Gamma- (and X-) ray spectrometry

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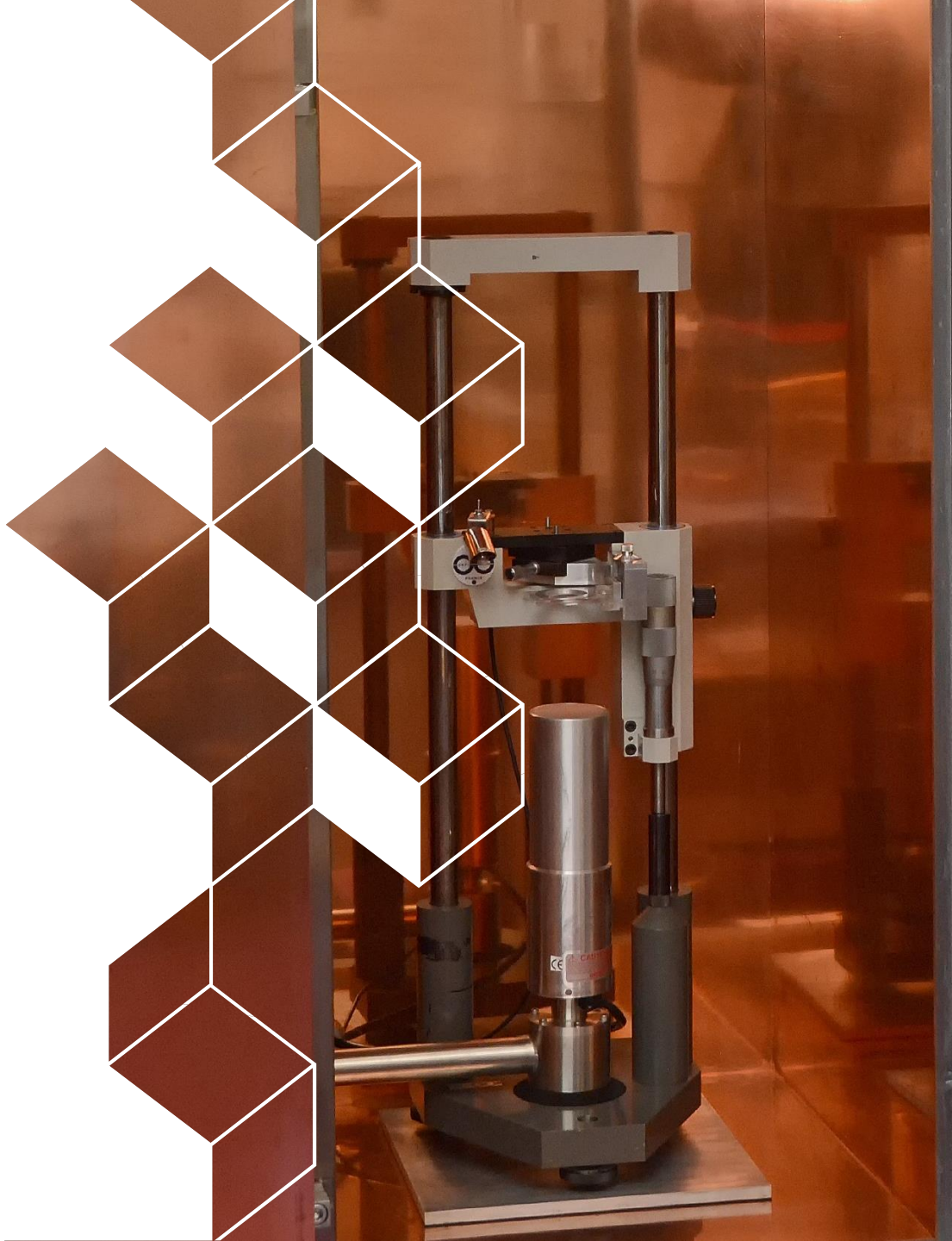


Outlook

1. Efficiency calibration
   a) Calibration curves fitting
   b) Net peak area determination
   c) Monte Carlo approach

2. Corrective factors
   a) Coincidence summing
   b) Geometry transfer

3. Absolute or relative photon emission intensities

4. Examples
   a) Photon emission intensities in the decay of ^{59}Fe
   b) Impurity in ^{93m}Nb

(Full-energy peak) Efficiency calibration

Gamma spectrometry -> identification and quantification of radionuclides

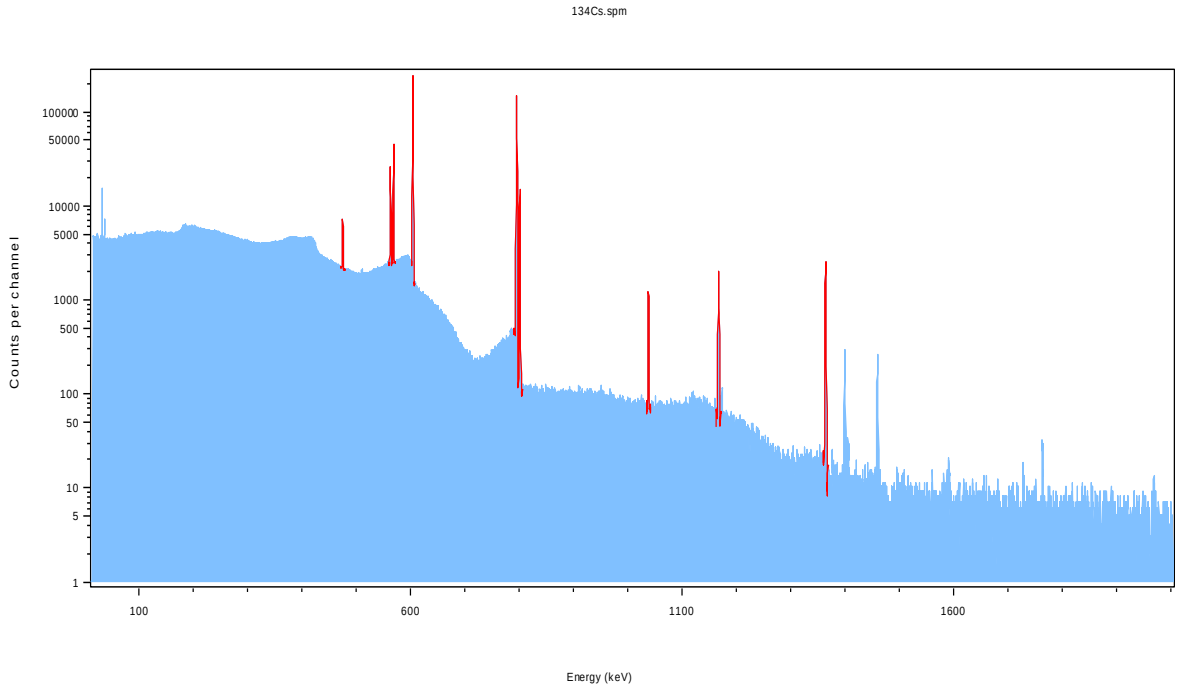
-> photon emission intensities

Basic equations

$$N_i = A \cdot I_i \cdot \varepsilon_i \cdot t \cdot Corr$$

$$I_i = \frac{N_i \cdot Corr'}{A \cdot \ \varepsilon_i \cdot t}$$

$$\varepsilon_i = \frac{N_i \cdot Corr'}{A \cdot I_i \cdot t}$$



$A\ (Bq)$: reference source activity

Measured data:

E_1 ... ... En (Energy)

N_1 ... ... N_n (Net peak area)

Tabulated data:

I_1 ... ... In (Emission intensity)

Full-energy peak efficiency:

ε_1 ... ... ε_n

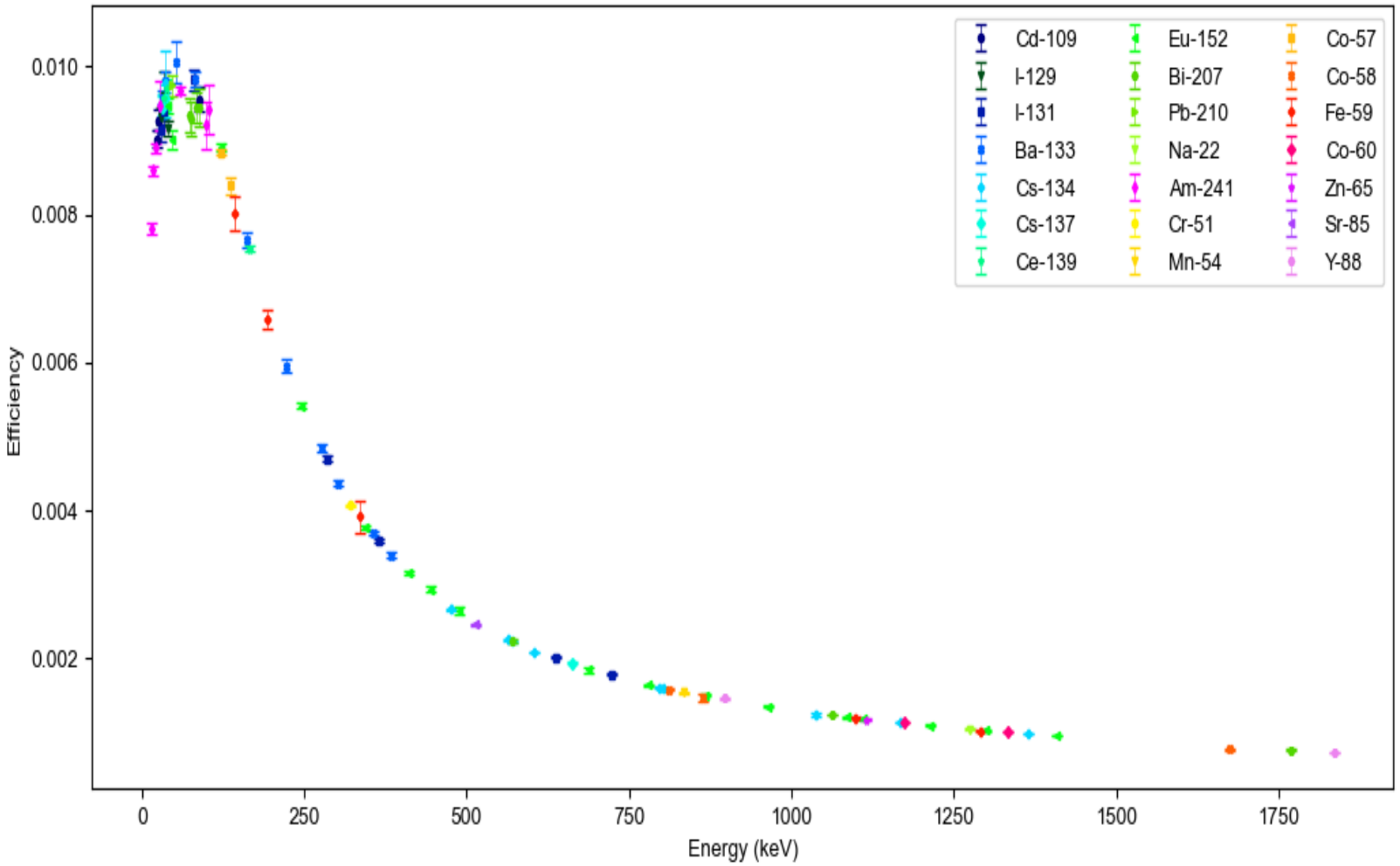
Efficiency calibration

$$\varepsilon_i = \frac{N_i \cdot Corr'}{A \cdot I_i \cdot t}$$

Typical radionuclides used for efficiency calibration:

- monoenergetic:
  ^{137}Cs, ^{54}Mn, ^{51}Cr, …
- multigamma:
  ^{152}Eu, ^{133}Ba, ^{60}Co, ^{88}Y …

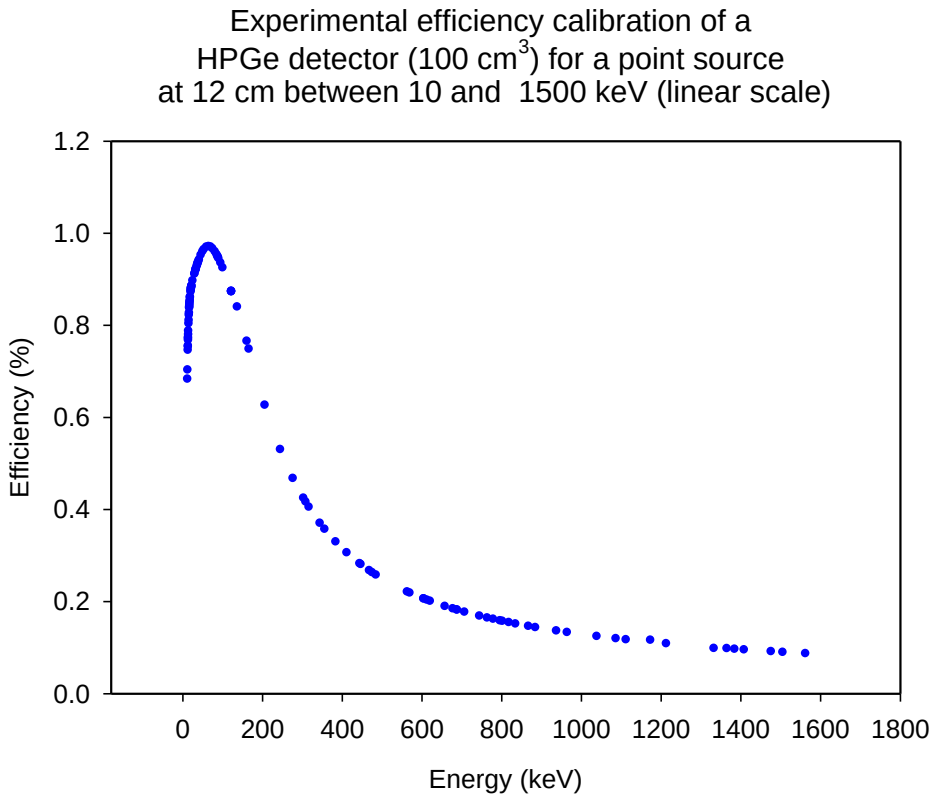
Example of experimental data obtained using 21 standard radionuclides in the energy range from 20 keV to 1836 keV

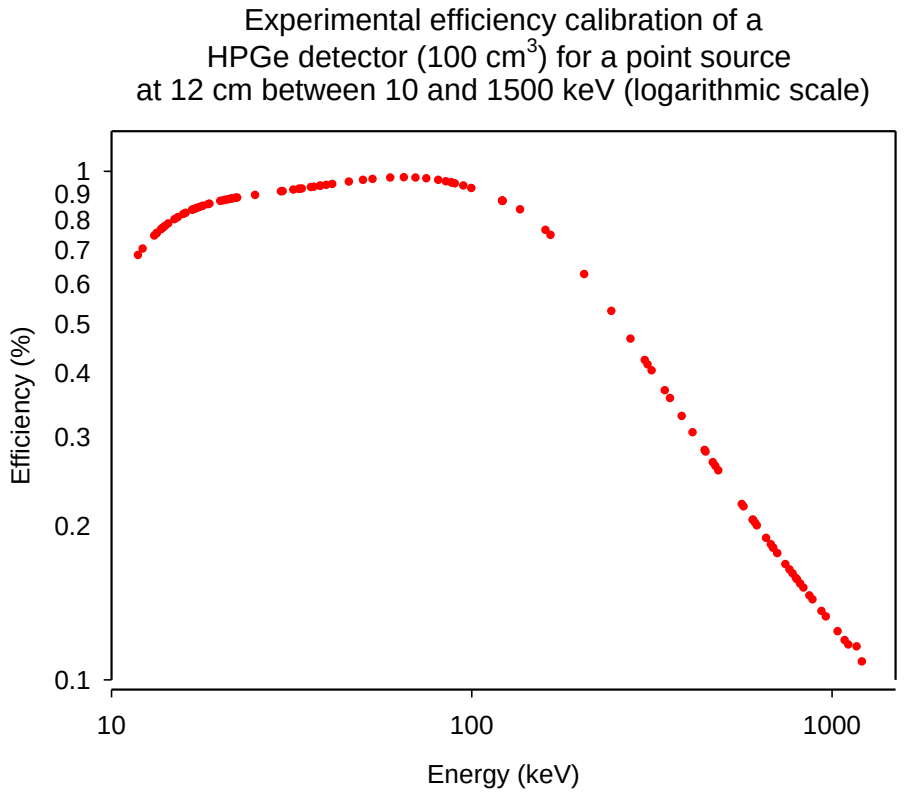


Efficiency calibration

$$\varepsilon_i = \frac{N_i \cdot Corr'}{A \cdot I_i \cdot t}$$

Mathematical fitting:
Determination of the best fitted function to a given set of experimental data (energy, efficiency)





In logarithmic scale, the shape is smoother than in linear scale

Efficiency calibration

$$\varepsilon_i = \frac{N_i \cdot Corr'}{A \cdot I_i \cdot t}$$

Functions most frequently used: polynomial in the log-log scale:

$$\ln \varepsilon(E) = \sum_{i=0}^{n} a_i \cdot (\ln E)^i$$

$$\ln \varepsilon(E) = \sum_{i=0}^{n} a_i \cdot E^{-i}$$

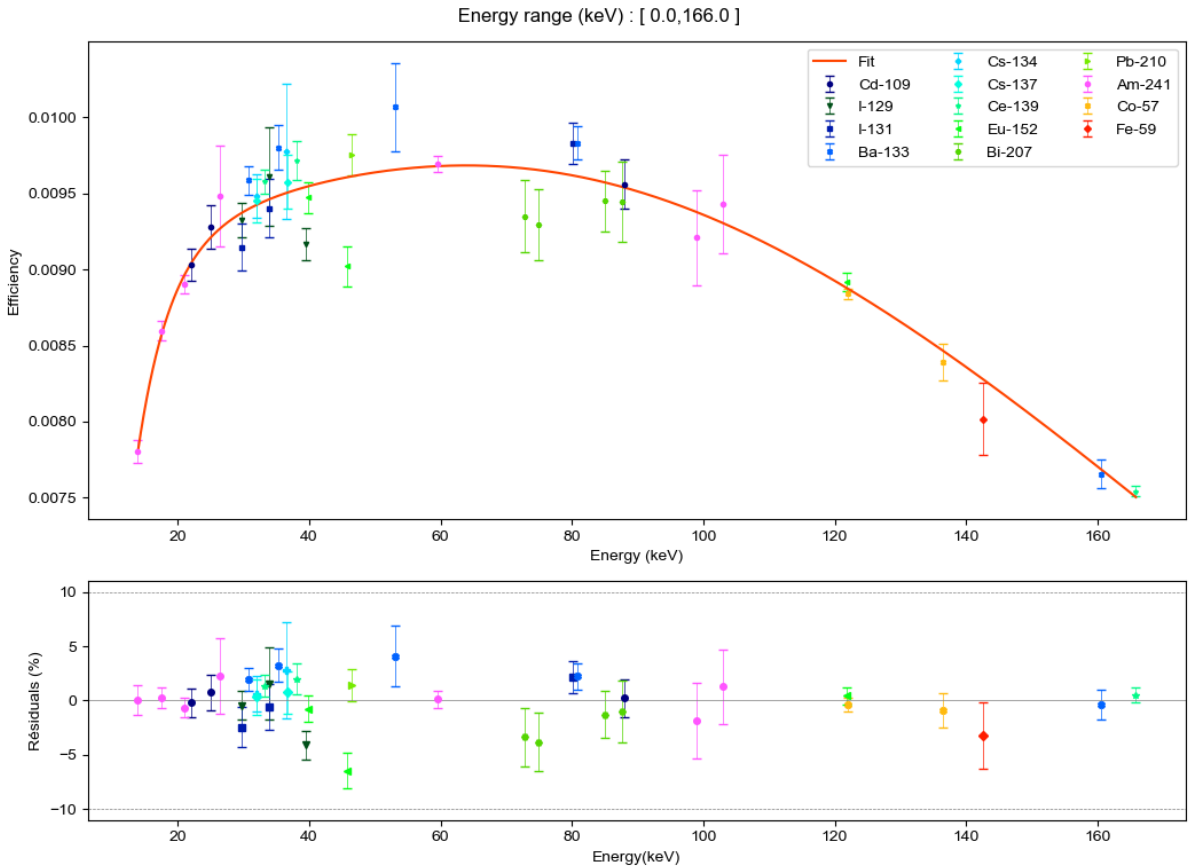
Remarks:

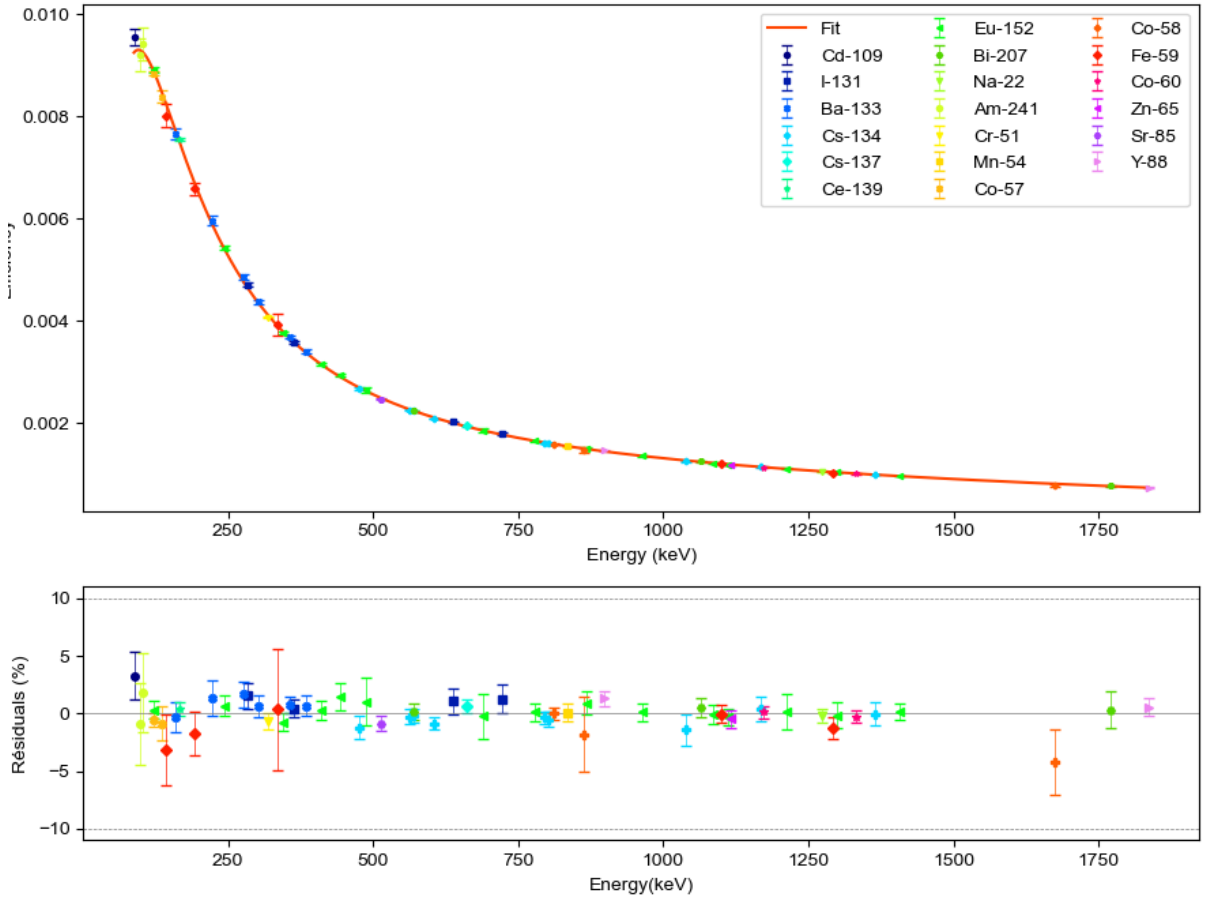
- a_i coefficients are determined using a least-squares fitting method
- experimental data must be **weighted** (uncertainties)
- **correlation** between input data
- the polynomial degree (n) must be adjusted depending on the number of experimental data (p): n << p
- in some case two different functions can be used with a cross point
- check the resulting fitted curves!

Efficiency calibration

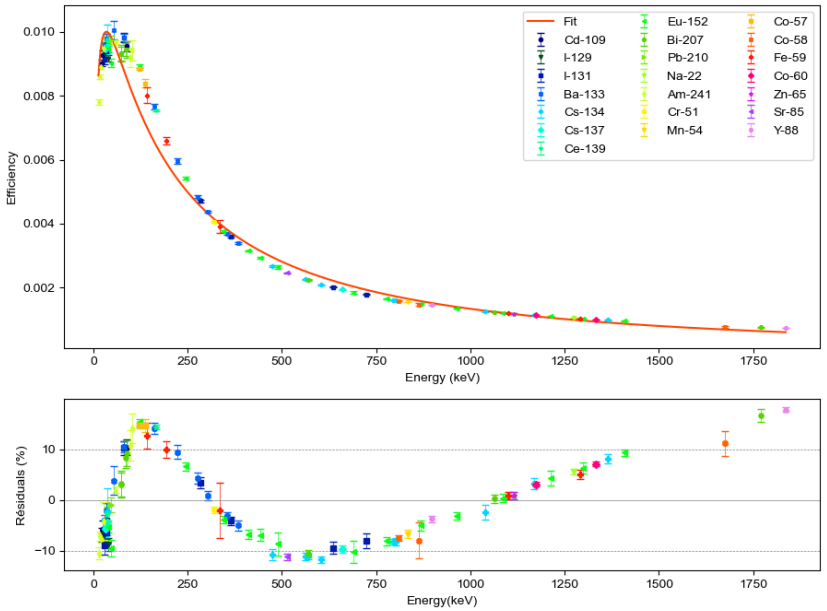
$$\varepsilon_n = \frac{N_n \cdot Corr'}{A \cdot I_n \cdot t}$$

Fitted curves using two energy ranges

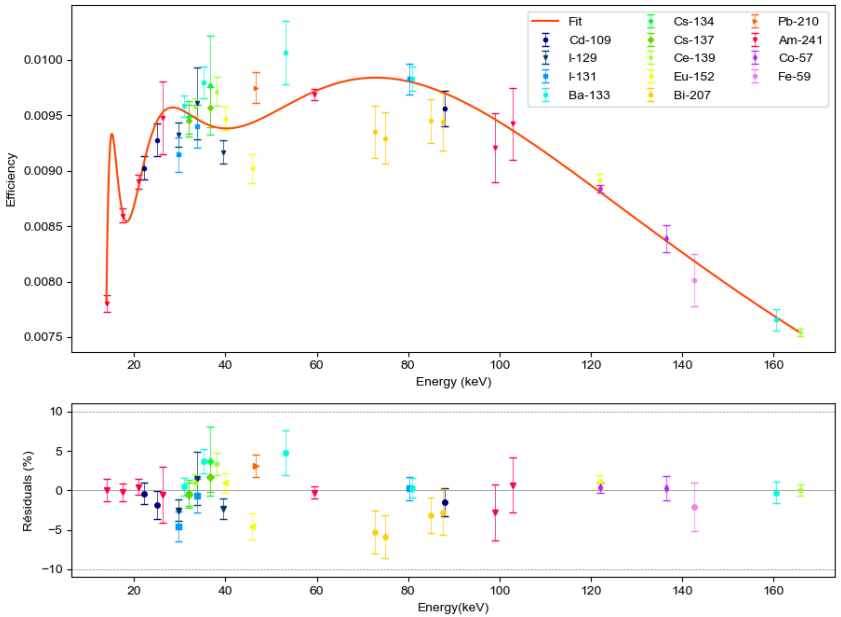




Efficiency calibration



Fitting with a polynomial of degree 3:
too few coefficients



Fitting with a polynomial of degree 7:
too many coefficients

Efficiency fitting should be visually checked (polynomial degree adapted to the number of experimental points: n << p)

In the case of cross point, avoid zones with important inflexion

Efficiency calibration: associated uncertainties

$$\varepsilon_i = \frac{N_i \cdot Corr'}{A \cdot I_i \cdot t} = \frac{N_i \cdot \prod C_{ij}}{A \cdot I_i \cdot t}$$

$\prod C_{ij}$: product of the corrective factors

$$\frac{u(\varepsilon_i)}{\varepsilon_i} = \left[\frac{u^2(N_i)}{N_i^2} + \frac{u^2(A)}{A^2} + \frac{u^2(I_i)}{I_i^2} + \sum_j \frac{u^2(C_{ij})}{C_{ij}^2} \right]^{1/2}$$

Main uncertainties:

- reference activity (calibrated source)
- photon emission intensities (data table mentioned)

Example: ^{137}Cs

Relative standard uncertainty of the activity: 0.5%

Relative standard uncertainty of the emission intensity: 0.34%

No corrective factor

Relative uncertainty on peak area (%)	Relative combined standard uncertainty on FEP efficiency (%)
1	1.17
0.1	0.61

The fitting procedure, in the absence of constraints, can lead to lower uncertainties: is this relevant? (systematic correlations not considered)

At LNHB, we have chosen to consider the lower experimental uncertainty as an additional uncertainty that is quadratically added to the uncertainty resulting from the fitting procedure.

Net peak area

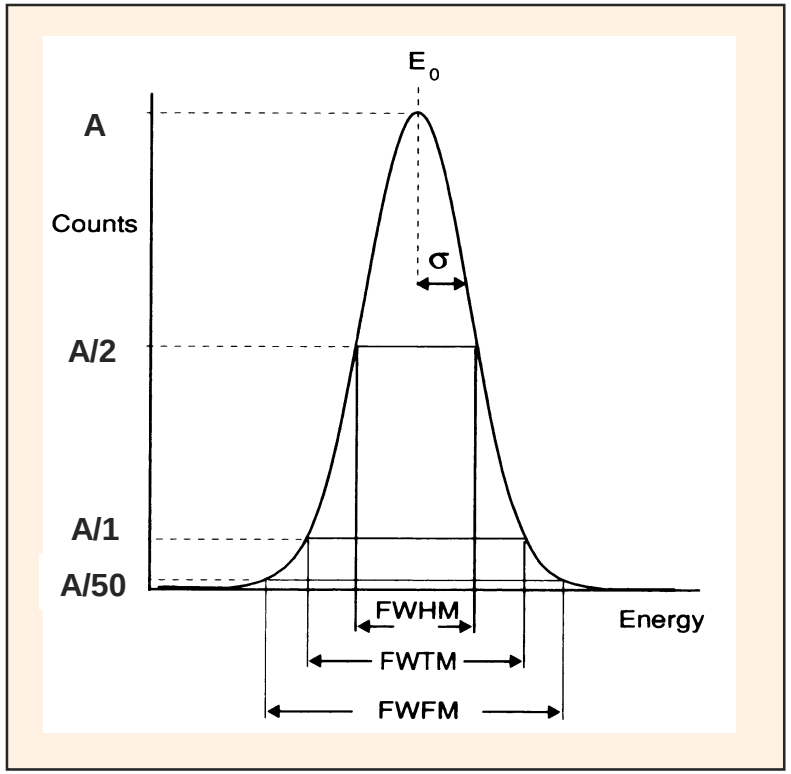
The net peak area, N_n, is the main input to derive efficiencies or photon emission intensities.

In first approximation the peak shape is Gaussian.

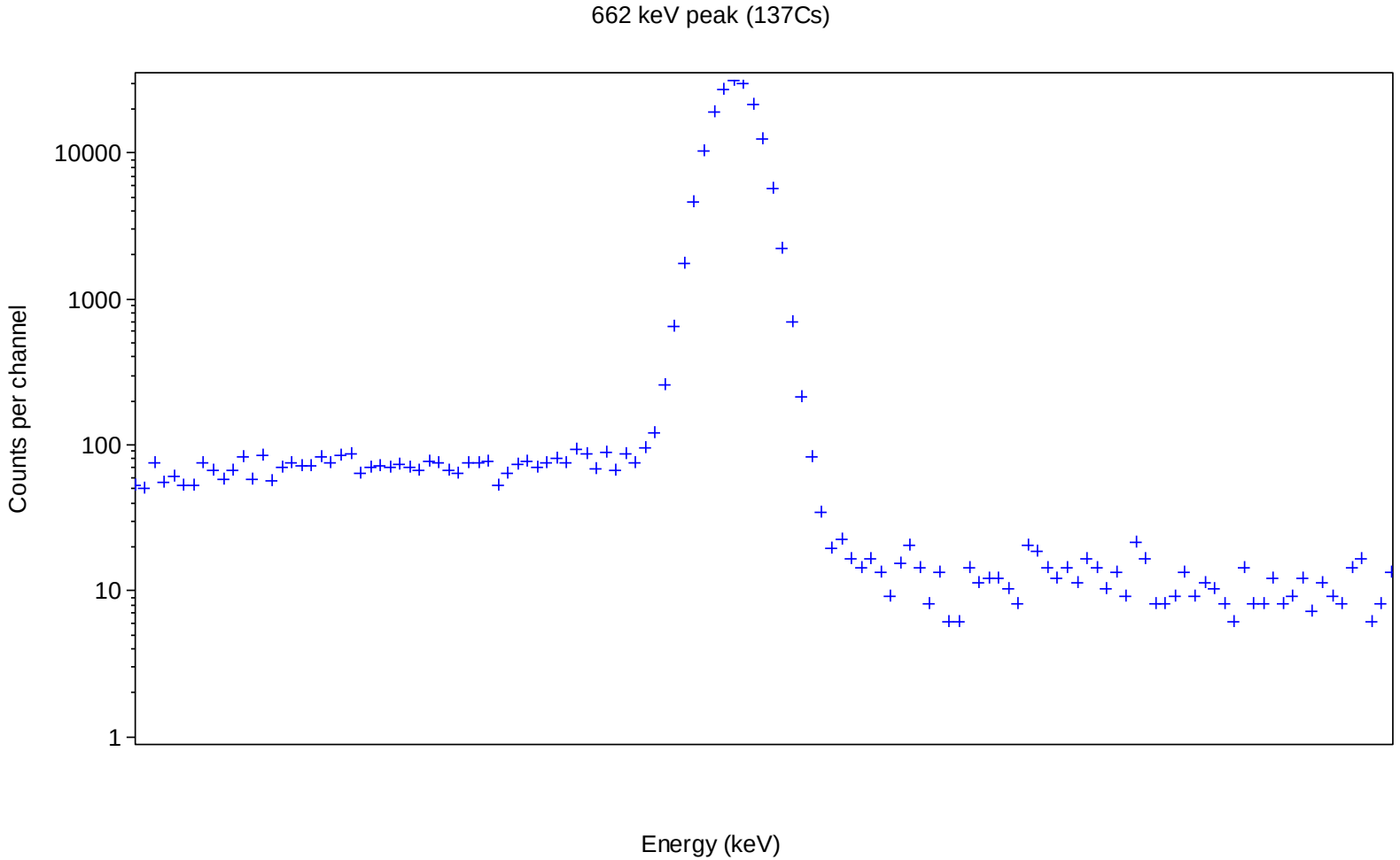
Two main approaches to determine the net peak area:

- Summing all channels and subtracting the background,

- Mathematical fitting using a peak model.



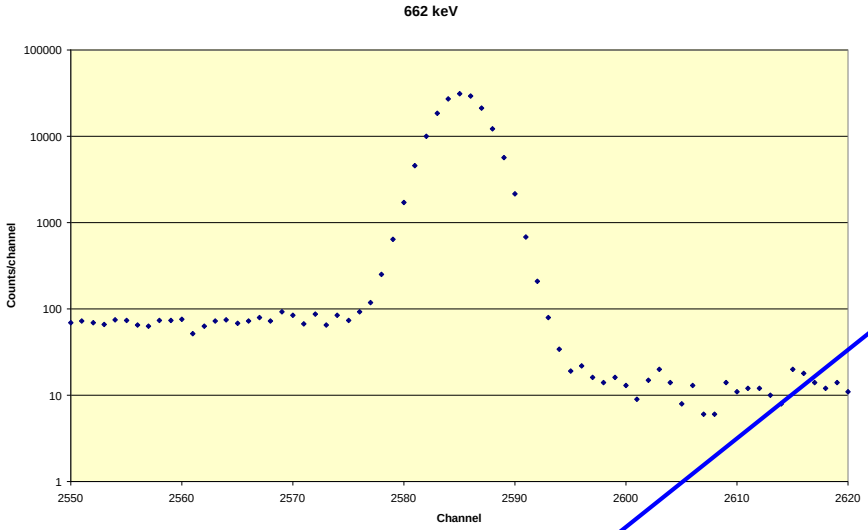
Net peak area

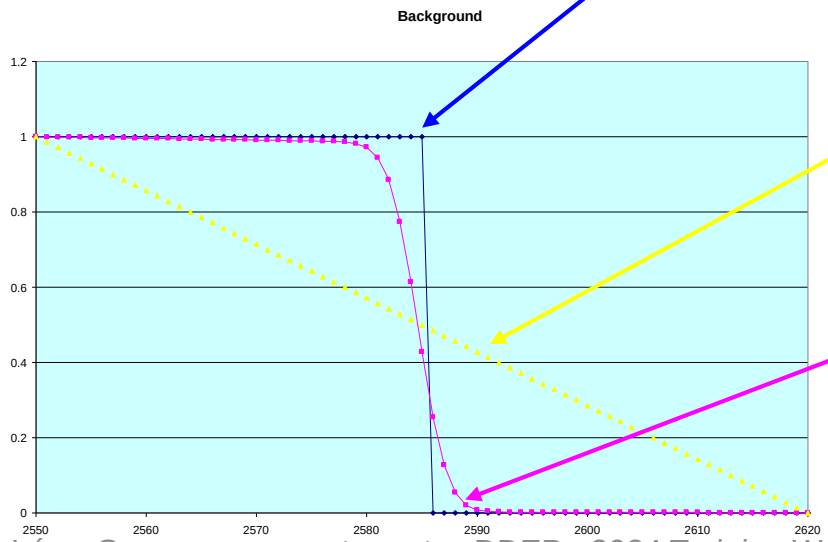


Peak superimposed on a continuous background

Net peak area: background

Peak with energy E_0 in region [f,I] first channel = f ; last channel = I





Step:

F(i) = 1 for E < E0

F(i) = 0 for E > E0

Linear: $$F(i) = 1 - \frac{(i - f)}{(I - f)}$$

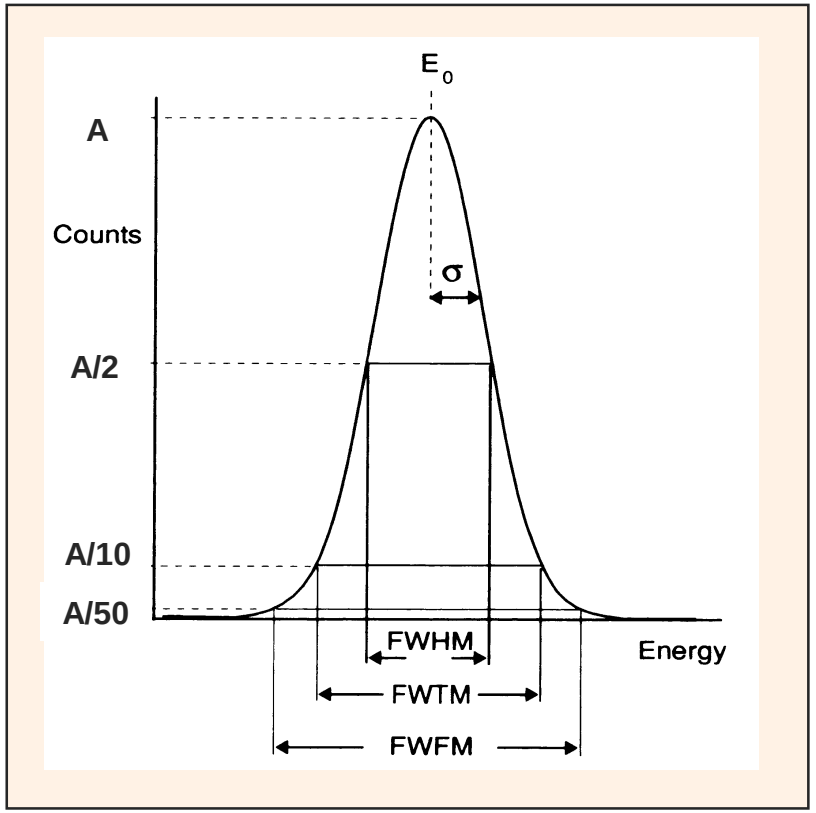
Galton curve: $$F(i) = 1 - \frac{\sum_{f}^{i} Y_i}{\sum_{f}^{I} Y_i}$$

Net peak area: mathematical fitting

Approximation: peak = Gaussian

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

E_0 = energy, A = amplitude, σ = standard deviation



Gaussian total area:

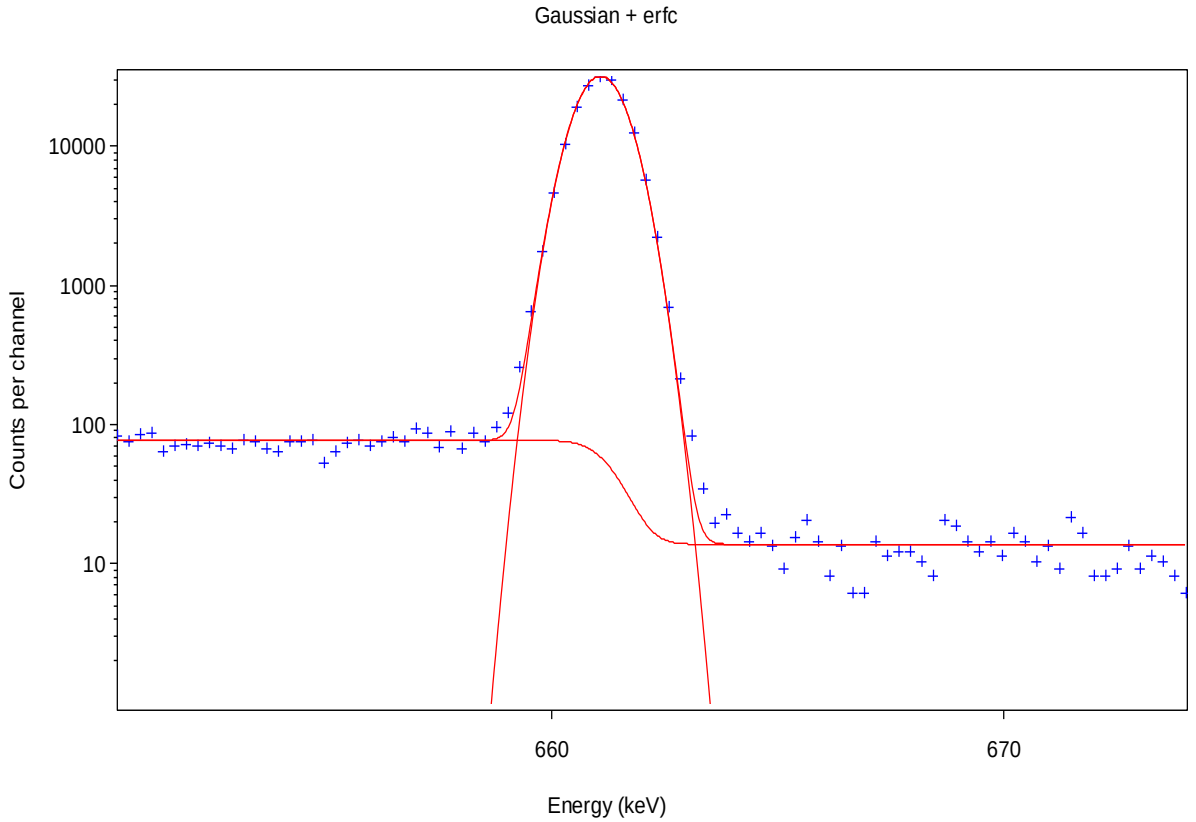
$$S[-\infty, +\infty] = \sqrt{2 \cdot \pi} \cdot \sigma \cdot A$$

$$S[-3\sigma, +3\sigma] = 0.997 \cdot S[-\infty, +\infty]$$

Approximation (without peak fitting):

Sum of channels in the region [-3σ, +3 σ] around the peak centroid = 99.7% of the Gaussian total area

Net peak area



Background subtracted as a Galton curve and peak fitted with a Gaussian function:

fitted parameters: $A = 31360$ $\sigma = 0.53$ $\rightarrow$ $N = 164\,200$

Using a linear background and summing of channel contents: $N = 164\,600$

Net peak area: overlapping peaks

If peaks have about the same width: individuals areas can be obtained from the total net area weighted by the relative amplitude of each peak.

Case of the 511-keV region:

Example: ^{85}Sr

(Gamma at 514 keV)

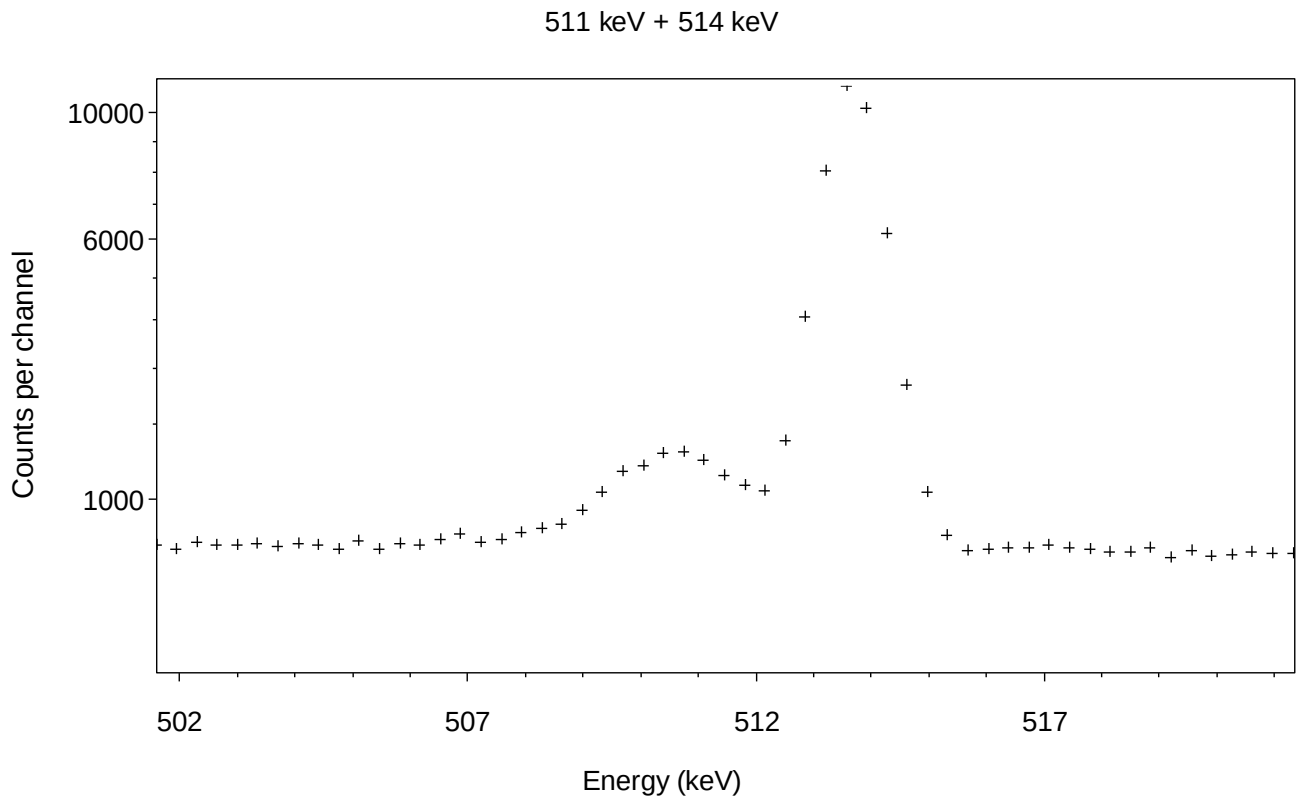
Total net area = 48500

A(511) = 1 000

A(514) = 10 000

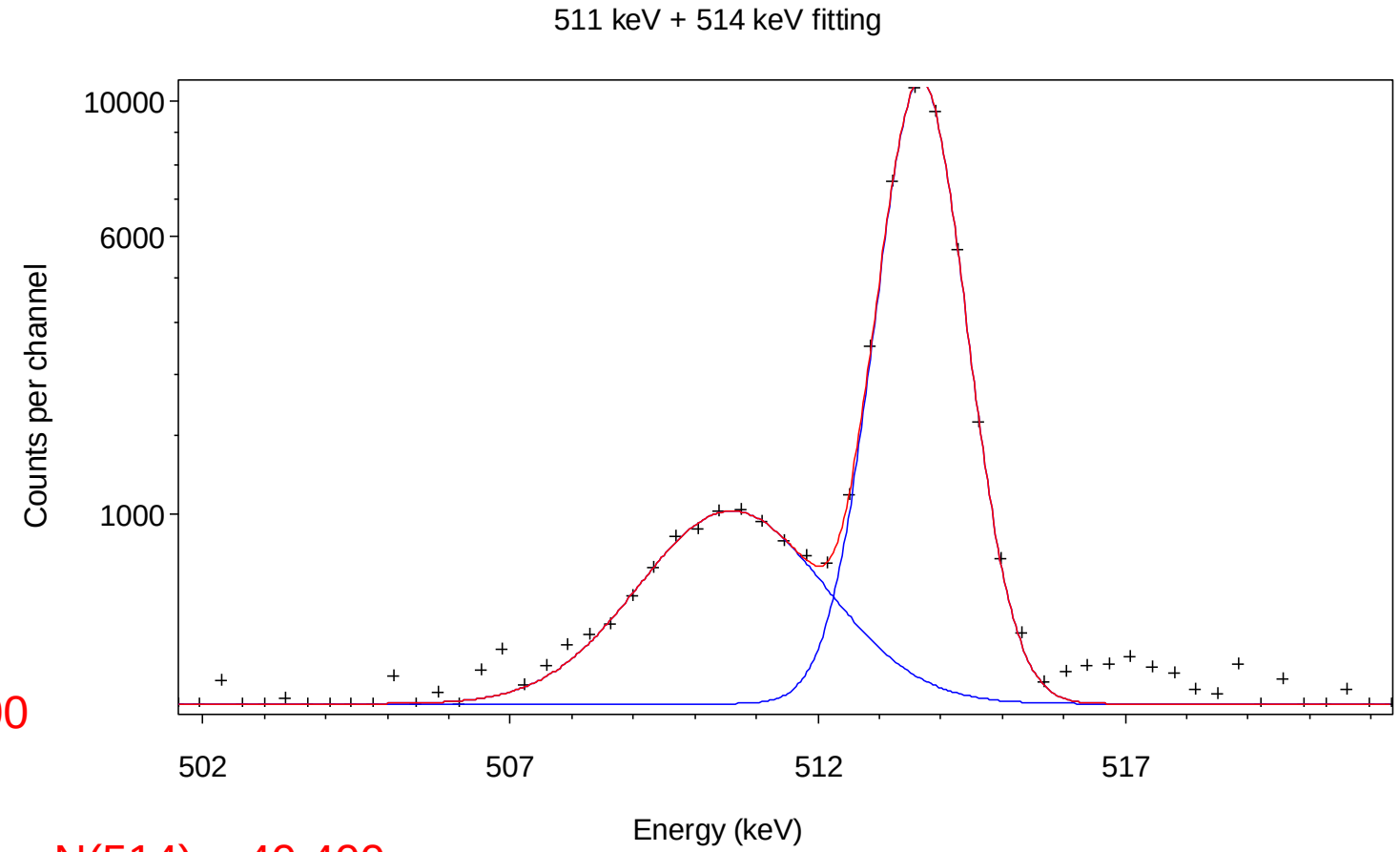
N(511) = 4 400

N(514) = 44 100



Net peak area: overlapping peaks

511 keV (annihilation peak) is much larger than other full-energy peaks



Summing procedure:

N(511) = 4 400 N(514) = 44 100

Fitting procedure: N(511) = 7 730 N(514) = 40 400

σ = 1.07 σ = 0.53

Bias on the 514 keV area = 9% !!!

Net peak area: overlapping peaks

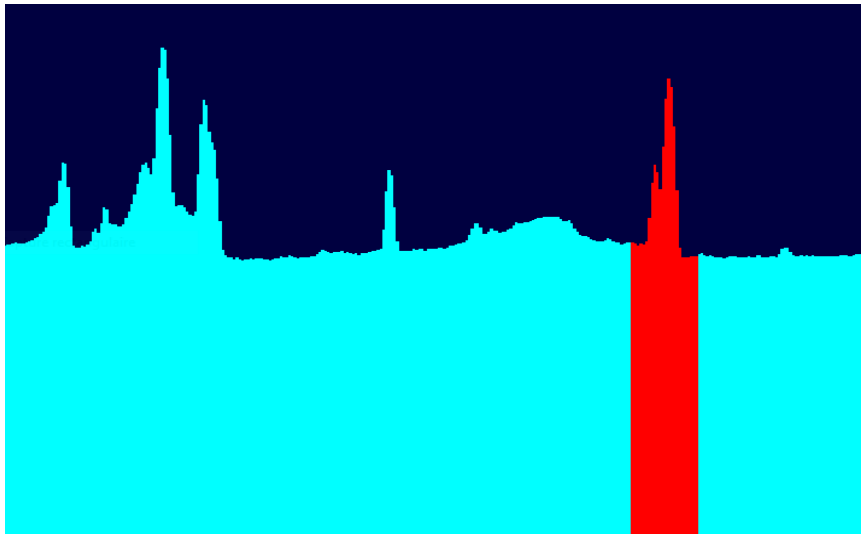
Importance of the detection **energy resolution**

^{133}Ba spectrum (79 keV- 80 keV doublet)

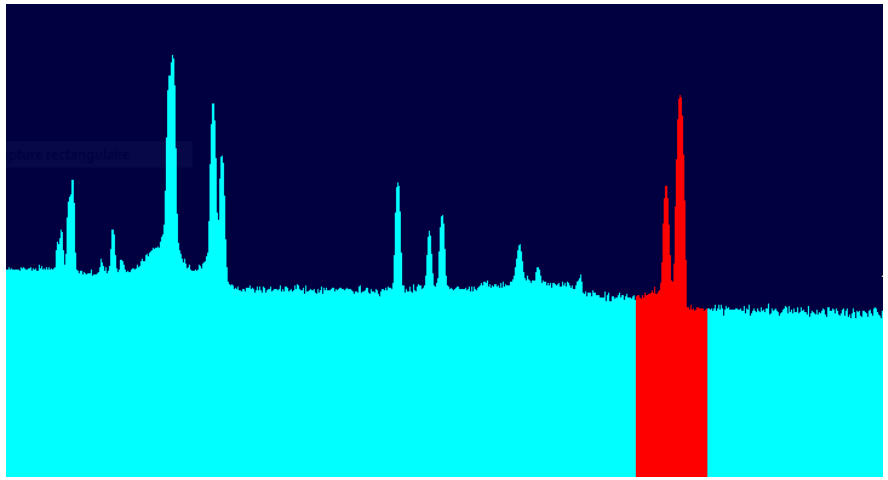
Energy window:15 keV – 100 keV

The high energy resolution enables for better deconvolution to accurately determine peak areas.

N-type HPGe detector (coaxial)



N-type HPGe detector (planar)



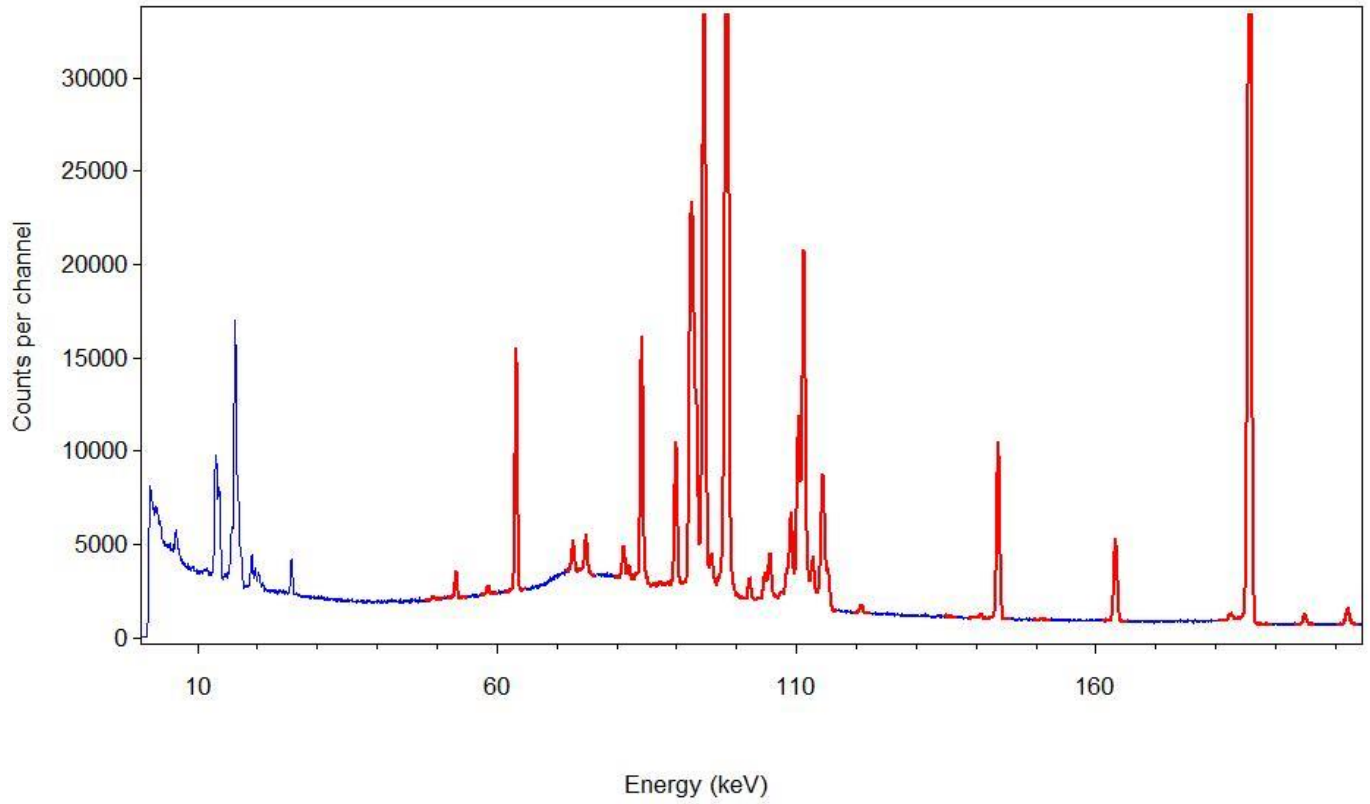
Net peak area: overlapping peaks

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

$^{238}U \rightarrow {}^{234}Th \rightarrow {}^{234}Pa^{m} \rightarrow {}^{234}Pa \rightarrow {}^{234}U$

$^{235}U \rightarrow {}^{231}Th \rightarrow {}^{231}Pa$

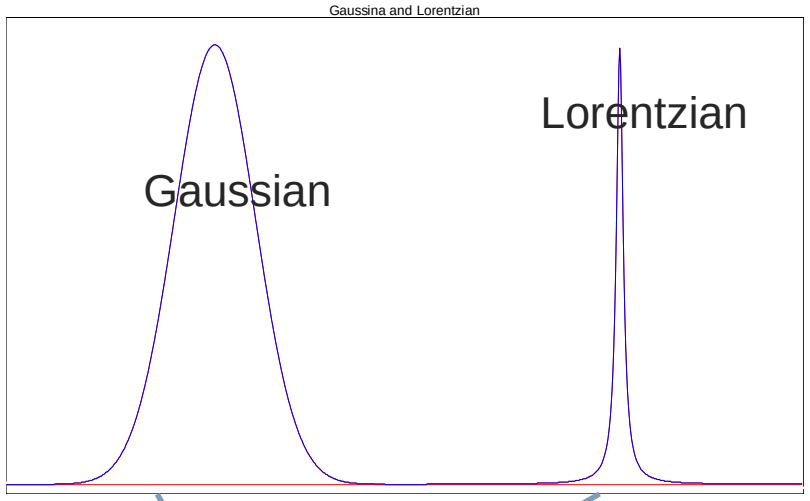
Spectra include **gamma** lines from different radionuclides **and X-rays** from U, Pa and Th



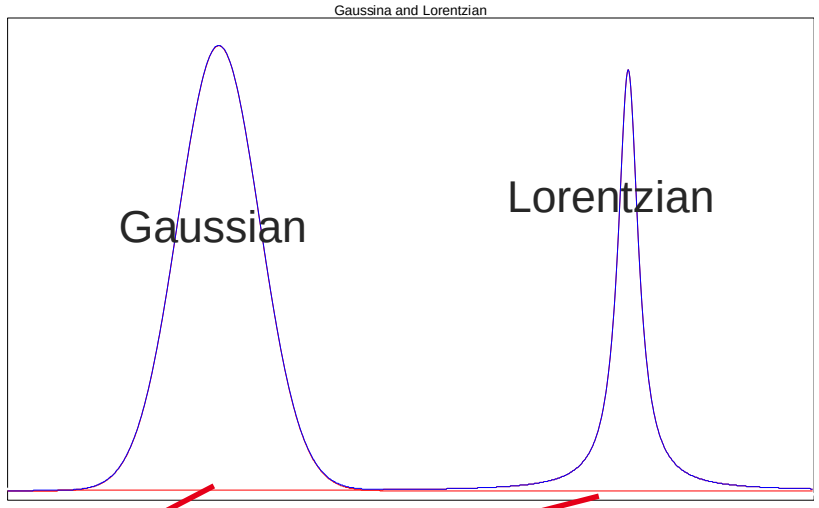
Net peak area: X-ray natural linewidth

Shape of photon emission lines: *Lorentzian (natural linewidth)*

Widening of the detection chain: *Gaussian*



σ = 50 eV – Γ = 10 eV



σ = 50 eV – Γ = 30 eV

Peak = *Lorentzian ⊗Gaussian (Voigt profile)*

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

Net peak area: X-ray natural linewidth

Shape of photon emission lines: *Lorentzian (natural linewidth)*
Widening of the detection chain: *Gaussian*

Element	Z	Kα X-ray energy	Kα X-ray natural width (eV)	FWHM (eV)	X-ray linewidth /FWHM
Nickel	28	7.45	1.95	155	0.013
Cadmium	48	22.98	9.9	200	0.049
Lead	82	72.80	66.8	350	0.19
Uranium	92	94.65	105.4	415	0.25

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

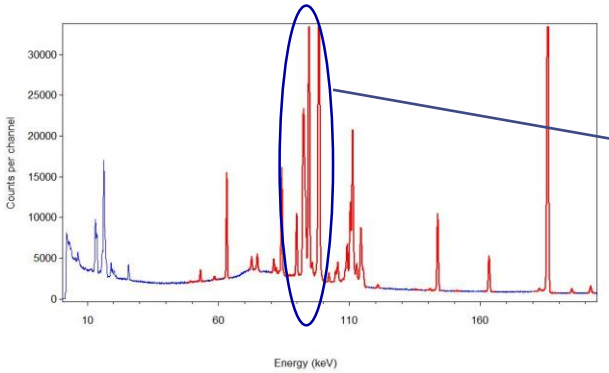
Net peak area: overlapping peaks

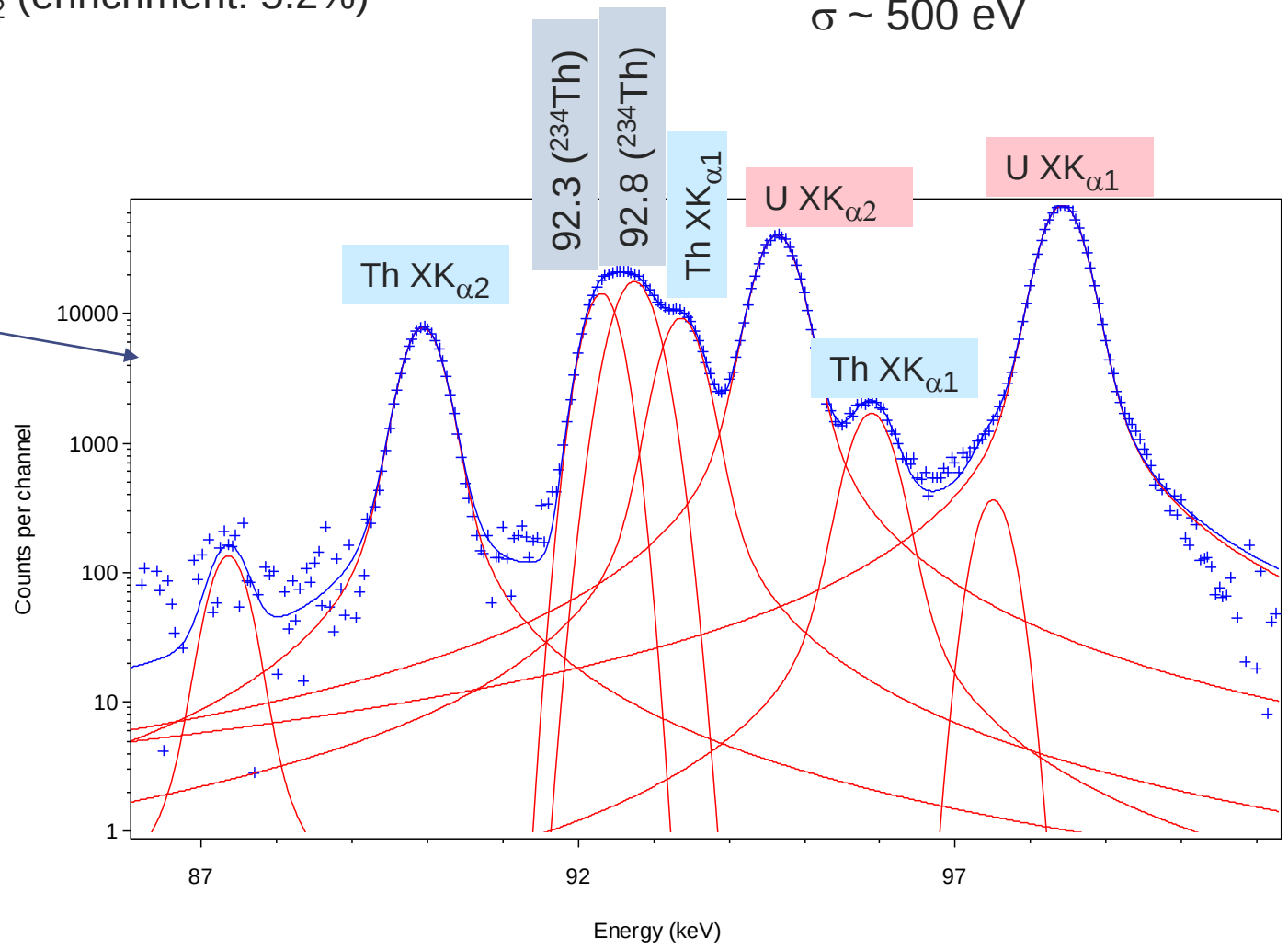
Example of processing of K X-rays of UO_2 (enrichment: 5.2%)

$^{238}U \rightarrow {}^{234}Th \rightarrow {}^{234}Pa^{m} \rightarrow {}^{234}Pa \rightarrow {}^{234}U$

$^{235}U \rightarrow {}^{231}Th \rightarrow {}^{231}Pa$

Γ ~ 100 eV
σ ~ 500 eV





Important for accurate determination of photon emission intensities

Monte Carlo approach

Detection efficiency can be derived using Monte Carlo simulation

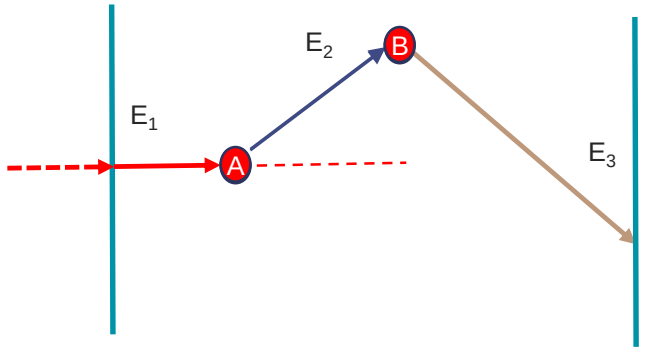
Mathematical method in which the problem of interest is solved by using a model based on random numbers sampled from appropriate distributions.

For the calculation of detection efficiency, the simulation must take into account:

- Physical effects occurring in the source-detector geometry
- Sampling distributions corresponding to particle-matter interactions probabilities

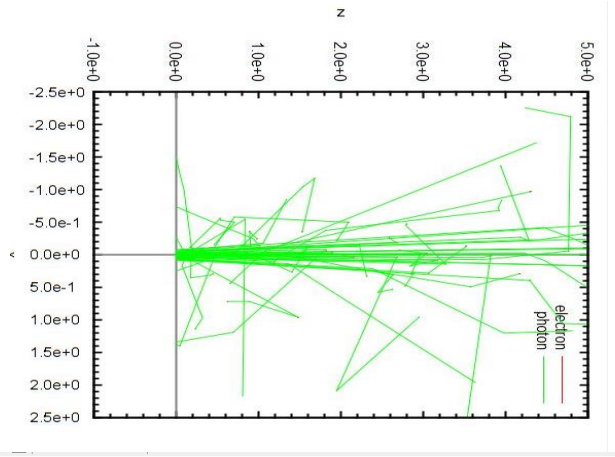
The history (trajectory) of a particle is considered as a random sequence of free paths ending in an interaction (photoelectric, Compton, Rayleigh, pair production) where the particle:

- changes direction,
- loses energy,
- and (often) produces secondary particles.



Monte Carlo approach

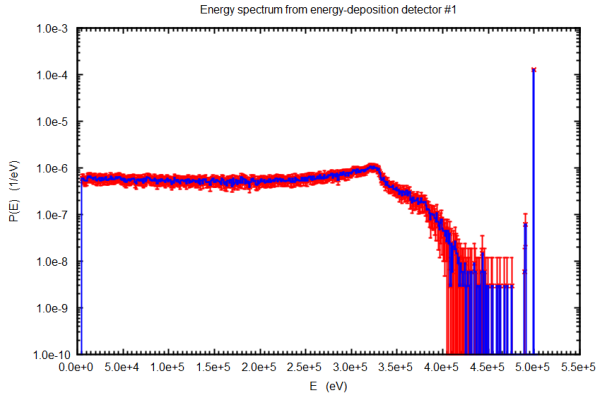
The energy deposited in the sensitive volume by the primary photon and all secondary particles is summed up for obtaining the detector signal associated with the primary photon.



Two kinds of code:

- Generalist codes:
  - GEANT4, MCNP, PENELOPE, EGS, TRIPOLI-4, etc.

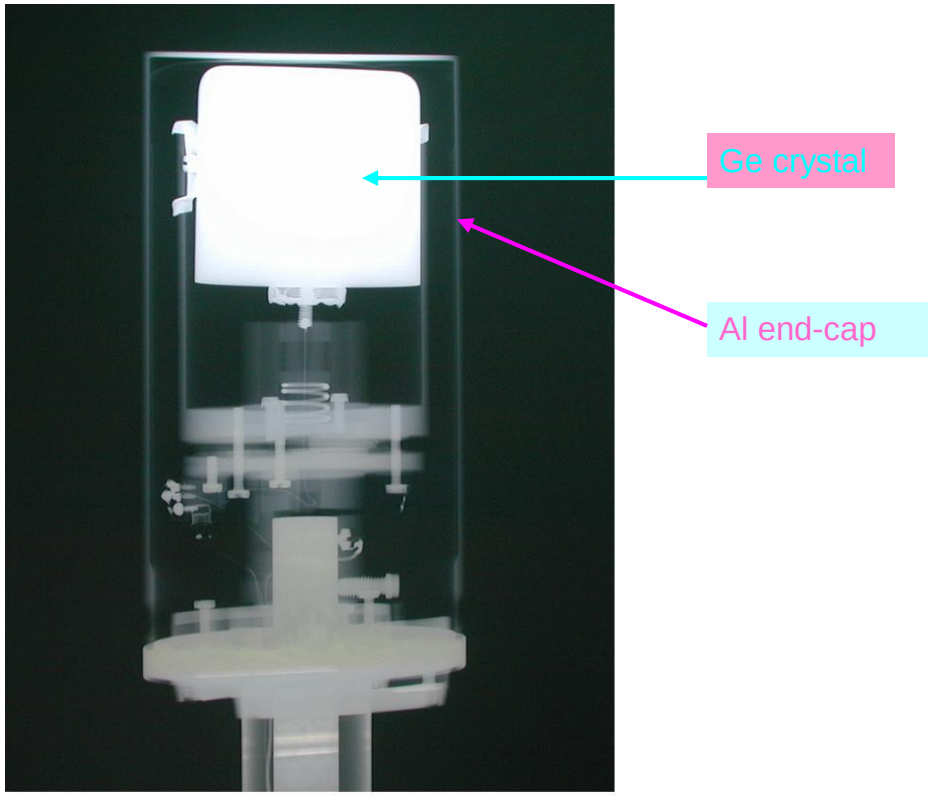
- Specific codes (for gamma-ray spectrometry):
  - GESPECOR, DETEFF, EFFTRAN
  - ISOCS™ (commercial tool)



Monte Carlo approach

This requires accurate description of the geometry (source, detector, environing materials)
Difficulty: internal composition of the detector

- crystal-window distance
- rounded shape
- exact crystal position (any shift/inclination)
- exact dead layer thickness (for the low-energy range)



Difficult to accurately get **absolute** efficiencies

Validation of MC results with experimental measurements is mandatory

Interest for efficiency transfer corrective factors

Corrective factors $\varepsilon_n = \frac{N_n \cdot Corr'}{A \cdot I_n \cdot t}$

Corr' must include:

- correction for radioactive decay from the date of the reference activity (A): $C_T = exp\left(-\ln(2) \cdot \frac{(T_m - T_0)}{T_{1/2}}\right)$

T_m: acquisition start time
T_0: reference time
$T_{1/2}$: radionuclide half-life

- correction for radioactive decay during the measurement (real time): $C_{Dec} = \frac{\ln(2) \cdot \frac{T_r}{T_{1/2}}}{1 - exp\left(-\ln(2) \cdot \frac{T_r}{T_{1/2}}\right)}$

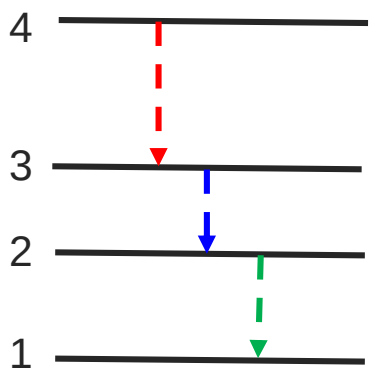
T_r: real acquisition time

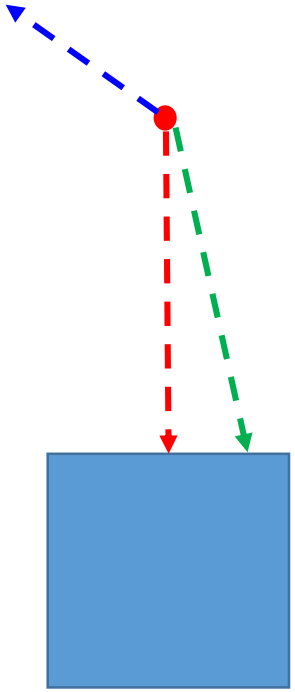
- correction for **coincidence summing**

- correction for **geometry transfer** (if applicable)

Corrective factors: true coincidence summing

Radionuclide decaying with **cascades gamma transitions**





Also with X-rays and with Compton-scattered photons

Two (or more) photons can reach simultaneously the detector

- general change in the spectrum shape
- loss of events in full-energy peaks (summing-out)
- gain of events in FEP corresponding to transitions between non consecutive levels (summing-in)

Corrective factors must be applied to take this effect into account

Depend on the radionuclide decay scheme and detection efficiency (geometry), not on the activity level

Important for measurements in close geometry (environmental samples)

Corrective factors: true coincidence summing

These can be computed with

Dedicated codes:

GESPECOR (Univ. Bucharest)
EFFTRAN (Tim Vidmar – SCK-Mol) https://efftran.github.io/
ETNA (LNHB)
…

Full Monte Carlo codes:

PENELOPE 2018 (including DDEP data base)
GEANT4
EGS
MCNP
…

They depend on:

- the decay scheme (importance of taking into account X-rays for a N-type detector)
- the detection efficiency (large source-to-detector distances reduce this effect)

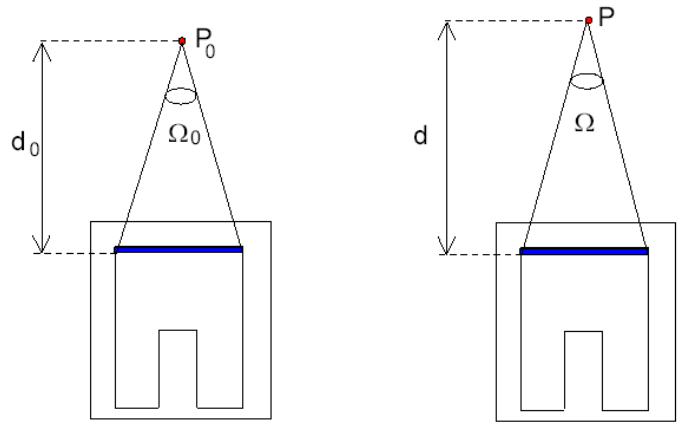
Coincidence summing corrective factors: examples

Radionuclide	Energy (keV)	Point source at 1 cm	Point source at 10 cm	50 cm^3 volume (contact)	500 cm^3 volume (contact)
^{56}Co	846.8	1.241	1.018	1.122	1.060
	1238.3	1.317	1.023	1.167	1.080
	2598.5	1.169	1.011	1.065	1.031
^{57}Co	122.1	1.136	1.007	1.004	1.001
	136.5	0.941	0.995	0.978	0.992
^{60}Co	1173.2	1.092	1.008	1.061	1.030
	1332.5	1.095	1.008	1.062	1.031
^{88}Y	898.0	1.193	1.013	1.069	1.032
	1836.1	1.211	1.014	1.079	1.036
^{133}Ba	81.0	1.306	1.019	1.173	1.082
	356.0	1.246	1.014	1.114	1.046
^{139}Ce	165.9	1.110	1.006	1.054	1.023
^{152}Eu	121.8	1.263	1.018	1.157	1.076
	778.9	1.154	1.011	1.092	1.043
	1408.0	1.277	1.016	1.133	1.054

Corrective factors: geometry transfer

Case where there is a change in the geometrical conditions of measurement, e.g:

- position of the source
- interposition of a screen
- shape of the source
- etc.



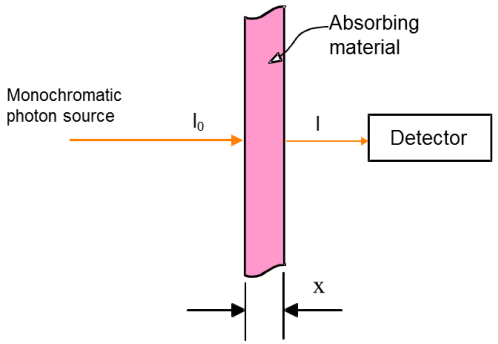
Change of distance → modification of the detection solid angle.

$$\mathcal{E}(E,P) = \mathcal{E}(E,P_0).\frac{\Omega(P)}{\Omega(P_0)}$$

Absorbing screen (important for low-energy photons): the corrective factor
- can be measured (transmission experiment),
- can be calculated if the material composition is well known

$$C_{Abs}(E) = exp(-\mu(E) \cdot x)$$

($\mu(E)$: linear attenuation coefficient of the material for energy E)



Corrective factors: geometry transfer

Change of the shape of the source: volume sources are subject to self-attenuation

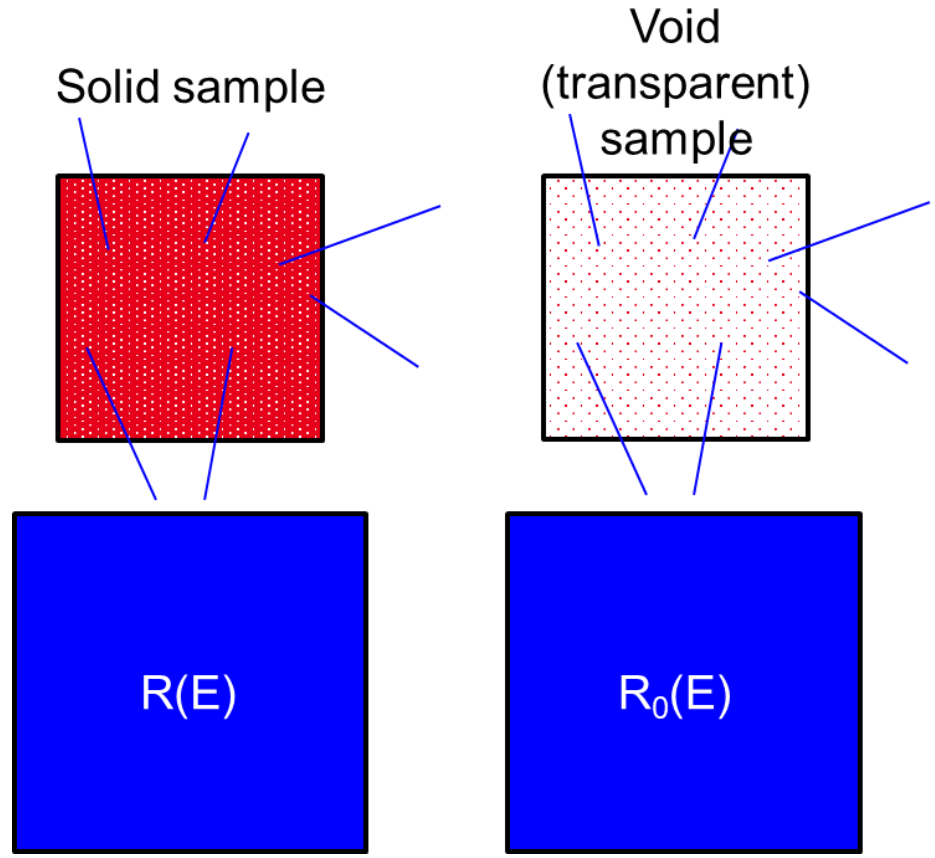
Number of photons exiting the sample: I
(What is recorded by the detector)

Number of photons emitted within the sample (activity) : I_0
(What we wish to determine)

$$C_{att} = \frac{I}{I_0}$$

Self-attenuation depends:

- on the composition of the sample material,
- on the sample dimensions and
- on the sample-to-detector distance



Corrective factors: efficiency transfer

As the coincidence summing corrective factors, the geometry transfer factors can be computed with

Dedicated codes:

- GESPECOR (Univ. Bucharest)
- EFFTRAN (Tim Vidmar – SCK-Mol) https://efftran.github.io/
- ETNA (LNHB)
- …

Full Monte Carlo codes:

- PENELOPE 2018
- GEANT4
- EGS
- MCNP
- …

Large variations in geometry should be avoided, as the associated uncertainties would lead to significant uncertainties in the results (efficiency, activity, photon emission intensity).

Photon emission intensities

$$I_i = \frac{N_i \cdot Corr_i{}'}{A \cdot \varepsilon_i \cdot t}$$

Efficiency calibration known → determination of photon emission intensities: 2 possibilities

- $A(Bq)$ know: absolute emission intensities $$I_{iA} = \frac{N_i \cdot Corr_i{}'}{A \cdot \varepsilon_i \cdot t}$$

- $A(Bq)$ unknow: relative emission intensities using the most intense peak (M) as reference

$$I_{MR} = \frac{N_M \cdot Corr_M{}'}{A \cdot \varepsilon_M \cdot t} = 100\%$$

$$I_{iR}(\%) = \frac{\varepsilon_M}{N_M \cdot Corr_M{}'} \cdot \frac{N_i \cdot Corr_i{}'}{\varepsilon_i}$$

Photon emission intensities: associated uncertainties

$$I_{iA} = \frac{N_i \cdot Corr_{i'}}{A \cdot \varepsilon_i \cdot t} = \frac{N_i \cdot \prod C_{ij}}{A \cdot \varepsilon_i \cdot t}$$

$\prod C_{ij}$: product of the corrective factors

$$\frac{\mathrm{u}(I_i A)}{I_{iA}} = \left[\frac{u^2(N_i)}{N_i^2} + \frac{u^2(\varepsilon_i)}{\varepsilon_i^2} + \frac{u^2(\mathrm{A})}{A} + \sum_j \frac{u^2(C_{ij})}{C_{ij}^2}\right]^{1/2}$$

$$I_{iR}(\%) = \frac{\varepsilon_M}{N_M \cdot \prod C_{MK}} \cdot \frac{N_i \cdot \prod C_{ij}}{\varepsilon_i}$$

$$\frac{\mathrm{u}(I_{iR})}{I_{iR}} = \left[\frac{u^2(N_i)}{N_i^2} + \frac{u^2(N_M)}{N_M^2} + \frac{u^2(\varepsilon_i)}{\varepsilon_i^2} + \frac{u^2(\varepsilon_M)}{\varepsilon_M^2} + \sum_j \frac{u^2(C_{ij})}{C_{ij}^2} + \sum_k \frac{u^2(C_{Mk})}{C_{Mk}^2} + u(\varepsilon_i, \varepsilon_M)\right]^{1/2}$$

$u(\varepsilon_1, \varepsilon_2)$: covariance due to correlation between efficiency values

Important factors to be checked by the evaluator

Efficiency calibration clearly mentioned

Uncertainty on efficiency hardly lower than 0.5%

Experimental spectrum provided: all peaks should be clearly identified (escape peaks)

Background must be taken into account

Low or moderate count rate (avoid pile-up effect)

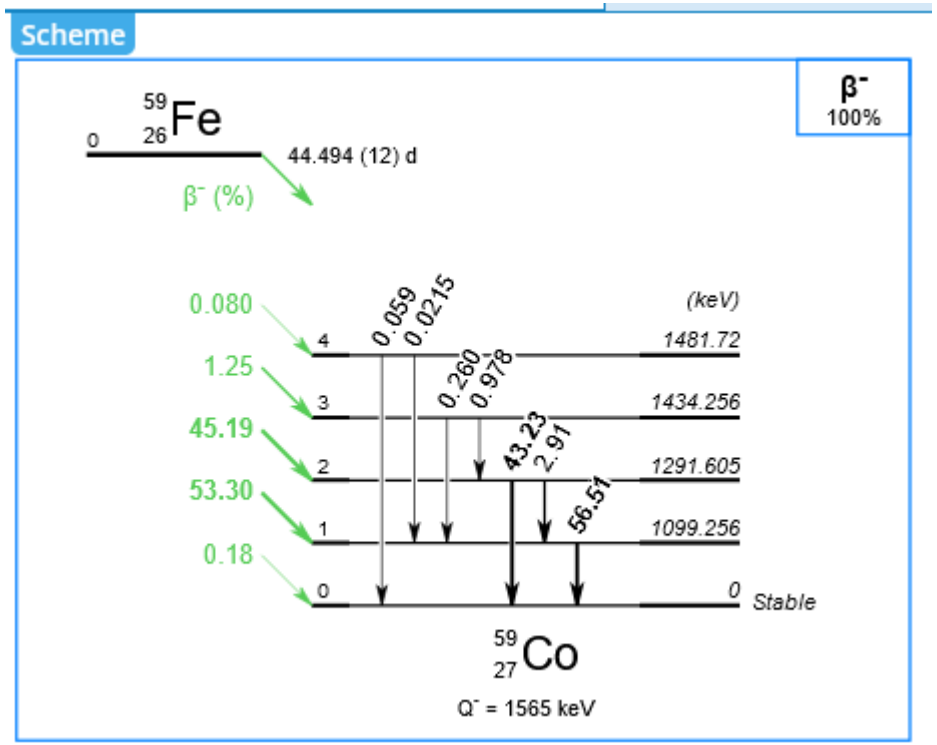
Peak area determined with the same method for calibration and measurement

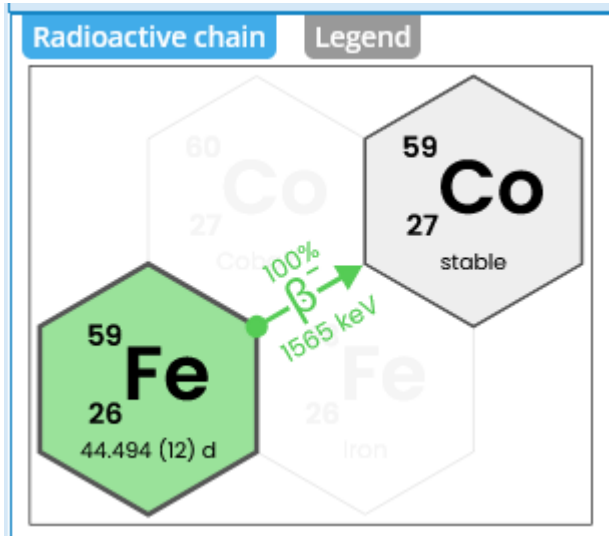
Corrective factors clearly mentioned and quantified

Example of peak fitting is interesting for processing of complex regions

Reference activity measurement method (checking of impurities)

Example of ^{59}Fe





^{59}Fe:
Beta- decay
Half-life: 44.494 (12) d
7 gamma transitions

Data and emissions file (ASCII text format): Fe-59.lara.txt

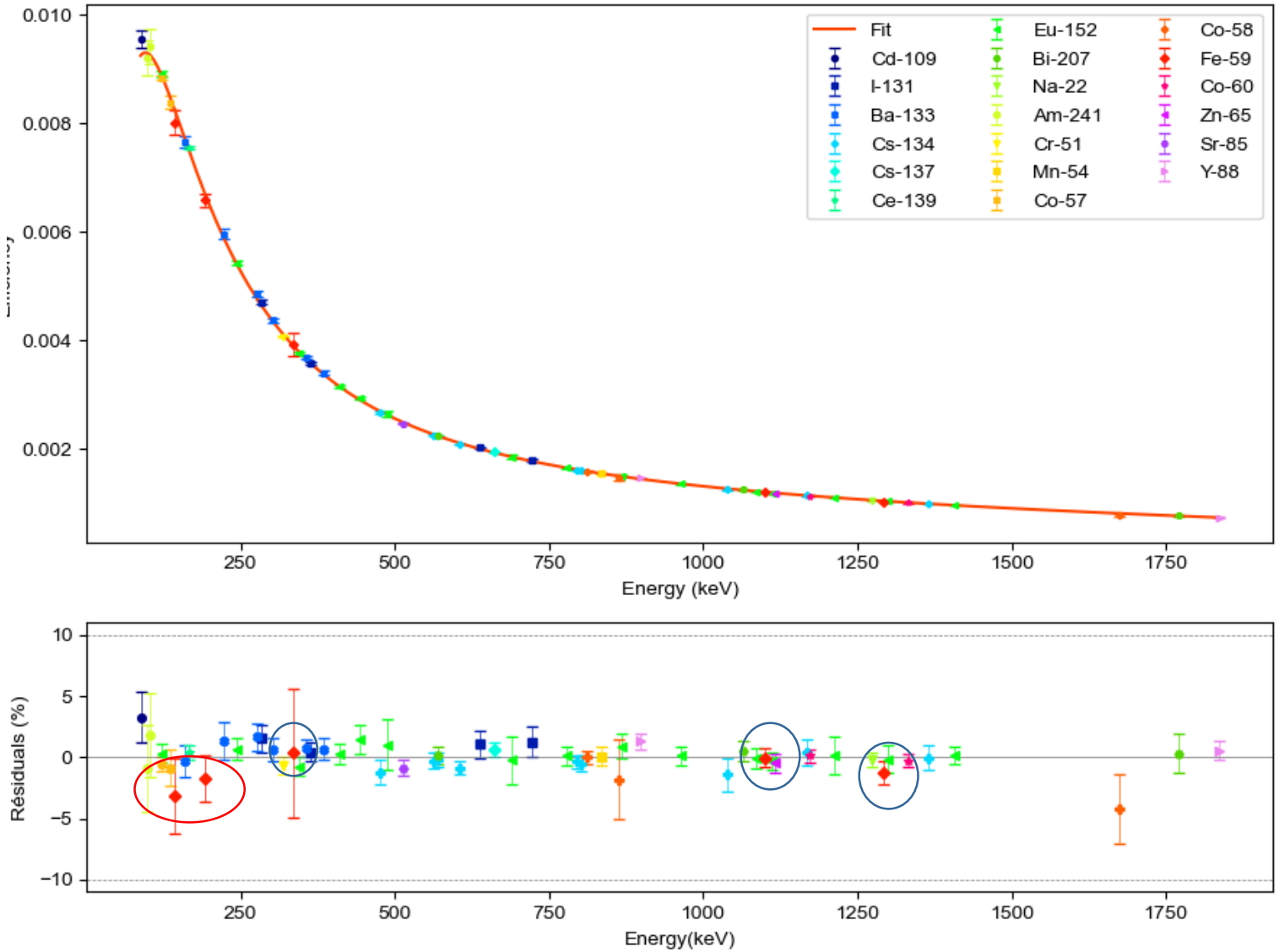
Emissions

Intensity threshold: **0.2%**
Coincidence threshold: **1%**
Emissions (**5** lines out of **12**) sorted by decreasing intensity

Energy (keV)	Intensity (%)	Type	Origin*	Levels Start*	Levels End*	Possible coincidence with (keV) / Possible sum of (levels)
1 099.245 (3)	56.51 (31)	γ	Co-59	1	0	192.349 (Σ=1 291.590)
1 291.590 (6)	43.23 (33)	γ	Co-59	2	0	(2→1)+(1→0)
192.349 (5)	2.91 (5)	γ	Co-59	2	1	1 099.245 (Σ=1 291.590)
142.651 (2)	0.978 (28)	γ	Co-59	3	2	
335.000 (6)	0.260 (11)	γ	Co-59	3	1	

Example of ^{59}Fe

Experimental calibration using ^{59}Fe



The points corresponding to low-energy peaks appear to be inaccurate

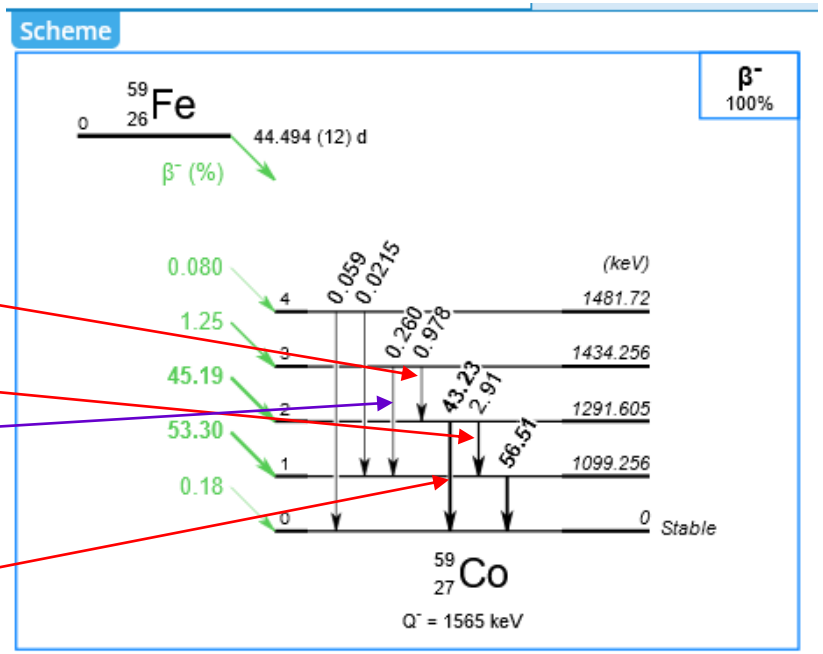
Experimental calibration using ^{59}Fe

Example of ^{59}Fe

Experimental calibration without ^{59}Fe

$$I_i = \frac{N_i \cdot Corr'}{A \cdot \varepsilon_i \cdot t}$$

Energy (keV)	Tabulated emission intensity (%)	Measured emission intensity (%)	Relative uncertainty (tabulated)	Relative difference (%)
142.651	0.978 (28)	0.946 (8)	2.86	-3.28
199.349	2.91 (5)	2.848 (18)	1.72	-2.11
355.000	0.260 (11)	0.278 (9)	4.23	6.83
1 099.245	56.51 (31)	56.51 (28)	0.55	0.00
1291.590	43.23 (33)	42.73 (22)	0.76	-1.15



Question on the decay scheme balance?

Impurities

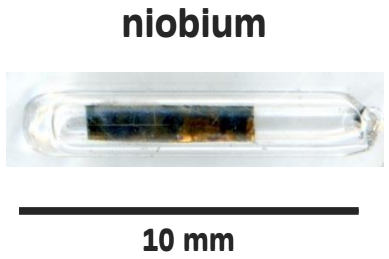
Gamma spectrometry is also used to **quantify gamma-ray impurities**

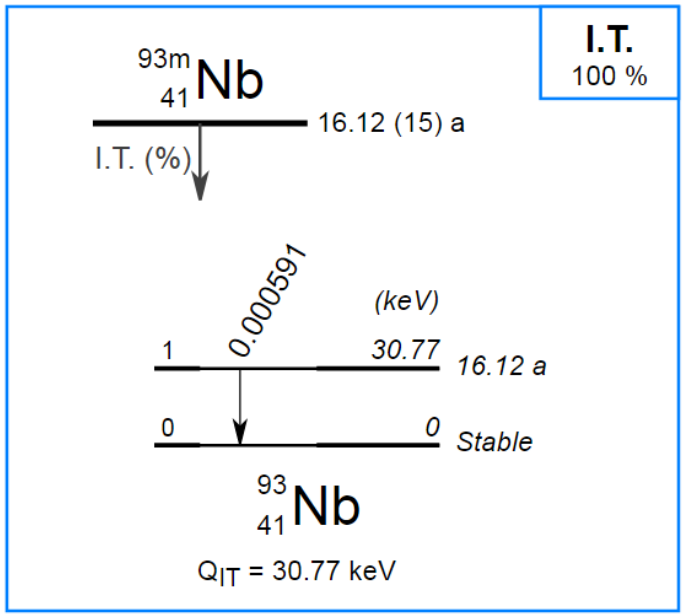
Important for absolute techniques to measure activity in the frame of radionuclides standardisation

Recent example: activity of ^{93m}Nb using the TDCR method

Niobium dosimeters are used in reactors to characterise neutron fluxes in the energy range around **1 MeV**.

- Information based on the activity of ^{93m}Nb resulting from the activation reaction **^{93}Nb(n, n')**.
- ^{93m}Nb decays by isomeric transition with a strong internal conversion coefficient.





^{93m}Nb decay scheme

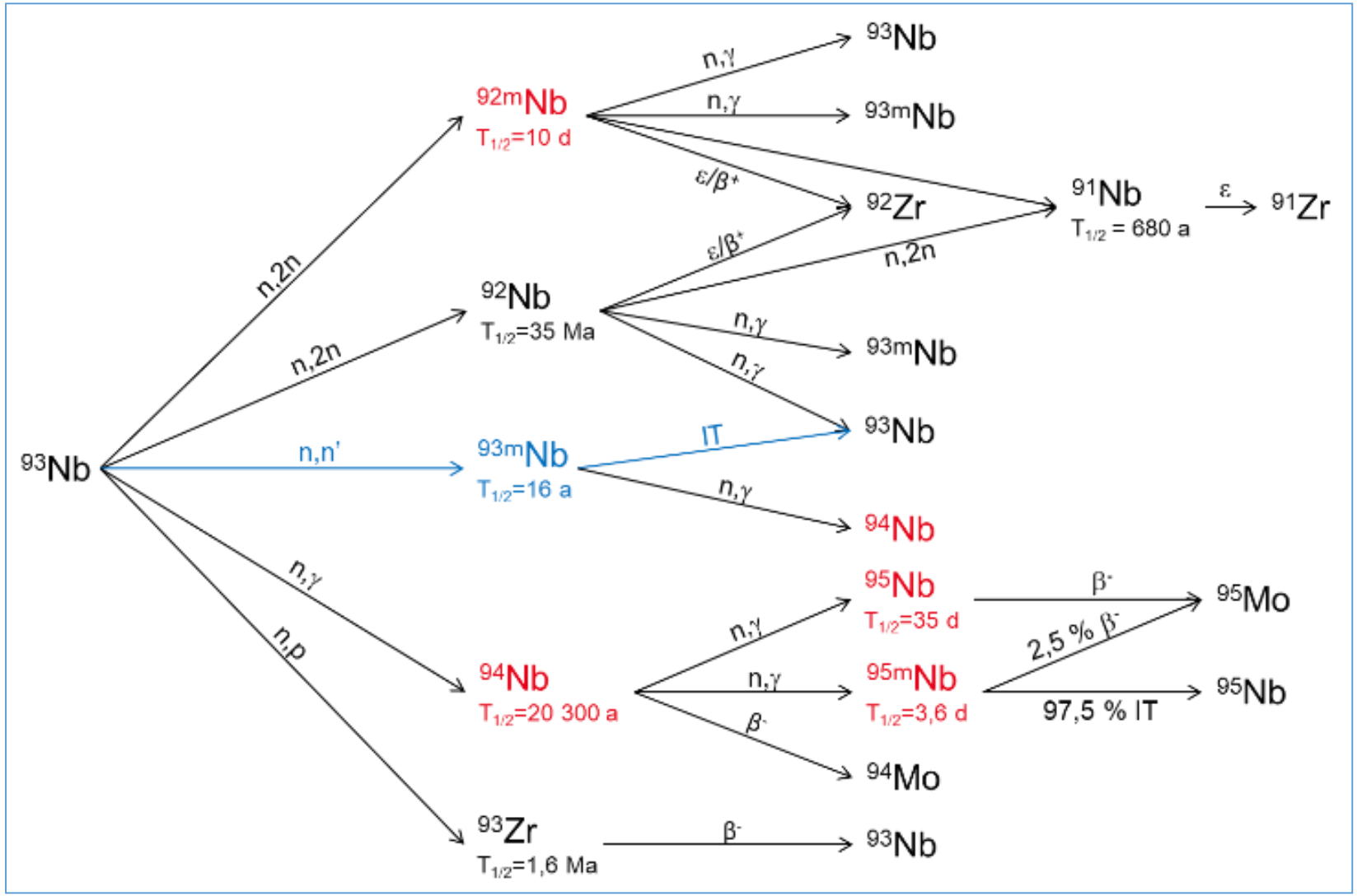
Activated Nb impurities

Activation products from ^{93}Nb
(n, 2n) (n, γ), (n, p) reactions

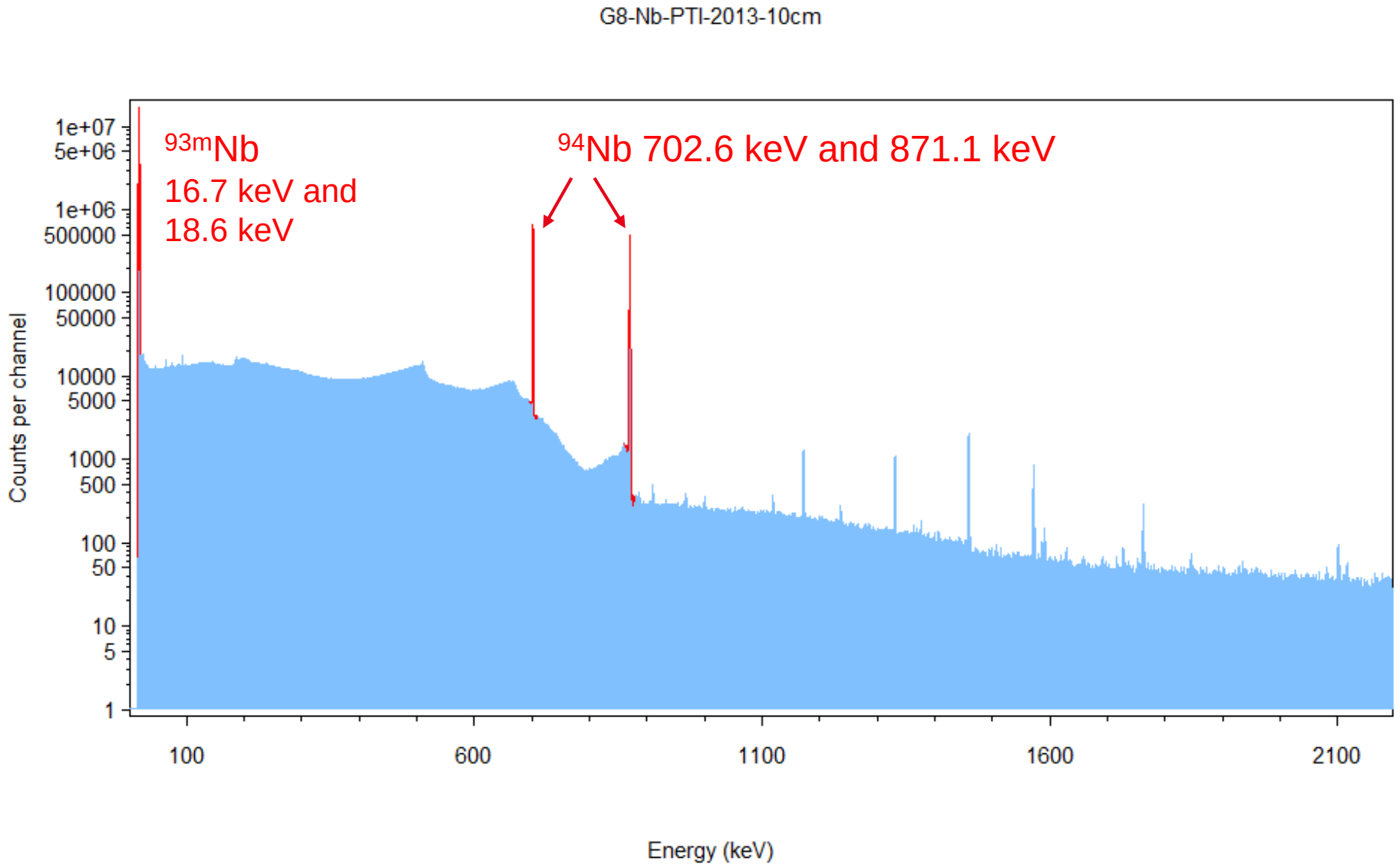
Peaks due to the decay of Nb activation products

- Gamma rays*
- X rays (Zr, Mo)
- **Nb K-X rays**

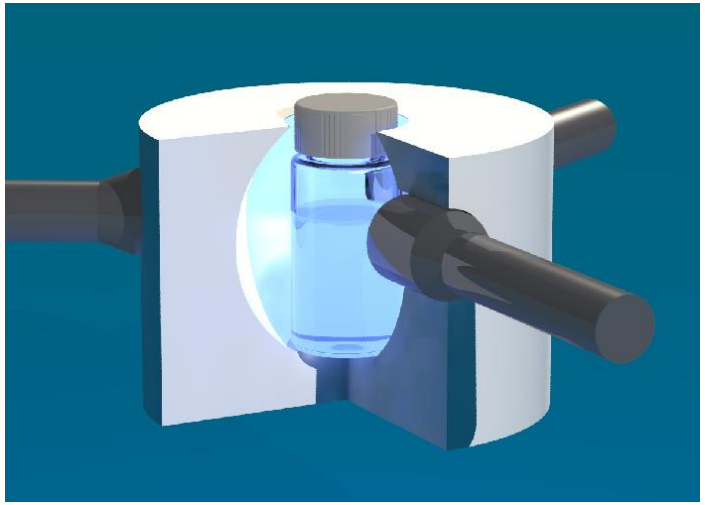
*Quantification using gamma-ray spectrometry



Impurities: gamma-ray spectrum of ^{93m}Nb



Activity measurement



Liquid solution prepared from dissolved niobium metal foil

TDCR liquid scintillation counting system

Detection efficiency for ^{94}Nb is the same as ^{93m}Nb -> corrective factor directly proportional to the relative activities.

Here the **corrective factor for the ^{93m}Nb activity is 2.4%** !

Thank you for your attention

Reference:

Uncertainties in gamma-ray spectrometry, Metrologia (52- Vol3) 2015 https://doi.org/10.1088/1681-7575/aa853b