



list
cea tech

FOCUS ON MEASUREMENTS AND CORRECTION FACTORS FOR LOW-ENERGY X- AND GAMMA-RAYS



GammaRay X Webinar – October 20, 2021
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A FEW WORDS ABOUT LNHB

Laboratoire National
Henri Becquerel



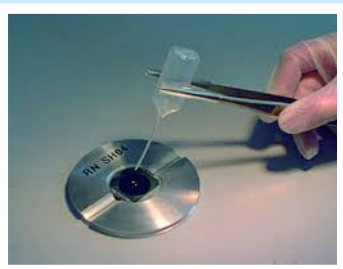
Atomic and Alternative Energy Commission (CEA)
French Metrology Institute (LNE)

In charge of radioactivity standards (Bq, Gy)

Activity laboratory (Bq): 21 permanent staff (4 PhDs – 1 post-doc)

RADIOACTIVITY METROLOGY AT LNHB

- Goal: Develop and disseminate national radioactivity standards adapted to users' needs
- Means: measurement equipment's, radiochemistry, sources preparation, Monte Carlo simulation, redundant methods
- Proficiencies: specific know-how, experience, tradition



Sources preparation



Neutron flux



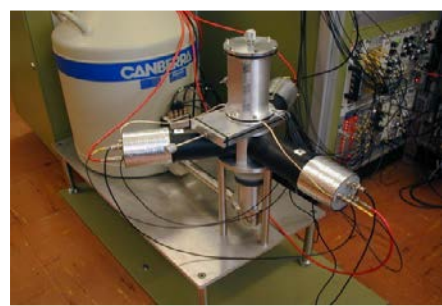
Radioactive gaz standards



Monochromatic X-ray source



TDCR counter
(Liquid Scintillation Counting)



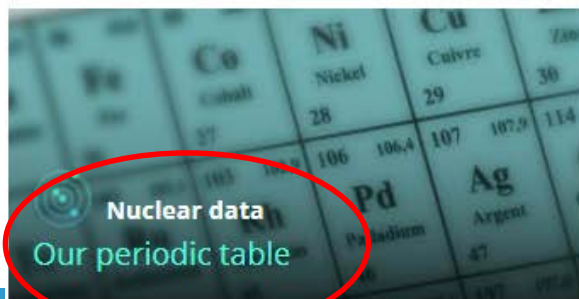
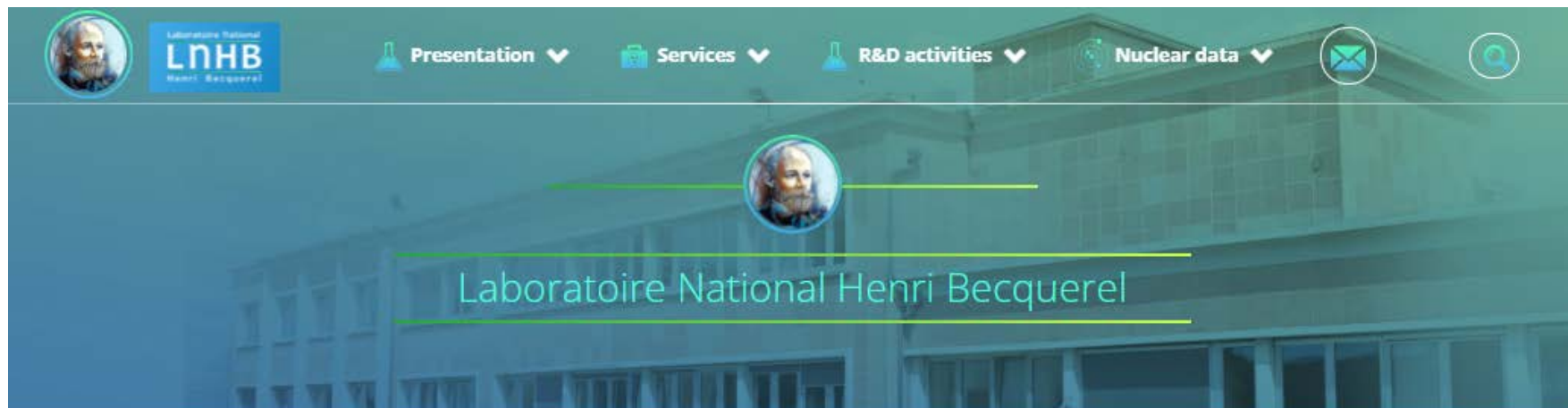
4π β - γ coincidence counting



Cryogenic detectors

Development of software

BETASHAPE
COLEGRAM
ETNA
etc





Atomic and Nuclear data

Many applications, such as applied research or detector calibration, require knowledge of the atomic and nuclear data that follow radioactive decay, e.g. half-life, decay modes and branching ratios, the energies and intensities of the various emissions, etc.

In order to provide users with carefully recommended data, an international working group (Decay Data Evaluation Project, [DDEP](#)) was created in 1995. The update of the recommended data pages is performed by the Laboratoire National Henri Becquerel.

Below are the **tables of recommended data** for more than 200 radionuclides with detailed comments describing how these values were obtained.

Tables of recommended data

DDEP Evaluations

More specifically for users of alpha and gamma spectrometry, we have also developed **Nucléide-Lara**.

The online application makes it possible to consult and query the decay data for more than 400 radionuclides (half-life, decay mode, emission energy and intensity, decay scheme...)

Nucléide-Lara Application

Practical tool

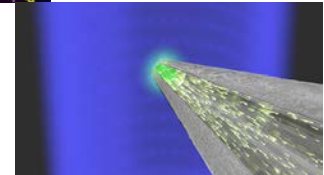
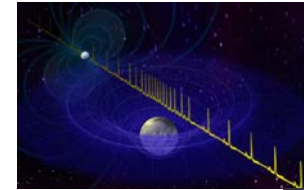
Laboratoire National
LNB
Henri Becquerel

- **Radionuclide decay data and efficiency calibration**
 - Efficiency calibration in the energy range from 20 keV to 120 keV
 - Radionuclide decay data
- **Low-energy spectra main features**
 - Full-energy peaks shape
 - Scattering
 - Escape peaks
 - Self-fluorescence
- **Self-attenuation**

Radionuclide decay data and efficiency calibration

RADIONUCLIDES OF INTEREST IN THE LOW-ENERGY RANGE

- Environmental monitoring (^{210}Pb , noble gases)
- Nuclear fuel cycle control (ores, reprocessing solutions, monitoring of nuclear materials, etc.)
- Reactor dosimetry ($^{93\text{m}}\text{Nb}$, $^{103\text{m}}\text{Rh}$)
- Medical applications (diagnostic, therapy)
- Fundamental research (astrophysics, X-ray applications)



- Some radionuclides of interest

- ✓ ^{241}Am (59 keV)
- ✓ ^{210}Pb (45 keV)
- ✓ ^{129}I , ^{133}I , ^{133}Xe
- ✓ L X-rays of actinides (10 keV – 20 keV)
- ✓ X-ray emitting radionuclides ($^{93\text{m}}\text{Nb}$, $^{103\text{m}}\text{Rh}$) (17 keV -23 keV)

- Photon emission intensities measurements and efficiency calibration directly linked:

$$\varepsilon(E) = \frac{N(E)}{A \cdot I(E) \cdot t} \prod_i C_i \quad I(E) = \frac{N(E)}{A \cdot \varepsilon(E) \cdot t} \prod_i C_i$$

N(E): Net peak area

A: source activity

I(E): photon emission intensity

t: counting time (live time)

Ci: corrective factors

- Efficiency calibration in the energy range 20 keV – 80 keV
- Few radionuclides for calibration

EFFICIENCY CALIBRATION WITH STANDARD SOURCES

- Photon emission intensities measurements and efficiency calibration directly linked:

$$\varepsilon(E) = \frac{N(E)}{A \cdot I(E) \cdot t} \prod_i C_i \quad I(E) = \frac{N(E)}{A \cdot \varepsilon(E) \cdot t} \prod_i C_i$$

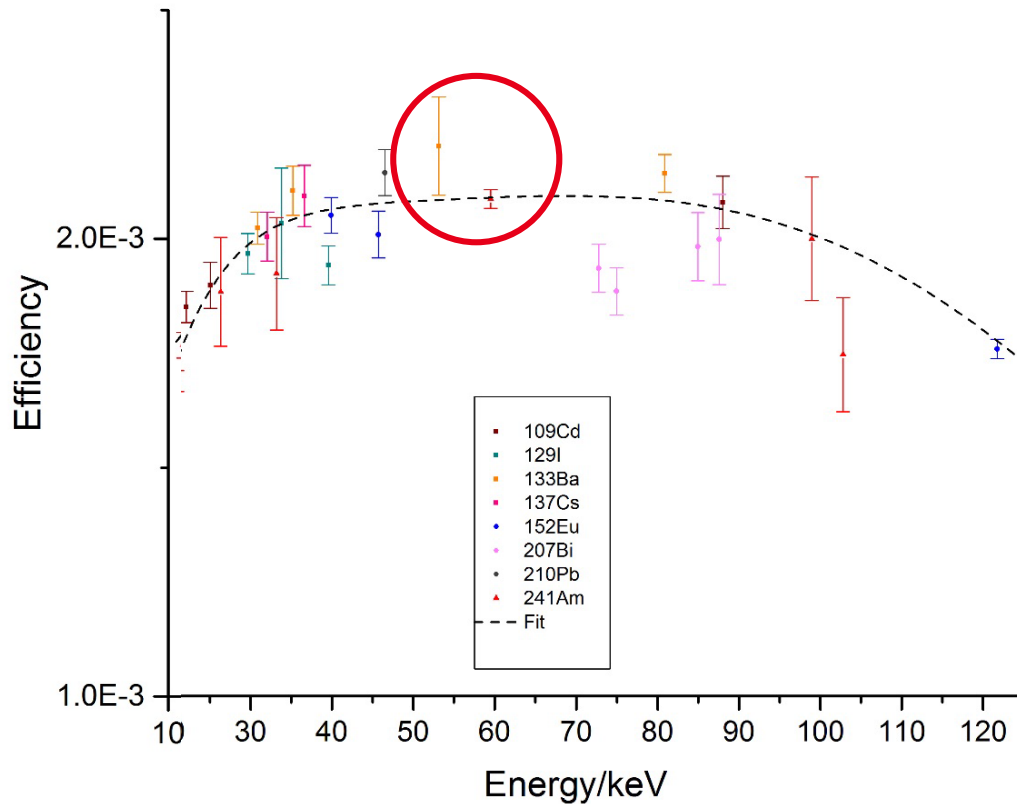
- Efficient
- Few radionuclides

Radionuclide	Energy of γ -rays (keV)	Energy of $K\alpha$ X-rays (keV)	Energy of $K\beta$ X-rays (keV)
^{129}I	39.6	29.5 – 29.8	33.7 – 34.5
^{210}Pb	46.5		
^{133}Ba	53.2 – 79.6+81.0	30.6 – 31.0	35.1 – 35.9
^{241}Am	59.5		
^{109}Cd	88.0	22.0 – 22.2	25.0 – 25.5
^{113}Sn		24.0 – 24.2	27.3 – 27.9
^{137}Cs		31.8 – 32.2	36.5
^{139}Ce		33.0 – 33.4	37.9 – 38.8
$^{166\text{m}}\text{Ho}$	80.6	48.2 – 49.1	55.7 – 57.3

EFFICIENCY CALIBRATION WITH STANDARD SOURCES

- Efficiency calibration with point sources + mathematical fitting
 - (log-log polynomial – covariances)

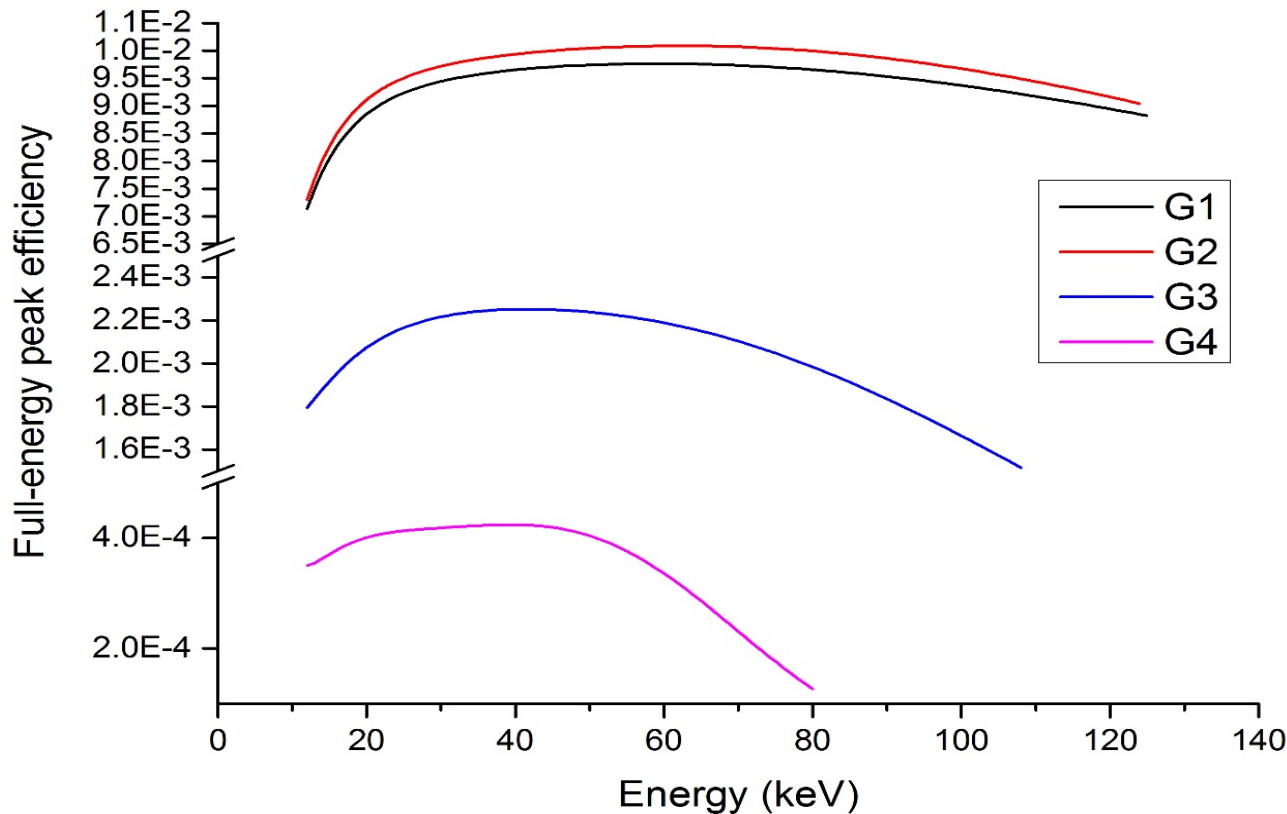
$E > 120$ keV: 0.5 %



5 keV < E < 120 keV: 2 %
(at best)

Systematic deviations

EFFICIENCY CALIBRATION OF HPGe DETECTORS

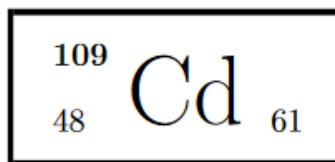
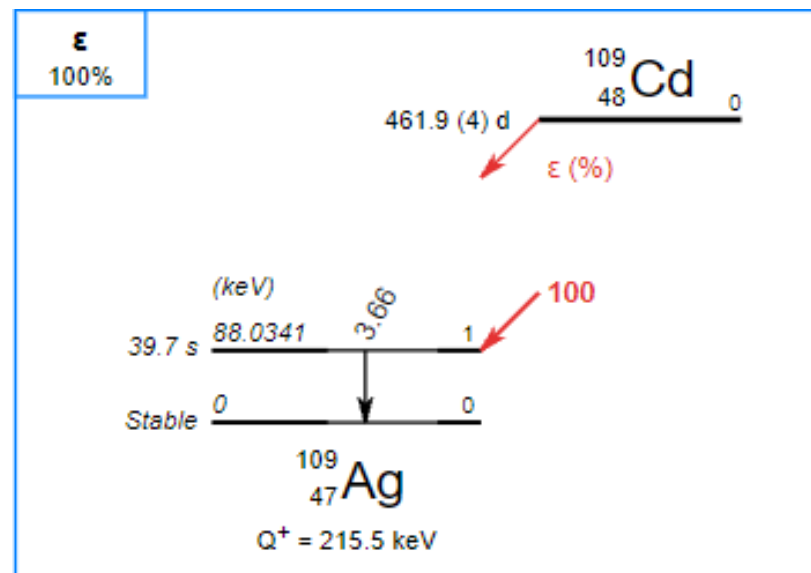


Maximum of the efficiency calibration curve depending on the detector size and composition (dead layer)

Not well known
(Use Monte Carlo simulation?)

- DDEP evaluation: example ^{109}Cd

LNE – LNHB/CEA Table de Radionucléides



1 Decay Scheme

Cd-109 decays by electron capture to the isomeric state (88 keV) of Ag-109 .

Le cadmium 109 se désintègre uniquement par capture électronique vers l'état isomérique de l'argent 109 (88 keV).

RADIONUCLIDE DECAY DATA

¹⁰⁹Cd - Comments on evaluation of decay data by M.M. Bé, E. Schönfeld

This evaluation was first completed by E. Schönfeld in 1996, and was completely reviewed by M.M. Bé in November 2014. New half-life and γ intensity measurement results and improved calculations of the internal conversion coefficients were included.

5. Photon Emissions

5.1 X-ray Emissions

The total KX- ray emission intensity is (assuming there is no EC transition to the ground state):

$$I_{KX} = \alpha_K \{P_K + [\alpha_K / (1 + \alpha_i)]\} \times 100 \% = 102,2 (9) \%$$

The measured values of the ratio I_{KX} / I_γ are summarized below and compared with the value deduced from the decay scheme parameters.

	I_{KX} / I_γ	Reference
1	33,8 (7) ^(O)	Wapstra and van der Eijk (1957Wa05) - outlier
2	26,2 (6)	Leutz <i>et al.</i> (1965Le06)
3	22,2 (6) ^(O)	Jansen and Wapstra (1966Ja01) - outlier
4	29,1 (10)	Freedman <i>et al.</i> (1966Fr12)
5	30 (4)	Foin <i>et al.</i> (1968Fo03)
6	26,2 (5)	Campbell and McNelles (1972Ca16)
7	27,0 (3)	Dragoun <i>et al.</i> (1976Dr07) – superseded by 1979PI04
8	27,3 (6)	Plch <i>et al.</i> (1979PI04)
9	27,34 (27)	Hoppes and Schima (1982HoZF)
10	27,9 (4)	Egorov <i>et al.</i> (1989Eg**)
11	27,7 (5)	Unweighted mean
12	27,25 (29)	Weighted mean; reduced- $\chi^2 = 2,4$; Crit. $\chi^2 = 2,8$
13	27,92 (45)	Present evaluation using the above equation together with the adopted values of ω_K , P_K , α_K , α_T and I_γ

2.4% relative difference

X-RAY EMISSION INTENSITIES

K X-ray emission accompanies atomic relaxation consecutive to a vacancy created in the electronic K shell either by electron capture or internal conversion

Internal conversion $I_{XK} = \frac{\alpha_K}{1 + \alpha_T} \omega_K P_\gamma$

Electron capture $I_{XK} = \omega_K P_{eK}$

K Fluorescence yield: $\omega_K = \frac{I_{XK}}{I_{XK} + I_{AK}}$

I_{XK} : K X-ray emission intensity

I_{AK} : K Auger electron emission intensity

P_γ : Probability of the gamma transition

P_{eK} : Probability of electron capture in K shell

ω_K : **Compilation from Schönfeld (compilation)**

α_K, α_T : Internal conversion coefficients – Theoretical calculation (BrIcc code)

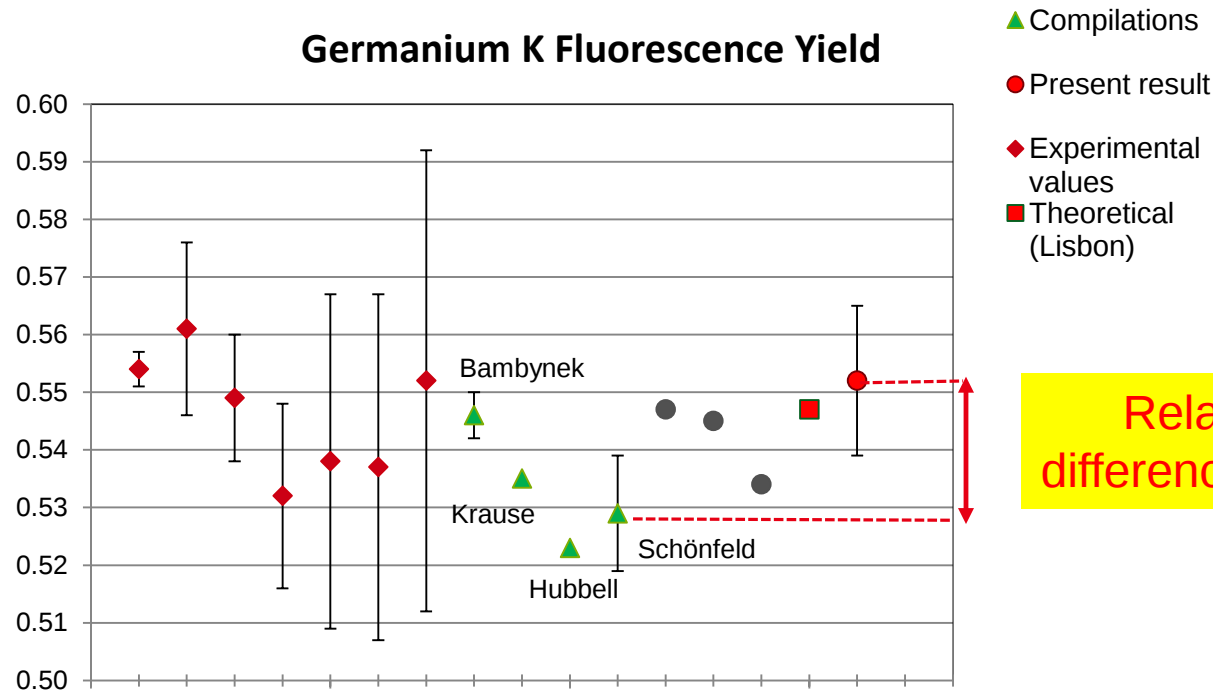
} Balance of the decay
scheme or calculation

2008Ki07 T. Kibédi, T.W. Burrows, M.B. Trzhaskovskaya, P.M. Davidson, C.W. Nestor, Jr.

'Evaluation of theoretical conversion coefficients using BrIcc' Nucl. Instr. and Meth. A 589 (2008) 202-229

X-RAY EMISSION INTENSITIES

- X-ray emission intensities are often derived from calculation → Relative standard deviation > 2%
- Relative standard deviation of K fluorescence yields: 2%
- Example of germanium: recent measurement and calculation of K fluorescence yield

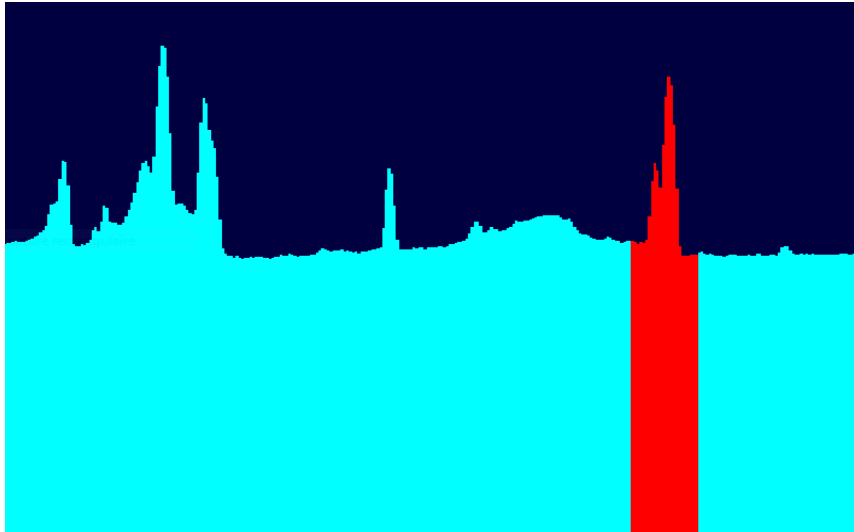


Relative
difference > 4 %

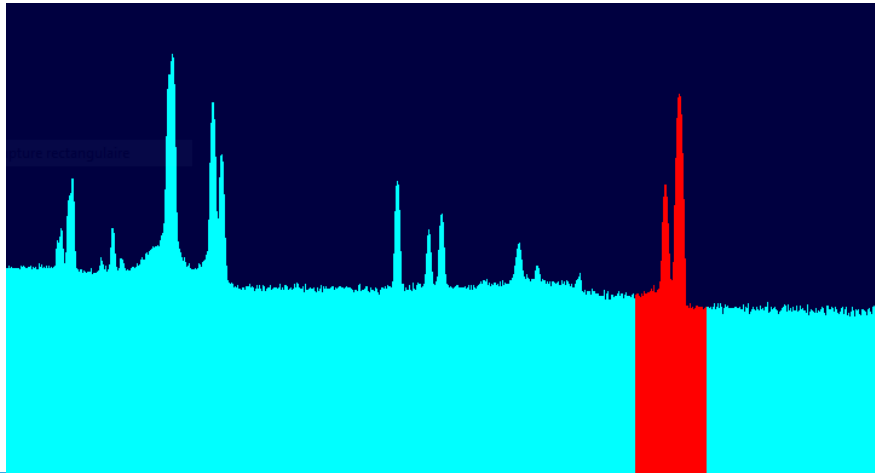
Low-energy spectra main features

EXAMPLE: DOUBLE PEAK AT 79-81 keV (^{133}Ba)

Energy range 15 keV – 100 keV



N-type HPGe detector (coaxial)



N-type HPGe detector (planar)

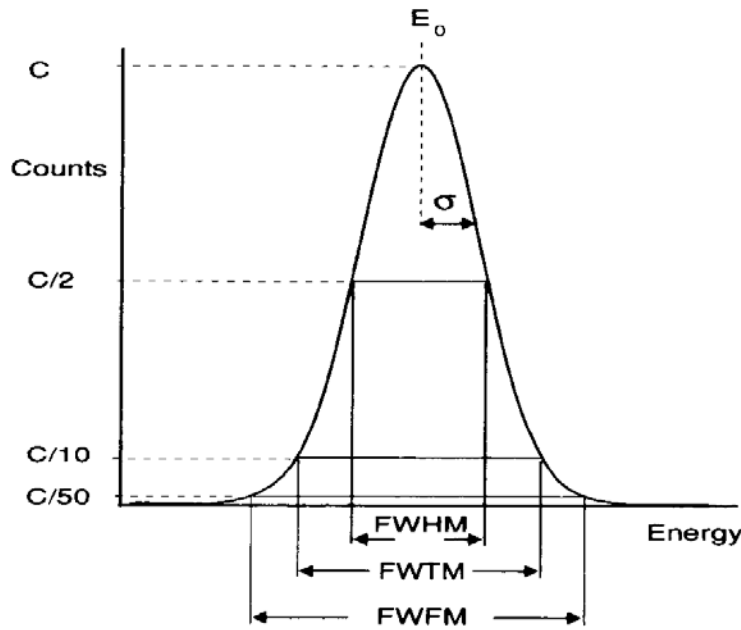
$$\varepsilon(E) = \frac{N(E)}{A \cdot I(E) \cdot t} \prod_i C_i \quad \text{or} \quad A = \frac{N(E)}{I(E) \cdot \varepsilon(E) \cdot t} \prod_i C_i$$

- Quantitative result directly linked to the full-energy peak area
 - ✓ Direct impact on the final uncertainty
- Determining the net peak area may need specific processing
- Dedicated processing taking into account:
 - ✓ Full-width at half maximum versus the energy;
 - ✓ Overlapping;
 - ✓ Natural width of incident photons;
 - ✓ Scattering effects.

ENERGY RESOLUTION

- Photon emission initially monoenergetic, but at the detector level, **widening** due to:
 - Energy drifts
 - Gaussian effects
 - ✓ Fluctuation of the number of charge carriers,
 - ✓ Preamplifier and shaping (electronics) noise,
 - Non-gaussian effects
 - ✓ Collection of the pairs charges (mobility of the electron/hole)
 - ✓ Pile-up
- In spectra: 'peaks' with defined width and shape more or less symmetrical
- First approximation: Gaussian shape

ENERGY RESOLUTION: GAUSSIAN DISTRIBUTION



Distribution centered on energy E_0 with standard deviation σ

FWHM (full-width at half-maximum): ΔE

σ and ΔE are linked: $\Delta E = 2.355 \sigma$

Energy resolution characterizes the ability of the spectrometer:

- to separate peaks with close energies (resolving power),
- and to reach low detection limits.

FWTM (Full Width at Tenth of Maximum)

FWFM (Full Width at Fiftieth of Maximum)

For a Gaussian peak (ideal case):

FWTM / FWHM = 1.82

FWFM / FWHM = 2.38

$$G(E) = A \cdot \exp\left(-\frac{(E - E_0)^2}{2\sigma^2}\right)$$

Peak area: $S = \sqrt{2\pi} \sigma \cdot A$

Information on the resulting peak shape, especially on the low-energy part (left side) (charge collection defects)

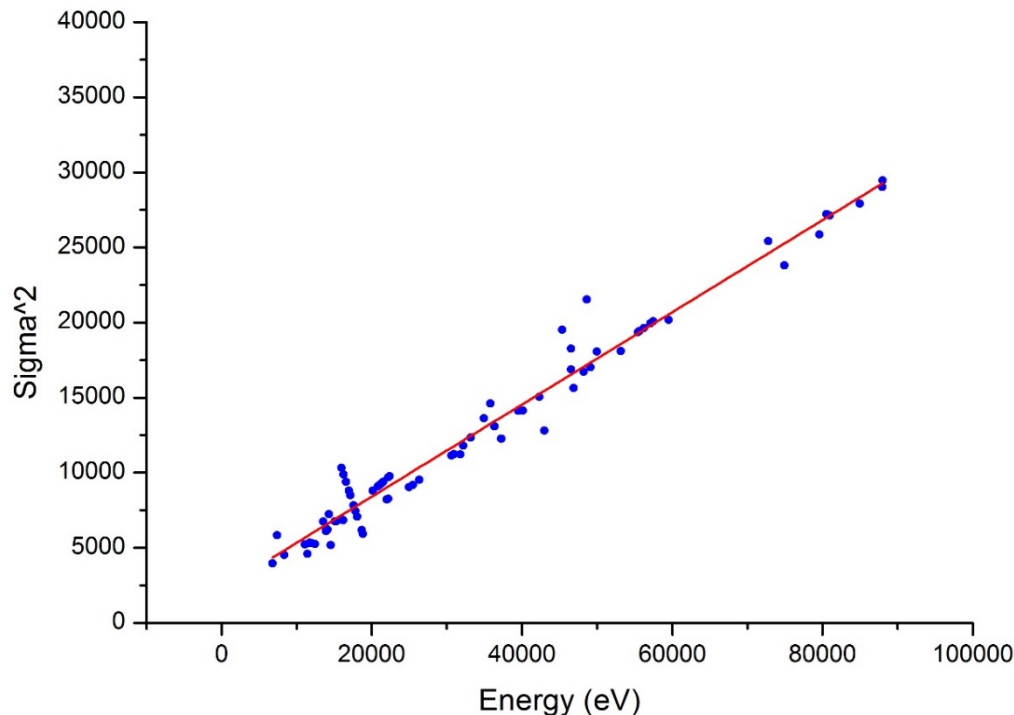
ENERGY RESOLUTION: FULL WIDTH AT HALF MAXIMUM

- Gaussian standard deviation depends on the photon energy, E :

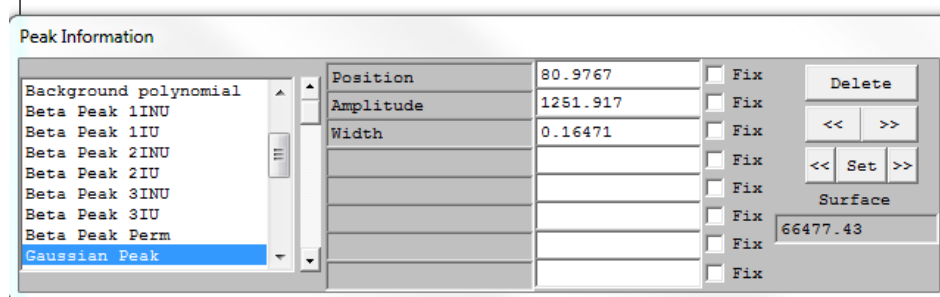
$$\sigma^2(E) = \sigma_0^2 + F \cdot w \cdot E$$

w = mean pair creation energy

F = Fano factor



With $w=2.97$ eV $\rightarrow F=0.103$

EXAMPLE OF PEAK PROCESSING (^{133}Ba – 79-81 keV)

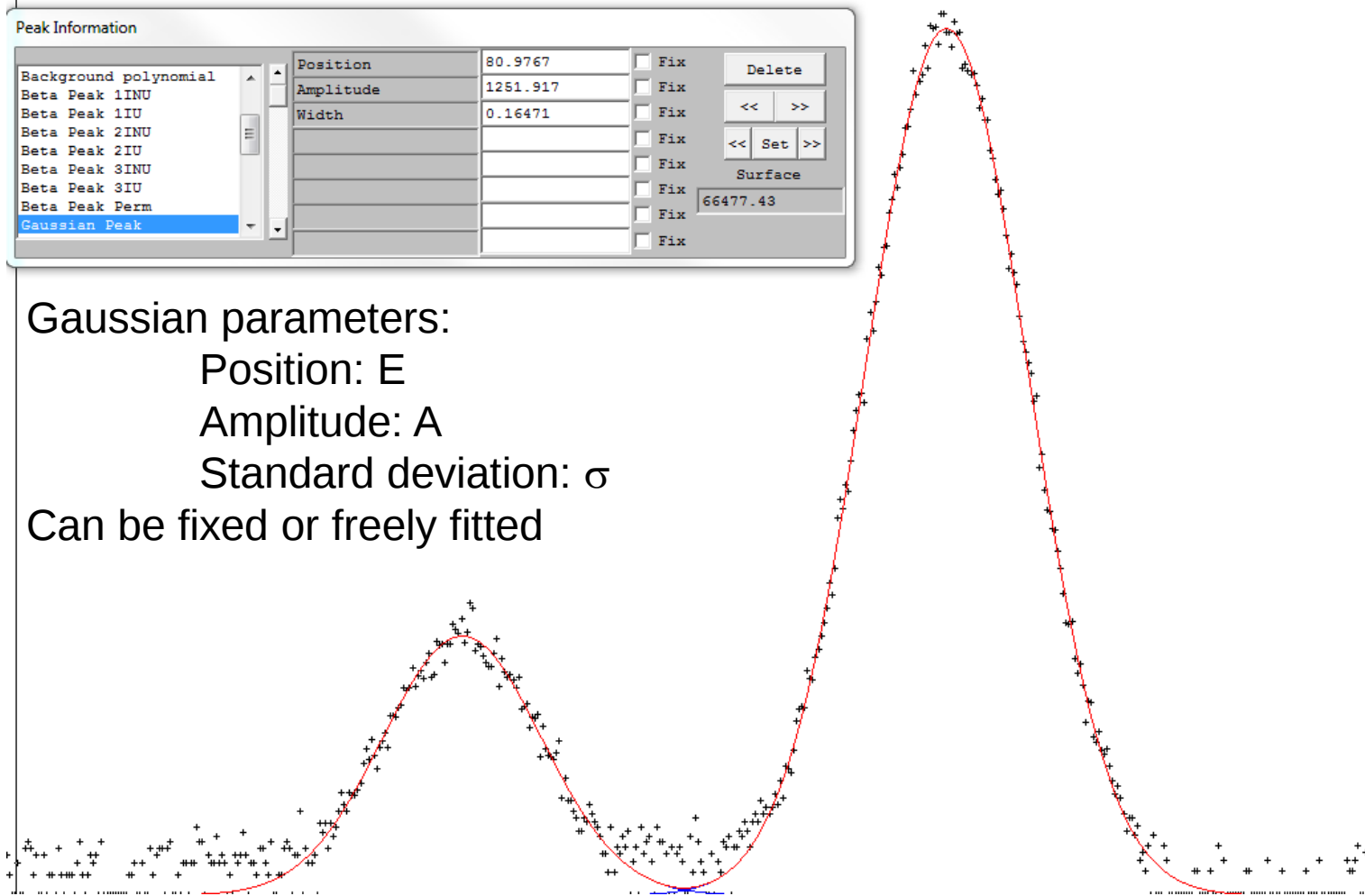
Gaussian parameters:

Position: E

Amplitude: A

Standard deviation: σ

Can be fixed or freely fitted

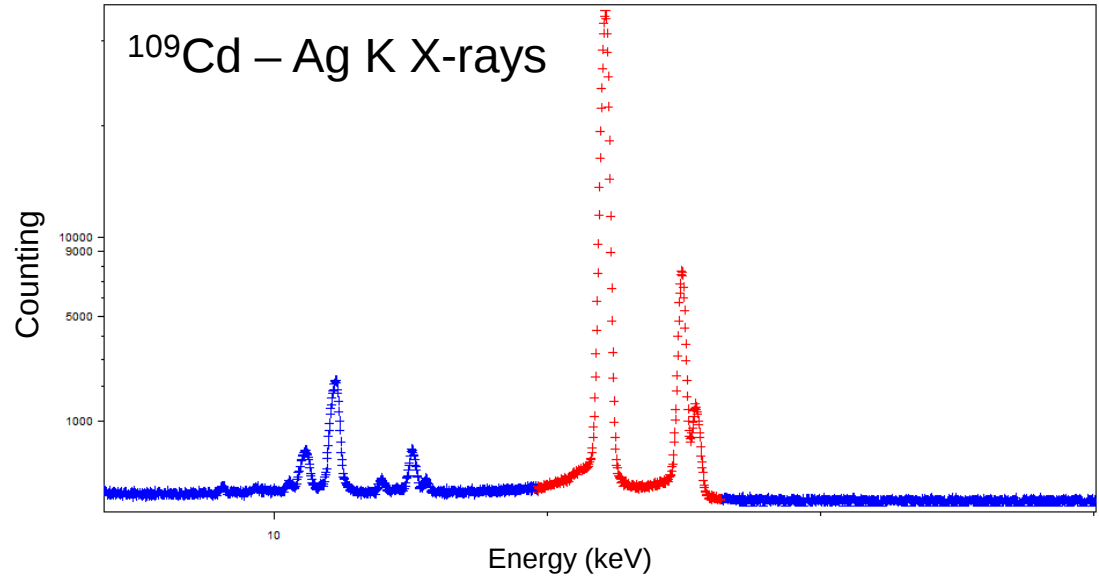


COLEGRAM, a flexible user-friendly software for processing of ionizing radiation spectra,
Y. Ménesguen and M.-C. Lépy, Nuclear Inst. and Methods in Physics Research, A 1003 (2021) 165341,
<https://doi.org/10.1016/j.nima.2021.165341>

X-RAY SPECTRA: Example around 20 keV

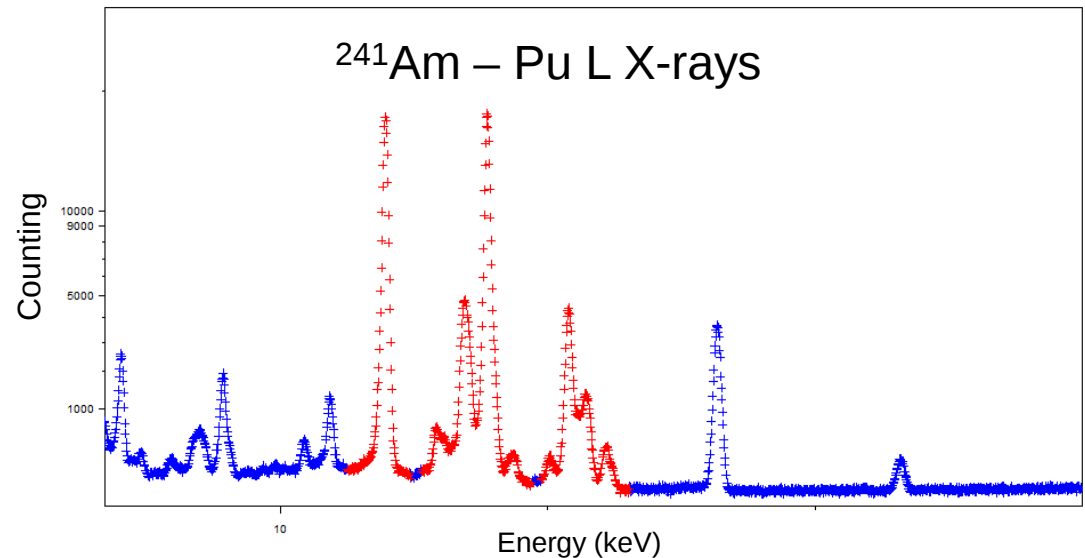
- **K X-rays: 2 groups**

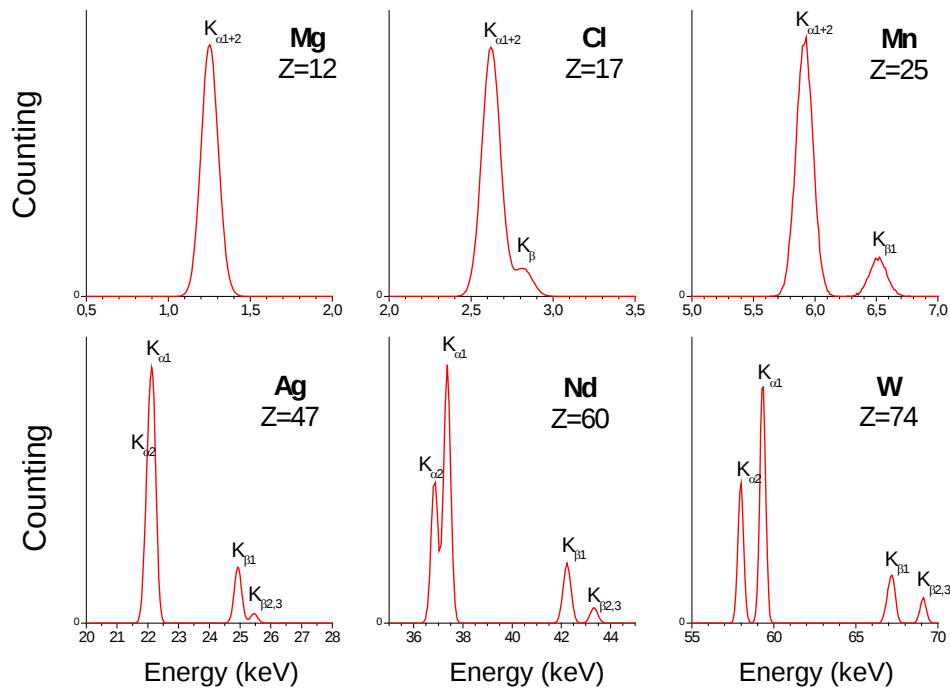
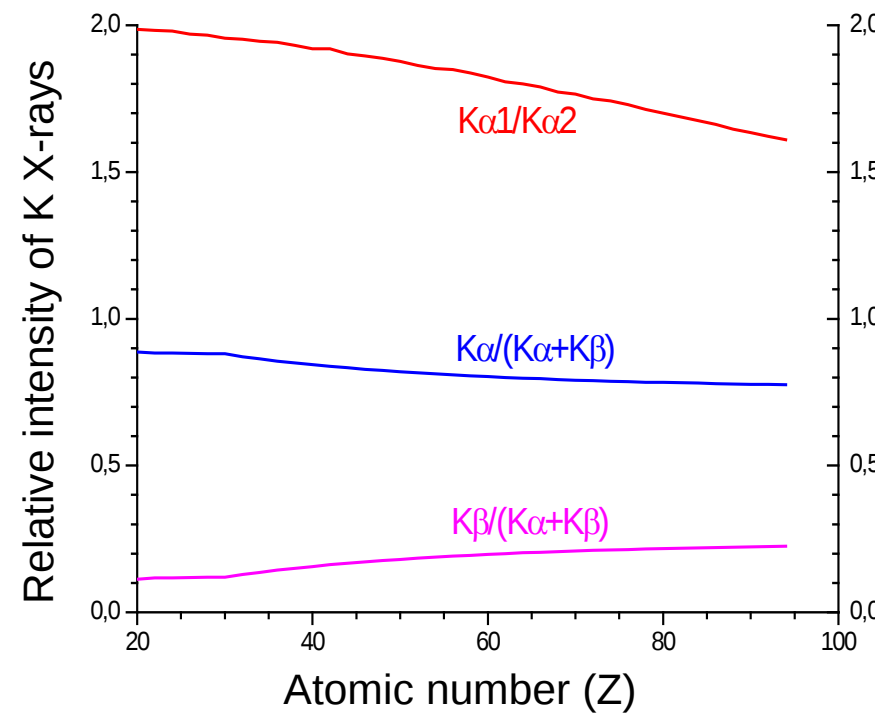
- ✓ Kalpha 2,1
- ✓ Kbeta1,2
- ✓ Escape peaks



- **L X-rays: 3 main groups**

- ✓ Lalpha
- ✓ Lbeta
- ✓ Lgamma







Photons = transitions between excited levels

Gamma = nuclear levels

X = atomic levels

Heisenberg uncertainty principle

$$\Delta E \cdot \Delta t \geq \frac{h}{2\pi} (= 6.582\,122 \cdot 10^{-16} \text{ eV.s})$$

Δt uncertainty of the life time of the energy level

ΔE uncertainty of the energy

Uncertainties on the energy levels of the transition → photon emission has an energy with

uncertainty : finite line shape that is Lorentzian (Γ): $L(E) = \frac{\Gamma/2\pi}{(E-E_0)^2 + (\Gamma/2)^2}$

Gamma-ray line width = some 10^{-3} eV (maximum)

X-ray lines : some eV

Detector widening (Gaussian) : some hundreds of eV

Width of emitted X-ray: sum of width of the two levels of the transition

Element	Z	K X-ray energy (keV)	Level width (eV)		K α 2 X-ray width (eV)	FWHM (eV)	X-ray linewidth /FWHM
			K	L2			
Nickel	28	7.45	1.44	0.52	1.95	155	0.013
Cadmium	48	22.98	7.28	2.62	9.9	200	0.049
Plomb	82	72.80	60.4	6.5	66.8	350	0.19
Uranium	92	94.65	96.1	9.3	105.4	415	0.25

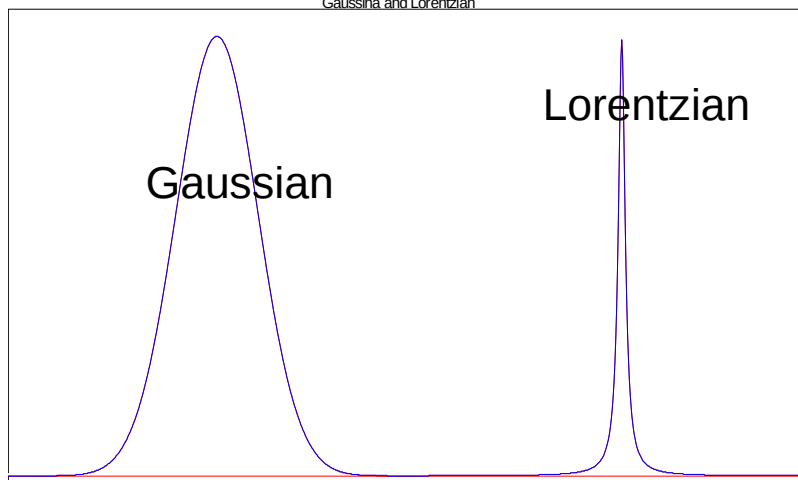
For high Z elements, the natural linewidth is not negligible compared to the energy resolution of the detector

Shape of emission lines : Lorentzian

Widening of the detector : Gaussian

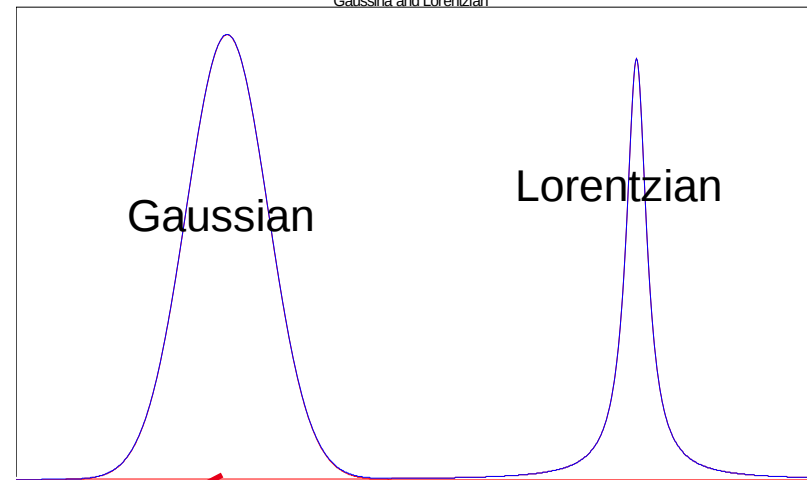
X-RAY NATURAL LINEWIDTH

Gaussian and Lorentzian



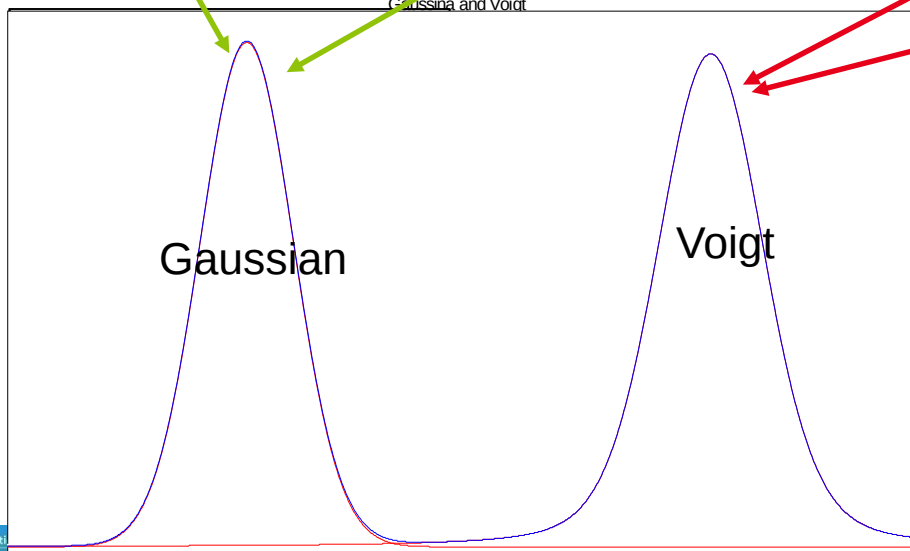
$$\sigma = 50 \text{ eV} - \Gamma = 10 \text{ eV}$$

Gaussian and Lorentzian



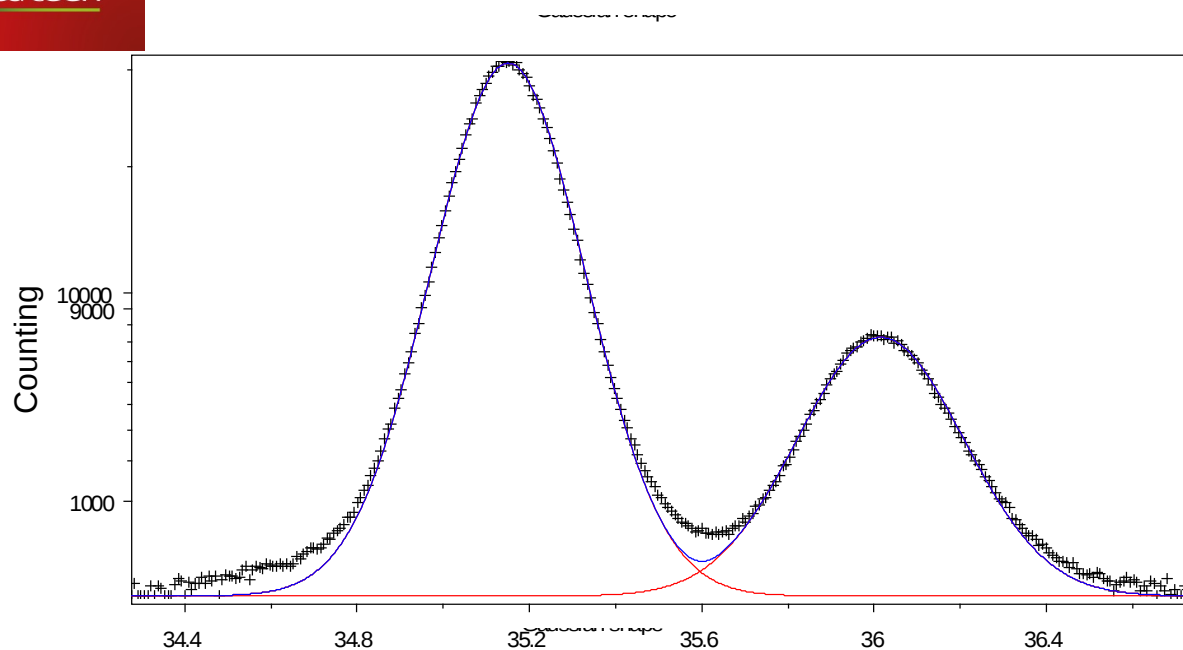
$$\sigma = 50 \text{ eV} - \Gamma = 30 \text{ eV}$$

Gaussian and Voigt

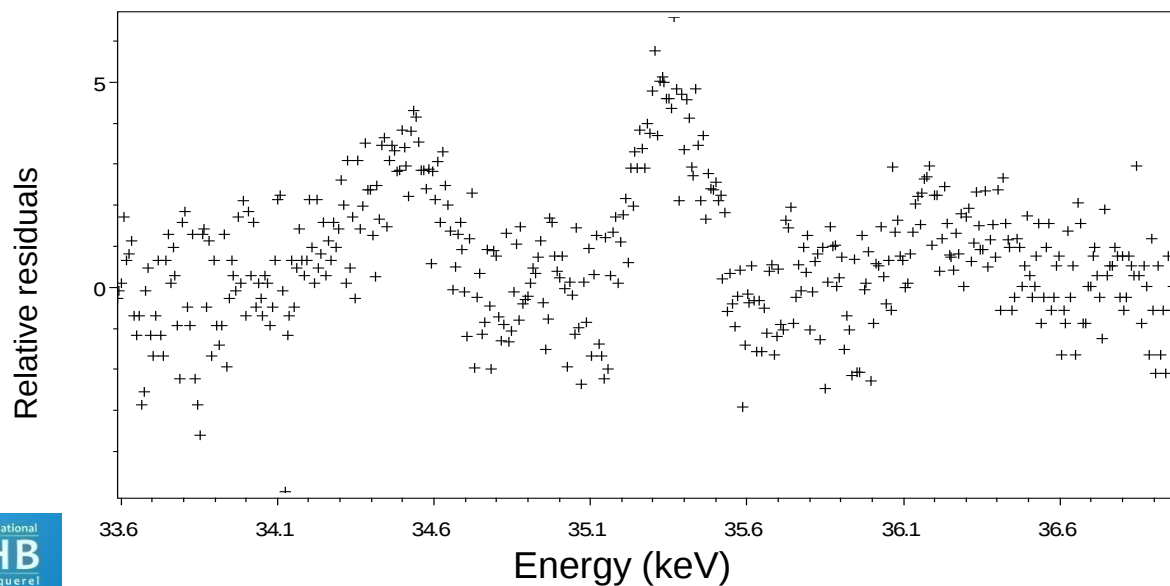


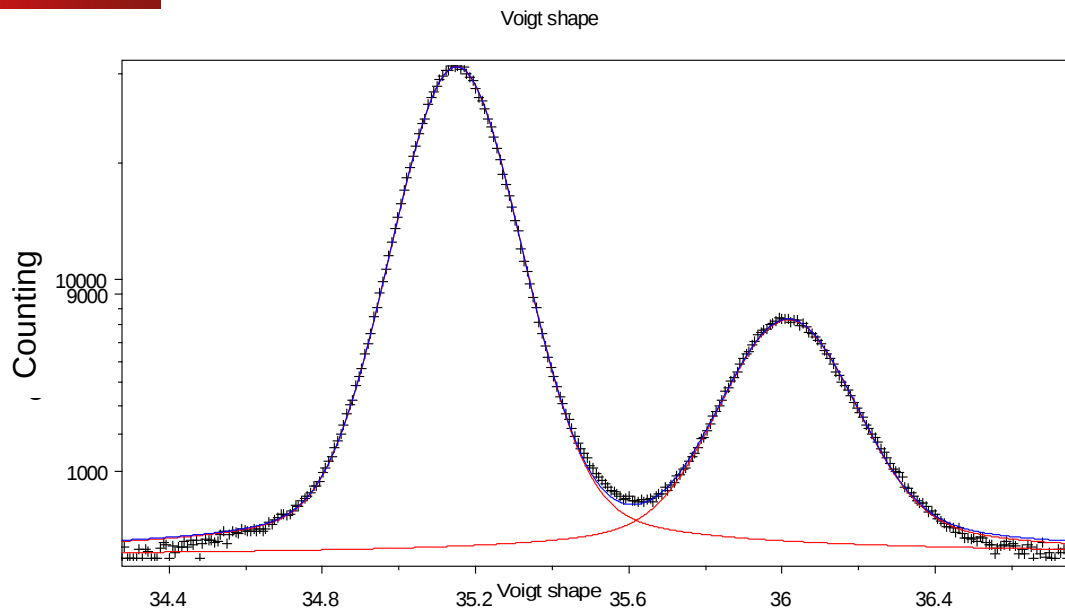
Peak = *Lorentzian* $\otimes$ *Gaussian*
(Voigt profile)

$$V(E) = \int_{-\infty}^{+\infty} L(E') \cdot G(E - E') \cdot dE'$$



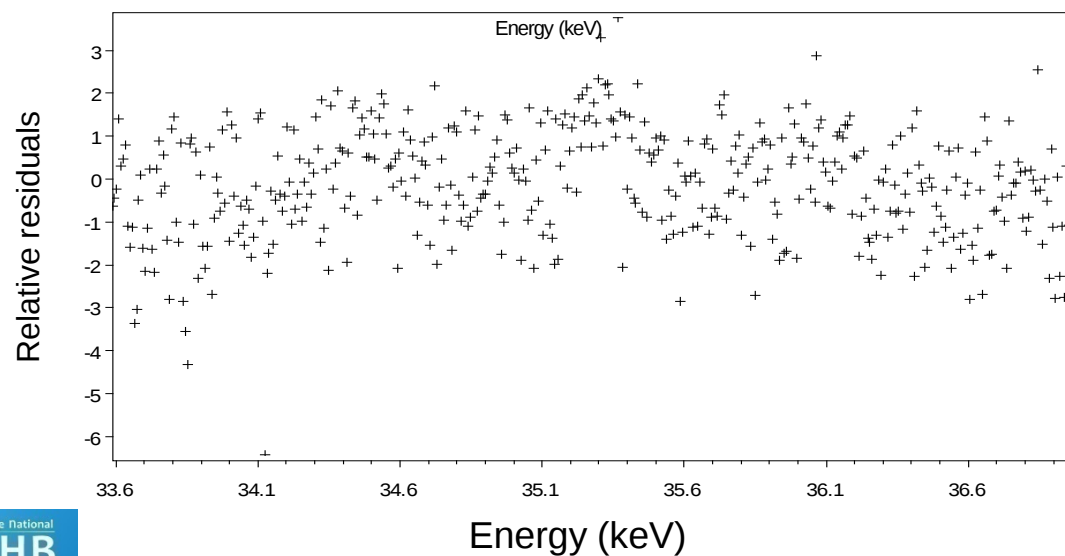
$^{133}\text{Ba} - \text{Cs K}\beta$
Gauss function
 $\chi^2 = 3.3$





$^{133}\text{Ba} - \text{Cs } K\beta$
Voigt function
Tabulated * values for Γ

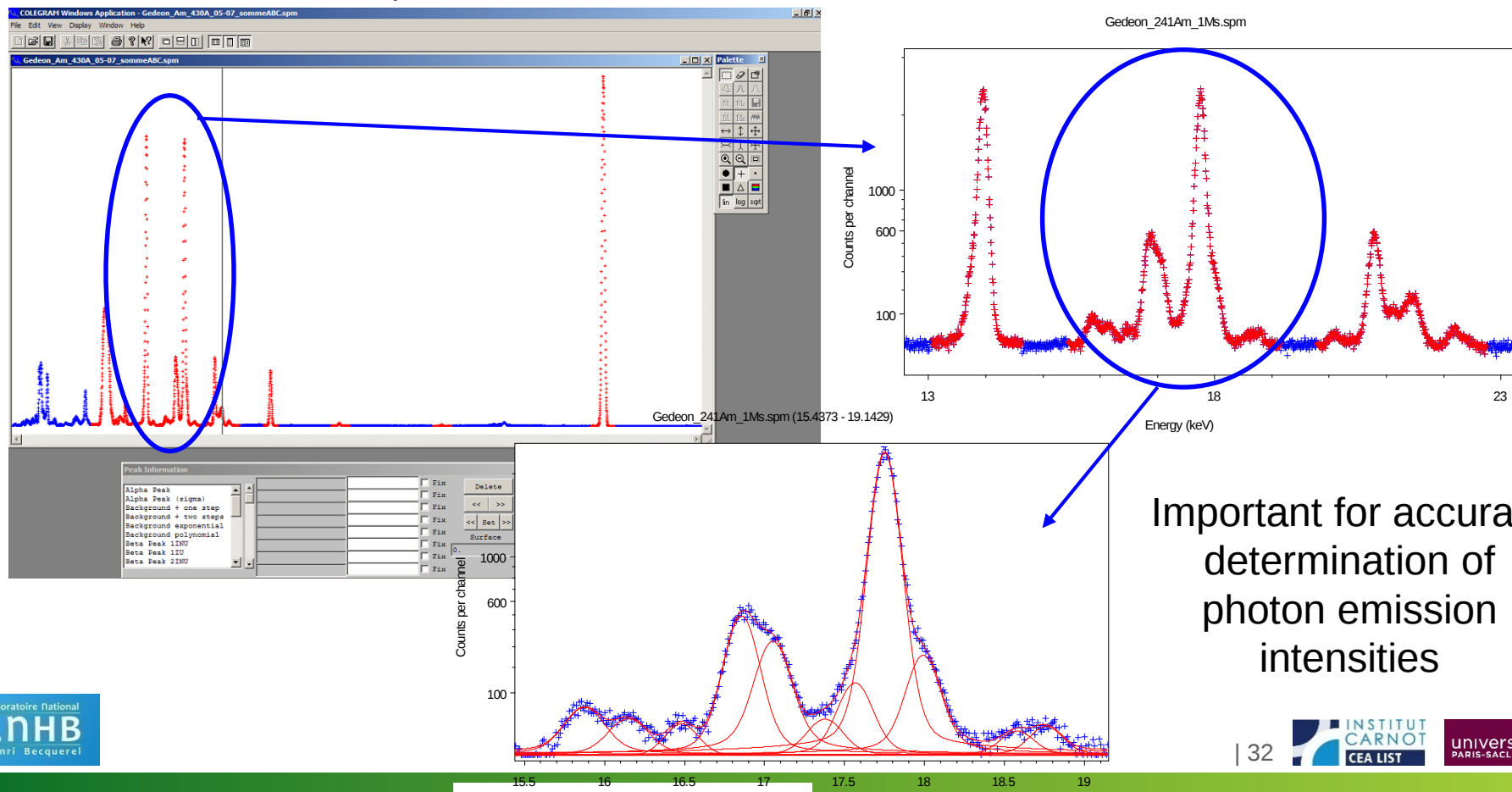
$\chi^2 = 1.65$



*Campbell, J.L., Papp, T., 2001,
Widths of the atomic K-N7 levels,
Atomic Data and Nuclear Data Tables 77, 1–56.

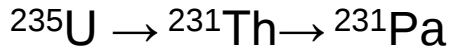
Example of processing of L X-rays of Pu (^{241}Am)

^{241}Am spectrum



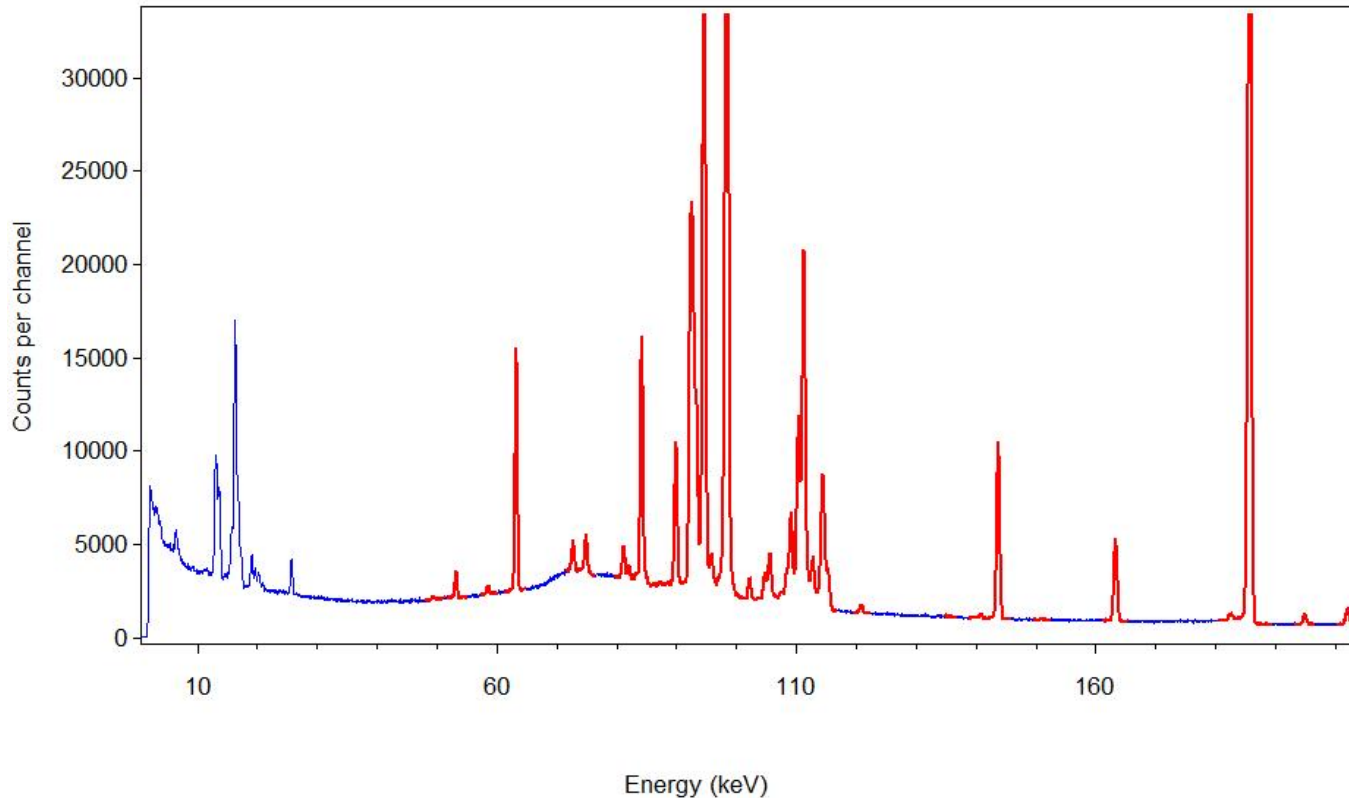
Important for accurate determination of photon emission intensities

Example of processing of UO_2 spectrum in the 10 keV – 200 keV energy range (enrichment: 5.2%)



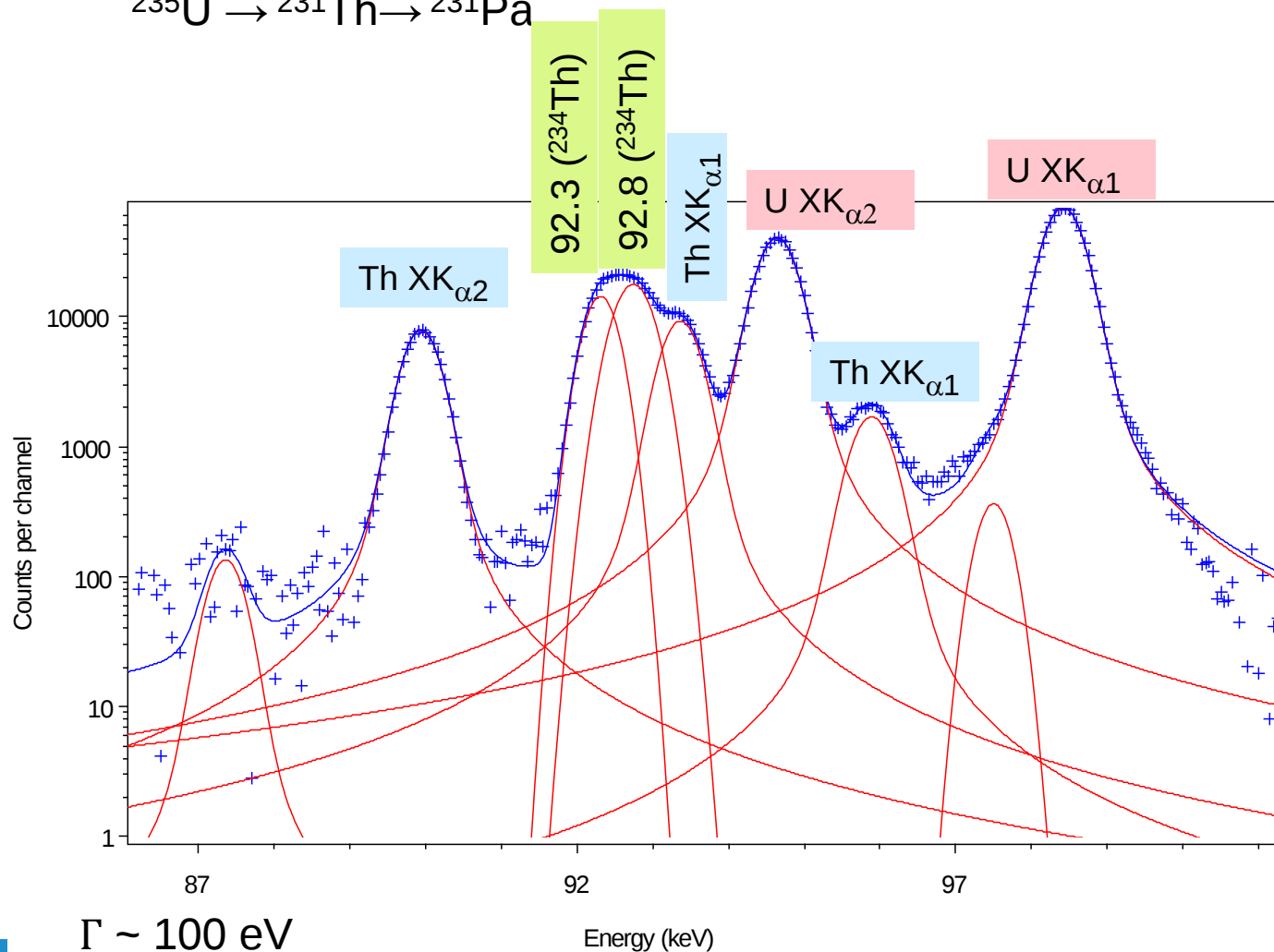
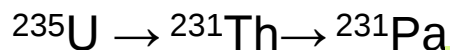
Spectra include **gamma** lines from different radionuclides

and **X-rays** from U, Pa and Th



DETAILED PROCESSING OF SPECTRA

Example of processing of K X-rays of UO_2 (enrichment: 5.2%)



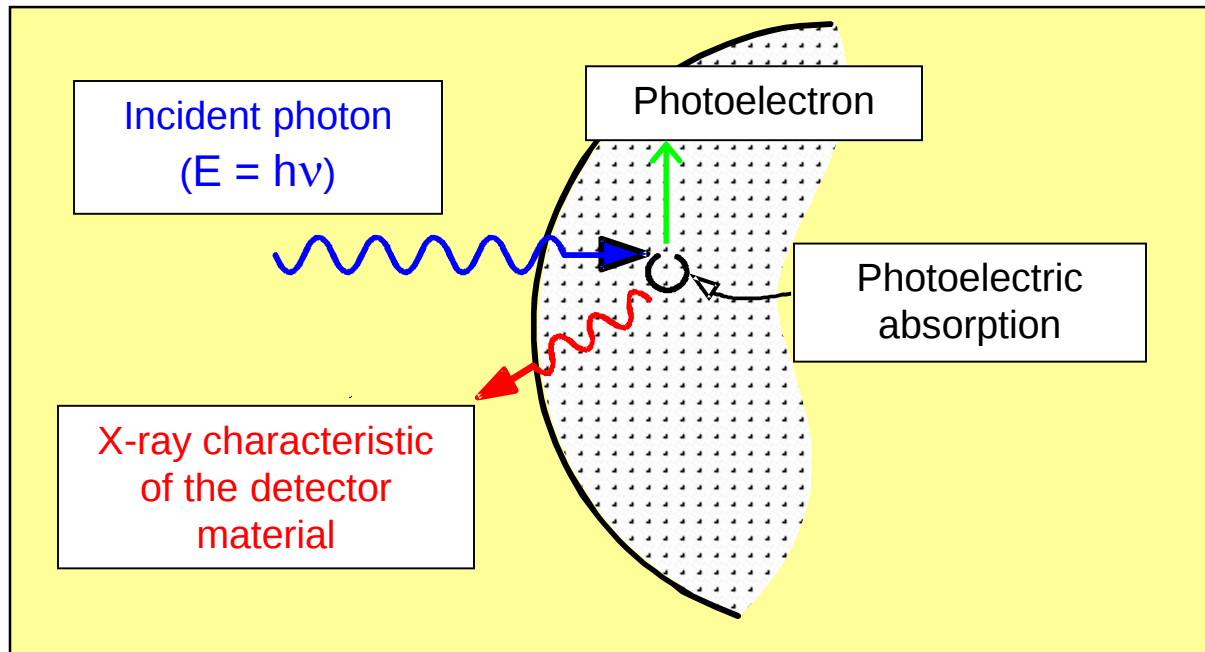
Important for
accurate
determination
of photon
emission
intensities

$\Gamma \sim 100 \text{ eV}$

$\sigma \sim 500 \text{ eV}$

ESCAPE PEAKS

Effect consecutive to the photoelectric absorption: ejection of the photoelectron is accompanied by atomic relaxation (rearrangement of the electronic shells) with emission of X-ray photons characteristic of the detector material (Ge, Si...)

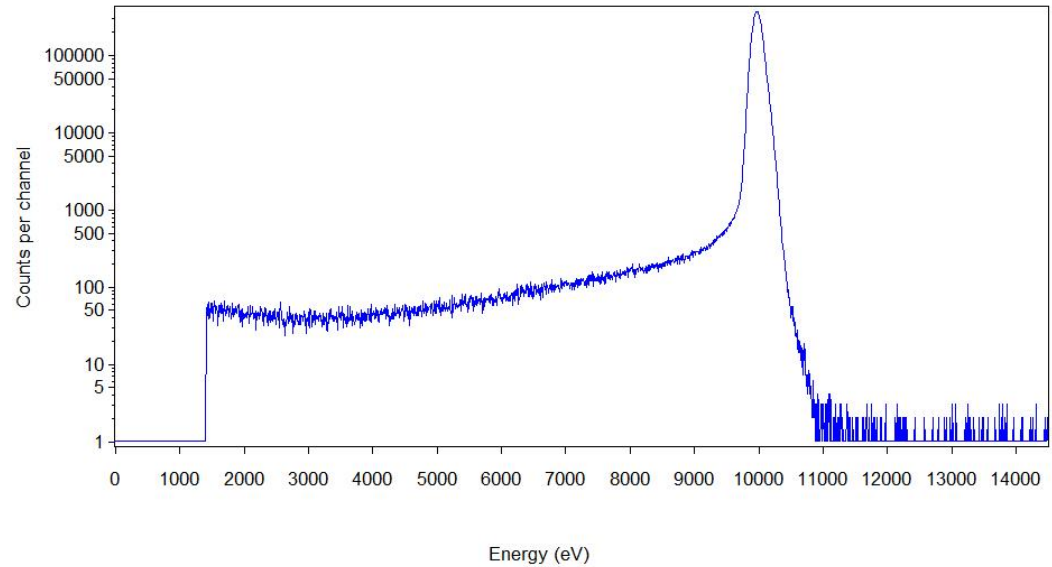


ESCAPE PEAKS

K binding energy of Ge: 11 keV

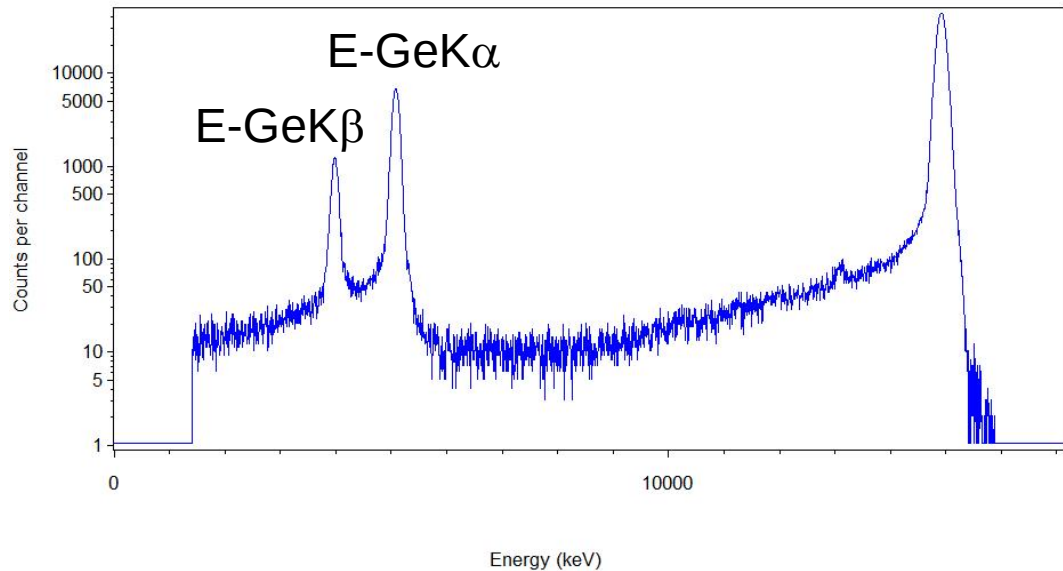
Below 11 keV: No escape

Monochromatic photons 10 keV



Monochromatic photons 15 keV

Above 11 keV :
Escape of characteristic
Ge X-rays



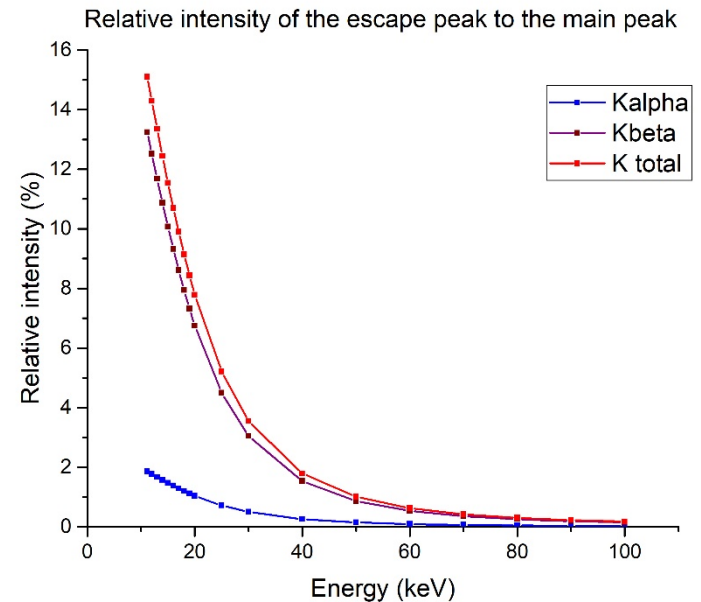
◆ If photoelectric absorption occurs close to the detector surface, Ge characteristic X-rays may escape from the detector active volume → only part of the incident energy is recorded:

→ Escape peak(s) at $E - E_x$

- Ge : $E_{xK\alpha} = 9.9 \text{ keV}$ $E_{xK\beta} = 11.0 \text{ keV}$ (a few percent)
- Si : $E_{xK} = 1.74 \text{ keV}$ (a few per thousand)

• Escape effect in Ge is more intense

- Smaller interaction depth,
- Larger fluorescence yield,
- Larger energy of characteristic X-rays.



SELF FLUORESCENCE (RARE)

Fluorescence induced in the source material by incident energies $>$ binding energy of the source material

Example : Nb dosimeter – activation $\rightarrow$ ^{93m}Nb and impurities (^{182}Ta).

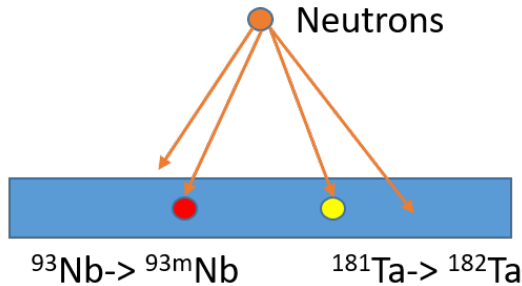


Tantalus' torment

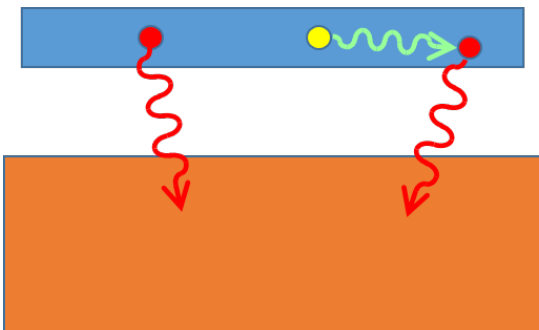
Niobium is from Niobé, Tantalus' daughter (son of Zeus) in Greek mythology, as tantalum, which was first discovered, is always mixed with niobium.

Decay of ^{182}Ta (mainly K X-rays of W) induces photoelectric effect in Nb
 $\rightarrow$ **additional K X-rays of niobium** superimposed
on K X-rays from the decay of ^{93m}Nb

SELF FLUORESCENCE (RARE)



Activation in a nuclear reactor of niobium dosimeter with Ta impurities



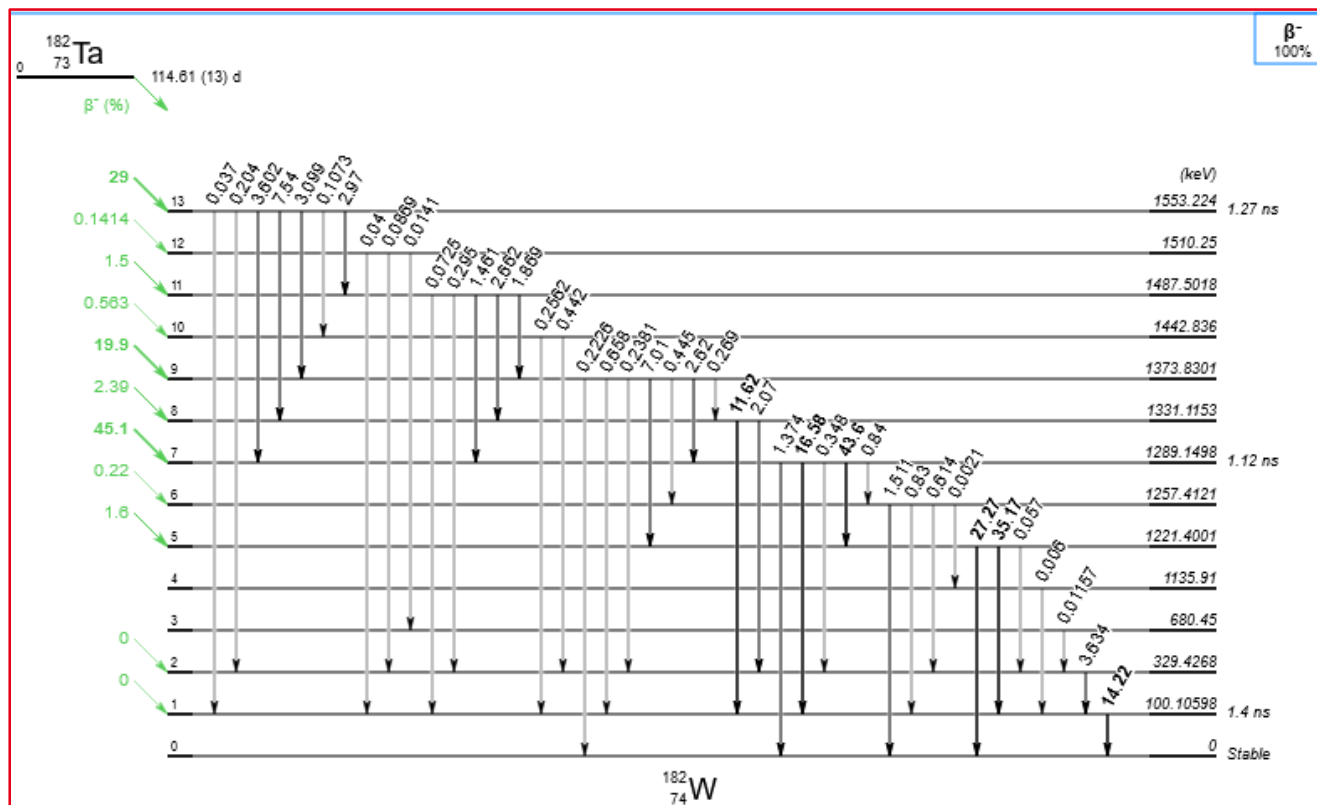
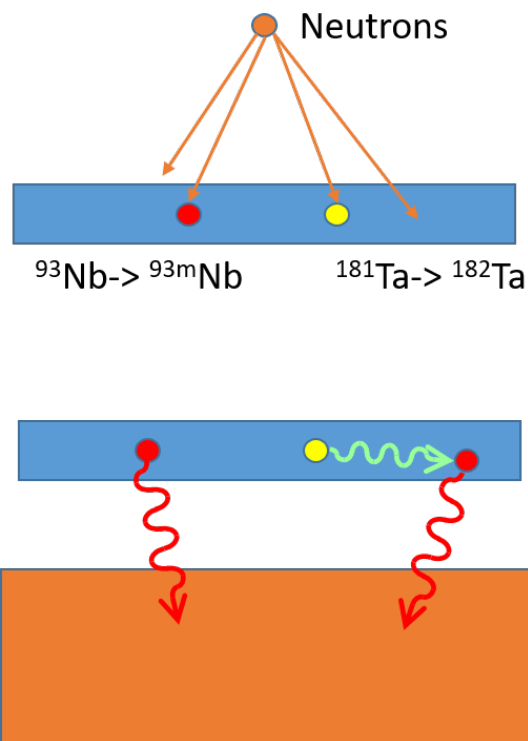
Photoelectric effect (photons from ^{182}Ta decay) in niobium

X-ray spectrometer

Needs to calculate a corrective factor for this extra-counting Monte Carlo simulation

Here, about 5% photons/ Bq ^{182}Ta

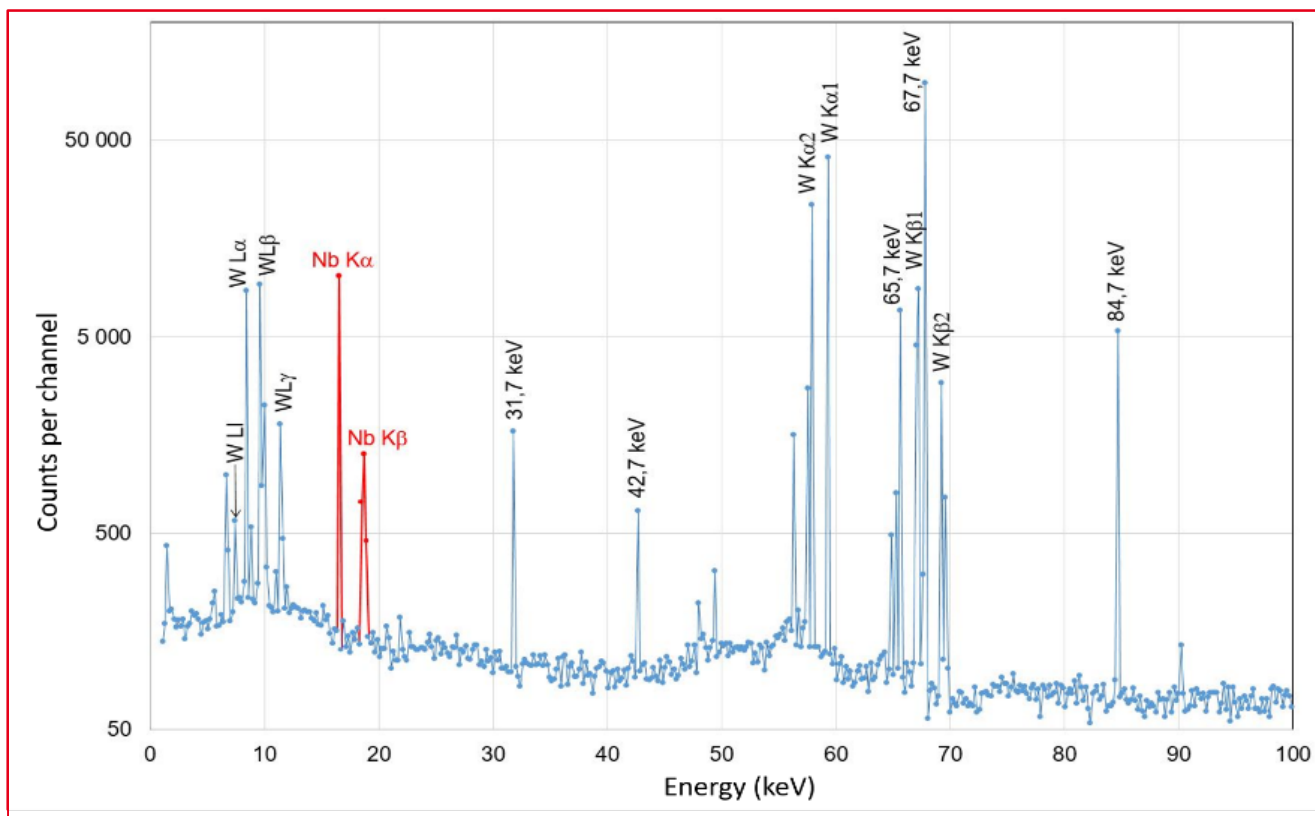
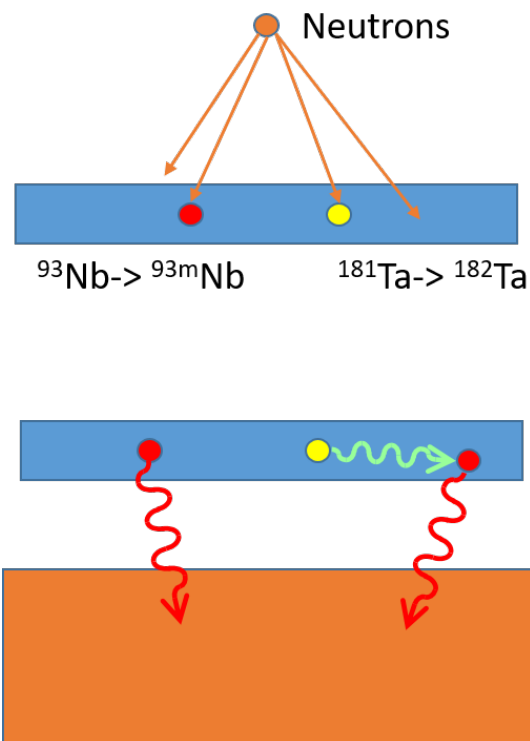
SELF FLUORESCENCE (RARE)



Needs to calculate a corrective factor for this extra-counting
Monte Carlo simulation

Here, about 5% photons/ Bq ^{182}Ta

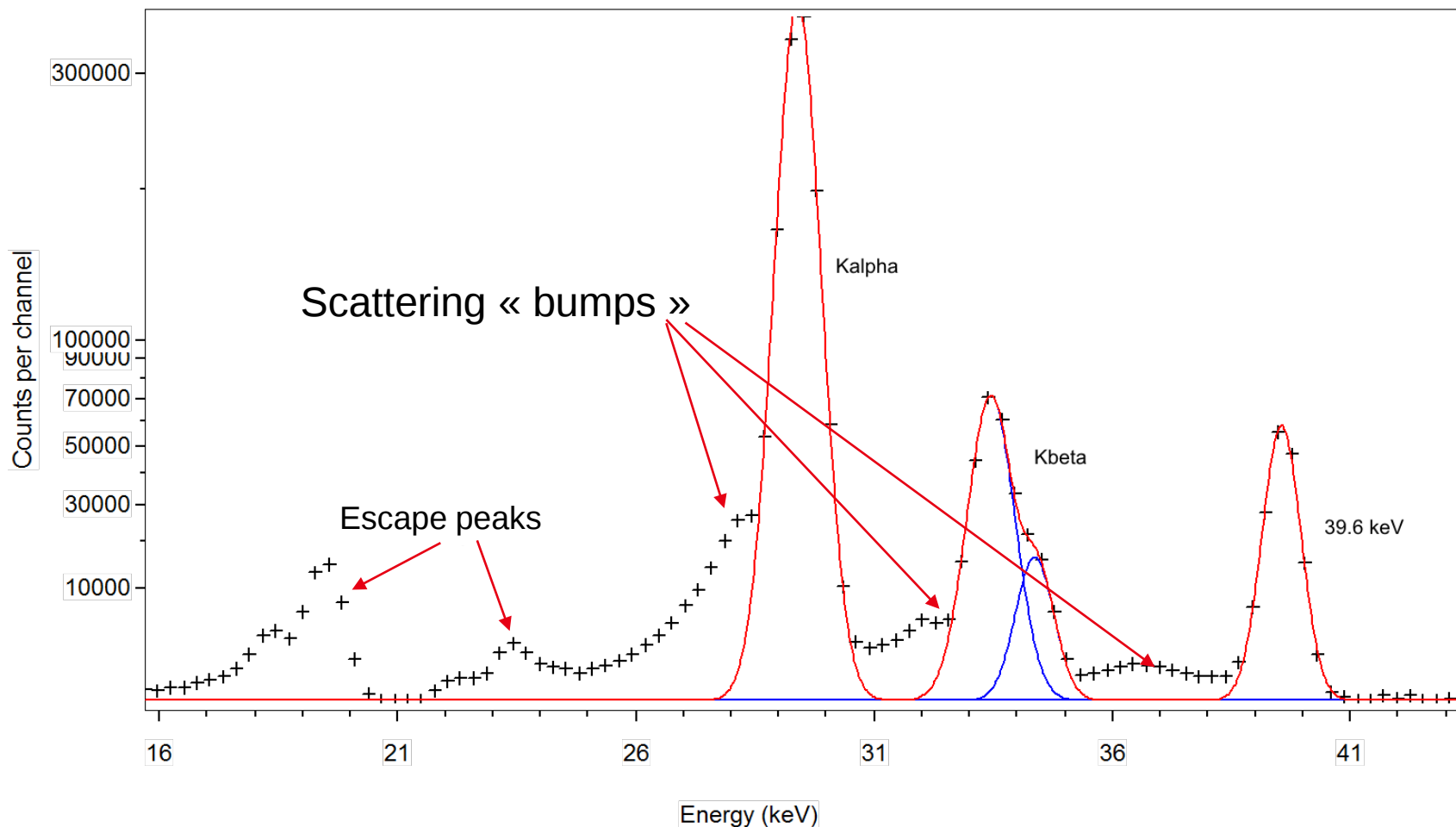
SELF FLUORESCENCE (RARE)



Needs to calculate a corrective factor for this extra-counting
Monte Carlo simulation

Here, about 5% photons/ Bq ^{182}Ta

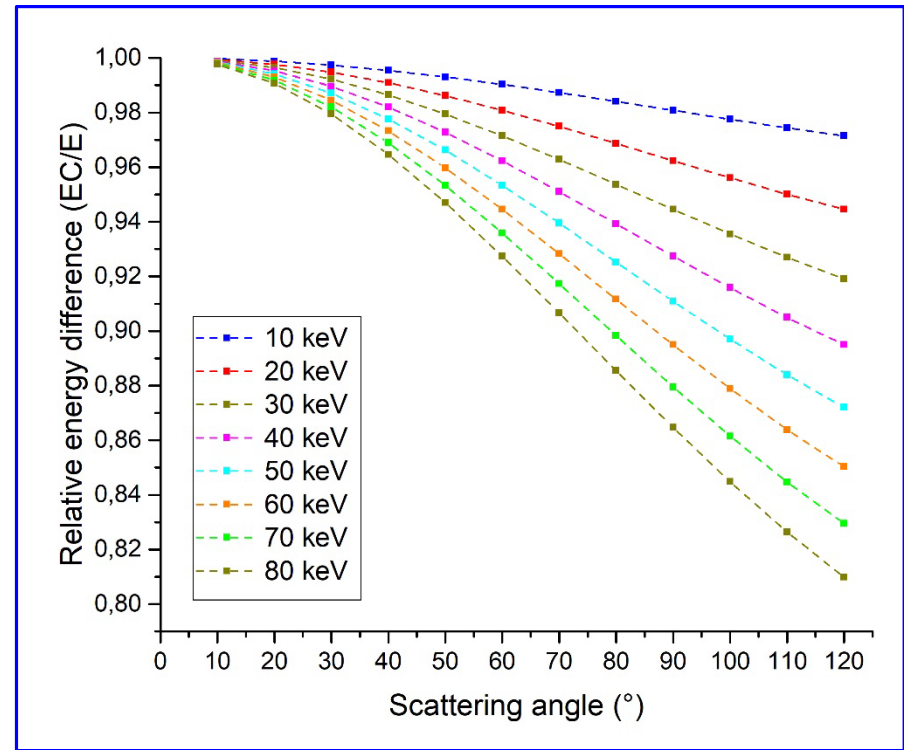
Scattering

Spectrum of ^{129}I 

Position and intensity of « bumps » depend on energy
and on source-detector geometry

$$E_c = \frac{E}{1 + \alpha(1 - \cos \theta)}$$

- E= Energy of incident photon
- E_c = Energy of scattered photon (Compton effect)
- θ = Scattering angle
- $\alpha = \frac{E}{511}$



- This effect should be taken into account but depends on:
 - Energy range,
 - Energy resolution of the spectrometer
 - Source-detector geometry (+ shielding and envionring materials)

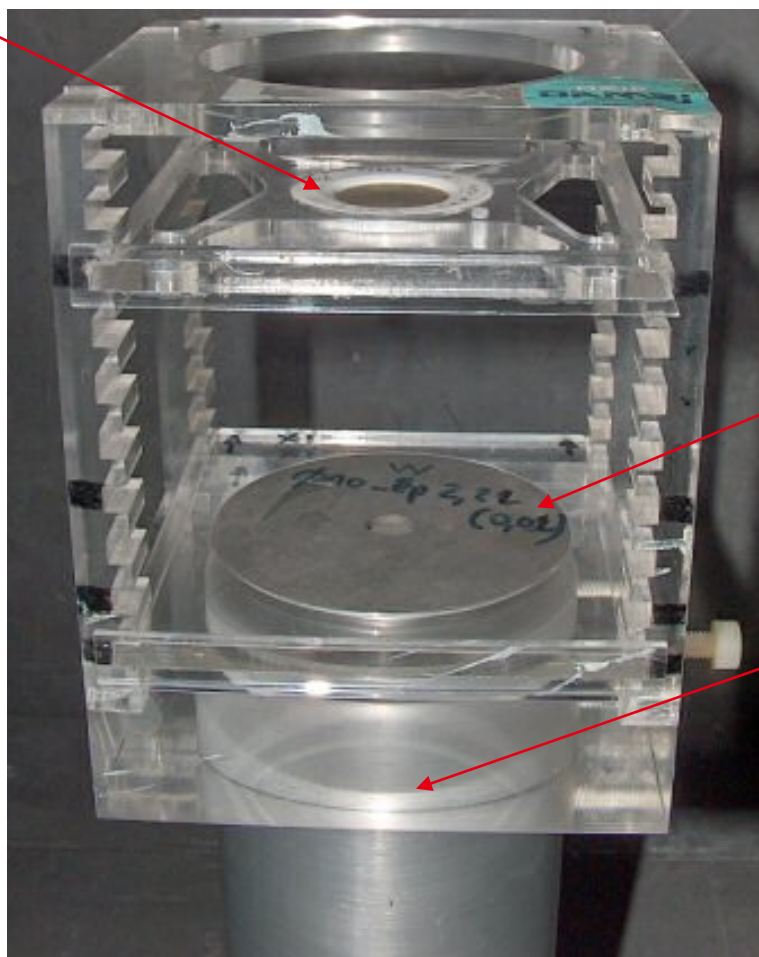
J, Plagnard, C, Hamon, M,C, Lépy, 2008, *Study of scattering effects in low-energy gamma-ray spectrometry*, Applied Radiation and Isotopes 66, 769–773,

EXPERIMENTAL STUDY

Goal : Determine the contribution of elements envrioning the radioactive source to the scattering bump



Successive measurements withdrawing elements around the radioactive source radioactive (^{109}Cd) :



Tungsten collimator

Detector = N-type planar HPGe
Active area = 300 mm²
Thickness = 10 mm
FWHM at 6 keV = 170 eV

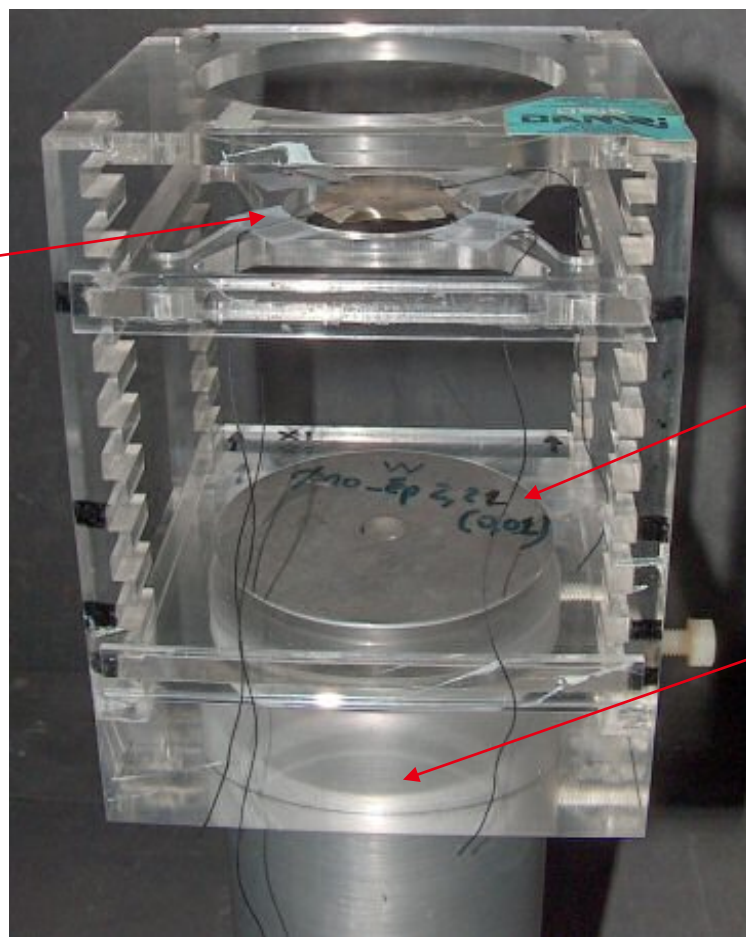
EXPERIMENTAL STUDY

Goal : Determine the contribution of elements envrioning the radioactive source to the scattering bump



Successive measurements withdrawing elements around the radioactive source radioactive (^{109}Cd) :

Source without plastic ring



Tungsten collimator

Detector = N-type planar HPGe
Active area = 300 mm²
Thickness = 10 mm
FWHM at 6 keV = 170 eV

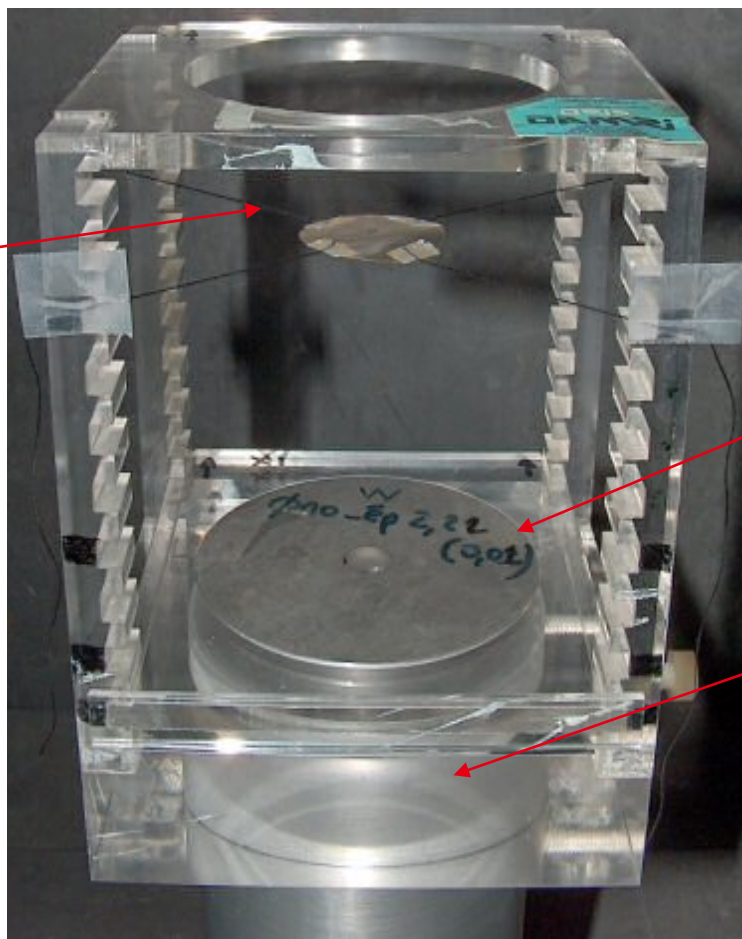
EXPERIMENTAL STUDY

Goal : Determine the contribution of elements envrioning the radioactive source to the scattering bump



Successive measurements withdrawing elements around the radioactive source radioactive (^{109}Cd) :

Source without plastic ring
maintained with wires
(no plastic drawer)



Tungsten collimator

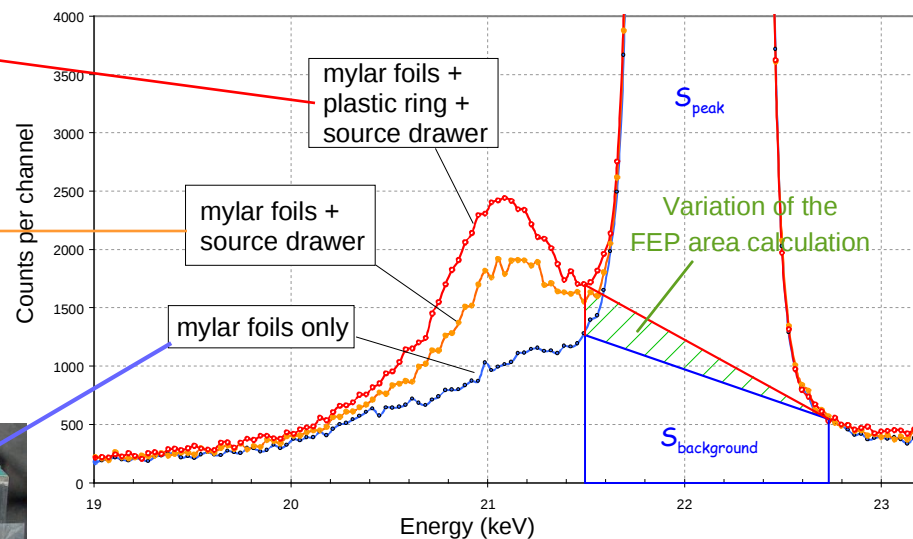
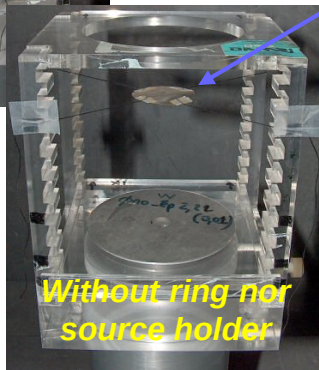
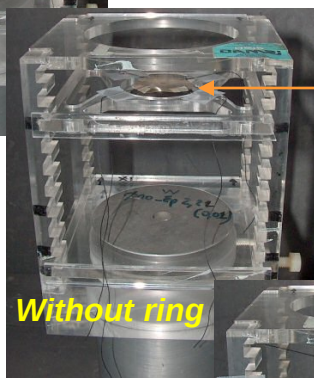
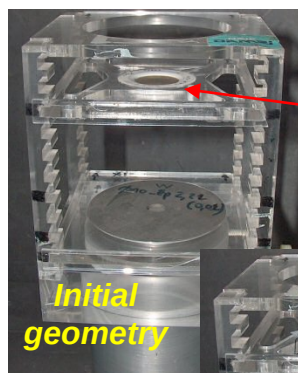
Detector = N-type planar HPGe
Active area = 300 mm²
Thickness = 10 mm
FWHM at 6 keV = 170 eV

EXPERIMENTAL STUDY

Goal : Determine the contribution of elements envrioning the radioactive source to the scattering bump



Successive measurements withdrawing elements around the radioactive source radioactive (^{109}Cd) :



Detector = N-type planar HPGe
Active area = 300 mm²
Thickness = 10 mm
FWHM at 6 keV = 170 eV

Experimental calibration using standard sources (LNHB)

Point sources

Volume sources :

Liquid in plastic container
(standardized geometries: SG15, SG 50 and SG 500)



Samples to be measured:

Volume sources :

different matrices in plastic container

Gaz in stainless steel container

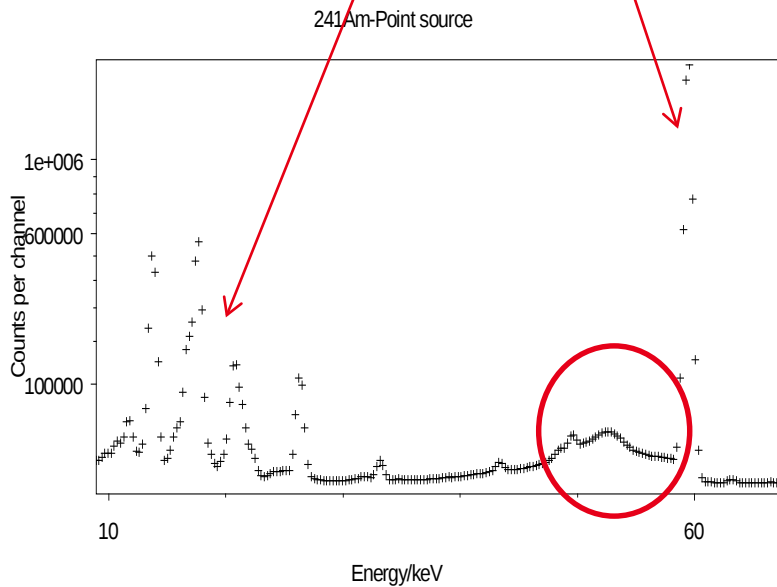


Efficiency transfer ?

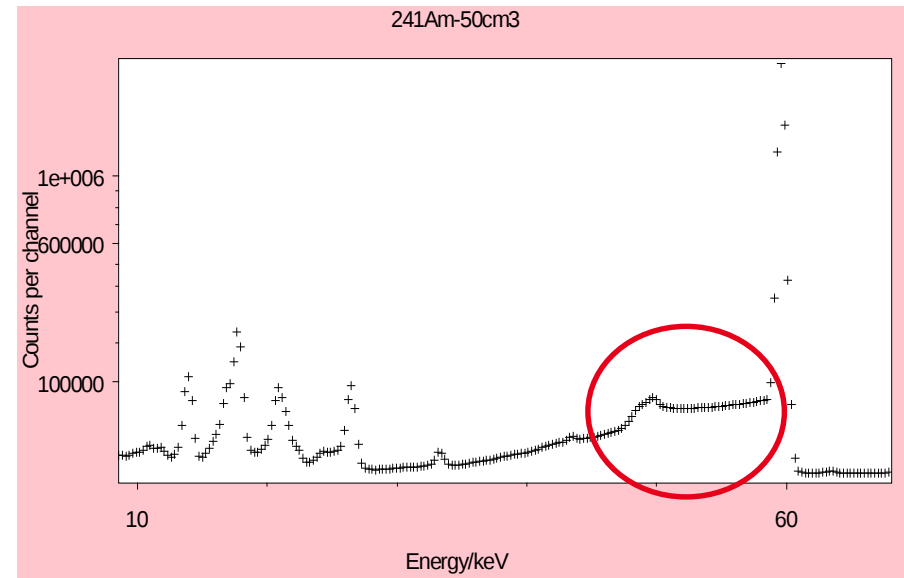
COMPARISON OF SPECTRA

Spectra recorded with standard sources:

^{241}Am : L X-rays + 59.5 keV



Point source at 10 cm



Volume source (50 cm³) at 10 cm

Scattering depends on the geometry

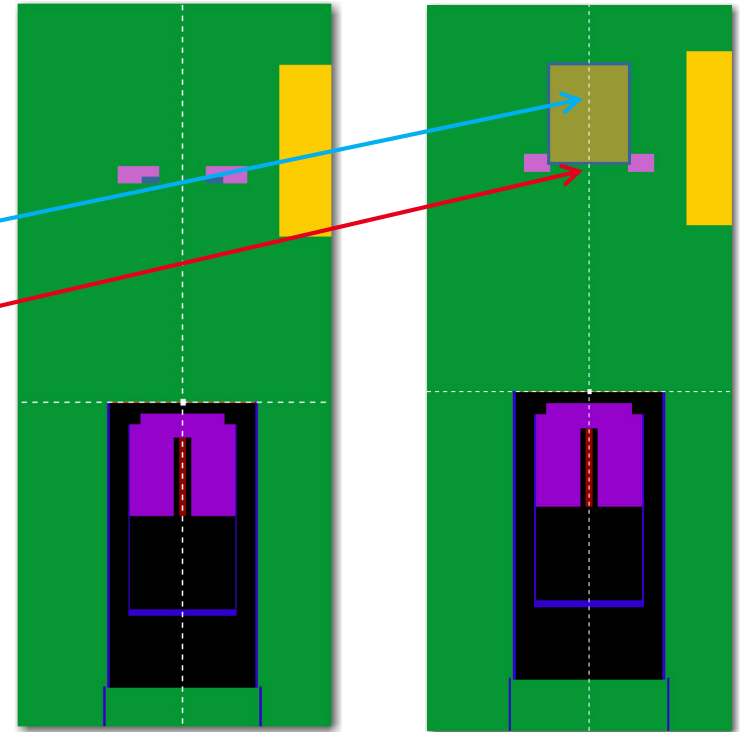
PENELOPE* code

Result : Energy deposition in selected “bodies”

Body 18 : source material (or Mylar® film)

Body 19 : container (or plastic ring)

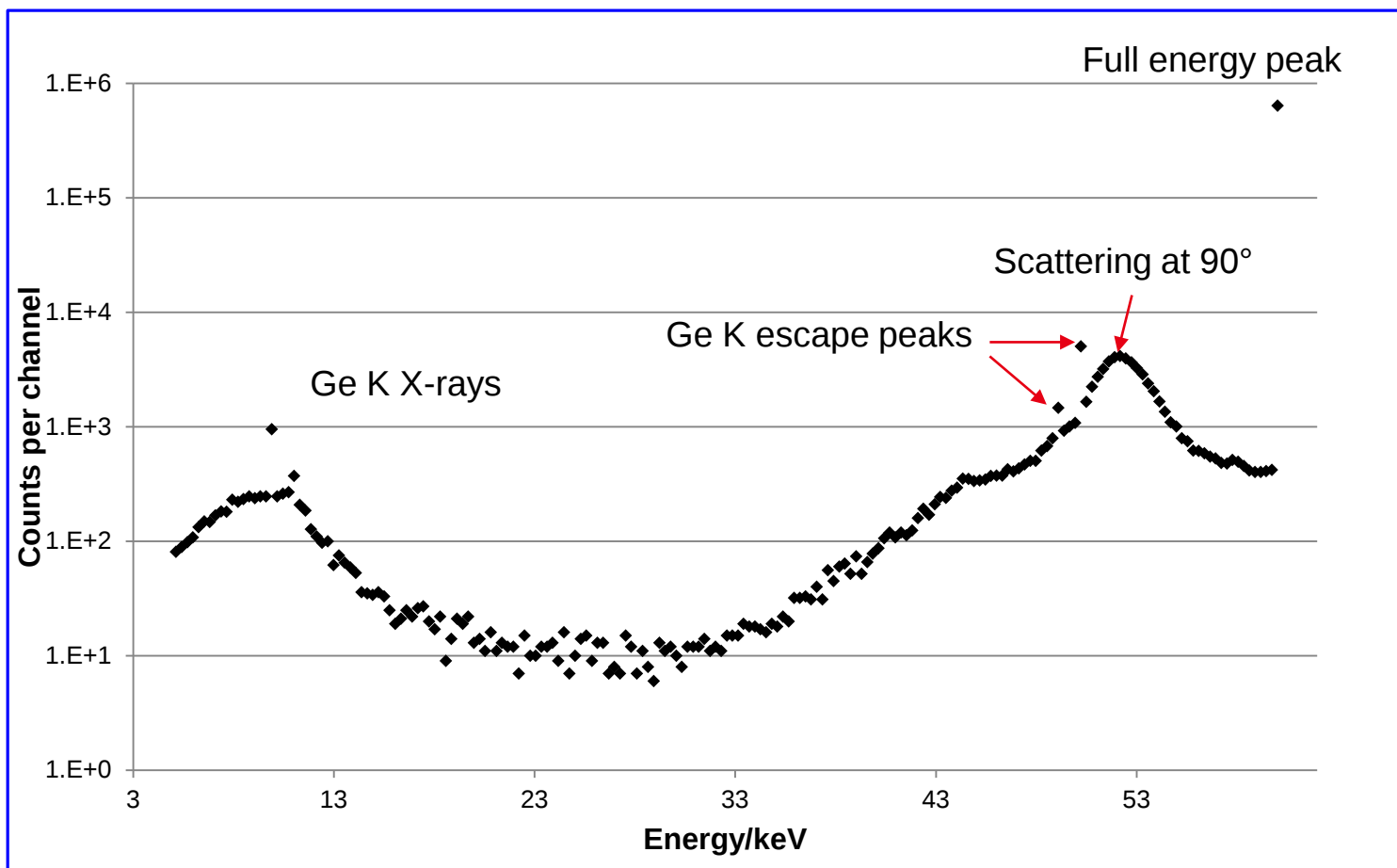
(scattering angles around $\pi/2$)



*Salvat, F. “PENELOPE-2018: A code System for Monte Carlo Simulation of Electron and Photon Transport”
Document NEA/MBDAV/R(2019)1, OECD Nuclear Energy Agency, Boulogne-Billancourt, France, 2019. Available from
<http://www.nea.fr/lists/penelope.html>.

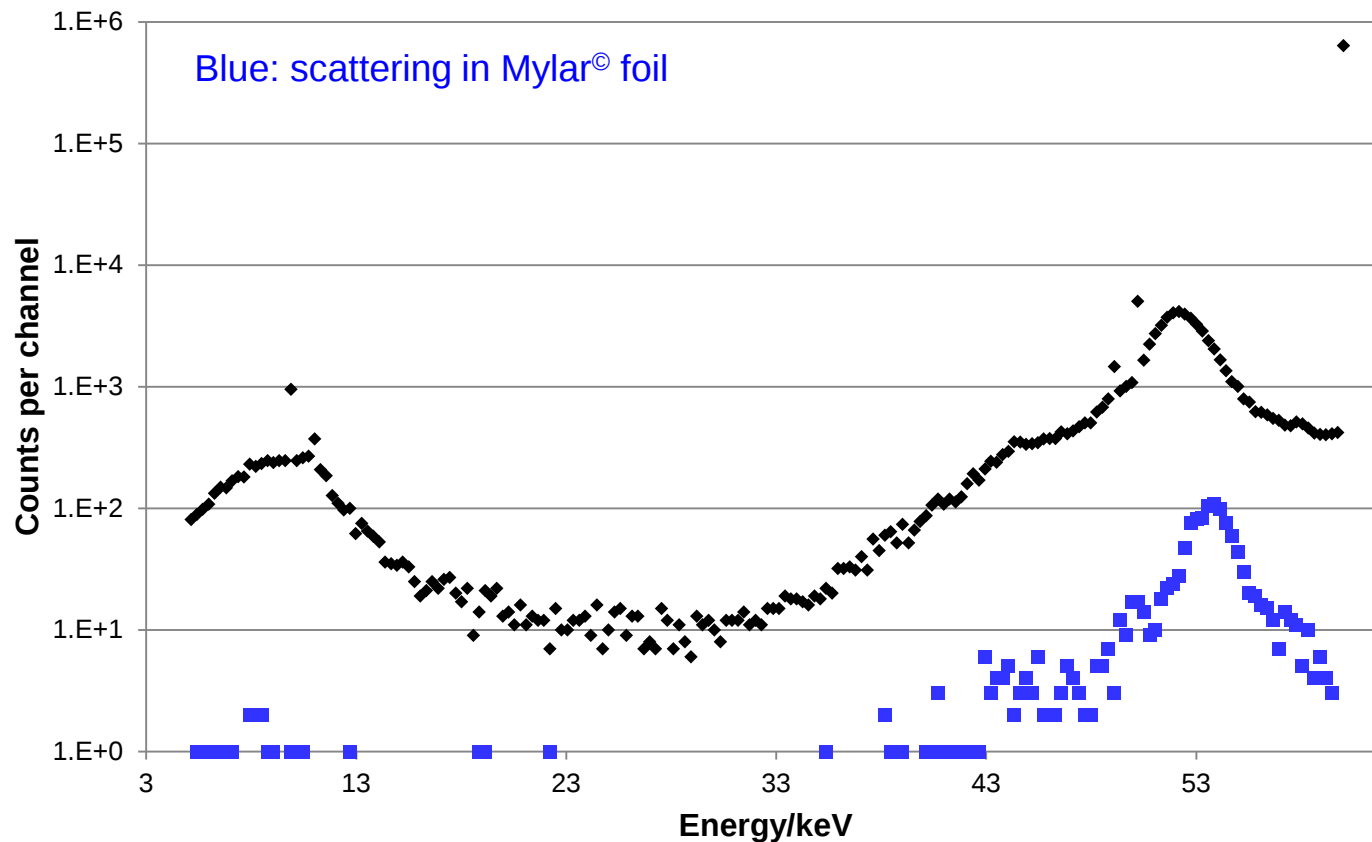
MONTE CARLO SIMULATION

Monte Carlo simulation for 60 keV photons
Point source at 10 cm



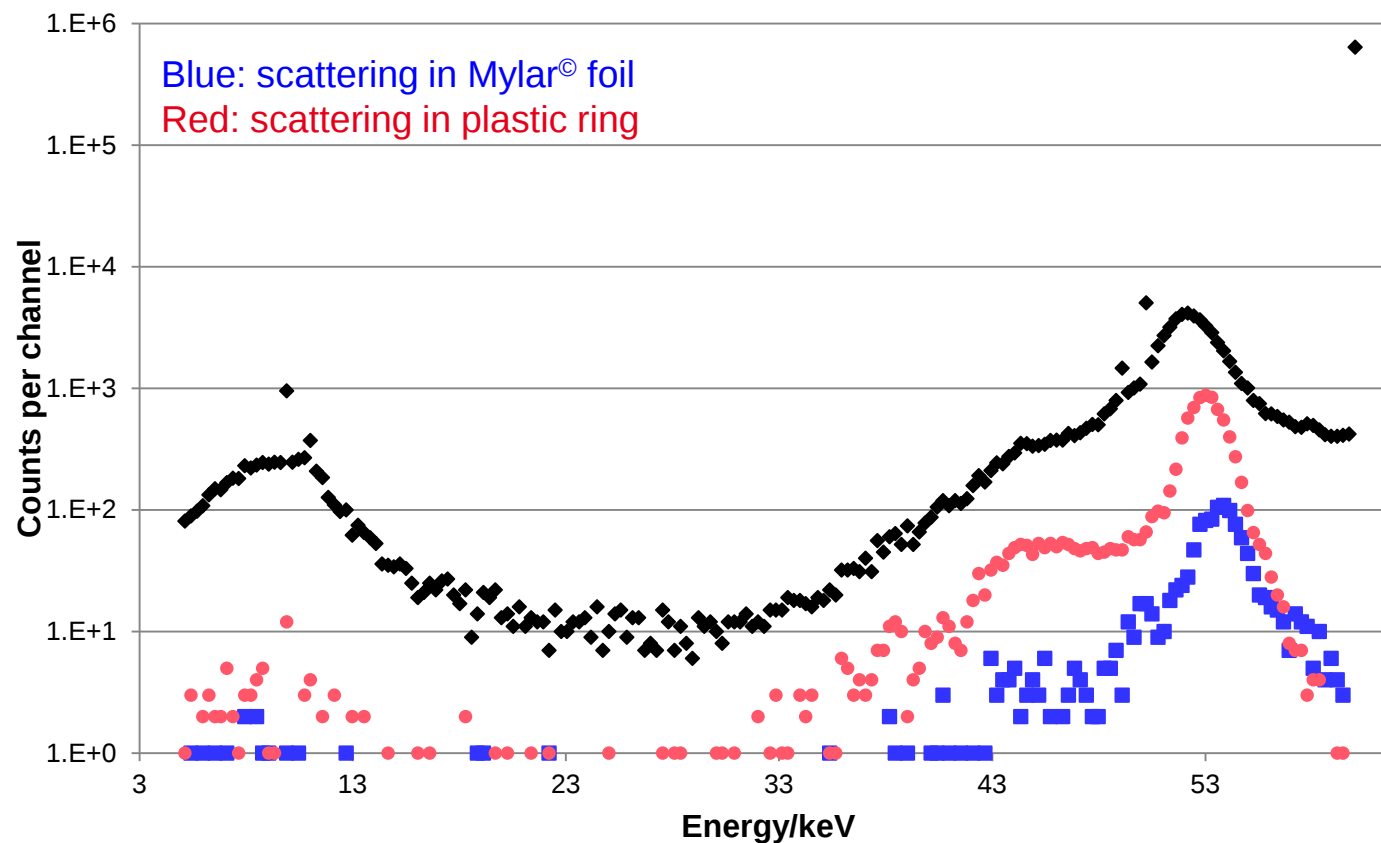
MONTE CARLO SIMULATION

Monte Carlo simulation : 60 keV photons
Point source at 10 cm from the detector window



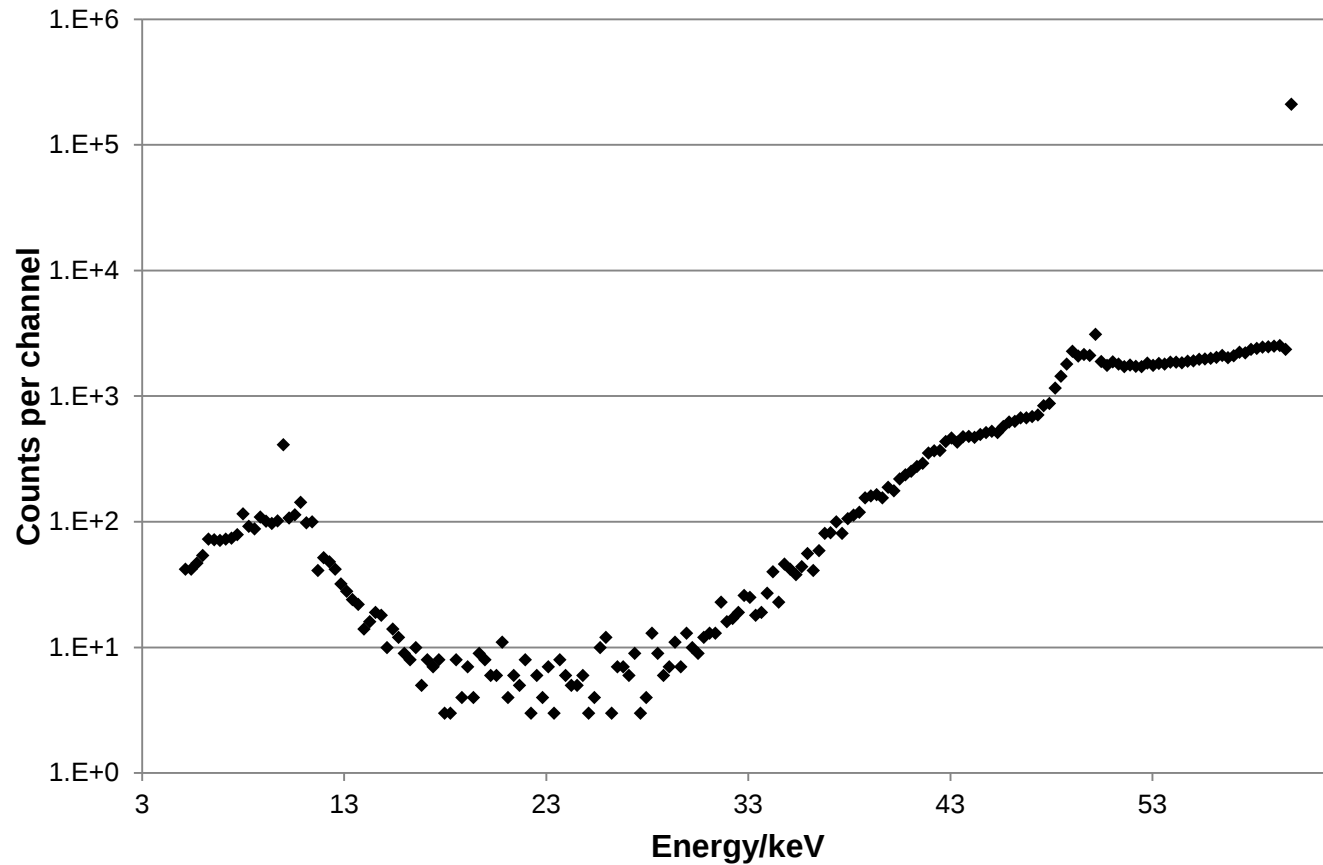
SIMULATION DE MONTE CARLO

Monte Carlo simulation : 60 keV photons
Point source at 10 cm from the detector window



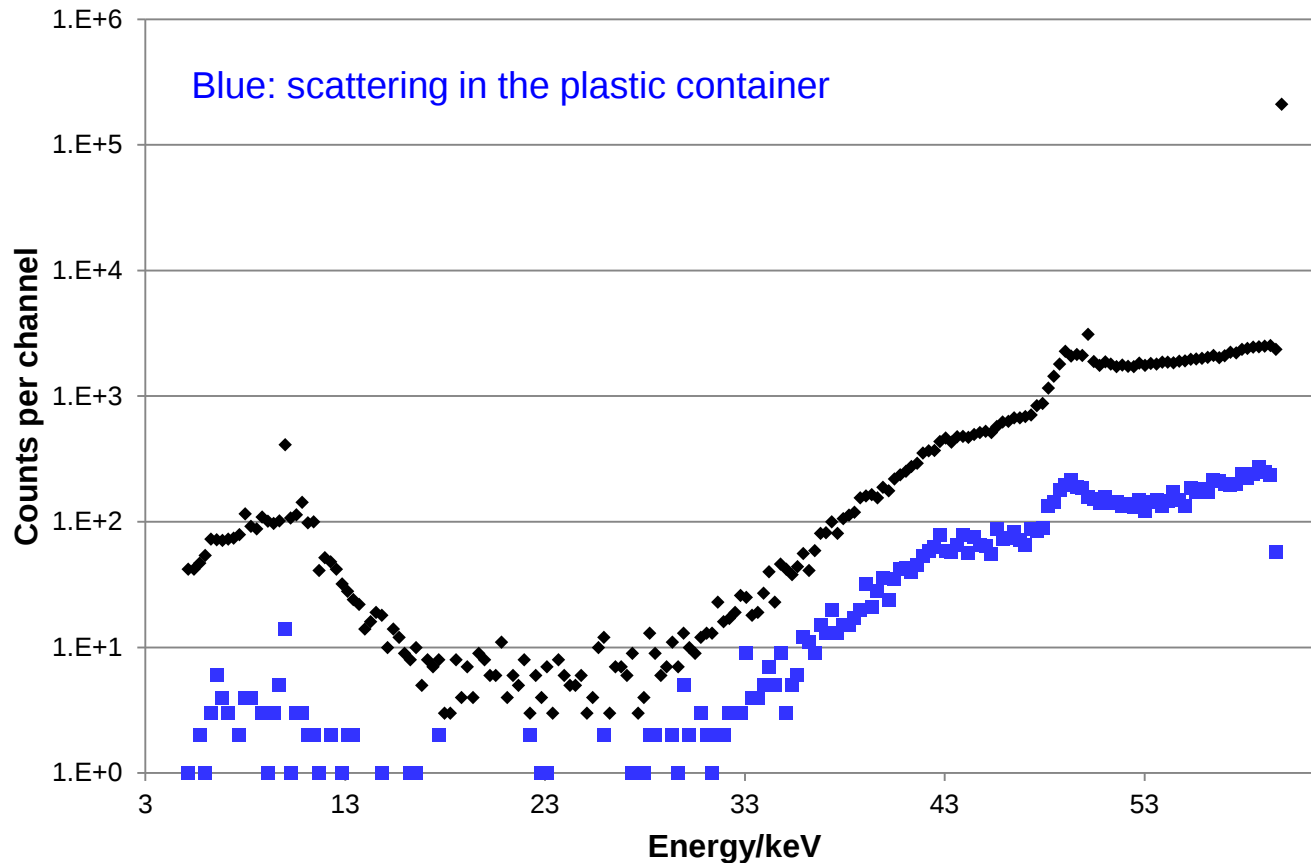
SIMULATION DE MONTE CARLO

Monte Carlo simulation for 60 keV photons
Liquid (H_2O) in a 50 cm³ plastic container at 10 cm from the detector window



SIMULATION DE MONTE CARLO

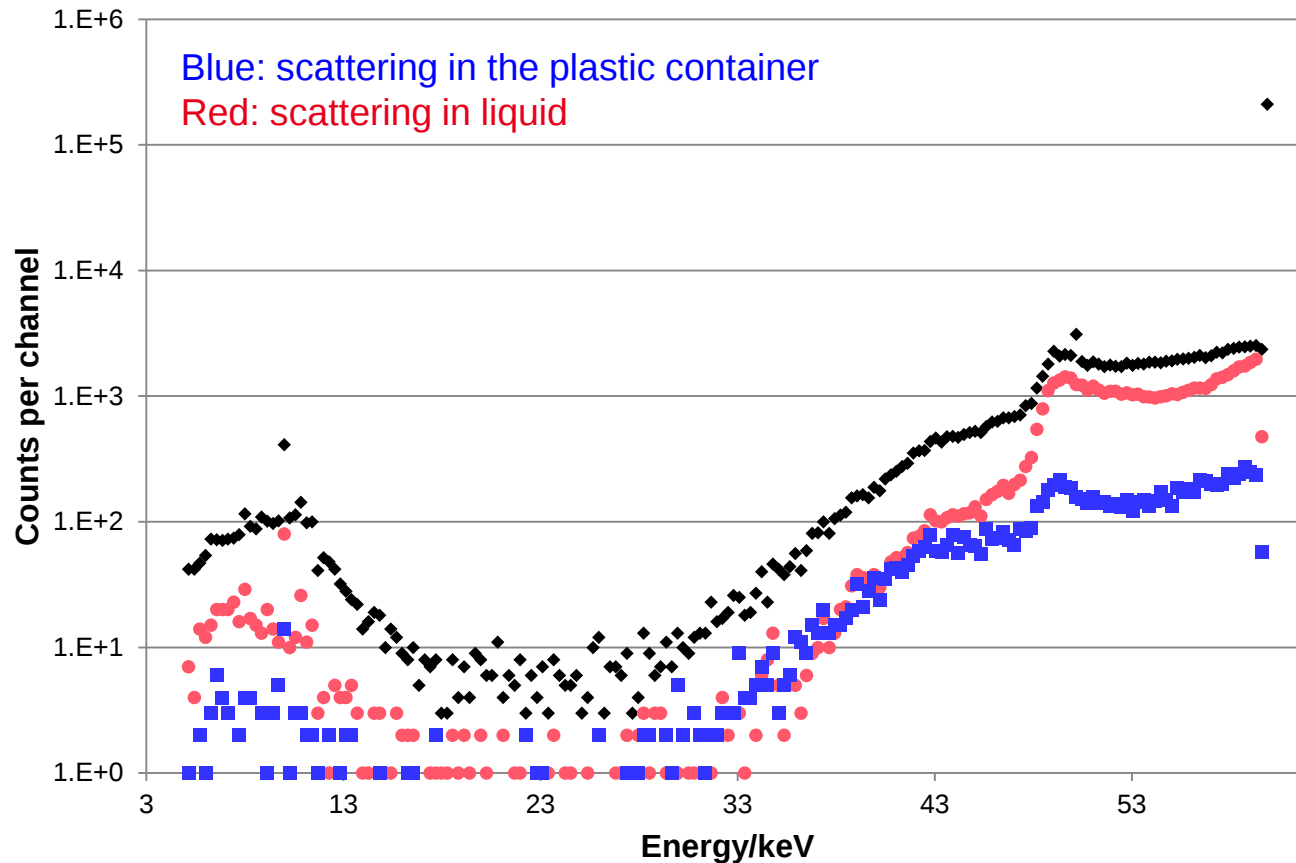
Monte Carlo simulation for 60 keV photons
Liquid (H_2O) in a 50 cm³ plastic container at 10 cm from the detector window



SIMULATION DE MONTE CARLO

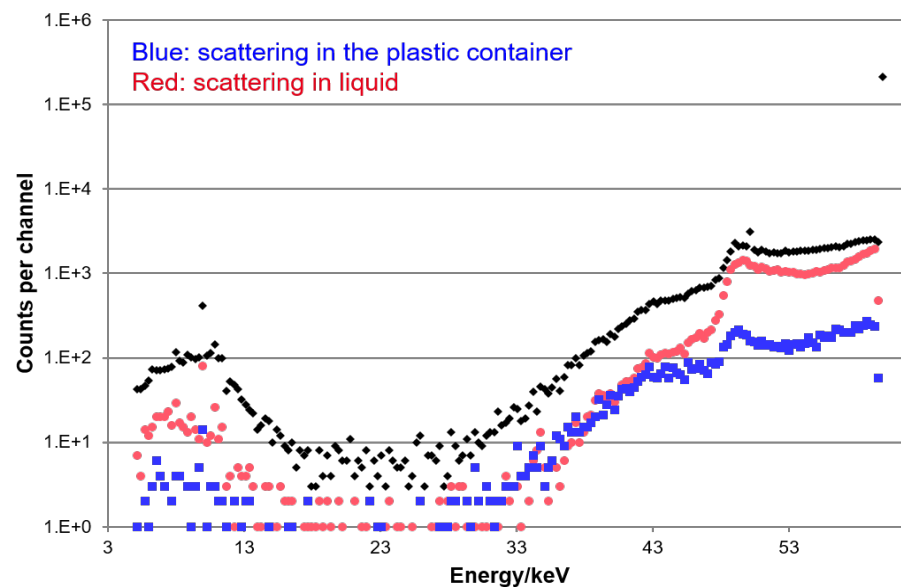
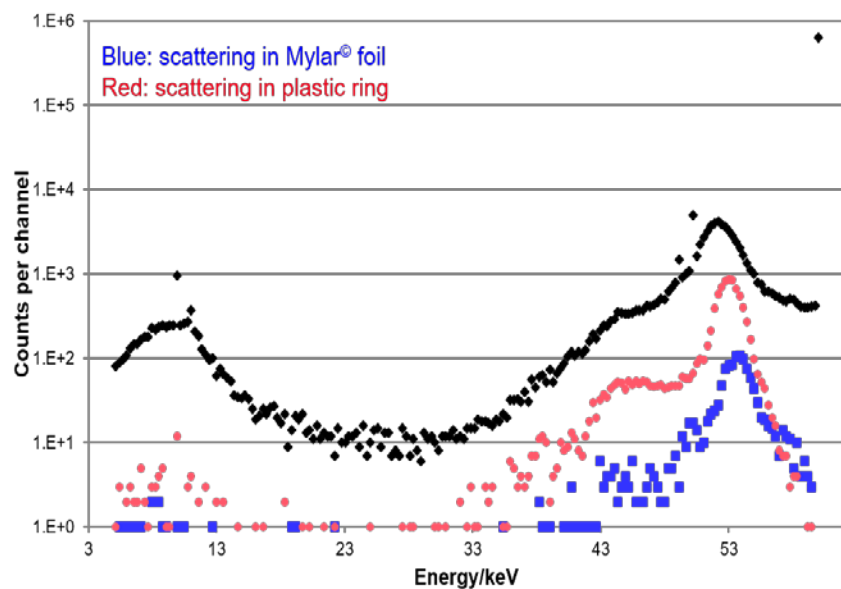
Monte Carlo simulation for 60 keV photons

Liquid (H_2O) in a 50 cm³ plastic container at 10 cm from the detector window

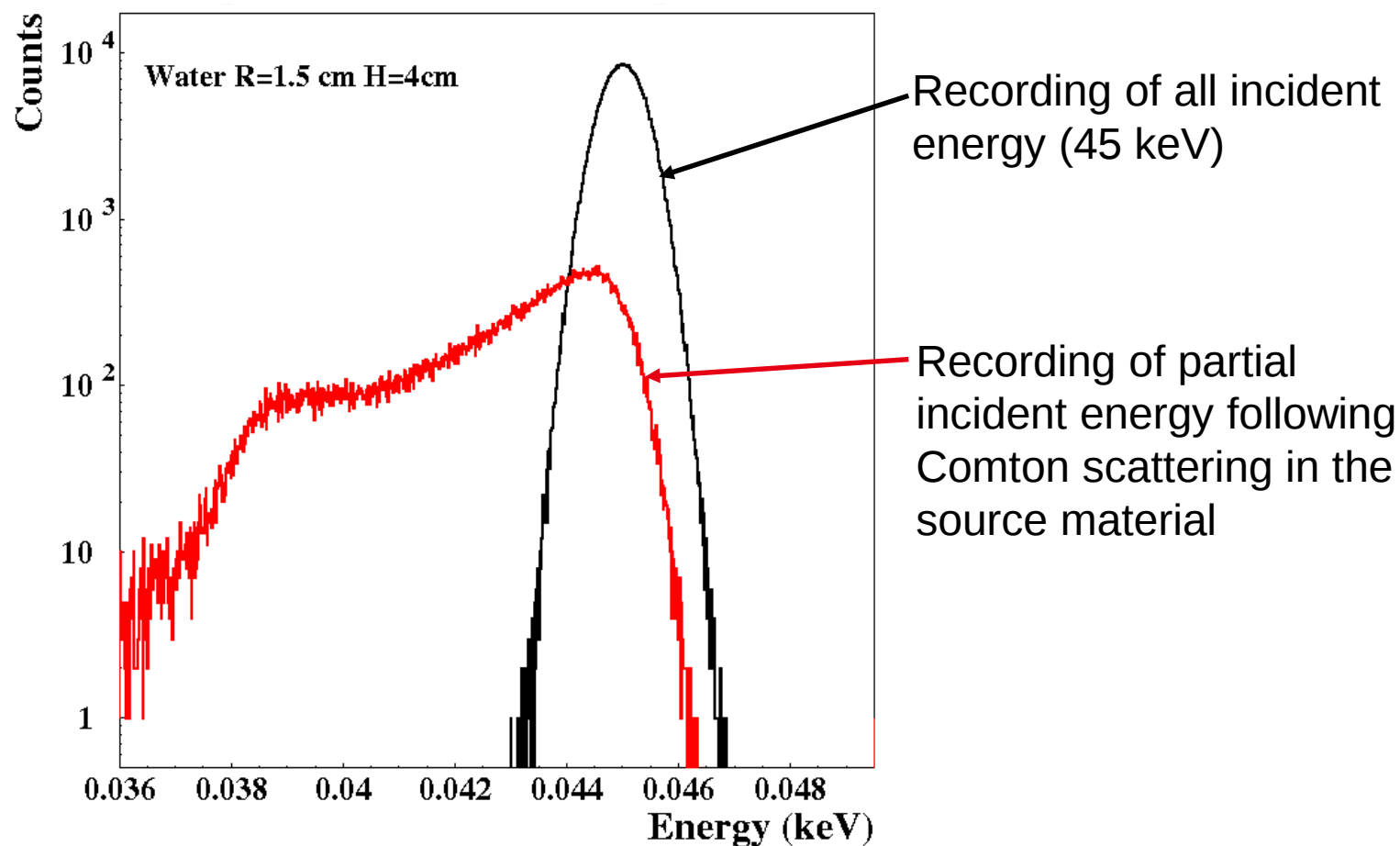


SIMULATION DE MONTE CARLO

Monte Carlo simulation for 60 keV photons



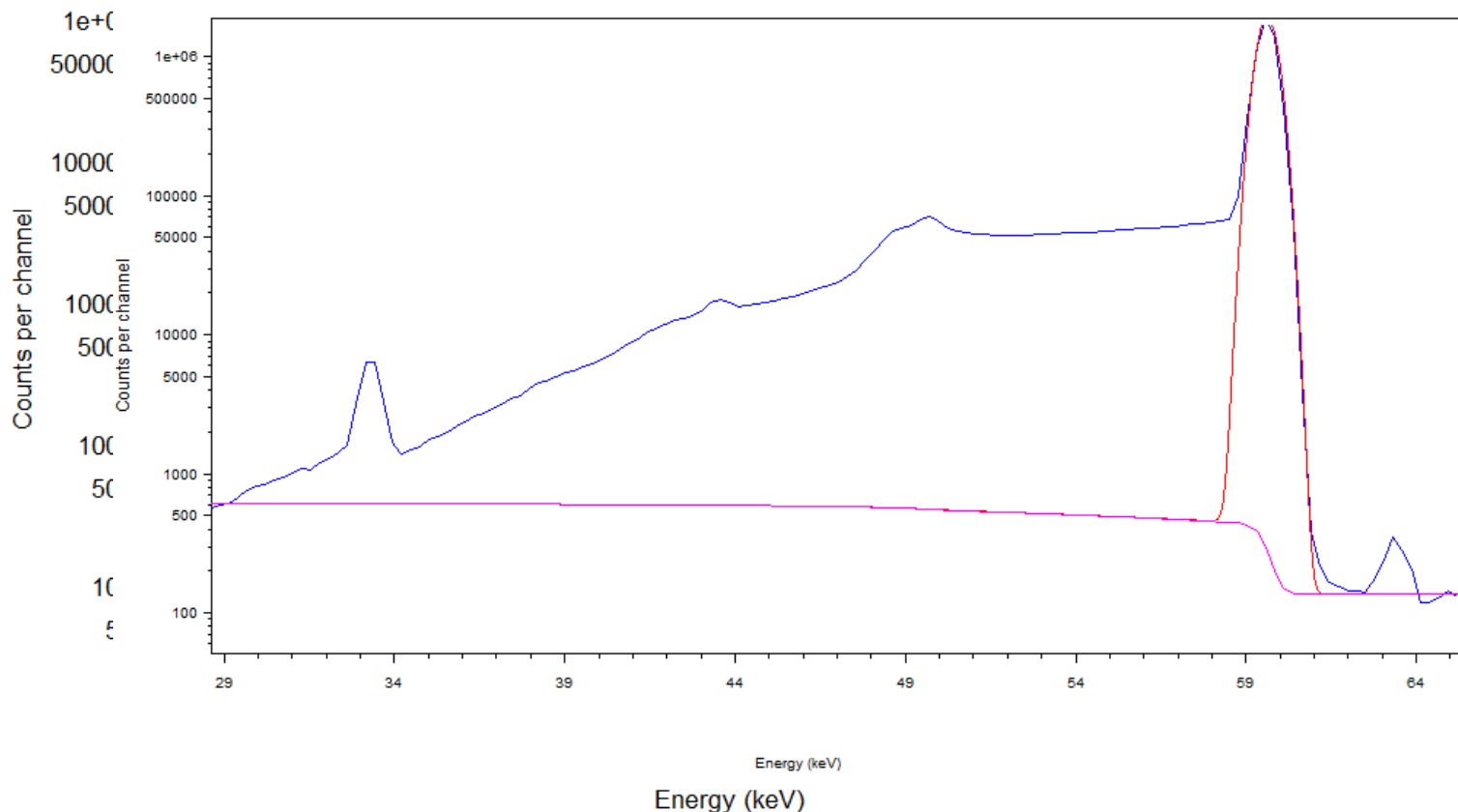
RESULTING SPECTRA



Picture « stolen » to Prof. Octavian Sima (Univ. Bucharest)

HOW TO DETERMINE THE FULL-ENERGY PEAK AREA ?

^{241}Am - Volume source (50 cm^3) at 10 cm



Subtraction of a linear background ? Which limits ?

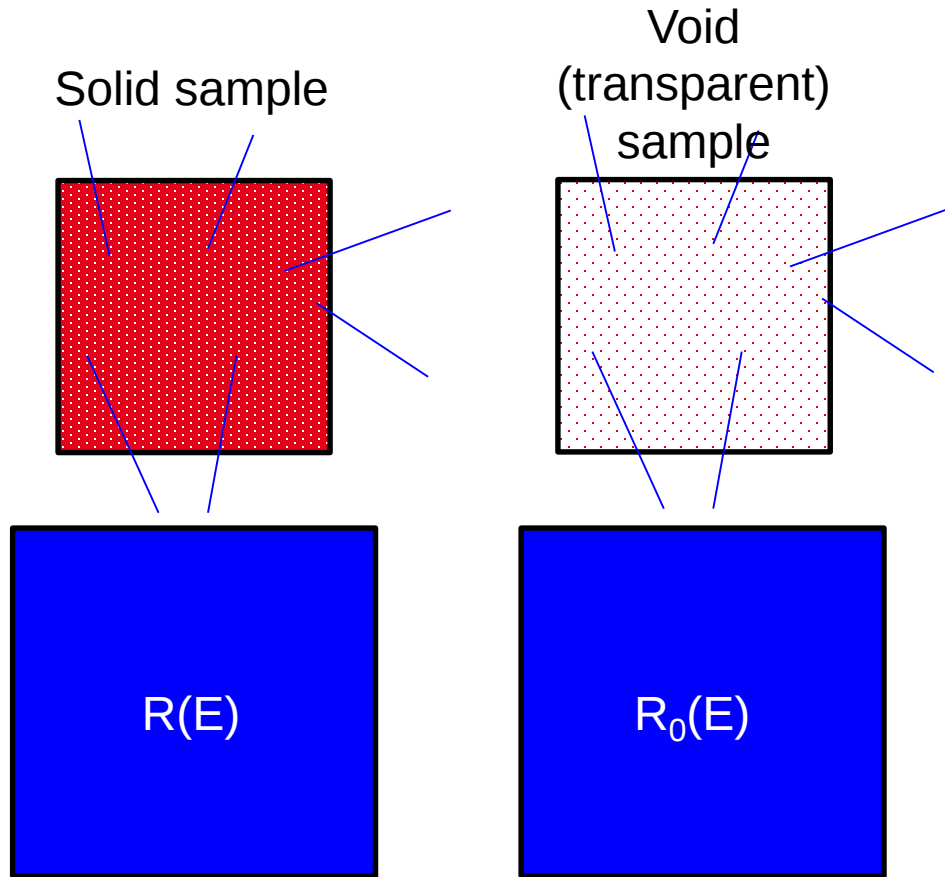
Gross: 5 444 932 → 4 954 364

Gross: 5 748 933 → 5 089 688

Gaussian fitting:
5 276 141

Self-attenuation

Self-attenuation in a volume sample

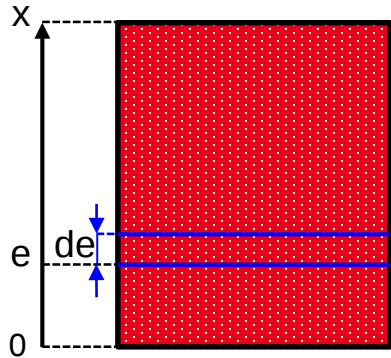


For the solid sample, the number of exiting photons is lower than the number of emitted photons

Count rate for the solid sample
< count rate for a void sample
($R(E) < R_0(E)$)

$$\text{Self-attenuation factor: } F(E, m) = \frac{\varepsilon(E, m)}{\varepsilon(E, 0)}$$

Self-attenuation in a volume sample



Intrinsic photon flux: I_0 (What you wish to know to determine the sample activity)

Exiting photon flux: I (What is recorded by the detector)

For a thin layer, with thickness de : $dI_0 = \frac{I_0}{x} \cdot de$

Only true for an homogeneous material !

This partial photon flux is attenuated through thickness e :

$$dI = \frac{I_0}{x} \cdot \exp(-\mu \cdot e) de$$

μ : linear attenuation coefficient (cm^{-1})

For the whole volume with thickness x :

$$I = \frac{I_0}{x} \int_0^x \exp(-\mu \cdot e) de = I_0 \cdot \frac{1 - \exp(-\mu \cdot x)}{\mu \cdot x}$$

Self-attenuation in a volume sample

Intrinsic photon flux: I_0 (What you wish to know to determine the sample activity)

Exiting photon flux: I (What is recorded by the detector)

$$I = I_0 \cdot \frac{1 - \exp(-\mu \cdot x)}{\mu \cdot x}$$

Self-attenuation factor: $C_{att} = \frac{1 - \exp(-\mu \cdot x)}{\mu \cdot x}$

Approximation for a thin source ($\mu \cdot x < 1$):

$$I = I_0 \cdot \frac{1 - \mu \cdot x}{2}$$

$$C_{att} = \frac{1 - \mu \cdot x}{2}$$

Self-attenuation in a volume sample

Self-attenuation factor:
$$C_{att} = \frac{1 - \exp(-\mu \cdot x)}{\mu \cdot x}$$

If the measured sample is subject to attenuation and the calibration source is not, a correction factor must be applied **to the peak area** that is : C_{att}^{-1}

$$C_{self} = C_{att}^{-1} = \frac{\mu \cdot x}{1 - \exp(-\mu \cdot x)}$$

If both are subject to self-attenuation, the correction factor to the peak area of the measured sample is the ratio of the self attenuation for each material:

$$C_{self} = \frac{(C_{att})_{mes}^{-1}}{(C_{att})_{cal}^{-1}} = \frac{\left[\frac{\mu_{mes} \cdot x_{mes}}{1 - \exp(-\mu_{mes} \cdot x_{mes})} \right]}{\left[\frac{\mu_{cal} \cdot x_{cal}}{1 - \exp(-\mu_{cal} \cdot x_{cal})} \right]} = \frac{\mu_{mes} \cdot x_{mes}}{\mu_{cal} \cdot x_{cal}} \cdot \frac{1 - \exp(-\mu_{cal} \cdot x_{cal})}{1 - \exp(-\mu_{mes} \cdot x_{mes})}$$

Mes: measured sample
Cal: calibration source

Self-attenuation in a volume sample

Self-attenuation factor:
$$C_{att} = \frac{1 - \exp(-\mu \cdot x)}{\mu \cdot x}$$

Can also be used to compute the transfer factor from an efficiency calibration established reference material to provide efficiency for measurement in different geometry

$$\varepsilon_{mes} = \varepsilon_{cal} \cdot f_{self}$$

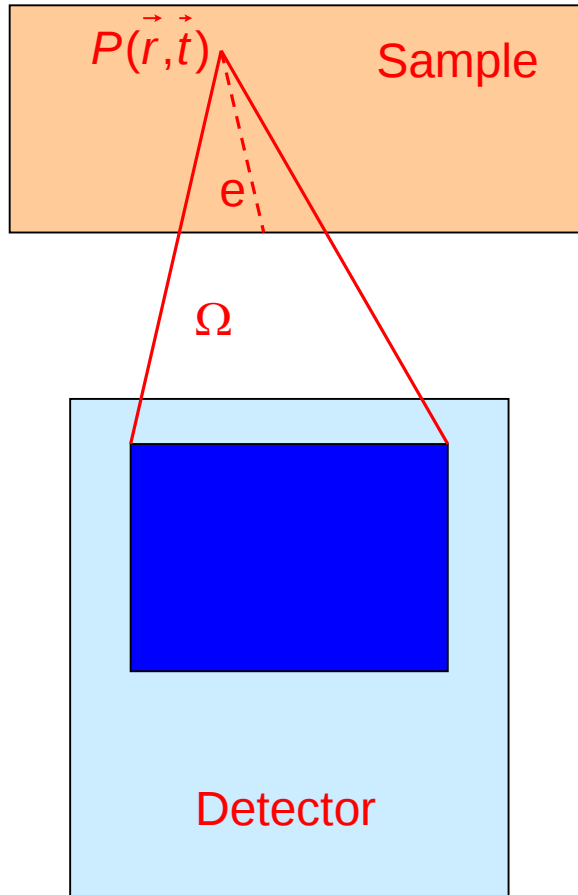
Thus the **efficiency transfer factor** is:

$$f_{self} = \frac{(C_{att})_{mes}}{(C_{att})_{cal}} = \frac{\left[\frac{1 - \exp(-\mu_{mes} \cdot x_{mes})}{\mu_{mes} \cdot x_{mes}} \right]}{\left[\frac{1 - \exp(-\mu_{cal} \cdot x_{cal})}{\mu_{cal} \cdot x_{cal}} \right]} = \frac{\mu_{cal} \cdot x_{cal}}{\mu_{mes} \cdot x_{mes}} \cdot \frac{1 - \exp(-\mu_{mes} \cdot x_{mes})}{1 - \exp(-\mu_{cal} \cdot x_{cal})}$$

Mes: measured sample
Cal: calibration source

SELF-ATTENUATION: GENERAL FORMULA

All possible trajectories for each point of the volume sample must be taken into account → integration over solid angle and sample volume:



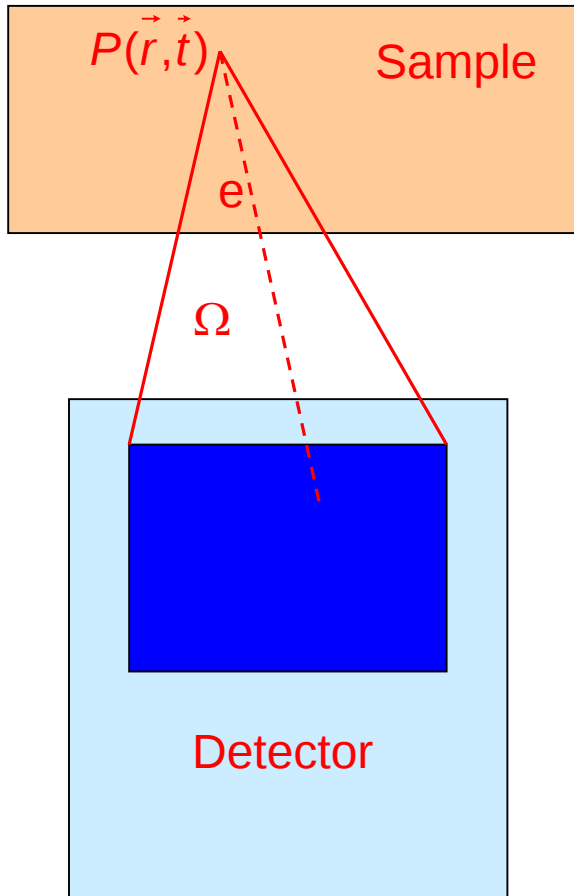
$$C_{att} = \frac{\int_V dV \int_{\Omega} \exp(-\mu(E) \cdot e(\vec{r} \cdot \vec{t})) \cdot d\Omega}{\int_V dV \int_{\Omega} d\Omega}$$

Point P with position $\vec{r}$, and emission direction $\vec{t}$
e: path in the sample matrix

Add the container absorption and probability of full-absorption of the photon energy in the detector active volume

SELF-ATTENUATION: GENERAL FORMULA

Taking into account probability of absorption in the detector:



$$C_{att} = \frac{\int_V dV \int_{\Omega} \exp(-\mu(E) \cdot e(\vec{r} \cdot \vec{t})) \cdot T(E, \vec{r}, \vec{t}) \cdot P(E, \vec{r}, \vec{t}) \cdot d\Omega}{\int_V dV \int_{\Omega} T(E, \vec{r}, \vec{t}) \cdot P(E, \vec{r}, \vec{t}) \cdot d\Omega}$$

Denominator = « self attenuation » for a transparent sample

$\mu(E)$: Attenuation coefficient of the sample material for energy E

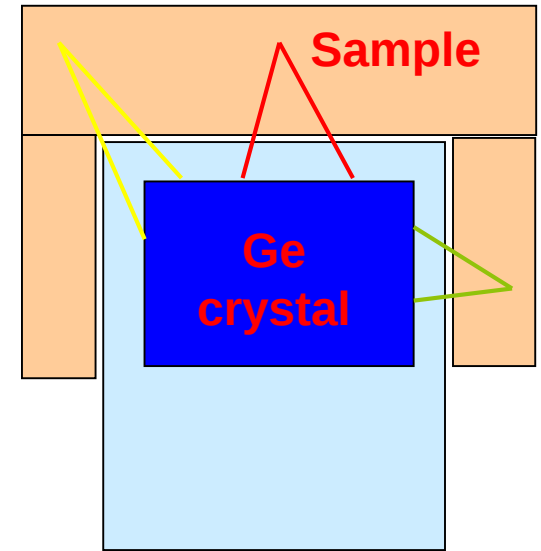
e : Path through the sample

T : Transmission through absorbers (container, detector window, etc.)

P : Probability of full-energy absorption in the detector

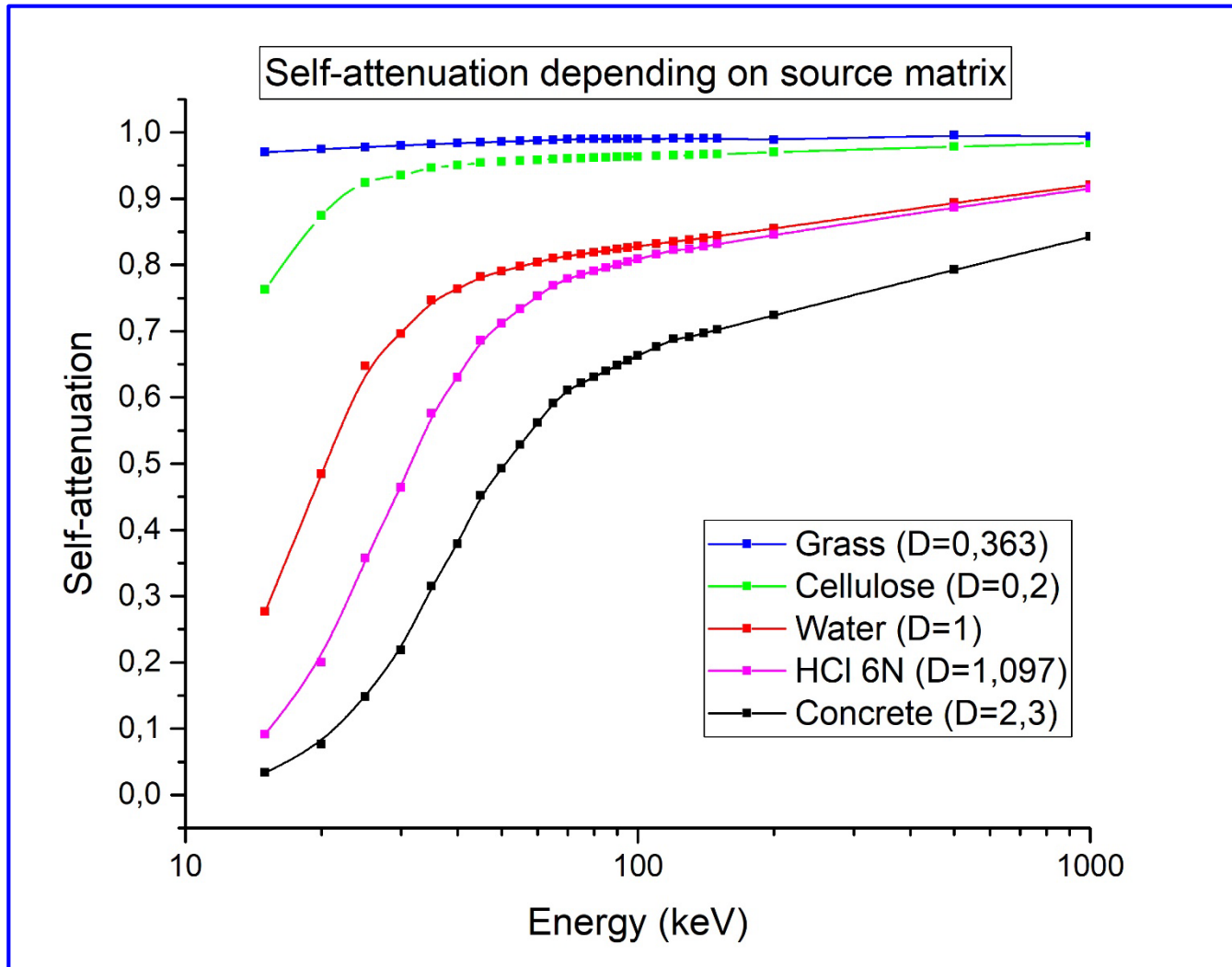
MONTE CARLO SIMULATION

- Self attenuation can be computed using Monte Carlo simulation
 - General codes (GEANT, MCNP, PENELOPE, etc.)
 - Dedicated software (DETEFF, EFFTRAN, GESPECOR, etc.)
- Any geometry (including non-cylindrical symmetry) can be considered
- Can include scattering in the source and environing materials
- Time-consuming? Dedicated software are optimized!



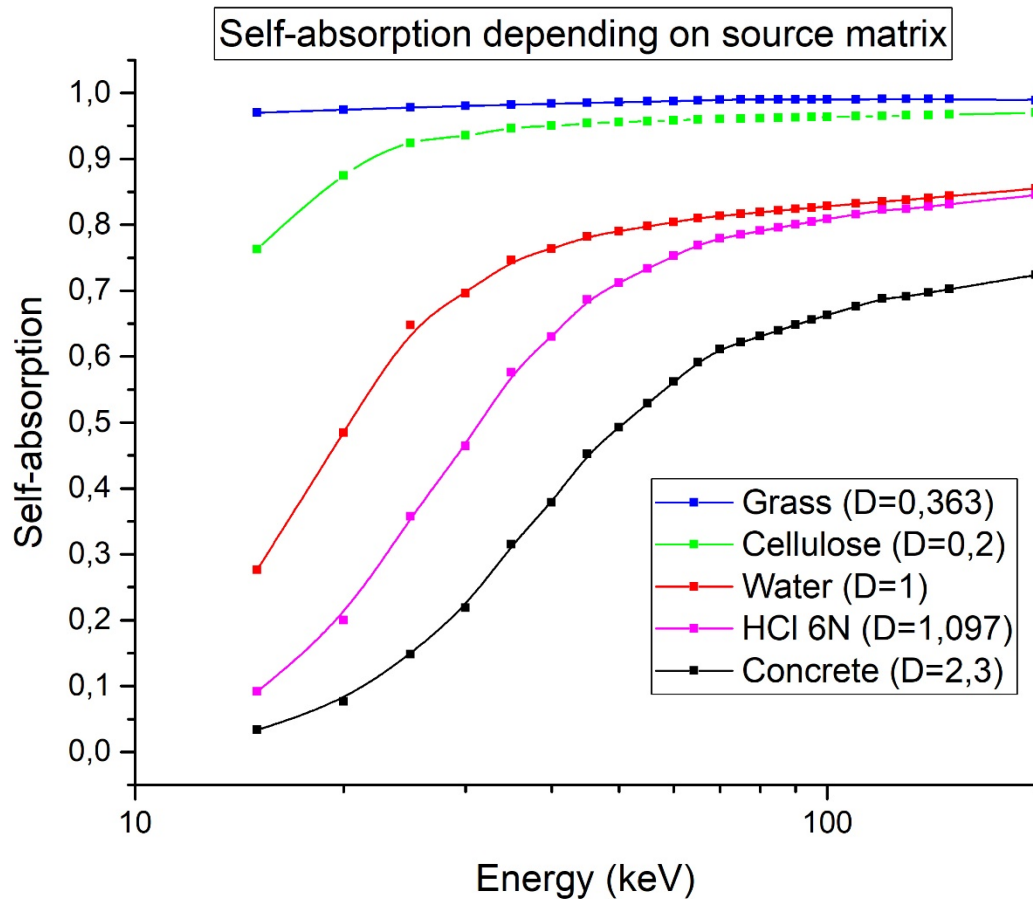
SELF-ATTENUATION: EXAMPLES

Volume source at 10 cm



SELF-ATTENUATION: EXAMPLES

Volume source at 10 cm (zoom)



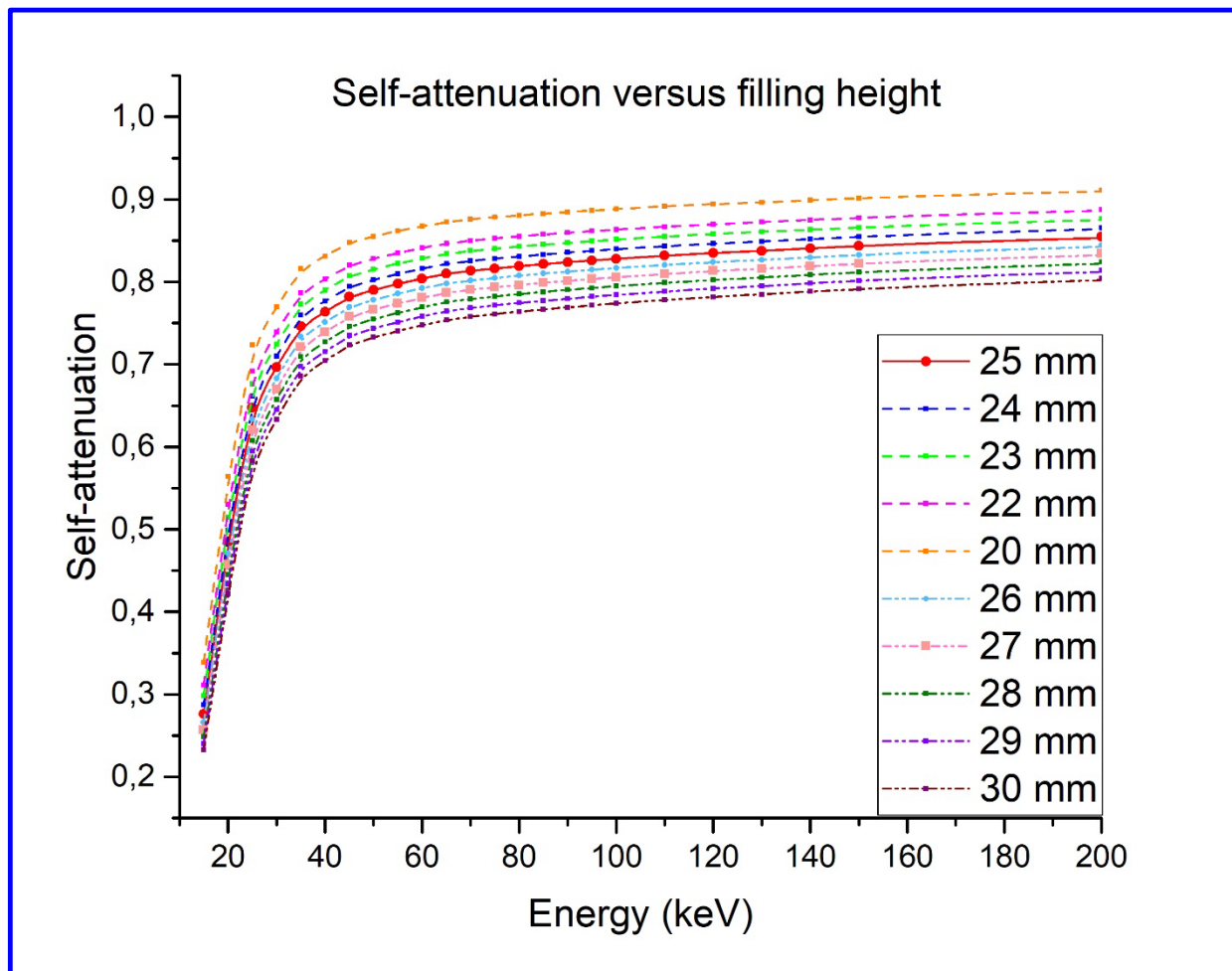
Water source at 10 cm

Relative difference for 2 mm change

100 keV: 2.8%

50 keV: 3%

20 keV : 5%-6%

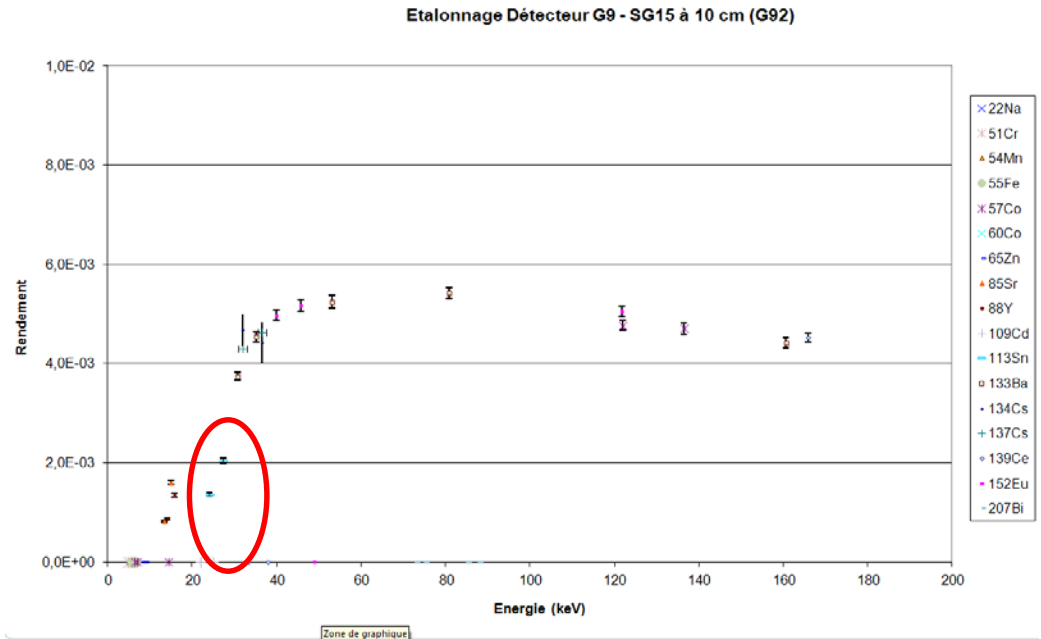


SELF-ATTENUATION IN THE SOURCE MATERIAL

For volume samples: attenuation of emitted photons within the source
Example: calibration using liquid sources (50 cm³ vials)

Chemical composition of the liquid solution depending on the radionuclide (chemical element for stability): generally HCl 0.1N.

Reference	HCl 0.1 N
⁷⁵ Se, ¹²⁹ I, ¹³¹ I	H ₂ O
¹⁰⁹ Cd	HCl 1N
¹¹³ Sn	HCl 6N
¹²⁵ Sb	HCl 2N
¹⁵² Eu, ^{166m} Ho	HCl 1N
²⁴¹ Am, ²³⁹ Pu	HNO ₃ 1N
²¹⁰ Pb	HNO ₃ 0.1N

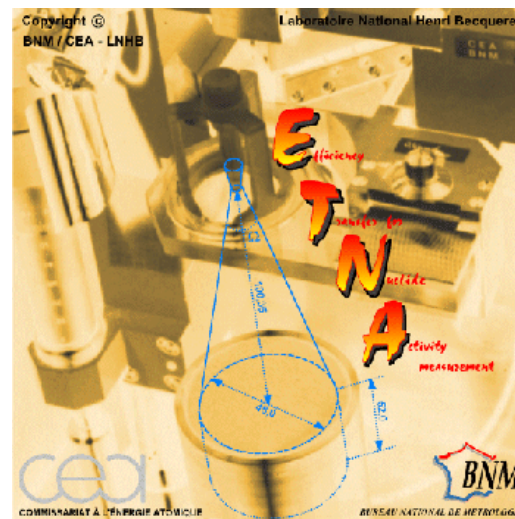
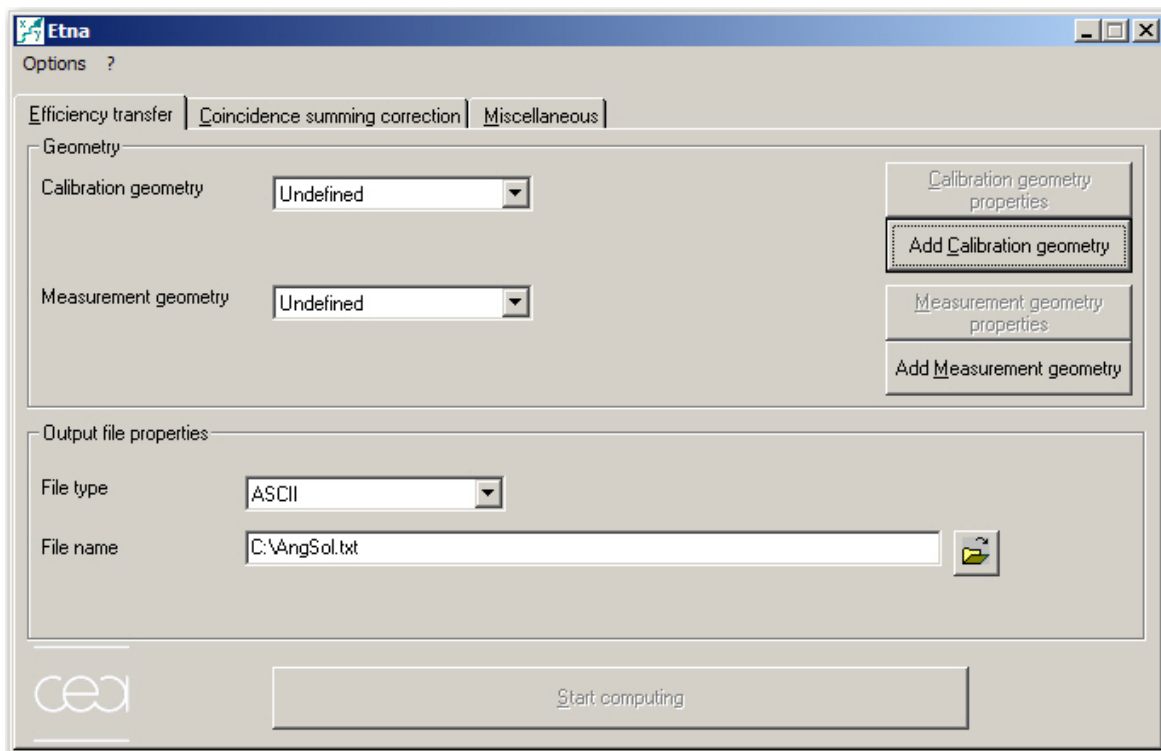


Important for energies < 200 keV

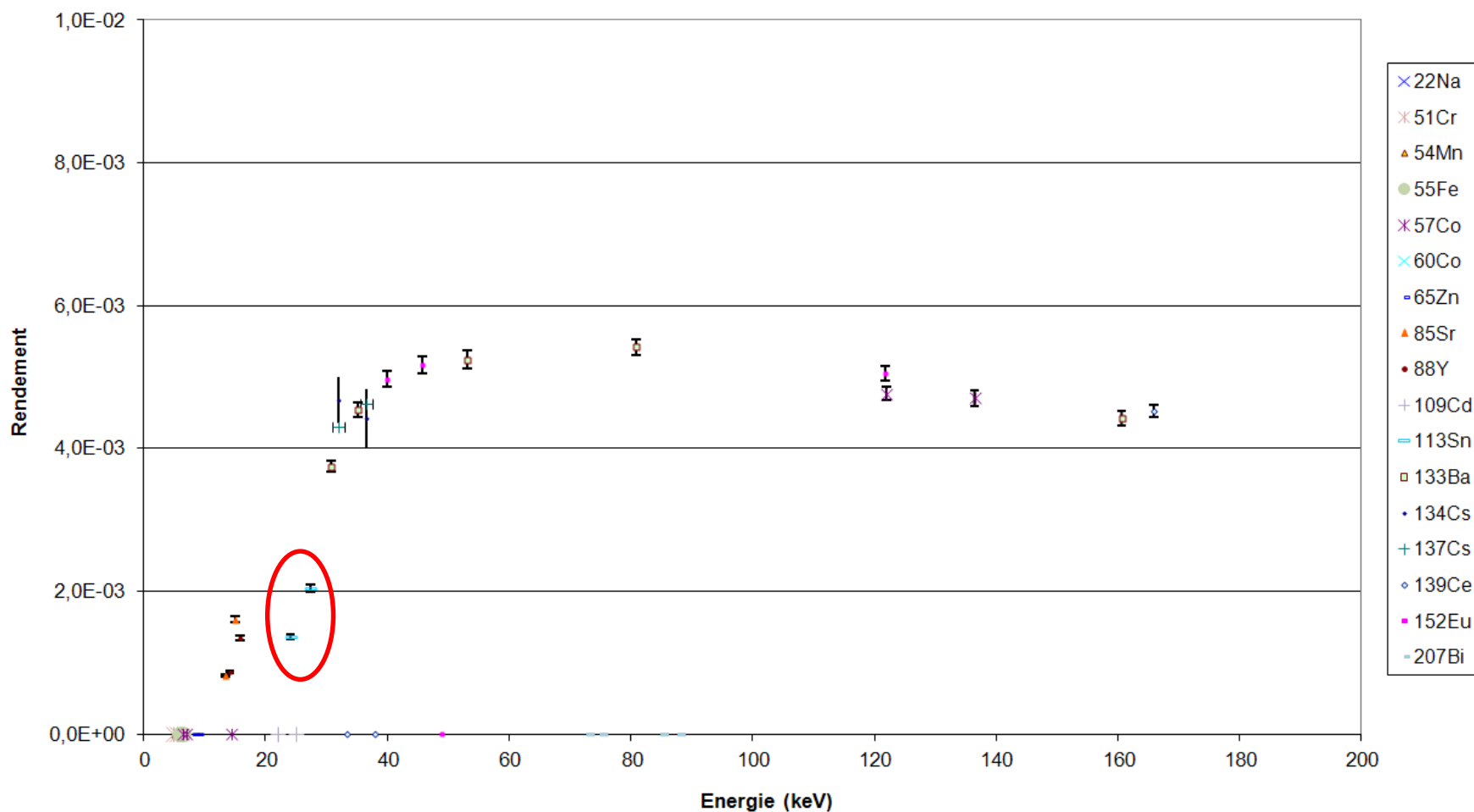
SELF-ATTENUATION IN THE SOURCE MATERIAL EFFICIENCY TRANSFER

ETNA is a software for computing efficiency transfer and coincidence summing corrections for gamma-ray spectrometry.

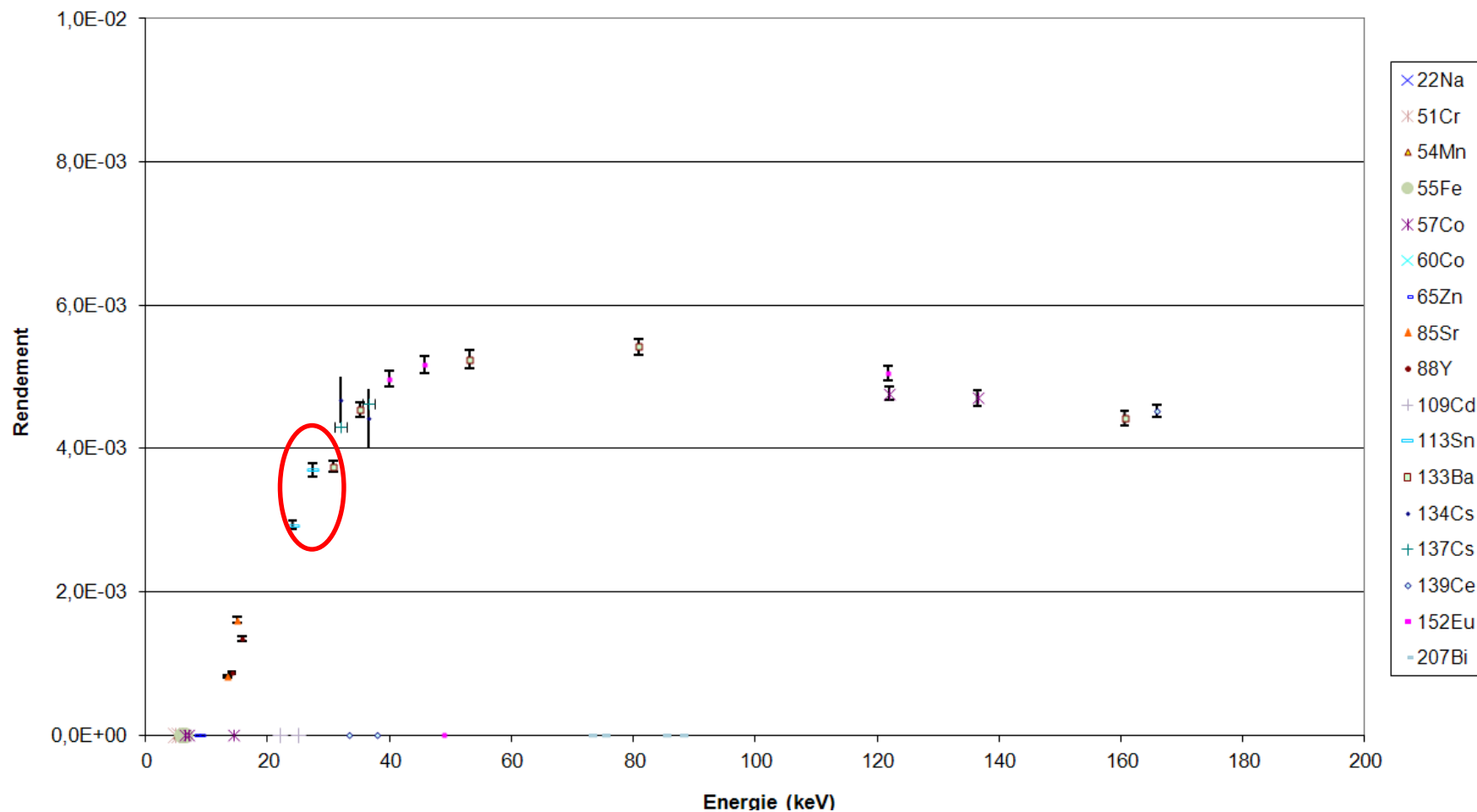
The software has been developed at the Laboratoire National Henri Becquerel and is available upon request.



Etalonnage Détecteur G9 - SG15 à 10 cm (G92)



Etalonnage Détecteur G9 - SG15 à 10 cm (G92)



Different methods to calculate self-attenuation correction factors

- Empirical methods – simplified computing
- Analytical approach
 - ANGLE
 - ETNA , etc.
- Monte Carlo simulation
 - DETEFF
 - EFFTRAN
 - GESPECOR
 - General codes (GEANT, PENELOPE, MCNP)
- Difficulty : knowledge of the mass attenuation coefficients
(exact composition of the sample material)

- **Radionuclide decay data and efficiency calibration**
 - Few radionuclides available
 - Decay data poorly known/correlations
 - Systematic deviations
- **Low-energy spectra main features**
 - Peak response function (energy resolution, X/gamma-ray)
 - Escape peaks
 - Self-fluorescence (rare)
 - Scattering phenomena in the source and surrounding materials (unavoidable)
- **Self-attenuation**
 - Depending on the energy
 - Depending on the geometry

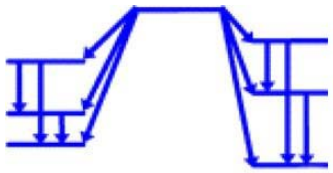
Low-energy range: interesting area ! Lot of fun !



THANK YOU FOR YOUR ATTENTION

Commissariat à l'énergie atomique et aux énergies alternatives
Institut List | CEA SACLAY NANO-INNOV | BAT, 861 – PC142
91191 Gif-sur-Yvette Cedex - FRANCE
www-list,cea.fr

Établissement public à caractère industriel et commercial | RCS Paris B 775 685 019



ICRM GSWG

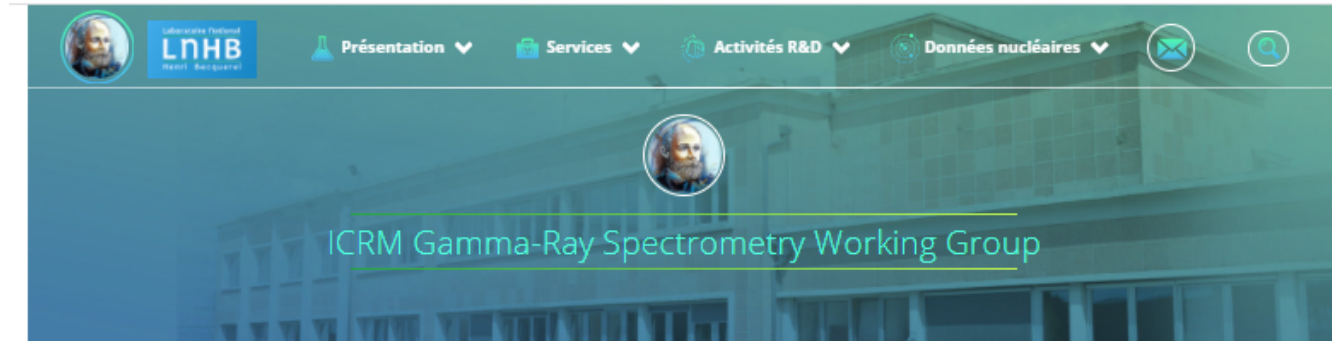
International
Committee for
Radionuclide
Metrology

ICRM Gamma Spectrometry Working Group

Web page : http://www.lnhb.fr/icrm_gs_wg/

Different information on the WG actions

- WG meeting presentations and actions status
- Training course presentations
- Practical information
- Etc.



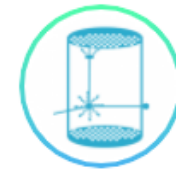
The Gamma-Ray Spectrometry Working Group is devoted to the development of the metrological aspects of gamma-ray spectrometry and its applications. This includes, but is not restricted to: measurement techniques and equipment, determination of photon emission intensities, detector efficiency calibrations (including Monte Carlo methods), coincidence-summing corrections, uncertainties, correlations, new instrumentation, and X-ray spectrometry.



[Meetings & Workshops](#)



[WG actions \(Past & on-going\)](#)



[Monte Carlo benchmarks](#)



[Past exercises & Publications](#)



[Events & Courses](#)



[Practical info & Useful links](#)



ICRM Gamma Spectrometry Working Group

Web page : http://www.lnhb.fr/icrm_gs_wg/

- On going actions:
 - Benchmark for Monte Carlo simulation
 - Efficiency (completed)
 - Coincidence summing corrections (on-going)
 - Calculation of detection limits (leader: Milton van Rooy)
 - Self-attenuation (just starting)
- Next International Committee for Radionuclide Metrology (ICRM) conferences
 - ✓ The ICRM Low Level Radioactivity Measurement Techniques conference will take place from May 2 to May 6, 2022 at Gran Sasso National Lab (Italy)
 - ✓ The 2023 ICRM conference will take place in Bucharest (Romania)