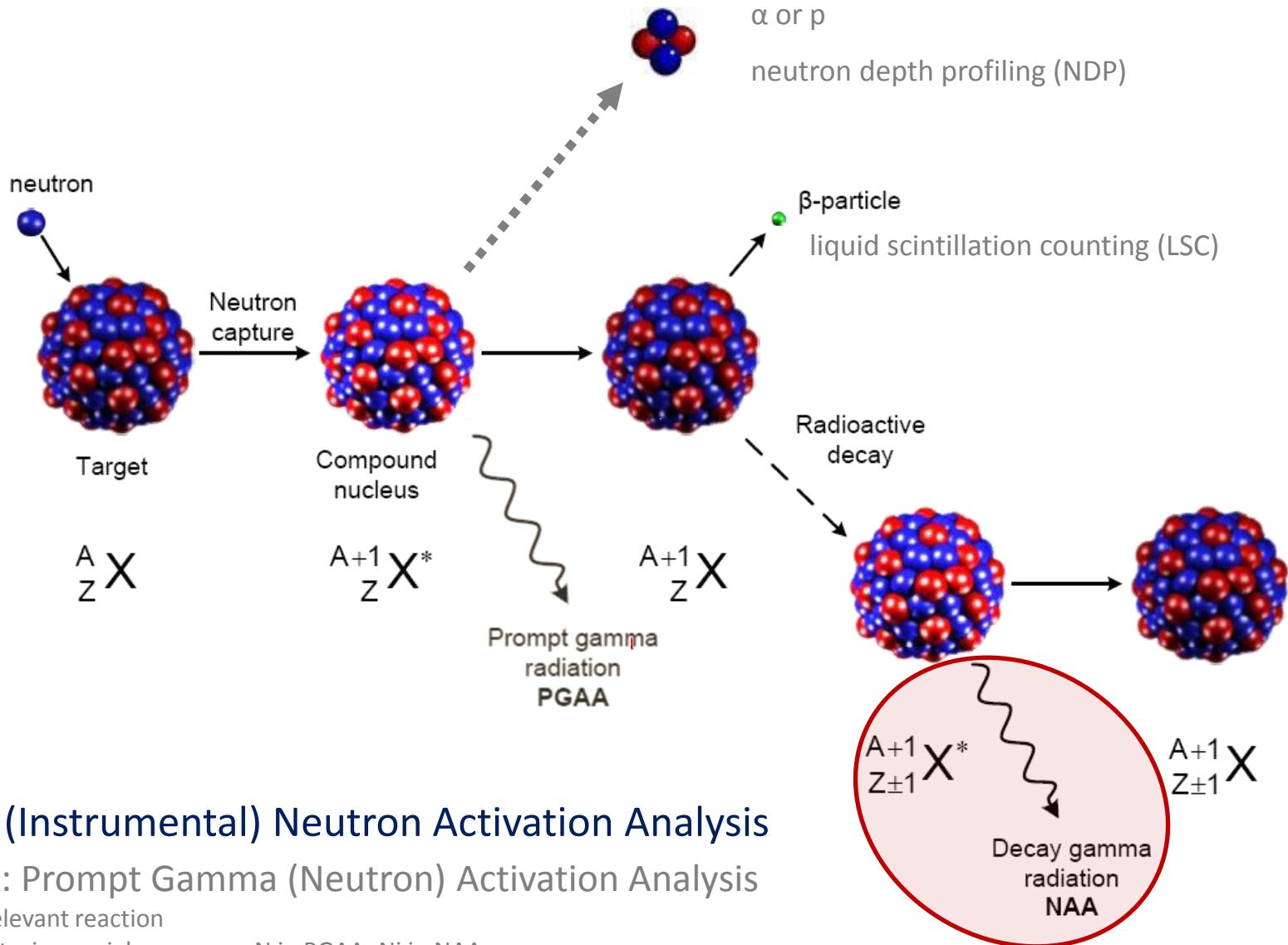


DEVA Workshop: Neutron Activation Analysis

C. Stieghorst, Zs. Révay



MLZ is a cooperation between:



(I)NAA: (Instrumental) Neutron Activation Analysis

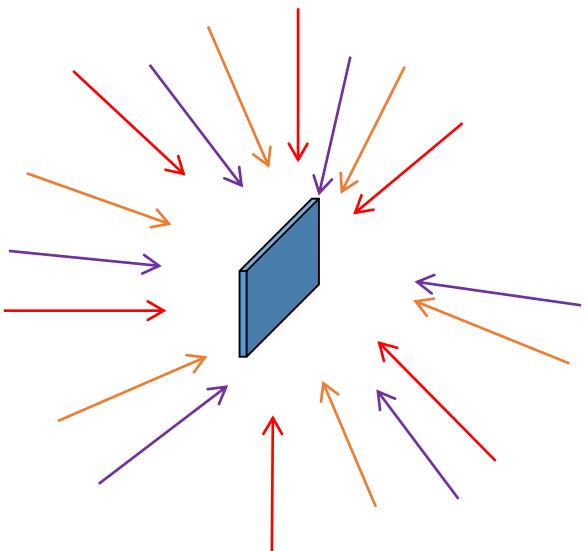
PG(N)AA: Prompt Gamma (Neutron) Activation Analysis

(n, γ) most relevant reaction

(n, p) , (n, α) etc. in special cases, e.g. N in PGAA, Ni in NAA

(n, n') FaNGaS

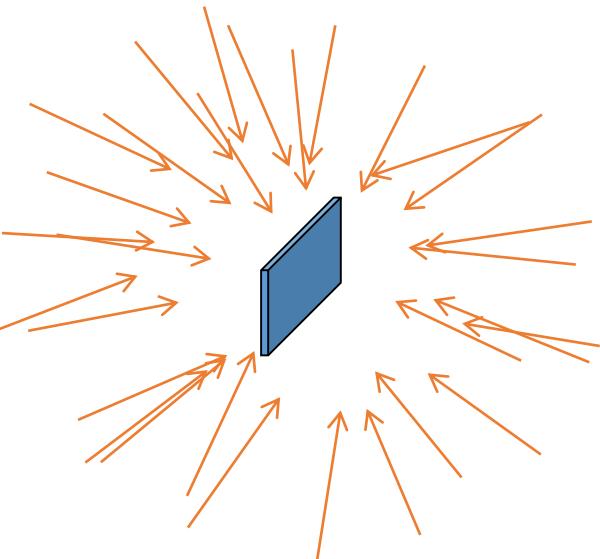
“classic in-core” NAA → instrument NAA



thermal, epithermal,
fast neutrons

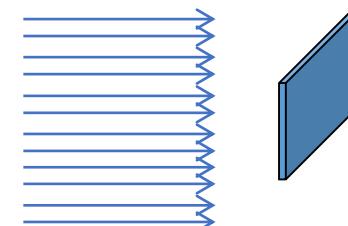
- orders of magnitude higher flux than in-beam
- → very low DLs (+)

FRM II



well-thermalized spectrum

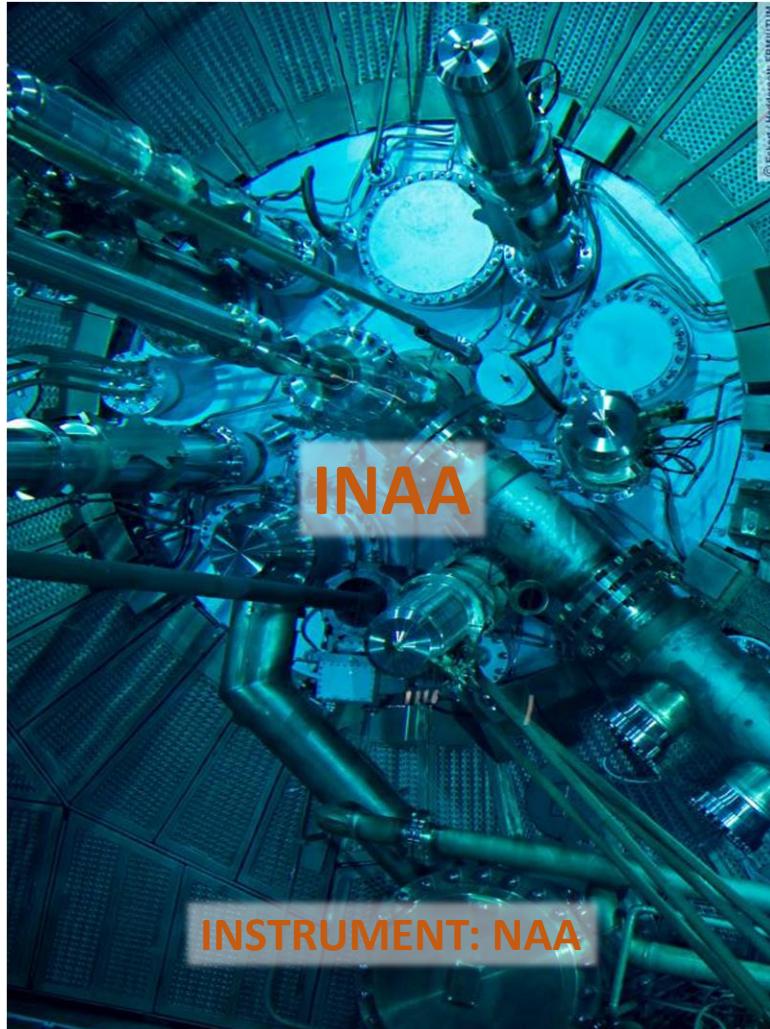
in-beam NAA → option at instrument PGAA



only parallel cold
neutrons

- large samples possible (+)
- attenuation is easy to calculate (+)
- much lower flux (-)

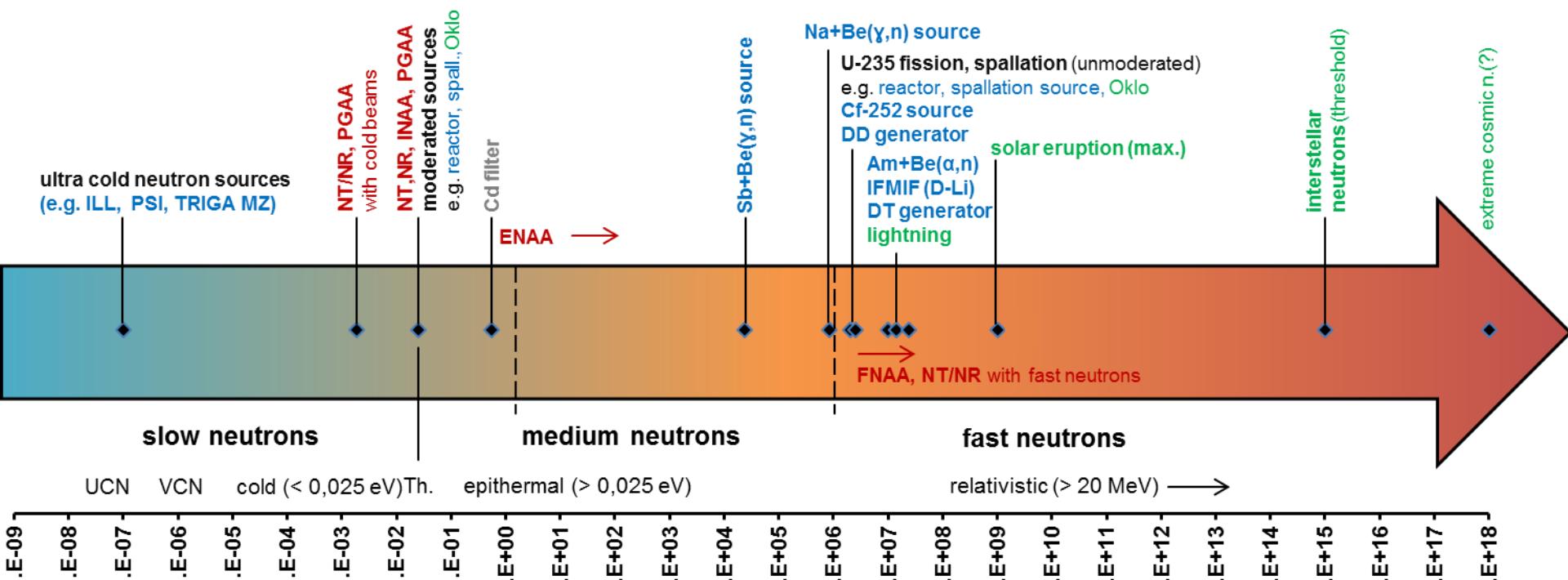
Core-near Positions + RCM Lab



Neutron Guide Hall West: NL4b



Neutron energy range used for elemental analysis:



Methods of chemical analysis with neutrons:

INAA / NAA: instrumental neutron activation analysis incl. cNAA, ib-NAA, ...
RNAA: radiochemical NAA
ENAA: epithermal NAA
FNAA: fast NAA
PGAA / PGNAA: prompt gamma(-ray neutron) activation analysis
PGAI-NT: Prompt Gamma-ray Activation Imaging and neutron tomography
NDP: neutron depth profiling
 further methods: NRA, neutron-induced LSC, ...

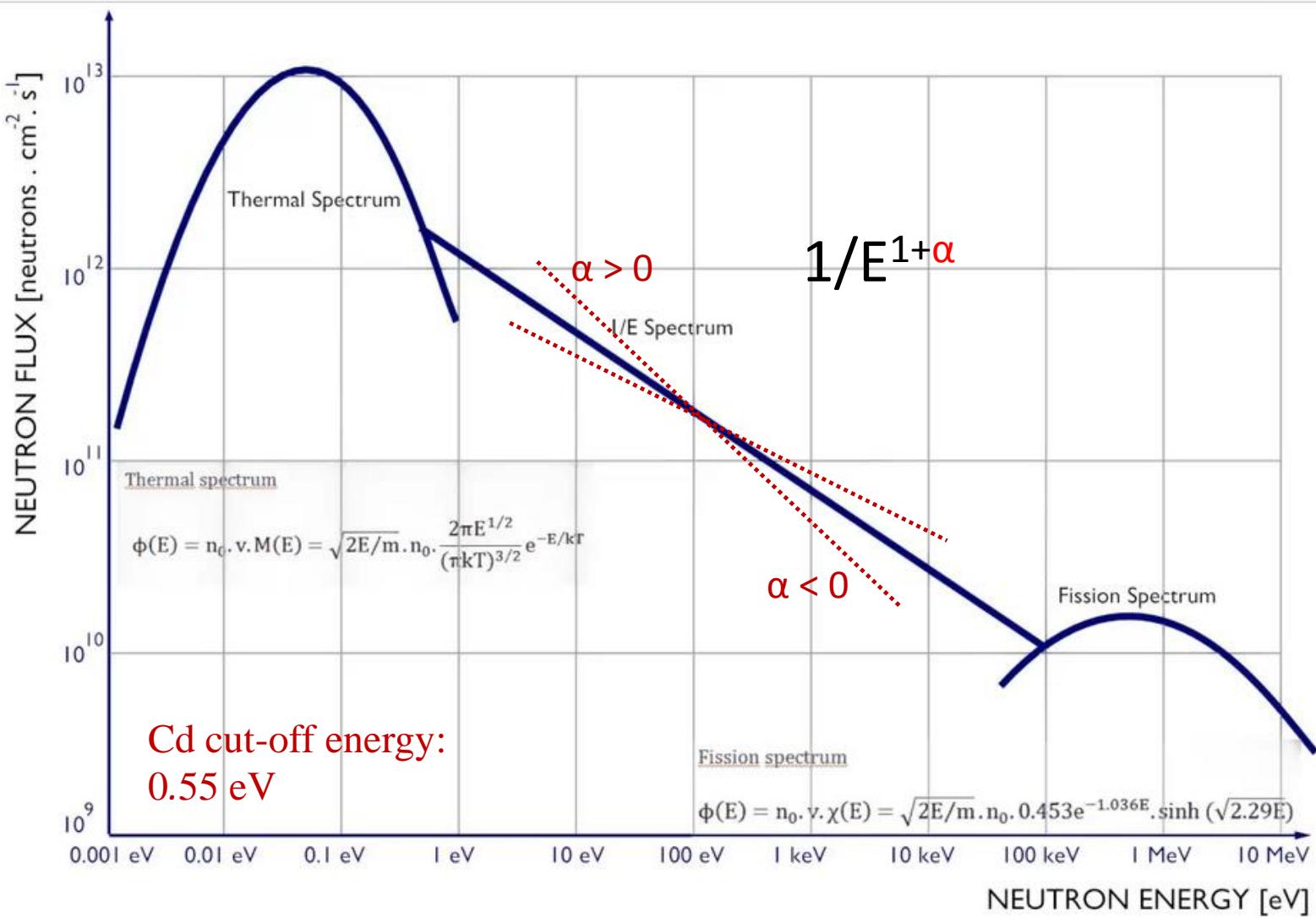
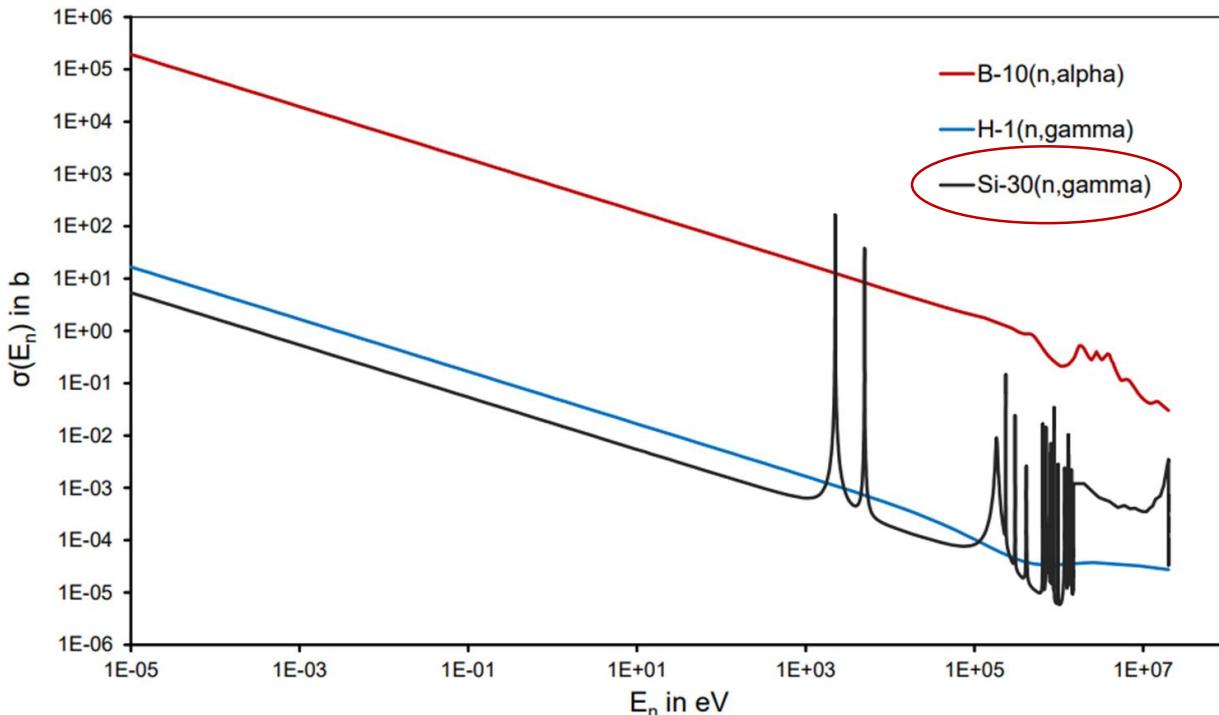


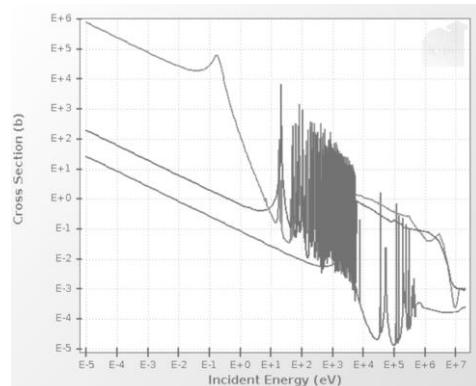
Figure: <https://nuclear-power.com>



1/v law

Capture probability / reaction rate is highest for thermal and esp. cold neutrons.

Moderation to lower temperature makes sense in most cases.



Cd filter for ENAA :

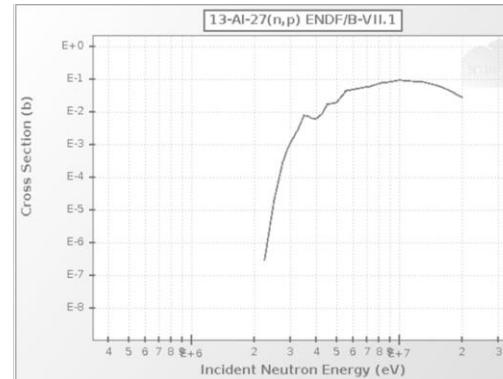
Cd-113

Na-23 ($I_0 / \sigma_0 \approx 0.4$)

Sb-123 ($I_0 / \sigma_0 \approx 30$)

$I_0: 0.55\text{eV}-1\text{ MeV}$

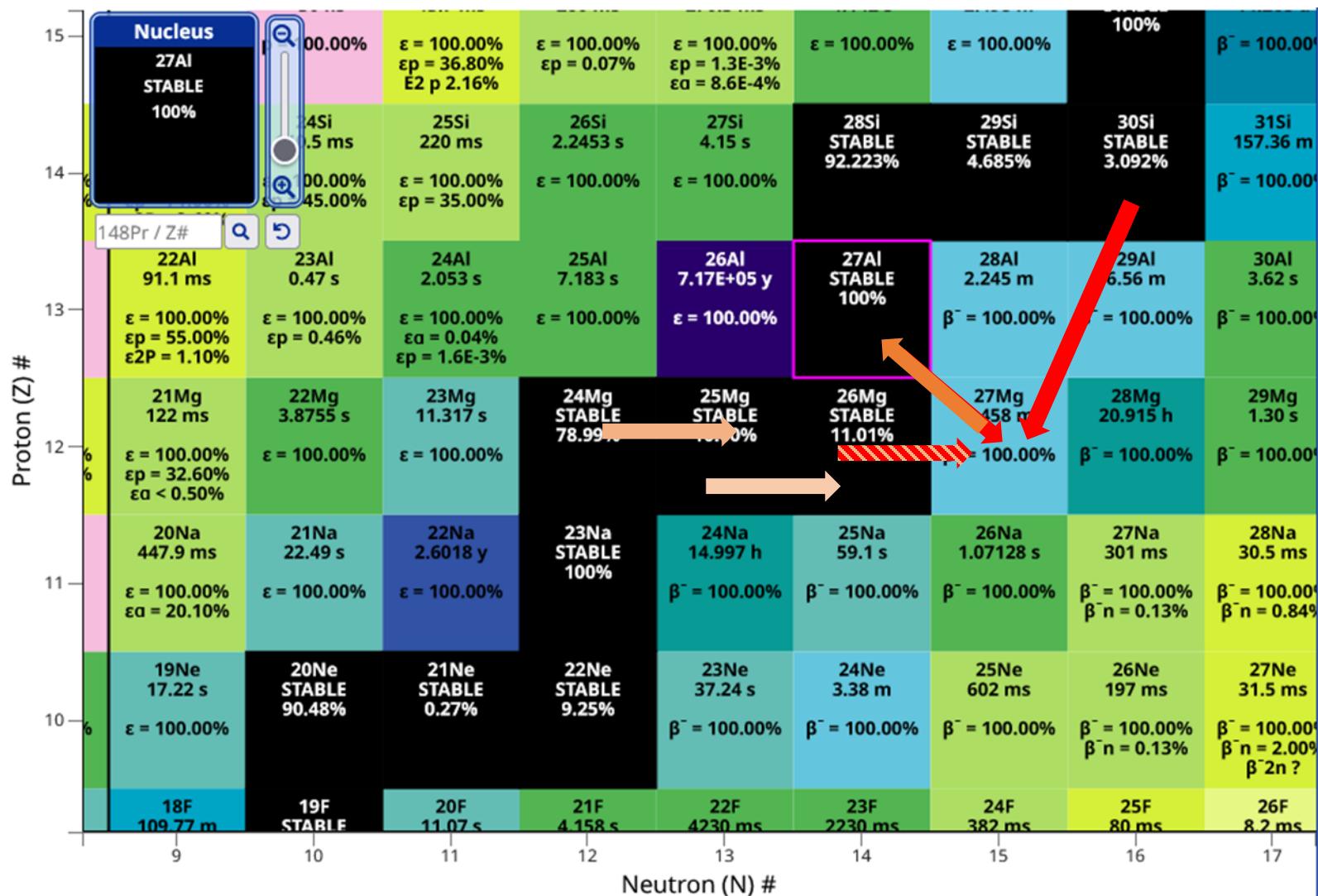
$\sigma_0: 0.025\text{eV}$



Threshold reactions do not occur in PGAA (with cold / thermal neutron beams). But they can disturb INAA and are used for FNAA.

NAA

PGAA: mostly (n, γ) NAA: mostly (n, γ) + β -decay + γ



<https://www.nndc.bnl.gov/nudat3/>

Chemical analysis with neutrons such as NAA enables a unique combination of advantages:

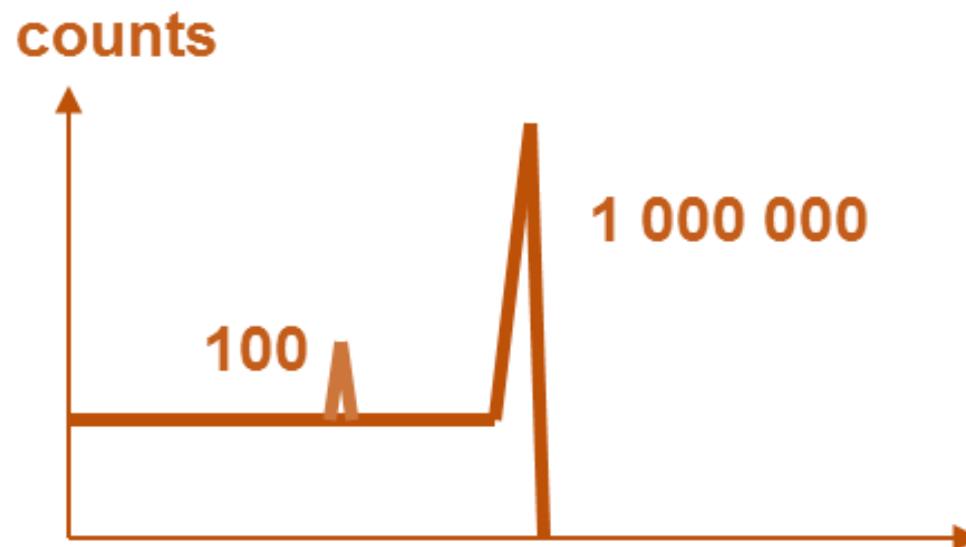
- ✓ Panoramic analysis with **very low DLs** for many elements.
 - ✓ No (or not much) sample preparation / non- or low-destructive.
 - ✓ High grade of matrix independency.
 - ✓ **High dynamic range.**
 - ✓ Real bulk method.
-
- ✓ NAA can be a so-called **primary method of measurement** (CCQM).
→ Its results can be deduced to fundamental constants using SI units (high traceability), also high reliability, high precision.

High dynamic range

PGAA

1 mg H together with 1 g Cl
(10 mg water in 1 g CCl_4)

1 mg Cl together with 1 g H
(1 mg Cl in 10 g water)

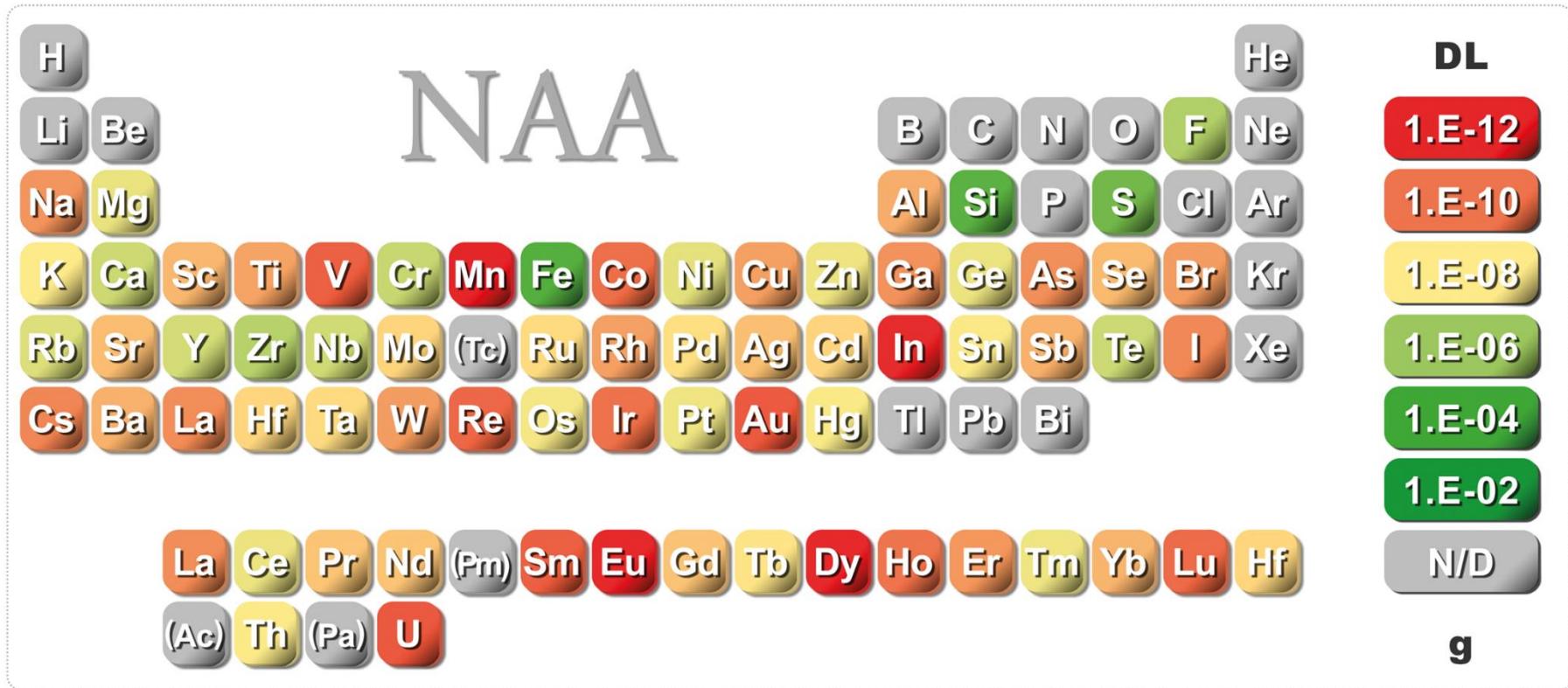


NAA

The HDR can be further increased by an irradiation-measurement plan adjusted to the half-lives of the activation products.

Parameters:
irradiation time, decay time, counting time, detector-sample distance.

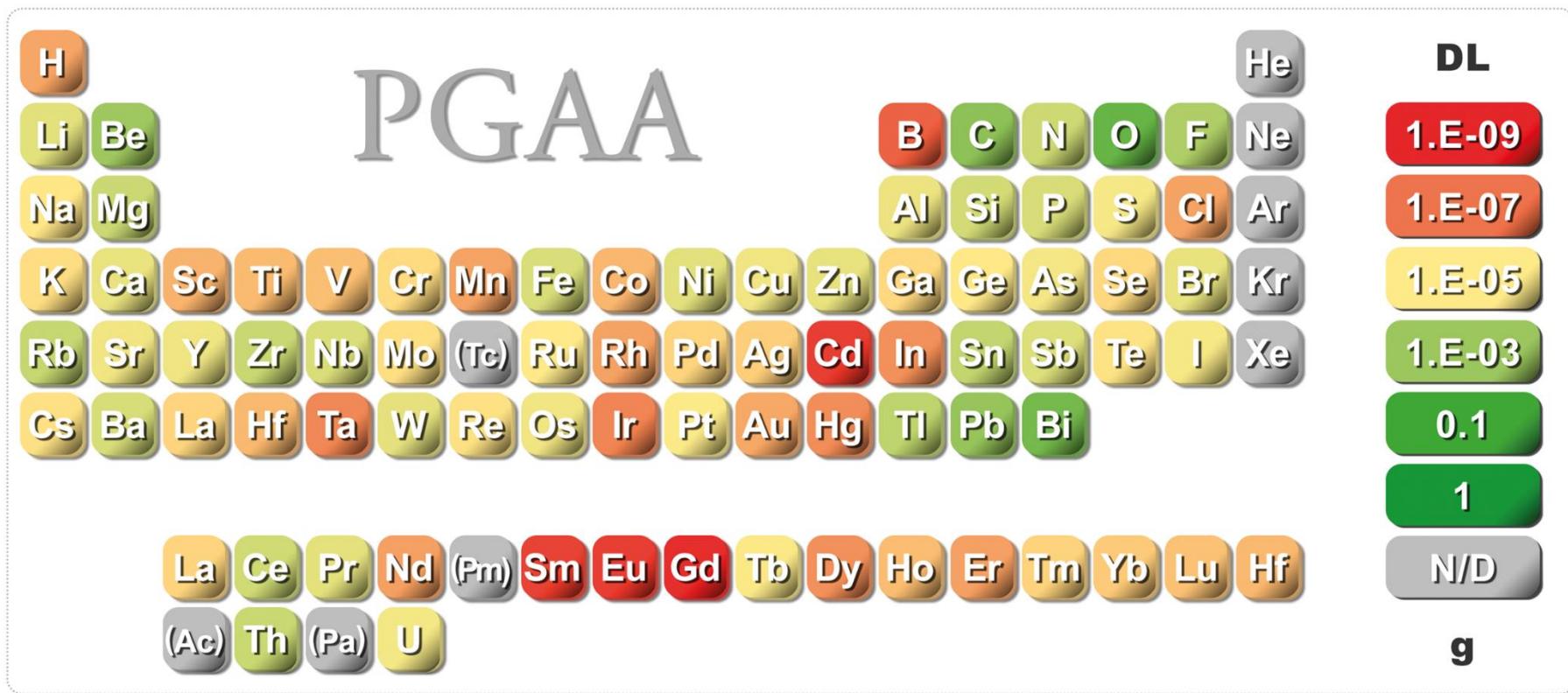
Sensitivity of INAA (typical geological material)



In-beam NAA: DL in g / INAA: DL in mg (ppt/ppq level possible!)

INAA = Instrumental Neutron Activation Analysis (no chem. treatment)

Sensitivity of PGAA (typical geological material)



Complementary Methods!

Lichen checking air pollution (PGAA + ibNAA)

Z	El	M	m meas	unc %	m Bkg	unc %	m net	ox. st.	m ox	unc %	mol%	unc %	w% el/el	unc %	
1	H	1.008	1.18E-2	0.4	2.44E-7	1.7	1.18E-2	1	0.11	0.4	48.6	2.2	6.17	4.	
5	B	10.81	1.14E-6	0.4	3.87E-9	0.9	1.13E-6	3	3.65E-6	0.4	4.35 ppm	2.2	5.92 ppm	4.	
6	C	12.01	0.107	3.1	3.50E-2	3.0	0.07	4	0.26	5.	25	4.	38	5.	
7	N	14.01	2.32E-3	2.1			0.0	2.32E-3	5	8.95E-3	2.1	0.69	3.0	1.21	5.
13	Al	26.98	7.32E-4	2.1	1.38E-4	2.0	5.93E-4	3	1.12E-3	2.6	0.091	3.4	0.31	5.	
14	Si	28.09	1.85E-3	3.1			0.0	1.85E-3	4	3.95E-3	3.1	0.27	3.7	0.97	5.
16	S	32.07	2.47E-4	2.6			0.0	2.47E-4	6	6.17E-4	2.6	320 ppm	3.3	0.129	5.
17	Cl	35.45	9.91E-5	1.7	9.27E-8	2.4	9.90E-5	-1	9.90E-5	1.7	116 ppm	2.7	0.052	4.	
19	K	39.1	9.92E-4	1.7			0.0	9.92E-4	1	1.20E-3	1.7	0.105	2.7	0.52	4.
22	Ti	47.87	3.50E-5	3.0			0.0	3.50E-5	4	5.84E-5	3.0	30 ppm	3.7	180 ppm	5.
25	Mn	54.94	5.05E-6	7.			0.0	5.05E-6	3	7.25E-6	7.	3.8 ppm	8.	26 ppm	9.
48	Cd	112.4	3.06E-8	4.0			0.0	3.06E-8	2	3.49E-8	4.0	0.0113ppm	5.	0.16 ppm	6.
62	Sm	150.4	3.73E-8	2.8			0.0	3.73E-8	3	4.33E-8	2.8	0.0103ppm	3.5	0.19 ppm	5.
64	Gd	157.3	4.45E-8	6.			0.0	4.45E-8	3	5.13E-8	6.	0.012ppm	6.	0.23 ppm	7.
		0													
8	O	16	0.096	7.			0.0	0.10	-2		7.	25	6.	50	4.
		0													
11	Na	22.99	5.22E-5	0.7			0.0	5.22E-5	1	7.03E-5	0.7	94 ppm	2.3	273 ppm	4.
17	Cl	35.45	1.02E-4	3.0			2.4	1.02E-4	-1	1.02E-4	3.0	120 ppm	3.7	0.053	5.
20	Ca	40.08	5.54E-3	13.			0.0	5.54E-3	2	7.75E-3	13.	0.6	13.	2.9	13.
21	Sc	44.96	3.36E-8	4.			0.0	3.36E-8	3	5.15E-8	4.	0.031ppm	5.	0.18 ppm	6.
23	V	50.94	6.64E-7	23.			0.0	6.64E-7	5	1.19E-6	23.	0.5 ppm	23.	3 ppm	23.
25	Mn	54.94	5.06E-6	2.2			0.0	5.06E-6	3	7.28E-6	2.2	3.8 ppm	3.1	26 ppm	5.
26	Fe	55.85	1.00E-4	15.			8.	1.00E-4	3	1.43E-4	15.	70 ppm	15.	0.05	15.
27	Co	58.93	1.25E-7	12.			0.0	1.25E-7	2	1.59E-7	12.	0.09 ppm	13.	0.7 ppm	13.
35	Br	79.9	2.00E-6	8.			0.0	2.00E-6	-1	2.00E-6	8.	1.0 ppm	9.	10 ppm	9.
38	Sr	87.62	1.09E-5	8.			0.0	1.09E-5	2	1.29E-5	8.	5.2 ppm	8.	57 ppm	9.
63	Eu	152	7.21E-9	8.			0.0	7.21E-9	3	8.35E-9	8.	0.0020ppm	9.	0.038ppm	9.
		0													



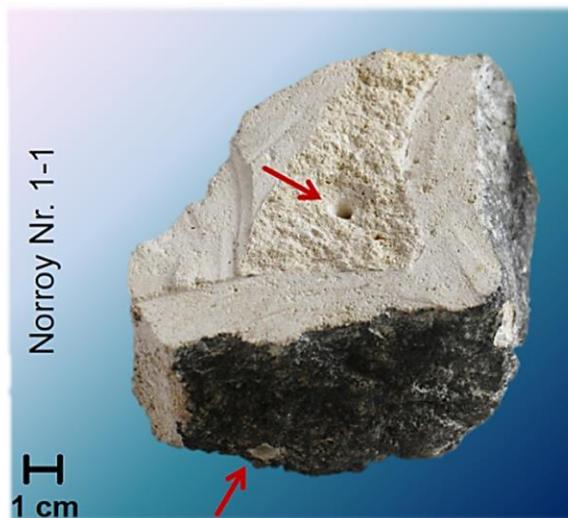
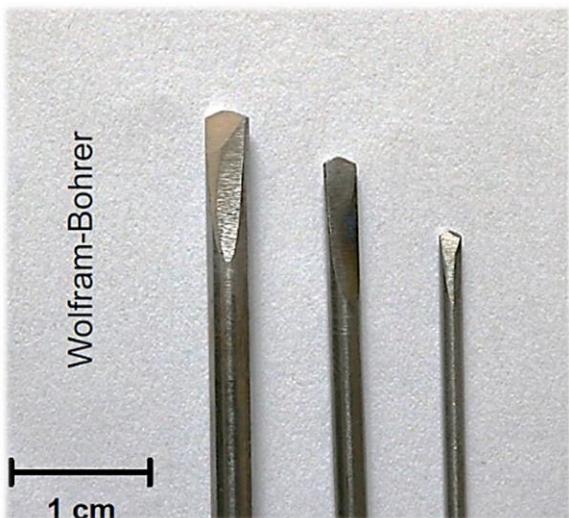
PGAA

stoichiometry



NAA





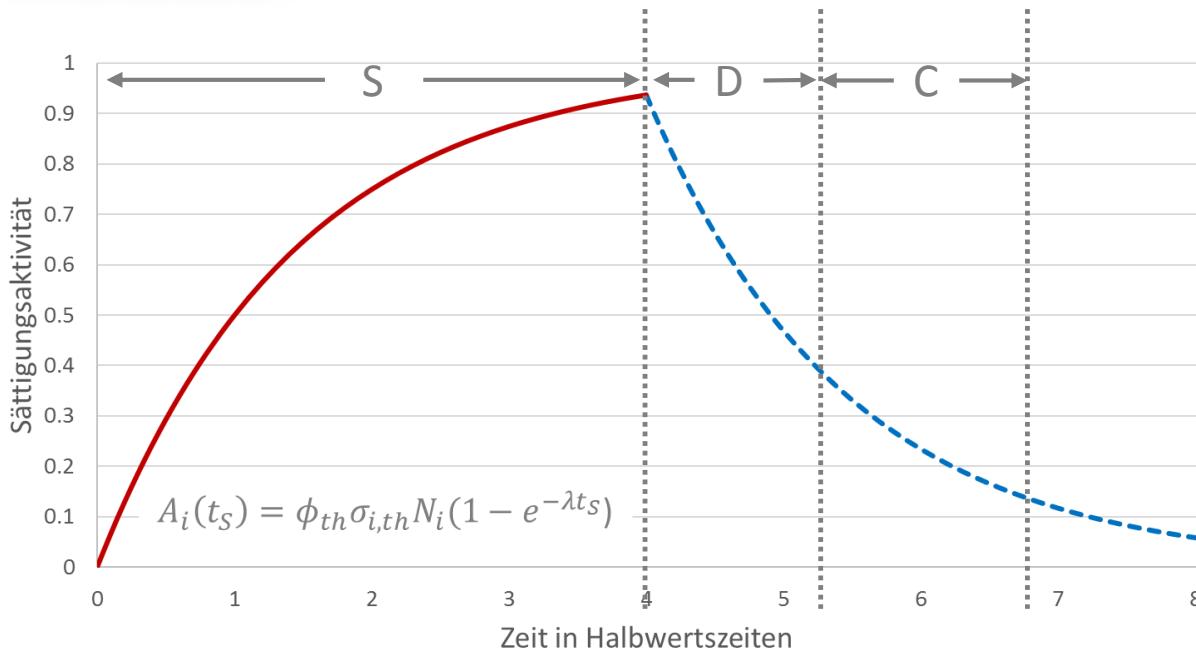
Sample preparation:

- Very sensitive to contaminations!
- Precise weighing necessary (typical sample mass in mg or μg range).

irradiation (S):



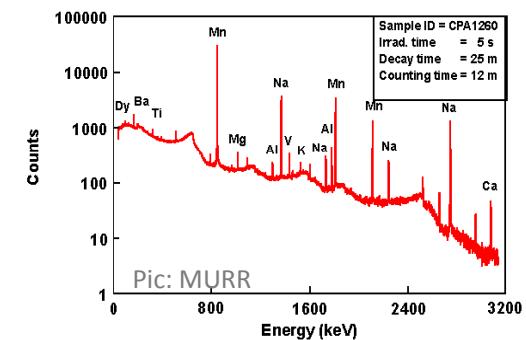
transport + decay /
cooling time (D)



$$S = 1 - e^{-\lambda t_{act}}$$

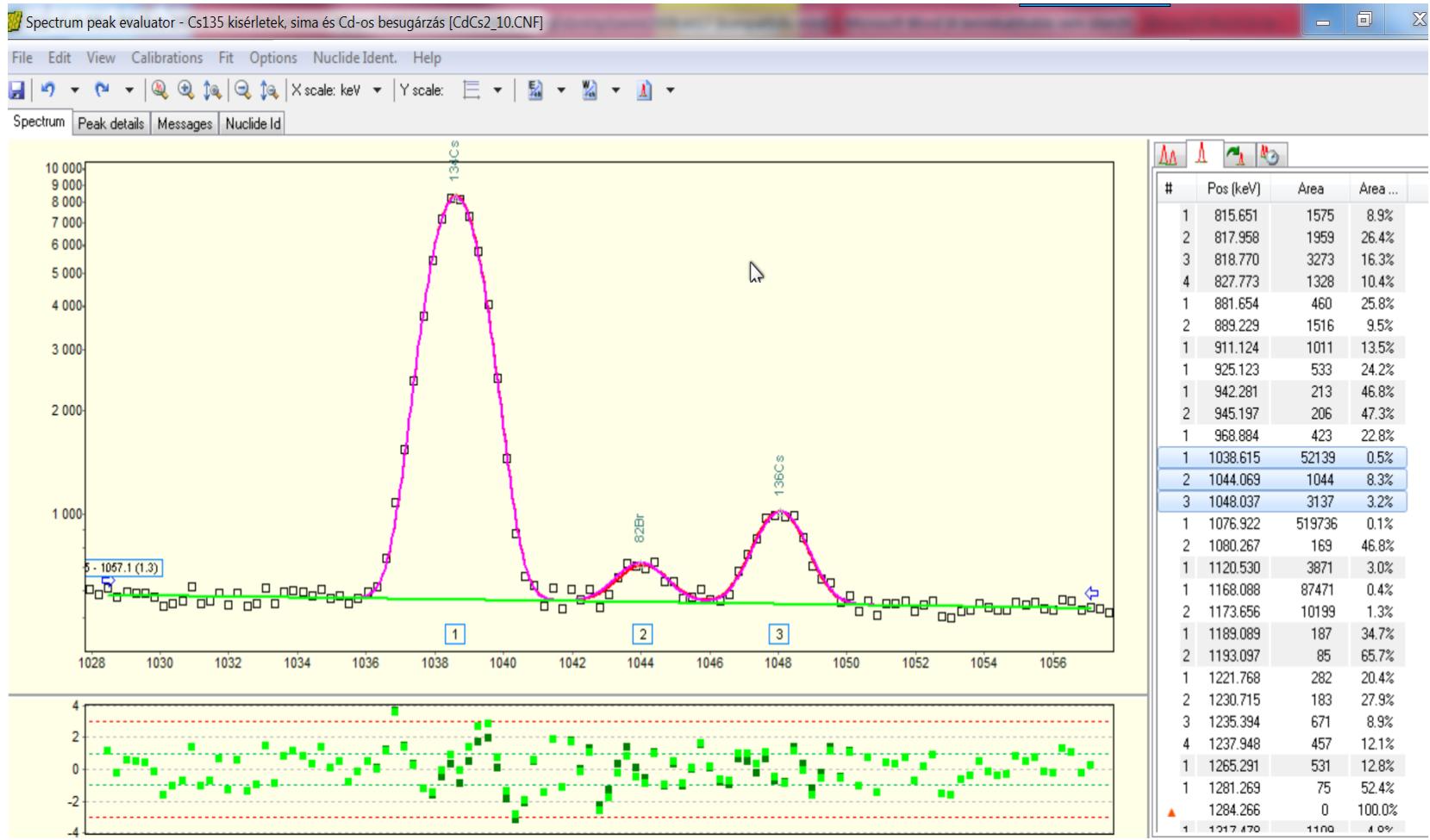
$$\text{peak area } A' = A/\text{SDC}$$

measurement (C):



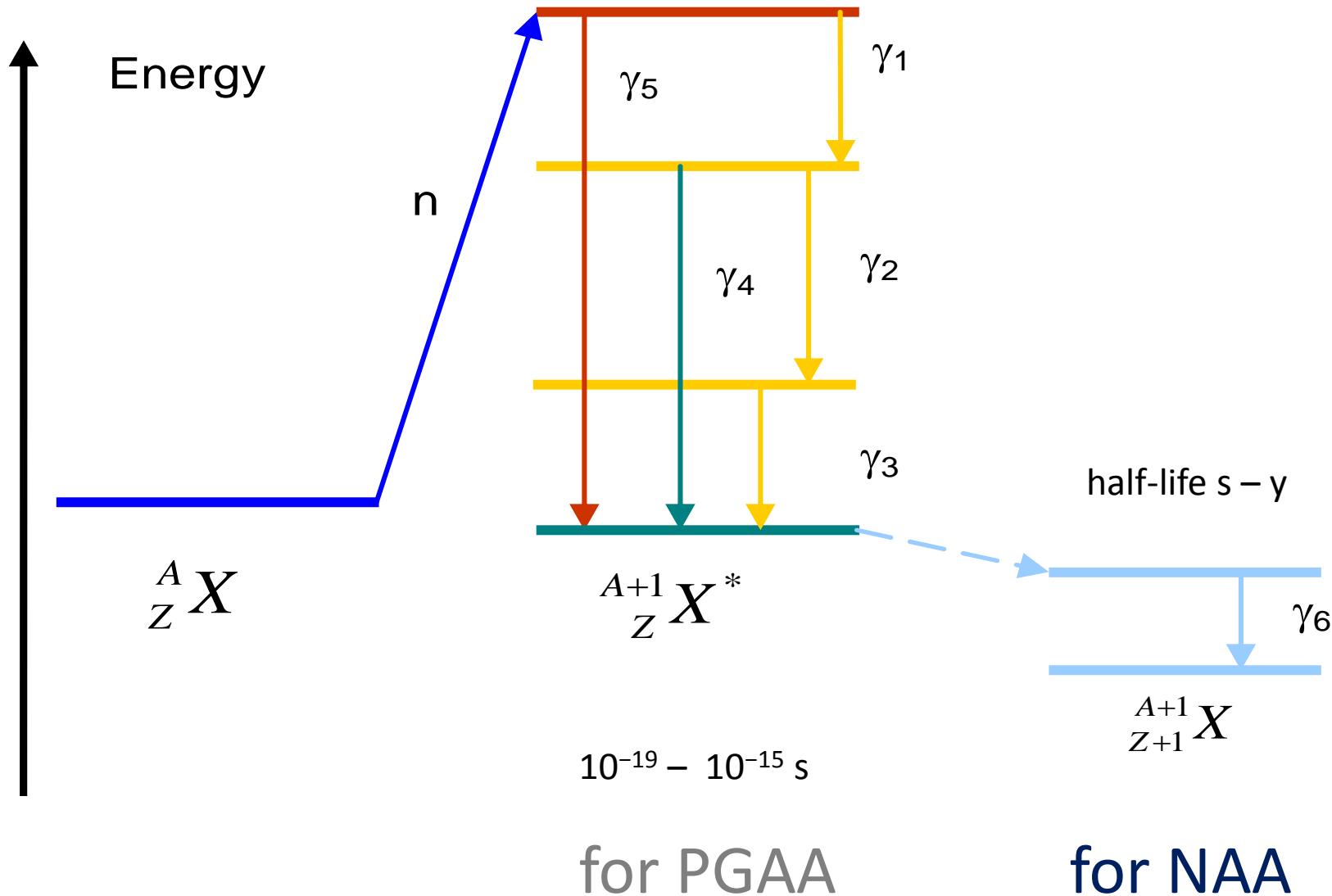
short + long irradiation

A few activation products are present in more than one measurement → „intrinsic control“!

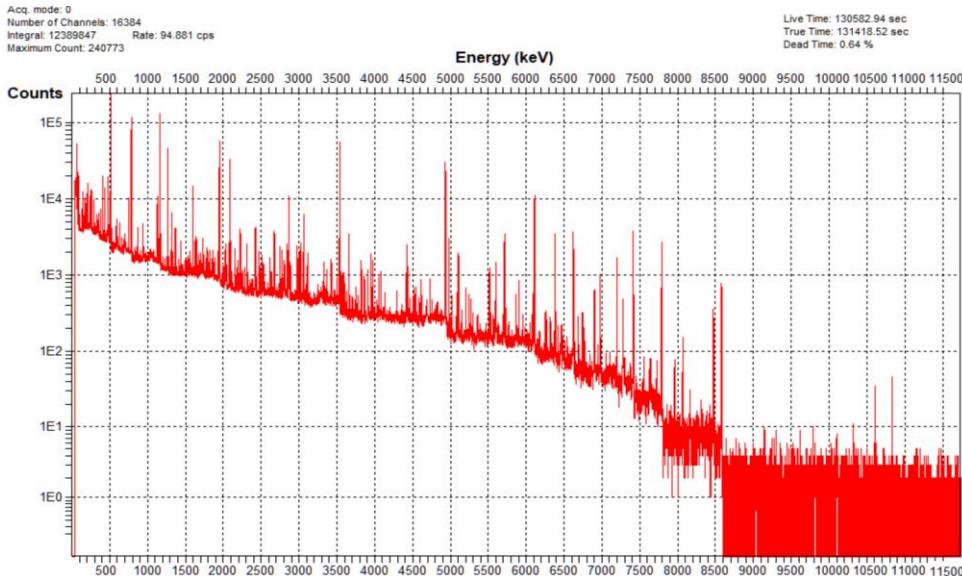


Evaluation step 1: Peak fitting with Hyperlab!

NAA

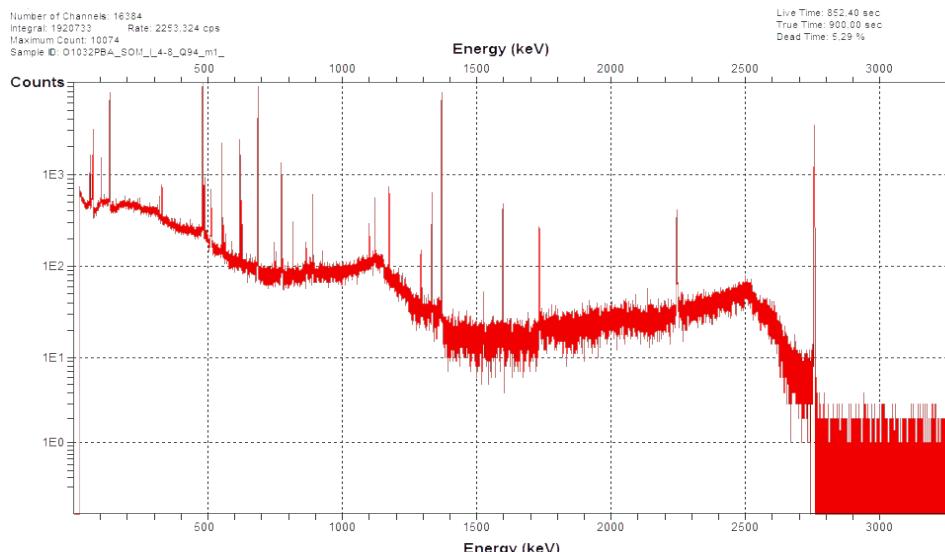


Pic: L. Szentmiklosi



PGAA

- Gamma energy range up to 12 MeV
- Complicated spectrum with up to 10^3 peaks
- Peak shape Gauss-like
- Baseline decreasing towards high energies
- Poisson statistics
- Peak positions -> identifying the elements
- Peak areas-> determining quantities



NAA

- Gamma energy range up to 2 (4) MeV
- Spectrum with up to 10^2 peaks
- Peak shape Gauss-like
- Baseline decreasing towards high energies
- Non-Poisson statistics (if changing count rate)
- Peak positions -> identifying the elements
- Peak areas, half lives -> determining quantities

$$\frac{N_p/t_m}{(N_p/t_m)^*} = \frac{w}{w^*} \cdot \frac{S \cdot D \cdot C}{S^* \cdot D^* \cdot C^*} \cdot \left(\frac{M^*}{M} \cdot \frac{\theta}{\theta^*} \cdot \frac{\gamma}{\gamma^*} \cdot \frac{\sigma_0}{\sigma_0^*} \right) \cdot \frac{f+Q_0}{f+Q_0^*} \cdot \frac{\varepsilon_p}{\varepsilon_p^*}$$

k_0 : universal nuclear constant
*: comperator

$$c_x(ppm) = \frac{\frac{N_{p,x}}{t_m \cdot S \cdot D \cdot C \cdot W}}{A_{sp,Au}} \cdot \frac{1}{k_{0,Au}(x)} \cdot \frac{f+Q_{0,Au}(\alpha)}{f+Q_{0,x}(\alpha)} \cdot \frac{\varepsilon_{p,Au}}{\varepsilon_{p,x}} \cdot 10^6$$

OR:

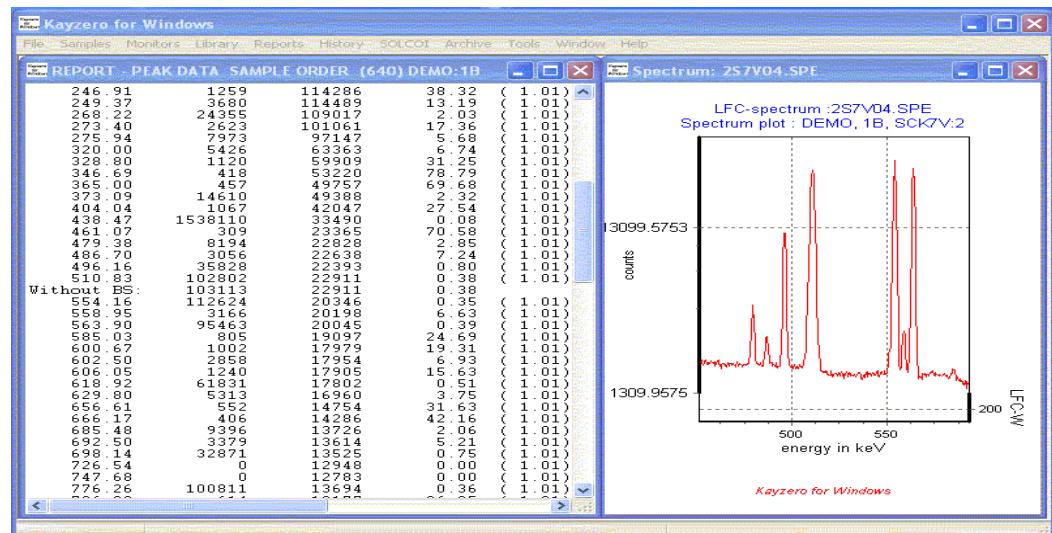
$$\frac{c_x}{c_{std}} = \frac{a_x}{a_{std}}$$

$$f = \frac{\Phi_{th}}{\Phi_e},$$

$$Q_0 = \frac{I_0}{\sigma_{th}},$$

$$A_{sp,x} = N_{p,x}/t_m \cdot S \cdot D \cdot C \cdot W$$

α : epitermal shape factor

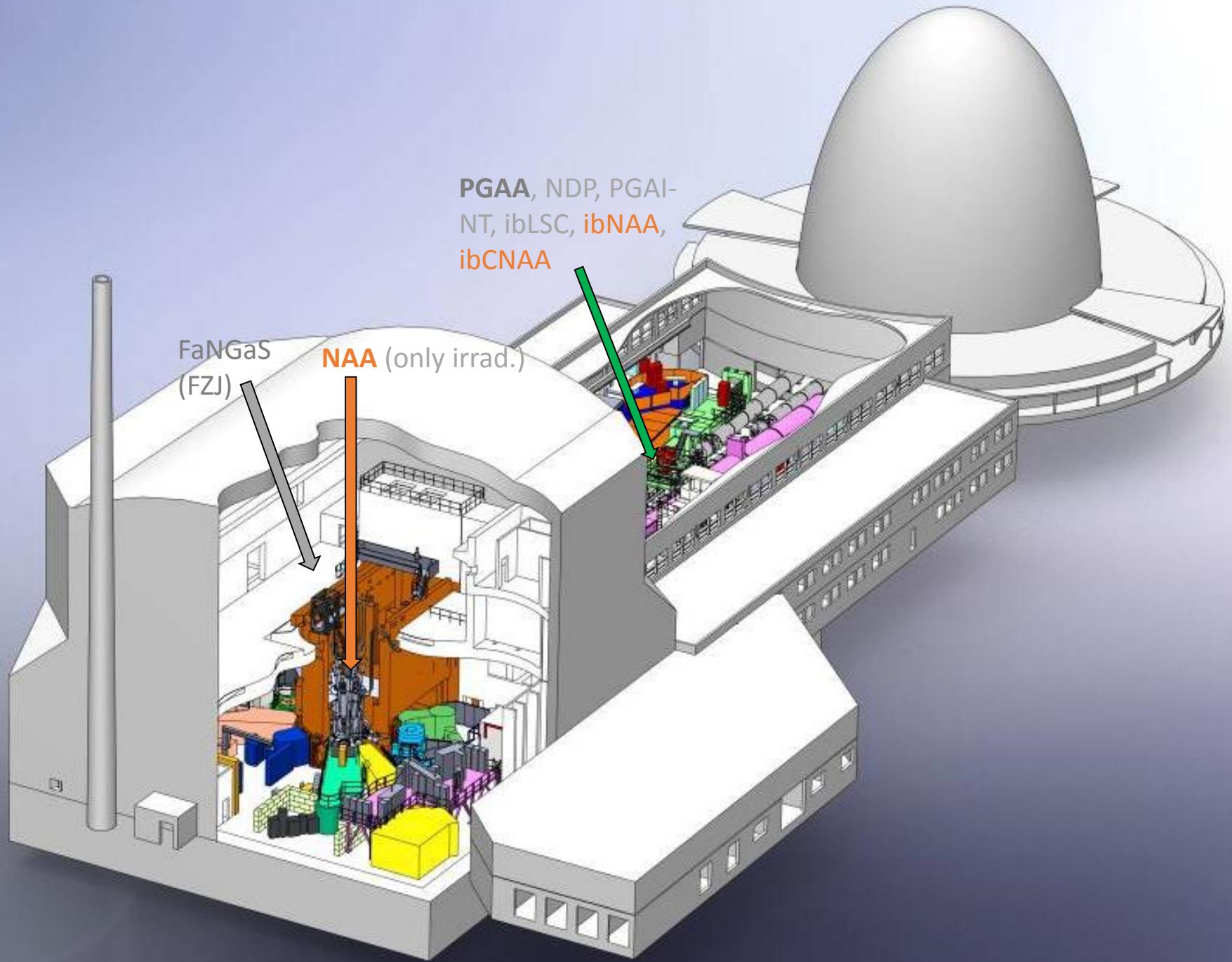


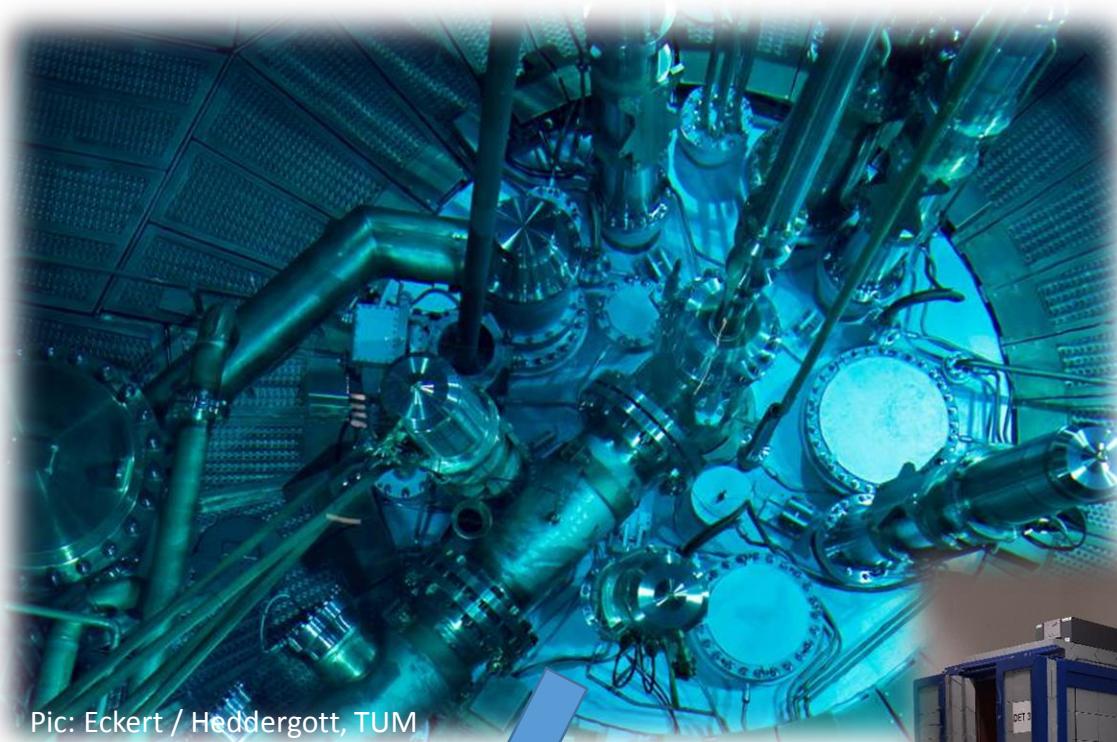
Evaluation step 2: Calculations with Kayzero!

Corrections for calculating the concentration:

- background (only special cases, e.g. Co-60)
- blank (if not re-packaged)
- gamma absorption (large samples)
- neutron self-absorption (large samples)
- burn-up (only long-time with high flux)
- dead-time
- true-coincidence correction (if sample is close to the detector)
- interferences
- threshold reactions (at FRM II mostly not relevant)
- fission

Evaluation step 2: Calculations with Kayzero!





Pic: Eckert / Heddergott, TUM

FRM II:
Flux for NAA
up to $\sim 10^{14} \text{ cm}^{-2}\text{s}^{-1}$



Repackaging



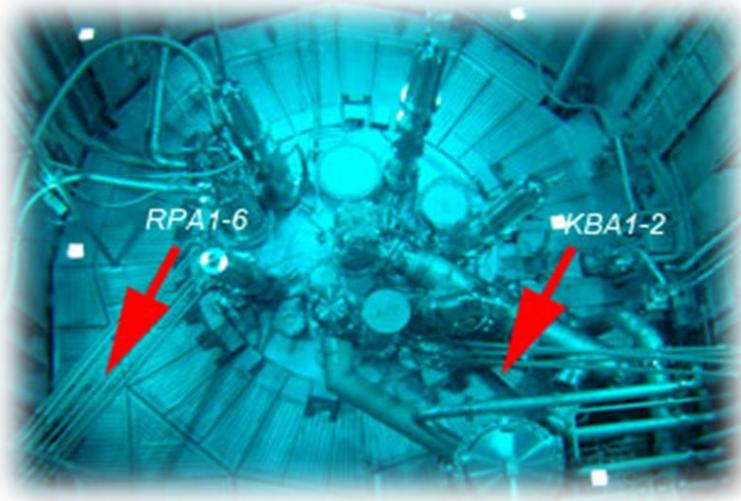
Counting stations
in the RCM

Radiochemistry Munich (RCM)

NAA is now an instrument and is fully integrated into the user system.

➤ **Irradiation**

- using rabbit system (transfer to RCM few mins in total):
 $5 \times 10^{12} - 7 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$
- capsule irradiation
- $f = \text{very high}$



➤ **Acquisition of gamma-ray spectra**

- 3 HPGe detectors with digital spectrometers
(Lynx, Canberra)

➤ **Evaluation:** Hyperlab + Kayzero (using k_0 method)

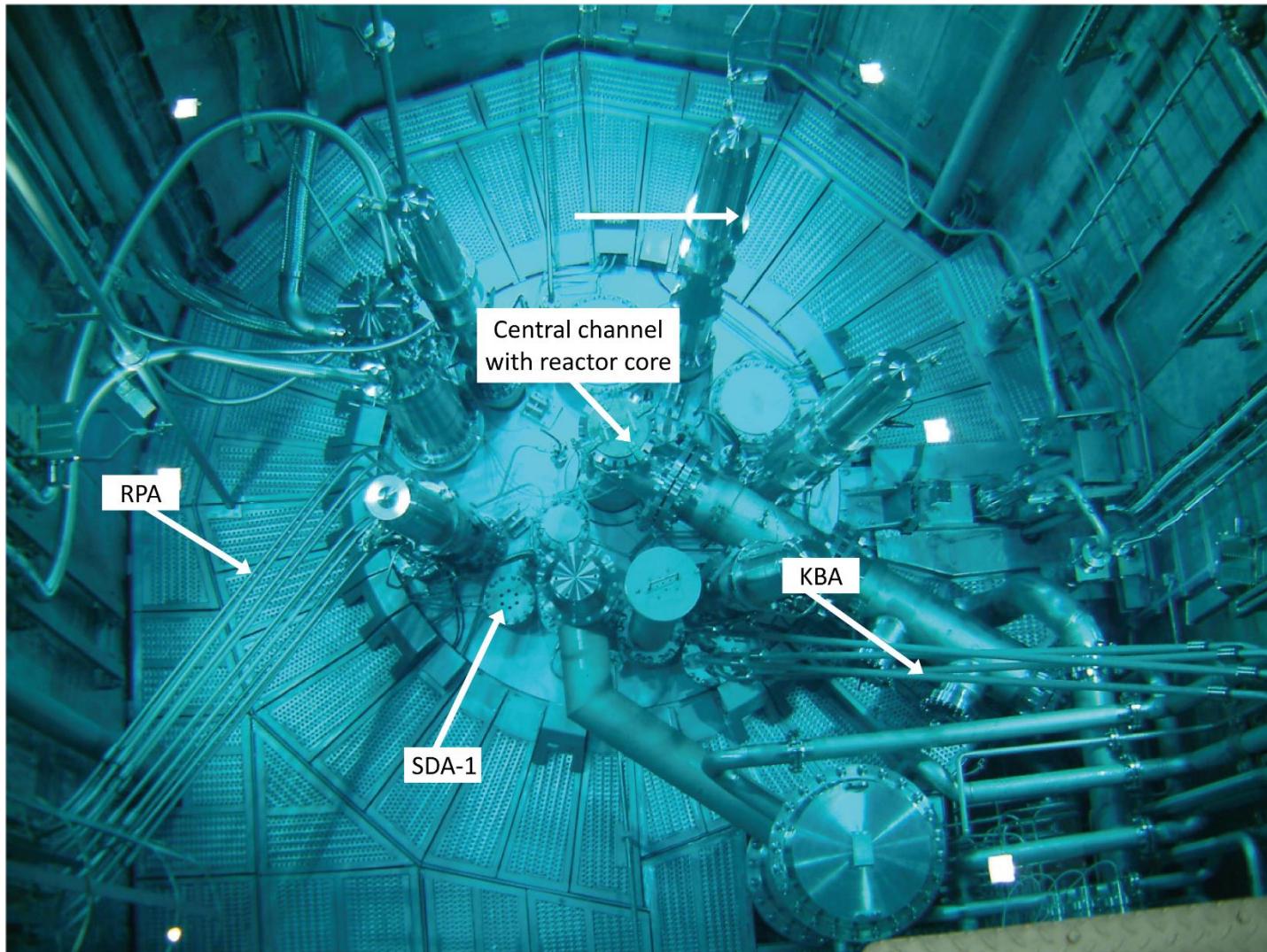
➤ **New standard protocol for a complete analysis:**

- I. Short irradiation (few mins)
2 countings within 30 mins
- II. Long irradiation (~1h)
2 countings 3 and 10 days later

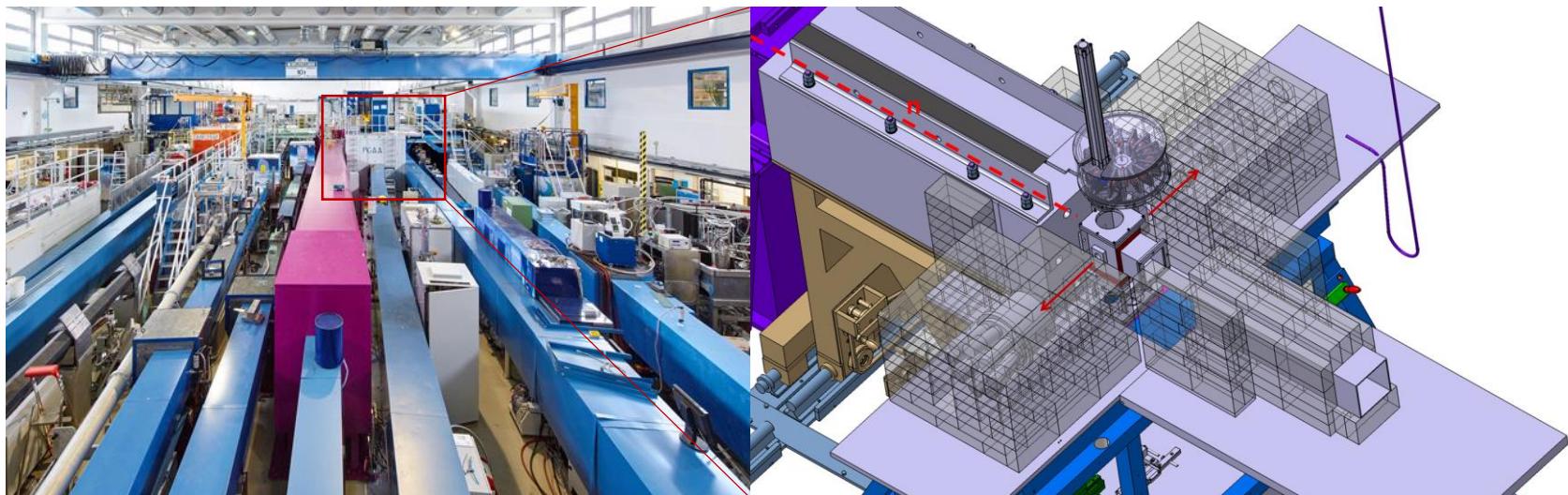
➤ **Throughput:** 2-4 samples/day (only weekdays)



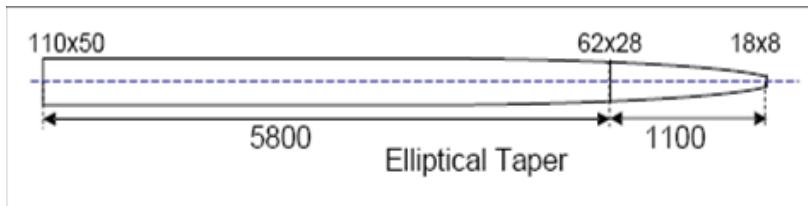
Close cooperation with reactor operation (V. Hutanu).



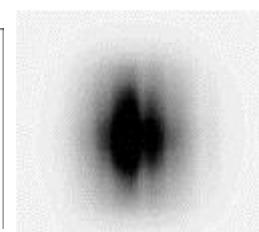
Position	ϕ_{th} (1/cm ² s)	ϕ_{epi} (1/cm ² s)	ϕ_f (1/cm ² s)	f (th/epi)
RPA1	3,6E+13	6,7E+09	2,0E+09	5300
RPA2	1,5E+13	3,2E+09	4,1E+08	4800
RPA3	4,8E+12	7,6E+08	7,2E+07	6400
RPA4	7,3E+13	2,4E+10	5,6E+11	3000
RPA5	3,9E+13	1,2E+10	5,0E+09	3400
RPA6	7,1E+12	1,2E+09	1,5E+08	5700
KBA1-1	1,3E+14	2,6E+11	3,9E+11	500
KBA1-2	9,3E+13	9,9E+10	2,0E+11	940
KBA2-1	1,1E+14	7,5E+10	2,1E+11	1500
KBA2-2	7,7E+13	3,9E+10	1,0E+11	2000
SDA1	1,2E+13	1,0E+09	1,5E+10	12000



5.8 m elliptical guide
1.1 m elliptical extension (removeable)



At focal point:
 $6 \times 10^{10} \text{ cm}^{-2} \text{ s}^{-1}$

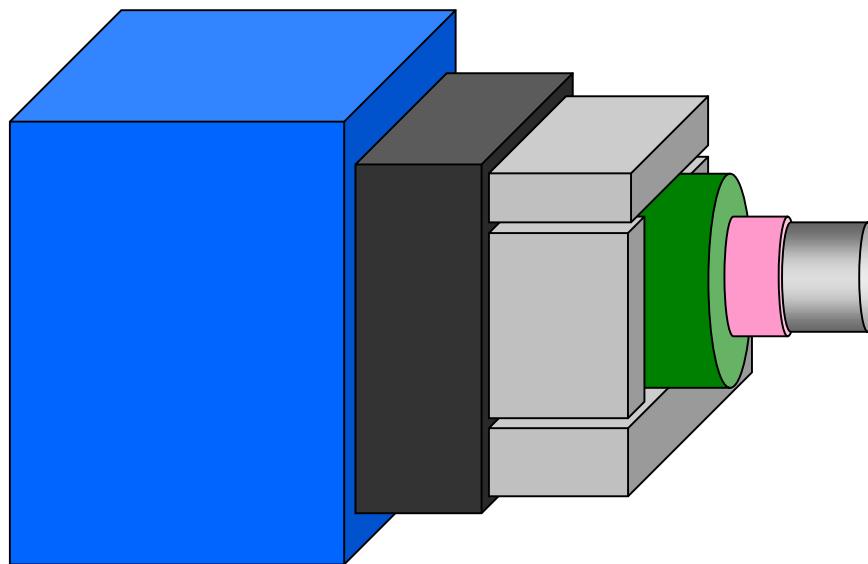


Recent situation
(no cold source
available): Factor
of 20 less

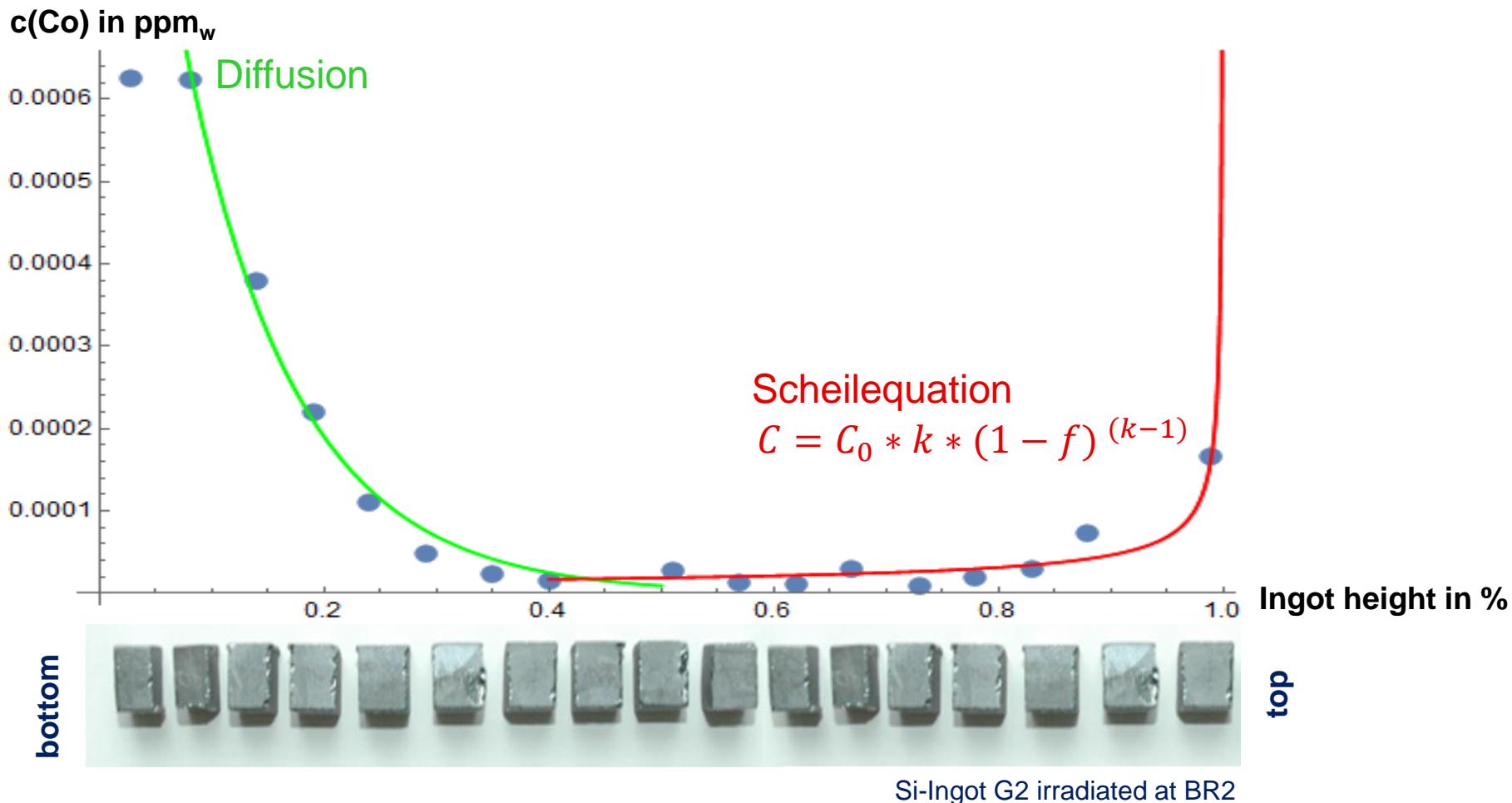
Area:
 $1.5 \times 1 \text{ cm}^2$

Other facilities
 $\sim 10^8 \text{ cm}^{-2} \text{ s}^{-1}$

- HPGe detector
- ^6Li -containing plastic (2.5 mm)
- Scintillator annulus (Compton and cosmic muon suppression) was NaI, now BGO
- 10 cm of lead
- 5mm boron rubber (40% B_4C)
- 5cm boron plastic (20% H_3BO_3)



Concentration profile of 3d-transition metals in Si-Ingot:



Karches, B., Welter, K., Stieghorst, C. et al. Instrumental determination of phosphorus in silicon for photovoltaics by β spectroscopy: a new approach. *J Radioanal Nucl Chem* **311**, 541–548 (2017). <https://doi.org/10.1007/s10967-016-5051-7>

Thank you for your attention!

Many thanks to the colleagues at BNC, Budapest and TRIGA Mainz!

