

Lawrence Berkeley National Laboratory  
Nuclear Science Division

# Tungsten capture $\gamma$ -ray analysis



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# Improving the neutron-capture $\gamma$ -ray spectrum



## Goal

Improve and build upon spectrum of known  $W \gamma$  rays in neutron data libraries

Improvements to the **Evaluated Gamma-ray Activation File** (EGAF) and **Evaluated Nuclear Structure Data File** (ENSDF) databases: clarification of nuclear structural problems

Changes to the ENSDF file will also be reflected in other databases: **Reference Input Parameter Library** (RIPL)

## Method

Use the statistical-decay code DICEBOX to model thermal-capture  $\gamma$  cascade and tune input parameters to experimental data from the Evaluated Gamma-ray Activation File (EGAF)

Ensure DICEBOX input files are consistent the latest developments in the Evaluated Nuclear Structure Data File (ENSDF) – and beyond?

## Deliverables

Improved  $\gamma$ -ray spectra and level schemes: new  $\gamma$  rays; new levels; clarification of nuclear structure issues

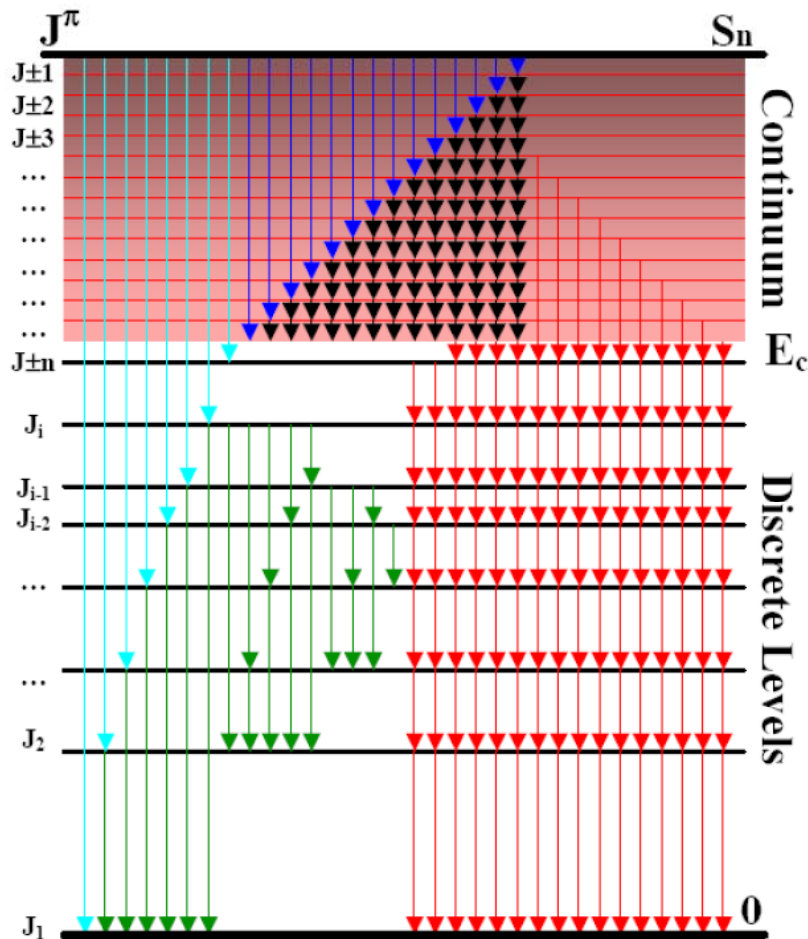
Improved EGAF, ENSDF, RIPL (and ENDF) evaluations with primary  $\gamma$  rays

Total radiative thermal neutron-capture cross sections

# Simulations of $\gamma$ -ray cascades following the thermal neutron-capture process



## DICEBOX Monte Carlo Code



DICEBOX generates  $(n,\gamma)$  level scheme simulations (nuclear realizations) based on statistical model level densities  $\rho(E_j, J^\pi_j)$  and  $\gamma$ -ray transition probabilities  $\Gamma_{if}$  where

- All levels and  $\gamma$ -rays below  $E_{crit}$  are taken from experiment.
- All levels and  $\gamma$ -rays above  $E_{crit}$  are generated randomly from level density and PSF models
- Primary  $\gamma$ -ray cross sections are taken from experiment when known.

Typically 30,000 capture state  $\gamma$ -ray decay cascades are randomly generated for each nuclear realization.

50 separate realizations are usually averaged to get the statistical variation in the simulated level feedings.

# critical energy / cut-off energy



reaction-model calculations (DICEBOX): complete discrete level schemes @  
/ excitation energy; specify all possible outgoing reaction channels and test  
level-density models (which replace discrete levels in the quasicontinuum  
higher-excitation energy)

completeness: specified energy and spin window within which all levels are  
characterized by unique energy, spin and parity values, and all  $\gamma$ - and particle-  
decay branches are known

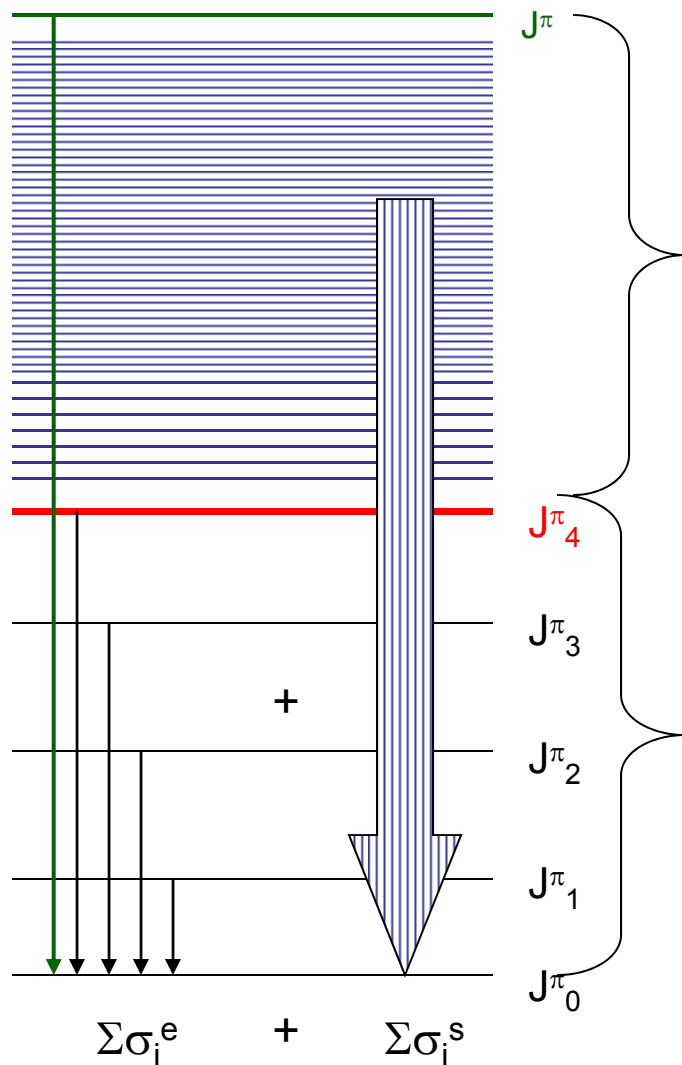
results for  $W$  evaluations presented according to a defined upper-energy limit  
(i) the **critical energy**  $E_{\text{crit}}$ ; (ii) the **cut-off energy**  $E_c$

$E_{\text{crit}}$ : upper-energy limit of levels characterized by **unique** spin and parity where  
 $\gamma$ -ray branching is known

$E_c$ : an empirically-determined maximum cut-off energy below which all level  
scheme information is either **complete** or has only a **very weak influence** on the  
calculation

degree of completeness varies largely over nuclear landscape:  $E_{\text{crit}}$  and  $E_c$  must  
be defined separately on an individual isotope basis

# al capture cross section



- New thermal neutron capture cross sections,  $\sigma_0$
- Sum of
  - Measured experimental gamma cross sections which feed the ground state (primary+feeding below  $E_{crit}$ ) ( $\Sigma \sigma_i^e$ )
  - Modeled population feeding from continuum to ground state ( $\Sigma \sigma_j^s$ )
- $\sigma_0 = \Sigma \sigma_i^e + \Sigma \sigma_j^s$

# Evaluation of the W isotopes



Elemental tungsten occurs naturally in 5 isotopic forms:

- $^{180}\text{W}$  (0.12%) very little  $(n,\gamma)$  information, not considered in evaluation
- $^{182}\text{W}$  (26.50%)
- $^{183}\text{W}$  (14.31%)
- $^{184}\text{W}$  (30.64%)
- $^{186}\text{W}$  (28.43%)

**Stable  $^{182-186}\text{W}$  all have large natural abundances, therefore, evaluation is needed for all isotopes**

For all W data evaluations:

consider compound system i.e.  $^A\text{W}(n,\gamma)^{A+1}\text{W}$

$^{A+1}\text{W}$  is the compound system

calculations assuming 10 nuclear realizations, with 100,000 capture-state  $\gamma$ -ray decay cascades generated per realization

# CEBOX: simulation cf. experiment



CEBOX calculates theoretical level feedings to all excited states in the input file

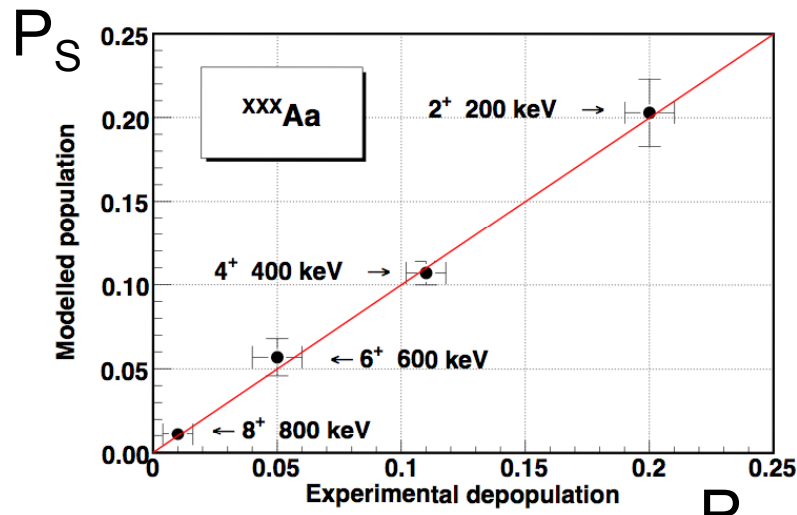
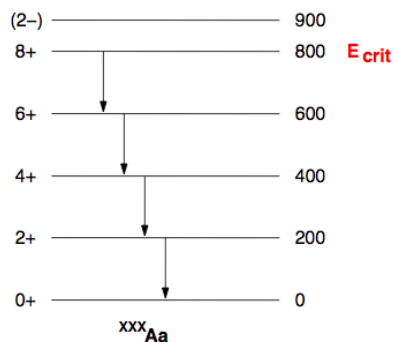
Modelled populations ( $P_S$ ) can then be compared to measured experimental depopulations

$P_E = P_S \Rightarrow$  statistical model accurately describes nuclear properties

Consider hypothetical nucleus  $^{XXX}Aa$ : 4 excited states in rotational sequence;  $E_{crit} = 800$  keV

Critical energy ( $E_{crit}$ ): highest level where all structure information ( $\gamma$  branching,  $J^\pi$ ) is known

Increasing transition probability upon descending band as consequence of decay process i.e. larger  $\gamma$ -decay branch is observed from states with decreasing excitation energy

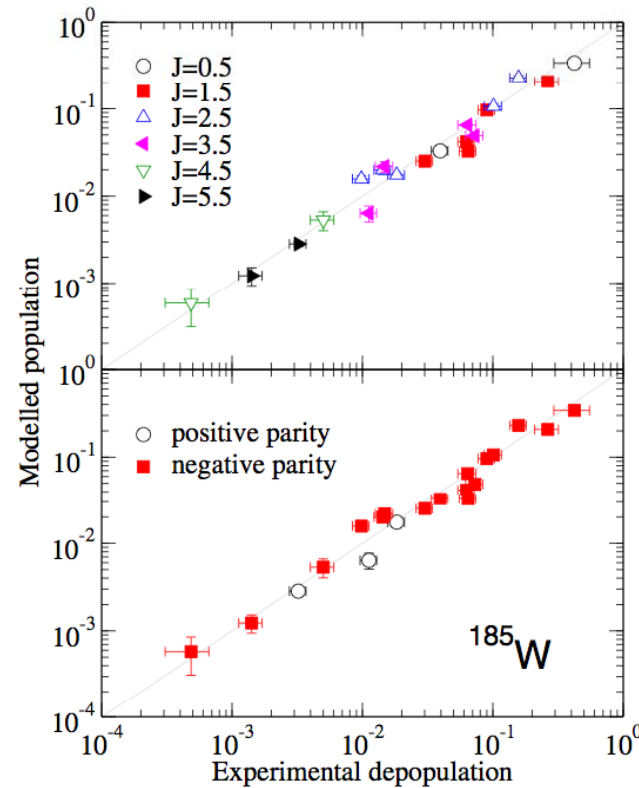
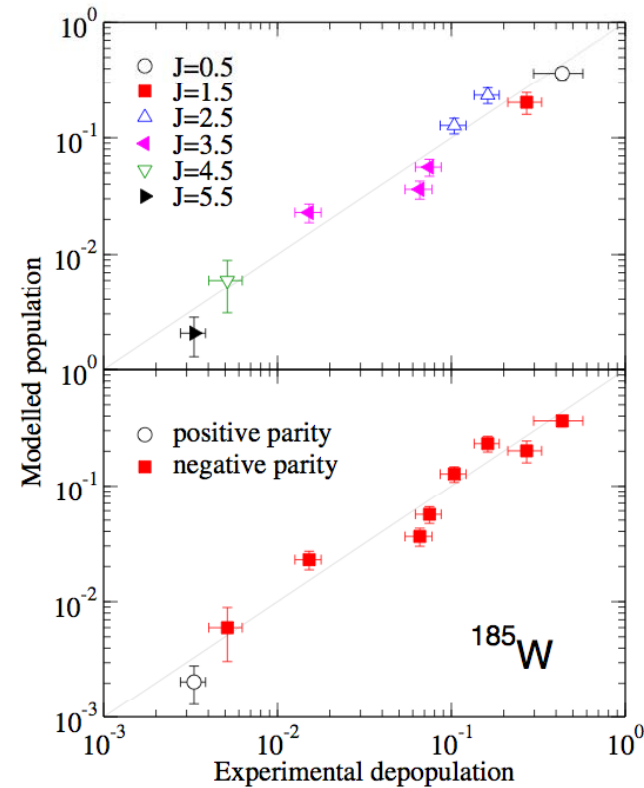


# $W$ capture- $\gamma$ analysis: $E_{\text{crit}}$ Vs $E_c$



$E_{\text{crit}} = 332.1 \text{ keV}; N = 10$

$E_c = 827.1 \text{ keV}; N = 28$



- $E_{\text{crit}}$ : 243 keV (RIPL)  $\rightarrow$  332 keV (EGAF)
- 9 new  $\gamma$  rays placed in level scheme
- $(13/2^+)$  isomer @ 384 keV prevents raising  $E_{\text{crit}}$

- $E_c$ : 332 keV  $\rightarrow$  827 keV
- 36 new  $\gamma$  rays placed in level scheme
- 3 new levels placed in level scheme



# Final results: improving the RIPL file



Confirmation/determination of  
relative  $J^\pi$  assignments

Analysis with  $E_c$  @ 827 keV:

14  $\nu$ -lying levels; 14 with ( $J^\pi$ )

Legend:

111/2 keV:  $J^\pi = (9/2^-)$

111/2 keV:  $J^\pi = (9/2^-)$

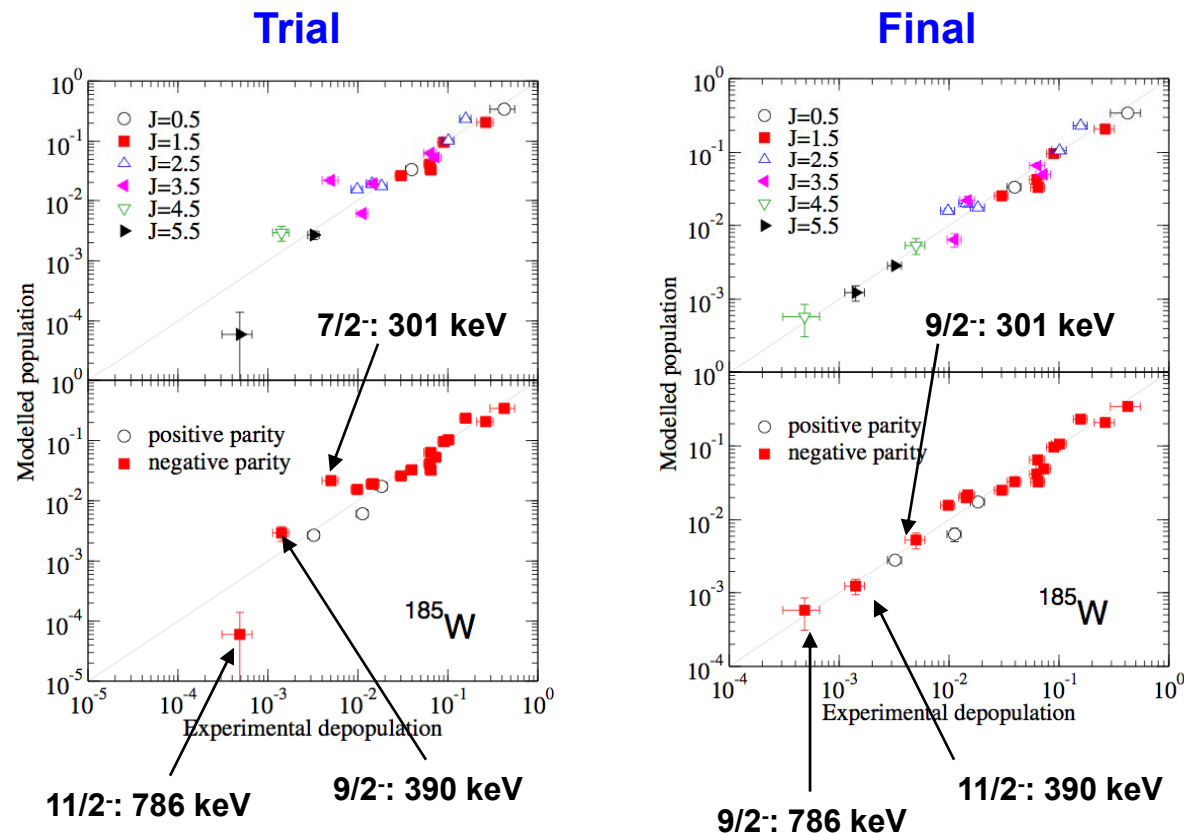
111/2 keV:  $J^\pi = (9/2^-)$

DICEBOX:

111/2 keV:  $J^\pi = 9/2^-$

111/2 keV:  $J^\pi = 11/2^-$

111/2 keV:  $J^\pi = 9/2^-$



Changes/improvements need to be made to the RIPL file based on DICEBOX analysis for  $^{185}\text{W}$

# : determining the capture-state composition



$E_c = 1431 \text{ keV}; N=20$

$(n,\gamma)^{184}\text{W}$

Capture on odd-A  $^{183}\text{W}$  target  
ground-state spin  $J_{gs}^\pi = \frac{1}{2}^-$

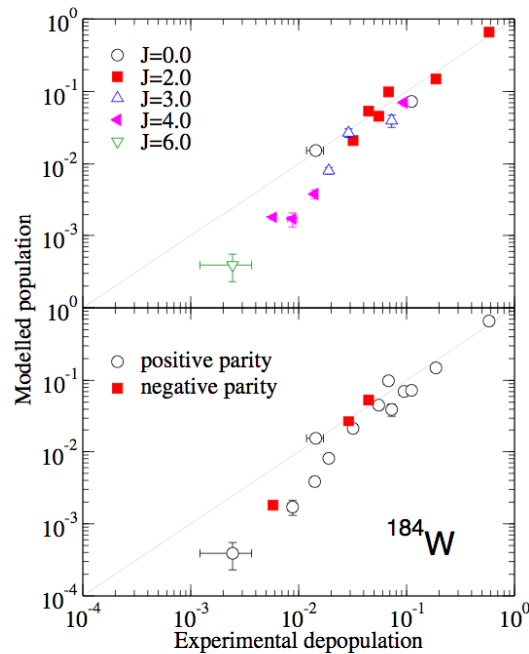
Capture state will have spin of  
position  $J = J_{gs}^\pi \pm \frac{1}{2}$

Capture-state composition:

0- or 1- in  $^{184}\text{W}$  compound

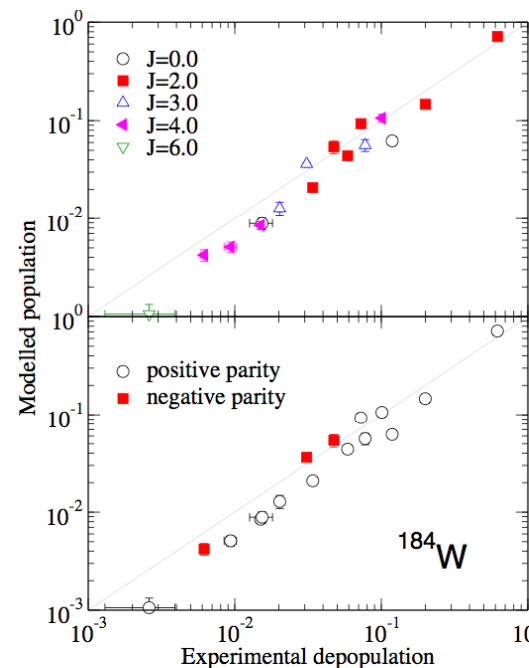
priority 1- neutron resonances

EGLO / BSFG



CS: 0-(80%) 1-(20%)

EGLO / BSFG



CS: 0-(0%) 1-(100%)

Higher-spin capture state (1-) favors population of high-spin states in  $^{184}\text{W}$

# al radiative capture cross sections for W isotopes



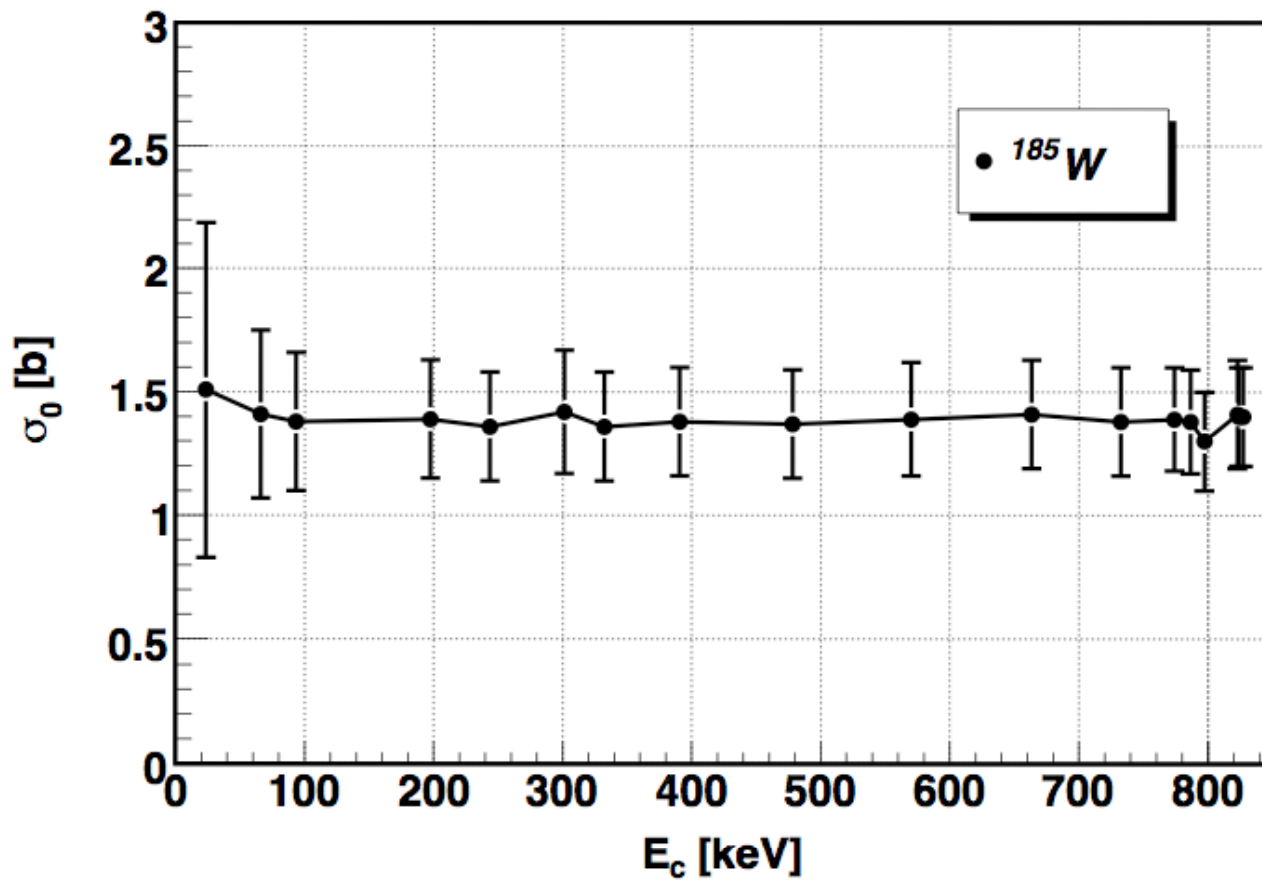
$$\sigma_0 = \sum \sigma_{\gamma}^{\text{exp}}(\text{GS}) + \sum \sigma_{\gamma}^{\text{sim}}(\text{GS})$$

Isotope (compound)	$\sigma_0$ [b] adopted	$\sigma_0$ [b] $E_{\text{crit}}$	$\sigma_0$ [b] $E_c$
$^{183}\text{W}: ^{182}\text{W}(n,\gamma)$	20(2)	18.0(7)	18.4(8)
$^{184}\text{W}: ^{183}\text{W}(n,\gamma)$	10.3(2)*	7.9(3)	8.0(2)
$^{185}\text{W}: ^{184}\text{W}(n,\gamma)$	1.8(2)	1.4(2)	1.4(2)
$^{187}\text{W}: ^{186}\text{W}(n,\gamma)$	34.9(2)	33.6(21)	34.1(5)

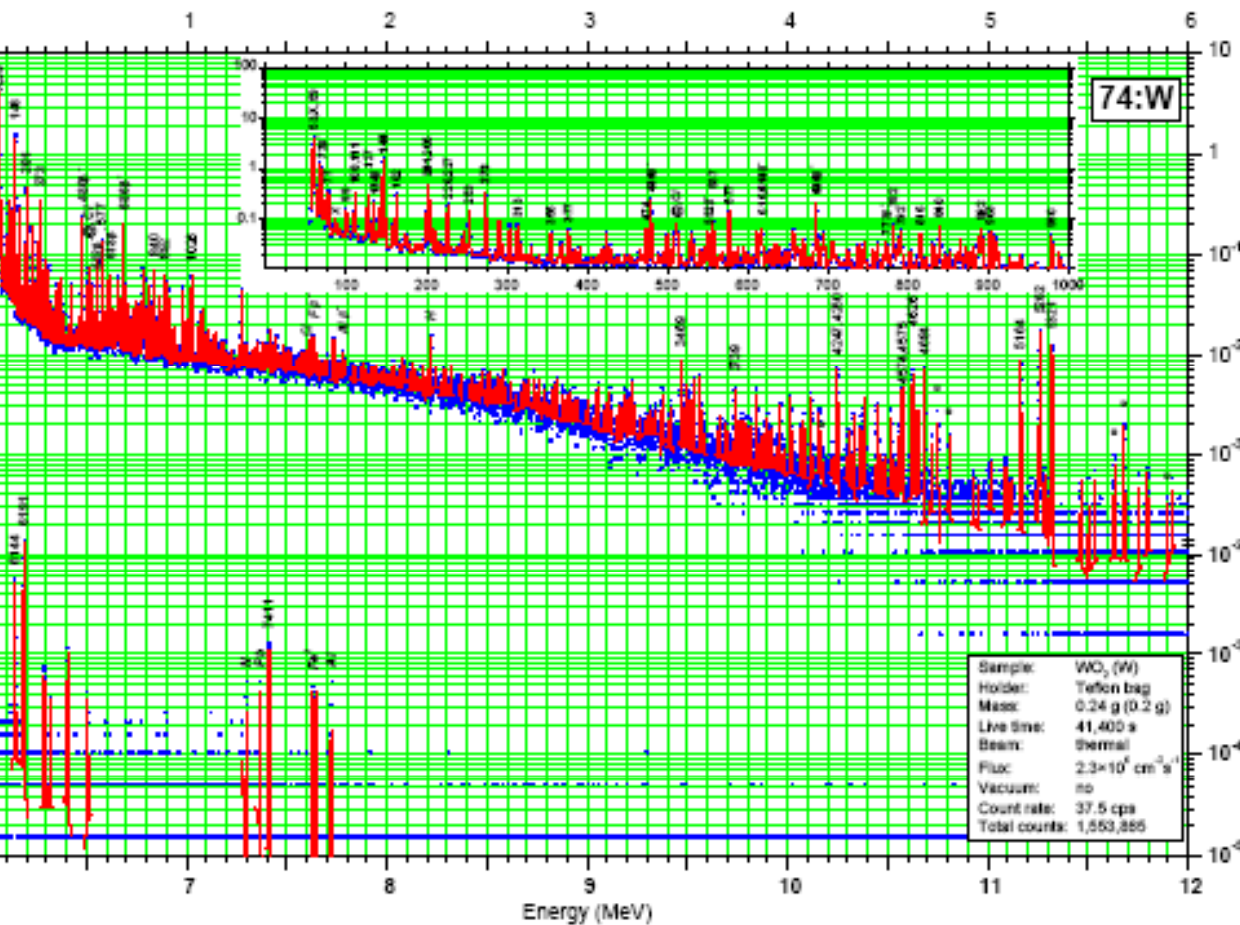
EGAF:  $\sigma_0 = 7.78(14)$  b

**PRELIMINARY !!!**

as a function of  $E_c$



# Experimental $^{74}\text{W}$ capture- $\gamma$ spectrum



- Na<sub>2</sub>WO<sub>6</sub> (s) sodium tungstate calibration
- WO<sub>2</sub> (s) tungsten oxide target

- $^{180}\text{W}(n,\gamma)^{181}\text{W}$   
[0.12 %]
- $^{182}\text{W}(n,\gamma)^{183}\text{W}$   
[26.50 %]
- $^{183}\text{W}(n,\gamma)^{184}\text{W}$   
[14.31 %]
- $^{184}\text{W}(n,\gamma)^{185}\text{W}$   
[30.64 %]
- $^{186}\text{W}(n,\gamma)^{187}\text{W}$   
[28.43 %]

ENSDF  $\Rightarrow$   $\gamma$  lines in individual W compound systems

Separated-W targets: definitive assignments: weaker signatures

# Summary and future work



CEBOX: useful tool for simulating  $\gamma$ -ray emission following thermal neutron capture on the W isotopes

Evaluation work on the W isotopes assuming an upper-energy limit defined by  $E_{\text{crit}}$  published in the following works:

*"Gamma spectrum from neutron capture on tungsten isotopes"*, published as proceedings to ND2010 by A.M. Hurst *et al.* in J. Kor. Nucl. Soc. (2010)

*"Data evaluation methods and improvements to the neutron-capture  $\gamma$ -ray spectrum"*, published as proceedings to UBC2010 by A. M. Hurst *et al.*

Several improvements to the EGAF and ENSDF (RIPL) file:  $^{185}\text{W}$  and  $^{187}\text{W}$  yields new information and confirmation/proposal of firm  $J^\pi$  assignments

Future: extensive EGAF measurements on all stable isotopically-enriched W targets (including  $^{180}\text{W}$ !) at the Budapest Reactor (mid-November 2010)

Supplemental W(d,p) measurements at 88" Cyclotron @ LBNL using LIBRACE to unravel structural problems in the W isotopes that otherwise prevent raising of  $E_{\text{crit}} \Rightarrow$  improvements to the RIPL file

forthcoming publication in Physical Review C discussing the W evaluation with

# Collaborators

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**BNL** – **A. M. Hurst**, R. B. Firestone, S. Basunia

**Budapest Reactor** – Zs. Revay, T. Belgya, L. Szentmiklósi

**LNL** – B. W. Sleaford, N. C. Summers, J. E. Escher

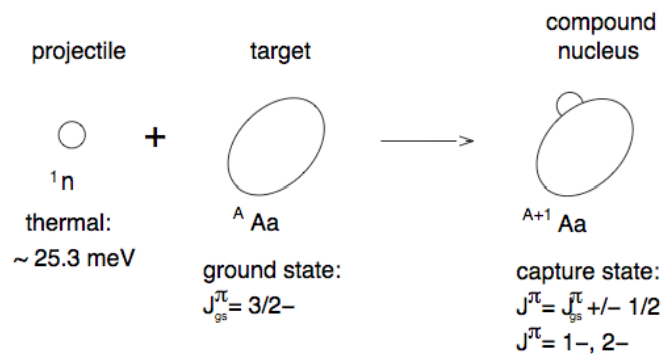
**Charles University (Prague)** – M. Krticka

**Seoul National University** – H. Choi

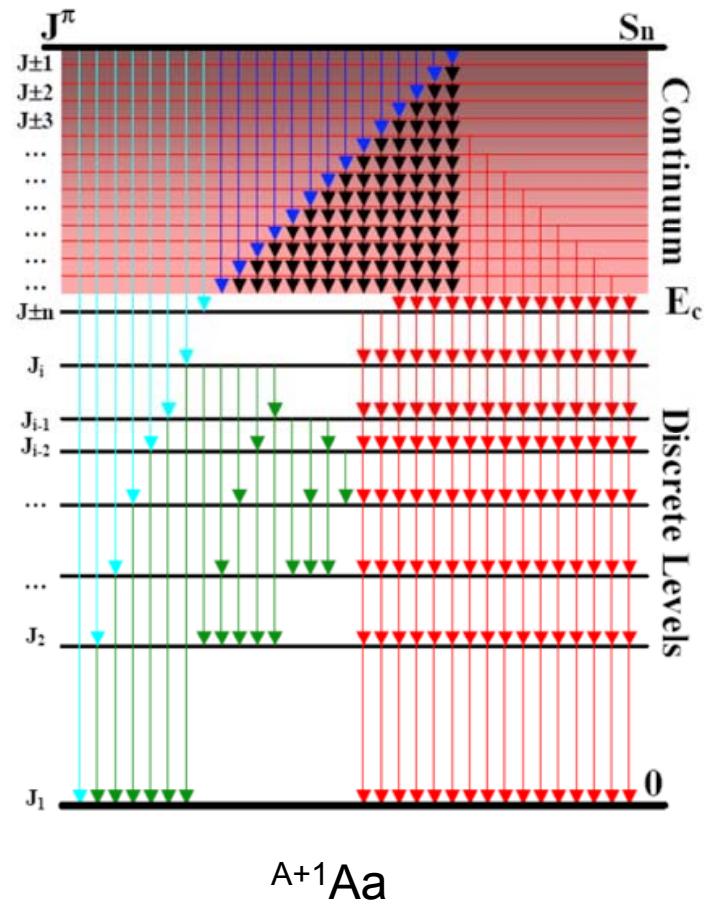
**North Carolina State University** – D. Dashdorj

**AEA** – R. Capote, A. Nichols

# Thermal neutron capture



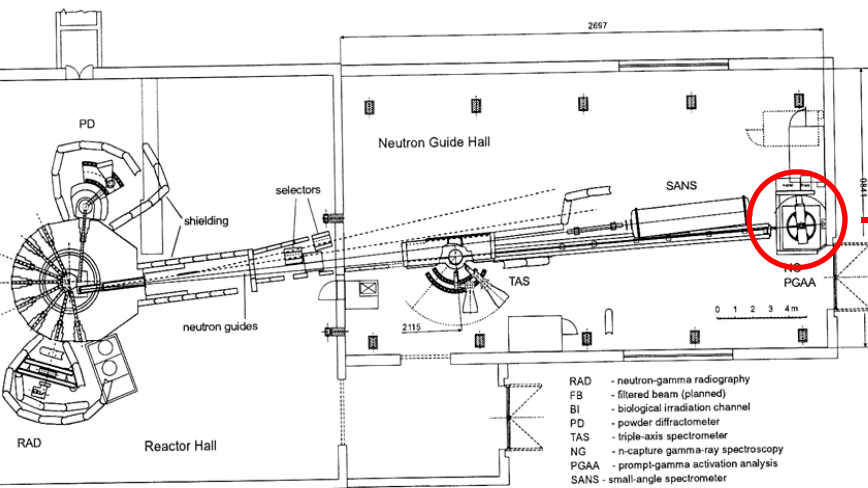
neutron separation energy



- Compound system formed at high energy
- Neutron separation energy
- Beneath threshold for particle evaporation
- Compound system deexcites through  $\gamma$ -ray emission
- Simulate this process using the Monte Carlo code DICEBOX



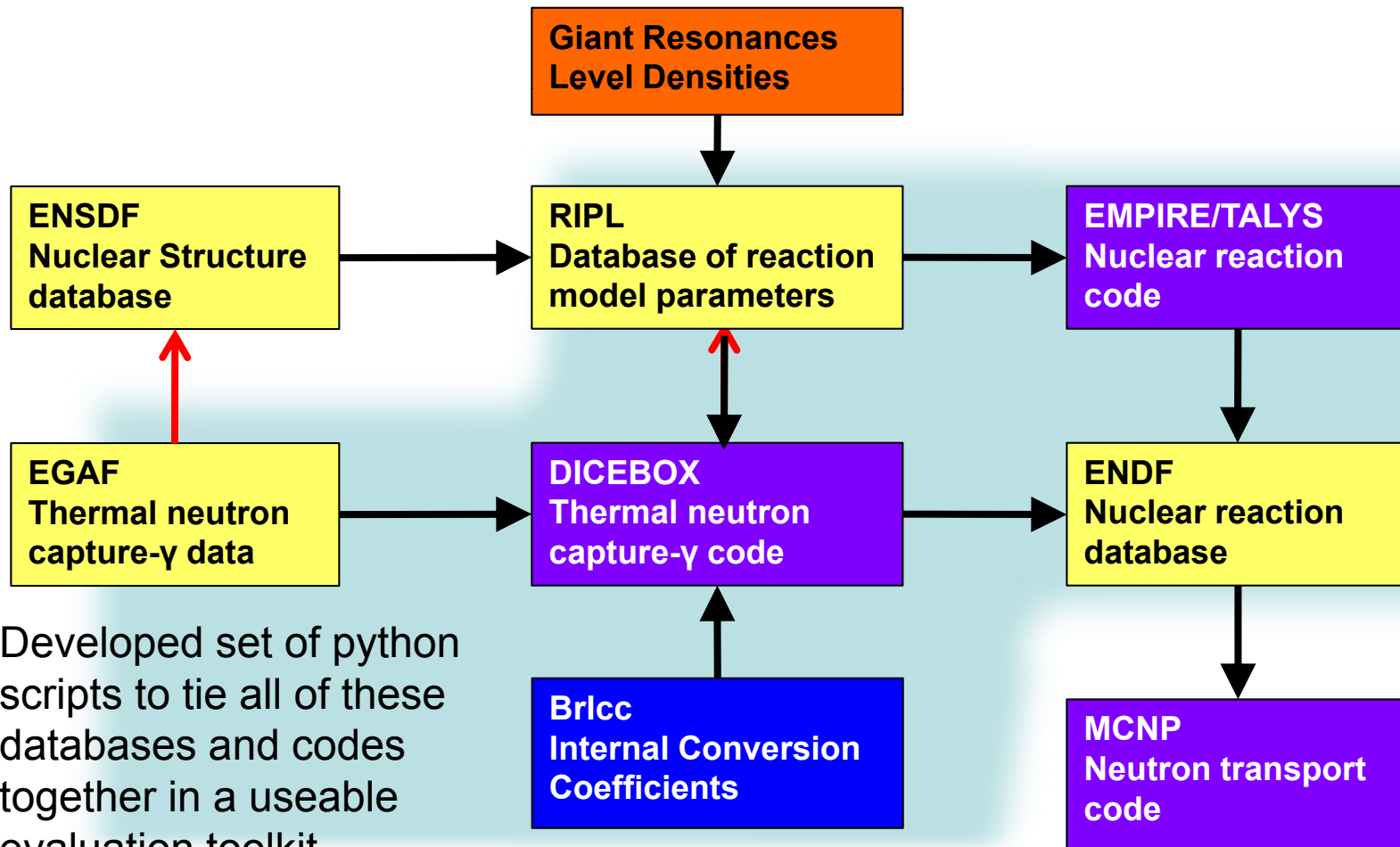
# AF Project: Budapest Reactor



- 10 MW Budapest reactor
- Guided-thermal neutron beam:  
thermal flux  $\sim 10^6 \text{ cm}^{-2}\text{s}^{-1}$   
cold flux  $\sim 10^8 \text{ cm}^{-2}\text{s}^{-1}$
- PGAA (Prompt Gamma Activation Analysis) experimental station is located  $\sim 30 \text{ m}$  from reactor wall
- Primary and secondary capture  $\gamma$  rays measured in low-background
- Compton-suppressed HPGe detector (closed-end coaxial) located 23.5 cm from target



# Databases and codes used in evaluation process

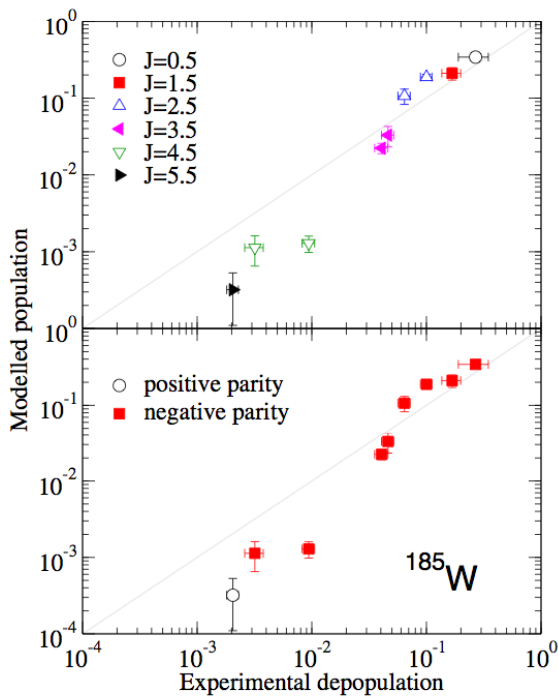


Developed set of python scripts to tie all of these databases and codes together in a useable evaluation toolkit

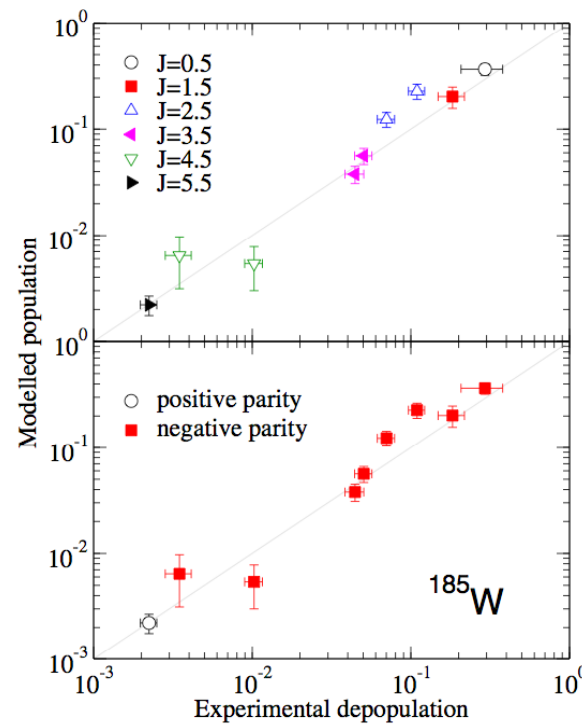
# V results: obtaining best fit



## BA / CTF

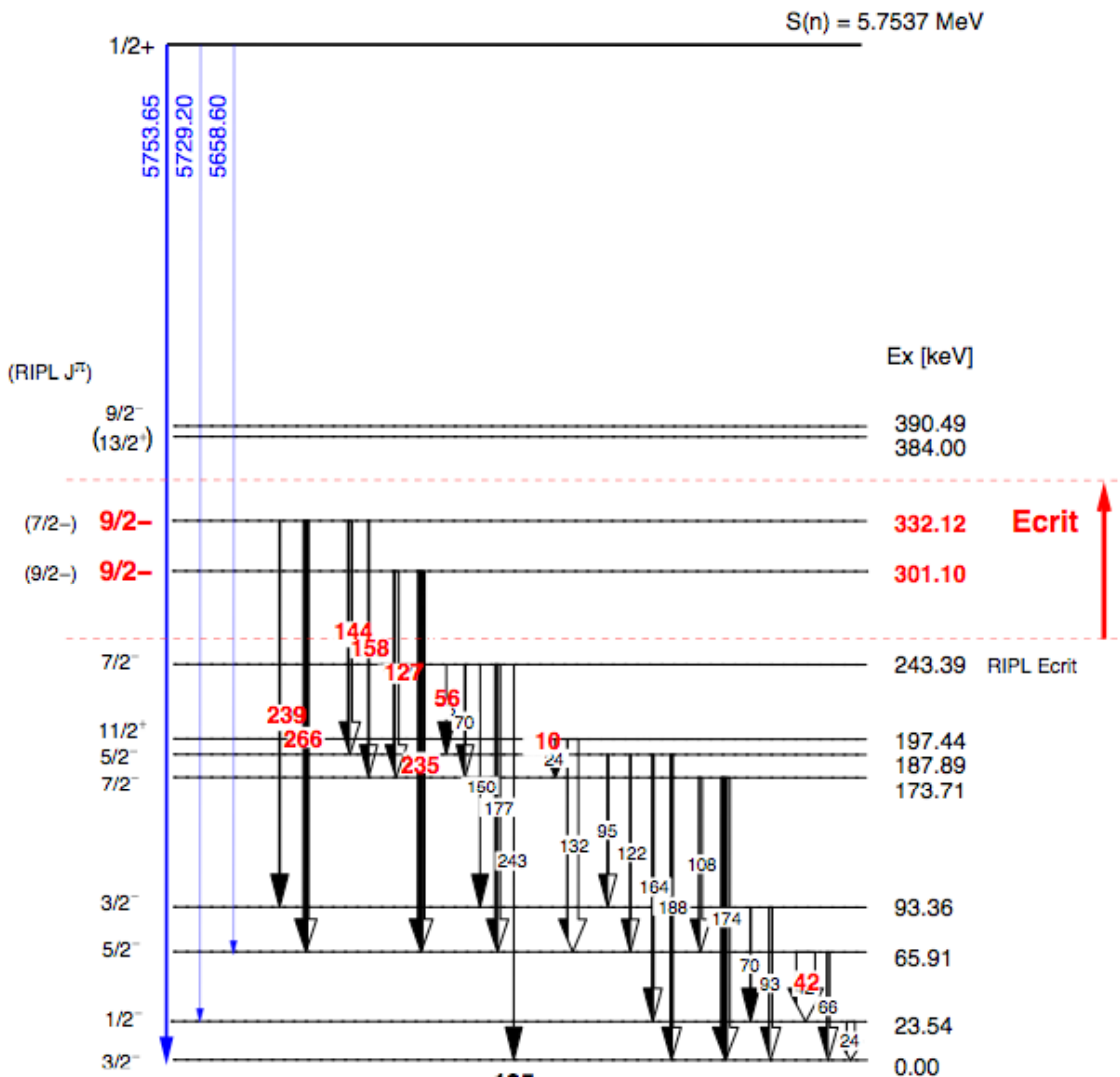


## EGLO / BSFG



**Best fit using EGLO PSF and BSFG level density**

# V summary: new level scheme



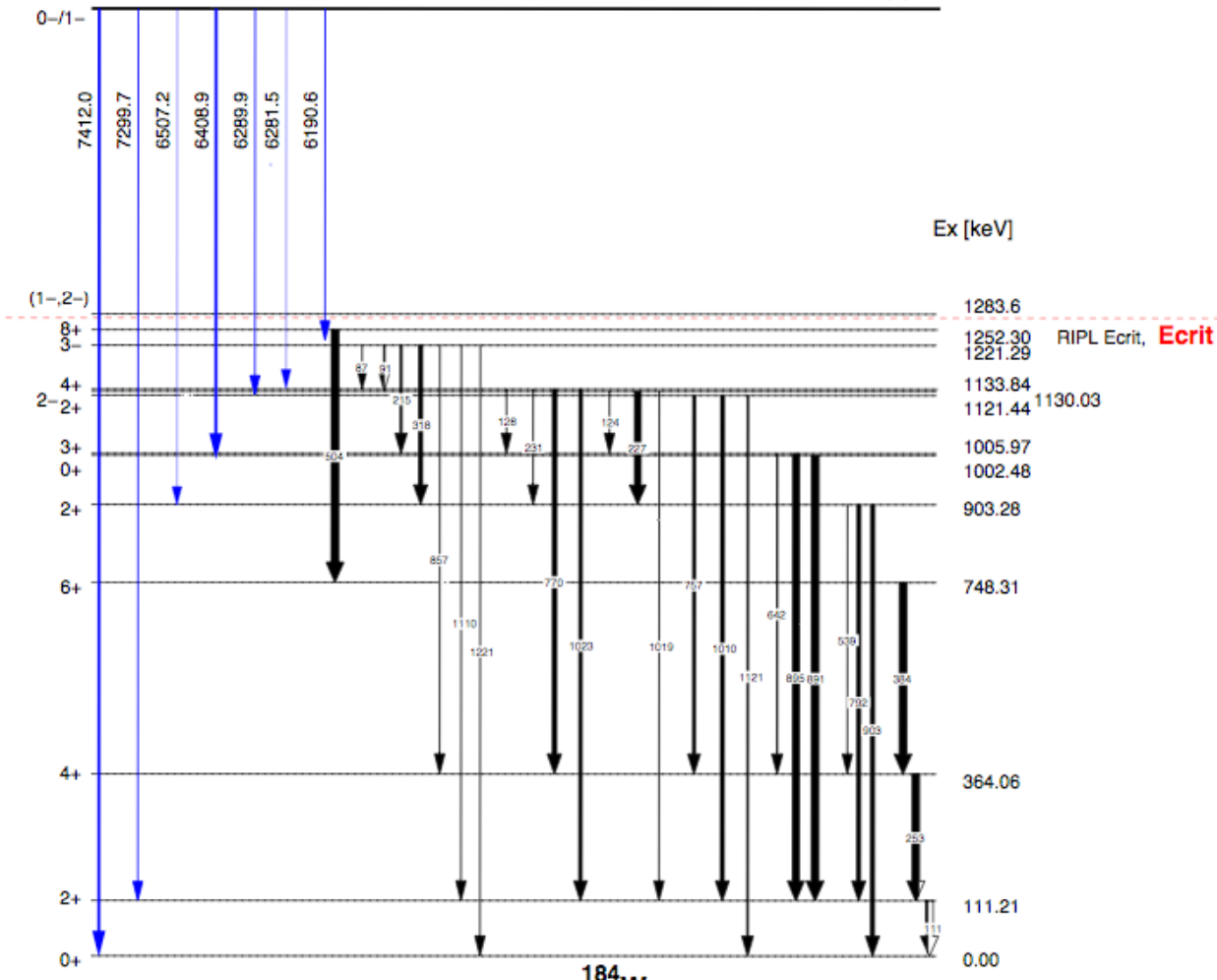
## RIPL improvements:

- 2 new  $J^\pi$  firmly established
- $E_{\text{crit}}$  raised by  $\sim 90$  keV with 2 new levels
- 9 new  $\gamma$  rays added to the decay scheme

# V decay scheme



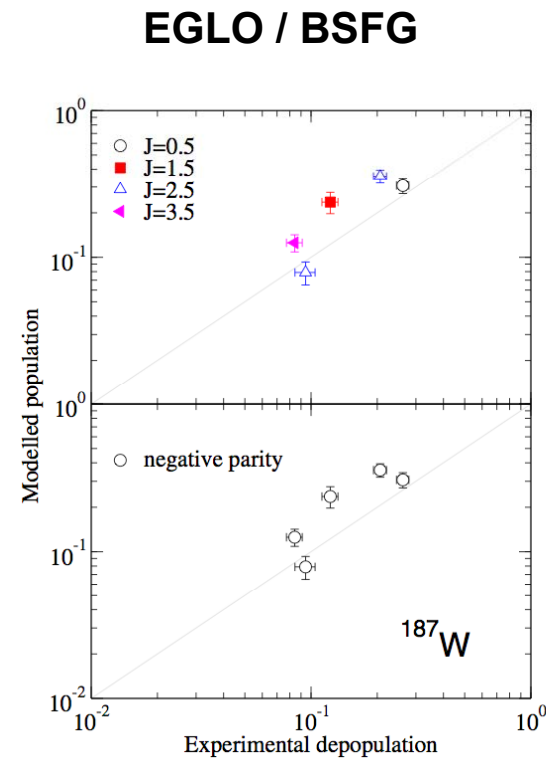
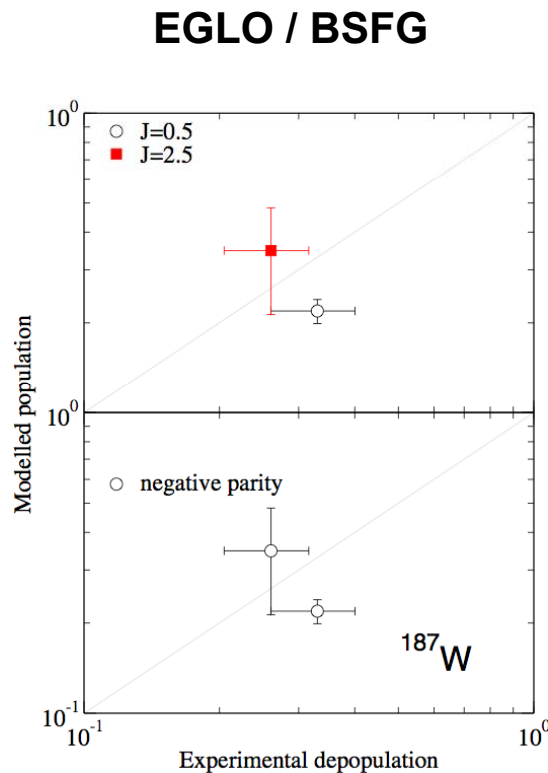
$S(n) = 7.41173 \text{ MeV}$



# Final results: improving the RIPL file

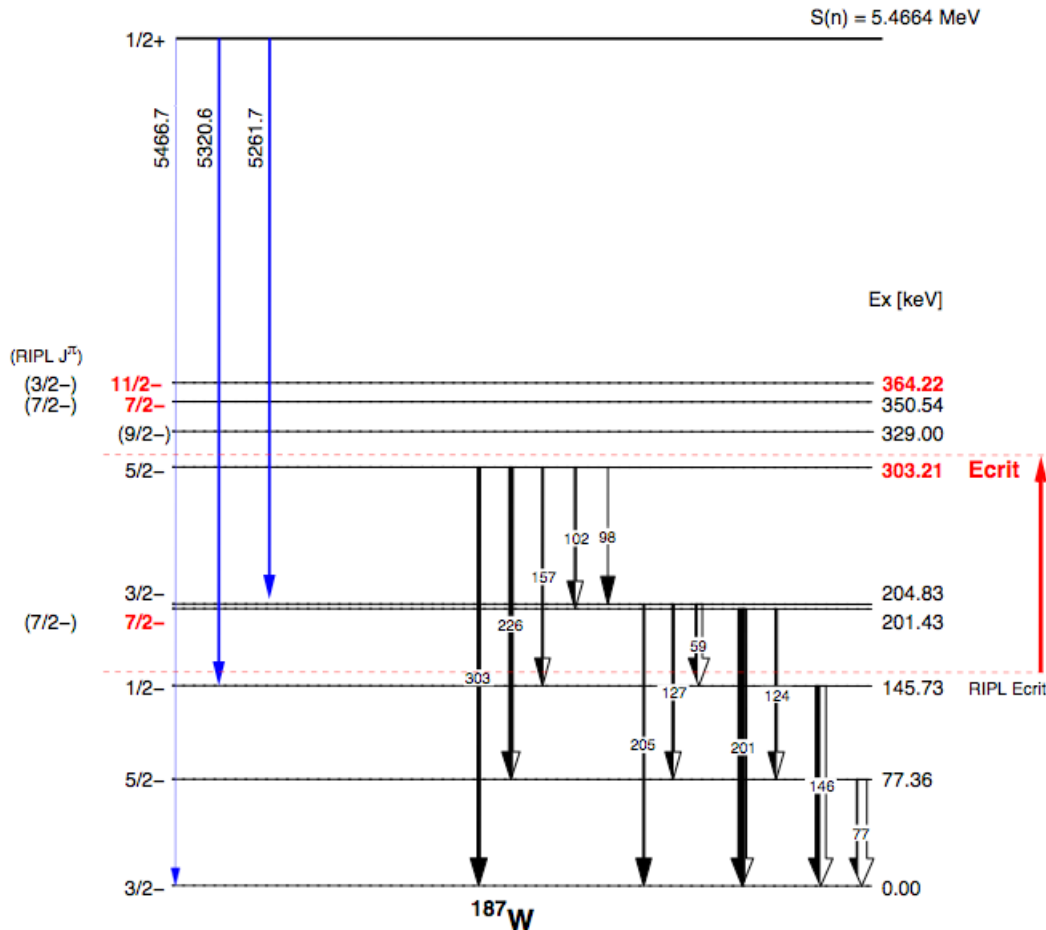


$(n,\gamma)^{187}\text{W}$   
Previous E<sub>crit</sub> in RIPL: 145.7 keV  
RIPL: (7/2<sup>-</sup>) @ 201.4 keV  
fit: 7/2<sup>-</sup> @ 201.4 keV  
E<sub>crit</sub> to 303.2 keV



**Changes/improvements need to be made to the RIPL file based on DICEBOX analysis for  $^{187}\text{W}$**

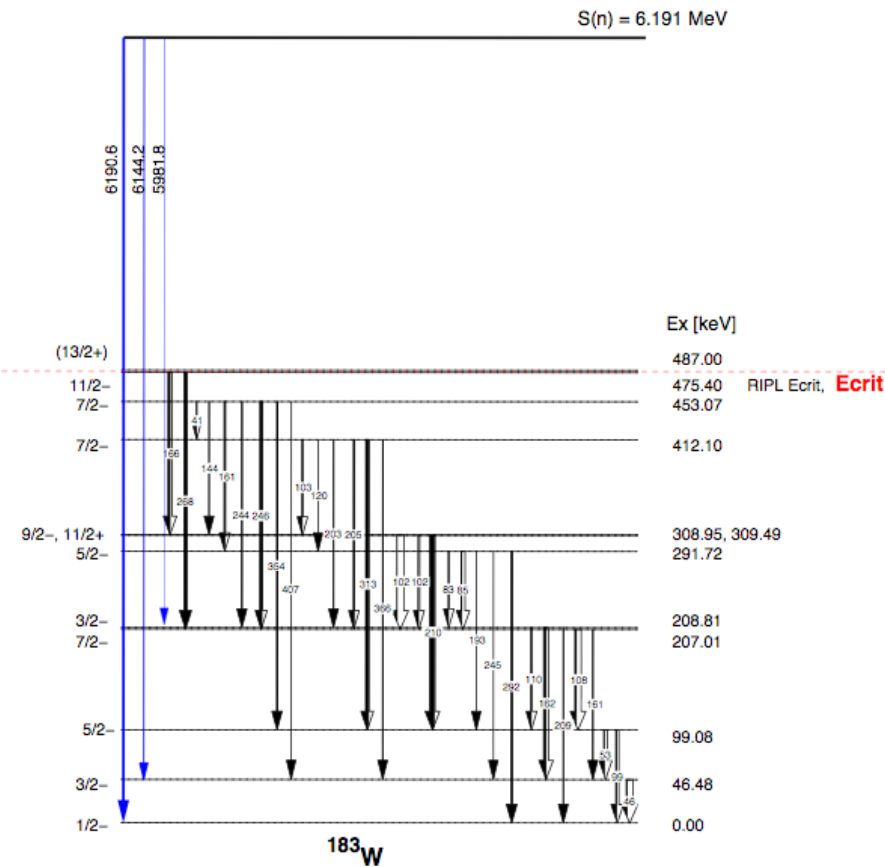
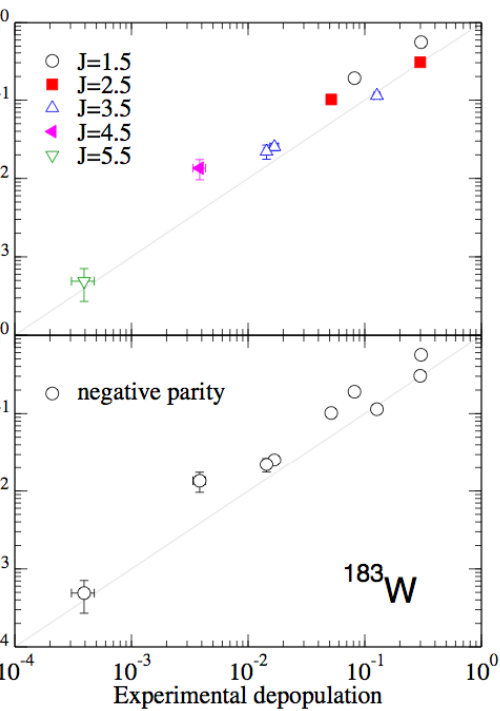
# V summary: new decay scheme



## RIPL improvements:

- $J^\pi$  firmly established as 7/2- @ 201.4 keV
- $E_{\text{crit}}$  raised by ~160 keV with 3 new levels

# V results



No new  $\gamma$  information from tentative (13/2<sup>+</sup>) isomer

$E_{\text{crit}}$  remains unchanged at 475.4 keV i.e. old RIPL value





# What is EGAF ?

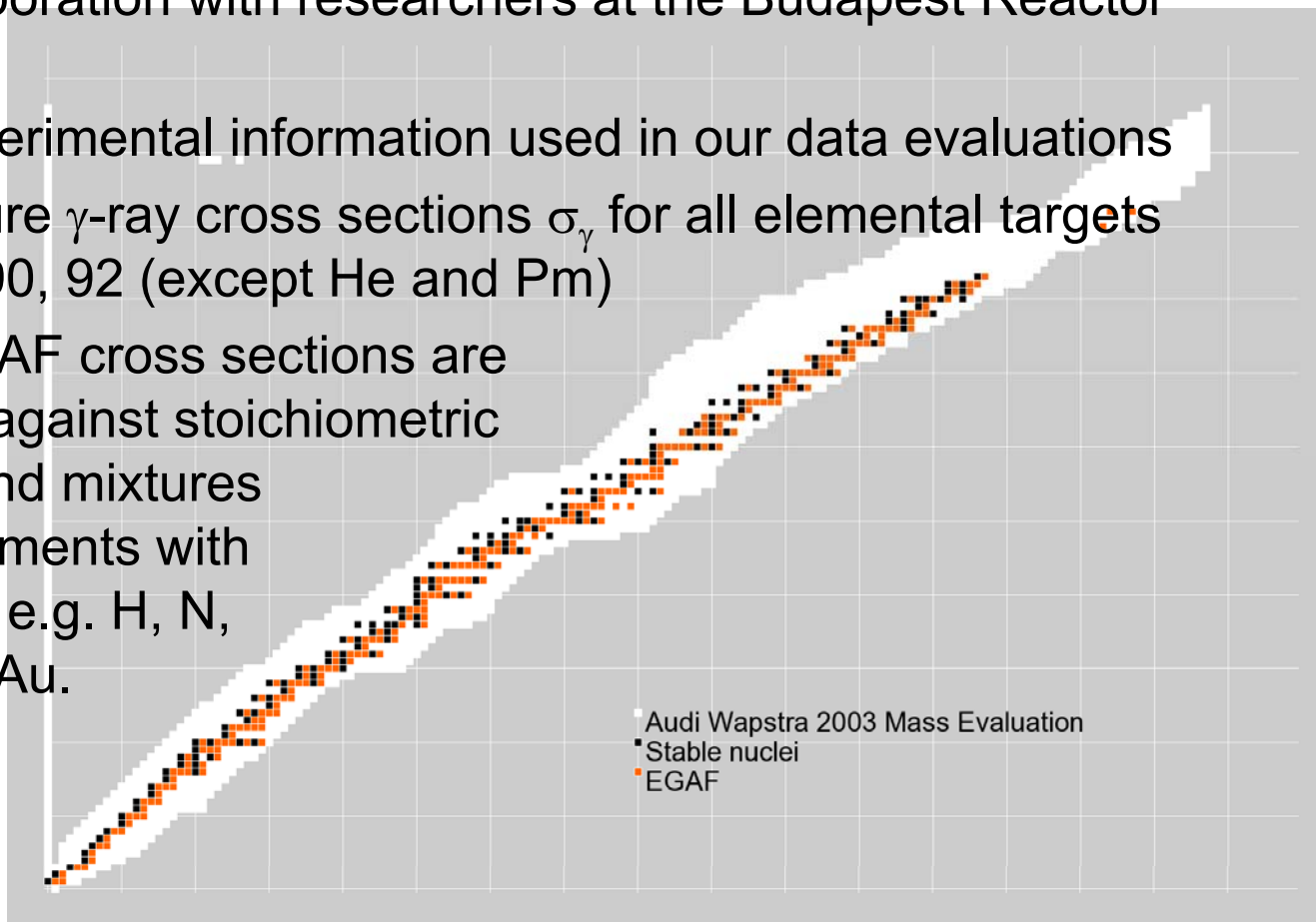


**EGAF - Evaluated Gamma-ray Activation File** developed at LBNL in collaboration with researchers at the Budapest Reactor and the IAEA.

Source of experimental information used in our data evaluations

Neutron-capture  $\gamma$ -ray cross sections  $\sigma_\gamma$  for all elemental targets with  $Z=1-83, 90, 92$  (except He and Pm)

Measured EGAF cross sections are standardized against stoichiometric compounds and mixtures containing elements with well-known  $\sigma_\gamma$  e.g. H, N, Cl, S, Na, Ti, Au.



# EA/LBNL EGAF $\sigma_\gamma$ Database

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A Coordinated Research Project was started in 2000 to evaluate  $\sigma_\gamma$  data

Evaluated Gamma-ray Activation File (EGAF) - 13,000  $\gamma$ -rays, 79 elements

*Database of Prompt Gamma Rays from Slow Neutron Capture for Elemental Analysis*, R.B. Firestone, et al, IAEA STI/PUB/1263, 251 pp (2007); online at: <http://www-pub.iaea.org/MTCD/publications/PubDetails.asp?pubId=7030>

*Handbook of Prompt Gamma Activation Analysis with Neutron Beams*, edited G.L. Molnar (Kluwer Publishers, 2004).

A Prompt Gamma-ray Activation Analysis Viewer:

<http://www-nds.iaea.org/pgaa/pgaa7/index.html>

Thermal Neutron Capture Gamma-ray Data: <http://ie.lbl.gov/ng.html>

# EGAF and ENSDF



Experimental spectroscopic EGAF properties can be established by comparison with evaluated nuclear data

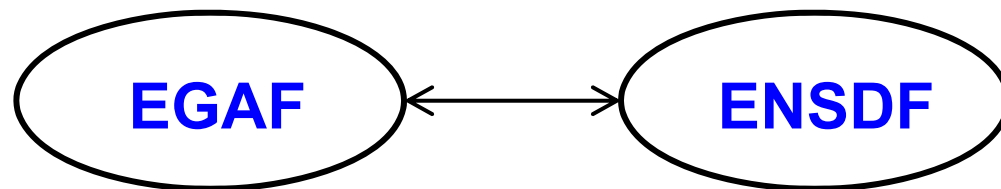
Nuclear structure information derived from independent experimental investigations

Experimental results are reviewed from literature in the public domain

Reaction-evaporation, few-nucleon transfer, Coulomb excitation, beta decay, etc.

Experimental measurements/results are used to collate an experimental database of nuclear data

**ENSDF – Evaluated Nuclear Structure Data File**



**Spectroscopic information pertaining to individual isotopes in the ENSDF repository can be used to identify individual isotopic features in the elemental EGAF capture- $\gamma$  spectrum**

# Application of ENSDF: generation of RIPL



project and ENSDF comprise data complimentary to each repository

step: information in ENSDF used to create a nuclear reaction database called

## **RIPL:- Reference Input Parameter Library**

database also pulls in information from other sources e.g. parameterization of experimentally  
determined giant multipole resonances and level density parameterizations.

Empirical-model calculations in this work are being used to improve knowledge of the decay  
and to determine radiative capture cross sections

Empirical-model calculations call information directly from the RIPL databases

Experimental energy-level information (e.g. energy,  $J\pi$ ) and  $\gamma$ -ray information (e.g. branching  
ratios) is taken from the **RIPL-3** database

Electric multipole parameterization sets are taken from the **RIPL-2** database, level density  
parameterization is taken from Von Egidy [PRC **72**, 044311 (2005)]

CAUTION: RIPL is derived from ENSDF, inaccurate or out-of-date information may carry through

CAUTION: decisions are made in RIPL on ambiguous spins (e.g. "(1/2-,3/2-)"); often random!

CAUTION: 10+ years may have passed since the most recent ENSDF evaluation for a certain

# ENSDF and RIPL databases



## ENSDF

*Evaluated Nuclear Structure Data File*, an electronic database of evaluated experimental nuclear structure data maintained by the National Nuclear Data Center, Brookhaven National Laboratory; online at: <http://www.nndc.bnl.gov/>

## RIPL

R. Capote *et al.*, *RIPL – Reference Input Parameter Library for Calculation of Nuclear Reactions and Nuclear Data Evaluations*, Nuclear Data Sheets 110 (2009) 3107.

T. Belgya *et al.*, *Handbook for calculations of nuclear reaction data, RIPL-2*, IAEA-TECDOC-1506 (IAEA, Vienna, 2006); online at: <http://www-nds.iaea.org/ripl2/>

### Two important points:

Improvements to the RIPL file (and ENSDF database) are ongoing

Statistical-model calculations can provide clarity on energy-level information e.g. alternative  $J\pi$  assignments

Statistical-model calculations can also test the validity of “firm assignments”

All established experimental data can also be used to test the statistical model

# anning DICEBOX



ut file contains structure information extracted from RIPL databases

ut file contains internal conversion coefficient data from BrIcc

oice of level density model:

- CTF (constant temperature formula)

- BSFG (back-shifted Fermi gas)

parameterization from Von Egidy

oice of models of photon strength functions for E1, M1 and E2 transitions e.g.

- SP (single particle) [E1, M1, E2]

- BA (Brink-Axel) [E1]

- KMF (Kademanski, Markushev, Furman) [E1]

- GLO (generalized Lorentzian) [E1]

- SSF (scissors + spin-flip) [M1]

- Islv (isoscalar + isovector) [E2]

r of importance: **E1>>M1>>E2**. Parameterization from RIPL where available (E1) or literature (M1, E2)

**Python scripts developed by Neil Summers can now automate generation of DICEBOX input files**

# Adopted models



Back-shifted Fermi gas (level density) with Von Egidy parameterization

$$\rho(E, J) = f(J) \frac{\exp(2\sqrt{a(E - E_1)})}{12\sqrt{2}\sigma_c a^{1/4} (E - E_1)^{5/4}}$$

*[Till von Egidy and Dorel Bucurescu, Phys. Rev. C 72, 044311 (2005)]*

Generalized Lorentzian E1 PSF with RIPL parameterization (experimental data)

$$f_{GLO}^{E1}(E_\gamma, \Theta) = \frac{1}{3\left(\frac{hc}{2}\right)^2} \left[ \frac{E_\gamma \Gamma_G(E_\gamma, \Theta)}{(E_\gamma^2 - E_G^2)^2 + E_\gamma^2 \Gamma_G^2(E_\gamma, \Theta)} + F_K \frac{4\pi^2 \Theta^2 \Gamma_G}{E_\gamma^5} \right] \sigma_G \Gamma_G$$

Resonance-shape parameters:

$E_G$  (energy centroid),  $\Gamma_G$  (resonance width),  $\sigma_G$  (resonance cross section)

Transitions + spin-flip M1 PSF with parameterization from Yb isotopes

*[Agvaanluvsan et al., Phys. Rev. C 70, 054611 (2004)]*

Transitions + Isovector E2 PSF or single particle