LA-UR-13-27602

Fundamental and Applied Nuclear Fission Research at LANL

Patrick Talou

T-2, Nuclear Physics Group, Los Alamos National Laboratory
New Mexico, USA

INT 13-3
Quantitative Large Amplitude Shape Dynamics:
Fission and Heavy Ion Fusion

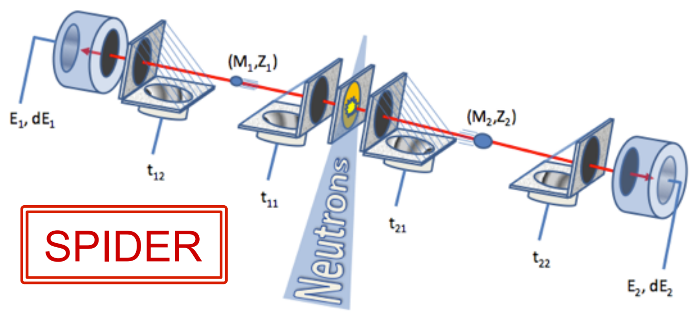
Talk Outline

- **Introduction:** a “Renaissance” of Nuclear Fission Research at LANL
  - New theoretical efforts, new experimental devices, why?
  - Both fundamental and applied science is being carried out
- **Goals**
  - From phenomenological and adjusted to more fundamental and predictive
  - “Putting it together”
- **Topics of research**
  - Fission cross-sections
  - Fission fragment yields
  - Post-scission: prompt fission neutrons and γ rays; β-delayed n and γ rays.
  - Fission recycling in nuclear astrophysics
  - UQ associated with all data to be delivered

“Renaissance” of Nuclear Fission Research at LANL

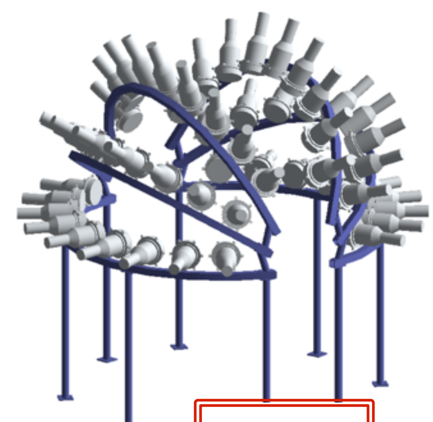
- **Los Alamos Neutron Science Center:**
  - Time-Projection Chamber, DANCE, Chi-Nu, SPIDER
- **T-2 Theory & Modeling efforts**
  - Fission cross sections, fission fragment yields, prompt fission neutrons and γ rays, β-delayed neutrons and γ rays, astrophysics reaction networks
- **X-CP Transport Calculations**
- **Uncertainty Quantification**

Time-Projection Chamber



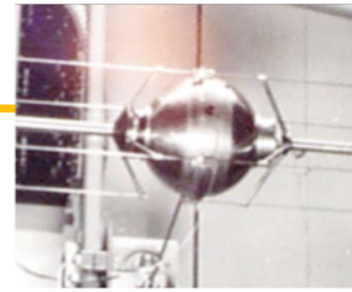
SPIDER

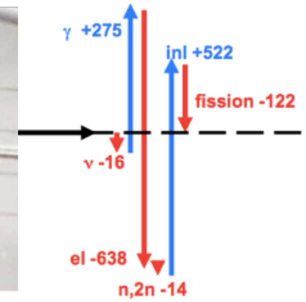
DANCE



Chi-Nu

Why?





- **We don't know everything yet!... Duh…**
  - Not such a trivial statement
  - Quality of evaluated nuclear data– oversold
  - Compensating errors in integral benchmarks (e.g., k_{eff} in Jezebel)
  - Lack of predictive power
- **New applications and new requirements for existing applications**
  - Future reactors (new fuel compositions, new geometries, etc.)
  - Existing fuel cycle (safety, waste management, etc.)
  - Non-proliferation, attribution, etc.
  - Astrophysics (reaction networks)
  - Uncertainty Quantification

- **New capabilities: Experimental & Computing**
- **Fundamental & Applied Research**

Two overarching goals

From
Phenomenological and Adjusted
to
more Fundamental and Predictive

“Putting it all together”

Fission cross sections
Fission fragment yields
Fission fragment angular distributions
Prompt fission neutrons
Prompt fission photons
(β-delayed neutrons and photons)

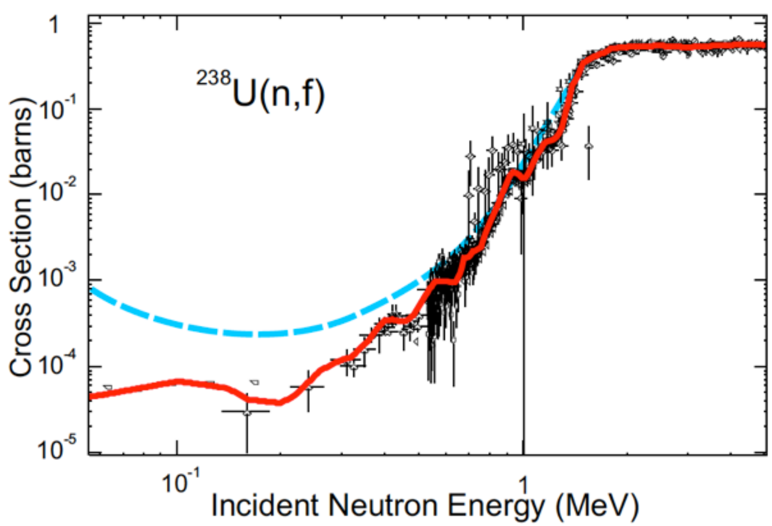
(right now, a different model for each fission data type)

FISSION CROSS SECTIONS

Fission Cross Sections

Status of Evaluated Models & Data

- **Fission Cross Sections**
  - Compound nucleus (Hauser-Feshbach) reaction theory
  - Non-statistical corrections (pre-equilibrium, width fluctuation corrections, …)
  - Multi-modal fission
  - Approximate treatments of class-II states
  - Fission barrier heights and widths, level densities, etc., fitted to measured cross sections



M.Herman *et al.*, Nuclear Data Sheets **108**, 2655 (2007)

Limitations?

- One-dimension
- Fitted barrier heights, widths, transition states, level densities, …
- Not predictive
- Lack of consistency between different isotopes and entrance channels, e.g., (γ,f), (t,pf)

Fission Transition States

- Original idea of A. Bohr (1959) following the experimental observation of strong anisotropies in fission fragment angular distributions

$$d\sigma_f(\theta) = \sum_J \sum_{M=-J}^{J} \sigma(JM) \sum_{K=0}^{J} \frac{\Gamma_f(JK)}{\Gamma(J)} \frac{2J+1}{4} \left(\left|\mathcal{D}^J_{MK}(\theta)\right|^2 + \left|\mathcal{D}^J_{M-K}(\theta)\right|^2 \right) sin\theta d\theta$$

s-wave neutrons on even target: $W(KI) = \frac{1}{4}(2I+1)\left[|d^I_{1/2,K}(\theta)|^2 + |d^I_{-1/2,K}(\theta)|^2\right]$

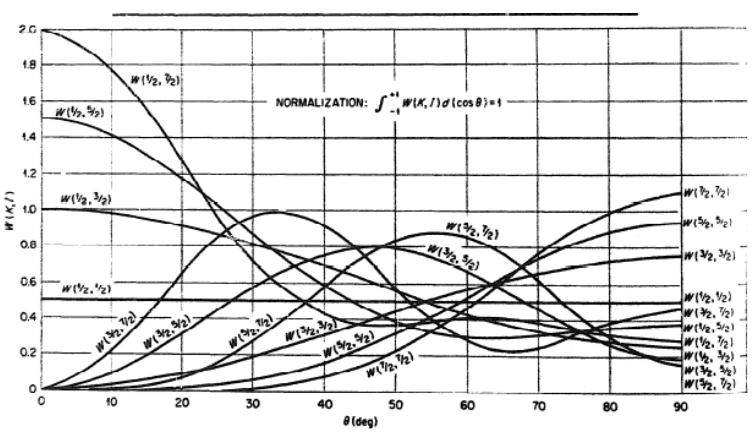
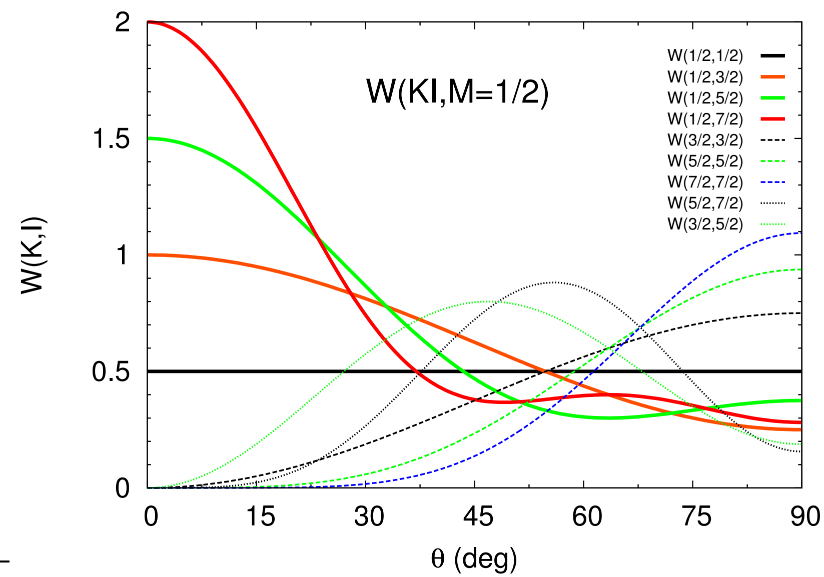
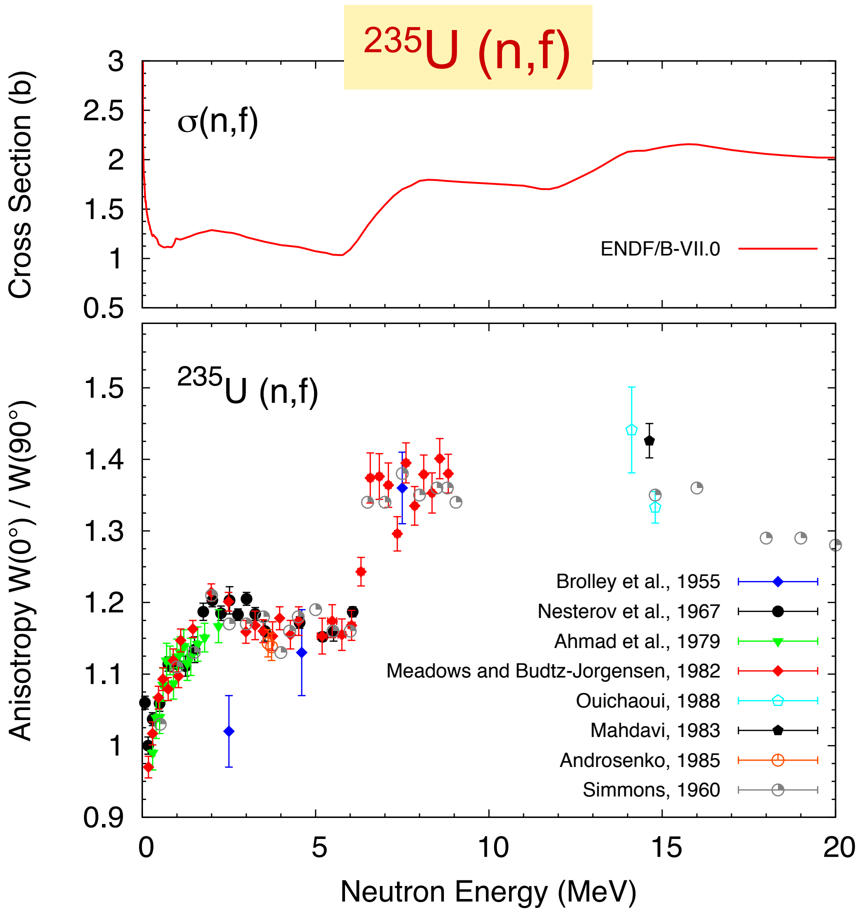


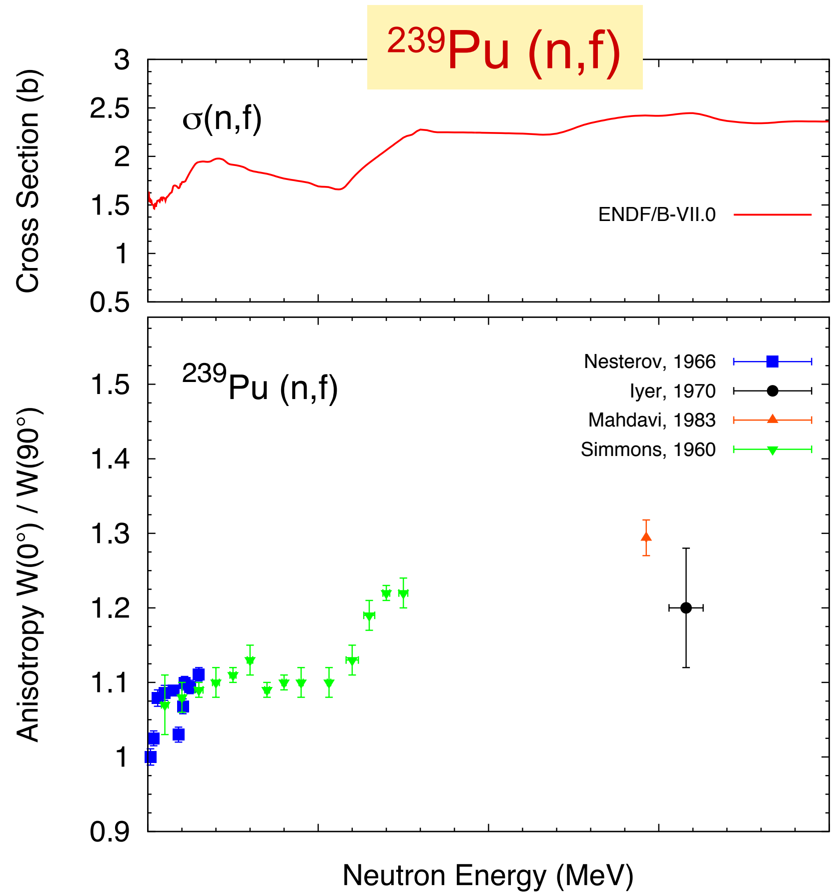
Fig. 6. Theoretical fission fragment angular distributions for fission through pure rotational states, *W(KI)*.



Lamphere, Nucl. Phys. **A38**, 661 (1962)

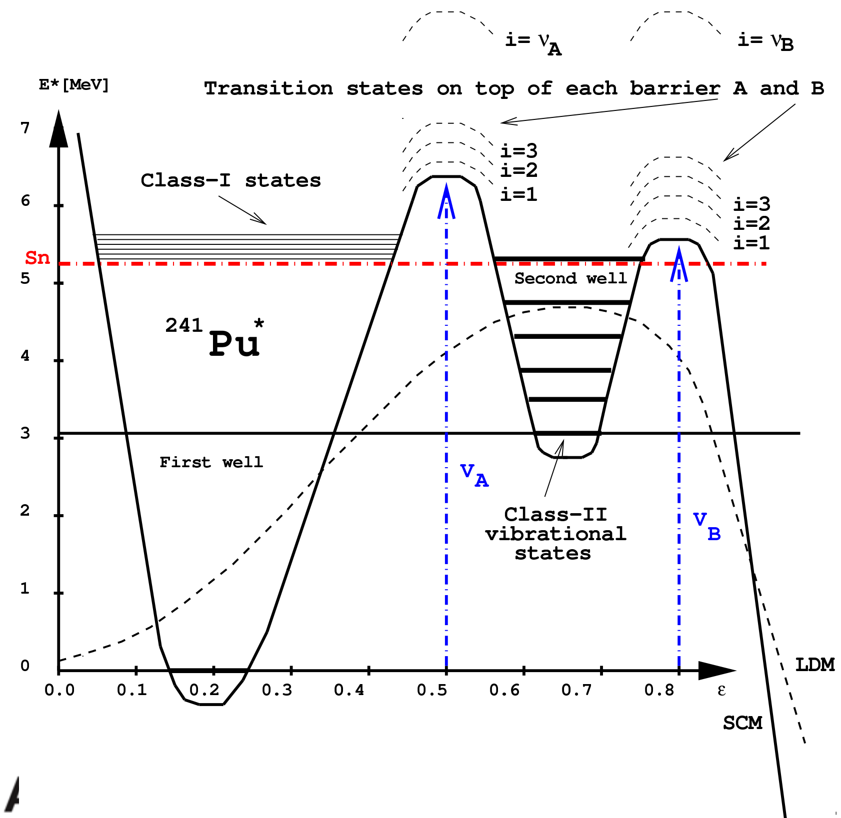
Anisotropy with increasing E*

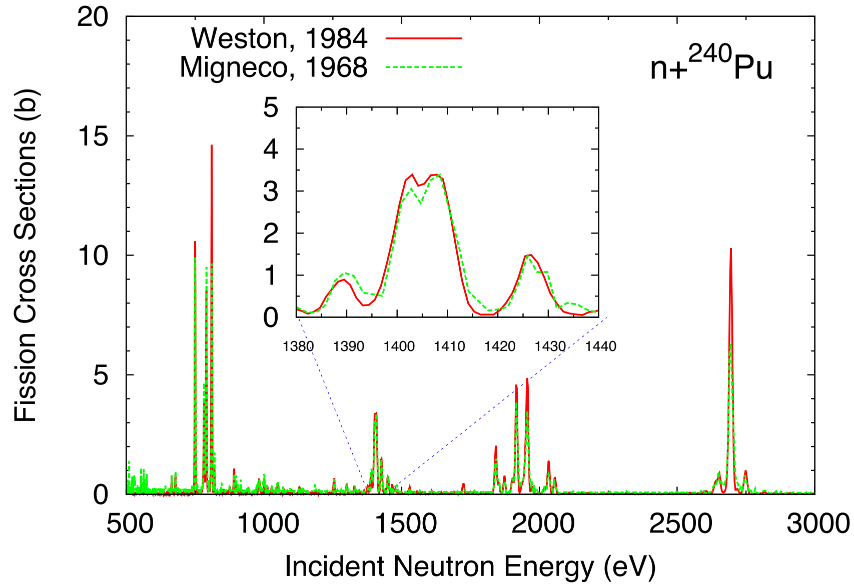




R-Matrix Approach to Fission Cross Section Modeling

- S.Bjørnholm and J.E.Lynn, Rev. Mod. Phys. **52**, 725 (1980)
- Presence of 2nd well → Coupling between class-I and class-II states





R-Matrix (cont'd)

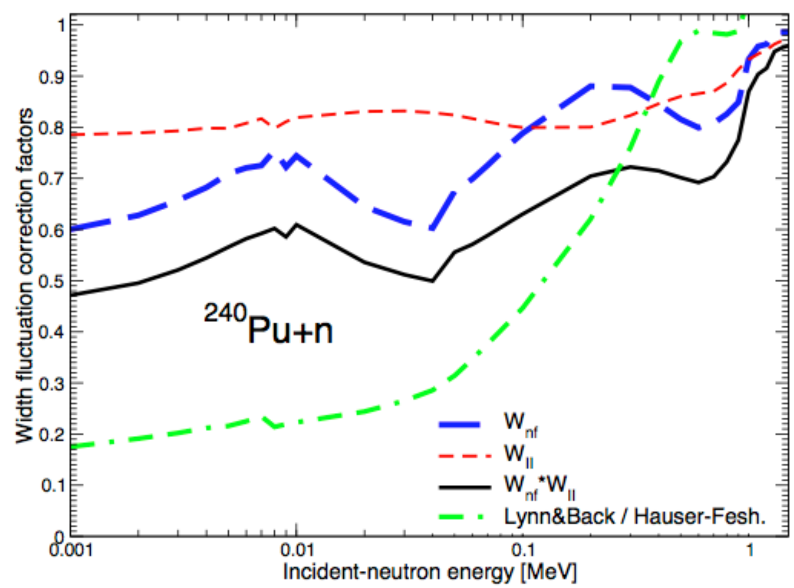
- **Hauser-Feshbach formula has to be modified**

$$\sigma_{nf} = \sum_{J^\pi} \sigma_c^{J^\pi} \times \frac{T_f}{\sum_c T_c} \times W_{cf} \times W_{II}$$

Width Fluctuation Correction Factor

Correction Factor due to 2nd well

- **Many approximate solutions exist**
- **Most accurate approach:** Monte Carlo sampling of Class-I and Class-II states characteristics, and of their coupling matrix elements
- Consistent approach throughout a suite of Pu isotopes: O.Bouland, J.E.Lynn, P.Talou, to appear in Phys. Rev. C (2013)

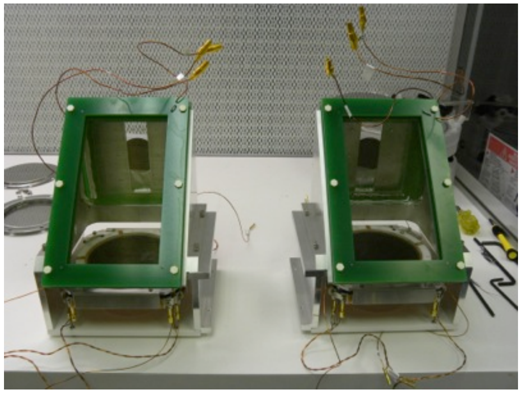


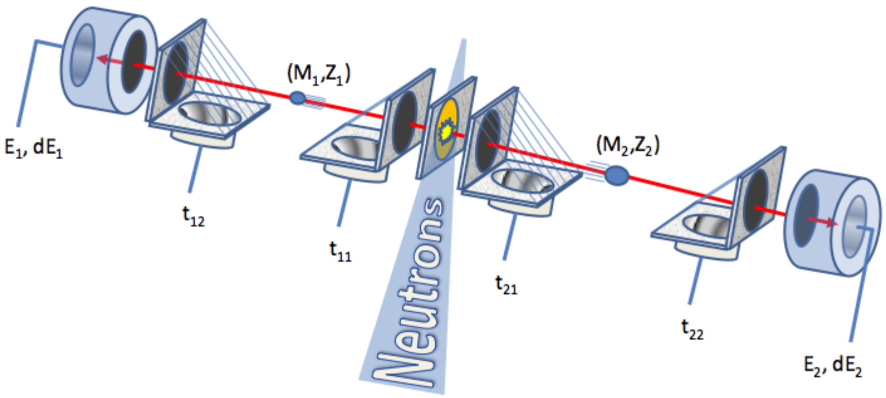
FISSION FRAGMENT YIELDS

See talks by Arnie Sierk, Jørgen Randrup, Peter Möller, Noël Dubray, Nicolas Schunck, etc.

SPectrometer for Ion DEtermination in fission Research (SPIDER) at LANSCE- F.Tovesson et al.

- 2E-2v method
- Time-of-Flight
- Ionization chambers to measure E of FF (0.5-1% energy resolution); dE/E to estimate Z
- Multiple detectors to increase efficiency

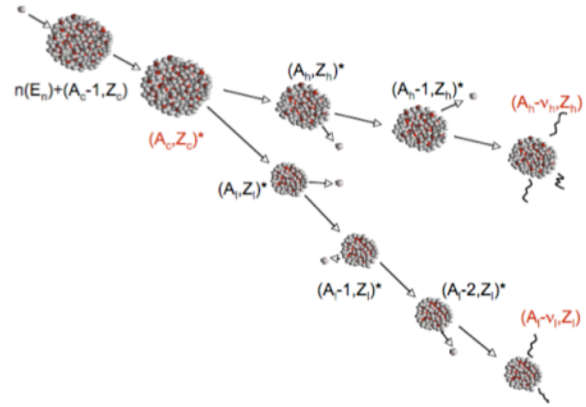




PROMPT FISSION NEUTRONS AND PHOTONS

Prompt Fission Neutrons and Photons

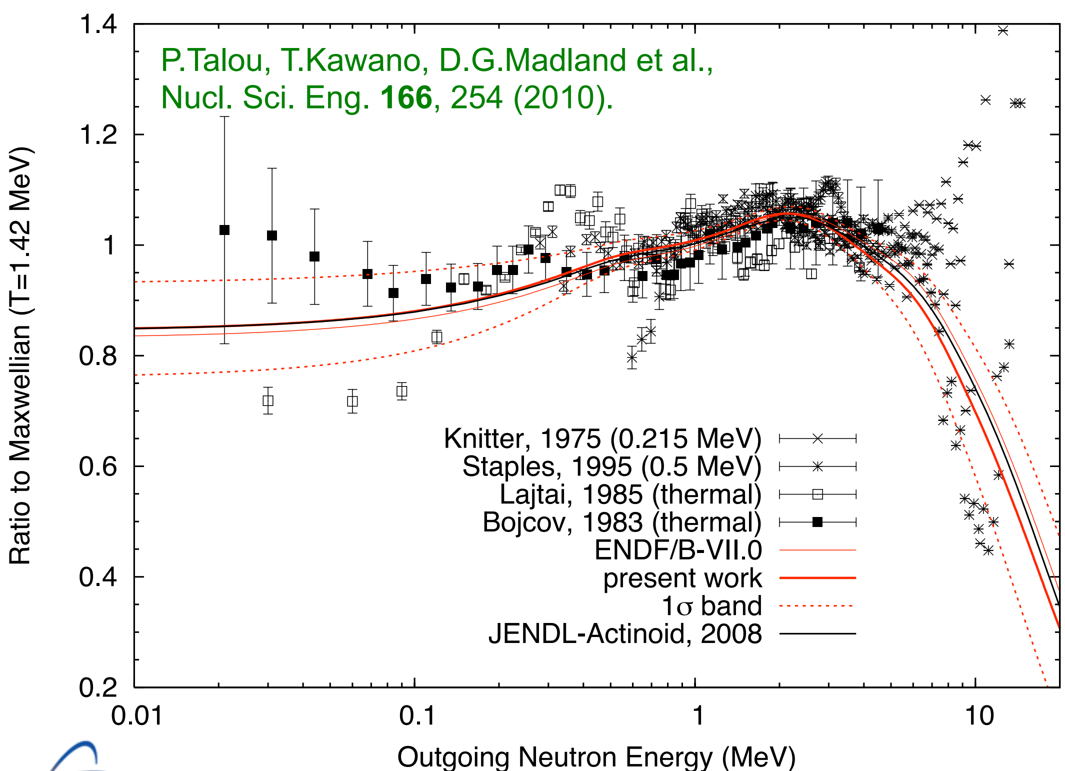
Status of Evaluated Models & Data

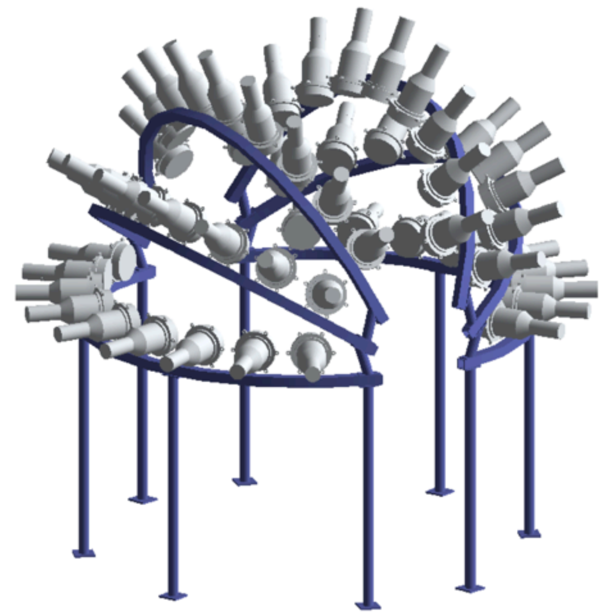


- **Prompt fission neutrons**
  - "Los Alamos" or "Madland-Nix" model
    D.G.Madland and J.R.Nix, Nucl. Sci. Eng. 81, 213 (1982)
  - Fits with Watt spectra
- **Prompt fission photons**
  - Experimental data only
- **Recent review of existing experimental data**
  - Neudecker *et al.*, LA-UR-13-24743, revealed many problems with past experiments
- **International efforts underway**
  - IAEA Coordinated Research Project on "Prompt fission neutron spectra of actinides", IAEA Secretary: R.Capote-Noy.
  - CIELO International Evaluation Project

Prompt Fission Neutron Spectrum (PFNS)

- **Chi-Nu experimental efforts to measure low- and high-energy tails of the PFNS**





- n+^{239}Pu PFNS for E_n from 0.5 to 30 MeV
- 5% uncertainty between 50 keV and 12 MeV emitted neutron energy
- Double ToF experiment + angular info
- ~ 60 Liquid scintillators + ^{6}Li-glass detectors

Theoretical & Evaluation Work on PFNS

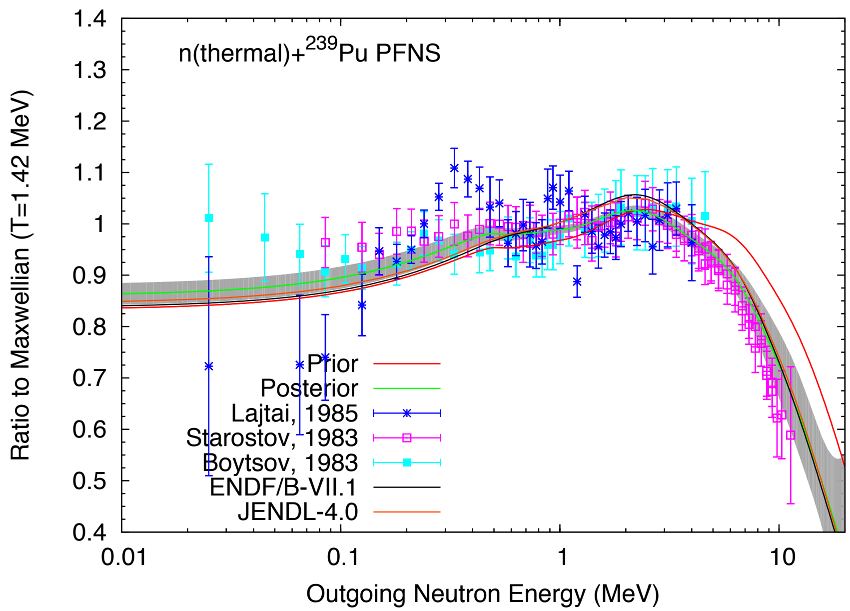
- **Original and extended versions**
  D.G.Madland and J.R.Nix, Nucl. Sci. Eng. **81**, 213 (1982)

$$N(E) = \frac{1}{2\sqrt{E_f}T_m^2}\frac{1}{1+b/3}\int_{(\sqrt{E}-\sqrt{E_f})^2}^{(\sqrt{E}+\sqrt{E_f})^2} d\epsilon\sigma_c(\epsilon)\sqrt{\epsilon}\left(1+b\frac{(E-\epsilon-E_f)^2}{4\epsilon E_f}\right)\int_0^{T_m} dTk(T)T\exp(-\epsilon/T)$$

- Bayesian statistical technique to combine experimental data and model calculations
  M.E.Rising, P.Talou, T.Kawano and A.K.Prinja, Nucl. Sci. & Eng. 175, 81-93 (2013).

- Very successful model used in most evaluated libraries

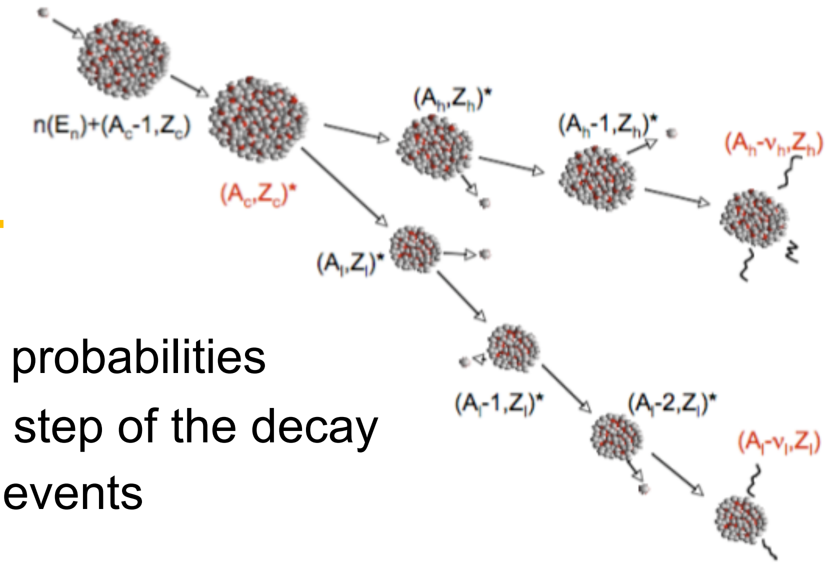
- Limitations:
  - Only average spectrum and multiplicity
  - Strong physical assumptions
  - Difficult to extend to other quantities



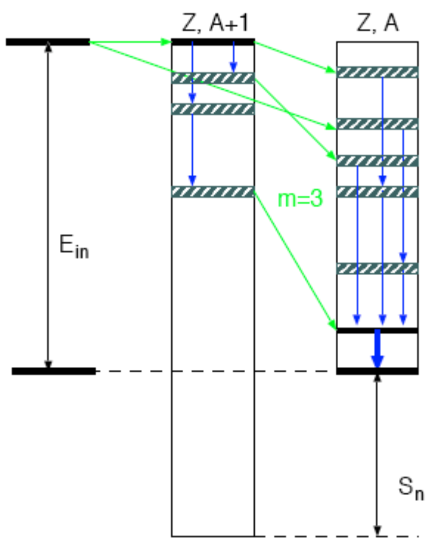
Monte Carlo Hauser-Feshbach Simulations

- **Each fission fragment = Compound nucleus with E*~10-15 MeV**
- **Hauser-Feshbach statistical theory**
  - Two main open emission channels: neutrons and photons
    Light charged particle emissions are negligible due to Coulomb barrier
- **LANL Code: CGM/F**
  - Deterministic and Monte Carlo modes
  - Written in C++, MPI-parallel instructions
  - Similar to DICEBOX at lowest excitation energies
  - Other *similar* codes: FREYA (LBNL-LLNL), FIFRELIN (CEA), GEF (Schmidt), …
- **Calculated Quantities:**
  - Deterministic: Average γ-ray spectrum and multiplicity
  - Monte Carlo: Set of histories with exact decay path → distributions, correlations, etc.

CGMF: The Mechanics



- Hauser-Feshbach Statistical Theory
- Calculate neutron and photon emission probabilities
- Sample probability distributions at each step of the decay
- Record Monte Carlo histories of fission events
- Perform statistical analyses of results



✧ CGMF –i 98252 –e 0.0 –n 100000 ➔ output: history file:

```
43 107 18.6565 5.5 1 104.368 2 6 0 0 0 1 1 1 2 3 3
4.382 0.410 0.242 2.450
0.819 1.890 0.801 1.946 0.275 1.622 0.423 1.827 0.160 2.905 0.077 0.766
55 145 14.7974 9.5 -1 77.0161 2 5 0 0 0 1 2 3 3 3
1.126 1.597 0.900 0.989
0.635 0.359 0.637 2.227 0.810 0.750 0.282 1.098 0.090 1.332
```

Z, A, U_i, J_i, π_i, KE_i, N_ν, N_γ, …
Neutrons: ε^1_{cm}, θ^1_{cm}, E^1_{lab}, θ^1_{lab}, ε^2_{cm}, θ^2_{cm}, E^2_{lab}, θ^2_{lab},…
Gammas: ε^1_{cm}, θ^1_{cm}, E^1_{lab}, θ^1_{lab}, ε^2_{cm}, θ^2_{cm}, E^2_{lab}, θ^2_{lab}, …

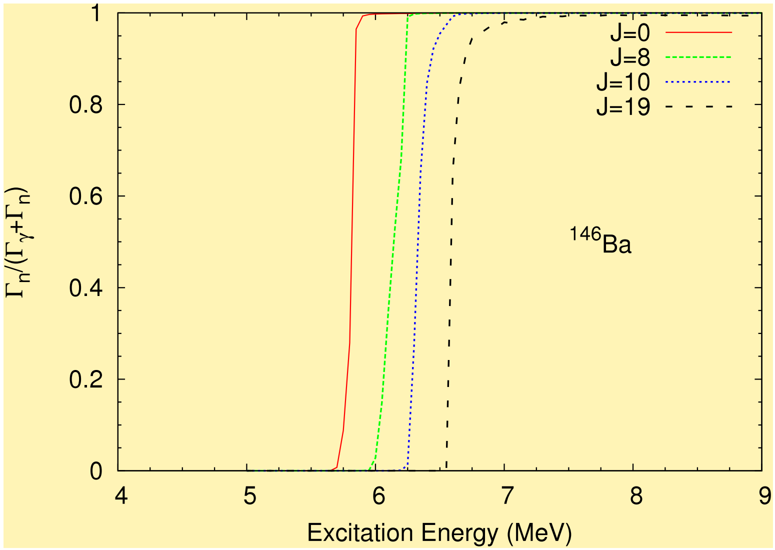
times 100,000 times 2, in this case

Prompt Neutron and Photon Emissions

- **Gamma emission:**
  - Gamma-ray strength function $f_{Xl}(\epsilon_\gamma)$
  - Transmission coefficients:

$$T_{Xl}(\epsilon_\gamma) = 2\pi\epsilon_\gamma^{2l+1} f_{Xl}(\epsilon_\gamma)$$

  - E1, M1, E2 only



- **Neutrons:**
  - Optical model calculations $T_n = 1 - |S_{nn}|^2$
  - Koning-Delaroche (spherical) OMP, 2003
  - Transmission coefficients:

- **Emission Probabilities:**

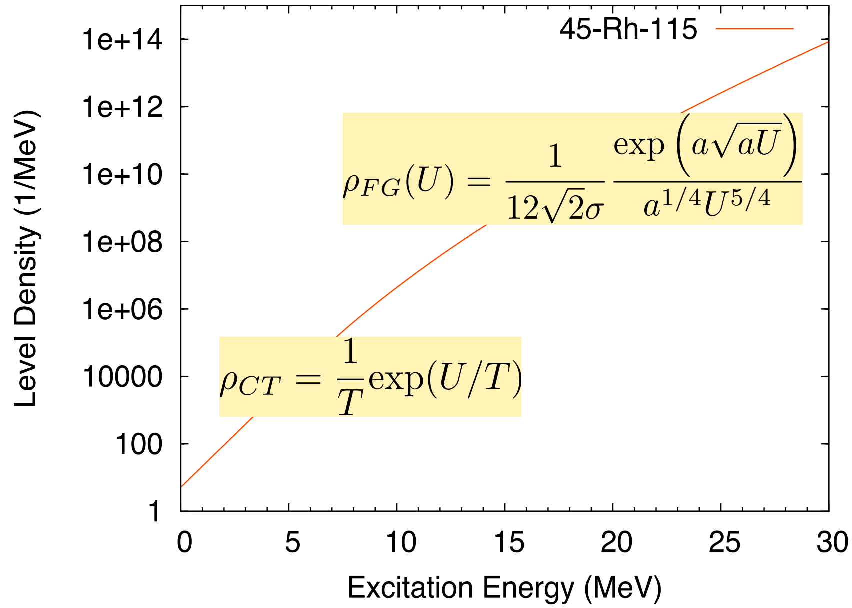
$$P(\epsilon_\gamma)dE \propto T_\gamma(\epsilon_\gamma)\rho(Z, A, E - \epsilon_\gamma)dE$$

$$P(\epsilon_n)dE \propto T_n(\epsilon_n)\rho(Z, A-1, E - \epsilon_n - S_n)dE$$

Level Density & Low-Lying Nuclear Structure

- **Gilbert-Cameron-Ignatyuk**
  - Constant-temperature at low E*
  - Fermi-gas at higher E*

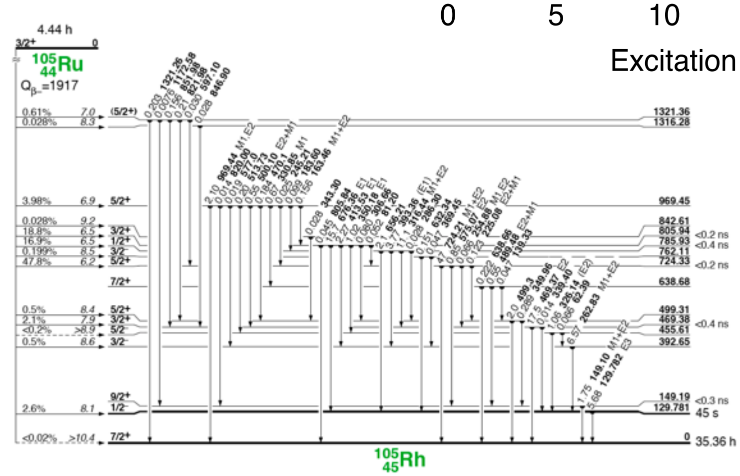
- **Poorly known low-lying levels for many isotopes of interest**



Most probable fission fragment for A=115

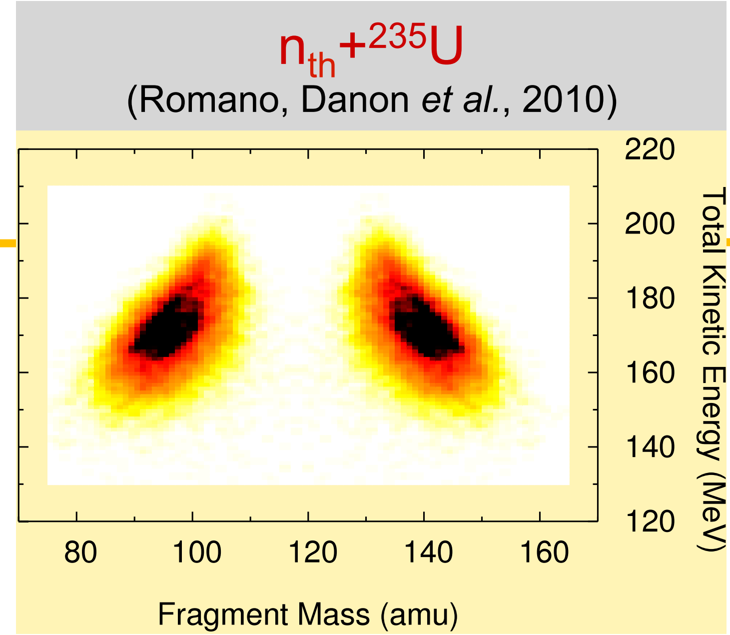
(7/2+) 0.99 s

$^{115}_{45}$Rh β−



Stable isotope ^{105}Rh ?

Primary Fission Fragment Yields



- **Theoretical Predictions:**
  - FRLDM + random walk (Randrup-Möller) + Langevin (Sierk), …
  - HFB: Younes *et al.* (LLNL), Dubray *et al.* (CEA-BRC), …
- **For now, use of experimental data**
  - Y(A,TKE) often inferred from post-neutron emission fragment yields
  - Very limited data available, mostly for thermal neutrons and spontaneous fission
  - Questionable results at higher excitation energies (lack of $<\nu>$(A,TKE) knowledge)
- **Initial conditions:**
  - $\rho_i(U,J,\pi)$
  - Excitation energy partitioning between the two fragments
  - Production of fragment angular momentum

Fission Fragment Initial Conditions in **Excitation Energy** and **Angular Momentum**

- **Initial Excitation Energies**
  - For a given fragment pair $$TXE = Q - TKE = \left(E^*_{int} + E^*_{def} + E^*_{\perp}\right)_{L,H}$$
  - Sharing of TXE between light and heavy fragments → $\boxed{R_T} = T_L/T_H$

- **Initial Angular Momentum Distributions**
  - Total angular momentum conservation $$\vec{J}_{tot} = \vec{J}_L + \vec{J}_H + \vec{l}$$
  - Initial distribution in fragments:

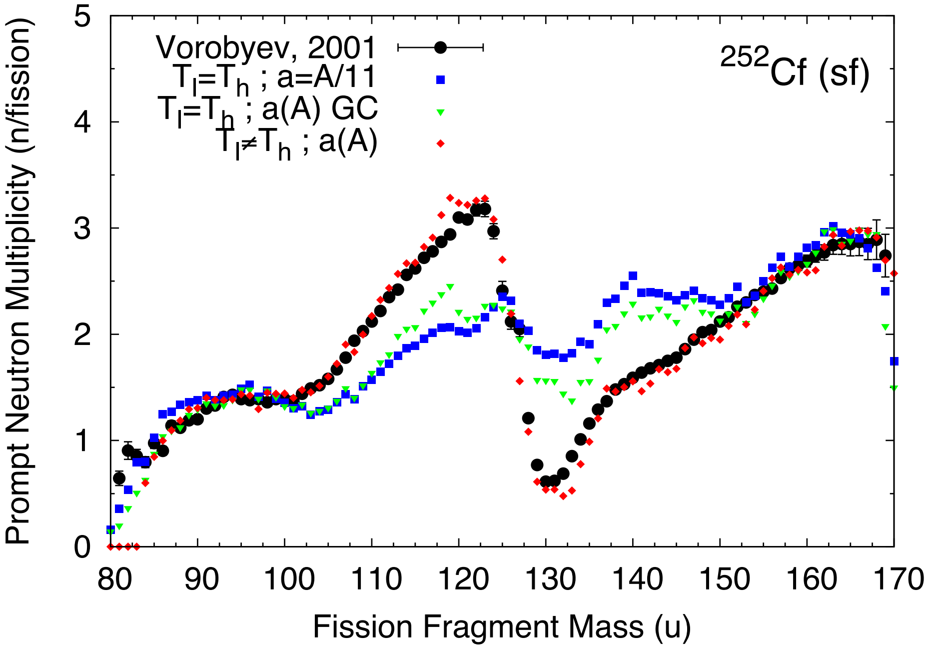
$$P(J) \propto (2J+1)\exp\left[-\frac{J(J+1)}{2B^2(Z,A,T)}\right]$$

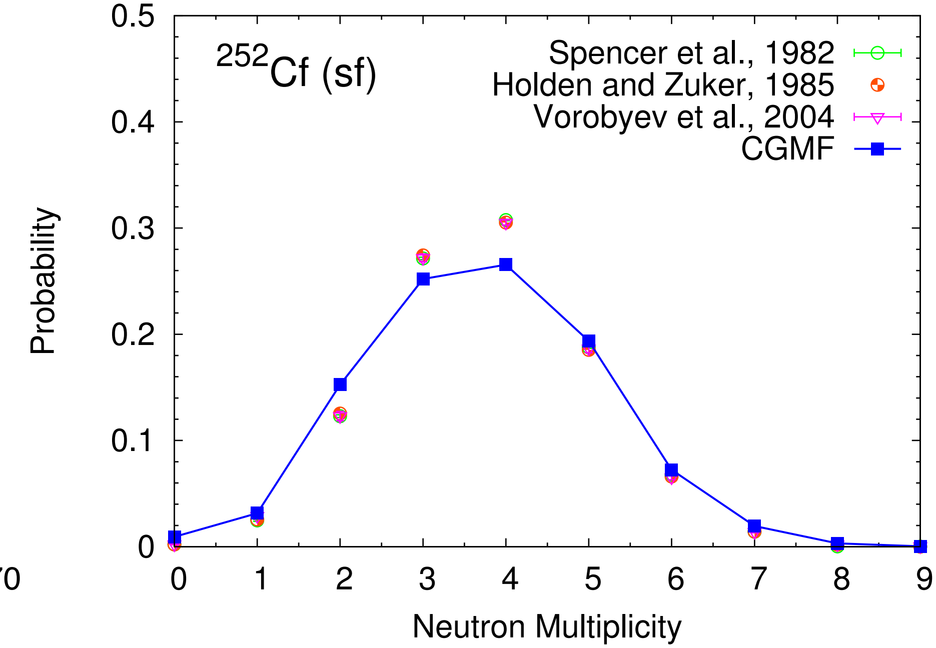
with $B^2(Z,A,T) = \boxed{\alpha}\frac{\mathcal{I}_0(A,Z)T}{\hbar^2}$

I_0 is the moment of inertia of the fragment (A,Z) in its ground-state.

Prompt Neutron Multiplicity <ν>(A) and P(ν)

Example of ^{252}Cf (sf)

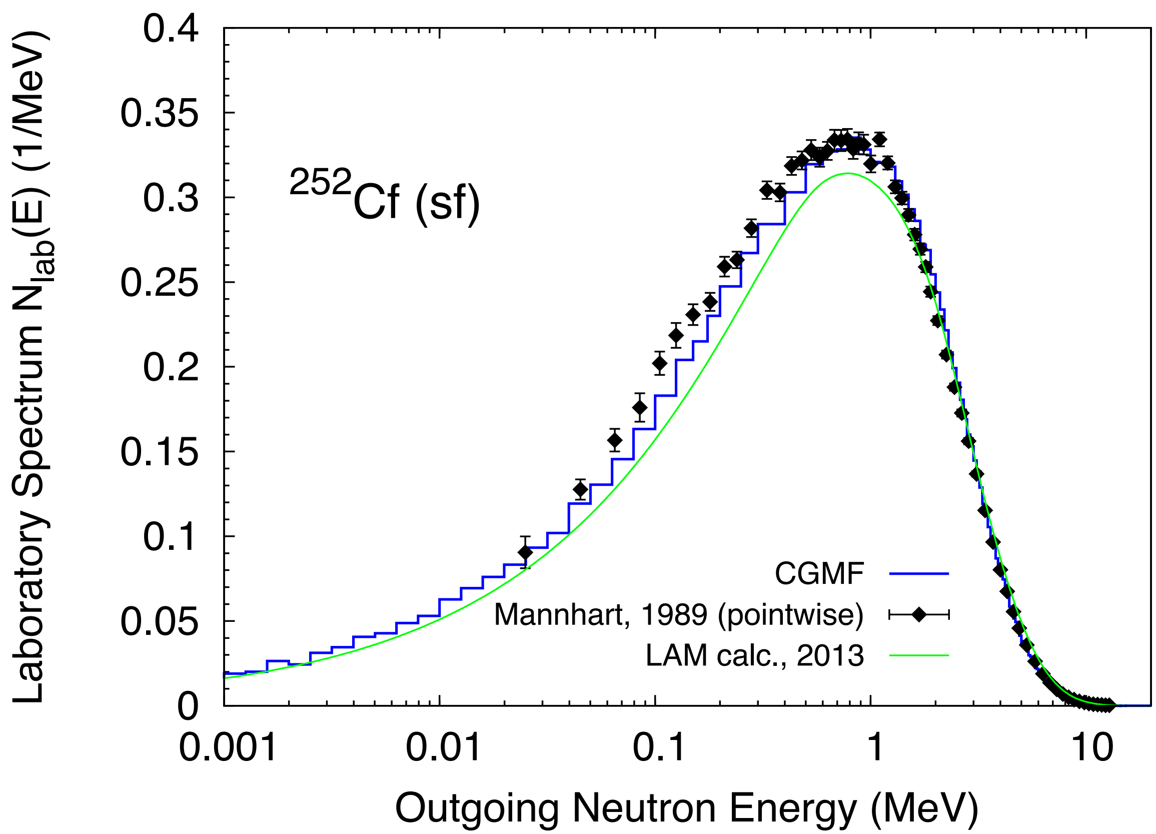




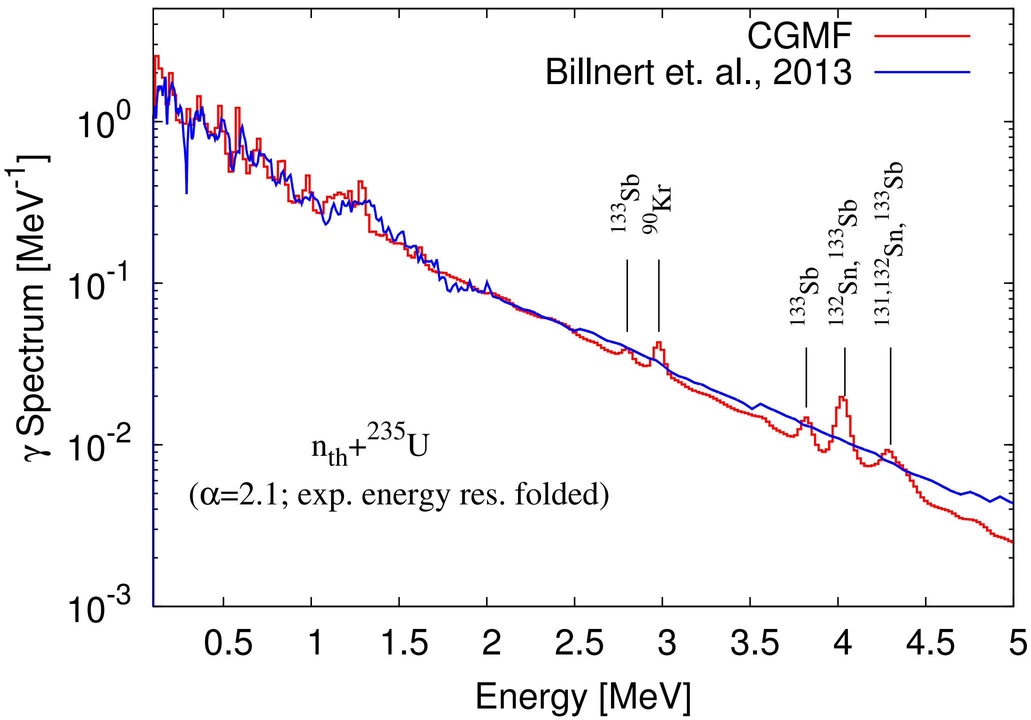
$<\nu>_{calc}=3.78$ vs. $<\nu>_{std}=3.7606$

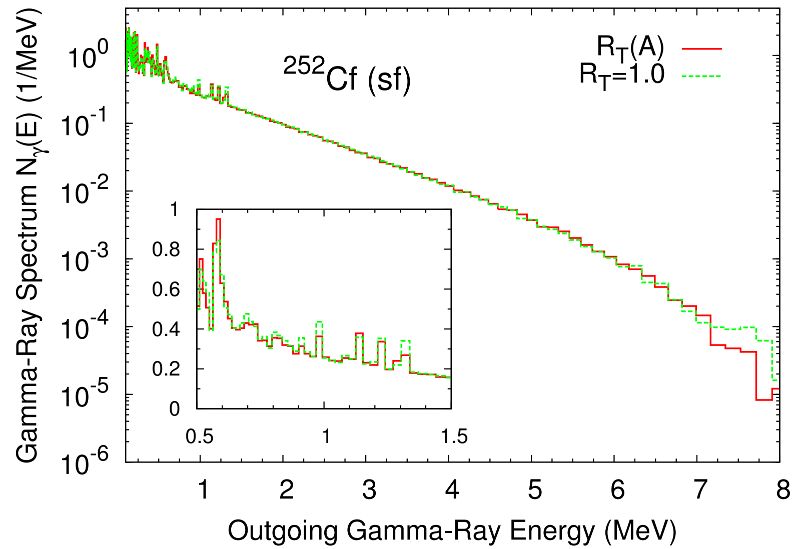
Average Prompt Fission Neutron Spectrum

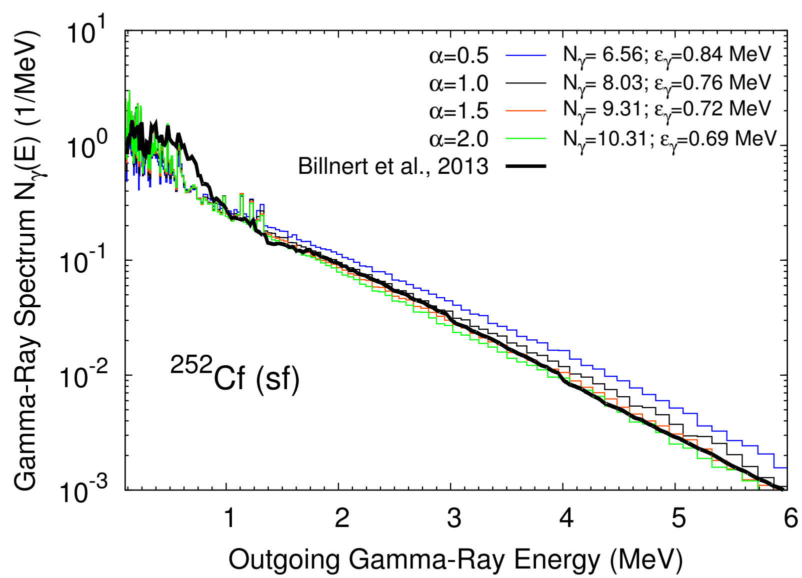
- ^{252}Cf (sf) PFNS is a “**standard**” (Mannhart, 1989)
- Difficulty to reproduce low-outgoing energy tail
- CGMF calculations better at low-energy but too soft at high energies



Prompt Photon Spectrum

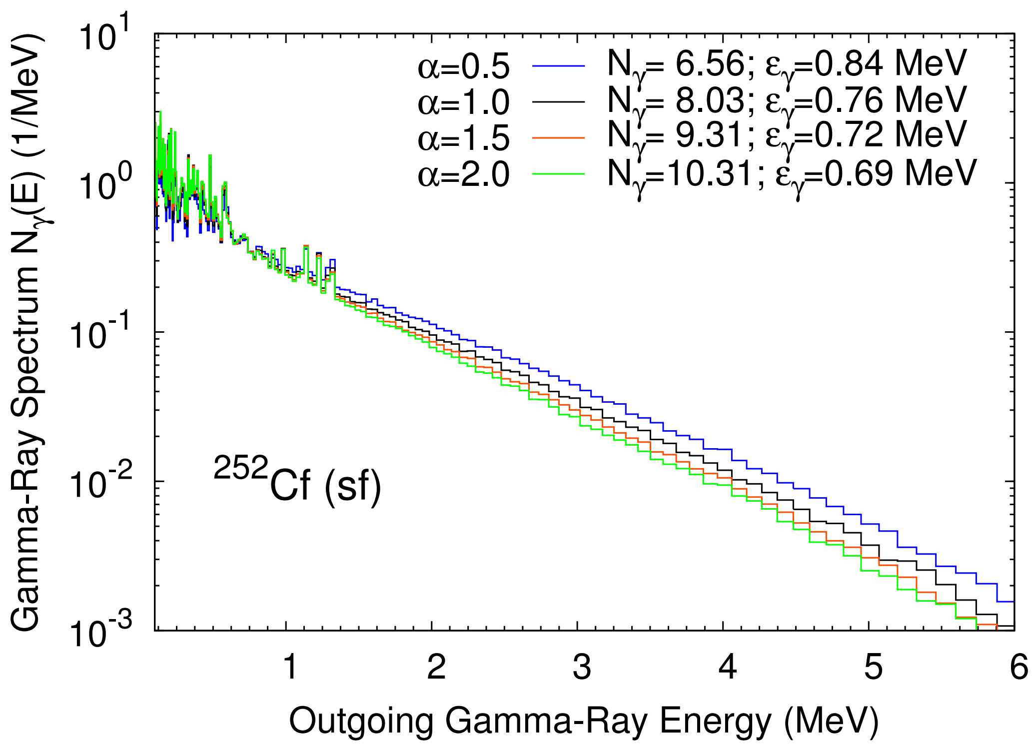


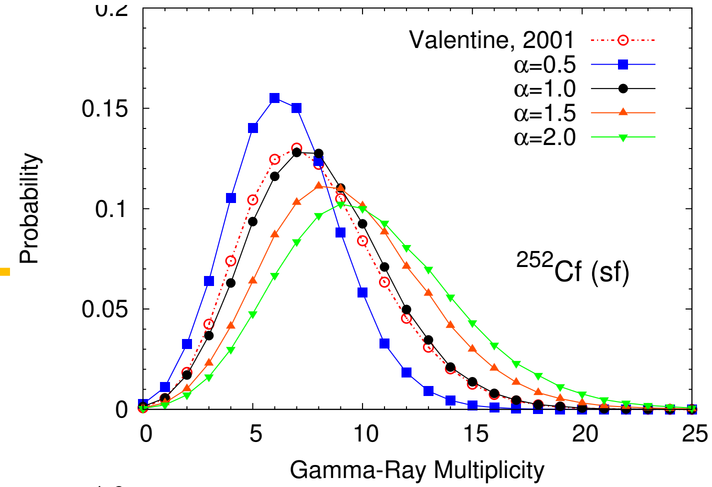


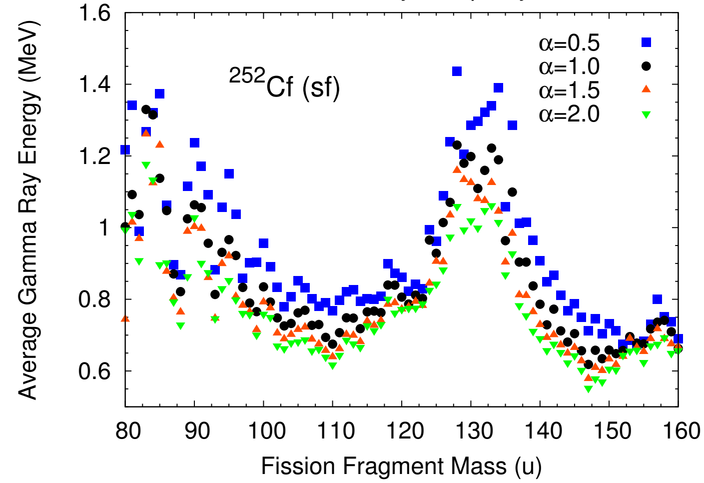


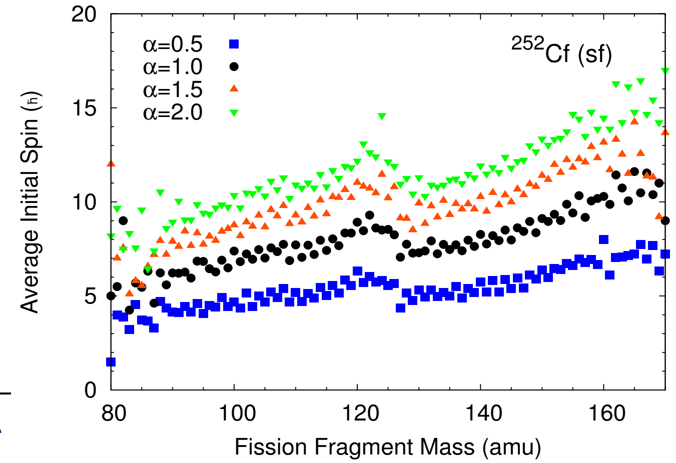
Impact of α (<J$_i$>)

- **The α-parameter impacts directly the spin-dependent initial population of the fragments.**





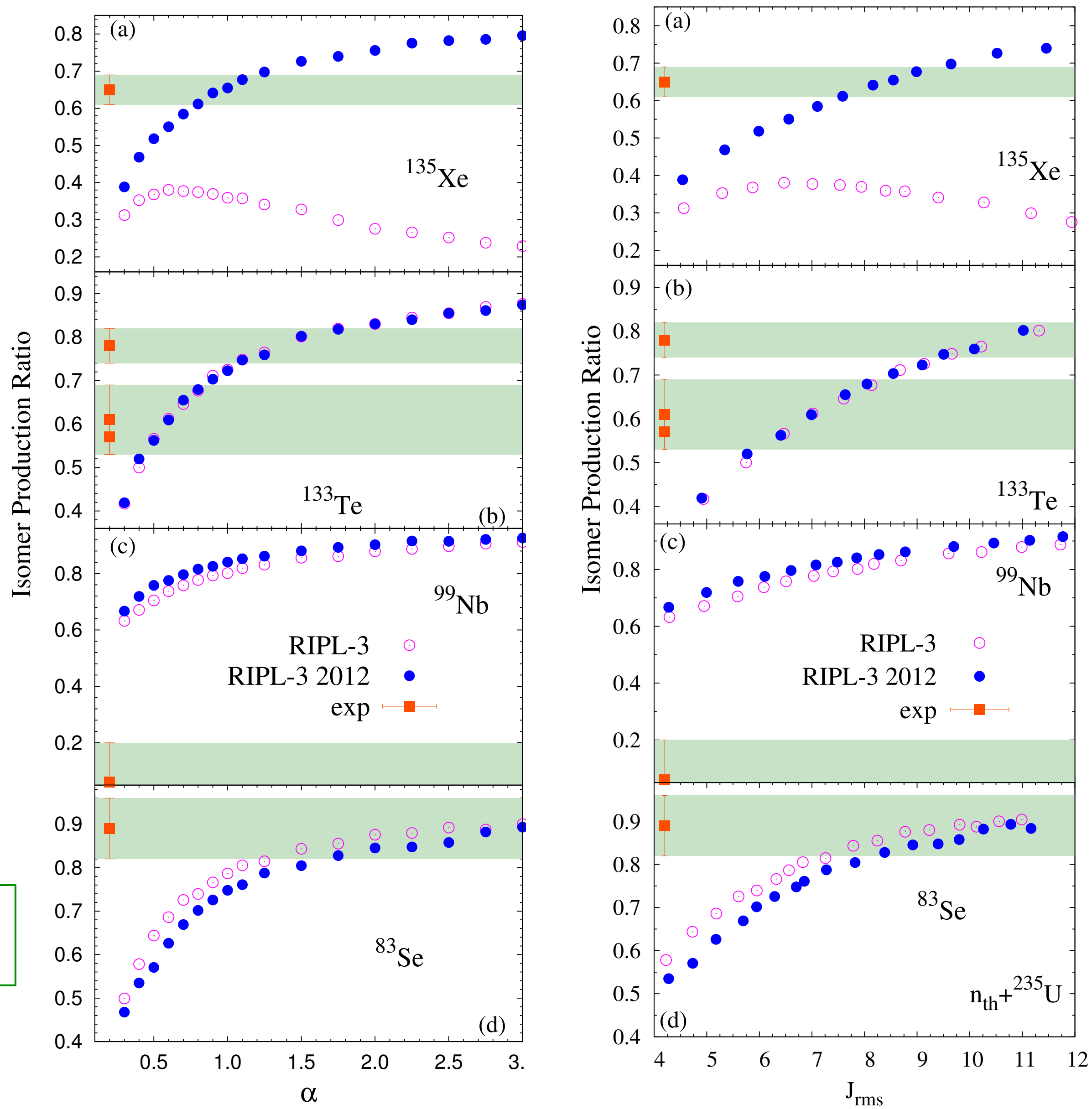


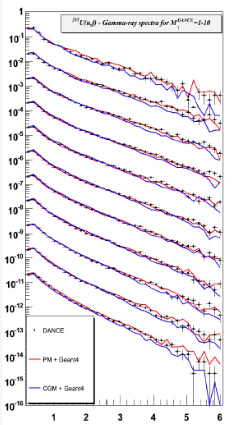


Isomeric Ratios

- Using measured ratios of isomer to ground-state to infer initial J_{rms}
- Very mixed results
- Very sensitive to (often unknown) detailed nuclear structure

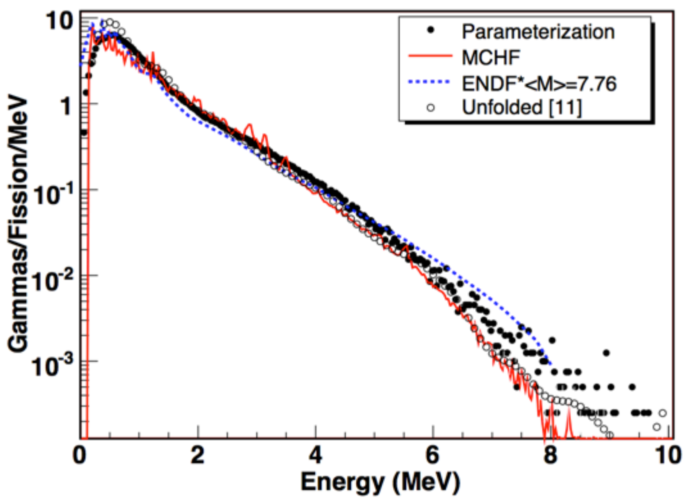
Stetcu, Talou, Kawano, Jandel, to appear in Phys. Rev. C (2013)



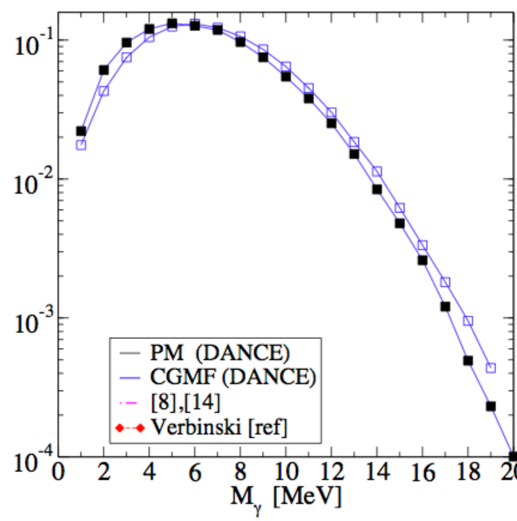


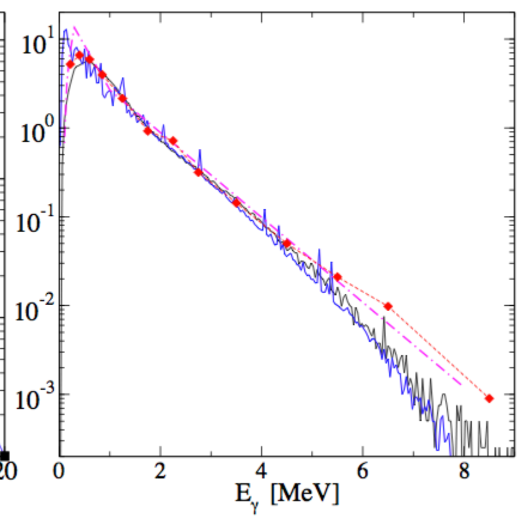
Prompt Gamma Rays in DANCE

^{239}Pu (n,f)



^{235}U (n,f)





J.Ullmann *et al.*,
"Prompt Gamma-Ray Production in Neutron-Induced Fission of 239Pu", Phys. Rev. C **87**, 044607 (2013).

M.Jandel *et al.*,
"Prompt Gamma-Ray Emission in Neutron-Induced Fission of 235U," to be submitted to PRC.

Final Remarks

- **A lot of research work on nuclear fission being performed**
- **For both fundamental research and applied needs**
- **Some examples:**
  - **Fission Cross Sections**
    - Fundamental: determine fission probabilities from robust determination of fission paths characteristics (level densities, transition states, K-mixing, etc.)
    - Applied: need predictive, consistent, and accurate capabilities (strong impact on integral benchmarks)
  - **Fission Fragment Yields**
    - Fundamental: stringent tests for static and dynamic fission models
    - Applied: fission product yields as a diagnostic for Pu burnup
  - **Prompt Fission Neutrons and Photons**
    - Fundamental: compound nucleus physics, dynamic vs. evaporation, scission neutrons?
    - Applied: strong impact on benchmarks (part of the compensating error picture)

LANL Collaborators

- T-2:
  T.Kawano, I.Stetcu, P.Möller, A.J.Sierk, A.C.Hayes, J.E.Lynn*, D.G.Madland*
- X-CP:
  M.B.Chadwick, M.C.White, J.Lestone, M.E.Rising
- LANSCE-NS:
  F.Tovesson, A.Laptev, R.Meharchand, R.C.Haight, H-Y. Lee, A.Couture, S. Mosby
- C-NR:
  M.Jandel, J.Wilhelmy*

**LANL Consultants*