New Methods for the Determination of Total Radiative Thermal Neutron Cross Sections (σ_0)

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Collaborators

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Measurements

At the Budapest Reactor we measured thermal neutron γ-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 σ_γ

Budapest Prompt Gamma-ray Facility

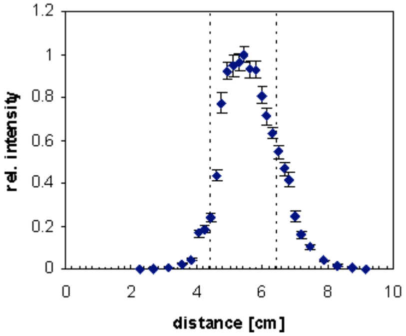
N-type coaxial HPGE detector
(25%,1.8 keV@1332)

BGO Compton shield

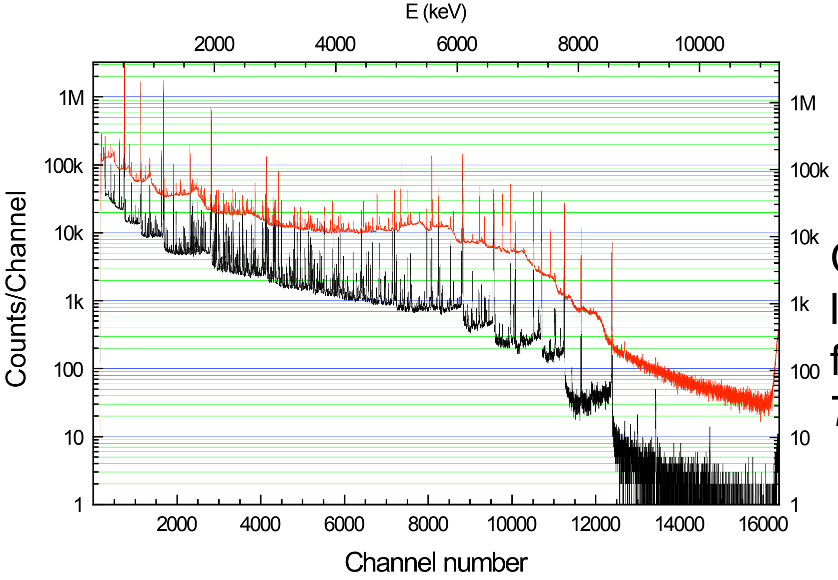
Thermal beam – 2×10^6 n·$s^{-1}$$cm^{-2}$

Cold beam – 5×10^7 n·$s^{-1}$$cm^{-2}$

Neutron
beam



Beam profile at the target position



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

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Internal Cross Section Calibration

- Stoichiometric compounds containing elements with well-known cross sections

H, N, Cl, S, Na, Ti, Au

e.g. KCl, $(CH_2)_n$, $Pb(NO_3)_2$, Tl_2SO_4

- Homogenous mixtures

Aqueous (H_2O) or acid (20% HCl) solutions, mixed powders (TiO_2)

- Cross section of activation products

^{19}F, ^{28}Al, ^{100}Tc, ^{235}U

IAEA/EGAF σ_γ Database

An IAEA Coordinated Research Project was established in 2000 to evaluate the Budapest k_0/σ_γ measurements and other literature data. This led to the Evaluated Gamma-ray Activation File (EGAF).

EGAF contains over 13,000 γ-rays from 79 elements and is the only source of precise k_0/σ_γ values. These results are available 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.

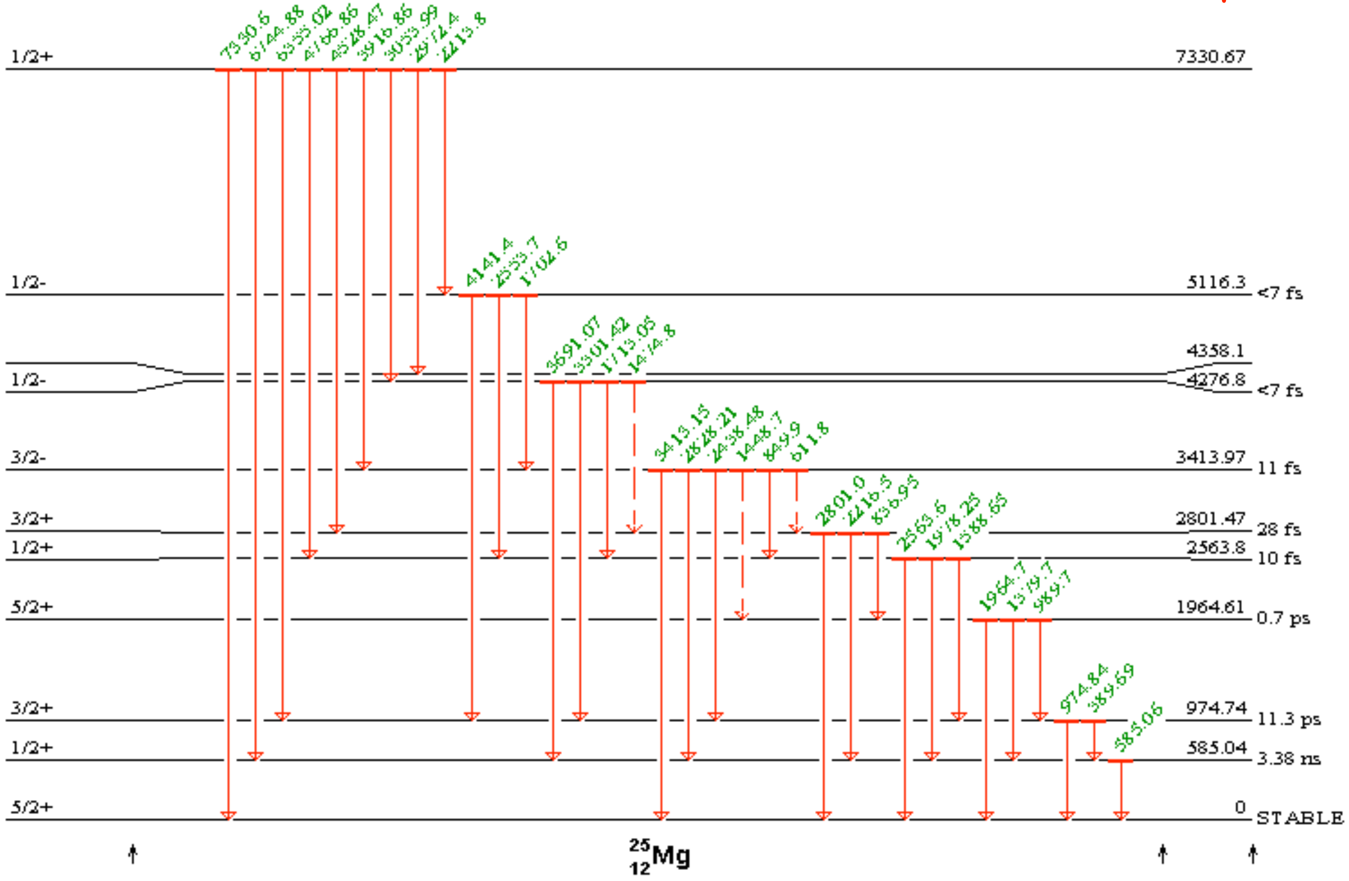
Handbook of Prompt Gamma Activation Analysis with Neutron Beams, Zs. Revay, T. Belgya, R.M. Lindstrom, Ch. Yonezawa, D.L. Anderson, Zs. Kasztovsky, and R.B. Firestone, edited by G.L. Molnar (Kluwer Publishers, 2004).

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 σ_0 – Low Z

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



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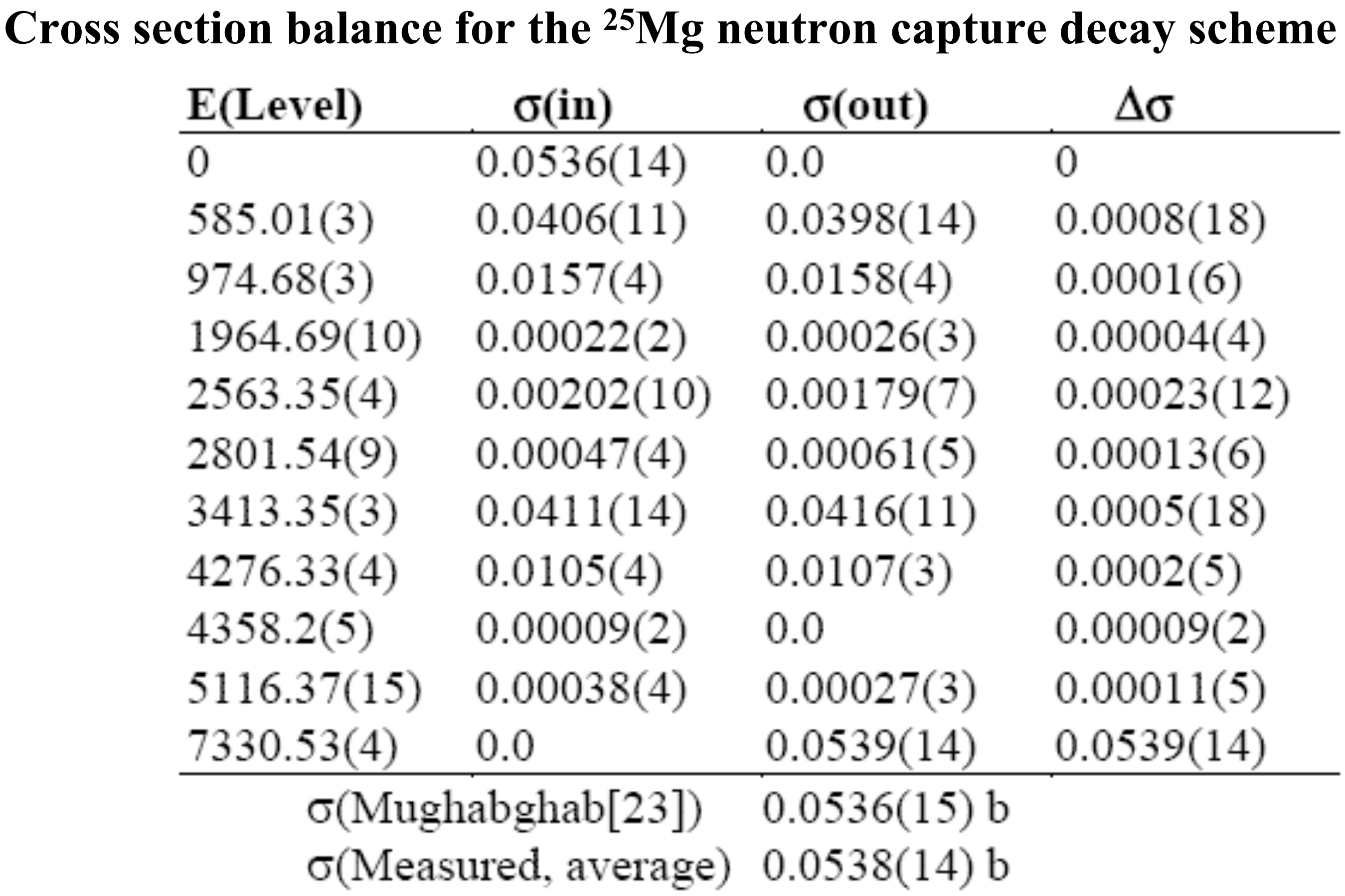
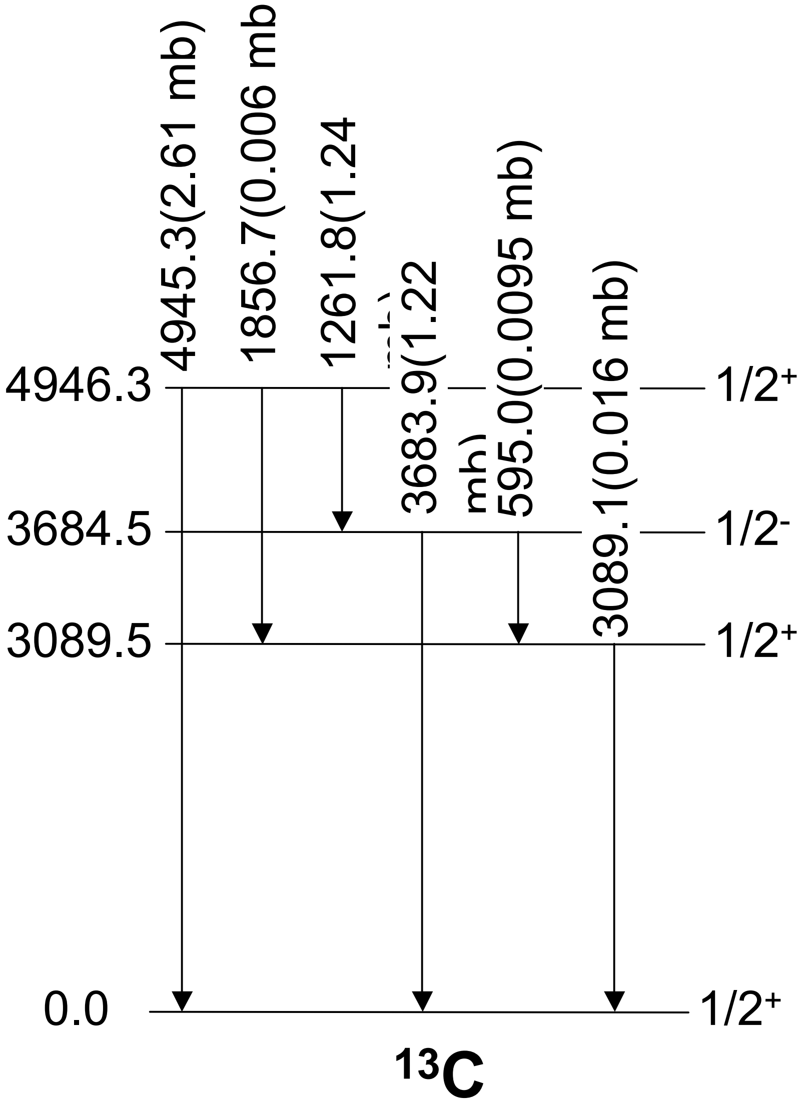
Example – $^{24}Mg(n,\gamma)^{25}Mg$



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

E(Level)	σ(in)	σ(out)	Δσ
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)
	σ(Mughabghab[23])	0.0536(15) b	
	σ(Measured, average)	0.0538(14) b	

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



Level	σ_γ(In)	σ_γ(out)
4946.3		3.86
3684.5	1.24	1.23
3089.5	0.016	0.016
0.0	3.85	

$^{12}C(n,\gamma)$ σ_0 Measurements

Reference	**σ_0(mb)**
Prestwich(1981)	3.50(16)
Jurney(1963)	3.53(7)
Nichols (1960)	3.57(3)
Sagot (1963)	3.72(15)
Starr (1962)	3.83(6)
Koechlin (1957)	3.85(15)
Yonezawa (2003)	4.01(15)
This work*	3.86(6) mb
Mughabghab(2006)	3.53(7) mb

* Average of measurements with various stoichiometric carbon compounds.

$(^{15}NH_2)_2{}^{13}CO$(Urea) Analysis

- Urea sample enriched to >99% in ^{13}C, ^{15}N
- Hydrogen internal standard (σ_0=0.3326(7) b)

	Total radiative cross section σ_0	
Isotope	**Mughabgab (2006)**	**This work**
^{13}C	1.37±0.04 mb	1.50±0.03 mb
^{15}N	24±8 μb	39±3 μb

Summary of σ_0 results for low-Z isotopes

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

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

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

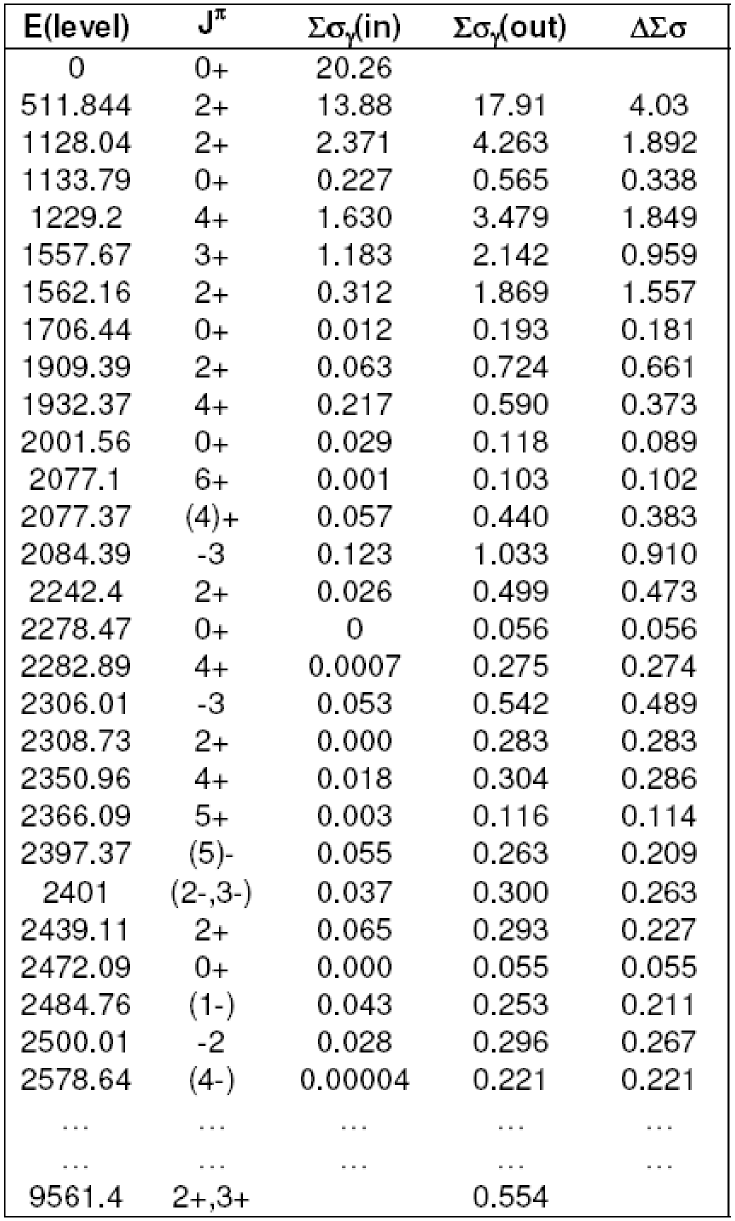
Analysis of σ_0 for heavier isotopes

For most isotopes with Z≥20 the neutron capture decay schemes are incomplete

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

What to do?

^{105}Pd(n,γ)^{106}Pd



E(level)	J^π	$\Sigma\sigma_\gamma$(in)	$\Sigma\sigma_\gamma$(out)	$\Delta\Sigma\sigma$
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, $\Sigma\sigma_\gamma$(out), is complete.

The observed cross section populating these states $\Sigma\sigma_\gamma$(in) is incomplete due to unresolved continuum γ-rays.

σ_0(tot)=21.0±1.5 b*

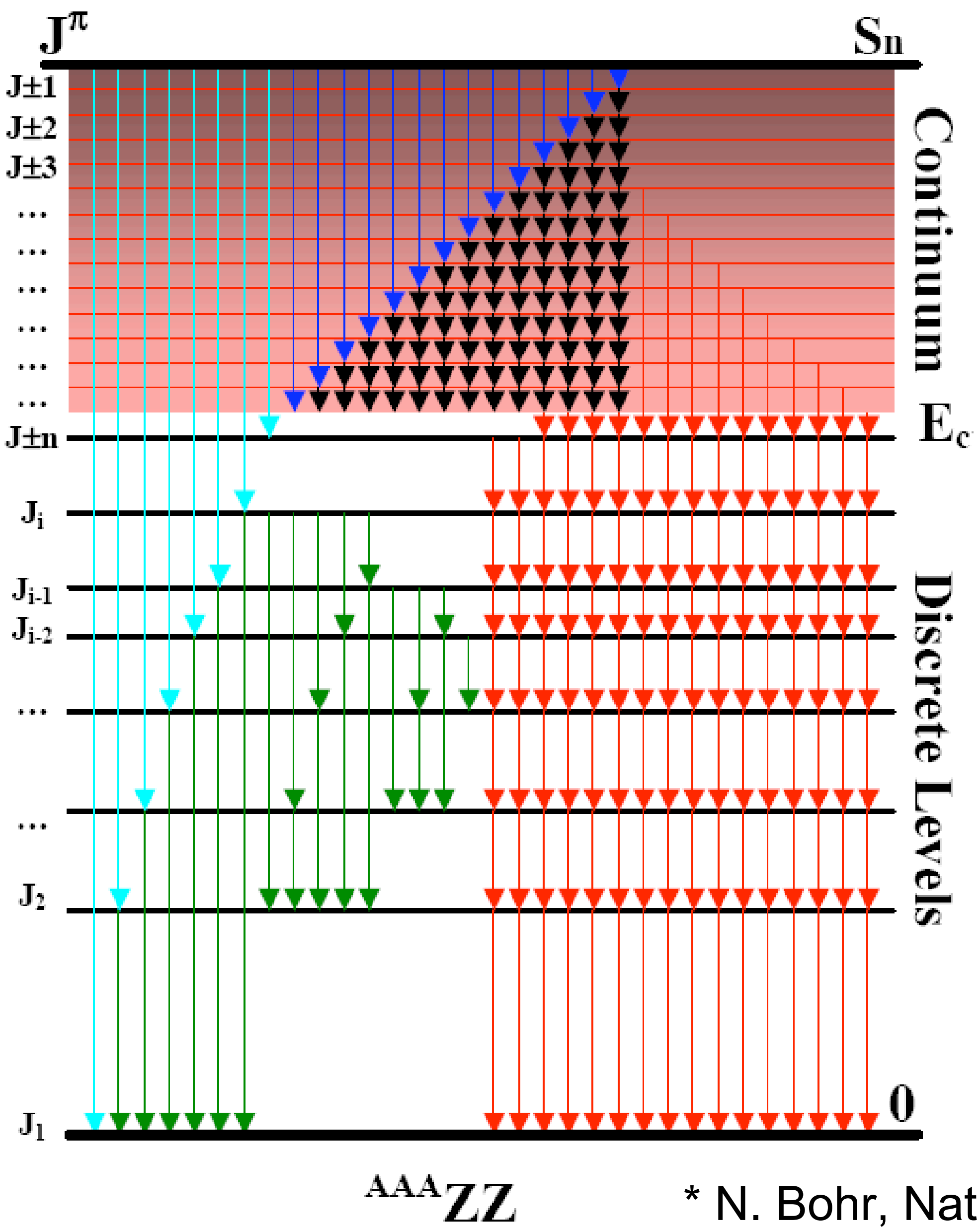
Mughabghab (2006)

Statistical Model Calculations

The continuum contribution can be calculated assuming level density and γ-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
   a) Brink-Axel E1
   $$f^{(E1)}_{\mathrm{BA}}(E_\gamma) = \frac{1}{3(\pi\hbar c)^2} \frac{\sigma_G E_\gamma \Gamma_G^2}{\left(E_\gamma^2 - E_G^2\right)^2 + E_\gamma^2 \Gamma_G^2}$$
   b) Other E1 models, Kadmenskii *et al* (KMF), Generalized Laurentzian (GLO), …
   c) Single particle (SP), spin flip (SF), fixed M1, or f^{E1}/f^{M1}=5-7
   d) Single particle E2, $f^{E2}(\mathrm{SP}) = 5\times10^{-11} E_\gamma^5$

DICEBOX Monte Carlo Code



The DICEBOX algorithm is based on a generalization of the extreme statistical model embodying Bohr's idea of a compound nucleus*.

1. Generate system of Γ_{if} describing the level scheme decay properties (nuclear realization) where
   - a) All levels and γ-rays below E_{crit} are taken from experiment.
   - b) All levels and γ-rays above E_{crit} are generated randomly from level density and PSF models
   - c) Primary γ-ray cross sections are taken from experiment when known.
2. Randomly generate capture state γ-ray decay cascades (30,000)
3. Perform numerous (50) realizations and average to get statistical variation of the models.

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* N. Bohr, Nature **137**, 344 (1936)

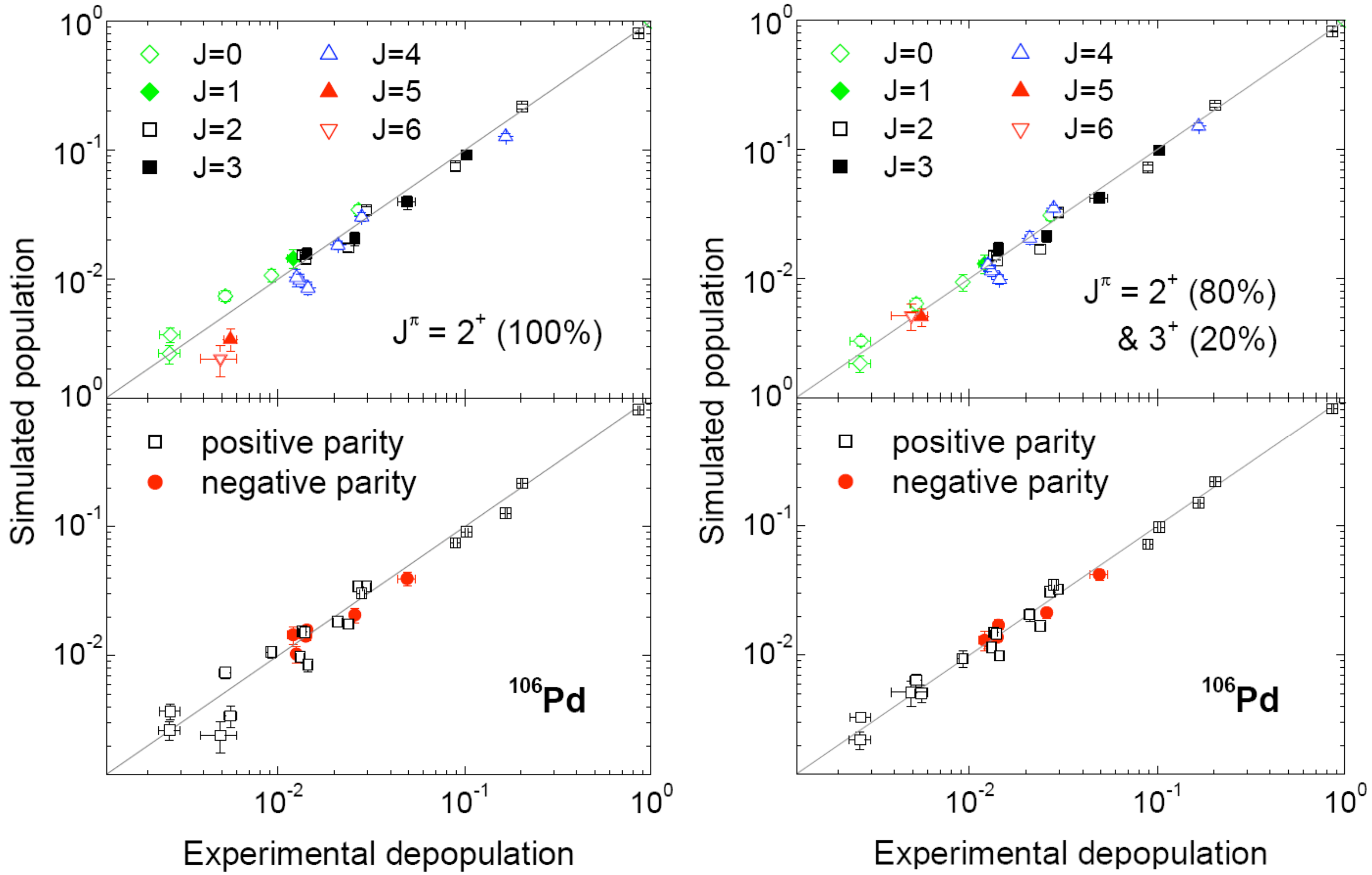
Statistical Model Selection

Model dependence of the total capture state width Γ_γ^{tot}

^{105}Pd	E1-PSF*	M1-PSF	$\rho(E,J)$	Γ_γ^{tot}
	Brink-Axel	SP	CTF	410±47
	Brink-Axel	SF	CTF	352±42
	KMF	SP	BSFG	201±14
	KMF	SF	BSFG	172±12
Best fit →	GLO	SP	BSFG	156±8
	GLO	SF	BSFG	126±8
	Experiment (Mughabghab, 2006)			148±10

* Giant dipole resonance parameters from Dietrich and Berman, At. Data Nucl Data Tables **38**, 199 (1988).

Comparison of $^{105}Pd(n,\gamma)^{106}Pd$ DICEBOX $\Sigma\sigma_\gamma$ (in) with Experimental $\Sigma\sigma_\gamma$(out)



$\sigma_0=\sigma_\gamma(GS)_{expt}+\sigma_\gamma(GS)_{calc}=20.3\pm0.3\ b+1.4\pm0.3\ b = 21.7\pm0.5\ b$

σ_0(Mughabghab, 2006)=21.0±1.5 b

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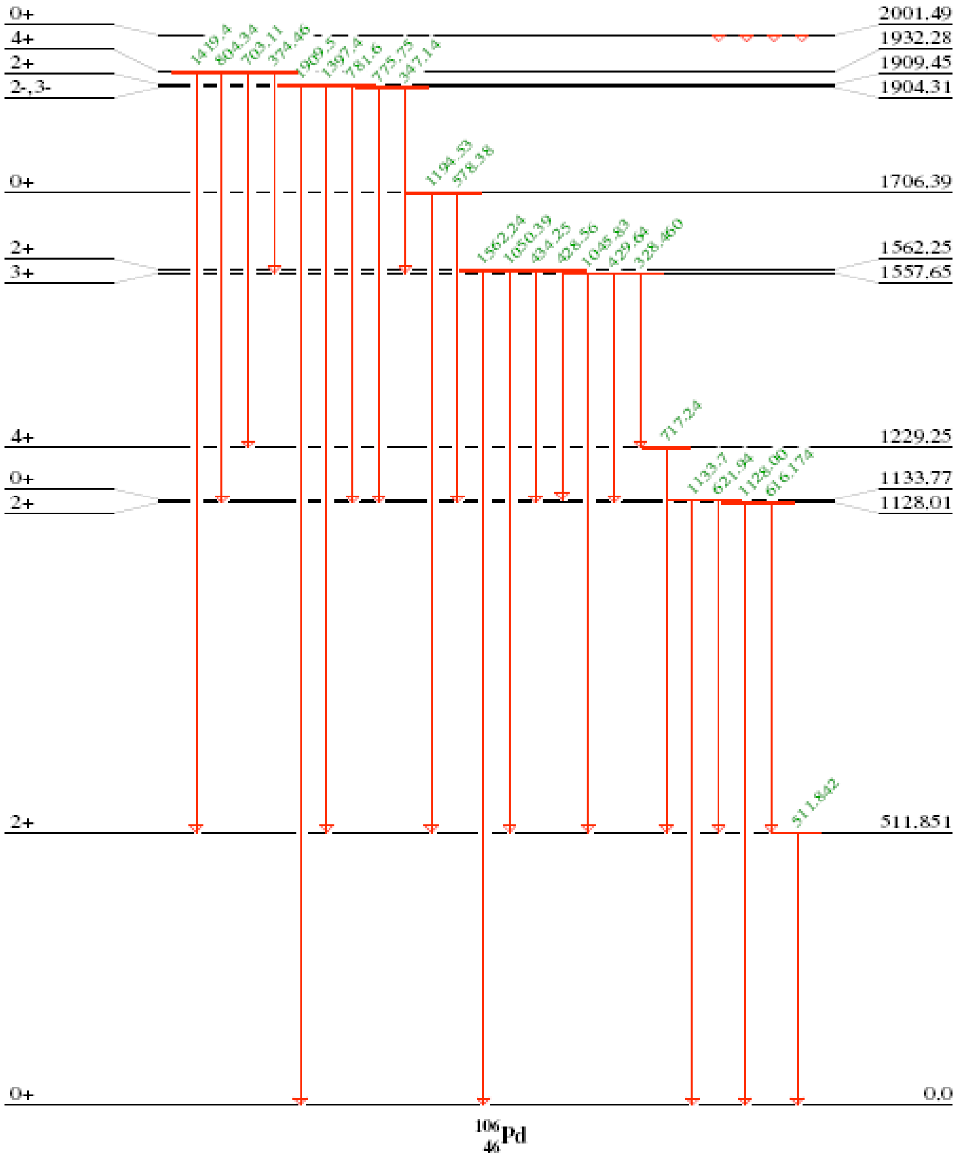
Determination of σ_0 for ^{104}Pd(n,γ)^{105}Pd

When only limited experimental data is available, level feedings(%) can be calculated with DICEBOX and σ_0 determined for each observed level.

PSF		**LD**			**Level Feeding(%)**		
E1	**M1**	**ρ(E,J)**	**GDER**	**E_{crit}(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)
				Average	**24(9)**	**4.2(12)**	**10(3)**
				σ_γ^{expt} (b)	0.145(13)	0.040(8)	0.099(18)
				σ_0 (b)	**0.60(23)**	**0.95(35)**	**0.99(35)**
				Average	**0.77(17) b**		

Mughabghab, 2006 0.65(30) b

Nuclear Structure Errors



Wrong level placement

A level assigned at 1904.3 keV by the Ritz principal is populated by $\sigma_\gamma(\mathrm{out})_{\mathrm{expt}}$=0.12 b, which is inconsistent with 1.13 b predicted by DICEBOX.

Placement of the 347- and 776-keV γ-rays out of the 1909.3 keV level gives $\sigma_\gamma(\mathrm{out})_{\mathrm{expt}}$=0.62 b, which is consistent with 0.83 b predicted by DICEBOX for that level.

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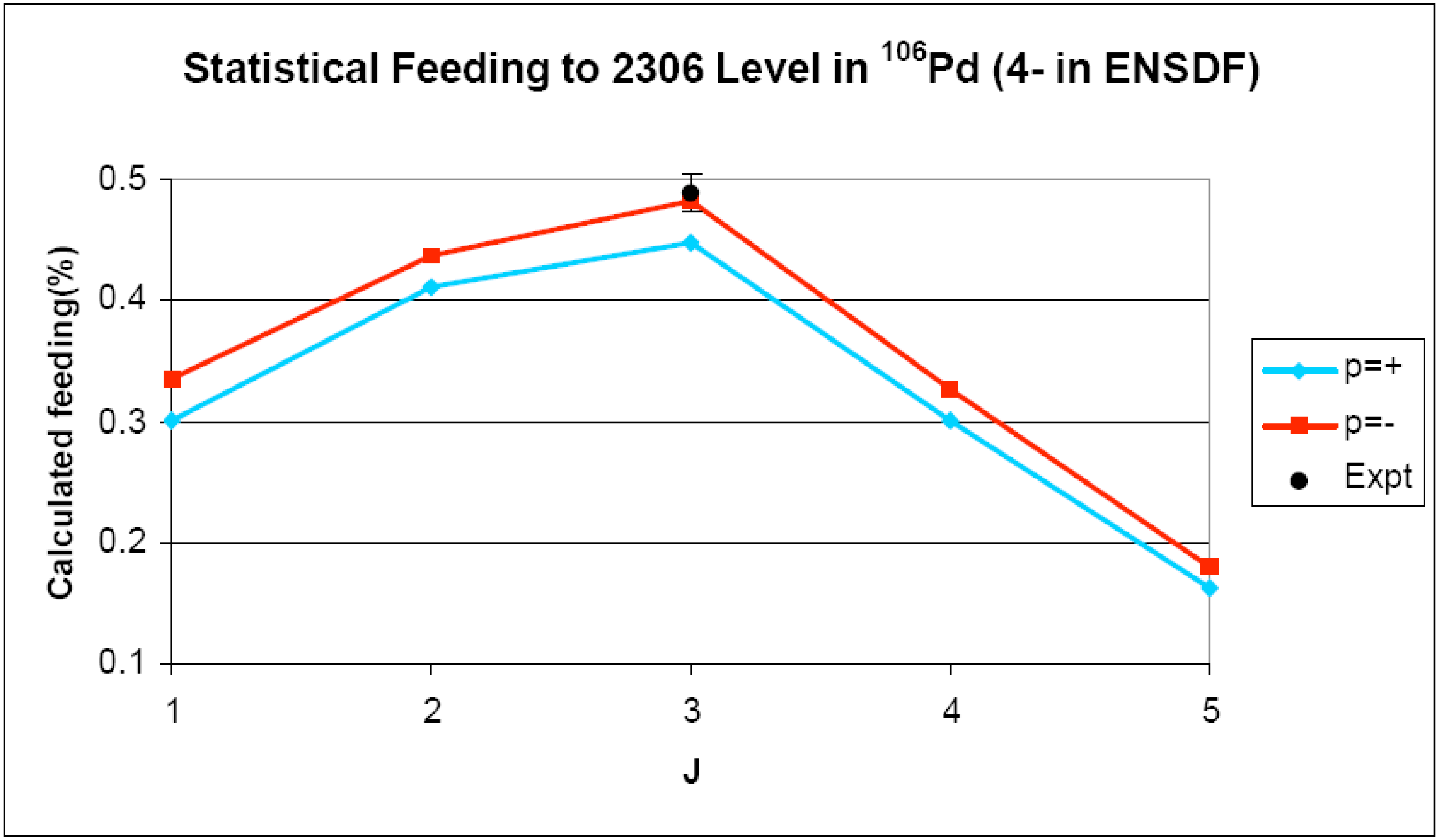
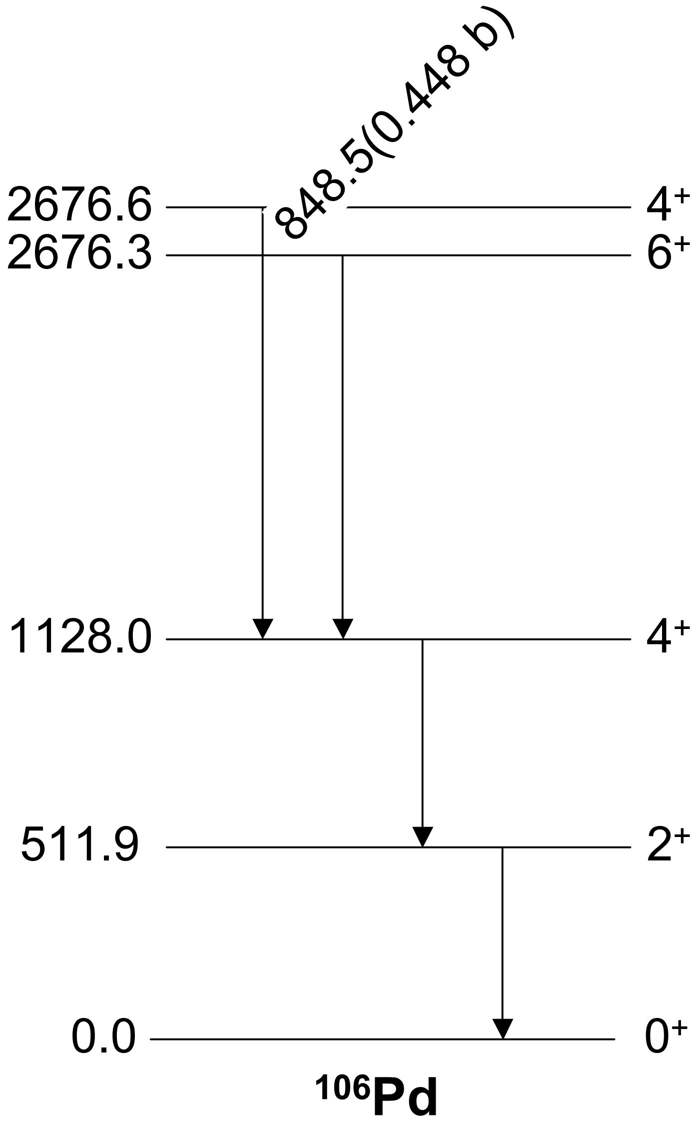
Statistical J^{π} Assignment



Dividing unresolved γ-ray intensities



848.5 keV doublet can be resolved from calculated population of 4^+ and 6^+ levels.

	E_γ	Transition	Cross Section
1.	848.5 keV	$4^+ \rightarrow 4^+$	0.345±0.022 b
2.	848.5 keV	$6^+ \rightarrow 4^+$	0.103±0.022 b

Pd σ_0 results

Reaction	σ_0(literature) (barns)	σ_0(this work) (barns)
^{102}Pd(n,γ)^{103}Pd	1.6±0.2	1.1±0.4
^{104}Pd(n,γ)^{105}Pd	0.65±0.30	0.77±0.17
^{105}Pd(n,γ)^{106}Pd	21.0±1.5	21.7±0.5
^{106}Pd(n,γ)^{107}Pd	0.30±0.03	0.36±0.10
^{108}Pd(n,γ)^{109}Pd	7.6±0.5	7.2±0.5
^{108}Pd(n,γ)^{109}Pd^{m}	0.185±0.010	0.185±0.011
^{110}Pd(n,γ)^{111}Pd	0.70±0.17	0.34±0.10

Conclusions

- The Budapest Reactor thermal neutron beam σ_γ measurements directly yield new, precise σ_0 values, free of corrections for contributions from reactor (fast) neutrons for low-Z isotopes.
- Accurate σ_0 values can be determined for high-Z isotopes from σ_γ measurements plus statistical calculations using the DICEBOX Monte Carlo code to account for the continuum contribution.
- Analysis of existing data from the Budapest Reactor will allow us to produce a new, self-consistent set of thermal neutron capture radiative cross sections from nearly all isotopes.