Realistic Reaction Evaluations for Fission Products Off Stability

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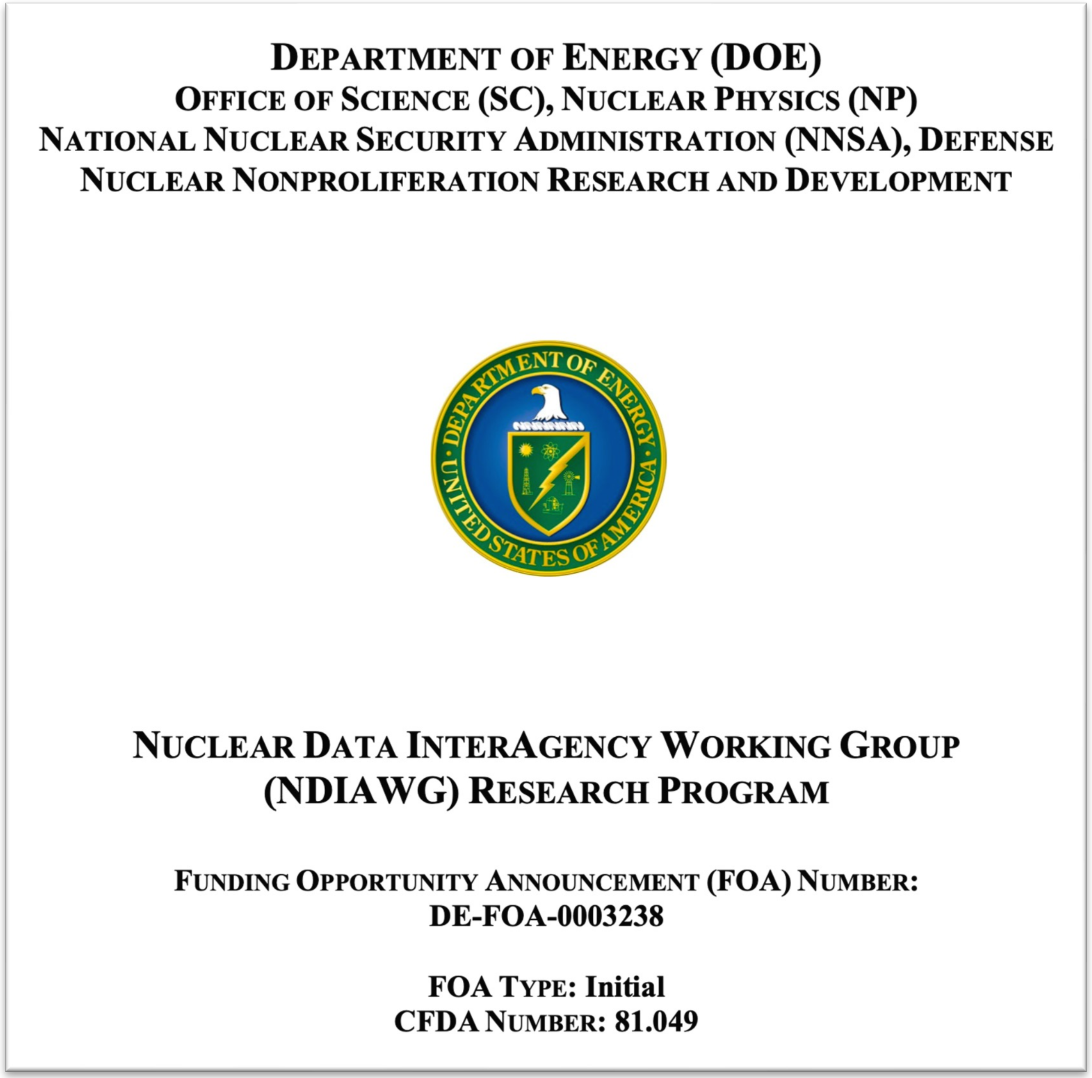
Outline

- Project Impact and Goals
- Team Introduction
- Planned Methodology
    - Fast region
    - Resonance region
    - Experimental component
- Status and perspectives


DEPARTMENT OF ENERGY (DOE)
OFFICE OF SCIENCE (SC), NUCLEAR PHYSICS (NP)
NATIONAL NUCLEAR SECURITY ADMINISTRATION (NNSA), DEFENSE NUCLEAR NONPROLIFERATION RESEARCH AND DEVELOPMENT

NUCLEAR DATA INTERAGENCY WORKING GROUP (NDIAWG) RESEARCH PROGRAM

FUNDING OPPORTUNITY ANNOUNCEMENT (FOA) NUMBER: DE-FOA-0003238

FOA TYPE: Initial
CFDA NUMBER: 81.049

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Project impact on NA-22 and other programs

- Applications such **nonproliferation, post-detonation forensics, spent-fuel assay, reactor burnup and design**, as well as **astrophysics**, rely on the accurate description of the neutron interaction with unstable fission products.
- Current cross-section descriptions of these nuclei are either non-existent or based on simplified assumptions, leading to unquantified impacts on predicted cross-sections.
- By project completion, more **predictive/realistic** new nuclear data will be produced, improving the **reliability** of applications involving **fission products off stability!**

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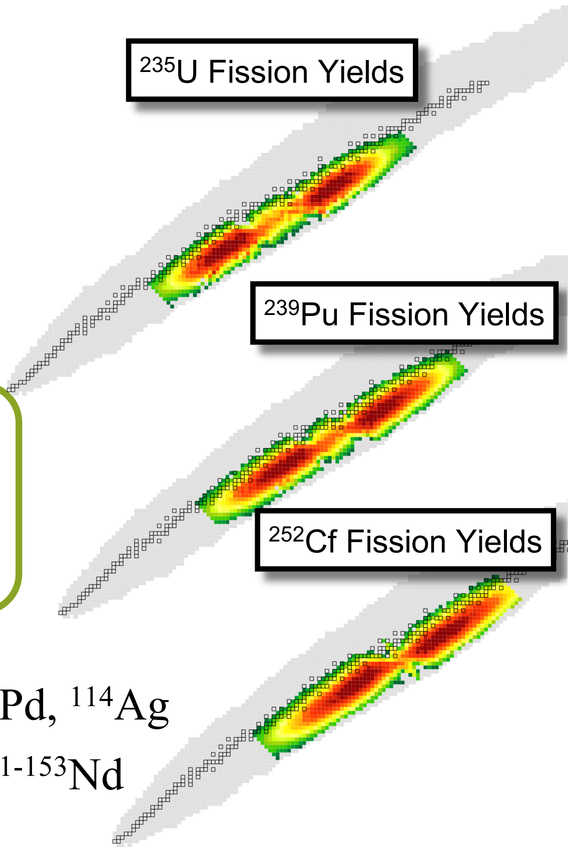
Project Goals and Objectives



To provide evaluated files for neutron-induced reactions on fission products off-stability and submit them to the ENDF/B nuclear data library

The **primary-goal nuclei** (mostly produced by ^{235}U fission):

- <u>1st Fission yield bump</u>: $^{87-89}$Br, $^{88-92}$Kr, $^{91-94}$Rb, $^{92-97}$Sr, $^{95-99}$Y, $^{97-102}$Zr, $^{101-103}$Nb
- <u>2nd Fission yield bump</u>: $^{131-133}$Sb, $^{132-136}$Te, $^{135-138}$I, $^{136-141}$Xe, $^{139-143}$Cs, $^{141-146}$Ba, $^{144-145}$La, $^{147-148}$Ce

Secondary goal (main fission products from ^{239}Pu and ^{252}Cf):

- <u>1st Fission yield bump</u>: ^{94,100}Y, ^{96,103}Zr, ^{99,100,104,105}Nb, $^{102-108}$Mo, $^{105-110}$Tc, $^{107-112}$Ru, $^{110-114}$Rh, $^{112-116}$Pd, ^{114}Ag
- <u>2nd Fission yield bump</u>: ^{131}Te, ^{134}I, ^{135}Xe, ^{137,138,144}Cs, ^{140}Ba, $^{143,146-148}$La, ^{145,146,149,150}Ce, $^{149-152}$Pr, $^{151-153}$Nd

Stretch goal (whole isotopic chain of fission products from ^{235}U, ^{239}Pu, and ^{252}Cf):

- ^{66}V, $^{66-67}$Cr, $^{66-71}$Mn, $^{66-75}$Fe, $^{66-77}$Co, $^{66-80}$Ni, $^{66-82}$Cu, $^{66-85}$Zn, $^{68-87}$Ga, $^{70-90}$Ge, $^{72-92}$As, $^{75-95}$Se, $^{77-98}$Br, $^{79-101}$Kr, $^{81,83-103}$Rb, $^{83-106}$Sr, $^{87-109}$Y, $^{88-112}$Zr, $^{91-114}$Nb, $^{93-117}$Mo, $^{97-119}$Tc, $^{98-121,124}$Ru, $^{101-125}$Rh, $^{103-126,128}$Pd, $^{106-132}$Ag, $^{108-134}$Cd, $^{111-137}$In, $^{113-139}$Sn, $^{118-140}$Sb, $^{120-143}$Te, $^{123,125,126,128-145}$I, $^{125,128,130-148}$Xe, $^{131-151}$Cs, $^{132-153}$Ba, $^{135,137-155}$La, $^{137-157}$Ce, $^{139-159}$Pr, $^{142-161}$Nd, $^{144-163}$Pm, $^{147-165}$Sm, $^{149,151-168}$Eu, $^{152-170}$Gd, $^{155-172}$Tb, $^{157-172}$Dy, $^{161-172}$Ho, $^{162-172}$Er, $^{165-172}$Tm, $^{168-172}$Yb, $^{171-172}$Lu

<u>Project will be successful if primary goal is achieved</u>. *However*, when the methods are well-stablished, generalization to secondary and stretch goals should be possible with relative low effort.

Team Introduction

	Team Member	Past/Current Leveraging Activities	Project Role
	Gustavo Nobre (BNL, PI)	• Led many previous evaluations, on and off stability • Reaction model developer with many published works related to deformed nuclei, predictive models, and machine learning. • ENDF/B library manager and EMPIRE co-developer	• Project Coordination • Lead fast region calculations, mentoring postdoc • Complete evaluated file assembly and submit to ENDF/B library
	David Brown (BNL)	• Extensive work and experience on resolved and unresolved resonances and analytical methods related cross-section probabilities and synthetic resonance generation. • ENDF evaluator and NNDC and CSEWG chair	• Lead resonance treatment • Implement transitions between different energy regions
	Kyle Wendt (LLNL)	• Theoretical nuclear physicist with extensive experience in modeling low energy phenomena. • Lead develop on nuclear data UQ suite at LLNL • Theory/AI team co-lead on SI-LDRD on ML for nuclear data	• Lead ML effort to provide cross-section priors off-stability for threshold reactions
	Alexander Voinov (OU)	• Experimentalist with extensive experience in nuclear level density measurements	• Perform experiments and data analysis for stable nuclei in the mass region of fission products
	Aman Sharma (LLNL, postdoc)	• Postdoc working on LLNL SI-LDRD on ML for nuclear data • Experience on both experimental and theoretic physics, with an emphasis on UQ in both context. • Has conducted experiments and evaluations as PhD student.	• Will work with Gustavo Nobre to learn about EMPIRE and to perform most of fast-region calculations.
	Shusen Liu (LLNL)	• Machine intelligence scientist with extensive experience on signal modeling and interpretable machine learning. • Theory/AI team co-lead on SI-LDRD on ML for nuclear data.	• Will adapt the LLNL ML to specific needs of this project.
	Emanuel Chimanski (BNL)	• Evaluator and model developer, with published works on microscopic models and preequilibrium • EMPIRE co-developer	• Fast-region preequilibrium modeling

Donny Hornback: BNL NA-22 POC

Approach in fast region

Which nuclei are we focusing on in this project?

- <u>1st Hump</u>: $^{87-89}$Br, $^{88-92}$Kr, $^{91-94}$Rb, $^{92-97}$Sr, $^{95-99}$Y, $^{97-102}$Zr, $^{101-103}$Nb
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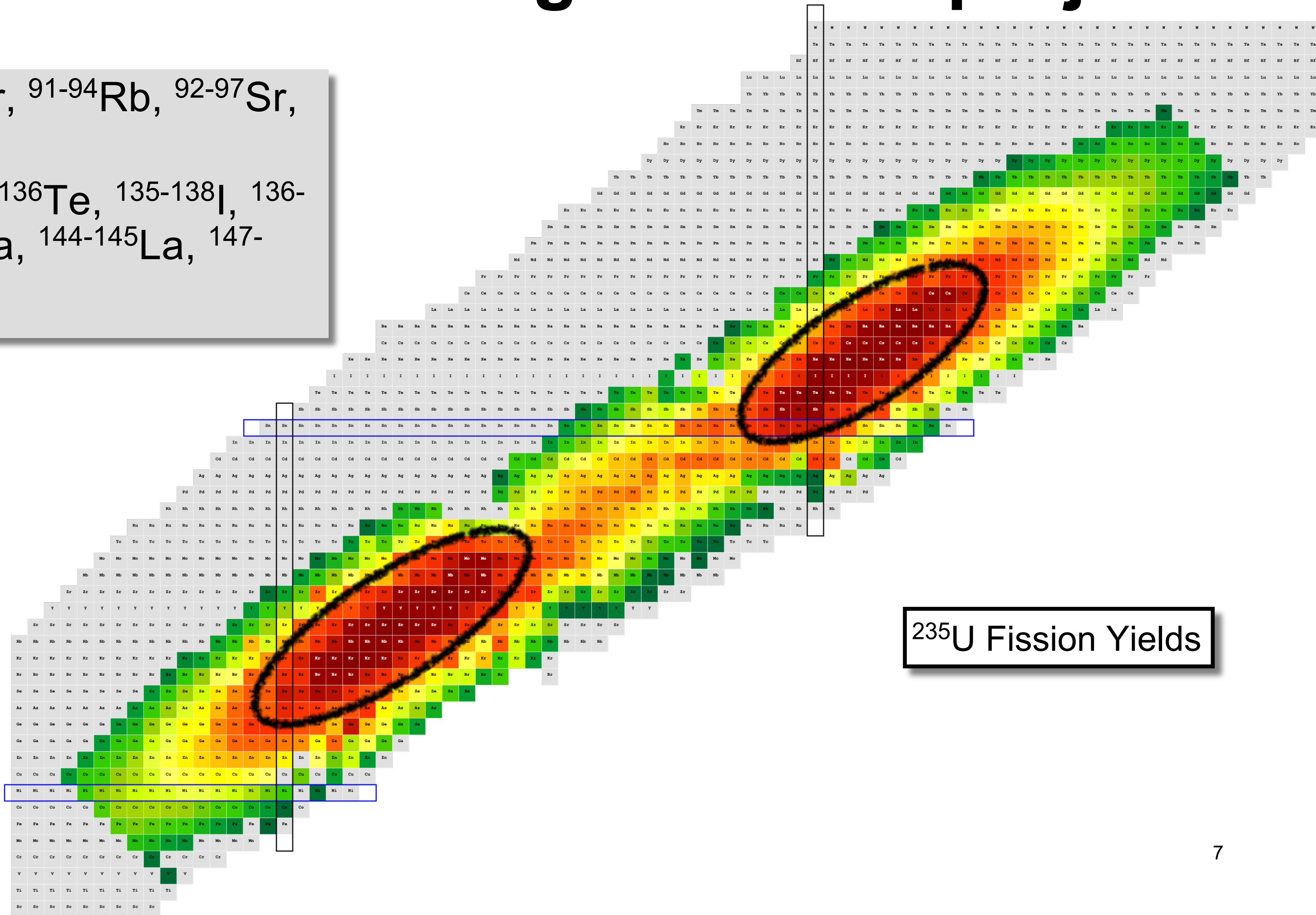

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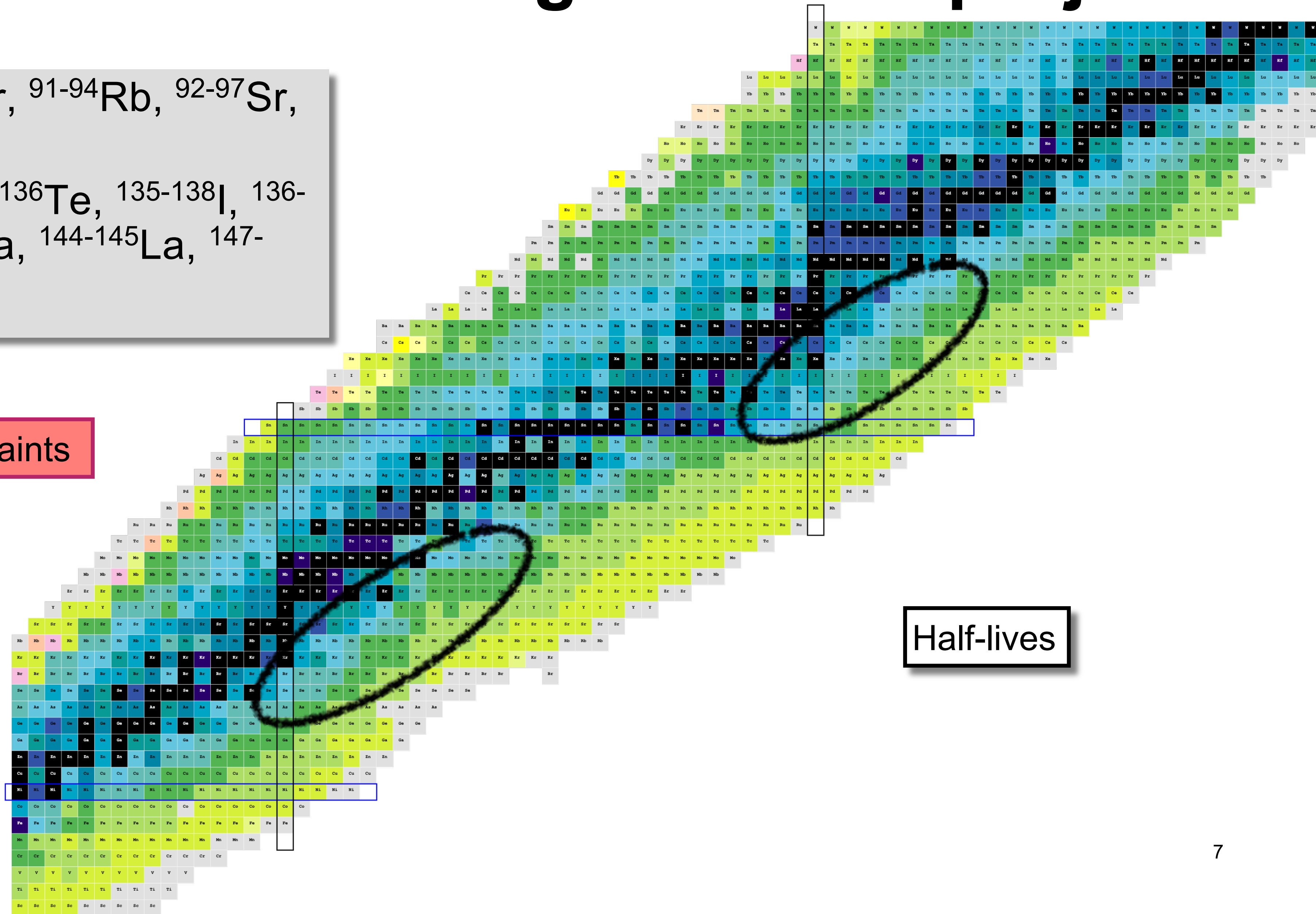
Off-stability = few data constraints



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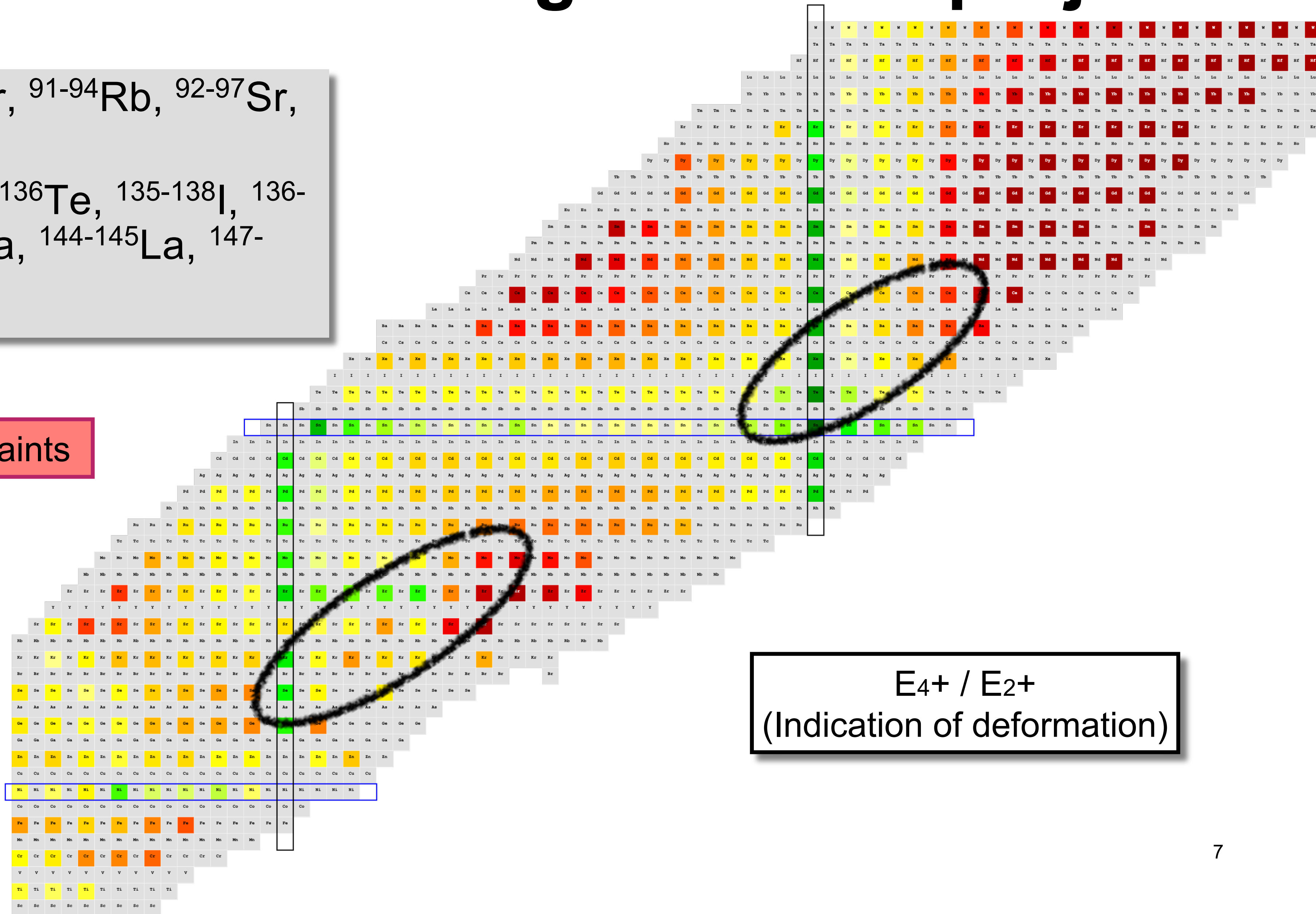
Many are (highly-)deformed!



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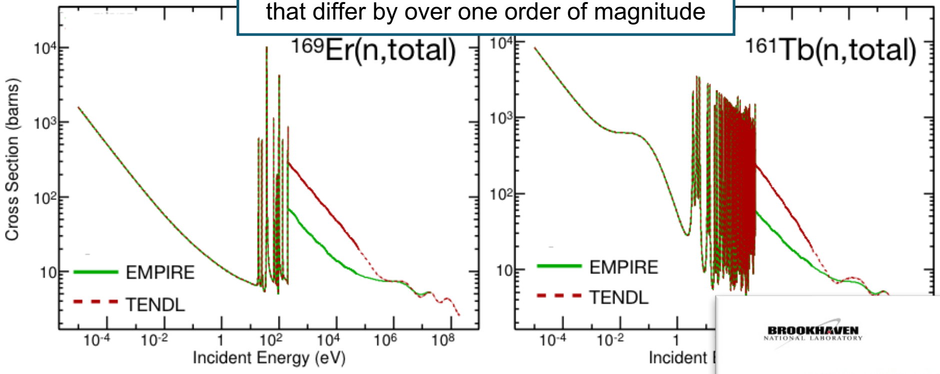
Leveraging previous experience with deformed nuclei

- "Predictive" adiabatic coupled-channels approach - takes as input:
  - A global/regional spherical OMP
  - Deformation parameters
- Deformation treatment changes total/elastic cross sections by orders of magnitude
- Indirectly impacts capture

Different models can predict cross sections that differ by over one order of magnitude



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Derivation of an optical potential for statically deformed rare-earth nuclei from a global spherical potential

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Comparison between EMPIRE and TENDL

G. P. Nobre

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USDOE Office of Science (SC), Nuclear Physics (NP) (SC-26)

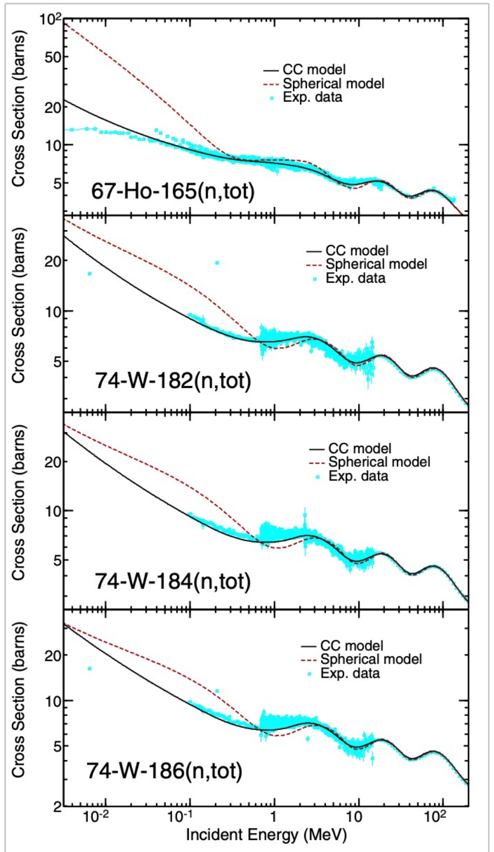
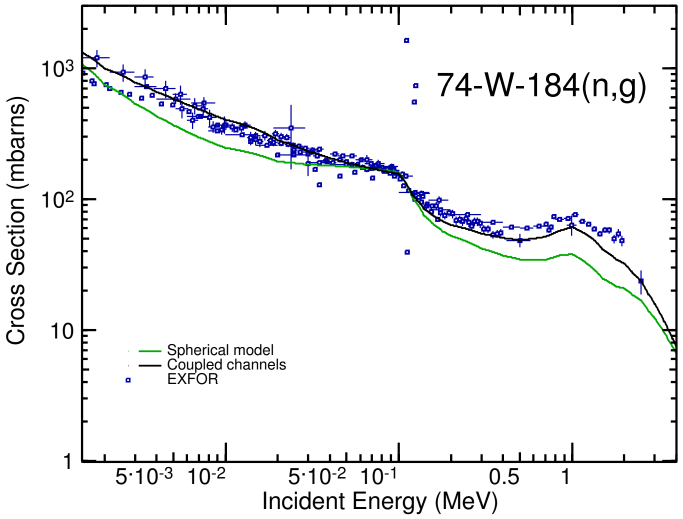
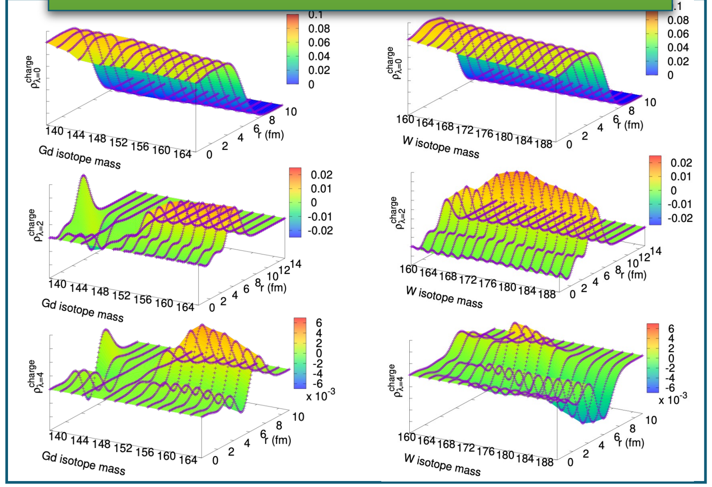


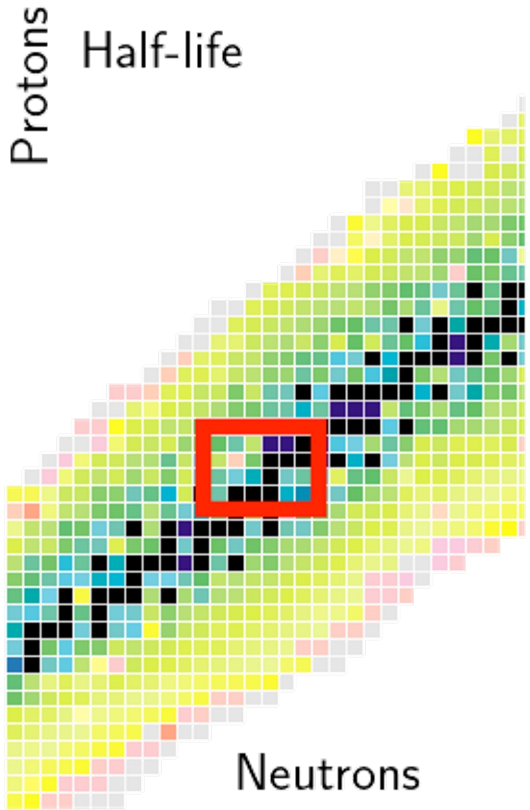
FIG. 1. (Color online) Total cross sections for neutrons scattered by a ^{165}Ho and ^{182,184,186}W targets for incident energies ranging from as low as ≈3 keV to as high as 200 MeV, which is the upper limit of validity for the KD optical potential [2]. The solid black curves correspond to the predictions of our CC model, while the dashed red curves are the results of calculations within the spherical model. The experimental data were taken from the EXFOR nuclear data library [39].

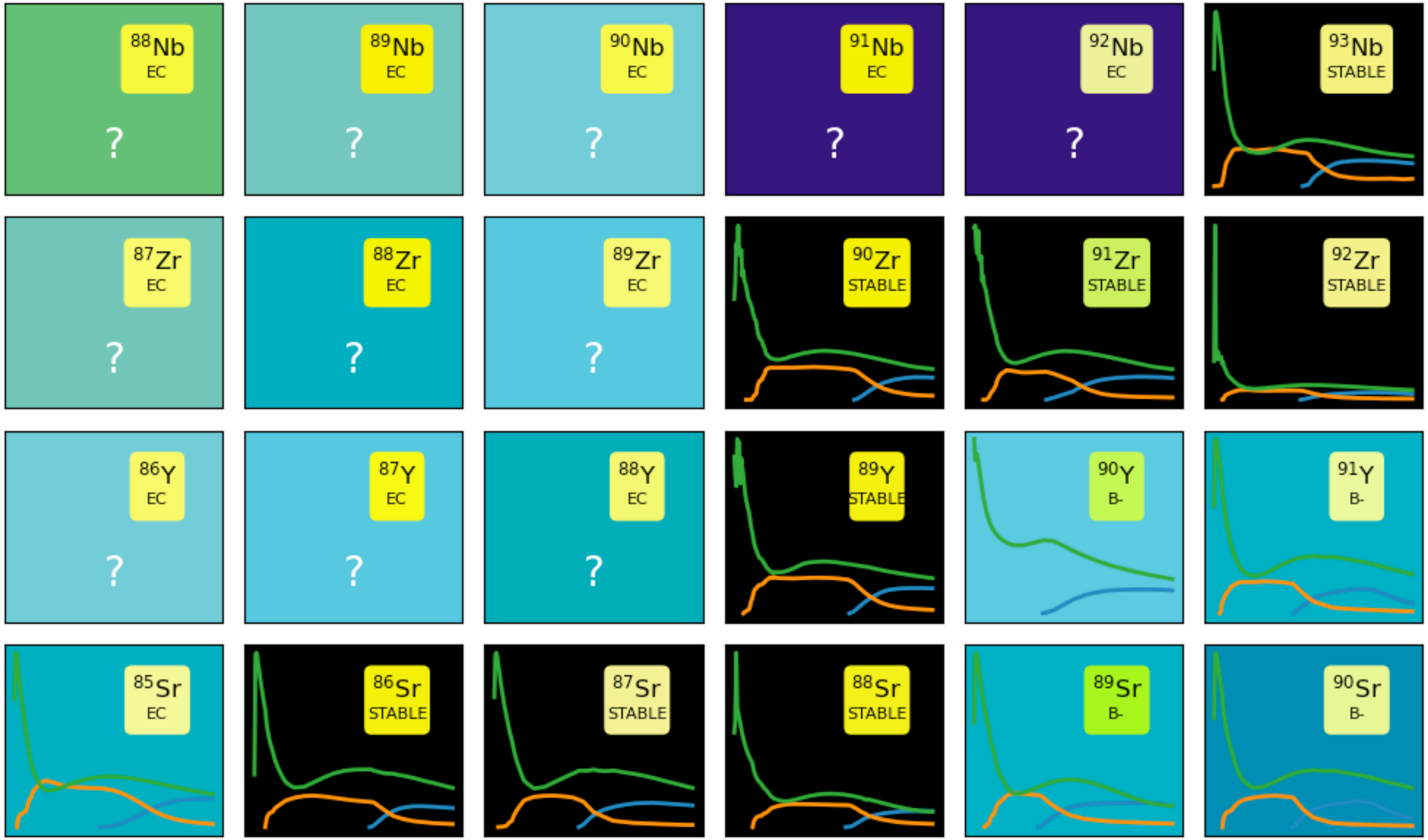


Previous exploration into extending approach to rare-earths off stability



We will also leverage existing calculations and measurements to learn how cross sections transform across the nuclear chart





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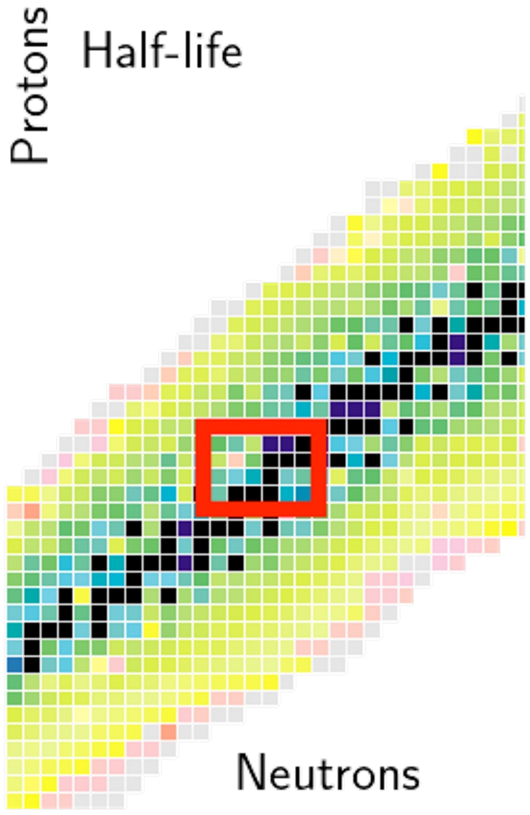

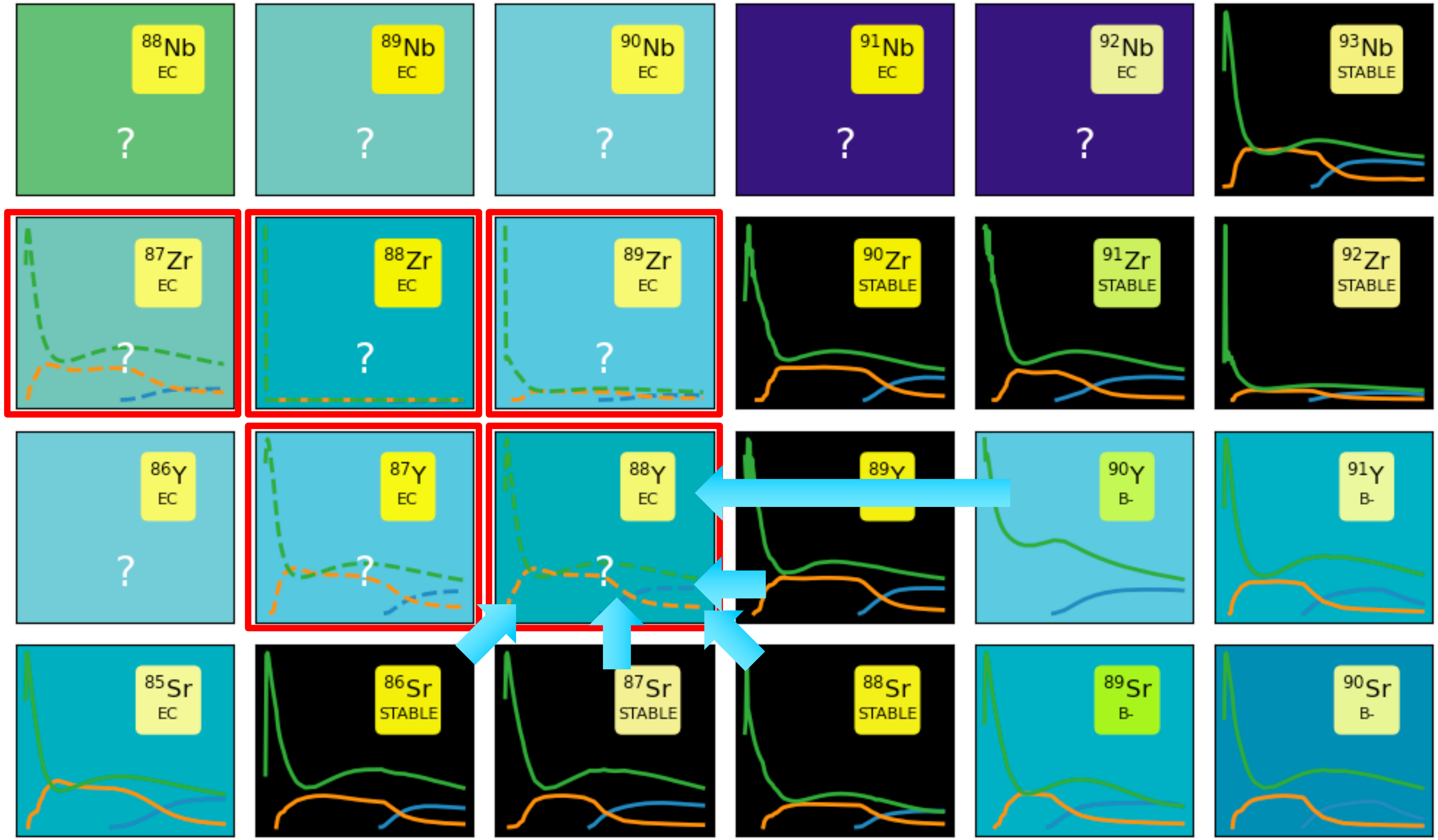

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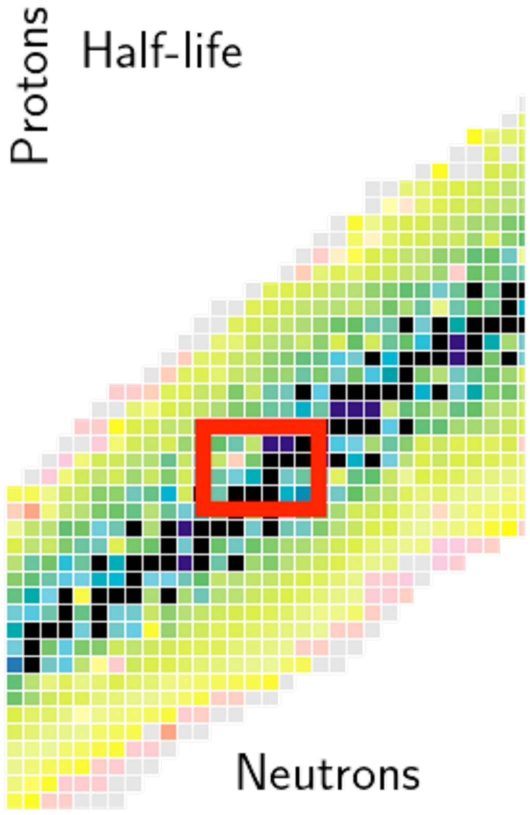
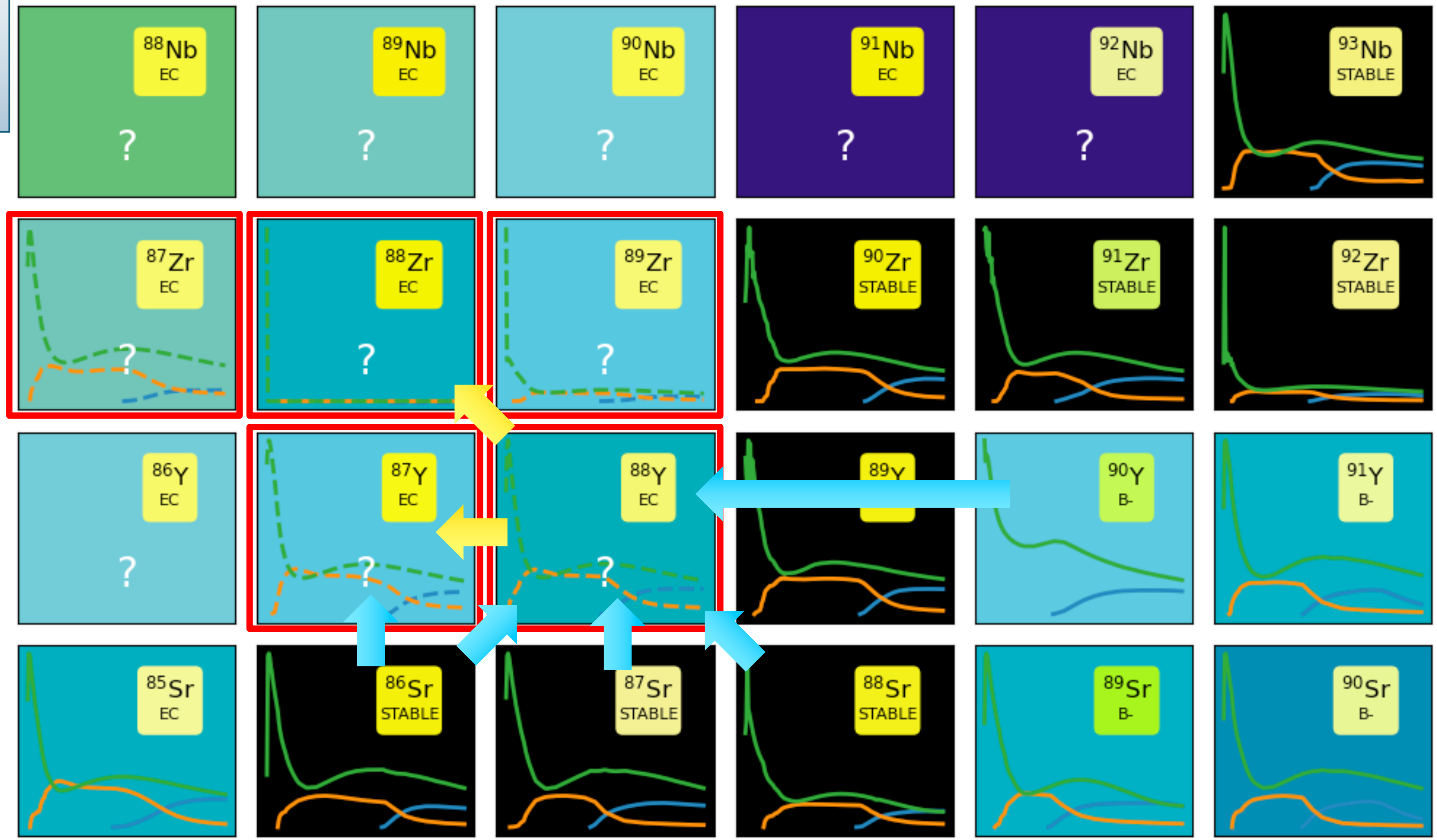
Aim to obtain “systematic” priors for evaluations on unstable nuclei.

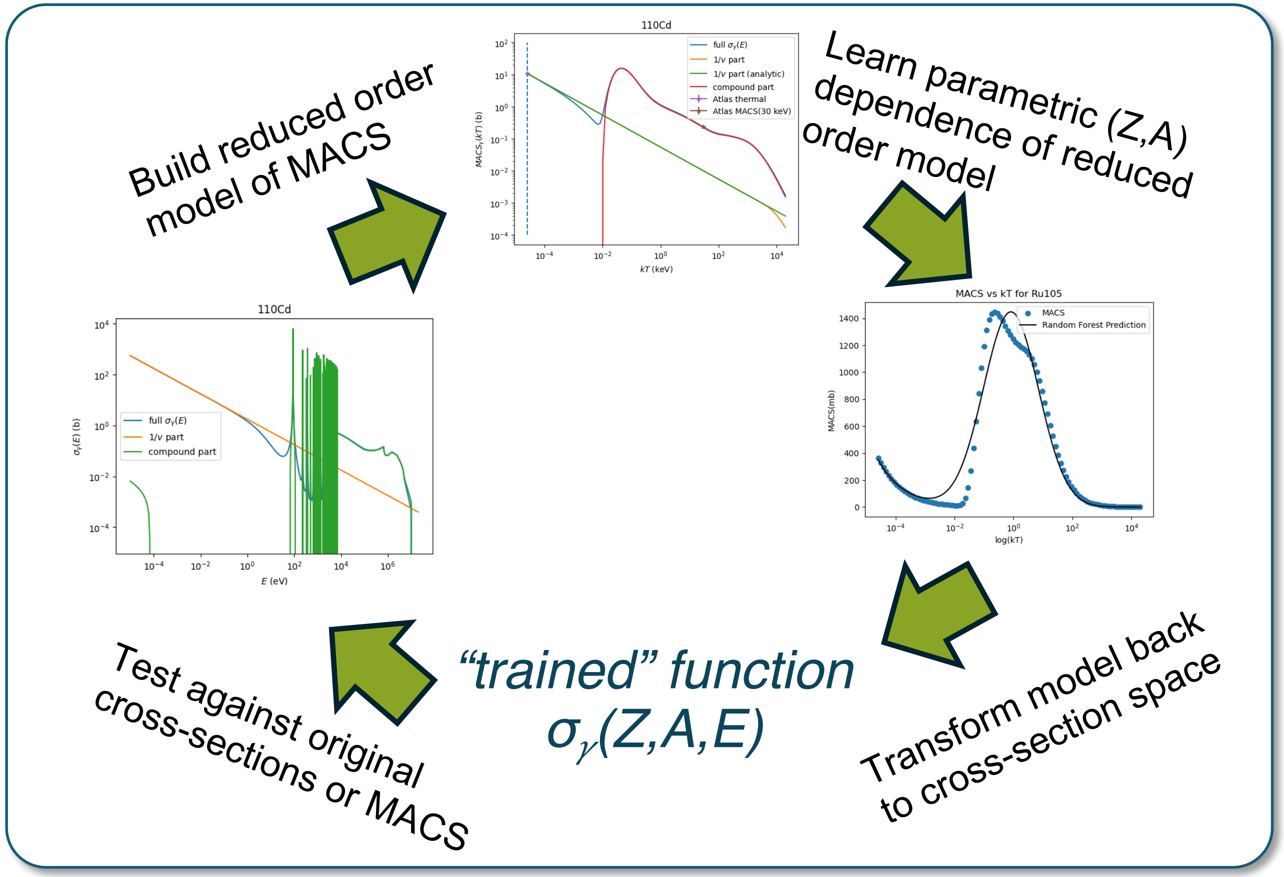




Approach in resonance region

Capture cross-section averages & fluctuations

- There are **too many** resonances.
- It is not possible to predict their position or width
- We focus on an "*average*" cross section and some *probability distribution* that captures the size of fluctuations



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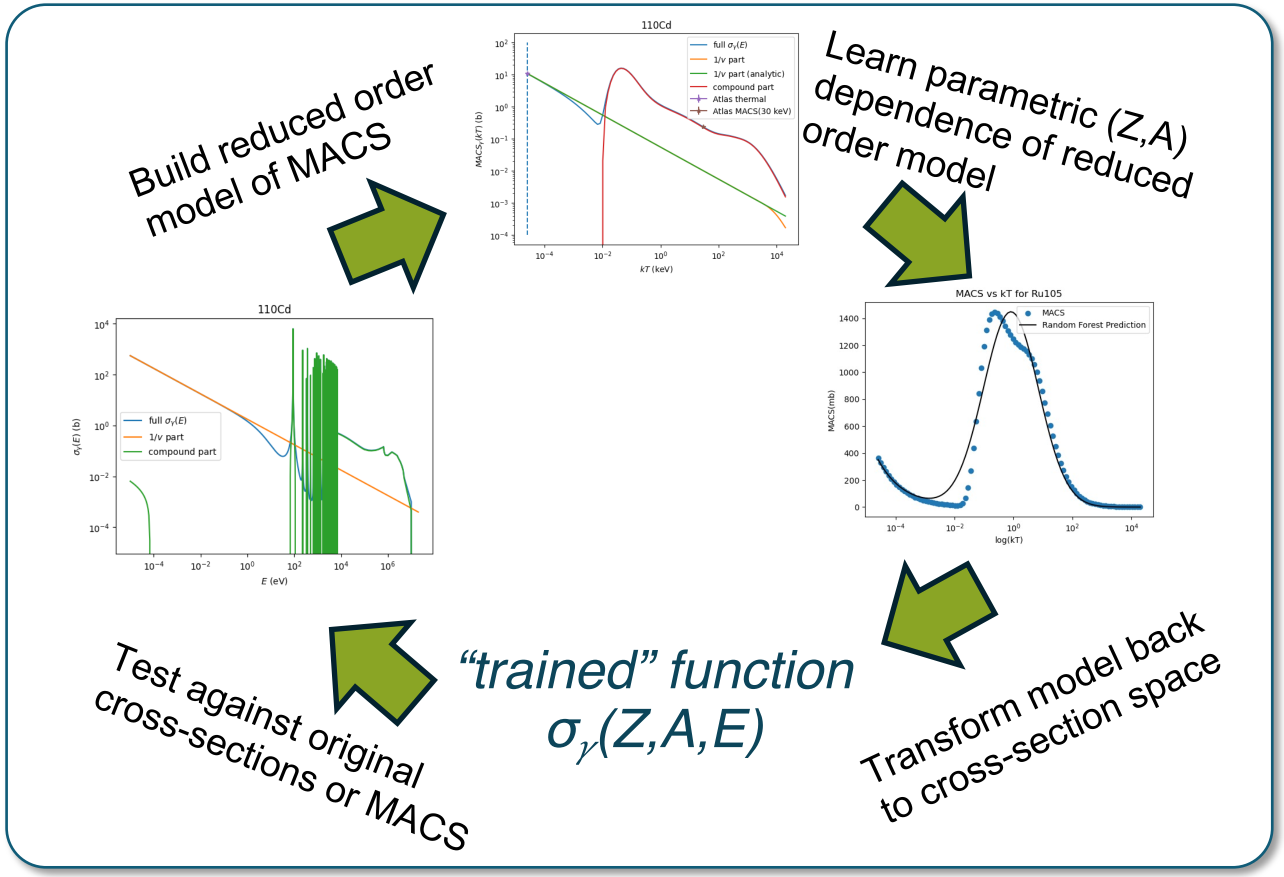

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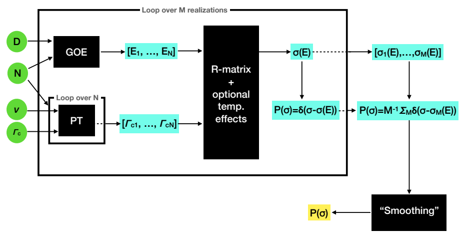
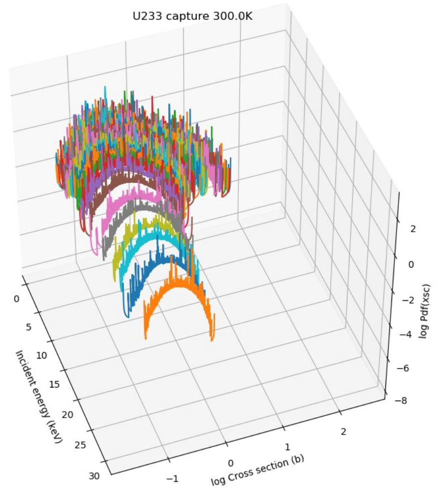
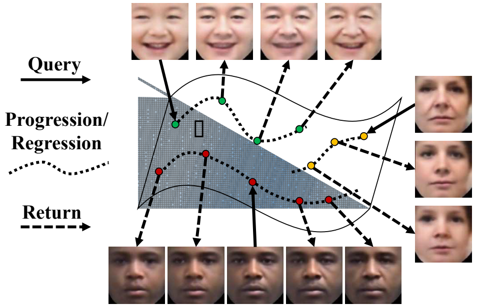


Use FUDGE as a generative model to simulate cross-section probability distribution function (PDF)

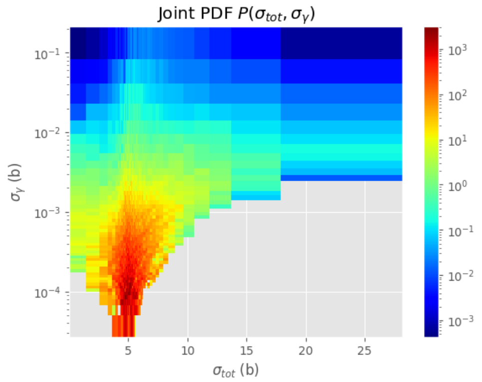
Leveraging preliminary work under NCSP AM-6 and NA-22 Intentional Forensics Venture



^{238}U(n,g) cross-section PDF



Use age-progression software to learn the temperature (and energy?) dependence of the cross-section PDF



Alternatively, we can use the PDFs directly with estimates of resonance spacings & widths

Experimental costraints: Nuclear Level Densities

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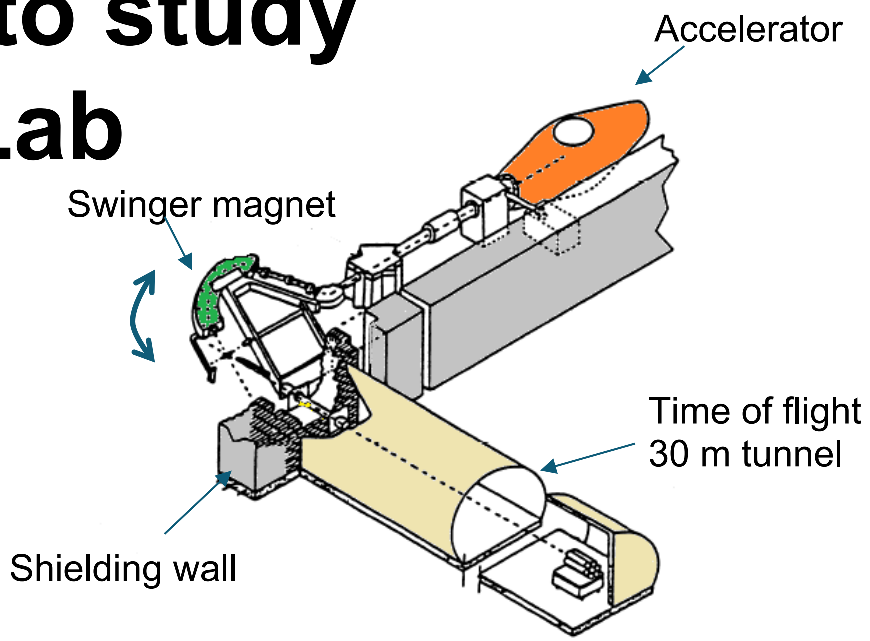
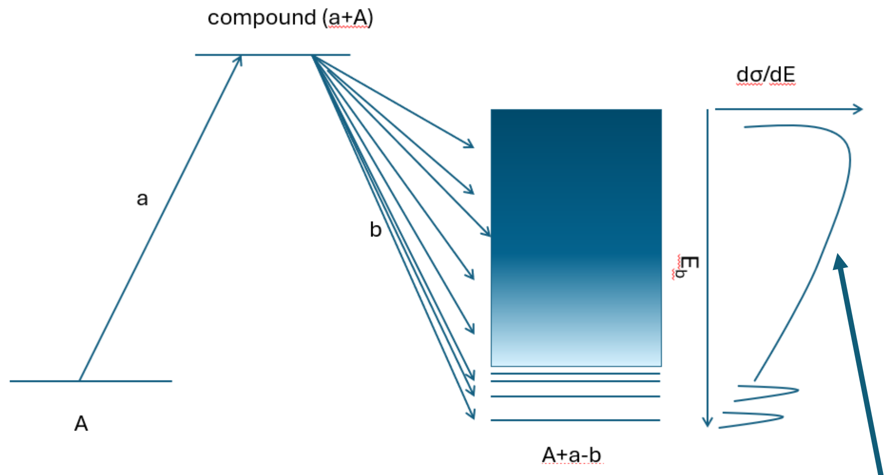
Particle evaporation technique to study level densities at the Edwards Lab

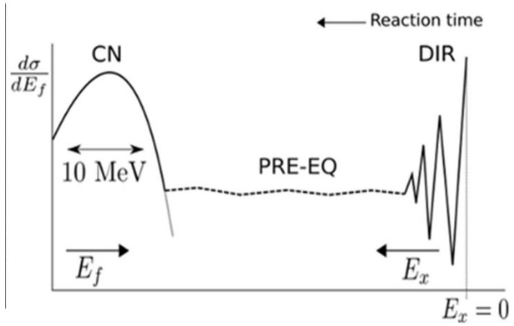


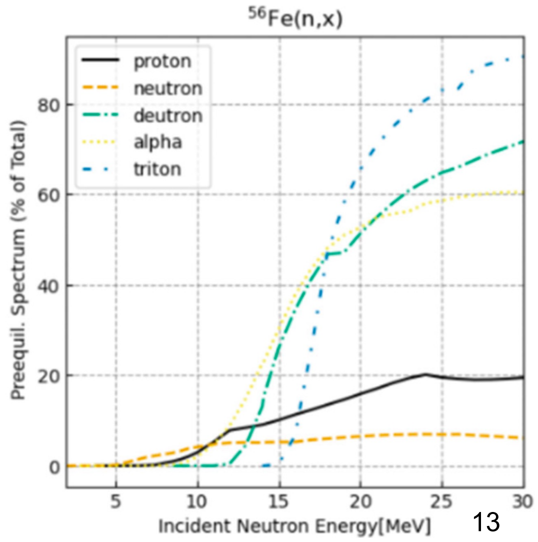
Goal: to improve systematics of the level density model parameters, which is a **key ingredient in evaluation models**, in the mass region of fission products thereby enhancing the **predictive power** of level density models for nuclear data evaluations when **extrapolating to nuclei off stability**.



We will use (p,n) and (d,n) reactions to measure neutron dσ/dE spectra

- **Preequilibrium** emissions might contribute **at high energies**
- We will use experimental information to constrain microscopic PE models
- Increase confidence in the description at stable isotopes before applying to unstable ones





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Status and perspectives

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Current status

- Currently in the beginning of Q2 of 2nd year (out of 3)
- Initial effort was dedicated to coordination and laying out groundwork
- Most of technical effort has focused on
  - Building the infrastructure
    - Creating (multiple) database(s) of nuclear deformations
    - Script to create EMPIRE inputs for all relevant nuclei and run them on the NNDC cluster, generating ENDF-6 formatted files
    - Integrated EMPIRE inputs with deformation data bases and with preliminary ML-generated shape cross sections
  - Preliminary calculations and comparisons
  - First experimental campaign
    - Measurement of ^{92,96}Zr(p,n) evaporation neutron spectra
    - Data analysis

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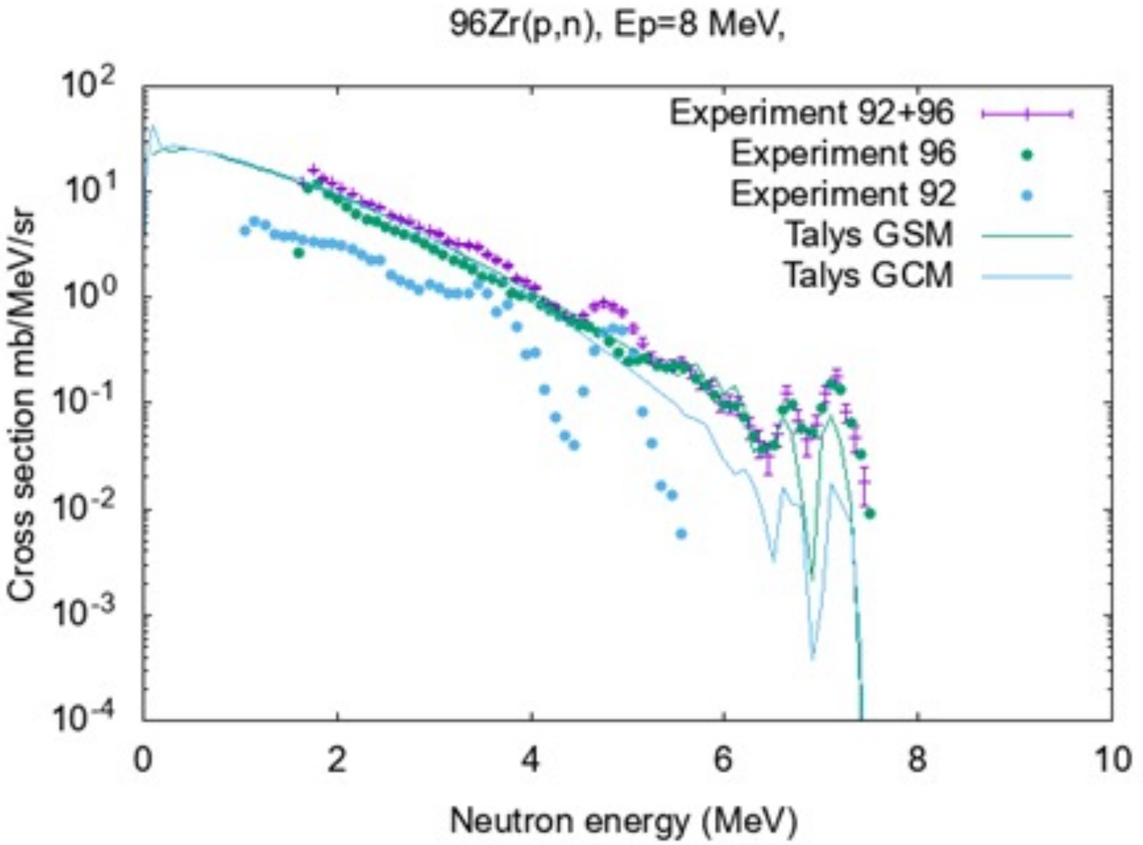
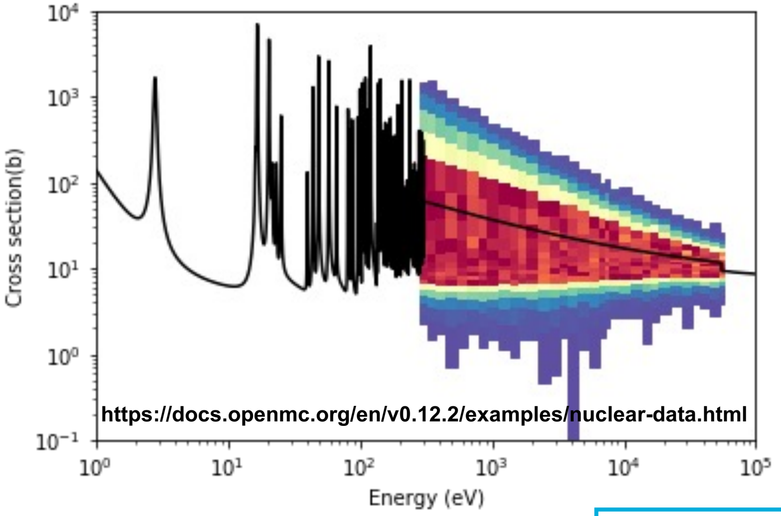
Level density measurements

- First experimental campaign at Ohio University
  - Measurement of $^{96}Zr(p,n)$ evaporation neutron spectra
  - Data analysis revealed peaks coming from ^{92}Zr present in the ^{96}Zr-enriched sample
  - Performed additional measurement of $^{92}Zr(p,n)$ to quantify the contamination in the original measurement.
- Different LD models will lead to different agreements with data
  - Measurements will help constrain LD

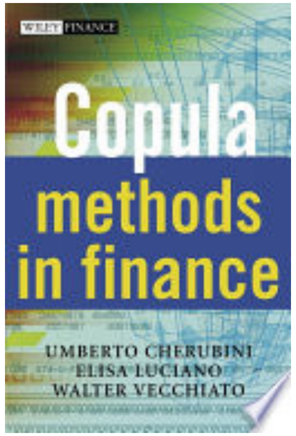


Developments in resonance range

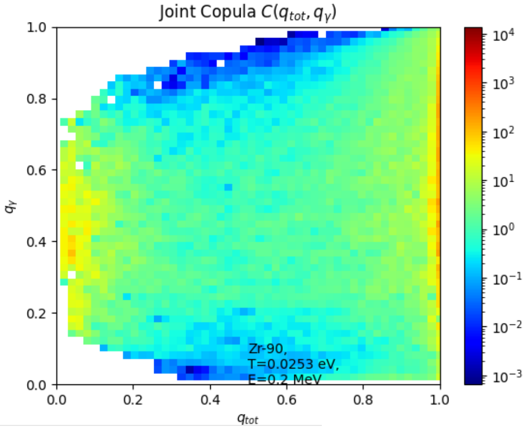
If we “don’t know anything” we must treat the problem probabilistically

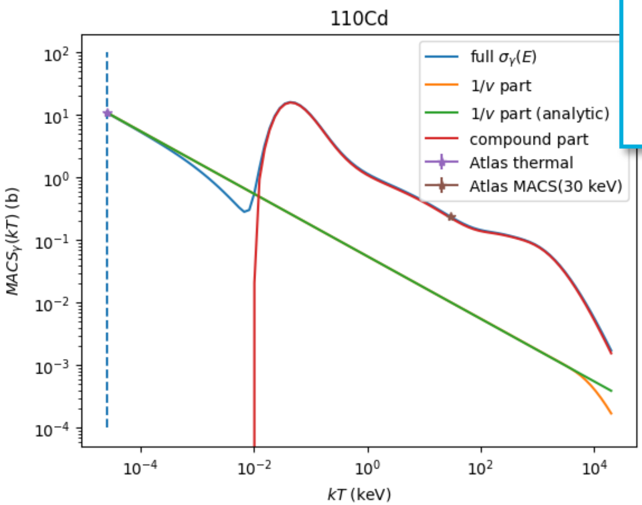


- Exploring the alternative approaches from the usual probability tables
- Replaces PDF by cumulative functions: copula*



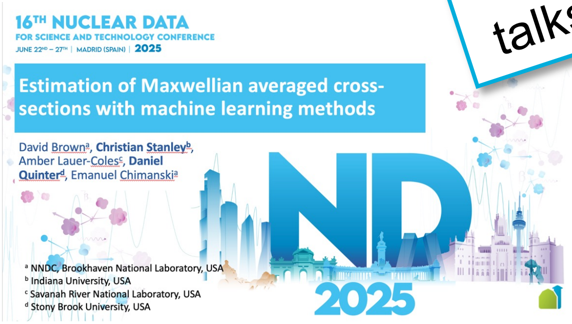
Cherubini, Luciano, Vecchiato. *Copula Methods in Finance.* John Wiley & Sons, Ltd, 2004





- Developing model for MACS functional

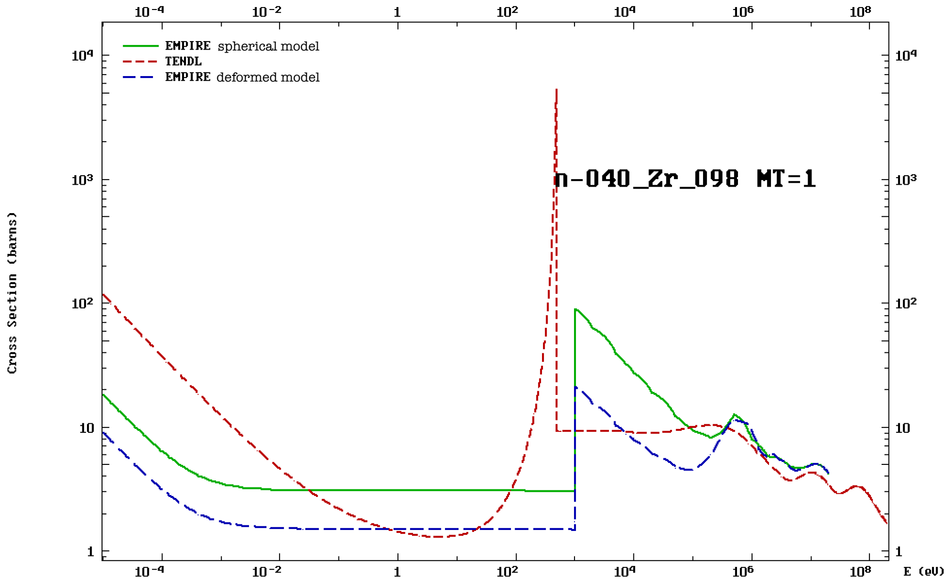
See D. Brown’s talks at ND 2025

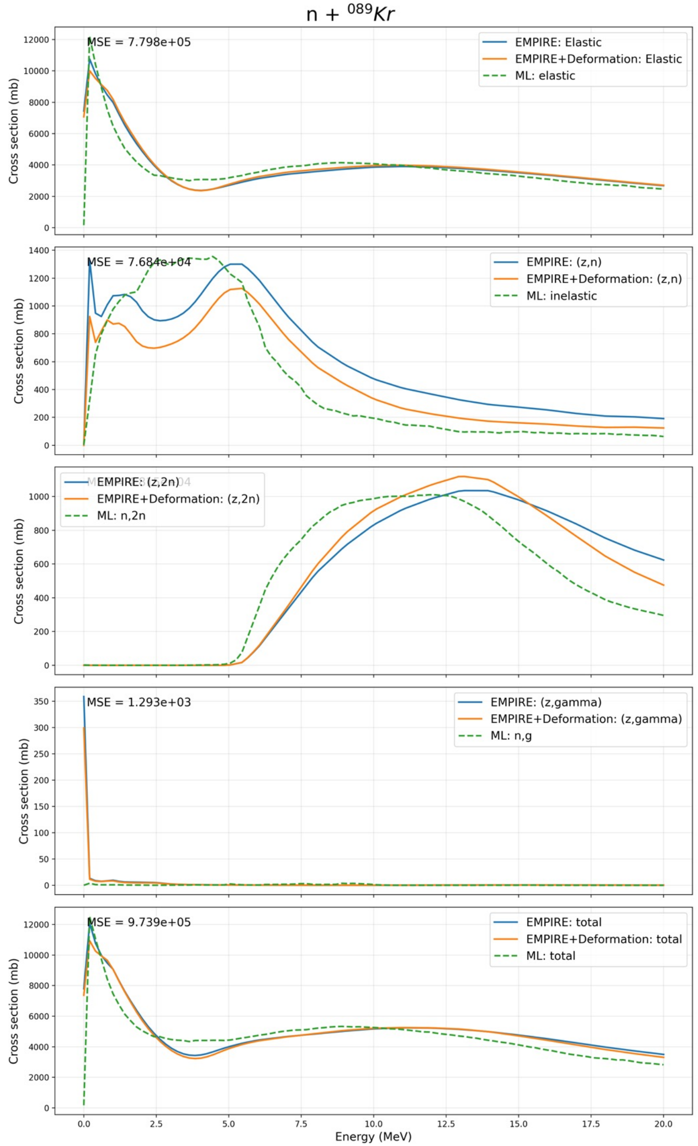


*In probability theory and statistics, a copula is a multivariate cumulative distribution function for which the marginal probability distribution of each variable is uniform on the interval [0, 1]. Copulas are used to describe / model the dependence (inter-correlation) between random variables. (Source: Wikipedia)

Developments in the fast neutron range

Preliminary coupled–channels calculations, testing impact of deformations





Built capability to plot and compare model calculations with first round of ML-extrapolated cross-sections shapes.

Next steps:

- Improve ML c.s., training on ENDF rather than TENDL
- Use these c.s. shapes as priors for model fits

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Perspectives

- Next steps:
  - Train ML age progression to obtain functional for c.s. distributions in resonance region
  - Improve library of ML-extrapolated cross-section priors and implement fits
  - Perform new round of level density measurements through evaporation spectra
  - Generation of preliminary evaluated files
- Challenges in defining potential validation avenues and/or integral testing
  - Spent fuel, activation
  - Reactor depletion, modeling of burnup transient states
  - Astrophysics
  - …?
- Feedback, especially regarding model approaches and validation are appreciated

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