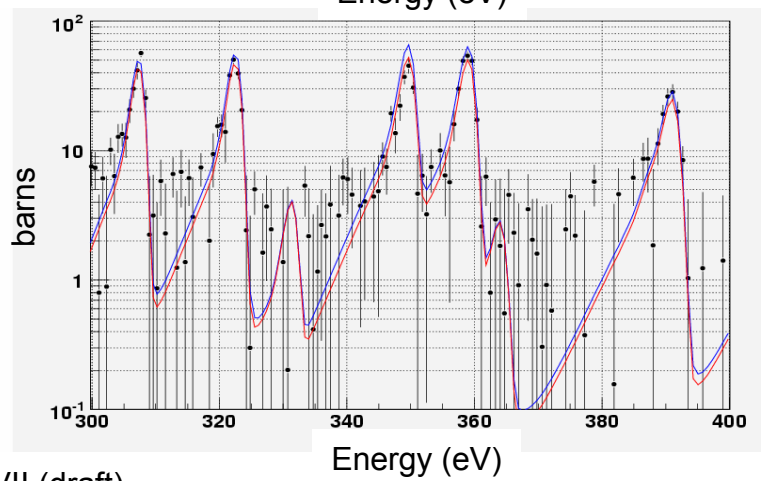
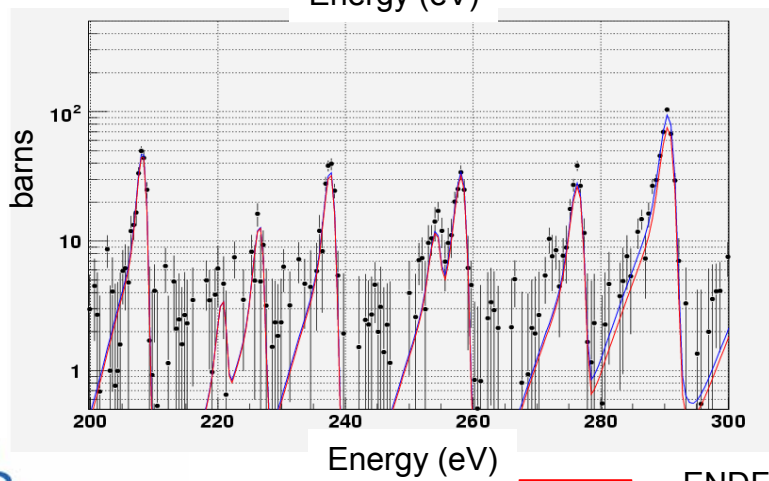
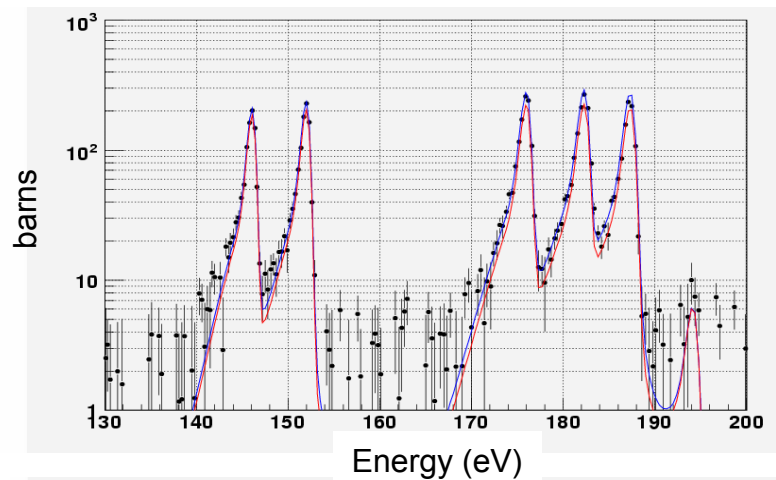
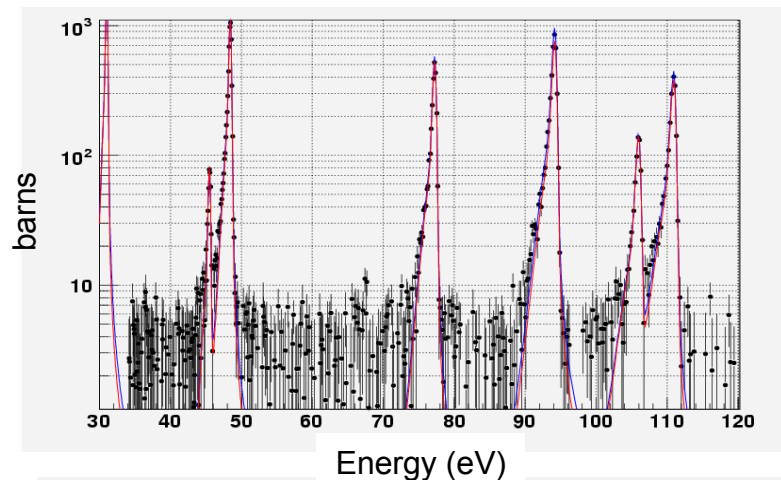
- 34 firm J assignments for previously unassigned
- 8 firm assignments where only tentative assignments
- 14 resonances < 1 keV without firm J
 - (9 < 700 eV)
- 6 firm assignments disagree with Sukhoruchkin
- 6 previously firm resonances shown to be doublets

Actual assignments were made using combinations of various multiplicities rather than $\langle M \rangle$

Non-statistical effects?

- Distribution of $J=3,4$ reduced neutron widths
 - Agree with each other
 - Disagree with Porter-Thomas
 (Different conclusion from Gledenov and Koehler – incorrect spin assignments)
- Combined $J=3,4$ distribution
 - En < 350 eV follow Porter-Thomas ($\nu=1$)
 - 350 < En < 700 eV, not PT ($\nu=2.39$)
- Is result statistically significant?

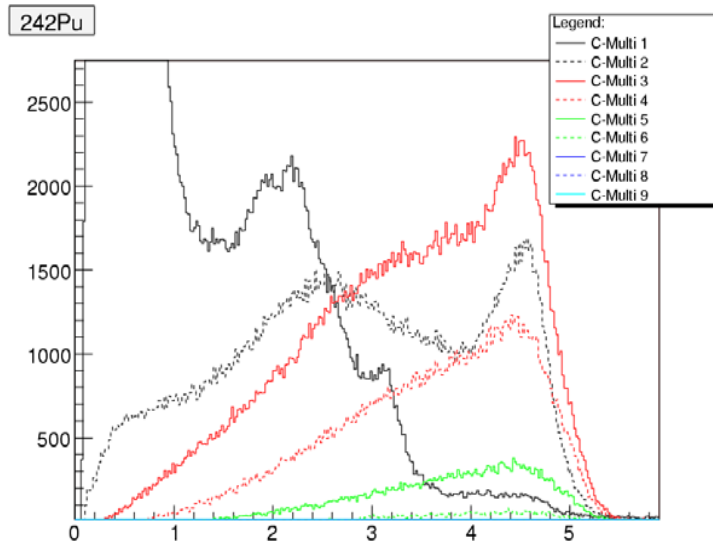
$^{234}\text{U}(n,\gamma)$ with broadened ENDF resonance parameters



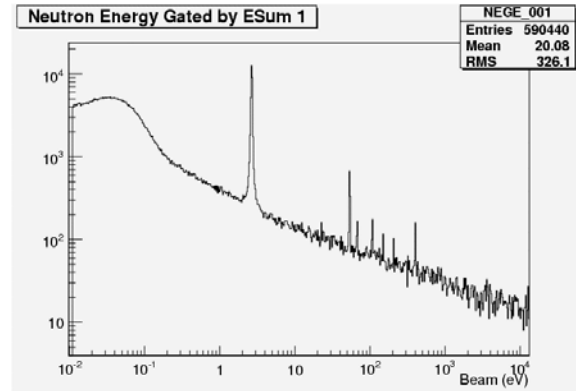
— ENDFB-VII (draft)
— ENDFB-VI

- SAMMY calc w/ "NTOF" res func
- No correction for fission resonances

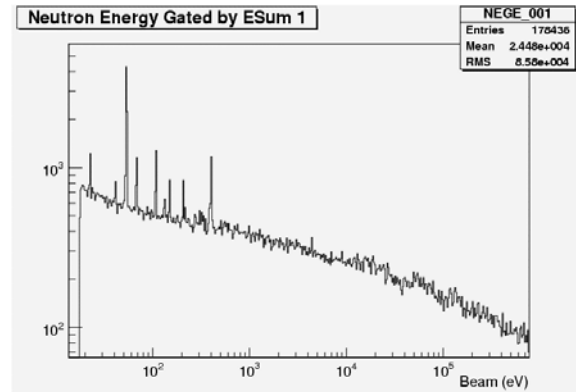
$^{242}\text{Pu}(n,\gamma)$, 0.7 mg



Sum energy (MeV) deposited in the crystal ball, if (n, γ) dominated



Yield as function of neutron energy (eV), gated on Q-value for $^{242}\text{Pu}(n,\gamma)$



Neutron Capture Cross sections of ^{236}U and ^{234}U

Neutron-capture cross sections on U isotope chain are important

Measurements on ^{234}U

- $Q(n,\gamma) = 5.30$ MeV
- $4.0 < E_{\text{sum}} < 5.5$ MeV, $\text{Mult} \geq 3$
- Target: 1.08 mg on 2, 2.5 μm Ti foils
- Normalized to Thermal (100 b) and Barr (absolute)
- Background Subtractions:
 - Target out
 - Fission
 - Gamma scattering (20%)
- Integral of 5.16 eV resonance
 - Thermal normalization = 1770 b-eV
 - Barr normalization = 1740 b-eV
 - ENDF/B-6 = 2830 b-eV
 - ENDF/B-7 = 2738 b-eV

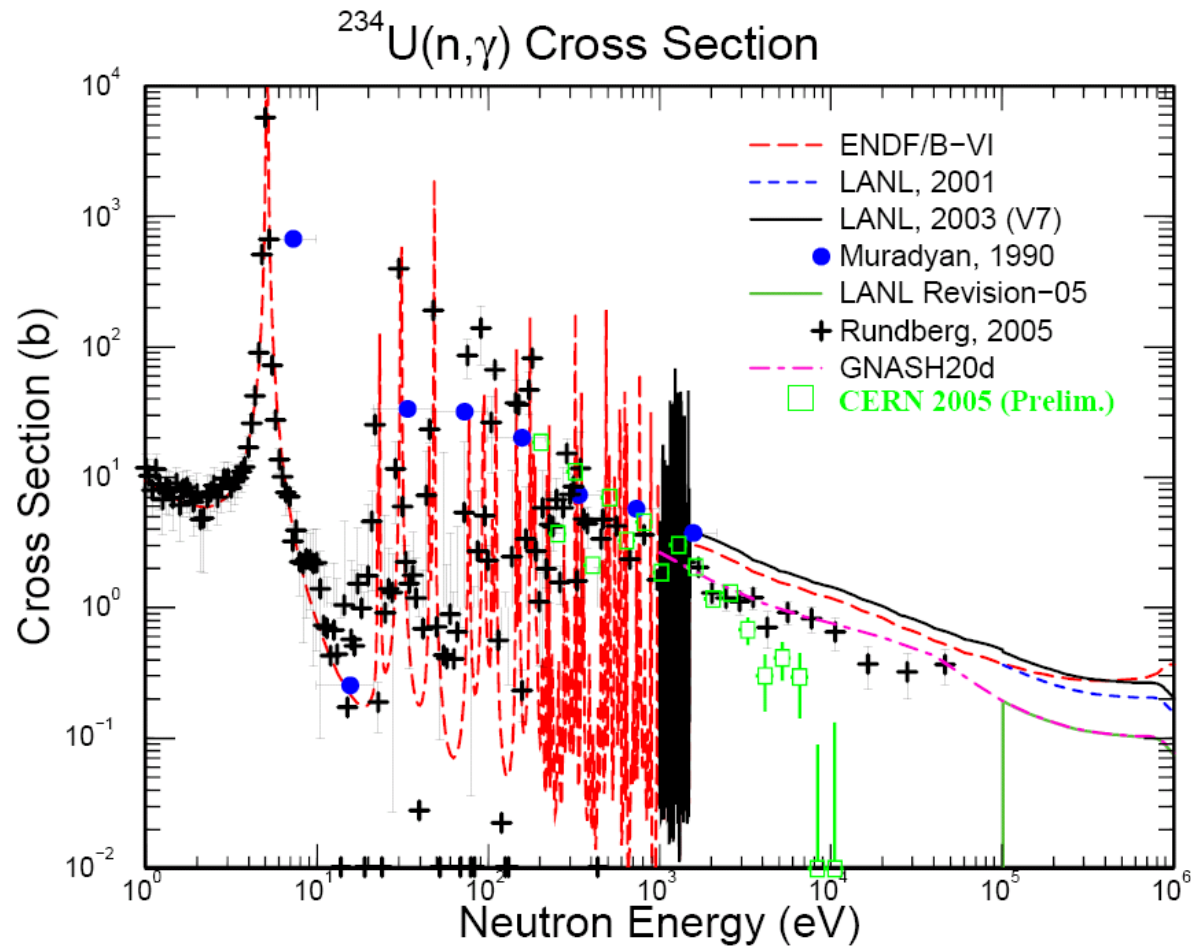
$^{236}\text{U}(n,\gamma)$ and $^{234}\text{U}(n,\gamma)$ not well known

- Several measurements of ^{236}U , but recent measurements above 1 keV differ by factor of 2
- Very few measurements on ^{234}U , this is first high-resolution measurement

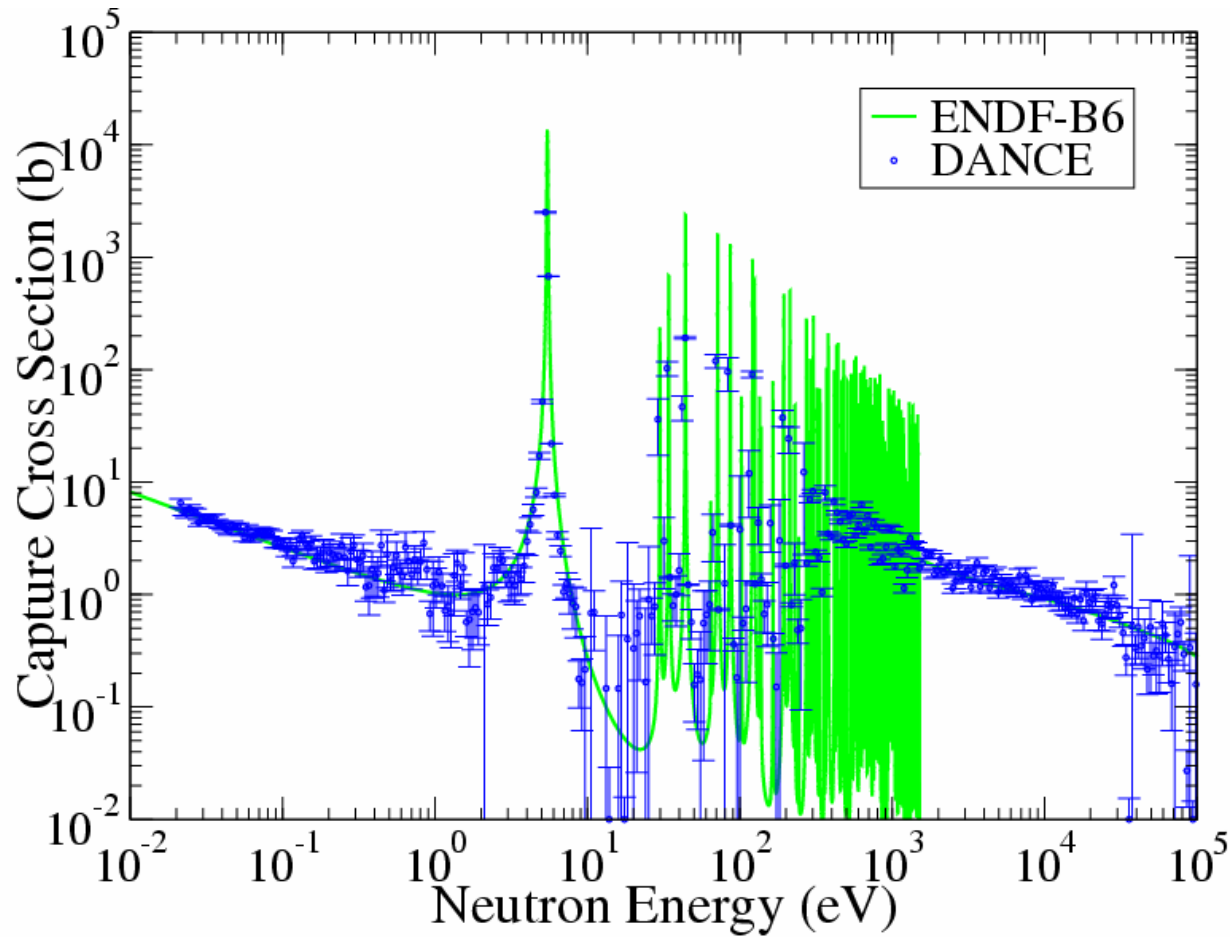
Measurements on ^{236}U

- $Q(n,\gamma) = 5.13$ MeV
- $4.0 < E_{\text{sum}} < 5.5$ MeV, $\text{Mult} \geq 3$
- Target: 0.49 mg on 2, 2.5 μm Ti foils
- Normalized to Thermal (5.1 b) and Barr (absolute)
- Background Subtractions:
 - Target out
 - Fission
 - Gamma scattering (20%)
- Integral of 5.45 eV resonance
 - Thermal normalization, Barr normalization, and ENDF/B-6 all agree

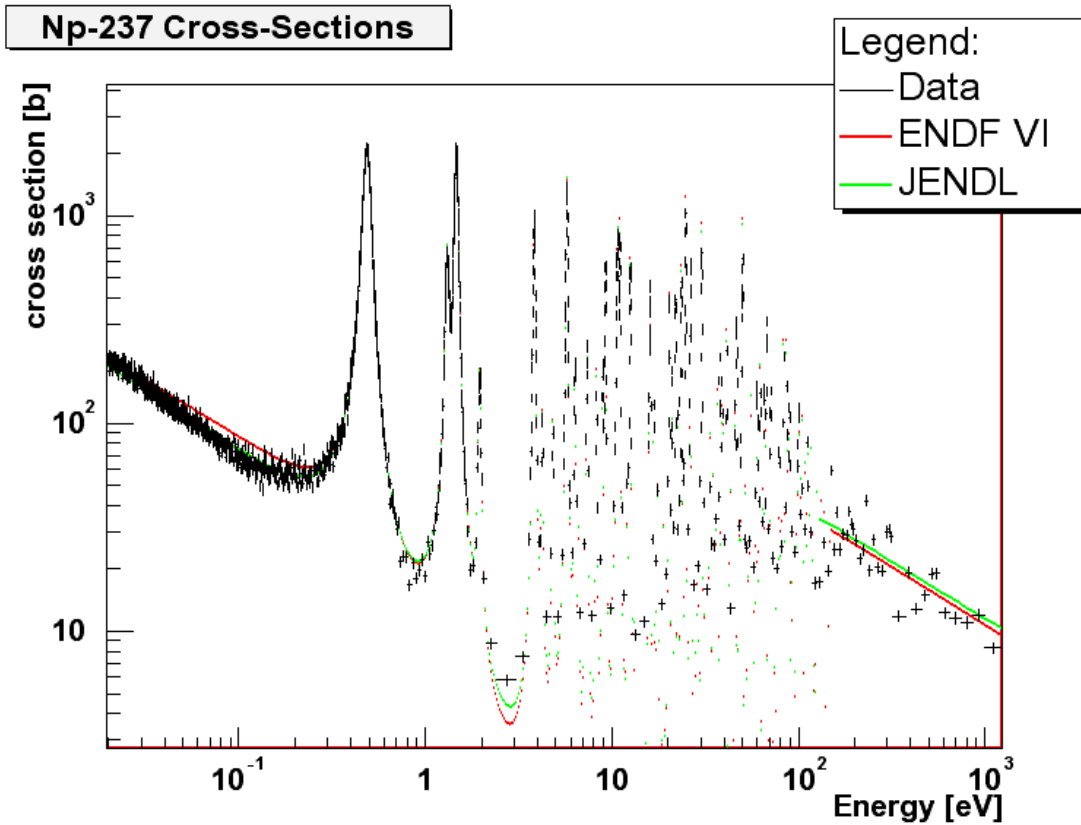
$^{234}\text{U}(n,\gamma)$ Cross Section



$^{236}\text{U}(n,\gamma)$ Cross Section from DANCE

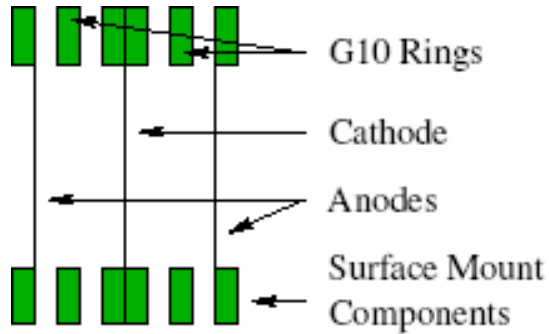


$^{237}\text{Np}(n,\gamma)$

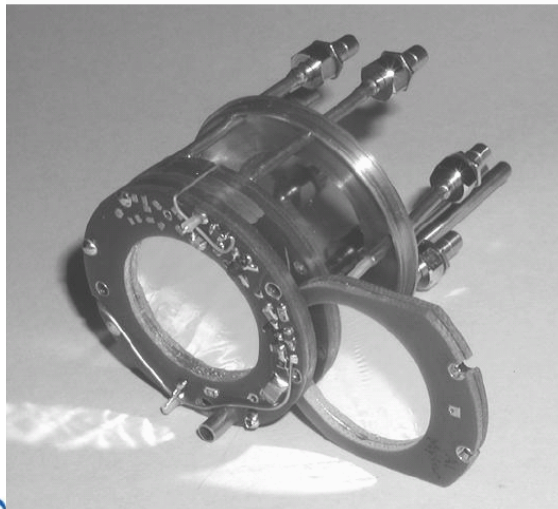


Target: 0.44 mg ^{237}Np in 6.4 mm diameter (1.4 mg/cm^2)
Existing data above 1 keV discrepant

PPAC Detector for Capture and σ_γ/σ_f Measurements



Parallel-Plate
Avalanche
Counter



Close-up of PPAC
showing removable
cathode/target assembly

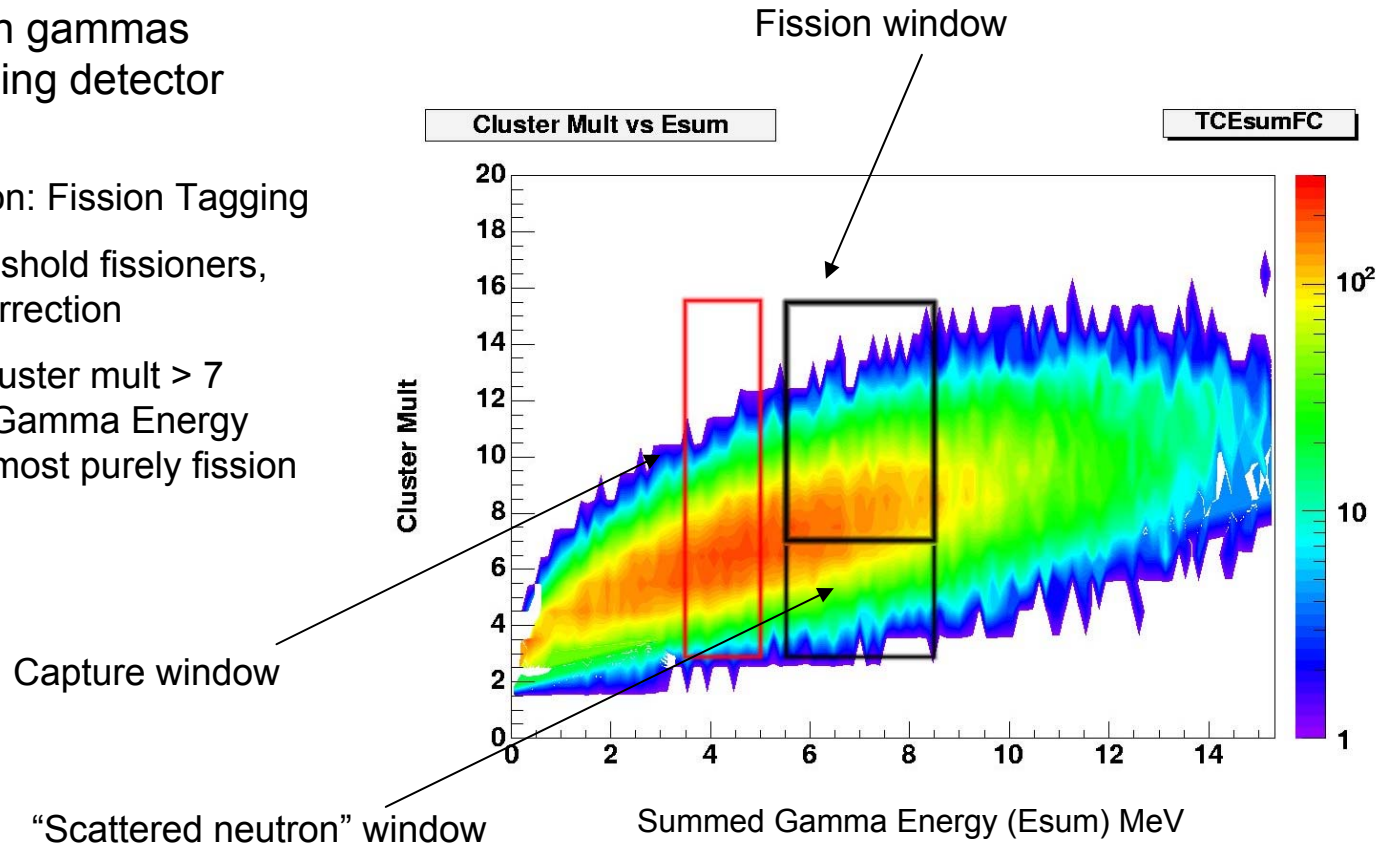


PPAC Assembly with gas lines and signal
cables ready for insertion into DANCE center

Fission correction for fissile nuclides

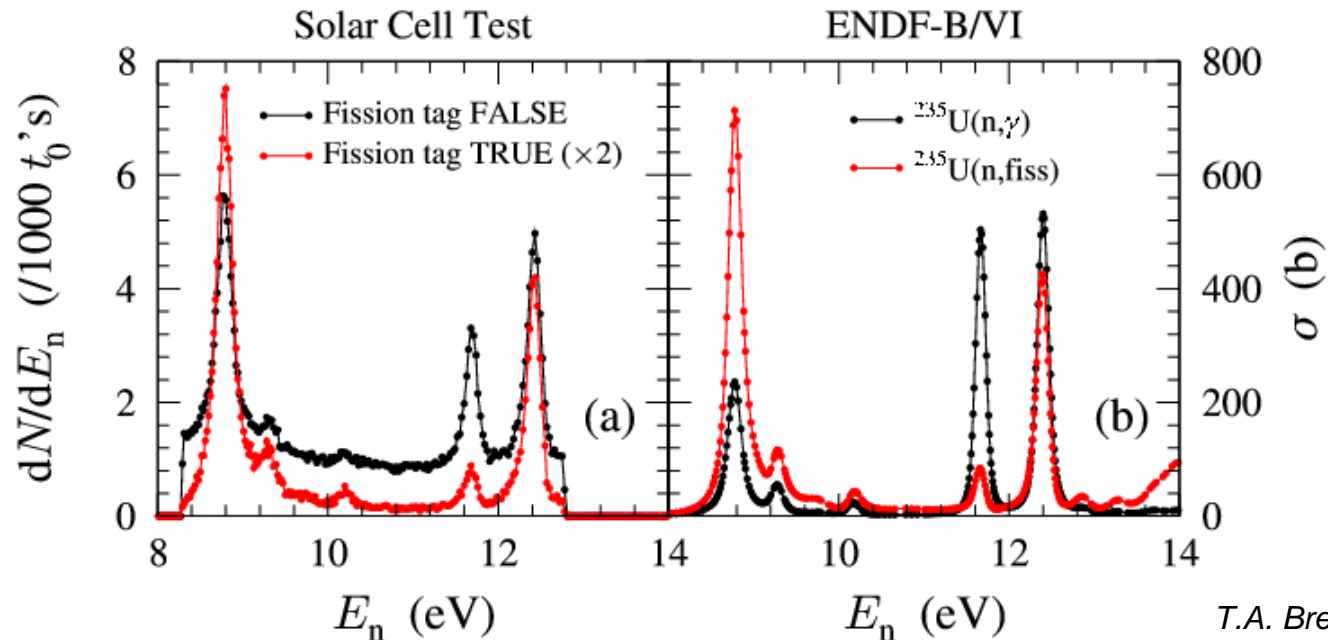
^{235}U fission gammas
Fission-tagging detector

- Best Correction: Fission Tagging
- For many threshold fissioners, can make correction
- Events with cluster mult > 7 and Summed Gamma Energy > 6 MeV are almost purely fission



Background from fission gammas can be determined by normalizing ^{235}U spectrum

Test measurements with a fission-tagging detector

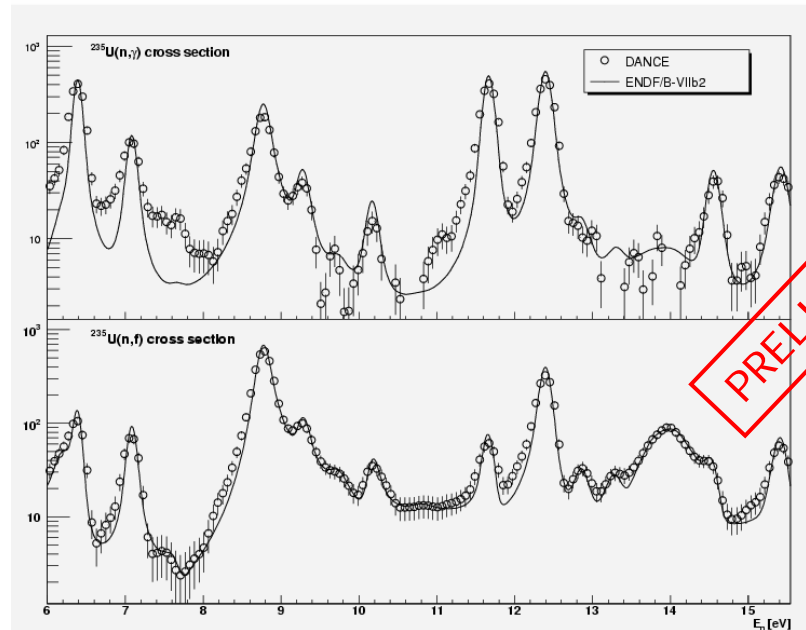


T.A. Bredeweg, et al.

- Study:
 - Fission-to-capture ratios (“alpha”)
 - Gamma emission following fission
- “Proof-of-principle” experiment used “thin” ^{235}U deposit on silicon solar cell (T. Ethvignot, et al.)

▪ Present: Thin gas fission chamber -- PPAC

PPAC Detector for Capture and σ_γ/σ_f Measurements



- Target: 460 μg ^{235}U (99.89%) in 0.7 cm deposit (1.2 mg/cm^2) electrodeposited on metalized mylar(flashed with 0.25 μg Ti on deposit side, 0.10 μg on other side)
- (n, γ) data has $5.5 < E_{\text{sum}}(\text{MeV}) < 7.5$, Multiplicity ≥ 4
- PPAC fission tag has 78% efficiency
- (n, γ) corrected for fission by subtracting 0.22 X fission spectrum
- Approximate normalization to ENDF/B-VI resonances

Analysis of DANCE Data is in Progress on Many Nuclides

$^{94,95}\text{Mo}$ (S. Sheets, NC State Univ.)

^{143}Nd , ^{149}Sm (P. Koehler, ORNL)

$^{152,154,157,160}\text{Gd}$ (W. Parker, Livermore)

$^{151,153}\text{Eu}$ (U. Agvanluvsaan, Livermore)

^{151}Sm (R. Reifarh, Los Alamos)

$^{203,205}\text{Tl}$ (A. Couture, Los Alamos)

^{235}U PPAC (T. Bredeweg, M. Jandel, Los Alamos)

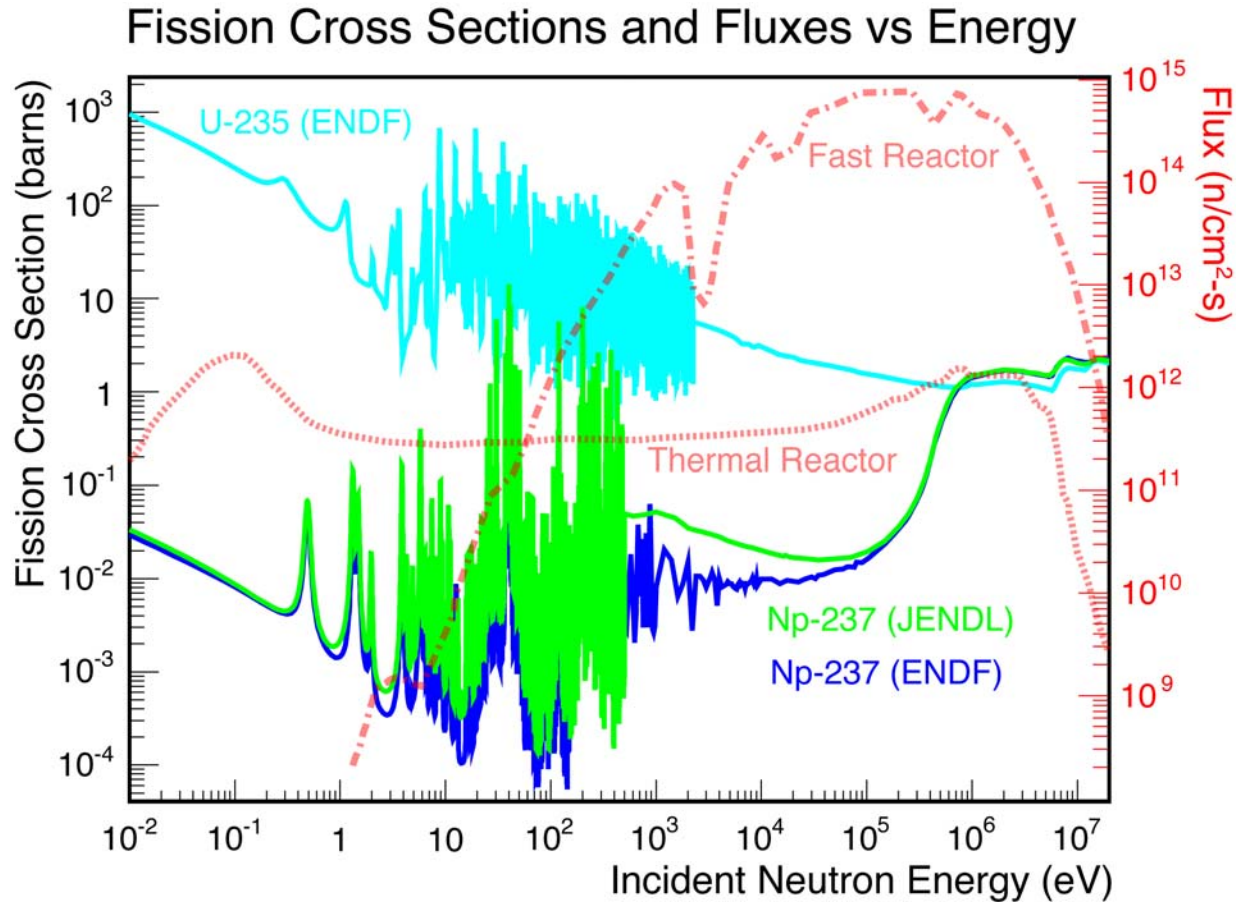
$^{240,242}\text{Pu}$ (A. Couture, R. Reifarh, Los Alamos)

$^{241,243}\text{Am}$ (T. Bredeweg, M. Jandel, Los Alamos)

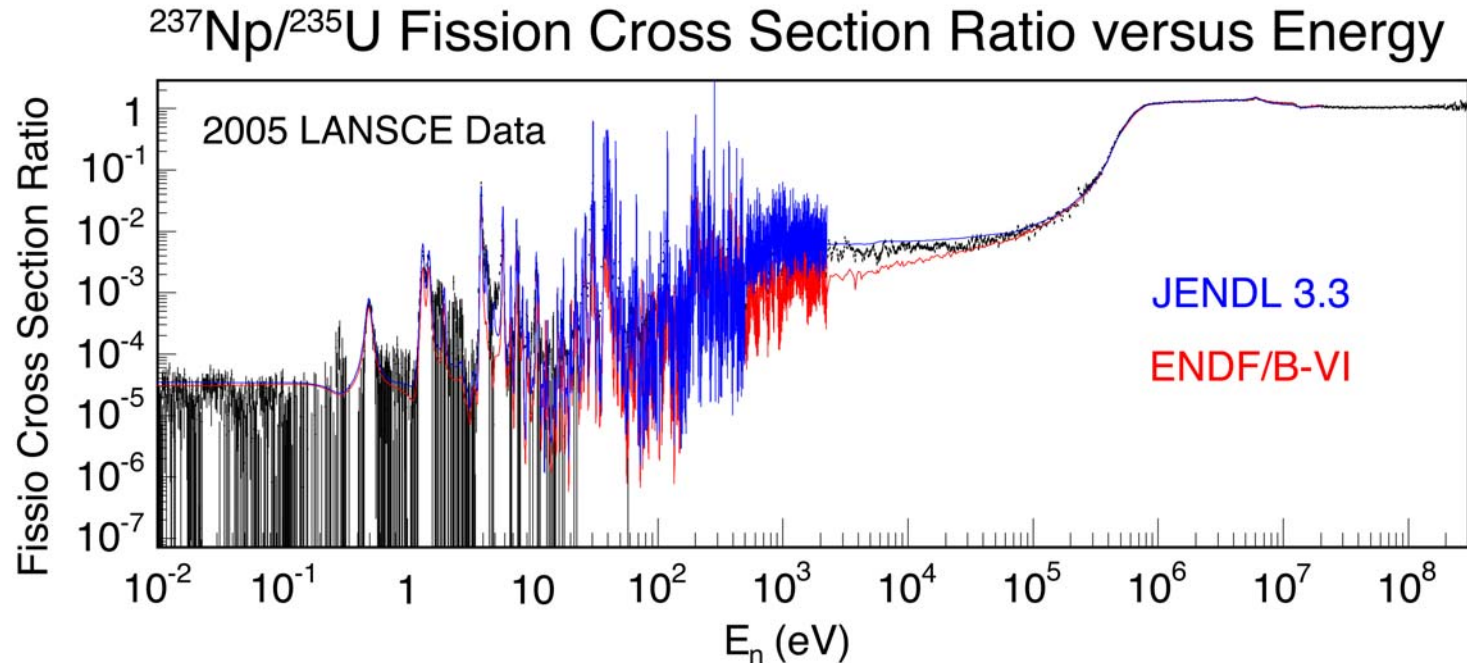
$^{242\text{m}}\text{Am}$ PPAC (R. Macri, Livermore, M. Jandel, Los Alamos)

Fission Cross Sections

Discrepancies in $^{237}\text{Np}(n,f)$ exist in “fast” region between major nuclear data libraries



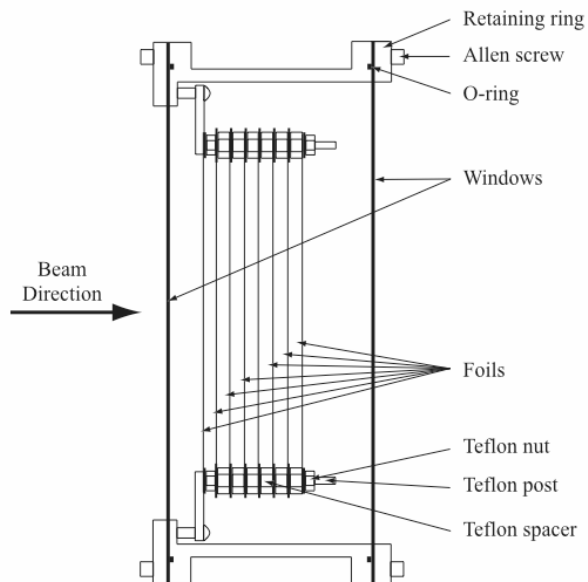
$^{237}\text{Np}(n,f)$ is the first completed measurement



**Analysis includes detailed
covariances (with T-16)**

**Contacts:
Tony Hill
Fredrik Tovesson**

Parallel-plate fission ionization chamber and gridded ion chamber



Parallel plate ionization chamber (PPIC)

- Commonly used for flux monitoring
- Detects on fission fragment per even
- Holds up to 4 samples



Double gridded ionization chamber

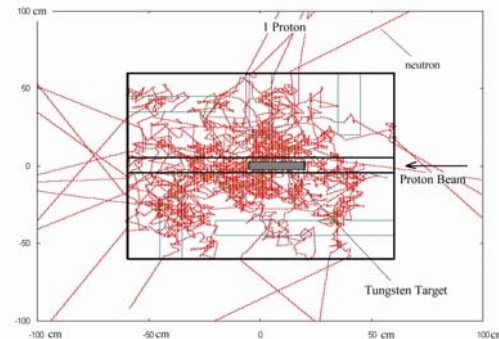
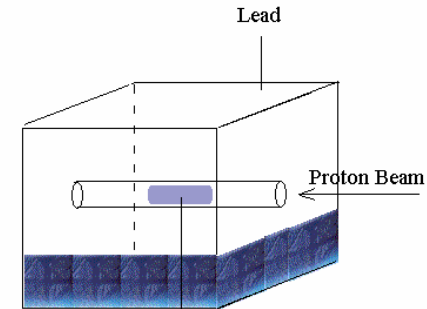
- On loan from IRMM, Geel.
- Detects both fission fragments, improving alpha-separation for highly active targets

LANSCCE fission measurements in the future

- Low energy (Lujan) data have been collected/calibrated for
 - U233, Pu239, Pu240, Pu242 thermal $< E_n < 200$ keV
- High energy (WNR) data will be taken this run cycle on
 - U233, Pu239, Pu240 and Pu242 100 keV $< E_n < 200$ MeV
- Complete analyses by summer `07
- Actinides of interest for future fission measurements:
 - Am241, Am242, Am242m, Cm242, Cm243, Np238, Pu238

Fission Cross Sections On Very Small Samples

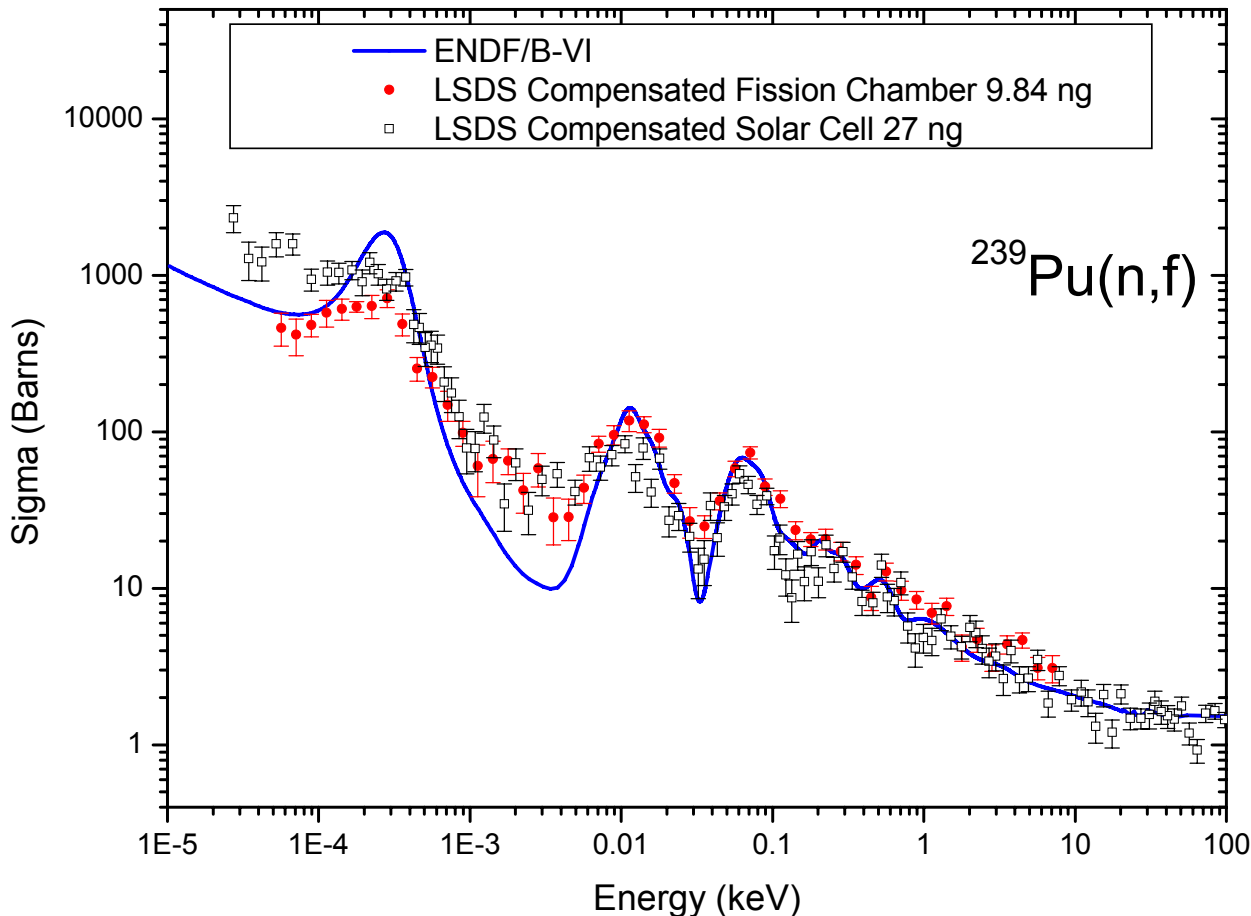
A Lead Slowing-Down Spectrometer is under development, driven by 800 MeV protons from the PSR



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

**Contact:
Bob Haight**

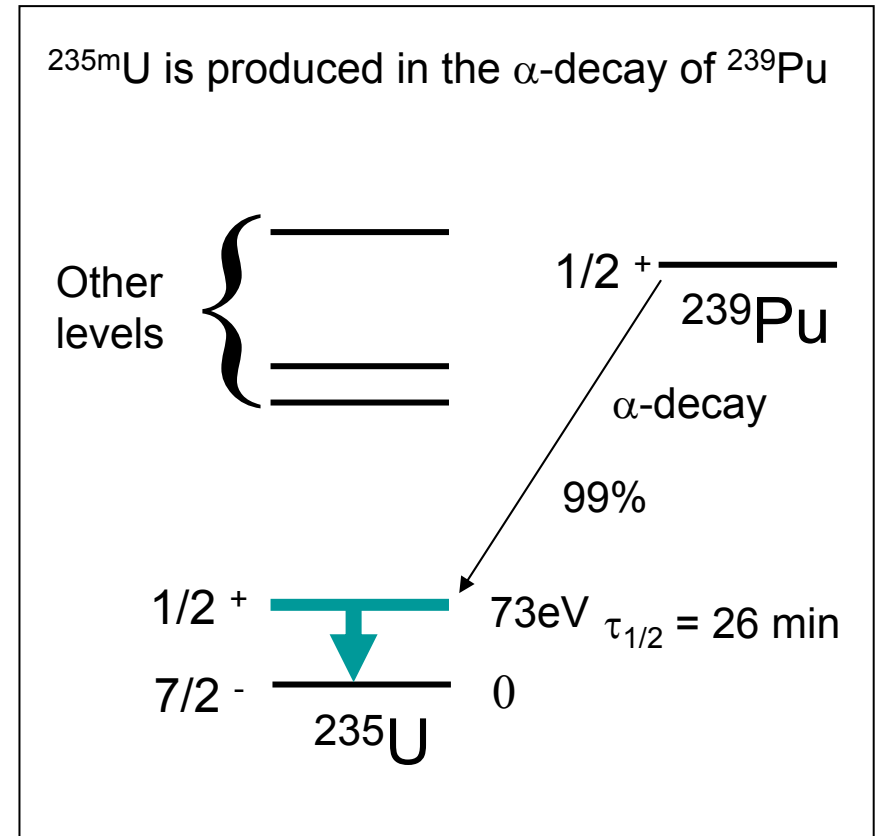
With the LSDS, we measured the neutron-induced fission cross section on ^{239}Pu with sub- μg samples



- **Sample size of 9.87 ng can be studied**
- **Good results up to 100 keV**
- **Still plan to measure fission cross section of $^{235\text{m}}\text{U}$ (26 minutes) after solving chemistry challenge**

First excited (isomeric) state of ^{235}U is produced in decay of ^{239}Pu

- $^{235\text{m}}\text{U}$
 - 26 min half-life
 - 73eV
 - Decays by internal conversion
 - 99% of ^{239}Pu decays populate $^{235\text{m}}\text{U}$
 - 5 gm of Pu will produce 10ng of $^{235\text{m}}\text{U}$
- Fast extraction of $^{235\text{m}}\text{U}$ will be required
- To measure this small cross section, it is necessary to increase the neutron flux by using a lead-slowing down spectrometer (LSDS)



Developments at LANSCE

- **Technical issues**
 - High power amplifier tube availability and quality (Burle 7835) → **problem solved for the present**
 - LANSCE-Refurbishment (“LANSCE-R”)
- **Funding problems (LANL contract → \$175M shortfall for this FY) → less running for LANSCE**
 - Electricity costs
- **Personnel changes**
 - Paul Lisowski (former LANSCE Director) to Washington to head GNEP
 - Kurt Schoenberg acting LANSCE Director
- **Reorganization of Lab and LANSCE**

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
- N,Z – LANL, Ohio U
- DANCE – LANL, LLNL, ORNL, INL, Colorado School of Mines, FZK Karlsruhe
- LSDS – LANL, LLNL, Bruyères-le-Châtel, RPI
- Fission – LANL, IRMM, LLNL, INL
- Others – MIT, Kentucky, Kyushu, Harvard,...