Needs for Nuclear Reactions on Actinides

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Nuclear Data for National Security

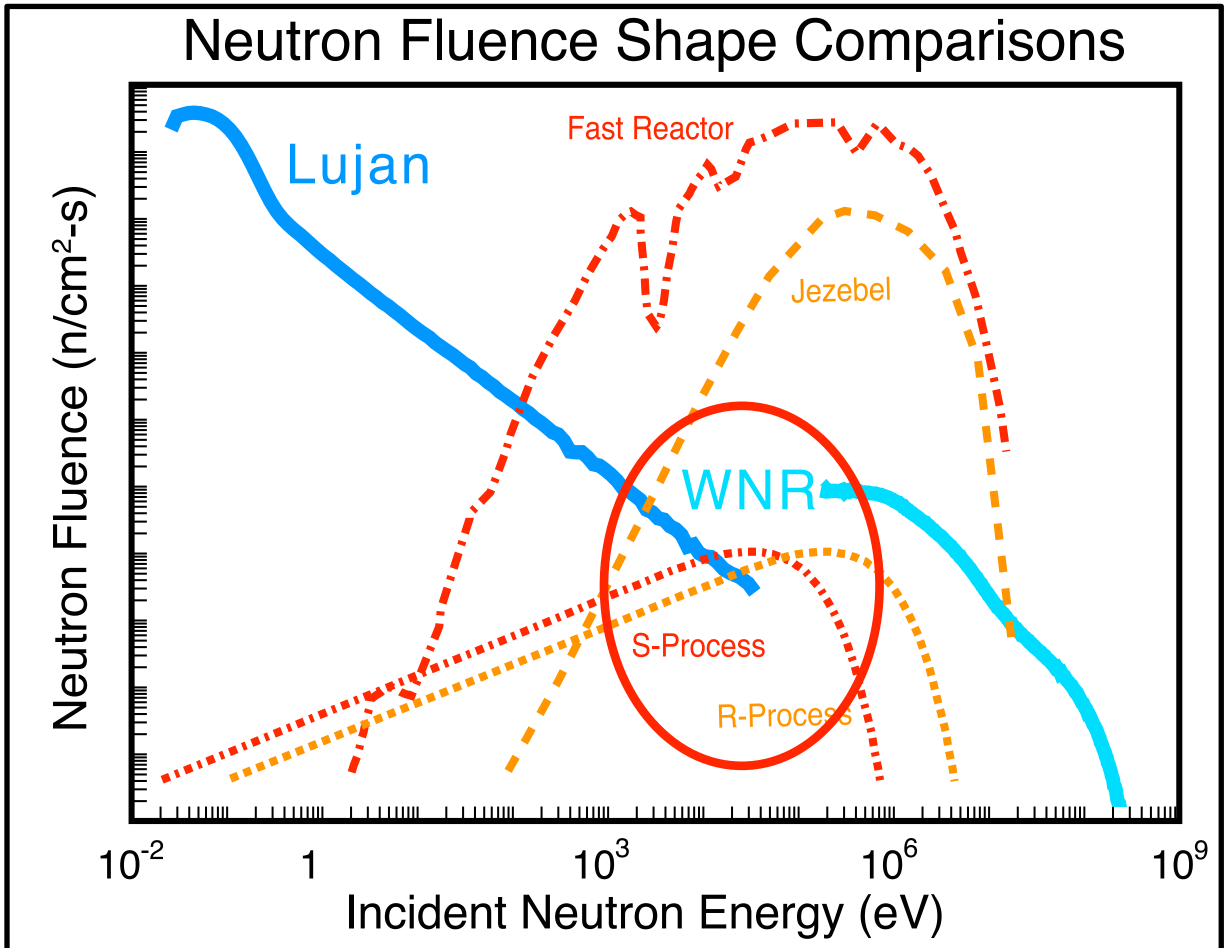
Many important topics will be discussed later:

- Detection
- Forensics
- HEDP opportunities
- Fission product yields; minor actinides & off-stability species, ….

Instead, I will focus here on nuclear criticality & transport, where advances are needed for stewardship (including NDSE diagnostic) & for advanced fast reactors:

- Capture, Inelastic & elastic scattering, fission (esp. PFNS spectra) in "intermediate energy range" – 1-500 keV

Gaps in our understanding of intermediate energy nuclear reactions (~1 keV – 500 keV)



Redesign spallation moderation target to increase fluence in intermediate energy region

Historically we model intermediate energy criticality benchmarks more poorly than simple fast benchmarks

- They involve more scattering, a more complex transport, and are more sensitive to inelastic, elastic, reactions
- Neutron-incident reactions in the 1-500 keV region are often less well understood, *e.g*. actinide capture reactions, inelastic scattering
- Reaction rates (fission, n2n, …) modeled more poorly here too

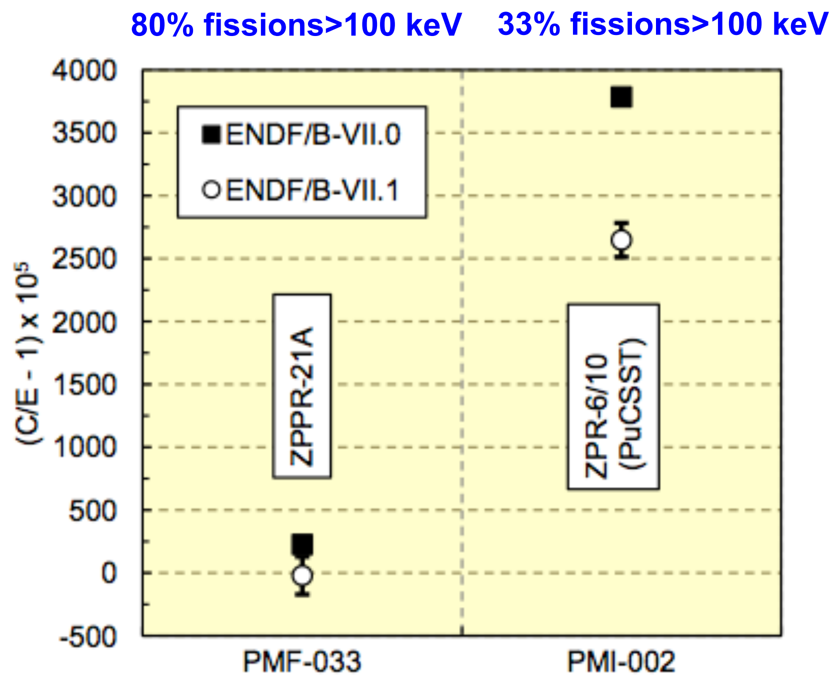


FIG. 24: MCNP Calculations with As-Built models for Pu metal FAST and INTER ZPR/ZPPR Assemblies.

ENDF performs less well in intermediate energy spectra than in fast spectra, for reaction rates Flattop critical assembly

- Fission reaction rates, including threshold fissioners, measured by LANL radiochemists

- Fast discrepancy ~ 6%

- Intermediate discrepancy ~ 12%

TABLE XVI: Measured and Calculated Fission Rate Ratios for Selected Actinides in Flattop-25 by Barr *et al.* [15]. Data for the uranium isotopes and ^{239}Pu are ratioed to ^{235}U(n,f), the remaining results are ratioed to ^{239}Pu(n,f). The measurement location for those data given in the top half of the Table are near the center of the assembly (r=1.11 cm), data given in the bottom half of the Table are from the tamper region (r=13.97 cm). As these data have not been published previously, we also include the measured spectral indices in the second column of this Table. A generic 5% uncertainty is judged appropriate for these data, but the values tabulated are given to the precision used in internal LANL documents.

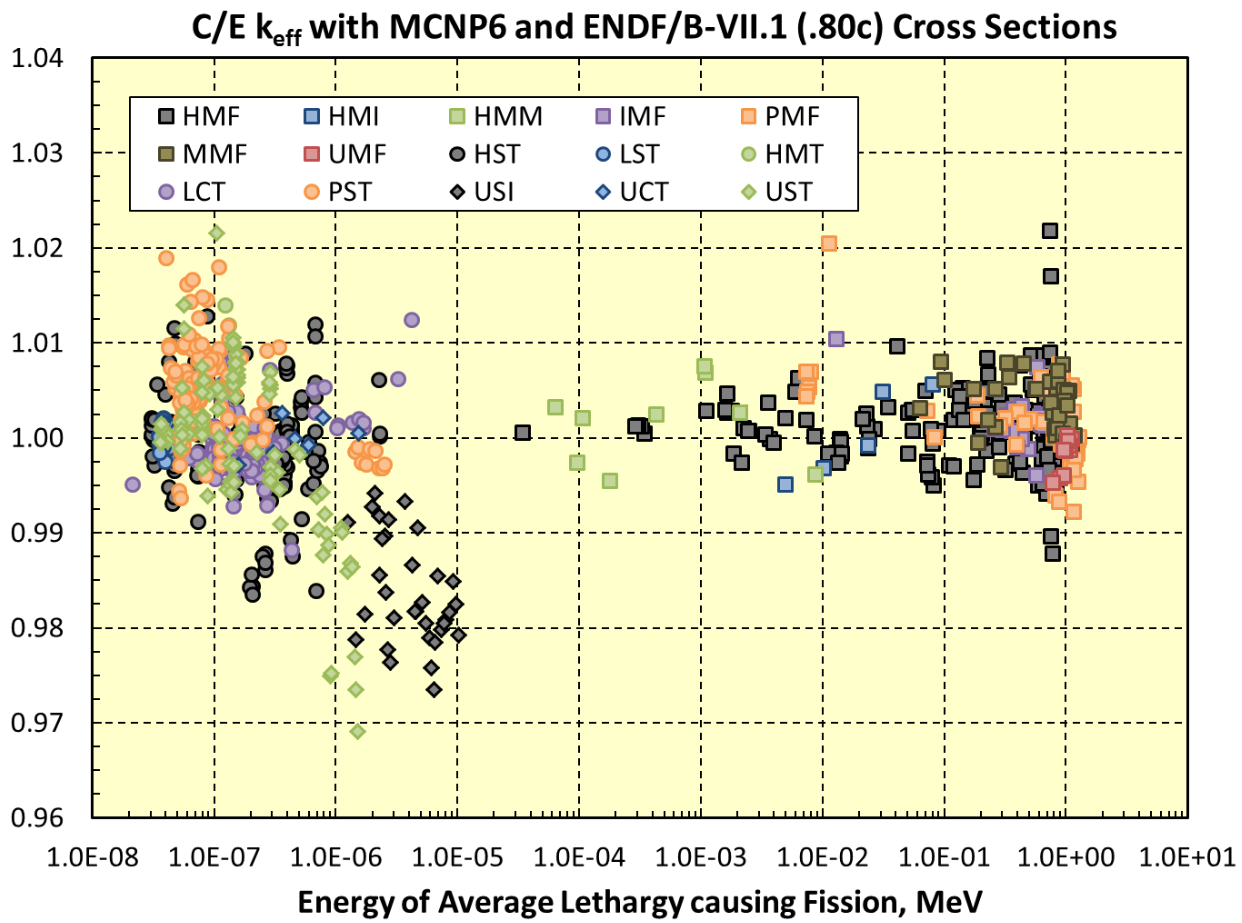
Center region (fast)

Outer region (intermediate)

Reaction	Measured Spectral Index	ENDF/B-VII.0 C/E	ENDF/B-VII.1 C/E
^{233}U(n,f)	0.3155	0.921(46)	0.922(46)
^{237}U(n,f)	0.537	0.832(42)	0.892(45)
^{238}U(n,f)	0.1397	1.029(51)	1.030(51)
^{239}Pu(n,f)	1.307	1.039(52)	1.039(52)
^{238}Pu(n,f)	1.002	0.967(48)	0.950(47)
^{240}Pu(n,f)	0.549	1.043(52)	1.026(51)
^{241}Pu(n,f)	1.073	0.911(46)	0.911(46)
^{242}Pu(n,f)	0.482	0.961(48)	0.984(49)
^{241}Am(n,f)	0.577	0.918(46)	0.914(46)
^{233}U(n,f)	0.08	0.669(33)	0.672(34)
^{237}U(n,f)	0.391	1.018(51)	0.973(49)
^{238}U(n,f)	0.02487	0.832(42)	0.832(42)
^{239}Pu(n,f)	1.145	0.985(49)	0.985(49)
^{238}Pu(n,f)	0.708	0.968(48)	0.946(47)
^{240}Pu(n,f)	0.26	0.899(45)	0.870(43)
^{241}Pu(n,f)	1.251	0.954(48)	0.953(48)
^{242}Pu(n,f)	0.19	0.845(42)	0.871(44)
^{241}Am(n,f)	0.184	0.793(40)	0.784(39)

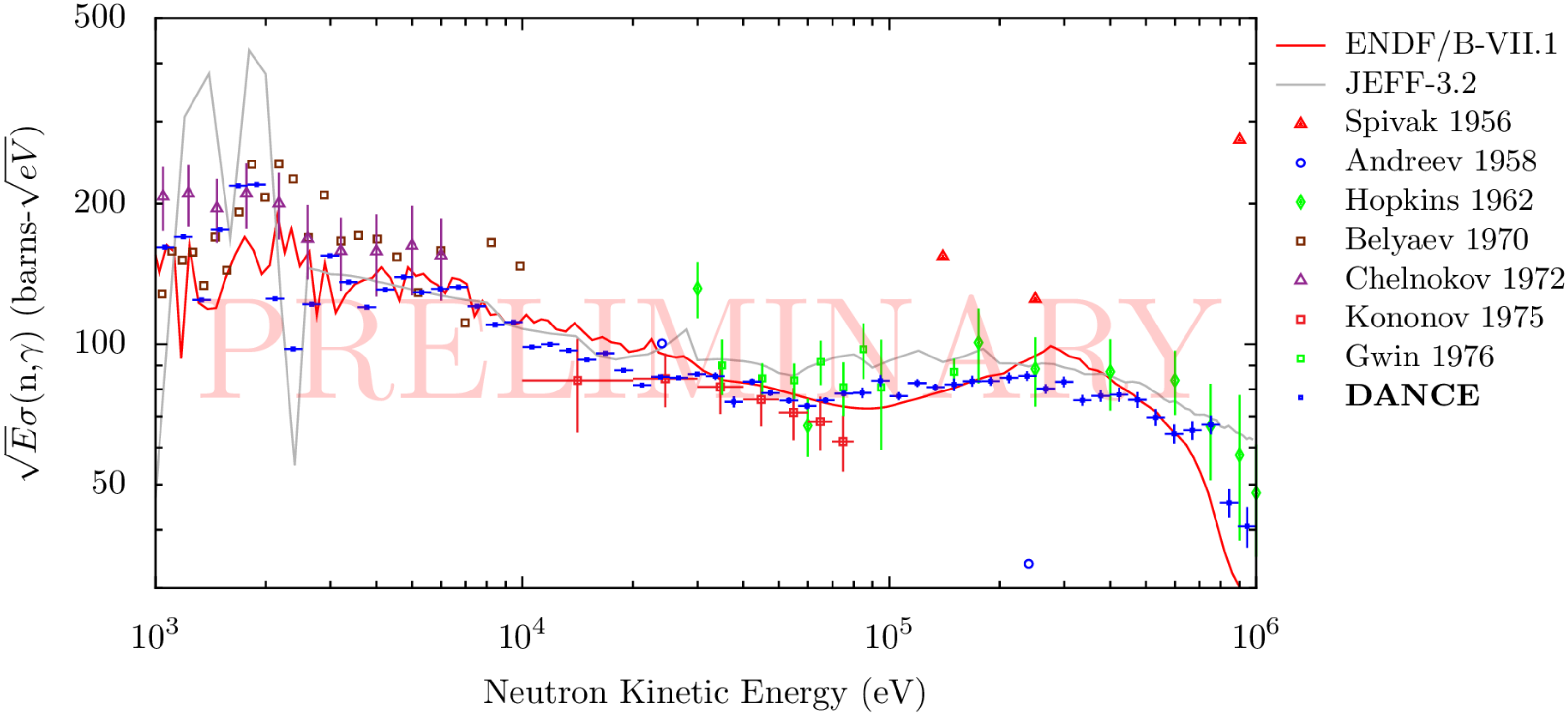
Survey of ICSBEP eigenvalue calculations (Kahler, LANL)

- Energy of Average Lethargy values below 1.0E-6 are largely "THERM" systems; values above 1.0E-1 are largely "FAST" systems.
- Fewer data exist the "INTERmediate" energy range.

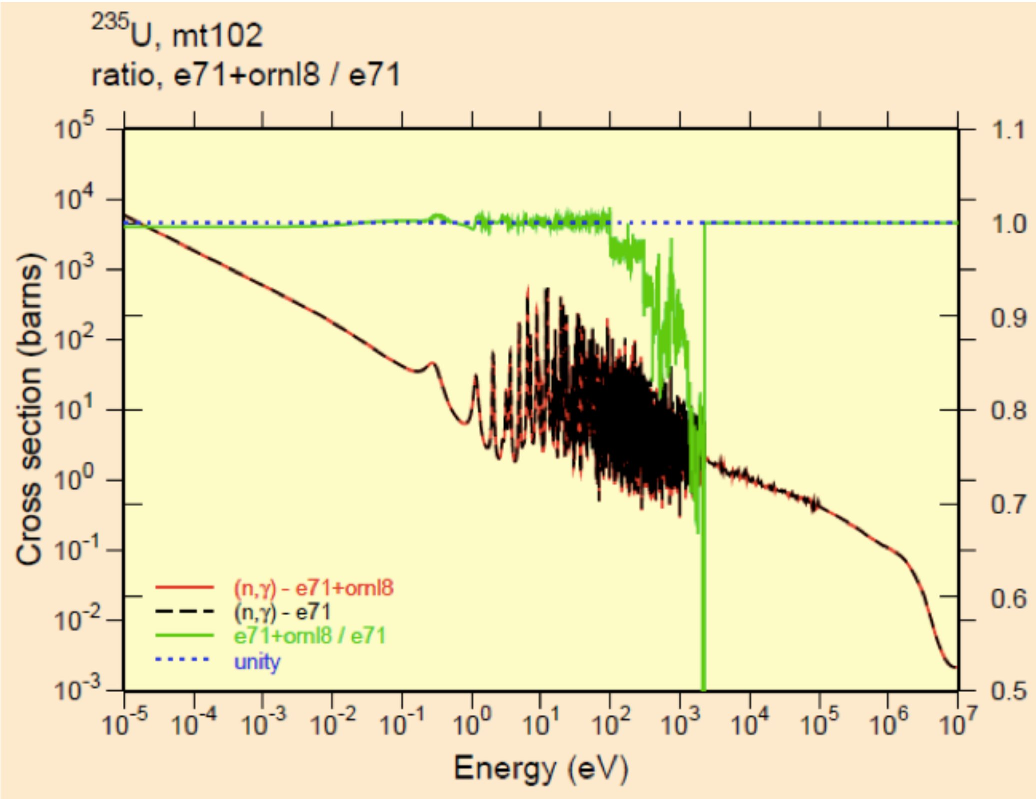


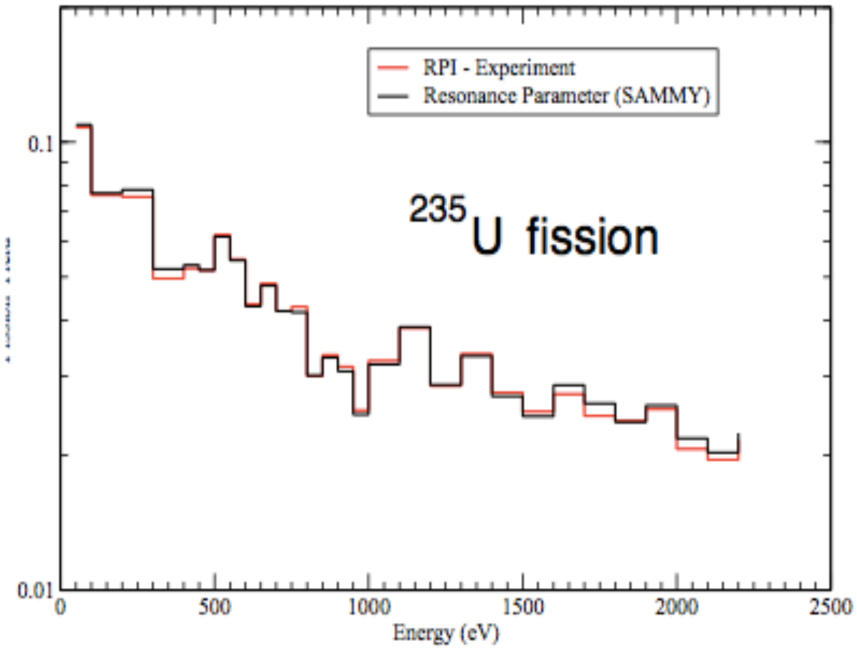
Radiative neutron capture is known poorly in the >1 keV region for many important actinides, including 239Pu

- Not measured since the 1960s at Los Alamos
- New DANCE effort started, but more work is needed

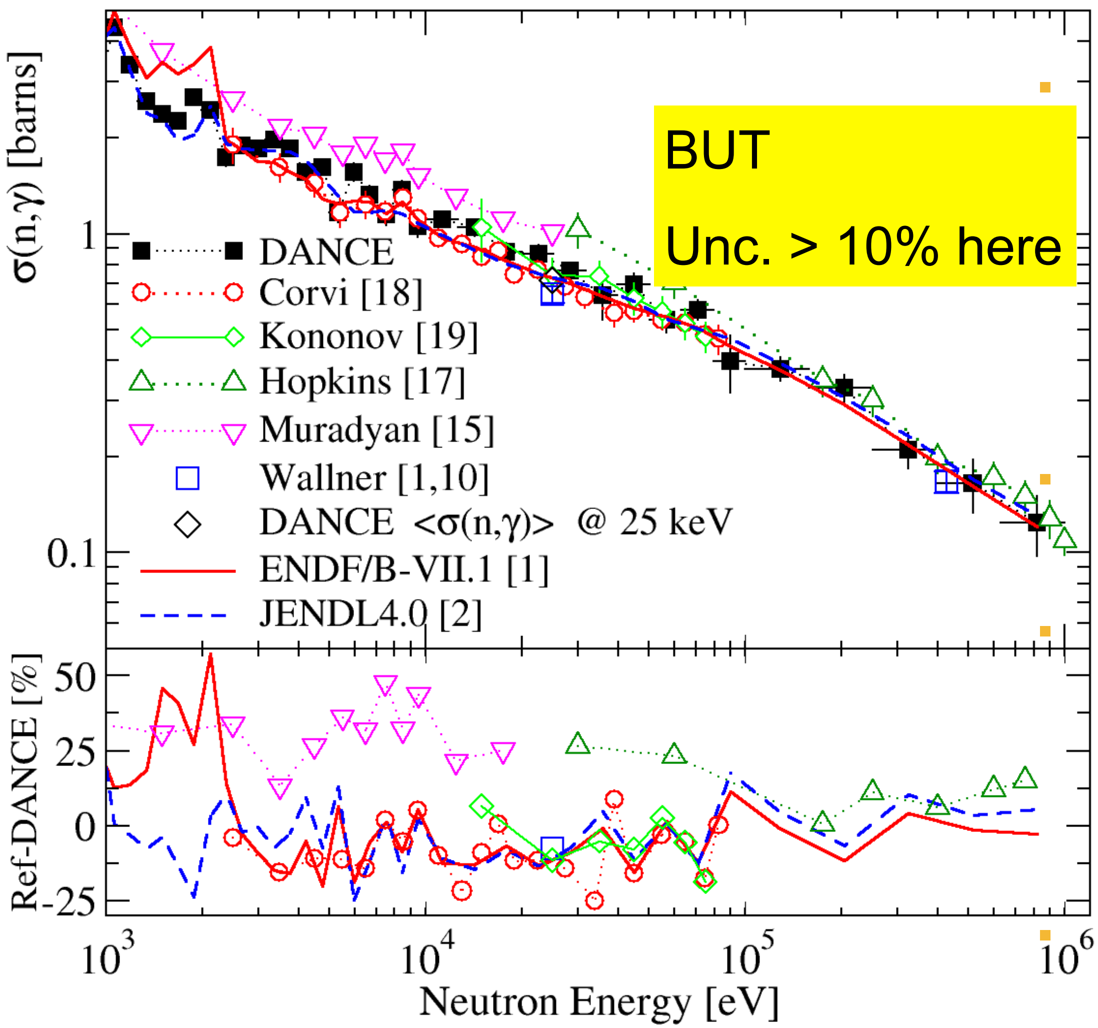


^{235}U capture: DANCE & RPI data solved the 0.5-2.5 MeV region questions (But questions above 2.5 keV still)





^{235}U capture: we need more accurate data in the 2.5 keV - MeV region



Jandel's ratio method helped

Precision <3% achieved using simultaneous rate determination;

- Rates of U5(ng) and U5(nf)
- The same target → same n flux for both reactions

Being implemented for ^{239}Pu (S. Mosby *et al.*)

But … discrepancies with Wallner AMS data at 25 keV; and ~10% uncertainties > 100 keV

NEUANCE detector?

neutron elastic & inelastic scattering – cross sections & angular distributions

- Fast reactor studies show that improving our understanding of scattering on 238U is one of the high priorities
  - WPEC/Subgroup report, Salvatores, Palmiotti, et al.

  - Labs need t better understand 235U and 239Pu scattering too, for:
    - Transport, leakage, “precise” understanding of criticality

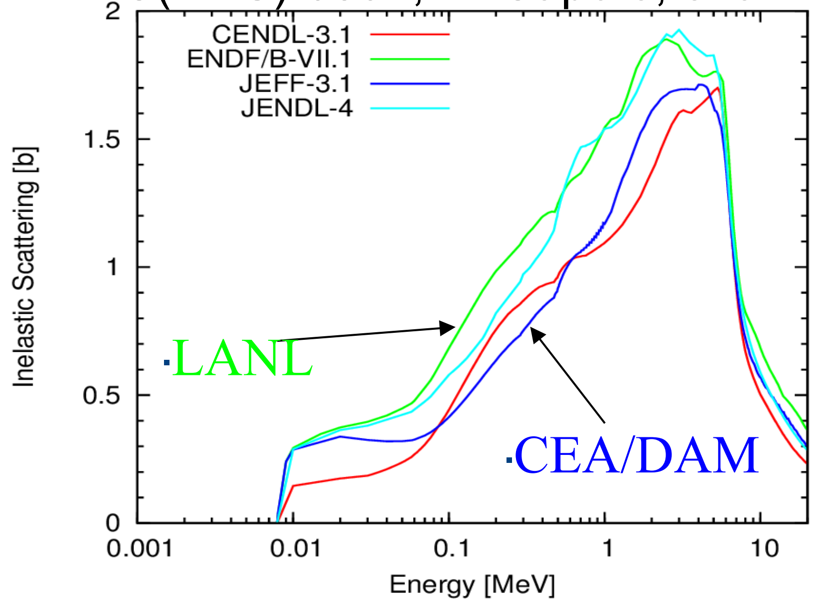
neutron scattering – cross sections & angular distributions

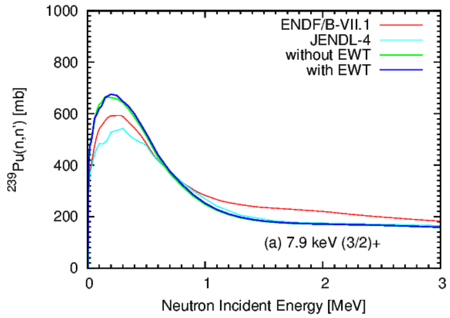
- IAEA Technical Meeting on Model Calculation for Major Actinides Summary report published: INDC(NDS)-0597, R. Capote, et al.

ENDF & CEA/BRC evaluations perform equally well for Jezebel keff prediction, but large compensating errors between scattering, PFNS, etc

Theory plays a key role here. Kawano et al working on various refinements to actinide scattering theory

Experimental measurements? Danon's semi-integral scattering measurements at RPI were successful for 238U & 56Fe. Develop capability at LANSCE for 239Pu and 235U?





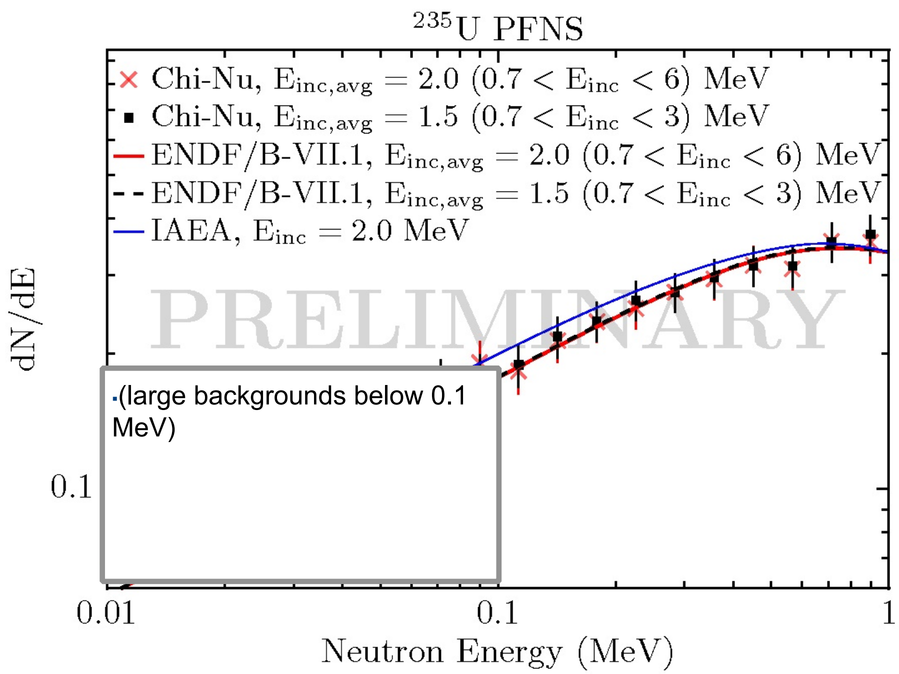
PFNS, prompt fission neutron scattering, is being actively studied across many countries

- ***Because there is an extremely large impact on criticality***
  - ***example: Major discussion/argument on whether the average energy for thermal ^{235}U PFNS is 2.03 MeV (ENDF) or 2.00 MeV (from latest IAEA & Talou analysis)***
  - ***Thermal ^{239}Pu PFNS poorly known too - impacts solution criticals significally***

- PFNS at thermal & fast energies
  - Indications from some experiments for a softer spectrum (high at low energies < 1 MeV; lower at high energies > 5 MeV)
    - *But there could be background scattering contamination*
  - Theories give quite a range of predictions…. Detailed fission data help (eg TKE as a function of A)
  - Recent LANL data on ^{235}U challenges this hypothesis

^{235}U : 2 LANL Experiments cover the whole emission energy range – Chi-nu (LANSCE) and NUEX (Lestone-Shores)

Eout < 1 MeV



Eout > 1 MeV

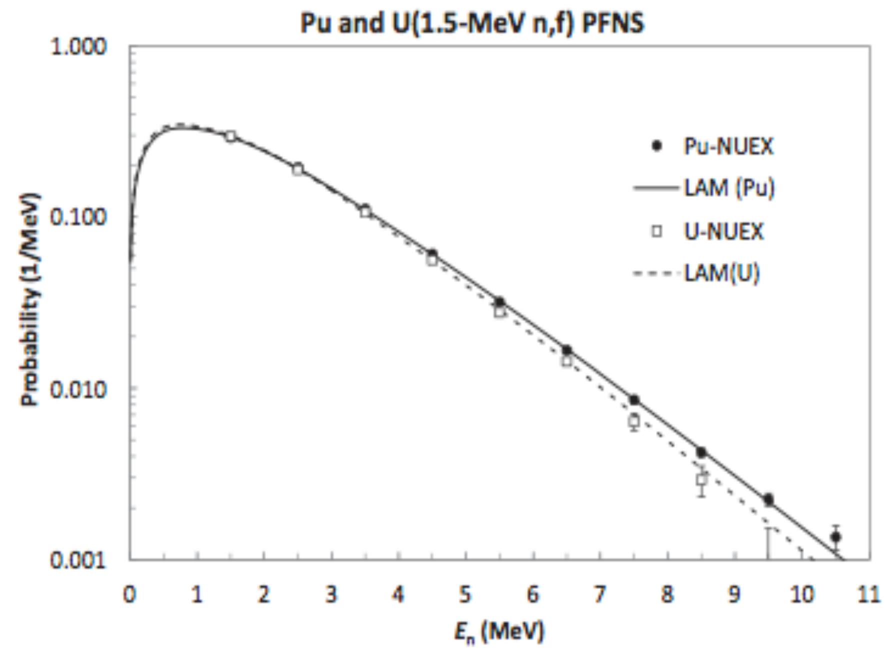
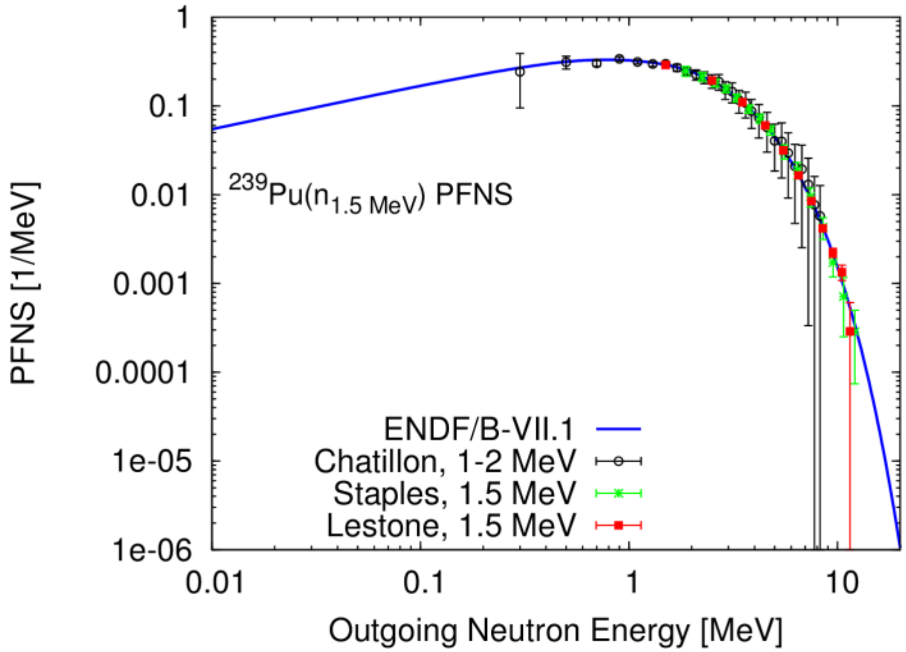
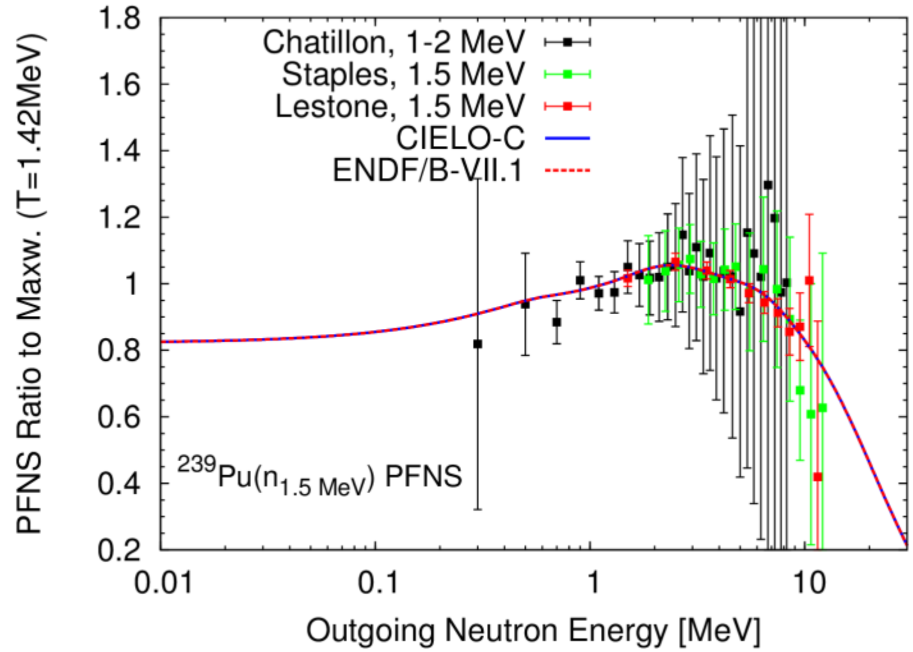


FIG. 3. The emission probabilities listed in Tables III and IV, and the corresponding 1.5-MeV n + ^{239}Pu and ^{235}U Los Alamos fission model fission-neutron energy spectra (curves).

239Pu PFNS at Einc = 1.5 MeV

Chi-nu data in coming years will address this



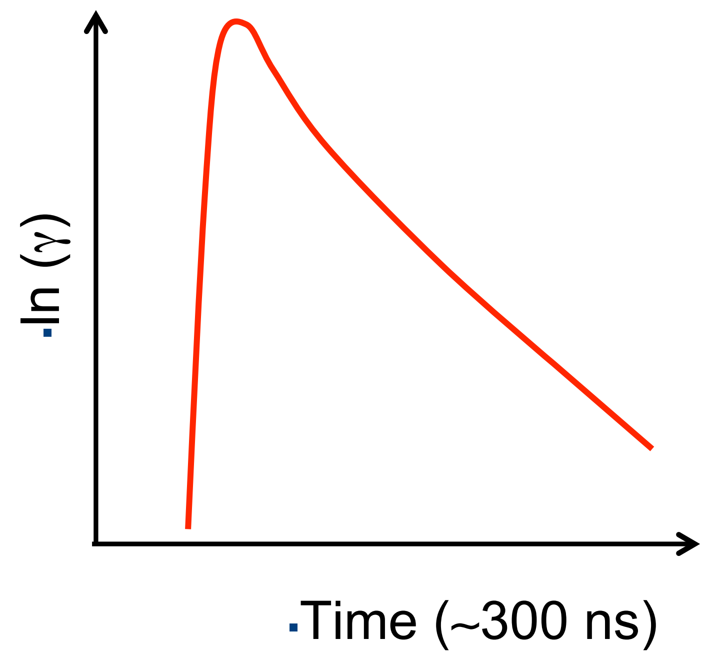


Fission Decay Chain Measurements Motivate Prompt Fission Gamma-Ray Data at LANSCE/Lujan

Traditional approaches

- 1/m plots (count rate v. control rod position, to identify asymptote & critical)
- Feynman variance of counts (doubles …) to infer multiplication and k-eff

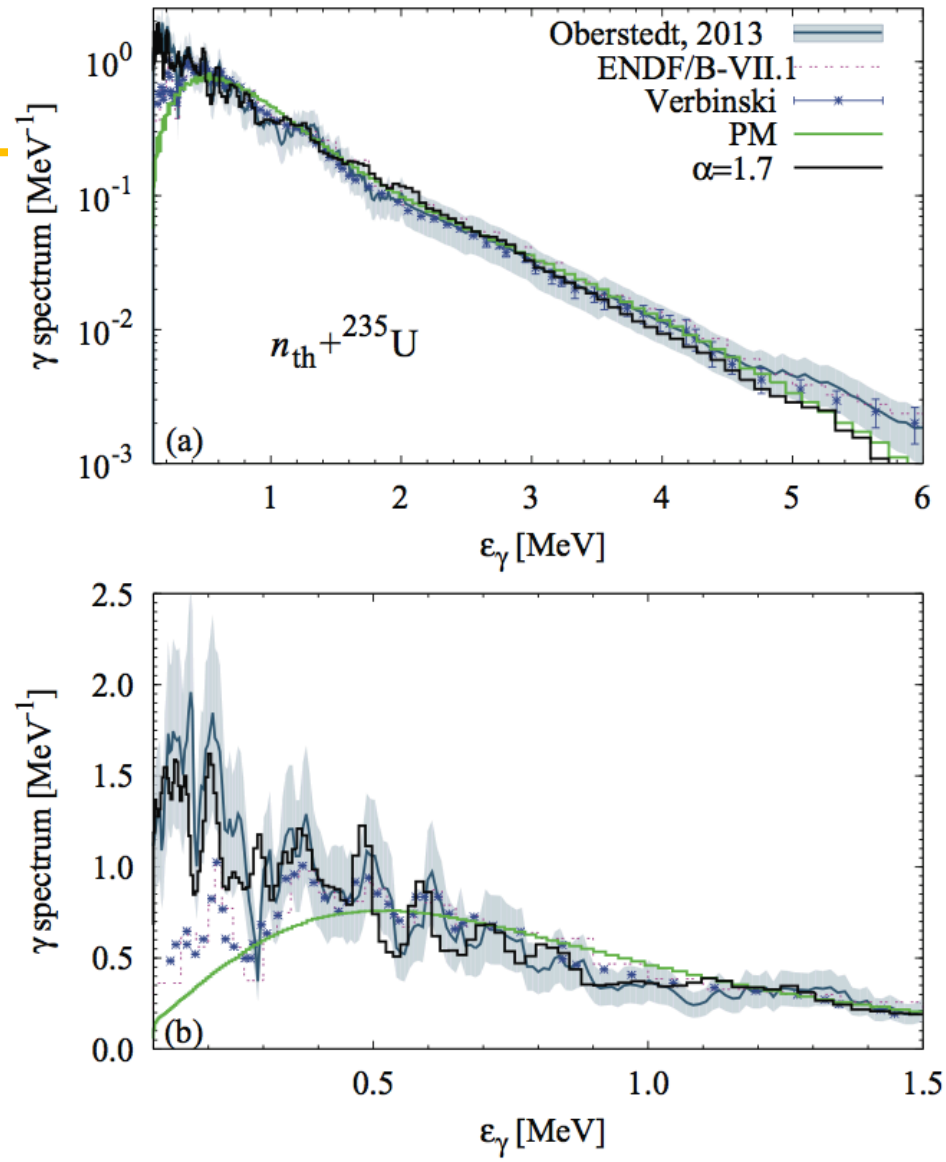
- Decay of fission chain via fission-gamma-rays
- Drives a better understanding of prompt-gamma-ray spectra, PFGS (studied by LLNL & LANL at LANSCE), & at IRMM/Geel



Prompt fission gamma-ray cascades

Results for n_{th}+^{235}U

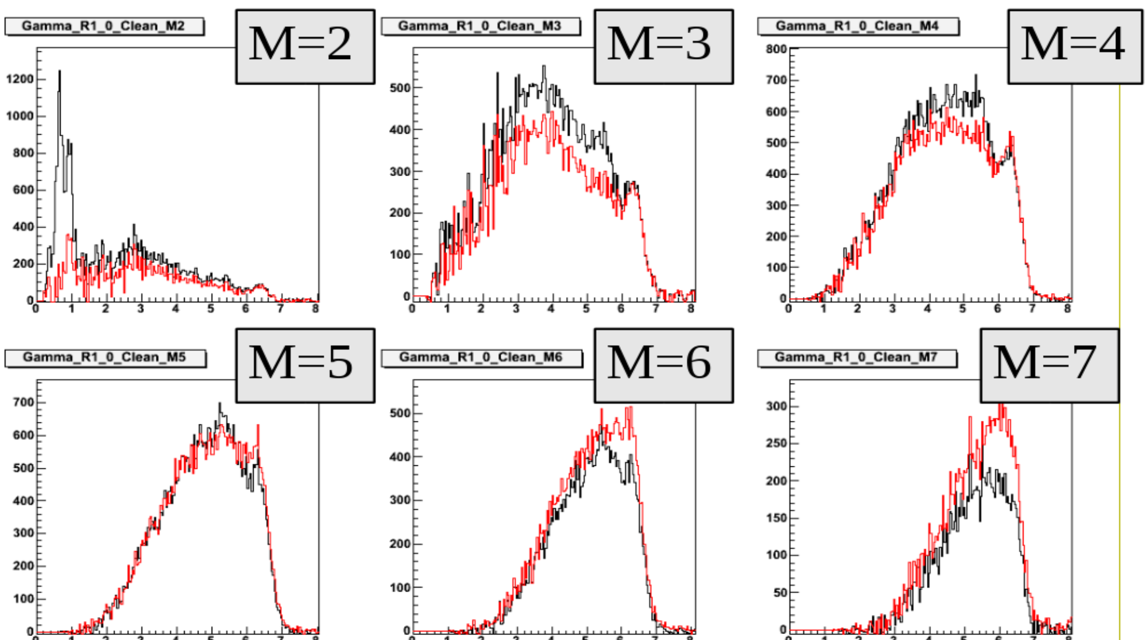
- Strong fluctuations of PFGS below 1 MeV, due to γ transitions in fission fragments
- Seen in IRMM experiments with LaBr detectors, but not at DANCE due to poor energy resolution
- Well reproduced by Monte Carlo simulations

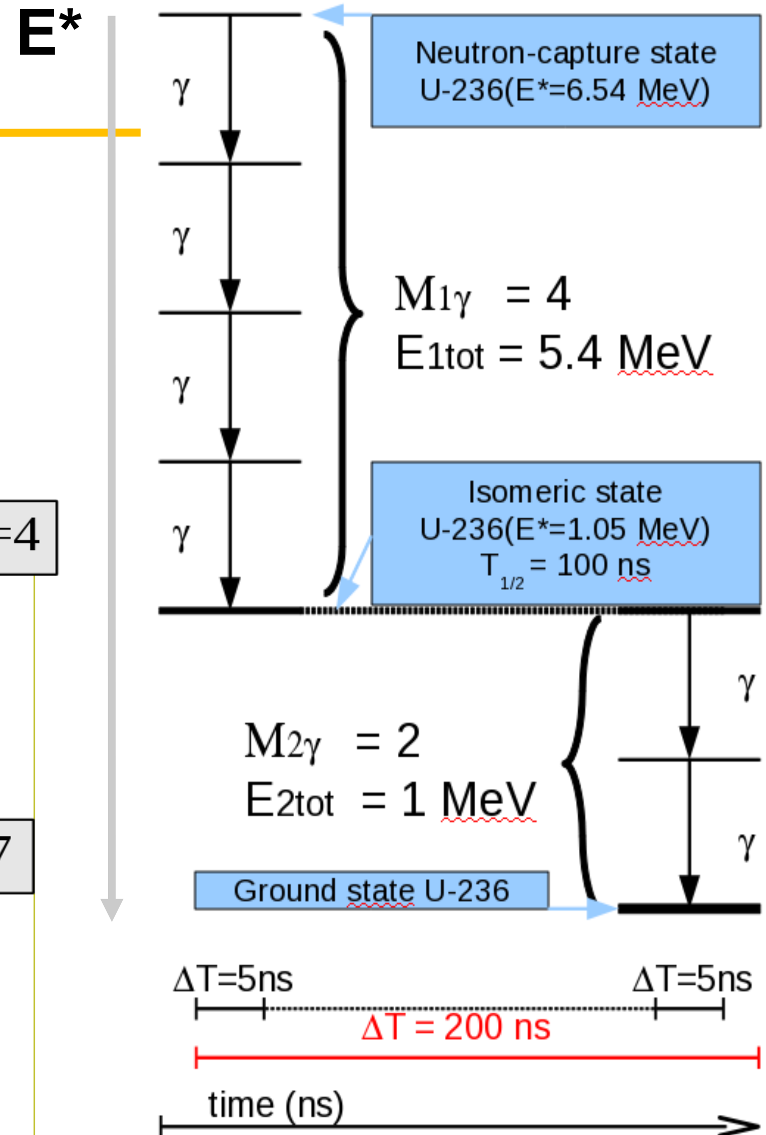


Short-Lived Isomeric states after U235+n

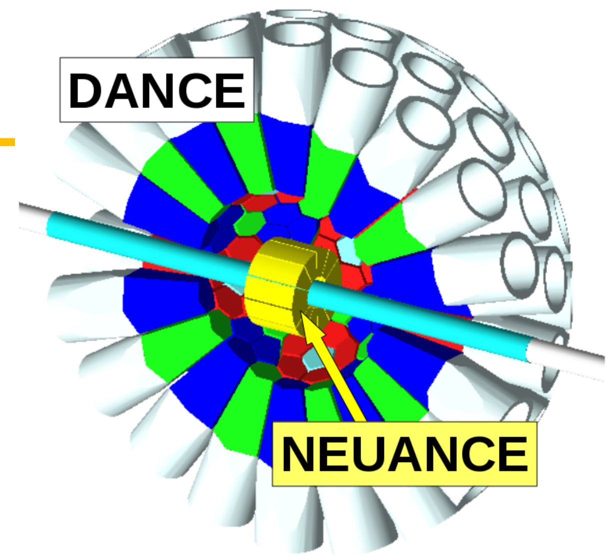
- During analysis of $^{235}U(n,\gamma)$ cross section we have found structure in the total gamma-ray energy E_{tot} spectra

M. Jandel et al., Phys Rev Lett 109, (2012)





Isomeric states after U-235+n

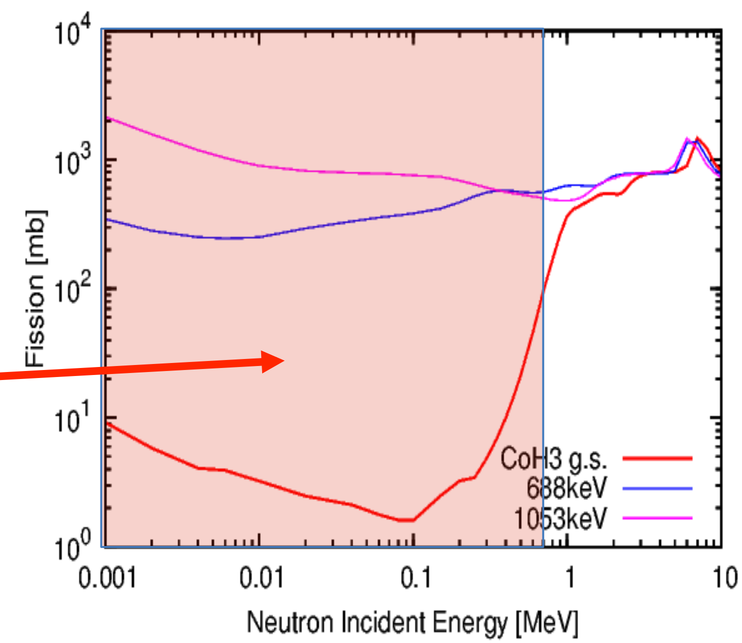


- In high neutron fluence the secondary reactions can occur
- $^{236}U^*$: 1024 keV (4-) $T_{1/2}$ = 100 ns
- $^{236}U^*$: 678 keV (1-) $T_{1/2}$ = 3.7 ns
- Current work addresses resonance region

- What is the population of these states after ^{235}U+n ?

- What are the n-reaction cross sections on these states ?

A. **Future – unresolved region En> 1keV**



Conclusions

Many questions remain for the major actinides

Impact of these uncertainties on our simulations is large

- Major gains if we could better understand these data

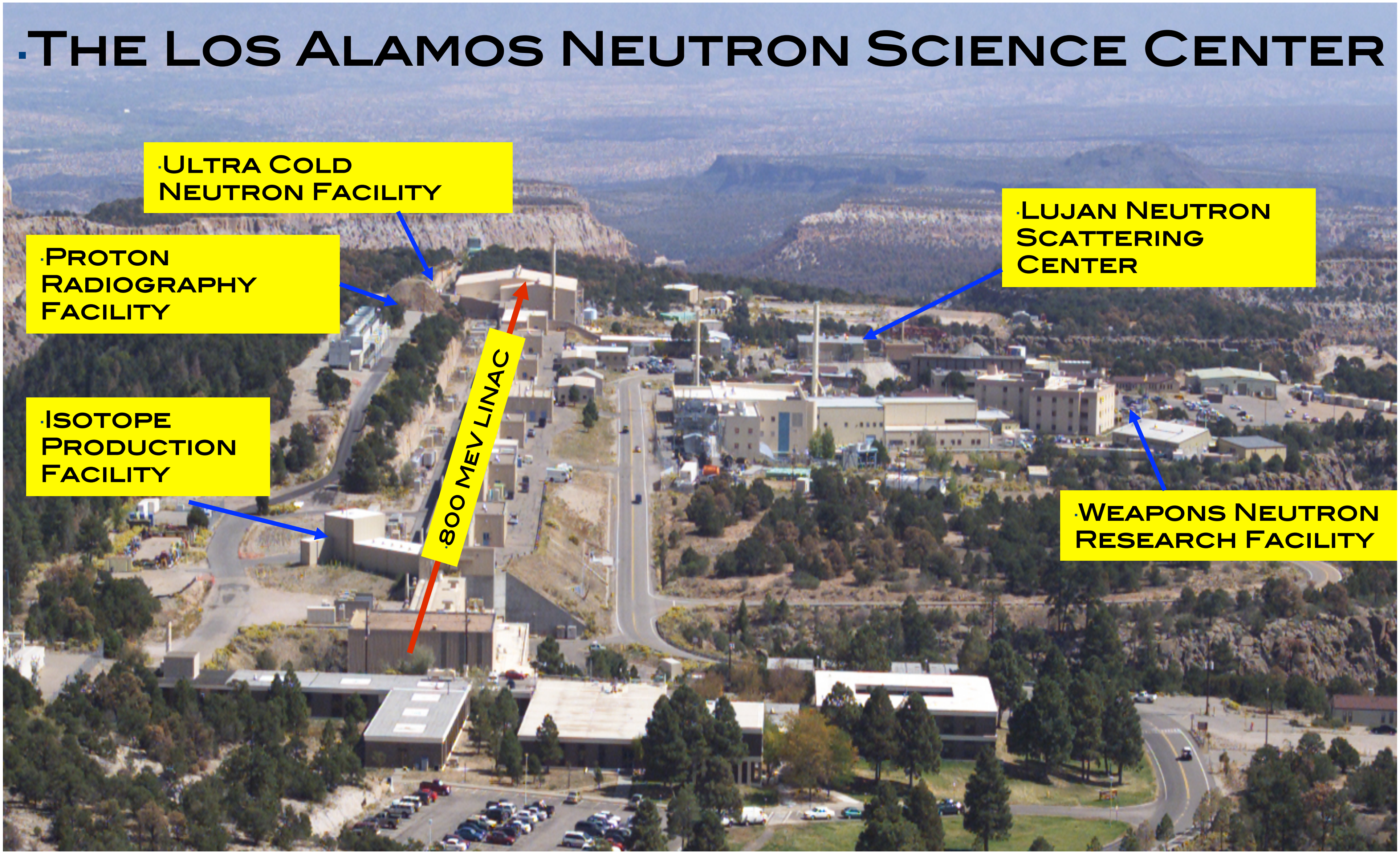
Backup

Optimization of a New Target at Lujan for Nuclear Science

- An opportunity now exists to optimize the present Lujan Center neutron spectrum to better cover the important intermediate neutron energy range between 100 eV to 2 MeV.

- Optimizations include:
  - Installation of a faster moderator which will enhance the neutron flux and energy resolution in this intermediate energy region
  - Changes to the pulse structure of the proton beam which includes producing a narrower proton pulse for better energy resolution and increasing the pulse repetition rate
  - Developing pulse stacking in the Proton Storage ring to increase the proton current. Initially, such a pulse-stacked pulse may be approximately 30 ns wide separated by 25 ms. If we store 4 pulses in the ring, the intensity will be approximately 95 uA with a pulse repetition rate of 160 Hz.

The Los Alamos Neutron Science Center



The issue:

- DOE/Office of Science pulled out of Lujan materials research