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Non-destructive analysis based on neutron induced reactions.
&
Thermal and epithermal neutron cross sections measurements and applications.

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Joint Research Centre (JRC)

Non-destructive analysis based on neutron induced reactions

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IRMM - Institute for Reference Materials and Measurements

Geel - Belgium

http://irmm.jrc.ec.europa.eu/

http://www.jrc.ec.europa.eu/

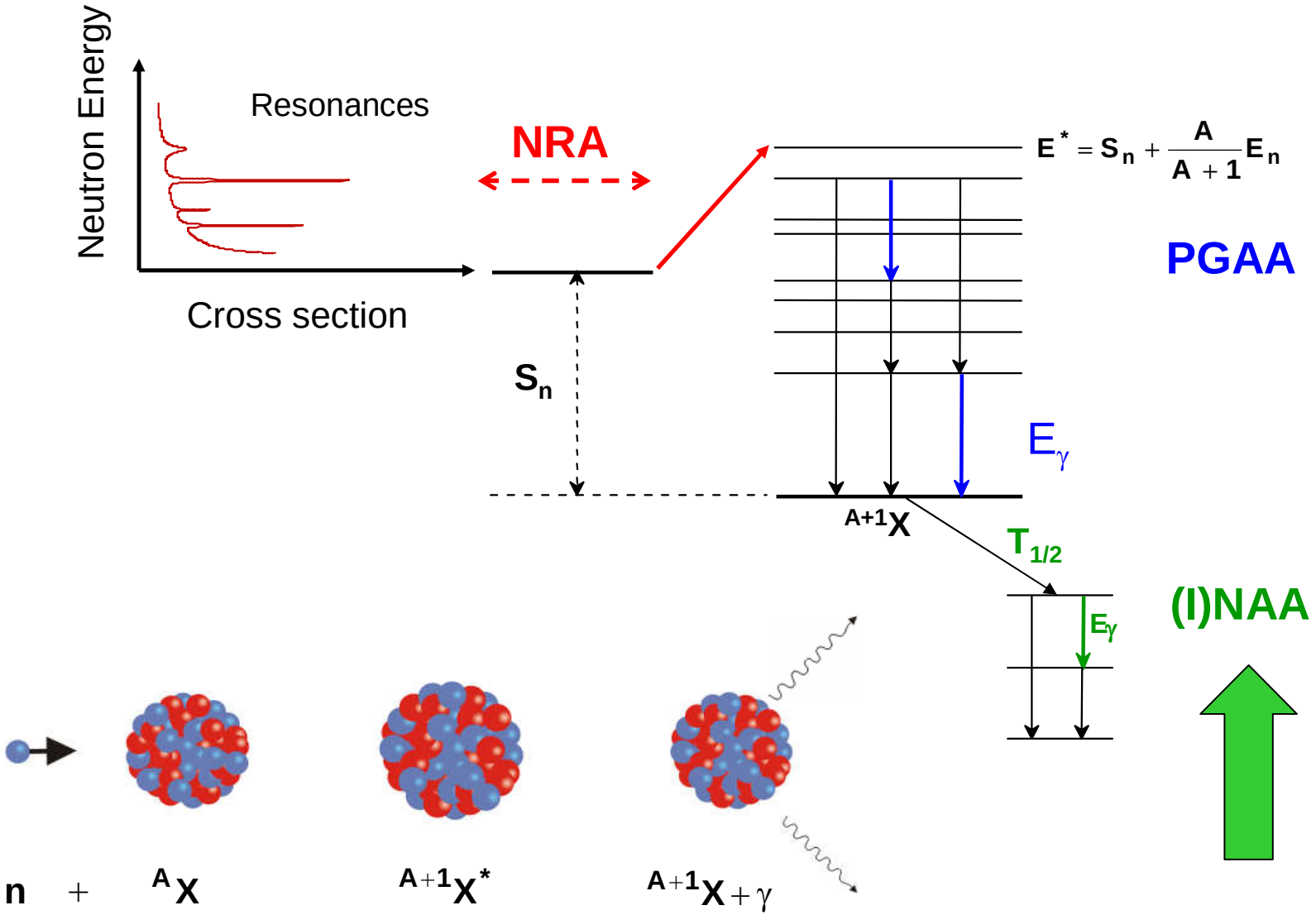
- **Neutron capture process**
  - Instrumental Neutron Activation Analysis (INAA)
  - Prompt Gamma Activation Analysis (PGAA)
  - Neutron Resonant Analysis (NRA)
    - Capture (NRCA)
    - Transmission (NRT)

- **Principles**
  - Physics and equations
  - Cross sections/Composition
- **Application to NDA**
- **Facilities/Examples**
- **Concluding Remarks**

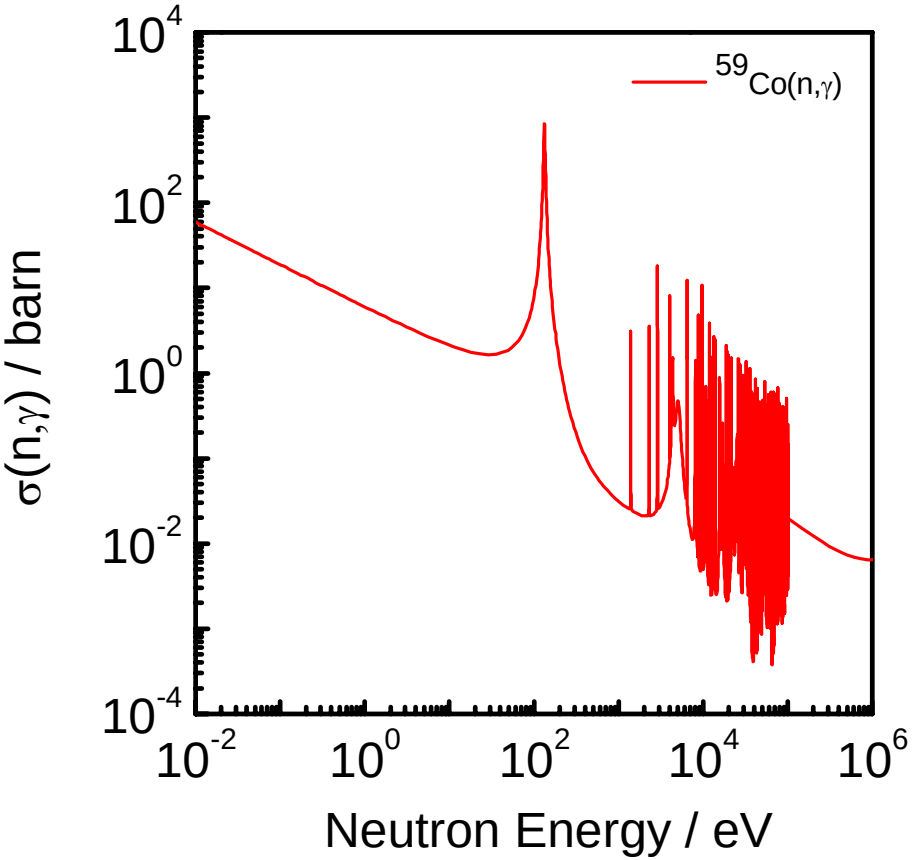
INAA - Overview

- **Principles/Quantities**
  - Neutron Capture Reactions
  - Reaction Rate
  - Flux shape
  - Westcott's g-factor and resonance integrals
- **INAA for NDA**
  - Determination of relative abundances
  - Standardization (k_0)
- **Facilities/Examples**

Neutron capture process



Capture cross section



The probability that a neutron interacts with a nucleus is expressed as a cross section σ, which has the dimension of an area

The unit of a cross section is taken as :

1 barn, 1 b = 10^{-24} cm^2

To calculate reaction probabilities we express the target thickness in atoms per barn :

$$n = \frac{0.6022}{m_A} \rho\, t$$

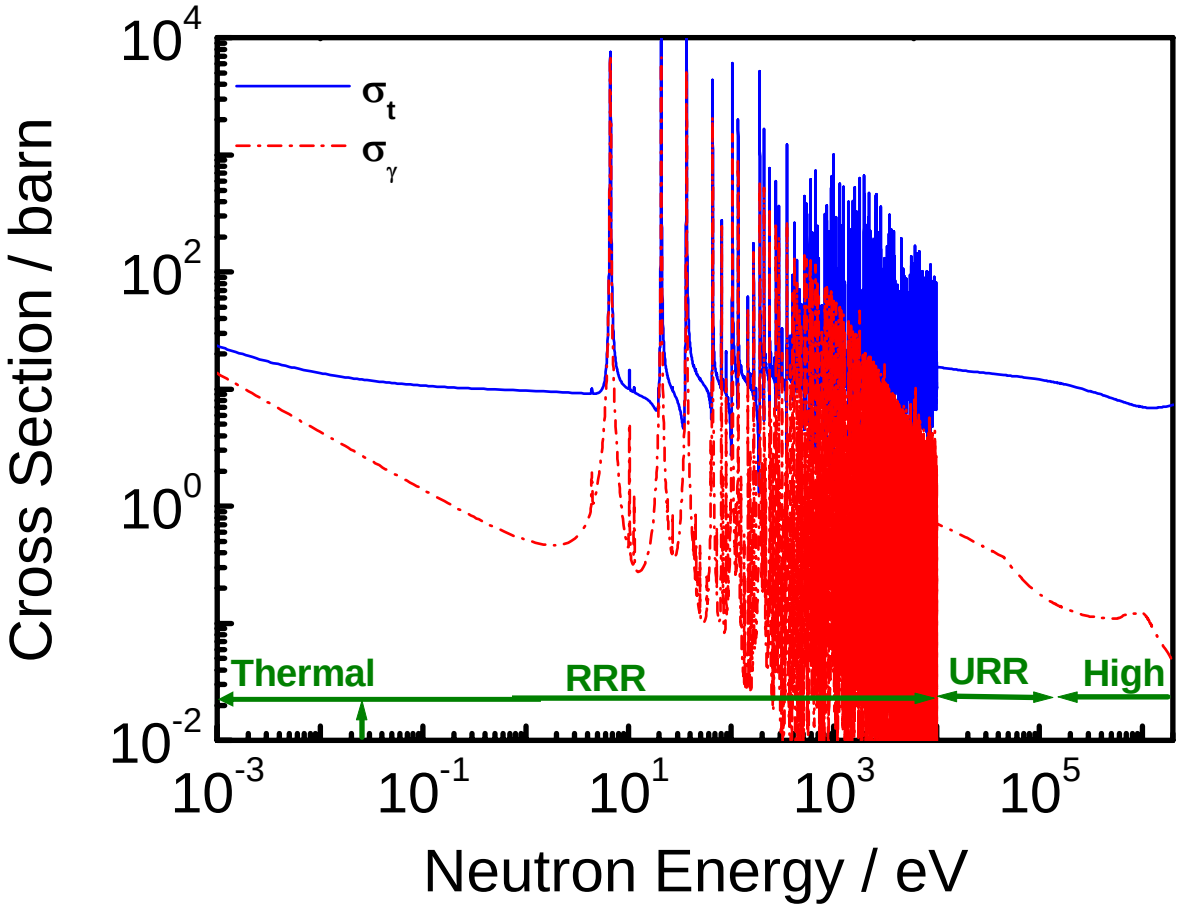
m_A : atomic mass

ρ : density in g/cm^3

t : thickness in cm

n : target thickness in at/b

Typical Cross Section shape (^{238}U)



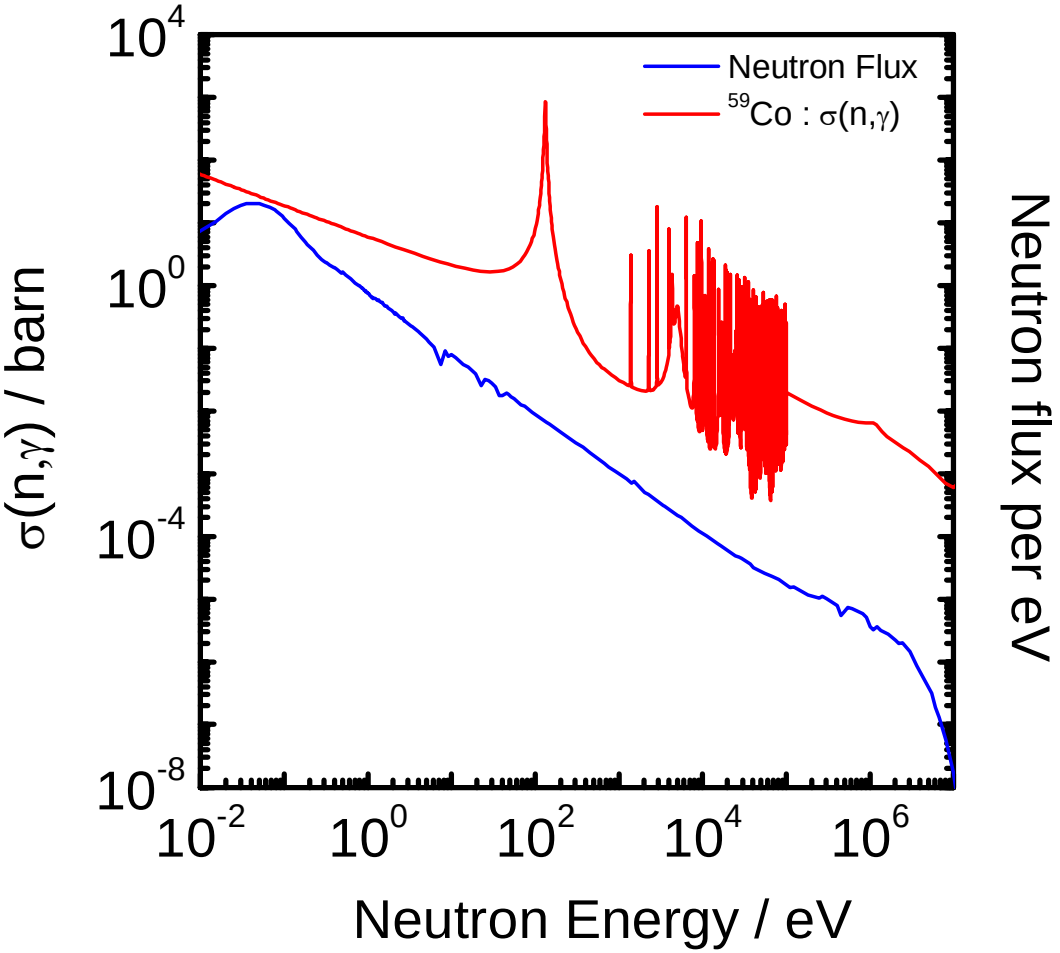
1/v part

RRR

URR

High energy part

Total reaction rate



The total reaction rate per atom:

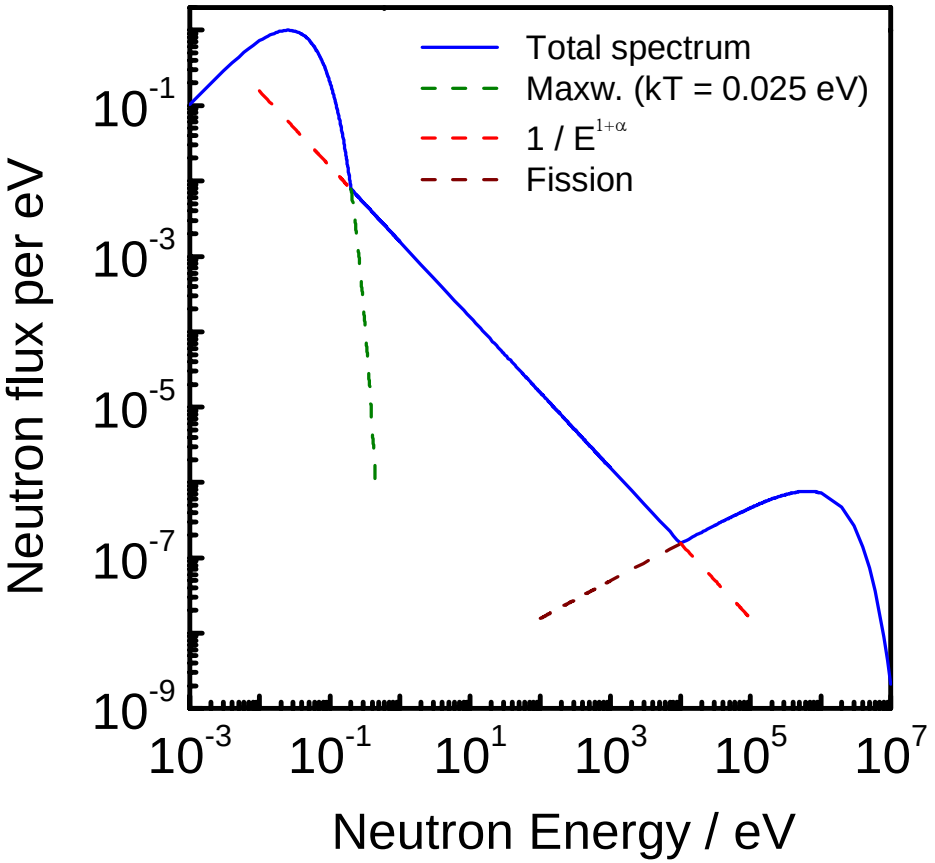
$$R = \int_0^\infty \varphi(E_n)\, \sigma_\gamma(E_n)\, dE_n$$

depends on:

$\varphi(E_n)$ the neutron flux and

$\sigma_\gamma(E_n)$ the capture cross section

Neutron flux in a thermal reactor



The neutron flux in a thermal reactor is a sum of three components

- **Maxwellian distribution with maximum at E_n =kT**
- **$1/E^{1+\alpha}$ distribution due to moderation process of the fast neutrons (epi-thermal spectrum)**
- **"Watt spectrum" of fission neutrons**

At a neutron guide, the neutron flux can be described by the thermal part only (e.g. PGAA at Budapest)

Reaction rate in a thermal reactor

The total reaction rate per atom is:

$$R = \int_0^\infty \varphi(E_n)\,\sigma(E_n)\,dE_n$$

To solve the integral one separates between the thermal and the epi-thermal region:

$$R = \int_0^\infty \sigma(E)\varphi(E)dE = \int_0^{E_{cd}} \sigma(E)\varphi(E)dE + \int_{E_{cd}}^\infty \sigma(E)\varphi(E)dE$$

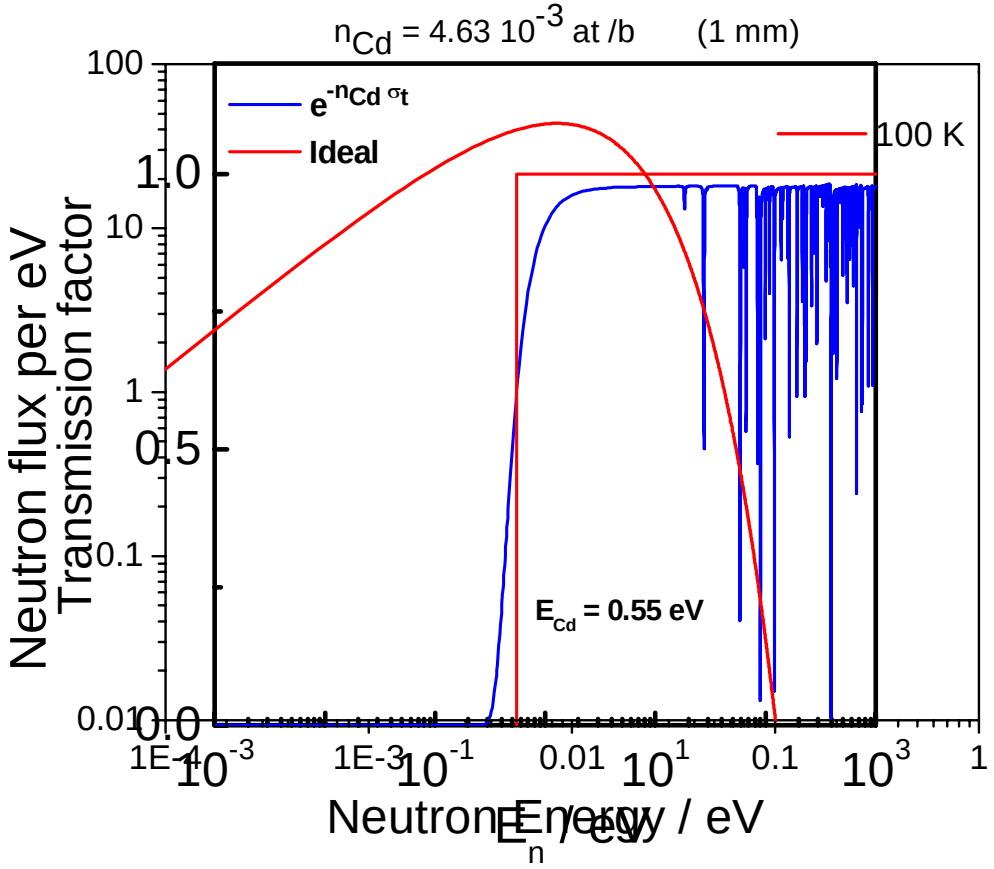
with E_{Cd} = 0.55 eV

Guided beam

$$R = \int_0^{E_{cd}} \sigma(E)\varphi(E)dE$$

Cd measurement

$$R = \int_{E_{cd}}^{\infty} \sigma(E)\varphi(E)dE$$



Reaction rate in a thermal reactor

$$R = \int_0^{\infty} \sigma(E)\varphi(E)dE = \int_0^{E_{cd}} \sigma(E)\varphi(E)dE + \int_{E_{cd}}^{\infty} \sigma(E)\varphi(E)dE$$

$$R = \varphi_t \sigma_0 g_w G_{th} + \varphi_e I_r G_r = \varphi_t \sigma_0 \left(g_w G_{th} + \frac{1}{f}\frac{I_r}{\sigma_0} G_r \right)$$

φ_t thermal flux

σ_0 thermal cross section at v=2200 m/s

g_w generalized g-factor: deviation of the cross section from 1/v

G_{th} thermal flux depression factor

φ_e epithermal flux

I_r effective resonance integral

G_r resonance self-shielding factor

f ratio of thermal to epithermal flux

$Q=I_r/\sigma_0$ ratio of the resonance integral and thermal cross section

Reaction rate in a thermal reactor

$$\int_0^{E_{cd}} \sigma(E)\varphi(E)dE = \varphi_t \sigma_0 g_w G_t$$

depends on the geometry, composition (σ)

$$\varphi_t = \frac{2}{\sqrt{\pi}} \int_0^{E_{cd}} \varphi(E)dE$$

$$\sigma_{th} = \frac{\int_0^{E_{cd}} \sigma(E)\varphi(E)dE}{\int_0^{E_{cd}} \varphi(E)dE}$$

$$g_w = \frac{\int_0^{E_{cd}} \sigma(E)\varphi(E)dE}{\frac{\sqrt{\pi}}{2}\varphi_t \sigma_0} = \frac{\int_0^{E_{cd}} \sigma(E)\varphi(E)dE}{\sigma_0 \int_0^{E_{cd}} \varphi(E)dE} = \frac{2}{\sqrt{\pi}} \frac{\sigma_{th}}{\sigma_0}$$

Reaction rate in a thermal reactor

$$g_w = \frac{\int_0^{E_{cd}} \sigma(E)\varphi(E)dE}{\frac{\sqrt{\pi}}{2}\varphi_t\sigma_0} = \frac{\int_0^{E_{cd}} \sigma(E)\varphi(E)dE}{\sigma_0 \int_0^{E_{cd}} \varphi(E)dE} = \frac{2}{\sqrt{\pi}}\frac{\sigma_{th}}{\sigma_0}$$

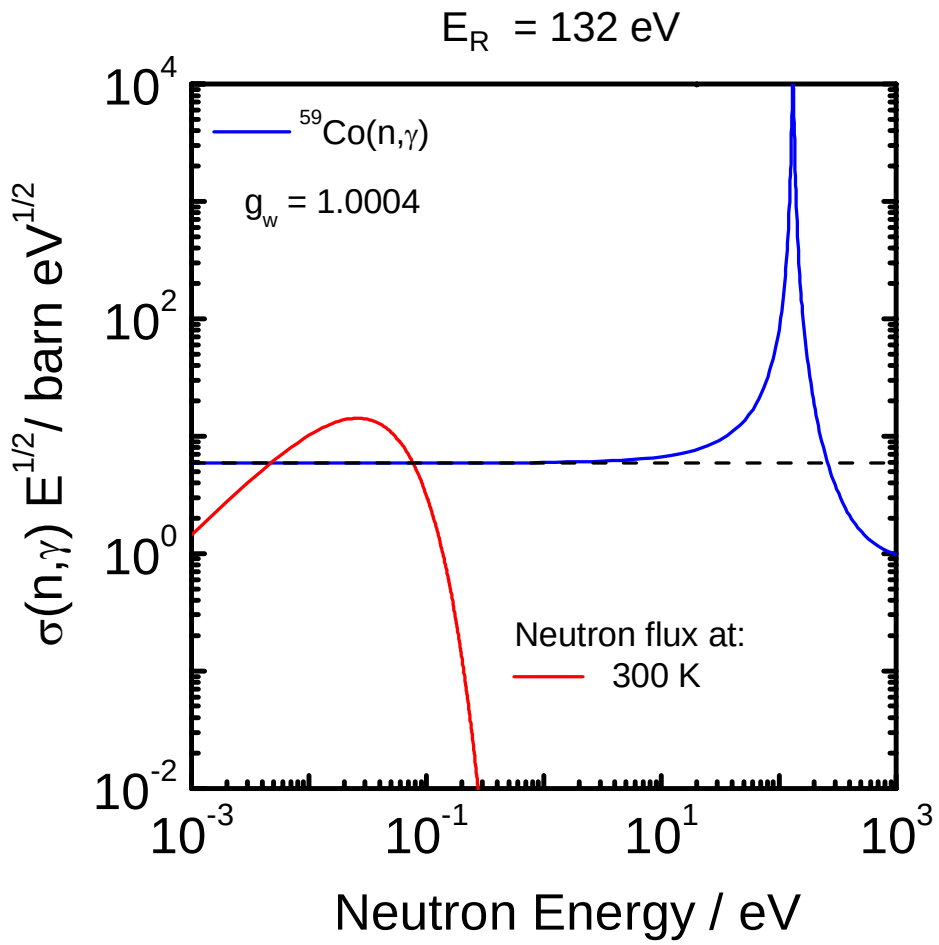
g_w depends on:

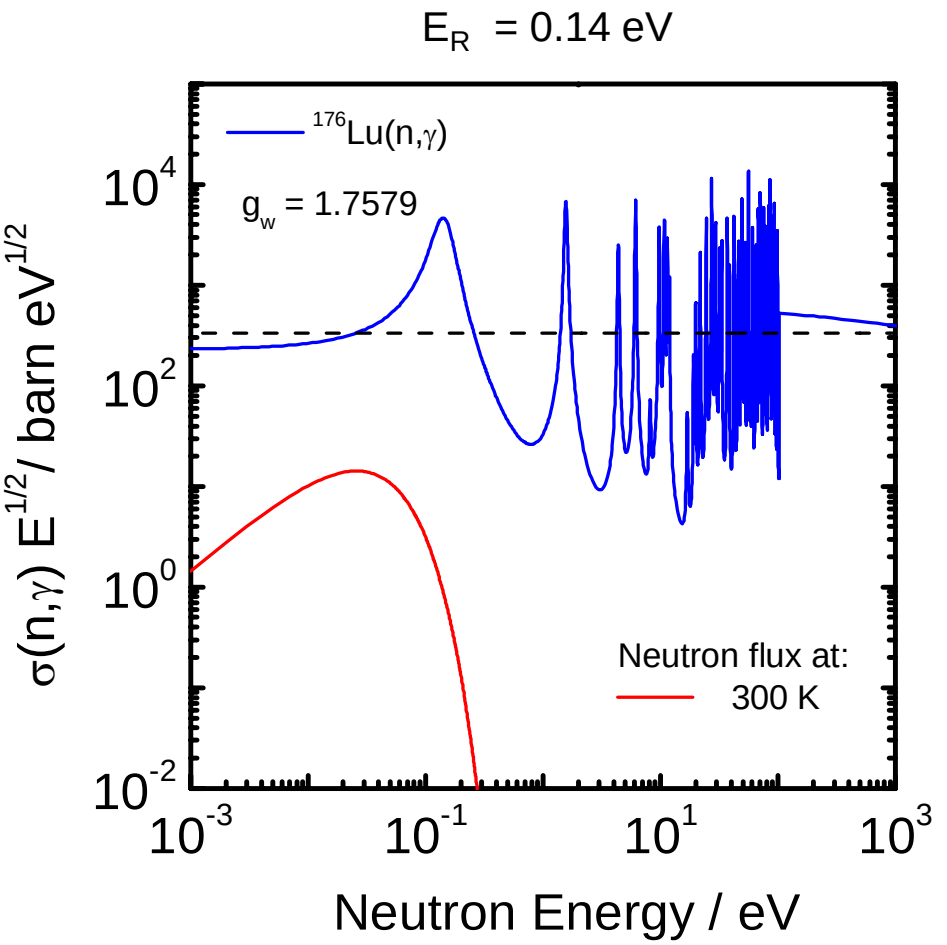
- **Flux shape (may not be Maxwellian)**
- **Cross section shape (may not be 1/v)**

If the flux has a Maxwellian distribution and If the cross section is 1/v

$$\sigma_{th} = \frac{\sqrt{\pi}}{2}\sigma_0\sqrt{\frac{T_0}{T}} \quad \Rightarrow \quad g_w = \sqrt{\frac{T_0}{T}}$$

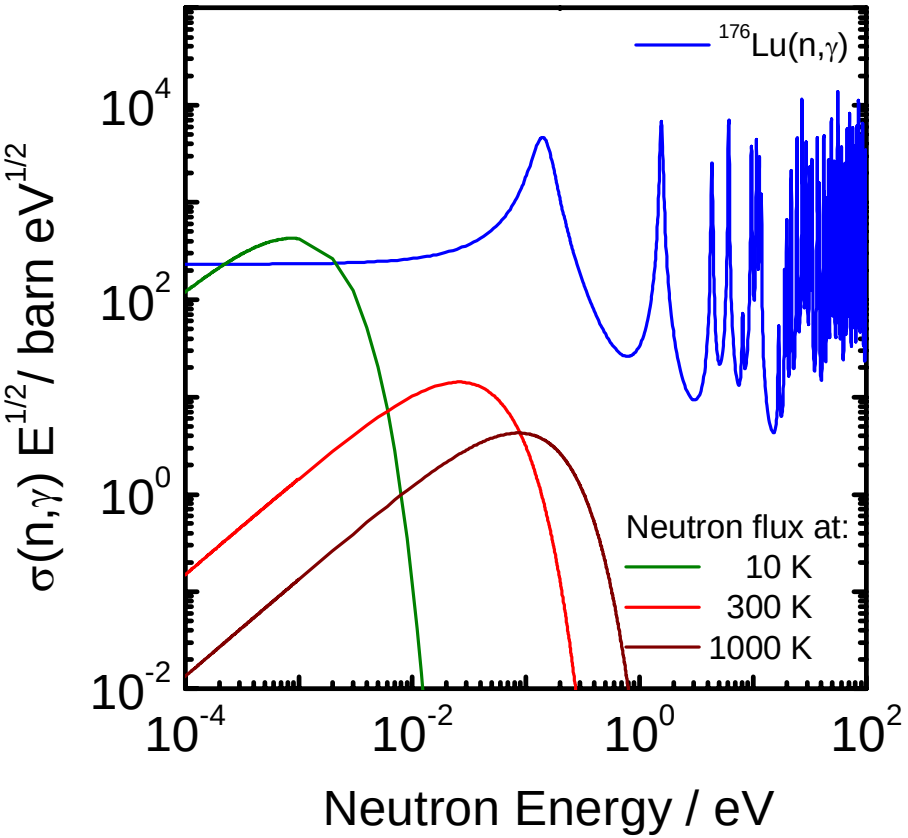
g_w - factor: deviation of 1/v behaviour #1

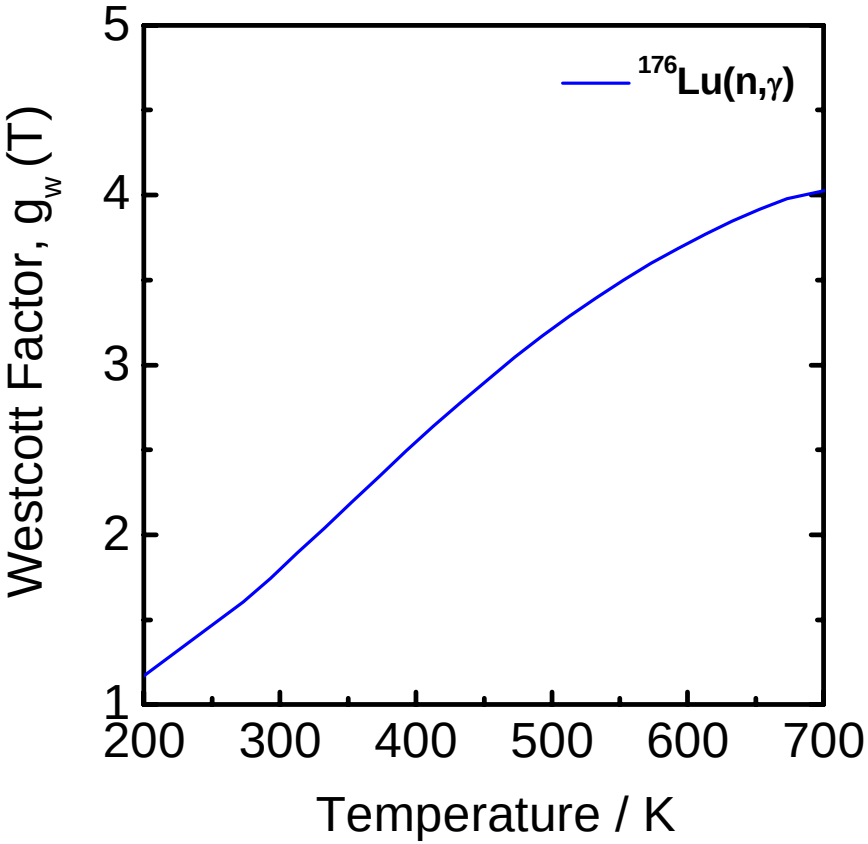




g_w - factor: deviation of 1/v behaviour #2

The Westcott g_w – factor is temperature dependent





Reaction rate in a thermal reactor

$$R = \varphi_t \sigma_0 g_w G_{th} + \varphi_e I_r G_r$$

$$\varphi_e I_r G_r = \int_{E_{cd}}^{\infty} \sigma(E)\varphi(E)dE$$

$$\varphi_e = \int_{E_{cd}}^{\infty} \varphi(E)dE$$

$$I_r = \frac{\int_{E_{cd}}^{\infty} \sigma(E)\varphi(E)dE}{\varphi_e}$$

Resonance integral

I_r depends on:

- $\sigma(E)$ (resonance parameters)
- $\varphi(E)$ (e.g. $E^{-(1+\alpha)}$)

Data base of σ_0, g_w-factor and I_r
Compilation by S.F.Mughabghab, BNL, USA
"Thermal neutron capture cross sections, resonance integrals and g_w-factors"
INDC(NDS) – 440
February 2003

So far, one atom
Real case N atoms

$$R_N = N\varphi_t\sigma_0\left(g_w G_{th} + \frac{1}{f}\frac{I_r}{\sigma_0}G_r\right)$$

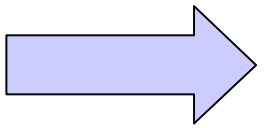
R_N from measurement

Knowing N, we can deduce σ (nuclear data)

Knowing σ (nuclear data), we can deduce N

How to measure the R_N

- Sample activation by neutron irradiation in reactor
- The sample is transferred into a counting station (Rabbit system)
- (Waiting time)
- Counting
- Isotope identification by means of the γ–ray energy
- High resolution detector needed
  - Ge(Li), HPGe
  - NaI
- Peak analysis
- Correlate peak area with R, isotope abundance



Simple reaction/decay (no branching…)

$$C_\gamma = \frac{N_{av} W \theta}{M} P_\gamma \varepsilon_{FE} t_m R \left(1 - e^{-\lambda t_i}\right) e^{-\lambda t_d} \frac{\left(1 - e^{-\lambda t_m}\right)}{e^{-\lambda t_m}}$$

C_γ Counts (DT,coinc)

W Sample mass

θ Isotopic abundance

M Molar mass

N_{av} Avogadro's number

P_γ γ-ray intensity

$$R = \varphi_t \sigma_0 \left(g_w G_{th} + \frac{1}{f} \frac{I_r}{\sigma_0} G_r \right)$$

t_i Irradiation Time

t_d Decay Time

t_m Counting Time

NAA for Non Destructive Analysis

$$W = \frac{C_\gamma}{t_m \left(1 - e^{-\lambda t_i}\right) e^{-\lambda t_d} \frac{\left(1 - e^{-\lambda t_m}\right)}{e^{-\lambda t_m}}} \frac{M}{N_{av} \theta} \frac{1}{R P_\gamma \varepsilon_{FE}}$$

– Absolute method (ε_{FE}, P_γ,σ_0 sources of uncertainty)

Relative abundance

$$\frac{W}{W_{ref}} = \frac{C_\gamma}{C_{\gamma,ref}} \frac{\varepsilon_{FE,ref}}{\varepsilon_{FE}} \frac{G_t g_w + G_f \frac{Q}{f}}{G_{t,ref} g_{w,ref} + G_{f,ref} \frac{Q_{ref}}{f}} \frac{\sigma_{0,ref}}{\sigma_0} \frac{M}{\theta P_\gamma} \frac{\theta_{ref} P_{\gamma,ref}}{M_{ref}}$$

Reference $^{197}Au(n,\gamma)$ E_γ=411.8 keV

k_0

accurately determined for many nuclides

depends only on the isotope

$$\frac{1}{k_0} = \frac{\sigma_{0,ref}}{\sigma_0} \frac{M}{M_{ref}} \frac{\theta_{ref} P_{\gamma,ref}}{\theta P_\gamma}$$

e.g. De Corte/Simonits (JRNC 133 (1989) 43)

N.B. It is actually a cross section measurement!

Relative abundance

$$\frac{W}{W_{ref}} = \frac{C_\gamma}{C_{\gamma,ref}} \frac{\varepsilon_{FE,ref}}{\varepsilon_{FE}} \frac{G_t g_w + G_f \frac{Q}{f}}{G_{t,ref} g_{w,ref} + G_{f,ref} \frac{Q_{ref}}{f}} \frac{1}{k_0}$$

Usually a ^{197}Au sample is co-irradiated with the sample to analyse

Experimental conditions

- ε_{FE}
- $\varphi(E)$ Q, g_w,f
- geometry G

$$\frac{G_t g_w + G_f \frac{Q}{f}}{G_{t,ref} g_{w,ref} + G_{f,ref} \frac{Q_{ref}}{f}}$$

Microscopic data

- $\sigma(E)$ Q, g_w, k_0

- **NIST Center for Neutron Research**
- **BNC (H)**
  - inorganic impurities in C_{60}
  - selenium in food
- **BR-1 at the SCK-CEN (B)**
- **INAA Laboratory at the Delft University of Technology (NL)**

and many others…

Pros:

- Multi elemental method
- High accuracy
- Low detection limits
- Most elements can be traced

Cons:

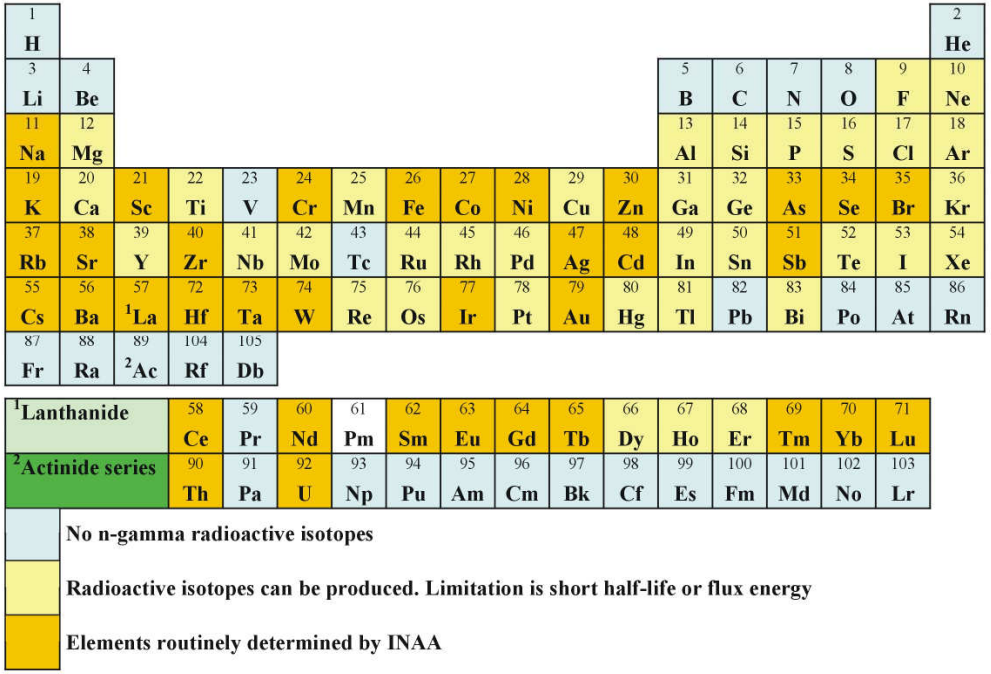
- Reactor (high flux) needed
- Activation, waiting time

Data needed:

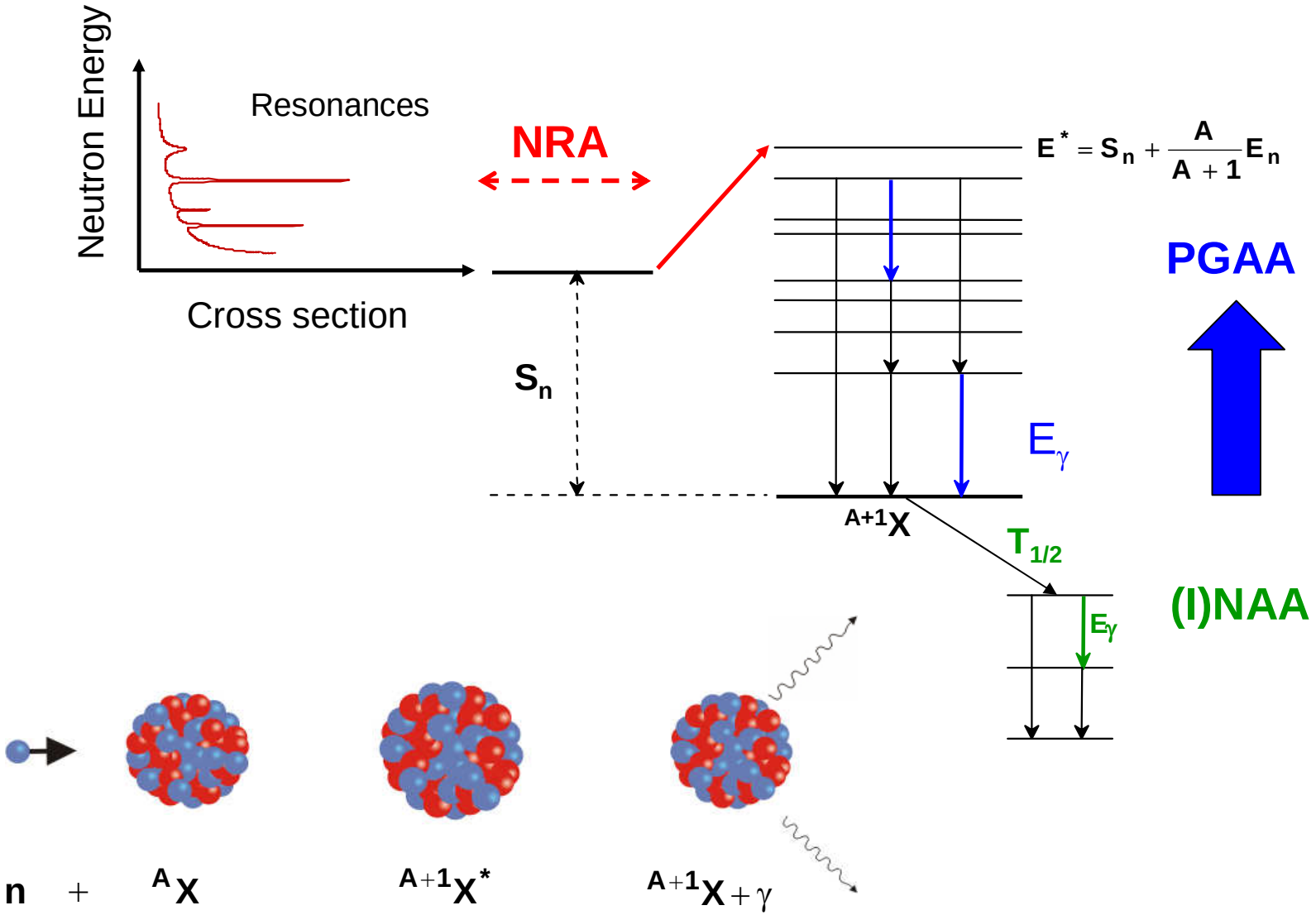
- k_0, g_w, l_r

OR

- Thermal cross sections
- Resonance Parameters



Neutron capture process



PGAA - Overview

- **Principles**
  - Basic equations
- **PGAA for NDA**
  - PGAA Standardization
  - PGAA at the BNC
    - Cold beam
    - Compton suppression system
  - Remarks
- **Examples**

Neutron Induced Prompt Gamma Activation Analyses (PGAA or PGNAA)

- Detection of prompt gamma radiation from a neutron induced nuclear reaction, usually radiative capture
- Isotope identification based on E_γ
- Isotope quantification based on I_γ
- Instantaneous method: results appear immediately
- Usually little or negligible residual activation of the sample

PGAA - principle

The total reaction rate per atom:

$$R = \int_0^{\infty} \varphi(E_n)\, \sigma_\gamma(E_n)\, dE_n$$

depends on:

- $\varphi(E_n)$ the neutron flux and
- $\sigma_\gamma(E_n)$ the capture cross section

For a guided (thermal or cold) neutron beam $R = G_{th} \varphi_t \sigma_{\gamma 0} g_w$

PGAA - principle

If we look at a given transition

$$R_i = G_{th}\varphi_t\sigma_{\gamma 0,i}g_w = G_{th}\varphi_t P_\gamma \sigma_{\gamma 0} g_w$$

where P_γ is the intensity of the transition

for N atoms

$$R_{N,i} = NG_{th}\varphi_t P_\gamma \sigma_{\gamma 0} g_w$$

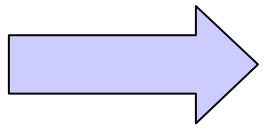
$R_{N,i}$ from measurement

Knowing N, we can deduce σ (nuclear data)

Knowing σ (nuclear data), we can deduce N

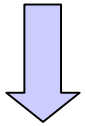
How to measure the R_N

- Sample by neutron irradiation (e.g. cold neutron beam)
- Counting
- Isotope identification by means of the γ–ray energy
- High resolution detector needed
  - Ge(Li), HPGe
  - NaI
- Peak analysis
- Correlate peak area with R, isotope abundance



non radioactive nucleus

$$C_\gamma = \frac{N_{av} W \theta}{M} P_\gamma \sigma_{\gamma 0} g_w G_{th} \varepsilon_{FE} t_m \varphi_t$$



C_γ	Counts (DT,coinc)
P_γ	γ-ray intensity
ε_{FE}	Full Energy peak detection efficiency
W	Sample mass
θ	Isotopic abundance
M	Molar mass
t_m	Counting Time

PGAA for Non Destructive Analysis

$$W = \frac{C_\gamma}{\varepsilon_{FE} t_m} \frac{M}{N_{av} \theta} \frac{1}{G_{th} \varphi_t P_\gamma \sigma_{\gamma 0} g_w}$$

– Absolute method (large uncertainties)

Relative measurements allow to reduce the uncertainty

– comparator (ref)

$$\frac{W}{W_{ref}} = \frac{C_\gamma}{C_{\gamma,ref}} \frac{\varepsilon_{FE,ref}}{\varepsilon_{FE}} \frac{g_w}{g_{w,ref}} \frac{\sigma_{0,ref}}{\sigma_0} \frac{M}{\theta P_\gamma} \frac{\theta_{ref} P_{\gamma,ref}}{M_{ref}} \rightarrow \frac{1}{k_0}$$

– homogeneous sample ⇒ G is the same

– efficiency ratio can be determined accurately (<1%)

– method standardization (Revay and Molnar, Radiochimica Acta 91, 361, 2003):

– k_0,σ_0 library

– Ultimate comparator: H σ_0 = 0.3326(7) b, 2.2 MeV

⇒ no standard samples needed

PGAA at the Budapest Neutron Centre

Why using a cold beam?

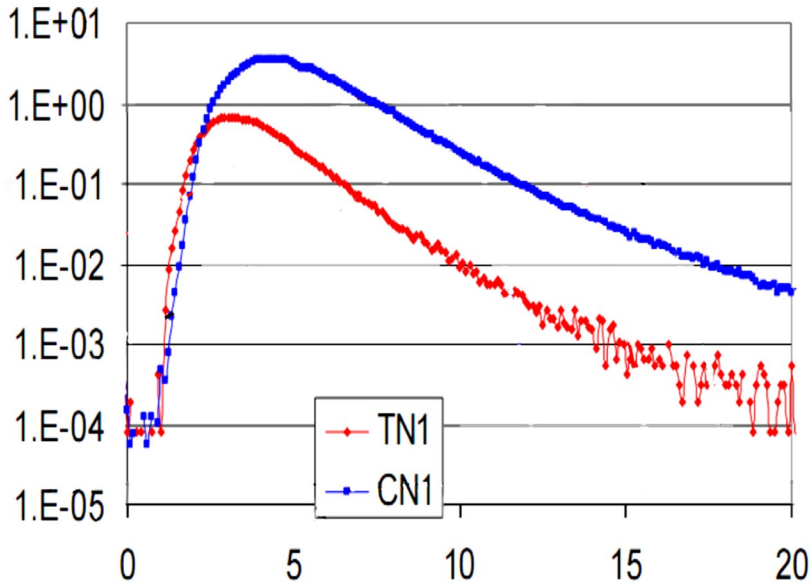
More neutron with lower energy

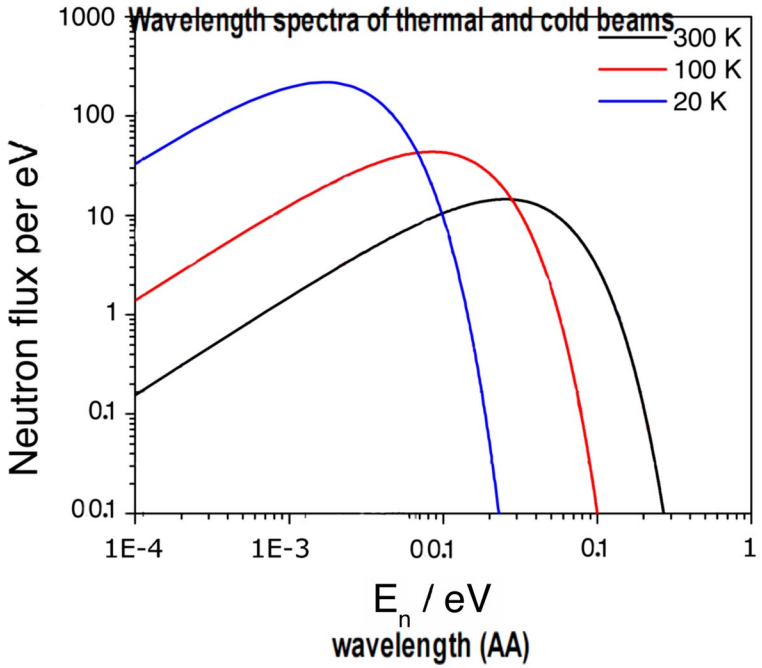
Cross section is higher

The count rate is higher

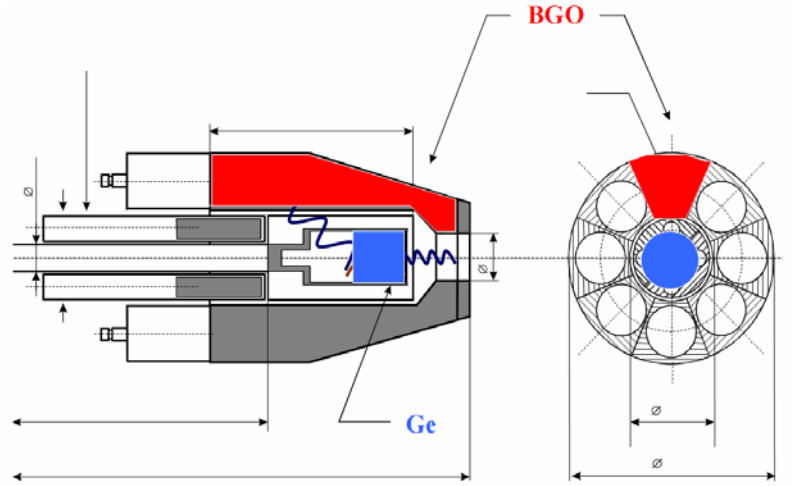
No epithermal neutrons

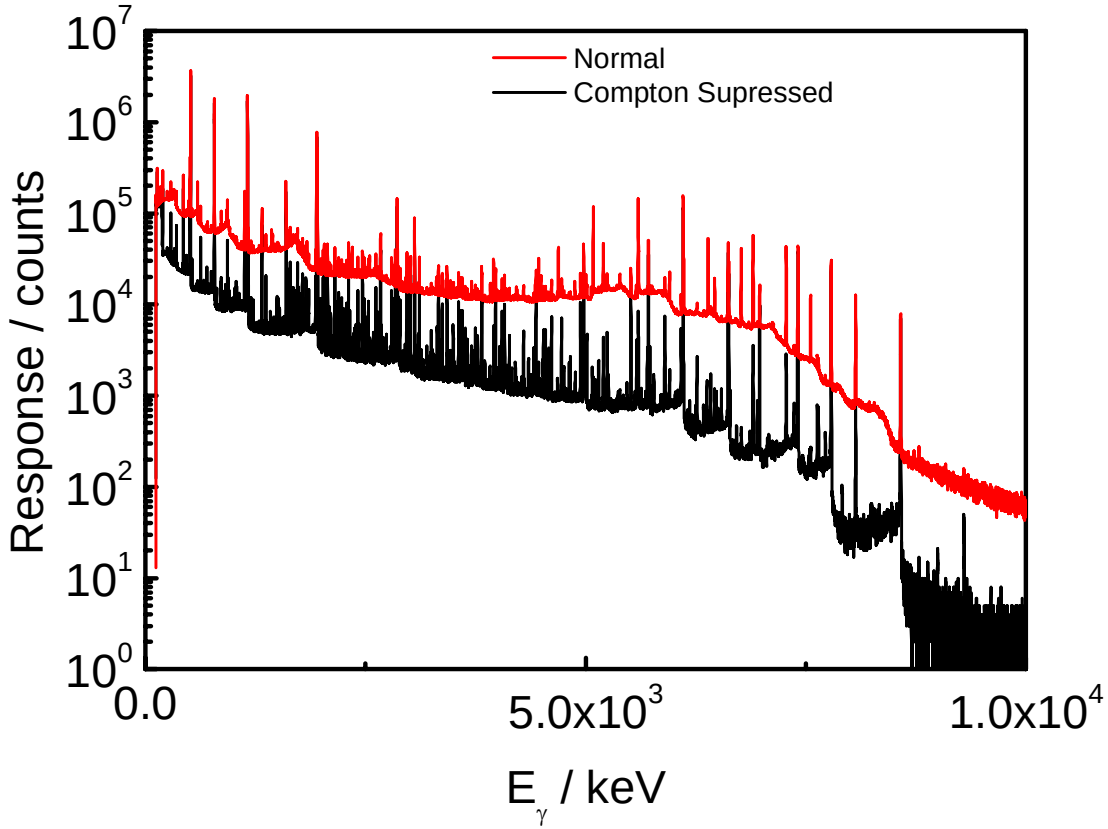
g_w is 1 in most cases





Why using Compton suppression system?





Peak to background improved

- **Neutron capture cross section measurements**
  - $^{nat}Pb, ^{206}Pb, ^{209}Bi, ^{127}I, ^{129}I$…
- **Rocks and minerals (Geology, Archaeology)**
  - lapislazuli composition (Cl, S content) ⇒ ore
- **Ceramics (Archaeology)**
- **Glasses Metals (Archaeology, Industry)**
  - Roman silver coins Cu/Ag ratio ⇒ period
- **Chemistry**
  - S in fullerene (C_{60})
- **Metals (Materials Research, Archaeology)**
- **Nuclear Materials (Safeguards, Transmutation)**
  - $^{235}U/^{238}U$ mass ratio

Pros:

- Multi elemental method
- Applicable in principle to all elements
- No special sample preparation

Cons:

- γ-ray spectra more complex than NAA

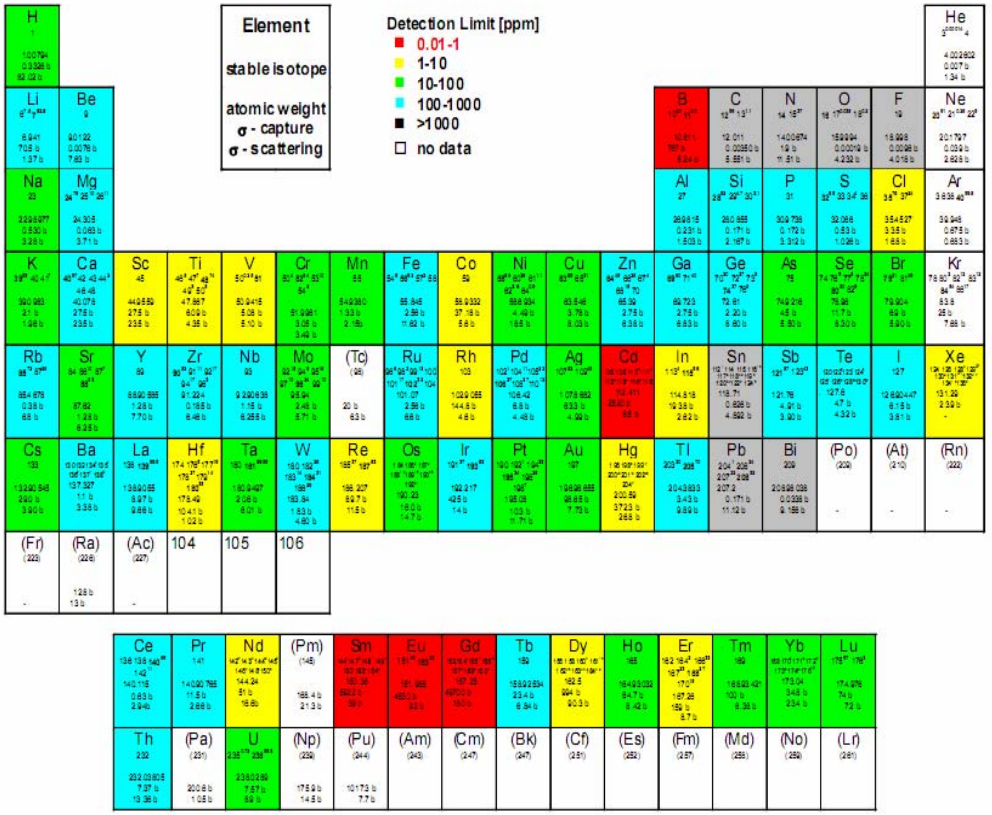
Data needed:

- g_w
- k_0

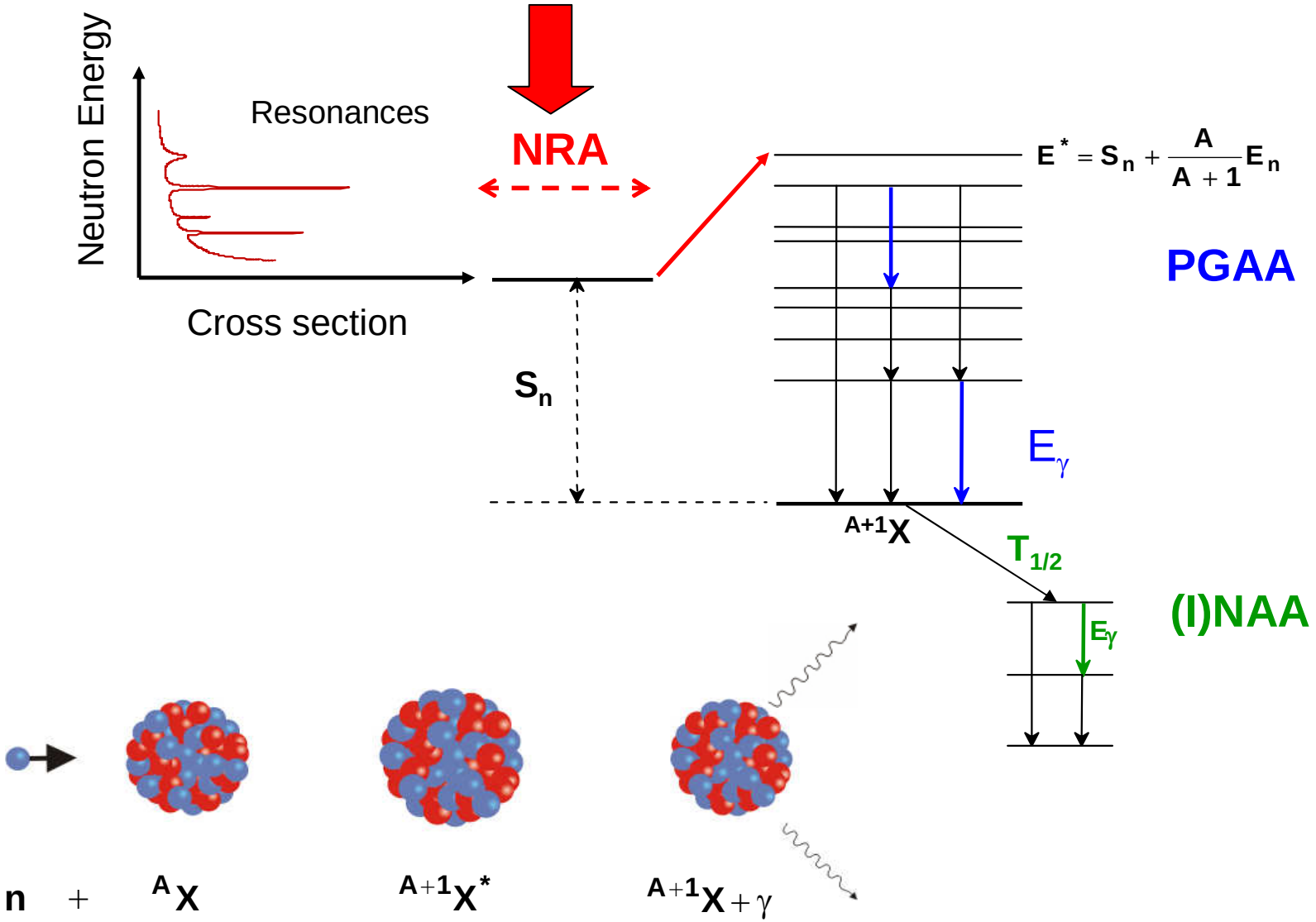
OR

- Thermal cross sections (Partial cross sections)

PGAA detection limits



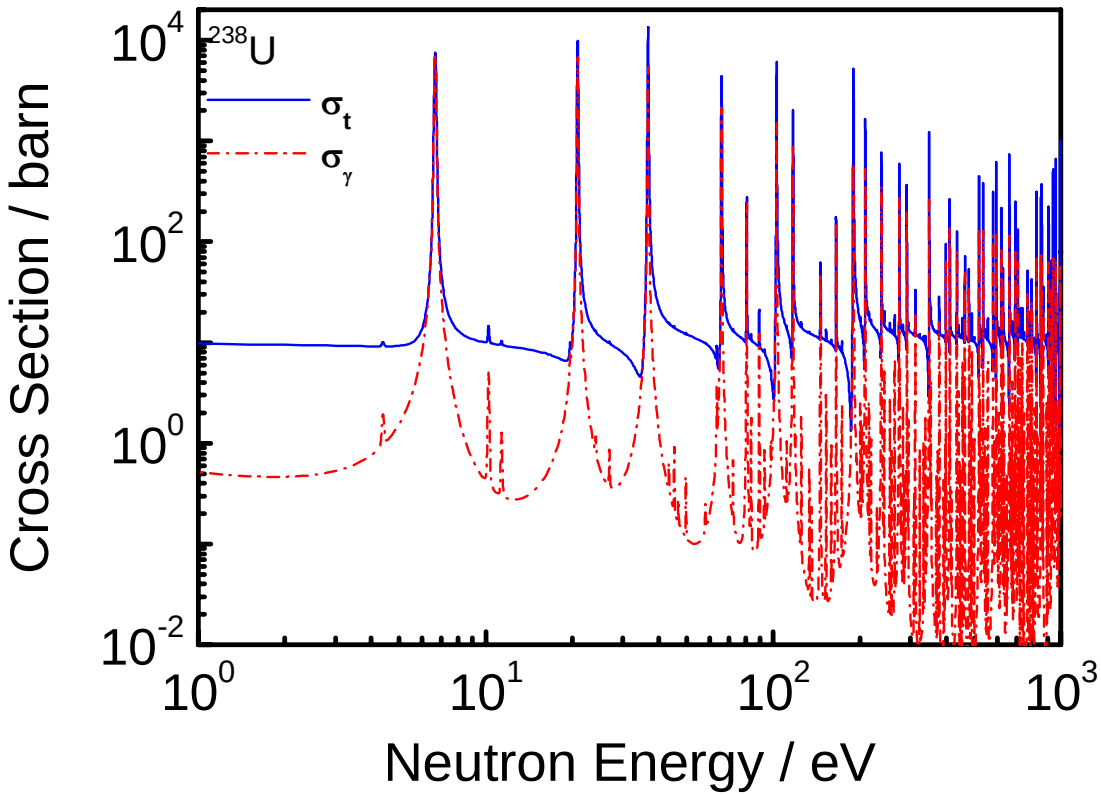
Neutron capture process



NRA - Layout

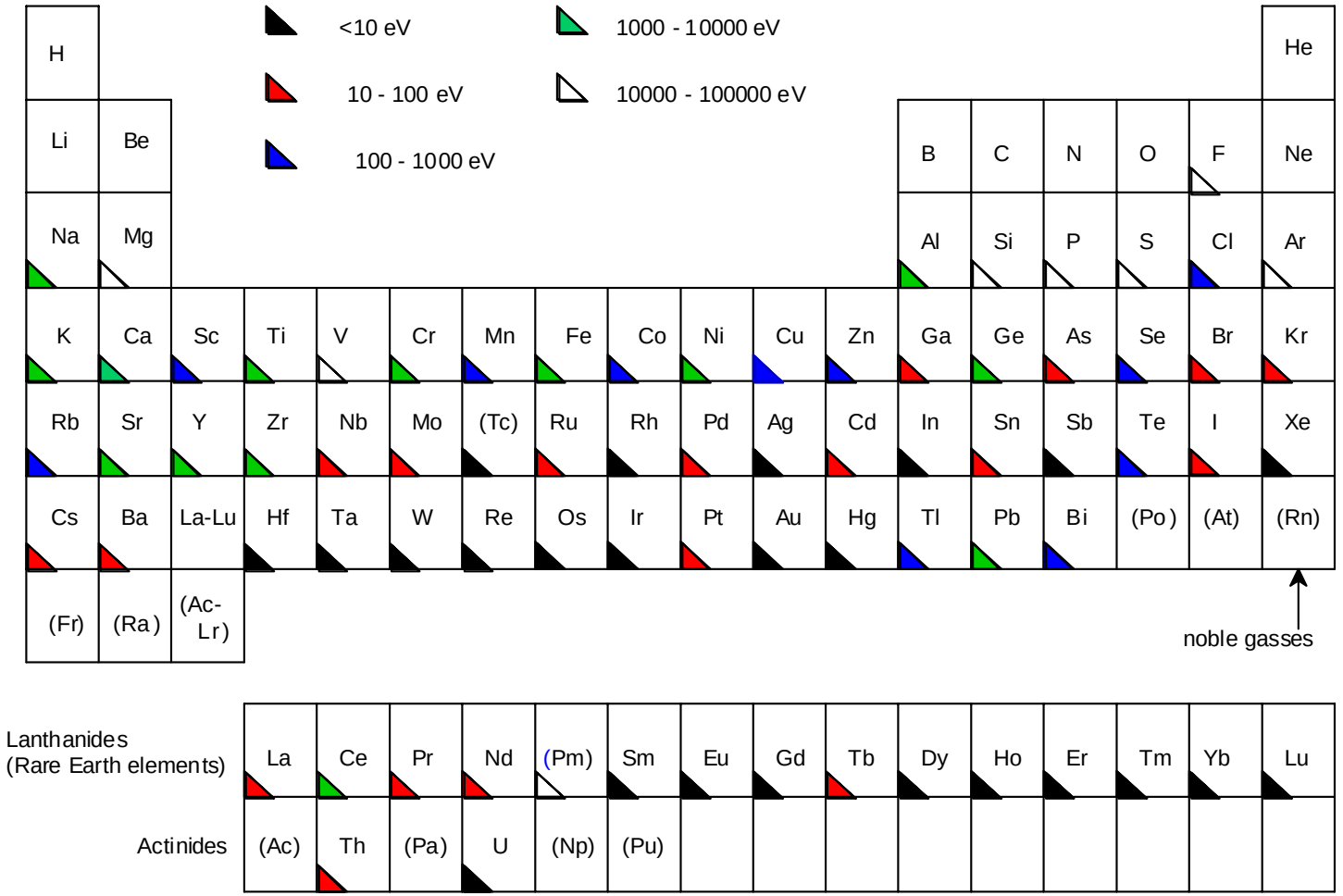
- Principle
  - Resonances for analysis of elemental composition
  - NRCA and NRT
- Data Analysis
  - Calibration approach
  - Methodological approach
- Applications
- NRA and PGAA
- ND for NRA
- Conclusions

Neutron Resonances for elemental identification



- Neutron resonances appear at given energies, specific for each nuclide
- Nuclides can be identified and the elemental (and isotopic) composition can be deduced
- Applicable for almost all elements
- No sample preparation required
- Non - Destructive
- Negligible residual activation

NRA feasibility



(I)NAA

Intense thermal neutron flux (irradiation in core, i.e. BR1)

Gamma detector: gamma ray energy resolution $\Rightarrow$ Ge-detectors

PGNAA

Intense neutron beam, (guided cold neutron beam Budapest, NIST)

Gamma detector: gamma ray energy resolution $\Rightarrow$ Ge-detectors

NRA

Pulsed white neutron beam (LINAC)

Gamma detector: good time resolution $\Rightarrow$ scintillators

Experimental Setup: Requirements

Pulsed Neutron Beam

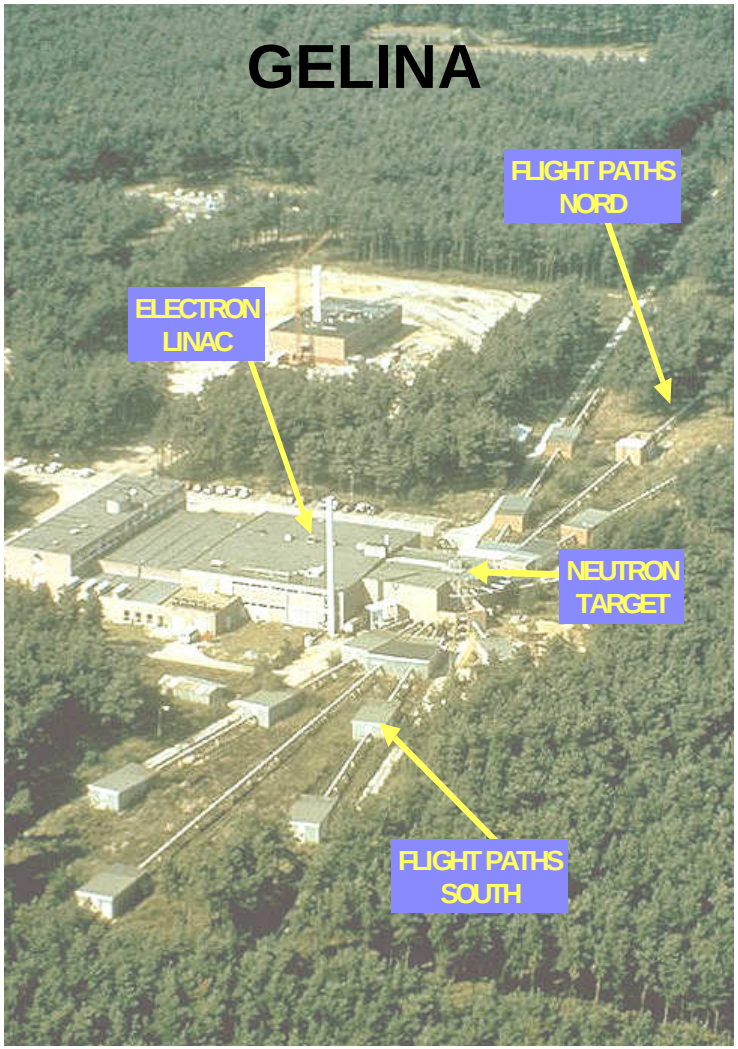
White Neutron Energy Spectrum

TOF $\Leftrightarrow$ E_n

$$E_n = \frac{1}{2} m_n v_n^2 = \frac{1}{2} m_n \left(\frac{L}{t} \right)^2$$

Good time resolution

High neutron flux



Optimum measurement conditions

Intensity

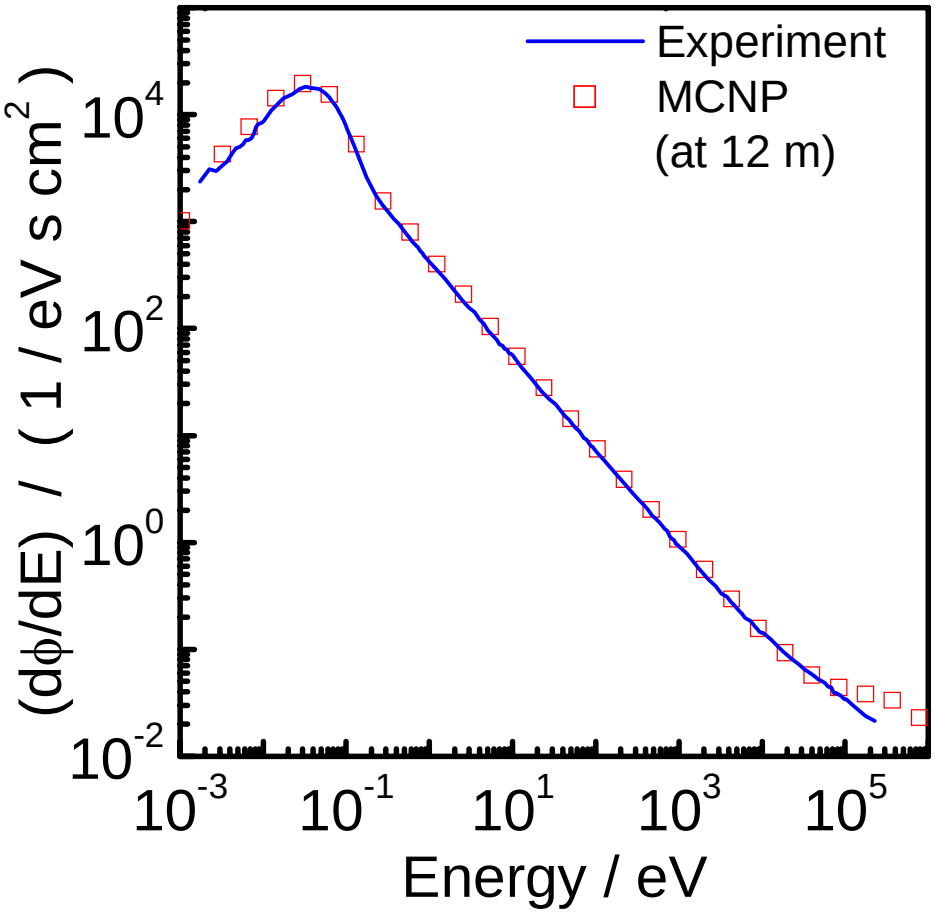
$$\varphi_n(E_n) \propto \frac{1}{L^2}$$

$L\uparrow \quad \Rightarrow \quad \varphi_n \downarrow$

Resolution

$$R_n = \frac{\Delta E_n}{E_n} = 2\sqrt{\frac{\Delta T_n}{T_n} + \frac{\Delta L}{L}}$$

$L\uparrow \Rightarrow R_n \downarrow$



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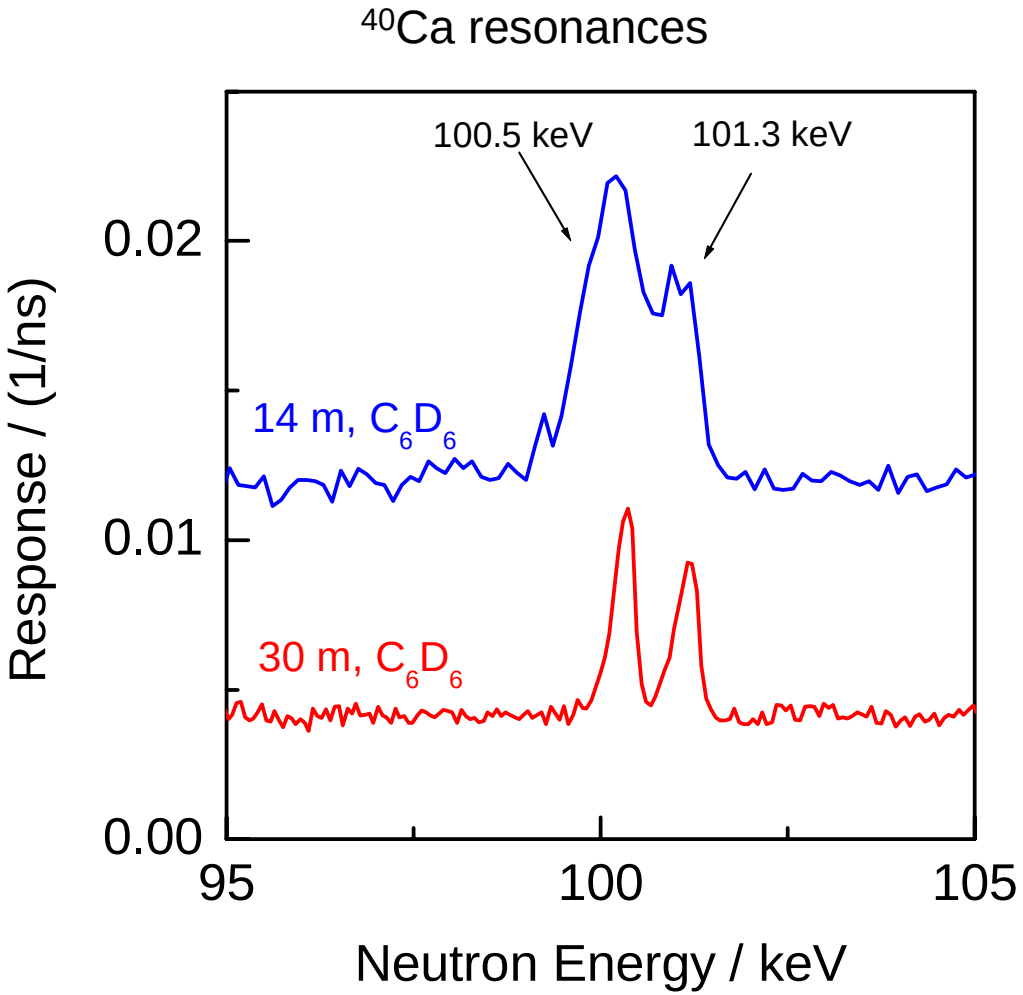
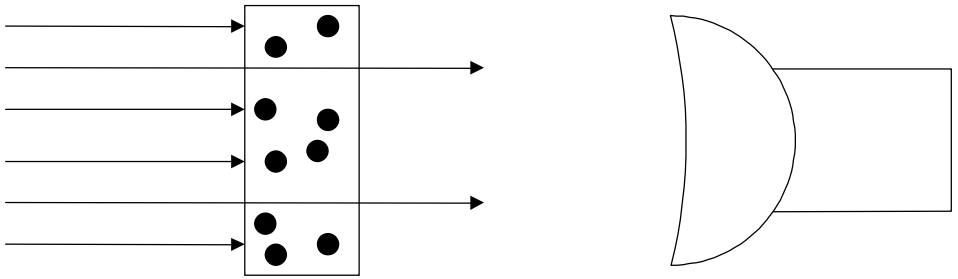


Perego R. et al, MTAA11, June 2004

Detector Response - Transmission (NRT)



$$T = \frac{C_{in}}{C_{out}} \cong e^{-n\sigma_{tot}}$$

no correction for efficiency

no normalisation required

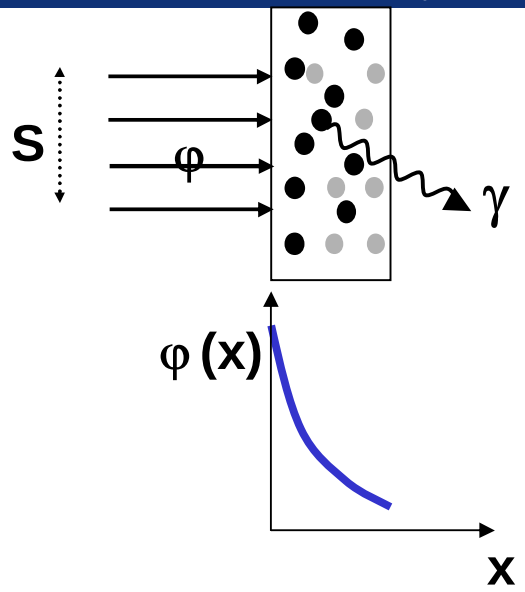
but good collimation required

- **All detected neutrons must have crossed the sample**
- **All neutrons scattered by the sample may not reach the detector**

low sensitivity (exponential + potential scattering)

Detector Response - Capture (NRCA)

$$R_c = \varepsilon_c\, n_x\, \sigma_\gamma\, F\, \varphi\, S$$



ε_c	detection efficiency for capture event
n_x	number of nuclei per cm^2 for nuclide x
σ_c	capture cross section
F	self-shielding factor
S	Effective area
φ	neutron flux

$$F = \left(\frac{1 - e^{-n\sigma_t}}{n\sigma_t} \right)$$

More sensitive but the data analysis is more complicated

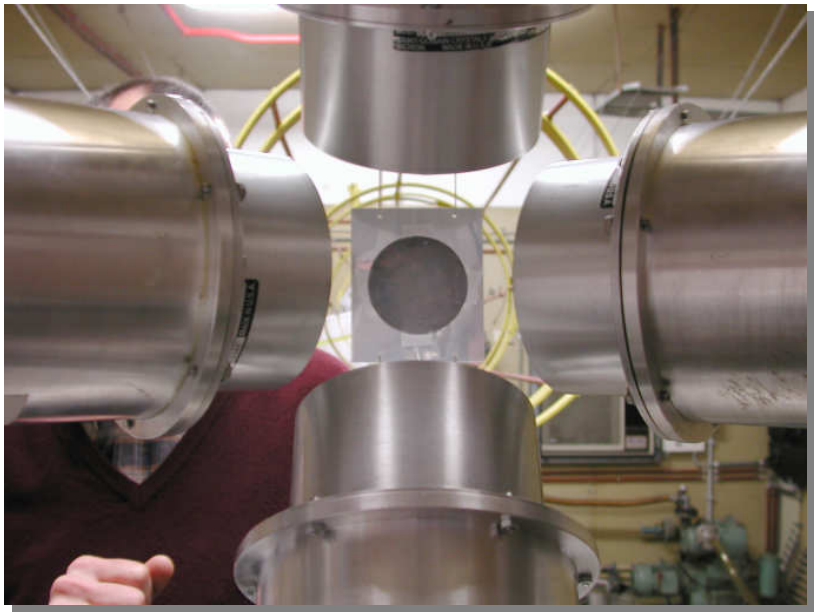
Detectors for NRCA and NRT

NRCA

Gamma Detectors - C_6D_6, YAP

Good time resolution (1 ns)

Low neutron sensitivity

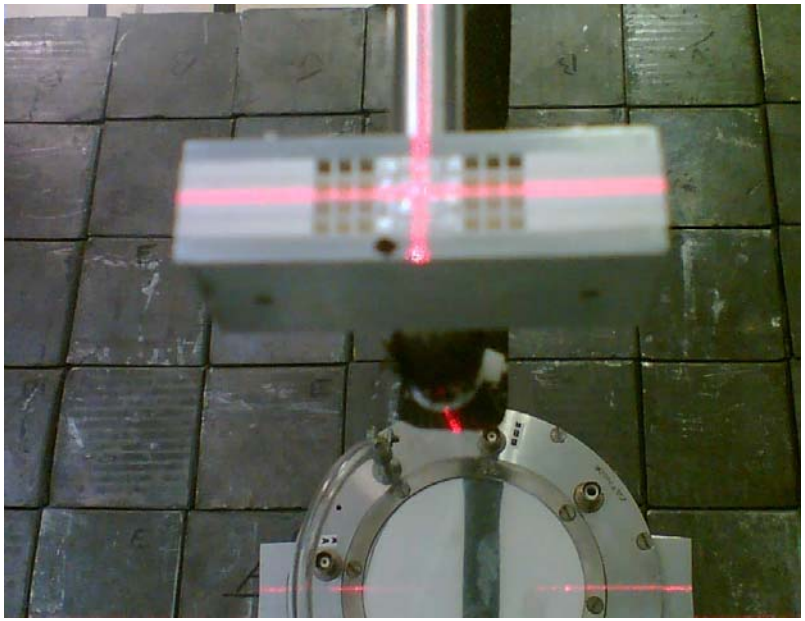


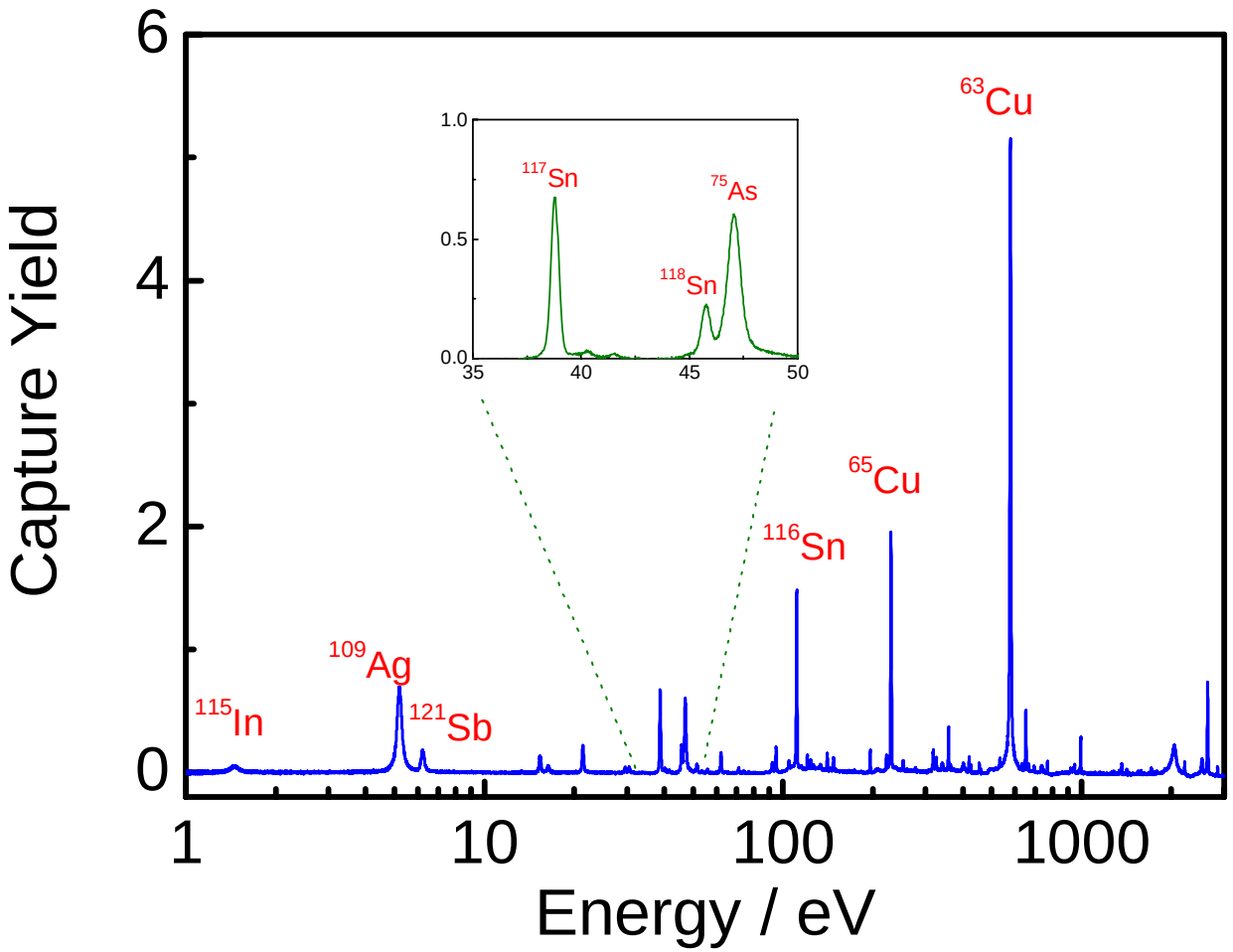
NRT

Neutron Detectors - Li glass NE905

Good time resolution

PSND (developed at CCLRC)





⇒ we can always define a region in the TOF (Energy) - spectrum where the resonance of interest dominates

Traditional approach
Area analysis

$$R_c = \varepsilon_c \, n_x \, \sigma_\gamma \, F \, \varphi \, S \quad \Rightarrow \quad C_c = \varepsilon_c \, n_x \, A_\gamma \, F \, \varphi \, S$$

$\Rightarrow$ only isolated resonances are used

Only valid for relatively weak resonances

neglect neutron scattering in the sample

Calibration

requires representative calibration samples to determine unknown and F

Limited in applications (no complex spectra, samples,…)

Limited use of information contained within the spectrum

However, Very successful (e.g. Archeology, H. Postma)

$$\frac{C_{r1,x}}{C_{r2,y}} = \frac{\varepsilon_{r1,x}}{\varepsilon_{r2,y}} \frac{F_{r1}}{F_{r2}} \frac{A_{\gamma,r1}}{A_{\gamma,r2}} \frac{\varphi_{r1}}{\varphi_{r2}} \frac{n_x}{n_y}$$

$$A_{\gamma,r} = 4.097 \times 10^6 \frac{g\Gamma_n\Gamma_\gamma}{E_r\Gamma} \text{(b)}$$

- **φ neutron flux (φ_{r1}/ φ_{r2}) (only shape required!!)**
  - $\Rightarrow$ Independent measurement by e.g. $^{10}B(n,\alpha)^7Li$
- **Resonance capture area ($A_{\gamma,r1}$ & $A_{\gamma r,2}$)**
  - $\Rightarrow$ From nuclear data libraries
- **ε detection efficiency for capture event (ε_{r1} / ε_{r2})**
  - $\Rightarrow$ By calibration with standard samples (no correction for attenuation in sample)
- **Self - shielding factors (F_{r1} / F_{r2})**
  - $\Rightarrow$ From combination of at least two resonances, with different strength

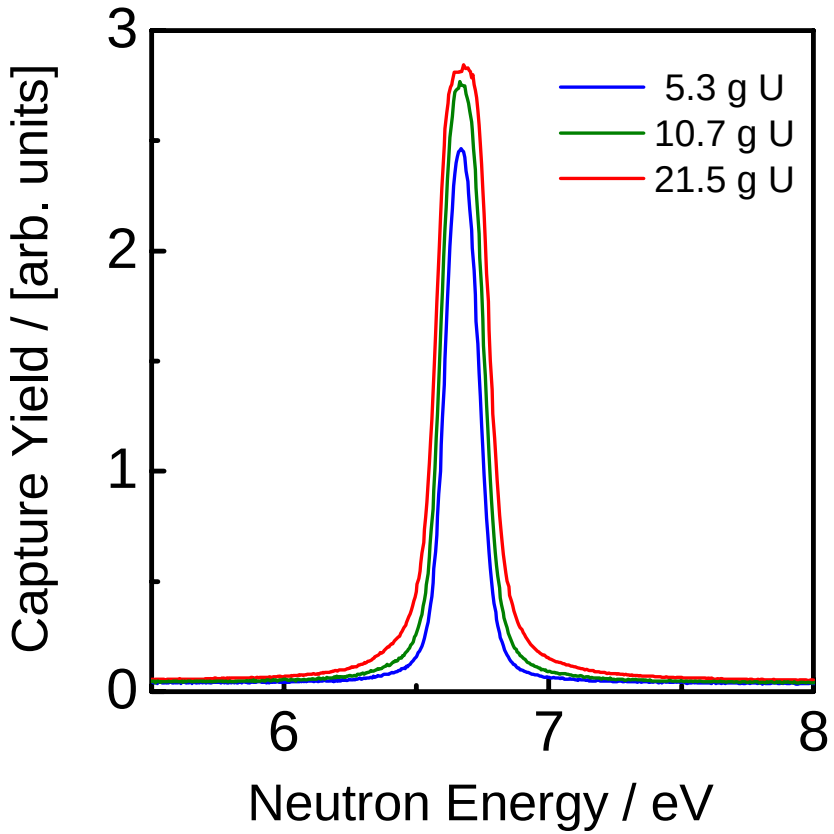
Self – shielding correction

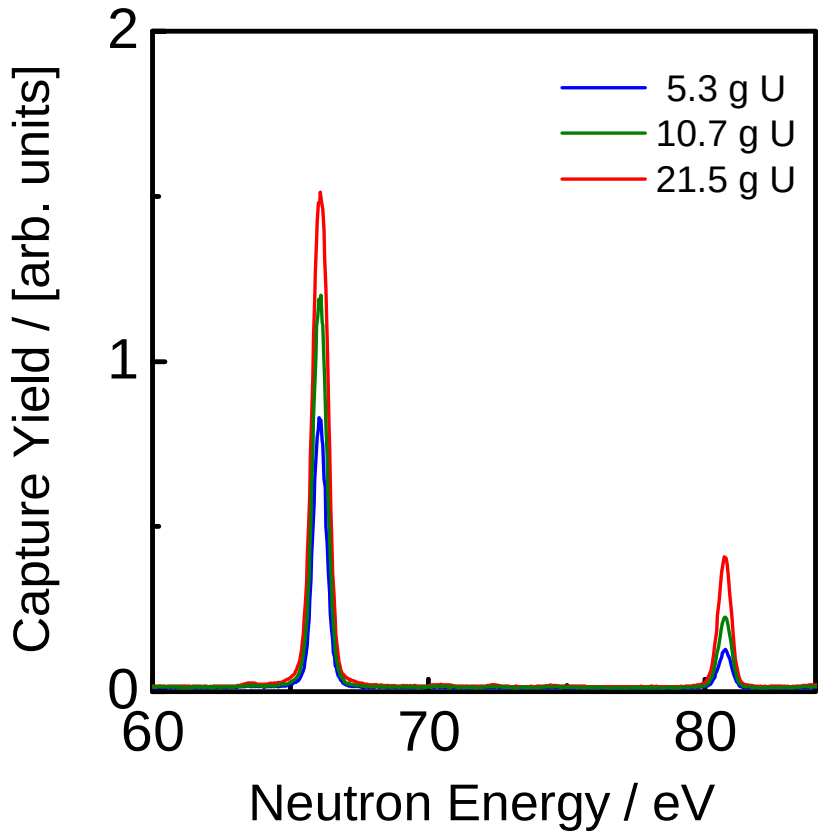
$$F_r = \left(\frac{1 - e^{-n\sigma_t}}{n\sigma_t} \right)$$

$n\sigma_t << 1 \Rightarrow F \cong 1$

for main elements use weaker resonances

for traces use stronger resonances





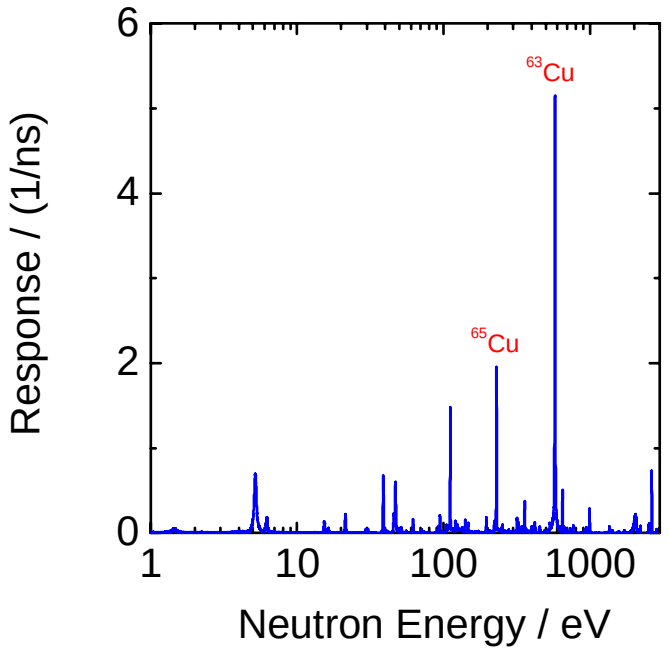
$\Rightarrow$ Thickness (n) from combination of resonances with different strength !!!

F_r from ratio of resonances with different strength

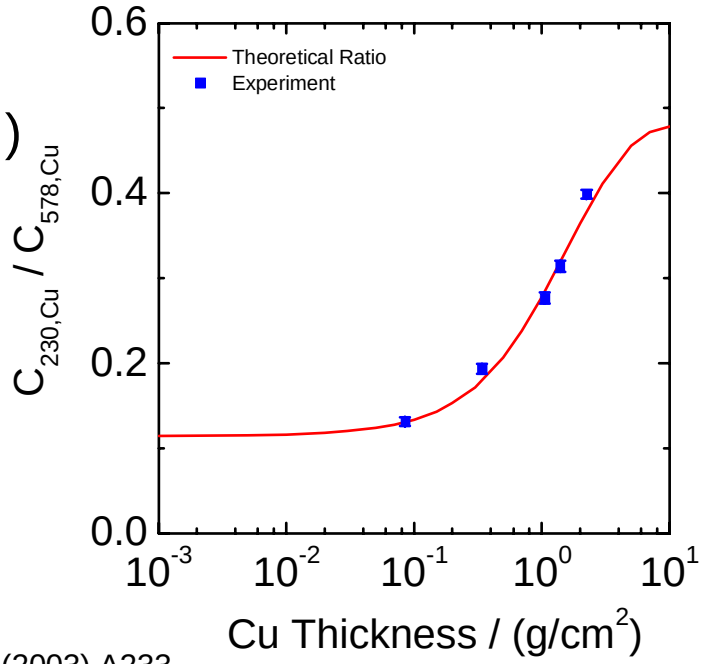
$$\frac{C_{r1,x}}{C_{r2,y}} = \frac{\varepsilon_{r1,x}}{\varepsilon_{r2,y}} \frac{\varphi_{r1}}{\varphi_{r2}} \frac{A_{\gamma,r1}}{A_{\gamma,r2}} \frac{F_{r1}(n_x,\sigma_{t1})}{F_{r2}(n_y,\sigma_{t2})}$$

independent of n

$$F_r = \left(\frac{1 - e^{-n\sigma_t}}{n\sigma_t} \right)$$



$$\frac{C_{r1,x}}{C_{r2,x}} = \frac{1}{K} f(n_x, \sigma_{t,r1}, \sigma_{t,r2})$$



H. Postma et al., Czech. J. Phys., 33 (2003) A233

Verification of self-shielding correction

Sn

39 eV ^{117}Sn

111 eV ^{116}Sn

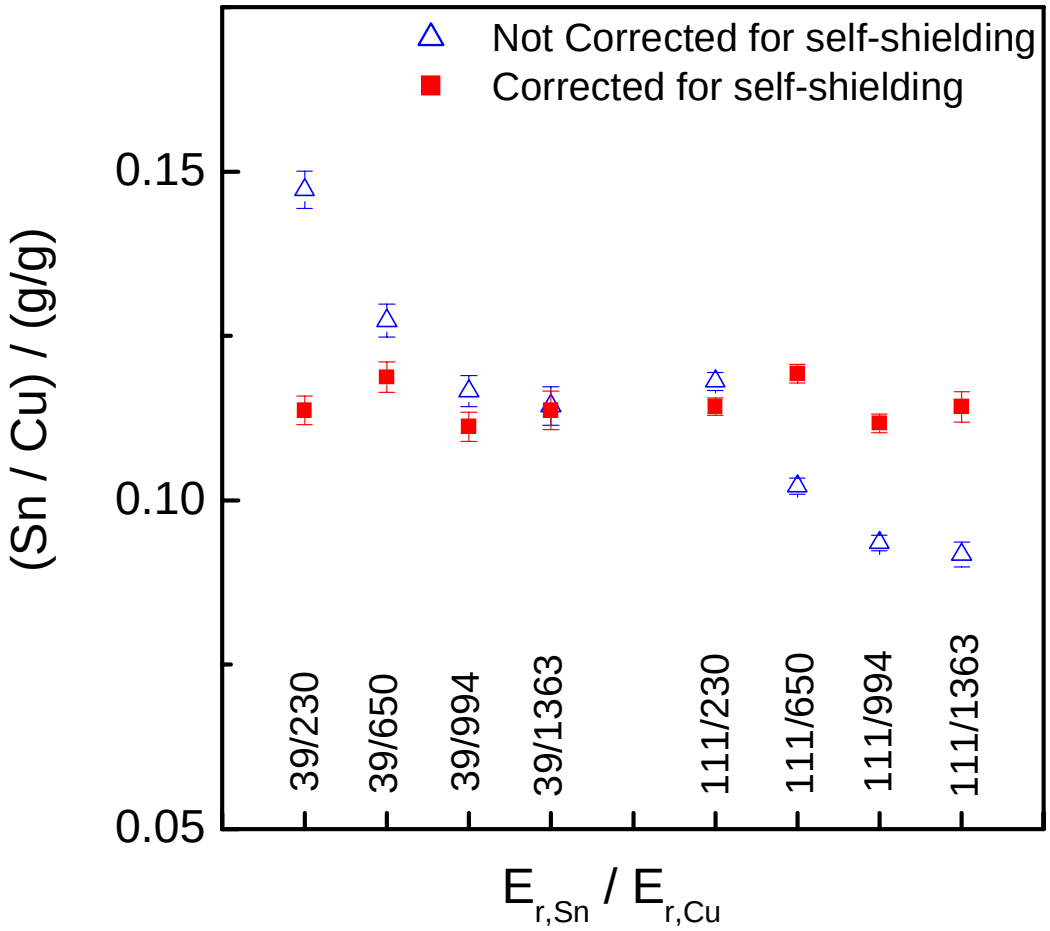
Cu

230 eV ^{65}Cu

650 eV ^{63}Cu

994 eV ^{63}Cu

1363 eV ^{65}Cu



H. Postma et al., MTAA11, 2004

Methodological Approach (REFIT, M. Moxon)

NRCA requires capture yield, Yc

Correction for the shape of the incoming neutron flux

Account for detection efficiency of the gamma event

(PHWT $\Rightarrow$ NIM A 577 (2007) 626-640)

Advantages:

Resonance Shape Analysis (R-matrix) vs area analysis

Correction for self - shielding and multiple scattering

Resolution of TOF - spectrometer and Doppler broadening

The whole energy spectrum can be used to assess the nuclide abundance

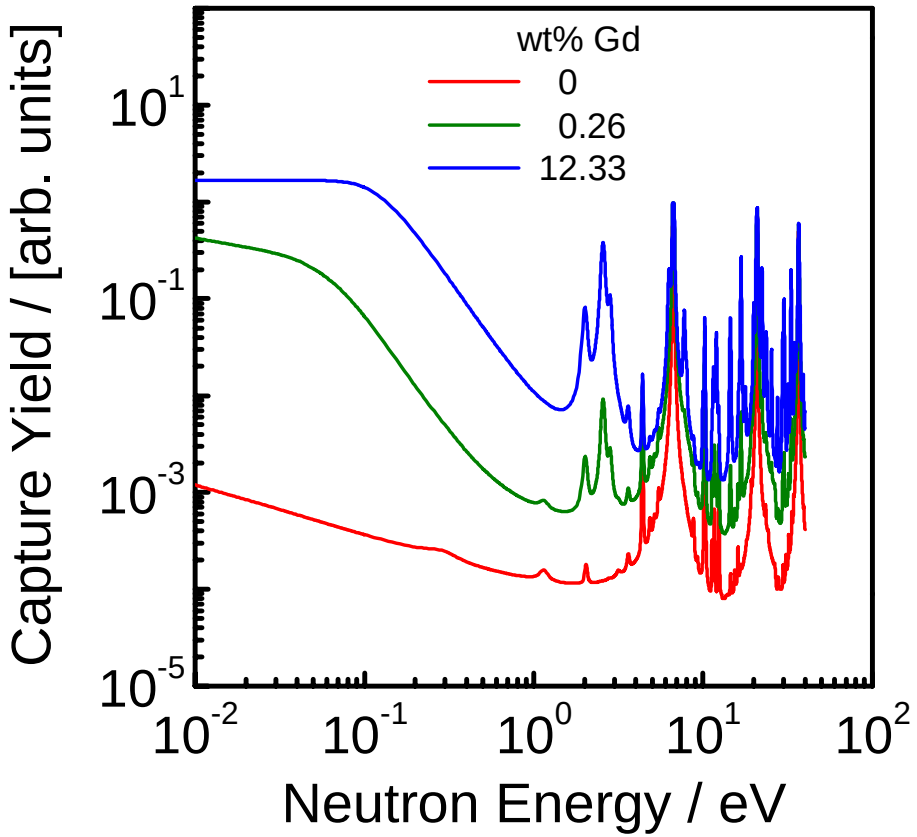
Simultaneous fit of several data sets (both NRT and NRCA)

Easy to handle multi-elemental homogenous samples

G is an analytical model describing the observed capture yield:

$$Y_c = G(R, T, RP, F, \mu, S_x^*, n_x, S_y^*, n_y, ...)$$

- R resolution of spectrometer
- T temperature (Doppler broad.)
- RP nuclear data to deduce σ_c, σ_t
- F self - shielding
- μ multiple scattering
- S_x^* binding energy nucleus x
- n_x atom density of nucleus x



⇒ **n_x, n_y, … : adjustable fitting parameters**

Archaeology

- Zn/Sn, *Cu/Pb* ratio
- *Ancient Charm Project (2D and 3D scanning)*

Trace Analysis

- *P/Ca in bones, Cl/Ca in marbles*

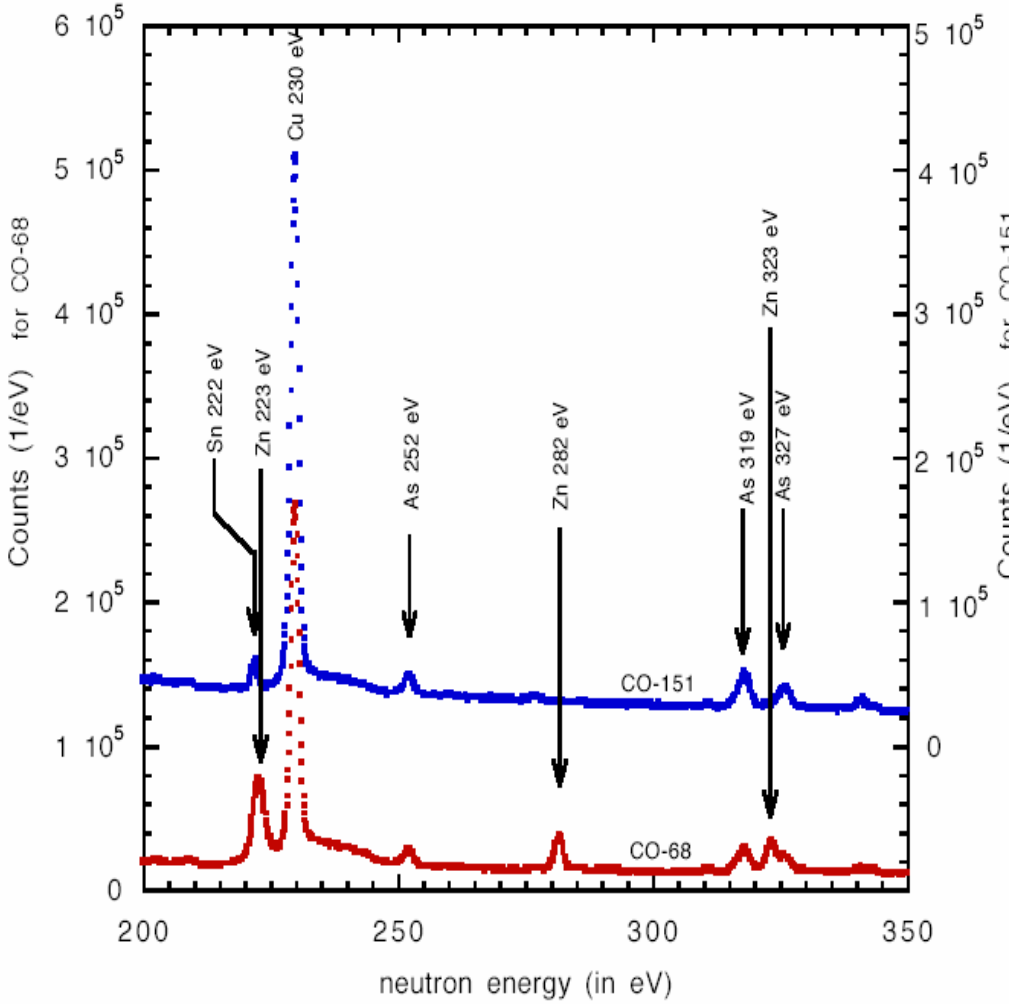
Determination of neutron poison in uranium

- Gd in U

Characterisation of reference materials

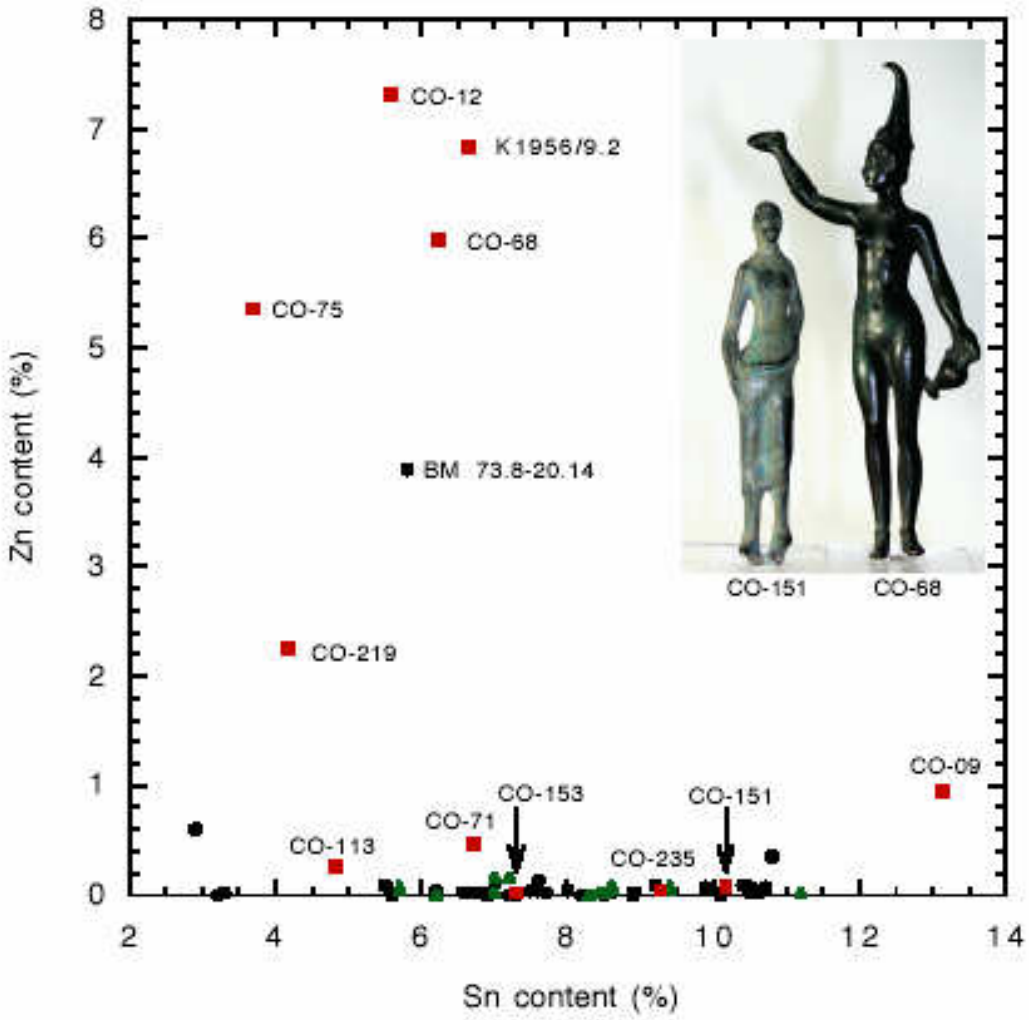
- Ag in Bi, Y_2O_3, Sb in Pb, ^{103}Rh, Pb_2I

Determination of the composition of special alloys



Postma H., Schillebeeckx P. and Halbertsma R.B., Archaeometry 46 (2004) 365

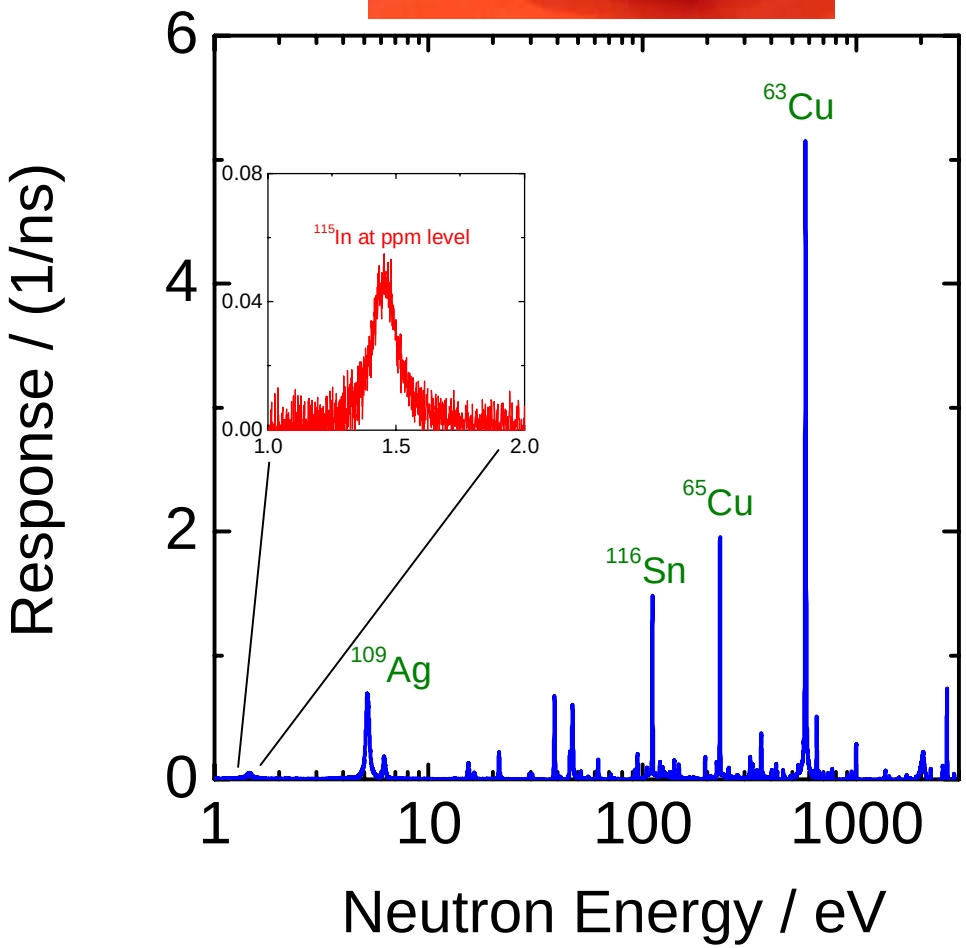
Archaeology: Authenticity of Etruscan artefacts



Archaeology NRCA of Geistingen Axes

Cat Nr.	Find spot	Element									
		Cu	Sn	As	Sb	Ag	Fe	Ni	Co	In	Pb
B550	Maastricht	94.30	0.06	0.67	1.63	0.54				5.1 ppm	< 2
B551	Maastricht	93.38	0.64	0.91	2.14	0.56				14.8 ppm	< 2
K787	Kleve	93.01	0.98	1.64	3.02	1.36				o.o.r	< 2
K788	Kleve	86.24	6.54	2.32	2.66	2.10			0.15	o.o.r	< 2
B562	Nijmegen	84.51	9.07	1.01	2.10	2.21	0.61		0.49	o.o.r	< 2
B557	Vierlingsbeek	80.93	12.72	0.06	0.01	0.04	6.20			o.o.r	< 2

Postma H. et al., Achaeologia protohistorica XIII (2005) 41-46



Element	Fractions (%)		Isotope	Resonance (eV)
Cu	77.76	(0.11)		
			^{63}Cu	579.0
			^{65}Cu	230.0
Sn	20.85	(0.10)		
			^{112}Sn	94.8
			^{116}Sn	111.2
			^{117}Sn	38.8
			^{118}Sn	45.7
			^{119}Sn	222.6
			^{120}Sn	427.5
			^{122}Sn	1756.0
			^{124}Sn	62.0
As	0.34	(0.01)		
			^{75}As	47.0
Sb	0.196	(0.021)		
			^{121}Sb	6.24
			^{123}Sb	21.4
Ag	0.090	(0.01)		
			^{107}Ag	16.3
			^{109}Ag	5.2
Fe	0.770	(0.09)		
			^{56}Fe	1147.4
In	0.0061	(0.0003)		
			^{115}In	1.46

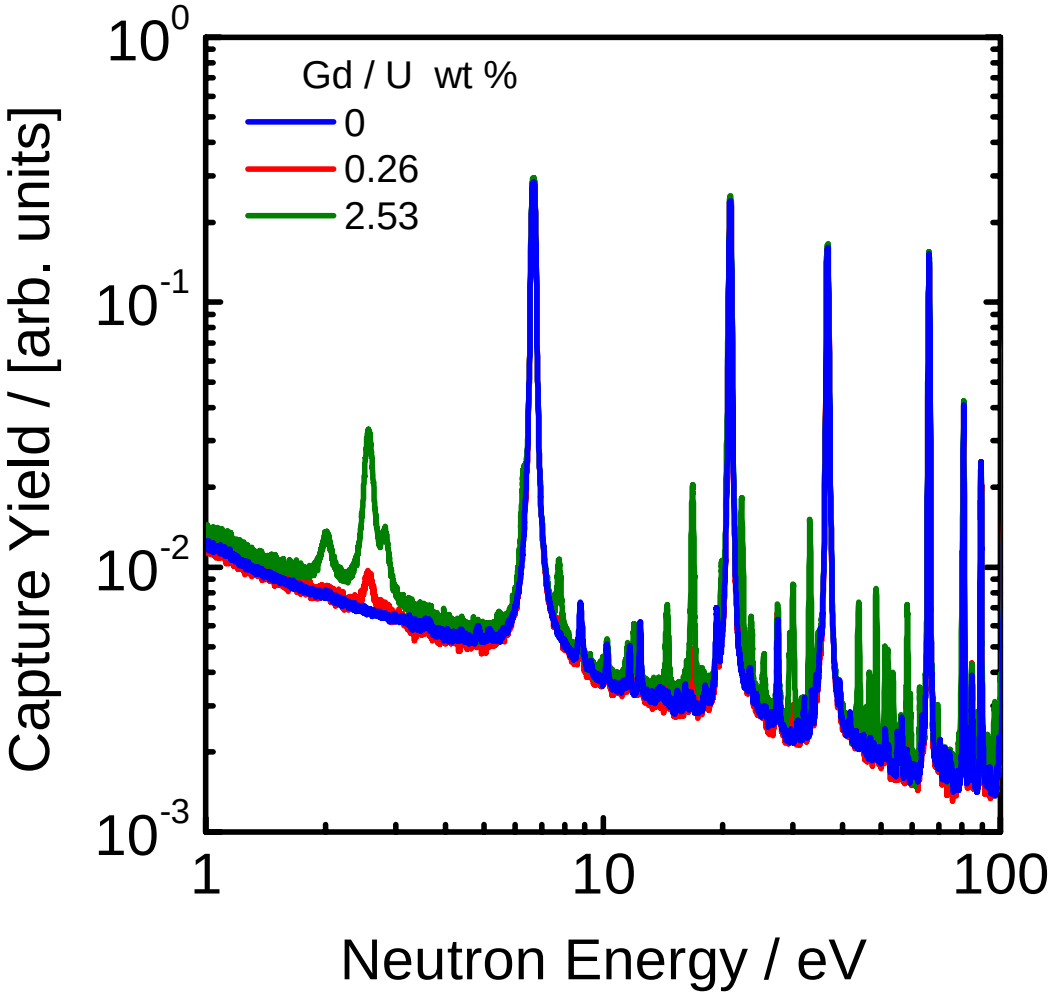
m_{NRCA} = 13.0 (0.5) g

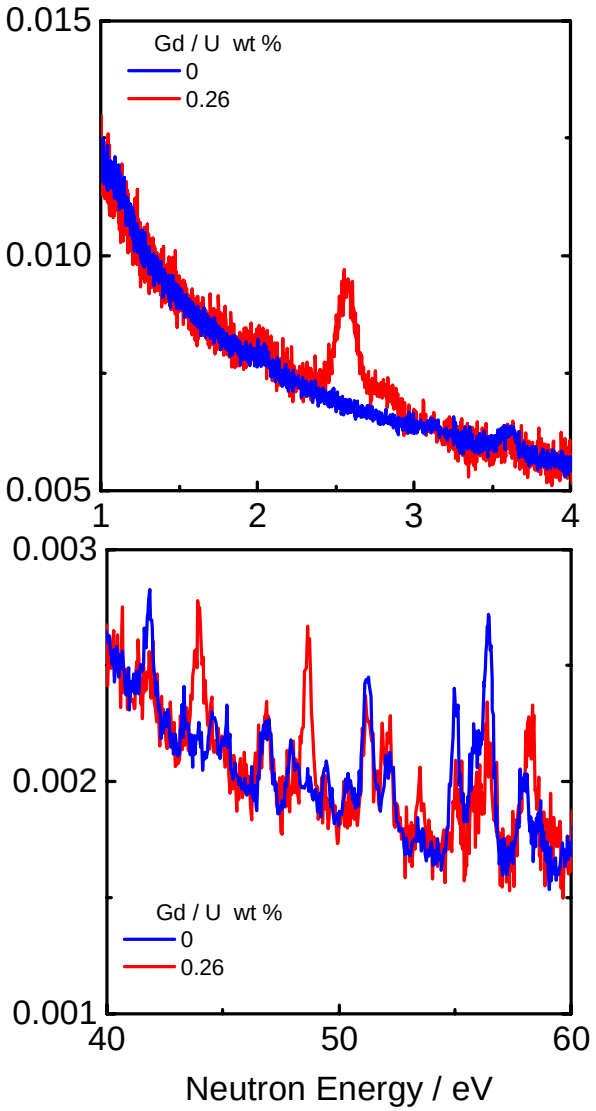
m_{weigth} = 13.25 g

Postma H. et al., Int. Symposium on Archaeometry, Amsterdam, April 2002

NRCA: Neutron poison in U samples

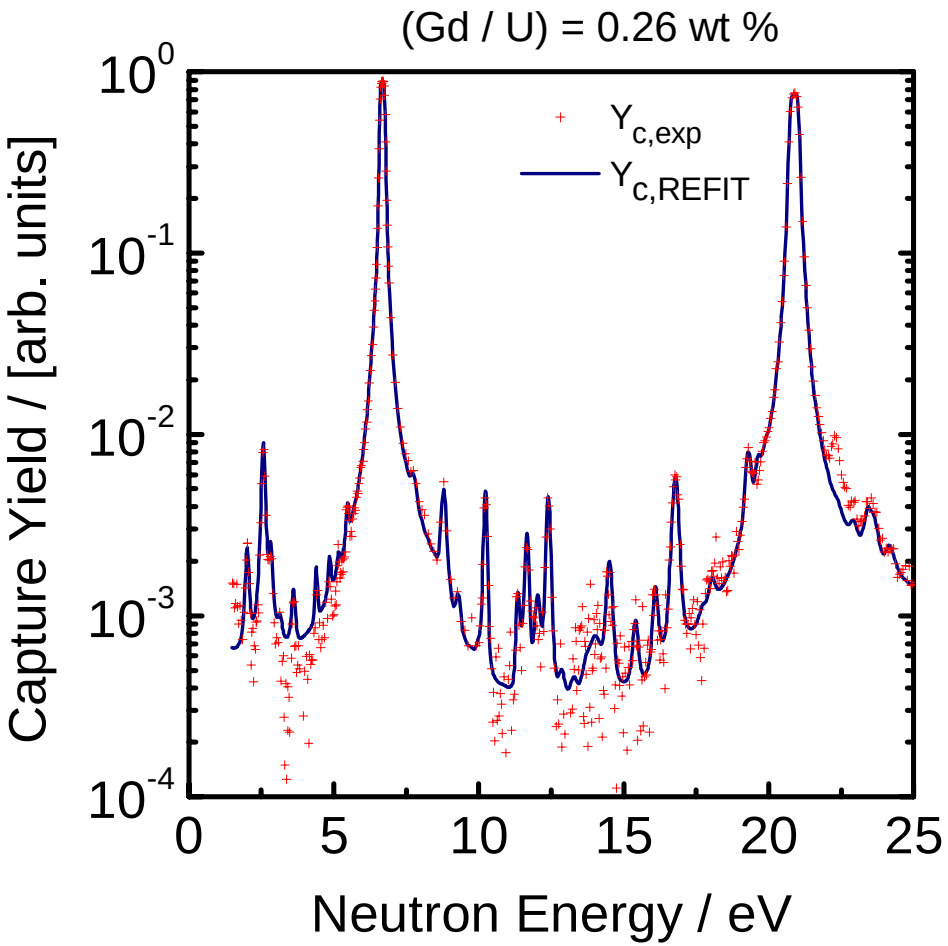
$^{dep}U_3O_8$ + $^{nat}Gd_2O_3$ powder

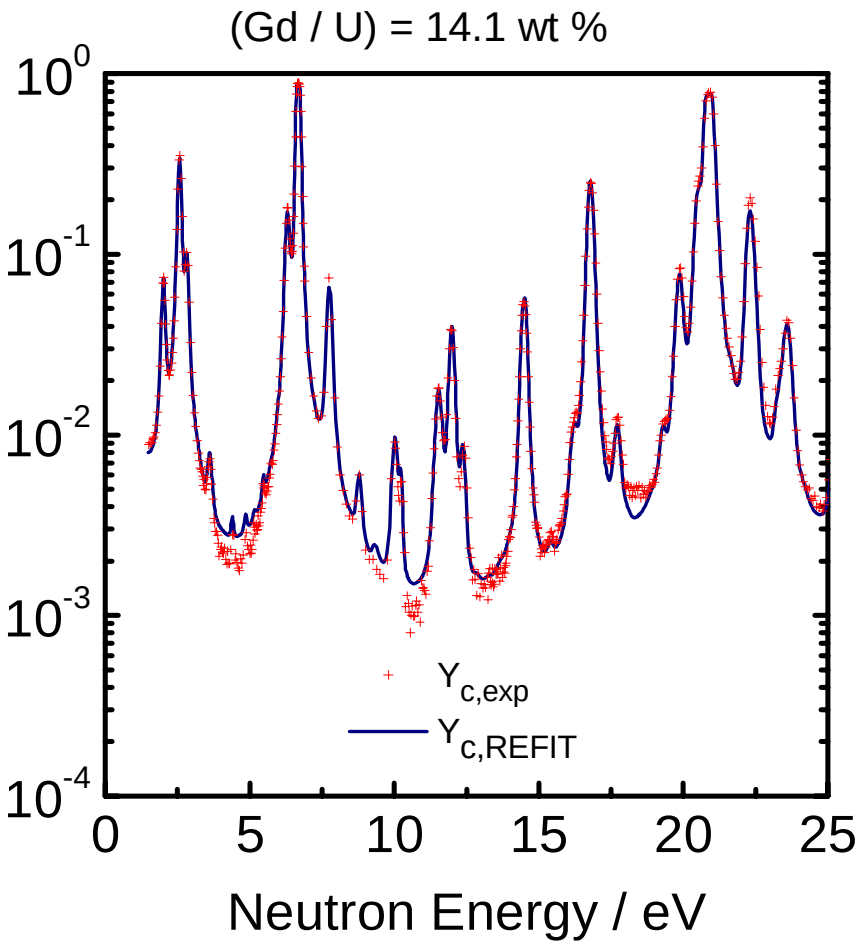




Determination of Gd in U - fit

NRCA: Neutron poison in U samples





Determination of Gd in U - results

NRCA: Neutron poison in U samples

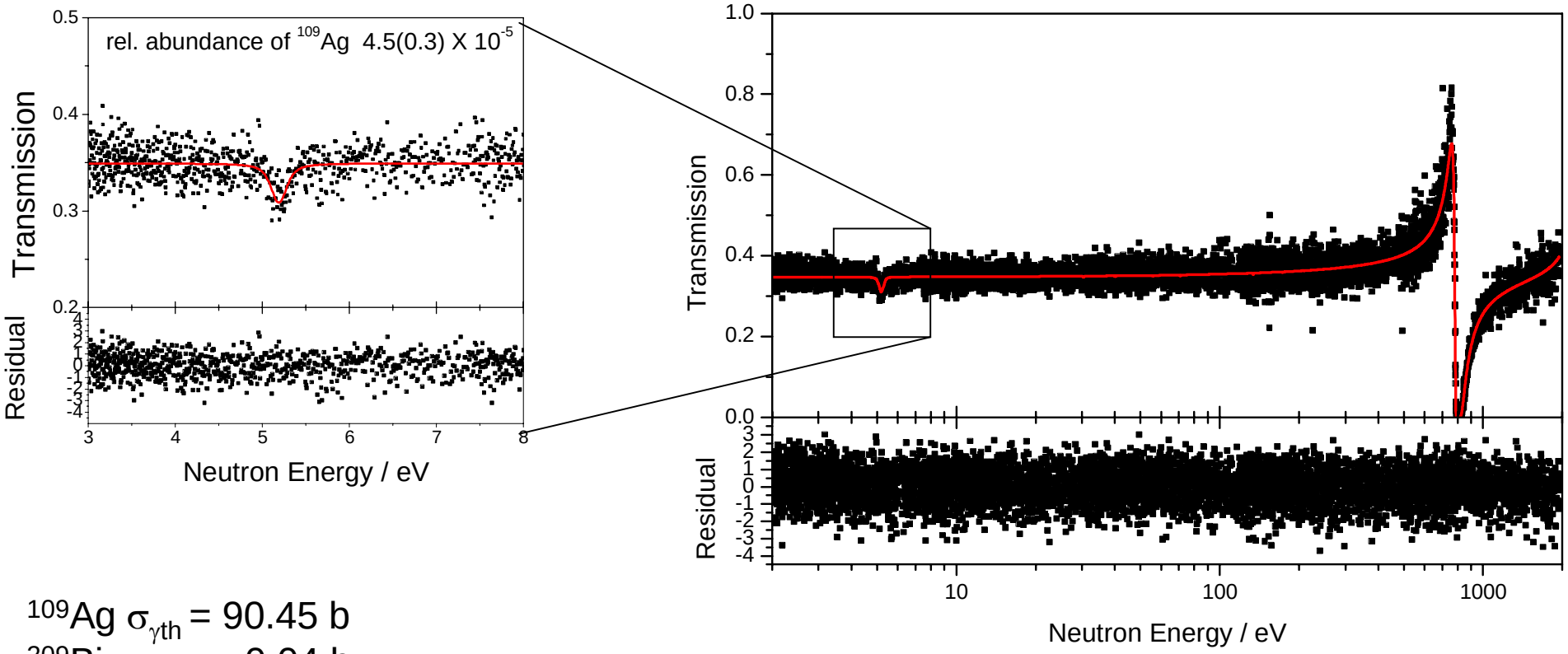
$$^{dep}U_3O_8 + {}^{nat}Gd_2O_3$$

U / g	Gd / g	n(^{155}Gd) / n(^{238}U)		n(^{157}Gd) / n(^{238}U)	
		declared abund.	NRCA	declared abund.	NRCA
20.988	0.0536	$5.77\ 10^{-4}$	$(5.76 \pm 0.04)\ 10^{-4}$	$6.10\ 10^{-4}$	$(6.59 \pm 0.07)\ 10^{-4}$
20.608	0.5206	$5.71\ 10^{-3}$	$(5.73 \pm 0.01)\ 10^{-3}$	$6.03\ 10^{-4}$	$(6.53 \pm 0.02)\ 10^{-3}$
18.656	2.6240	$3.13\ 10^{-2}$	$(3.14 \pm 0.01)\ 10^{-2}$	$3.36\ 10^{-2}$	$(3.51 \pm 0.03)\ 10^{-2}$

$\Rightarrow$ nuclear data ^{157}Gd

Ag in bismuth sample (NRT, S. Kopecky)

NRT for sample characterization



^{109}Ag $\sigma_{\gamma th}$ = 90.45 b

^{209}Bi $\sigma_{\gamma th}$ = 0.04 b

Without accounting for Ag (107 and 109) the $\sigma_{\gamma th}$ would overestimated by 15%

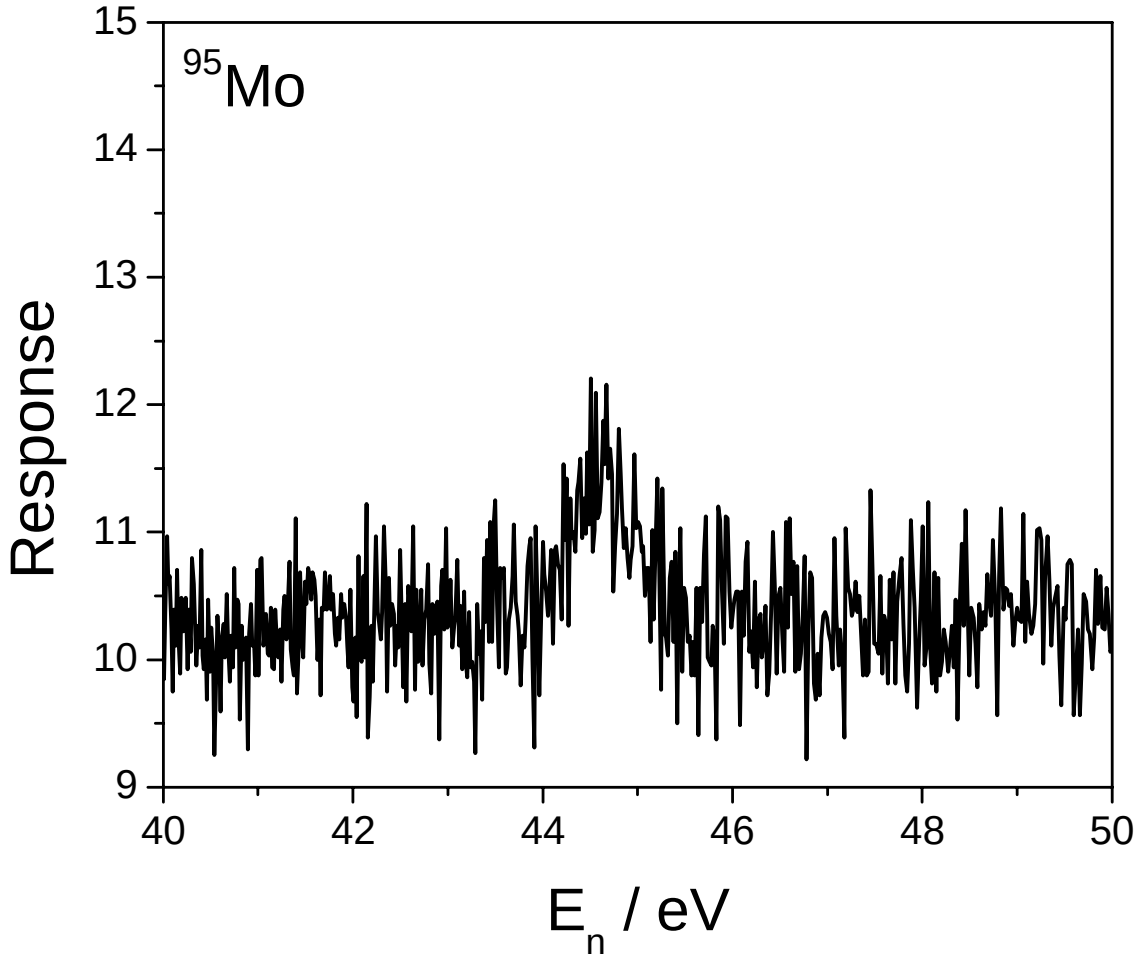
Impurities in Y_2O_3 matrix for AmO_2 samples

NRCA for sample characterization

Y_2O_3 matrix important to obtain uniform samples

Impurities found in Y_2O_3 matrix

⇒



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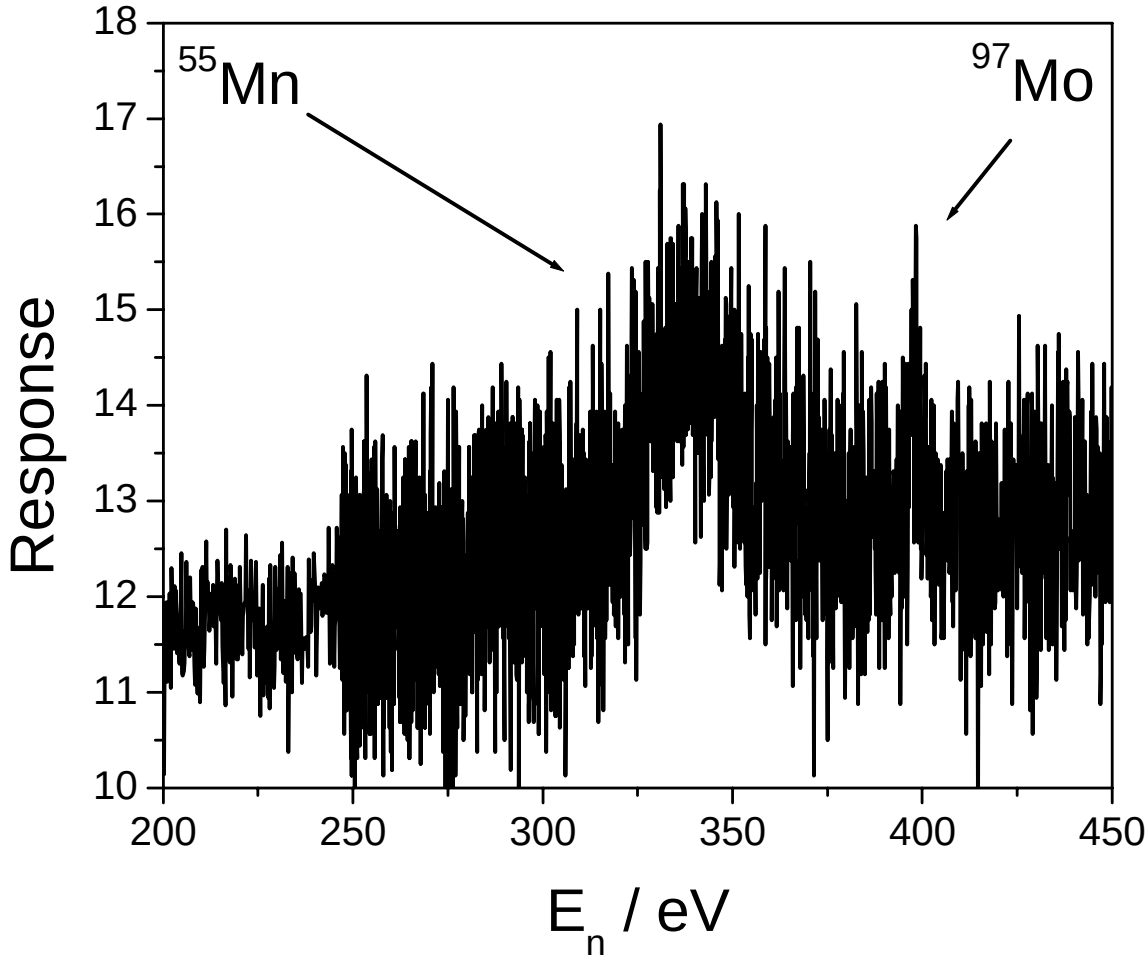

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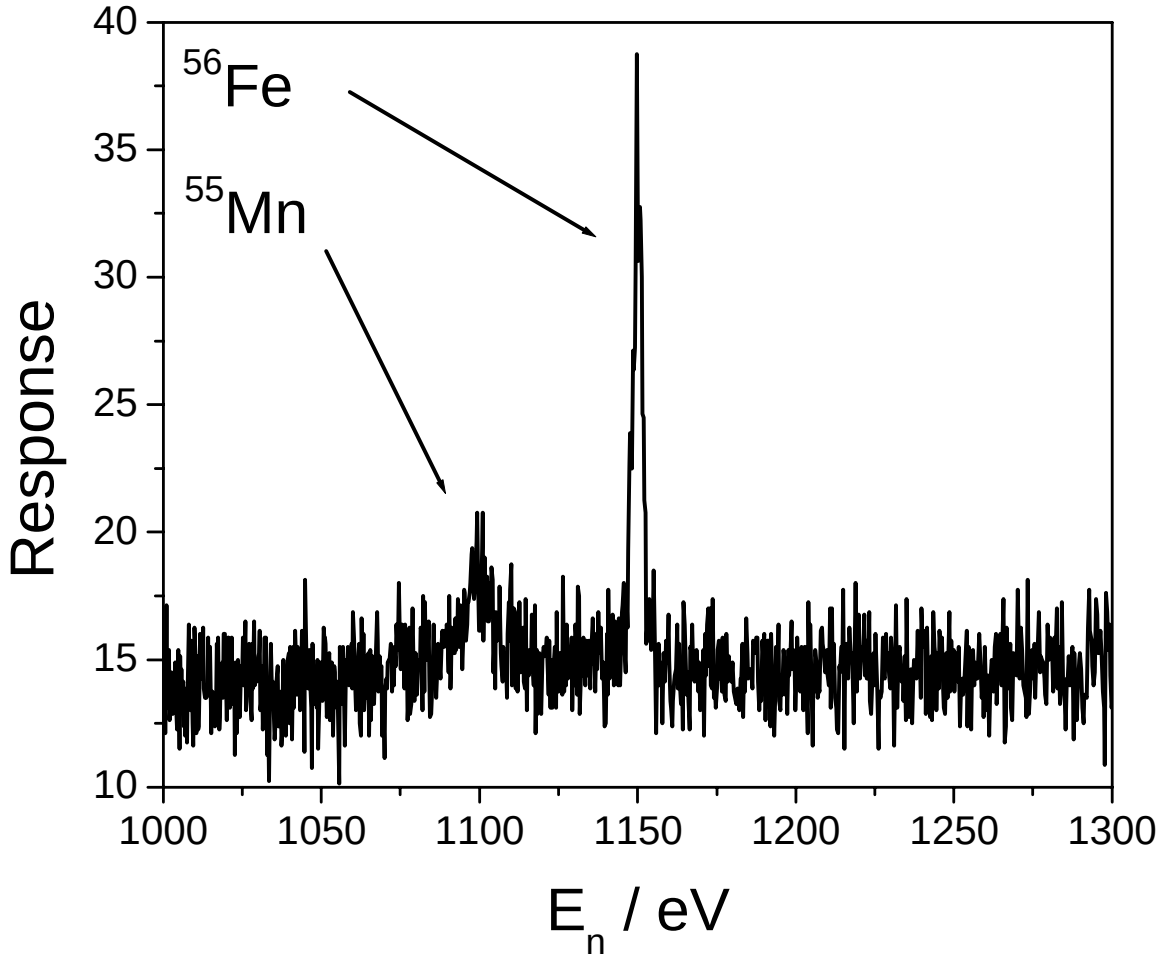

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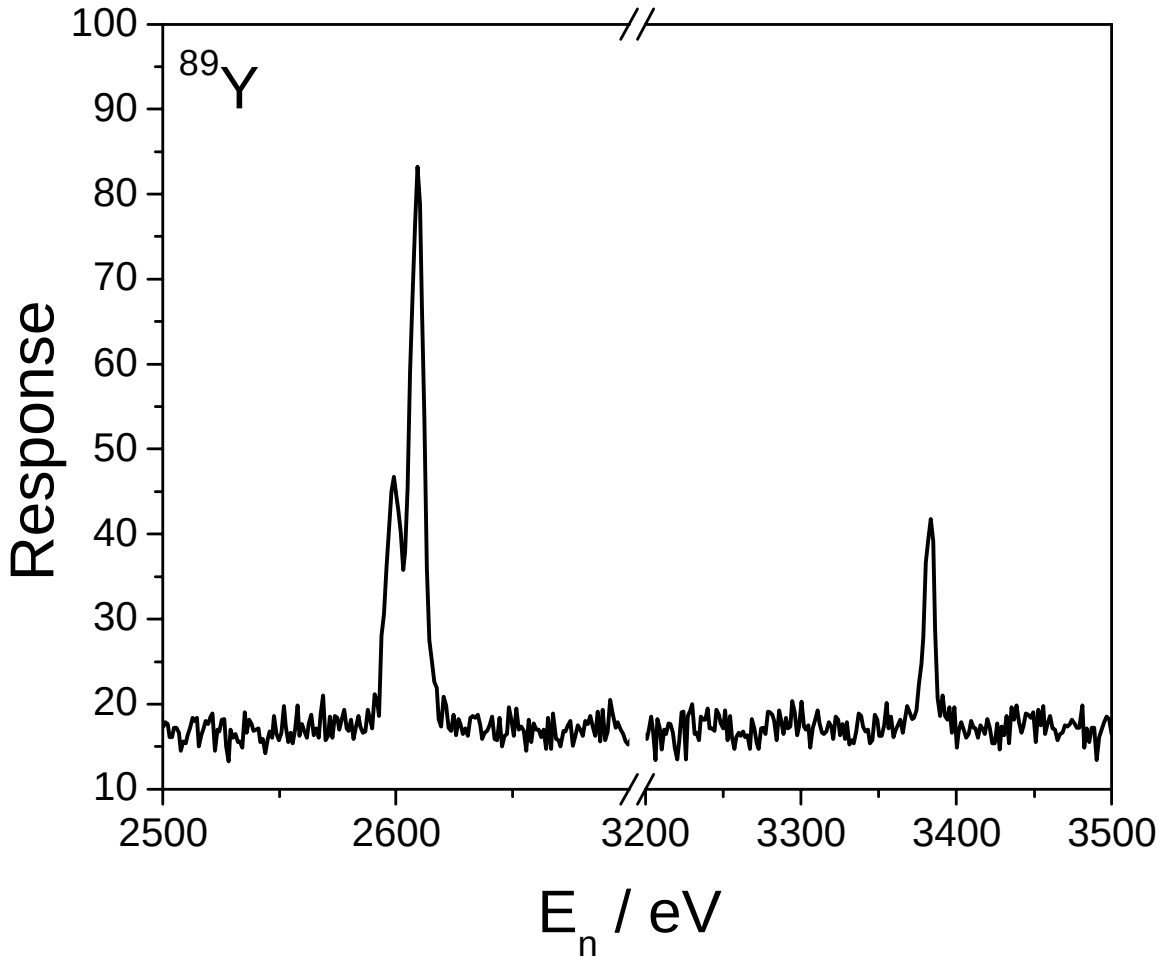

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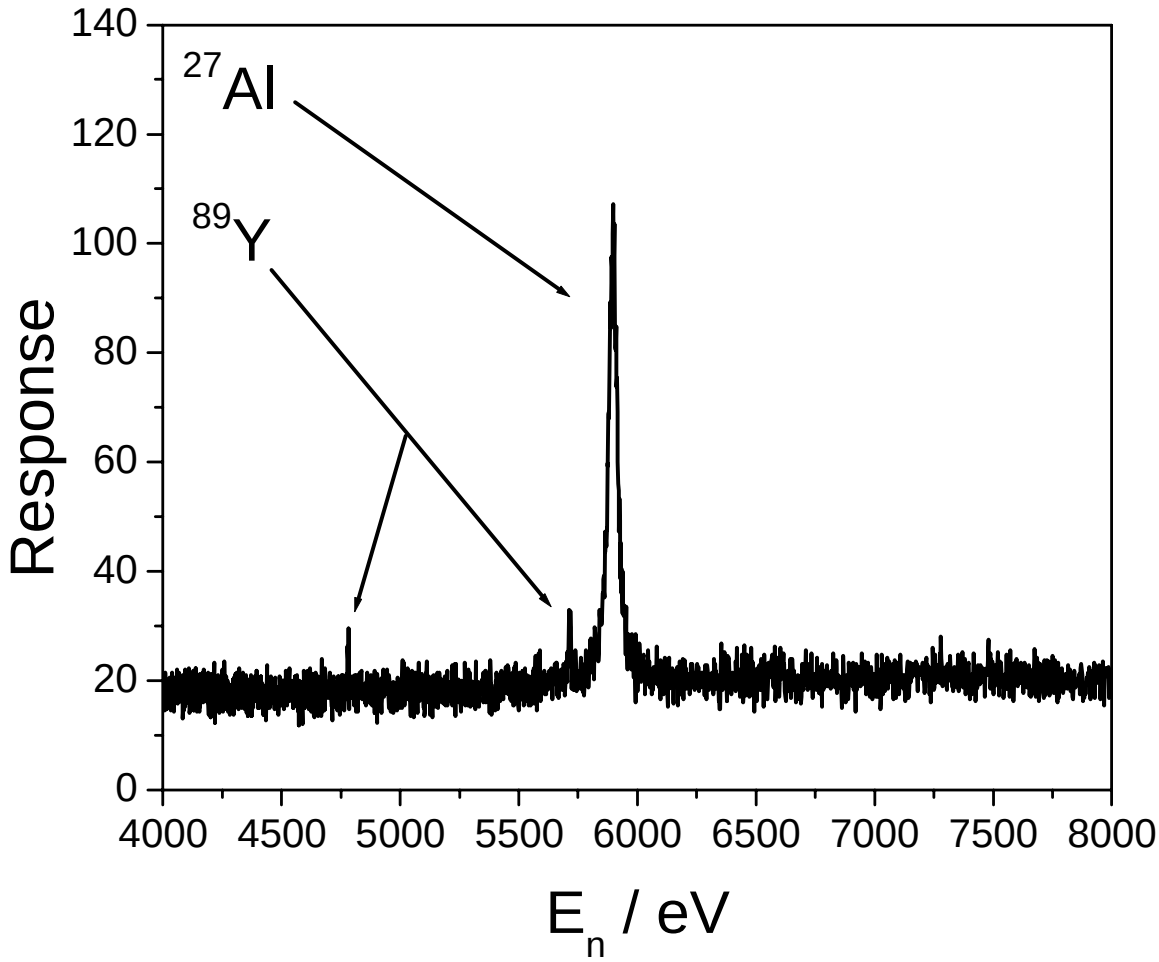
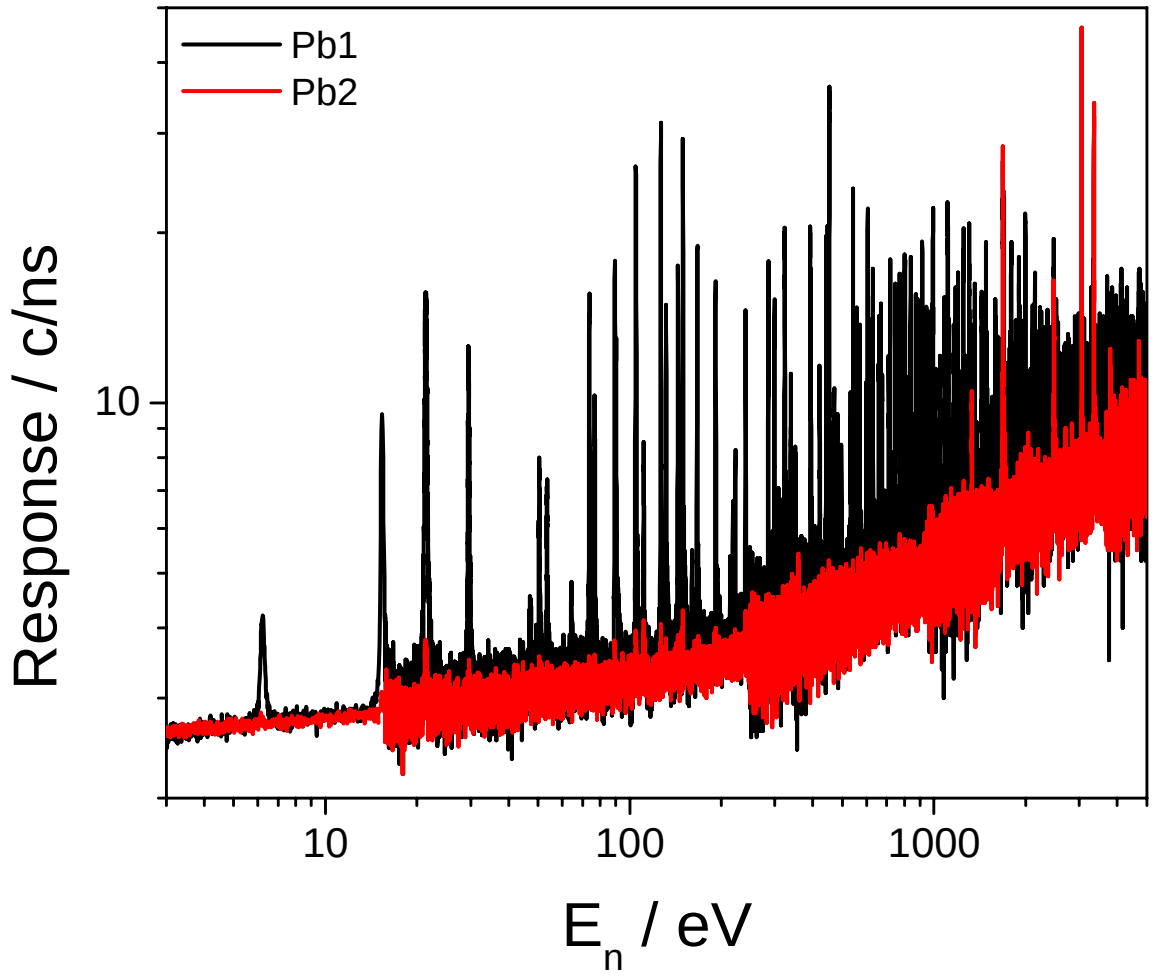


Pb shielding for

- Am sample
- Detectors

Sb-free lead is required to measure Am resonances



Characterisation of Reference Materials
^{103}Rh metal disc

	Natural abundance (wt %)	Relative Amount (wt %)	
^{103}Rh	100	99.5137	
^{181}Ta	99.988	0.0337	(0.0029)
^{191}Ir	37.3	0.0870	(0.0033)
^{193}Ir	62.7	0.1478	(0.0076)
^{182}W	26.3	0.0552	(0.0027)
^{183}W	14.3	0.0302	(0.0028)
^{186}W	28.6	0.0613	(0.0025)
^{197}Au	100	0.0059	(0.0011)

Impurities contribute for 0.5 % to the observed count rate in the thermal energy region

^{103}Rh(n_{th},γ) cross section is requested with an accuracy < 2%

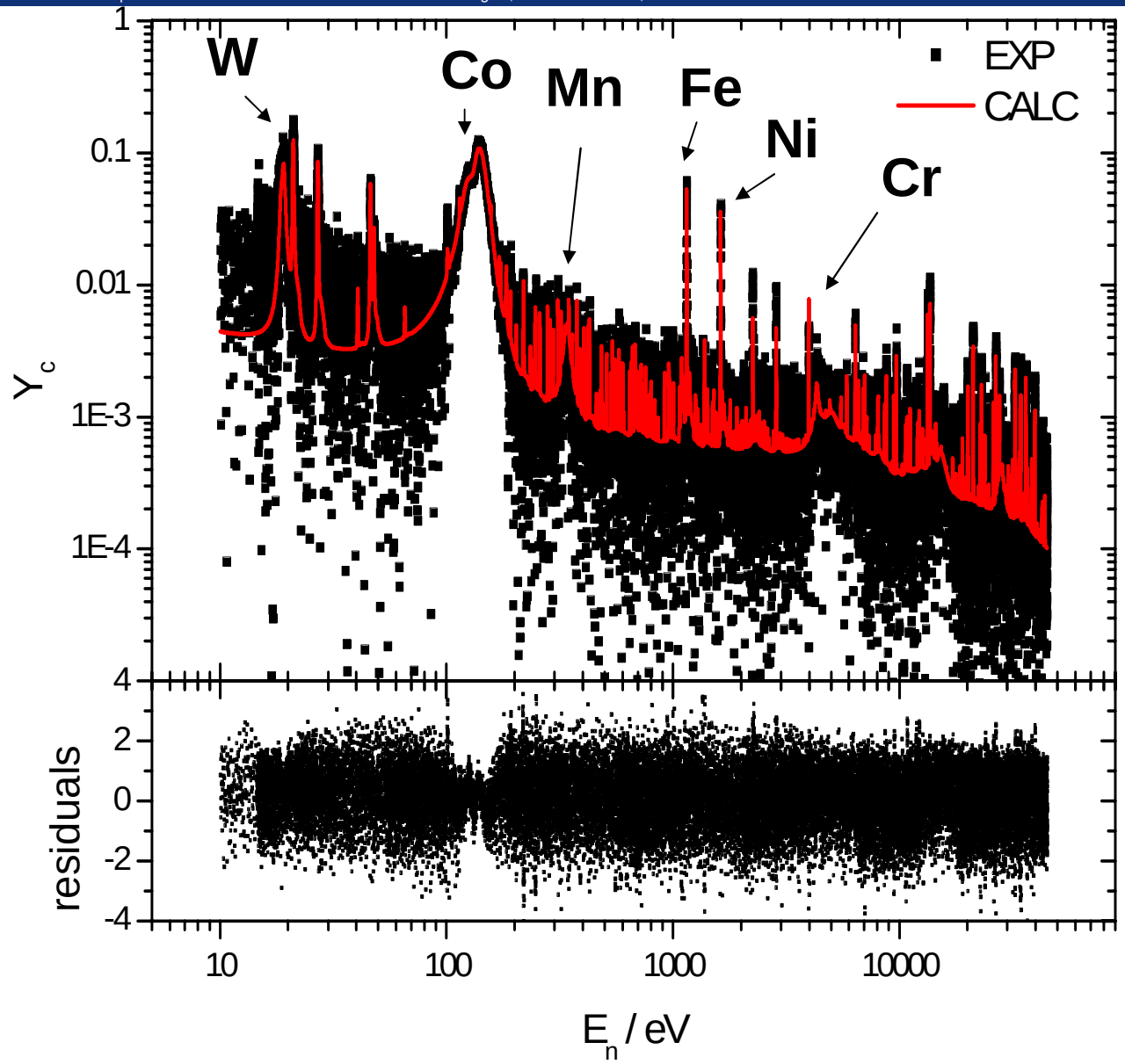
Postma H. et al.,, ND2004, Santa Fe, September 2004

Characterisation of Reference Materials PbI_2 samples from reprocessed fuel

Elememt		NRCA		ICP-MS		(I)NAA	
Iodine	Total	20.5	(0.8)	19.9	(0.5)		
	^{127}I	3.4	(0.1)	3.4	(0.1)	3.4	(0.1)
	^{129}I	17.1	(0.8)	16.5	(0.5)		
Lead	Total	53.5	(3.0)	59.5	(0.2)	51.1	(1.8)
	^{204}Pb	0.8					
	^{206}Pb	12.8	(0.5)				
	^{207}Pb	12.1	(0.3)				
	^{208}Pb	27.8	(3.0)				
Oxygen		15.2	(0.8)	14.5	(1.5)		
Sulfur		6.2	(0.4)				

Noguere G., NIMA 575 (2007) 476-488

NRCA: special alloys composition



% w	Z
0.738	Mn
4.36	W
24.38	Cr
20.77	Fe
33.6	Ni
14.27	Co
1.13	Si

Nuclear Data for NRA - Examples

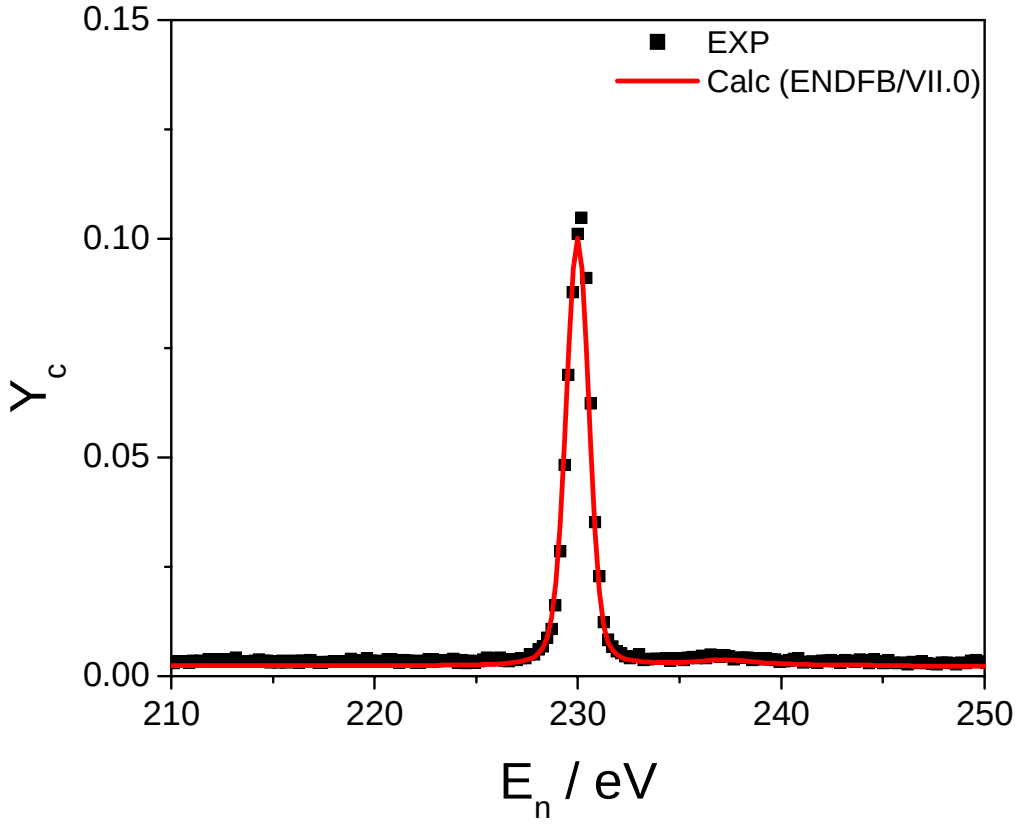
^{nat}Cu

Important/Abundant element in archeological objects

Resonance Parameters

E, Γ_n for 578 eV resonance are wrong in ENDFB.VII, JEFF, JENDL

Missing Resonances too



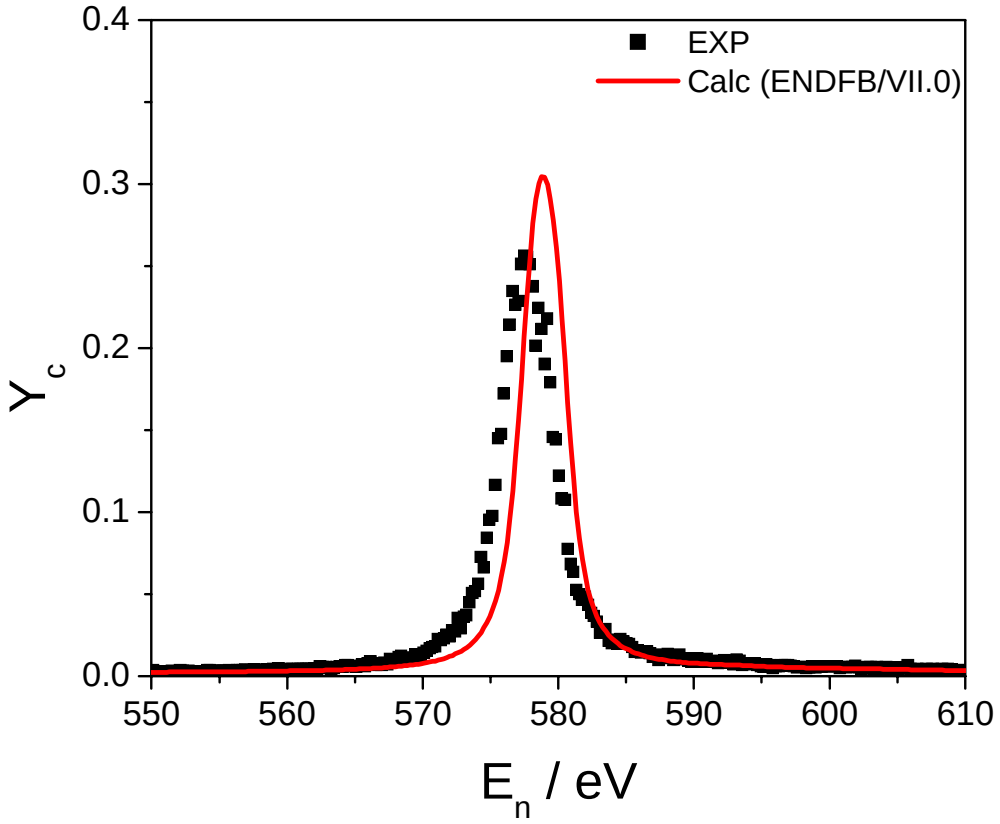
^{nat}Cu

Important/Abundant element in archeological objects

Resonance Parameters

E, Γ_n for 578 eV resonance are wrong in ENDFB.VII, JEFF, JENDL

Missing Resonances too



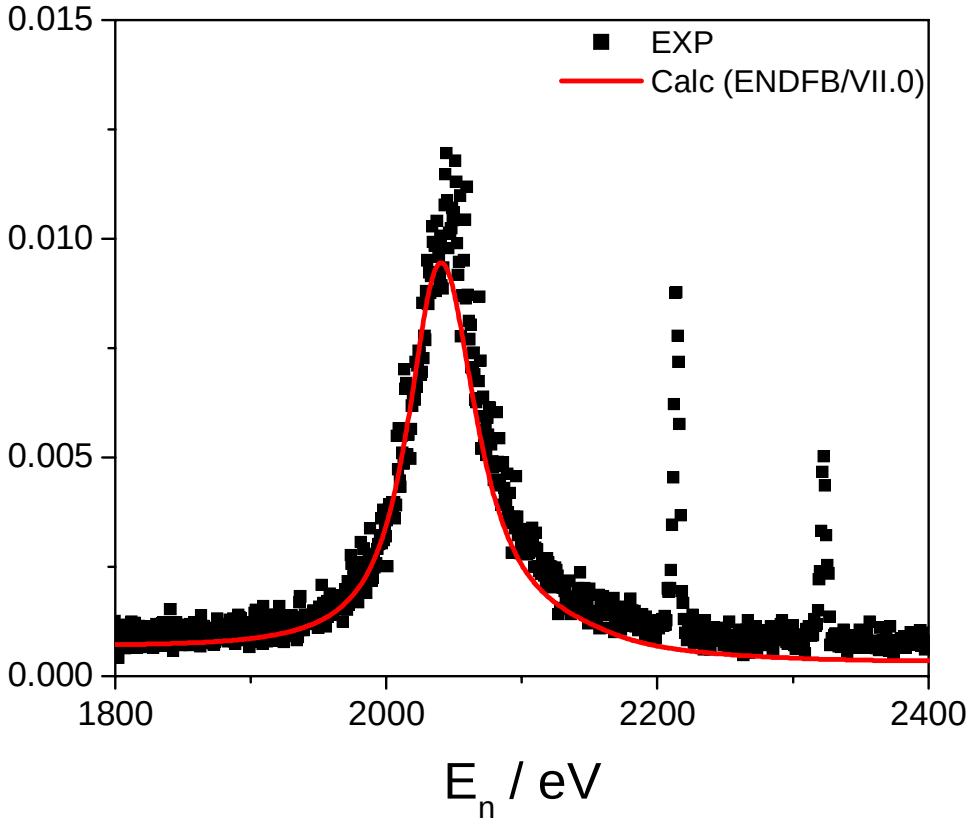
Nuclear Data for NRA - Examples

^{nat}Cu

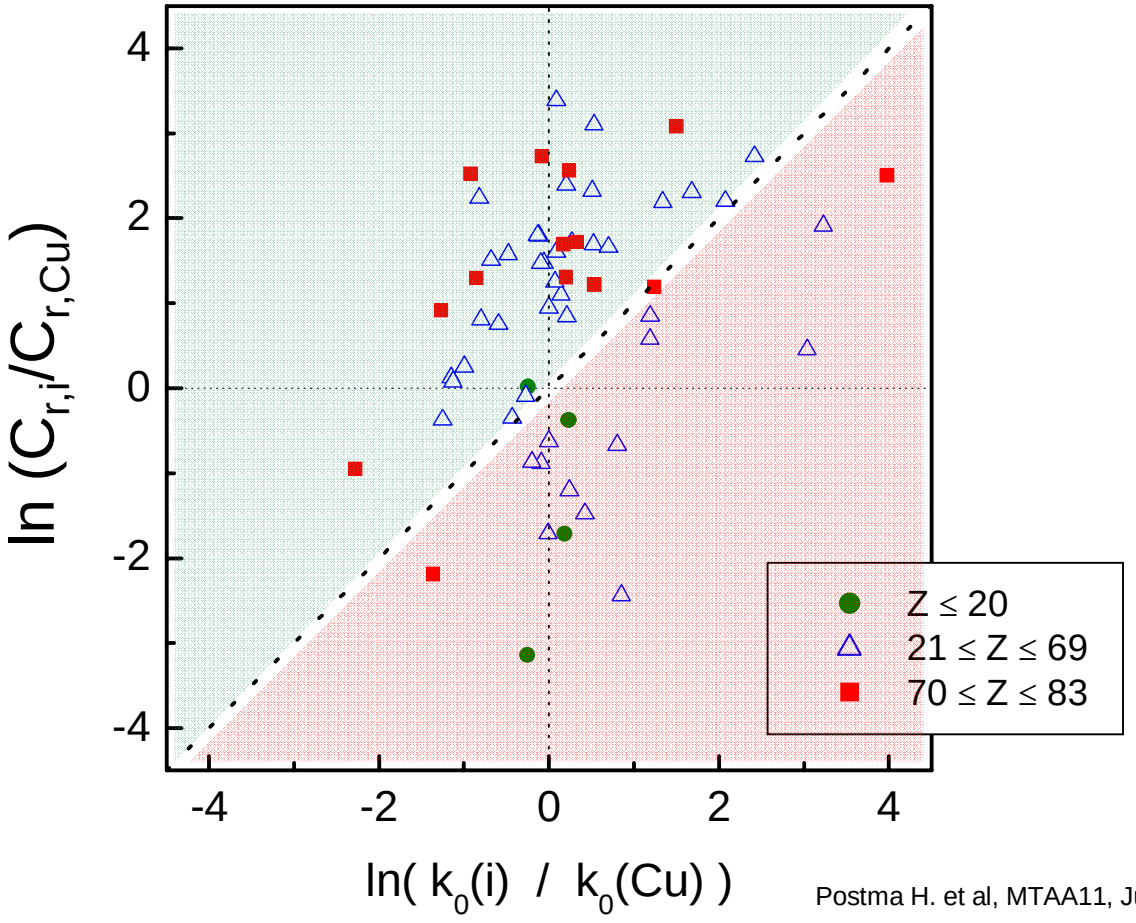
Important/Abundant element in archeological objects

Resonance Parameters E, Γ_n for 578 eV resonance are wrong in ENDFB.VII, JEFF, JENDL

Missing Resonances too



PGAA (at Budapest) and NRCA (at GELINA) Accuracy for Cu in a bronze artefact about 1%



Postma H. et al, MTAA11, June 2004

⇒ k_o and C_r relative to Cu

PGAA best for light elements

H, S, P, and K

NRCA best for heavy elements

As, Ag, Sb, Au and Pb

Both NRCA and PGAA are fully non-destructive methods for bulk analysis

The residual activation is negligible, especially for NRCA

PGAA is good for light elements, NRCA better for heavy elements

PGAA can be hampered by an unfavourable balance between the thermal capture cross sections of the elements

⇒ for NRCA we can always choose a region in the TOF-spectrum where the resonance of interest dominates

NRA - Remarks

Pros:

- Multi elemental method
- Applicable to all elements with resonances
- No special sample preparation
- Negligible residual activation

Cons:

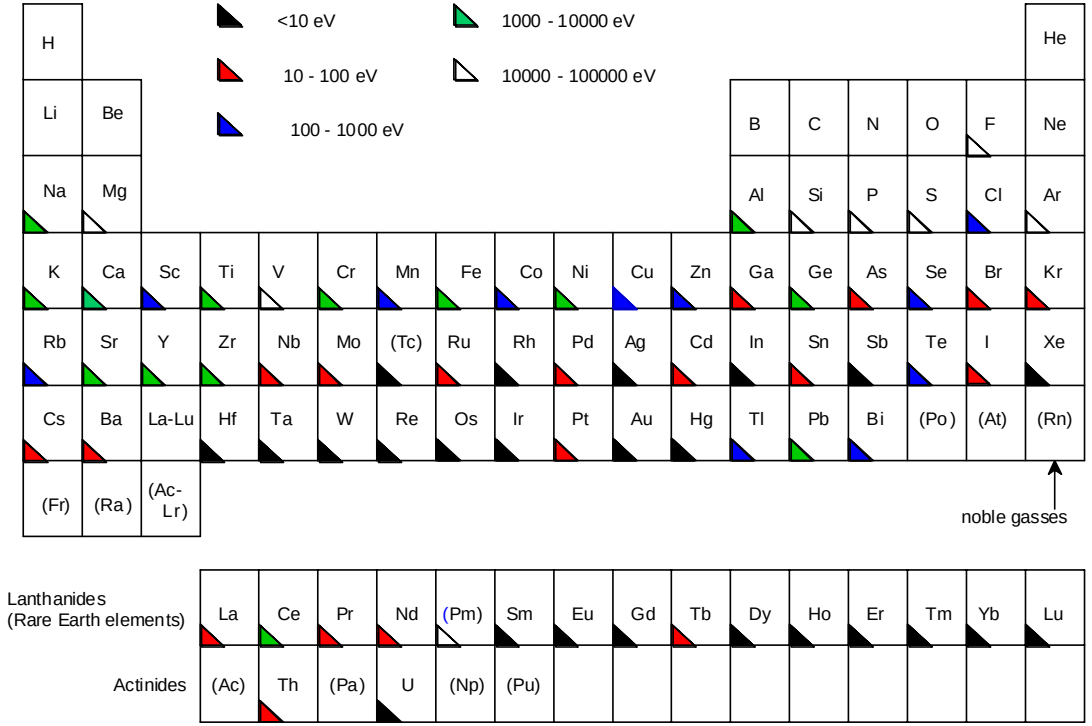
- Data analysis not easy
- Additional measurements with methodological approach

Data needed:

- A_γ

OR

- Resonance Parameters $\Rightarrow \sigma_\gamma(E), \sigma_t(E)$



- Collaborators: J.C. Drohe', J. Van Gils, R. Wynants
- ICTP/IAEA
- This work is supported by the European Commission's 6th Framework Programme in the frame of ANCIENT CHARM Work performed with financial support by the European Community "New and Emerging Science and Technology" Contract No 15311

Joint Research Centre (JRC)

Thermal and epithermal neutron cross sections measurements and applications

A. Borella[1], H. Postma[2], M. Moxon[3] and P. Schillebeeckx[1]

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IRMM - Institute for Reference Materials and Measurements

Geel - Belgium

http://irmm.jrc.ec.europa.eu/

http://www.jrc.ec.europa.eu/

Overview

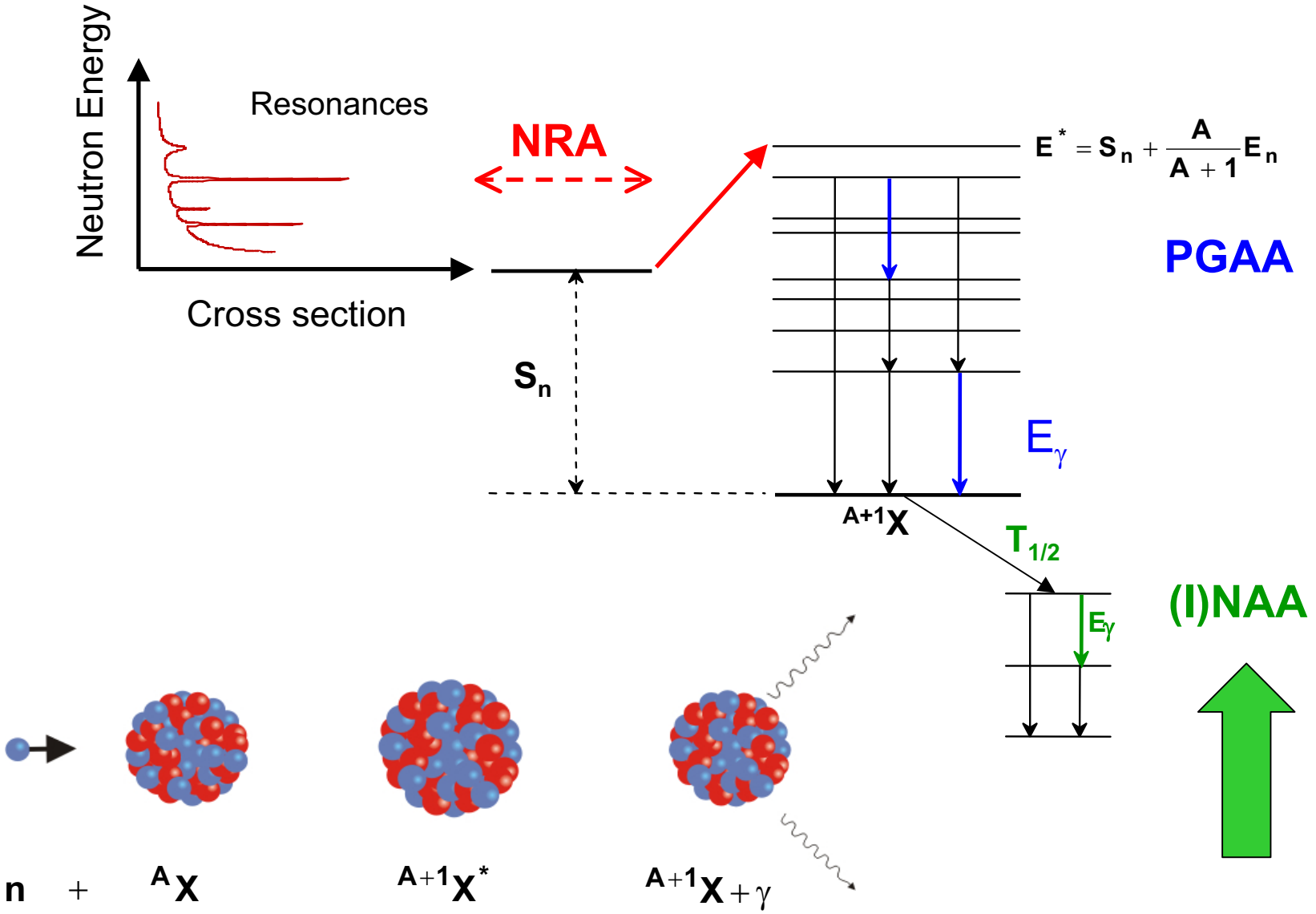
- **Neutron capture process**
  - Techniques originally developped for material analysis
  - Instrumental Neutron Activation Analysis (INAA)
  - Prompt Gamma Activation Analysis (PGAA)
  - Techniques originally developped for cross section measurements
  - Neutron Resonant Analysis (NRA)
    - Capture (NRCA)
    - Transmission (NRT)

- **Principles**
  - Physics and equations
- **Application to NDA/XS measurements**
- **Facilities/Examples**
- **Concluding Remarks**

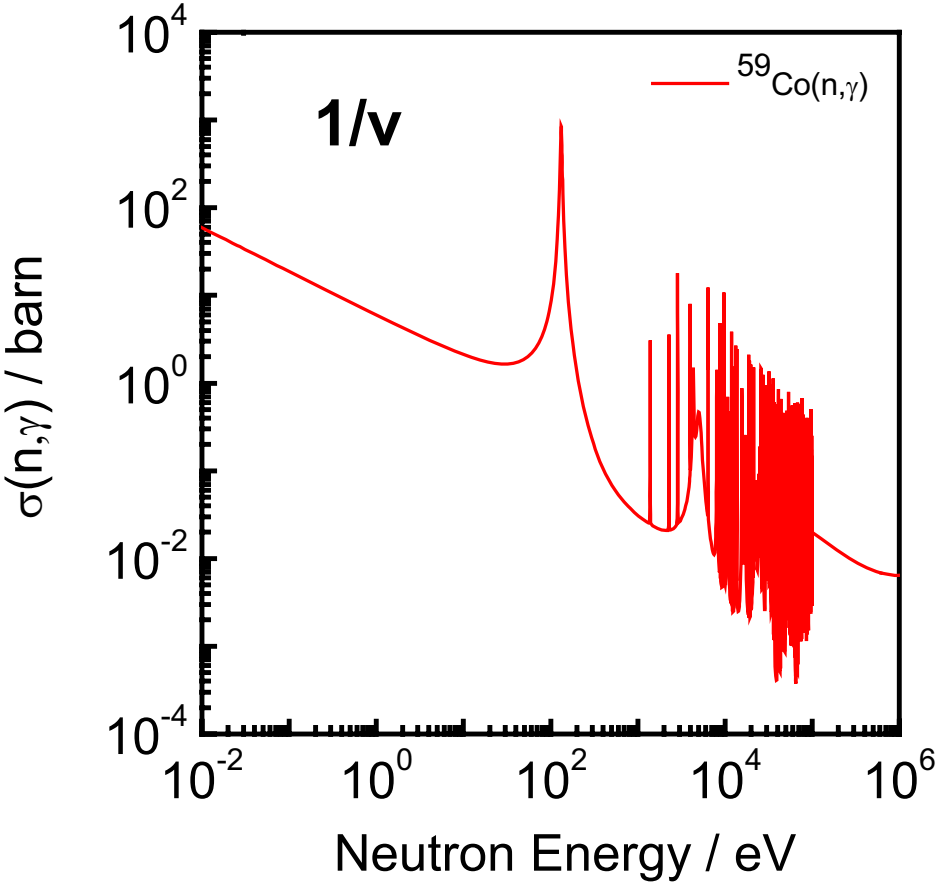
INAA - Overview

- **Principles/Quantities**
  - Neutron Capture Reactions
  - Reaction Rate
  - Flux shape
  - Generalised g-factor and resonance integrals
- **INAA for NDA**
  - Determination of relative abundances
  - Standardization (k_0)
- **Facilities/Examples**

Neutron capture process



Capture cross section

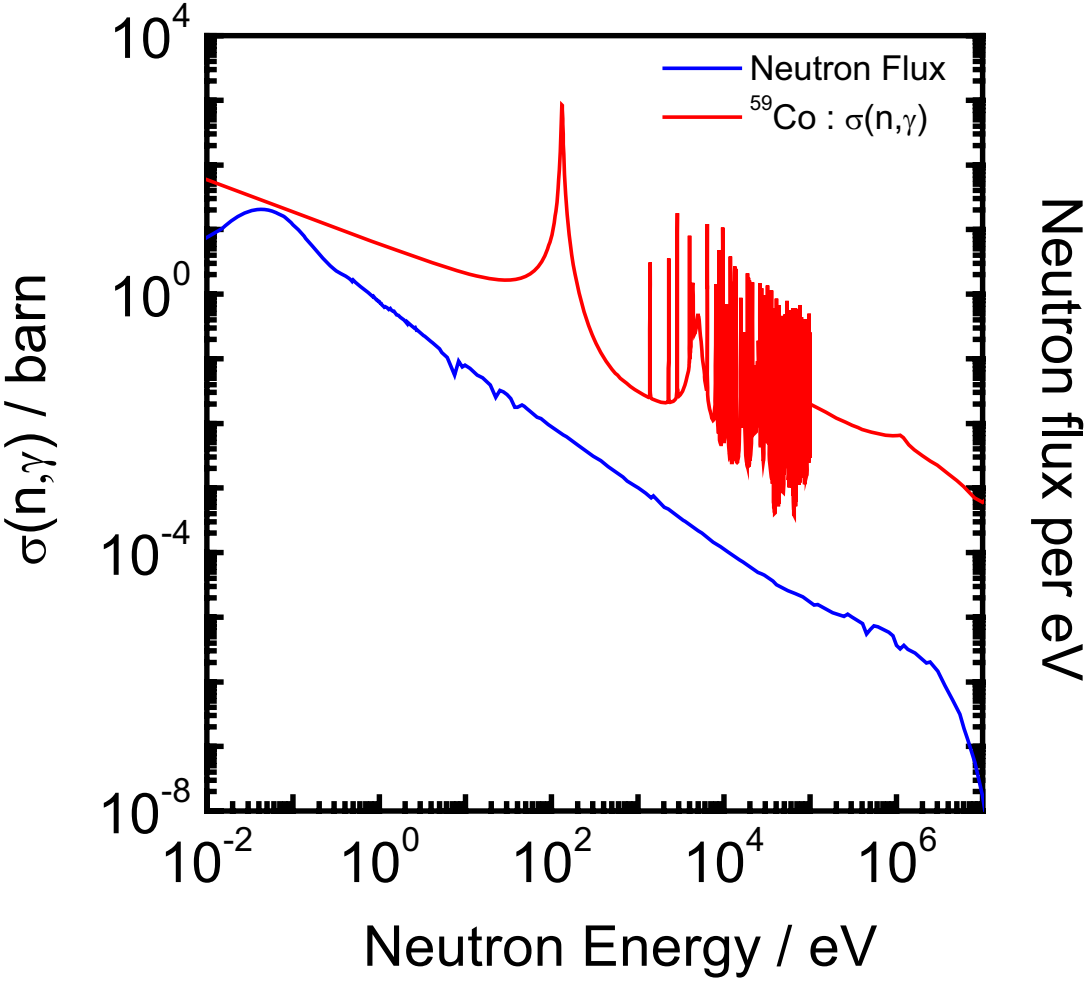


The probability that a neutron interacts with a nucleus is expressed as a cross section σ, which has the dimension of an area

The unit of a cross section is taken as :

1 barn, 1 b = 10^{-24} cm^2

Total reaction rate



The total reaction rate per atom:

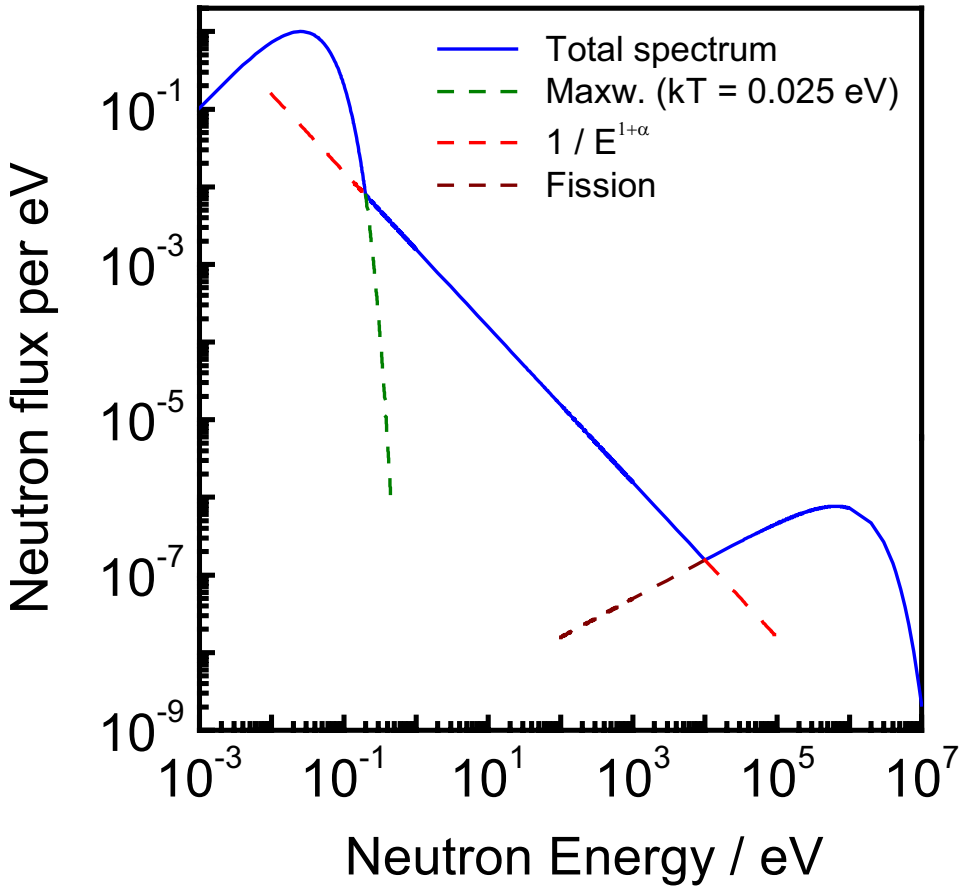
$$R = \int_0^\infty \varphi(E_n)\,\sigma_\gamma(E_n)\,dE_n$$

depends on:

$\varphi(E_n)$ the neutron flux and

$\sigma_\gamma(E_n)$ the capture cross section

Neutron flux in a thermal reactor



The neutron flux in a thermal reactor is a sum of three components

- **Maxwellian distribution with maximum at E_n =kT**
- **$1/E^{1+\alpha}$ distribution due to moderation process of the fast neutrons (epi-thermal spectrum)**
- **"Watt spectrum" of fission neutrons**

At a neutron guide, the neutron flux can be described by the thermal part only (e.g. PGAA at Budapest)

Reaction rate in a thermal reactor

The total reaction rate per atom is:

$$R = \int_0^{\infty} \varphi(E_n)\,\sigma(E_n)\,dE_n$$

To solve the integral one separates between the thermal and the epi-thermal region:

$$R = \int_0^{\infty} \sigma(E)\varphi(E)dE = \int_0^{E_{cd}} \sigma(E)\varphi(E)dE + \int_{E_{cd}}^{\infty} \sigma(E)\varphi(E)dE$$

with E_{Cd} = 0.55 eV

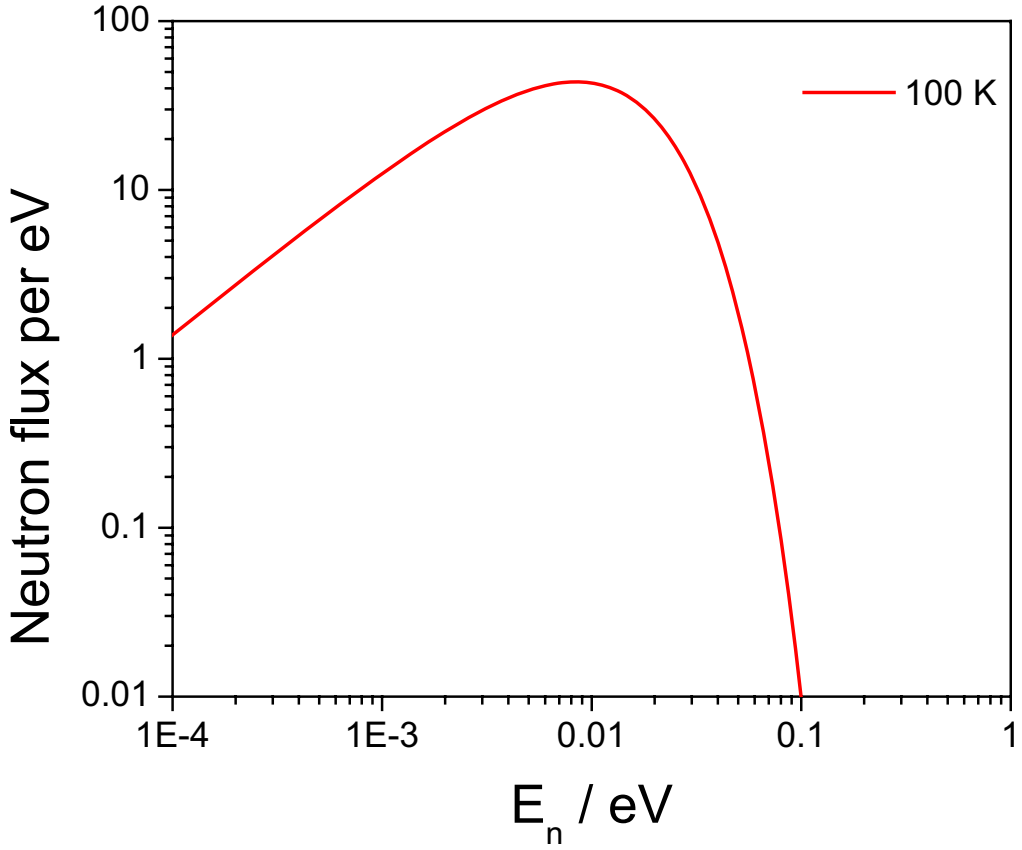
Reaction rate

Guided beam

$$R = \int_0^{E_{cd}} \sigma(E)\varphi(E)dE$$

Cd measurement

$$R = \int_{E_{cd}}^{\infty} \sigma(E)\varphi(E)dE$$

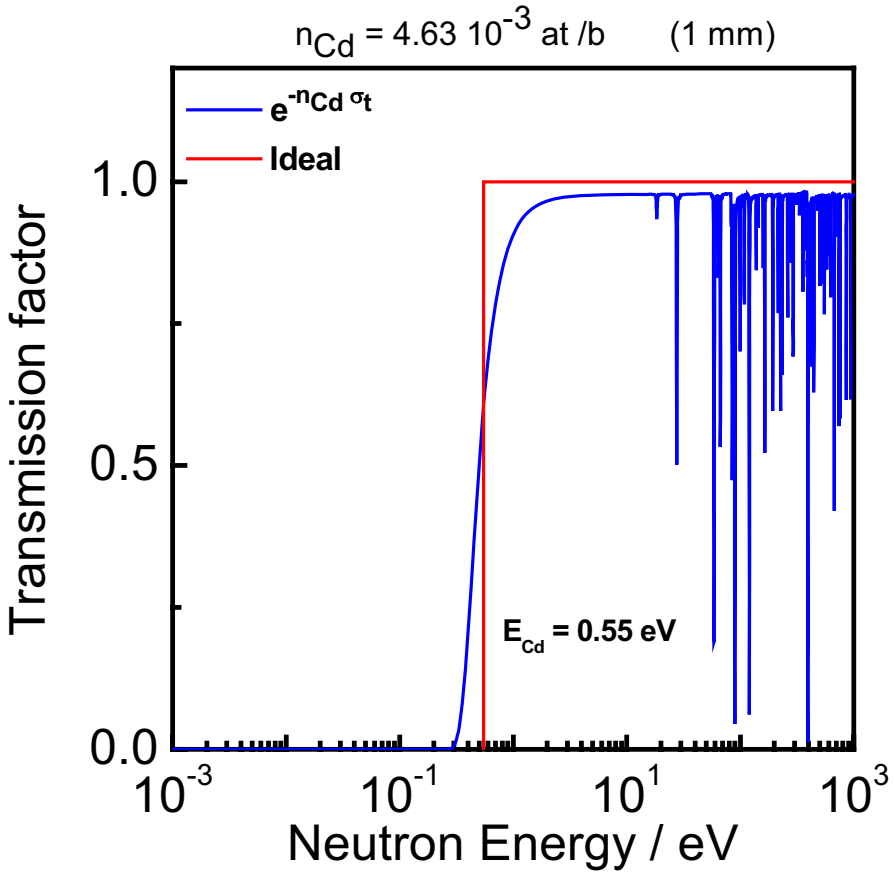


Guided beam

$$R = \int_0^{E_{cd}} \sigma(E)\varphi(E)dE$$

Cd measurement

$$R = \int_{E_{cd}}^{\infty} \sigma(E)\varphi(E)dE$$



Reaction rate in a thermal reactor

$$R = \int_0^{\infty} \sigma(E)\varphi(E)dE = \int_0^{E_{cd}} \sigma(E)\varphi(E)dE + \int_{E_{cd}}^{\infty} \sigma(E)\varphi(E)dE = R_{th} + R_{epi}$$

$$R_{th} = \varphi_t \sigma_0 g_w G_{th}$$

φ_t thermal flux
σ_0 thermal cross section at v=2200 m/s
g_w generalized g-factor: deviation of the cross section from 1/v
G_{th} thermal flux depression factor

Reaction rate in a thermal reactor

$$R = \int_0^{\infty} \sigma(E)\varphi(E)dE = \int_0^{E_{cd}} \sigma(E)\varphi(E)dE + \int_{E_{cd}}^{\infty} \sigma(E)\varphi(E)dE = R_{th} + R_{epi}$$

$$R_{epi} = \varphi_e I_r G_r = \varphi_t \sigma_0 \frac{1}{f} \frac{I_r}{\sigma_0} G_r$$

φ_e epithermal flux
I_r effective resonance integral
G_r resonance self-shielding factor
f ratio of thermal to epithermal flux
$Q=I_r/\sigma_0$ ratio of the resonance integral and thermal cross section

Reaction rate in a thermal reactor

$$R_{th} = \int_0^{E_{cd}} \sigma(E)\varphi(E)dE = \varphi_t \sigma_0 g_w G_t$$

depends on the geometry, composition (σ)

$$\varphi_t = \frac{2}{\sqrt{\pi}} \int_0^{E_{cd}} \varphi(E)dE$$

$$\sigma_{th} = \frac{\int_0^{E_{cd}} \sigma(E)\varphi(E)dE}{\int_0^{E_{cd}} \varphi(E)dE}$$

$$g_w = \frac{\int_0^{E_{cd}} \sigma(E)\varphi(E)dE}{\frac{\sqrt{\pi}}{2}\varphi_t \sigma_0} = \frac{\int_0^{E_{cd}} \sigma(E)\varphi(E)dE}{\sigma_0 \int_0^{E_{cd}} \varphi(E)dE} = \frac{2}{\sqrt{\pi}} \frac{\sigma_{th}}{\sigma_0}$$

Reaction rate in a thermal reactor

$$g_w = \frac{\int_0^{E_{cd}} \sigma(E)\varphi(E)dE}{\frac{\sqrt{\pi}}{2}\varphi_t\sigma_0} = \frac{\int_0^{E_{cd}} \sigma(E)\varphi(E)dE}{\sigma_0 \int_0^{E_{cd}} \varphi(E)dE} = \frac{2}{\sqrt{\pi}}\frac{\sigma_{th}}{\sigma_0}$$

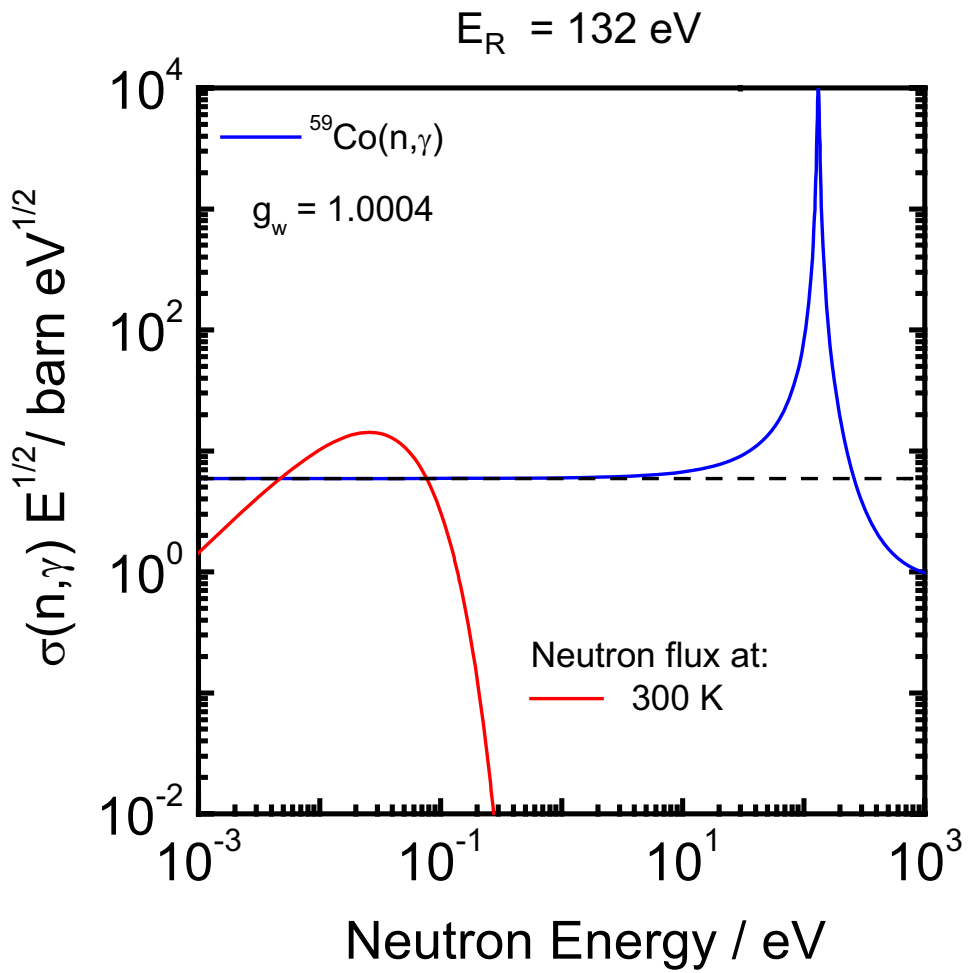
g_w depends on:

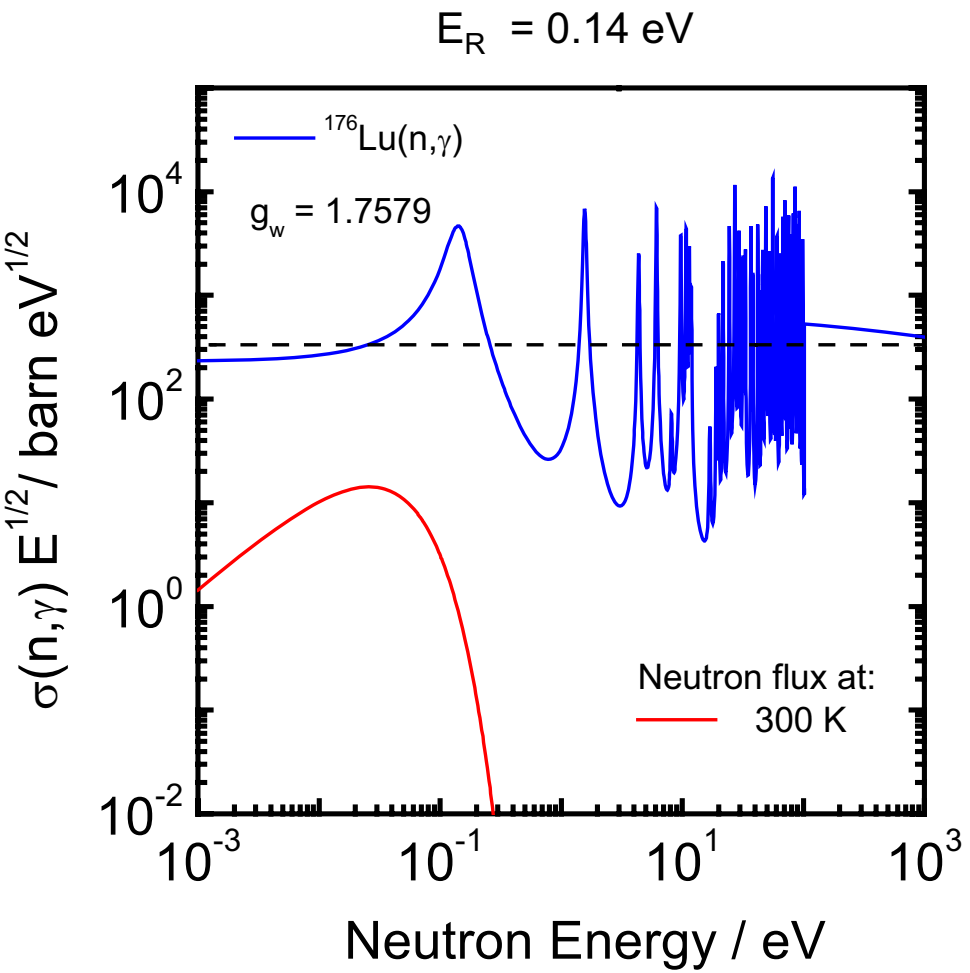
- **Flux shape (may not be Maxwellian)**
- **Cross section shape (may not be 1/v)**

If the flux has a Maxwellian distribution and If the cross section is 1/v

$$\sigma_{th} = \frac{\sqrt{\pi}}{2}\sigma_0\sqrt{\frac{T_0}{T}} \quad \Rightarrow \quad g_w = \sqrt{\frac{T_0}{T}}$$

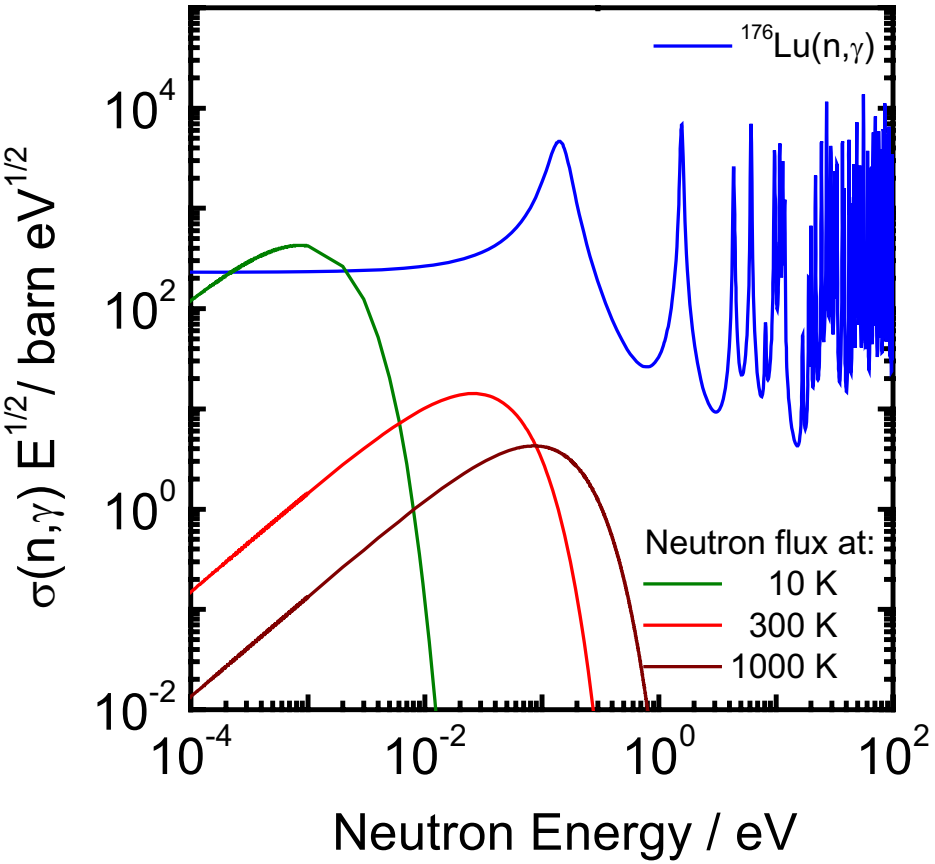
g_w - factor: deviation of 1/v behaviour #1

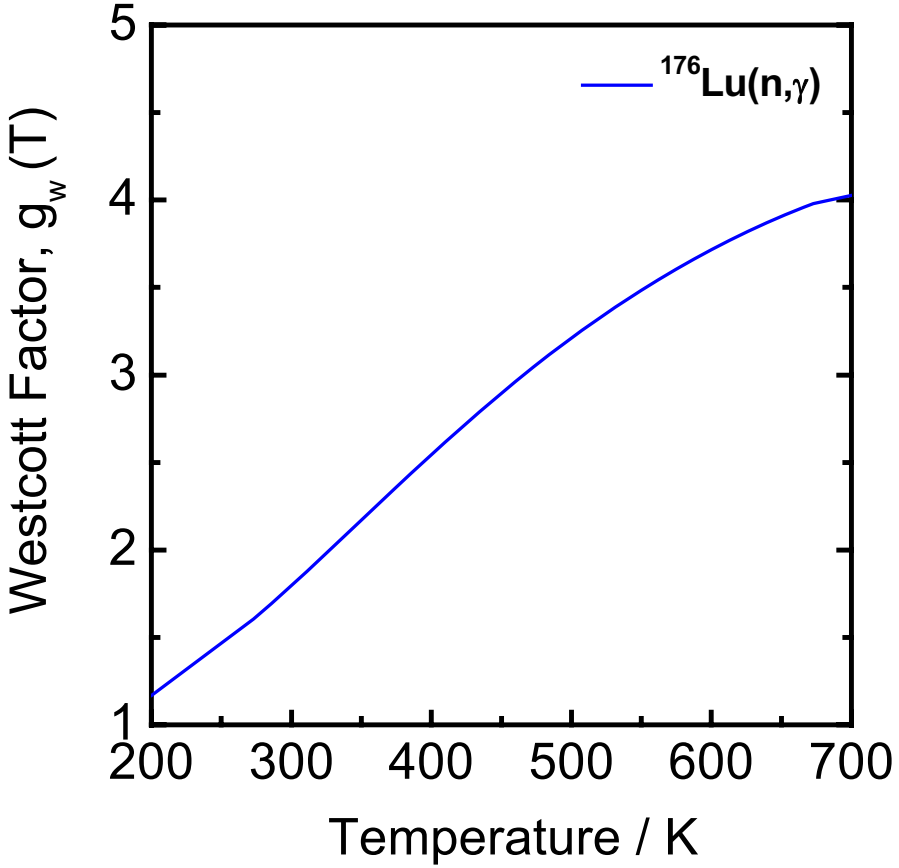




g_w - factor: deviation of 1/v behaviour #2

The Westcott g_w – factor is temperature dependent





Reaction rate in a thermal reactor

$$R = \varphi_t \sigma_0 g_w G_{th} + \varphi_e I_r G_r$$

$$\varphi_e I_r G_r = \int_{E_{cd}}^{\infty} \sigma(E)\varphi(E)dE$$

$$\varphi_e = \int_{E_{cd}}^{\infty} \varphi(E)dE$$

Resonance integral

I_r depends on:

- $\sigma(E)$ (resonance parameters)
- $\varphi(E)$ (e.g. $E^{-(1+\alpha)}$)

$$I_r = \frac{\int_{E_{cd}}^{\infty} \sigma(E)\varphi(E)dE}{\varphi_e}$$

Data base of σ_0, g_w-factor and I_r
Compilation by S.F.Mughabghab, BNL, USA
"Thermal neutron capture cross sections, resonance integrals and g_w-factors"
INDC(NDS) – 440
February 2003

So far, one atom
Real case N atoms

$$R_N = N\varphi_t\sigma_0\left(g_w G_{th} + \frac{1}{f}\frac{I_r}{\sigma_0}G_r\right)$$

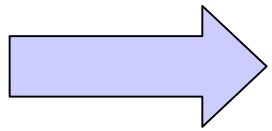
R_N from measurement

Knowing N, we can deduce σ (nuclear data)

Knowing σ (nuclear data), we can deduce N

How to measure the R_N

- Sample activation by neutron irradiation in reactor
- The sample is transferred into a counting station (Rabbit system)
- (Waiting time)
- Counting
- Isotope identification by means of the γ–ray energy
- High resolution detector needed
  - Ge(Li), HPGe
  - NaI
- Peak analysis
- Correlate peak area with R, isotope abundance



Simple reaction/decay (no branching…)

$$C_\gamma = \frac{N_{av} W \theta}{M} P_\gamma \varepsilon_{FE} R \left(1 - e^{-\lambda t_i}\right) e^{-\lambda t_d} \frac{\left(1 - e^{-\lambda t_m}\right)}{\lambda}$$

C_γ Counts (DT,coinc)

W Sample mass

θ Isotopic abundance

M Atomic mass

N_{av} Avogadro's number

P γ-ray intensity

$$R = \varphi_t \sigma_0 \left(g_w G_{th} + \frac{1}{f} \frac{I_r}{\sigma_0} G_r \right)$$

t_i Irradation Time

t_d Decay Time

t_m Counting Time

NAA for Non Destructive Analysis

$$W = \frac{C_\gamma}{\left(1 - e^{-\lambda t_i}\right) e^{-\lambda t_d} \frac{\left(1 - e^{-\lambda t_m}\right)}{\lambda} N_{av} \theta R P_\gamma \varepsilon_{FE}} \frac{M}{1}$$

– Absolute method (ε_{FE}, P_γ,σ_0 sources of uncertainty)

Relative abundance

$$\frac{W}{W_{ref}} = \frac{C_\gamma}{C_{\gamma,ref}} \frac{\varepsilon_{FE,ref}}{\varepsilon_{FE}} \frac{G_t g_w + G_f \frac{Q}{f}}{G_{t,ref} g_{w,ref} + G_{f,ref} \frac{Q_{ref}}{f}} \frac{\sigma_{0,ref}}{\sigma_0} \frac{M}{\theta P_\gamma} \frac{\theta_{ref} P_{\gamma,ref}}{M_{ref}}$$

Reference $^{197}Au(n,\gamma)$ E_γ=411.8 keV

k_0

accurately determined for many nuclides

depends only on the isotope

$$\frac{1}{k_0} = \frac{\sigma_{0,ref}}{\sigma_0} \frac{M}{M_{ref}} \frac{\theta_{ref} P_{\gamma,ref}}{\theta P_\gamma}$$

e.g. De Corte/Simonits (JRNC 133 (1989) 43)

N.B. It is actually a cross section measurement!

Relative abundance

$$\frac{W}{W_{ref}} = \frac{C_\gamma}{C_{\gamma,ref}} \frac{\varepsilon_{FE,ref}}{\varepsilon_{FE}} \frac{G_t g_w + G_f \frac{Q}{f}}{G_{t,ref} g_{w,ref} + G_{f,ref} \frac{Q_{ref}}{f}} \frac{1}{k_0}$$

Usually a ^{197}Au sample is co-irradiated with the sample to analyse

Experimental conditions

- ε_{FE}
- $\varphi(E)$ Q, g_w,f
- geometry G

$$\frac{G_t g_w + G_f \frac{Q}{f}}{G_{t,ref} g_{w,ref} + G_{f,ref} \frac{Q_{ref}}{f}}$$

Microscopic data

- $\sigma(E)$ Q, g_w, k_0

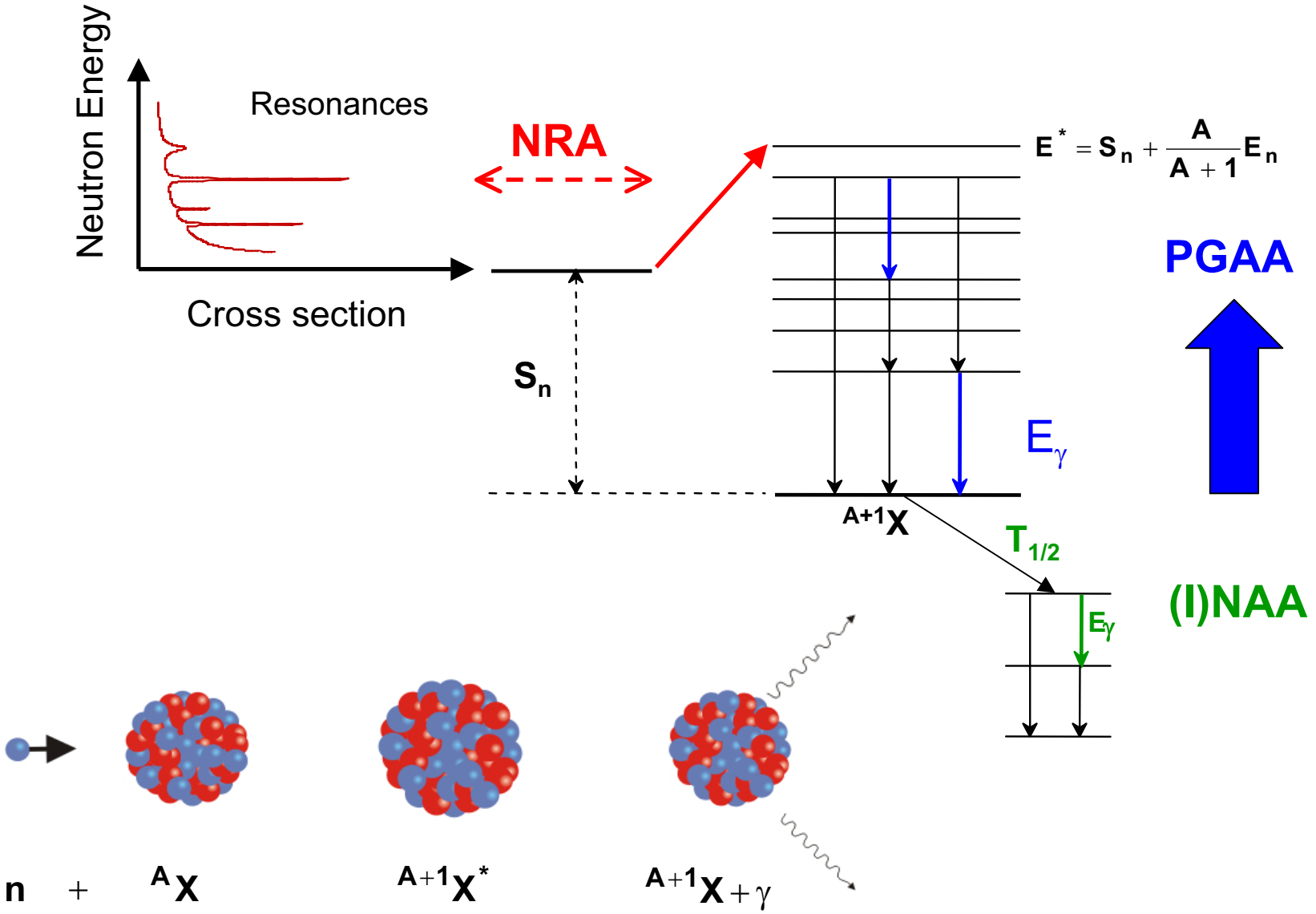
Facilities/Applications

- **NIST Center for Neutron Research**
- **BNC (H)**
  - inorganic impurities in C_{60}
  - selenium in food
- **BR-1 at the SCK-CEN (B)**

- **INAA Laboratory at the Delft University of Technology (NL)**

and many others…

Neutron capture process



PGAA - Overview

- **Principles**
  - Basic equations
- **PGAA**
  - PGAA at the BNC
    - Cold beam
    - Compton suppression system
  - PGAA Standardization (NDA)
  - Examples
  - PGAA for XS measurements
  - Examples (^{209}Bi, ^{206}Pb)

Neutron Induced Prompt Gamma Activation Analyses (PGAA or PGNAA)

- Detection of prompt gamma radiation from a neutron induced nuclear reaction, usually radiative capture
- Isotope identification based on E_γ
- Isotope quantification based on I_γ
- Instantaneous method: results appear immediately
- Usually little or negligible residual activation of the sample

PGAA - principle

The total reaction rate per atom:

$$R = \int_0^\infty \varphi(E_n)\,\sigma_\gamma(E_n)\,dE_n$$

depends on:

- $\varphi(E_n)$ the neutron flux and
- $\sigma_\gamma(E_n)$ the capture cross section

For a guided (thermal or cold) neutron beam $R = G_{th}\varphi_t\sigma_{\gamma 0}g_w$

PGAA - principle

If we look at a given transition

$$R_i = G_{th}\varphi_t\sigma_{\gamma 0,i}g_w = G_{th}\varphi_t P_\gamma \sigma_{\gamma 0} g_w$$

where P_γ is the intensity of the transition

for N atoms

$$R_{N,i} = NG_{th}\varphi_t P_\gamma \sigma_{\gamma 0} g_w$$

$R_{N,i}$ from measurement

Knowing N, we can deduce σ (nuclear data)

Knowing σ (nuclear data), we can deduce N

How to measure the R_N

- We look at the prompt-gamma emission after neutron irradiation (e.g. cold neutron beam)
- Isotope identification by means of the γ–ray energy
- High resolution detector needed
  - Ge(Li), HPGe
  - NaI
- Peak analysis
- Correlate peak area with R, isotope abundance/σ

PGAA at the Budapest Neutron Centre

Why using a cold beam?

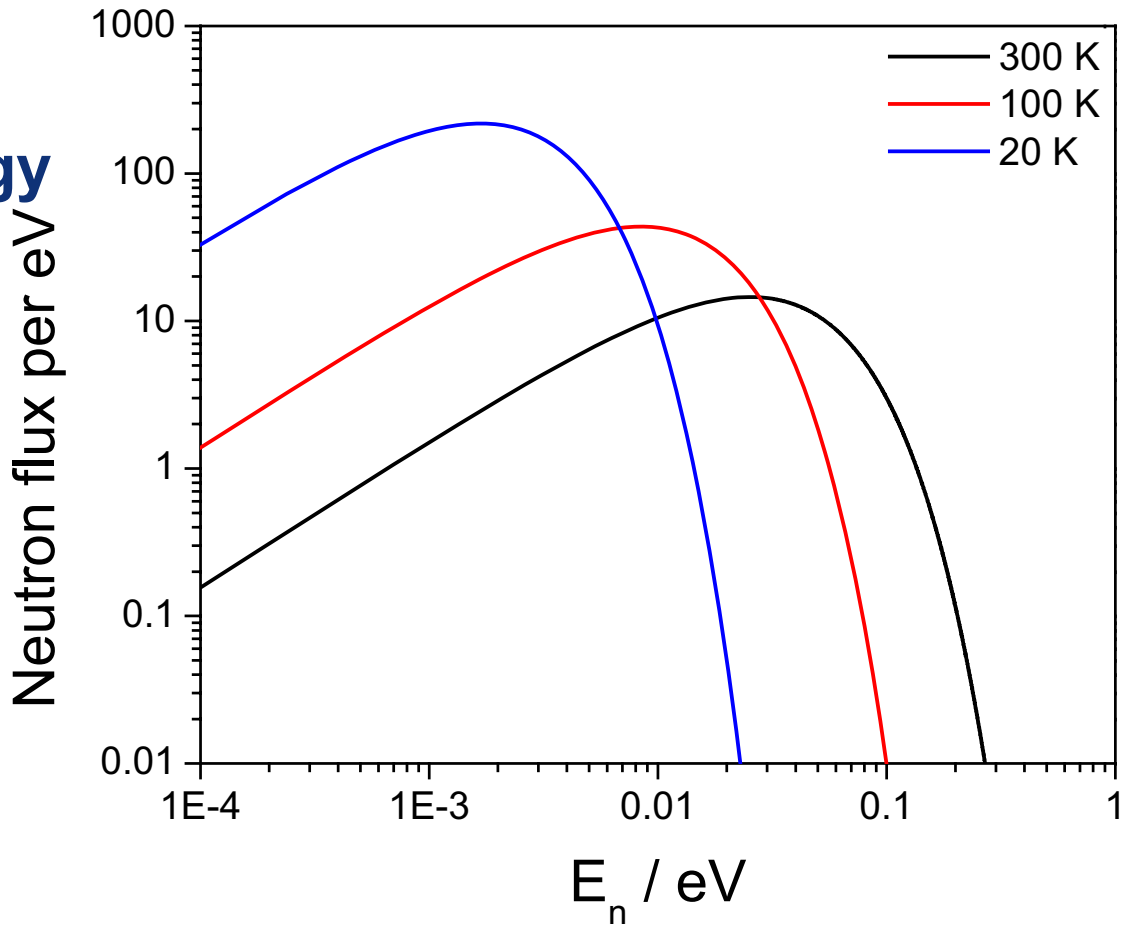
More neutron with lower energy

Cross section is higher

The count rate is higher

No epithermal neutrons

g_w is 1 in most cases



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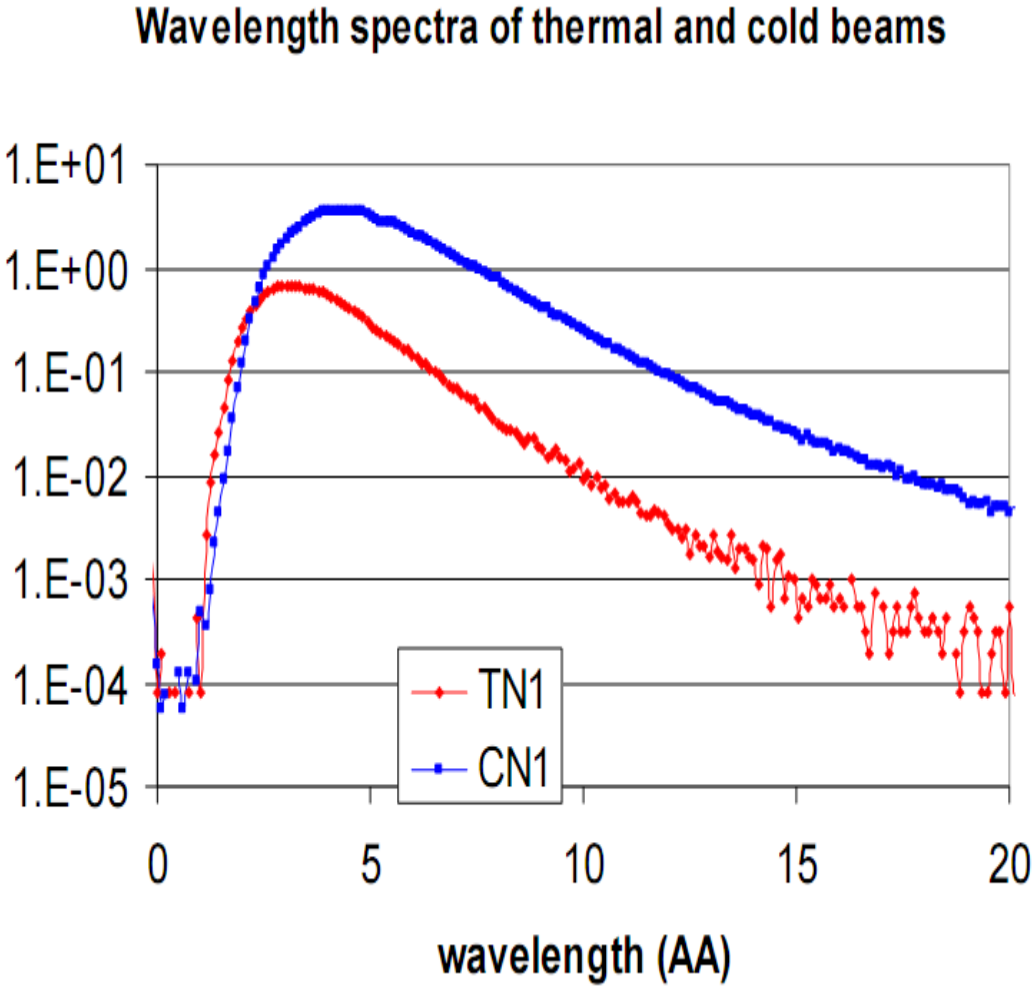
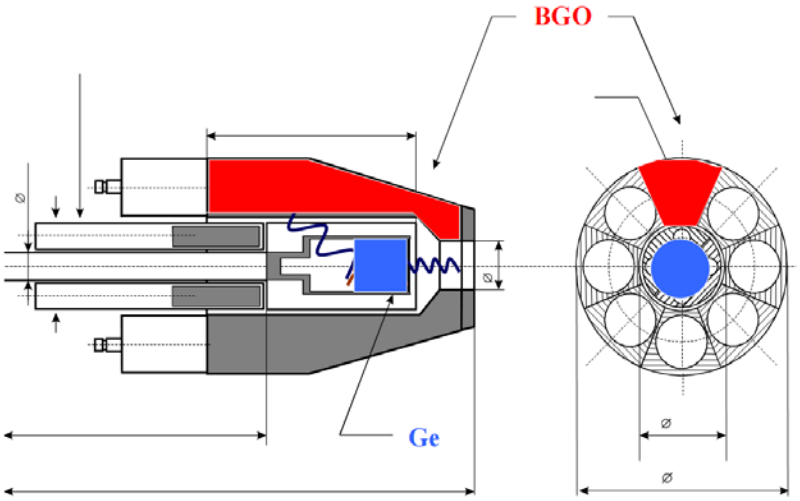
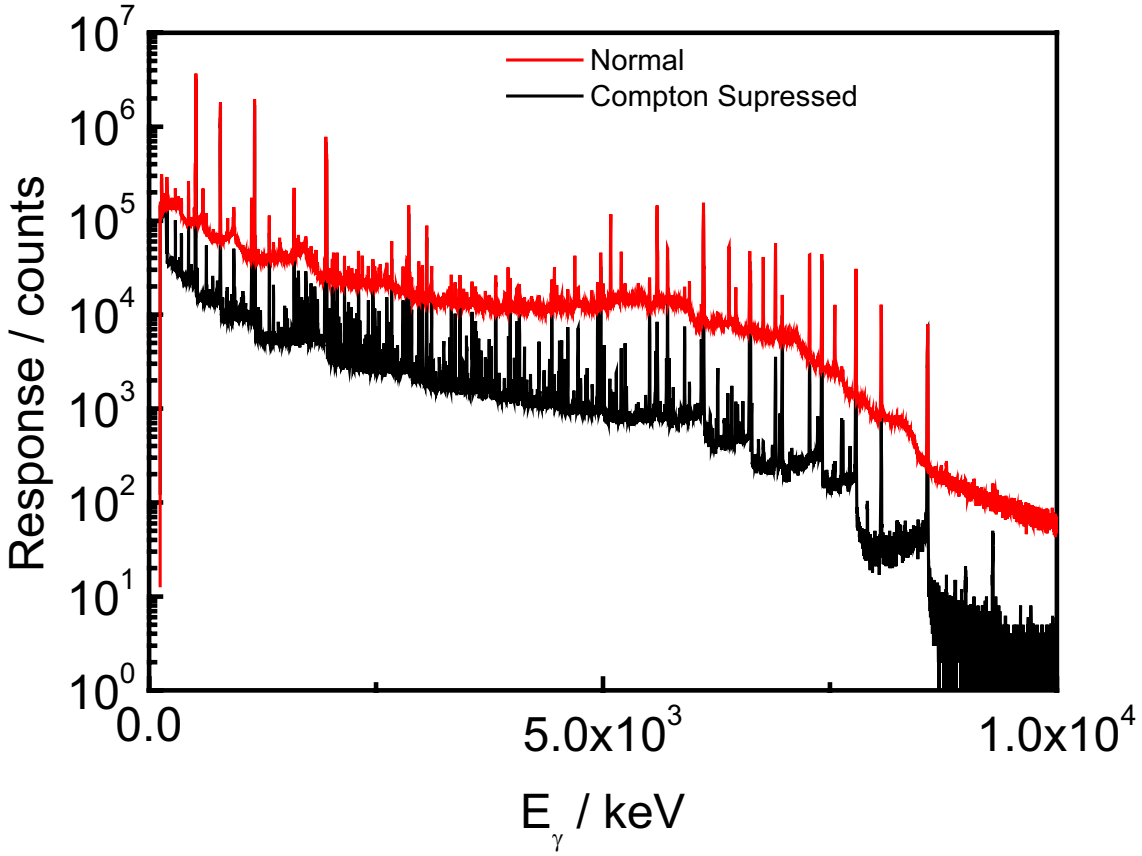


Why using Compton suppression system?

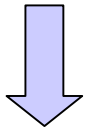




Peak to background improved

non radioactive nucleus

$$C_\gamma = \frac{N_{av} W \theta}{M} P_\gamma \sigma_{\gamma 0} g_w G_{th} \varepsilon_{FE} t_m \varphi_t$$



C_γ	Counts (DT,coinc)
P_γ	γ-ray intensity
ε_{FE}	Full Energy peak detection efficiency
W	Sample mass
θ	Isotopic abundance
M	Atomic mass
t_m	Counting Time

PGAA for Non Destructive Analysis

$$W = \frac{C_\gamma}{\varepsilon_{FE} t_m} \frac{M}{N_{av} \theta} \frac{1}{G_{th} \varphi_t P_\gamma \sigma_{\gamma 0} g_w}$$

- Absolute method (large uncertainties)

Relative measurements allow to reduce the uncertainty

– comparator (ref)

$$\frac{W}{W_{ref}} = \frac{C_\gamma}{C_{\gamma,ref}} \frac{\varepsilon_{FE,ref}}{\varepsilon_{FE}} \frac{g_w}{g_{w,ref}} \frac{\sigma_{0,ref}}{\sigma_0} \frac{M}{\theta P_\gamma} \frac{\theta_{ref} P_{\gamma,ref}}{M_{ref}} \rightarrow \frac{1}{k_0}$$

– homogeneous sample ⇒ G is the same

– efficiency ratio can be determined accurately (<1%)

– method standardization (Revay and Molnar, Radiochimica Acta 91, 361, 2003):

– k_0,σ_0 library

– Ultimate comparator: H σ_0 = 0.3326(7) b, 2.2 MeV

⇒ no standard samples needed

- **Neutron capture cross section measurements**
  - ^{nat}Pb, ^{206}Pb, ^{209}Bi, ^{127}I, ^{129}I…
- **Rocks and minerals (Geology, Archaeology)**
  - lapislazuli composition (Cl, S content) ⇒ ore
- **Ceramics (Archaeology)**
- **Glasses Metals (Archaeology, Industry)**
  - Roman silver coins Cu/Ag ratio ⇒ period
- **Chemistry**
  - S in fullerene (C_{60})
- **Metals (Materials Research, Archaeology)**
- **Nuclear Materials (Safeguards, Transmutation)**
  - $^{235}U/^{238}U$ mass ratio

non radioactive nucleus

$$C_\gamma = \frac{N_{av} W \theta}{M} P_\gamma \sigma_{\gamma 0} g_w G_{th} \varepsilon_{FE} t_m \varphi_t$$

- C_γ Counts (DT,coinc)
- P_γ γ-ray intensity
- ε_{FE} Full Energy peak detection efficiency
- W Sample mass
- θ Isotopic abundance
- M Atomic mass
- t_m Counting Time

$$n = \frac{N_{av} \theta W}{M}$$

Number of nuclei

$$\sigma_\gamma = P_\gamma \sigma_{\gamma 0}$$

Partial Capture cross section

non radioactive nucleus

$$C_\gamma = n\sigma_\gamma g_w G_{th} \varepsilon_{FE} t_m \varphi_t$$

C_γ	Counts (DT,coinc)
σ_γ	Partial γ-ray cross section
ε_{FE}	Full Energy peak detection efficiency
W	Sample mass
θ	Isotopic abundance
M	Atomic mass
t_m	Counting Time

PGAA for cross section measurements

$$\sigma_\gamma = \frac{C_\gamma}{\varepsilon_{FE} t_m} \frac{1}{n} \frac{1}{G_{th} \varphi_t g_w} \Rightarrow \sigma_\gamma = \frac{C_\gamma}{\varepsilon_{FE} t_m} \frac{1}{n} \frac{(\alpha_\gamma + 1)}{G_{th} \varphi_t g_w}$$

– Internal conversion coefficient, important for low energy transitions

Mixture

$$\frac{\sigma_{\gamma,1x}}{\sigma_{\gamma,2y}} = \frac{\left(P_\gamma \sigma_{\gamma 0}\right)_1}{\left(P_\gamma \sigma_{\gamma 0}\right)_2} = \frac{C_{\gamma,1}}{C_{\gamma,2}} \frac{\varepsilon_{FE,2}}{\varepsilon_{FE,1}} \frac{n_y}{n_x} \frac{\alpha_{\gamma 1}+1}{\alpha_{\gamma 2}+1} \frac{g_{w,2}}{g_{w,1}}$$

Relative partial cross section

Stoichiometric compounds

n_Y/n_X known

e.g. nitrate sample $Pb(NO_3)_2$, $Bi(NO_3)_3$, normalisation to $^{14}N(n_{th},\gamma)$ 1884 keV line

ε

Accounts also γ–ray attenuation in the sample, can be accessed experimentally
with measurements on ad-hoc sample or
with simulations (e.g. MCNP)

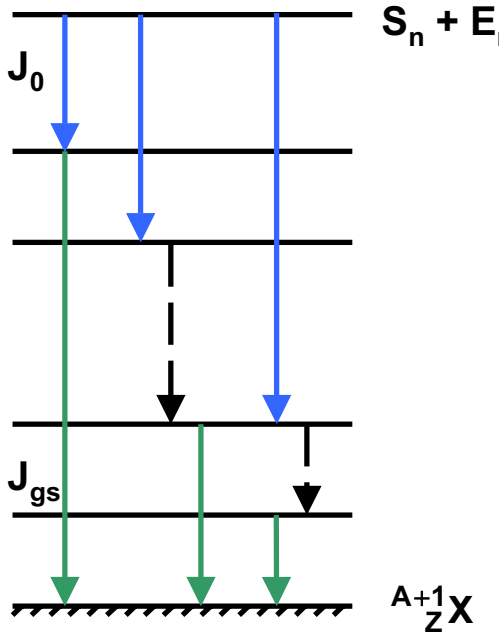
Adding partial cross section feeding/depopulating one state

For primary gamma's

$$\sigma_{\gamma 0} = \sum_{J_0} \sigma_{\gamma,i}$$

Gamma's to the ground state

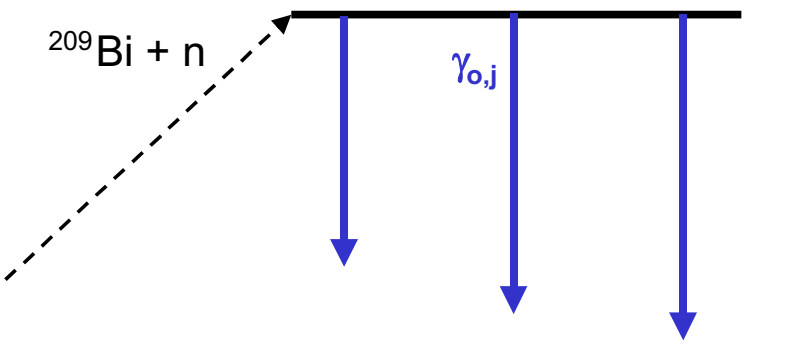
$$\sigma_{\gamma 0,g} = \sum_{J_{gs}} \sigma_{\gamma,i}$$



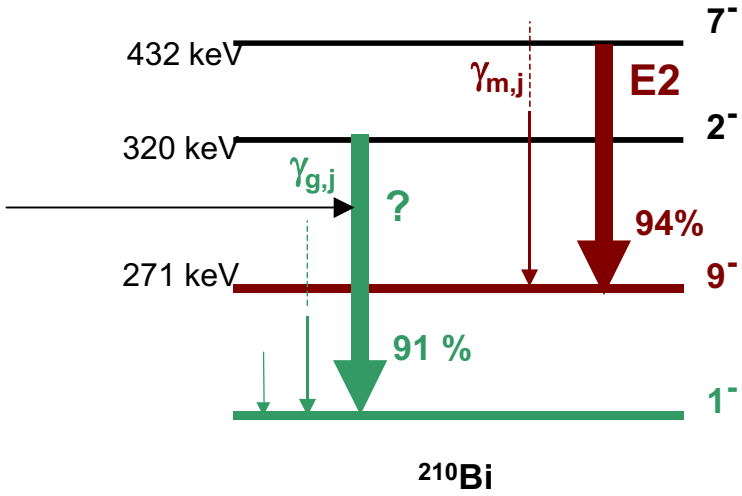
Alternative method: Total energy (all observed gamma's)

$$\sum_{J=all} \sigma_{\gamma,i} E_{\gamma,i} = \sigma_{\gamma 0} E^{*}$$

$^{209}Bi(n_{th},\gamma)$ cross sections



$$\sigma_{m+g} = \sum_j \sigma_{\gamma o,j}$$



$$\sigma_m = \sum_j \sigma_{\gamma m,j}$$

320 keV transition multipolarity unknown

- M1
- E2
- 50% E2 + 50% M1

$$\sigma_g = \sum_j \sigma_{\gamma g,j}$$

$^{209}Bi(n_{th},\gamma)$ results

Method	σ_{m+g} / mb	σ_m / mb	σ_g / mb
Primary γ's	34.2 (0.4)		
Feeding state	35.0 (1.4) [a]	**17.7 (0.7)**	17.3 (0.7) [a]
	37.0 (1.5) [b]		19.3 (0.8) [b]
	39.2 (1.6) [c]		**21.5 (0.9)** [c]
Total energy	39.8 (1.6) [a]	17.7 (0.7)	22.1 (1.4) [a]
	40.0 (1.6) [b]		22.3 (1.4) [b]
	40.1 (1.6) [c]		**22.4 (1.4)** [c]

320 keV transition

a 100% E2

b 50% M1 + 50% E2

c 100% M1

σ_{m+g}= 40.1 (1.2) mb

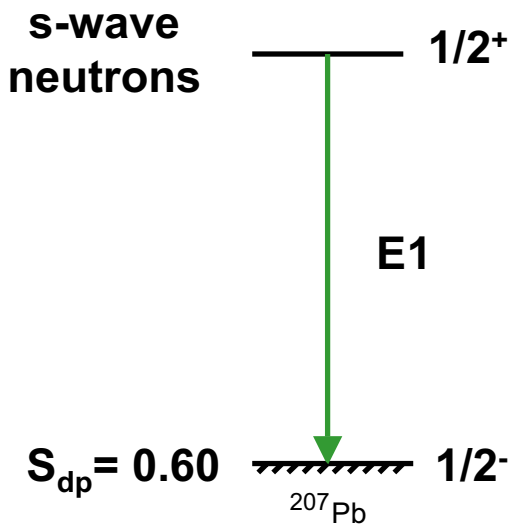
σ_{m+g}= σ_m+ σ_g for 320 keV transition M1

isomeric ratio at n_{th} is β = 1.27 (0.09)

$^{206}Pb(n_{th},\gamma)$ cross section

Phys. Rev. C 2002

$\sigma_\gamma(E_{th})$ = 27.3 (0.8) mb 26.3 (1.2) mb (Blackmon et al.)



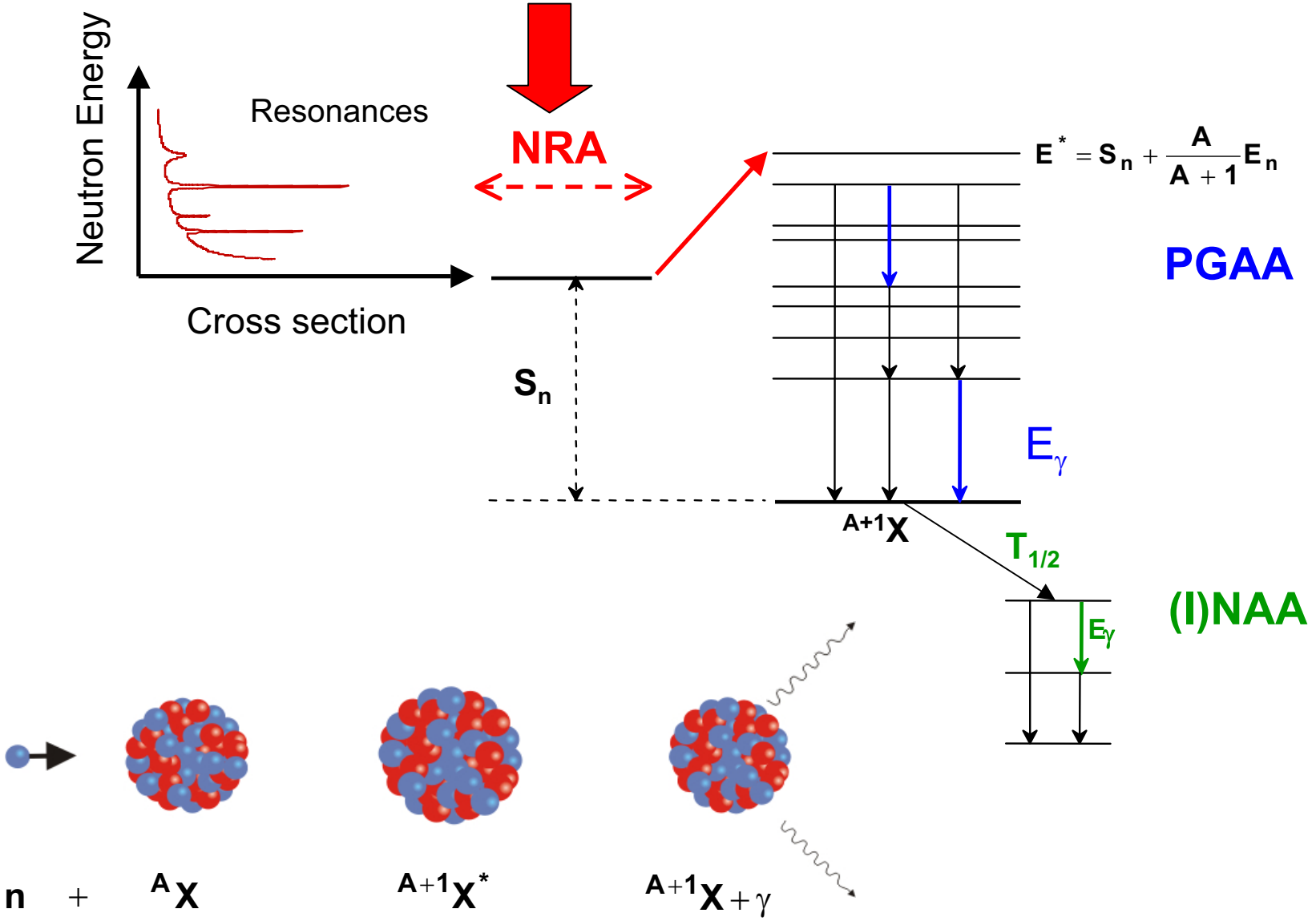
$\sigma_\gamma(E_{th})$ = s-wave resonances + direct capture

8.3 (0.2) mb + 14.0 mb

= 22.3 mb

⇒ Experimental evidence of direct capture in ^{206}Pb

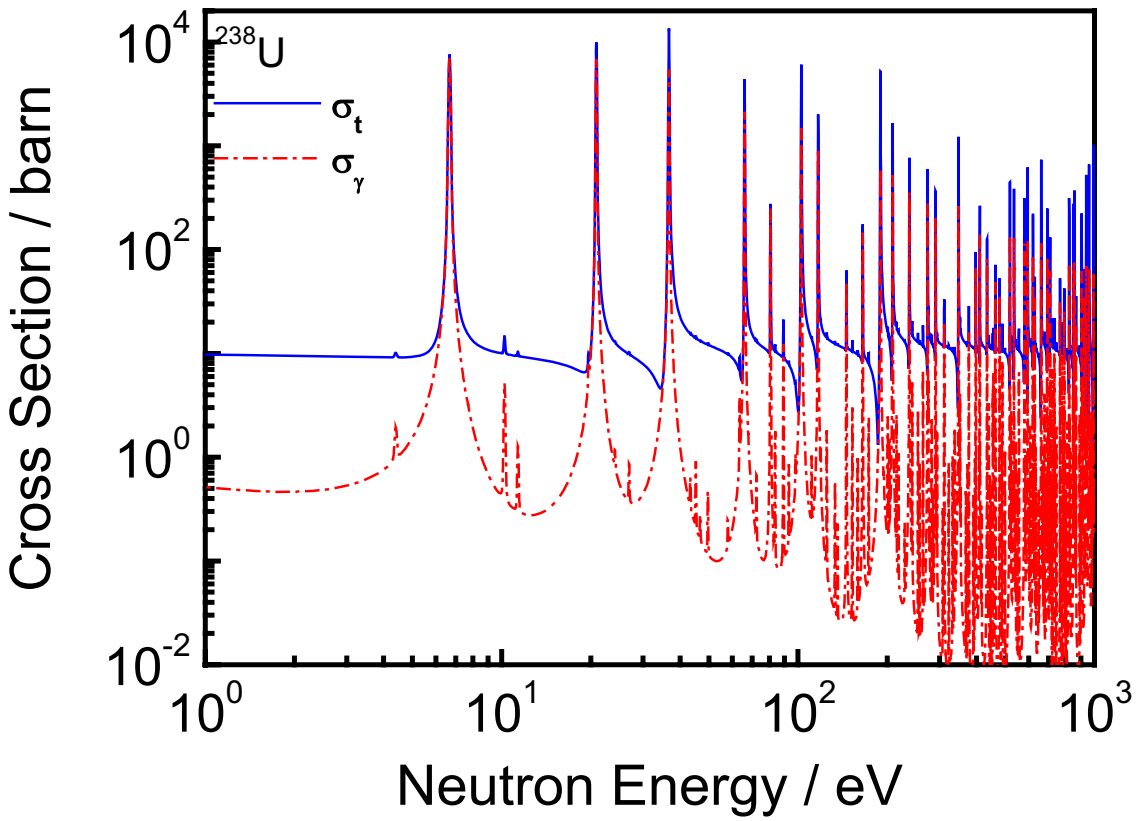
Neutron capture process



NRA - Layout

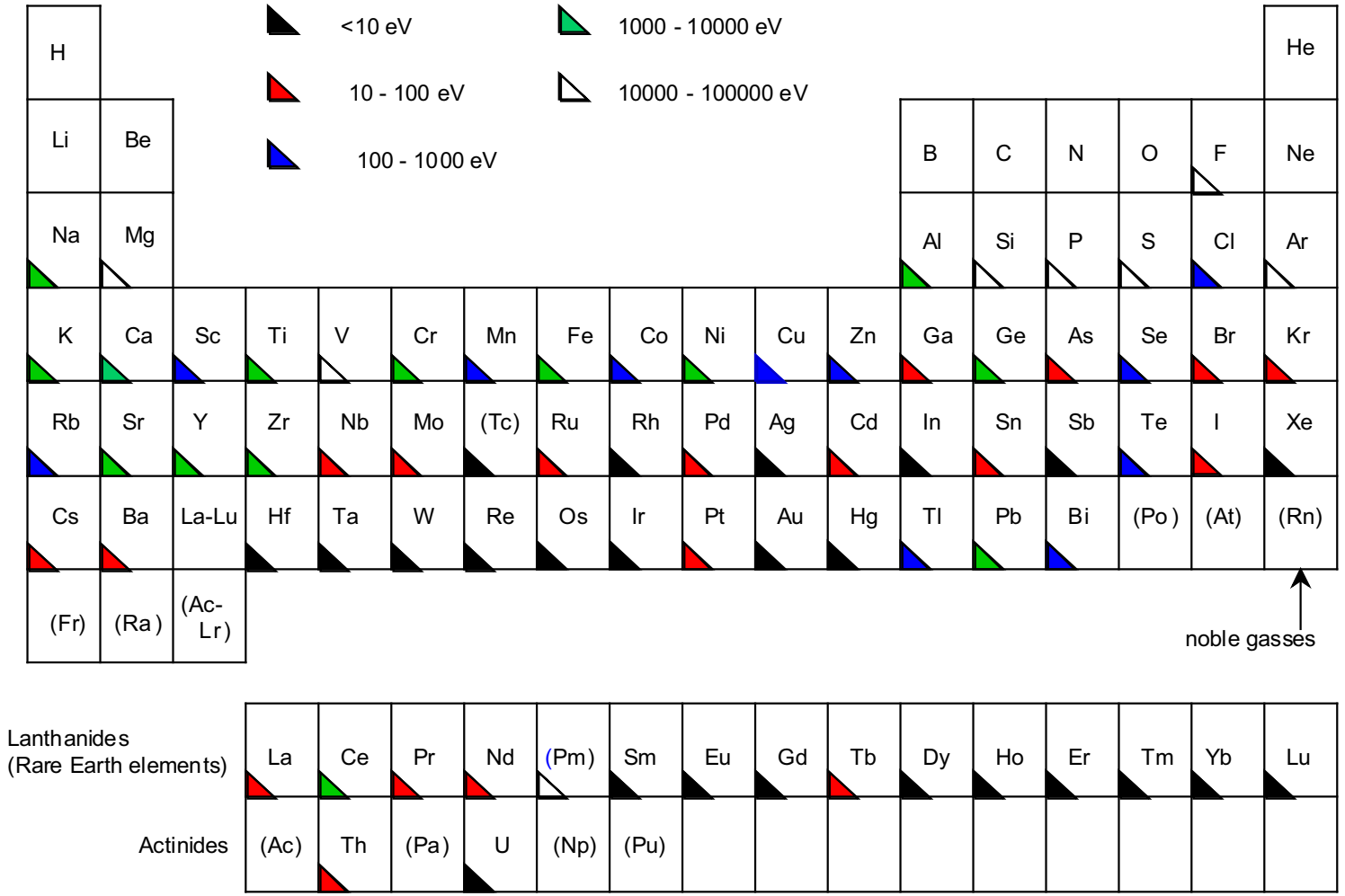
- Principle
  - Resonances for analysis of elemental composition
  - NRCA and NRT
- Data Analysis
  - Calibration approach
  - Methodological approach
- Applications
- NRA and PGAA
- Conclusions

Neutron Resonances for elemental identification



- Neutron resonances appear at given energies, specific for each nuclide
- Nuclides can be identified and the elemental (and isotopic) composition can be deduced
- Applicable for almost all elements
- No sample preparation required
- Non - Destructive
- Negligible residual activation

NRA feasibility



(I)NAA

Intense thermal neutron flux (irradiation in core, i.e. BR1)
Gamma detector: gamma ray energy resolution ⇒ Ge-detectors

PGNAA

Intense neutron beam, (guided cold neutron beam Budapest, NIST)
Gamma detector: gamma ray energy resolution ⇒ Ge-detectors

NRA

Pulsed white neutron beam (LINAC)
Gamma/Neutron detector: good time resolution ⇒ scintillators

Pulsed Neutron Beam

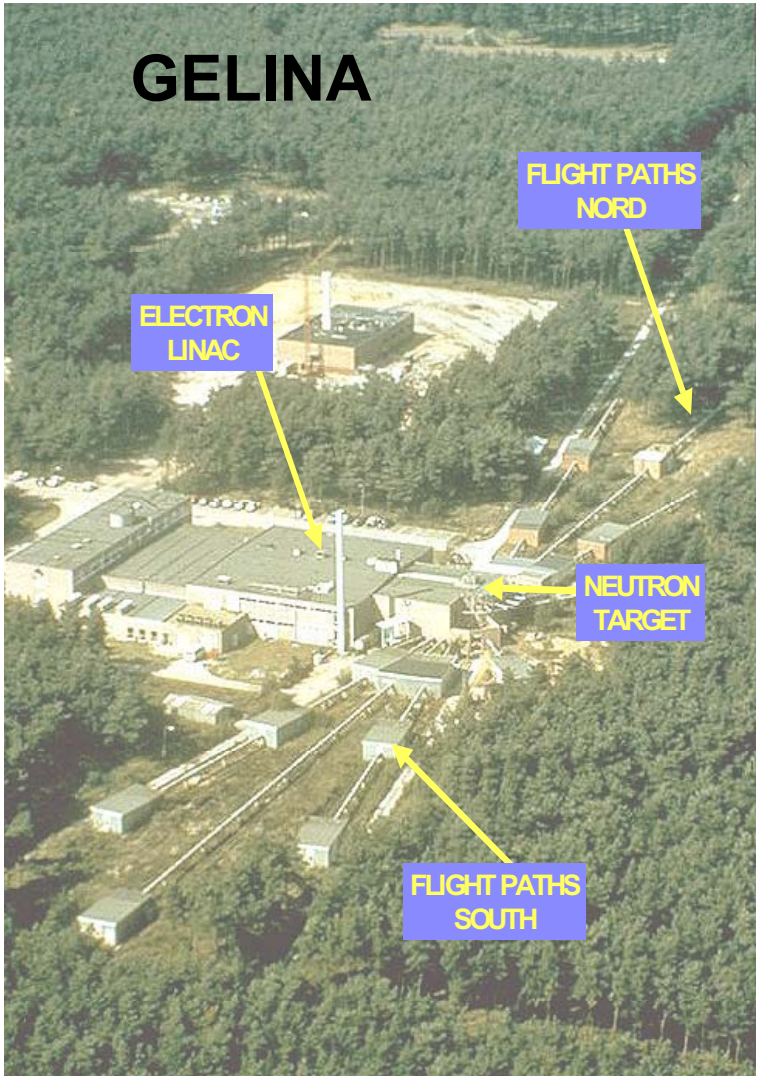
White Neutron Energy Spectrum

TOF $\Leftrightarrow$ E_n

$$E_n = \frac{1}{2} m_n v_n^2 = \frac{1}{2} m_n \left(\frac{L}{t}\right)^2$$

Good time resolution

High neutron flux



Optimum measurement conditions

Intensity

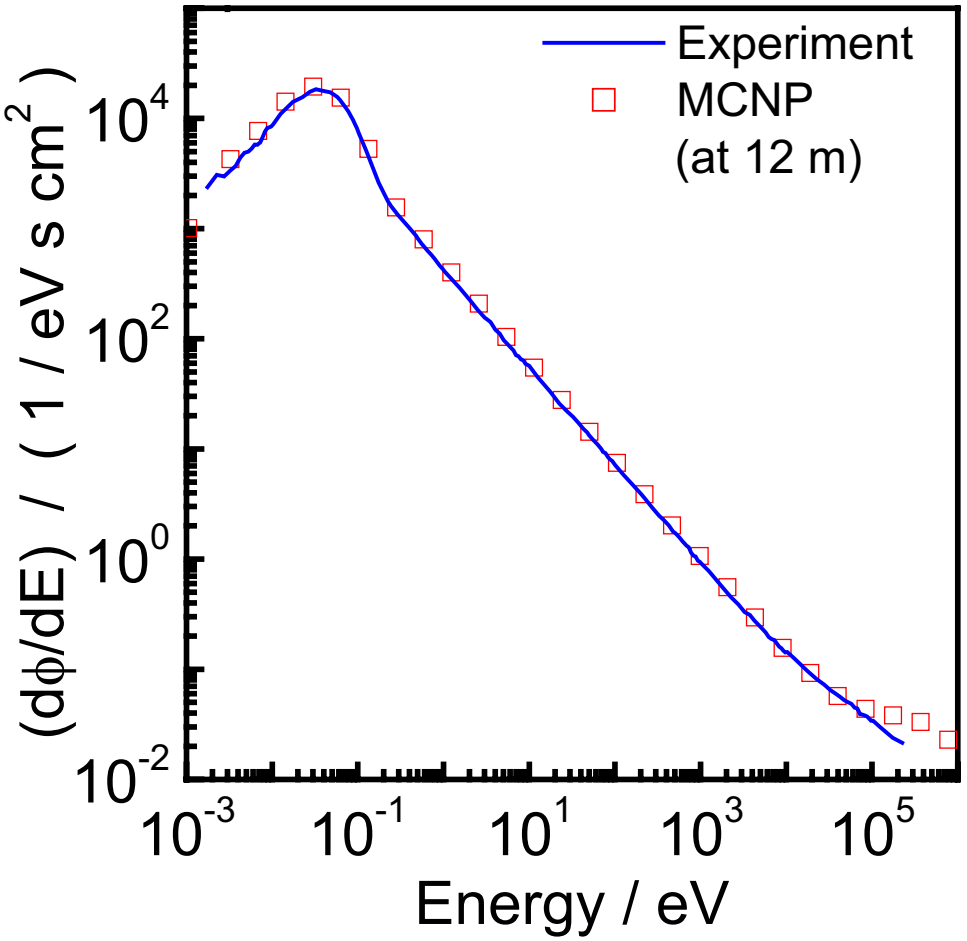
$$\varphi_n(E_n) \propto \frac{1}{L^2}$$

$L\uparrow \quad \Rightarrow \quad \varphi_n \downarrow$

Resolution

$$R_n = \frac{\Delta E_n}{E_n} = 2\sqrt{\frac{\Delta T_n}{T_n} + \frac{\Delta L}{L}}$$

$L\uparrow \Rightarrow R_n \downarrow$



Optimum measurement conditions

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$L\uparrow \quad \Rightarrow \quad \varphi_n \downarrow$

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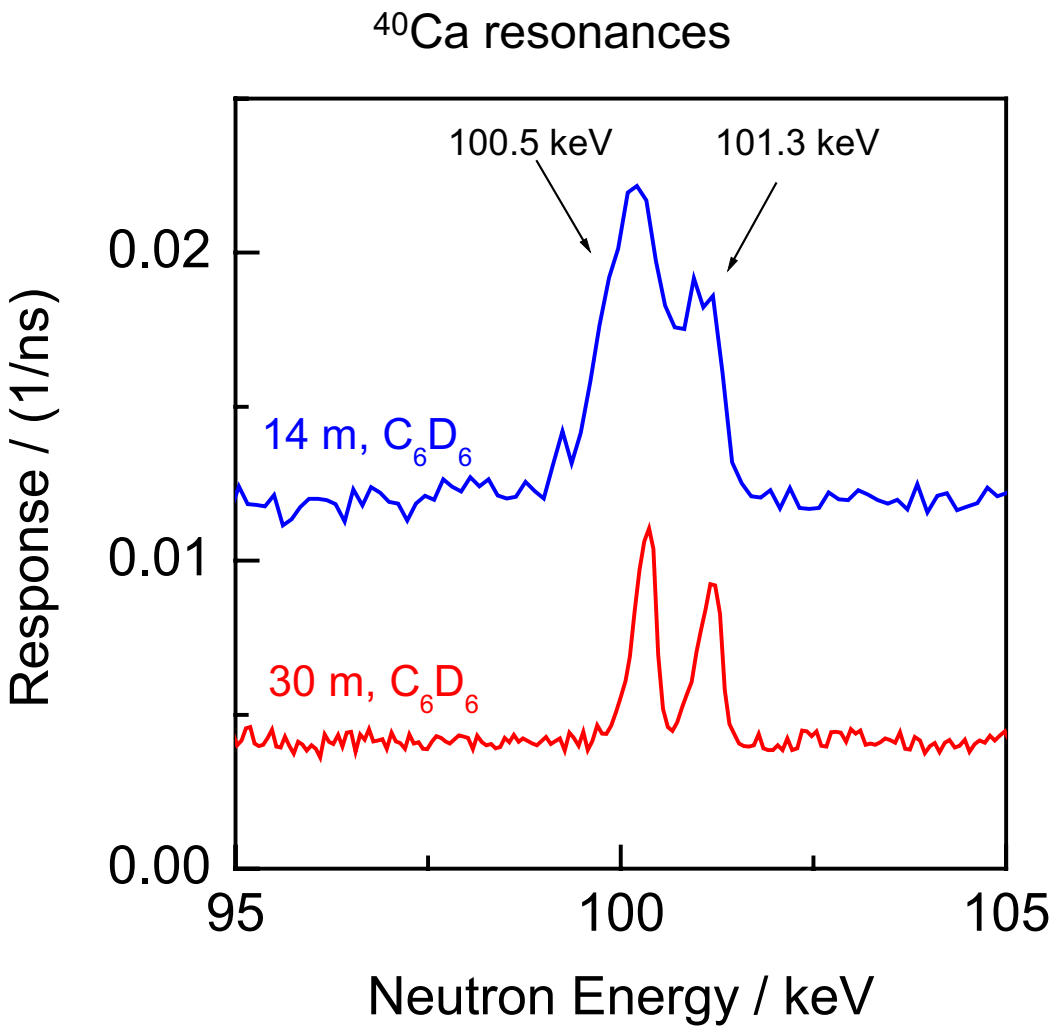
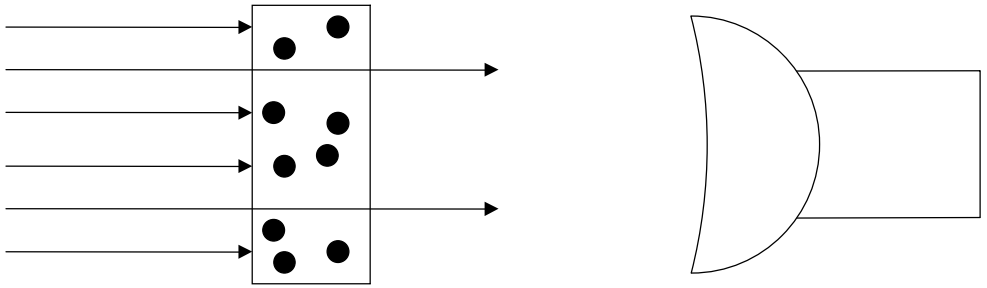


Perego R. et al, MTAA11, June 2004

Detector Response - Transmission (NRT)



Detector

$$T = \frac{C_{in}}{C_{out}} \cong e^{-n\sigma_{tot}}$$

no correction for efficiency

no normalisation required

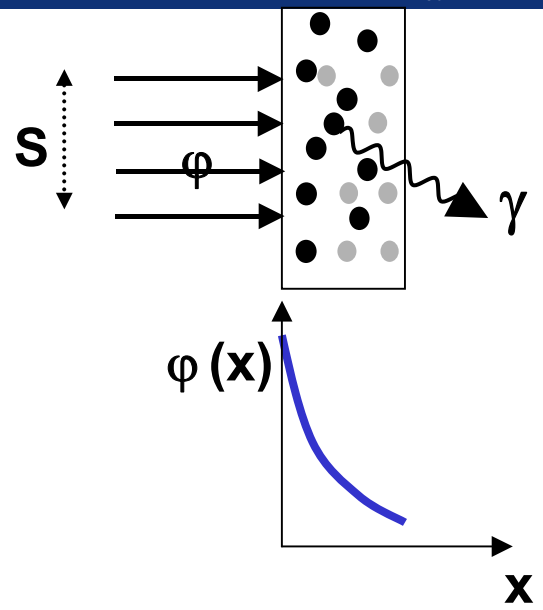
but good collimation required

- **All detected neutrons must have crossed the sample**
- **All neutrons scattered by the sample may not reach the detector**

low sensitivity (exponential + potential scattering)

Detector Response - Capture (NRCA)

$$R_c = \varepsilon_c\, n_x\, \sigma_\gamma\, F\, \varphi\, S$$



- ε_c detection efficiency for capture event
- n_x number of nuclei per cm^2 for nuclide x
- σ_c capture cross section
- **F** self-shielding factor
- **S** Effective area
- φ neutron flux

$$F = \left(\frac{1 - e^{-n\sigma_t}}{n\sigma_t} \right)$$

More sensitive but the data analysis is more complicated

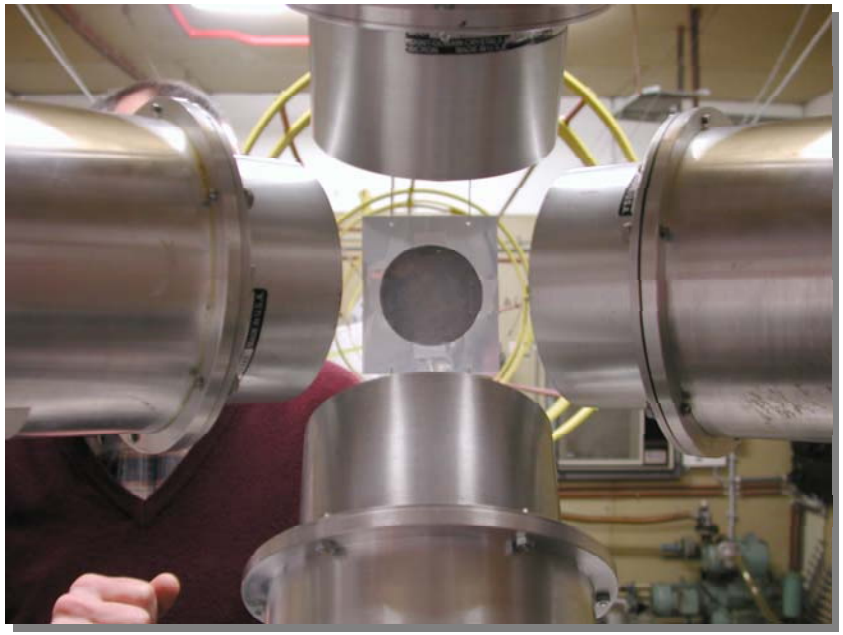
Detectors for NRCA and NRT

NRCA

Gamma Detectors - C_6D_6, YAP

Good time resolution (1 ns)

Low neutron sensitivity

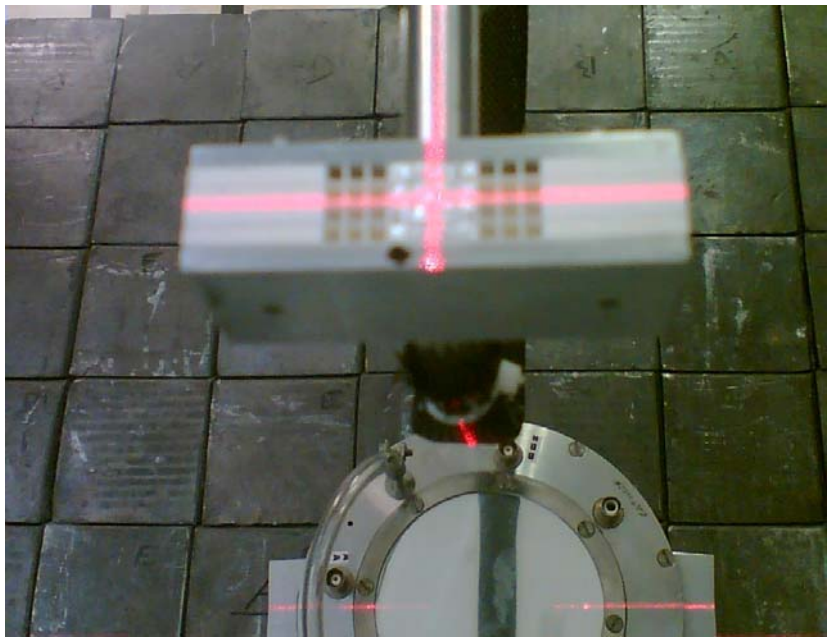


NRT

Neutron Detectors - Li glass NE905

Good time resolution

PSND (developed at CCLRC)



Traditional approach
Area analysis

$$R_c = \varepsilon_c \, n_x \, \sigma_\gamma \, F \, \varphi \, S \quad \Rightarrow \quad C_c = \varepsilon_c \, n_x \, A_\gamma \, F \, \varphi \, S$$

$\Rightarrow$ only isolated resonances are used

Only valid for relatively weak resonances

neglect neutron scattering in the sample

Calibration

requires representative calibration samples to determine unknown and F

Limited in applications (no complex spectra, samples,…)

Limited use of information contained within the spectrum

However, Very successful (e.g. Archeology, H. Postma)

$$\frac{C_{r1,x}}{C_{r2,y}} = \frac{\varepsilon_{r1,x}}{\varepsilon_{r2,y}} \frac{F_{r1}}{F_{r2}} \frac{A_{\gamma,r1}}{A_{\gamma,r2}} \frac{\varphi_{r1}}{\varphi_{r2}} \frac{n_x}{n_y}$$

$$A_{\gamma,r} = 4.097 \times 10^6 \frac{g\Gamma_n\Gamma_\gamma}{E_r\Gamma} \text{(b)}$$

- **φ neutron flux (φ_{r1}/ φ_{r2}) (only shape required!!)**
  $\Rightarrow$ Independent measurement by e.g. $^{10}B(n,\alpha)^7Li$
- **Resonance capture area ($A_{\gamma,r1}$ & $A_{\gamma r,2}$)**
  $\Rightarrow$ From nuclear data libraries
- **ε detection efficiency for capture event (ε_{r1} / ε_{r2})**
  $\Rightarrow$ By calibration with standard samples (no correction for attenuation in sample)
- **Self - shielding factors (F_{r1} / F_{r2})**
  $\Rightarrow F=F(n,\sigma_{tot})$

Self – shielding correction

$$F_r = \left(\frac{1 - e^{-n\sigma_t}}{n\sigma_t} \right)$$

$n\sigma_t << 1 \Rightarrow F \cong 1$

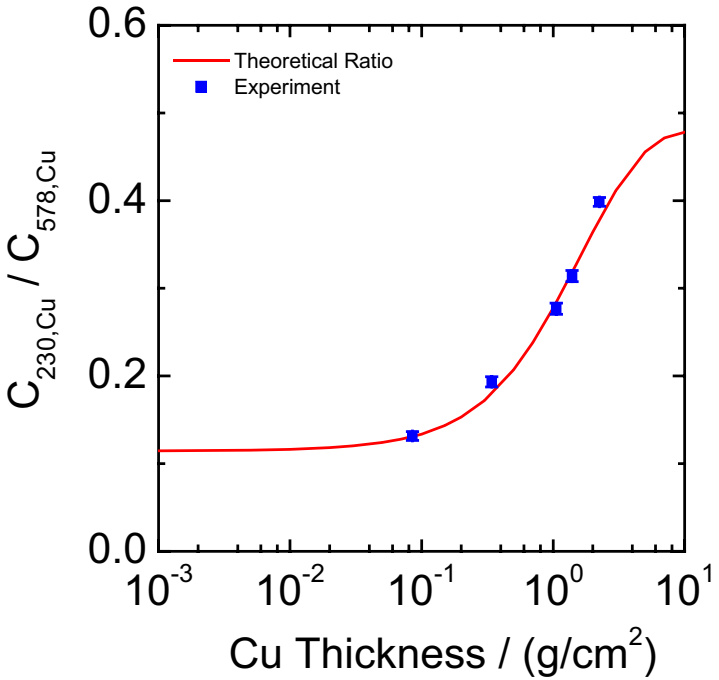
for main elements use weaker resonances

for traces use stronger resonances

Looking at resonances of the same element ($n_x = n_y$)

$$\frac{C_{r1,x}}{C_{r2,x}} = \underbrace{\frac{\varepsilon_{r1,x}}{\varepsilon_{r2,x}} \frac{\varphi_{r1}}{\varphi_{r2}} \frac{A_{\gamma,r1}}{A_{\gamma,r2}}}_{\text{independent of n}} \frac{F_{r1}(n_x, \sigma_{t1})}{F_{r2}(n_x, \sigma_{t2})}$$

$$\frac{C_{r1,x}}{C_{r2,x}} = \frac{1}{K} f(n_x, \sigma_{t,r1}, \sigma_{t,r2})$$



$\Rightarrow$ Thickness (n) from combination of resonances with different strength !!!

H. Postma et al., Czech. J. Phys., 33 (2003) A233

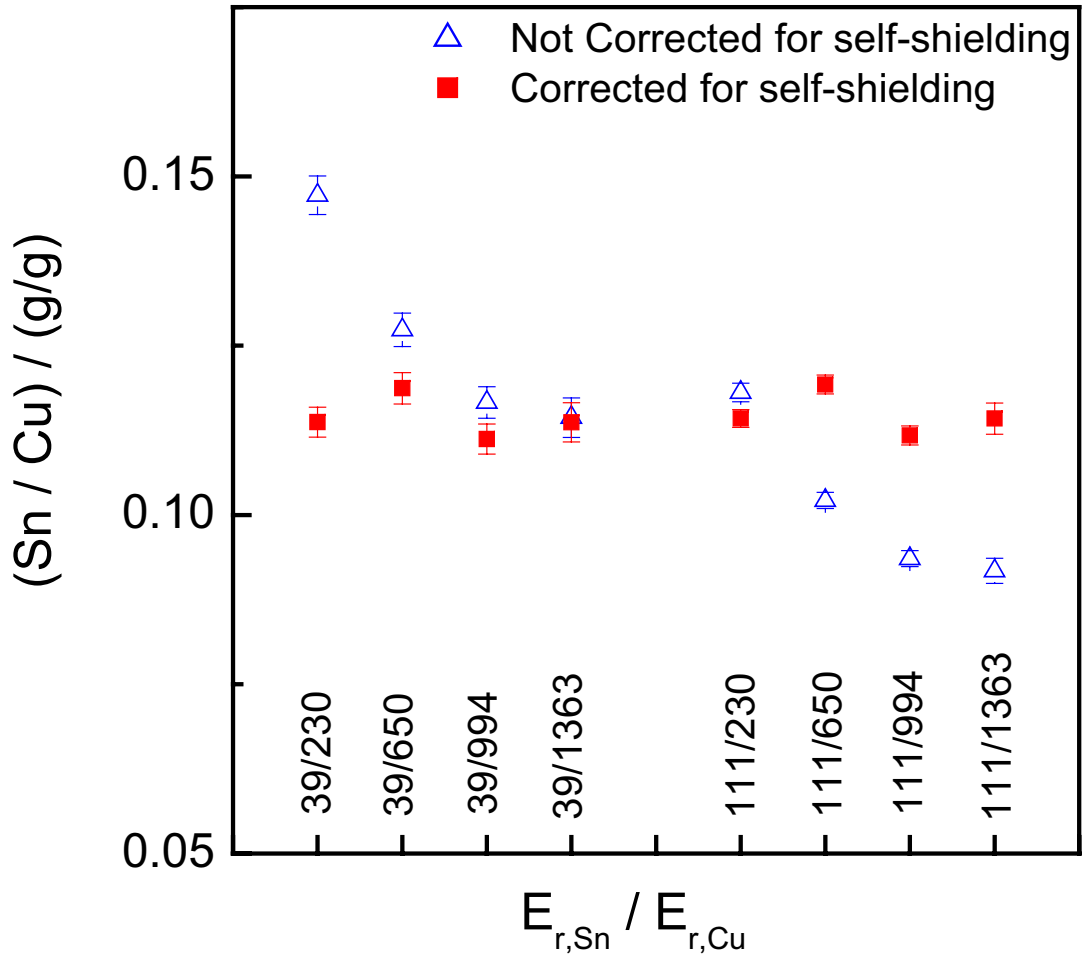
Verification of self-shielding correction

Sn

39 eV ^{117}Sn
111 eV ^{116}Sn

Cu

230 eV ^{65}Cu
650 eV ^{63}Cu
994 eV ^{63}Cu
1363 eV ^{65}Cu



H. Postma et al., MTAA11, 2004

Methodological Approach (REFIT, M. Moxon)

NRCA requires capture yield, Yc

Correction for the shape of the incoming neutron flux

Account for detection efficiency of the gamma event

(PHWT $\Rightarrow$ NIM A 577 (2007) 626-640)

Advantages:

Resonance Shape Analysis (R-matrix) vs area analysis

Correction for self - shielding and multiple scattering

Resolution of TOF - spectrometer and Doppler broadening

The whole energy spectrum can be used to assess the nuclide abundance

Simultaneous fit of several data sets (both NRT and NRCA)

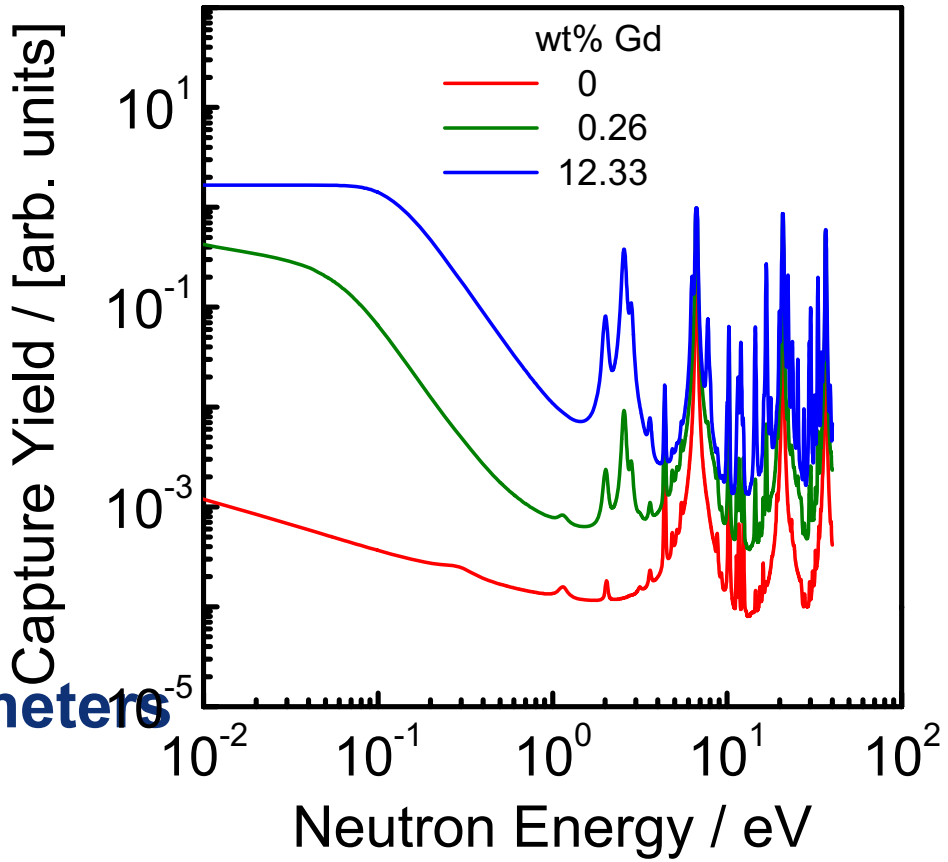
Easy to handle multi-elemental homogenous samples

Resonance Shape Analysis code: REFIT

G is an analytical model describing the observed capture yield:

$$Y_c = G(R, T, RP, F, \mu, S_x^*, n_x, S_y^*, n_y, \ldots)$$

- R resolution of spectrometer
- T temperature (Doppler broad.)
- RP nuclear data to deduce σ_c, σ_t
- F self - shielding
- μ multiple scattering
- S_x^* binding energy nucleus x
- n_x atom density of nucleus x



$\Rightarrow$ **n_x, n_y, … : adjustable fitting parameters**

Archaeology

- Zn/Sn, *Cu/Pb* ratio
- *Ancient Charm Project (2D and 3D scanning)*

Trace Analysis

- *P/Ca in bones, Cl/Ca in marbles*

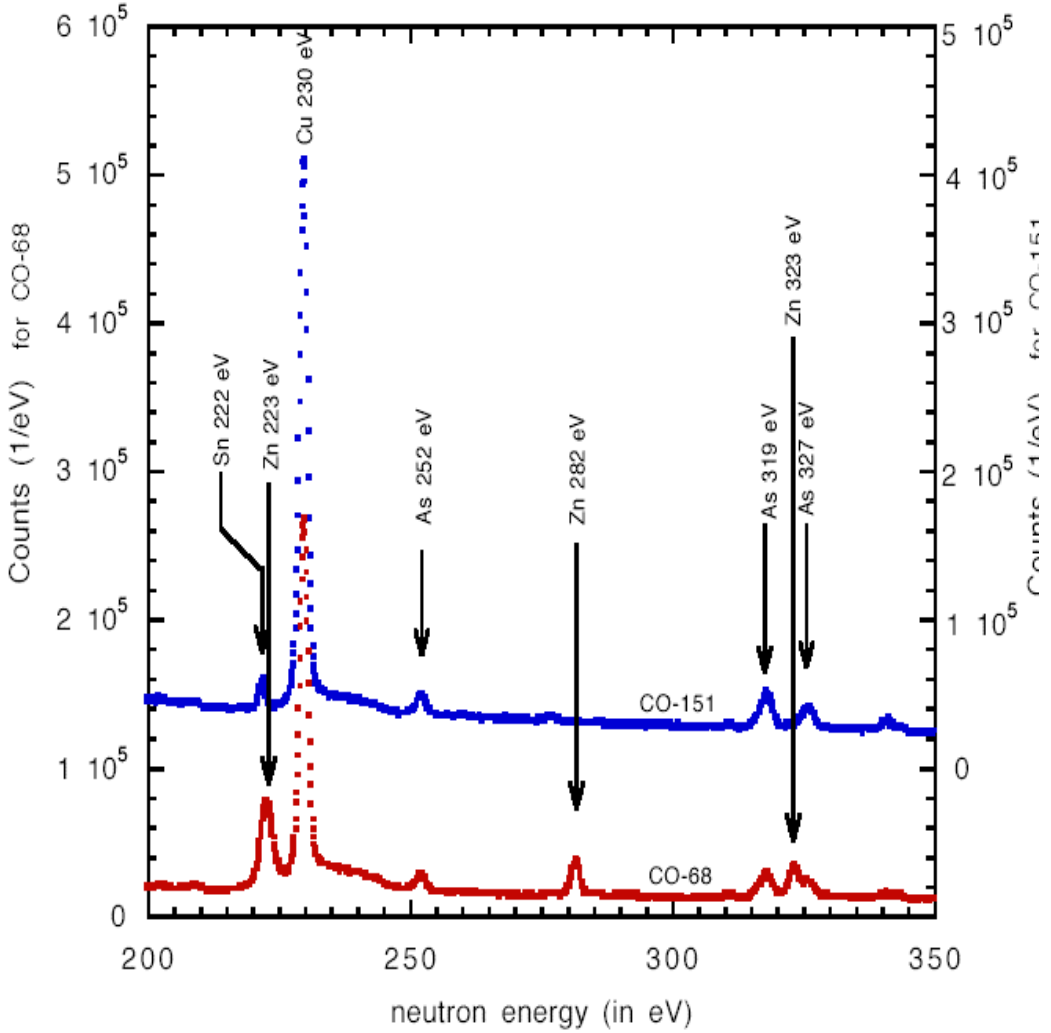
Determination of neutron poison in uranium

- Gd in U

Characterisation of reference materials

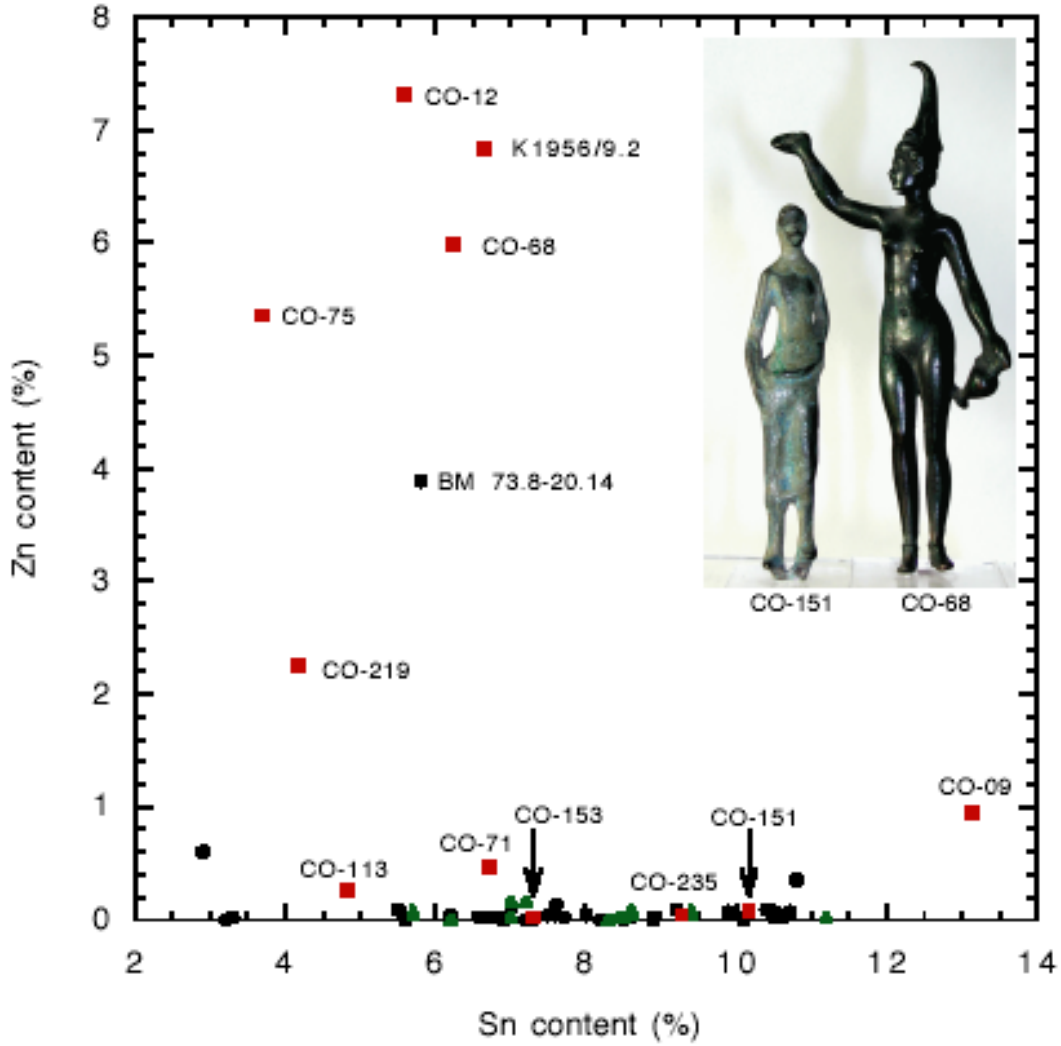
- Ag in Bi, Y_2O_3, *Sb in Pb,* ^{103}Rh, Pb_2I

Determination of the composition of special alloys



Postma H., Schillebeeckx P. and Halbertsma R.B., Archaeometry 46 (2004) 365

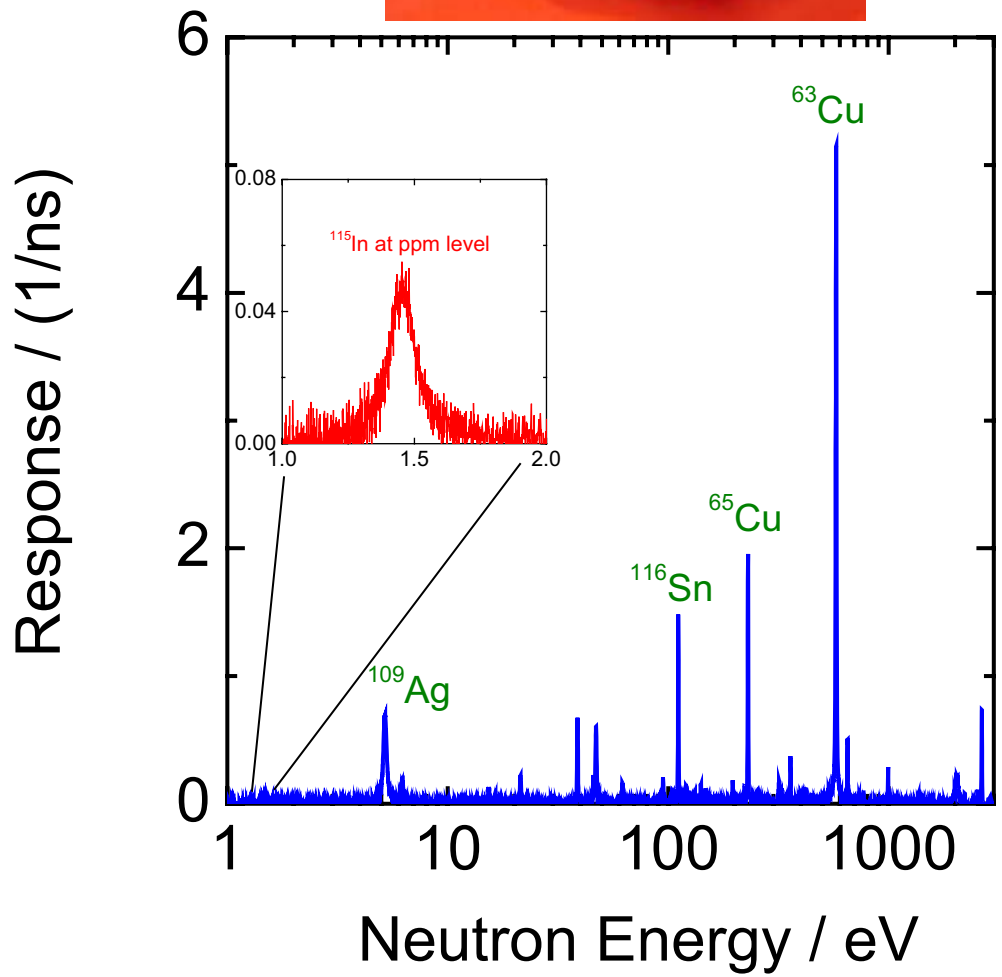
Archaeology: Authenticity of Etruscan artefacts



Archaeology NRCA of Geistingen Axes

Cat Nr.	Find spot	Element									
		Cu	Sn	As	Sb	Ag	Fe	Ni	Co	In	Pb
B550	Maastricht	94.30	0.06	0.67	1.63	0.54				5.1 ppm	< 2
B551	Maastricht	93.38	0.64	0.91	2.14	0.56				14.8 ppm	< 2
K787	Kleve	93.01	0.98	1.64	3.02	1.36				o.o.r	< 2
K788	Kleve	86.24	6.54	2.32	2.66	2.10			0.15	o.o.r	< 2
B562	Nijmegen	84.51	9.07	1.01	2.10	2.21	0.61		0.49	o.o.r	< 2
B557	Vierlingsbeek	80.93	12.72	0.06	0.01	0.04	6.20			o.o.r	< 2

Postma H. et al., Achaeologia protohistorica XIII (2005) 41-46



Element	Fractions (%)		Isotope	Resonance (eV)
Cu	**77.76**	**(0.11)**		
			^{63}Cu	579.0
			^{65}Cu	230.0
Sn	**20.85**	**(0.10)**		
			^{112}Sn	94.8
			^{116}Sn	111.2
			^{117}Sn	38.8
			^{118}Sn	45.7
			^{119}Sn	222.6
			^{120}Sn	427.5
			^{122}Sn	1756.0
			^{124}Sn	62.0
As	**0.34**	**(0.01)**		
			^{75}As	47.0
Sb	**0.196**	**(0.021)**		
			^{121}Sb	6.24
			^{123}Sb	21.4
Ag	**0.090**	**(0.01)**		
			^{107}Ag	16.3
			^{109}Ag	5.2
Fe	**0.770**	**(0.09)**		
			^{56}Fe	1147.4
In	**0.0061**	**(0.0003)**		
			^{115}In	1.46

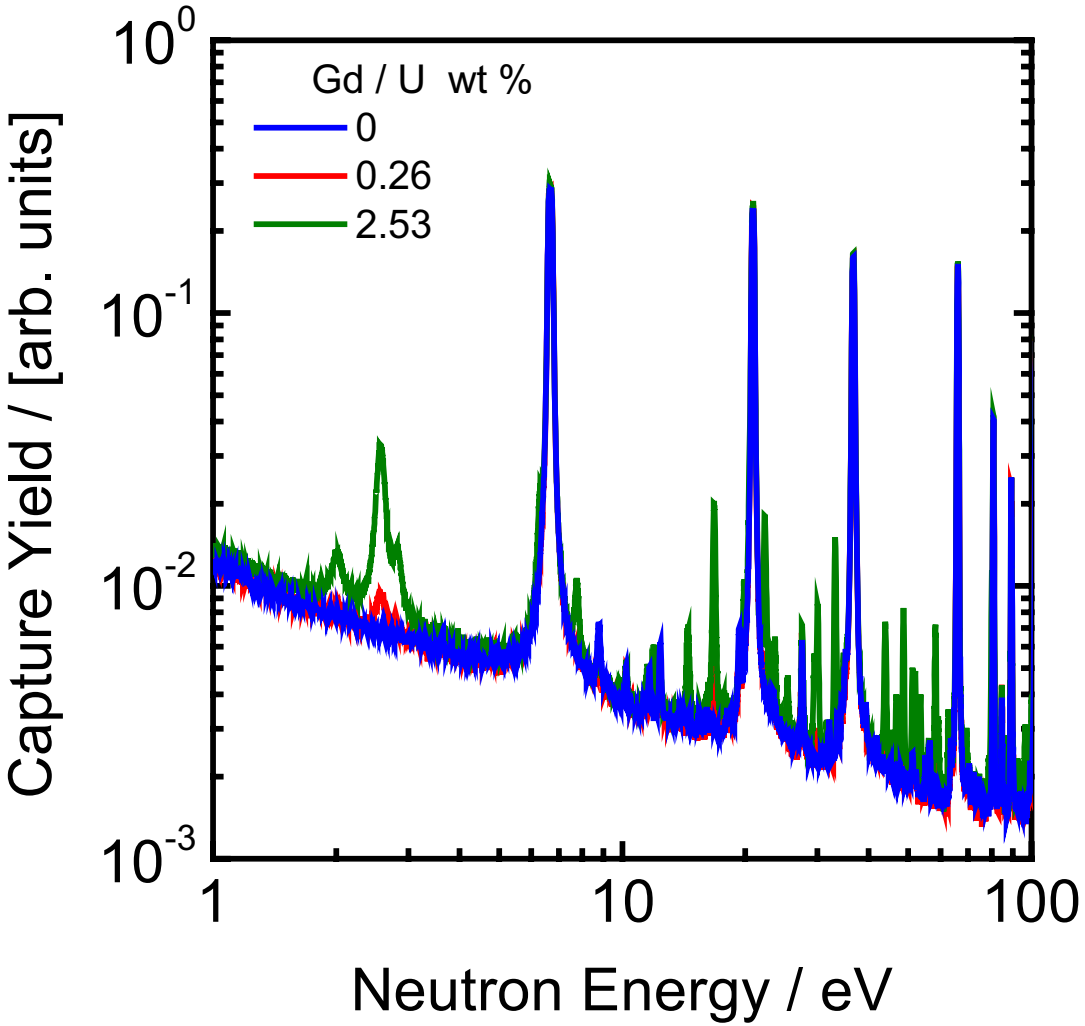
m_{NRCA} = 13.0 (0.5) g

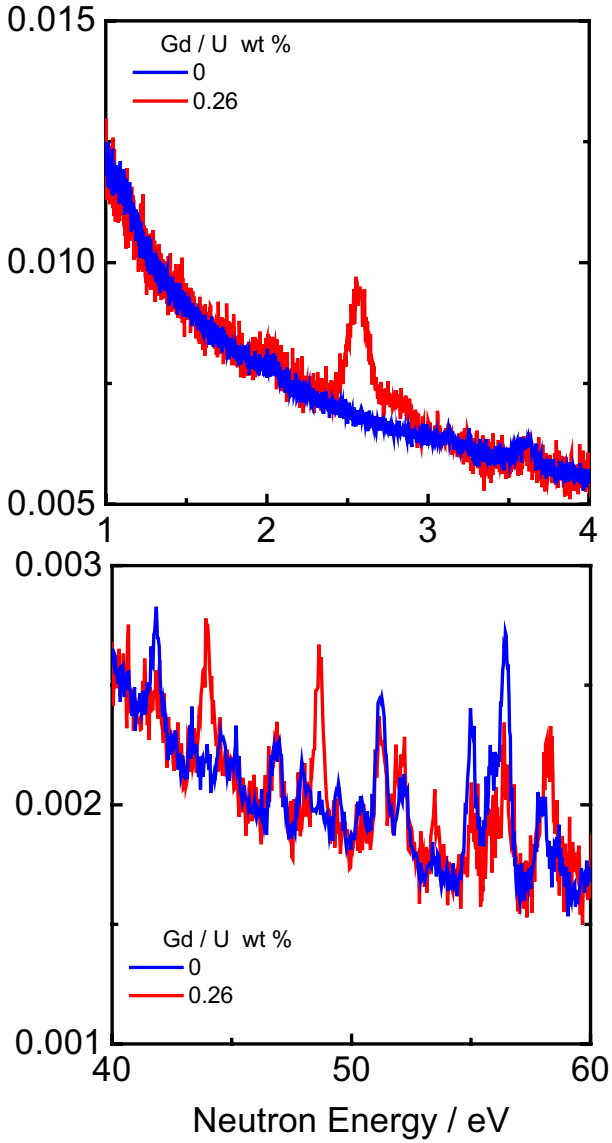
m_{weigth} = 13.25 g

Postma H. et al., Int. Symposium on Archaeometry, Amsterdam, April 2002

NRCA: Neutron poison in U samples

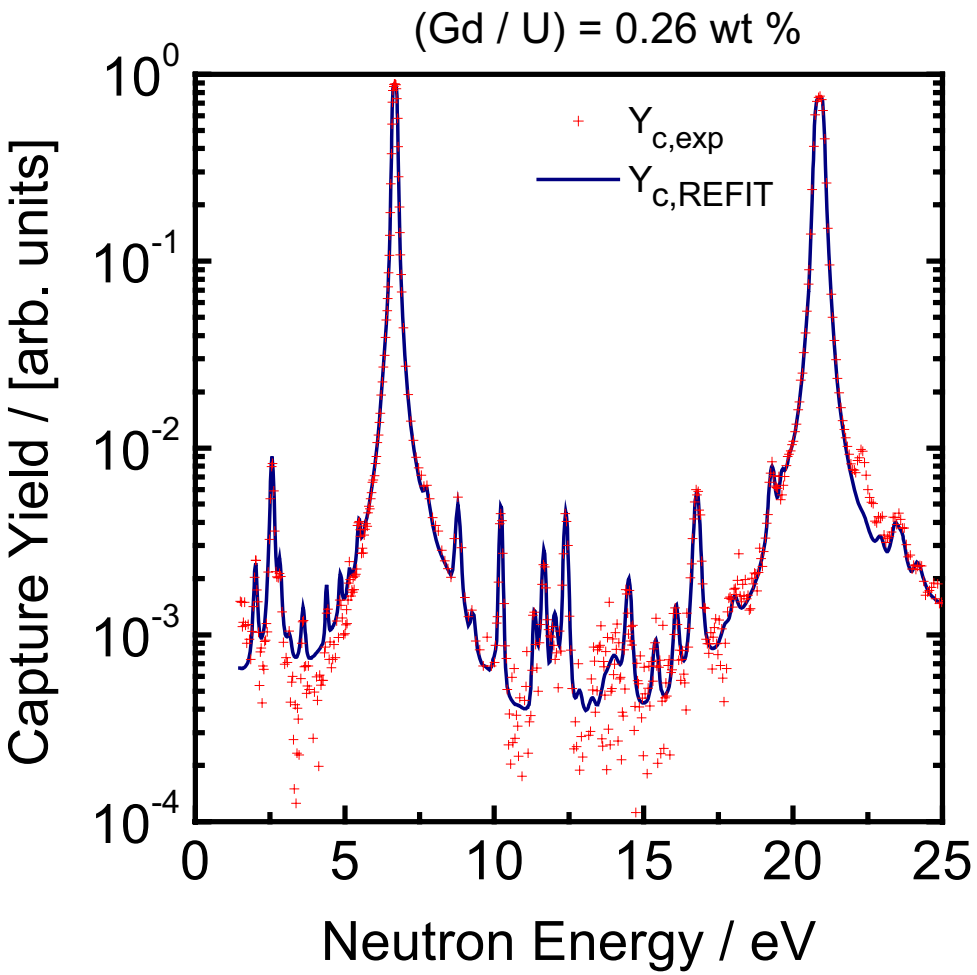
$^{dep}U_3O_8$ + $^{nat}Gd_2O_3$ powder

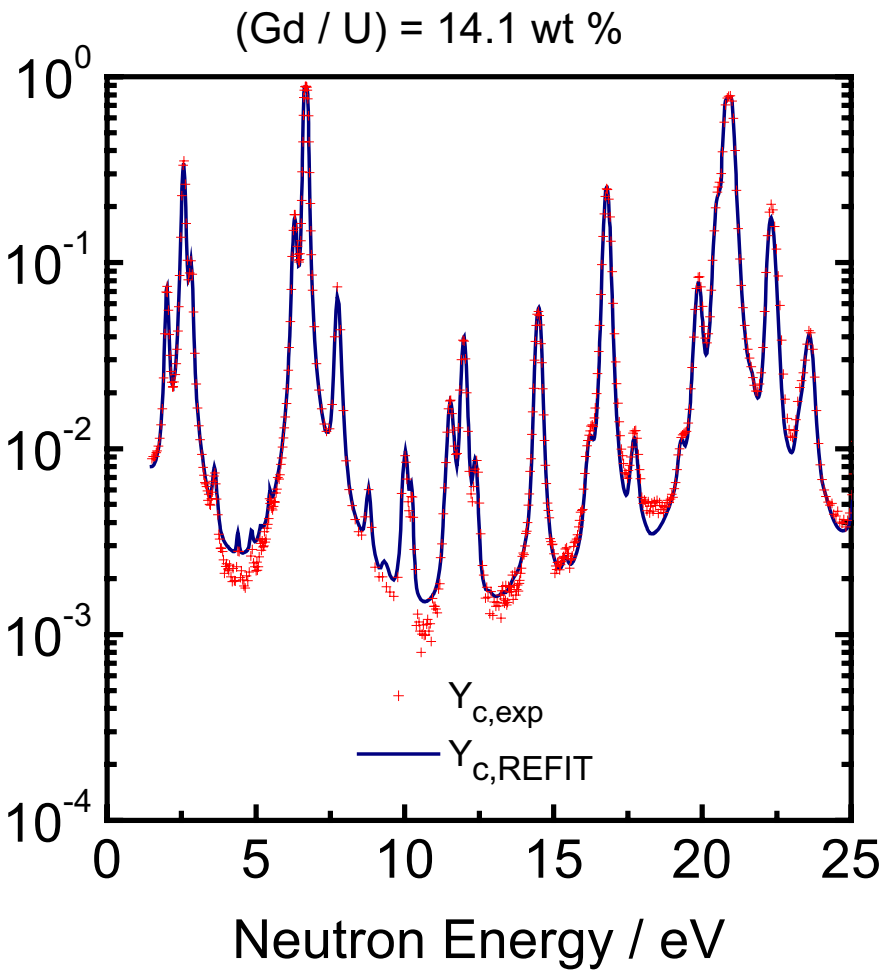




Determination of Gd in U - fit

NRCA: Neutron poison in U samples





NRCA: Neutron poison in U samples

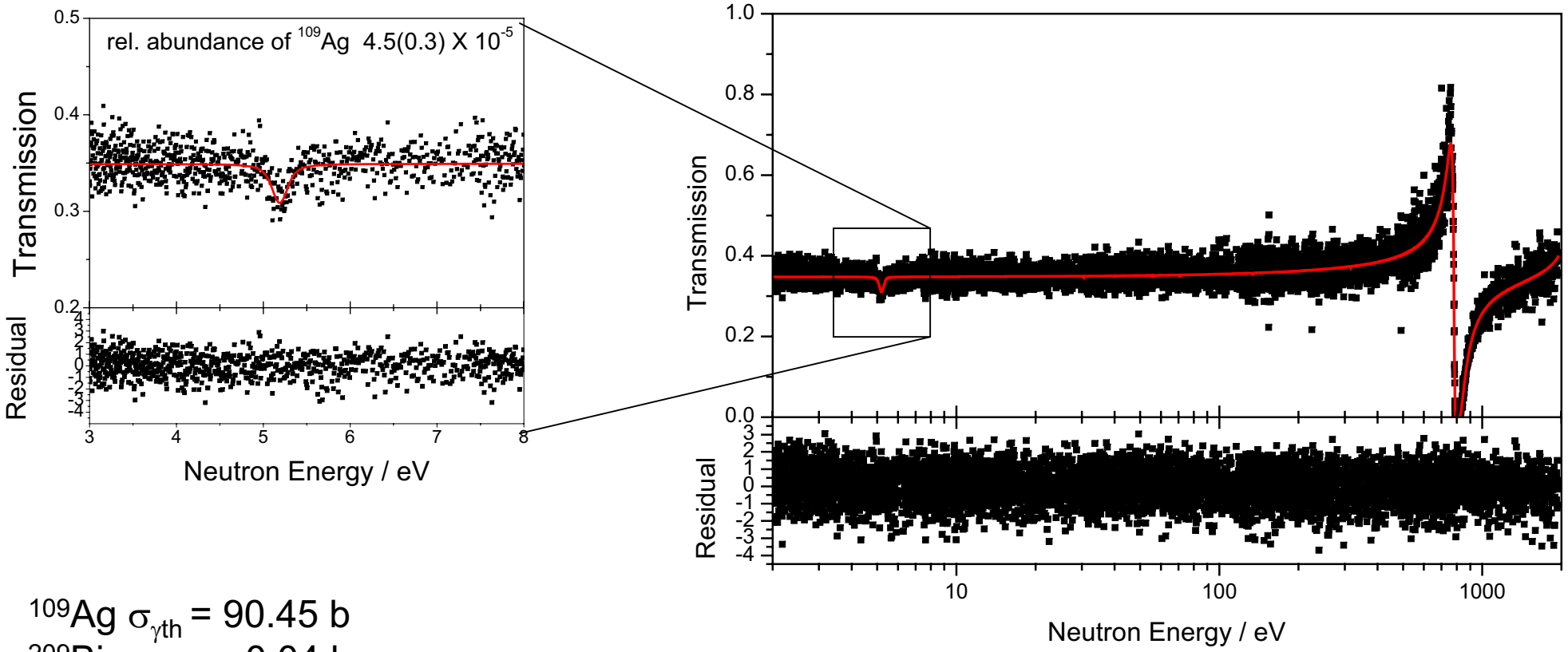
$$^{dep}U_3O_8 + {}^{nat}Gd_2O_3$$

U / g	Gd / g	n(^{155}Gd) / n(^{238}U)		n(^{157}Gd) / n(^{238}U)	
		declared abund.	NRCA	declared abund.	NRCA
20.988	0.0536	$5.77\ 10^{-4}$	$(5.76 \pm 0.04)\ 10^{-4}$	$6.10\ 10^{-4}$	$(6.59 \pm 0.07)\ 10^{-4}$
20.608	0.5206	$5.71\ 10^{-3}$	$(5.73 \pm 0.01)\ 10^{-3}$	$6.03\ 10^{-4}$	$(6.53 \pm 0.02)\ 10^{-3}$
18.656	2.6240	$3.13\ 10^{-2}$	$(3.14 \pm 0.01)\ 10^{-2}$	$3.36\ 10^{-2}$	$(3.51 \pm 0.03)\ 10^{-2}$

$\Rightarrow$ nuclear data ^{157}Gd

Ag in bismuth sample (NRT, S. Kopecky)

NRT for sample characterization



^{109}Ag $\sigma_{\gamma th}$ = 90.45 b

^{209}Bi $\sigma_{\gamma th}$ = 0.04 b

Without accounting for Ag (107 and 109) the $\sigma_{\gamma th}$ would overestimated by 15%

Characterisation of Reference Materials
^{103}Rh metal disc

	Natural abundance (wt %)	Relative Amount (wt %)	
^{103}Rh	100	99.5137	
^{181}Ta	99.988	0.0337	(0.0029)
^{191}Ir	37.3	0.0870	(0.0033)
^{193}Ir	62.7	0.1478	(0.0076)
^{182}W	26.3	0.0552	(0.0027)
^{183}W	14.3	0.0302	(0.0028)
^{186}W	28.6	0.0613	(0.0025)
^{197}Au	100	0.0059	(0.0011)

Impurities contribute for 0.5 % to the observed count rate in the thermal energy region

^{103}Rh(n_{th},γ) cross section is requested with an accuracy < 2%

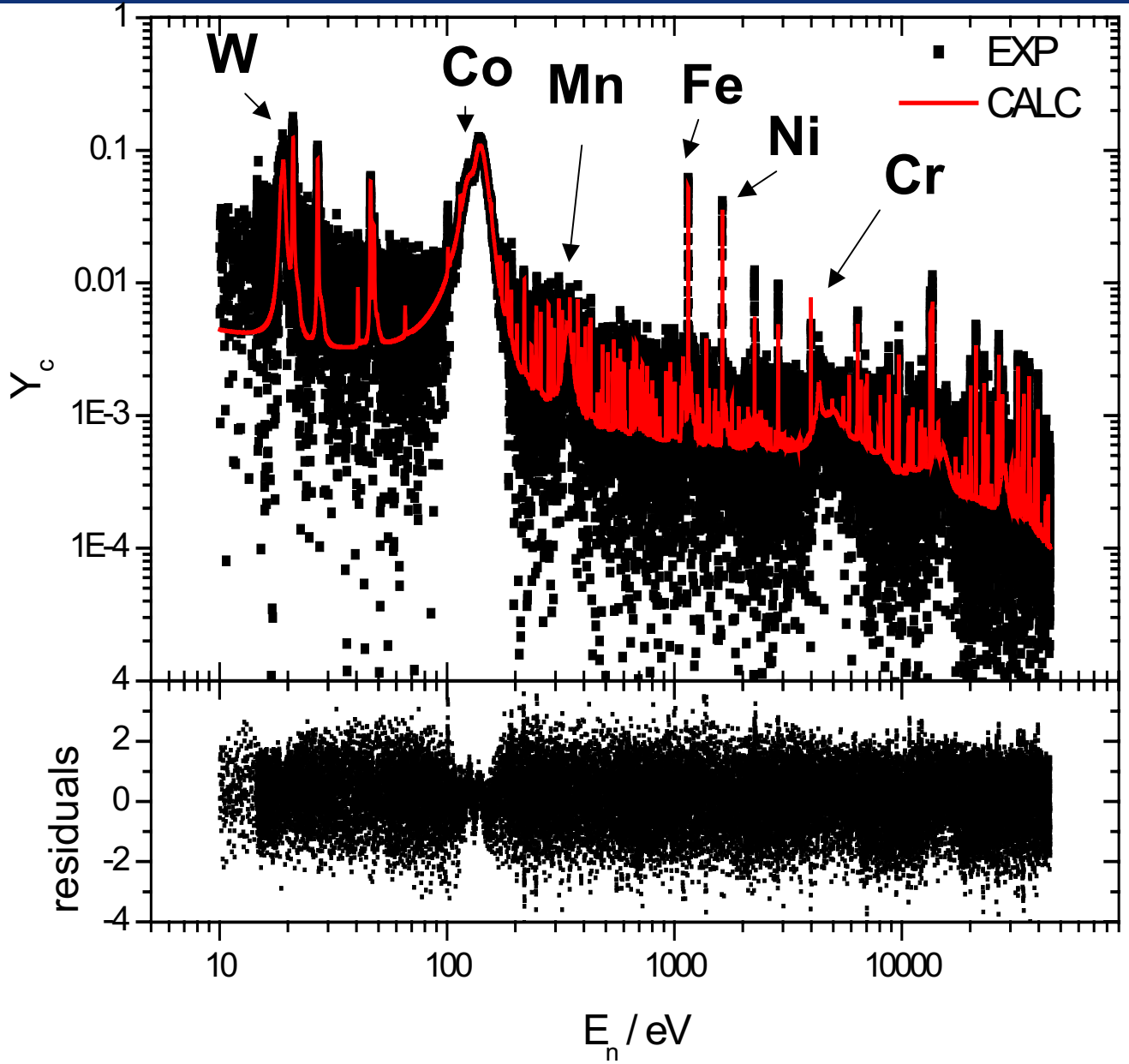
Postma H. et al.,, ND2004, Santa Fe, September 2004

Characterisation of Reference Materials PbI_2 samples from reprocessed fuel

Elememt		NRCA		ICP-MS		(I)NAA	
Iodine	Total	20.5	(0.8)	19.9	(0.5)		
	^{127}I	3.4	(0.1)	3.4	(0.1)	3.4	(0.1)
	^{129}I	17.1	(0.8)	16.5	(0.5)		
Lead	Total	53.5	(3.0)	59.5	(0.2)	51.1	(1.8)
	^{204}Pb	0.8					
	^{206}Pb	12.8	(0.5)				
	^{207}Pb	12.1	(0.3)				
	^{208}Pb	27.8	(3.0)				
Oxygen		15.2	(0.8)	14.5	(1.5)		
Sulfur		6.2	(0.4)				

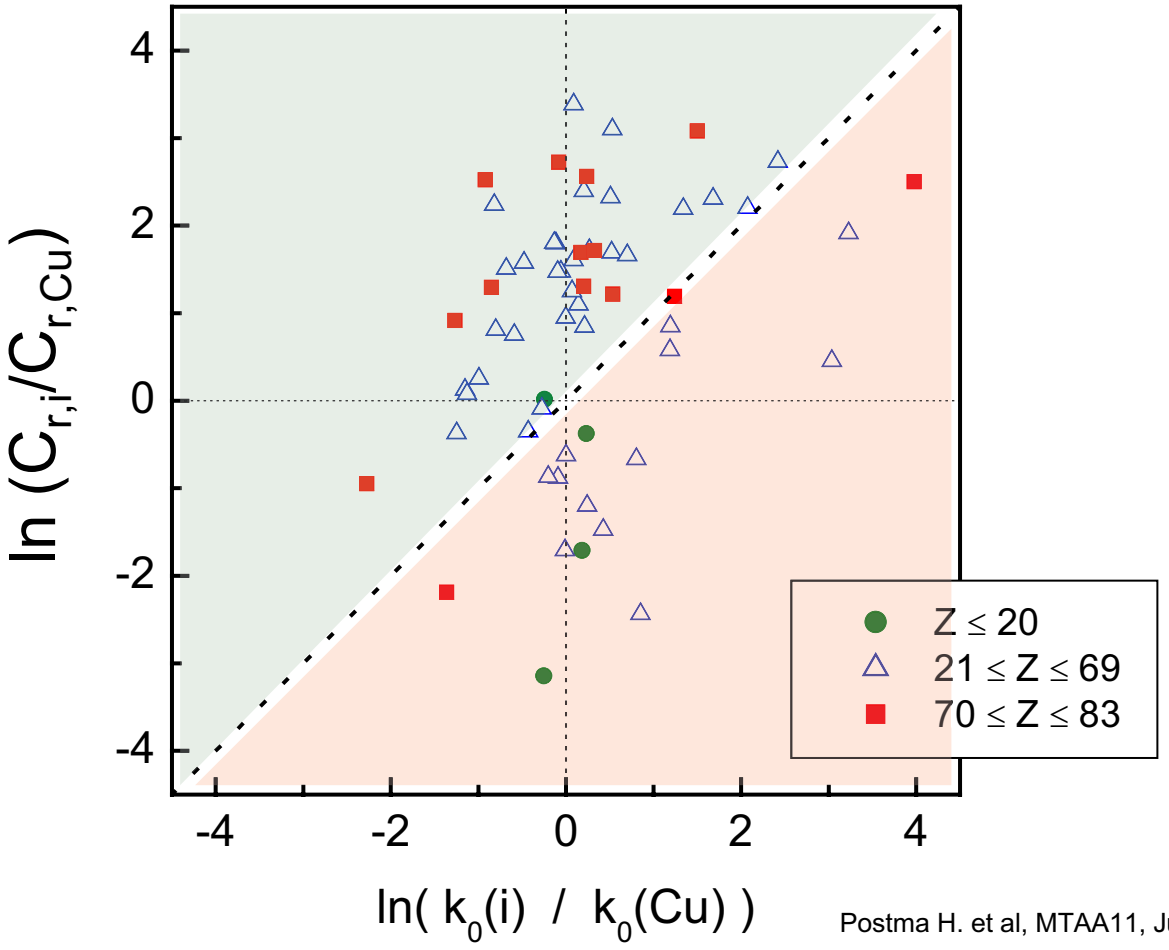
Noguere G., NIMA 575 (2007) 476-488

NRCA: special alloys composition



% w	Z
0.738	Mn
4.36	W
24.38	Cr
20.77	Fe
33.6	Ni
14.27	Co
1.13	Si

PGAA (at Budapest) and NRCA (at GELINA)
Accuracy for Cu in a bronze artefact about 1%



Postma H. et al, MTAA11, June 2004

⇒ k_o and C_r relative to Cu

PGAA best for light elements

H, S, P, and K

NRCA best for heavy elements

As, Ag, Sb, Au and Pb

Both NRCA and PGAA are fully non-destructive methods for bulk analysis

The residual activation is negligible, especially for NRCA

PGAA is good for light elements, NRCA better for heavy elements

PGAA can be hampered by an unfavourable balance between the thermal capture cross sections of the elements

⇒ for NRCA we can always choose a region in the TOF-spectrum where the resonance of interest dominates

Remarks - INAA

Pros:

- Multi elemental method
- High accuracy
- Low detection limits
- Most elements can be traced

Cons:

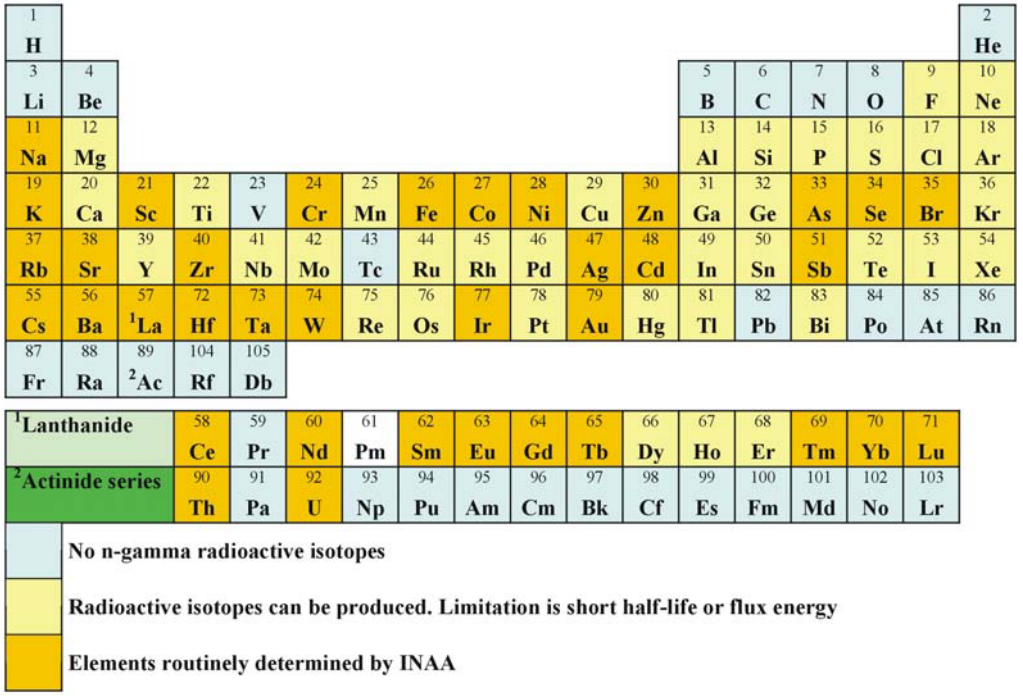
- Reactor (high flux) needed
- Activation, waiting time

Data needed:

- k_0, g_w, I_r

OR

- Thermal cross sections
- Resonance Parameters



Remarks - PGAA

Pros:

- Multi elemental method
- Applicable in principle to all elements
- No special sample preparation

Cons:

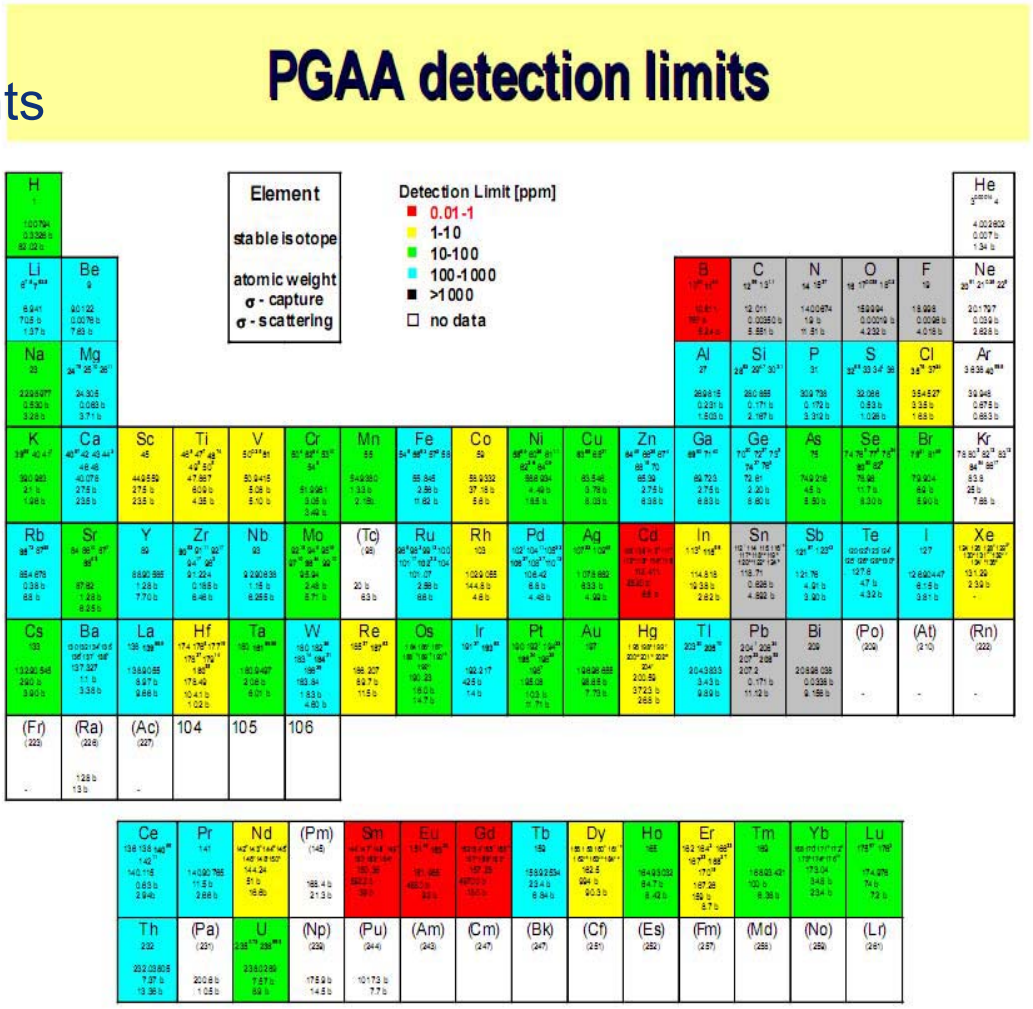
- γ-ray spectra more complex than NAA

Data needed:

- g_w
- k_0

OR

- Thermal cross sections (Partial cross sections)



Remarks - NRA

Pros:

- Multi elemental method
- Applicable to all elements with resonances
- No special sample preparation
- Negligible residual activation

Cons:

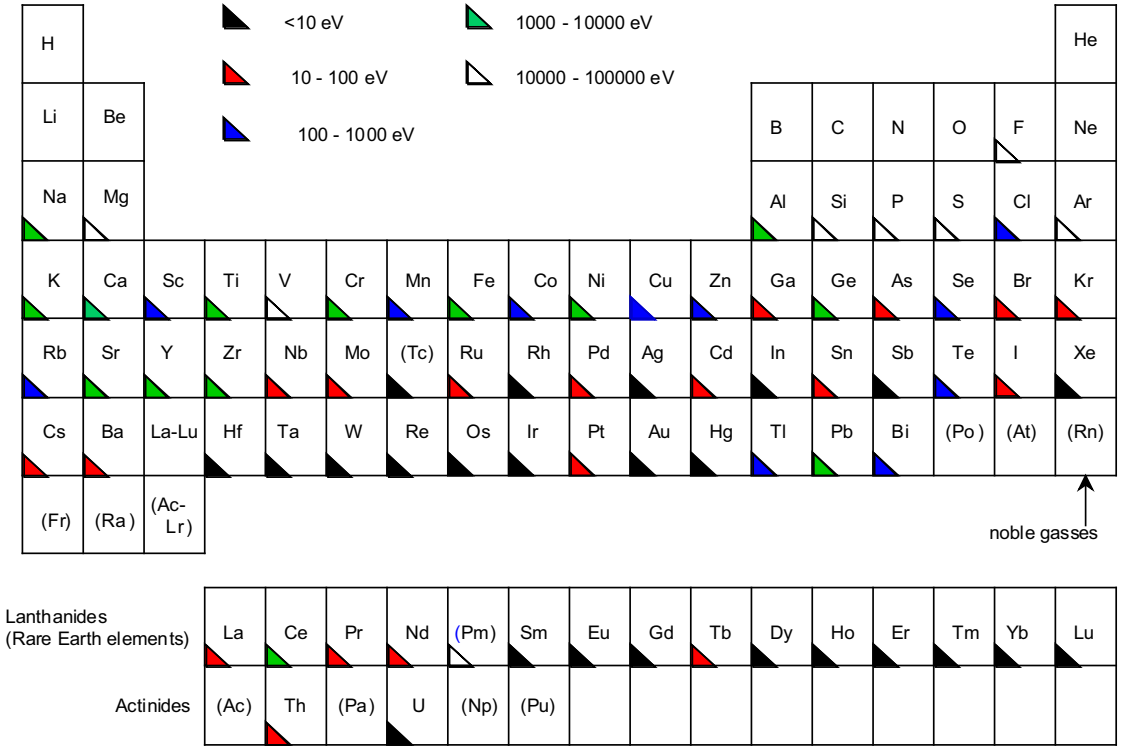
- Data analysis not easy
- Additional measurements with methodological approach

Data needed:

- A_γ

OR

- Resonance Parameters $\Rightarrow \sigma_\gamma(E), \sigma_t(E)$



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