Determination of capture cross sections in high-flux cold neutron beams

(n,γ) measurements in high-flux cold neutron beams

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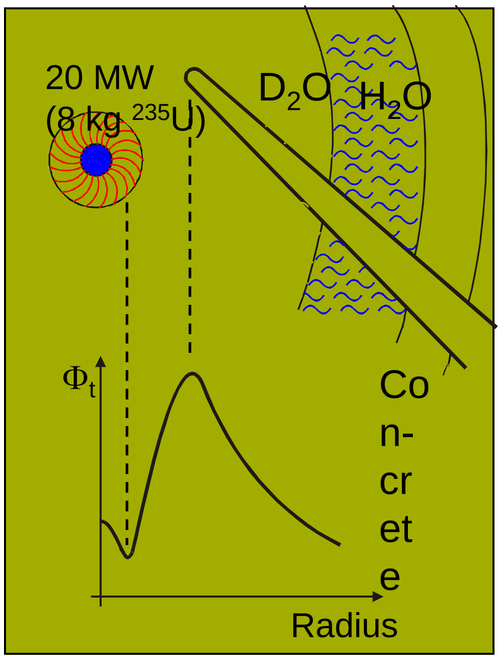
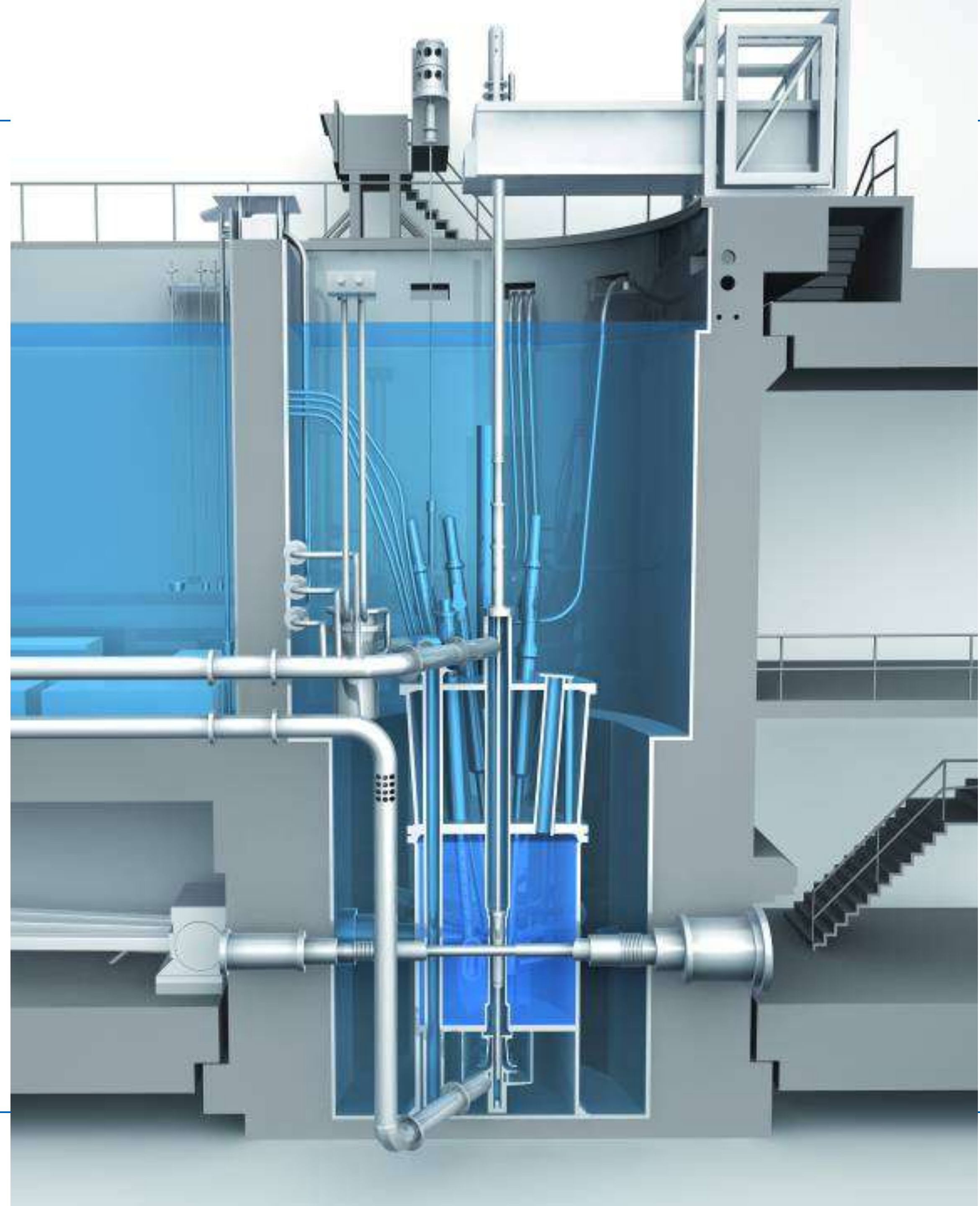
Earlier: Budapest Neutron Center

FRM II Reactor





The cold neutron source

Liquid deuterium moderator

Volume moderator vessel	25 liters
Volume of liquid D_2	~ 13 liters
Temperature	25 K

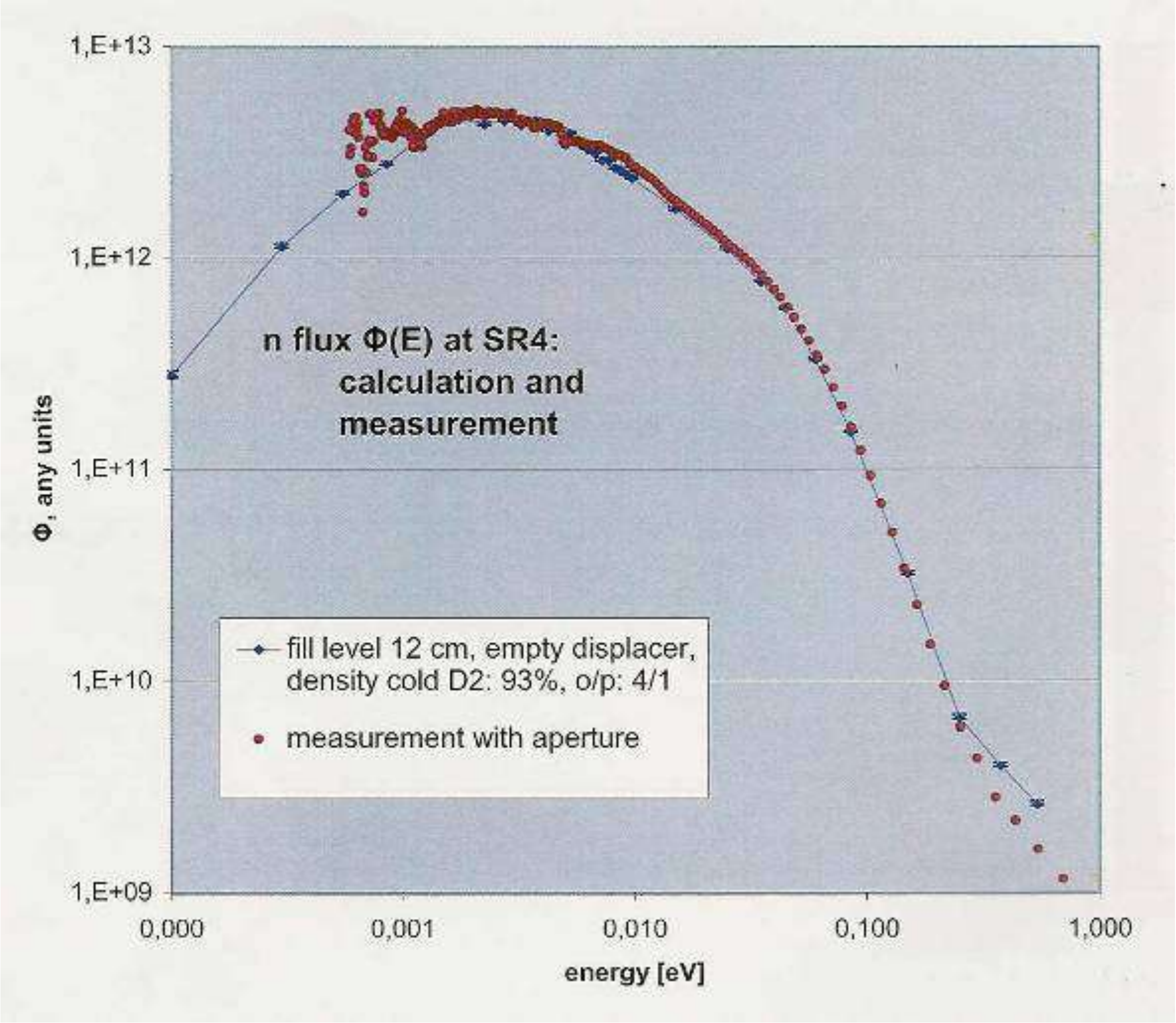
3 beam tubes
for cold neutron experiments

1 vertical beam tube is not in use

Cooling power total	~ 6 kW
in liquid D_2	2.7 kW

E. Gutsmiedl, Päthe, Chr. Müller, A. Scheuer

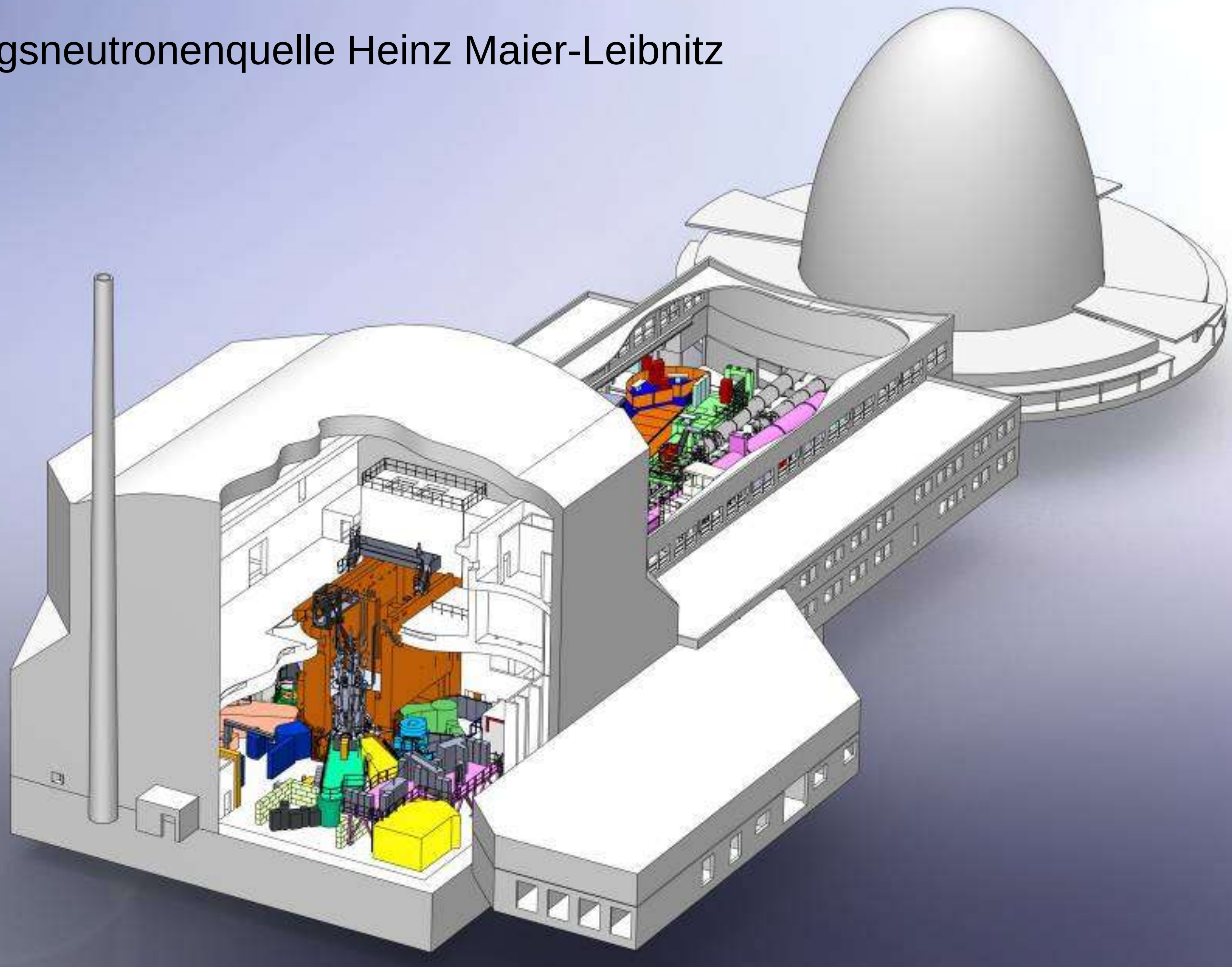
Cold neutron spectral Flux at 20 MW

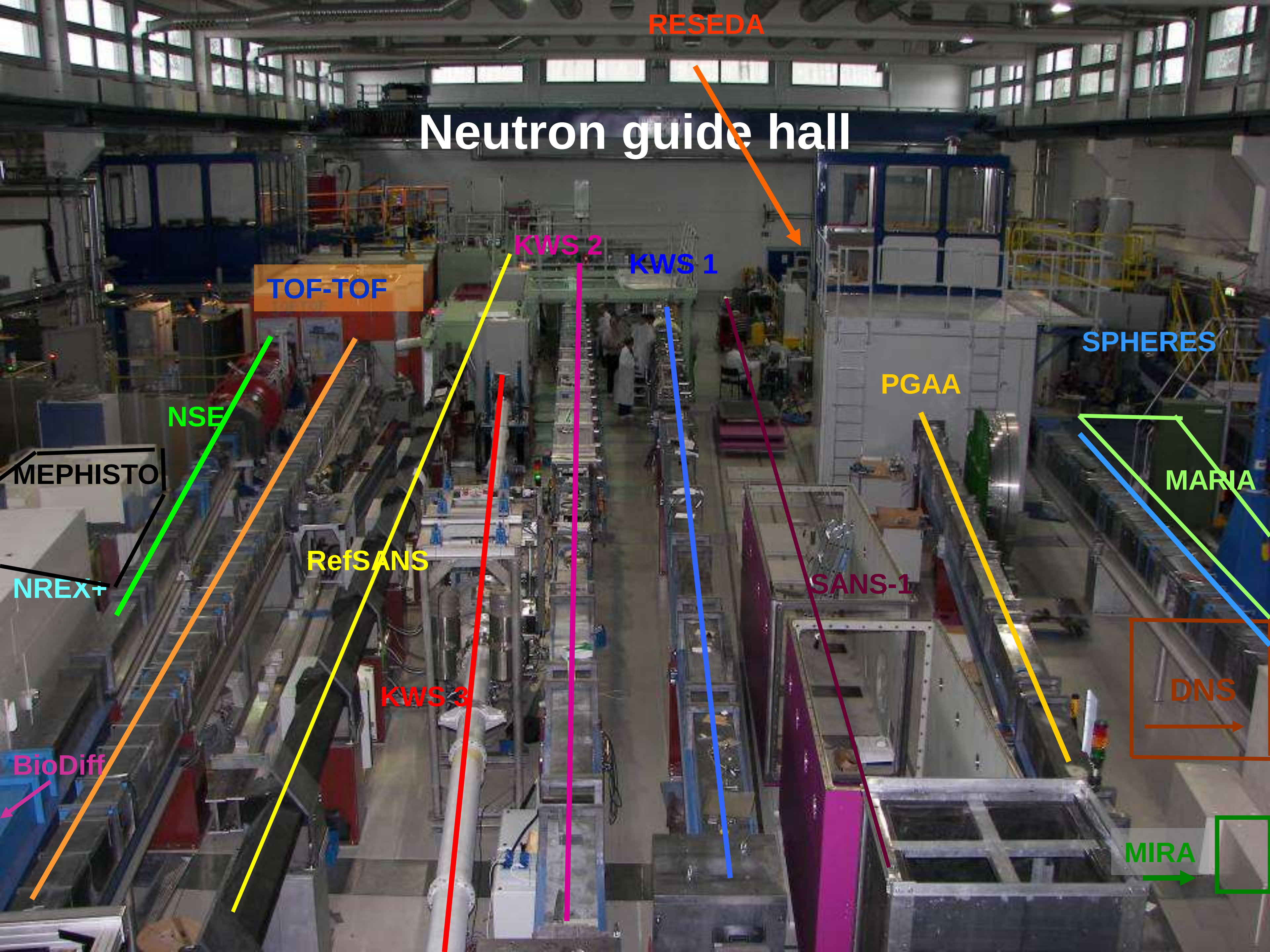


For guided cold neutrons:
$\Phi(\lambda)\lambda^2$
Cut-off below 2Å

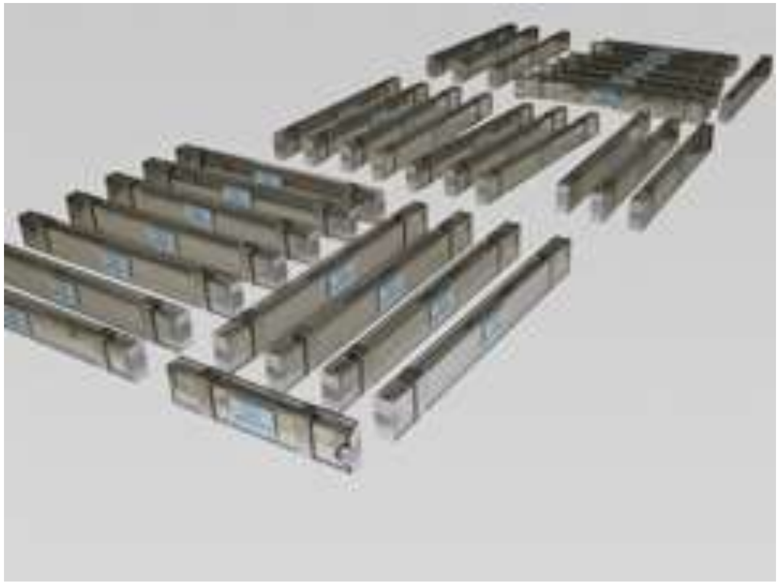
A. Röhrmoser, K. Zeitelhack

Forschungsneutronenquelle Heinz Maier-Leibnitz

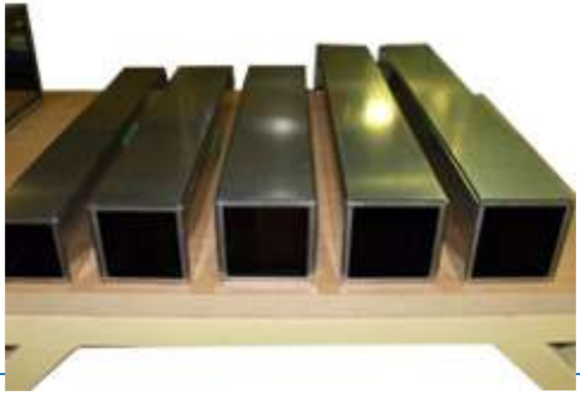



RESEDA
Neutron guide hall
KWS 2
KWS 1
TOF-TOF
SPHERES
PGAA
NSE
MEPHISTO
MARIA
RefSANS
NREX+
SANS-1
DNS
KWS 3
BioDiff
MIRA

3) Neutron guides



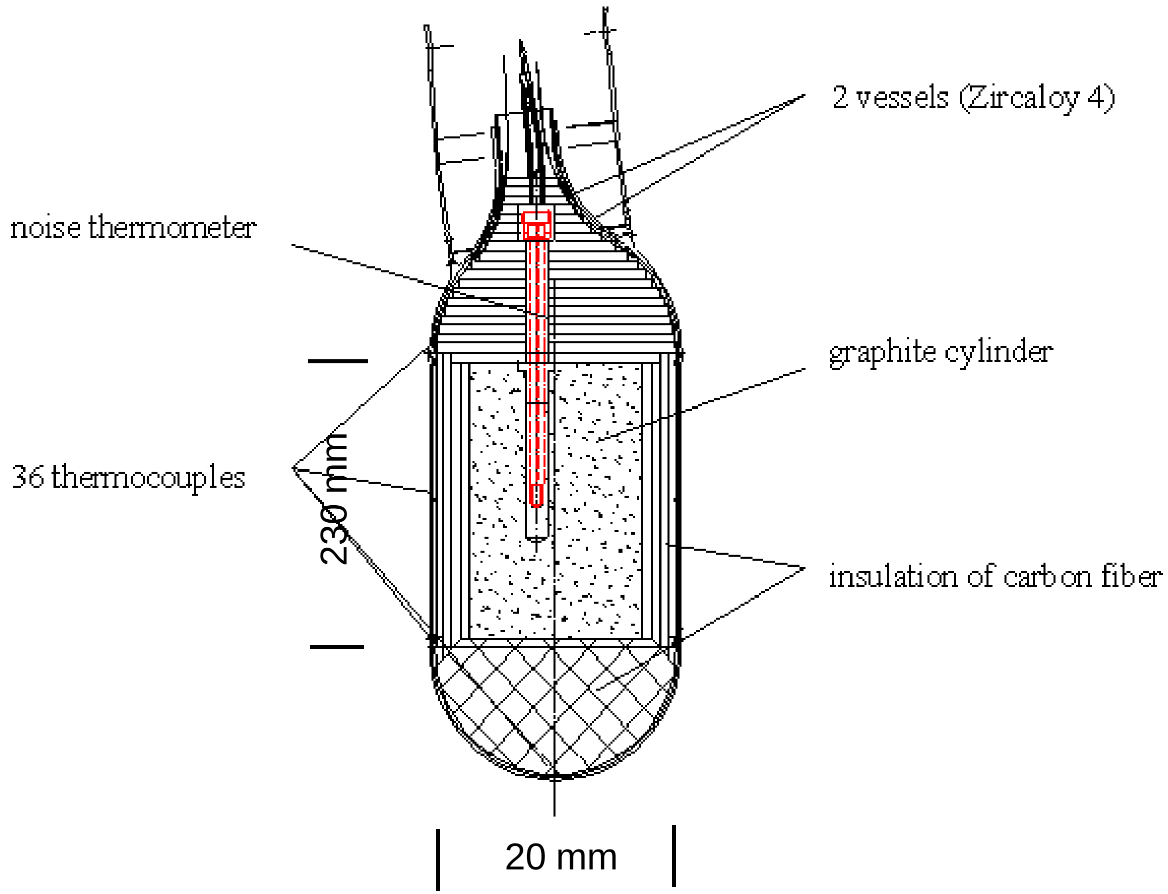
- Ni or supermirror guides
- relatively small losses
- **low background**



Facilities

- Guided cold beam: ~5meV
- Thermal beam: 25meV
- Hot beam: ~250meV
- Fast beam: fission spectrum

Graphite moderator of hot source (250meV)



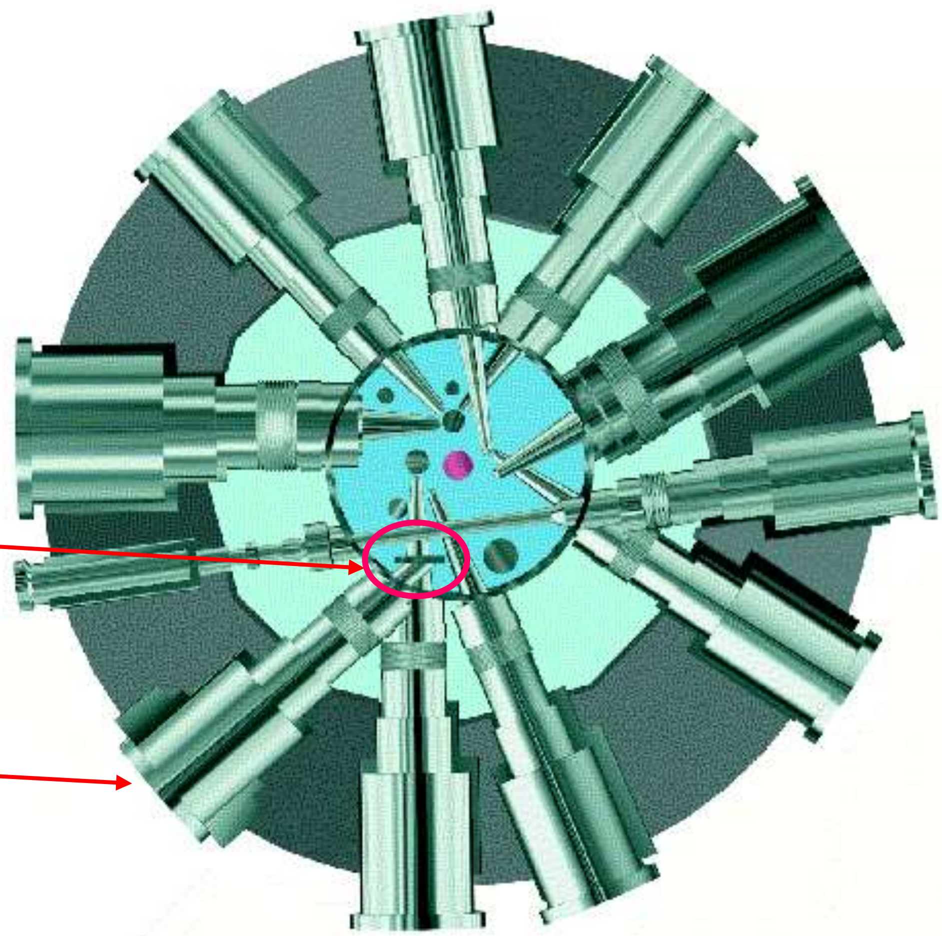
E. Gutsmiedl, Chr. Müller, A. Scheuer

Fast neutron channel
For (n,n')?

Reactor pool

Uranium converter

Beam tube SR10

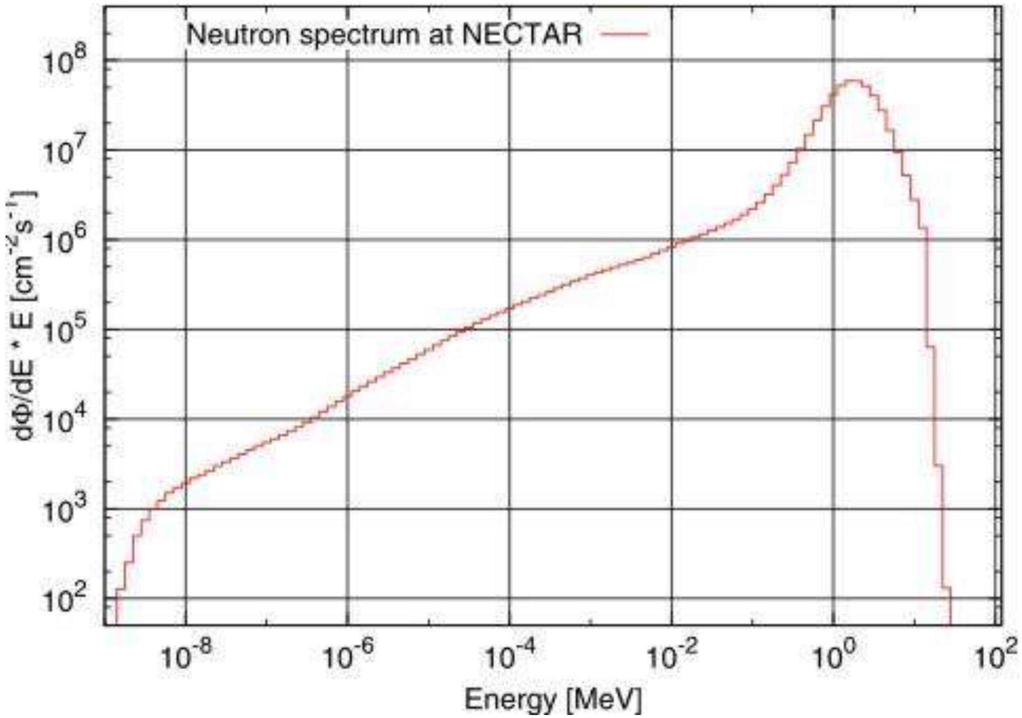


Beam quality: Fast neutron spectrum

- Thermal neutrons (without converter)
- Beam area: 23x18 cm²
- Φ_{th} : 3.9 x10^{9} s^{-1}cm^{-2}

- Fast neutrons (with converter)
- Beam area up to 30x20 cm (multi leaf collimator)
- Φ_{f}: 3.2 x10^{8} s^{-1}cm^{-2}
- Very small thermal neutron fraction
- Filters for adjustment of the n/γ-fraction

Unfolded spectrum

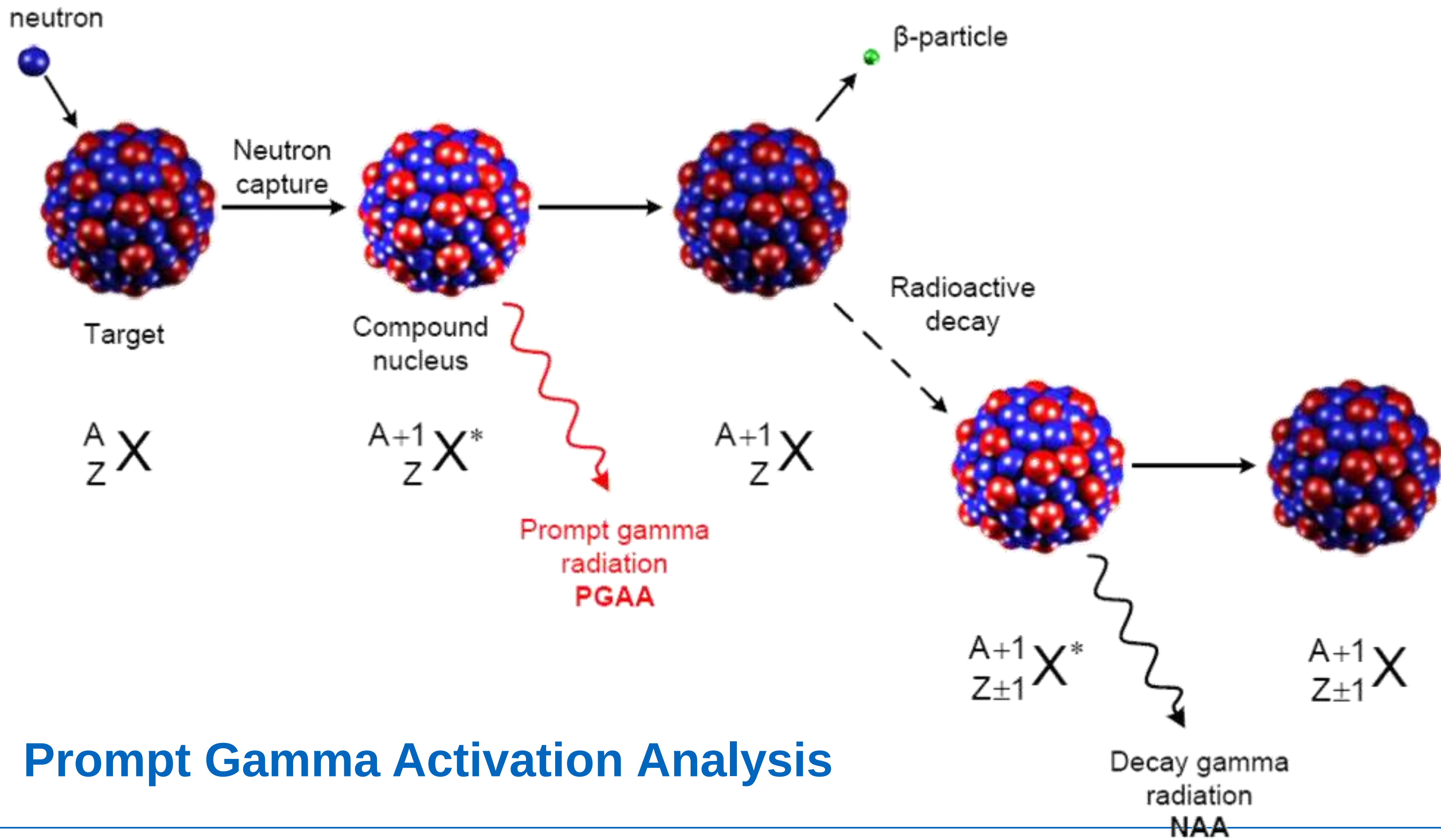


$\Longrightarrow$ Watt spectrum plus intermediate neutrons

F.M. Wagner, B. Loeper-Kabasakal, H. Breitkreutz, Th. Bücherl, S. Garni

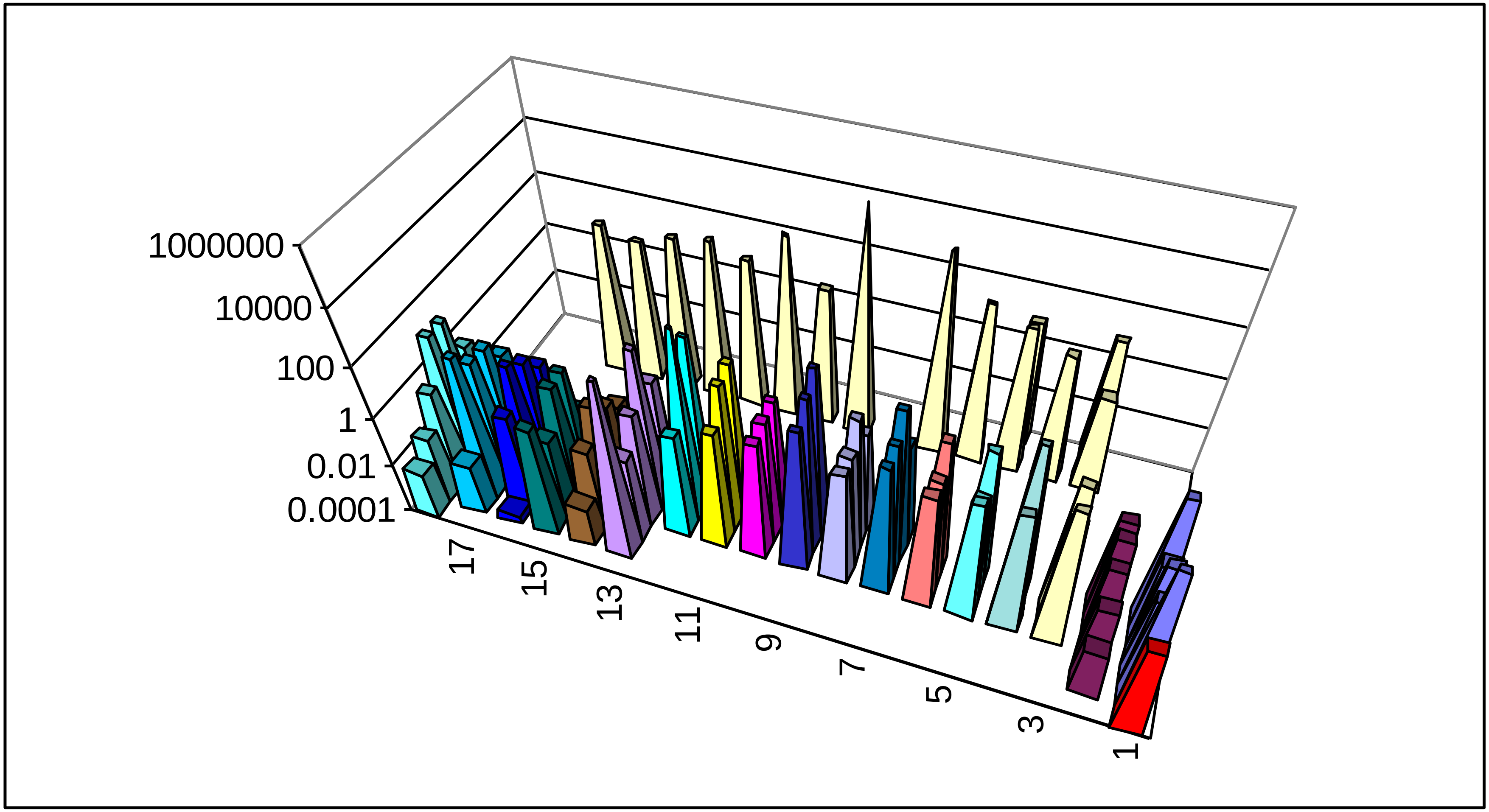
Prompt Gamma Activation Analysis
and
Garching PGAA facility

(n,γ) reaction



Prompt Gamma Activation Analysis

Neutron capture cross section

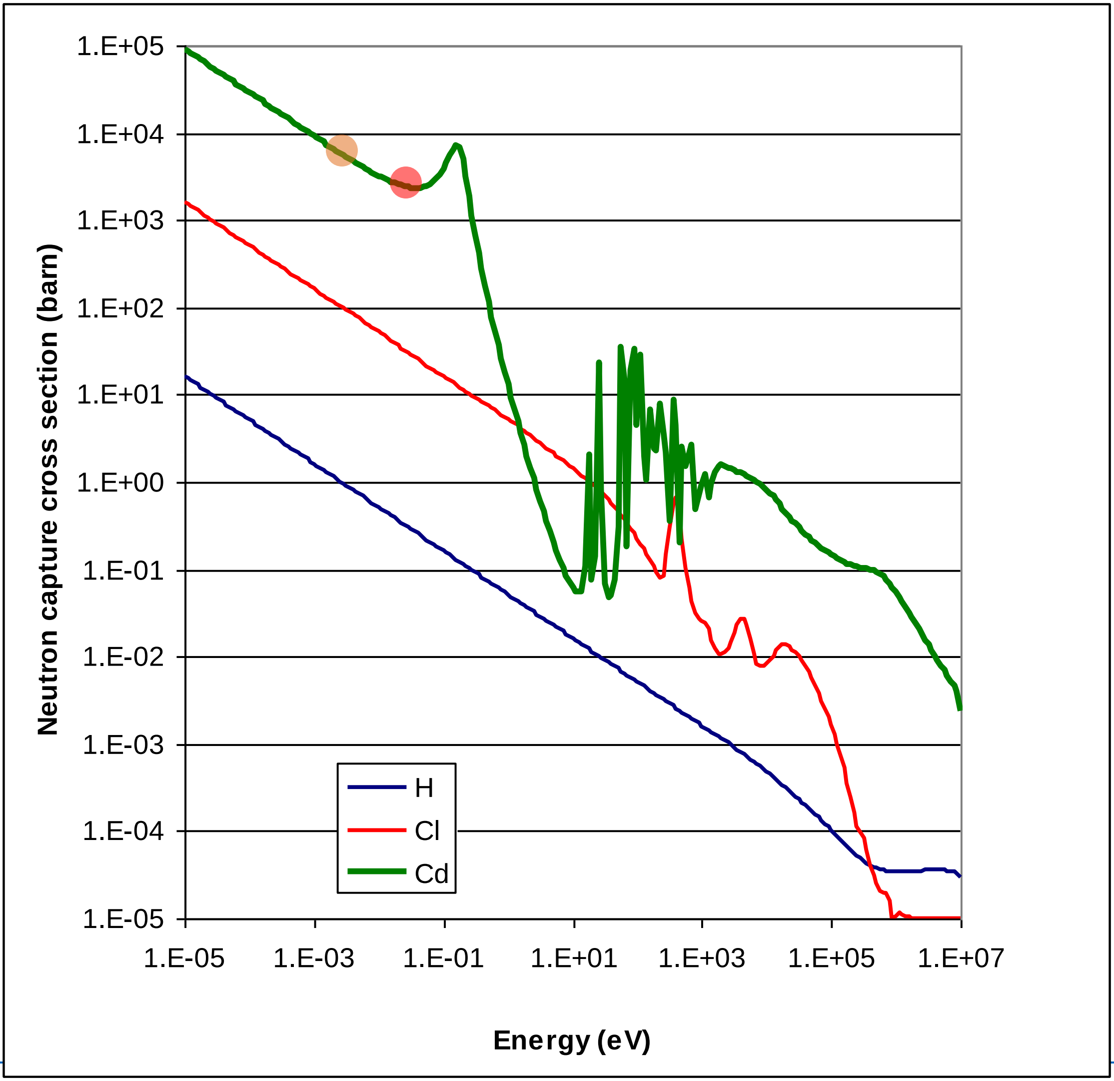


(n,γ) reaction with cold neutrons

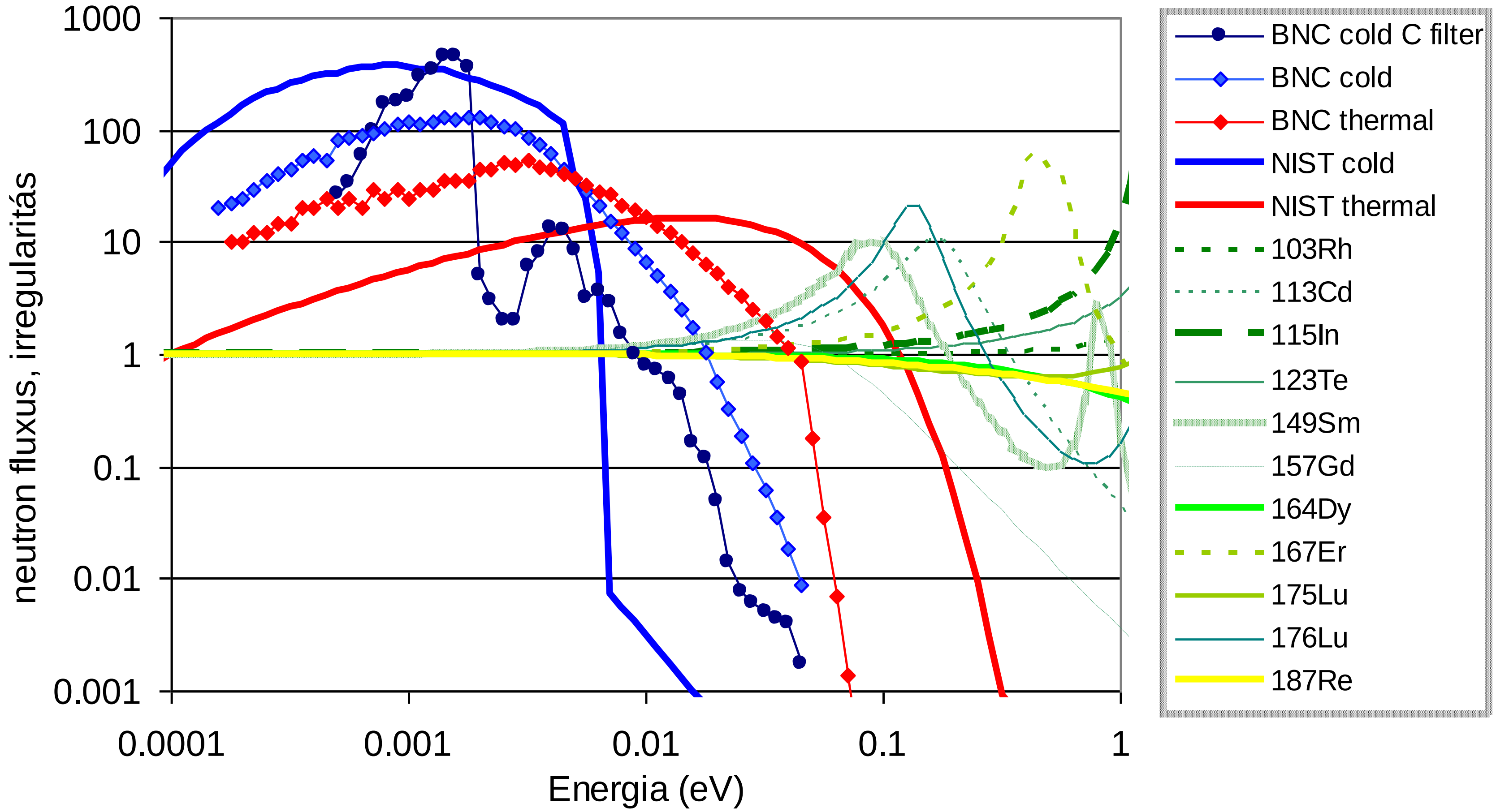
- De-excitation almost exactly from the binding energy
- Kinetic energy of neutrons <0.02eV
- All nuclides follow 1/*v* law
  - All cold beams excite the same way

2) Cross section
(probability of intraction)

- neutron capture cross section
- at low E: 1/*v* law
  - highest reaction rate for **cold neutrons**
- neutrons must be moderated
- resonance neutrons
  - RNCA (IRMM, Gel, Postma et al.)

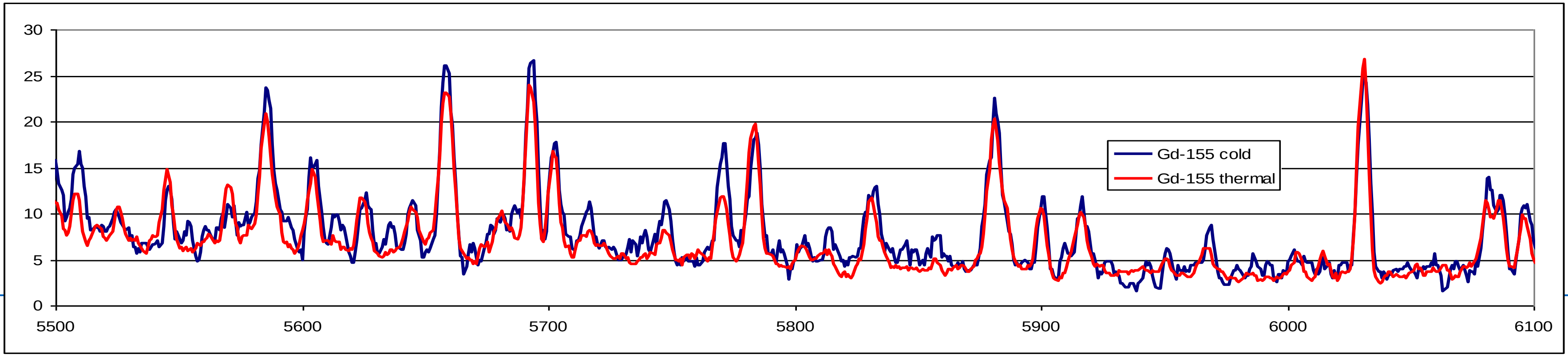


Activation of non 1/v nuclides



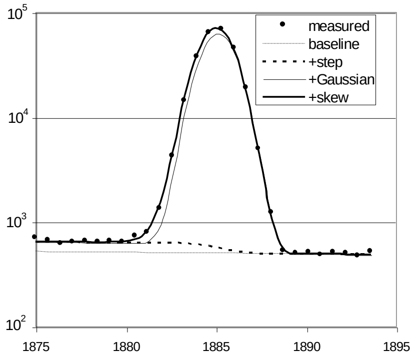
Radiative neutron capture

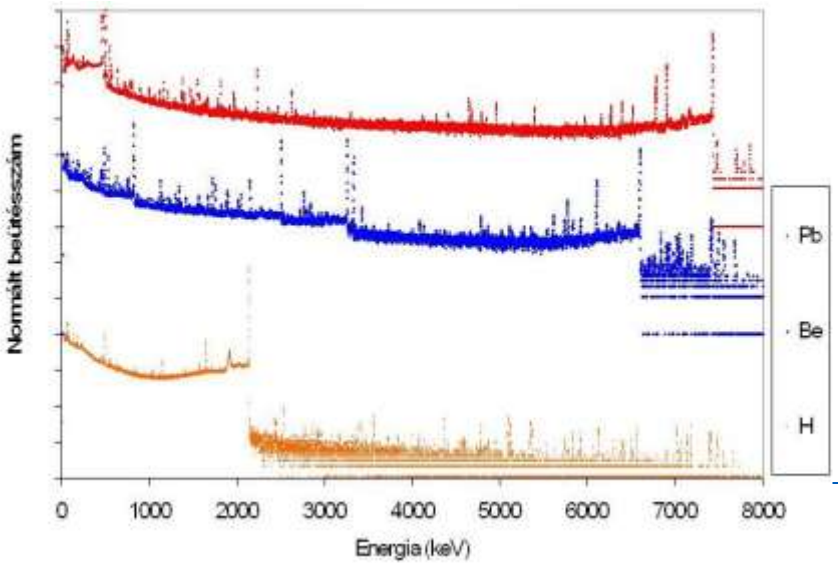
- Kinetic energy of neutron (<eV) is negligible compared to binding energy (S_n~MeV), or the detector resolution (~keV)
- Prompt de-excitation from capture state
  - Is the de-excitation independent of the beam energy?
  - Mostly yes, but close-lying resonance(s) can also be excited, from which de-excitation **can** be be different
    - Emission probabilities (mainly below S_n) may differ in different beams
      - ^{149}Sm, ^{157}Gd, ^{151}Eu, 10—20% deviations in thermal/cold beam
      - but not ^{113}Cd
      - Epithermal neutrons may cause further deviations (collimated beams)



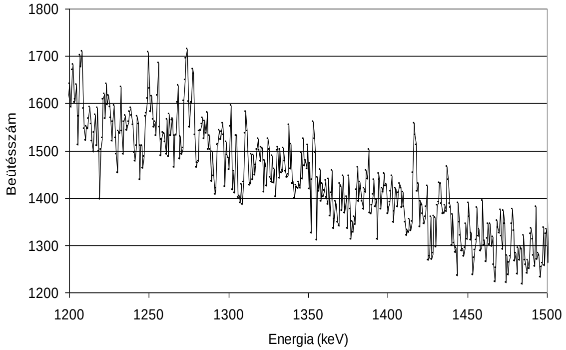
Spectrum types

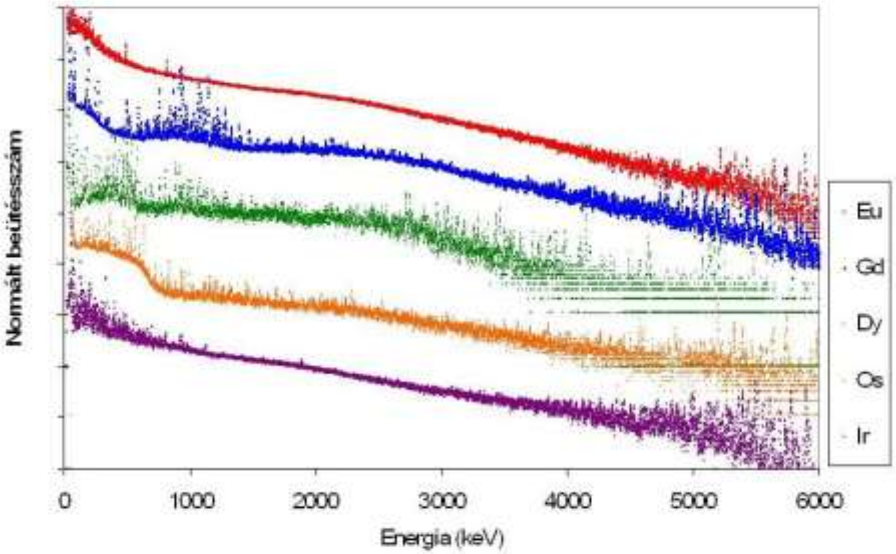
Type 1



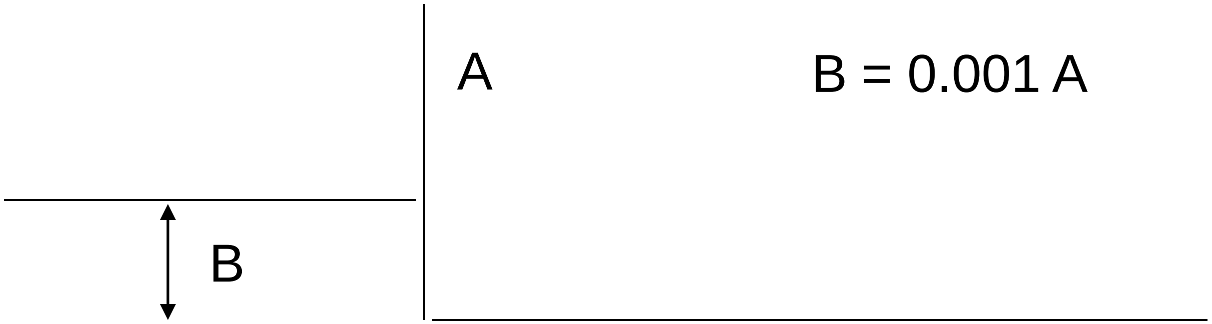


type 2





Simplified detector response function of collimated HPGe detectors



- Slope ~ peak density
- The missing peak areas can be estimated

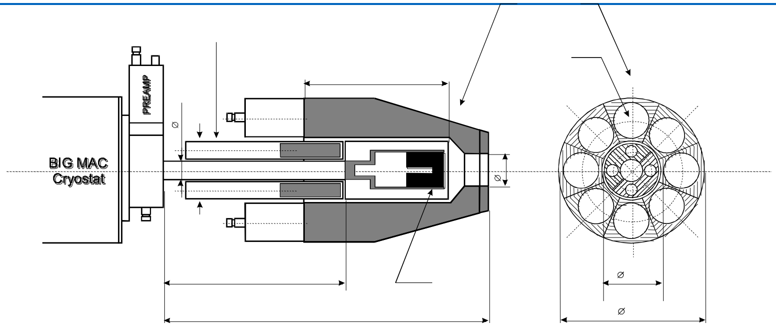
Properties of PGAA

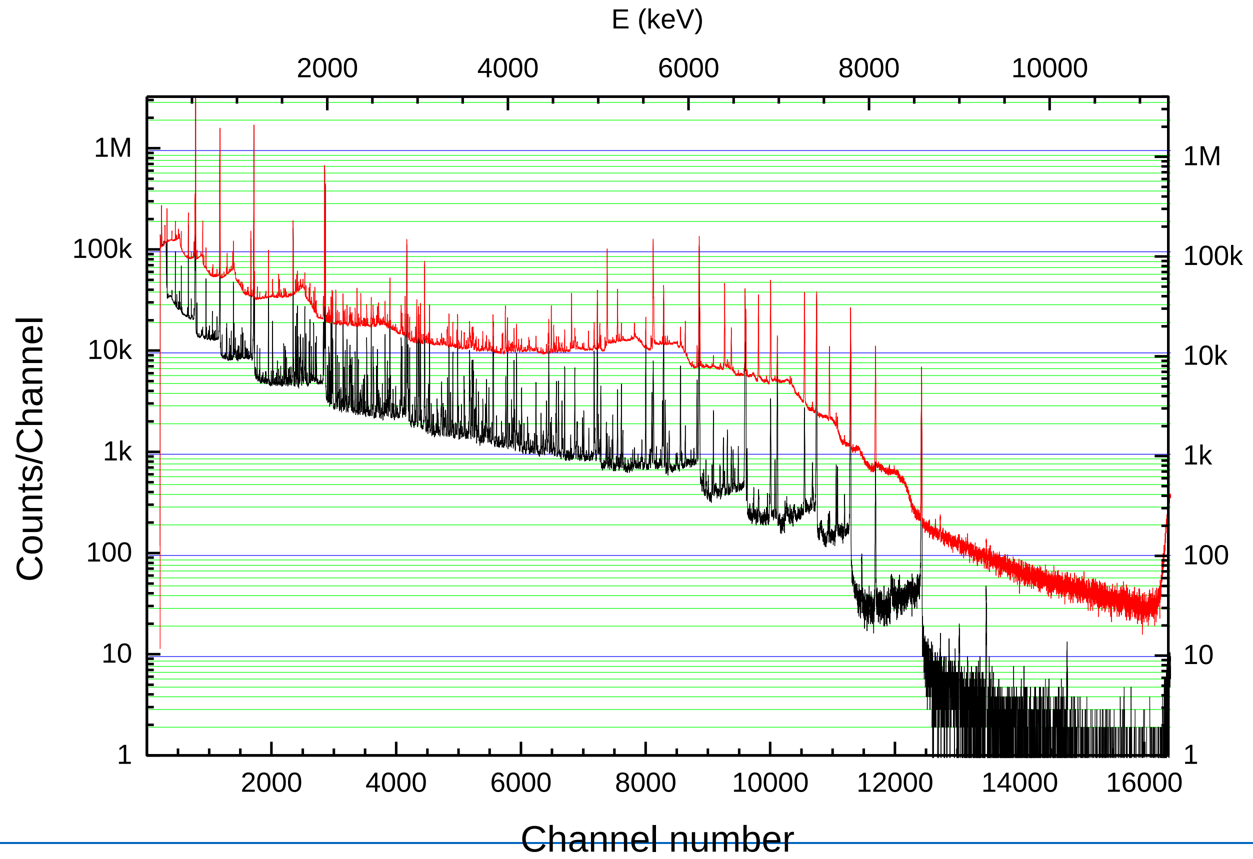
- Almost unattenuated penetration through material
  - Neutrons
  - Gammas
- Insensitive to details (surface, internal structure)
- Very linear signal: $A = n\, \Phi\, \sigma_\gamma$

  (activity = number of atoms × flux × cross-sec)

Active and passive shielding

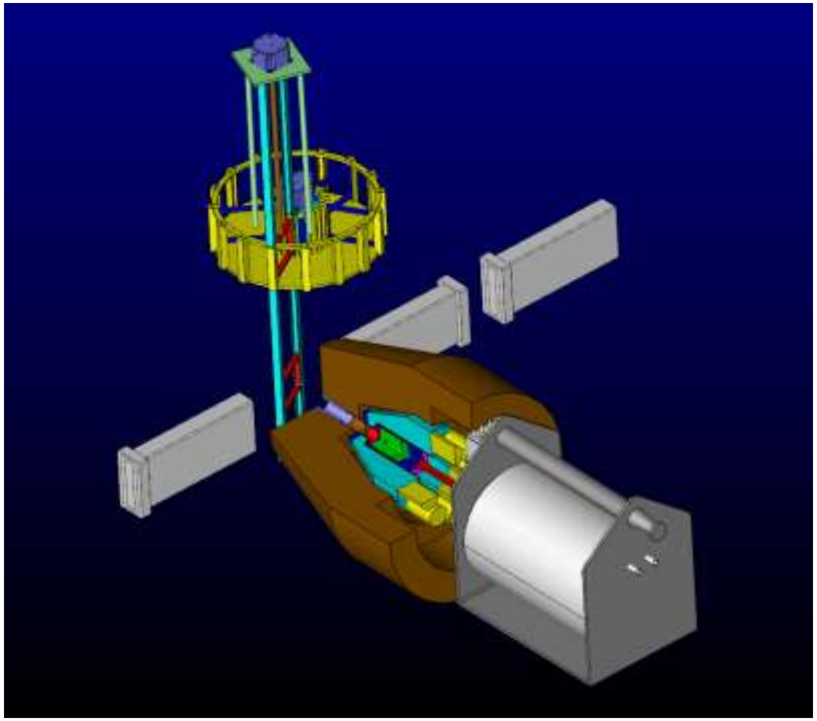
Compton suppression

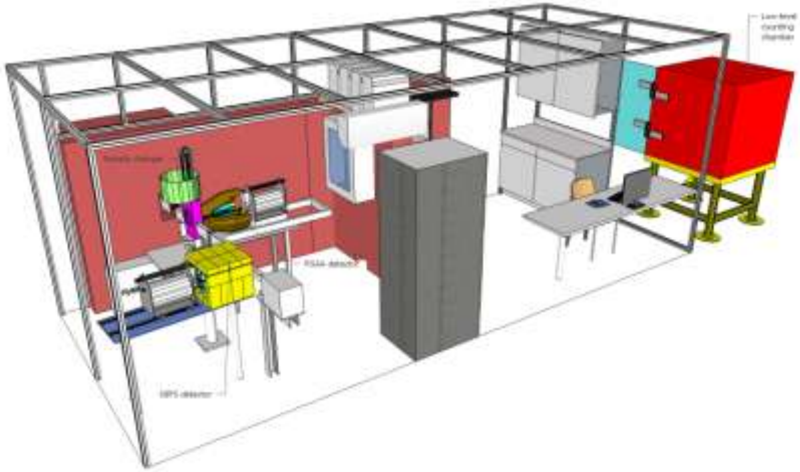




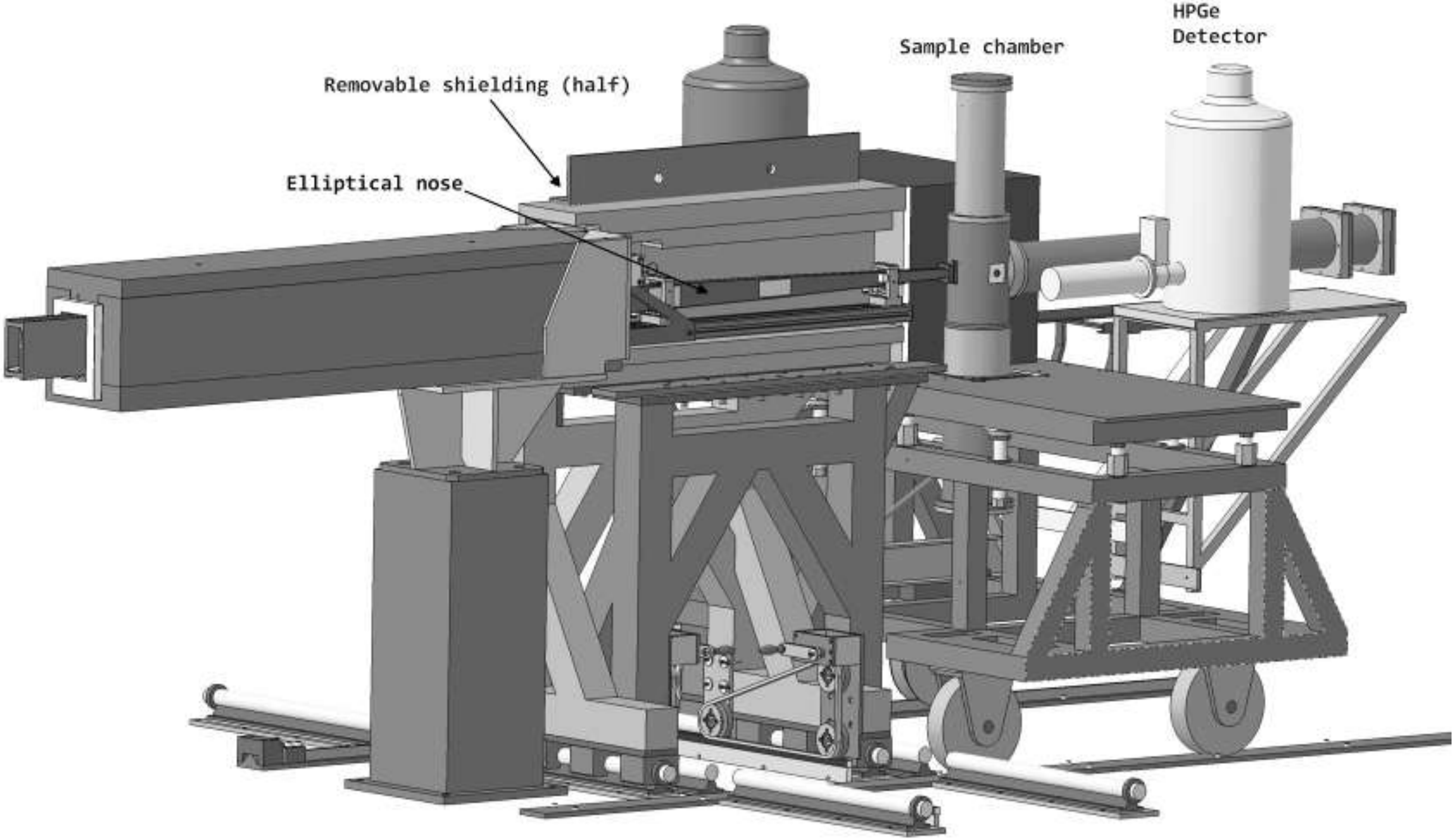
Budapest PGAA facility

- 10^8 cm^{-2} s^{-1}
- Cold beam
- Curved neutron guide
- At the end of the hall





PGAA facility at Garching

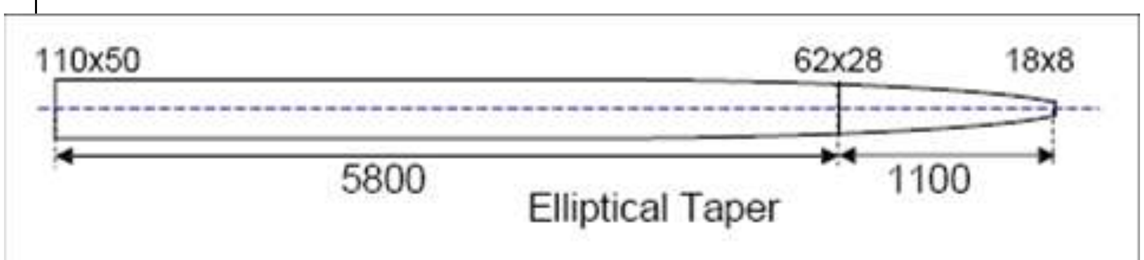


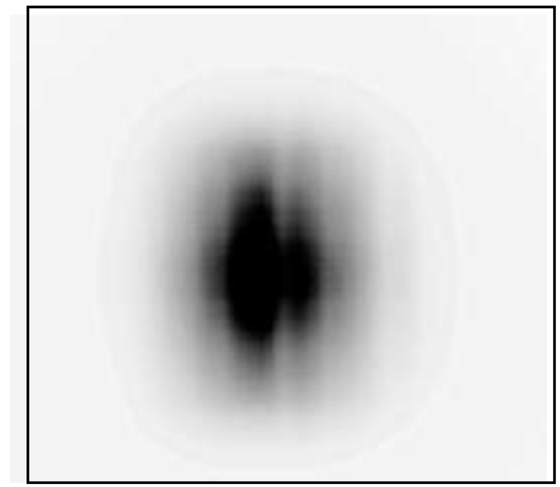
From L. Canella et al. NIM 636 (2010) 108

Latest reconstruction

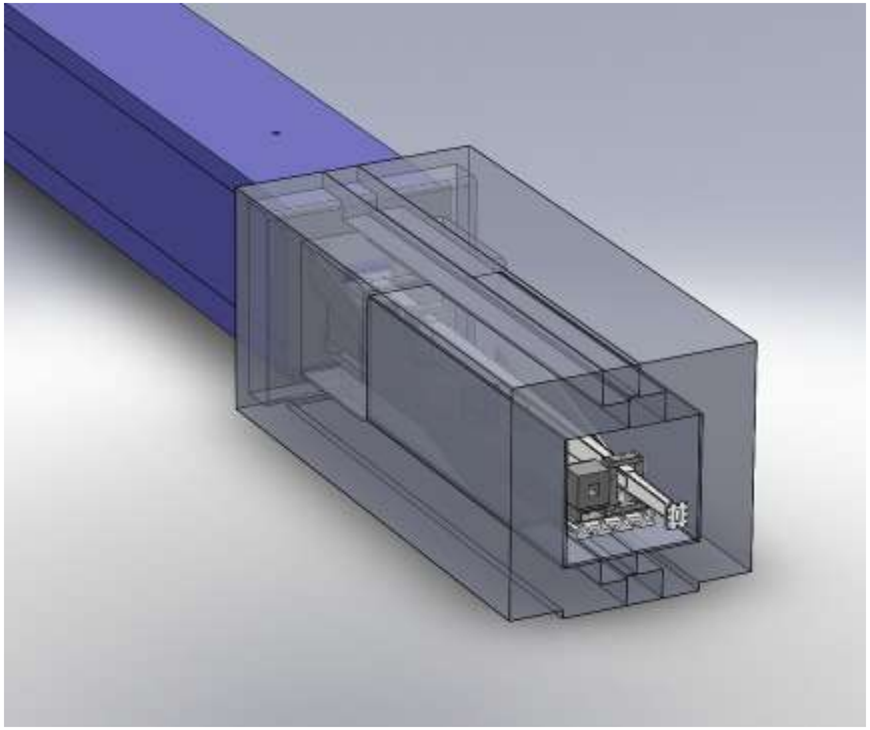
Focusing

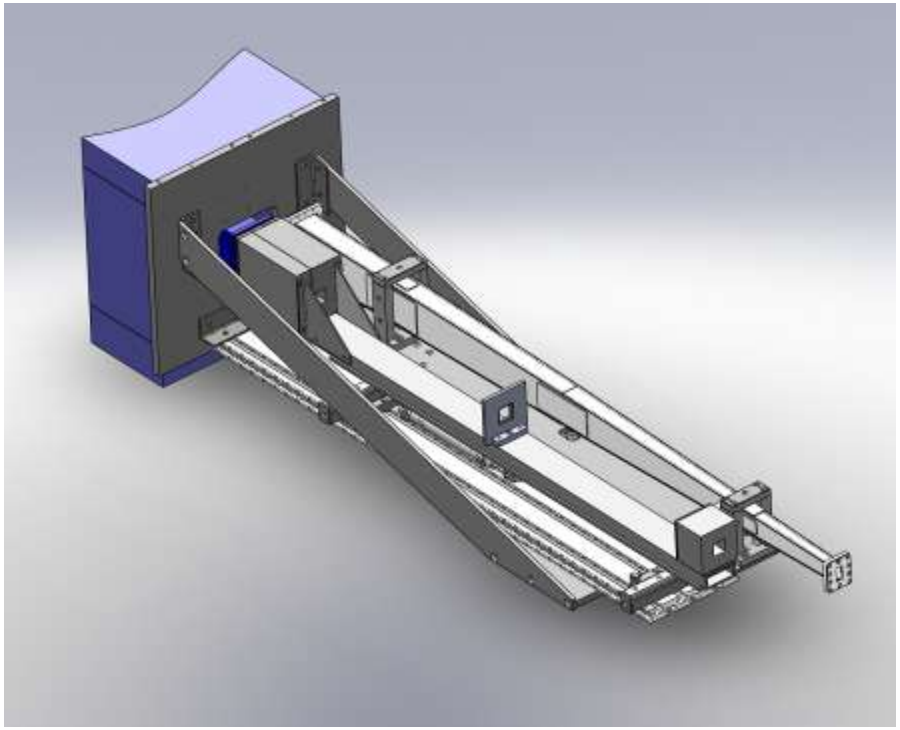
- 5.8 m elliptical guide
- 1.1 m elliptical extension (removeable)





Focusing guide interchangeable with a collimator

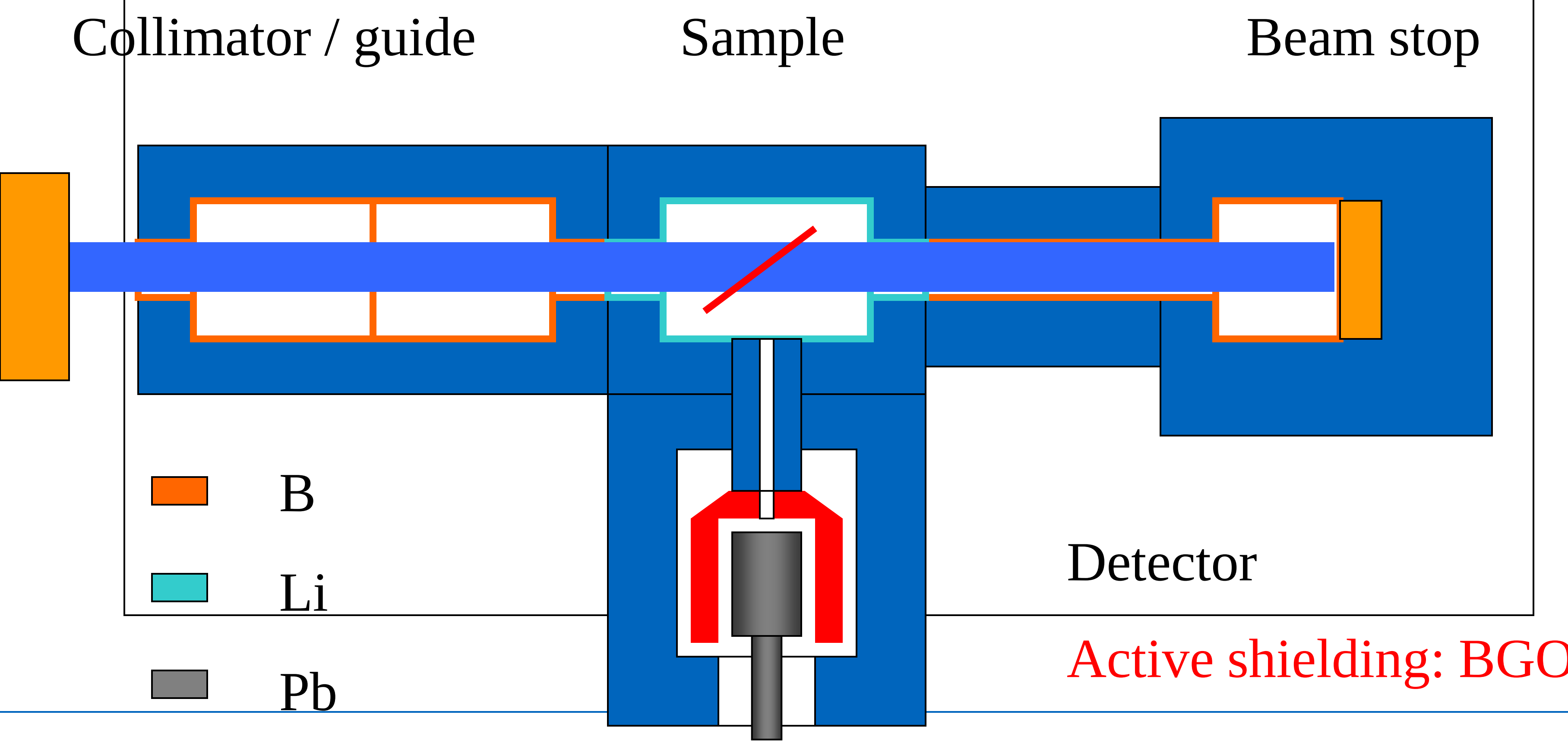




By Harald Türck

Shielding arrangements

Best arrangement: separate chambers



Complete shielding

Characteristics

- Highest flux: $4x10^{10}$ cm^{-2} s^{-1}
  - Earlier background ~7 000cps (max rate 10,000cps then)
  - New background 400 cps
- Working flux: $2x10^{9}$ cm^{-2} s^{-1}
  - Earlier background: 200 cps
  - New background: 11 cps
  - **Signal/background ratio: best in the world**

50,000

7000

500

200

20

Counting characteristics

- Flux: 4×10^{10} cm^{-2} s^{-1}
- Sample-to-detector distance: 33cm
- Counting efficiency: 10^{-4} — 10^{-5}
- Beam background: 400 cps
  - With sample holder and packing about 800cps
- Max. count rate: 50,000 cps
- Typical meas. time: 1000—100,000 s

Sample characteristics

- Min. sample mass: around 0.1mg
- Lowest cross section: 1μb

Radioactive samples:

- Max. activity: 10MBq
- Min. half-life: 100 years

Coincidence measurements

- Copton-suppressed detector with the efficiency of 10^{-4}
- Normal detector close to sample (efficiency ~ 0.01)
- The reaction rate is enought to have enough coinc. events
- No random (much less) random coinc.

Determination of Nuclear Data in Cold Neutron Beams

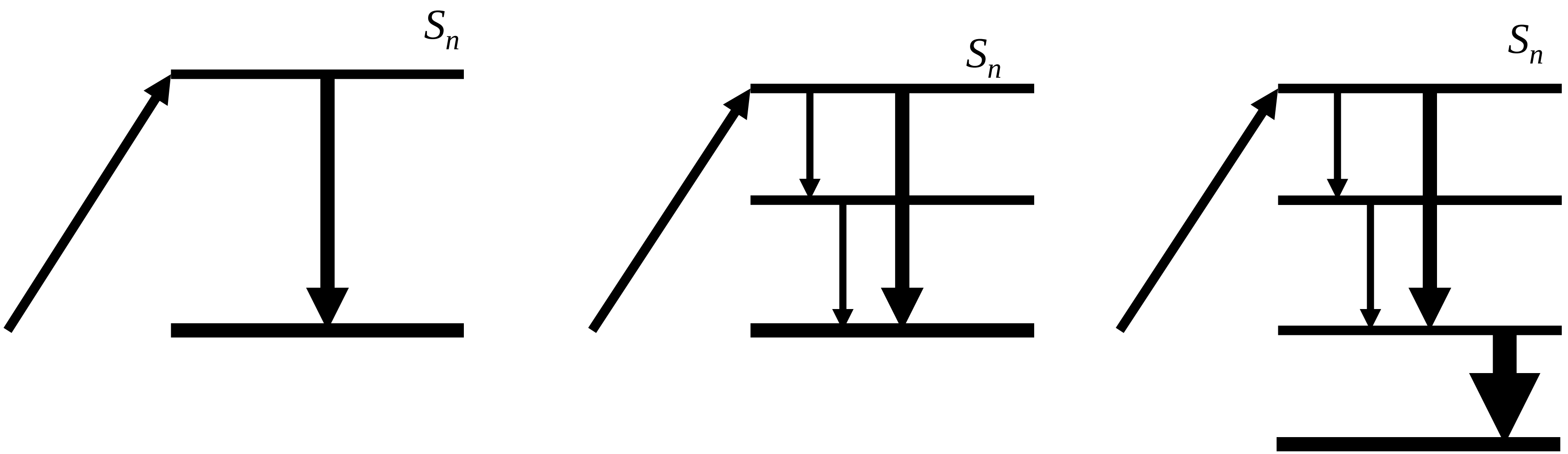
Capture-related cross sections

- Probability of neutron capture
  - Neutron capture cross-section: σ
  - Normally given for thermal energy (25meV): σ_0
- Probability for the emission of a given-energy gamma photon after neutron capture
  - Partial gamma-ray production cross-section: σ_γ

$$\sigma_\gamma = P_\gamma \, \theta \, \sigma$$

(emission probability × isotopic abundance × cross-section)

Radiative neutron capture
(n,γ) reaction



H, D

^{6}Li, ^{7}Li, ^{12}C

^{16}O

1) $\sigma = \sum E_\gamma / S_n \, \sigma_\gamma$

2) Sum of all primary transitions

3) Sum of all ground-state transitions

4) From decay of ground state

S_n – bindig energy of neutron

Spectroscopy data

- Partial gamma-ray production cross-section
  - Emission probability (relative intensity)
  - Proportional to the mass of the nuclide
- Characteristic energy
  - Identifies the emitter nuclide
- Other important data
  - Neutron capture cross-section
  - Half-life
  - Wescott *g* factor

Absolute method

- Activation equation: $A = n\ \Phi\ \sigma_\gamma\ \varepsilon\ t$
- Unknown: A, n (mass), Φ, or ε

$$\frac{A}{\varepsilon t} = \left(\frac{m}{M} N_A \right) \Phi_0 \left(\sigma_0\, \theta\, P_\gamma \right)$$

  - Peak area, efficiency, time
  - Mass, molar weight, (Avogadro constant)
  - Thermal equivalent flux
  - Partial gamma-ray production cross-section

Relative method

- If we know nothing of the beam…
- In case of homogeneous sample
- Activation: $A = n\,\Phi\,\sigma_\gamma\,\varepsilon\,t$

$$\frac{A_1/\varepsilon_1}{A_2/\varepsilon_2} = \frac{n_1}{n_2}\frac{\sigma_{\gamma,1}}{\sigma_{\gamma,2}}$$

  - Peak-area ratio
  - Efficiency ratio
  - Molar ratio (mass ratio)
  - Partial cross-section ratio

Comparator methods

- Activation equation: $A = n\,\Phi\,\sigma_\gamma\,\varepsilon\,t$
- Metal foil measurements
- Flux monitoring
- Before, during, after measurement

PGAA spectroscopy library

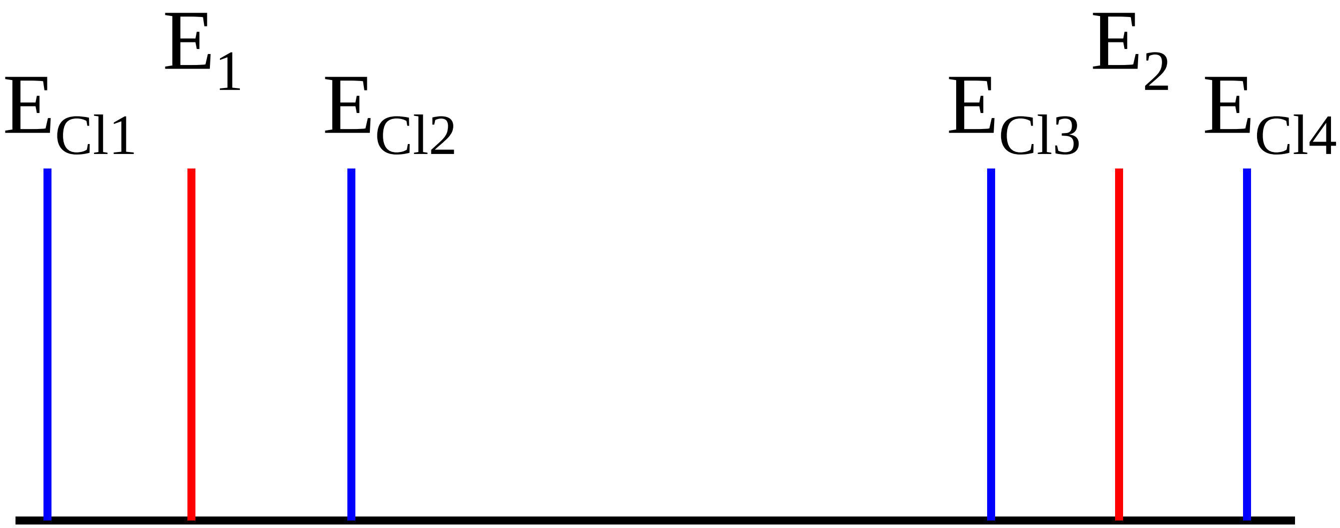
1. Elemental spectra: to obtain ...
   - Relative positions
   - Relative intensities
2. Energy calibration: to absolutize energy scale
   - 2 energies det-d for the 2-point E-calibration
   - Non-linearity
3. Standardization: to absolutize intensity scale
   - efficiency ratios
   - compounds or mixtures

1. Elemental spectra

- Elements
  - Metals
- Oxides
  - Nonmetals
  - Metals
- Other simple compound with low-Xsec elements
  - Carbonates, hydroxides, carbides, nitrates etc.

2. Energy calibration

- Energy difference method
- Crystal spectrometer data for ^{35}Cl
- Element measured in presence of Cl



3. Standardization

- Stoichimetric compounds
  - Chlorides, nitrates
- Homogeneous mixtures
  - Water solution
  - Water-TiO_2-XO suspension

3. Comparators

- C, N, O, F, S, Cl, Fe (Au)
- Ultimate comparator: H
  - 2223.255 keV 0.3326(7) barn
- One-, or two-step standardization

$$\frac{A_1 / \varepsilon_1}{A_2 / \varepsilon_2} = \frac{n_1}{n_2} \frac{\sigma_{\gamma,1}}{\sigma_{\gamma,2}}$$

Elements measured in Budapest 1997-2000

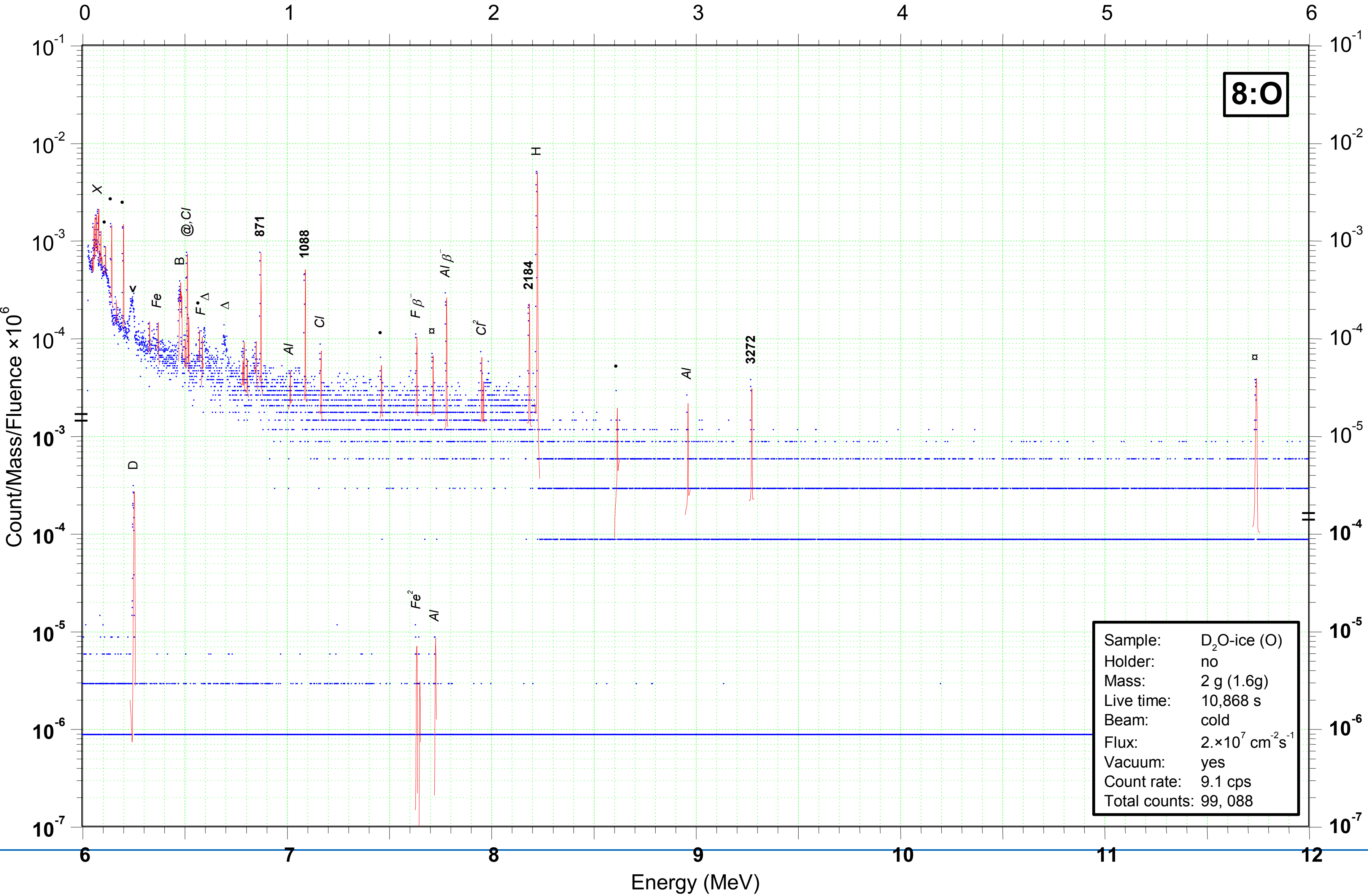
1 H O			1 D *O*														2 He
3 Li CO_3,*C-F*	4 Be * O											5 B *C*, H-O	6 C ** H	7 N C-D-O, NO_3	8 O H, Be	9 F C	10 Ne
11 Na * CO_3,C-H-O	12 Mg *											13 Al ** O	14 Si * O N	15 P * **O**	16 S **	17 Cl **C,C-H**	18 Ar *
19 K HCO_3	20 Ca * O CO_3	21 Sc O	22 Ti ** O	23 V O	24 Cr * O-H	25 Mn * O	26 Fe **	27 Co *	28 Ni **	29 Cu * O	30 Zn * O	31 Ga **	32 Ge * O	33 As O	34 Se * O-H	35 Br * C-H	36 Kr *
37 Rb O CO_3	38 Sr CO_3	39 Y O	40 Zr O	41 Nb O	42 Mo **	43 (Tc)	44 Ru **	45 Rh * C-H	46 Pd *	47 Ag **	48 Cd **	49 In *	50 Sn **	51 Sb O	52 Te **	53 I * **C-H**	54 Xe F
55 Cs O	56 Ba OH,CO_3	57 La O	72 Hf * O	73 Ta * O	74 W O	75 Re *	76 Os * O C-H	77 Ir * O	78 Pt *	79 Au *	80 Hg ** O	81 Tl *	82 Pb **	83 Bi **	84 (Po)	85 (At)	86 (Rn)
87 (Fr)	88 (Ra)	89 (Ac)															

58 Ce O C-H-O	59 Pr O	60 Nd O	61 (Pm)	62 Sm **O**	63 Eu **O**	64 Gd **O**	65 Tb O	66 Dy *	67 Ho O	68 Er **O**	69 Tm O	70 Yb O	71 Lu O
90 Th NO_3	91 (Pa)	92 U ***O*** C-H-O											

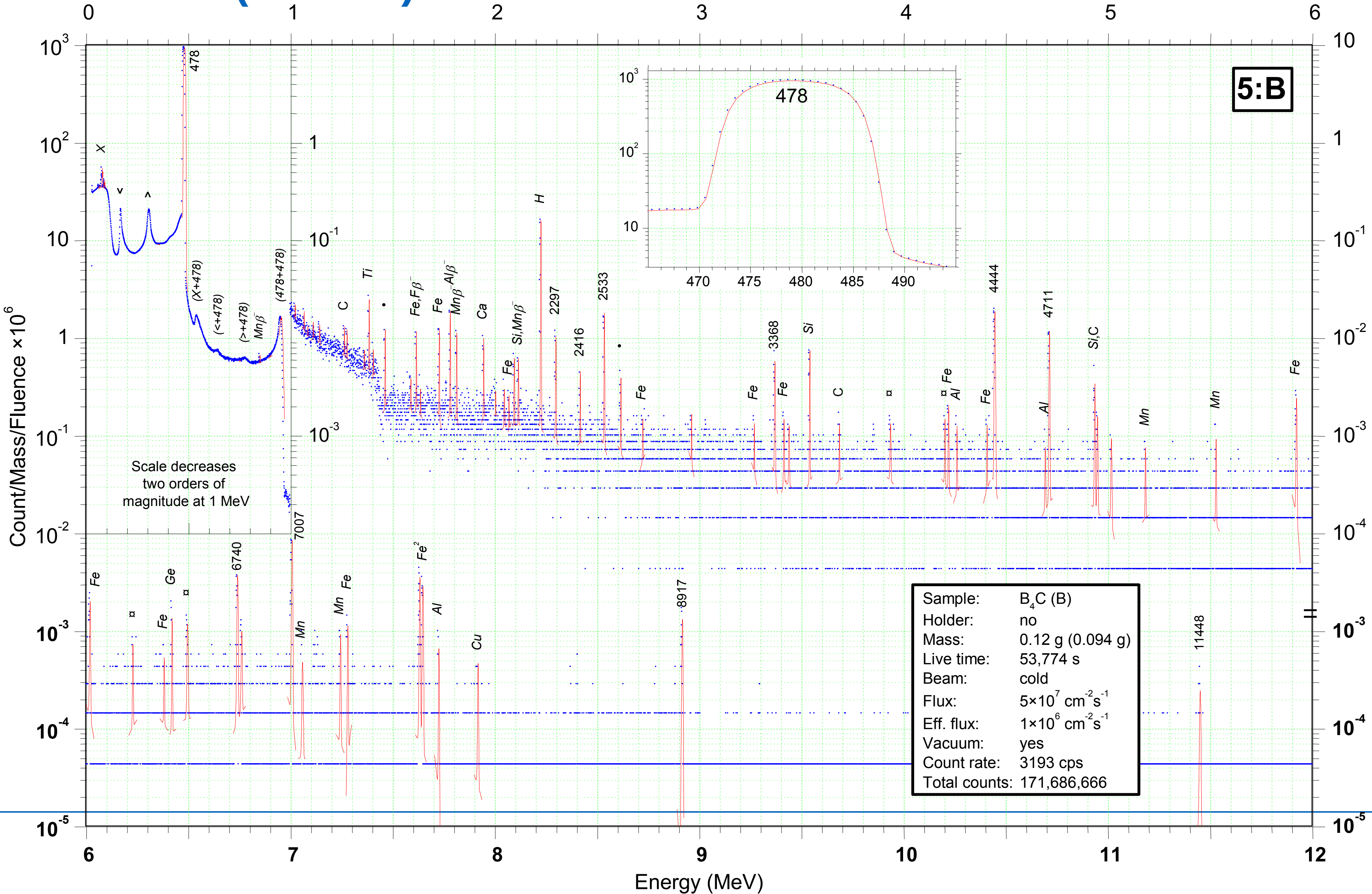
PGAA library

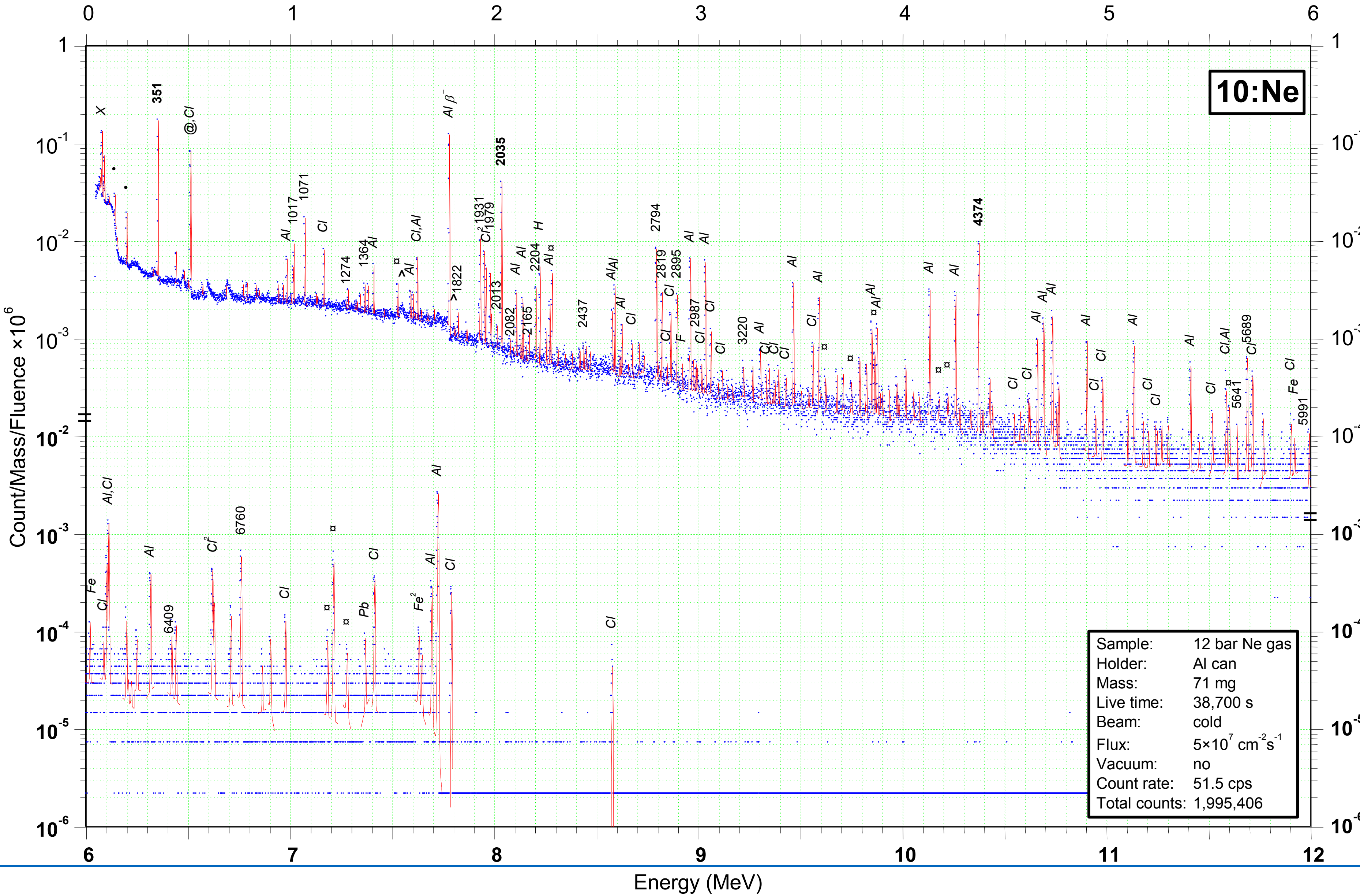
Z	El	A	MW	#	E	dE	σ	dσ %	RI	Area	cps/g
1	H	1	1.01	1	2223.259	0.019	0.3326	0.2	100.00	100.00	64.183
1	H	2	1.01	2	6250.204	0.098	0.000492	5.0	0.15	5.00	0.0286
3	Li	6	6.94	5	477.586	0.050	0.001399	5.9	3.52	10.14	0.1218
3	Li	7	6.94	2	980.559	0.046	0.004365	5.1	10.97	18.74	0.2251
3	Li	7	6.94	3	1051.817	0.048	0.004364	5.1	10.97	17.83	0.2141
3	Li	7	6.94	1	2032.310	0.070	0.0398	5.0	100.00	100.00	1.2007
3	Li	6	6.94	6	6769.633	0.263	0.001354	6.5	3.40	0.84	0.0101
3	Li	6	6.94	4	7246.800	0.275	0.002106	8.4	5.29	1.17	0.014
4	Be	9	9.01	4	853.631	0.011	0.00165	8.9	26.69	100.00	0.0723
4	Be	9	9.01	3	2590.014	0.025	0.00188	8.9	30.41	49.08	0.0355
4	Be	9	9.01	2	3367.484	0.035	0.002924	8.9	47.30	58.96	0.0427
4	Be	9	9.01	5	3443.421	0.036	0.000993	8.9	16.06	19.54	0.0141
4	Be	9	9.01	6	5956.602	0.092	0.000146	9.1	2.36	1.41	0.001
4	Be	9	9.01	1	6809.579	0.099	0.006181	9.0	100.00	48.52	0.0351
5	B	10	10.81	1	477.600	5.000	712.5	0.3	100.00	100.00	39806
6	C	12	12.01	2	1261.708	0.057	0.00123	2.7	45.58	100.00	0.0306
6	C	12	12.01	3	3684.016	0.069	0.001175	3.5	43.53	38.02	0.0116
6	C	12	12.01	1	4945.302	0.066	0.002699	2.9	100.00	60.55	0.0186
7	N	14	14.01	22	583.567	0.031	0.000429	3.3	1.81	6.93	0.0159
7	N	14	14.01	12	1678.244	0.029	0.006254	1.5	26.34	47.15	0.1085
7	N	14	14.01	18	1681.174	0.043	0.001296	2.7	5.46	9.76	0.0225
7	N	14	14.01	21	1853.944	0.052	0.000474	4.5	2.00	3.31	0.0076
7	N	14	14.01	5	1884.853	0.031	0.0145	1.3	61.07	100.00	0.2301
7	N	14	14.01	24	1988.532	0.077	0.000294	5.8	1.24	1.94	0.0045
7	N	14	14.01	15	1999.693	0.032	0.003208	1.7	13.51	21.12	0.0486
7	N	14	14.01	13	2520.446	0.039	0.004246	1.8	17.88	22.98	0.0529

PGAA spectrum of O (heavy water ice in vacuum)



Boron (carbide)

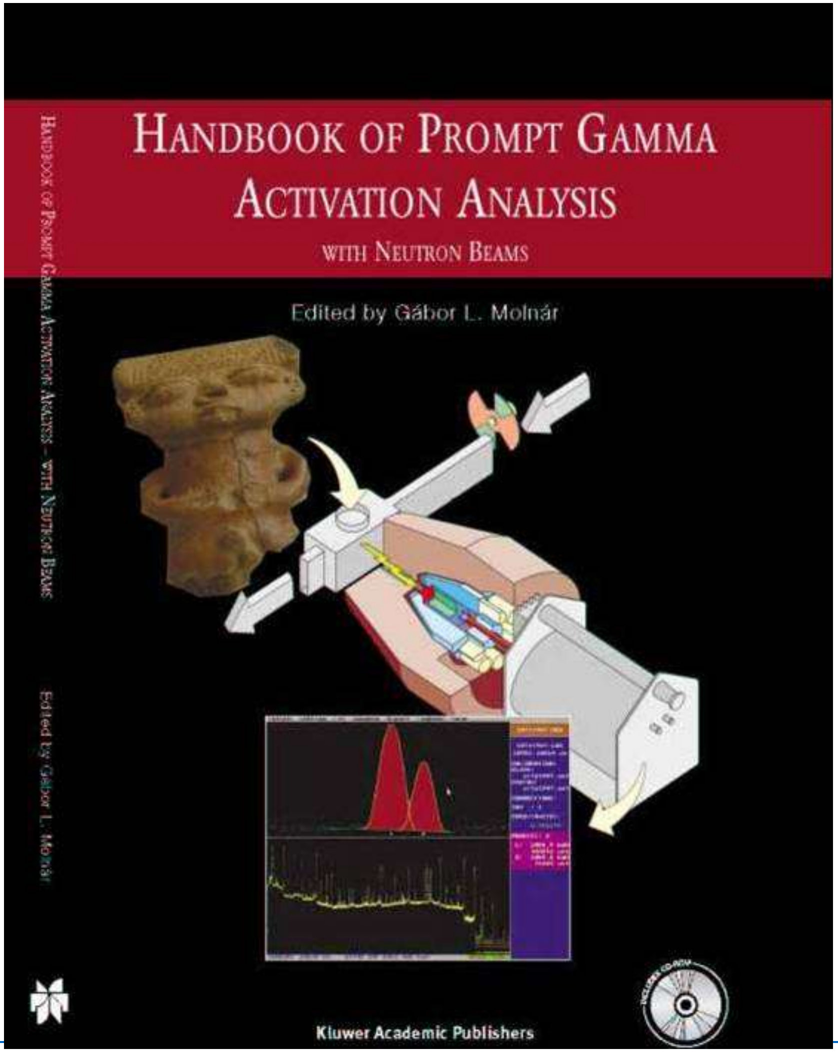



10:Ne
Count/Mass/Fluence ×10^6
Energy (MeV)
351
2035
4374
Sample: 12 bar Ne gas
Holder: Al can
Mass: 71 mg
Live time: 38,700 s
Beam: cold
Flux: 5×10^7 cm^-2 s^-1
Vacuum: no
Count rate: 51.5 cps
Total counts: 1,995,406

Compilations (by Rick Firestone)

- Analytical database
  - Handbook
- For nucl. physics
  - EGAF

- Further measurements
- Re-evaluation
- Publication



Measurement of low-cross-section nuclides

- Initiated by Rick Firestone
- Started in Budapest and NIST (Washington)
- Stable isotopes Z<10
- Some cross sec. data are really poor quality

Cross-sections of light nuclides

Nuclide	Abu. %	Xsec (mb)
^{1}H	99.985	332.6±0.2%
^{2}D	0.015	0.52 ±1.3%
^{3}He	1.4ppm	0.03±30%
^{3}He(n,p)		5333 barn
^{4}He	99.9999	0
^{6}Li	7.5	39 ± 8%
^{6}Li(n,α)		940 barn
^{7}Li	92.5	45 ± 7%
^{9}Be	100	8.8 ± 11%
^{10}B	19.9	500±40%
^{10}B(n,α)		3837 barn
^{11}B	80.1	5.5±60%

Nuclide	Abu. %	Xsec (mb)
^{12}C	98.89	3.53±2%
^{13}C	1.11	1.37±3%
^{14}N	99.634	79.8±2%
^{14}N(n,p)		1830 barn
^{15}N	0.366	24±33%
^{16}O	99.762	0.19±10%
^{17}O	0.038	0.54±17%
^{17}O(n,α)		235 barn
^{18}O	0.2	0.16±7%
^{19}F	100	9.6±5%

Experiments at FRM II

D	Heavy water (99.98—92%) + comparator
He	^{3}He – spin filter chamber (planned)
Li	nat. Li, ^{7}LiF
Be	$Be(NO_3)_2$
B	H_3BO_3, $^{11}B_2O_3$
C	Urea: $((NH_2)_2CO)$, Urea-D, Urea-^{13}C ^{15}N, ^{13}C
N	Urea-D, Urea-^{13}C ^{15}N
O	Water, heavy water, $H_2{}^{18}O$, $H_2{}^{17\text{-}18}O$, $D_2{}^{18}O$
F	Teflon

Earlier results

Elem	literature	Budapest 1999	Bp-NIST 2004
^{1}H	0.3326(7)	standard	standard
^{2}D	0.000519(7)	0.000490(10)	0.000497(13)
^{12}C	0.00351(5)	0.00376(2)	0.00384(7)
^{14}N	0.0795(14)		0.0792
^{16}O	0.000190(19)	0.000180(15)	0.000184(3)

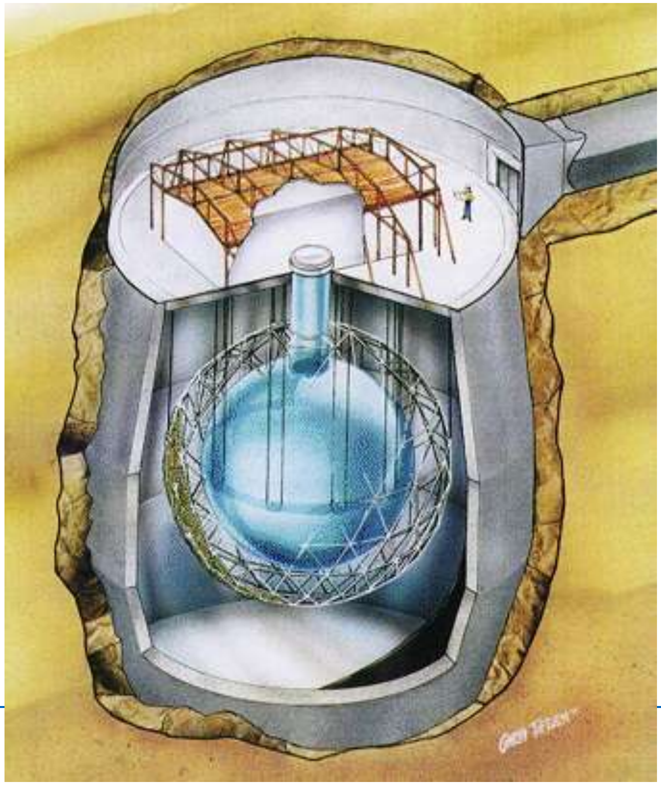
Uncertainties still too high

Effects of pollutants

- Systematic error caused by pollutants
  - D_2O: 100 ppm H_2O → 7.4 % virtual increase
  - C: 100 ppm N → 3.8 % virtual increase
  - C: 1 ppm Cd → 7.0 % virtual increase

Applications

- Better nuclear data
- Better PGA analysis
- Improved model calculations in reactor and in particle physics



Plans

- Other low-cross-section elements/nuclides (new proposal):

  Medium-Z elements:

  ^{27}Al, $^{90\text{-}92,94,96}$Zr,

  Heavy elements:

  $^{204,206\text{-}208}$Pb, ^{209}Bi, ...

TANDEM project

- Trans-Uranium Actinides' Nuclear Data – Evaluation and Measurement
- Collaboration of
  - Jülich (FZJ)
  - Garching (FRM II, TUM)
  - Budapest (CER, HAS)
  - Berkeley (LBNL)
- To improve (n,γ) data for trans-U actinides

Further calculations from the spectroscopy library (EGAF)

- Dicebox calulations
- Capture corss sections
  - K isotopes (Berkeley)
  - Pd isotopes (Berkeley, Livermore)
  - Cu (Berkeley)

Present measurements

- Gd
  - GdB_6
- Eu
  - $EuCl_3$
- Ge isotopes
  - Together with the Oslo group
- N calibration standard
  - Efficiency and level-scheme fit at the same time

Dead line for beam time applications: 25 July

Thank you