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international centre for theoretical physics

H4.SMR/1503 - 05

**WORKSHOP ON NUCLEAR DATA FOR SCIENCE AND
TECHNOLOGY: MATERIALS ANALYSIS**

(19 - 30 May 2003)

**Prompt gamma activation analysis
Part 1: Principles**

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**Workshop on Nuclear Data for Science and Technology:
Materials Analysis
ICTP Trieste, 19-30 May 2003**

Prompt gamma activation analysis Part 1: Principles

Gábor L. Molnár

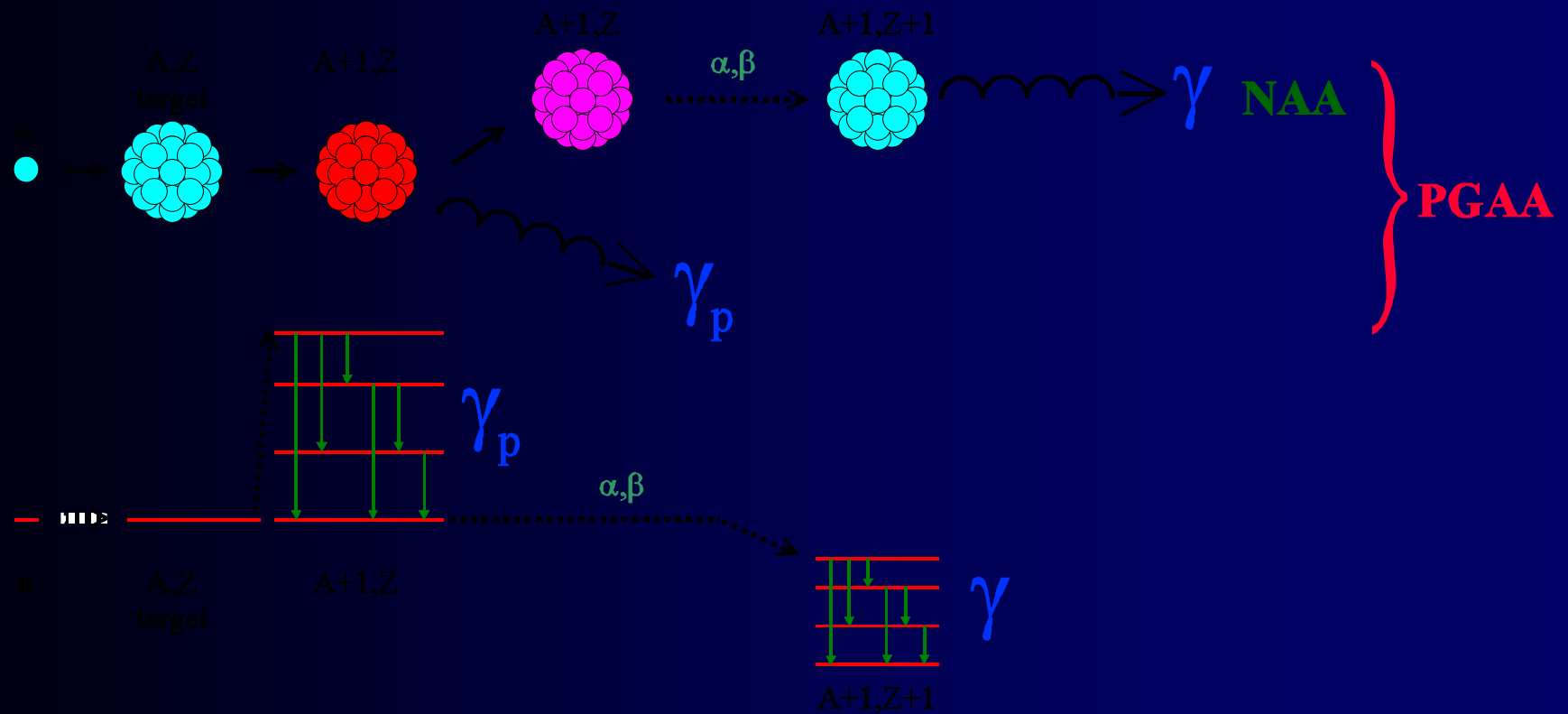
**Institute of Isotope and Surface Chemistry
Chemical Research Centre
Budapest, Hungary**

Neutron-induced prompt gamma activation analysis (PGAA or PGNAA)

- **Physical method:** detection of **prompt** gamma radiation from a neutron-induced nuclear reaction
 - radiative capture (all elements and isotopes)
 - fission (actinides)
- **Active method:** sample subjected to neutron radiation
- **Non-destructive method:** no (or little) residual radioactivity
- **Instantaneous method:** results appear promptly
- **Absolute method:** no standards needed

1. Physical basis of PGAA

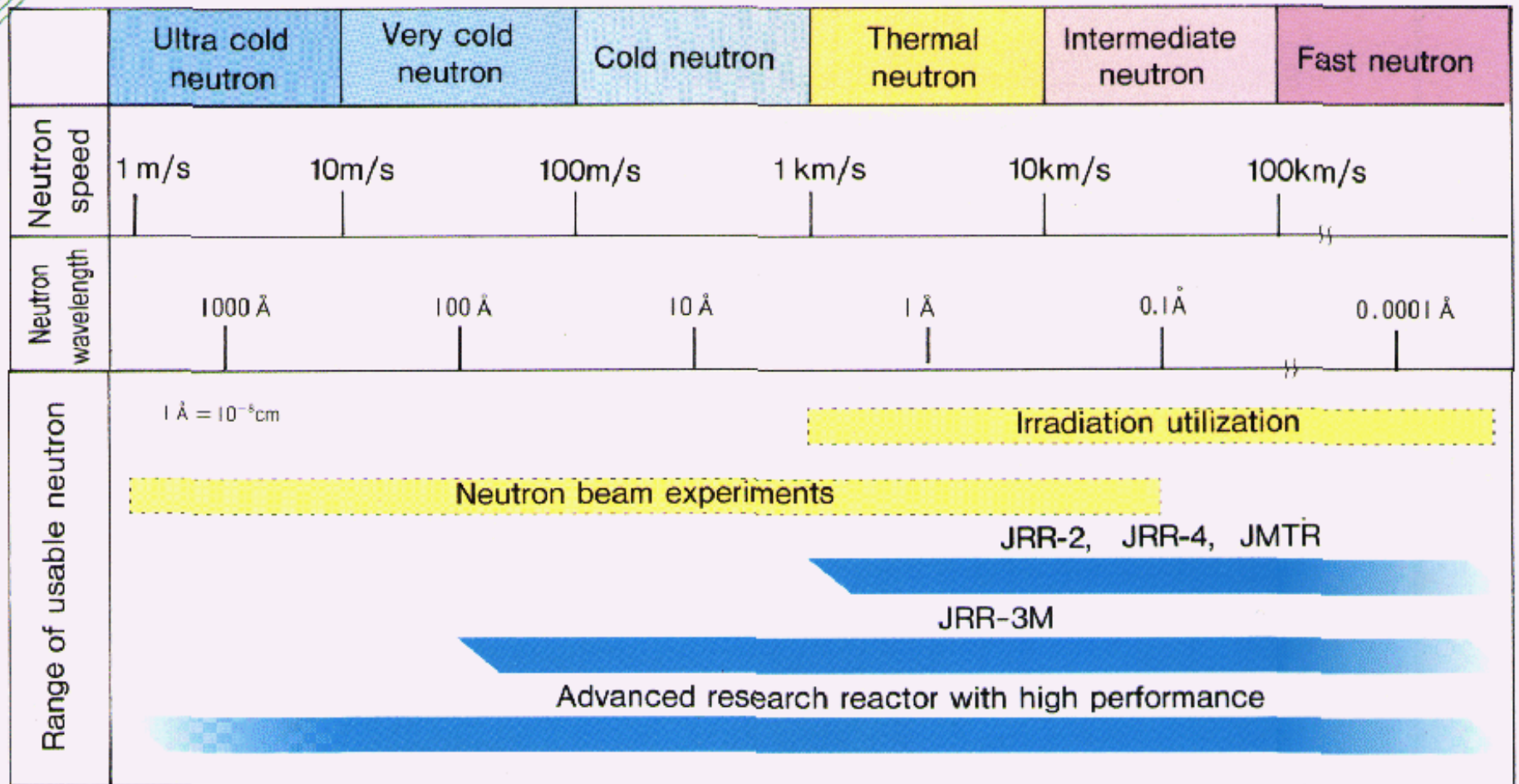
Neutron capture and subsequent beta decay



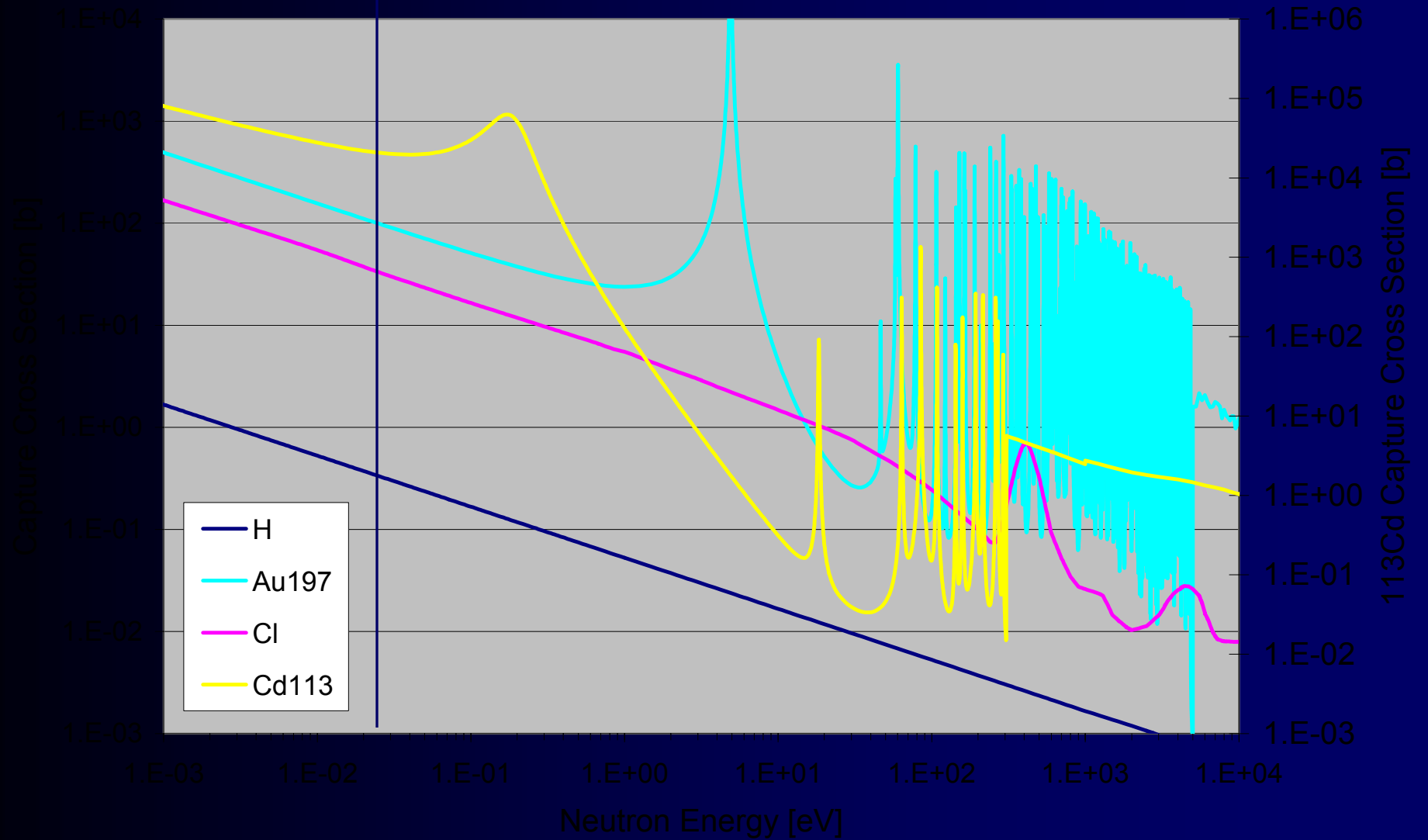
Basic reaction: Radiative neutron capture

- Most important reaction for slow neutrons
 - High cross sections (but vary widely!) >> all elements
 - Simple $1/v$ behavior (exceptions!) >> easy quantification
 - Reaction rate \sim amount & time >> increase sensitivity
- Complementary reactions at high/low energies
 - Inelastic scattering >> C, N, O, Pb
 - (n,p) etc. >> O, Pb (with NAA)
 - Low-energy fission >> actinides

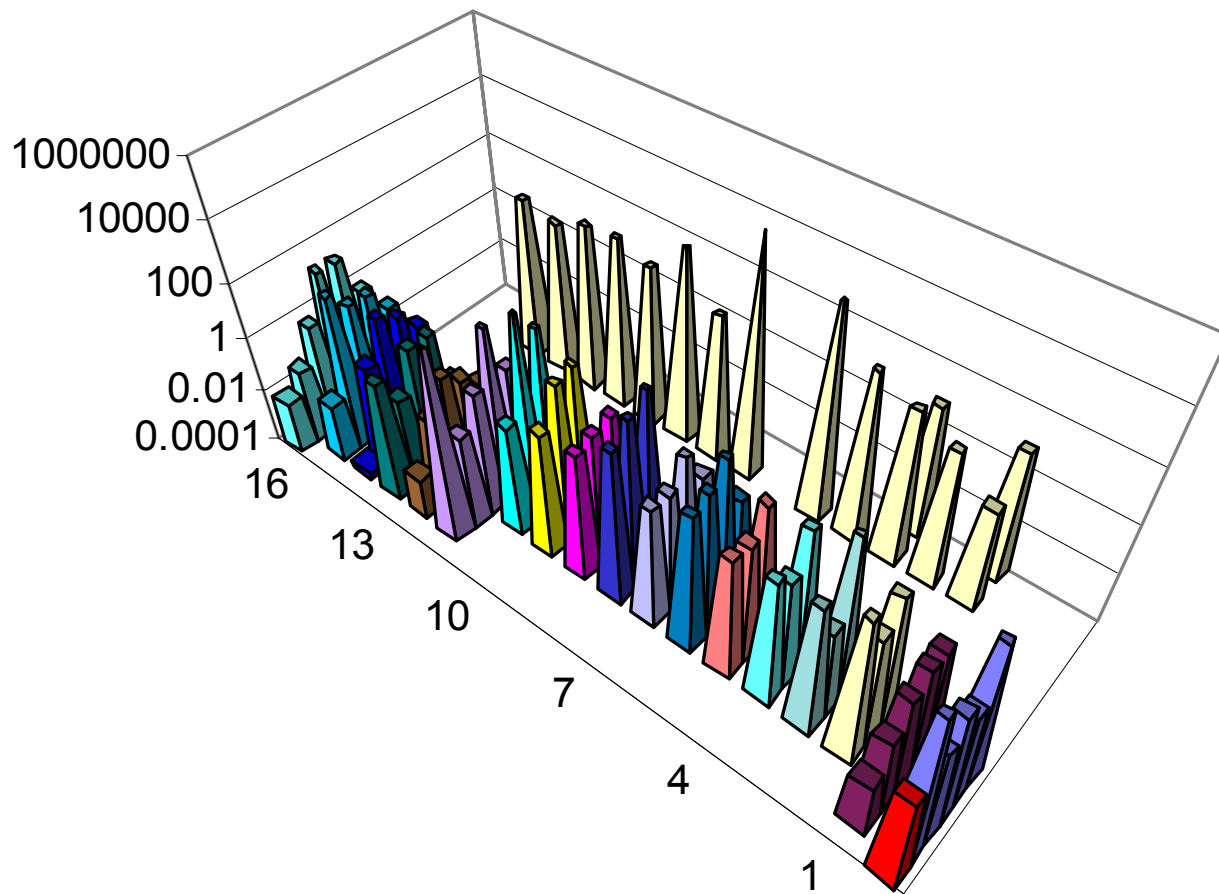
Various neutrons for use



Capture Cross Sections

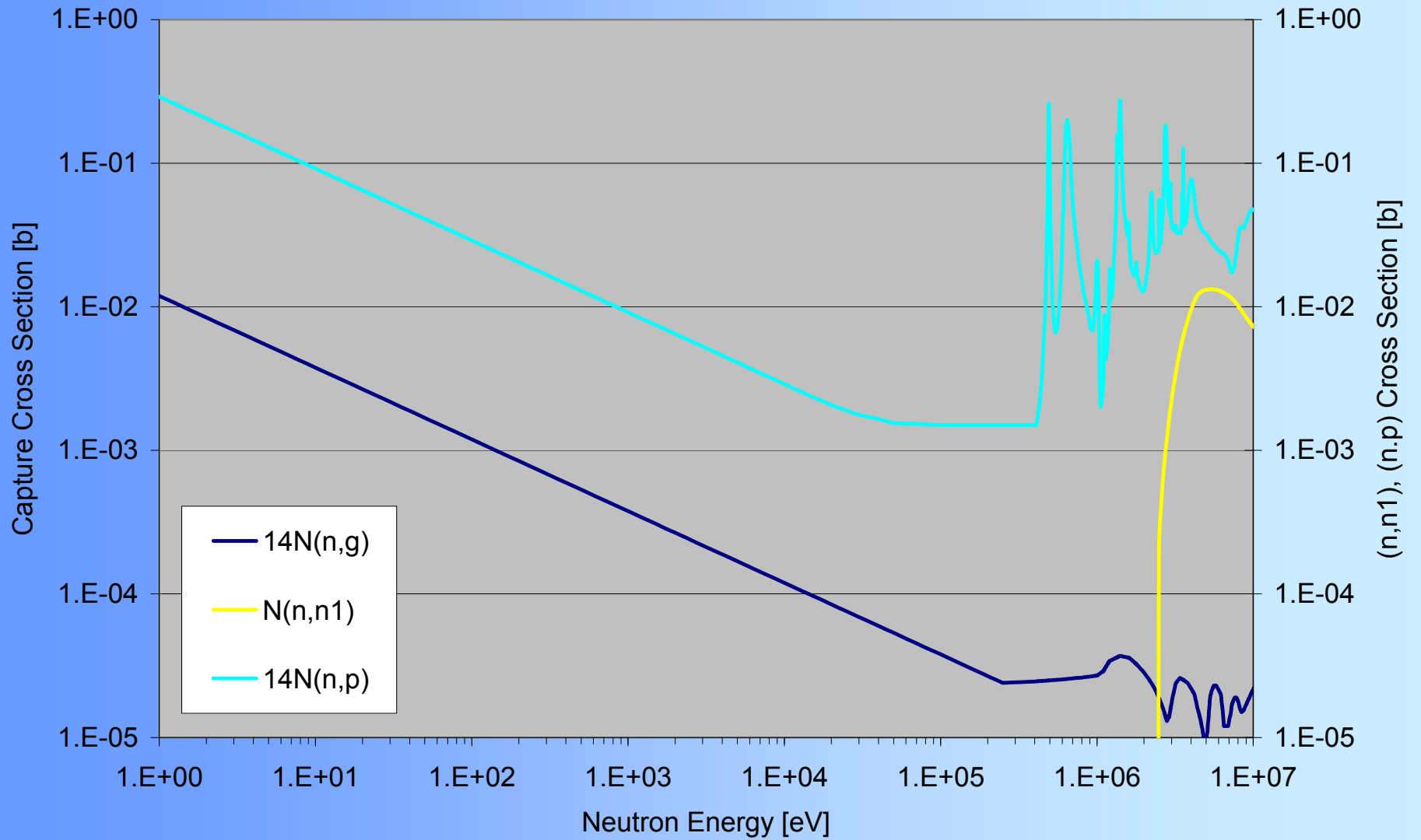


Thermal neutron capture cross sections



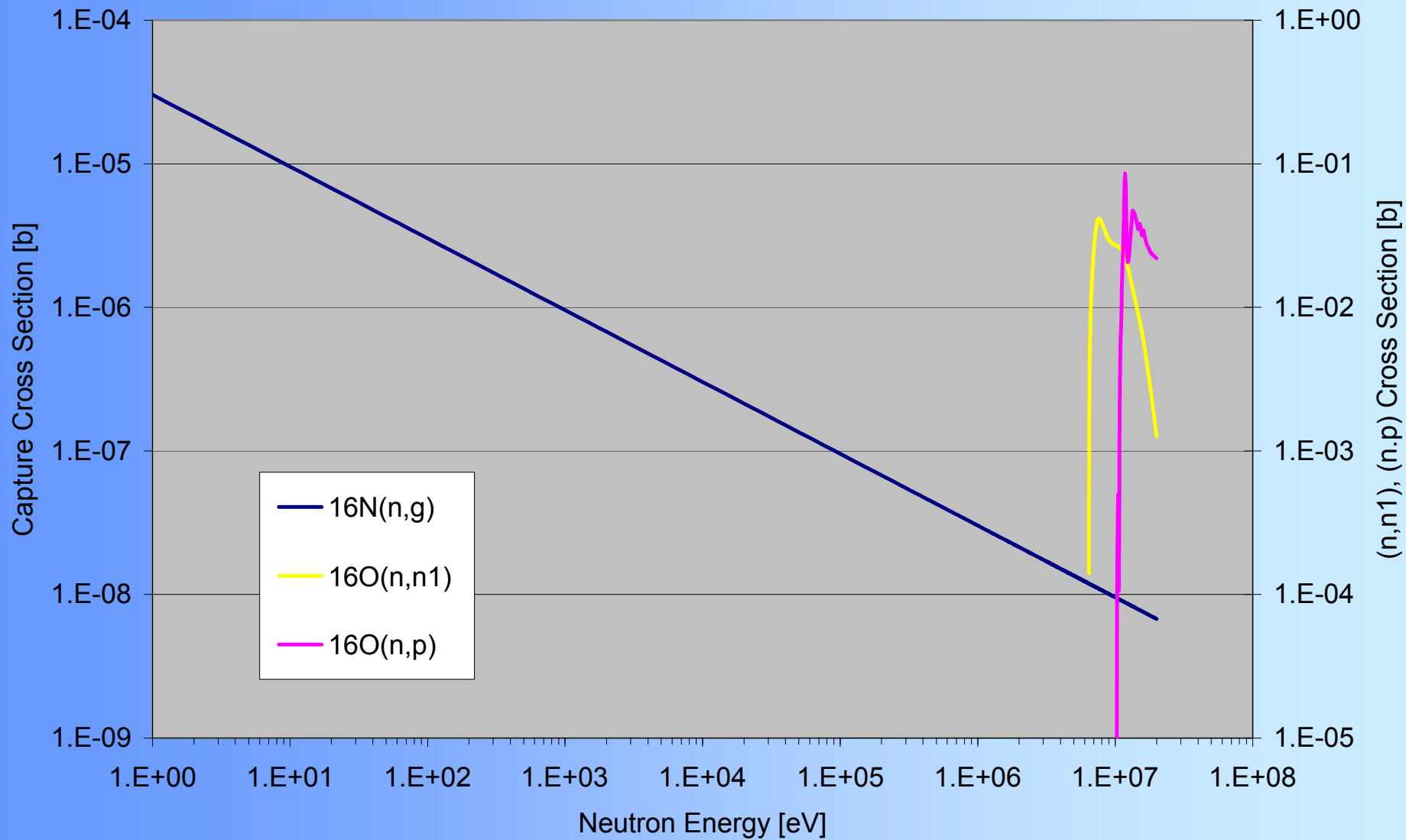
^{14}N Cross Sections

$$\sigma_{\text{th}} = 8 \times 10^{-2} \text{ b}$$



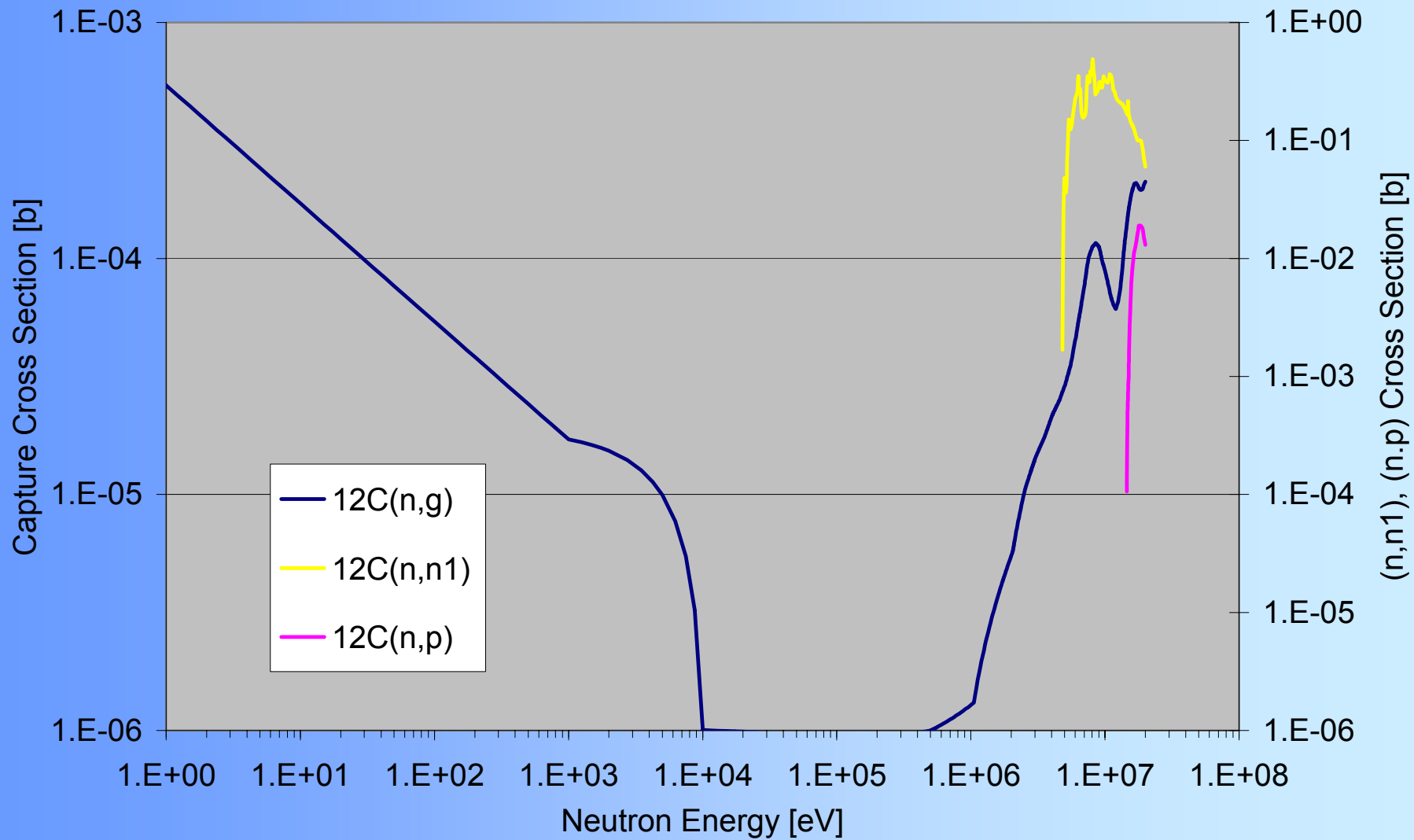
^{16}O Cross Sections

$$\sigma_{\text{th}} = 2 \times 10^{-4} \text{ b}$$

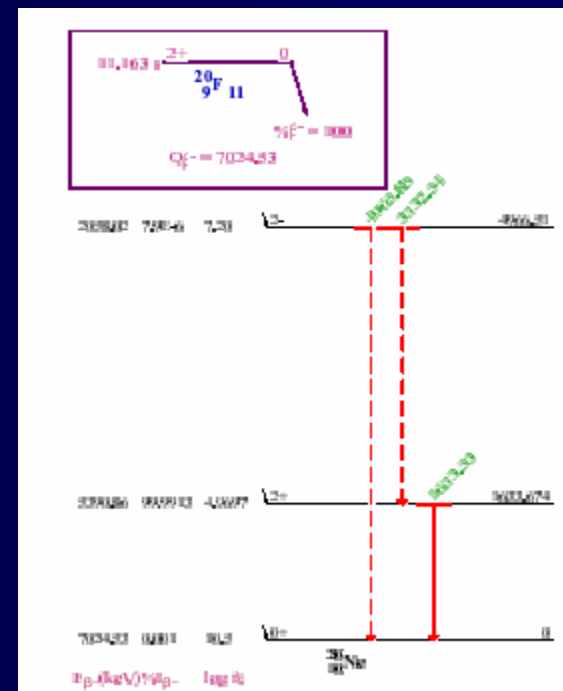
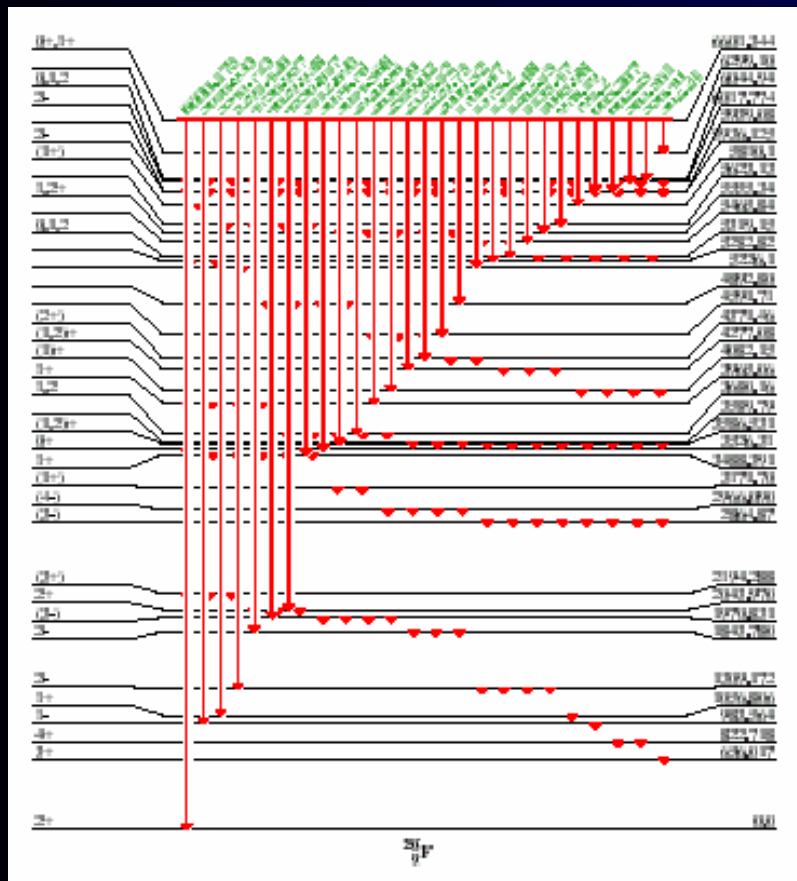


^{12}C Cross Sections

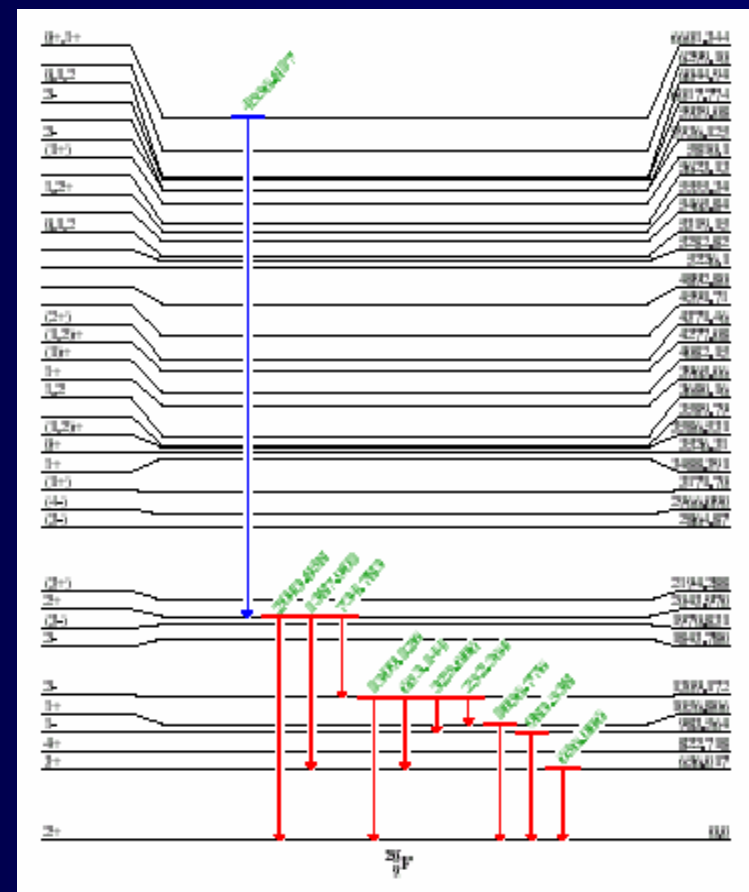
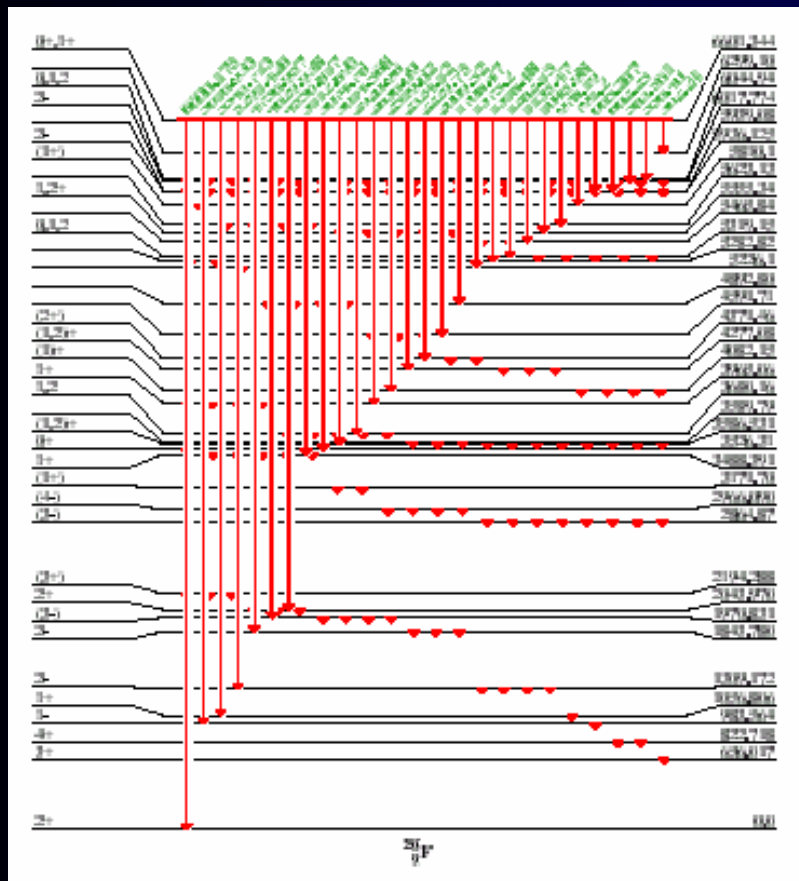
$$\sigma_{\text{th}} = 3.8 \times 10^{-3} \text{ b}$$



Nuclear Structure View of PGAA and NAA



Coincidence in PGAA

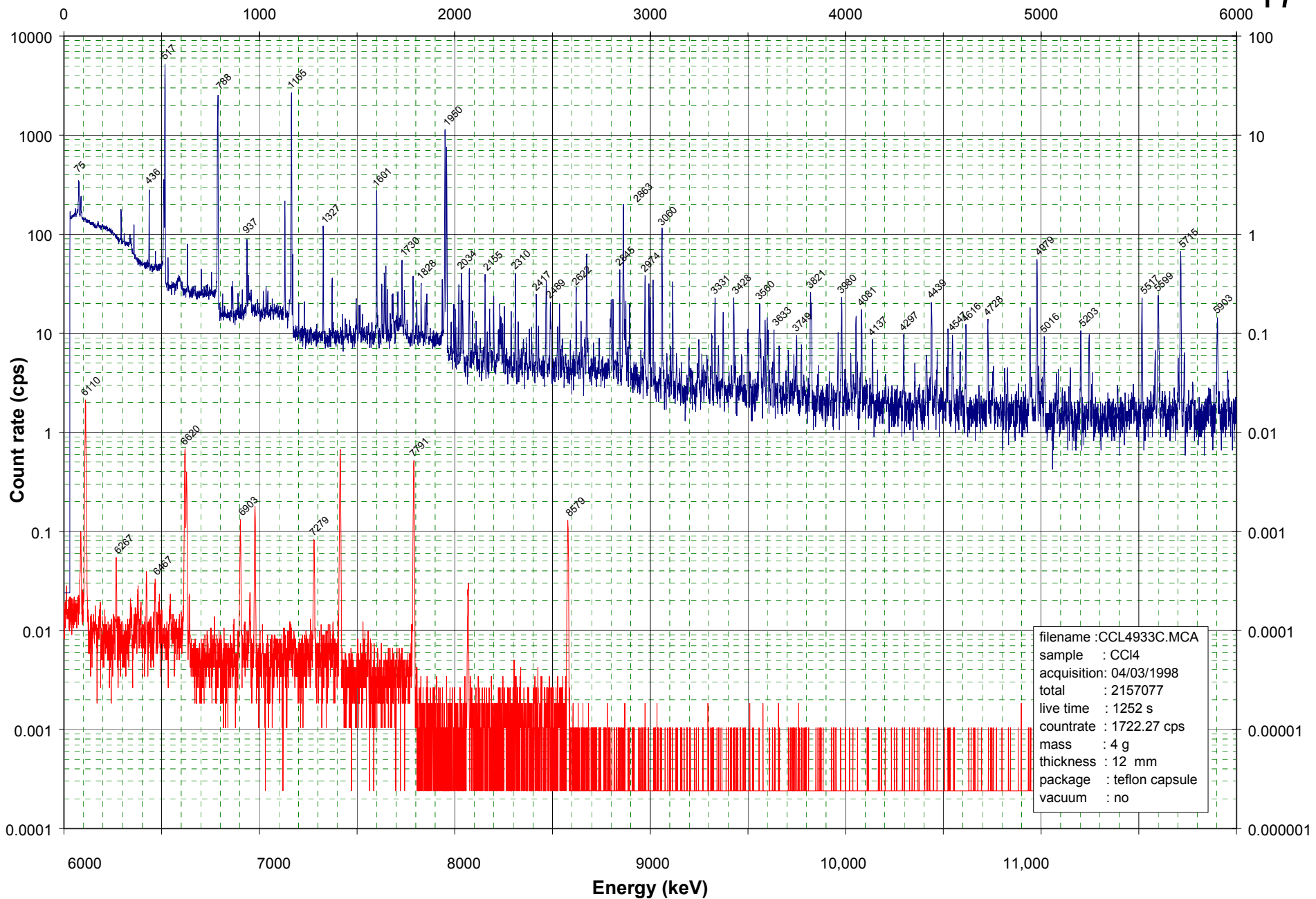


2. Features of PGAA

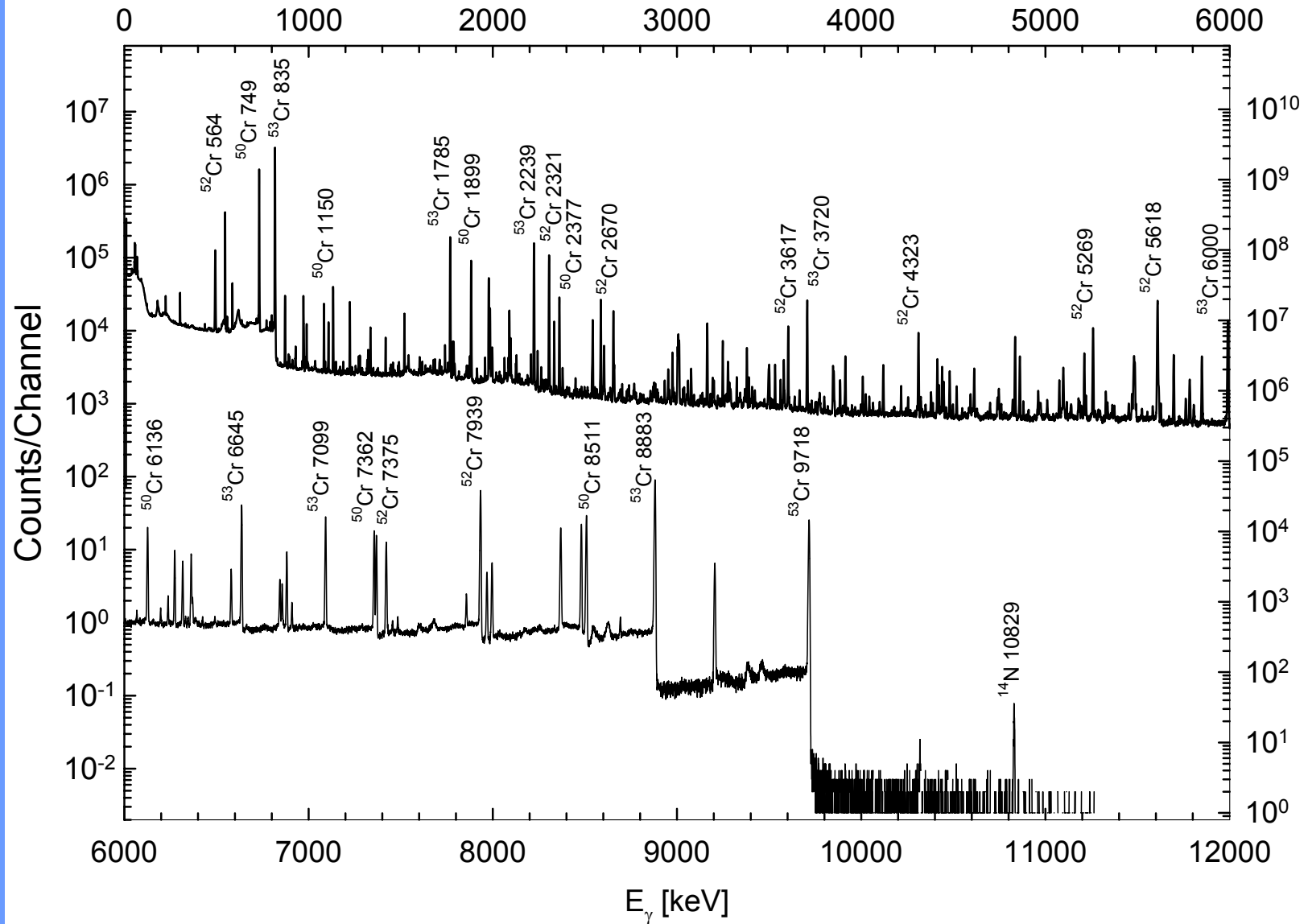
PGAA properties

- Gamma radiation is characteristic
 - energy → elements (isotopes)
 - intensity → quantity
- complicated gamma spectrum
- independent from chemical environment
- nondestructive
- no sample preparation, sample in any form
- extremely different sensitivities

PGAA spectrum of Cl



Cr capture spectrum



Reaction Rate

$$R = \int_{E_{\min}}^{E_{\max}} N_0 \sigma(E) \Phi(E) dE$$

σ - differential cross section (cm²),

Φ - flux cm⁻²·s⁻¹ eV⁻¹

N - number of target nuclides (~ mass)

$$N = m/M \theta N_A$$

For thermal neutrons

$$R_{th} = N_0 \sigma_0 \Phi_{th}$$

PGAA analysis

$$R_{\gamma} = N \sigma_{\gamma} \Phi$$

$$\sigma_{\gamma} = \sigma I_{\gamma}$$

R_{γ} - induced activity for a given gamma energy

N - number of atoms (~mass of component)

σ_{γ} - gamma ray production cross section

σ - capture reaction cross section

I_{γ} - gamma-ray intensity

Φ - neutron flux

DETERMINATION OF CHEMICAL COMPOSITION

$$A_E = m \cdot S \cdot t$$

$$S = \frac{N_A}{M} \cdot \theta \cdot \sigma_0 \cdot I_\gamma \cdot \Phi_0 \cdot \varepsilon(E_\gamma)$$

m : Mass of the element

S : Sensitivity

A_E : Peak area

N_A : Avogadro-number

M : Molar weight

θ : Isotopic abundance

σ_0 : Neutron capture cross-section

I_γ : Gamma-yield

Φ_0 : Neutron flux

$\varepsilon(E_\gamma)$: Detector efficiency

PGAA detection limits

Element																He																																																											
stable isotope																^3He , ^4He																																																											
atomic weight																																																																											
σ - capture																																																																											
σ - scattering																																																																											
Detection Limit [ppm]																																																																											
■ 0.01-1																																																																											
■ 1-10																																																																											
■ 10-100																																																																											
■ 100-1000																																																																											
■ >1000																																																																											
□ no data																																																																											
H 1 1.00794 0.3326 b 82.02 b	Li 6^7Li , 7^7Li , 6^9Li 6.941 7.05 b 1.37 b		Be 9 9.0122 0.0076 b 7.63 b	Na 23 22.98977 0.530 b 3.28 b		Mg 24^{24} , 25^{25} , 26^{26} 24.305 0.063 b 3.71 b		K 39^{39} , 40^{40} , 41^{41} 39.0983 2.1 b 1.96 b		Ca 40 40.078 2.75 b 2.35 b	Sc 45 44.9559 2.35 b	Ti 46^{46} , 47^{47} , 48^{48} 46.48 47.867 6.09 b 4.35 b	V 50^{50} , 51^{51} 50.9415 5.08 b 5.10 b	Cr 50^{52} , 52^{53} , 53^{53} 54 51.9961 3.05 b 3.49 b	Mn 55 54.9380 1.33 b 2.15b	Fe 54^56 , 56^{57} , 57^{58} 55.845 2.56 b 11.62 b	Co 59 58.9332 37.16 b 5.6 b	Ni 58^{60} , 60^{61} , 61^{61} , 62^{62} , 64^{63} 58.6934 4.49 b 1.85 b	Cu 63^{65} , 65^{67} 63.546 68.70 6.38 b	Zn 64^{66} , 66^{67} , 67^{67} 65.39 2.75 b 6.38 b	B 10^{10} , 11^{10} 10.811 767 b 5.24 b	C 12^{12} , 13^{13} 12.011 0.00350 b 5.551 b	N 14^{15} , 15^{17} 14.00674 1.9 b 11.51 b	O 16^{17} , 17^{18} , 18^{18} 15.9994 0.00019 b 4.232 b	F 19 18.998 0.0096 b 4.018 b	Ne 20^{20} , 21^{20} , 22^{22} 20.1797 0.039 b 2.628 b	Al 27 26.9815 0.231 b 1.503 b	Si 28^{28} , 29^{29} , 30^{31} 28.0855 0.171 b 2.167 b	P 31 30.9738 0.172 b 3.312 b	S 32^{32} , 33^{34} , 34^{36} 32.066 0.172 b 3.312 b	Cl 35^{35} , 37^{37} 35.4527 3.5 b 1.68 b	Ar 36^{38} , 40^{39} , 40^{40} 39.948 0.675 b 0.683 b	Kr 78^{80} , 82^{82} , 83^{83} , 84^{84} , 86^{87} 78.96 6.9 b 25 b 7.68 b	Rb 85^{87} , 87^{88} 85.4678 0.38 b 6.8 b	Sr 84^{86} , 86^{87} , 87^{87} , 88^{88} 87.62 1.28 b 6.25 b	Y 89 88.90585 7.70 b	Zr 90^{92} , 91^{91} , 92^{92} 91.224 9.290638 6.46 b	Nb 93 92.90638 1.15 b 6.255 b	Mo 92^{94} , 94^{95} , 95^{96} , 97^{98} , 98^{98} , 99^{99} 95.94 2.48 b 5.71 b	(Tc) (98) 20 b 6.3 b	Ru 96^{98} , 98^{99} , 99^{100} , 100^{101} , 101^{102} , 102^{104} 101.07 2.56 b 6.6 b	Rh 103 102.9055 14.48 b 4.6 b	Pd 102^{104} , 104^{105} , 105^{106} 106.42 6.8 b 4.48 b	Ag 107^{109} , 109^{110} 107.8682 6.33 b 4.99 b	Cd 106^{108} , 110^{111} , 111^{112} , 113^{114} , 114^{114} 112.411 2520 b 6.5 b	In 113^{115} , 115^{116} 114.818 193.8 b 2.62 b	Sn 112^{114} , 114^{115} , 116^{117} , 117^{118} , 118^{119} , 119^{120} , 120^{122} , 122^{124} 118.71 0.626 b 4.892 b	Sb 121^{123} , 123^{124} 121.76 4.91 b 3.90 b	Te 120^{122} , 123^{124} , 124^{125} , 125^{126} , 126^{128} , 128^{130} 127.6 4.7 b 4.32 b	I 127 126.90447 6.15 b 3.81 b	Xe 134^{136} , 136^{137} , 137^{138} , 138^{139} , 139^{141} , 141^{142} 131.29 2.39 b -	Cs 133 132.90545 2.90 b 3.90 b	Ba 130^{132} , 134^{135} , 135^{137} , 137^{138} 137.327 1.1 b 3.38 b	La 138 138.9055 8.97 b 9.66 b	Hf 174^{176} , 177^{179} , 178^{180} , 179^{181} 178.49 10.41 b 1.02 b	Ta 180^{182} , 182^{184} , 183^{184} 180.9497 183.84 18.3 b 4.60 b	W 182^{184} , 184^{186} , 186^{188} 183.84 18.3 b 4.60 b	Re 185^{187} , 187^{189} 186.207 8.97 b 11.5 b	Os 188^{190} , 189^{191} , 190^{192} , 191^{193} 190.23 190.23 16.0 b 14.7 b	Ir 191^{193} , 193^{195} 192.217 4.25 b 14 b	Pt 190^{192} , 192^{194} , 194^{196} 195.08 195.08 10.3 b 11.71 b	Au 197^{199} , 199^{201} 196.96655 98.65 b 7.73 b	Hg 196^{198} , 199^{200} , 200^{201} , 201^{202} , 202^{204} 200.59 372.3 b 2.68 b	Tl 203^{205} , 205^{206} 204.3833 3.43 b 9.89 b	Pb 204^{206} , 206^{208} , 207^{210} , 208^{212} 207.2 0.171 b 11.12 b	Bi 209 208.98038 0.0338 b 9.156 b	(Po) (209)	(At) (210)	(Rn) (222)	(Fr) (223) -	(Ra) (226) 12.8 b 13 b	(Ac) (227) -	104	105	106

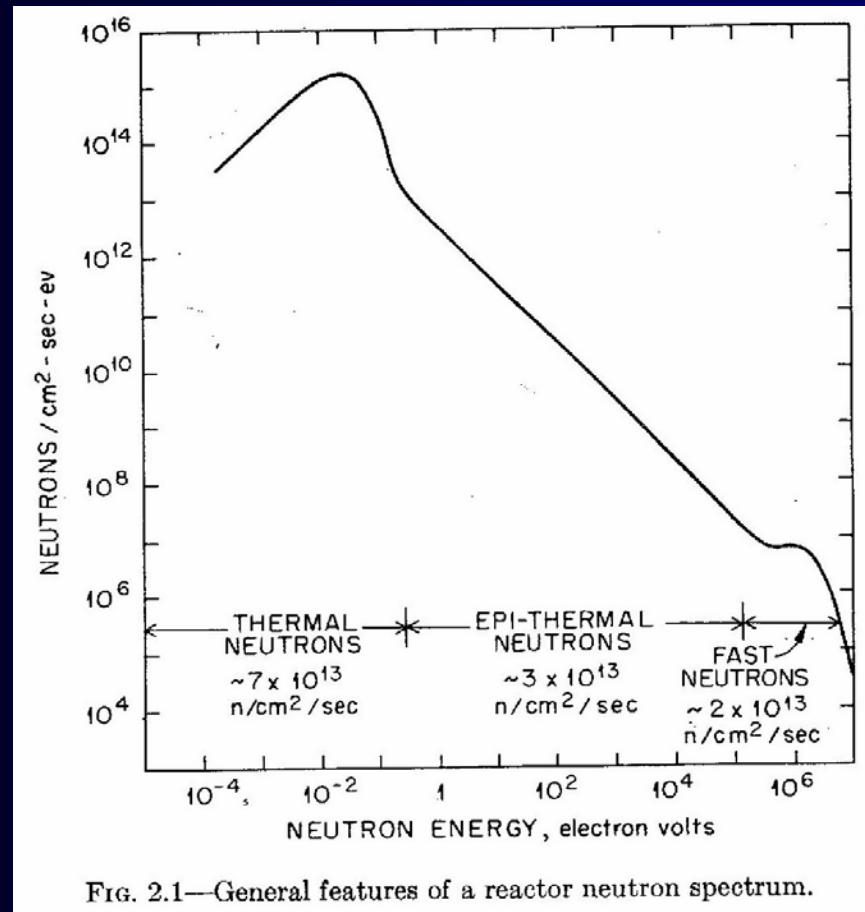
Ce 136^{138} , 140^{140} 142.1 140.115 0.63 b 2.94b	Pr 141 140.90765 11.5 b 2.66 b	Nd 142^{143} , 143^{144} , 144^{145} , 146^{148} , 150^{150} 144.24 51 b 16.6b	(Pm) (145) 168.4 b 21.3 b	Sm 144^{147} , 147^{148} , 149^{150} , 150^{152} , 152^{154} 150.36 5922 b 39 b	Eu 151^{153} , 153^{154} 151.965 4530 b 9.2 b	Gd 152^{154} , 155^{156} , 156^{157} , 157^{158} , 158^{160} 157.25 49700 b 180 b	Tb 159 158.92534 2.34 b 6.84 b	Dy 158^{160} , 160^{161} , 161^{163} , 162^{164} 162.5 994 b 90.3 b	Ho 165 164.93032 6.47 b 8.42 b	Er 162^{164} , 166^{167} , 166^{168} 170.24 167.26 8.7 b	Tm 169 168.93421 100 b 6.38 b	Yb 168^{170} , 171^{172} , 172^{173} , 173^{174} , 174^{176} 173.04 3.48 b 2.34 b	Lu 175^{177} 174.976 7.4 b 7.2 b
Th 232 23.203805 7.37 b 13.36 b	(Pa) (231) 200.6 b 10.5 b	U 235^{237} , 238^{238} 238.0289 7.57 b 8.9 b	(Np) (239) 175.9 b 14.5 b	(Pu) (244) 1017.3 b 7.7 b	(Am) (243)	(Cm) (247)	(Bk) (247)	(Cf) (251)	(Es) (252)	(Fm) (257)	(Md) (258)	(No) (259)	(Lr) (261)

3. PGAA methodology

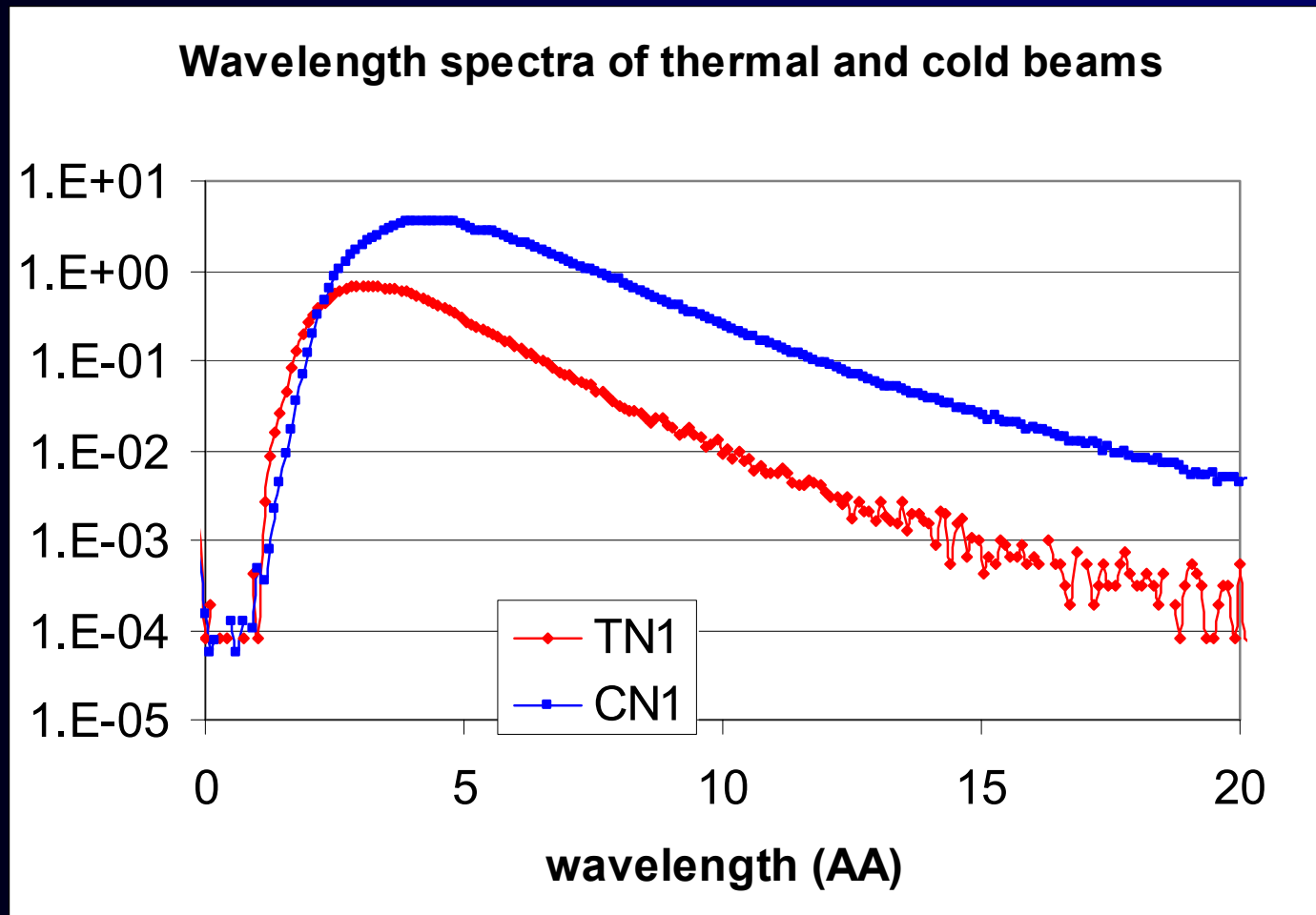
Neutron Sources

- Reactor
 - Thermal 25 meV
 - Cold 5 meV
- Isotopic sources
 - Fission (^{252}Cf) 1-5 MeV
 - Alpha (Pu-Be) 4-8 MeV
- Generators
 - d-D 3 MeV
 - d-T 14 MeV
- Photoneutron sources

Reactor neutron spectrum

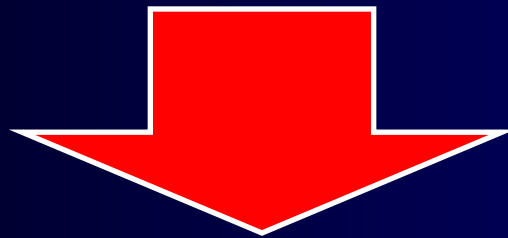


Cold and thermal neutron spectra



Advantages of cold neutron beams

- Higher flux
- every nuclide behaves regularly
 - (follows the $1/v$ -law)
- every nuclide has higher cross section

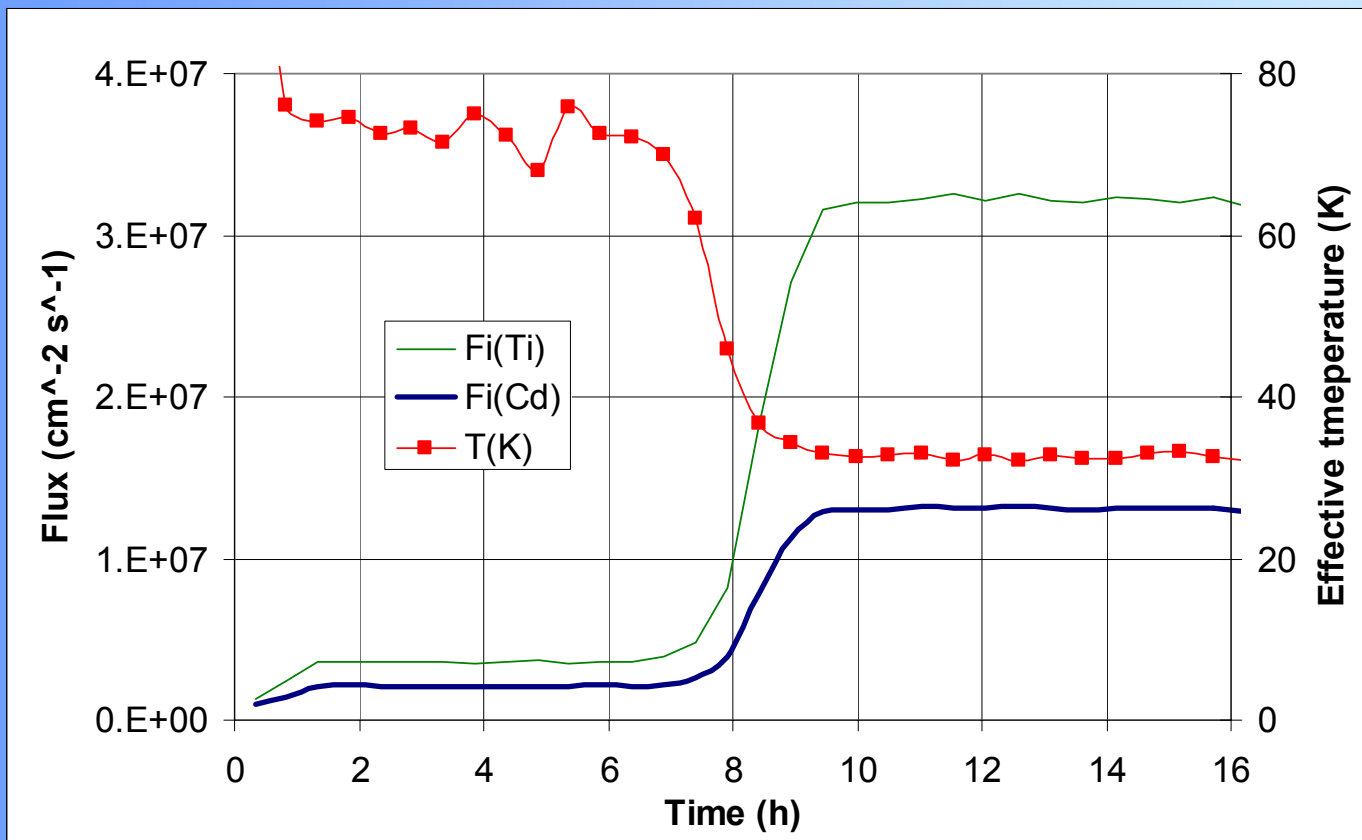


higher reaction rate

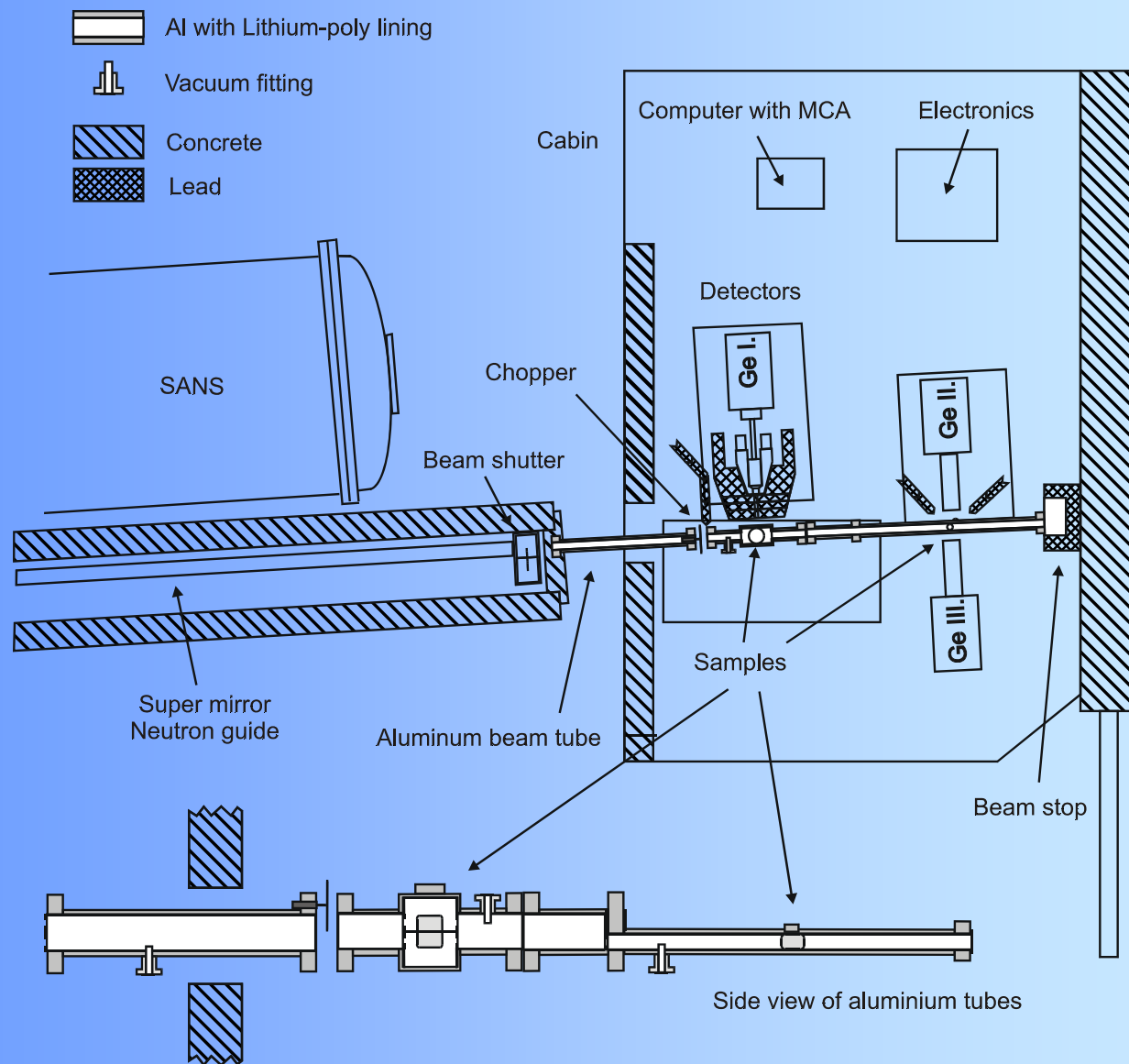
Neutron guides at Budapest



Starting of the cold source at Budapest Research Reactor



The PGAA facility at Budapest



PGAA system





NIPS (Neutron Induced Prompt Gamma Spectroscopy) system

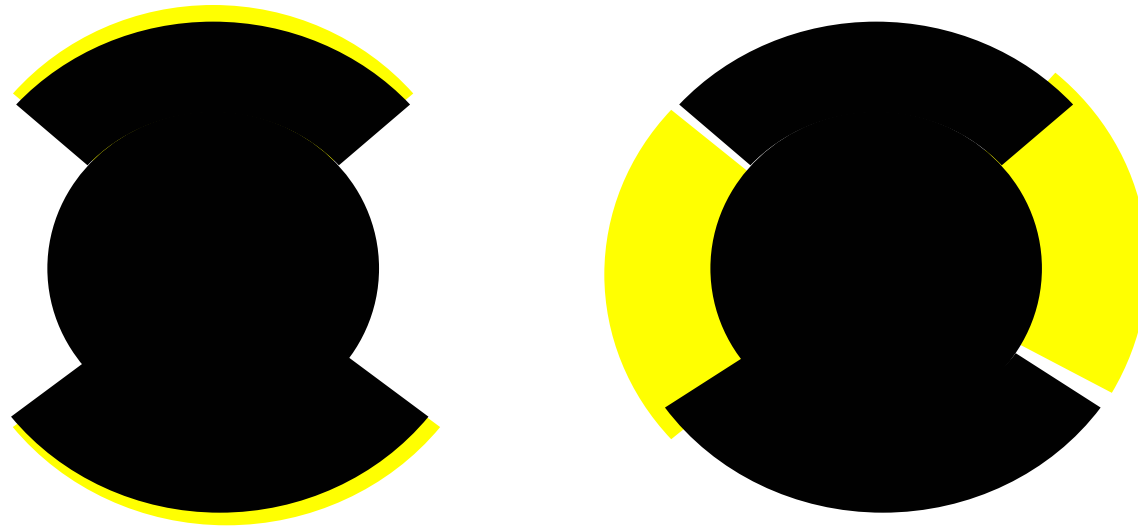


PGAA sample



BEAM CHOPPER

Li-6 and Gd-coated rotating disks

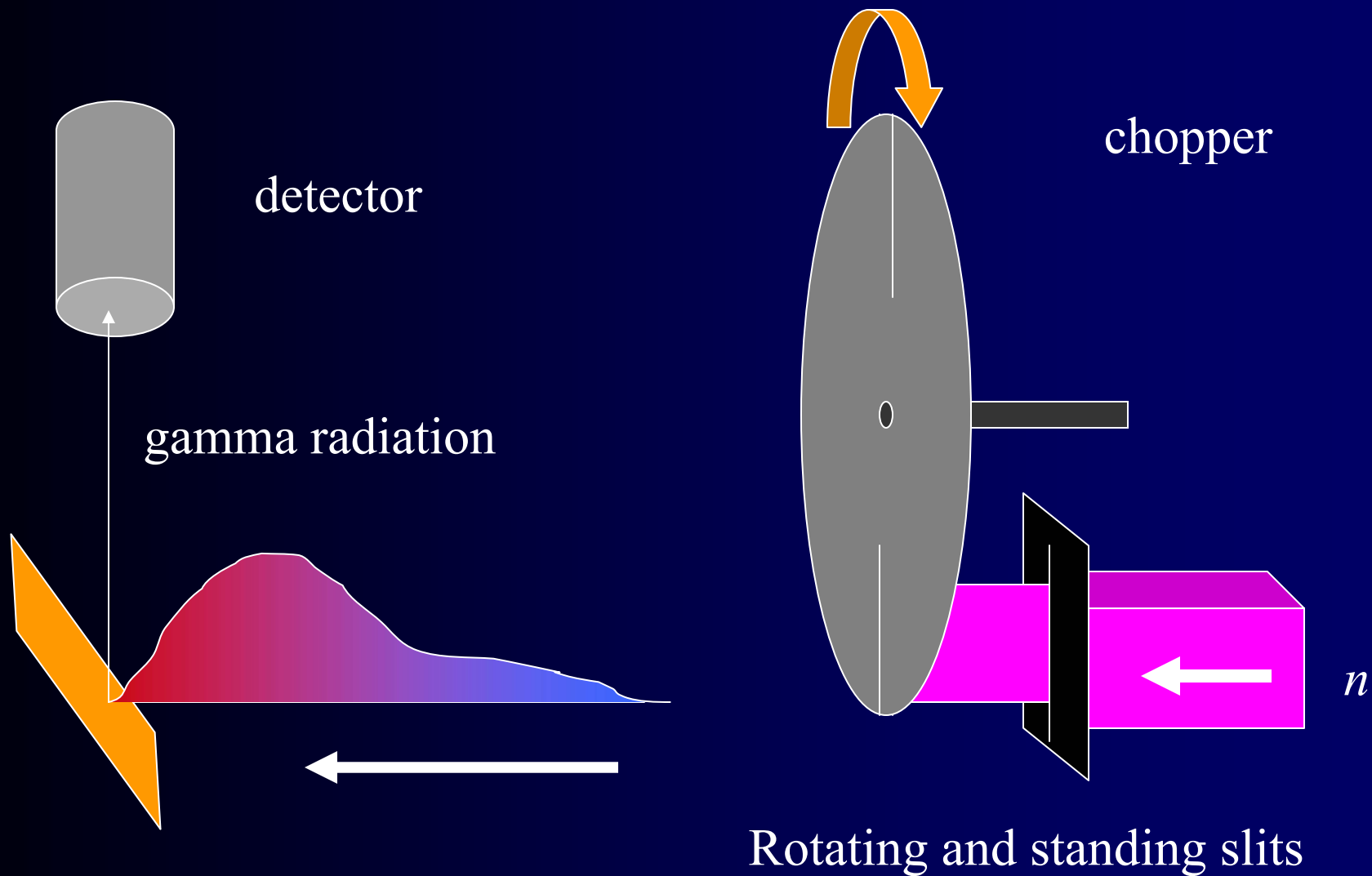


Gap width: 0.2 – 50%

- Variable frequency : 3 – 100 Hz

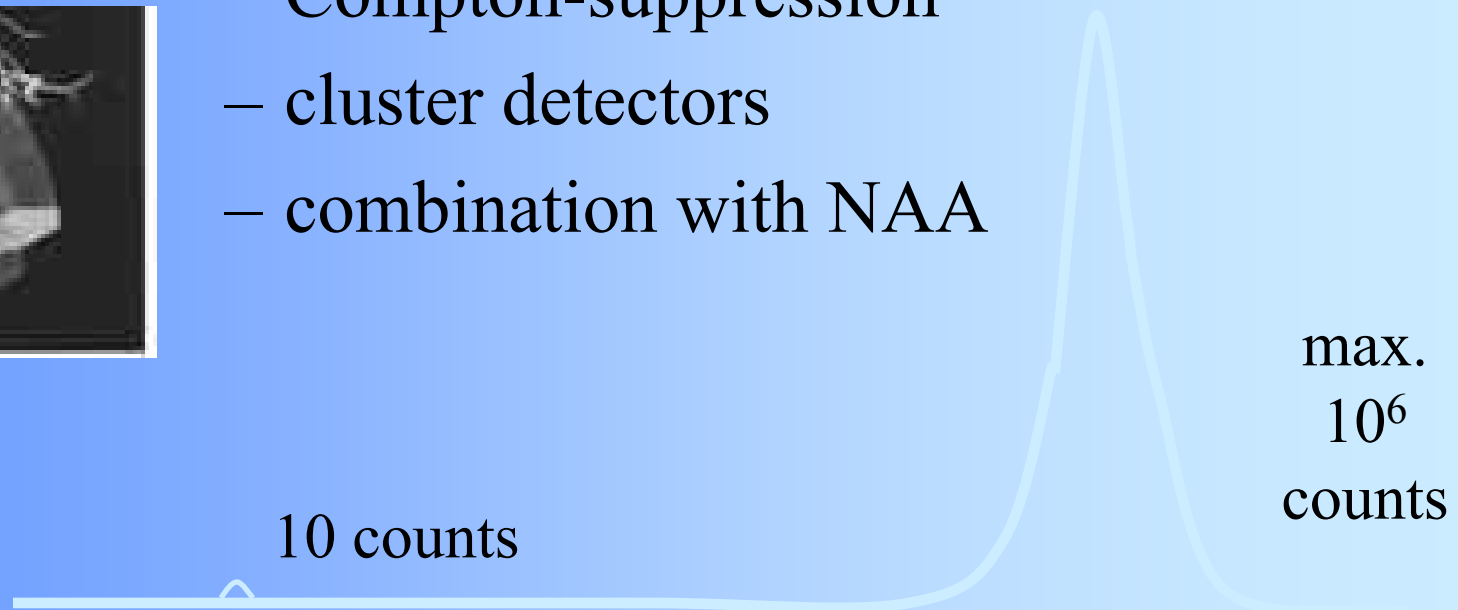
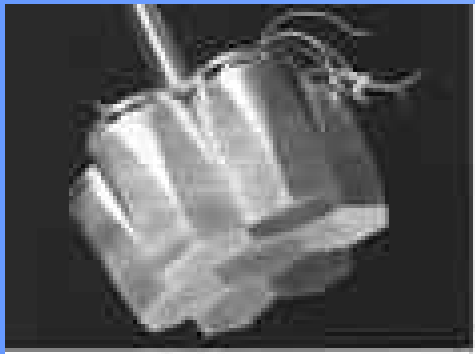
Possible cyclic activation of short-lived isotopes
(Na, F, Sc, Ge, Pd, Ag, In, Er, Hf, W, etc.)

Time of flight



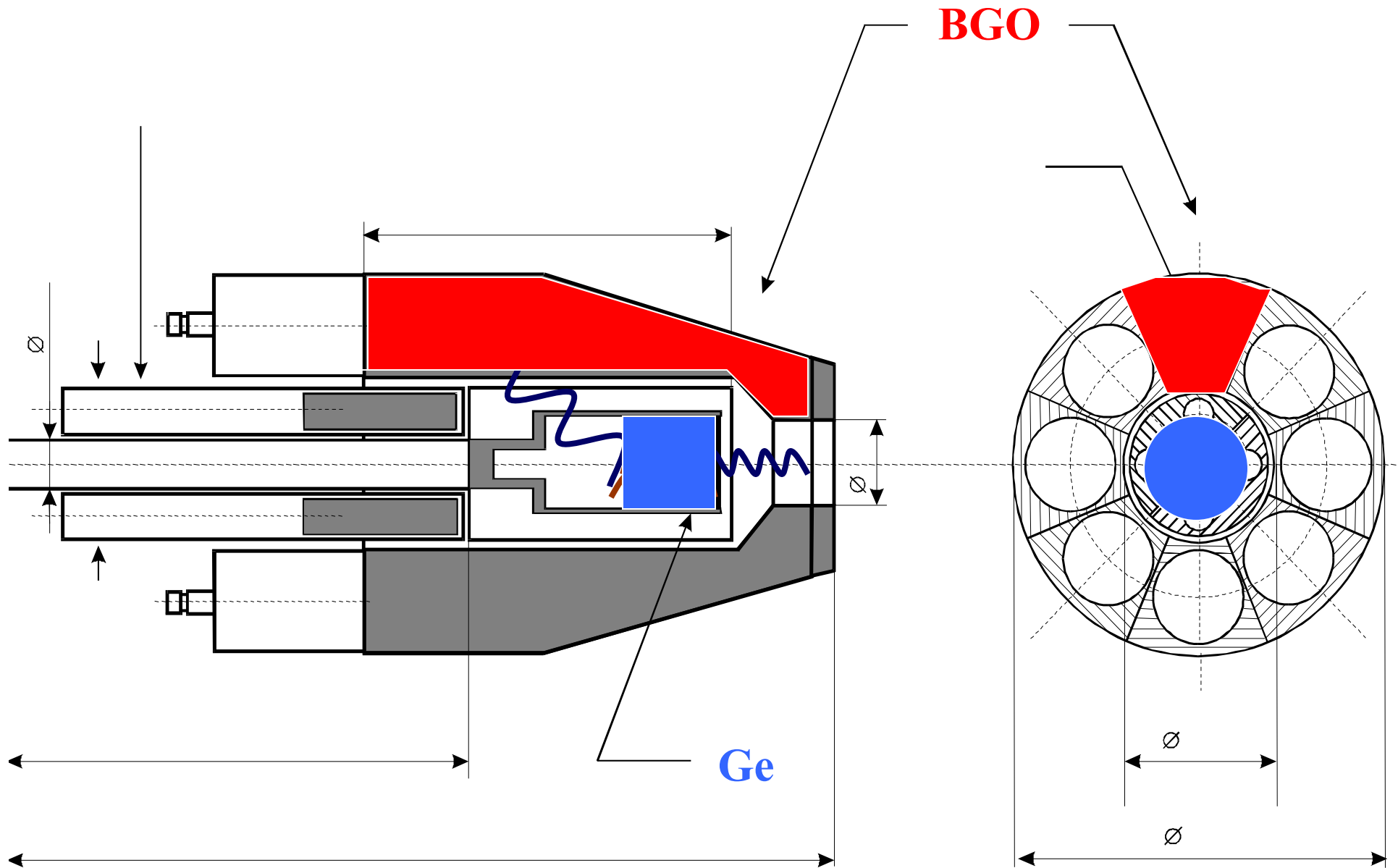
Gamma-ray spectroscopy

- Decreasing the background
 - Compton-suppression
 - cluster detectors
 - combination with NAA

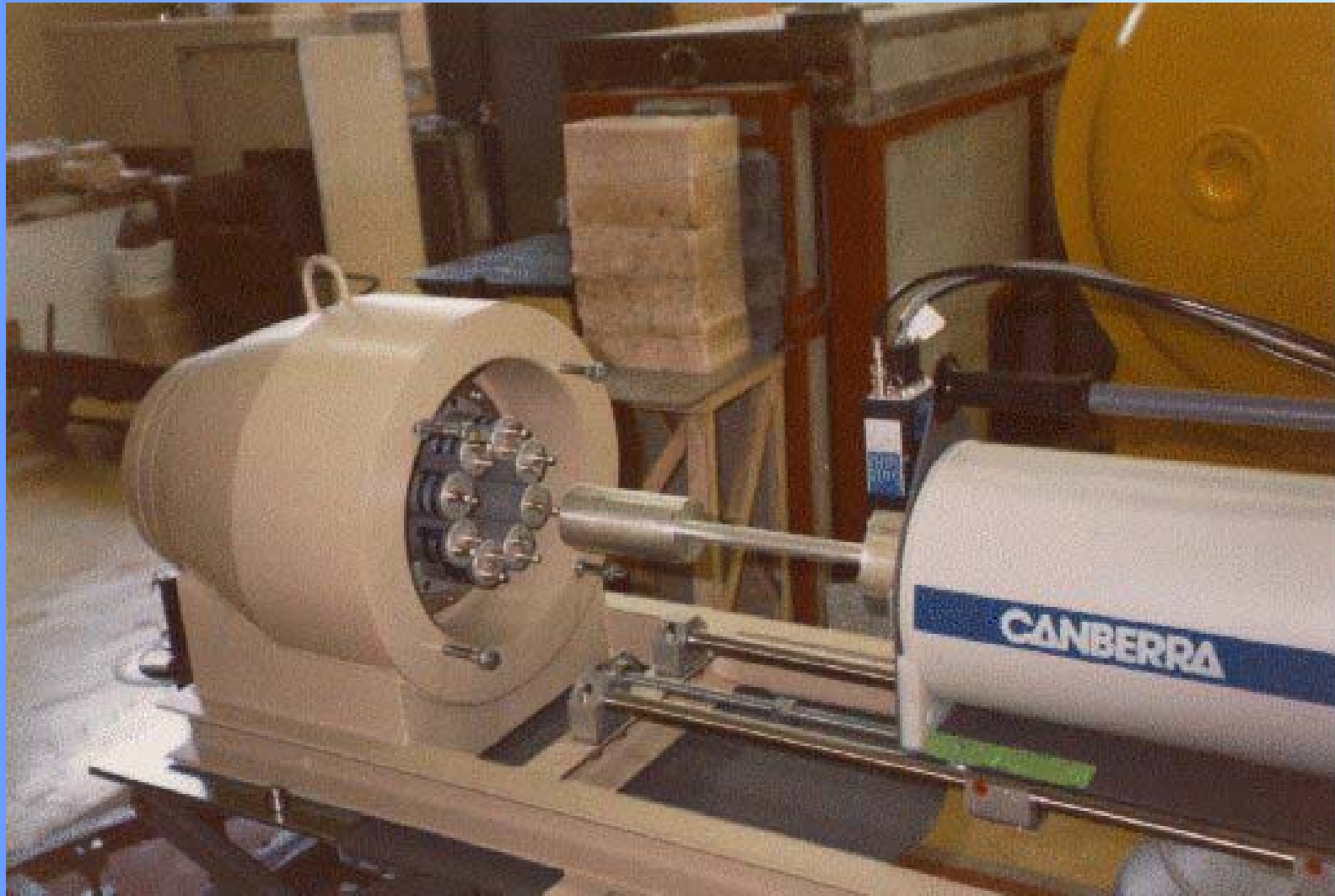


$$\mu_A/\mu_B = A_A/A_B = 10^{-5}$$

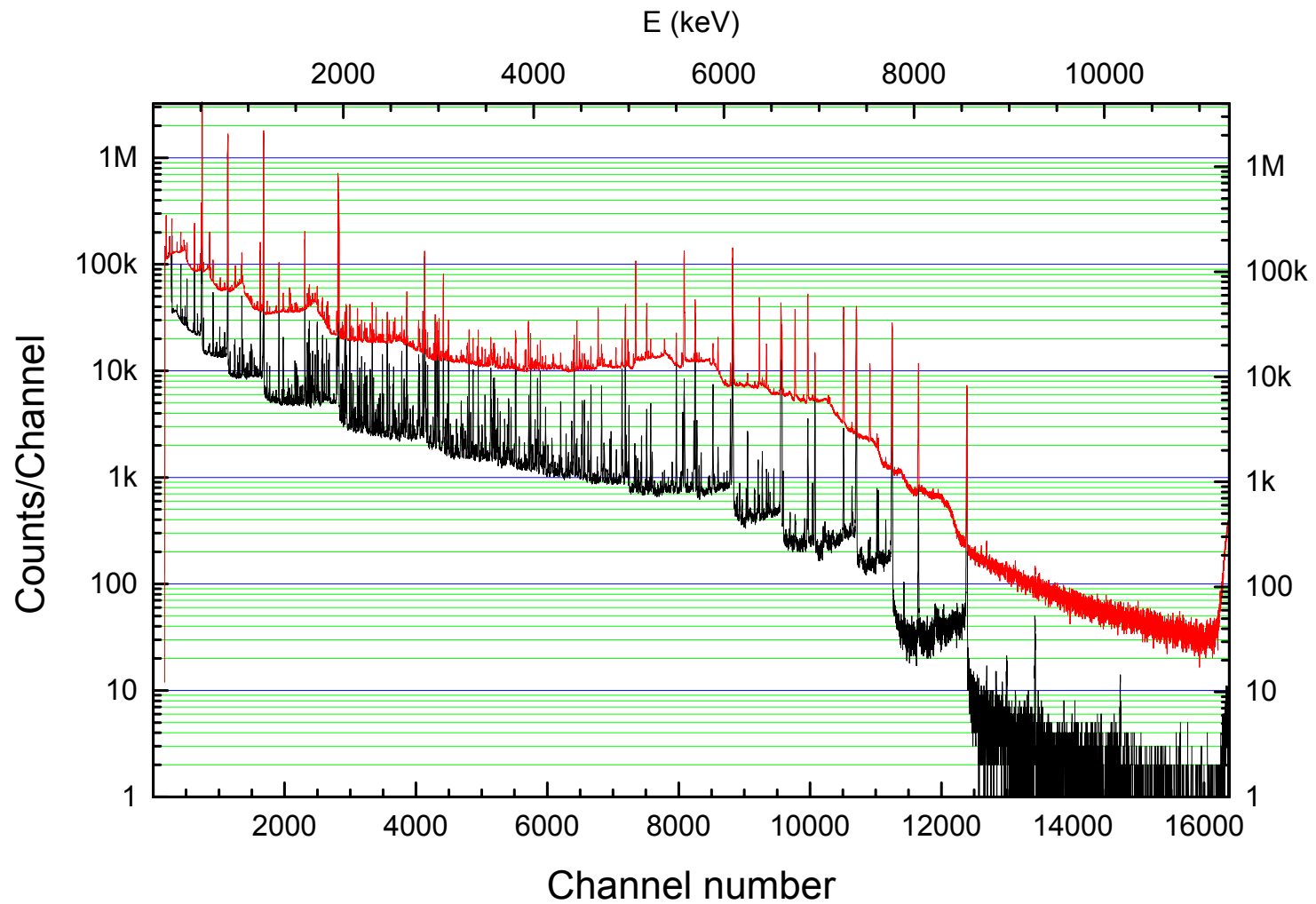
Compton-suppression system



Compton-suppressed detector



Background reduction via Compton-suppression



Composite detectors

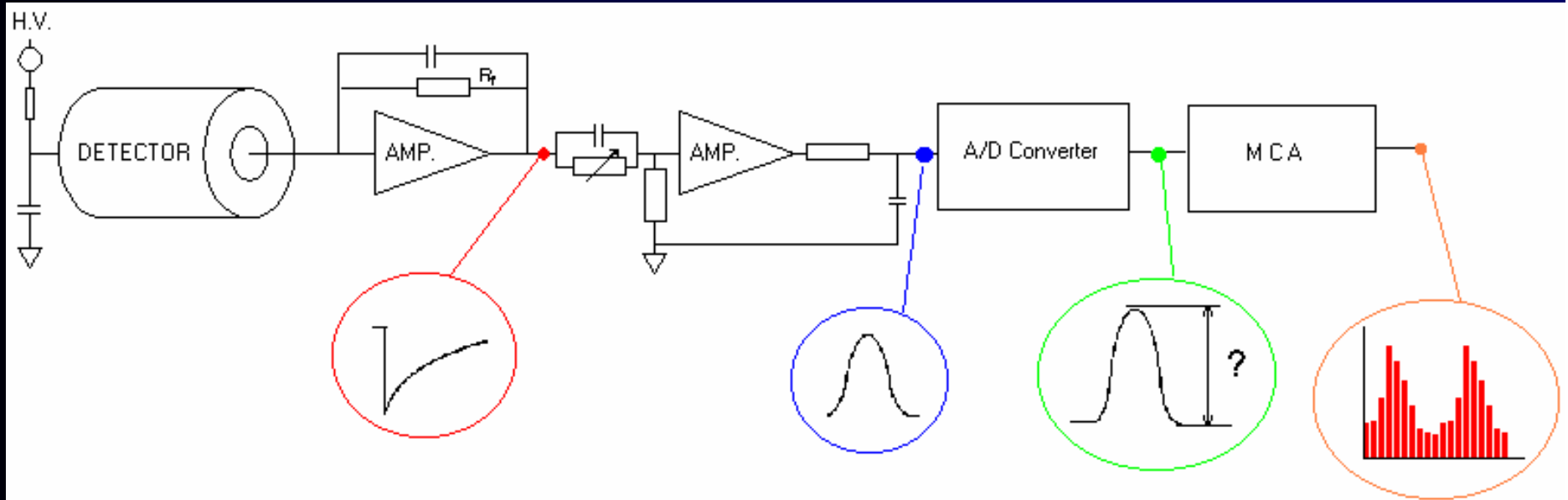
Clover



Cluster



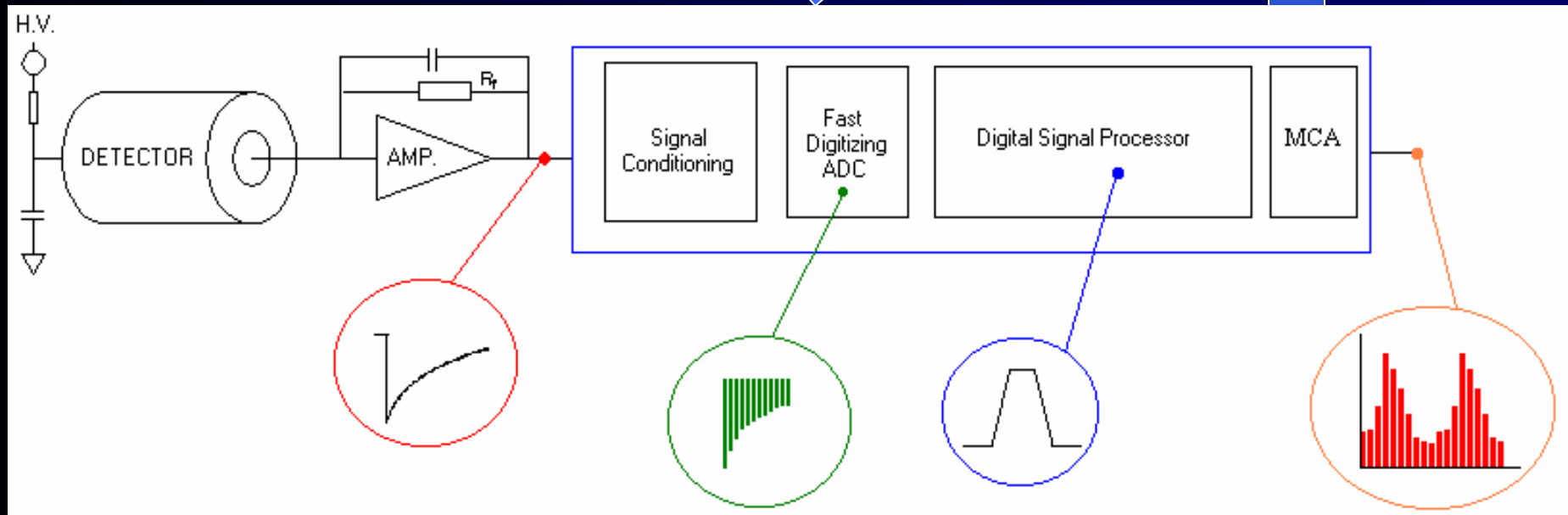
Analog signal processing chain



Preamplifier → Spectroscopy Amplifier → ADC → MCA

Digital signal processing

Digitization



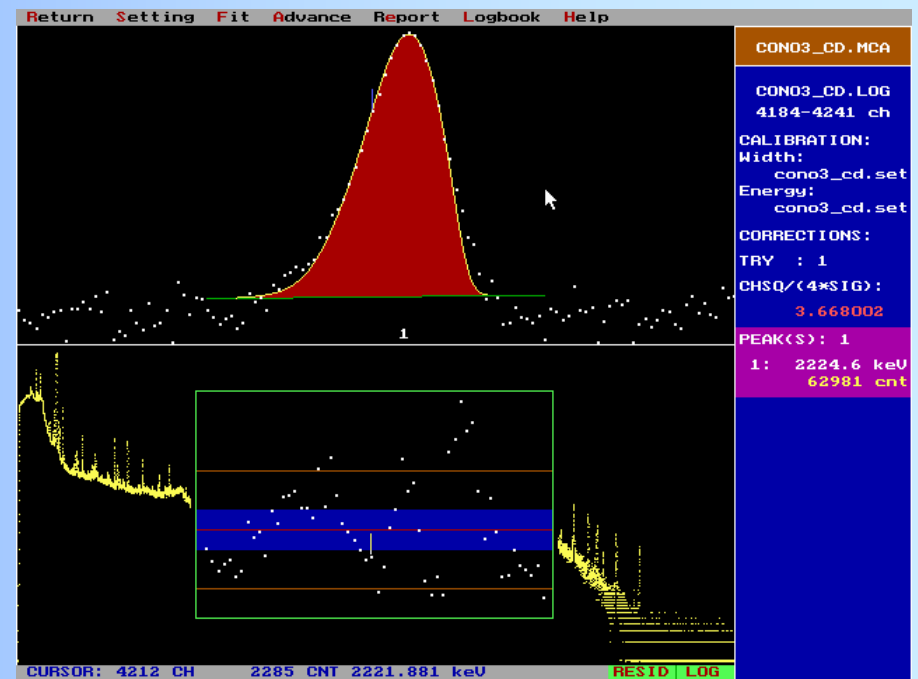
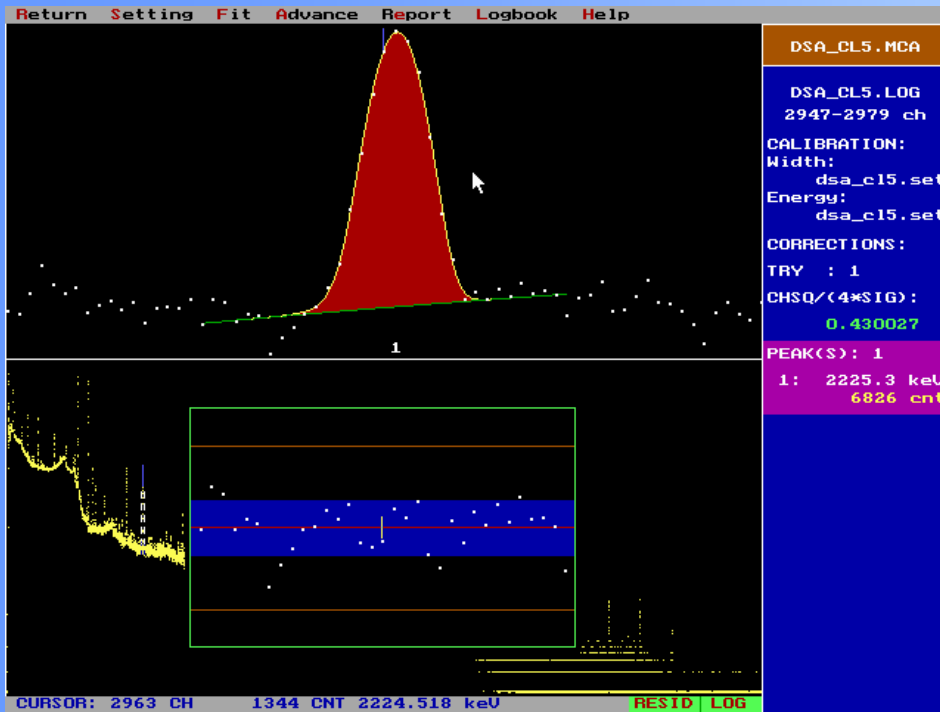
Preamplifier → Sampling ADC → DSP → MCA

Parameters of a spectrometer

- **Peak shape**
- **Resolution vs. energy and amplifier gain**
- **Time stability**
- **Detector efficiency**
- **Energy nonlinearity**

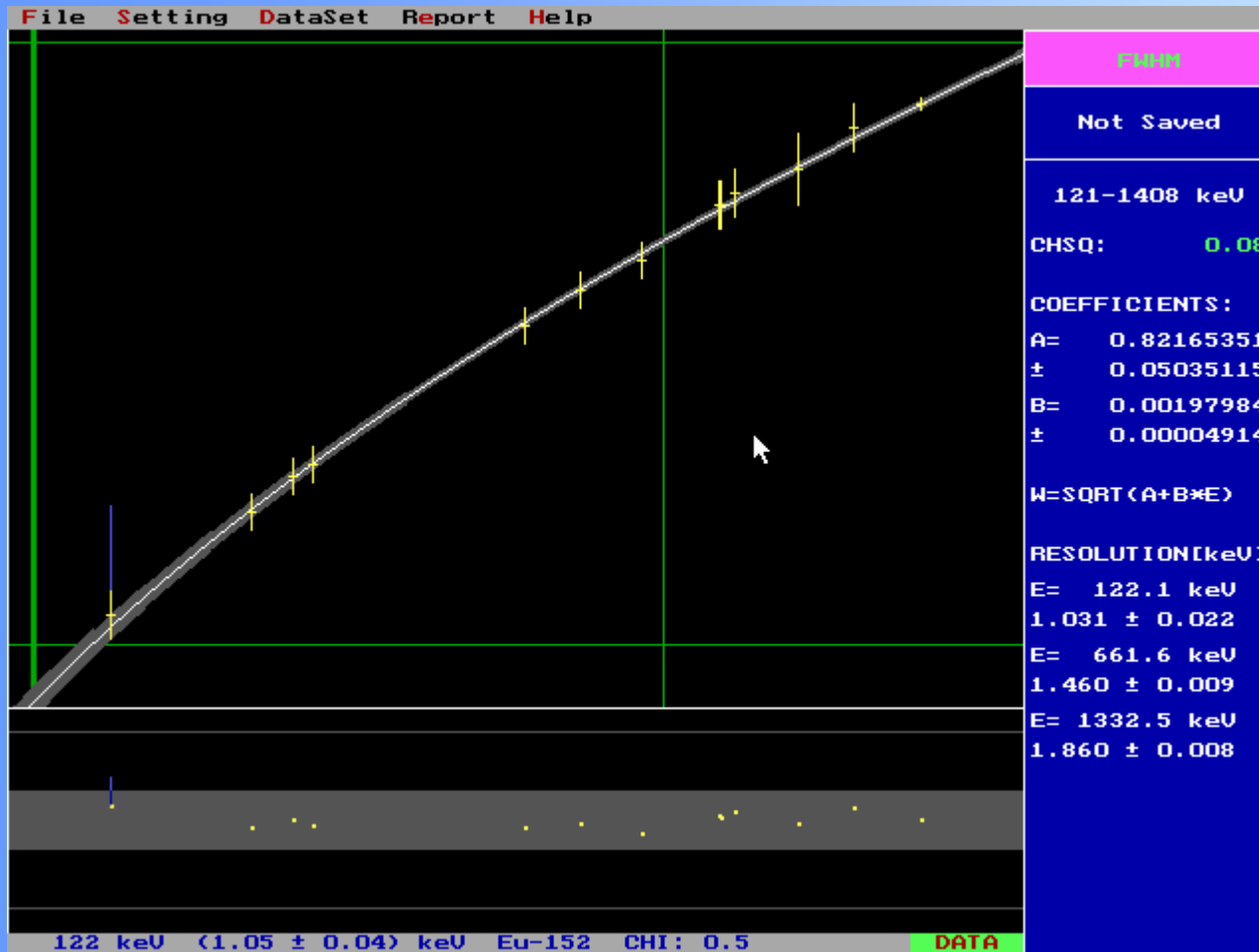
Peak shape

Optimum peak shape



Distorted peak shape

FWHM analysis



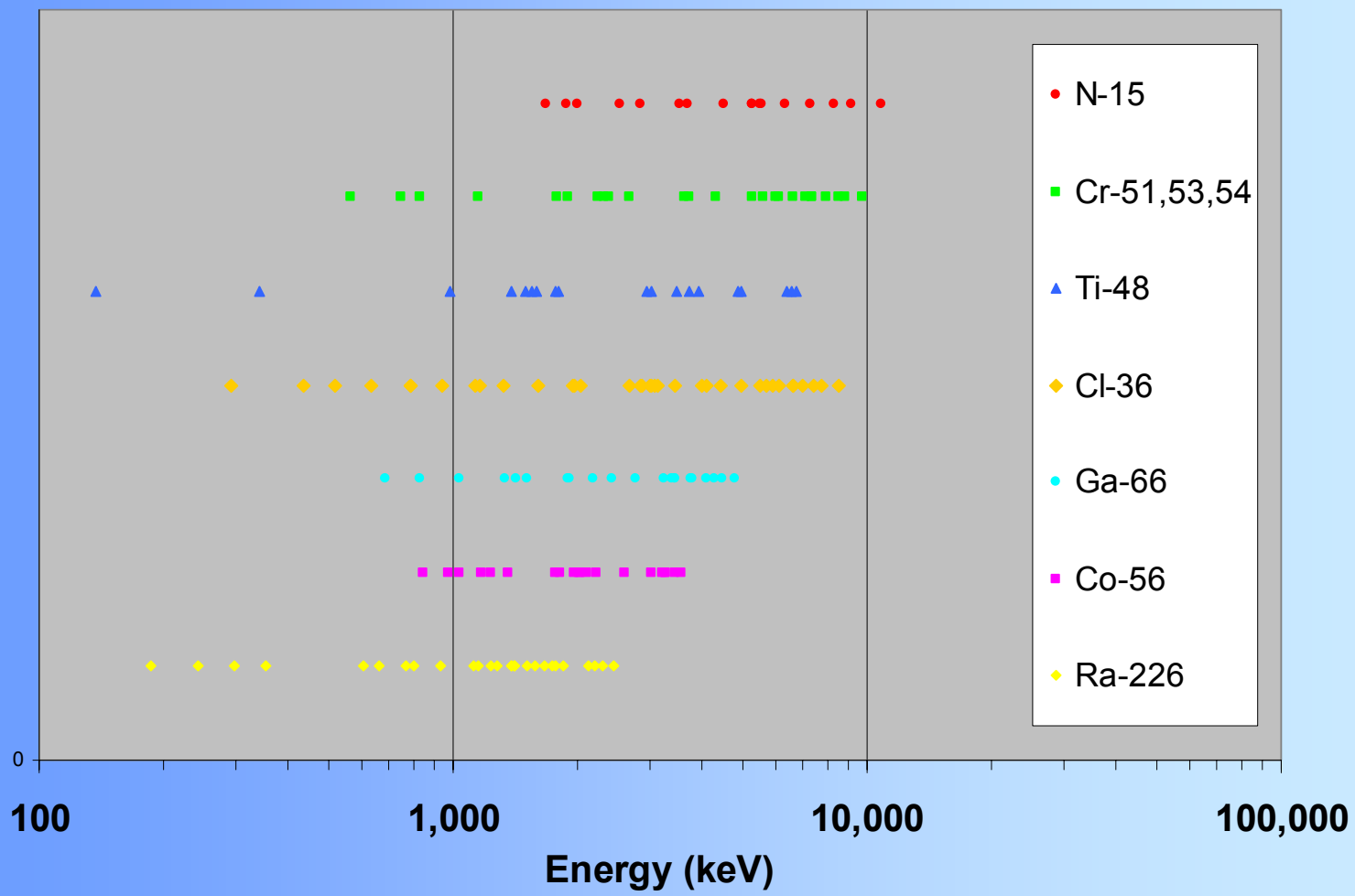
$$FWHM = \sqrt{a^2 + b^2 E}$$

if $E \rightarrow 0$:

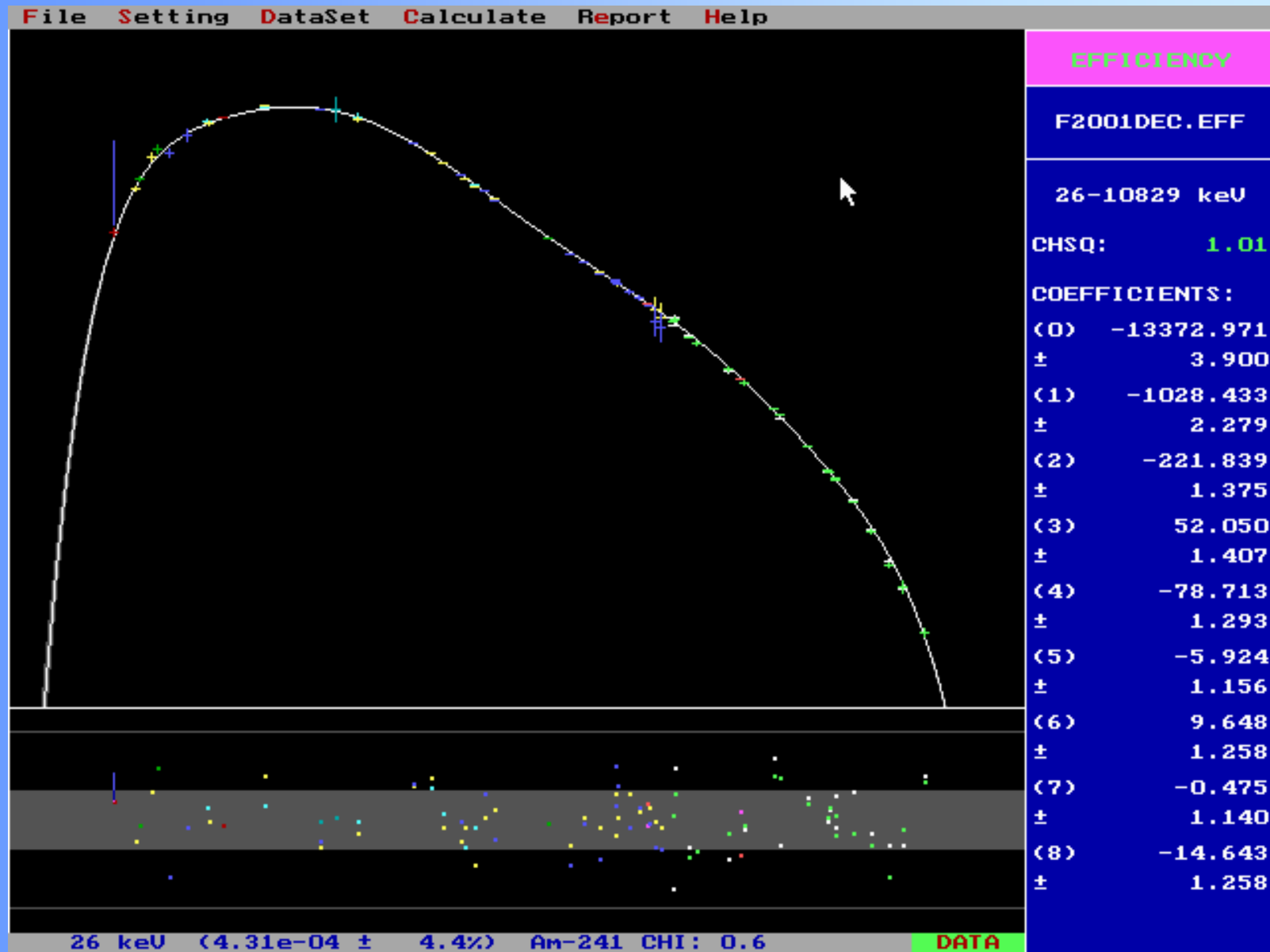
$$FWHM \rightarrow a$$

a : peak broadening
effect of electronics
in keV

Range of calibration sources

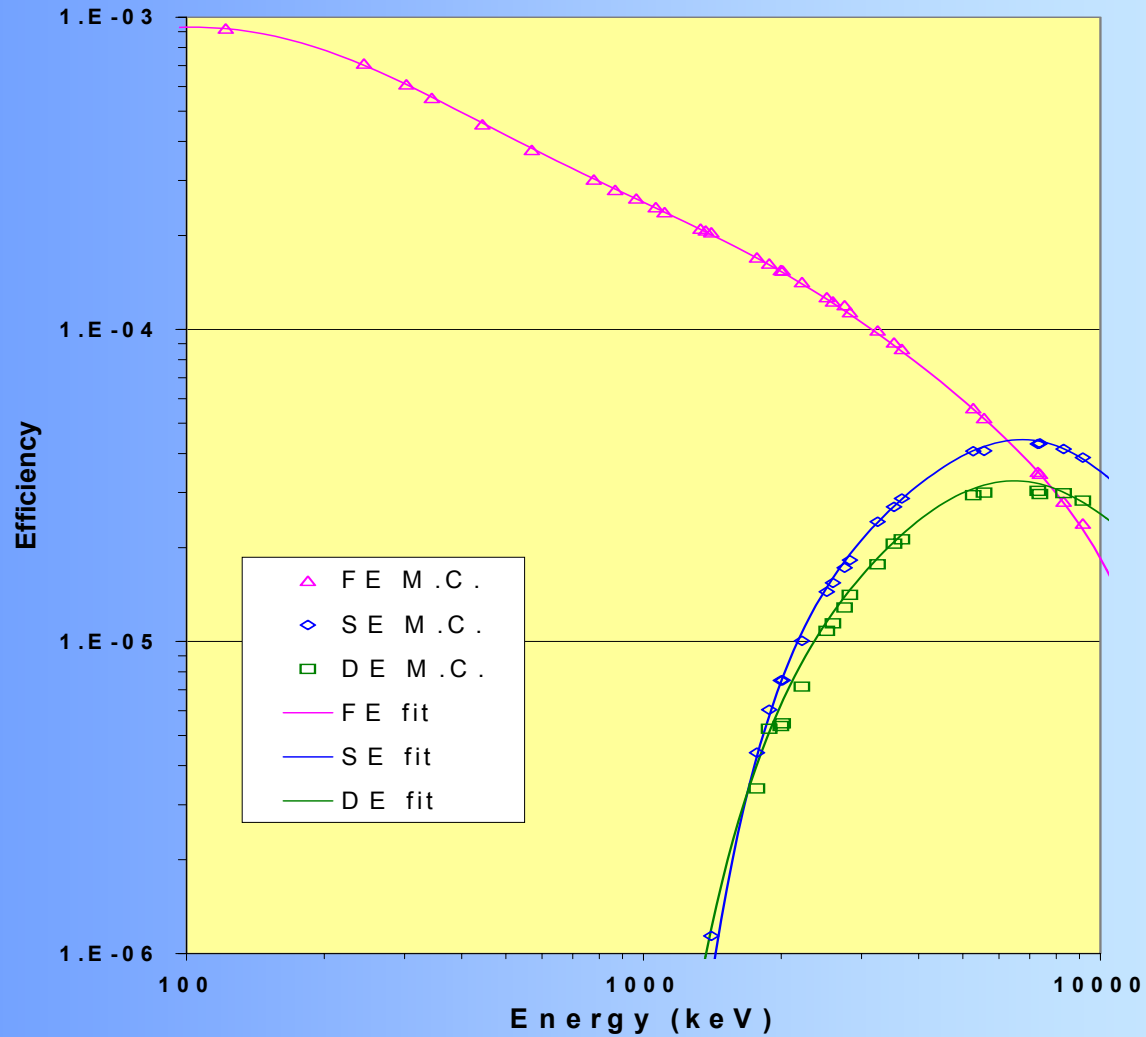


Efficiency calibration

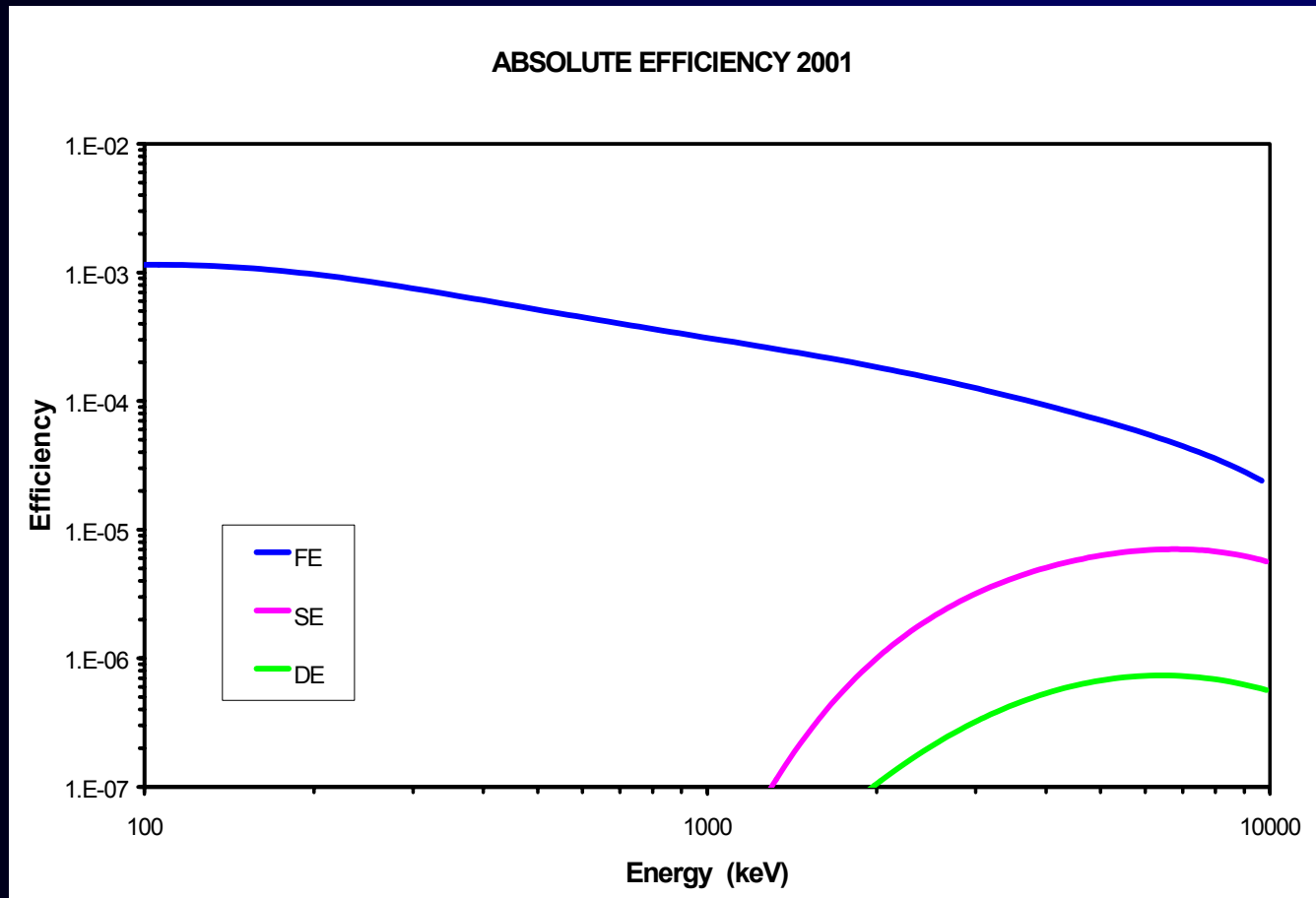


Monte Carlo simulation with CYLTRAN vs. Budapest efficiency

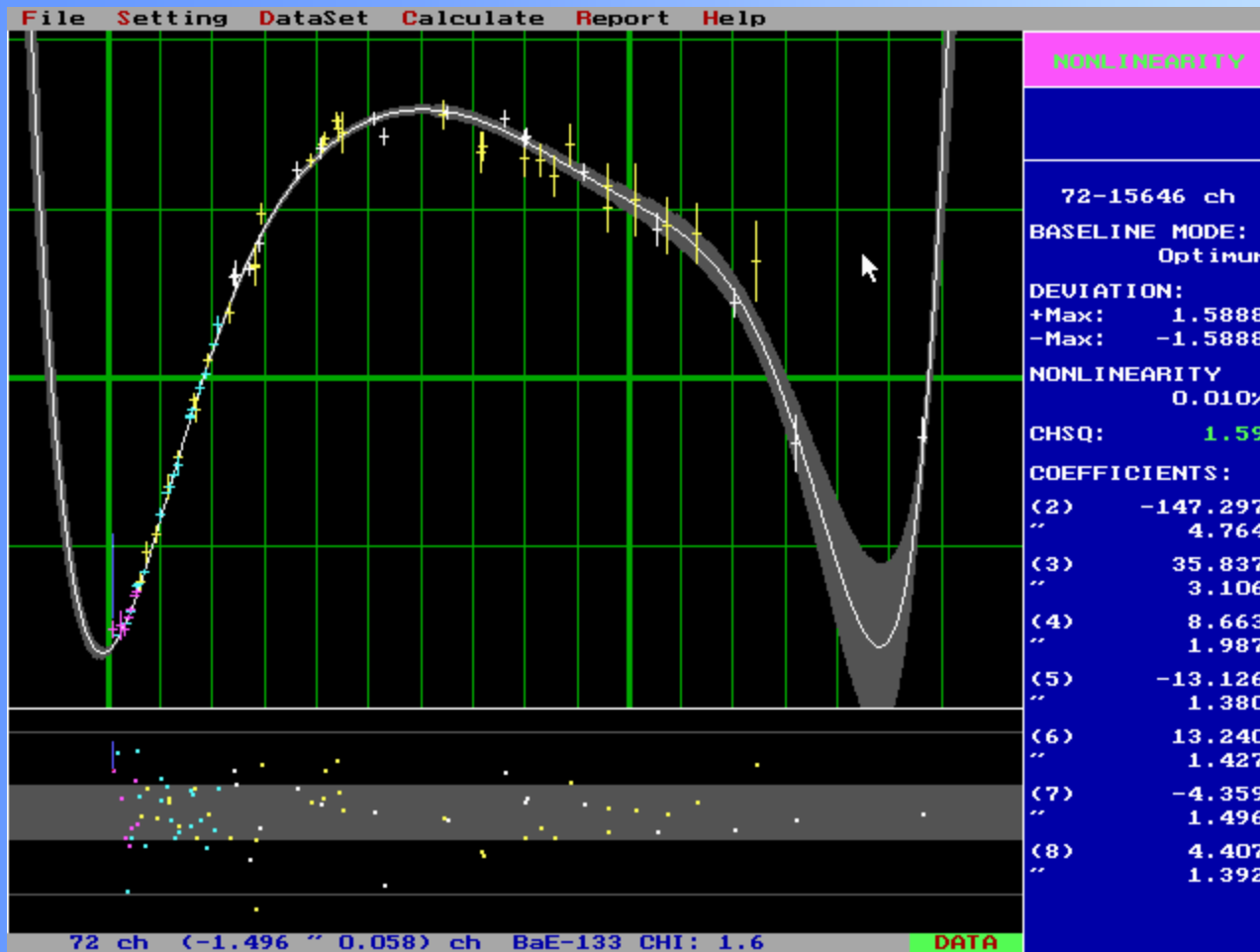
R. G. Helmer (INEEL)



Comparison of efficiencies Compton-suppressed mode

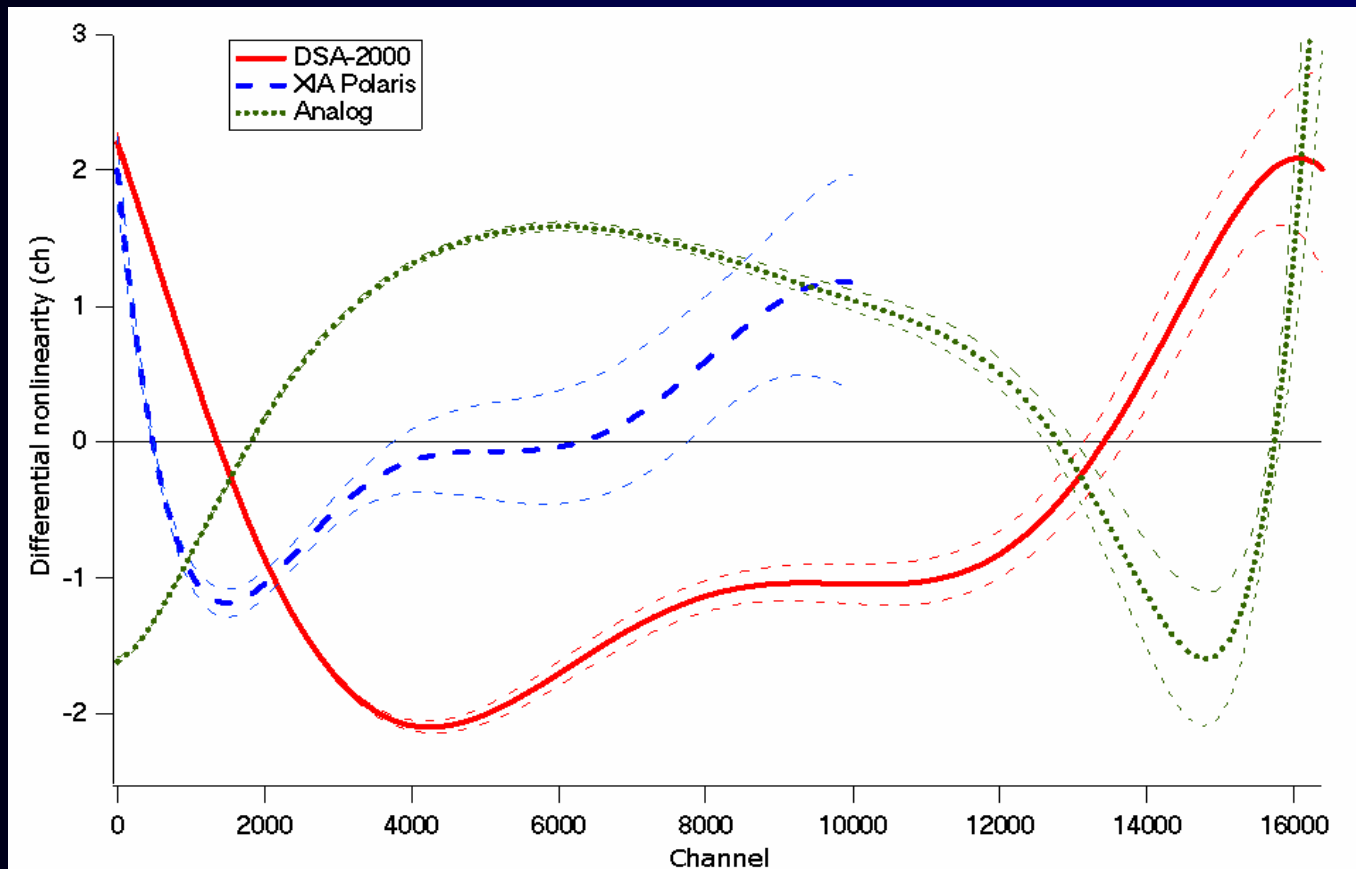


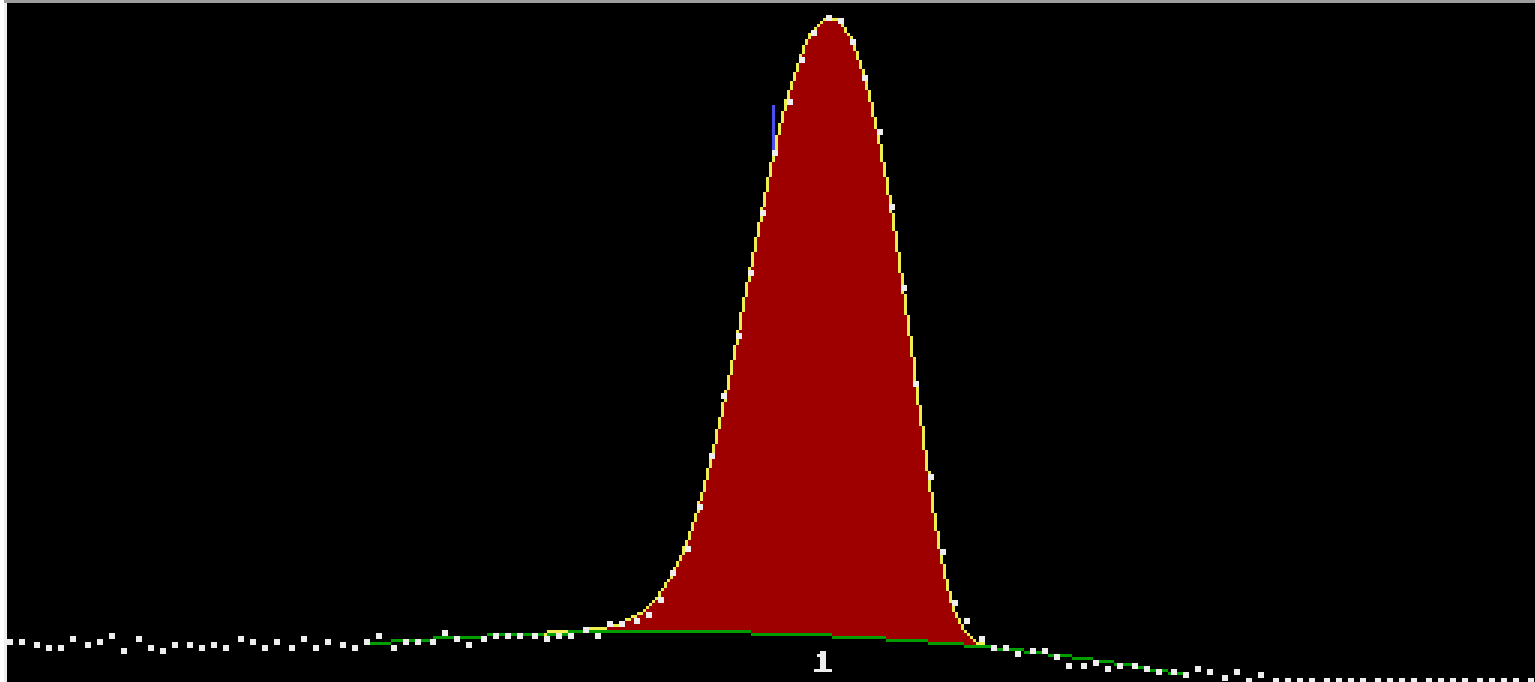
Nonlinearity analysis



- Measured with ^{152}Eu source and $\text{Cl}(n,\gamma)$
- 16k channels
- Nonlinear fit of orthogonal polynomials
- Centred around zero

Nonlinearity





PB8C_267.MCA

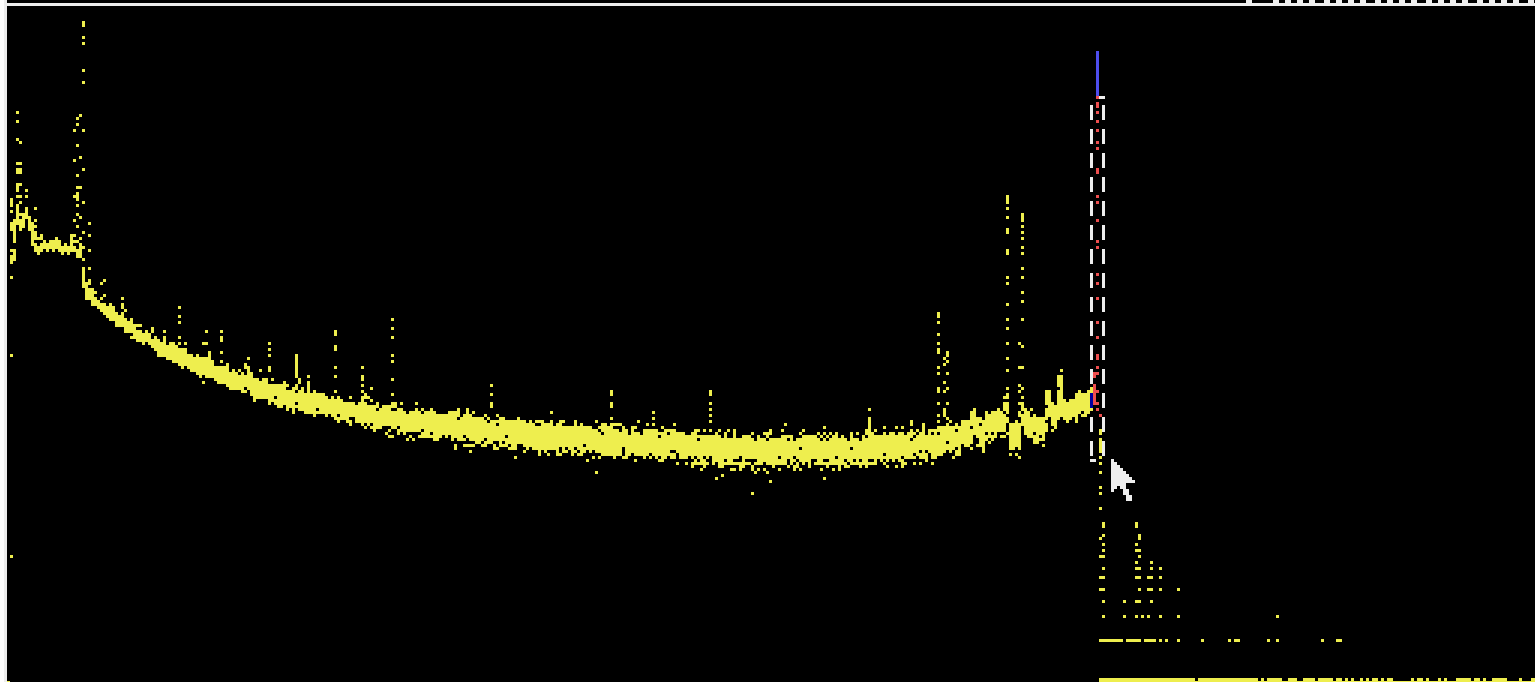
PB8C_267.LOG
11595-11659 ch

CALIBRATION:
Width: manual
Energy: manual

CORRECTIONS:
M98w.LIN

TRY : 1
CHSQ/(4*SIG) :
1.329229

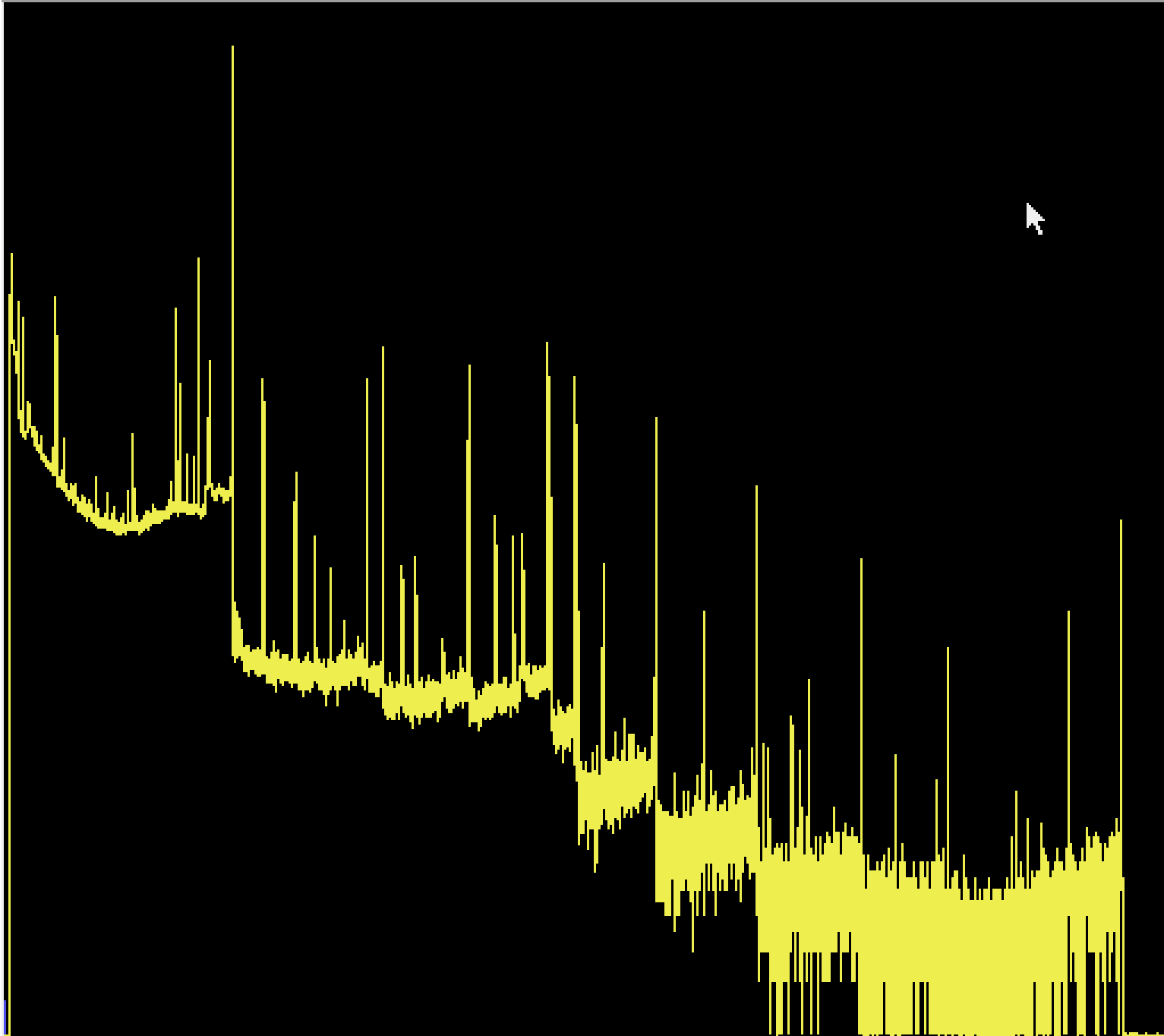
PEAK(S) : 1
1: 7647.9 keV
256936 cnt



CURSOR: 11627 CH 13122 CNT 7645.283 keV

SPECT LOG

<<REFITS



MEL_H66C.MCA

MCA #1
Tag No=953
MCA Mode: PHA+
COLLECTED AT
23:39:27
03-17-2000

ELAPSED TIME
Live=216721 sec
True=217492 sec

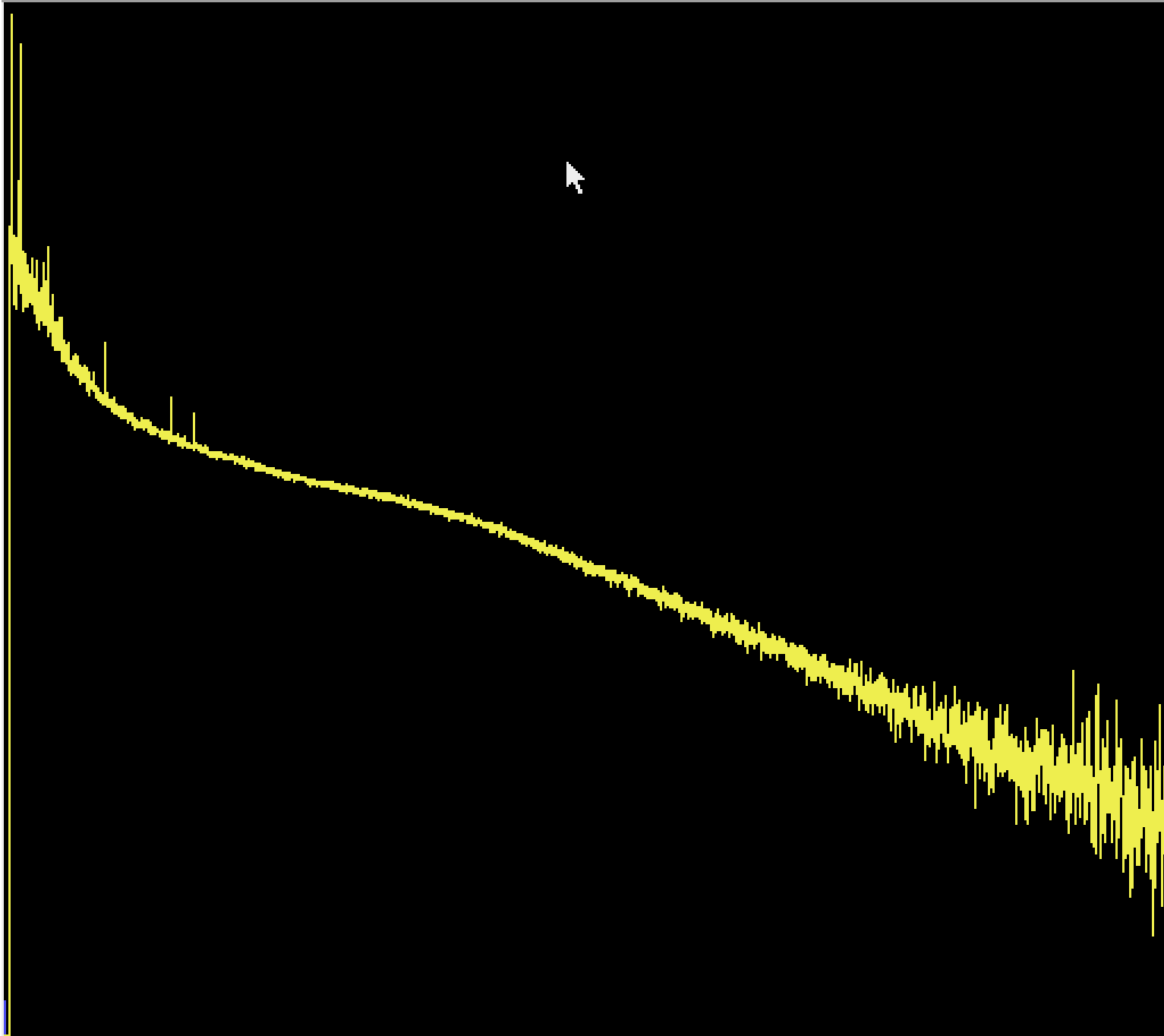
CALIBRATION:

ENERGY
Unit: keV
E0: 6.1266
E1: 0.6897

NO WIDTH

LENGTH:16384 CH
SUM: 10852693
MAX: 674198
MIN: 0

DISPLAY
FULL SPECTRUM



EUD_A77C.MCA

MCA #1
Tag No=1421
MCA Mode: PHA+
COLLECTED AT
13:49:39
06-29-1998

ELAPSED TIME
Live=5699 sec
True=6828 sec

CALIBRATION:

ENERGY
Unit: keV
E0: 1.6578
E1: 0.4455

NO WIDTH

LENGTH:16384 CH
SUM: 38605635
MAX: 2052606
MIN: 0

DISPLAY
FIRST HALF

4. Special techniques

Dynamic range

$$(\mu_A/\mu_B)_{\max} / (\mu_A/\mu_B)_{\min} \approx 10^6 \text{ — } 10^7$$

e.g. 1 mg Mn in 1g H₂O (~0.1 g H)

_____ 1 mg H₂O (0.1 mg H) in 1 g Mn

Can it be improved?

Simultaneous PGAA and NAA measurement with a chopper

- **Beam open**

prompt gamma rays

decay gamma rays

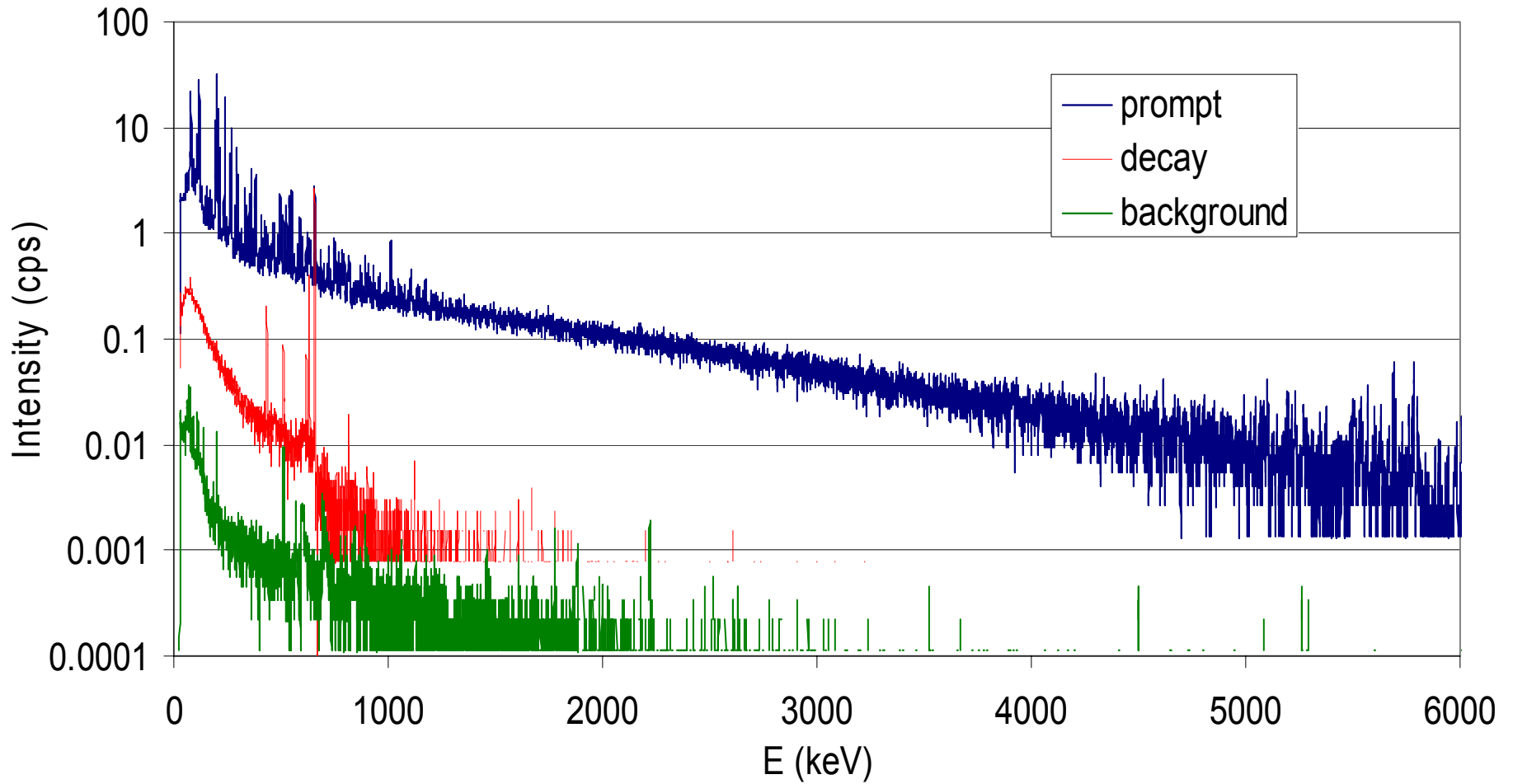
Usual PGAA spectrum

- **Beam closed**

only **decay** gamma rays

cyclic NAA spectrum

Spectra



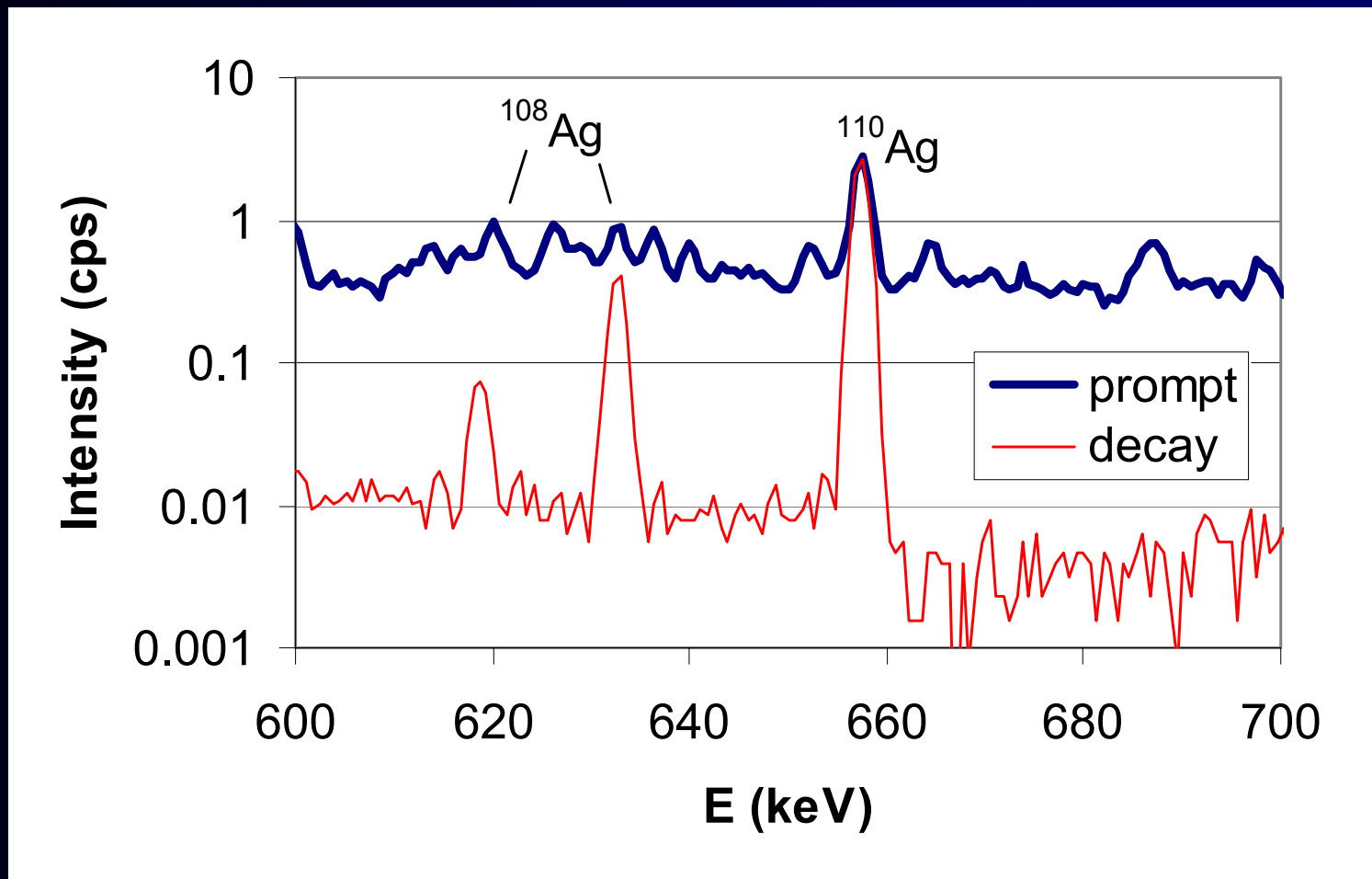
Elements with decay lines

< 1 s:	Na,
< 1 min:	F, Sc, Ge, Pd, Ag, In, Er, Hf, W,
< 10 min	Mg, Al, V, Cr, Se, Br, Rh, Dy, Ir,
< 1 h:	Ga, Rb, Sn, I, Pr, Nd, Ta, Re,
<1 day:	Mn, Cu, Sr, Cs, Ba, Eu, Lu,
longer:	As, Ru, La, Ce, Tb, Ho, Yb, Au,

Background

Name	Reactor	Beam	Vacuum	Chopper	Phase	Background (cps)
Room background	off	—	—	—	—	0.63
Beam-off background	on	off	—	—	—	1.5
Beam-on background (in vacuum)	on	on	yes	—	—	4.0
Beam background in air	on	on	no	—	—	5.6
Chopper background in prompt phase	on	on	no	on	prompt	5.3
Chopper background in decay phase	on	on	no	on	decay	4.6

Prompt and decay spectrum of Ag



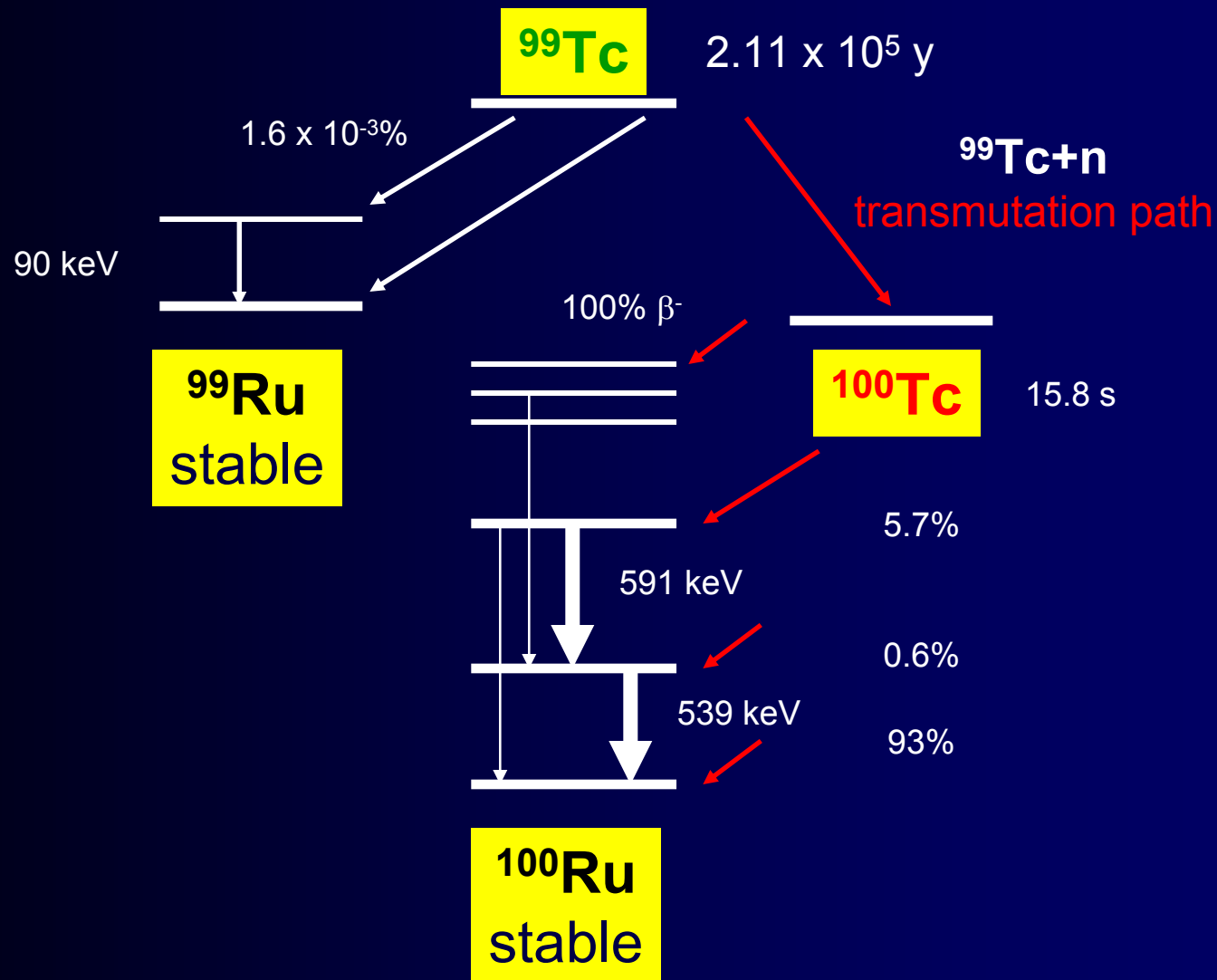
Mea- sure- ments

El	E (keV)	prompt spectrum		decay spectrum		gain
		rate	S/N	rate	S/N	
F	1633	1.63(2)	1300	1.63(6)	7000	5
Al	1779	2.62(1)	2000	2.61(1)	200,000	100
Sc	143	28(1)	200	29(1)	40,000	20
	147	46.8(7)	400			
V	125	20.0(2)	270			
	1434	9.3(1)	1200	8.8(4)	41,000	34
Cu	159	29.6(2)	200			
	1039	0.58(2)	90	0.55(2)	900	10
Ag	198	82(1)	62			
	658	7.16(15)	20	7.3(3)	1900	95
In	163	157(3)	53	146(6)	3700	70
Er	185	32(1)	400			
	208	14.6(8)	180	15.4(6)	11,000	60

Results

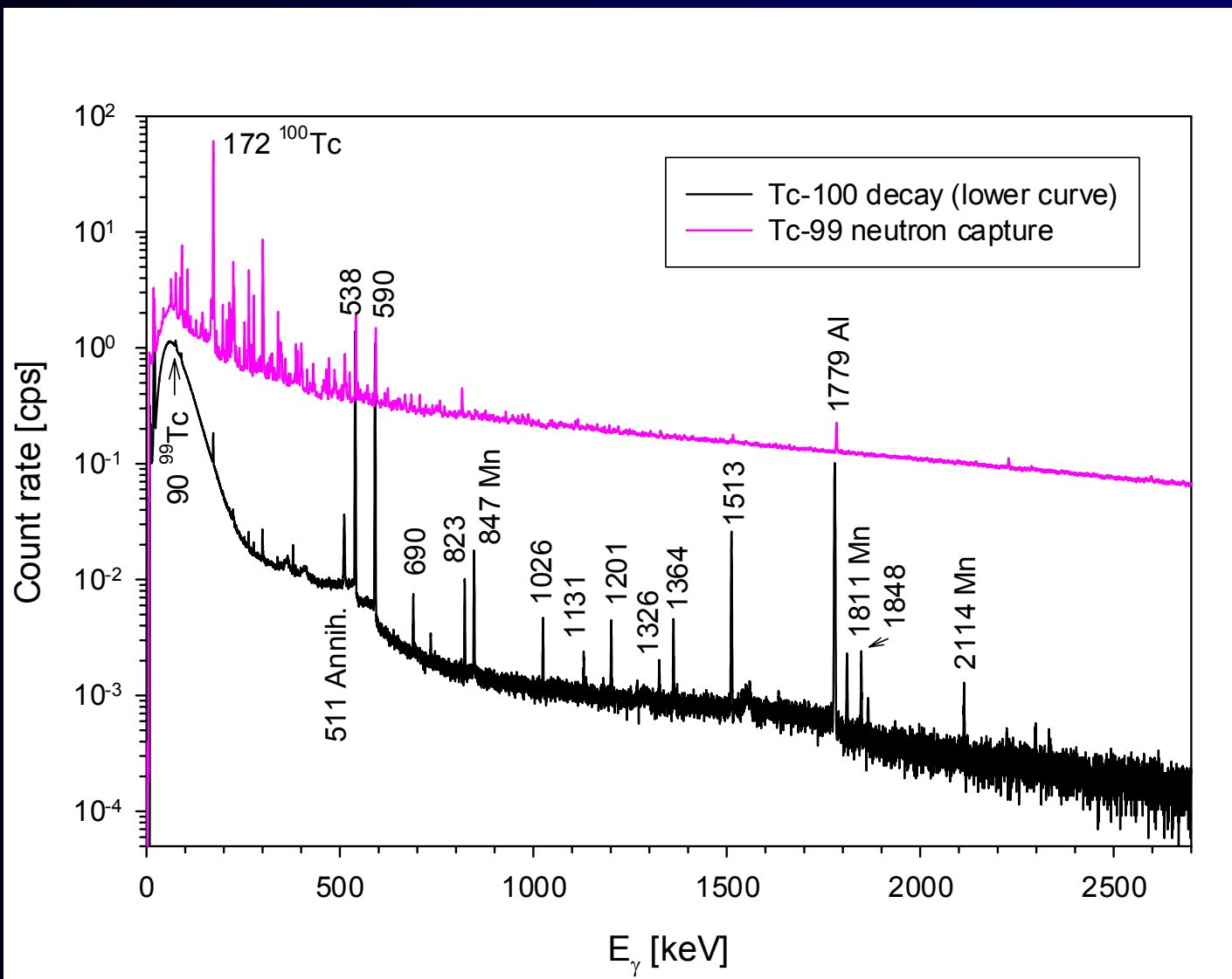
Element	Half-life	Energy (keV)	decay line σ_γ (barn)	prompt line σ_γ (barn)
F	11.16 s	1633	0.0093(3)	—
Al	2.24 m	1779	0.233(4)	<0.005
Sc	18.75 s	143	4.88(10)	<0.13
V	3.75 m	1434	5.20(10)	<0.3
Cu	5.12 m	1039	0.0600(12)	<0.0023
Ag	24.6 s	658	1.93(4)	<0.08
In	2.18 s	163	15.8(8)	<1.1
Er	2.27 s	208	2.15(9)	<0.18

Scheme of transmuted ^{99}Tc



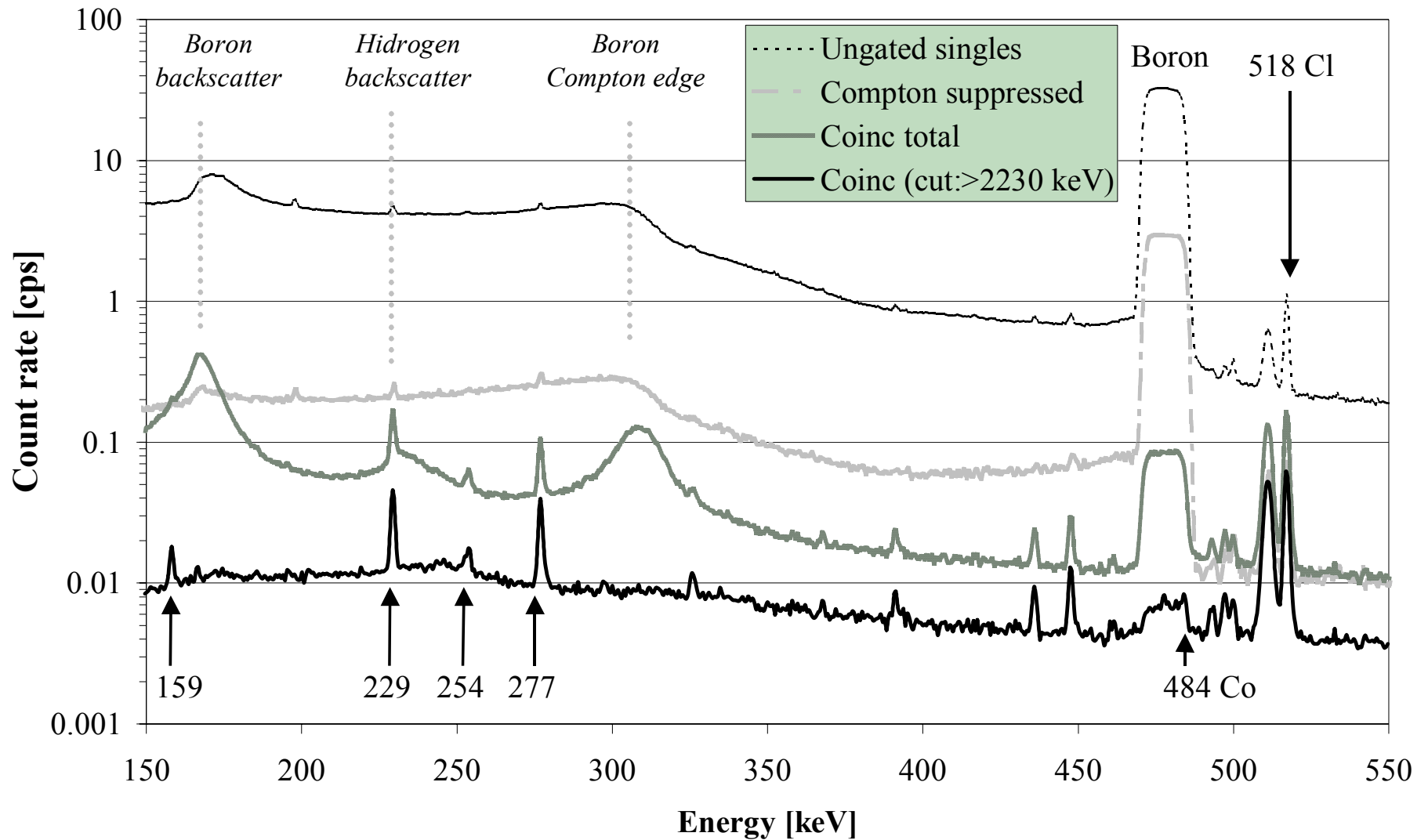
PGAA on a radioactive target

^{99}Tc chopped beam (n,γ) + decay spectra



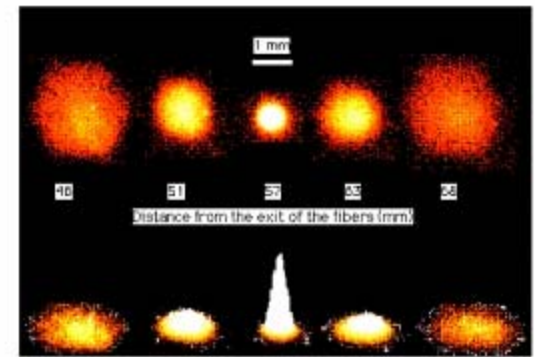
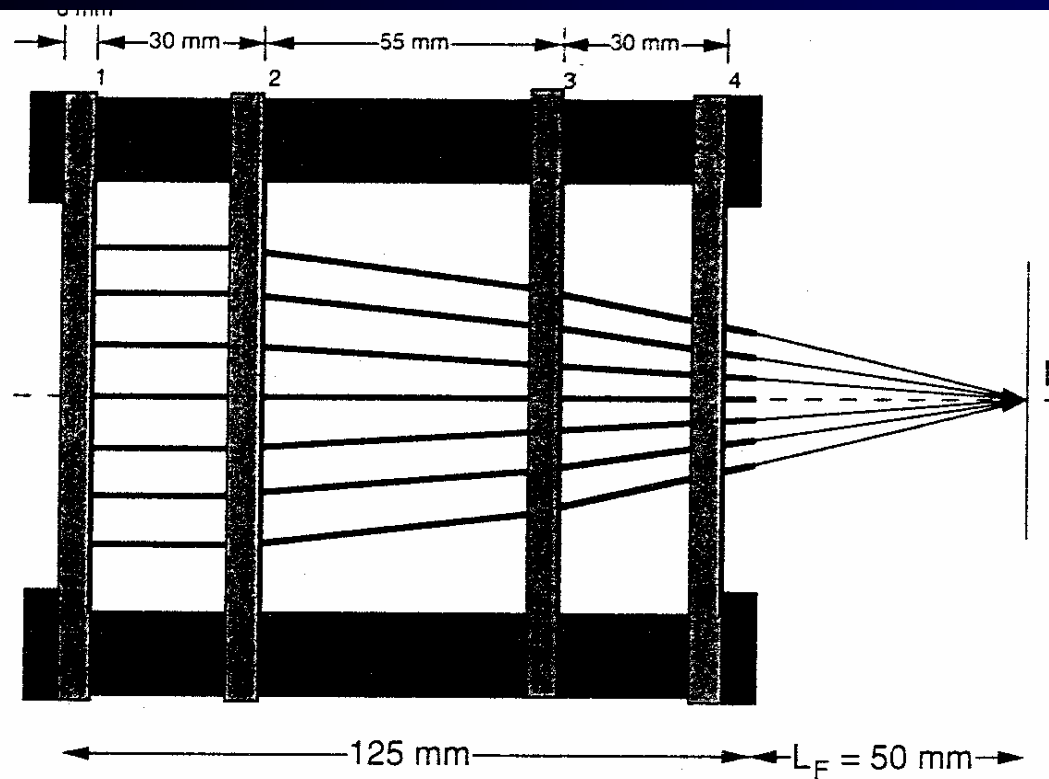
Increase of PGAA sensitivity by coincidence

9.35 mg $\text{CoCl}_2(2\text{H}_2\text{O})$ and 22.1 mg of H_3BO_3 / ml



Focused Beams with Cold Neutrons

Polycapillary neutron lens (NIST). 1763 glass fibres (0.5 mm), each with 1657 channels (9 μm) focus neutrons to 0.5 mm spot.



End of Part 1