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

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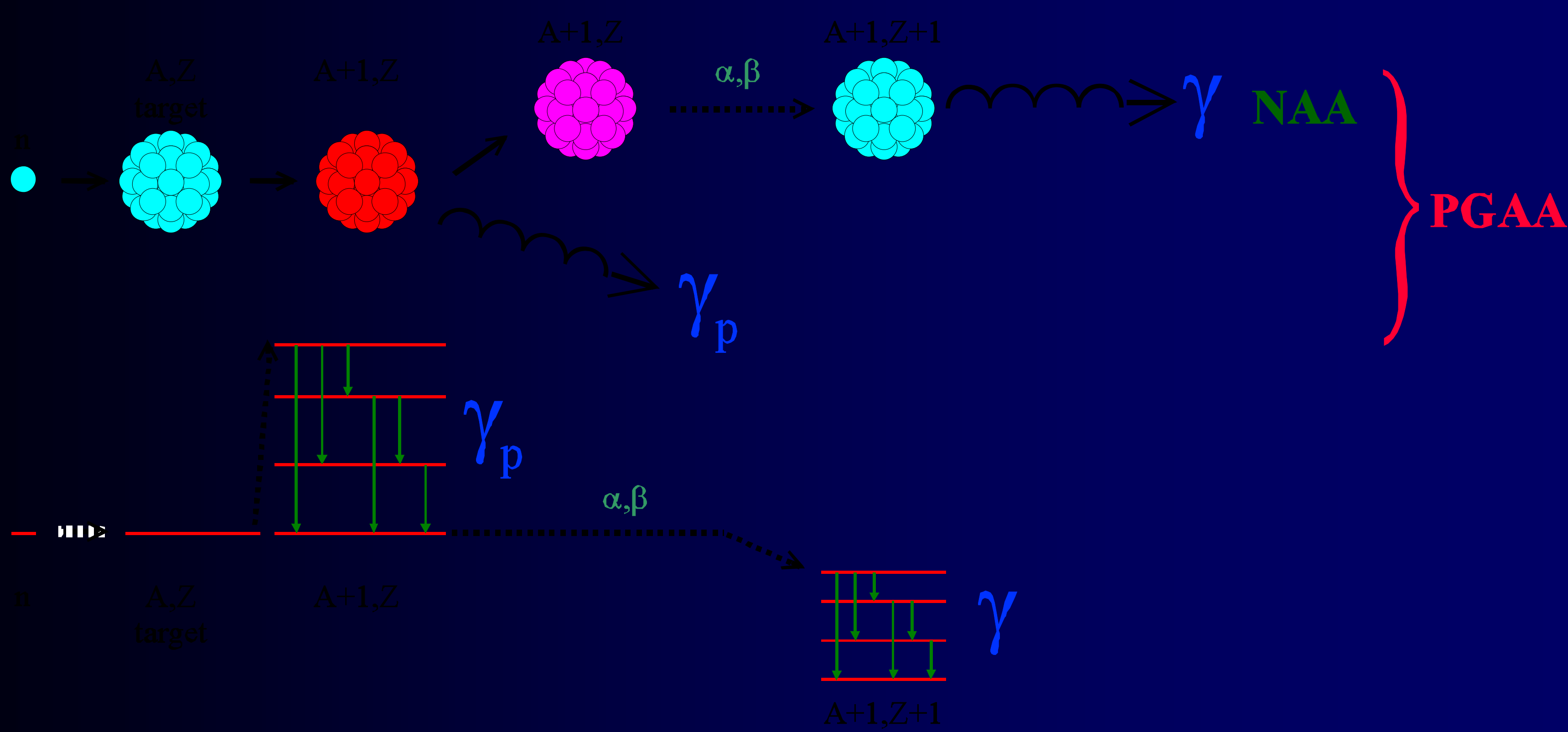
Neutron-induced prompt gamma activation analysis (PGAA or PGNAA)

- **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

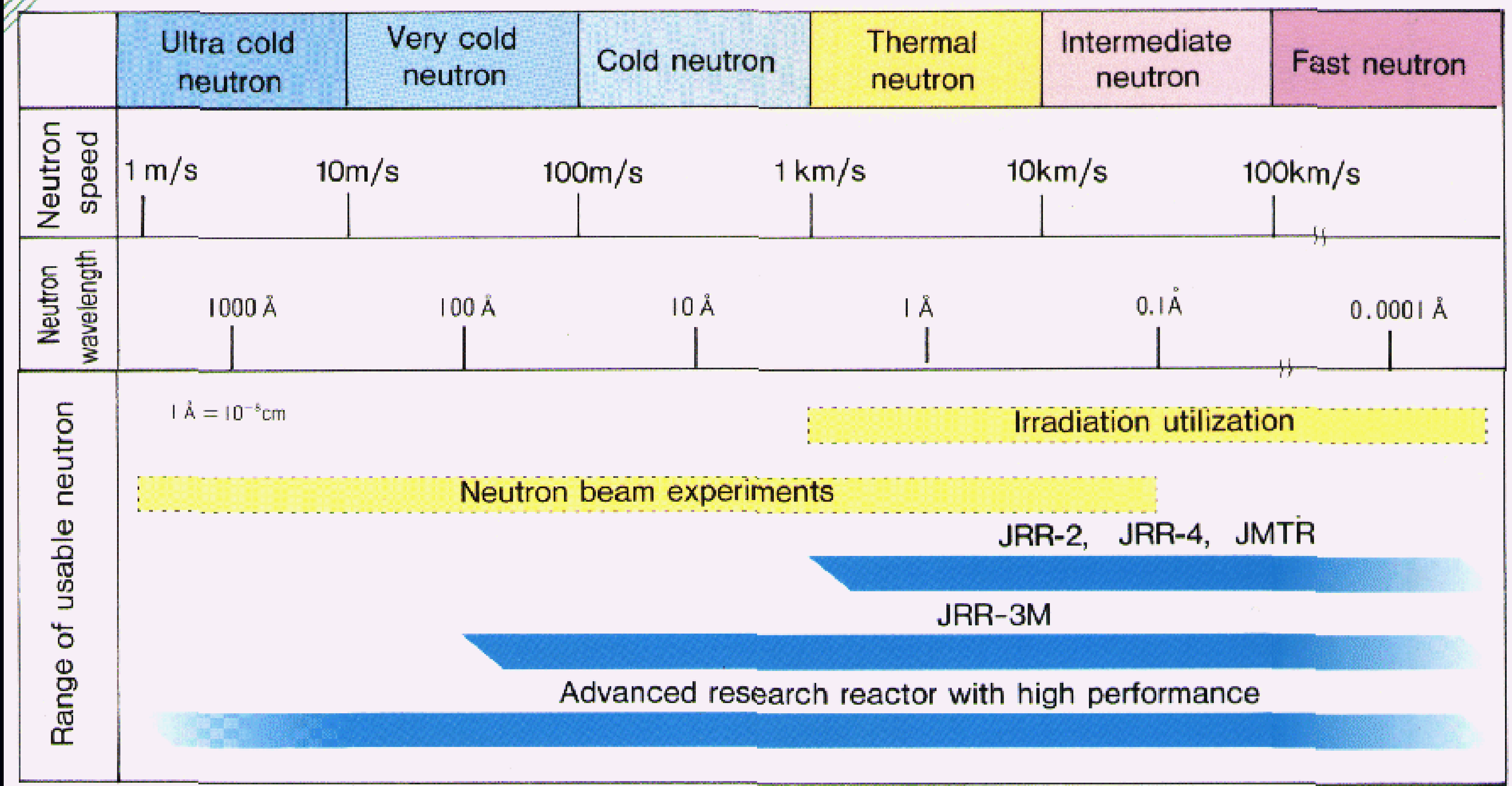
Neutron capture and subsequent beta decay



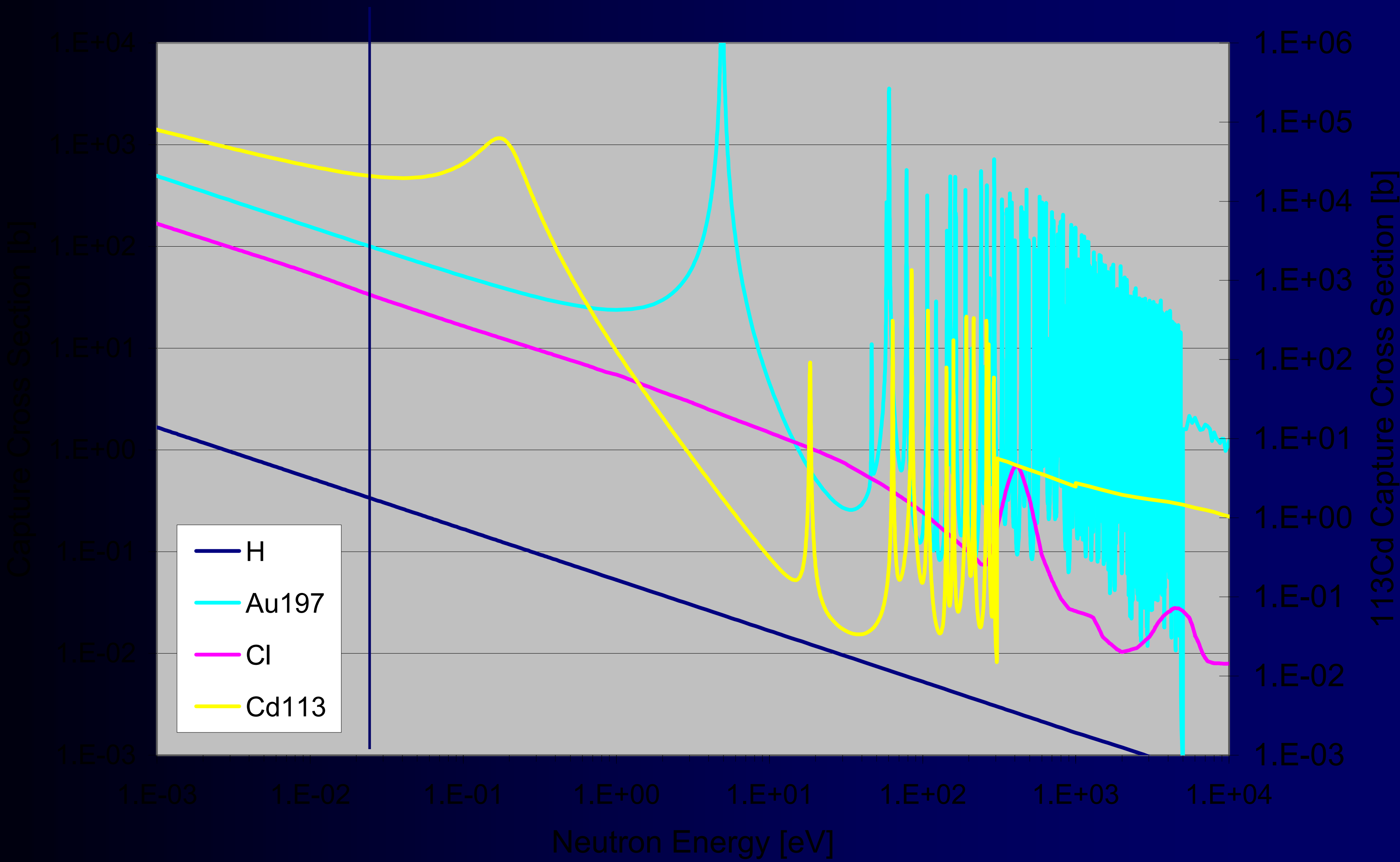
Basic reaction: Radiative neutron capture

- Most important reaction for slow neutrons
  - High cross sections (but vary widely!) >> all elements
  - Simple 1/v behavior (exceptions!) >> easy quantification
  - Reaction rate ~ 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

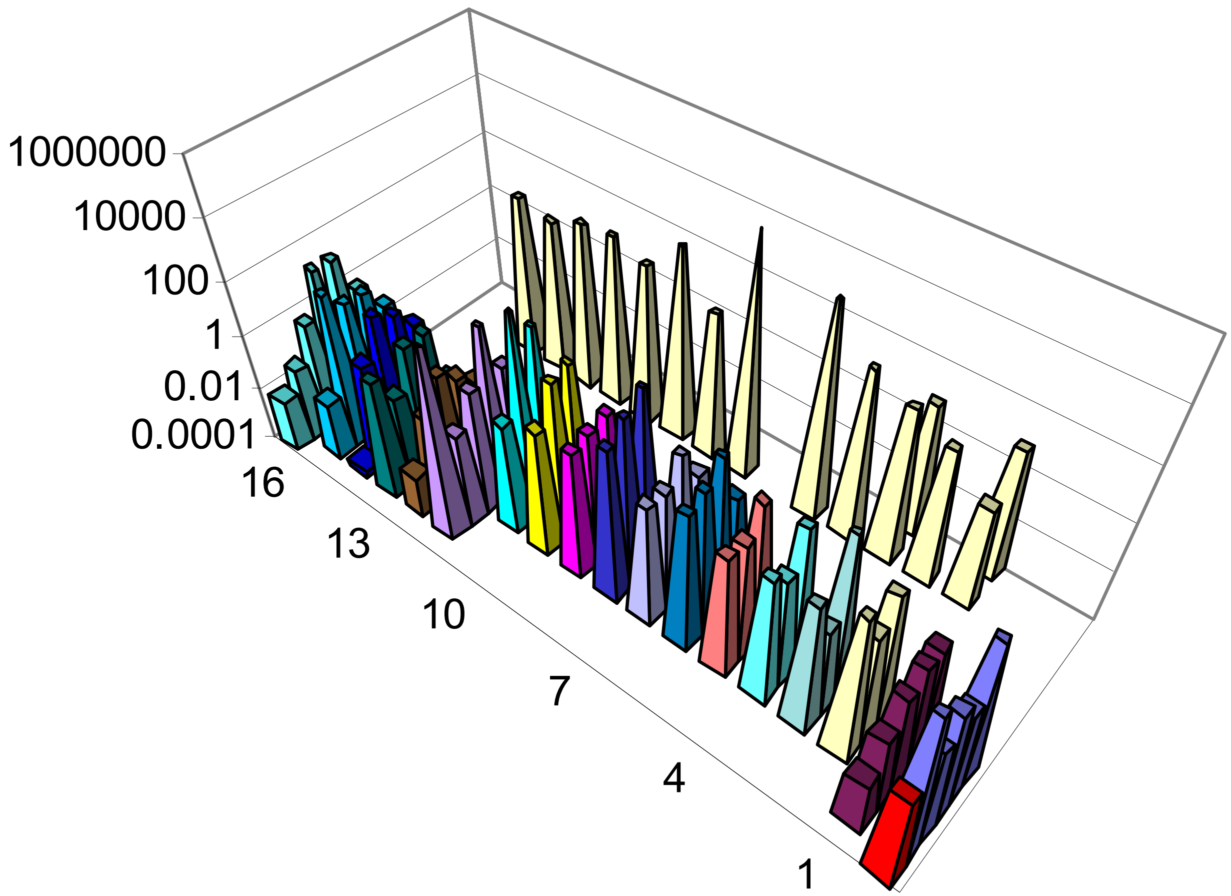
Various neutrons for use



Capture Cross Sections

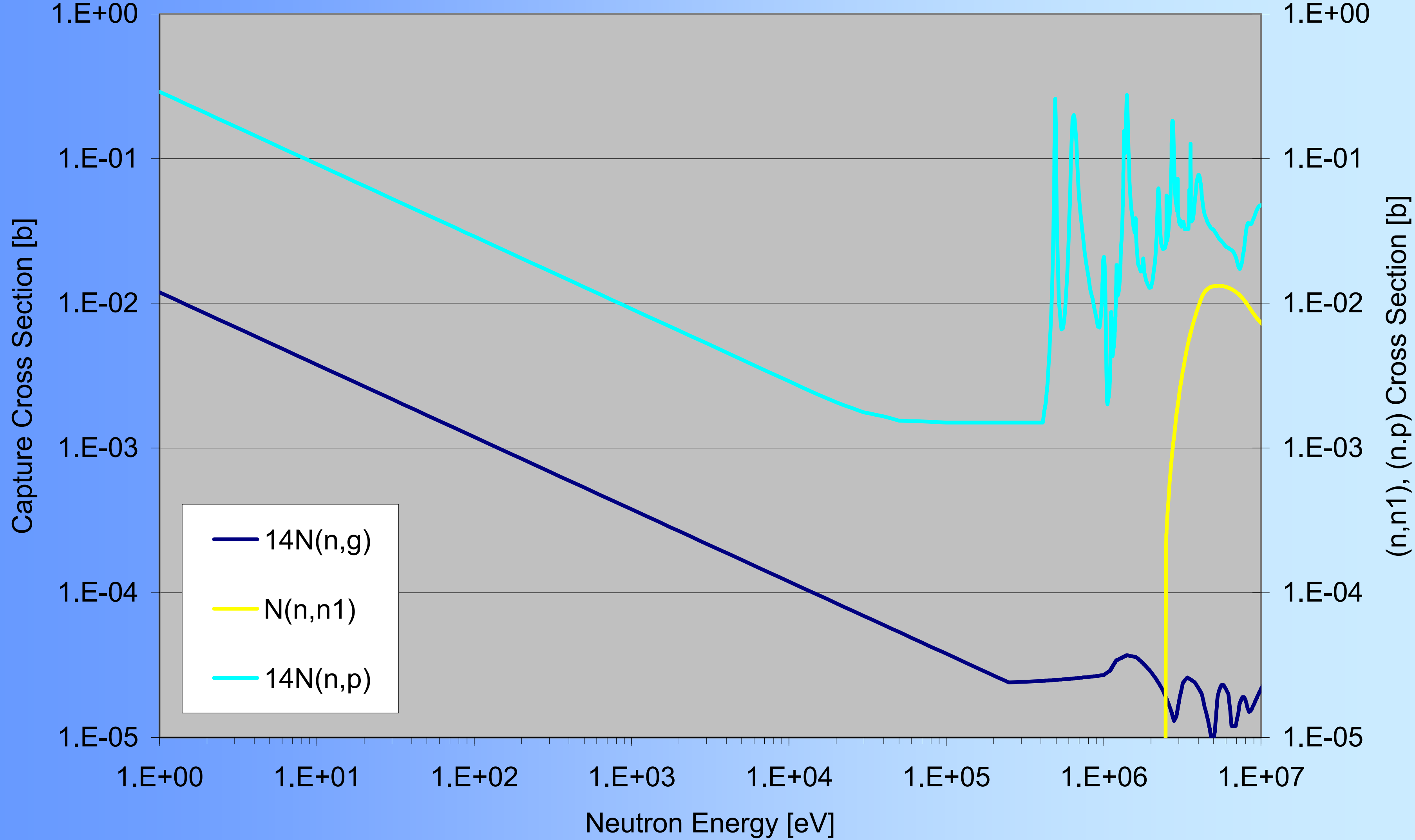


Thermal neutron capture cross sections



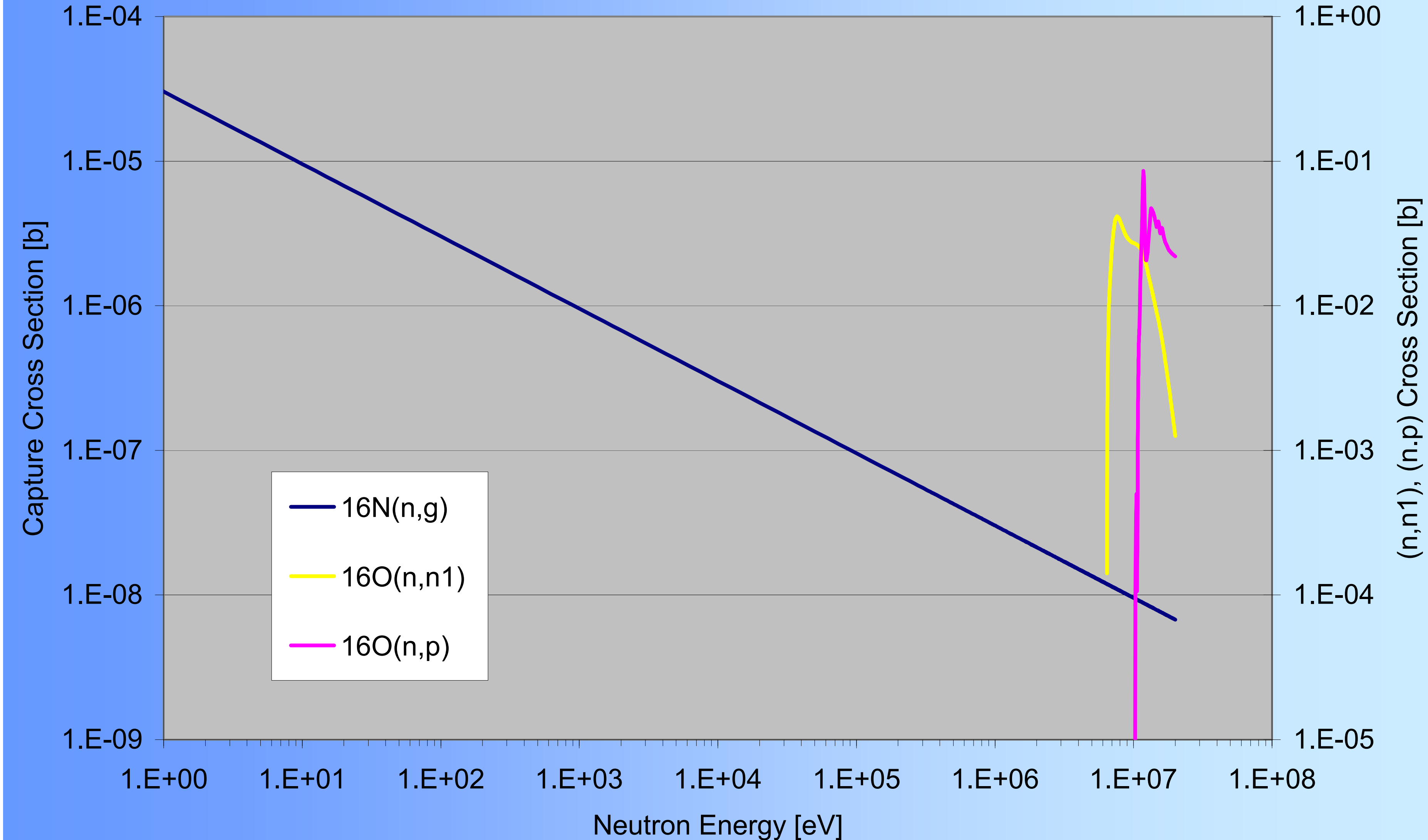
^{14}N Cross Sections

σ_{th}=8x10^{-2} b



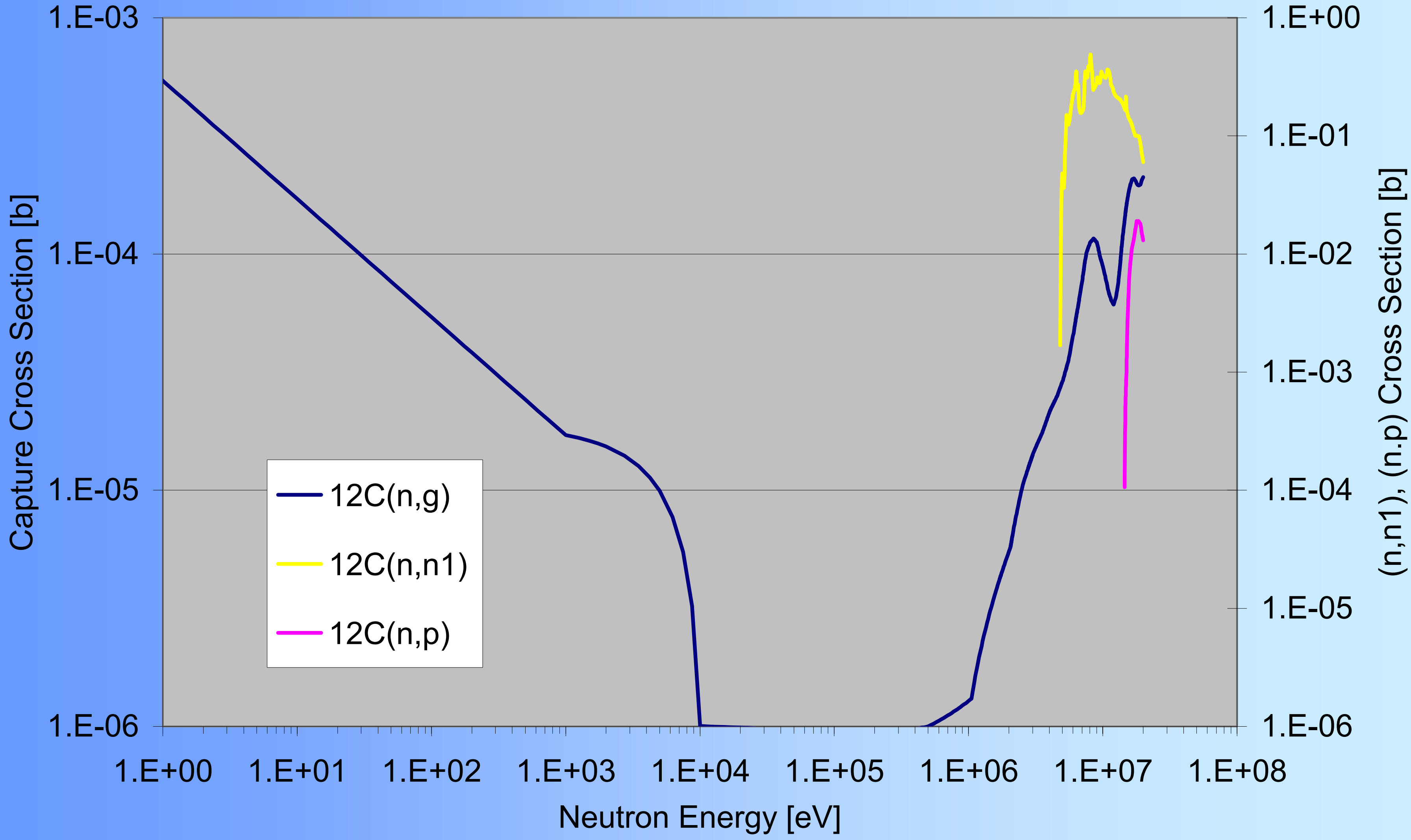
^{16}O Cross Sections

σ_{th}=2x10^{-4} b

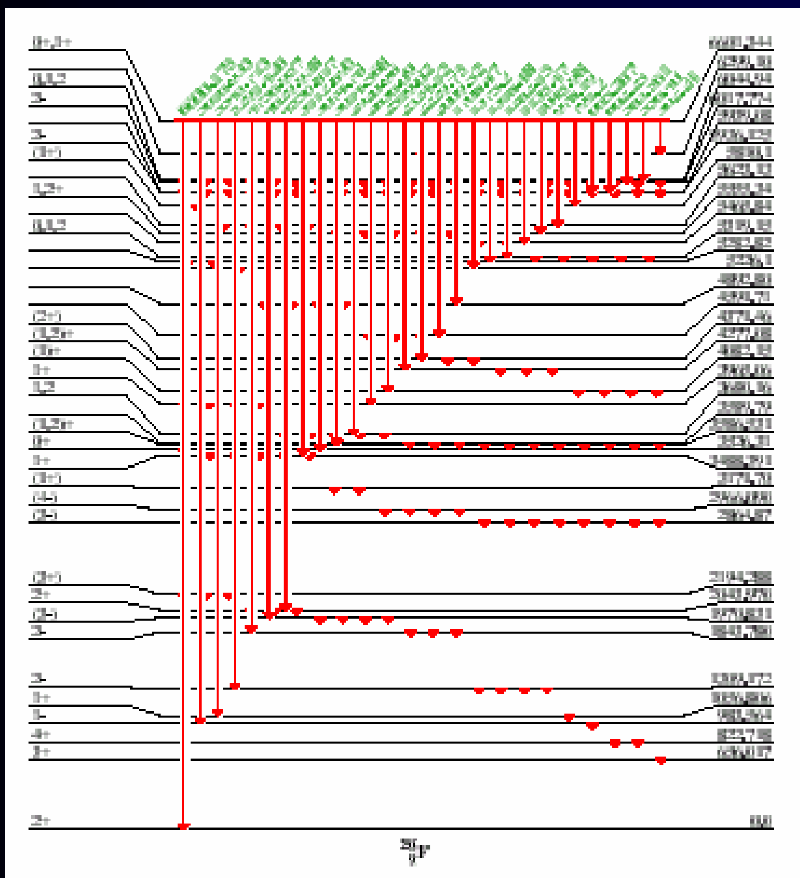


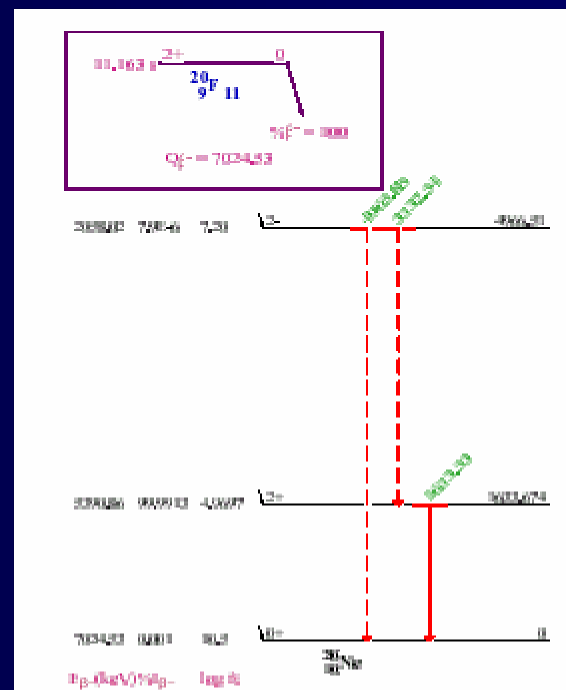
^{12}C Cross Sections

$\sigma_{th}=3.8x10^{-3}$ b

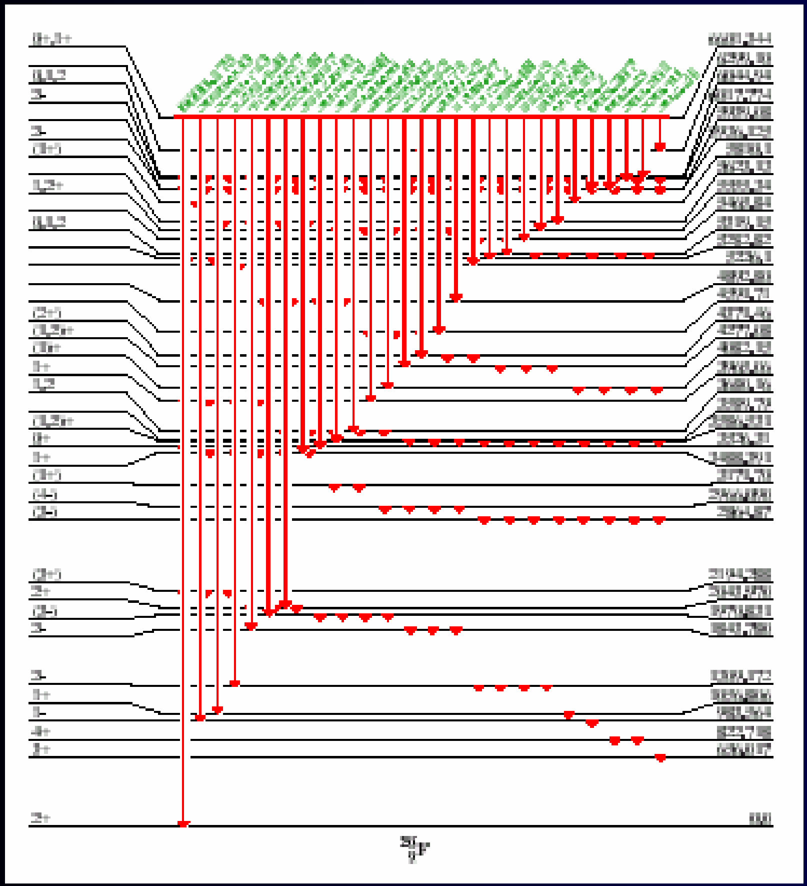


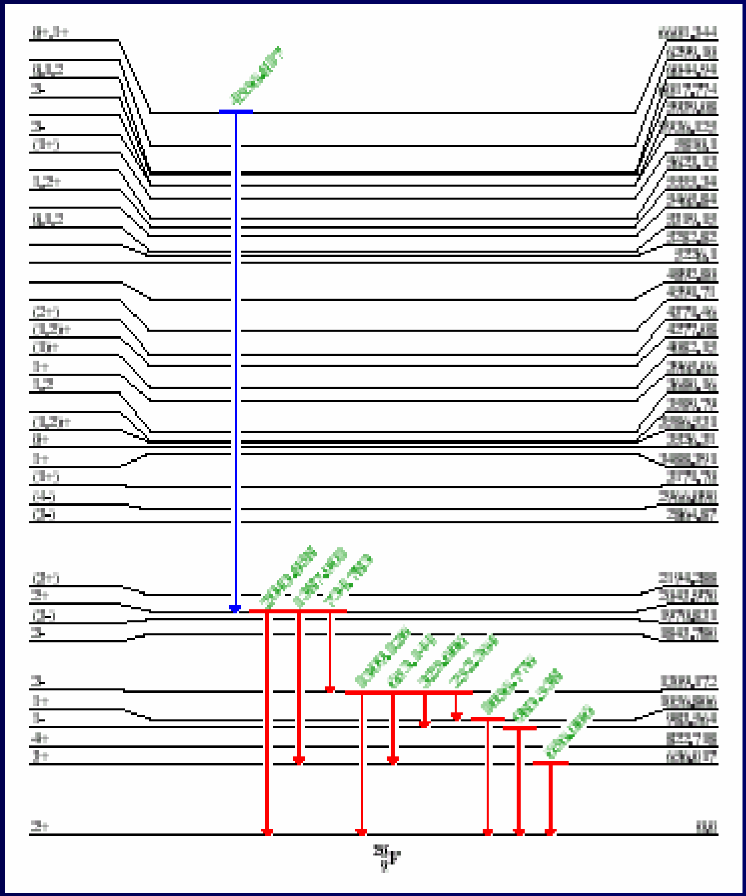
Nuclear Structure View of PGAA and NAA





Coincidence in PGAA

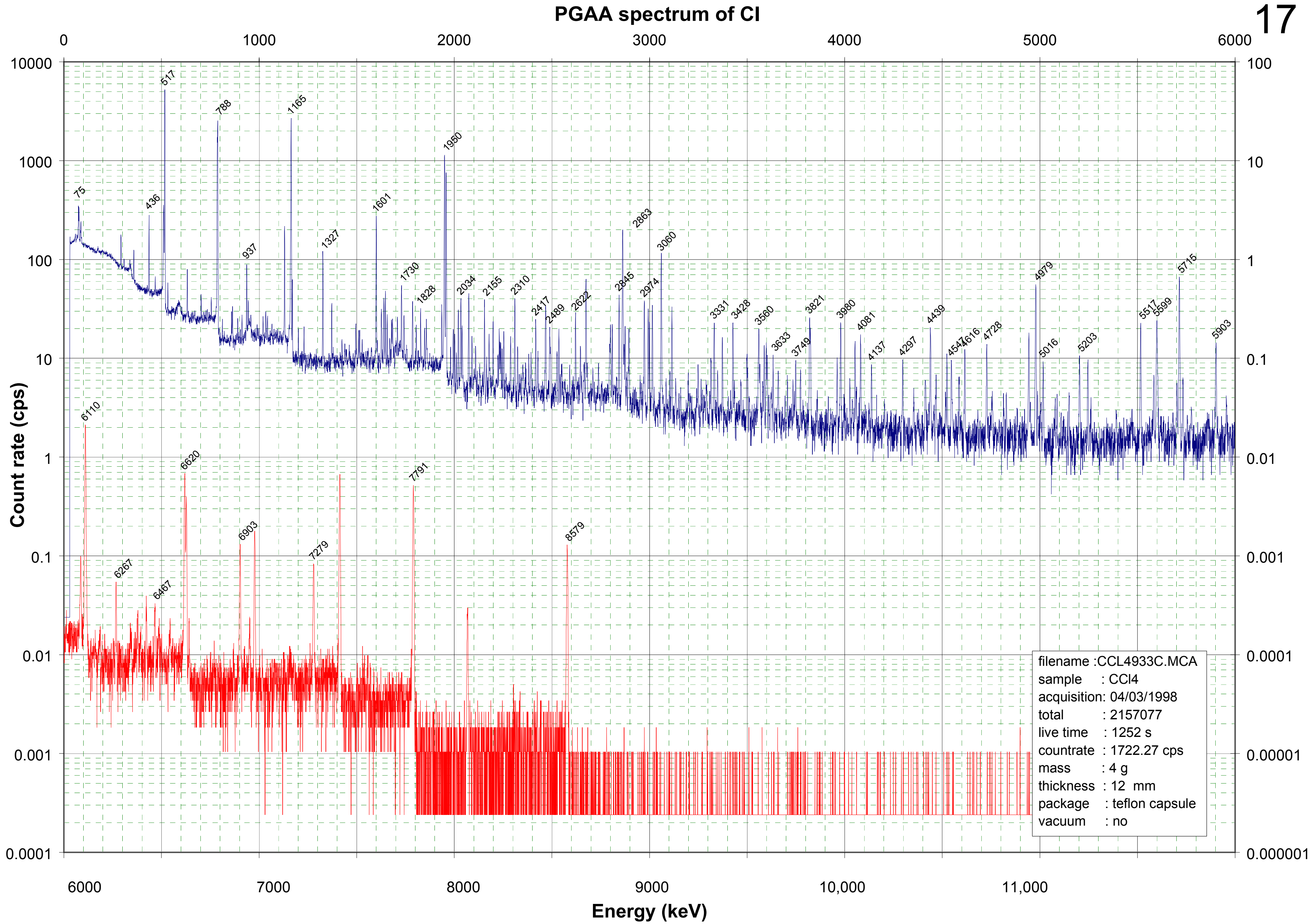




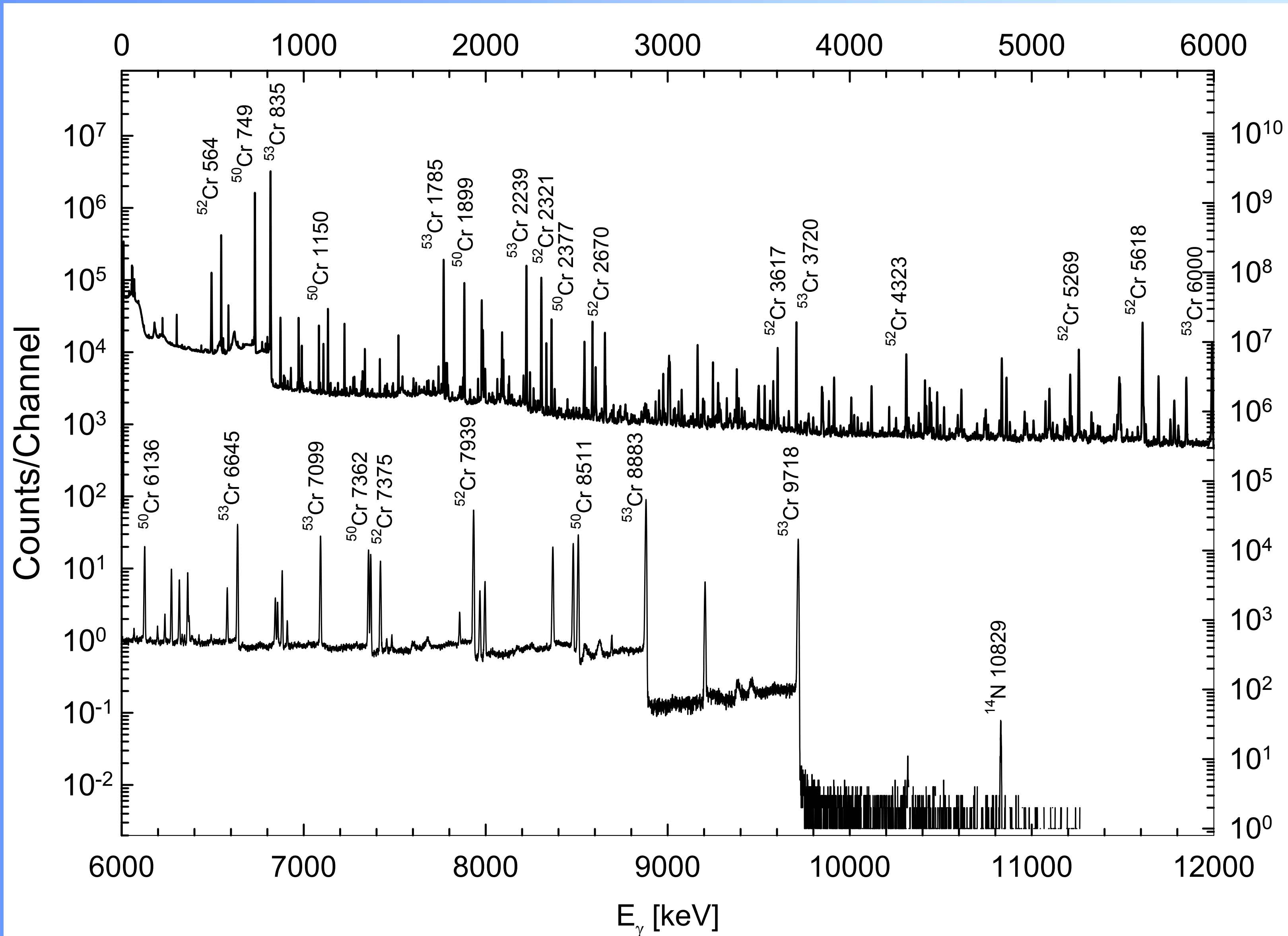
2. Features of PGAA

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
Count rate (cps)
Energy (keV)
75
436
517
788
937
1165
1327
1601
1730
1828
1950
2034
2155
2310
2417
2489
2622
2845
2863
2974
3060
3331
3428
3560
3633
3749
3821
3980
4081
4137
4297
4439
4616
4728
4979
5016
5203
5517
5599
5715
5903
6110
6267
6467
6620
6903
7279
7791
8579
filename :CCL4933C.MCA
sample : CCl4
acquisition: 04/03/1998
total : 2157077
live time : 1252 s
countrate : 1722.27 cps
mass : 4 g
thickness : 12 mm
package : teflon capsule
vacuum : no

Cr capture spectrum



Reaction Rate

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

σ - differential cross section (cm^2),

Φ - flux cm^{-2}·s^{-1} eV^{-1}

N - number of target nuclides (~ mass)

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

For thermal neutrons

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

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

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

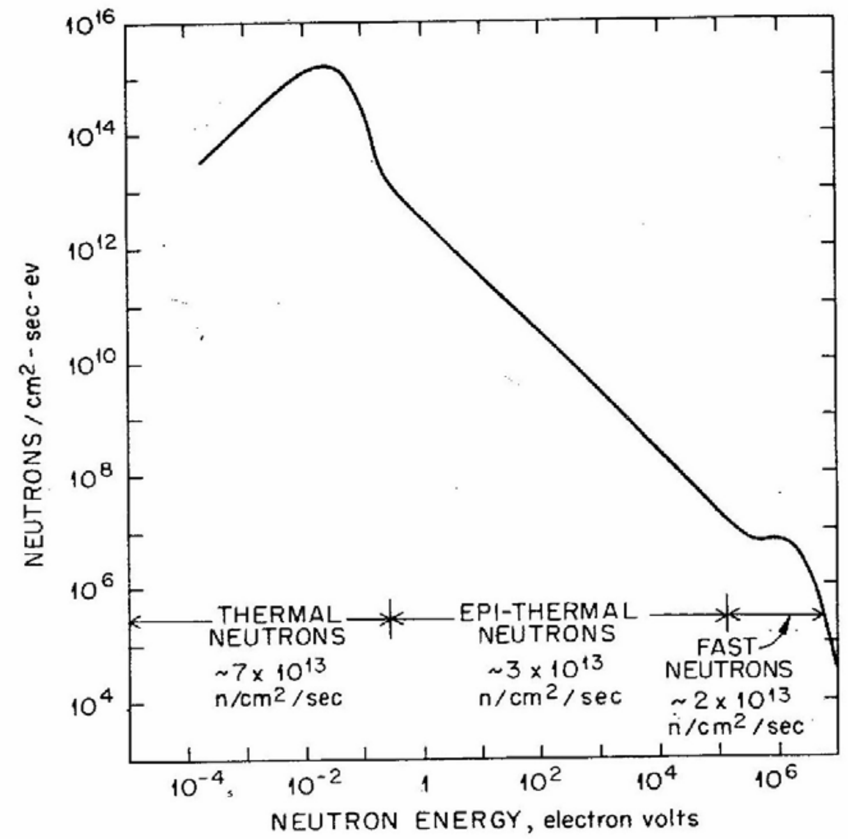
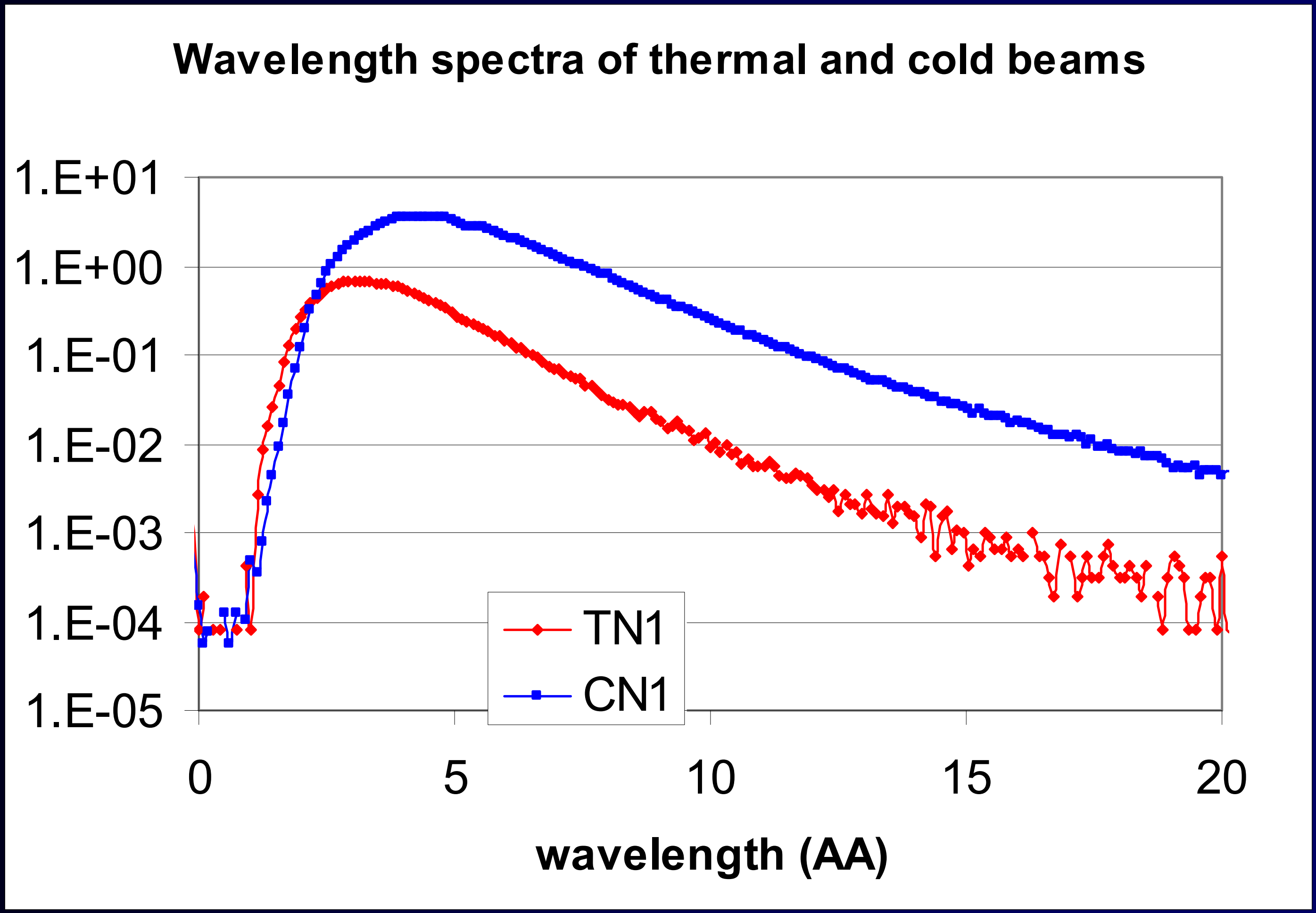


FIG. 2.1—General features of a reactor neutron spectrum.

Cold and thermal neutron spectra

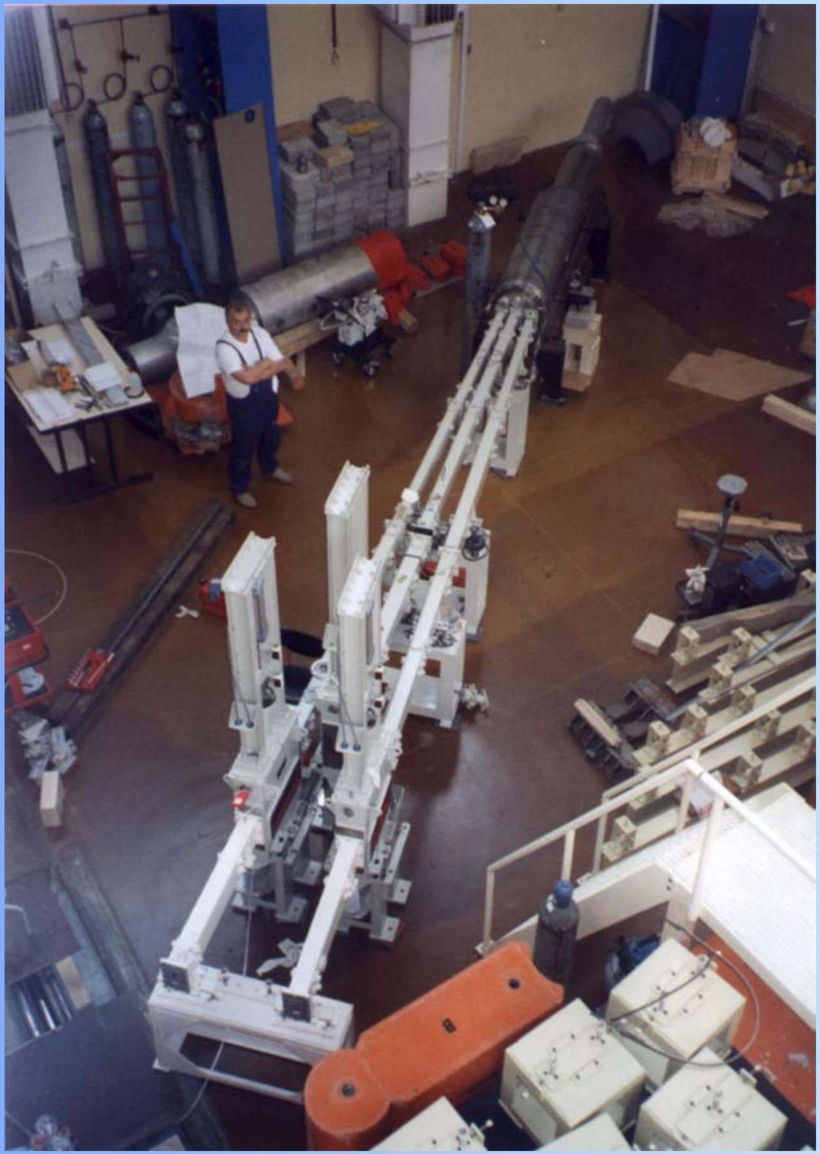


Advantages of cold neutron beams

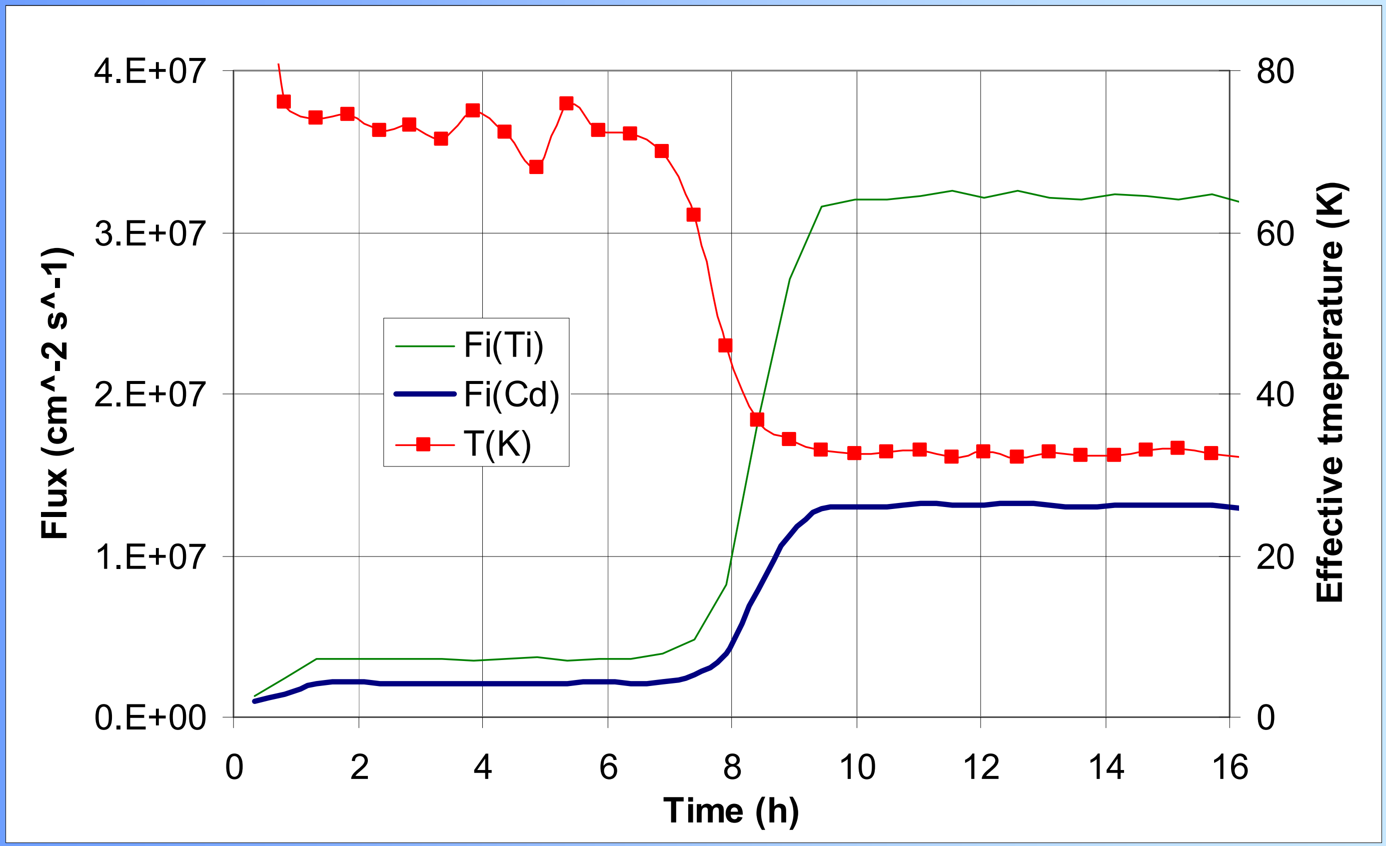
- Higher flux
- every nuclide behaves regularly
  – (follows the 1/v-law)
- every nuclide has higher cross section

higher reaction rate

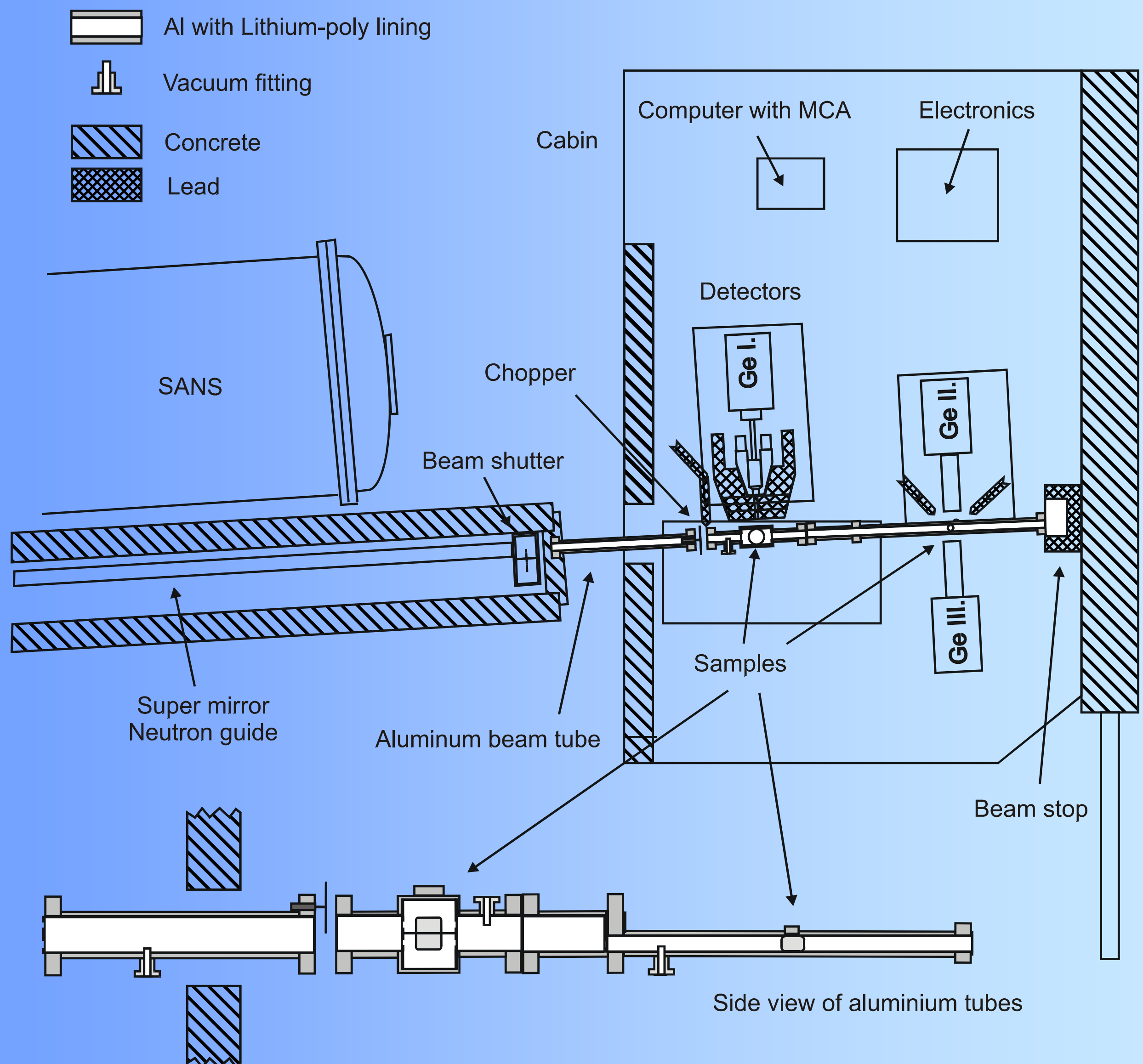
Neutron guides at Budapest



Starting of the cold source
at Budapest Research Reactor



The PGAA facility at Budapest



PGAA system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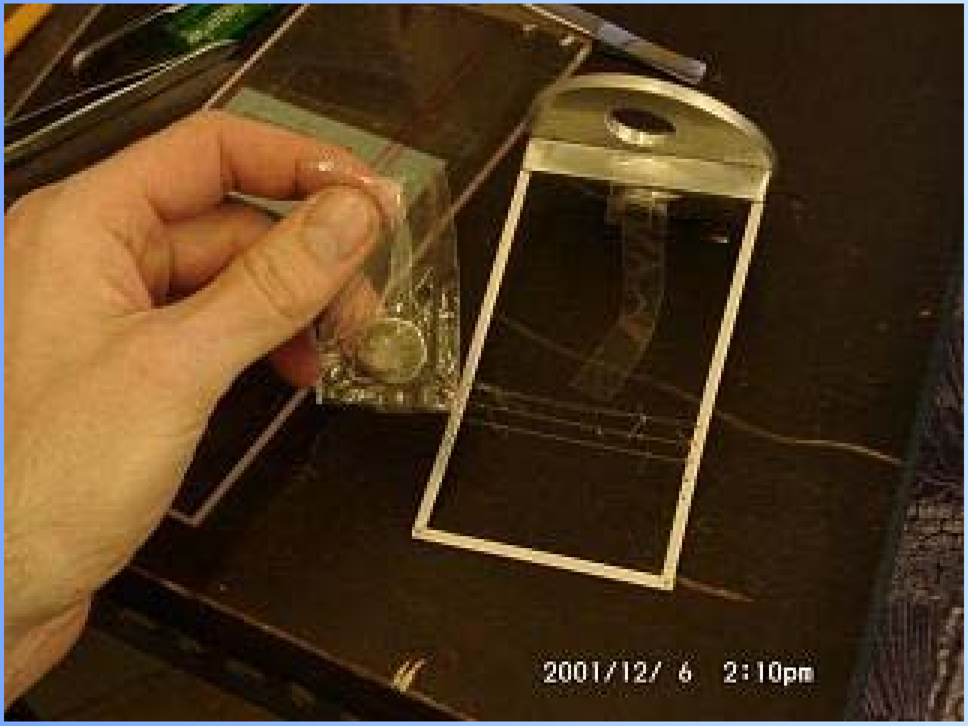
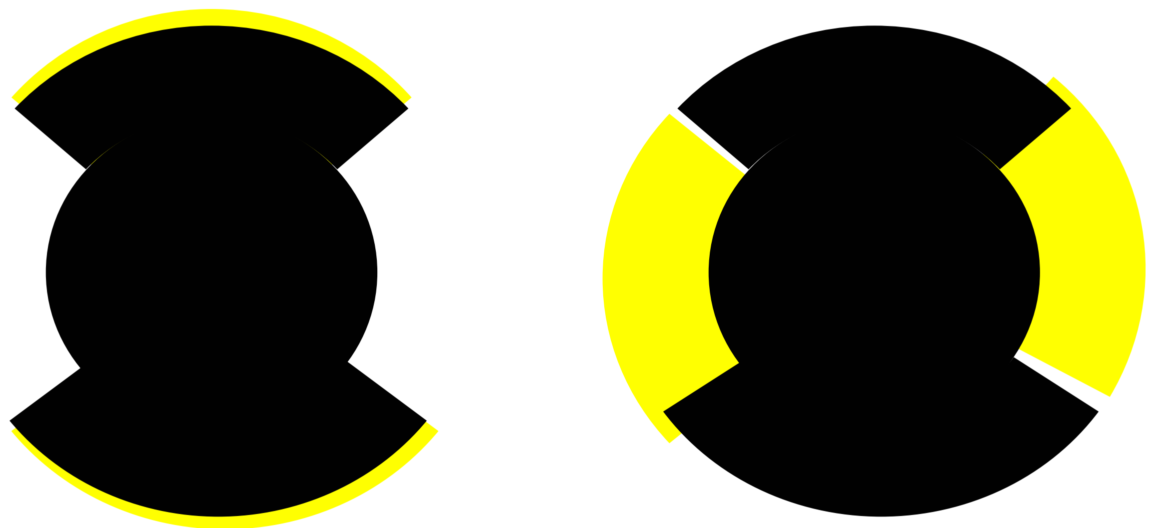
NIPS (Neutron Induced Prompt Gamma Spectroscopy) system

PGAA sample



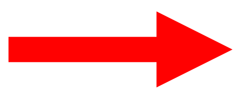
BEAM CHOPPER

Li-6 and Gd-coated rotating disks



Gap width: 0.2 – 50%

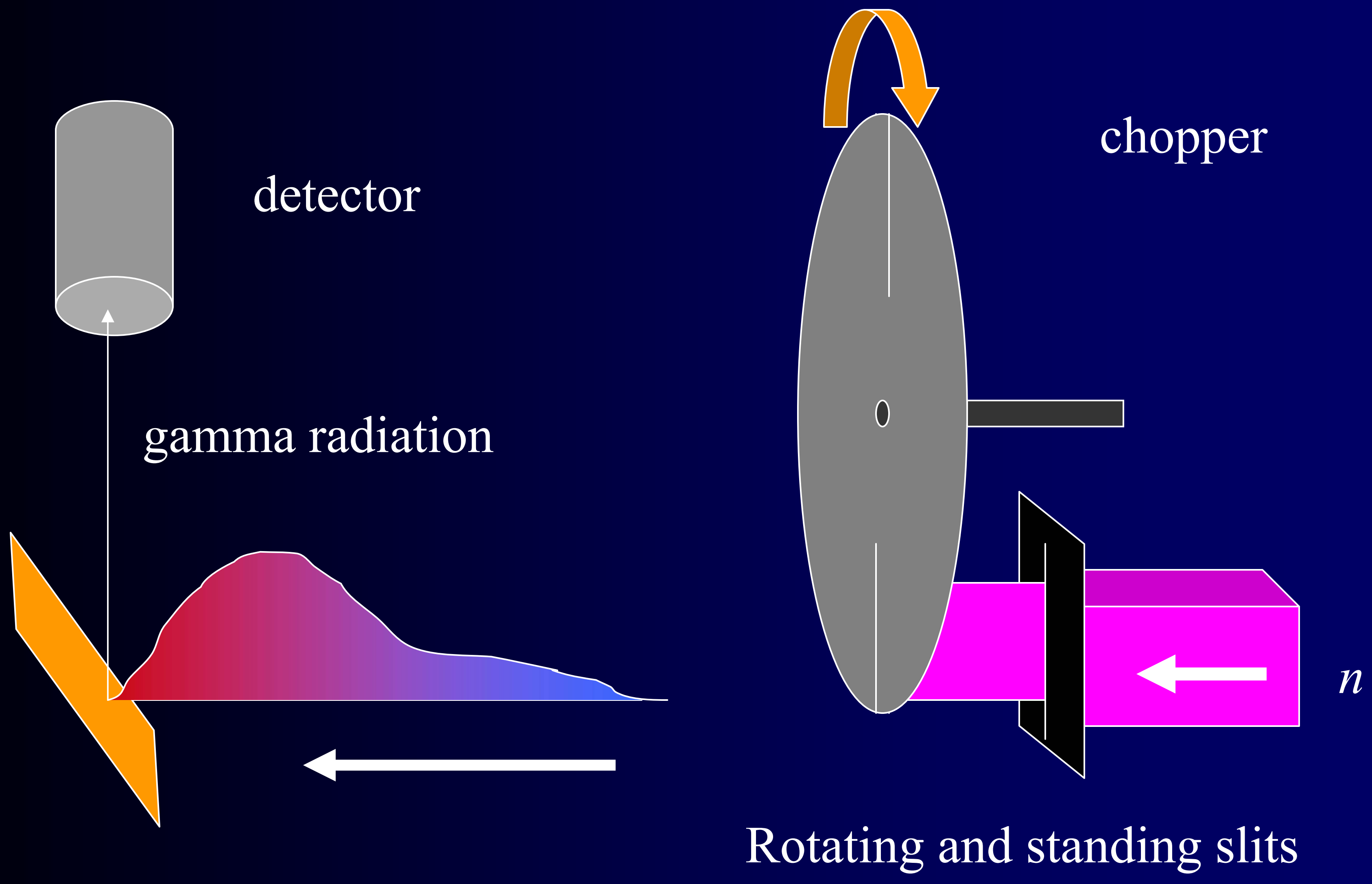
• Variable frequency : 3 – 100 Hz



Possible cyclic activation of short-lived isotopes

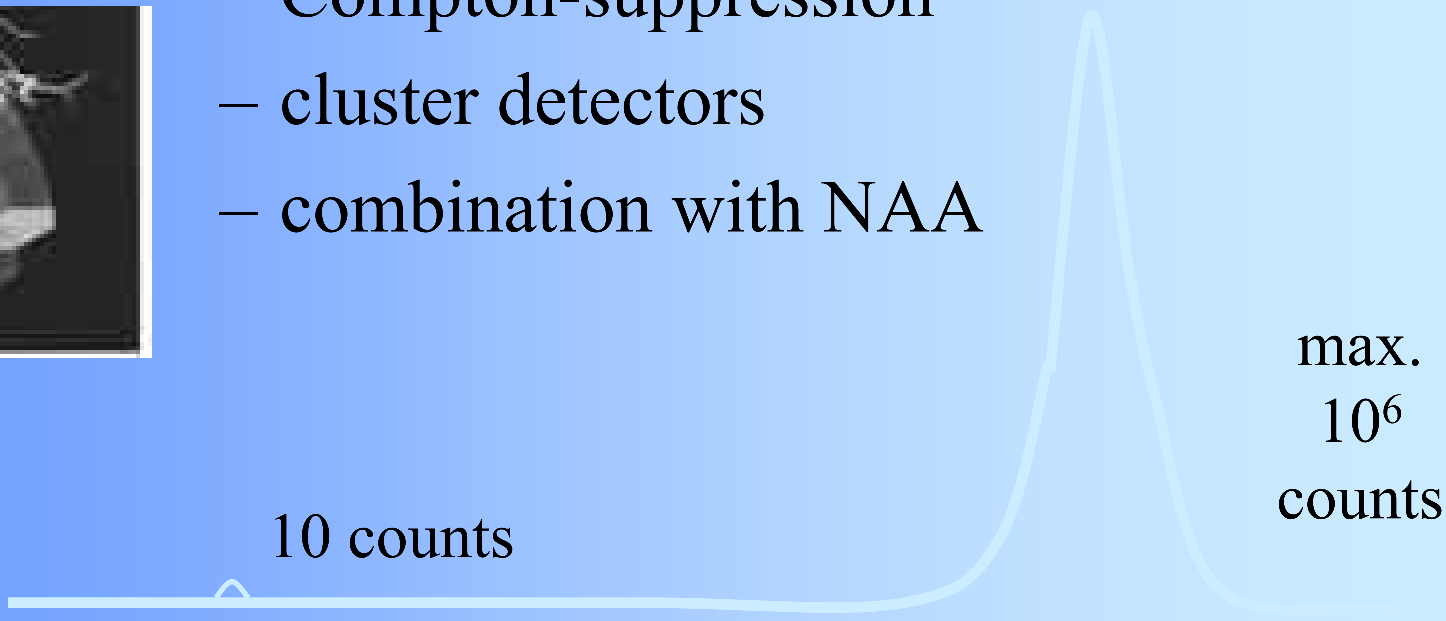
(Na, F, Sc, Ge, Pd, Ag, In, Er, Hf, W, etc.)

Time of flight



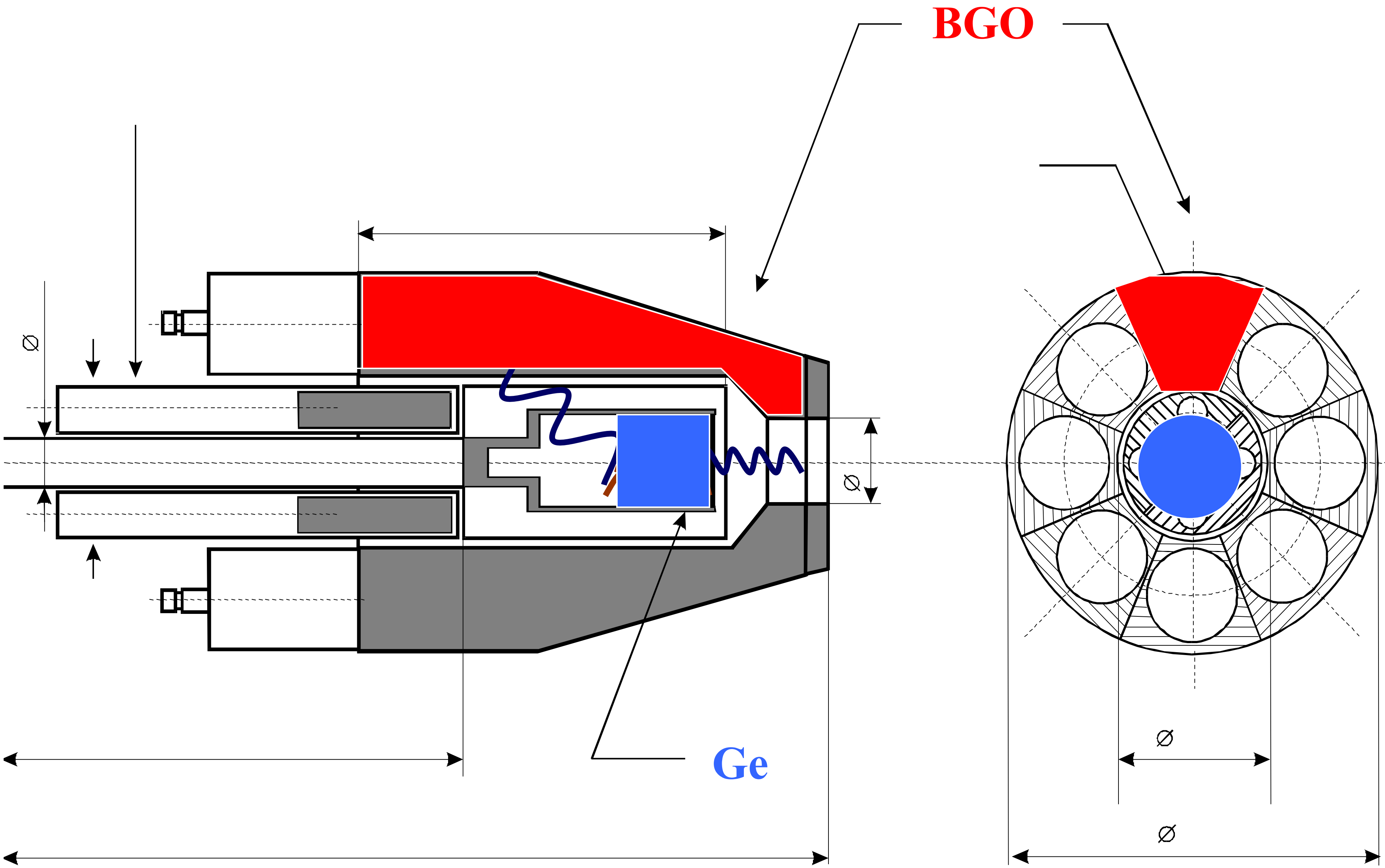
Gamma-ray spectroscopy

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

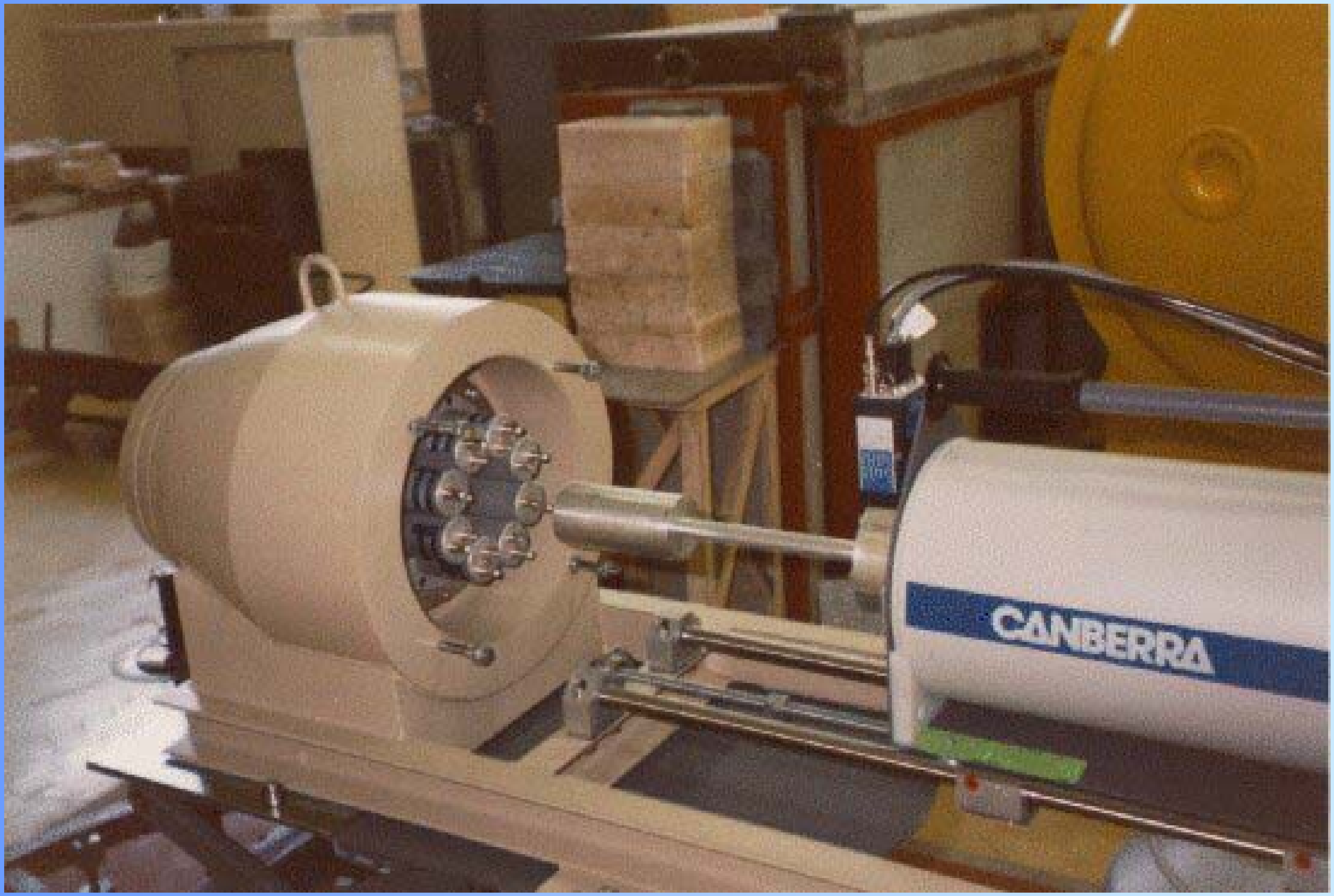


$$\mu_A/\mu_B = A_A/A_B = 10^{-5}$$

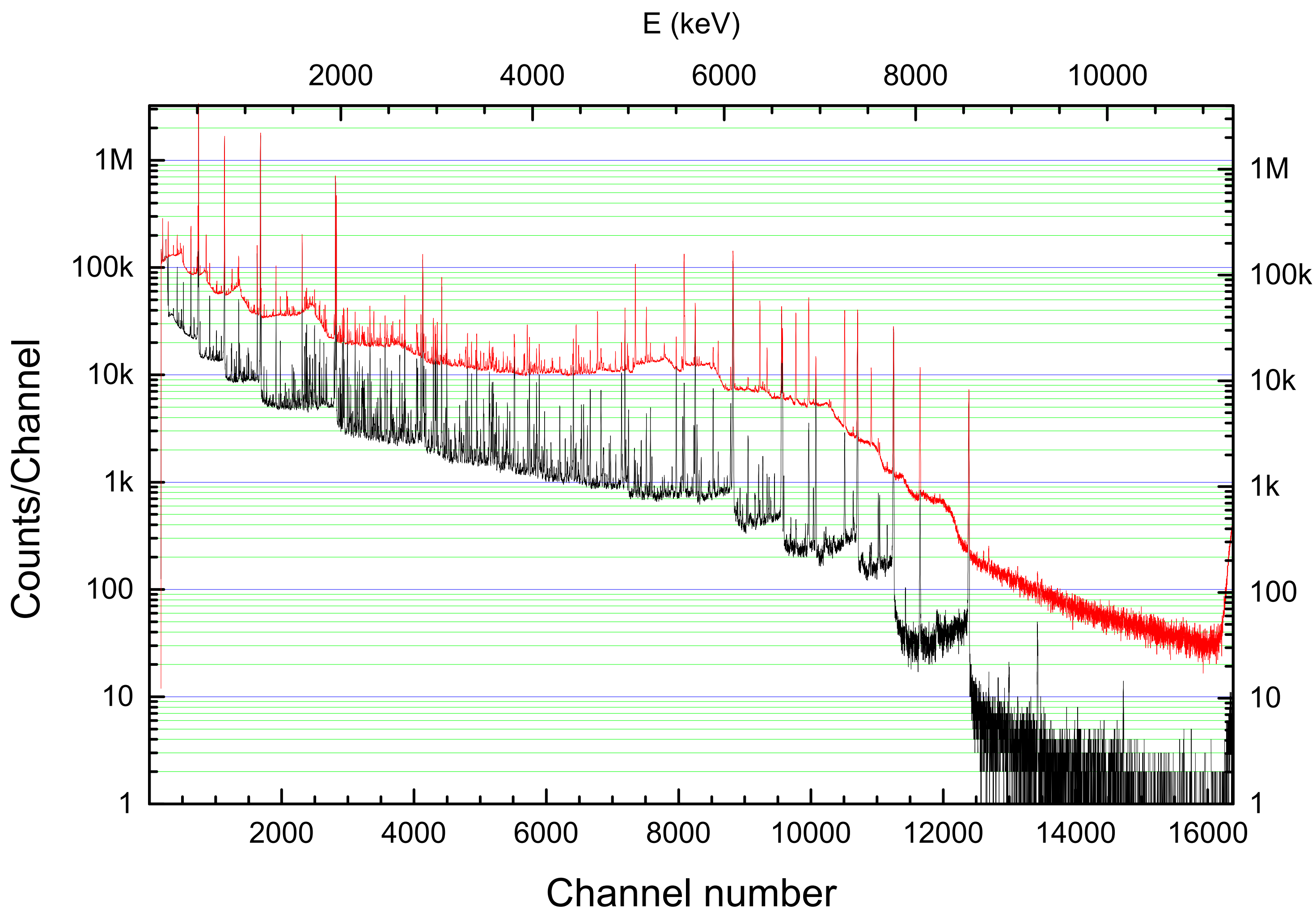
Compton-suppression system



Compton-suppressed detector



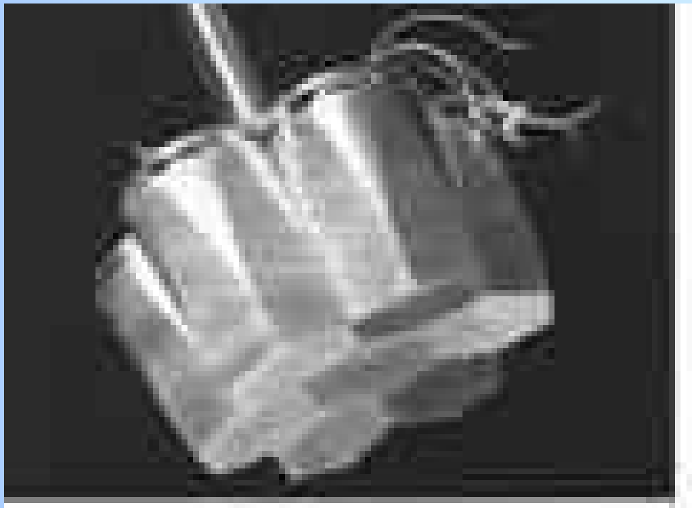
Background reduction via Compton-suppression



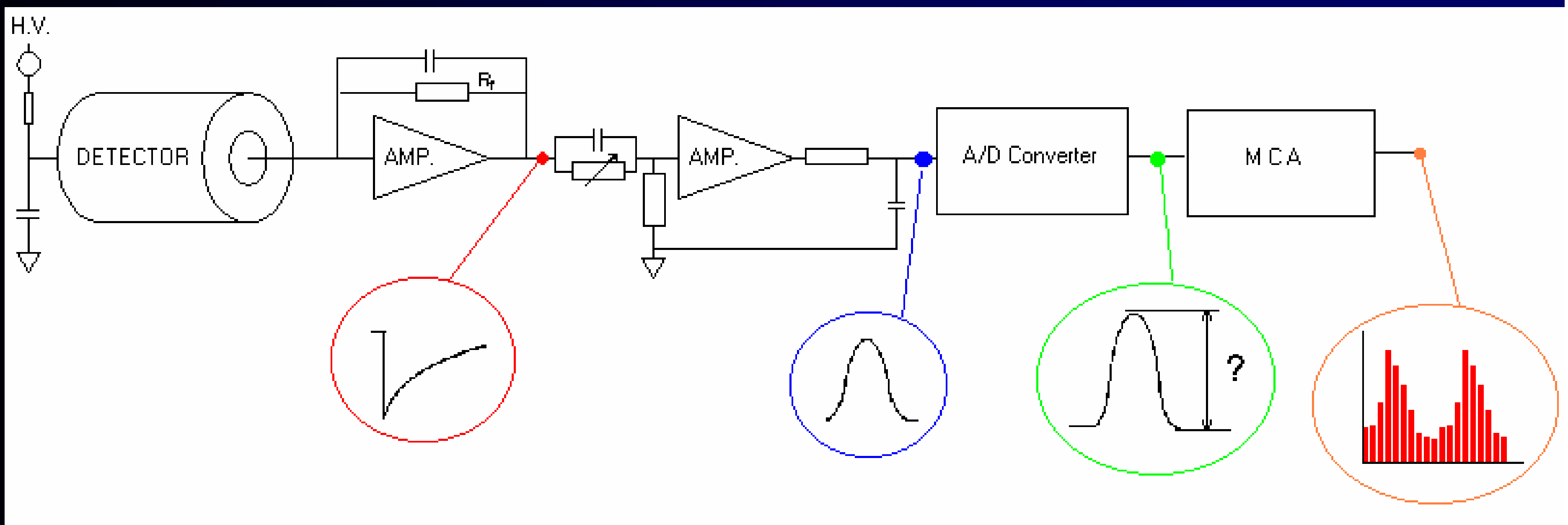
Composite detectors

Clover

Cluster

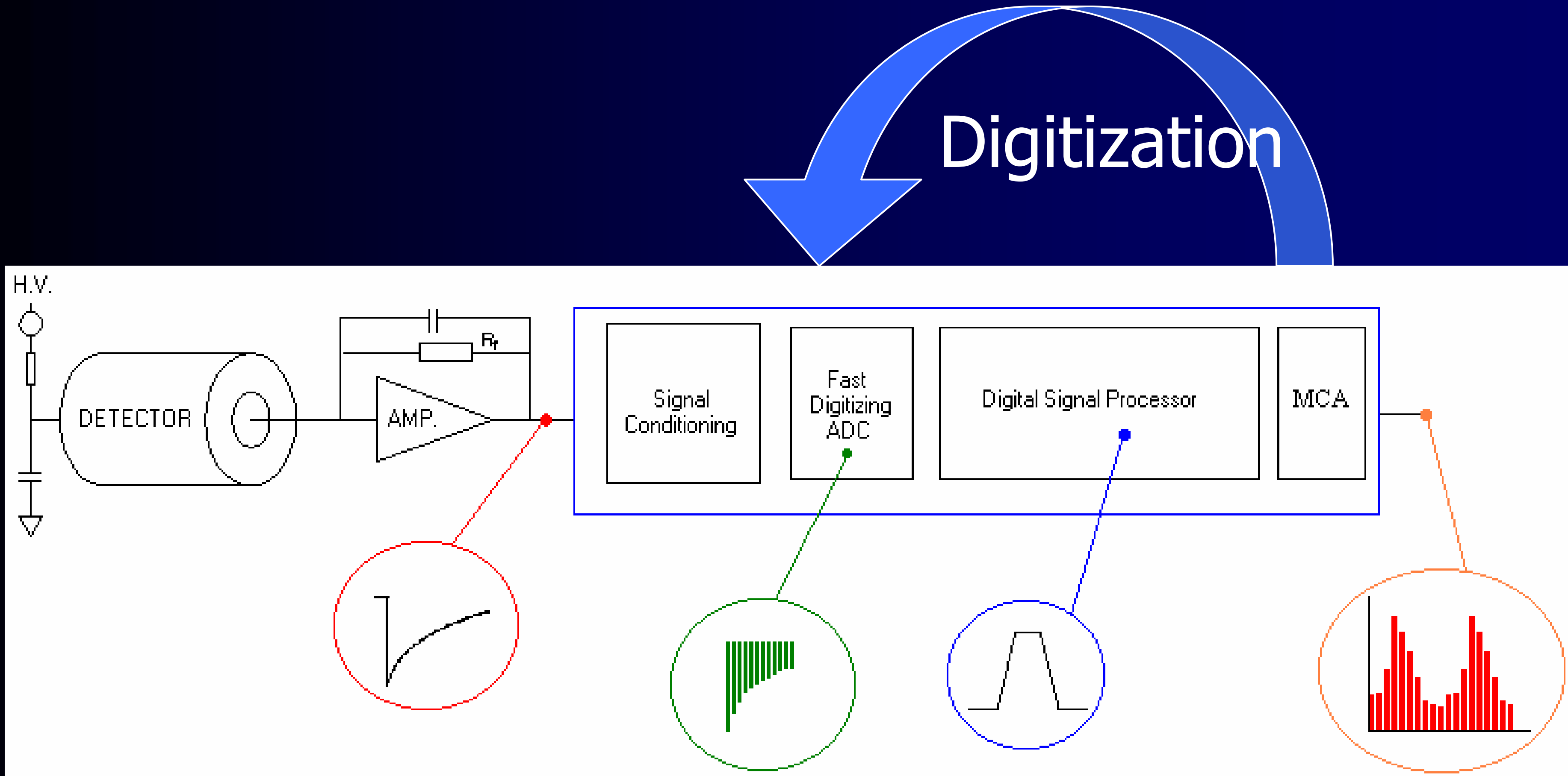


Analog signal processing chain



Preamplifier → Spectroscopy Amplifier → ADC → MCA

Digital signal processing

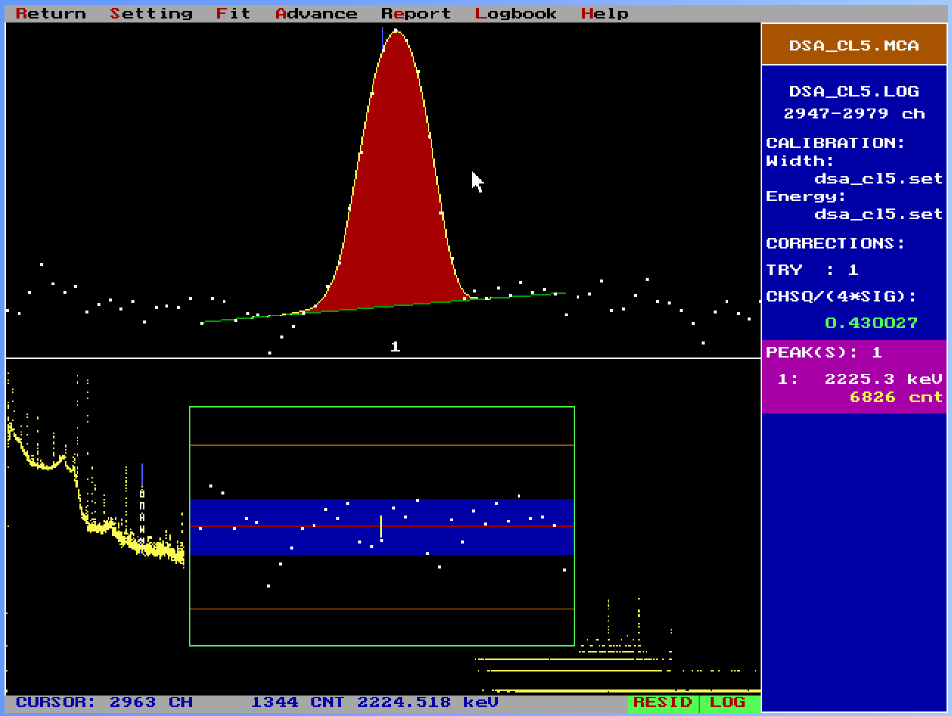


Preamplifier → Sampling ADC → DSP → MCA

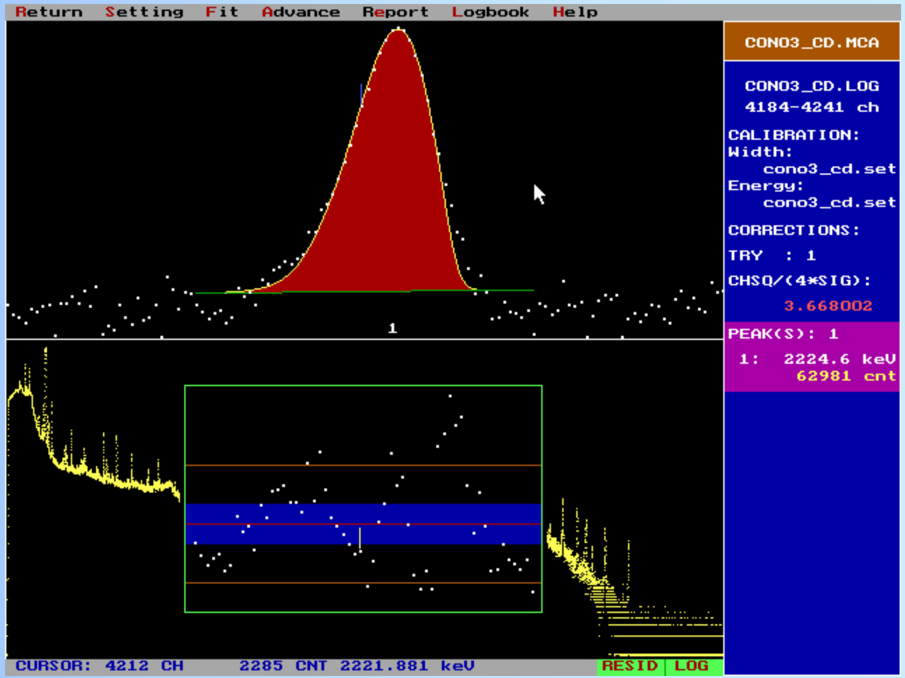
Parameters of a spectrometer

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

Peak shape

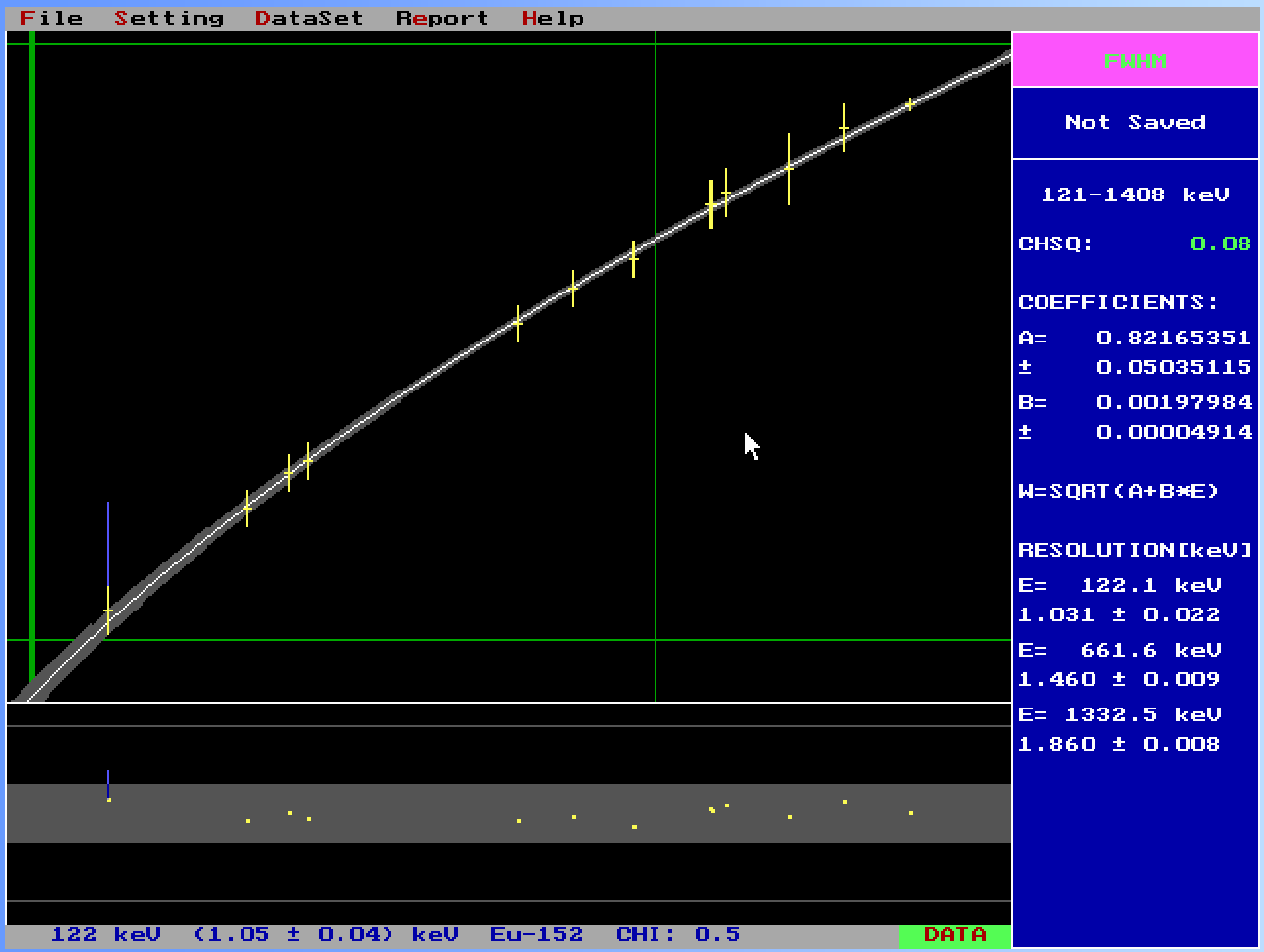


Optimum peak shape



Distorted peak shape

FWHM analysis



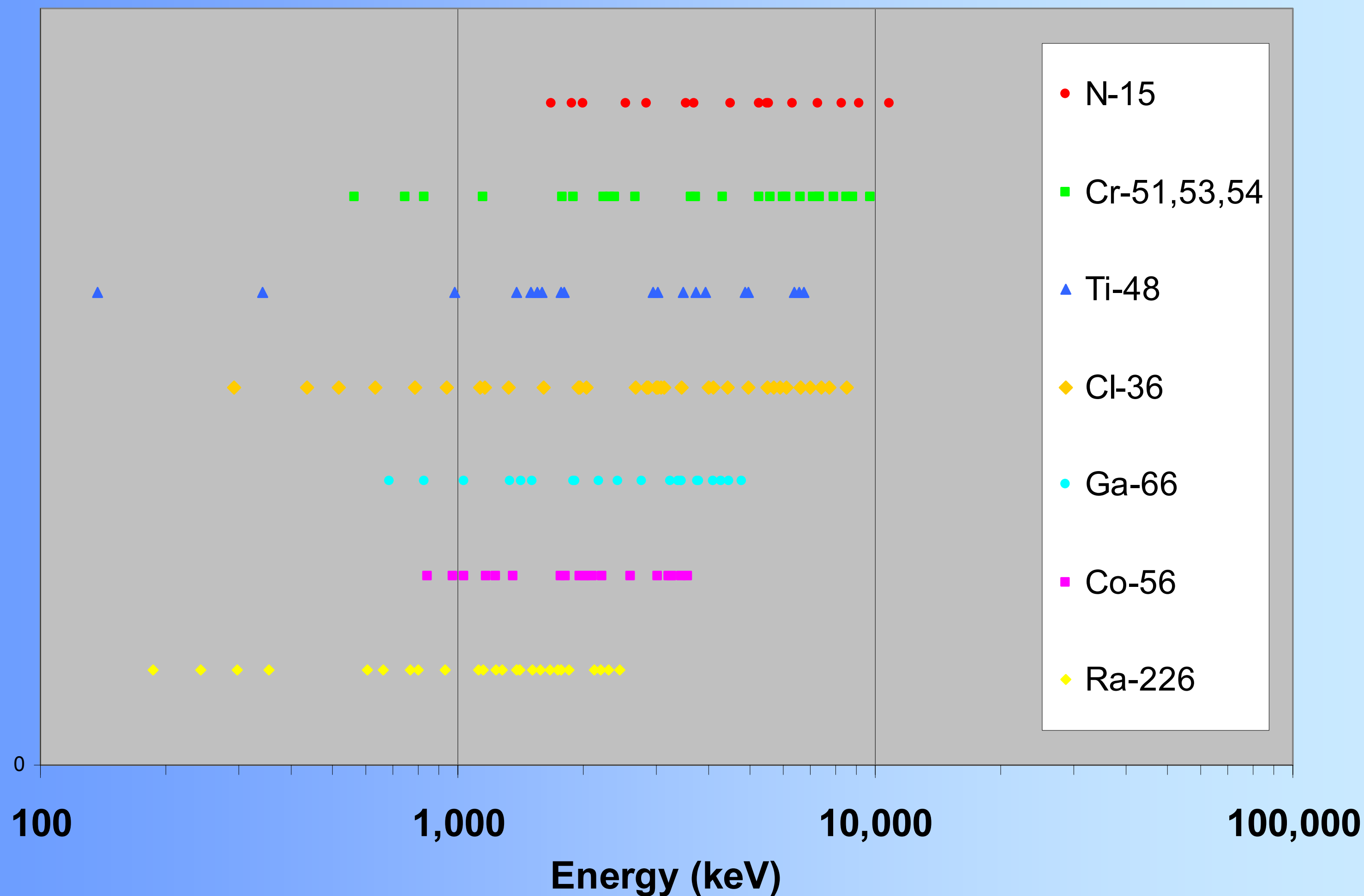
$$FWHM = \sqrt{a^2 + b^2 E}$$

$$if\ E \rightarrow 0:$$

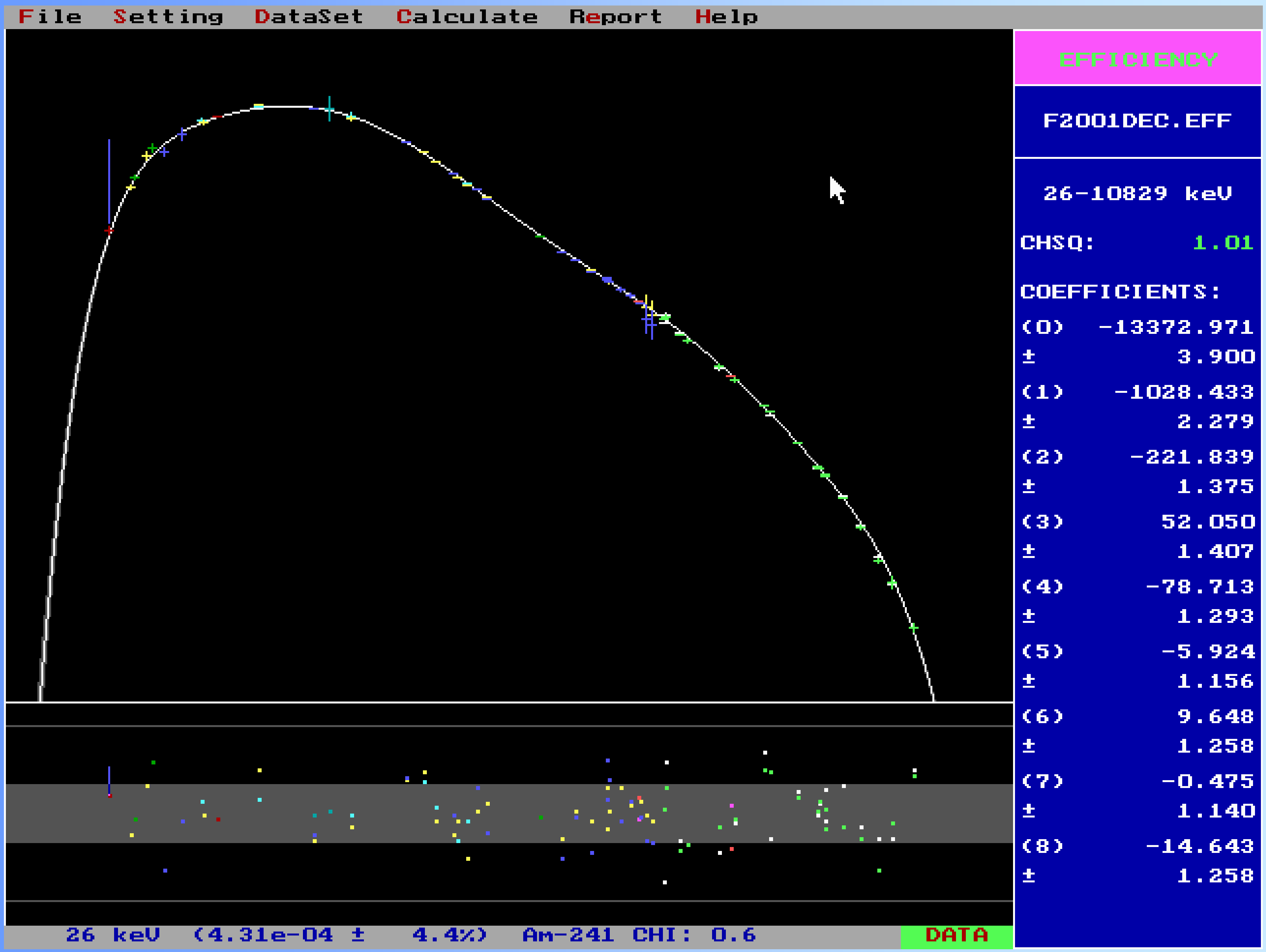
$$FWHM \rightarrow \boldsymbol{a}$$

a : peak broadening effect of electronics in keV

Range of calibration sources

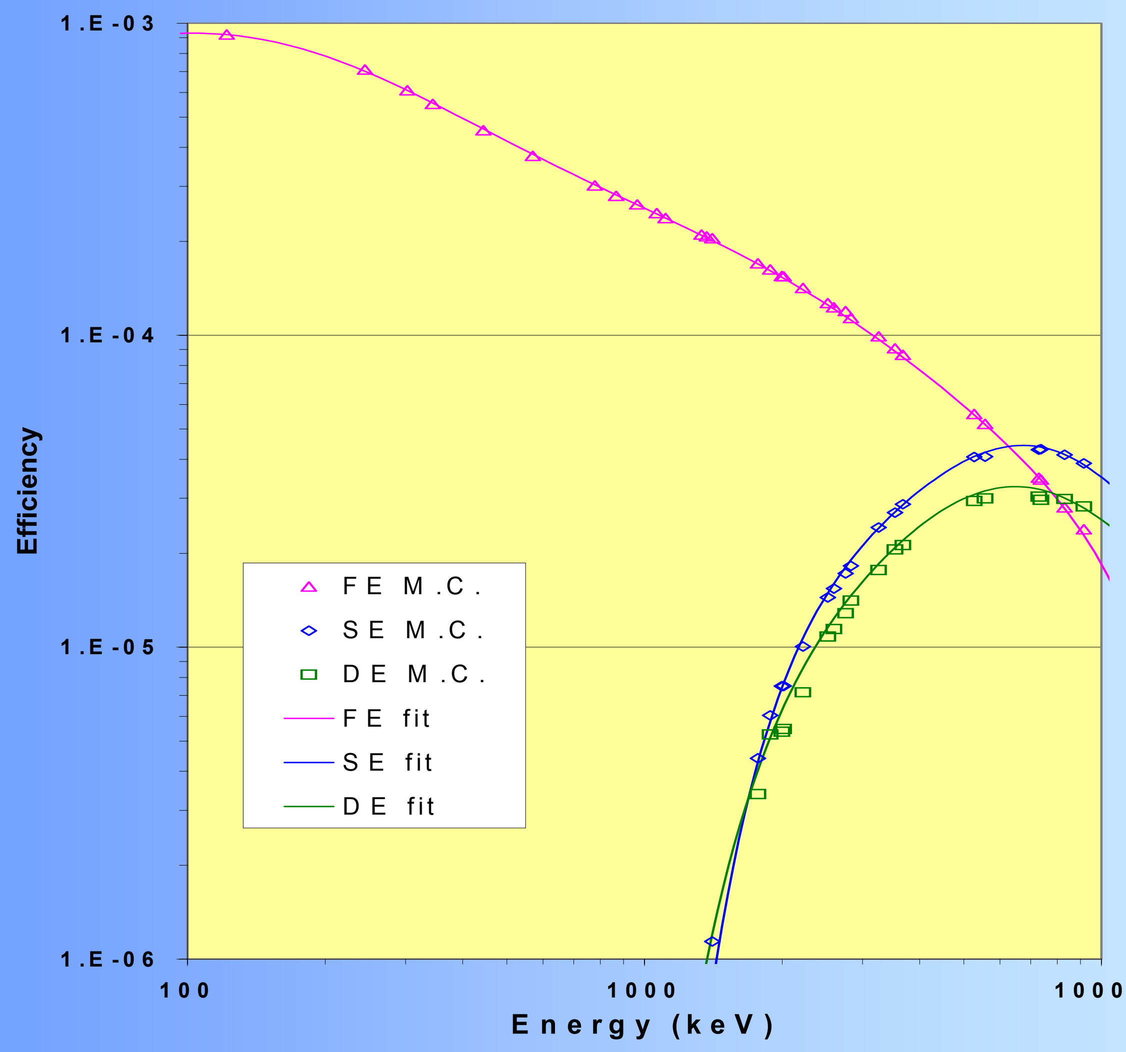


Efficiency calibration

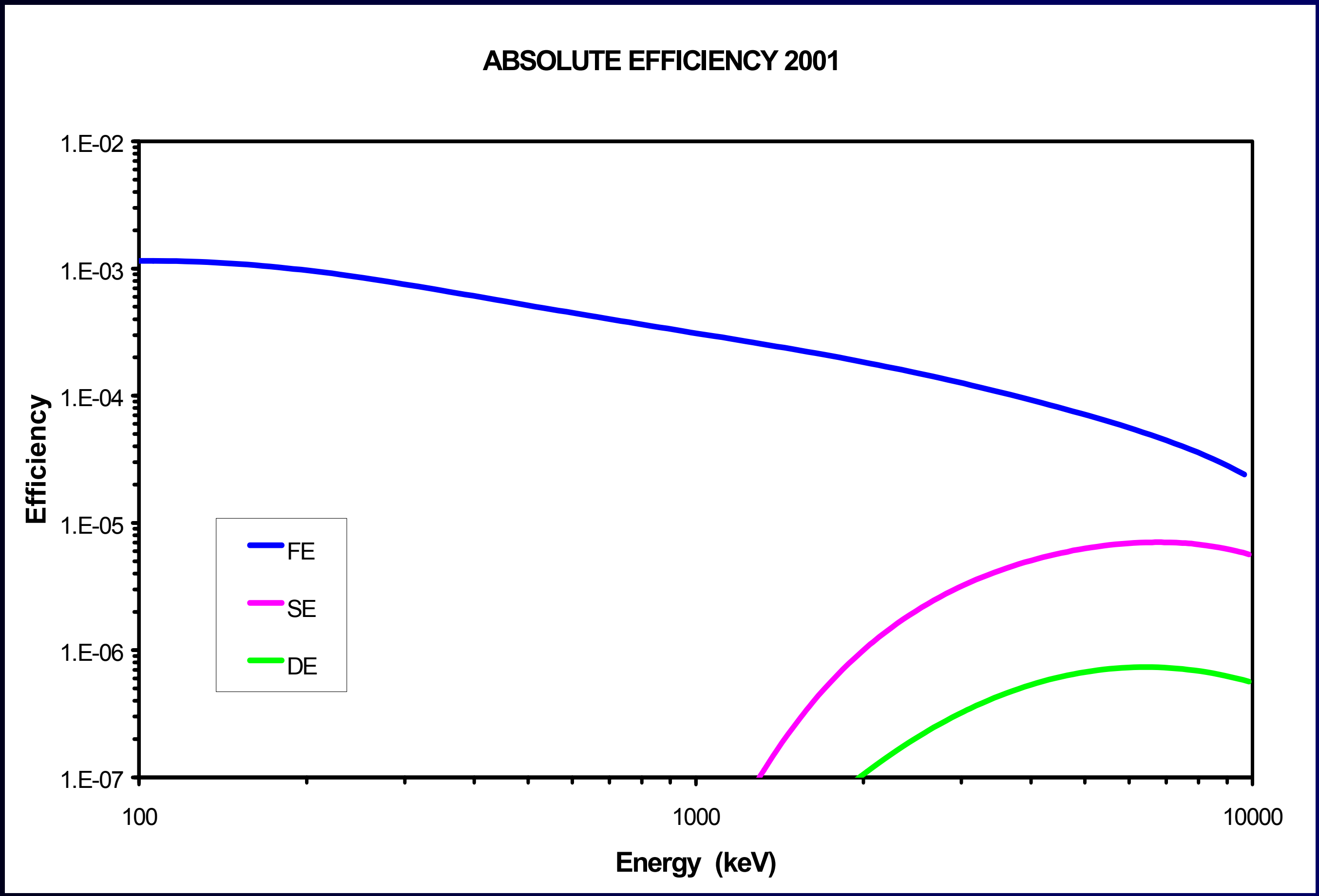


Monte Carlo simulation with CYLTRAN vs. Budapest efficiency

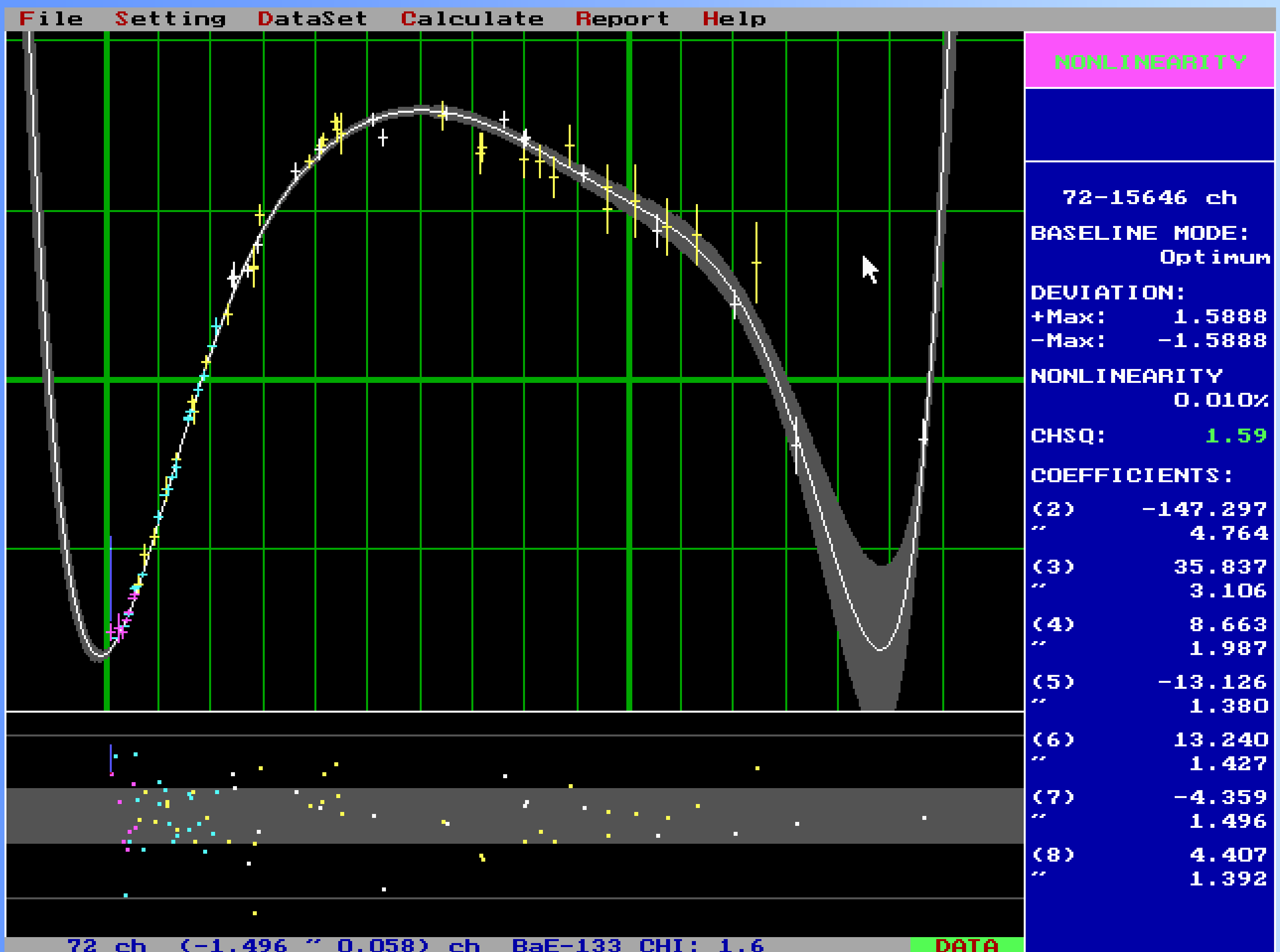
R. G. Helmer (INEEL)



Comparison of efficiencies
Compton-suppressed mode

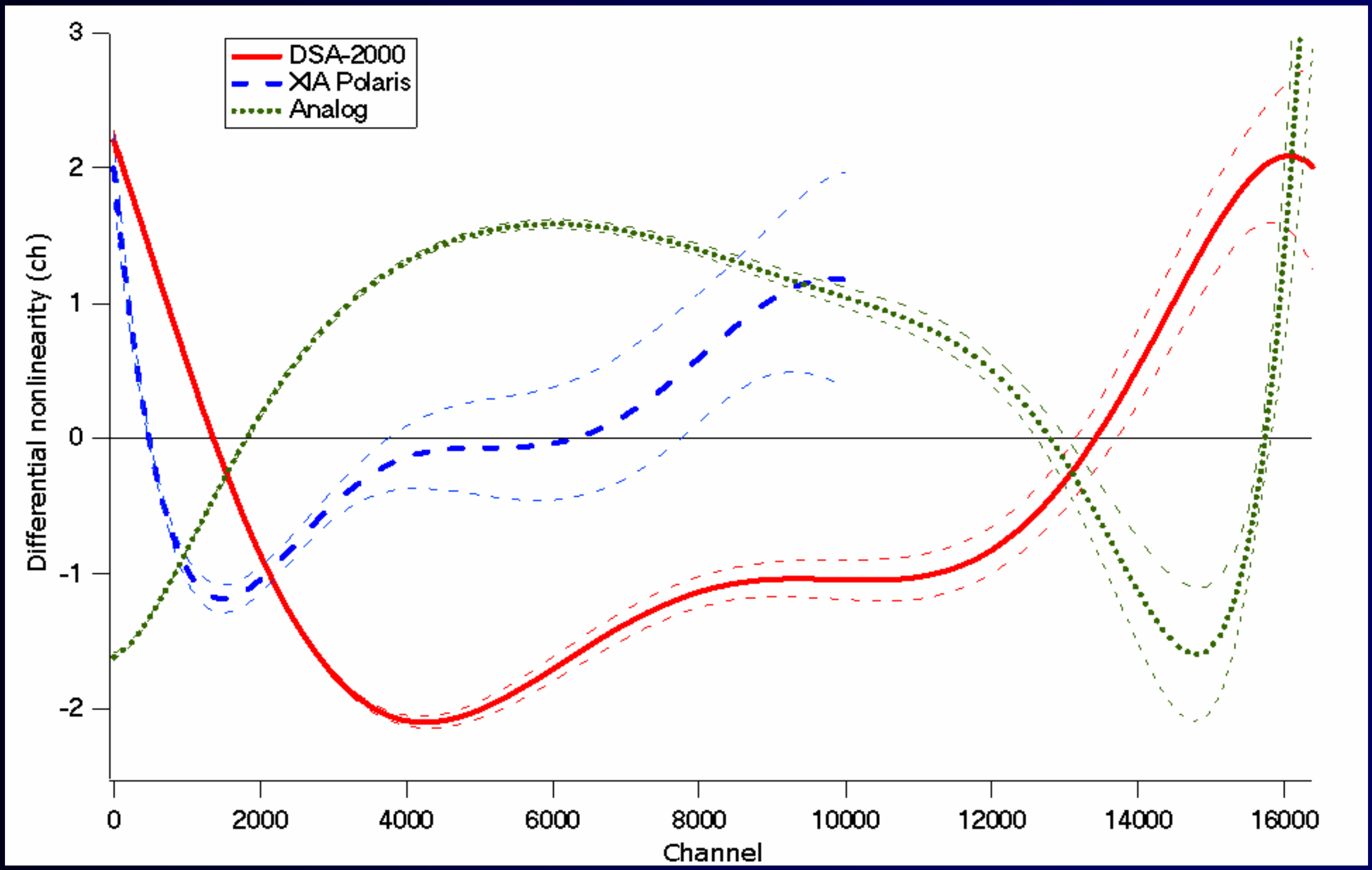


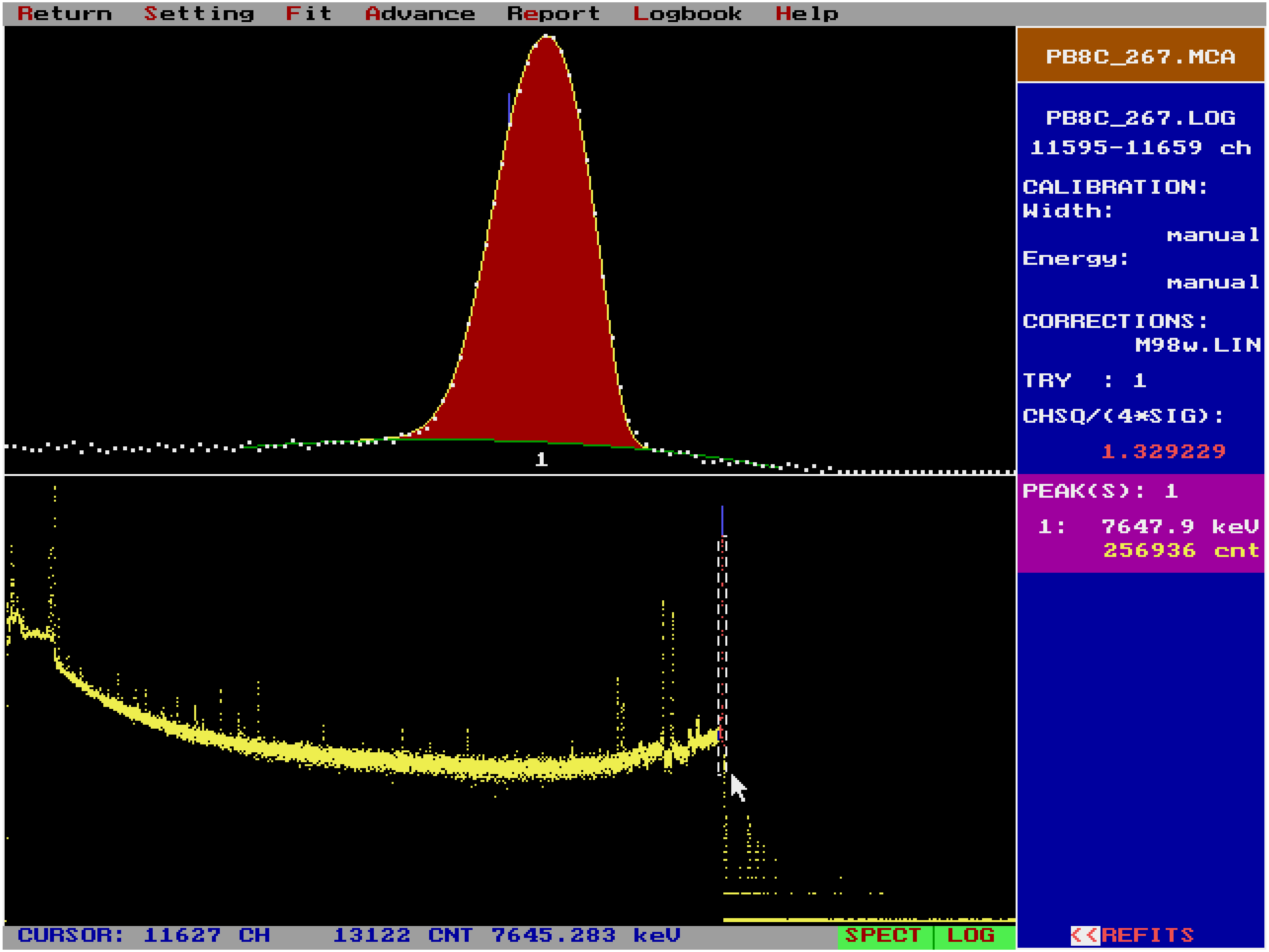
Nonlinearity analysis

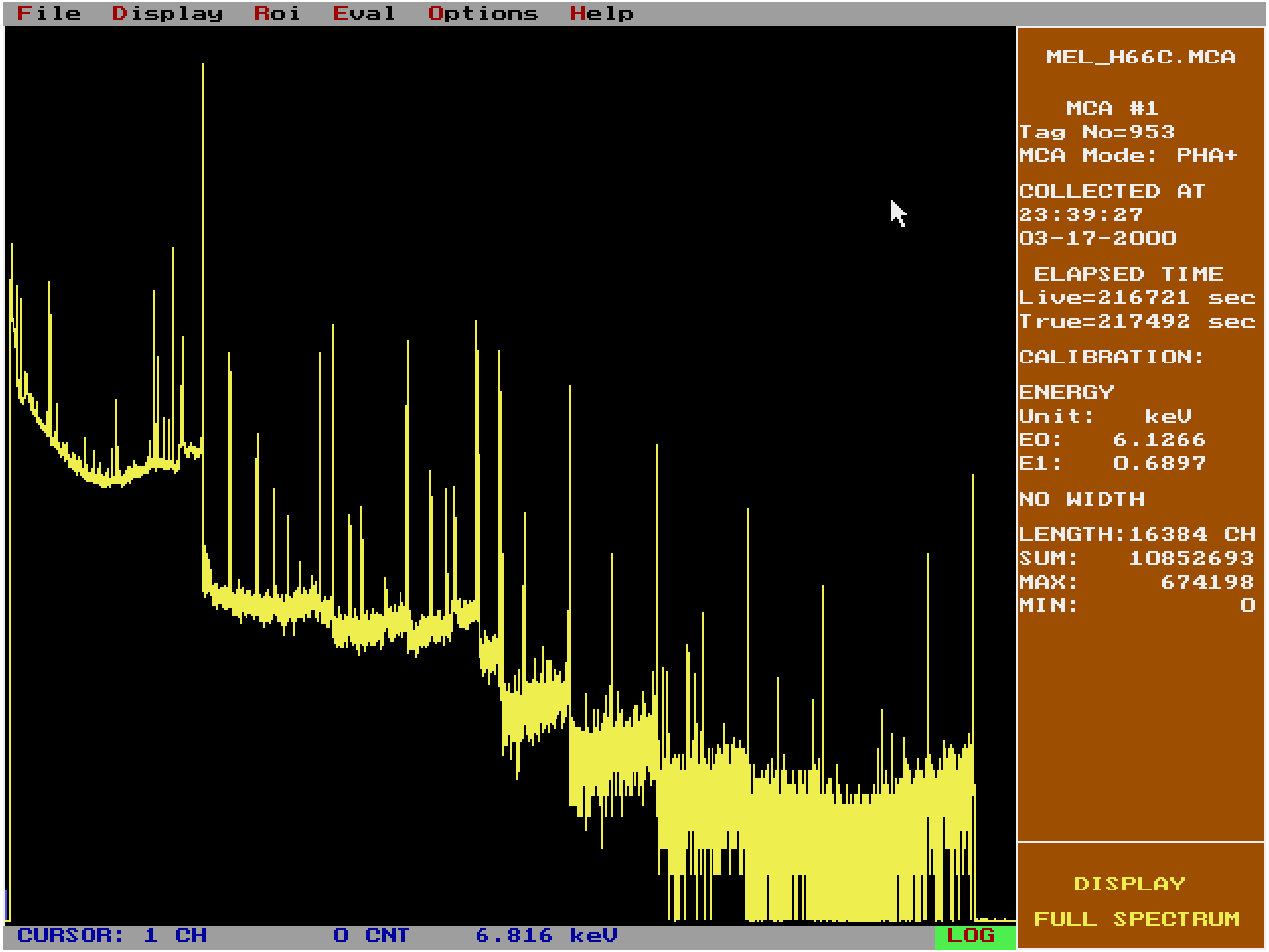


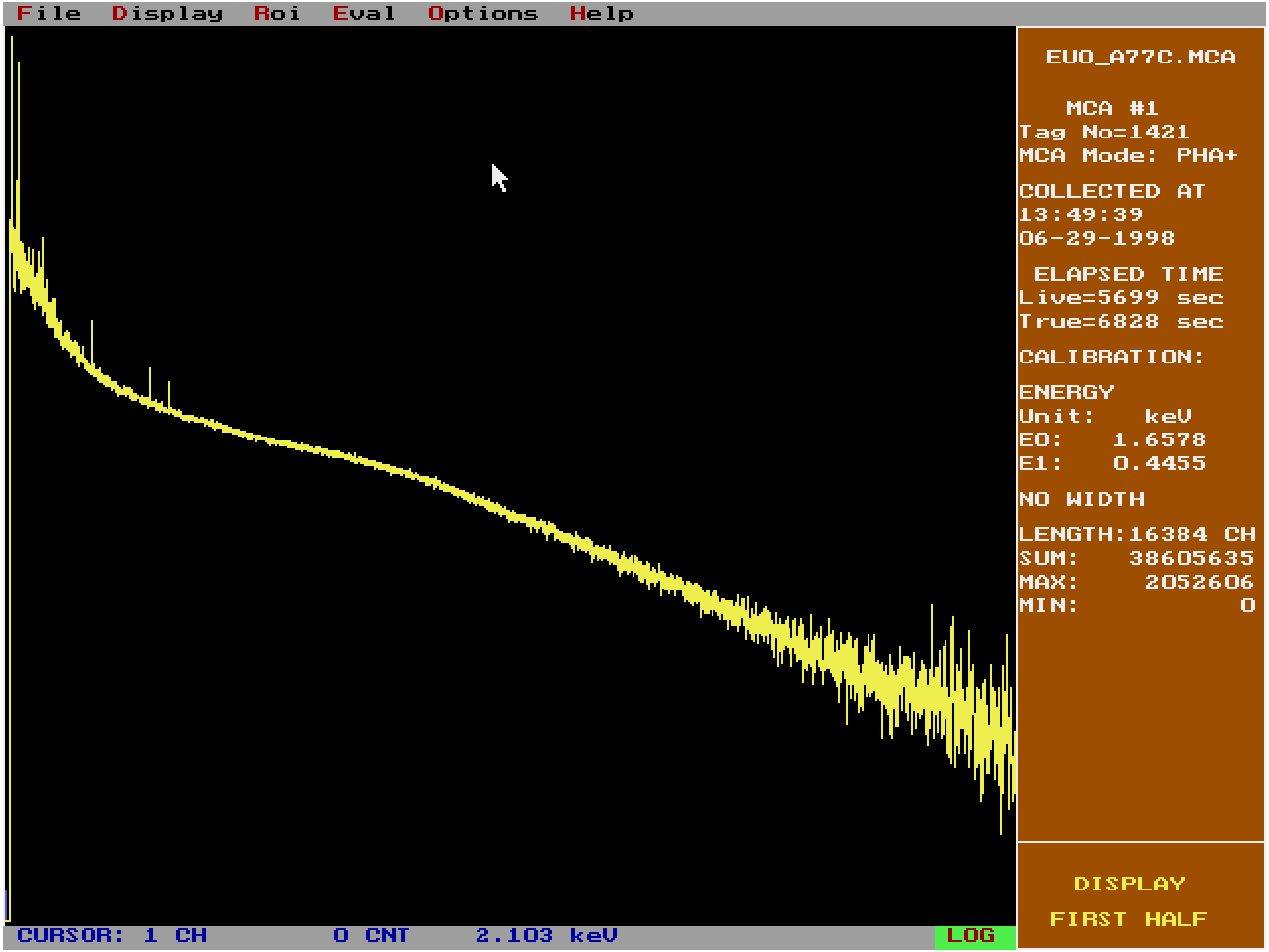
- Measured with ^{152}Eu source and Cl(n,γ)
- 16k channels
- Nonlinear fit of orthogonal olynomials
- Centred around zero

Nonlinearity




Return Setting Fit Advance Report Logbook Help
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
1
CURSOR: 11627 CH 13122 CNT 7645.283 keV
SPECT LOG
<<REFITS


File Display Roi Eval Options Help
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
CURSOR: 1 CH
0 CNT
6.816 keV
LOG


File Display Roi Eval Options Help
EUO_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
EO: 1.6578
E1: 0.4455
NO WIDTH
LENGTH:16384 CH
SUM: 38605635
MAX: 2052606
MIN: 0
DISPLAY
FIRST HALF
CURSOR: 1 CH 0 CNT 2.103 keV
LOG

4. Special techniques

Dynamic range

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

e.g. 1 mg Mn in 1g H_2O (~0.1 g H)

_____1 mg H_2O (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

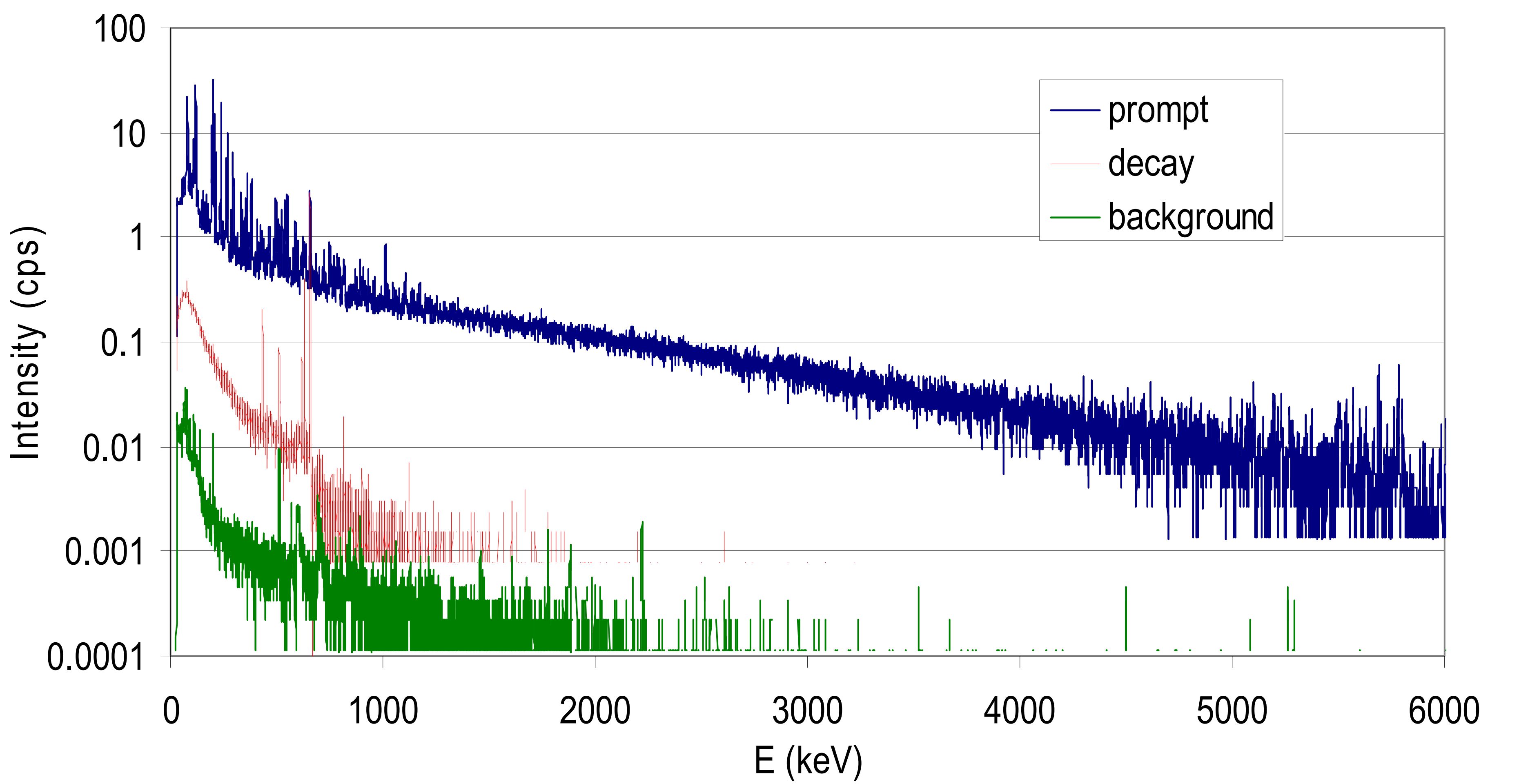
Usual PGAA spectrum

- **Beam closed**

only decay gamma rays

cyclic NAA spectrum

Spectra



100
10
1
0.1
0.01
0.001
0.0001
Intensity (cps)
0
1000
2000
3000
4000
5000
6000
E (keV)
prompt
decay
background

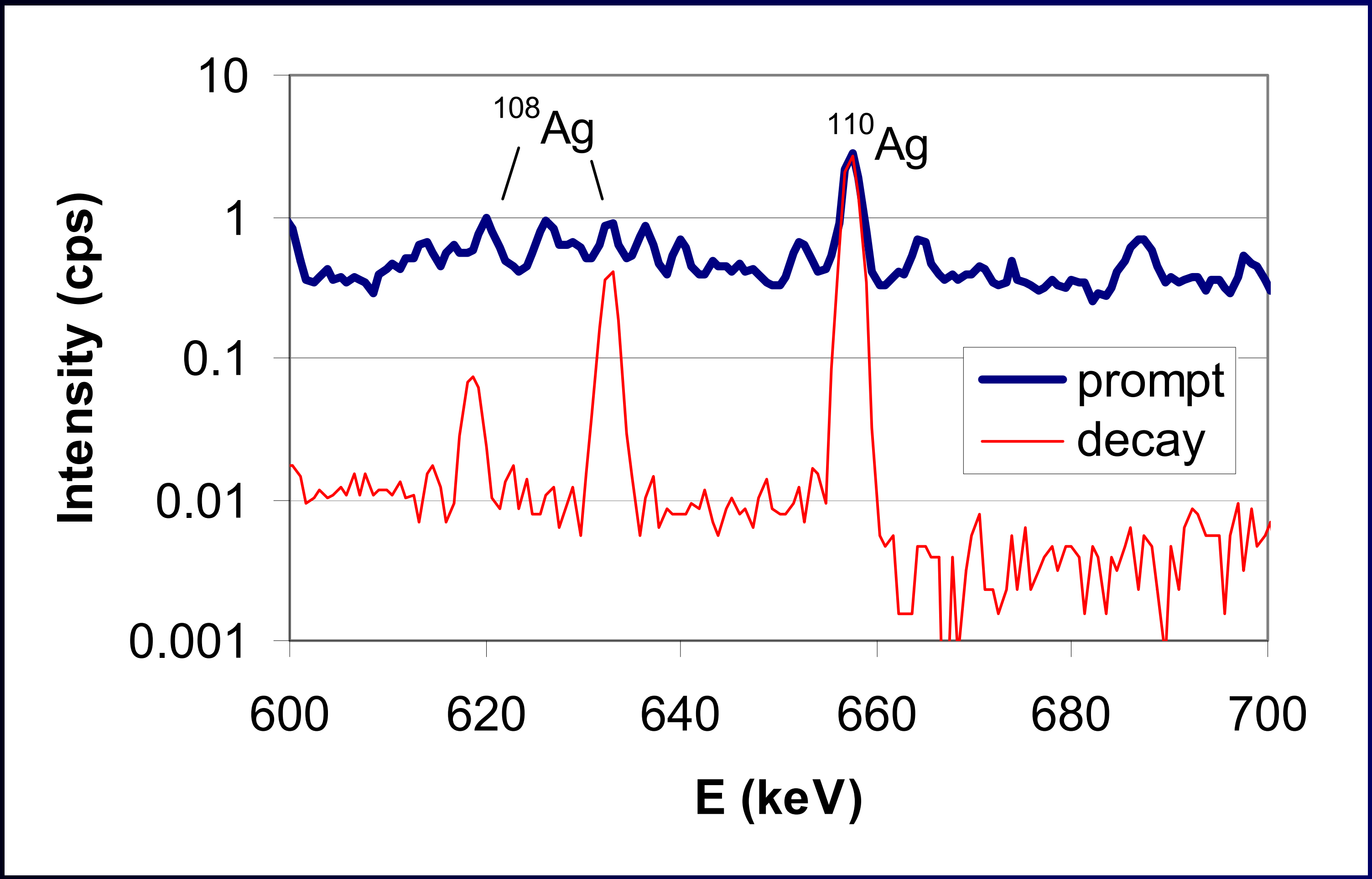
Elements with decay lines

< 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	Backgound (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



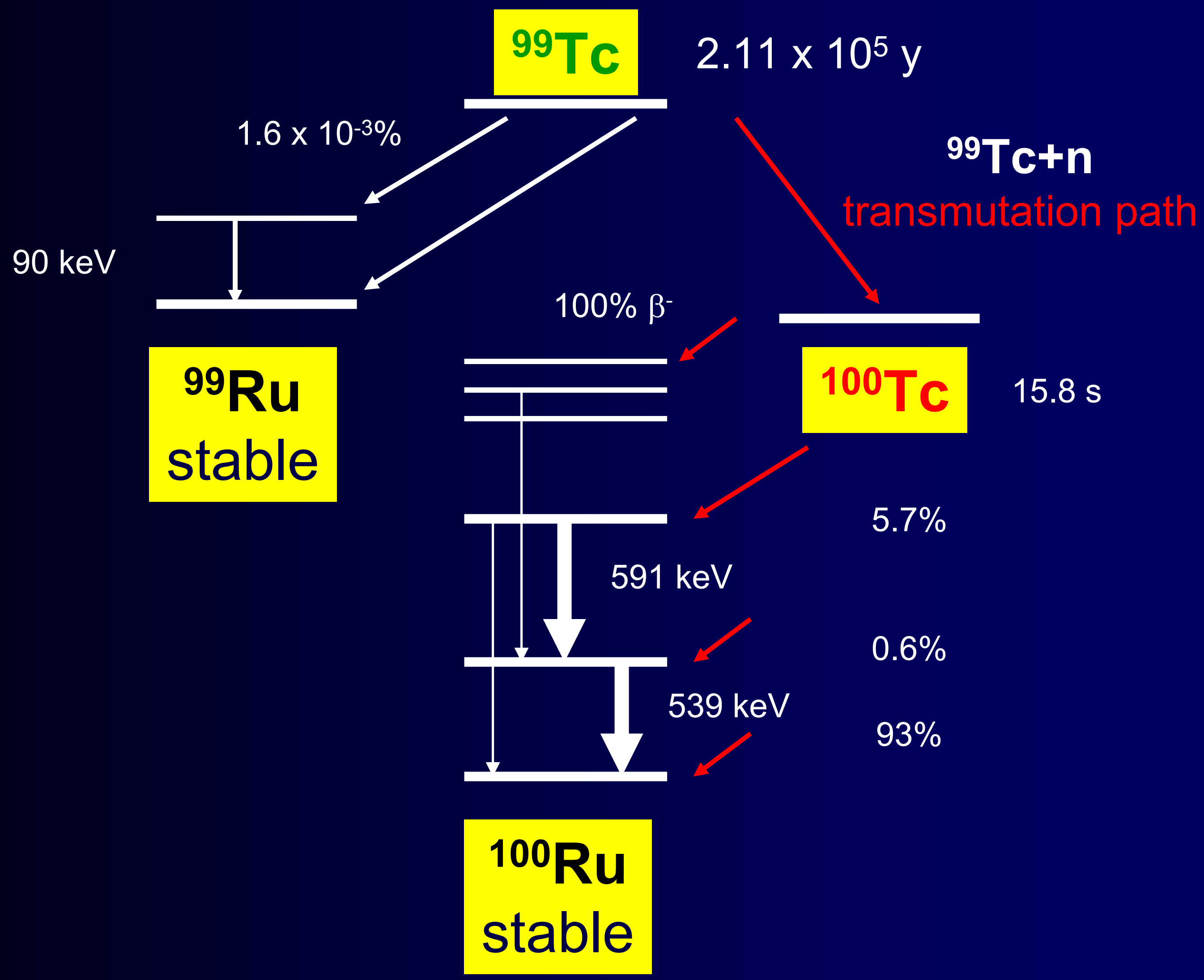
Mea-sure-ments

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

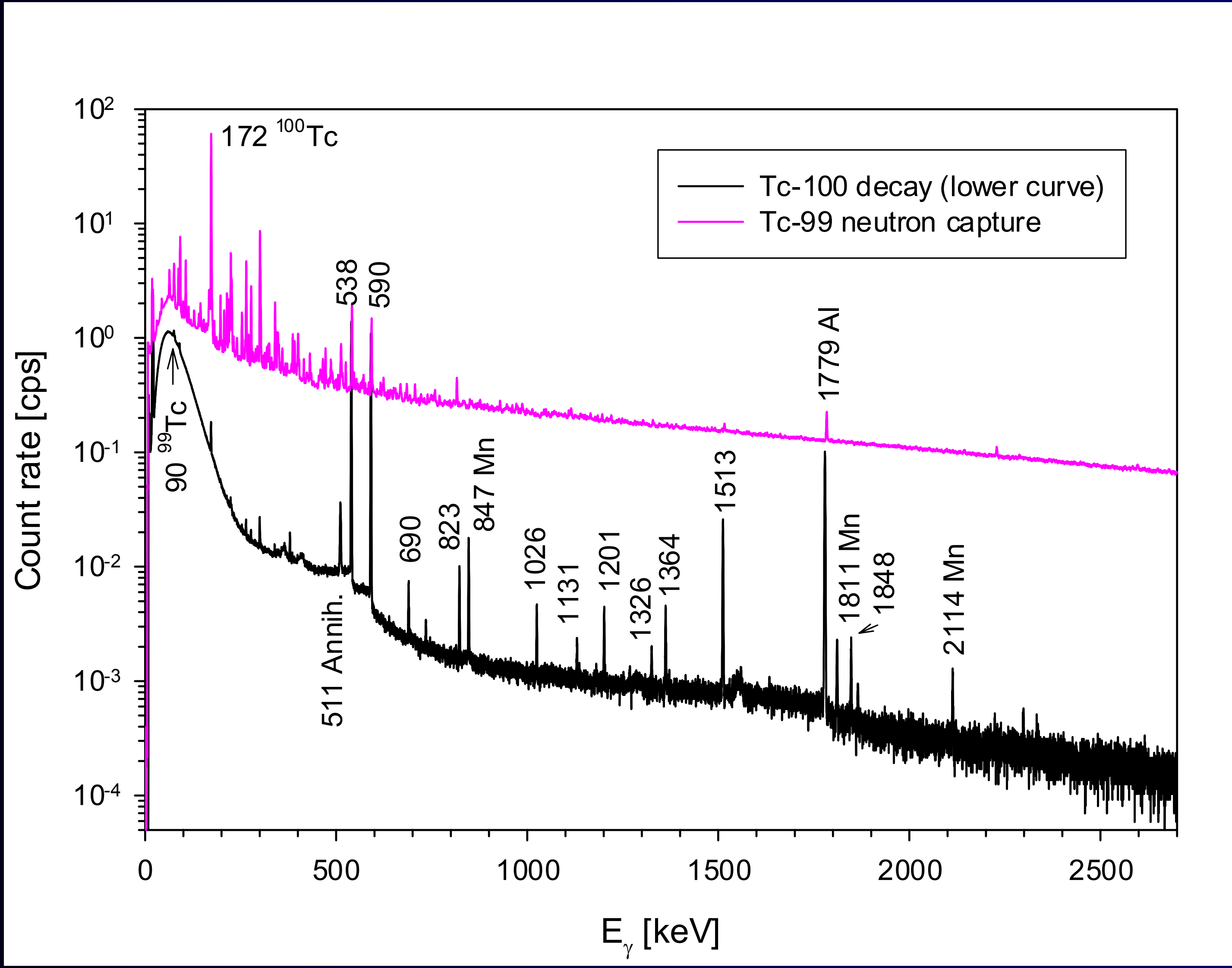
Element	Half-life	Energy (keV)	decay line σ_γ (barn)	prompt line σ_γ (barn)
F	11.16 s	1633	0.0093(3)	–
Al	2.24 m	1779	0.233(4)	<0.005
Sc	18.75 s	143	4.88(10)	<0.13
V	3.75 m	1434	5.20(10)	<0.3
Cu	5.12 m	1039	0.0600(12)	<0.0023
Ag	24.6 s	658	1.93(4)	<0.08
In	2.18 s	163	15.8(8)	<1.1
Er	2.27 s	208	2.15(9)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

Scheme of transmuting ^{99}Tc



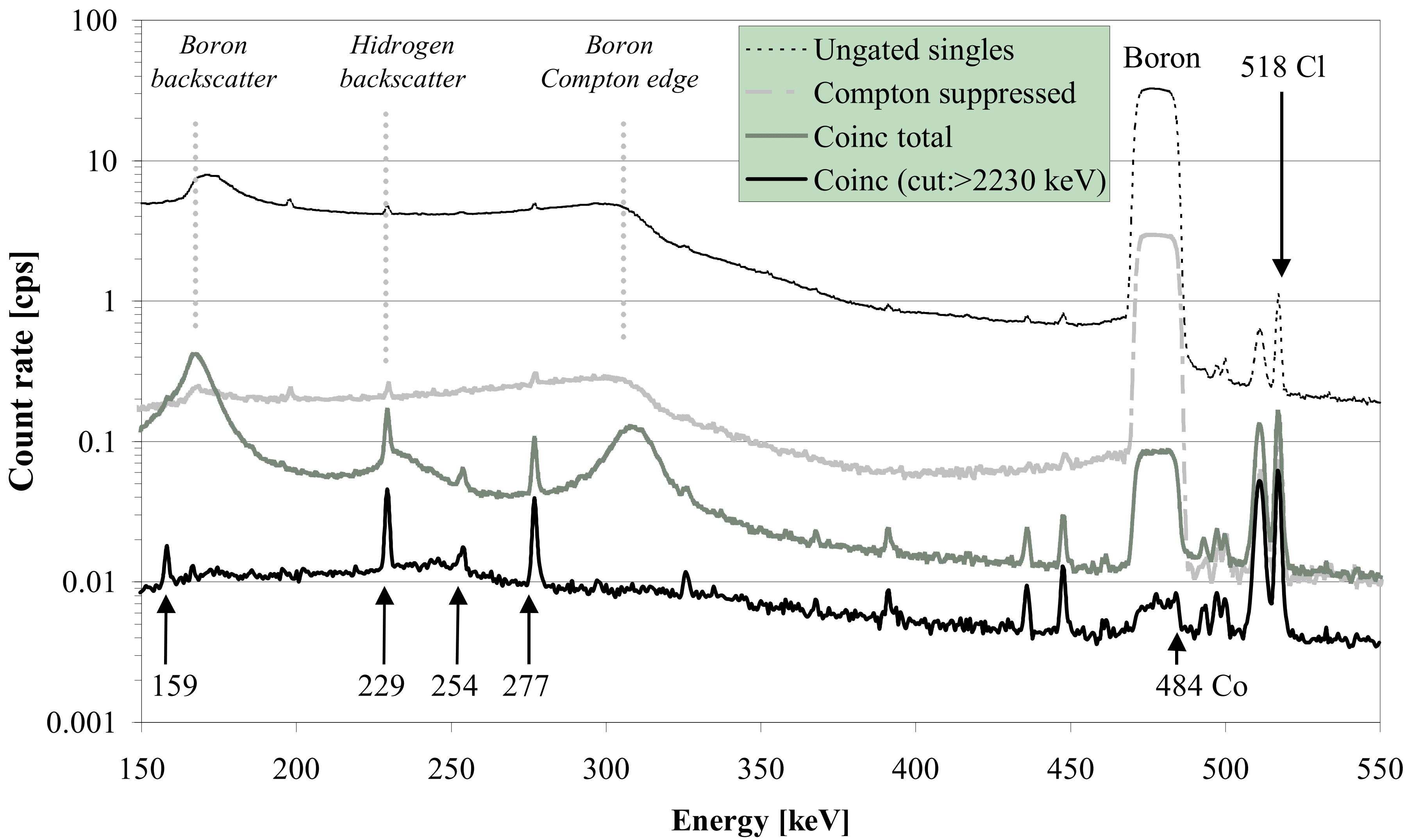
PGAA on a radioactive target

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



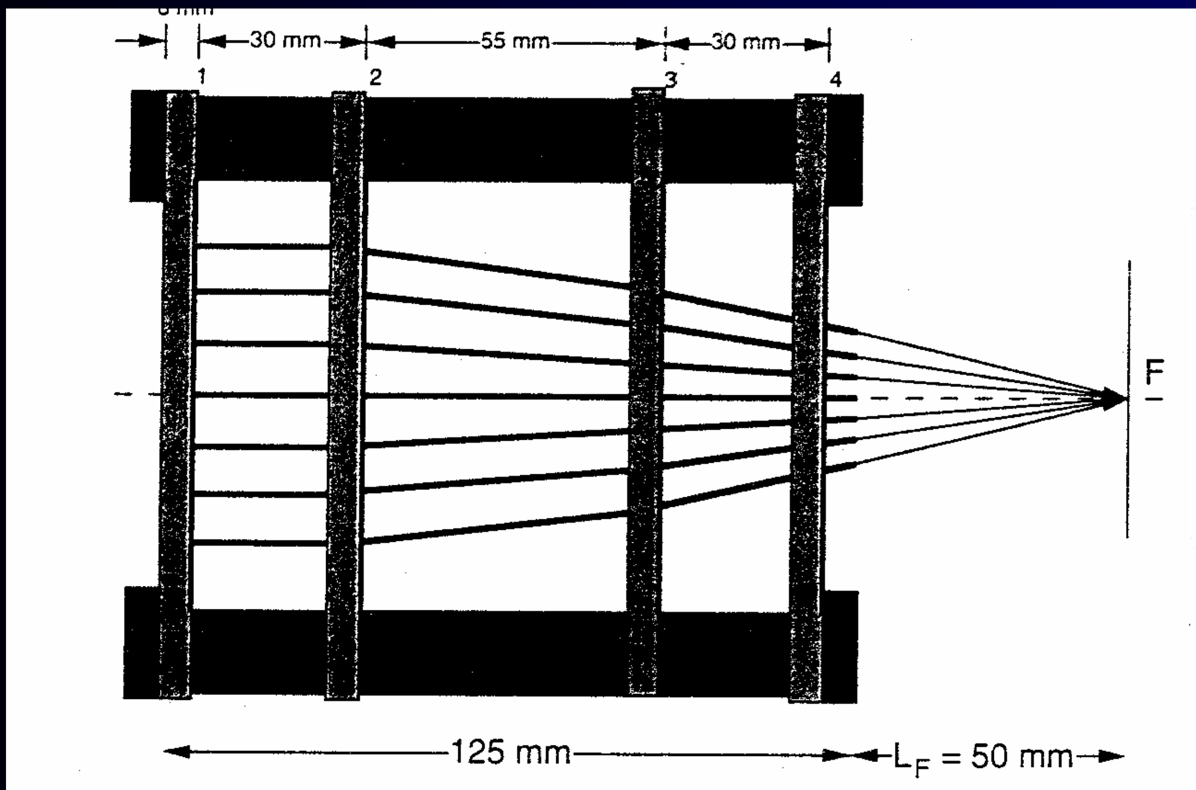
Increase of PGAA sensitivity by coincidence

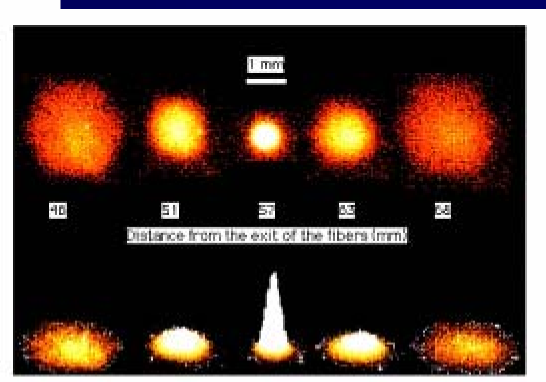
9.35 mg $CoCl_2(2H_2O)$ and 22.1 mg of H_3BO_3 / ml



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