

# Neutron Cross Section Measurements at LBNL

Richard B. Firestone  
Lawrence Berkeley National Laboratory

## Collaborators

Zs. Revay and T. Belgya (Budapest Reactor)

M. Krticka, F. Becvar (Charles University, Prague)

D. McNabb, B. Sleaford, U. Agvaanluvsan (LLNL)



# Measurements

At the Budapest Reactor we have measured thermal neutron  $\gamma$ -ray cross sections for all elements with  $Z=1-83, 92$  except He and Pm.

- Pure thermal guided neutron beam
- Internal standard calibrations
- Precision of  $<3\%$  for strong transitions
- IAEA sponsored evaluation of  $\sigma_{\gamma}$

# Budapest Prompt Gamma-ray Facility



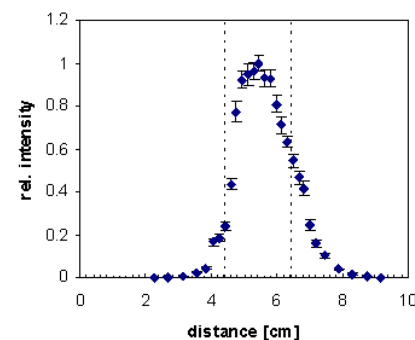
N-type coaxial HPGE detector  
(25%, 1.8 keV@1332)

BGO Compton shield

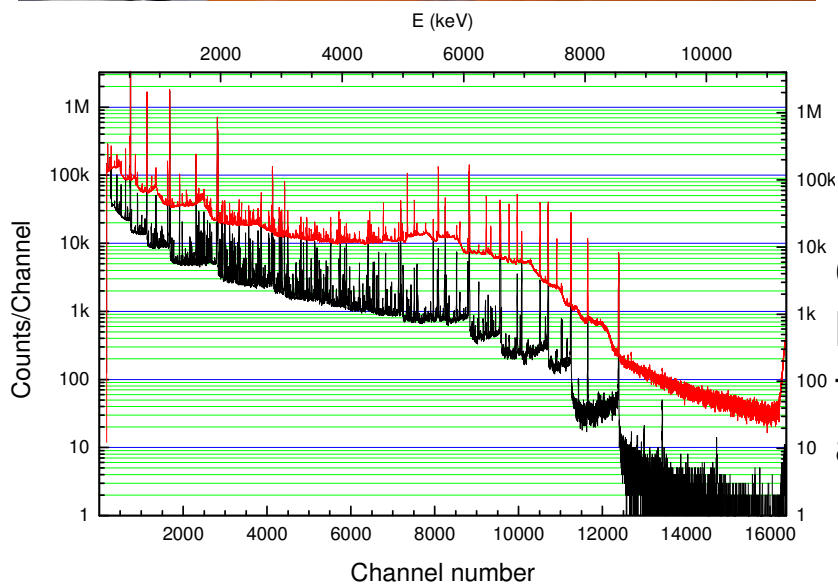
Thermal beam –  $2 \times 10^6 \text{ n} \cdot \text{s}^{-1} \text{cm}^{-2}$

Cold beam –  $5 \times 10^7 \text{ n} \cdot \text{s}^{-1} \text{cm}^{-2}$

Neutron  
beam



Beam profile at the target position



Compton suppression  
lowered background by a  
factor of  $\sim 5$ @1332 to  $\sim 40$   
at 7 MeV.

# Internal Cross Section Calibration

## Calibration Methods

- Stoichiometric compounds of well-known composition containing elements with well-known cross sections  
e.g. H,N,Cl,S,Na,Ti,Au,  $\rightarrow$  KCl,  $(\text{CH}_2)_n$ ,  $\text{Pb}(\text{NO}_3)_2$ ,  $\text{Ti}_2\text{SO}_4$
- Homogenous mixtures
  - Aqueous solutions ( $\text{H}_2\text{O}$ ) or acid solutions (20% HCl)
  - Mixed powders ( $\text{TiO}_2$ )
- Activation product cross section e.g.  $^{28}\text{Al}$ ,  $^{100}\text{Tc}$ ,  $^{235}\text{U}$

# IAEA/EGAF $\sigma_\gamma$ Database

The Evaluated Gamma-ray Activation File (EGAF) is the result of an IAEA CRP established in 1999 to evaluate  $k_0/\sigma_\gamma$  measurements at the Budapest Reactor and compare them with literature data. EGAF contains over 13,000  $\gamma$ -rays from 81 elements. These results are published in

*Database of Prompt Gamma Rays from Slow Neutron Capture for Elemental Analysis*, R.B. Firestone, H.D. Choi, R.M. Lindstrom, G.L. Molnar, S.F. Mughabghab, R. Paviotti-Corcuera, Zs. Revay, V. Zerkin, and C.M. Zhou, IAEA STI/PUB/1263, 251 pp (2007); on-line at <http://www-pub.iaea.org/MTCD/publications/PubDetails.asp?pubId=7030>.

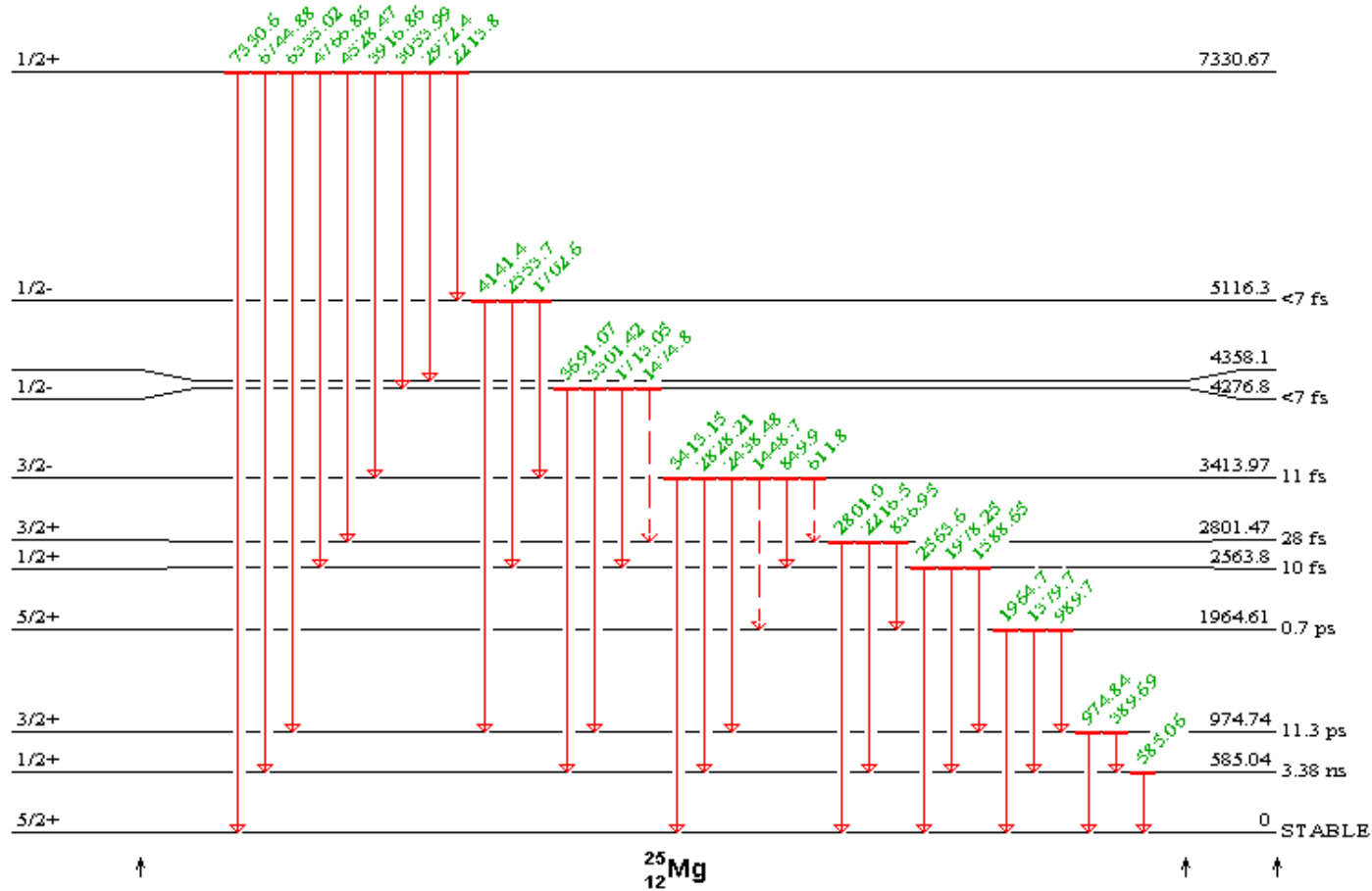
*Handbook of Prompt Gamma Activation Analysis with Neutron Beams*, Zs. Revay, T. Belgya, R.M. Lindstrom, Ch. Yonezawa, D.L. Anderson, Zs. Kasztovsky, and R.B. Firestone, edited by G.L. Molnar (Kluwer Publishers, 2004).

IAEA Prompt Gamma-ray Activation Analysis Viewer:  
<http://www-nds.iaea.org/pgaa/pgaa7/index.html>

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

# Total Thermal Neutron Radiative Cross Sections $\sigma_0$ – Low Z

For *complete decay schemes* the total thermal radiative neutron cross section  $\sigma_0 = \sum \sigma_{\gamma+e}(\text{GS}) = \sum \sigma_{\gamma+e}(\text{CS})$

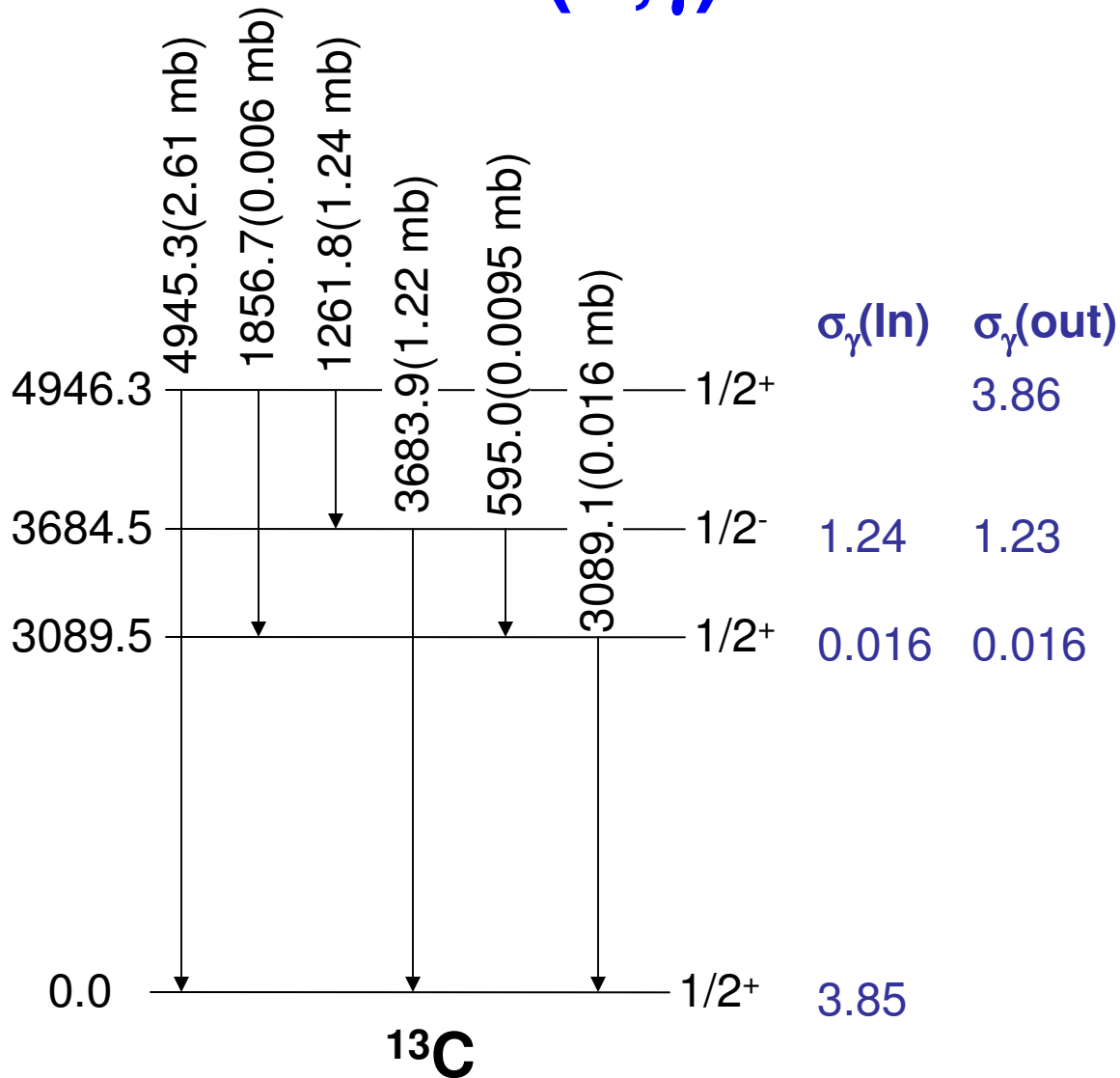


# Example – $^{24}\text{Mg}(n,\gamma)^{25}\text{Mg}$

Cross section balance for the  $^{25}\text{Mg}$  neutron capture decay scheme

E(Level)	$\sigma(\text{in})$	$\sigma(\text{out})$	$\Delta\sigma$
0	0.0536(14)	0.0	0
585.01(3)	0.0406(11)	0.0398(14)	0.0008(18)
974.68(3)	0.0157(4)	0.0158(4)	0.0001(6)
1964.69(10)	0.00022(2)	0.00026(3)	0.00004(4)
2563.35(4)	0.00202(10)	0.00179(7)	0.00023(12)
2801.54(9)	0.00047(4)	0.00061(5)	0.00013(6)
3413.35(3)	0.0411(14)	0.0416(11)	0.0005(18)
4276.33(4)	0.0105(4)	0.0107(3)	0.0002(5)
4358.2(5)	0.00009(2)	0.0	0.00009(2)
5116.37(15)	0.00038(4)	0.00027(3)	0.00011(5)
7330.53(4)	0.0	0.0539(14)	0.0539(14)
	$\sigma(\text{Mughabghab}[23])$	0.0536(15) b	
	$\sigma(\text{Measured, average})$	0.0538(14) b	

# $^{12}\text{C}(n,\gamma)^{13}\text{C}$ Discrepancy



## $^{12}\text{C}(n,\gamma) \sigma_0$ Measurements

Reference	$\sigma_0$ (mb)
Prestwich(1981)	3.50(16)
Jurney(1963)	3.53(7)
Nichols (1960)	3.57(3)
<b>Mughabghab(2006)</b>	<b>3.53(7) mb</b>
Sagot (1963)	3.72(15)
Starr (1962)	3.83(6)
Koechlin (1957)	3.85(15)
Yonezawa (2003)	4.01(15)
<b>This work*</b>	<b>3.86(6) mb</b>

\* Average of measurements with various stoichiometric carbon compounds.



# $(^{15}\text{NH}_2)_2^{13}\text{CO}$ (Urea) Analysis

- Urea sample enriched to >99% in  $^{13}\text{C}$ ,  $^{15}\text{N}$
- Hydrogen internal standard ( $\sigma_0=0.3326(7)$  b)

	<b>Total radiative cross section <math>\sigma_0</math></b>	
<b>Isotope</b>	<b>Mughabgab (2006)</b>	<b>This work</b>
$^{13}\text{C}$	$1.37 \pm 0.04$ mb	$1.50 \pm 0.03$ mb
$^{15}\text{N}$	$24 \pm 8$ $\mu\text{b}$	$39 \pm 3$ $\mu\text{b}$

# Summary of $\sigma_0$ results for low-Z isotopes

Isotope	$\sigma(\text{Atlas})^*$	$\sigma(\text{EGAF})$
$^1\text{H}$	332.6(7) mb	$\approx 332.6(7)$ mb
$^2\text{H}$	0.508(15) mb	0.492(25) mb
$^6\text{Li}$	38.5(30) mb	<b>52.6(22) mb</b>
$^7\text{Li}$	45.4(27) mb	45.7(9) mb
$^9\text{Be}$	8.49(34) mb	8.8(6) mb
$^{10}\text{B}$	305(16) mb	384 mb 8
$^{10}\text{B}(n,\alpha)$	3837(9) b	3820(135) b
$^{11}\text{B}$	5.5(33) mb	<b>11.4(10) mb</b>
$^{12}\text{C}$	3.53(7) mb	<b>3.89(6) mb</b>
$^{13}\text{C}$	1.37(4) mb	<b>1.50(3) mb</b>
$^{14}\text{N}$	80.1(6) mb	79.0(9) mb
$^{15}\text{N}$	24 $\mu\text{b}$ 8	<b>39 <math>\mu\text{b}</math> 3</b>
$^{16}\text{O}$	0.190(19) mb	0.189(8) mb
$^{19}\text{F}$	9.51(9) mb	9.50(11) mb
$^{23}\text{Na}$	517(4) mb	527(7) mb
$^{23}\text{Na}^m(472)$	400(30) mb	<b>478(4) mb</b>

Isotope	$\sigma(\text{Atlas})^*$	$\sigma(\text{EGAF})$
$^{24}\text{Mg}$	53.8(13) mb	53.7(14) mb
$^{25}\text{Mg}$	199(3) mb	197(5) mb
$^{26}\text{Mg}$	38.4(6) mb	37.7(13) mb
$^{27}\text{Al}$	231(3) mb	232(3) mb
$^{28}\text{Si}$	177(4) mb	<b>186(3) mb</b>
$^{29}\text{Si}$	119(3) mb	118(3) mb
$^{30}\text{Si}$	107(2) mb	<b>116(3) mb</b>
$^{31}\text{P}$	165(3) mb	167(5) mb
$^{32}\text{S}$	518(14) mb	536(8) mb
$^{33}\text{S}$	454(25) mb	461(15) mb
$^{34}\text{S}$	256(9) mb	<b>277(8) mb</b>
$^{35}\text{Cl}$	43.6(4) b	43.84(17) b
$^{37}\text{Cl}$	433(6) mb	<b>553(23) mb</b>
$^{39}\text{K}$	2.1(2) b	2.19(3) b
$^{40}\text{K}$	30(8) b	<b>92(8) b</b>
$^{41}\text{K}$	1.46(3) b	<b>1.73(2) b</b>

\*S.F. Mughabghab, Atlas of Neutron Resonances, Elsevier (2006).

# Analysis of $\sigma_0$ for heavier isotopes

For most isotopes with  $Z \geq 20$  the neutron capture decay schemes are incomplete

- High level density below the capture state
- Numerous unresolved continuum gamma rays

What to do?

# $^{105}\text{Pd}(n,\gamma)^{106}\text{Pd}$

E(level)	J <sup>π</sup>	Σσ <sub>γ</sub> (in)	Σσ <sub>γ</sub> (out)	ΔΣσ
0	0+	20.26		
511.844	2+	13.88	17.91	4.03
1128.04	2+	2.371	4.263	1.892
1133.79	0+	0.227	0.565	0.338
1229.2	4+	1.630	3.479	1.849
1557.67	3+	1.183	2.142	0.959
1562.16	2+	0.312	1.869	1.557
1706.44	0+	0.012	0.193	0.181
1909.39	2+	0.063	0.724	0.661
1932.37	4+	0.217	0.590	0.373
2001.56	0+	0.029	0.118	0.089
2077.1	6+	0.001	0.103	0.102
2077.37	(4)+	0.057	0.440	0.383
2084.39	-3	0.123	1.033	0.910
2242.4	2+	0.026	0.499	0.473
2278.47	0+	0	0.056	0.056
2282.89	4+	0.0007	0.275	0.274
2306.01	-3	0.053	0.542	0.489
2308.73	2+	0.000	0.283	0.283
2350.96	4+	0.018	0.304	0.286
2366.09	5+	0.003	0.116	0.114
2397.37	(5)-	0.055	0.263	0.209
2401	(2-,3-)	0.037	0.300	0.263
2439.11	2+	0.065	0.293	0.227
2472.09	0+	0.000	0.055	0.055
2484.76	(1-)	0.043	0.253	0.211
2500.01	-2	0.028	0.296	0.267
2578.64	(4-)	0.00004	0.221	0.221
...	...	...	...	...
...	...	...	...	...
9561.4	2+,3+		0.554	

The cross section deexciting low-lying states in higher-Z nuclei, Σσ<sub>γ</sub>(out), is complete.

The observed cross section populating these states Σσ<sub>γ</sub>(in) is incomplete due to unresolved continuum γ-rays.

$$\sigma_0(\text{tot}) = 21.0 \pm 1.5 \text{ b}$$

Mughabghab (2006)

# Statistical Model Calculations

The continuum contribution can be calculated assuming level density and  $\gamma$ -ray transition probability varies statistically leading to a random distribution of partial level widths  $\langle \Gamma_{if} \rangle = f^{XL}(E_\gamma, \xi) E_\gamma^3 / \rho(E_i, J^\pi_i)$  where

1. Level density  $\rho(E_i, J^\pi_i)$  can be described by

a) Constant temperature formula

$$\rho(E, J) = \frac{f(J)}{T} \exp\left(\frac{E - E_0}{T}\right)$$

b) Back-shifted Fermi Gas formula

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

2. Photon strength  $f^{XL}(E_\gamma, \xi) E_\gamma^3$  for multipolarity XL=E1, M1

a) E1: Brink-Axel

$$f_{BA}^{(E1)}(E_\gamma) = \frac{1}{3(\pi\hbar c)^2} \frac{\sigma_G E_\gamma \Gamma_G^2}{(E_\gamma^2 - E_G^2)^2 + E_\gamma^2 \Gamma_G^2}$$

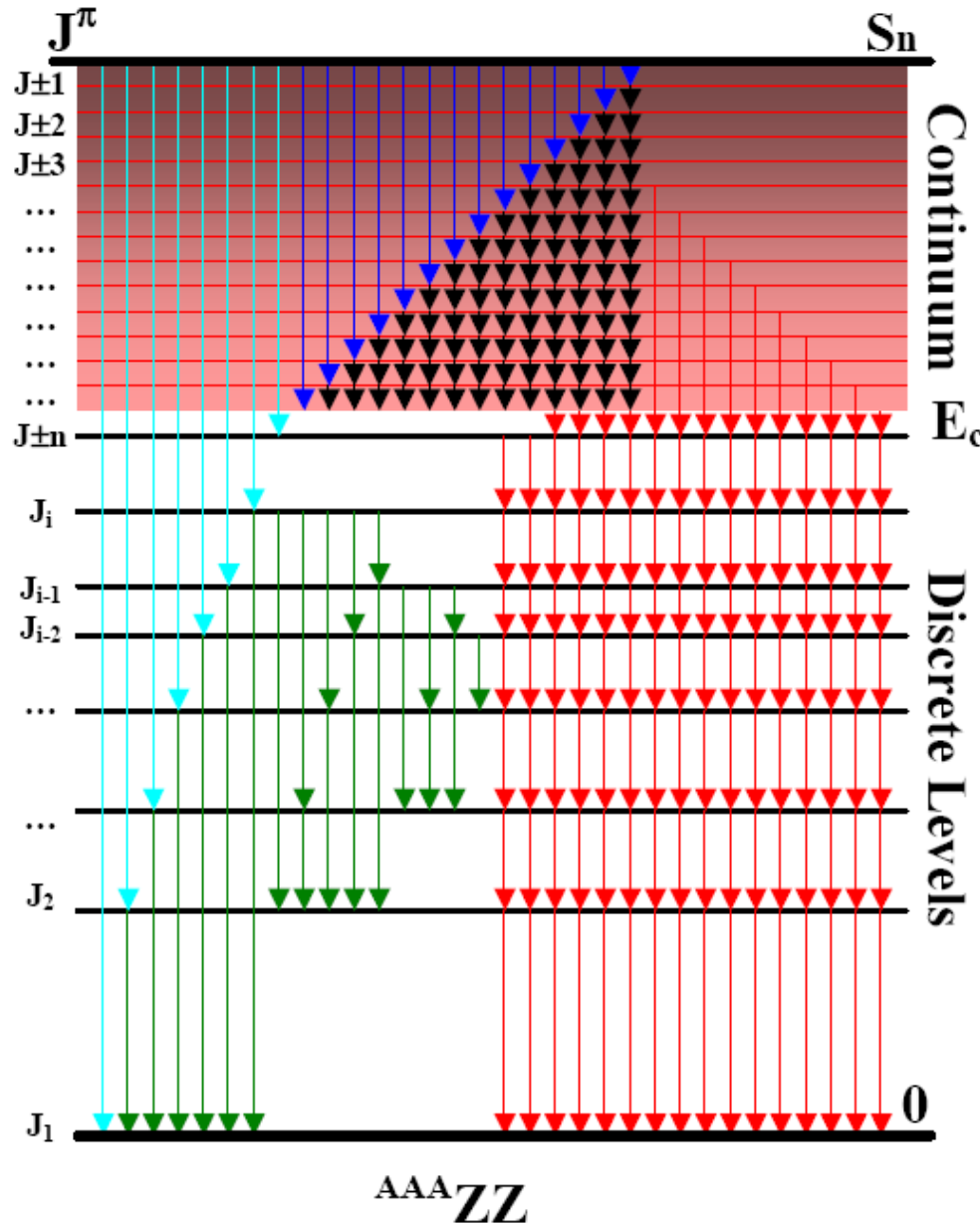
also, Kadenskii *et al* (KMF), Generalized Lorentzian (GLO).

GDER parameters: Dietrich, Berman(ATNDT) or Herman (Empire)

b) M1: Single particle,  $f^{E2}(SP) = 5 \times 10^4 E_\gamma^3$ , spin flip (SF), or  $f^{E1}/f^{M1} = 5-7$

c) E2: Single particle,  $f^{E2}(SP) = 1 \times 10^{-7} E_\gamma^5 / A^{4/3}$

# DICEBOX Monte Carlo Code



DICEBOX generates  $(n, \gamma)$  level scheme simulations (nuclear realizations) based on statistical model level densities  $\rho(E_i, J^\pi_i)$  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.

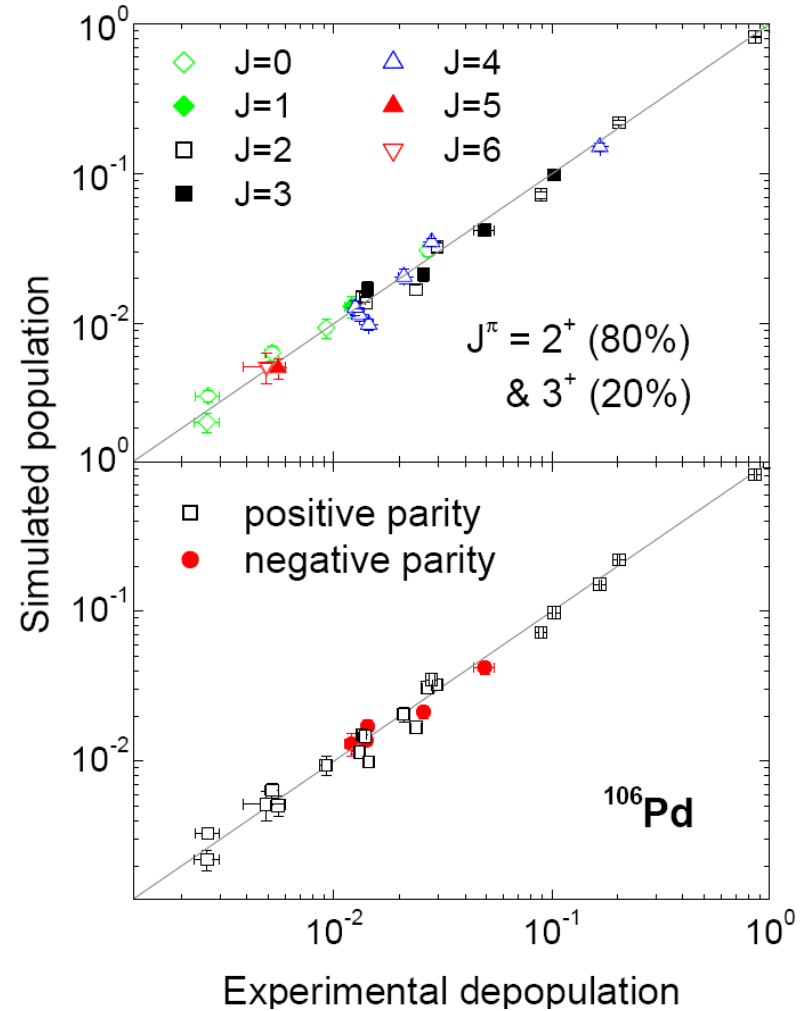
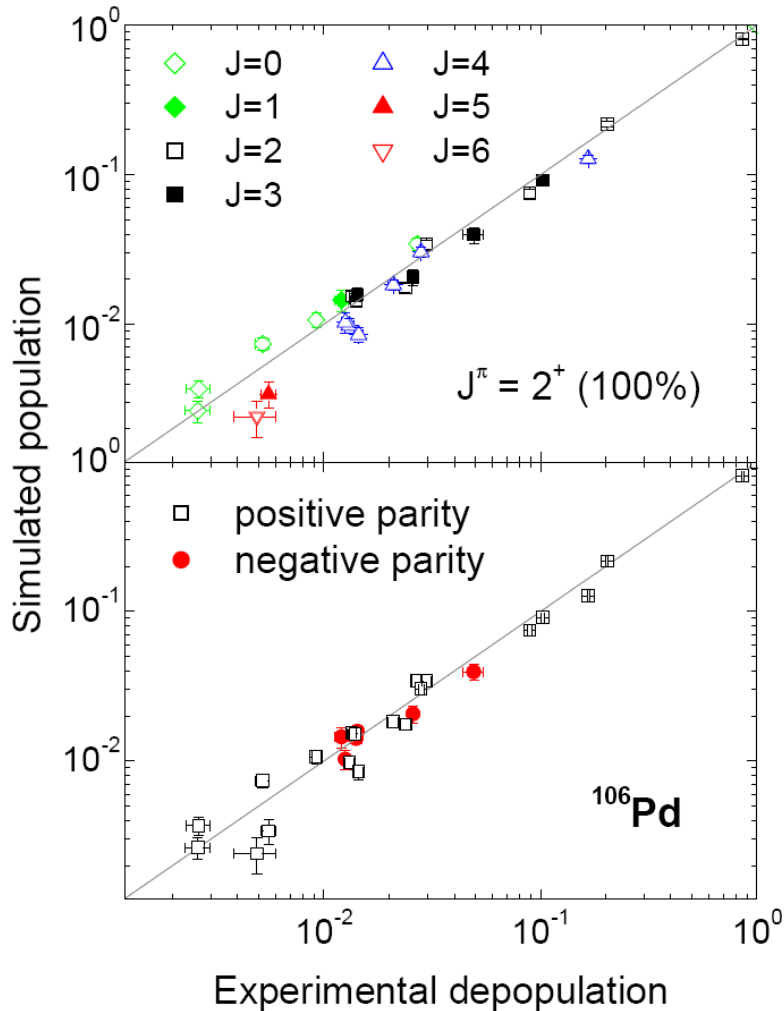
# Statistical Model Selection Example

Model dependence of the total capture state width  $\Gamma_{\gamma}^{\text{tot}}$

$^{105}\text{Pd}(n,\gamma)$	E1-PSF*	M1-PSF	$\rho(E,J)$	$\Gamma_{\gamma}^{\text{tot}}$
	Brink-Axel	SP	CTF	$410 \pm 47$
	Brink-Axel	SF	CTF	$352 \pm 42$
	KMF	SP	BSFG	$201 \pm 14$
	KMF	SF	BSFG	$172 \pm 12$
<b>Best fit</b> 	<b>GLO</b>	<b>SP</b>	<b>BSFG</b>	<b><math>156 \pm 8</math></b>
	GLO	SF	BSFG	$126 \pm 8$
	Experiment (Mughabghab, 2006)			<b><math>148 \pm 10</math></b>

\*GDER parameters from Dietrich and Berman, ADNDT **38**, 199 (1988).

# Comparison of $^{105}\text{Pd}(n,\gamma)$ simulated $\Sigma\sigma_{\gamma}(\text{in})$ with experimental $\Sigma\sigma_{\gamma}(\text{out})$



$\sigma_0 = \sigma_{\gamma}(\text{GS})_{\text{expt}} + \sigma_{\gamma}(\text{GS})_{\text{calc}} = 20.3 \pm 0.3 \text{ b} + 1.4 \pm 0.3 \text{ b} = 21.7 \pm 0.5 \text{ b}$   
 $\sigma_0(\text{Mughabghab, 2006}) = 21.0 \pm 1.5 \text{ b}$



# Determination of $\sigma_0$ for $^{104}\text{Pd}(n,\gamma)^{105}\text{Pd}$

If minimal experimental data is available  $\sigma_0$  can be calculated with DICEBOX independently for each observed level feeding.

PSF		LD			Level Feeding(%)		
E1	M1	$\rho(E,J)$	GDER	$E_{\text{crit}}(\text{keV})$	280	306	344
GLO	SP	BSFG	Dietrich	350	23(9)	4.3(13)	10(3)
KMF	SP	BSFG	Herman	350	26(9)	4.1(12)	10(3)
				<b>Average</b>	<b>24(9)</b>	<b>4.2(12)</b>	<b>10(3)</b>
				$\sigma_{\gamma}^{\text{expt}}(\text{b})$	0.145(13)	0.040(8)	0.099(18)
				$\sigma_0(\text{b})$	<b>0.60(23)</b>	<b>0.95(35)</b>	<b>0.99(35)</b>
				<b>Average</b>	<b>0.77(17) b</b>		

**Mughabghab, 2006**

**0.65(30) b**

## Pd $\sigma_0$ results\*

Reaction	$\sigma_0$ (literature) (barns)	$\sigma_0$ (this work) (barns)
$^{102}\text{Pd}(n,\gamma)^{103}\text{Pd}$	$1.6\pm 0.2$	$1.1\pm 0.4$
$^{104}\text{Pd}(n,\gamma)^{105}\text{Pd}$	$0.65\pm 0.30$	$0.77\pm 0.17$
$^{105}\text{Pd}(n,\gamma)^{106}\text{Pd}$	$21.0\pm 1.5$	$21.7\pm 0.5$
$^{106}\text{Pd}(n,\gamma)^{107}\text{Pd}$	$0.30\pm 0.03$	$0.36\pm 0.10$
$^{108}\text{Pd}(n,\gamma)^{109}\text{Pd}$	$7.6\pm 0.5$	$7.2\pm 0.5$
$^{108}\text{Pd}(n,\gamma)^{109}\text{Pd}^m$	$0.185\pm 0.010$	$0.185\pm 0.011$
$^{110}\text{Pd}(n,\gamma)^{111}\text{Pd}$	$0.70\pm 0.17$	$0.34\pm 0.10$

\* Submitted to Physical Review C.

# Future Directions

- The EGAF database will be expanded to include activation data in collaboration with the IAEA CRP *Reference Database for Neutron Activation Analysis*.
- Total thermal radiative neutron cross sections  $\sigma_0$  will be derived from EGAF data for all isotopes.
- The EGAF  $\sigma_0$  values will be compared with literature values and new evaluated  $\sigma_0$  values will be included in EGAF.
- Capture  $\gamma$ -ray spectra will be compared with DICEBOX calculations to find input parameters that best reproduce the continuum shape.
- EGAF will be extended to include continuum  $\gamma$ -rays.
- DICEBOX calculations will be extended to use the thermal parameters for calculations at higher neutron energies.
- ENDF format  $\gamma$ -ray libraries will be generated in collaboration with LLNL.