EFNUDAT

European Facilities for Nuclear Data Measurements

2nd EFNUDAT workshop

Determination of thermal radiative capture cross section

Tamás Belgya

Institute of Isotopes Hungarian Academy of Sciences, H-1525, POB 77, Budapest, Hungary

*E-mail: Belgya@IKI.KFKI.HU, http://www.iki.kfki.hu/nuclear/

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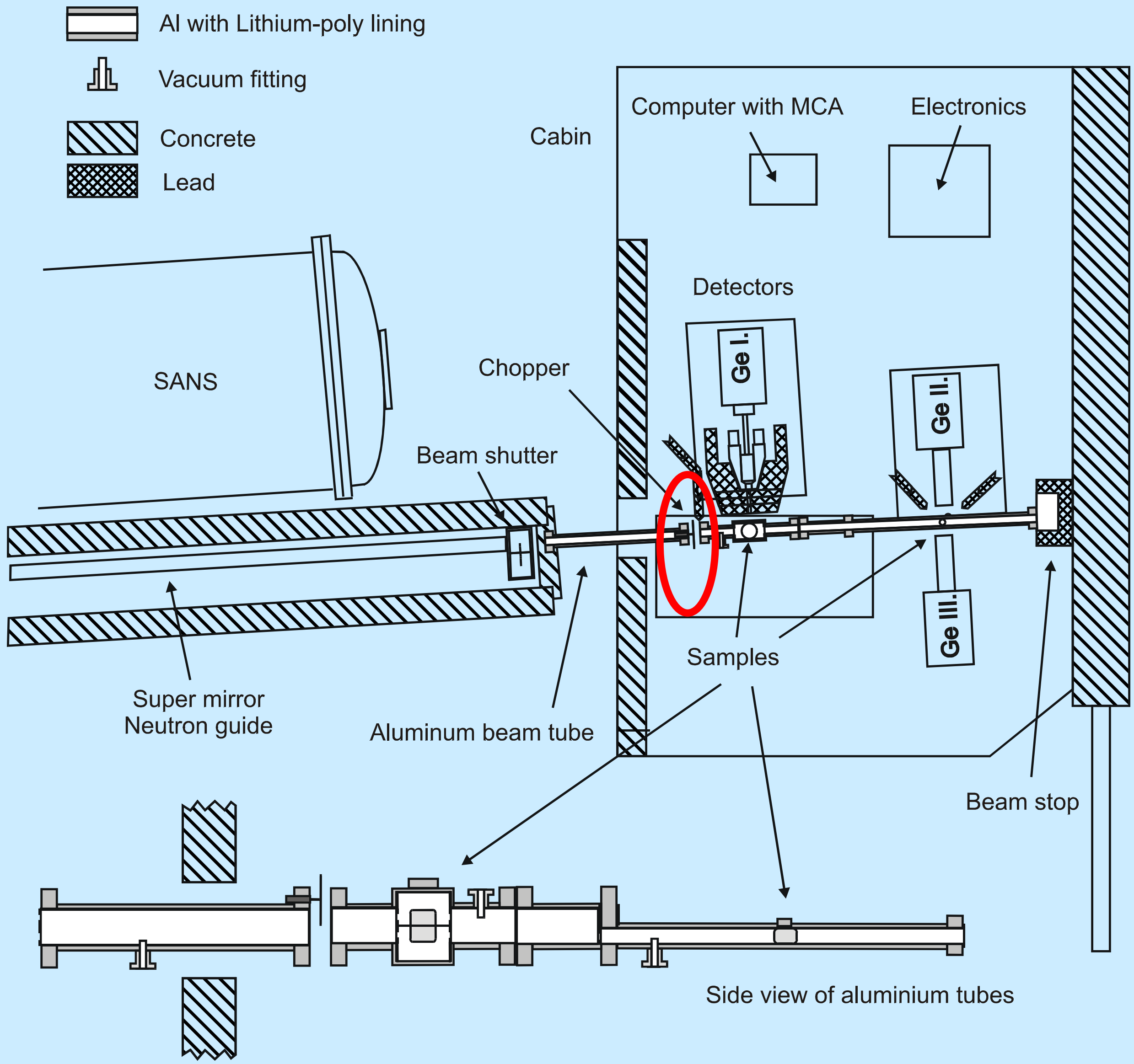
Content

- PGAA-NIPS experimental facilities at the Budapest
- Precision of internal calibration of partial γ-ray Xsections
- Crossing Intensity Sums (CIS)
  - Intensities of the $^{14}N(n,\gamma)$ high energy standard
- Determination of thermal neutron capture Xsections with γ-ray spectroscopy and its limitations
- Completeness from experimental point of view
  - An example
- Conclusion

The PGAA-NIPS facility



PGAA-NIPS facilities

Partial γ-ray production cross section
(standardization)

- Comparator method for prompt activation
  - Standardization (table) or dedicated standardization experiment
  - Provides thermal Xsection values for 1/v nuclei

$$\sigma_{\gamma x} = \sigma_{\gamma c} \frac{n_c}{n_x} \frac{A_{\gamma x}}{A_{\gamma c}} \frac{\varepsilon(E_{\gamma c})}{\varepsilon(E_{\gamma x})} \frac{f(E_{\gamma c})}{f(E_{\gamma x})}; \quad \sigma_\gamma = \theta P_\gamma \sigma_{th}$$

Partial γ-ray Xsection	σ_γ
Number of atoms (integer for chemical compounds)	n_x and n_c
Peak area	A
Detector efficiency	ε
Absorption correction	f
Enrichment	θ
Absolute γ-ray decay probability	P_γ

Precision of internal σ_γ calibration for enriched samples

$$\sigma_{\gamma x} = \sigma_{\gamma c} \; \frac{n_c}{n_x} \; \frac{A_{\gamma x}}{A_{\gamma c}} \; \frac{\varepsilon(E_{\gamma c})}{\varepsilon(E_{\gamma x})} \; \frac{f(E_{\gamma c})}{f(E_{\gamma x})}$$

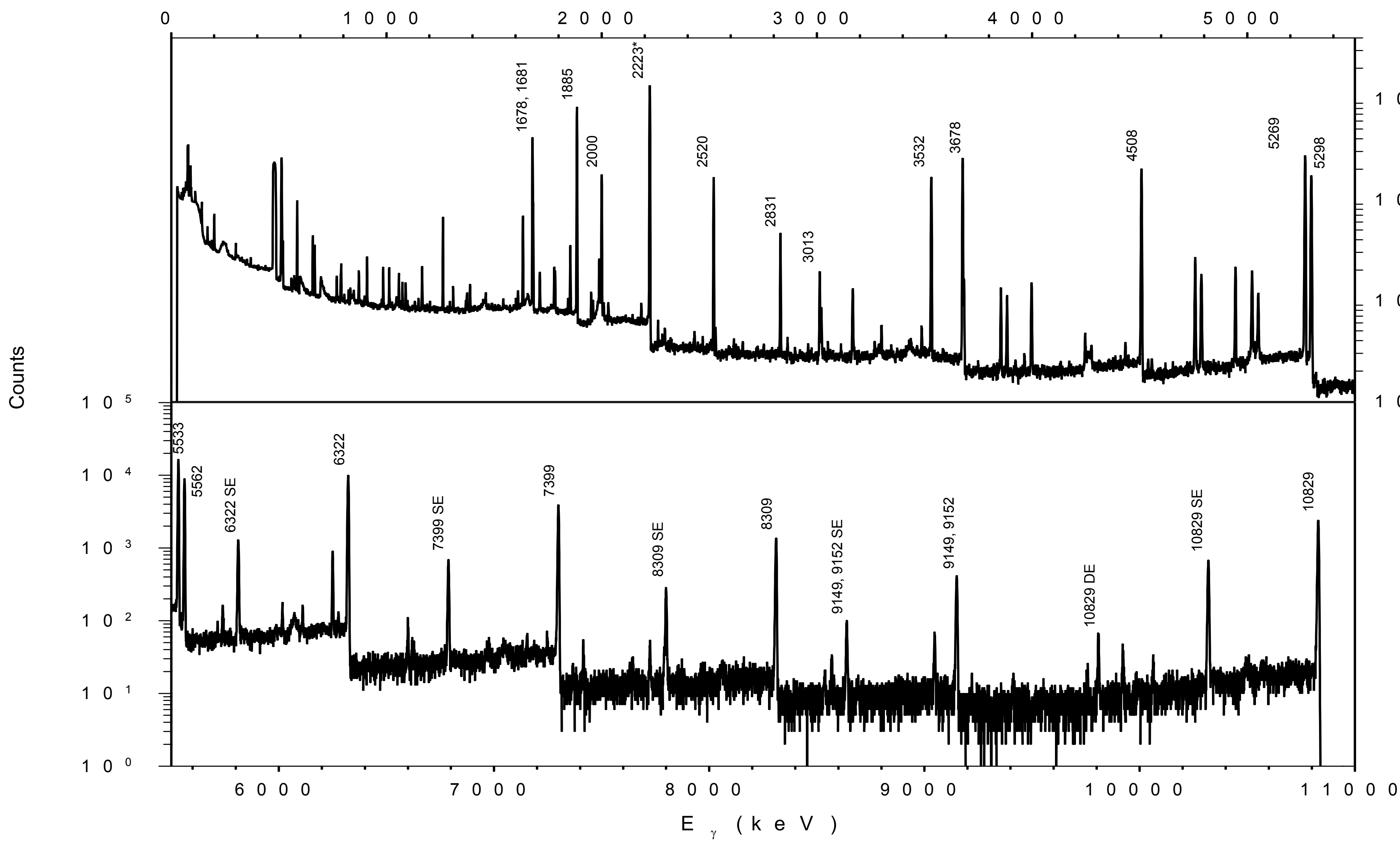
1 2 3 4 5

Factor number	Type of error
1. Derived from primary (H) or secondary (Cl, N…) standards (table or dedicated experiment)	Systematic (~ 1%)
2. Exactly 1	0
3. Uncertainty can be decreased by time or by count rate	Statistical
4. Uncertainty can be decreased to systematic level (Most important term which can be minimized by improving standards)	Statistical + systematic, 0 at pivot ($\equiv E_{\gamma c}$) point, (~ 1%)
5. Can be measured or calculated using attenuation models (thin sample minimizes the uncertainty)	Statistical + systematic or systematic (~ 1%)

Improvement of photo peak efficiency standards

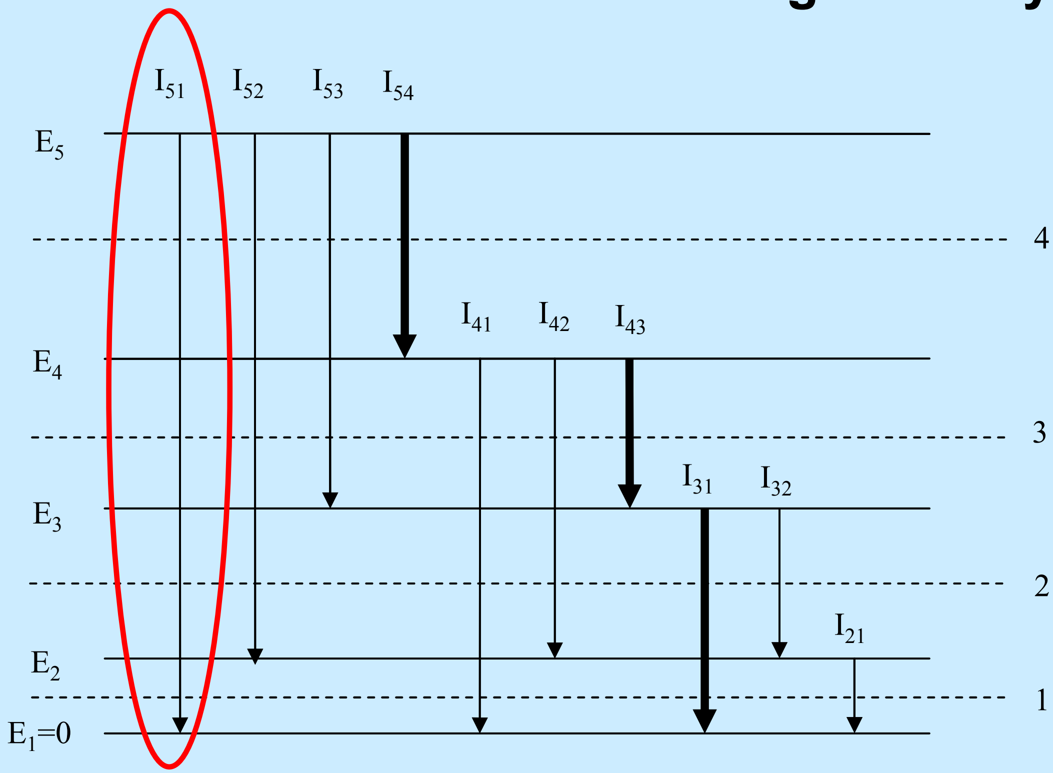
- High energy radioactive γ-ray sources
  - ^{56}Co, ^{66}Ga and ^{226}Ra
    - G.L. Molnár, Zs. Révay, T. Belgya., Rep.INDC(NDS)-437 (2002) 23
    - Baglin, C.M., E. Browne, E.B. Norman, G.L. Molnár, T. Belgya, Zs. Révay, and F. Szelecsényi, Nucl. Instrum. Methods **A 481** (2002) 365-377
    - Bé, M.M., V.P. Chechev, R. Dersch, O.A.M. Helene, R.G. Helmer, M. Herman, S. Hlavac, A. Marcinkowski, G.L. Molnár, A.L. Nichols, E. Schönfeld, V.R. Vanin, and M.J. Woods, Update of X-ray and gamma ray decay data standards for detector calibrations and other applications. Vol. I and II. 2007, Vienna: IAEA
- High energy capture sources
  - ^{14}N(n,γ)^{15}N primary source
    - Belgya, T., Improved accuracy of gamma-ray intensities from basic principles for the calibration reaction ^{14}N(n,γ)^{15}N. Phys. Rev. **C 74** (2006) 024603-1-8
  - ^{35}Cl(n,γ) and ^{NAT}Cr(n,γ) secondary sources (not yet fully compatible with N source)
    - Molnár, G.L., Z. Révay, and T. Belgya, Accurate absolute intensities for the ^{35}Cl(n,gamma) reaction gamma-ray standard. Nucl. Instrum. Methods **B 213** (2004) 32-35
    - Belgya, T. and G.L. Molnár, Accurate relative gamma-ray intensities from neutron capture on natural chromium. Nucl. Instrum. Methods **B 213** (2004) 29-31

$^{14}N(n,\gamma)$ spectrum



Determination of nitrogen intensities and detector efficiency function in one step

Crossing Intensity Sum (CIS)



$$I_{i,j} = A_{i,j}\,\varepsilon^{-1}(E_{i,j})$$

$$T_f = \sum_{\substack{i>f \\ j\le f}} I_{i,j} = \sum_{\substack{i>f \\ j\le f}} A_{i,j}\varepsilon^{-1}(E_{i,j}) = C \qquad f = 1,2,\ldots n-1$$

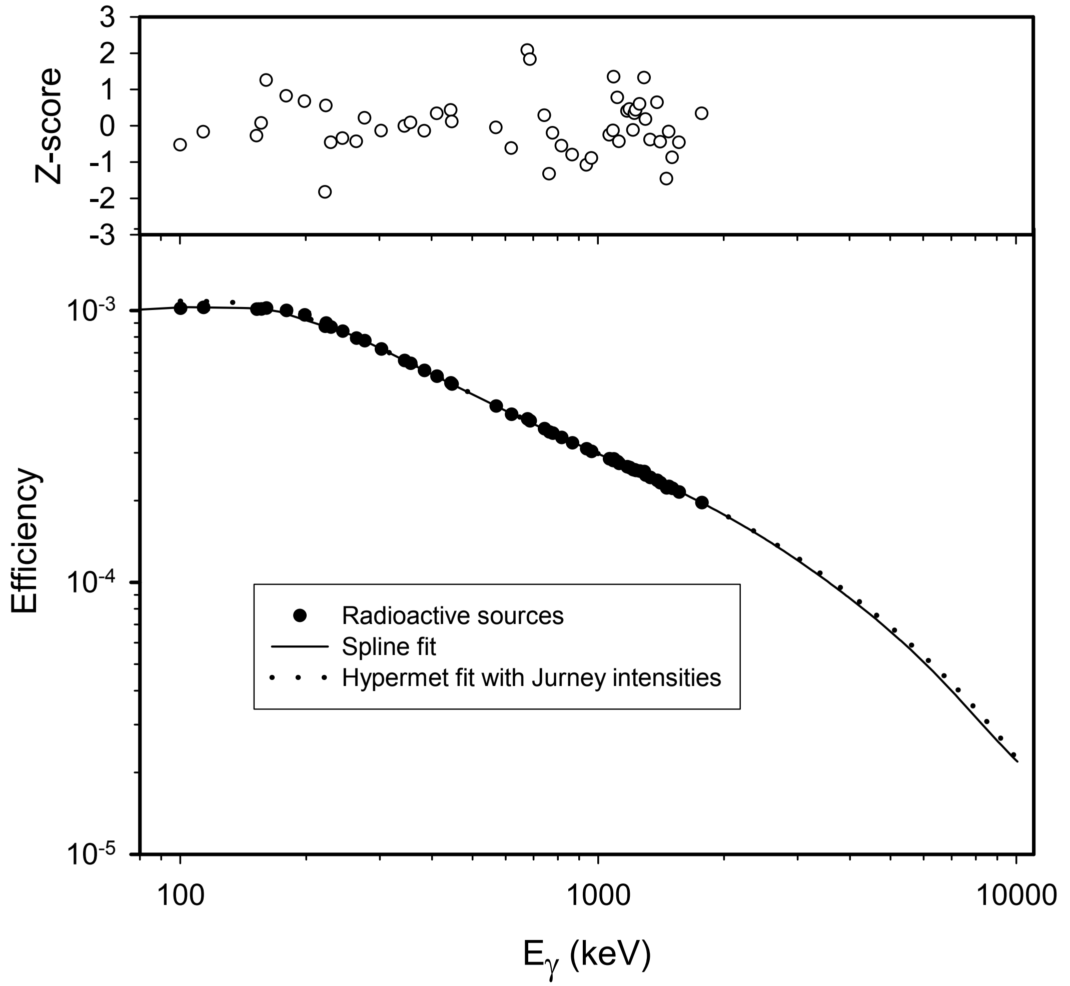
$$Q = \sum_{\substack{1\le f\le n-1 \\ 1\le s\le n-1}} (T_f - C)w_{f,s}(T_s - C) + \sum_m \frac{\left(\tilde{\varepsilon}_m^{-1} - \varepsilon^{-1}(E_{i,j})\right)^2}{\sigma_m^2}$$

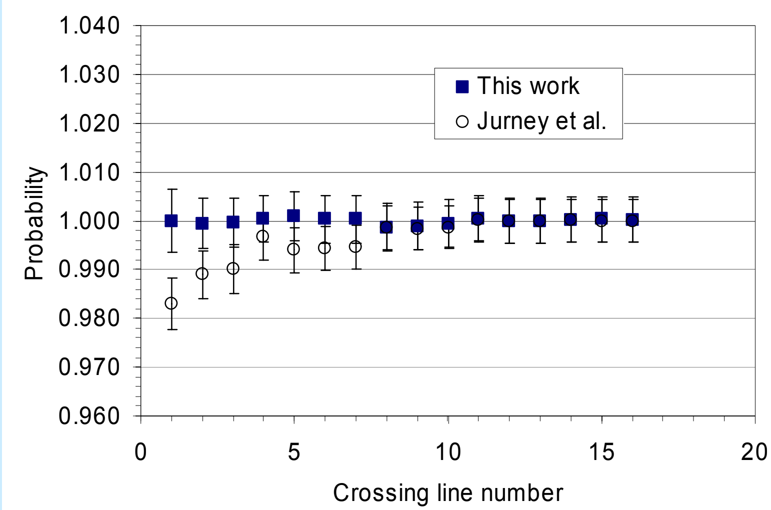
- Sums of crossing intensities are constant C
- Least-squares fit for inverse efficiency function ε^{-1} and C
- Input is peak areas and efficiencies at low energy
- CIS for line 1 ≡ intensity sum to the ground state
- CIS for line n-1 ≡ intensity sum for the primary transitions

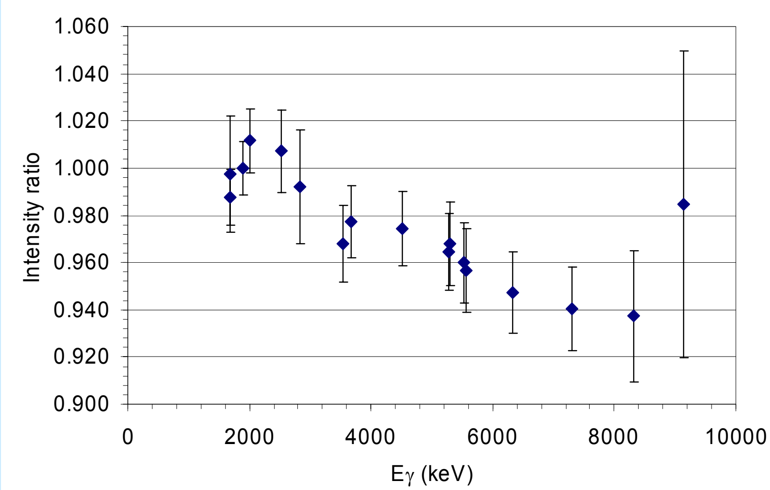
$$\text{New}: \quad \sigma_{th} = 80 - 83\ mb$$

$$\text{Mughabghab}: \quad \sigma_{th} = 79.8(14)\ mb$$

Results







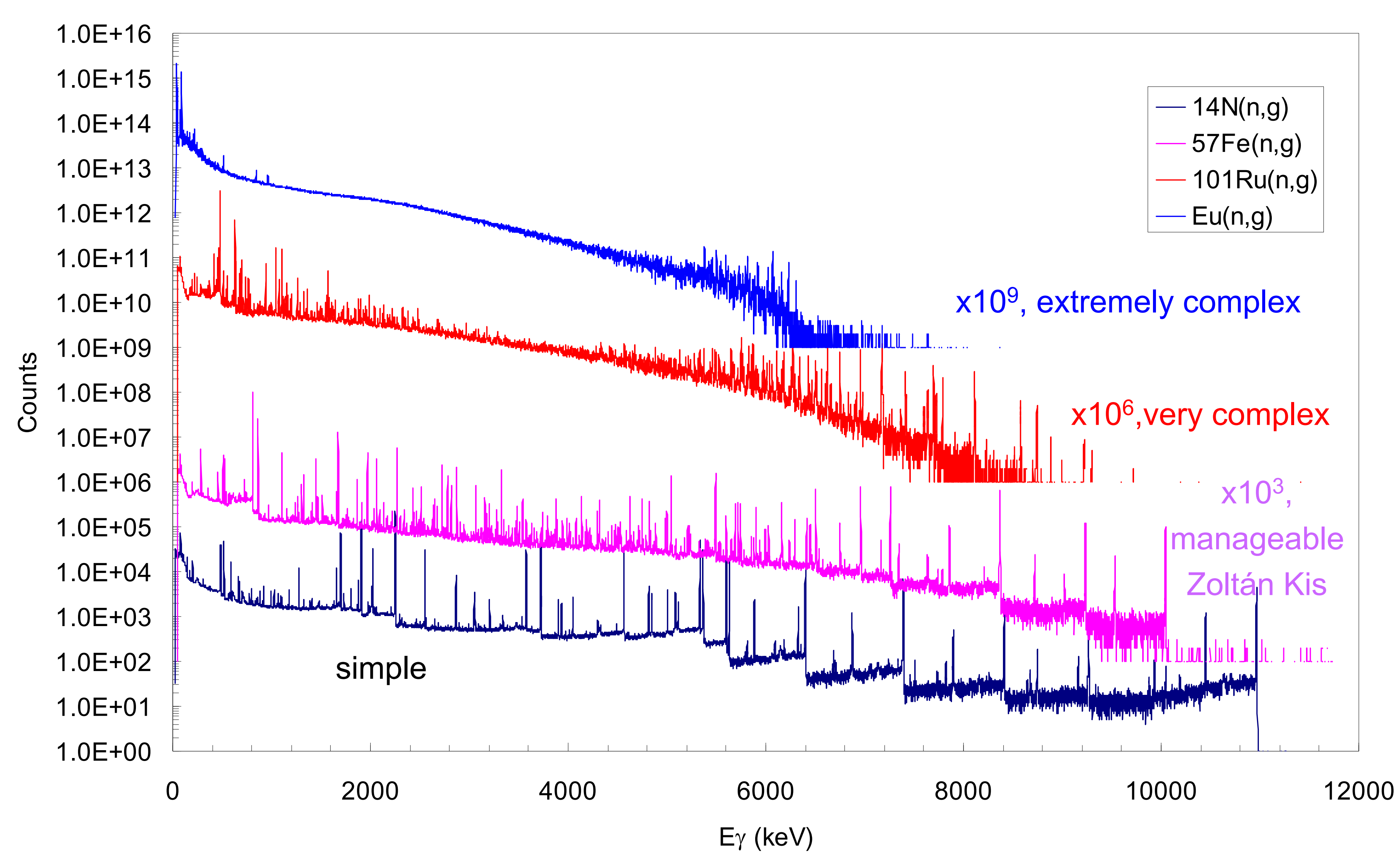
Ratio=Intensity Jurney / Intensity this work

Total neutron capture cross section from γ-spectroscopy

Method	Equation	Notes
1	$\sigma_{th} = \dfrac{\sigma_\gamma}{\theta P_\gamma}$	P_γ must be known, for example from beta decay if the captured nucleus is unstable.
2	$\sigma_{th} = \sum_{f=1}^{n-1} \sigma_{\gamma\, C \to f} (1+\alpha_f)(1+PCC_f)$	The sum of all primary transitions from the capture state can be used for nuclei with relatively simple decay scheme.
3	$\sigma_{th} = \sum_{i=2}^{n} \sigma_{\gamma\, i \to g.s.} (1+\alpha_i)(1+PCC_i)$	The sum of all ground state transitions can be used for nuclei with relatively simple decay scheme. Conversion coefficients α must be known.
4	Average of CISs: $Q = \min\left(\sum_{\substack{1 \le f \le n-1 \\ 1 \le s \le n-1}} (T_f - \sigma_{th}) w_{f,s} (T_s - \sigma_{th}) \right)$	Well balanced and relatively simple decay scheme. Conversion coefficients α must be known.
5	$\sigma_{th} = \sum_i E_i \sigma_{\gamma i} (1+\alpha_i)(1+PCC_i) / B_n$	The energy weighted sum can be used for any nuclei with resolved gamma-transitions. E_i is the energy of the transition, B_n is the binding energy and *PCC* is the pair conversion.

For which nuclei we can use that

(Simple and complex spectra)



Consequences

- Method 1 based on decay γ-rays or X-rays can be used for any complexity
  - Precision depends on the P_γ value
- Method 2-5 is applicable for nuclei with manageable decay scheme (~500 γ-rays)
  - Limitation can be the unobserved conversion electron intensities
- Method 5 is applicable for spectra with resolvable γ-lines (~ 700-800 γ-rays)
  - Limitation can be the unobserved conversion electron intensities
  - Similar to the weighting function method used with C_6D_6 detectors
- Above that quasi continuum of γ-rays appears in the spectra
  - A possible approach is stripping or deconvolution of spectra on the few percent level accuracy
  - C_6D_6 detector or total absorption detector (needs highly enriched sample, unobserved conversion electron can also be a problem)

Question of completeness from the experimental point of view

- The Q-value test (equivalent to method 5) was applied in the past to estimate completeness

$$B_n = \sum_{i \in observed\ \gamma} \frac{E_i \sigma_{\gamma i}}{\sigma_{th}}; \quad \sigma_{th} = \sum_{i \in observed\ \gamma} \frac{E_i \sigma_{\gamma i}}{B_n}$$

- This is still the best way to find out the degree of completeness, however we propose to use the inverse Q-value instead and compare it directly to other independent Xsection values
- There are experimental methods that do not depend on observing all of the γ-rays, they provide the independent Xsections (they usually have other problems)
  - Pile oscillator, activation, transmission and calorimeter

An example

$^{101}Ru(n,\gamma)$ reaction (proposed by ILL)

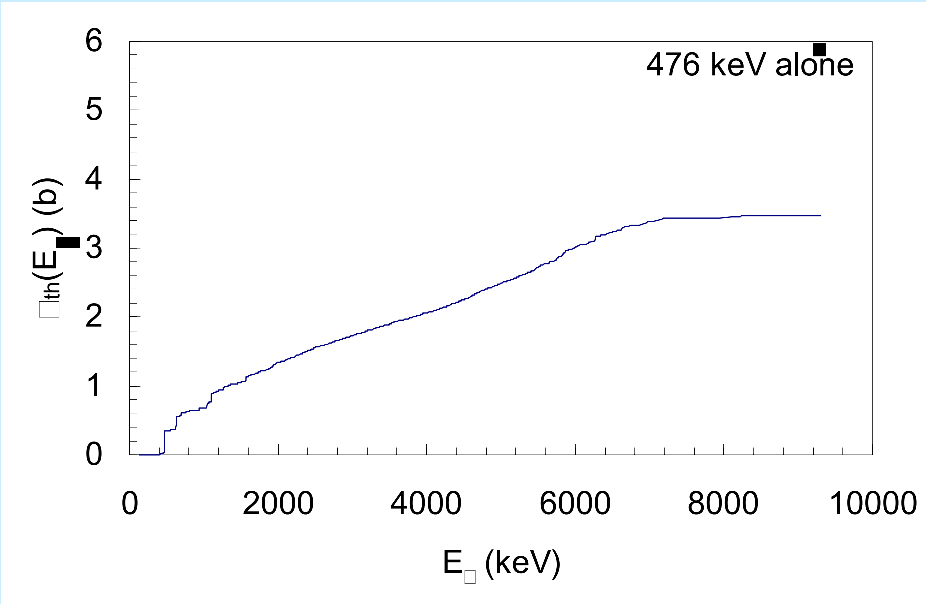
Earlier data:

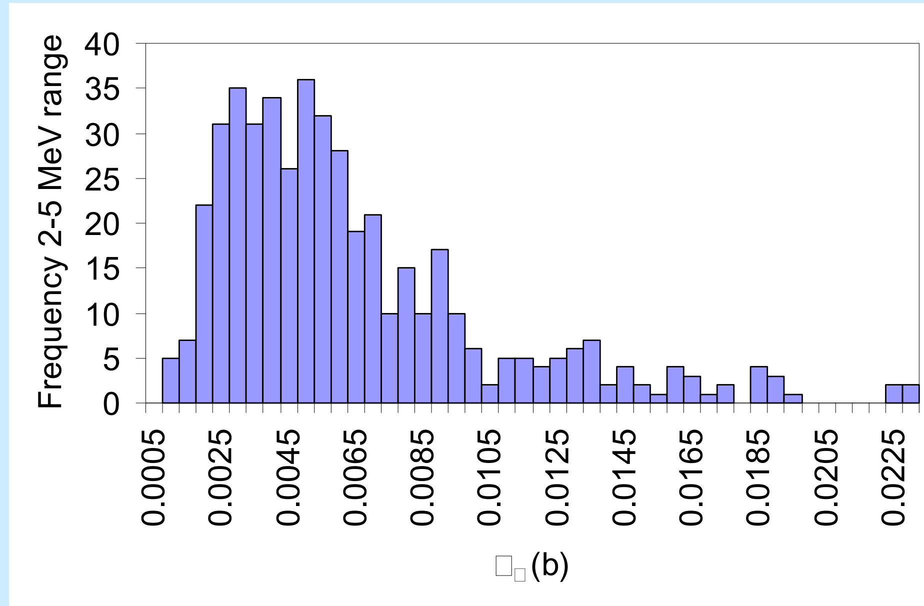
- No γ-rays in the ENSDF database
- EXFOR

σ_{th} (b)	Facility	Method	Author	year
3.4(9)	Reactor Internal McMaster	PGAA	Islam	1991
3.1(9)	Reactor Oakridge	Activation ??	Halperin	1964
5.5(1.4)	Reactor Oakridge	Mass spectrometry	Halperin	1965

The only independent or different method is the mass spectrometry

^{101}Ru(n,γ) reaction studied at our PGAA facility





Unobserved continuum is at least 40%

^{101}Ru g.s. spin $5/2^+ \rightarrow$ capture state spins 2^+ and 3^+

^{102}Ru: final state cumulative level number for spins 2,3,4,5 at 9.3 MeV $\sim 1.5 \times 10^5$

Minimum observed σ_γ in the 2-5 MeV range is 0.001 b

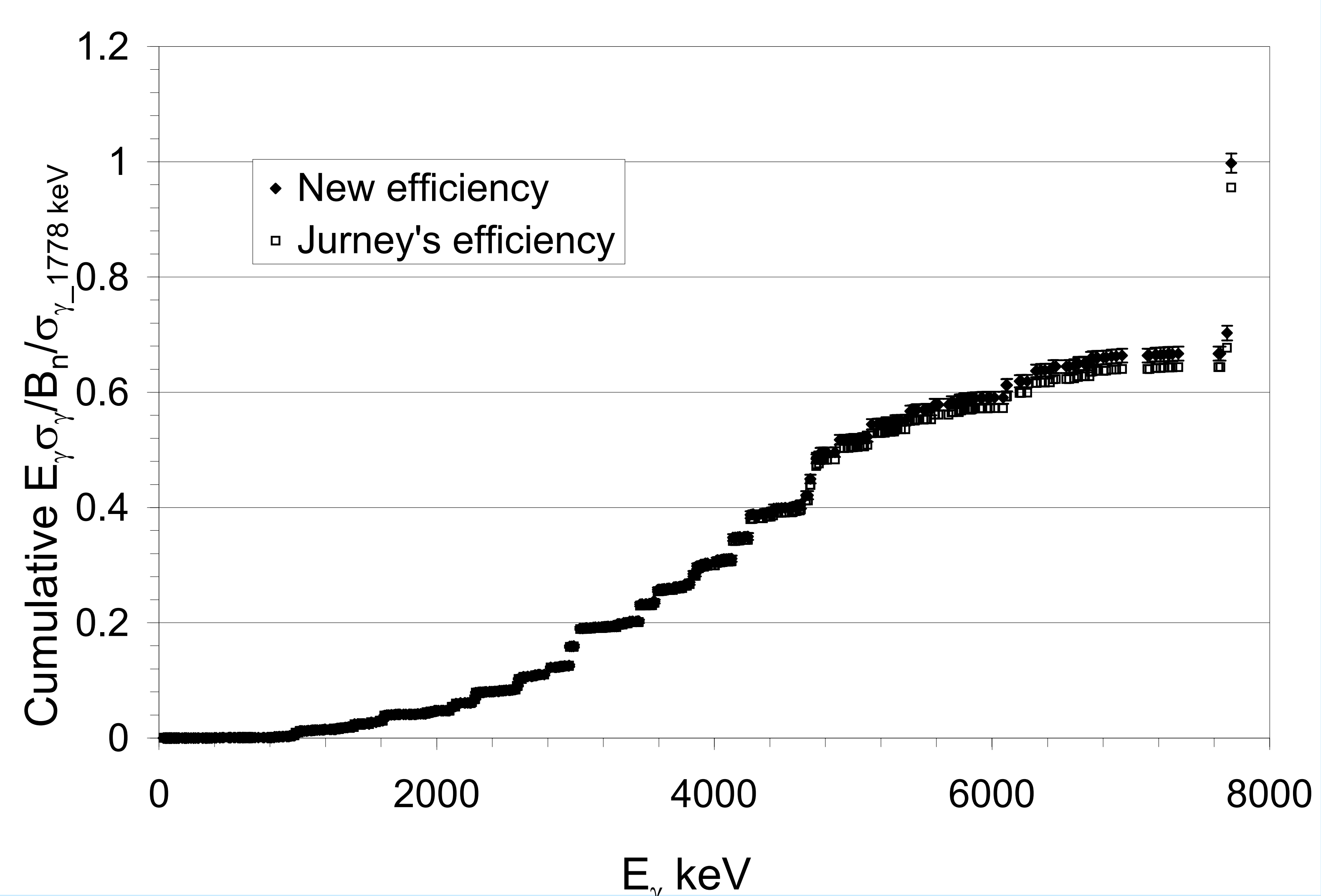
Estimate: maximum missing intensity = $1.5 \times 10^5 \times 0.001/4 = 37.5$ b ! $\rightarrow$
useless limit $\rightarrow$ We need to use better model (e.g. DICEBOX Frantisek Becvar)

Conclusions

- The PGAA-NIPS facilities provide excellent means for determination of σ_{th} and σ_γ
  - Good background conditions
  - Chopped neutron beam option
  - Firmly established methods
- Present methods can provide accurate cross sections for nuclei with moderated level density with PGAA
- Activation works for higher level density nuclei, but accuracy is limited by the accuracy of decay branching ratio
- Method outlined by Frantisek Becvar helps for nuclei with higher level density to determine capture cross sections
- Other experimental methods need to be worked out for high level density nuclei

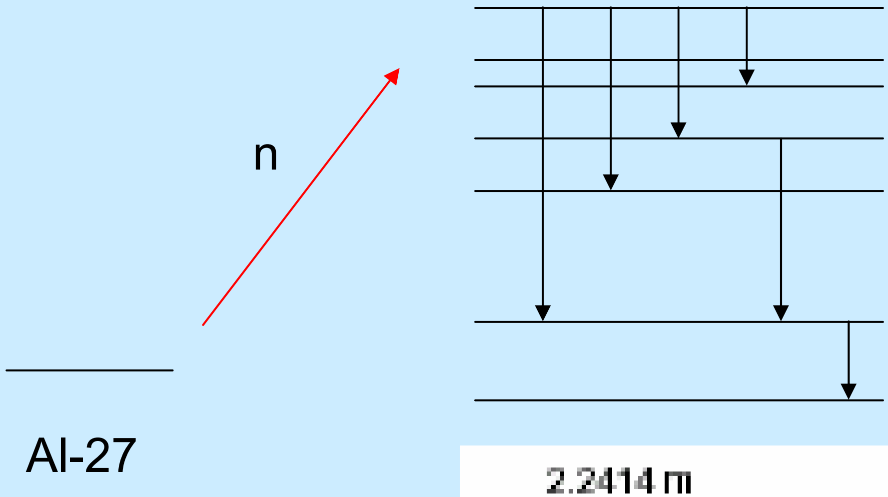
Thank you for your attention!

Test of nitrogen new intensities $^{27}Al(n,\gamma)$ reaction inverted *Q*-value



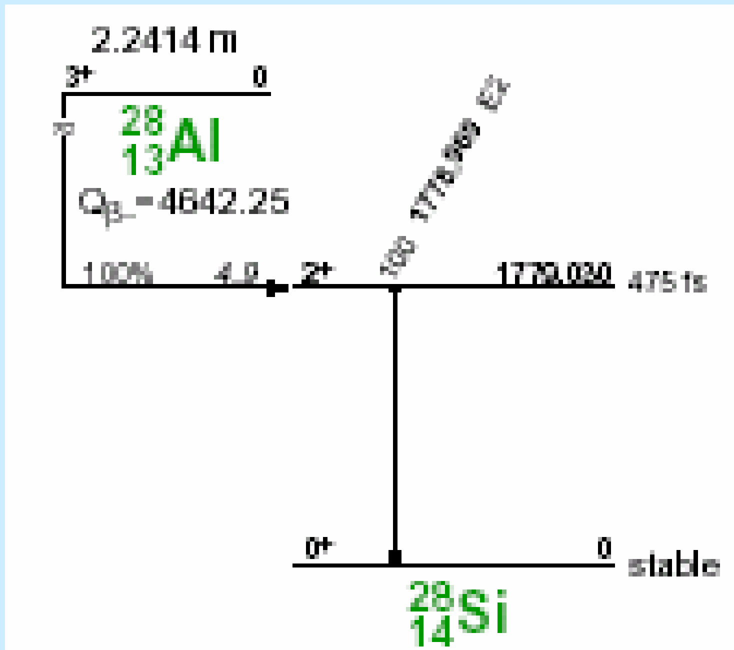
T. Belgya, Phys. Rev. C 74, 024603 (2006).

Test with $^{27}Al(n,\gamma)$



Method 4

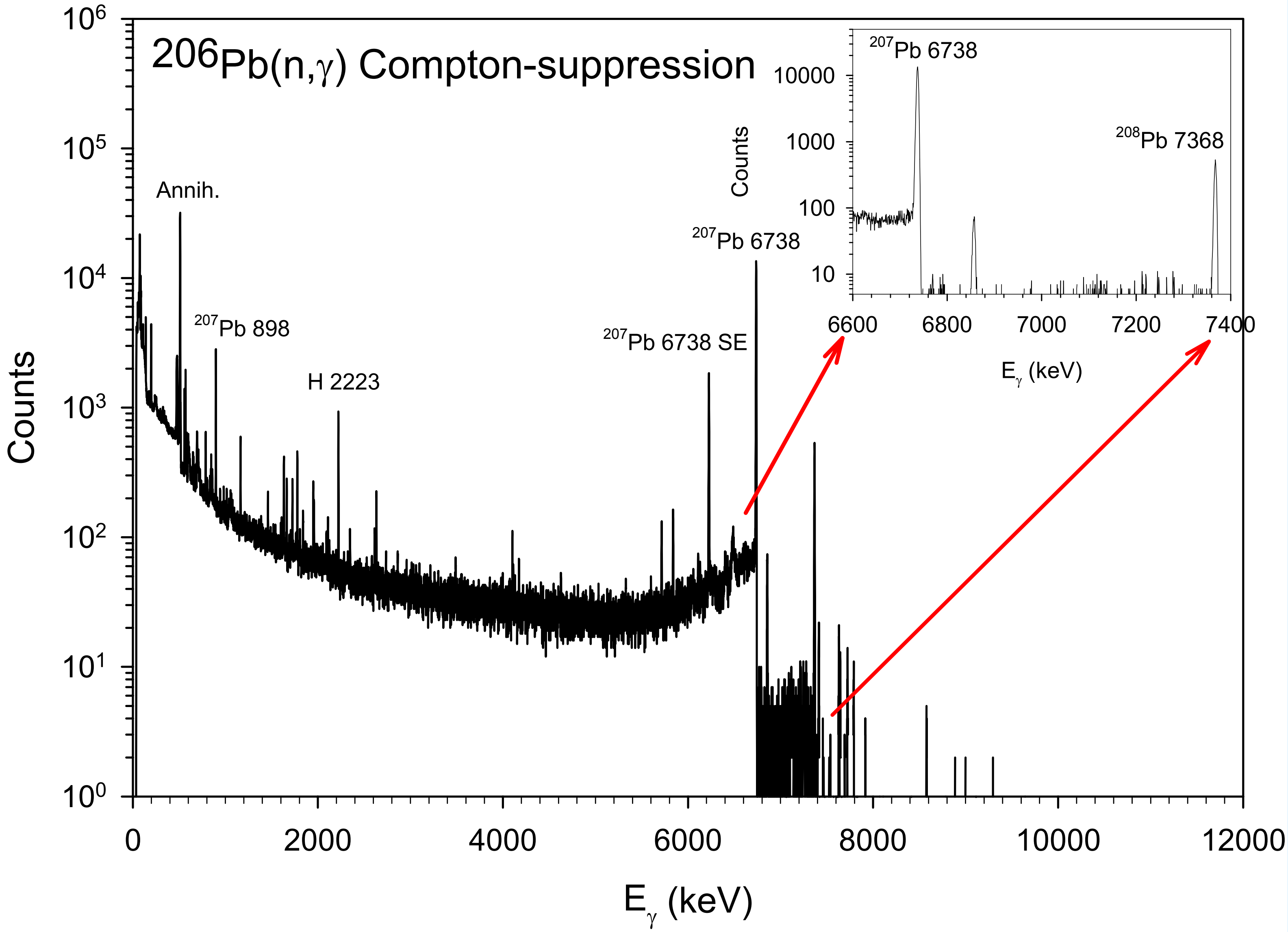
$\sigma_{th}=\Sigma\ E_i\sigma_{\gamma i}\ c_i/B_n$



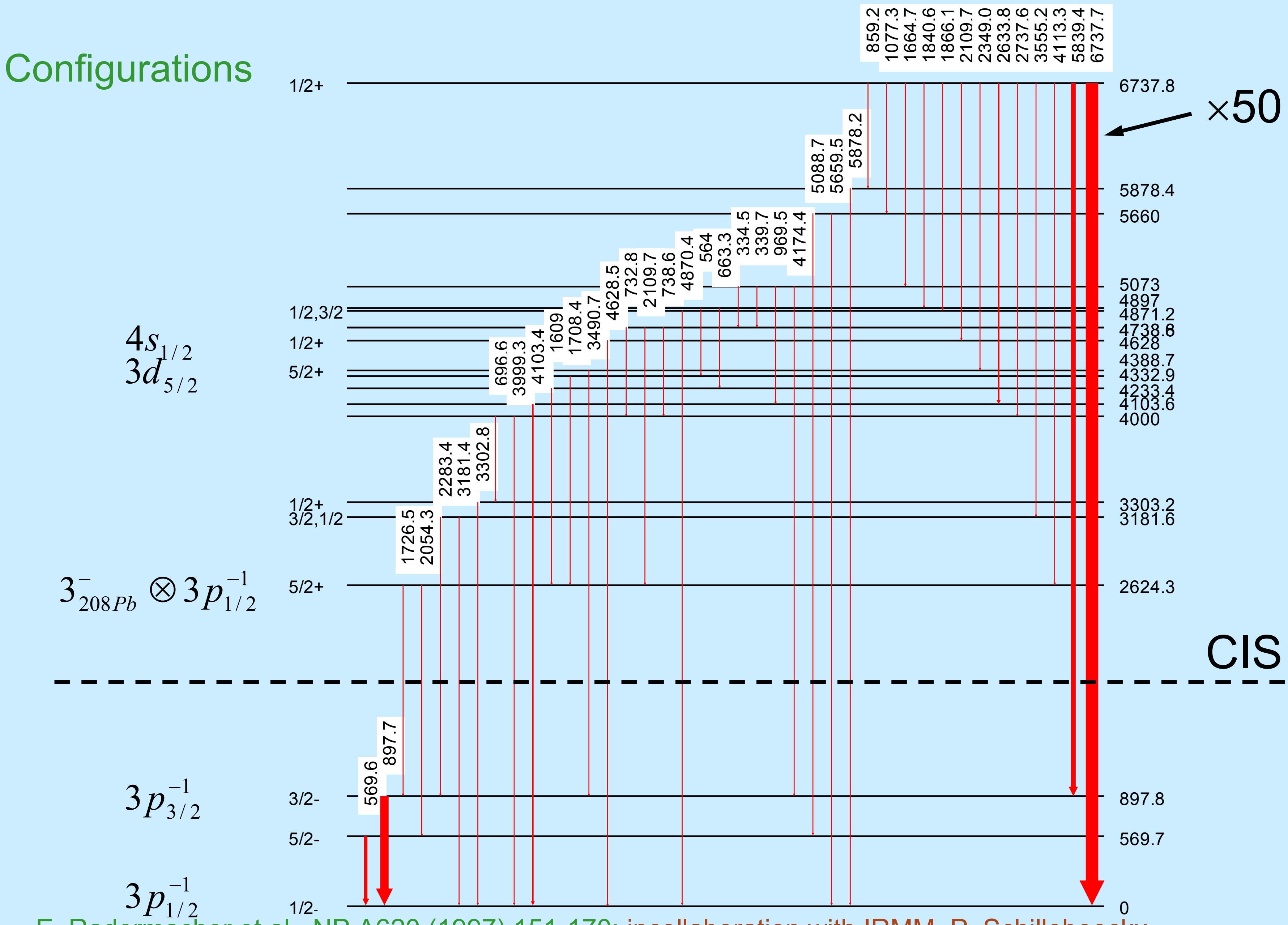
Method 1

$\sigma_{th}=\sigma_{\gamma 1779}$

^{206}Pb(n,γ) spectrum

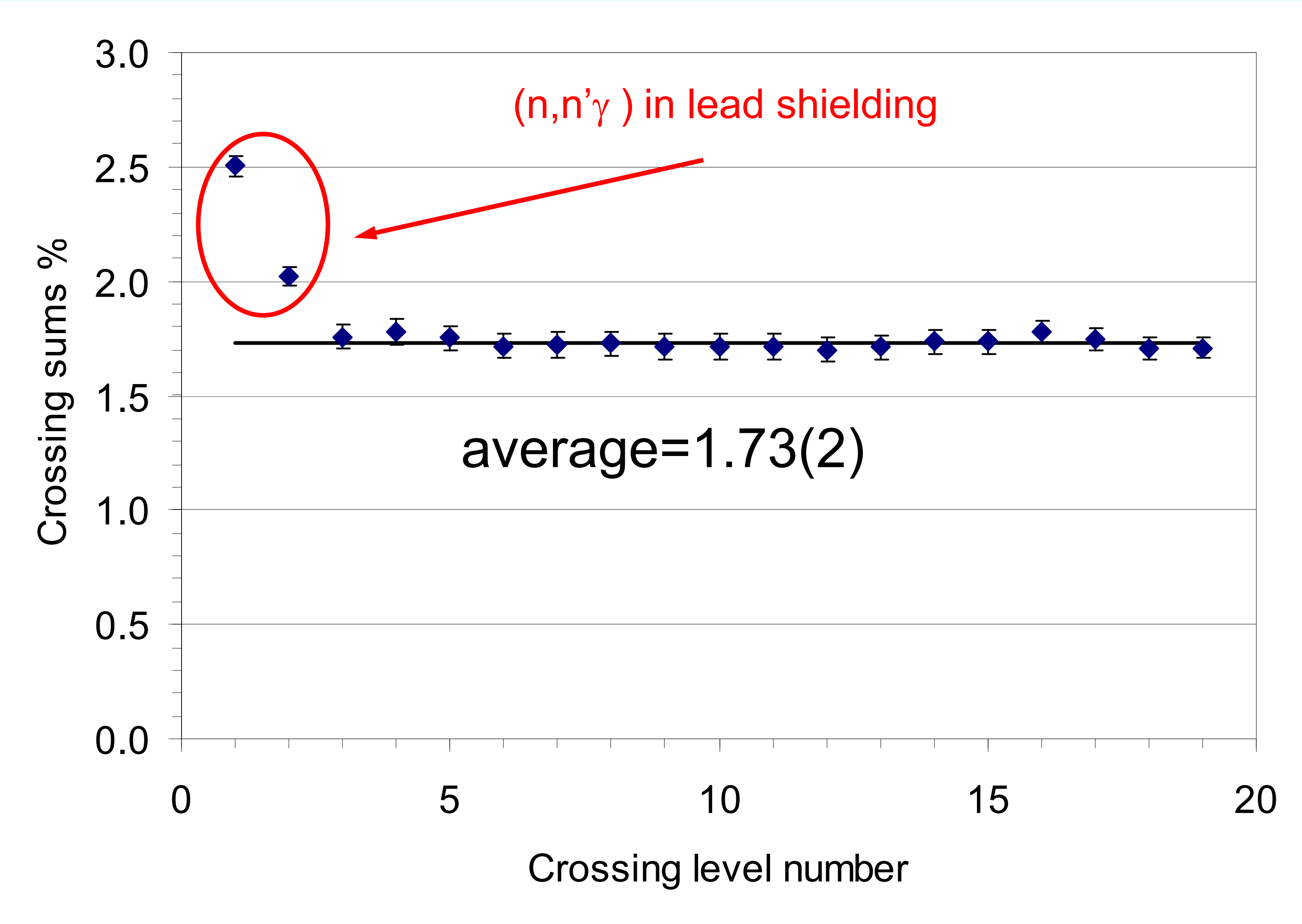


New neutron capture decay scheme of ^{207}Pb



E. Radermacher et al., NP A620 (1997) 151-170; incollaboration with IRMM, P. Schillebeeckx

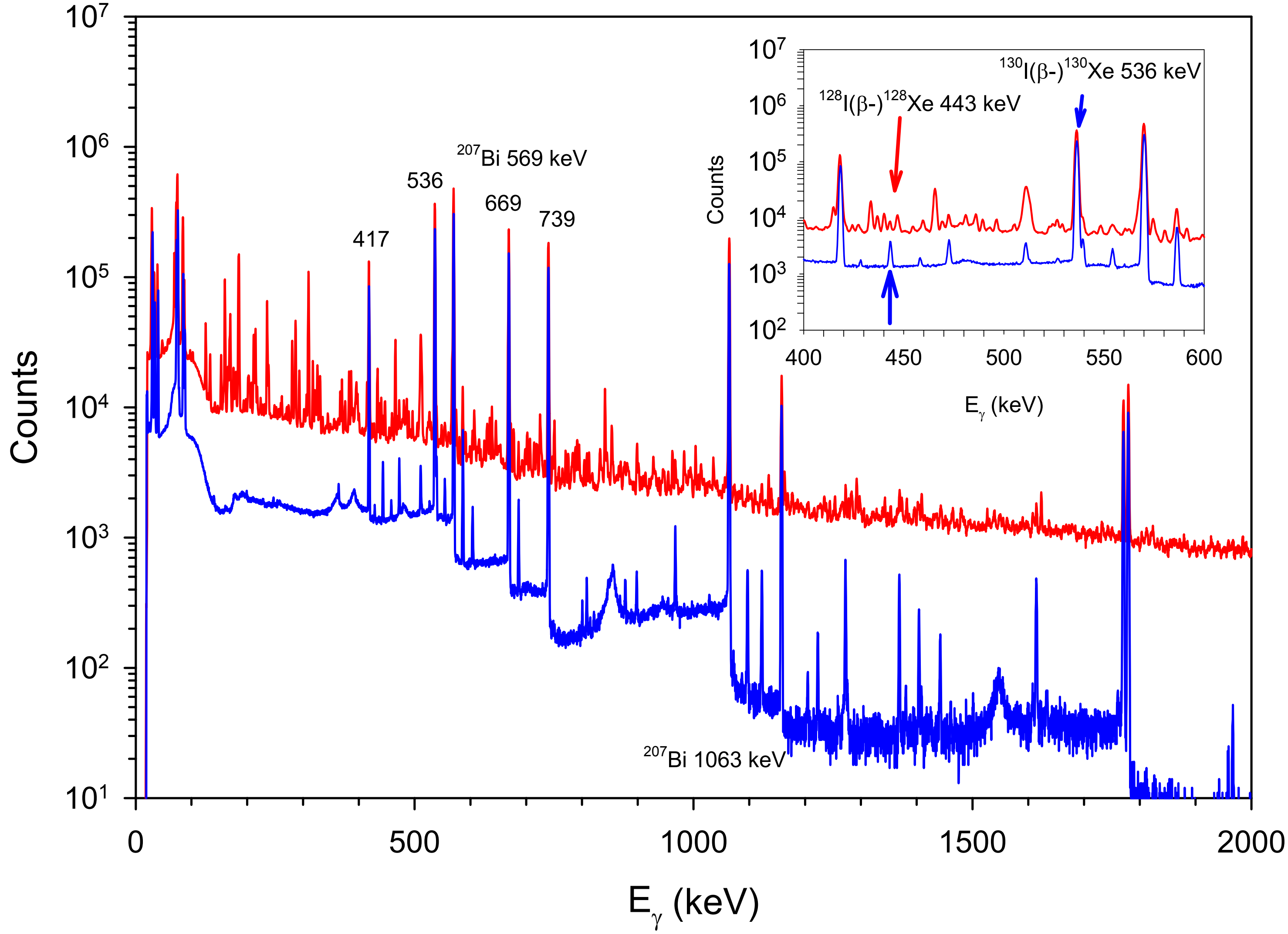
Crossing Intensity Sums for the decay scheme of ^{207}Pb



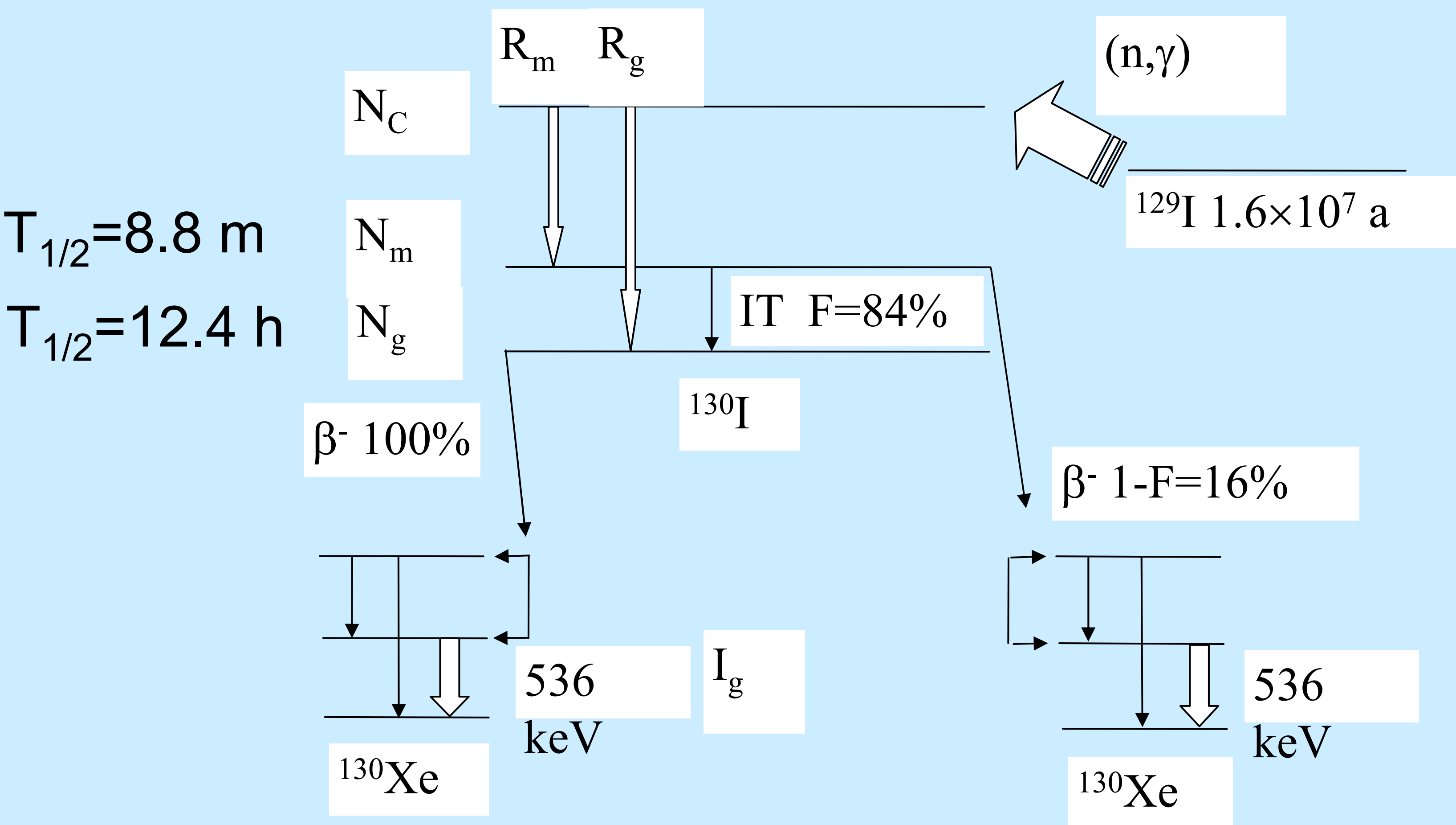
Results for $^{204,206,207}Pb$ Xsec

Isotope	This work (mb)	Mugabgab (mb)	Comment
^{204}Pb	482(20)	661(70)	preliminary
^{206}Pb	28.7(7)	26.6(12)	Increase is due to the N source
^{207}Pb	649(14)	625(30)	increase is due to the N source

^{127,129}I chopped beam (n,γ) spectra



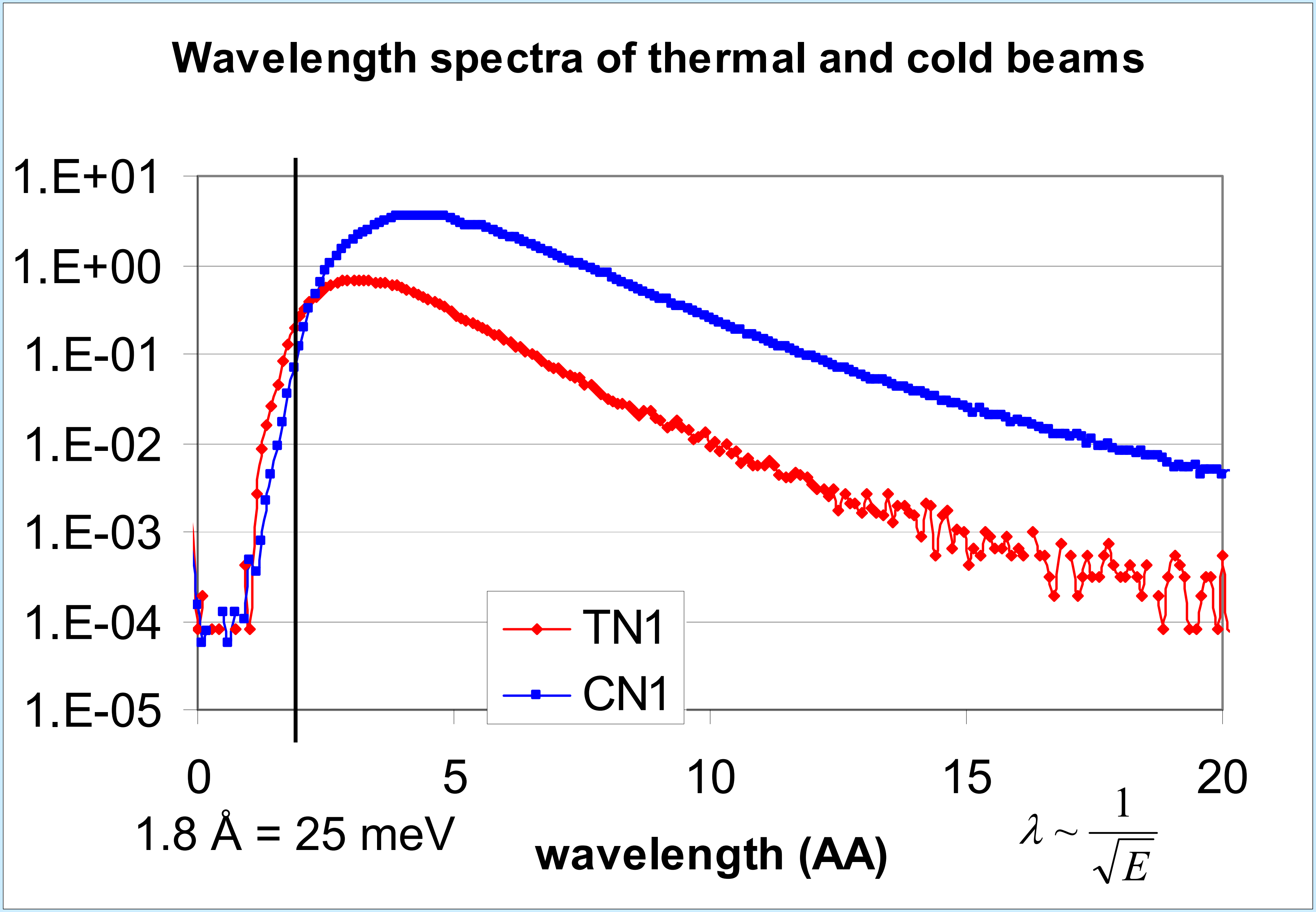
Simplified decay scheme of ^{130}I from literature



Results for ^{129}I Xsec

Year	Author	Method	σ_{th} (b)
1956	Purkayastha *et al.*	Activation reactor	35
1958	Roy *et al.*	Activation reactor	26.7(20)
1963	Pattenden *et al.*	TOF	28(2)
1969	Block *et al.*	TOF	31(4)
1983	Friedmann *et al.*	Activation reactor	33.9(19)
1996	Nakamura *et al.*	Activation reactor	30.3(12)
2007	Belgya et al.	Chopped cycl. act.	30.6(11)

Neutron energy is mostly below the first resonance



Total neutron capture cross section

another way

- Total energy detector concept, ε=E_γ or inverse Q value;

$$\sigma_{th} = \sum_i E_i \sigma_{\gamma i} (1+\alpha_i)(1+PCC_i)/B_n$$

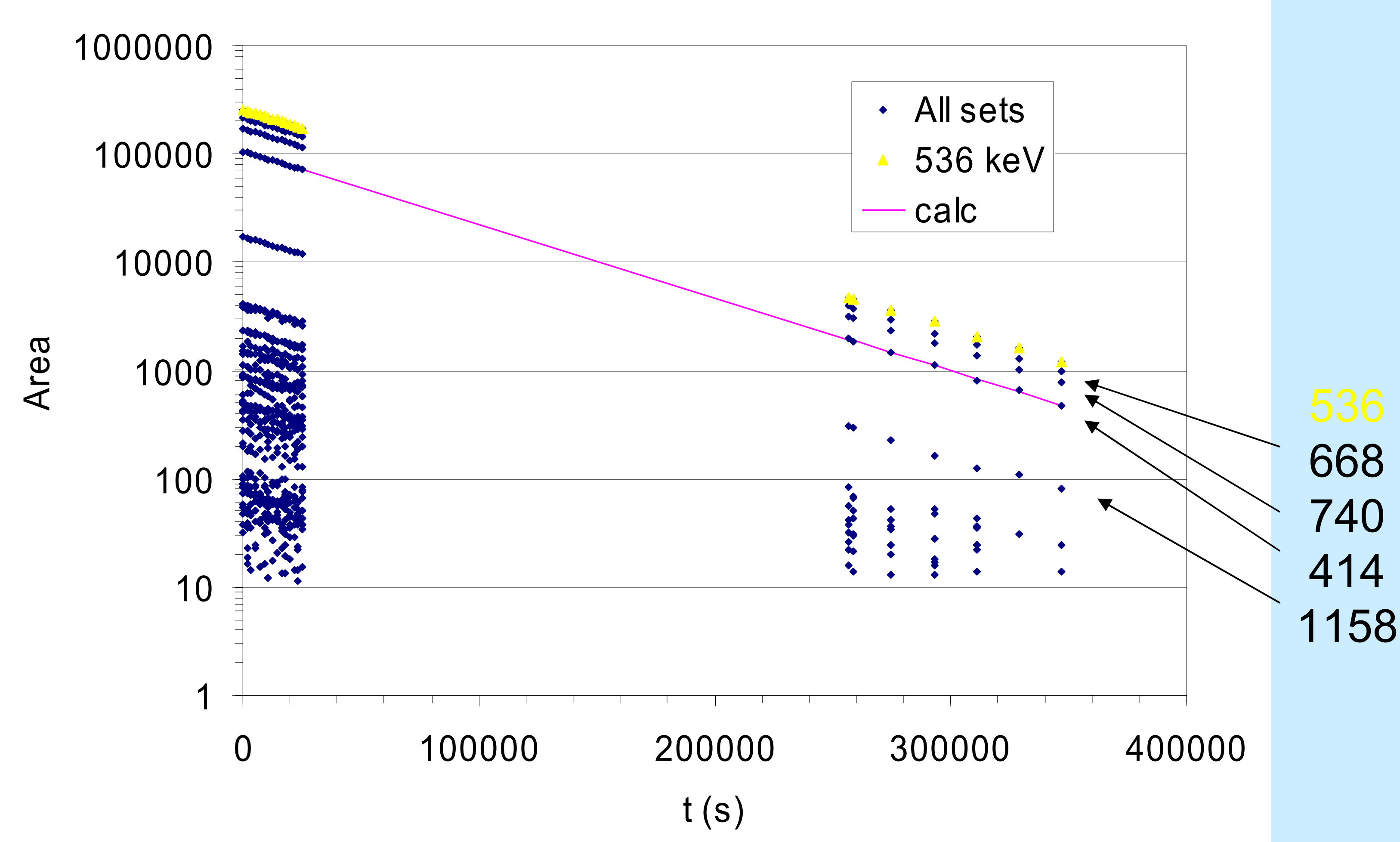
  - *i*= all transitions, (no decay scheme is needed)
  - Good identification is necessary, eg. ^{209}Bi, ^{206}Pb, ^{27}Al, etc.

Total neutron capture cross section

another way

- From decay gamma rays: $$\sigma_{th} = \frac{\sigma_\gamma}{\theta P_\gamma}$$
  - Continuous beam activation (traditional)
  - Chopped beam activation (new)
  - Absolute decay probability P_γ is needed, $^{238,235}U$, ^{232}Th, ^{129}I, ^{99}Tc, ^{27}Al, Te isotopes etc.,

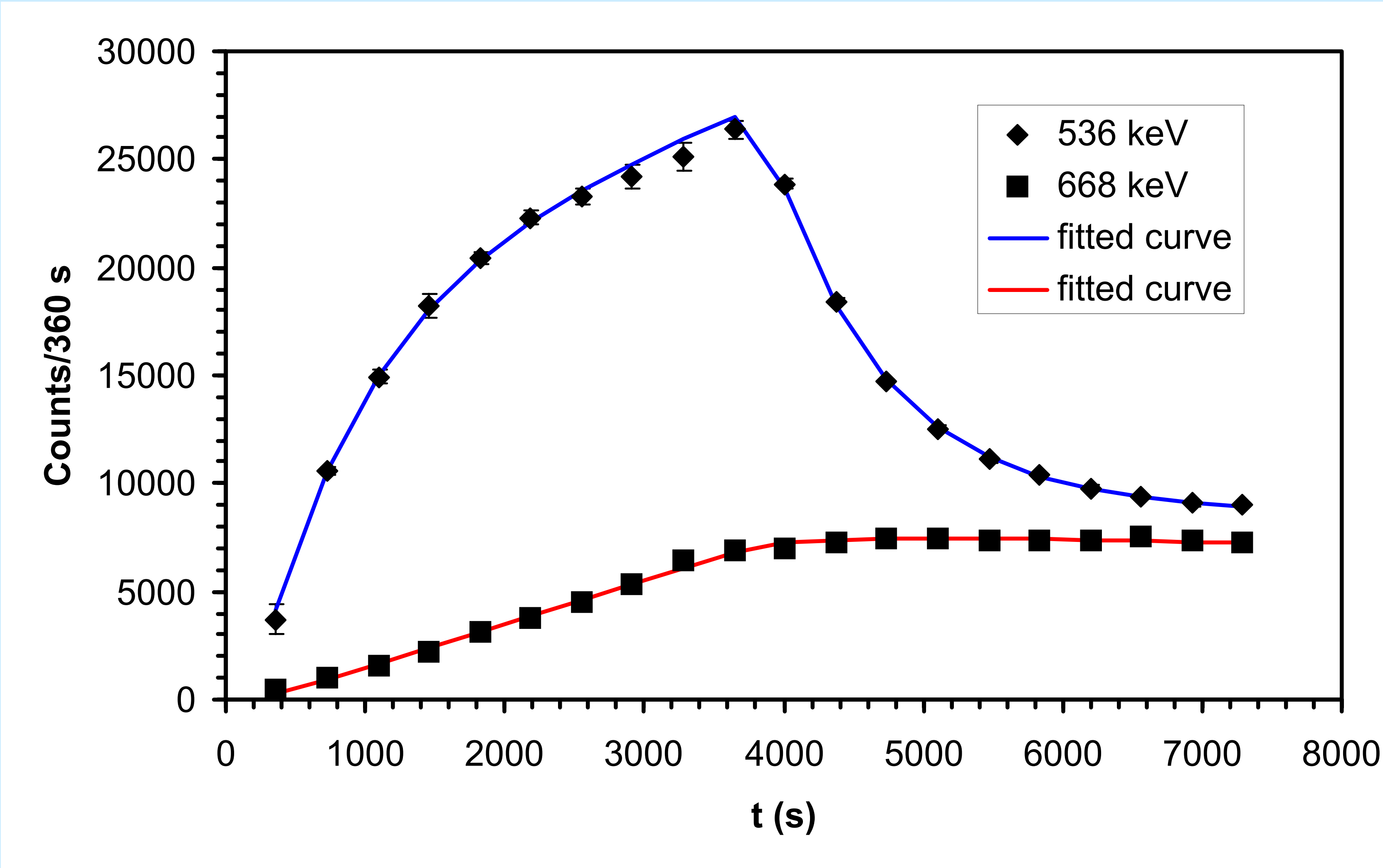
Long decay of ^{130}I ground state



E (keV)	T1/2 (h)	Uncertainty		mean	deviation	ENSDF	ucertainty
417	12.375	0.031		12.390	0.035	12.36	0.01
536	12.439	0.019					
668	12.360	0.022					
740	12.387	0.020					
1158	12.253	0.039					

Peak area of 536 and 668 keV $^{130}I(\beta-)^{130}Xe$ decay gamma rays during activation and decay

F=71%, R_m=36%, λ_m=1.279E-3, χ^2=1.25 **proposed new**



New fit results and uncertainties

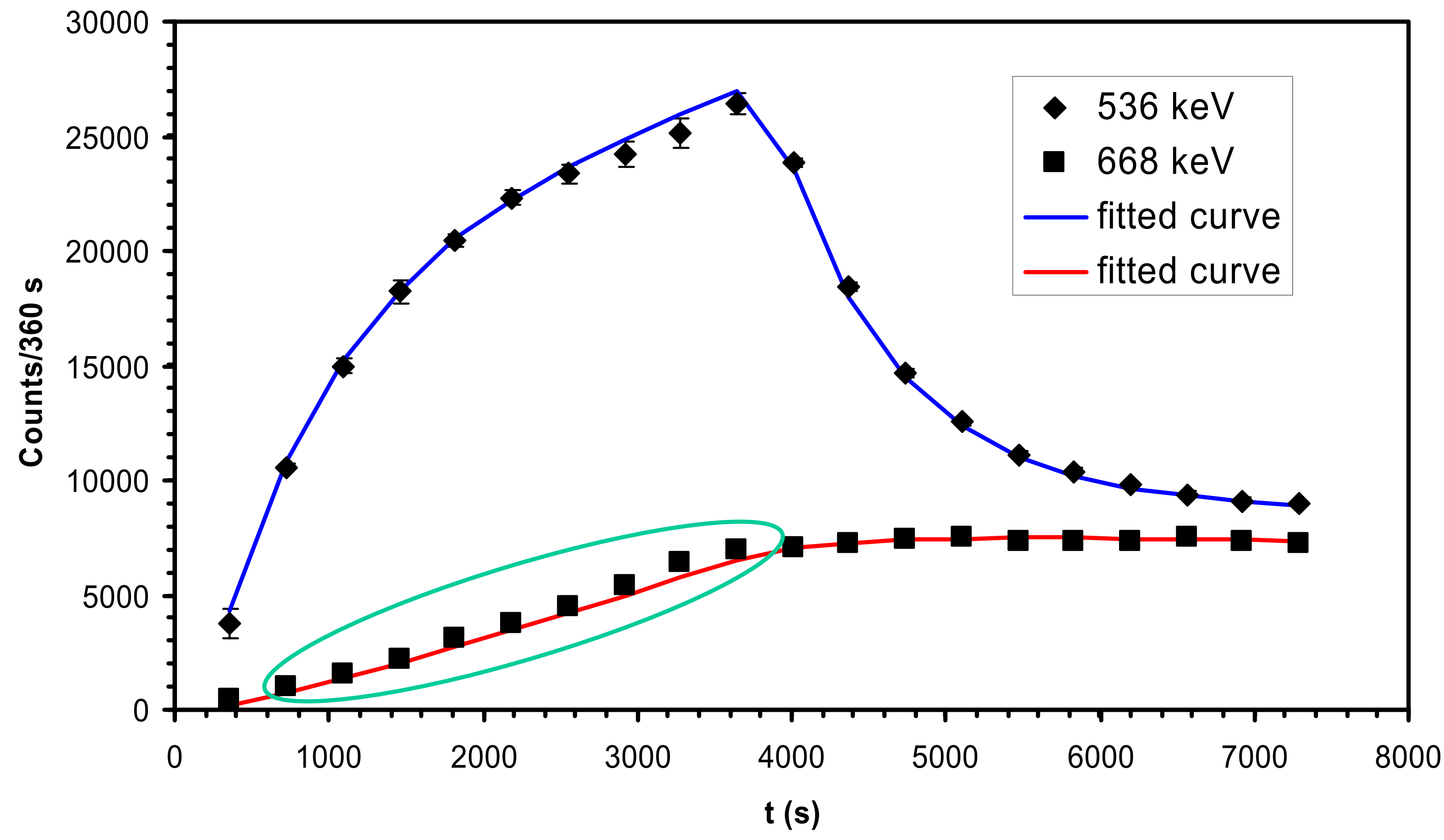
	Present work	Unc.	Rel. U.	Literature
d	516.9	8.0	1.55	-
R_m	0.355	0.008	2.14	0.60(9) a)
λ_m	1.279E-3	1.6E-05	1.27	1.316E-3(3E-6) b)
λ_g	1.558E-05	2.3E-07	1.49	fixed
F	0.709	0.009	1.26	0.83(3) a)
b_g	0.997	0.016	1.58	fixed
b_m	0.987	0.034	3.39	fixed
deadt_corr	0.0218	0.0013	6.10	-
b668g	0.832	0.012	1.39	-

a) P.K. Hopke, A.G.Jones, W.B. Walters, A. Prindle, R.A. Meyer, PRC 2 (1973) 745

b) S. Nakamura, H. Harada, T. Katoh, Z. Ogata, J. Nucl. Sci. Techn. 33 (1996) 283

Peak area of 536 and 668 keV $^{130}I(\beta\text{-})^{130}Xe$ decay gamma rays during activation and decay

F=83% (fixed), R_m=0.6, λ_m=1.344E-3 χ^2=4.5 Literature



Uncertainty budget

ISO Guide to the expression of Uncertainty in Measurement (GUM)

$$\sigma_{\gamma x} = \sigma_{\gamma c} \frac{n_c}{n_x} \frac{A_{\gamma x} / \varepsilon(E_{\gamma x}) / f(E_{\gamma x})}{A_{\gamma c} / \varepsilon(E_{\gamma c}) / f(E_{\gamma c})}$$

- Advantages of the relative internal calibration method
  - Absolute flux, inhomogeneity of sample and flux, multiple scattering (build up effect), dead time, energy distribution of flux, sample weight cancel out
- Uncertainty components for partial gamma ray cross sections
  - Uncertainty (1σ, δA) of the are *A* is obtained from peak fitting with Hypermet PC
  - Uncertainty efficiency ratio can be obtained from the correlation matrix of the efficiency fit. Relative uncertainty of the efficiency is about 0,5-1% in the 0.1-10 MeV range.
  - Gamma and neutron self absorption is calculated with numerical integration over simple shape of samples. For thin samples they are close to 1 and the estimated uncertainty is 5% of the difference from 1

A nice summary: Zs. Révay, Nucl. Instr. & Methods **A 564** (4-6), 688-697 (2006)

Uncertainty calculation example

Continuous beam

$$\sigma_{\gamma x}(1 \pm \delta\sigma_{\gamma x}) = \sigma_{\gamma c}\frac{A_{\gamma x}}{A_{\gamma c}}\frac{n_c\varepsilon(E_{\gamma c})f(E_{\gamma c})}{n_x\varepsilon(E_{\gamma x})f(E_{\gamma x})}\cdot$$

$$\left(1 \pm \sqrt{(\delta A_{\gamma x})^2 + (\delta A_{\gamma c})^2 + (\delta\varepsilon(E_{\gamma x}))^2 + (\delta\varepsilon(E_{\gamma c}))^2 - 2\delta\,\mathrm{cov}(\varepsilon(E_{\gamma x}),\varepsilon(E_{\gamma c})) + (\delta\sigma_c)^2 + \left(\delta\frac{\varepsilon(E_{\gamma x})}{\varepsilon(E_{\gamma c})}\right)^2}\right)$$

- Without correlation we get overestimate of the uncertainty
- For sum of partial cross sections the uncertainty calculus must be used, if we use simple re-normalization than the correlation is neglected
- For calculus of decay partial gamma-ray cross sections and uncertainties related to chopper methodology please see

Szentmiklósi, L., Z. Révay and T. Belgya (2006). *Measurement of partial gamma-ray production cross-sections and k_0 factors for radionuclides with chopped-beam PGAA*, Nucl. Instr. and Methods A **564**: 655-661.

Selected recent publications

Handbook of PGAA with neutron beams (Eds. G.L. Molnár, Kluwer Academic Publisher), 2004

^{99}Tc(n,γ):

G.L. Molnár et al., Radiochim. Acta **90** (2002) 479-482, T. Belgya et al.,Porc. of the enlargement workshop on Neutron Measurements and Evaluations for Applications (Eds. A.J.M. Plompen), 5-8 Nov. 2003, Budapest, Hungary, EUR Report 21100 EN, Luxembourg, ISBN 92-894-6041-5, 2004, 2004, pp. 159-163.

^{127,129}I(n, γ):

Belgya, T., G. L. Molnár, Z. Révay and J. Weil (2005). *Determination of thermal neutron capture cross sections using cold neutron beams*, 10th International Conference on Nuclear Data for Science and Technology, September 26 - October 1, 2004, Santa Fe, New Mexico, AIP 769, pp. 744-747

^{238}U(n, γ):

G.L. Molnár, Zs. Révay and T. Belgya, Nucl. Instr. Methods B 213, 389 (2004)

^{209}Bi(n, γ)

Borella, A., A. Moens, P. Schillebeeckx, R. Van Bijlen, G. L. Molnár, et al. (2005). *Determination of the 209Bi(n,g) capture cross section at a cold neutron beam*, Journal of Radioanalytical and Nuclear Chemistry **265**(2): 267-271.

Te isotopes:

I. Tomandl et al., Phys. Rev. C **68** (2003) 067602

Pb in progress

^{15}N, ^{208}Pb, ^{27}Al:

Belgya, T. (2006). *Improved accuracy of gamma-ray intensities from basic principles for the calibration reaction 14N(n,g)15N*, Physical Review C **74**: 024603; Belgya, T. (2007). *New gamma-ray intensities for the 14N(n,g)15N high energy standard and its influence on PGAA and on nuclear quantities*, Journal of Radioanalytical and Nuclear Chemistry: accepted

Pd:

Firestone, R. B., M. Kritcka, D. P. McNabb, B. W. Sleaford, U. Agvaanluvsan, et al. (2005). *Thermal neutron capture cross section of the palladium isotopes*, 12nd international Conference on Capture Gamma-Ray Spectroscopy and Related Topics, September 4-9, 2005 University of Notre Dame, Indiana, USA, API, pp.

Absolute FEP efficiency

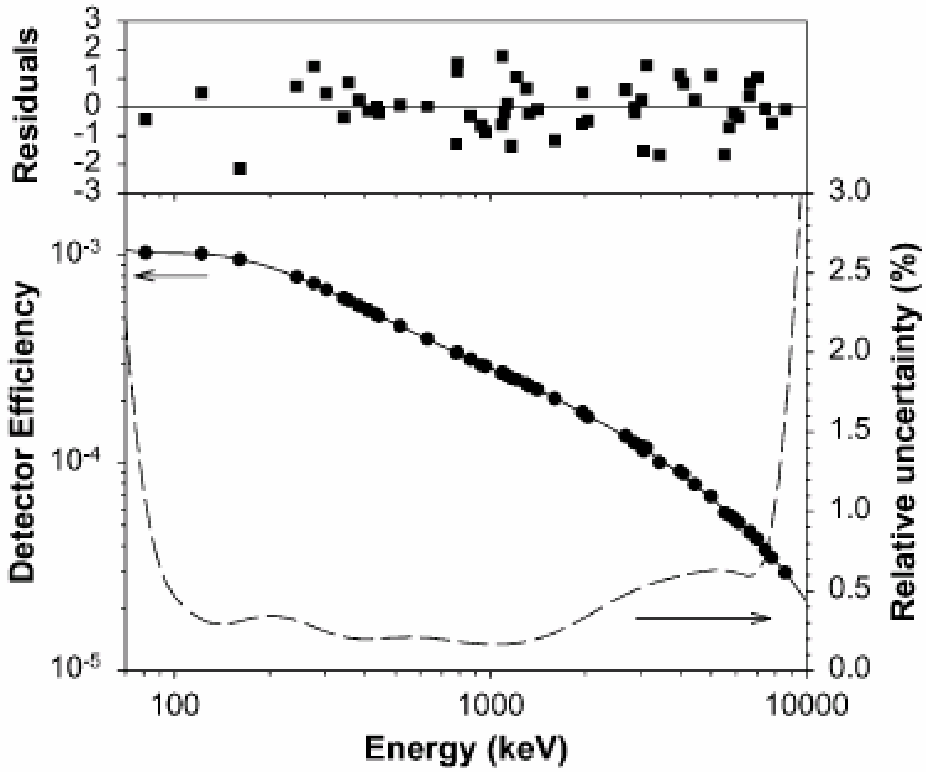
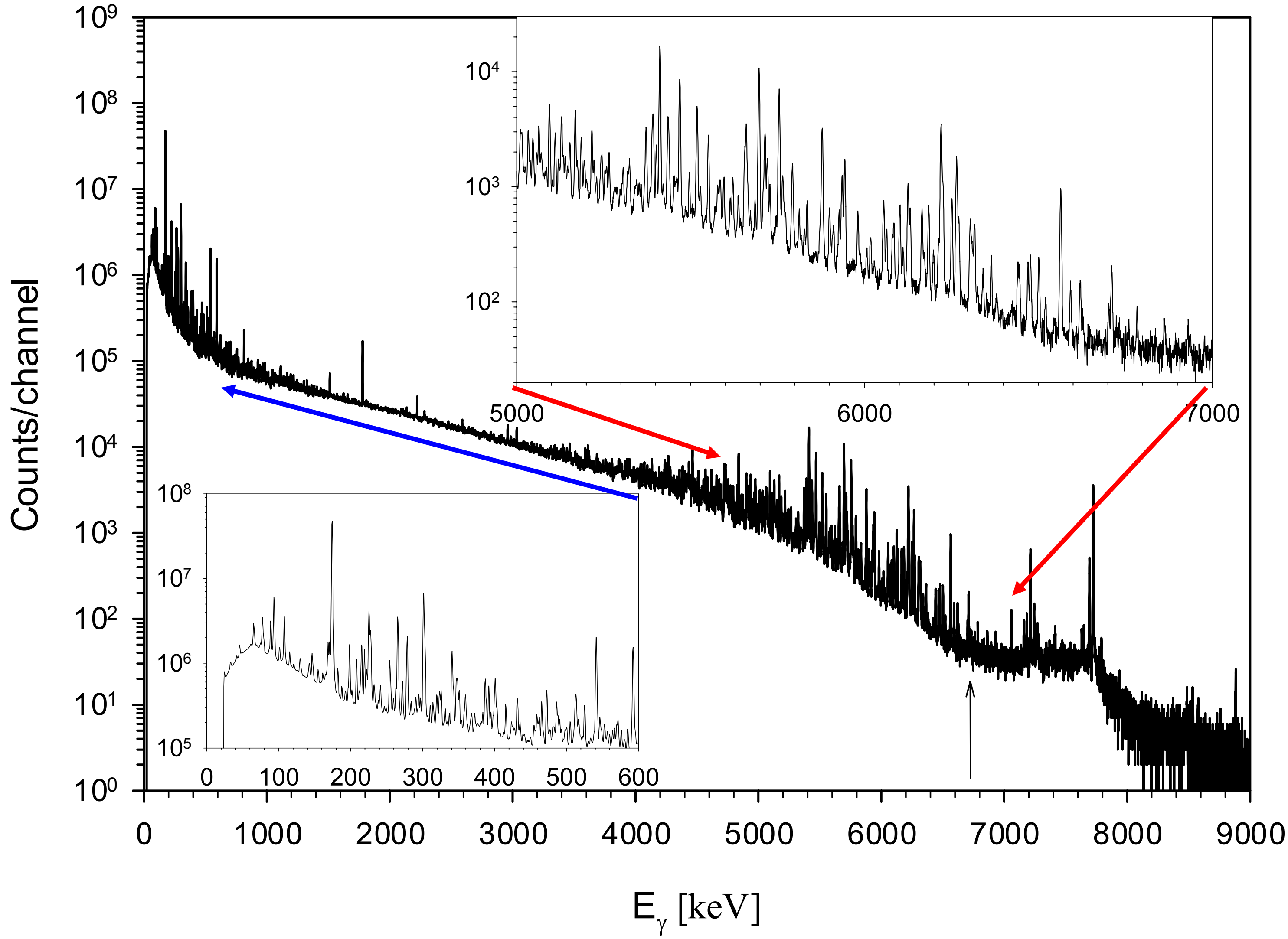


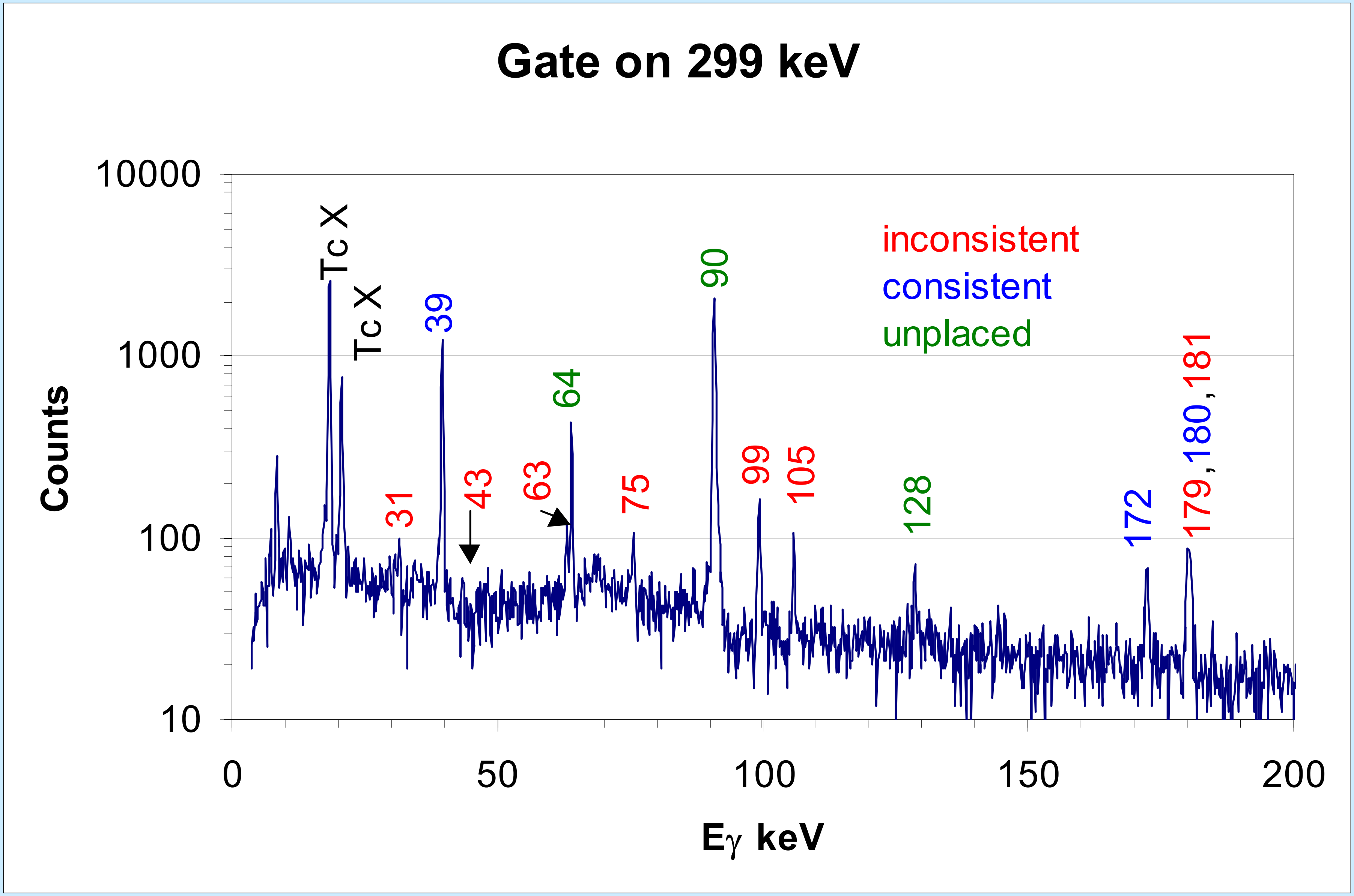
Fig. 4. Absolute full-energy peak efficiency (left-hand scale) as a function of γ-ray energy for the Budapest detector. The residuals of the fit to the data points (experimental value minus fitted value divided by the experimental uncertainty) are shown at the top of the figure. The dashed curve represents the relative standard uncertainties for the fitted curve (in percent, right-hand scale).

2002_NIMA_481_365-377_Baglin_66Ga

^{99}Tc 0.5 g sample (n,γ) spectrum



^{99}Tc (n,γ γ) prompt-coincidence spectrum



Partial γ-ray production cross sections of capture and decay lines for a ^{99}Tc target

E_γ (keV)	Origin	P_γ (γ/100 captures or decays)	σ_γ [a] (b)	Sensitivity (cps/mg)
172.1	^{99}Tc(n,γ)	67±6	16.61±0.15	3.0
223.4	^{99}Tc(n,γ)	6.1±0.6	1.472±0.013	0.24
263.5	^{99}Tc(n,γ)	5.9±0.5	1.425±0.012	0.21
539.5	^{100}Tc β^-	6.6±0.5 [b]	1.604±0.014	0.14
590.7	^{100}Tc β^-	5.3±0.5	1.296±0.011	0.10
89.5	^{99}Tc β^-	$(6.5\pm1.5)\times10^{-4}$ [c]		4.3×10^{-3}

G.L. Molnár, T. Belgya, Zs. Révay and S.M.Qaim, Radiochim. Acta **90**, 479-482 (2002)

Inferred total thermal-neutron capture cross section of ^{99}Tc

Method	Basis	σ (b)	Comment
^{100}Tc(β^-)^{100}Ru	539 γ	24.7±2.3	with Pγ Furutaka et al.
	591 γ	23.9 ±1.8	
	Average	*24.3 ±2.2*	*unweighted average*
^{99}Tc(n,γ)^{100}Tc	Σ σγ g.s.	21.21±0.17	lower limit

Literature: EXFOR database

H. Pomerance 1975	19±2 b	pile oscillator
R.B. Tattersall 1960	16 ±7 b	pile oscillator
N.J. Pattenden 1958	25 ±2 b	transmission
M. Lucas 1977	20 ±2 b	mass spectrometer
V.V. Ovechkin 1973	24 ±4 b	activation
H. Harada 1995	22.9 ±2.6 b	activation
Mughabgab 2003	20 ±1	evaluation INDC(NDS)-440

Parameters of the NIPS station

• Neutron beam cross section:	2.5×2.5 cm^2
• Thermal-equivalent flux at target:	$\approx 3 \times 10^7 \cdot cm^{-2}s^{-1}$
• Vacuum in target chamber (optional):	$\approx$1 mbar
• Form of target at room temperature:	Solid, powder, liquid, gas in pressure container
• Largest target dimensions:	$1.5 \times 1.5 \times 3.5$ cm^3
• γ-ray detector No.1	n-type coax. HPGe
• Relative efficiency:	13% at 1332 keV
• FWHM:	1.8 keV at 1332 keV
• γ-ray detector No 2.	n-type coax. HPGe
• Relative efficiency:	30% at 1332 keV
• FWHM:	1.9 keV at 1332 keV
• γ-ray detector No 3.	Planar HPGe
• FWHM:	0.6 keV at 122 keV

Parameters of the PGAA station

• beam cross section:	$\leq 2\times2$ cm^2
• Thermal-equivalent flux at target:	$\approx 5\times10^7$ cm^{-2}s^{-1}
• Vacuum in target chamber (optional):	≈1 mbar
• Form of target at room temperature:	Solid, powder, liquid, gas in pressure container
• Largest target dimensions:	$4\times4\times10$ cm^3
• γ-ray detector	n-type coax. HPGe, with BGO shield
• Distance from target to detector:	23.5 cm
• Relative efficiency:	25% at 1332 keV
• FWHM:	1.8 keV at 1332 keV
• Compton suppression enhancement:	≈5 (1332 keV) to ≈40 (7000 keV)

What's NIPS?

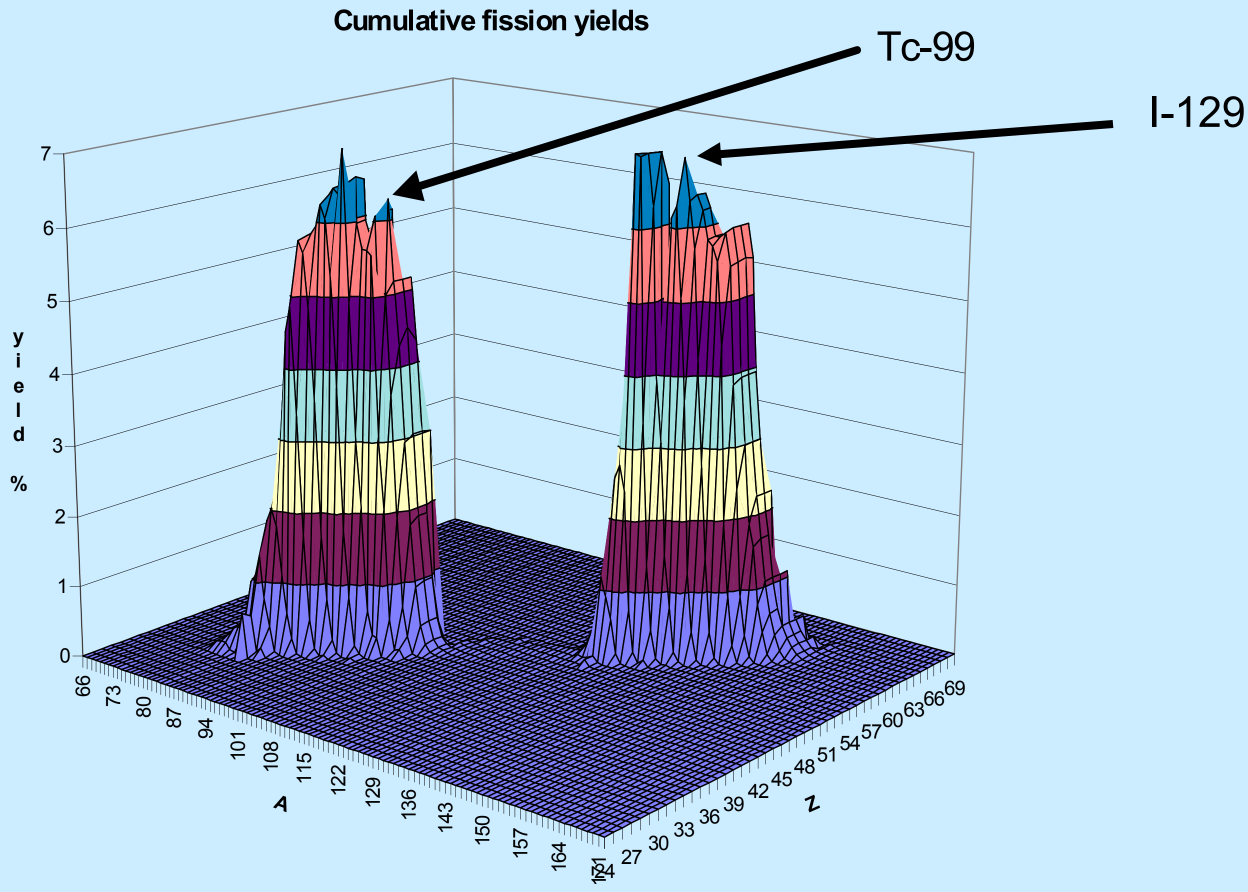
- Neutron Induced Prompt gamma-ray Spectroscopy
- Intent: To build a multipurpose experimental station
  - Close detector geometry (2.5 cm)
  - Place for more detectors (≥3)
  - Good shielding (^{6}Li-poly)
  - Multiparameter data acquisition

Publications

- P.P. Ember, T. Belgya, G.L. Molnár, *Improvement of capabilities of PGAA by coincidence techniques*, Appl. Radiat. Isot. 56 (2002) 535
- P.P. Ember, T. Belgya, J.L. Weil, G.L. Molnár, *Coincidence measurement setup for PGAA and nuclear sructure studies*, Appl. Radiat. Isot. (2002) In print
- T. Belgya, Zs. Révay, L. Szentmiklósi, M. Lakatos, J.L. Weil, *The application of a digital specrometer in PGAA*, IRRMA-V (2002)
- T. Belgya, G.L. Molnár, *Accurate relative gamma-ray intensities from neutron capture on natural chromium*, IRRMA-V (2002)
- G.L. Molnár, T. Belgya, Zs. Révay, S.M. Qaim, *Partial and total neutron capture cross section for non-destructive assay and transmutation monitoring of* ^{99}Tc, Radiochemia Acta, submitted

The ^{99}Tc

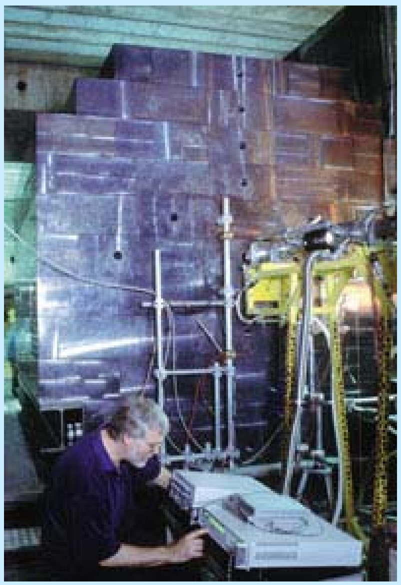
- One of the most important LLFF
- ^{99}Tc half-life: 210 000 years
- cumulative fission yield in reactor: 6.1%
- The (n,γ) reaction can efficiently destroy the Tc waste



^{99}Tc transmutation

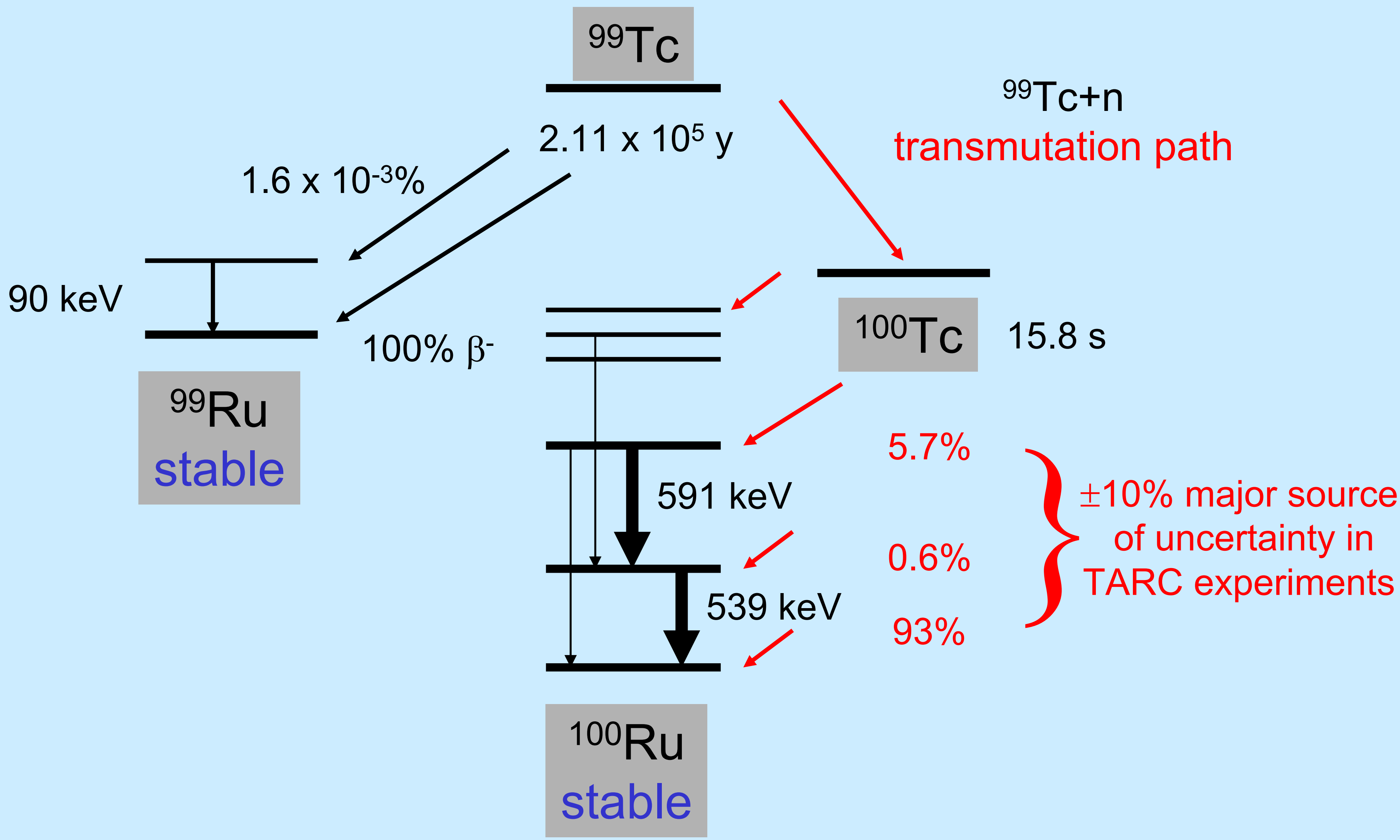
- TARC experiments at CERN to measure transmutation rates
- A. Abanades et al., Nucl. Instr. and Methods A 478 (2002) 577–730

CERN COURIER



Carlo Rubbia's TARC (Transmutation by Adiabatic Resonance Crossing) experiment at CERN. Accelerator-driven transmutation has emerged as a potentially complementary technology for radioactive waste handling by transmuting the longest-lived radioactive isotopes into short-lived or stable ones.

Scheme of transmuting ^{99}Tc



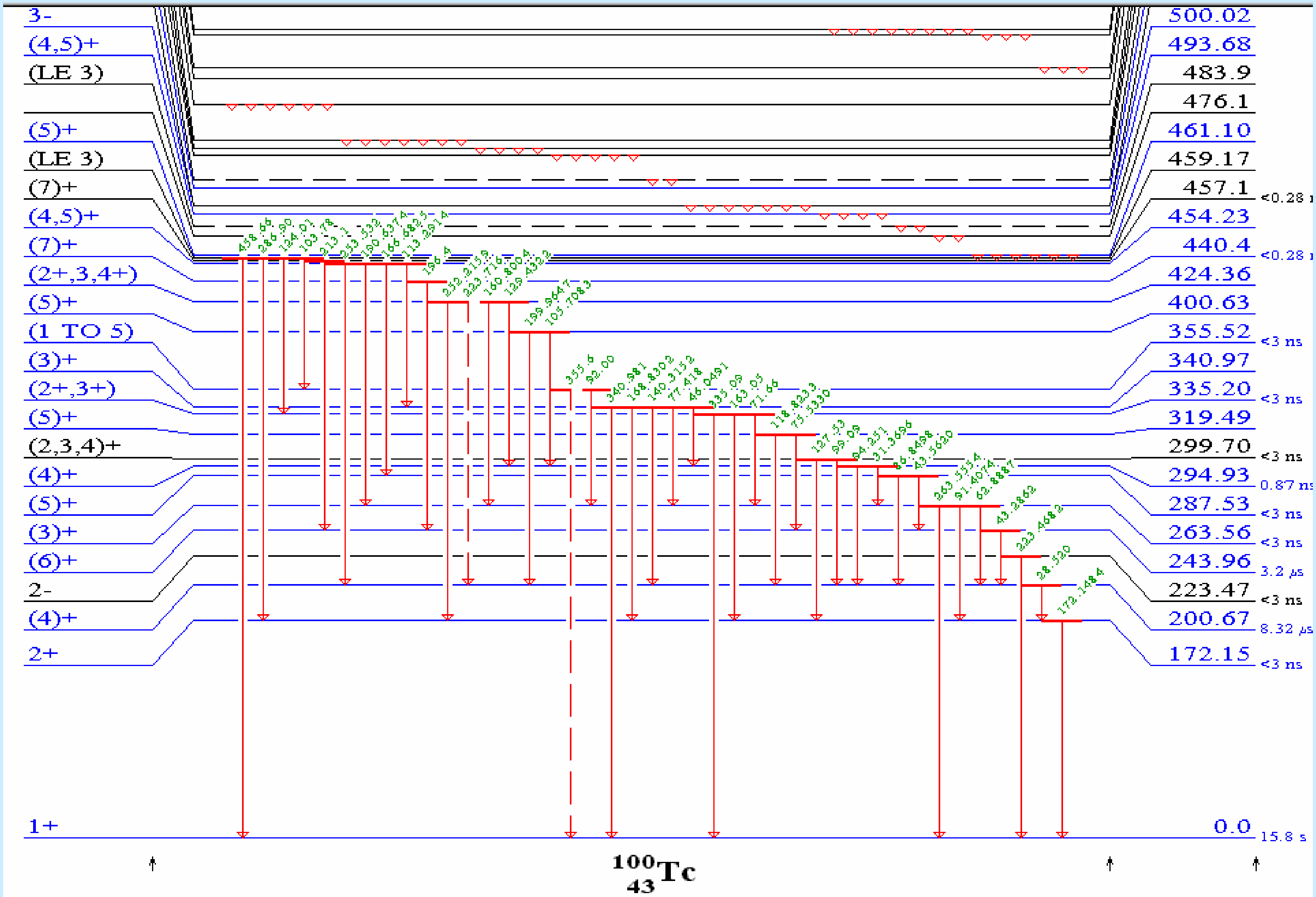
^{99}Tc measurements at our PGAA & NIPS facilities

✓NH_4TcO_4	PGAA (partial cross section)
✓^{99}Tc(n,γ)	PGAA (rel. γ intensities)
✓^{99}Tc(n,γ)	Chopped beam PGAA
	(^{100}Tc β^- decay rel. γ intensities)

Evaluation is in progress:

• ^{99}Tc(n,γ γ)	Coincidence (decay scheme)
• ^{99}Tc(d,p)^{100}Tc	High resolution proton spectrum
	(level scheme)
	measured at , TU Munich

^{100}Tc levels observed in (d,p) spectrum



Quantities we can measure and their role in nuclear waste transmutation

- Partial gamma-ray cross sections

  on-line transmutation yield measurement

  non-destructive assay of waste by PGAA

- Decay gamma-ray partial cross sections

  on- and off-line transmutation yield measurement

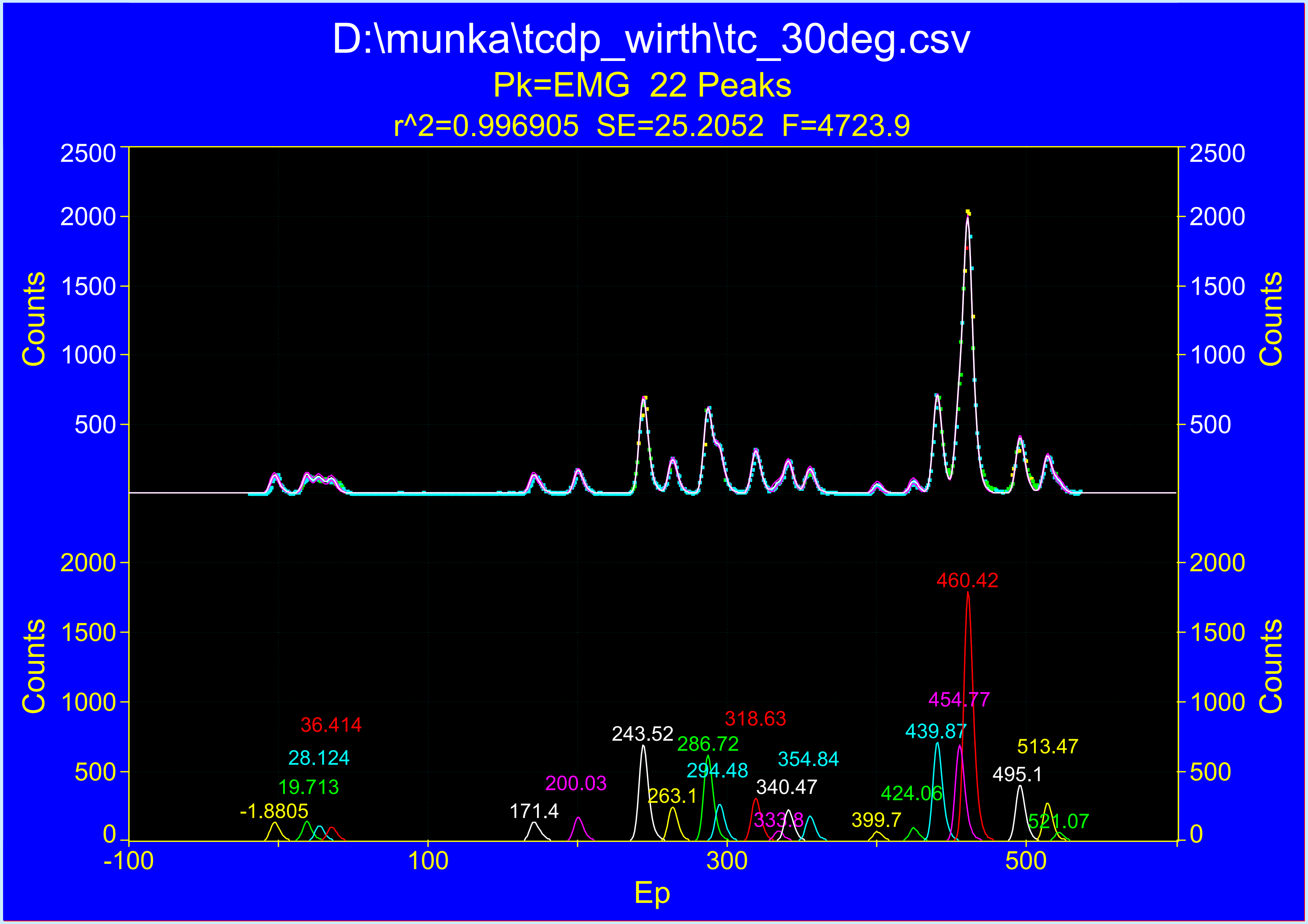
- Thermal neutron capture cross sections

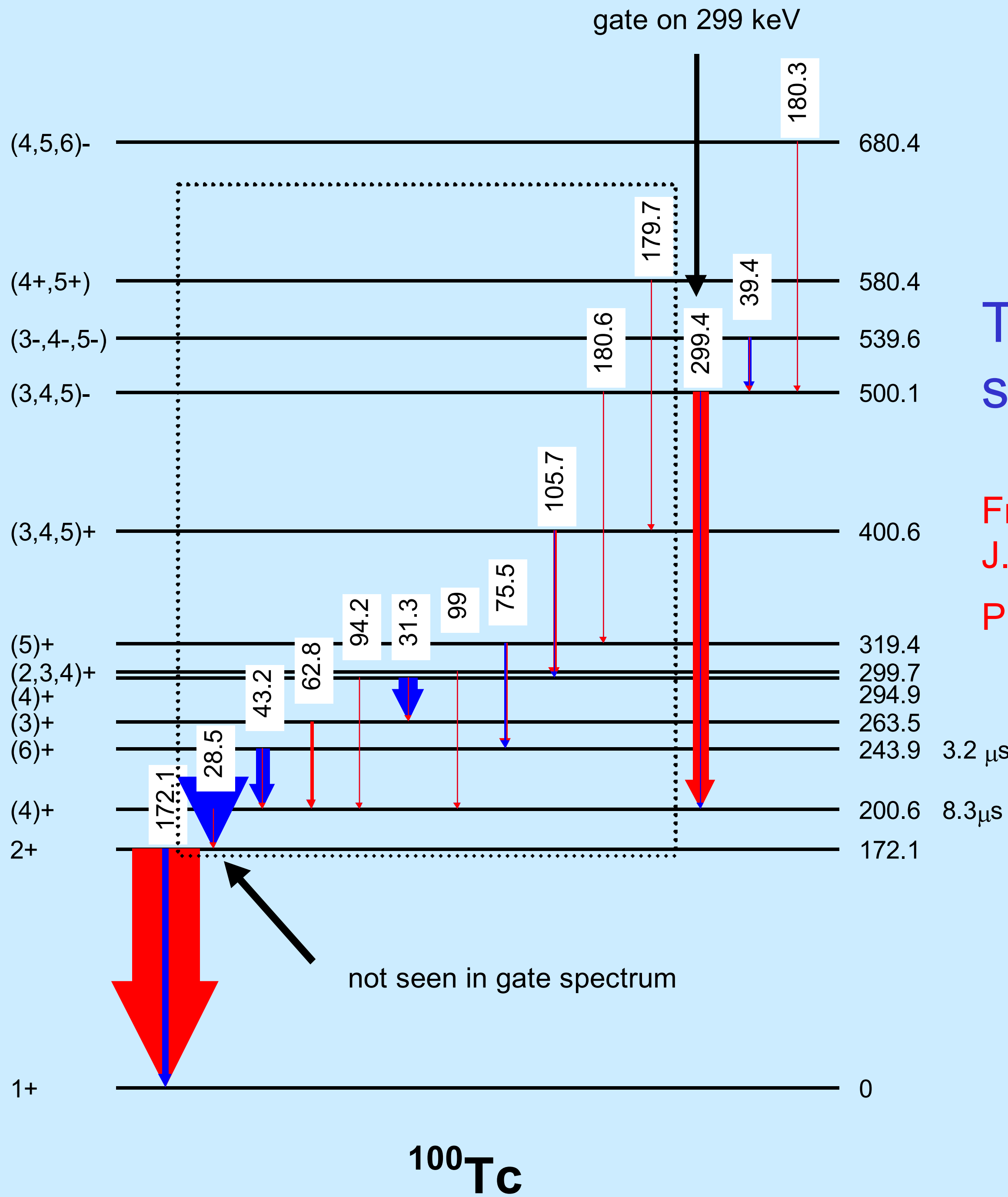
  transmutation yield calculations

  normalization point for differential cross section experiments (e.g. GELINA, Geel, Belgium, n-TOF, CERN)

Our goal is to measure these quantities with high precision

^{99}Tc (d,p) spectrum (München tandem)





Tc(n,γ) partial Level-scheme

From ENSDF &
J.A. Pinston et al. Nucl. Phys. **A321** (1979) 25

Why to measure nuclear waste, structural material capture cross sections?

- Waste is generated by power production
- Safe disposal → transmutation by ADS
- Problematic wastes are the Long-Lived Fission Fragments or LLFFs (^{99}Tc, ^{129}I,…) and minor actinides
- Structural materials are parts of ADS (Bi, Pb, ...)
- For optimization of transmutation yields, control of transmutation and detection of various isotopes in waste:

↓

Accurate γ-ray yields and thermal neutron cross sections are needed

Objectives

(n,γ) reactions

PGAA

Research

- Archaeology (IAEA)
- Geology
- Catalyst
- Material sciences
- In-beam Mössbauer
- Safeguard

Methodology

- PGAA library (IAEA, LBL)
- Chopped beam PGAA
- Standards (IAEA)

Cross sections

Research

- Xsections for ADS
- LLFF
  - ^{99}Tc, ^{129}I (IRMM)
- Fuel
  - ^{238,235}U,^{232}Th
- Structural mat.
  - ^{209}Bi, ^{204,206,207}Pb (IRMM)

Methodology

- Internal comparator
- Chopped beam

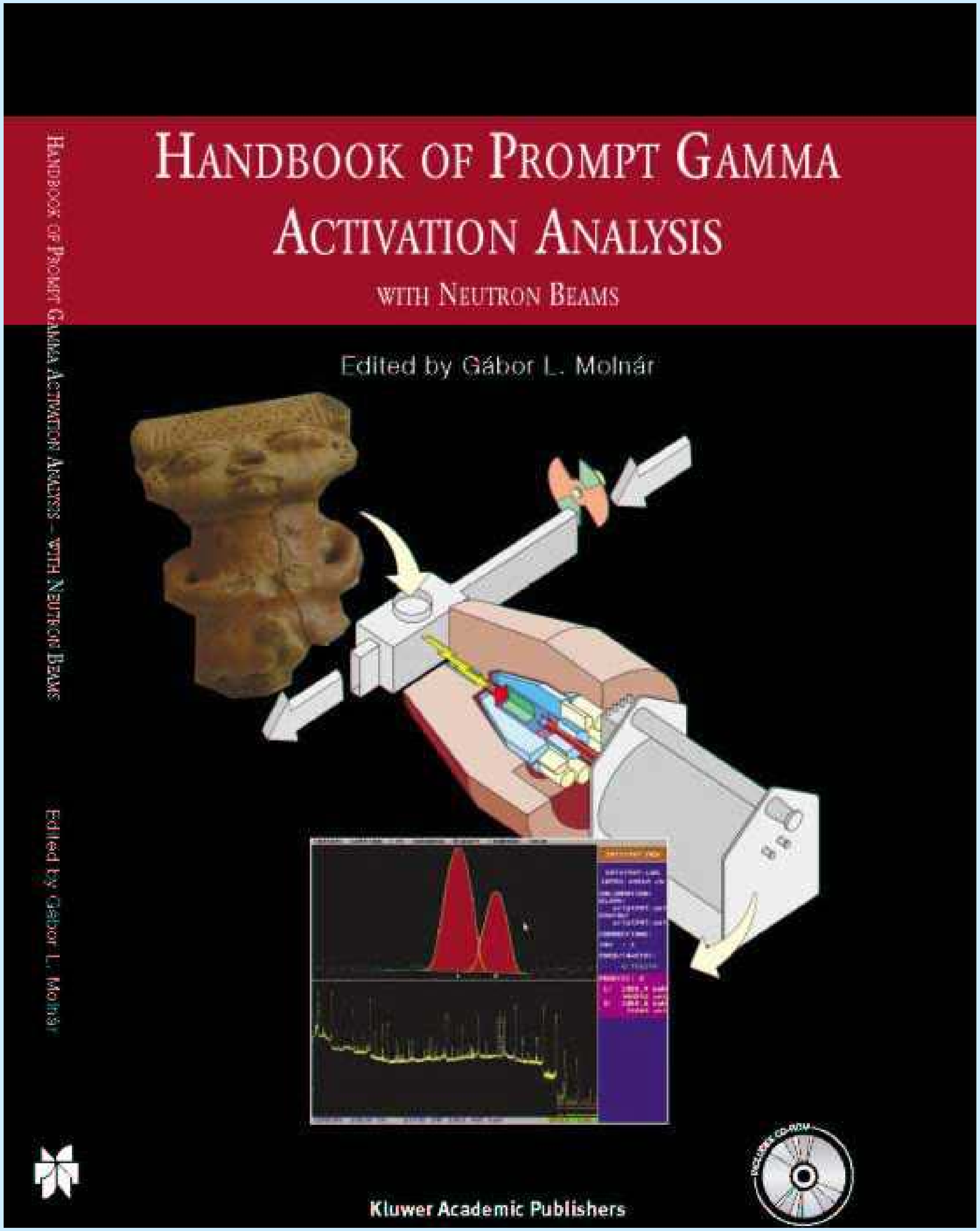
Nuclear Physics

Research

- Decay schemes
  - ^{204,206,207}Pb (IRMM)
  - ^{99}Tc
- Strength functions
  - ^{57}Fe (Oslo, Frank L.)
  - Mo, Yb, Gd
- Modeling Decay s.
  - ^{99}Tc

Methodology

- (n,γ), (n,$\gamma\gamma$)
- Monte Carlo



Editor: G. L. Molnár†

1. Principles of the PGAA method
(Zs. Révay, T. Belgya)

2. Beams and Facilities
(R.M. Lindstrom, Zs. Révay)

3. Samples and Standards
(R.M. Lindstrom, Ch. Yonezawa)

4. Gamma-Ray Spectrometry
(T. Belgya, Zs. Révay)

5. Quantitative Analysis
(Ch. Yonezawa)

6. Applications of PGAA with Neutron Beams
(D.L. Anderson, Zs. Kasztovszky)

7. Appendices Reference Data
(R.B. Firestone, G.L. Molnár, Zs. Révay)

+ CD supplement

General precision 1-5% for σ_γ

NH_4TcO_4 (n,γ) spectrum, H comparator