Fitting in gamma-ray spectrometry

Fitting in gamma-ray spectrometry

2 main applications:

Efficiency curves

Full-energy peaks

Experimental calibration : series of discrete values (E, ε(E))

Fitting function: to get an efficiency value for any energy (interpolation)

Checking of the consistency of the input data

- E.g: residuals versus radionuclide

Influence of correlations between input data

EXAMPLE OF EFFICIENCY CALCULATION

				^{133}Ba
Spectrum name:				
Geometry	G91			
Calibration distance (mm)	102,7	±	0,1	0,1
Source :	818A			

half-life	10,539	a	0,006
half-life	3,3258E+08	s	1,9E+05
half-life	5,5430E+06	min	3,2E+03
half-life	9,2383E+04	h	5,3E+01
half-life	3,8493E+03	j	2,2E+00
half-life	1,0539E+01	a	6,0E-03

1 a =	365,242	j
1 a =	3,2E+07	s

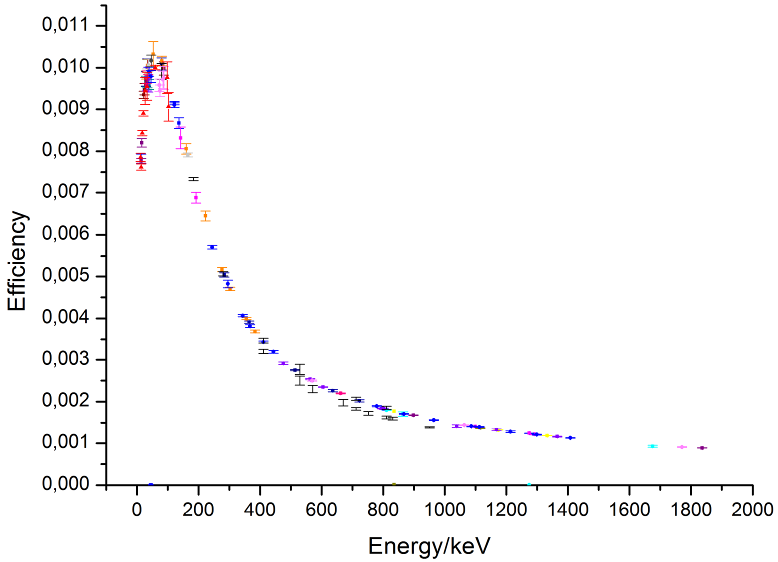
Date/time reference :	1/10/11 12:00	UTC	**Activity (Reference time)**	**19 757**	Bq	±	0,43	%	
Date/time measurement :	19/9/15 15:24	UTC	Corr. Radioactive decay :	1,2982		±	0,01	%	To get the counting at the reference data
Active time	50 000	s							
Real time	51 124	s	Corr. mesuring time:	1,00005		±	3,033E-06	%	To get the counting at the measurement starting time

Energy (keV)	Net pic area	Absolute unc.	Relative unc. (%)	Peak area with decay corrections	Relative unc. (%)	Intensity	unc (%)	Correction for coincidence.	unc (%)	Correction (other)	unc (%)	Efficiency	unc (%)	Absolute
30,85	7082934	4712	0,07	9195285	0,2	**0,962**	0,84	1,016	0,2	1,00	0,0	**9,829E-03**	1,0	9,7E-05
35,22	1709062	2315	0,14	2218757	0,2	**0,2269**	1,38	1,016	0,2	1,00	0,0	**1,006E-02**	1,5	0,00015
53,16	164030	592	0,36	212949	0,4	**0,0214**	2,80	1,025	0,3	1,00	0,0	**1,033E-02**	2,9	0,0003
80,90	2722682	1797	0,07	3534670	0,2	**0,3594**	0,99	1,020	0,2	1,00	0,0	**1,016E-02**	1,1	0,00011
160,61	39144	443	1,13	50818	1,1	**0,0064**	0,94	0,999	0,0	1,00	0,0	**8,057E-03**	1,5	0,00012
223,24	21386	288	1,35	27764	1,4	**0,00450**	1,11	1,032	0,3	1,00	0,0	**6,448E-03**	1,8	0,00012
276,40	272874	556	0,20	354253	0,3	**0,0713**	0,84	1,027	0,3	1,00	0,0	**5,164E-03**	1,0	5,3E-05
302,85	640941	830	0,13	832089	0,2	**0,1831**	0,60	1,023	0,2	1,00	0,0	**4,705E-03**	0,8	3,8E-05
356,01	1854400	1204	0,06	2407440	0,2	**0,6205**	0,31	1,017	0,2	1,00	0,0	**3,993E-03**	0,6	2,4E-05
383,85	250087	444	0,18	324671	0,3	**0,0894**	0,67	1,003	0,0	1,00	0,0	**3,687E-03**	0,8	3,1E-05

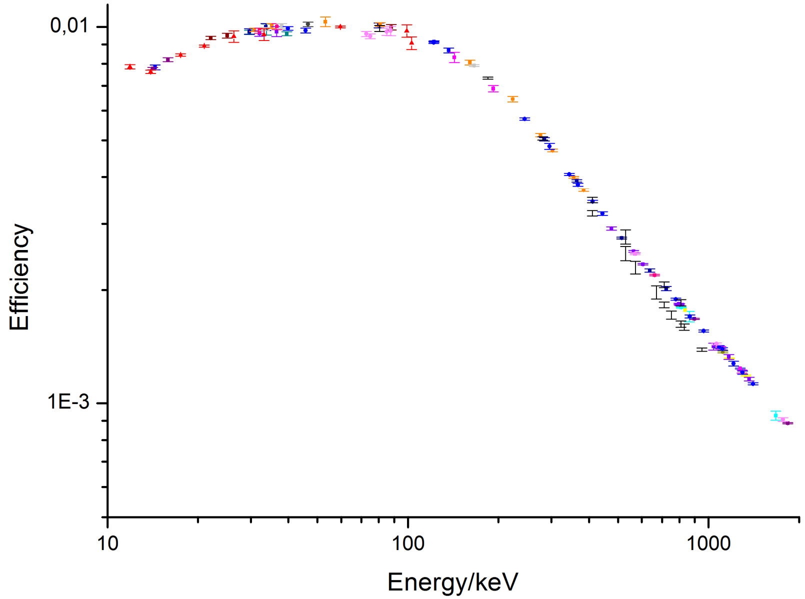
Decay data (Nucléide) KRI 2015

Efficiency calibration: fitting function

Experimental calibration of a HPGe detector with point sources at 10 cm from the detector window



Eff (E) = f(E)



Log(Eff (E)) = f(log(E))

Logarithmic scale : easier !

Efficiency calibration: fitting function

Frequently used fitting functions :

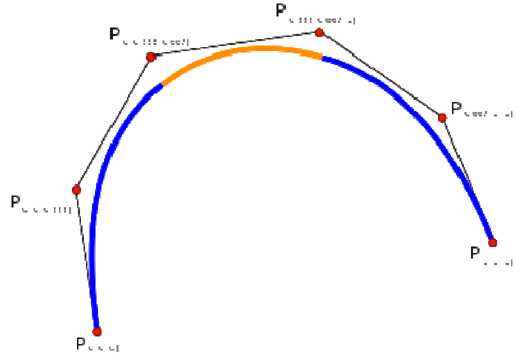
Log-log polynomial: $\ln\left(\varepsilon(E)\right) = \sum_{i=0}^{n} a_i \cdot (\ln(E))^i$

Log against 1/E: $\ln\left(\varepsilon(E)\right) = \sum_{i=0}^{n} a_i \cdot E^{-i}$

- a_i coefficients determined by a least squares fitting method
- Polynomial degree >> number of experimental data
- Two parts for the maximum region (E ~ 100 keV) “knee”

Other functions i.e $\varepsilon(E) = \frac{1}{E}\sum_{i=0}^{n} a_i \cdot \left(\ln\left(\frac{E}{E_0}\right)\right)^{i-1}$ ….

Efficiency calibration: fitting function



SPLINE functions

- defined piecewise by polynomials
- ability to approximate complex shapes
- avoid oscillations for high degrees

(PTB : H. Janssens- NIM A 286(3), 1990, 398-402).

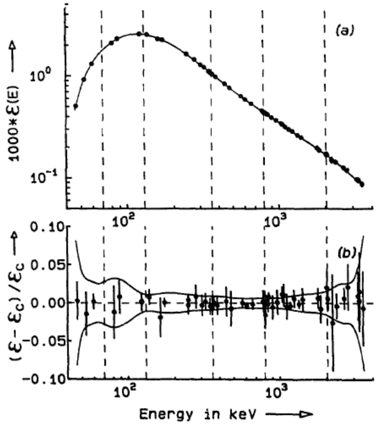


Fig. 1. (a) Experimental efficiency calibration data (dots) for a 67.5 cm³ Ge(Li) detector and fitted spline curve (solid line). (b) Residuals of the fit (dots) and threefold relative standard deviation of the spline curve (solid line). Positions of interior knots are indicated by vertical broken lines.

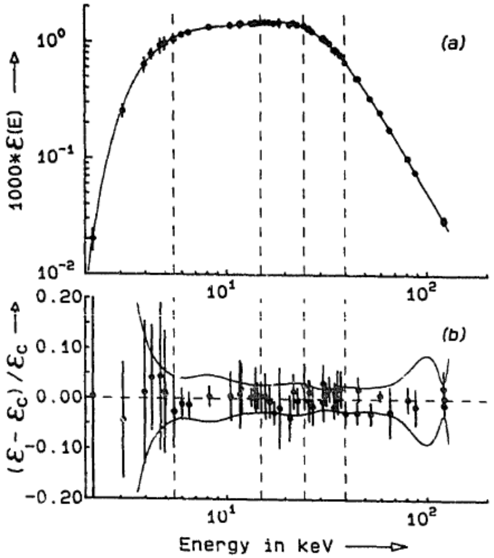


Fig. 2. Same as fig. 1. for a 200 mm^2 × 5 mm Si(Li) detector.

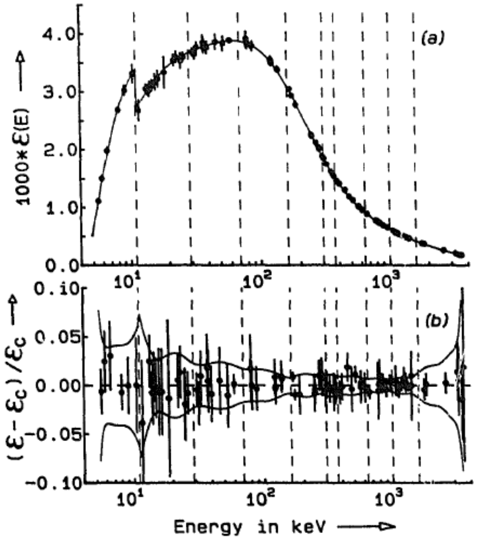
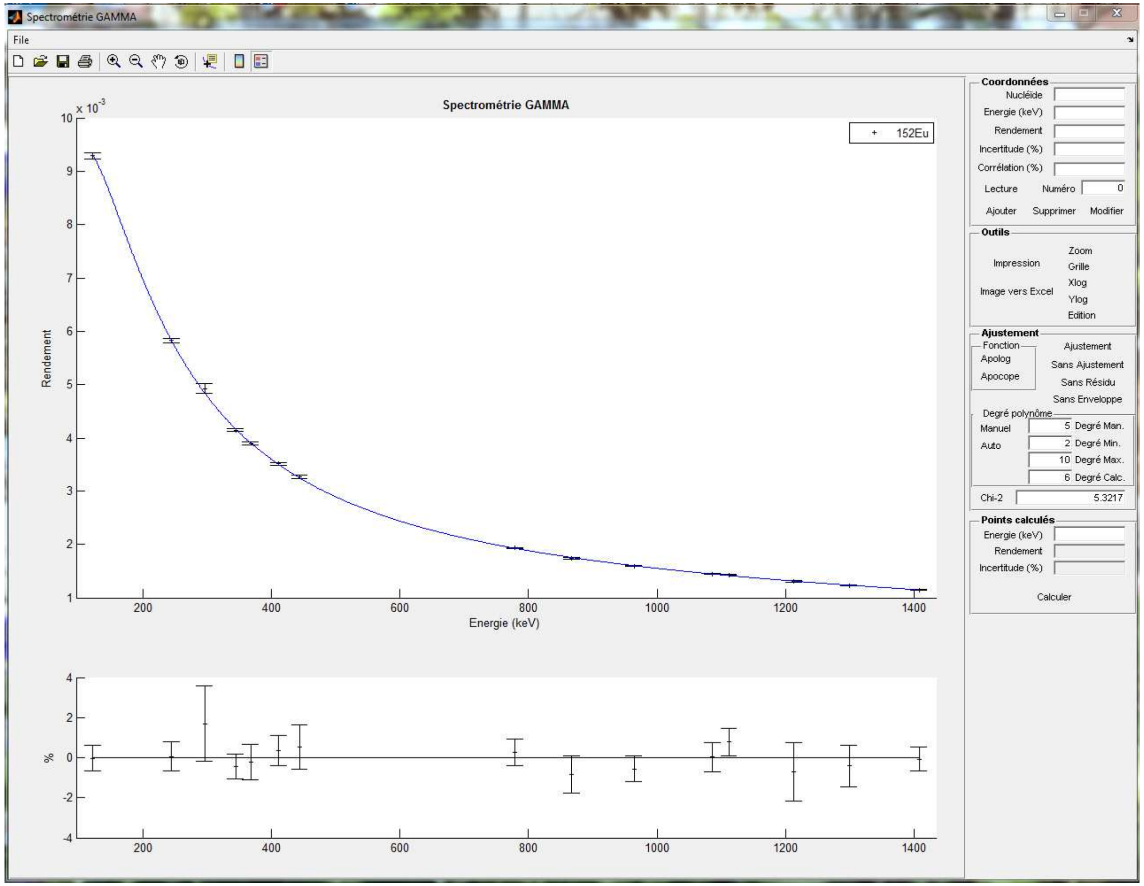


Fig. 3. Same as fig. 1 for a 127 cm^3 HPGe detector.

Fitting using a log-log function

^{152}Eu : 15 experimental values



Fitting using a log-log function :Influence of weighting

Example fitting with log-log function using 15 experimental values from calibration with ^{152}Eu:

Fit using a log-log polynomial (degree 4)

(1) without weighting

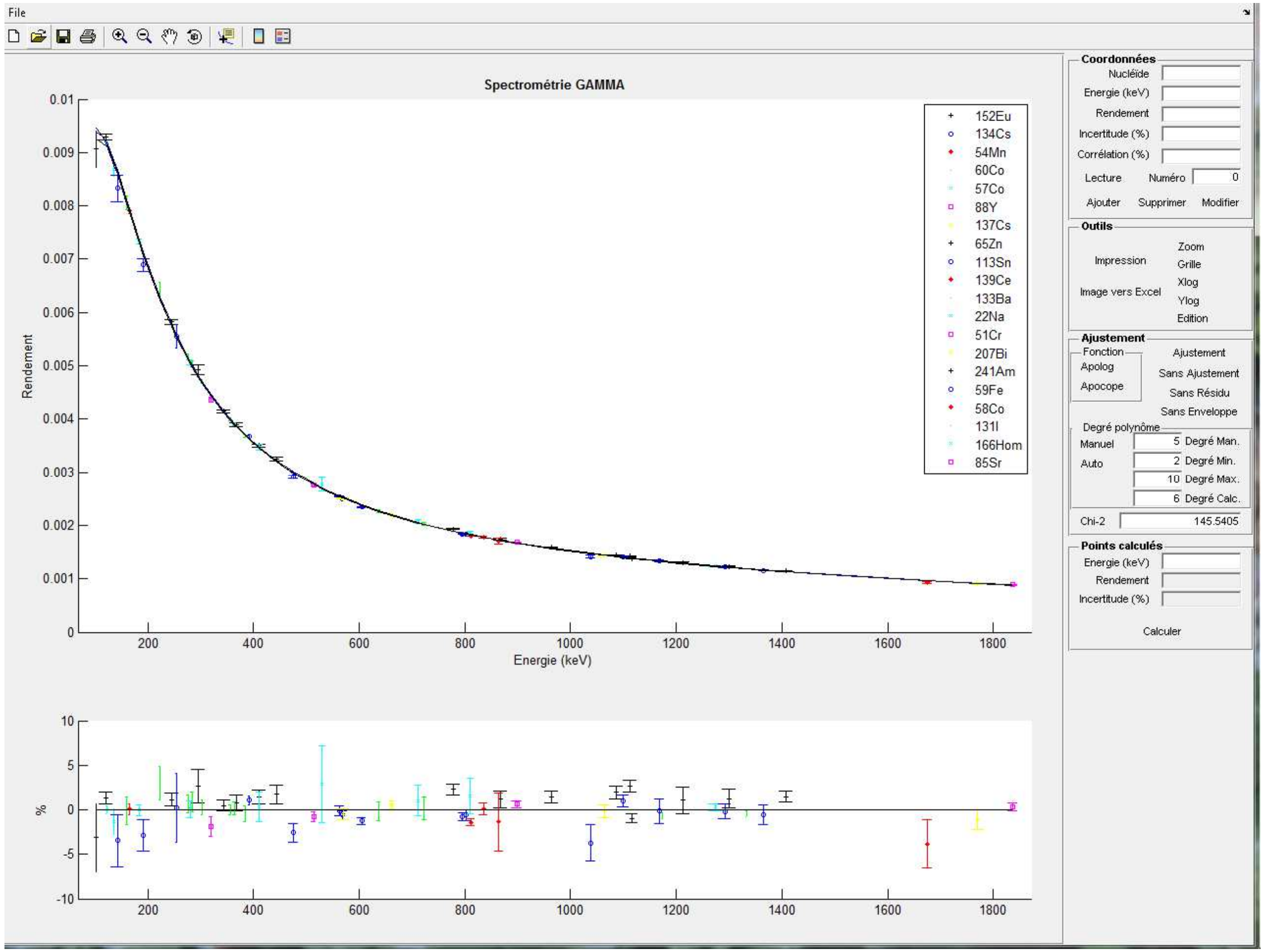
(2) with weighting

E (keV)	Experimental efficiency	Relative uncertainty (%)	Fitted efficiency (1)	Fitted efficiency (2)
121.8	8.93E-03	1.5	8.92E-03	8.93E-03
244.7	4.64E-03	1.8	4.69E-03	4.62E-03
344.3	2.95E-03	1.5	2.93E-03	2.96E-03
411.3	2.35E-03	3.6	2.30E-03	2.35E-03
444	2.15E-03	2.7	2.08E-03	2.13E-03
564.5	1.41E-03	12.3	1.54E-03	1.58E-03
688.6	1.26E-03	7.8	1.23E-03	1.25E-03
778.9	1.10E-03	1.5	1.08E-03	1.09E-03
867.4	9.80E-04	2.7	9.70E-04	9.70E-04
964.1	8.70E-04	1.5	8.70E-04	8.70E-04
1086.5	7.70E-04	1.8	7.70E-04	7.70E-04
1112.1	7.50E-04	1.8	7.50E-04	7.50E-04
1212.9	6.80E-04	5.4	6.90E-04	6.90E-04
1299.8	6.40E-04	5.1	6.40E-04	6.40E-04
1408	5.90E-04	1.5	5.90E-04	5.90E-04

Fitting using a log-log function : several radionuclides

^{152}Eu + other nuclides

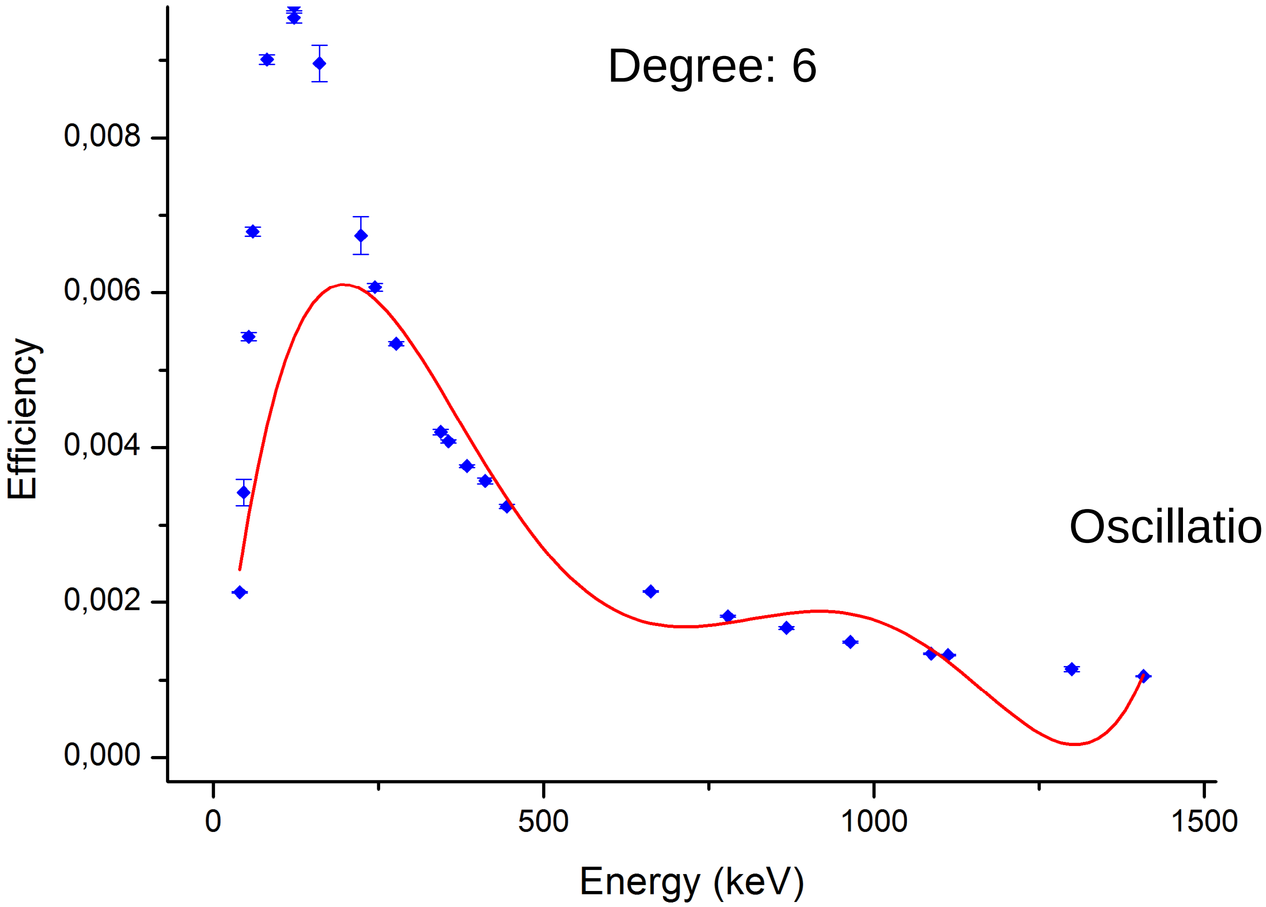
Deviation of ^{152}Eu (black)



Fitting using a log-log function : several radionuclides

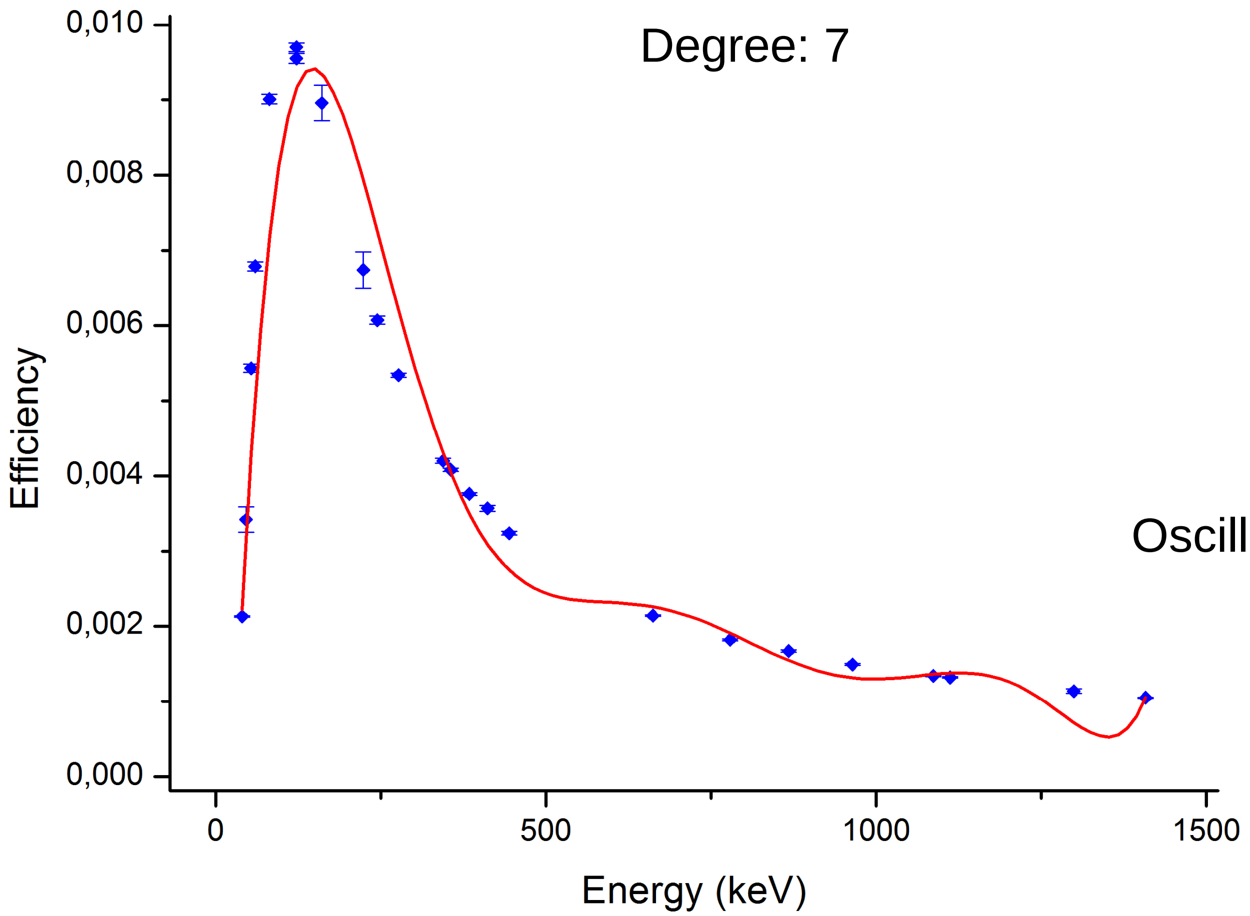
	^{152}Eu only		All data		
Energy (keV)	Efficiency (X 10^3)	Uncertainty (%)	Efficiency (X 10^3)	Uncertainty (%)	Ratio
120	9.320	0.924	9.200	0.500	1.013
150	8.551	1.614	8.403	0.476	1.018
200	6.972	1.345	6.876	0.463	1.014
250	5.705	0.909	5.646	0.421	1.010
300	4.778	0.791	4.736	0.405	1.009
400	3.590	0.784	3.553	0.386	1.010
500	2.889	0.844	2.850	0.374	1.014
600	2.435	0.899	2.394	0.373	1.017
700	2.117	0.882	2.077	0.373	1.019
750	1.991	0.851	1.953	0.372	1.020
800	1.881	0.816	1.844	0.369	1.020
900	1.698	0.763	1.664	0.362	1.020
1000	1.550	0.757	1.520	0.359	1.020
1100	1.428	0.778	1.402	0.360	1.019
1250	1.278	0.781	1.256	0.369	1.017
1500	1.085	1.113	1.070	0.386	1.014
1750	0.938	3.026	0.926	0.449	1.013
2000	0.820	6.466	0.808	0.706	1.014

Fitting using a log-log function : influence of polynomial degree



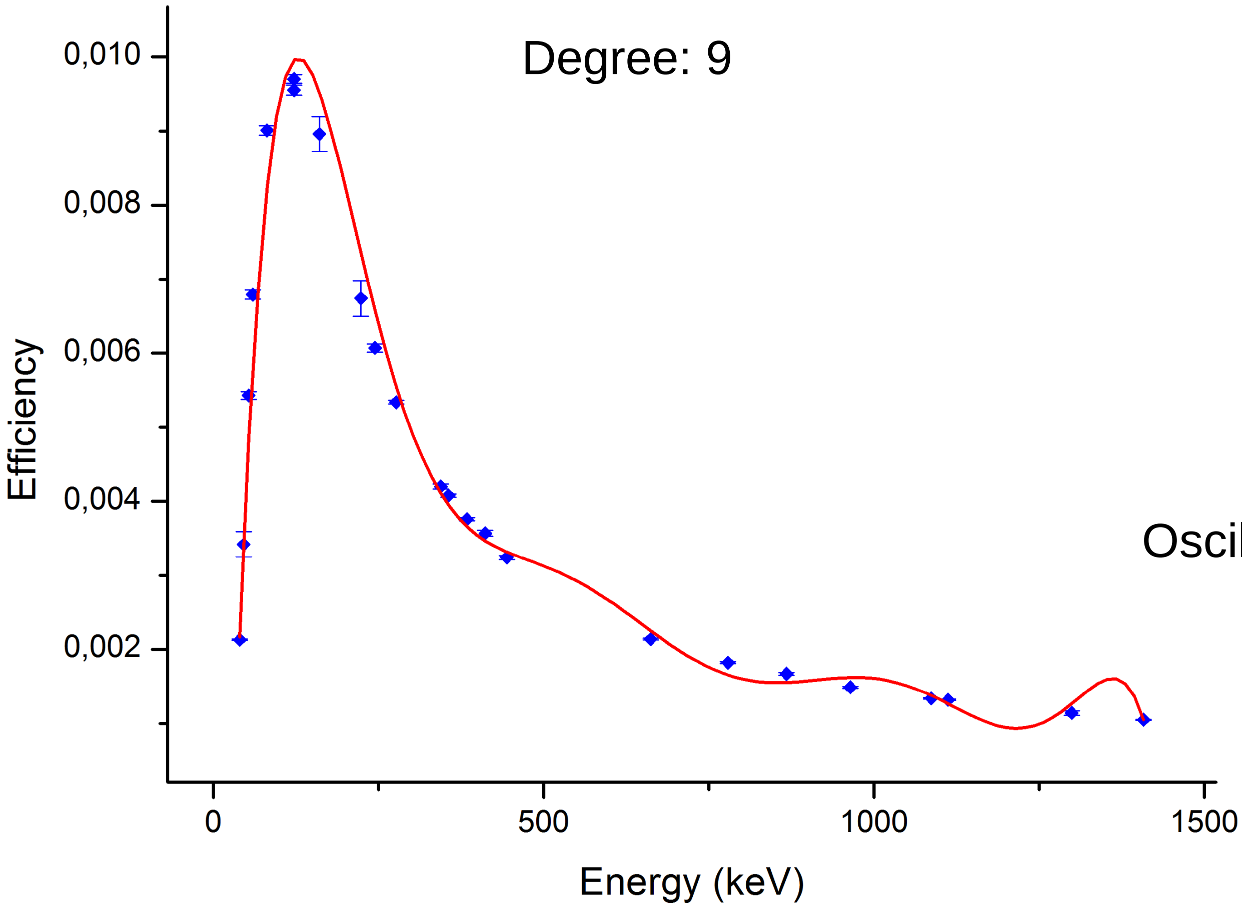
Oscillations !

Fitting using a log-log function : influence of polynomial degree



Oscillations !

Fitting using a log-log function : influence of polynomial degree



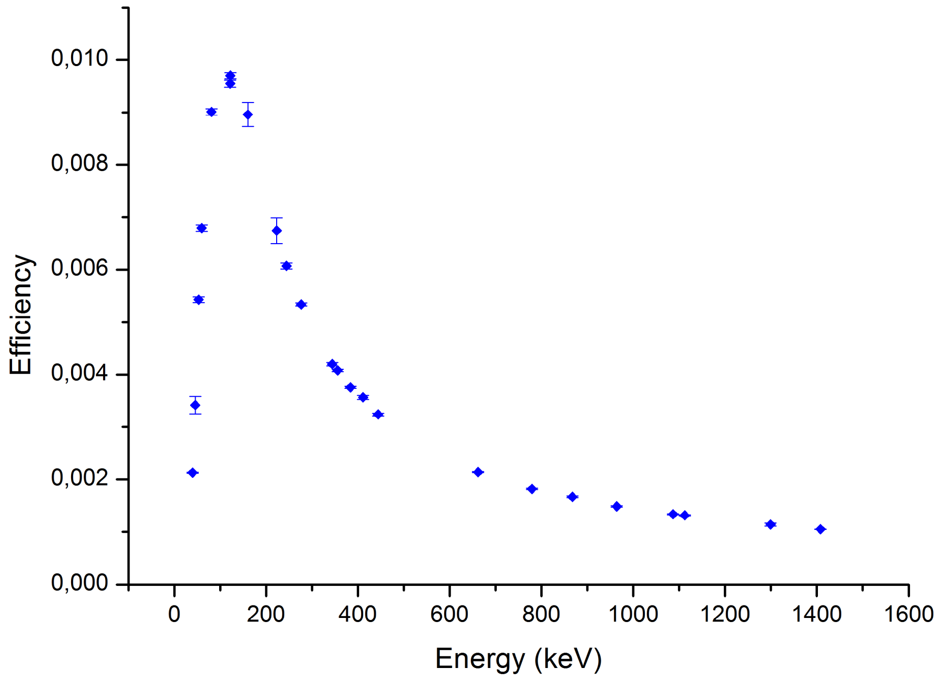
Oscillations !

Fitting using a log-log function : influence of input data

TEST CASE: SET OF EXPERIMENTAL DATA

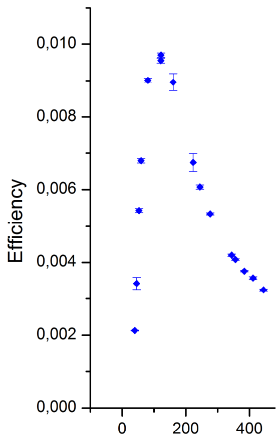
« Monoenergetic » : ^{241}Am, ^{57}Co, ^{137}Cs

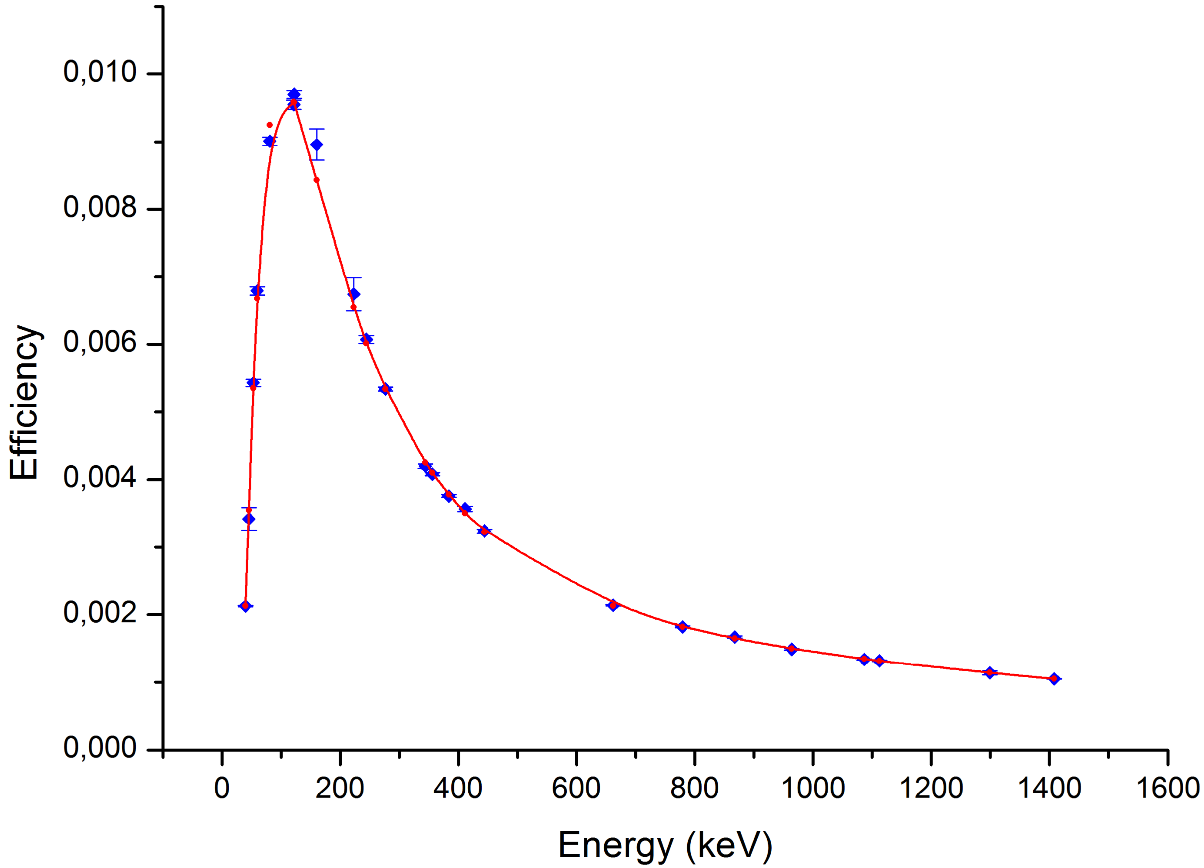
« Multigamma » : ^{133}Ba and ^{152}Eu



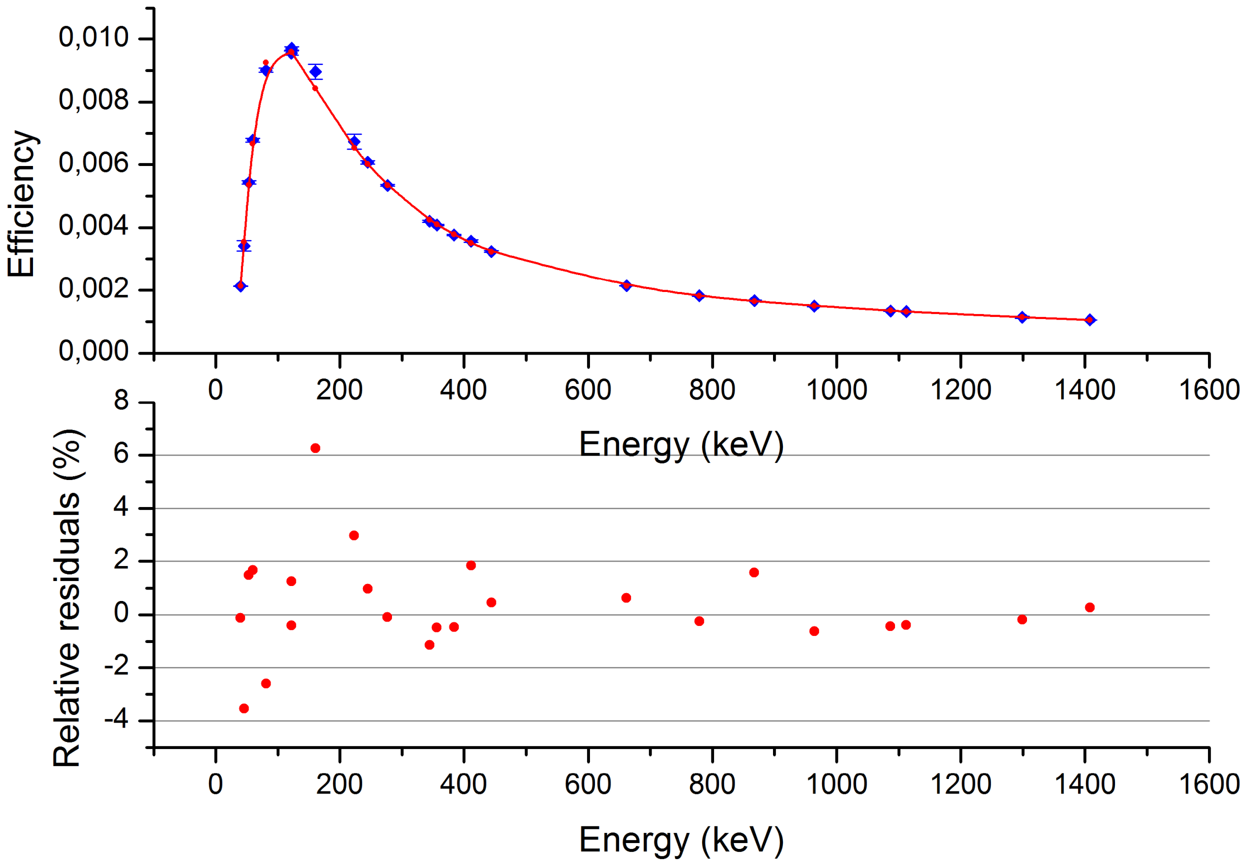
Radionuclide	Energy (keV)	Efficiency	Relative uncertainty (%)
152Eu	39.9	2.13E-03	0.423
152Eu	45.7	3.42E-03	4.971
133Ba	53.2	5.43E-03	0.994
241Am	59.6	6.79E-03	0.898
133Ba	81	9.01E-03	0.700
152Eu	121.8	9.55E-03	0.701
57Co	122.1	9.70E-03	0.598
133Ba	160.6	8.96E-03	2.600
133Ba	223.2	6.74E-03	3.605
152Eu	244.7	6.07E-03	0.906
133Ba	276.4	5.34E-03	0.506
152Eu	344.3	4.20E-03	0.809
133Ba	356	4.08E-03	0.490
133Ba	383.8	3.76E-03	0.505
152Eu	411.4	3.57E-03	1.092
152Eu	444	3.24E-03	0.802
137Cs	661.7	2.14E-03	0.514
152Eu	778.9	1.82E-03	0.716
152Eu	867.4	1.67E-03	1.016
152Eu	964	1.49E-03	0.805
152Eu	1086.6	1.34E-03	0.671
152Eu	1112.1	1.32E-03	0.608
152Eu	1299.1	1.14E-03	2.719
152Eu	1408	1.06E-03	0.569

FIT USING LOG/LOG FUNCTION





FIT USING LOG/LOG FUNCTION



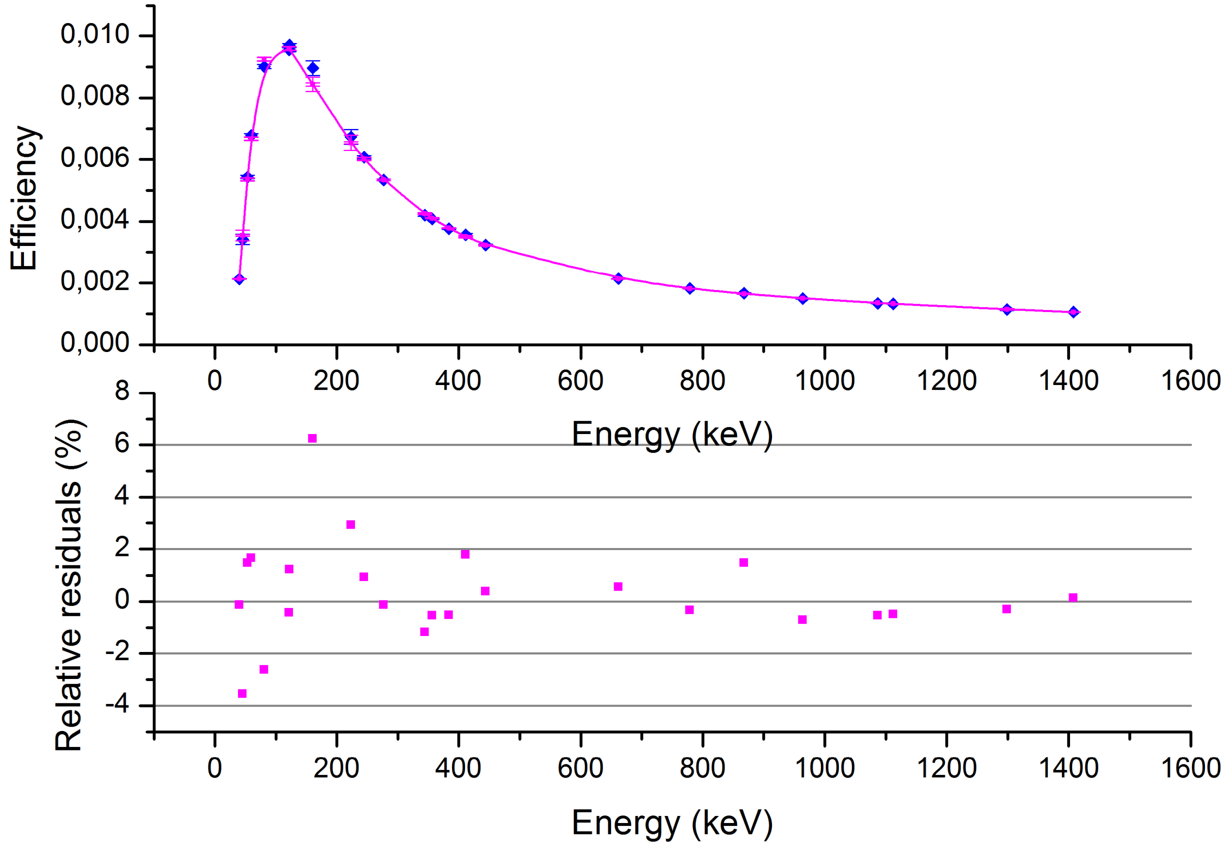
Energy	Efficiency	Relative uncertainty
50	0.004605	0.601
100	0.009827	0.413
136.54	0.009200	0.405
200	0.007190	0.399
302.8	0.004865	0.285
500	0.002842	0.337
1000	0.001451	0.351
1400	0.001059	0.536

FIT USING LOG/LOG FUNCTION

		Experimental data		Fitted data		
Radionuclide	**Energy (keV)**	**Efficiency**	**Relative uncertainty (%)**	**Efficiency**	**Relative uncertainty (%)**	**Relative residuals (%)**
152Eu	39.9	2.13E-03	0.423	0.002132	0.422	-0.12
152Eu	45.7	3.42E-03	4.971	0.003546	0.513	-3.54
133Ba	53.2	5.43E-03	0.994	0.005354	0.619	1.48
241Am	59.6	6.79E-03	0.898	0.006678	0.586	1.67
133Ba	81	9.01E-03	0.700	0.009247	0.432	-2.61
152Eu	121.8	9.55E-03	0.701	0.009590	0.408	-0.41
57Co	122.1	9.70E-03	0.598	0.009584	0.408	1.26
133Ba	160.6	8.96E-03	2.600	0.008432	0.404	6.27
133Ba	223.2	6.74E-03	3.605	0.006546	0.384	2.97
152Eu	244.7	6.07E-03	0.906	0.006014	0.362	0.97
133Ba	276.4	5.34E-03	0.506	0.005340	0.320	-0.09
152Eu	344.3	4.20E-03	0.809	0.004249	0.250	-1.14
133Ba	356	4.08E-03	0.490	0.004100	0.247	-0.48
133Ba	383.8	3.76E-03	0.505	0.003781	0.251	-0.46
152Eu	411.4	3.57E-03	1.092	0.003507	0.267	1.84
152Eu	444	3.24E-03	0.802	0.003229	0.294	0.45
137Cs	661.7	2.14E-03	0.514	0.002129	0.358	0.63
152Eu	778.9	1.82E-03	0.716	0.001820	0.324	-0.25
152Eu	867.4	1.67E-03	1.016	0.001648	0.318	1.57
152Eu	964	1.49E-03	0.805	0.001499	0.340	-0.62
152Eu	1086.6	1.34E-03	0.671	0.001348	0.368	-0.44
152Eu	1112.1	1.32E-03	0.608	0.001320	0.370	-0.39
152Eu	1299.1	1.14E-03	2.719	0.001142	0.388	-0.19
152Eu	1408	1.06E-03	0.569	0.001052	0.556	0.26

Uncertainty on fitted values twice lower than the lowest experimental uncertainties

FIT WITH UNCERTAINTY THRESHOLD



Energy	Efficiency	Relative uncertainty
50	0.004606	0.735
100	0.009829	0.590
136.54	0.009202	0.585
200	0.007193	0.581
302.8	0.004867	0.510
500	0.002844	0.540
1000	0.001453	0.549
1400	0.001060	0.683

Add minimum experimental uncertainty in quadrature

- Several energies from the same radionuclide:
- Activity
- Decay scheme (balancing the transition probabilities)

CORRELATION BETWEEN INPUT DATA
1 RADIONUCLIDE -> SEVERAL ENERGIES

$$\varepsilon = \frac{N_i}{A \cdot I \cdot t} \cdot C$$

Associated variance :

$$u_c^2(\varepsilon) = \left(\frac{1}{A \cdot I}\right)^2 \cdot u^2(N) + \left(\frac{N}{A^2 \cdot I}\right)^2 \cdot u^2(A) + \left(\frac{N}{A \cdot I^2}\right)^2 u^2(I)$$

In logarithmic scale :

$$u_c^2(\ln(\varepsilon)) = \frac{u^2(\varepsilon)}{\varepsilon^2} \Longleftrightarrow u_c^2(\ln(\varepsilon)) = \frac{u^2(N)}{N^2} + \frac{u^2(A)}{A^2} + \frac{u^2(I)}{I^2}$$

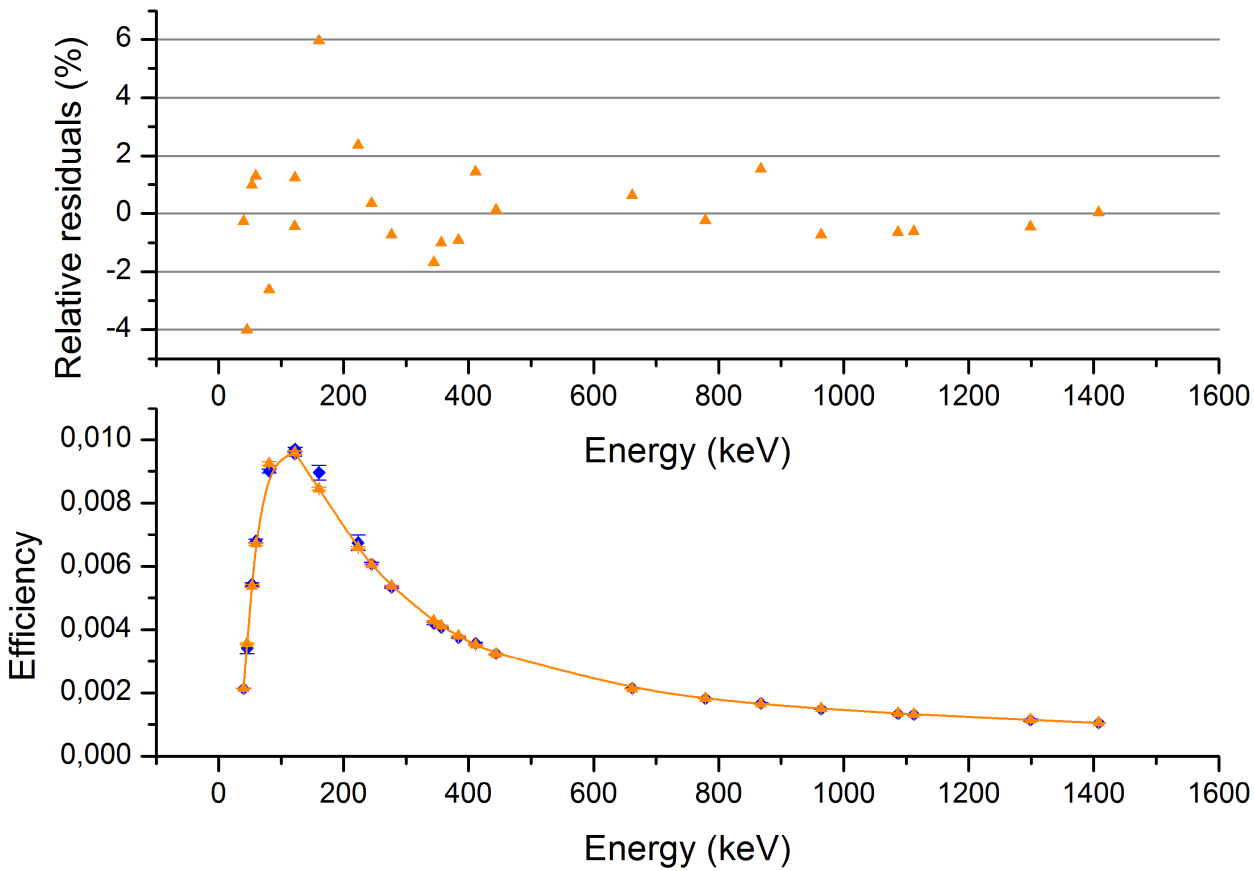
Covariance between two efficiency values (log scale) :

$$cov\left(\ln(\varepsilon_i), ln(\varepsilon_j)\right) = \frac{cov(\varepsilon_i, \varepsilon_j)}{\varepsilon_i \cdot \varepsilon_j} = \left[\frac{u^2(N_i)}{N_i^2} + \frac{u^2(I_i)}{I_i^2}\right] \cdot \delta_{ij} + \frac{u^2(A)}{A^2} + \frac{cov(I_i, I_j)}{I_i \cdot I_j}$$

Neglecting covariances between intensity values (unknown !)

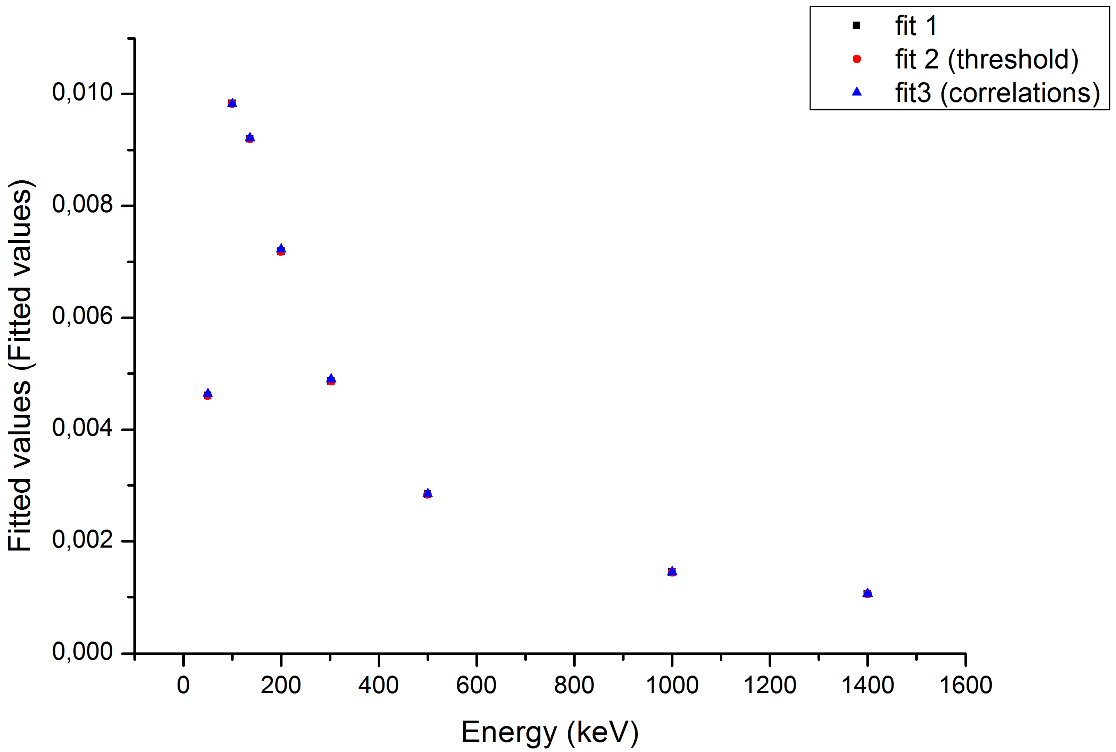
$$cov\left(\ln(\varepsilon_i), ln(\varepsilon_j)\right) = \frac{cov(\varepsilon_i, \varepsilon_j)}{\varepsilon_i \cdot \varepsilon_j} = \left[\frac{u^2(N_i)}{N_i^2} + \frac{u^2(I_i)}{I_i^2}\right] \cdot \delta_{ij} + \frac{u^2(A)}{A^2}$$

FIT WITH CORRELATIONS

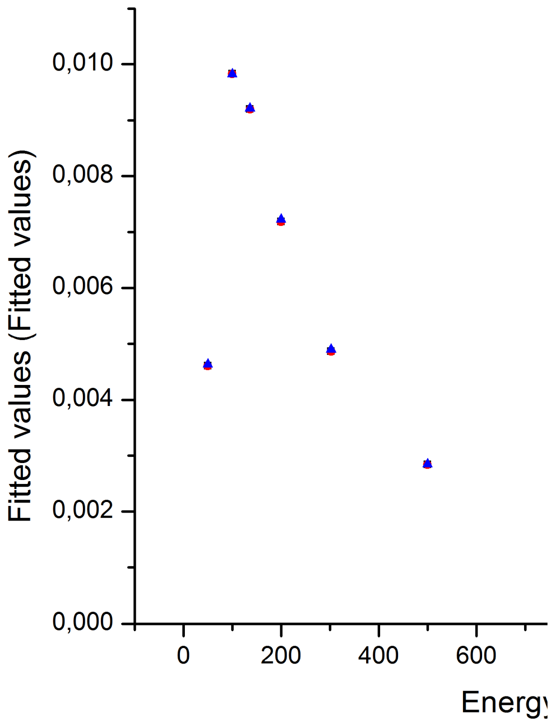


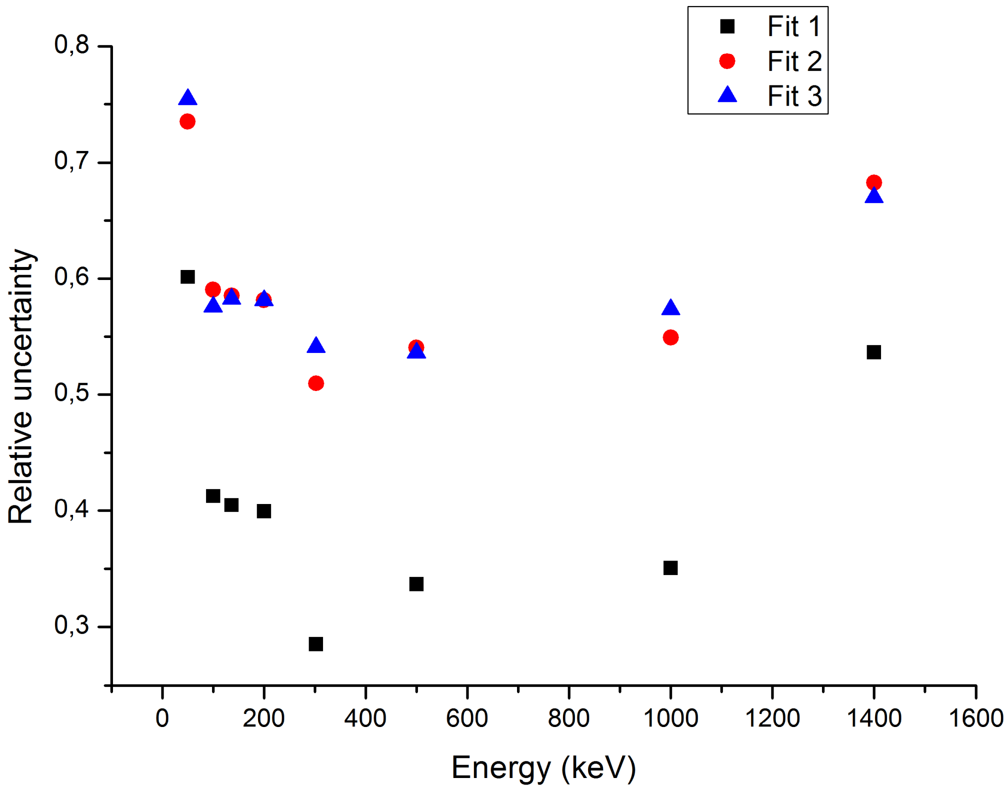
Energy	Efficiency	Relative uncertainty
50	0.004630	0.754
100	0.009822	0.576
136.54	0.009212	0.583
200	0.007227	0.581
302.8	0.004895	0.541
500	0.002847	0.536
1000	0.001453	0.573
1400	0.001061	0.670

COMPARISON OF CALCULATED VALUES FROM FITS



COMPARISON OF CALCULATED VALUES FROM FITS





Several nuclides

Weighting

Correlation

Avoid high polynomial degree

Several sections if strong inflexion

Check consistency

No extrapolation

Fitting in gamma-ray spectrometry

2 main applications:

Efficiency curves

Full-energy peaks

Approximation : peak = Gaussian

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

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

Gaussian area : $S[-\infty, +\infty] = (2\pi)^{1/2}\,\sigma A$

Sum of channels in the region $[-3\sigma, +3\sigma]$ around the peak centroïd = 99.7 % of the Gaussian total area (S)

Peak fitting

Peak with low-energy tailing : G(E) + T(E)

Tail = exponential background ⊗ Gaussian shape (detector widening)

$$T(E) = \int_{-\infty}^{E_0} A \cdot T \cdot \exp(\tau \cdot E) \cdot \exp\left[\frac{-(E - E_0)^2}{2\sigma^2}\right] \cdot dE$$

$$T(E) = A \cdot \frac{T}{2} \cdot \exp\left[(E - E_0)\tau + \frac{\sigma^2 \tau^2}{2}\right] \cdot \mathrm{erfc}\left[\frac{1}{\sqrt{2}} \cdot \left(\frac{(E - E_0)}{\sigma} + \sigma\tau\right)\right]$$

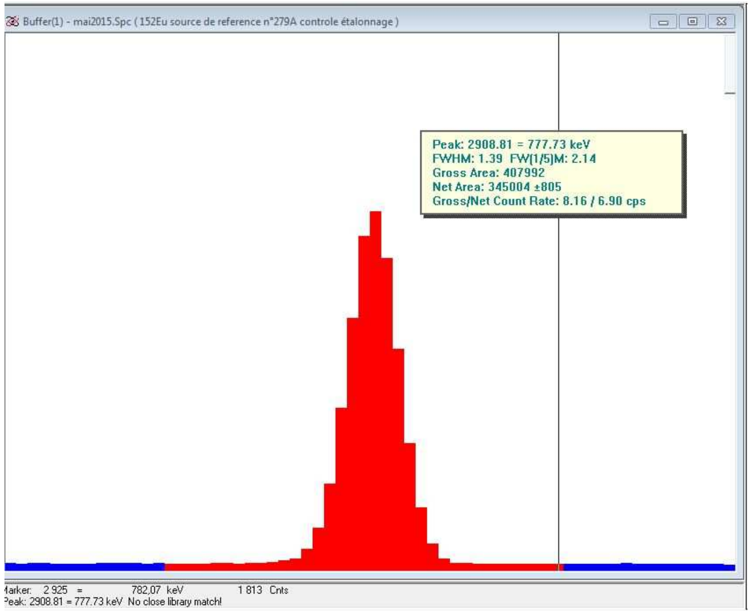
$$\mathrm{erfc}(x) = \frac{2}{\sqrt{\pi}} \int_x^{\infty} e^{-t^2}\, dt$$

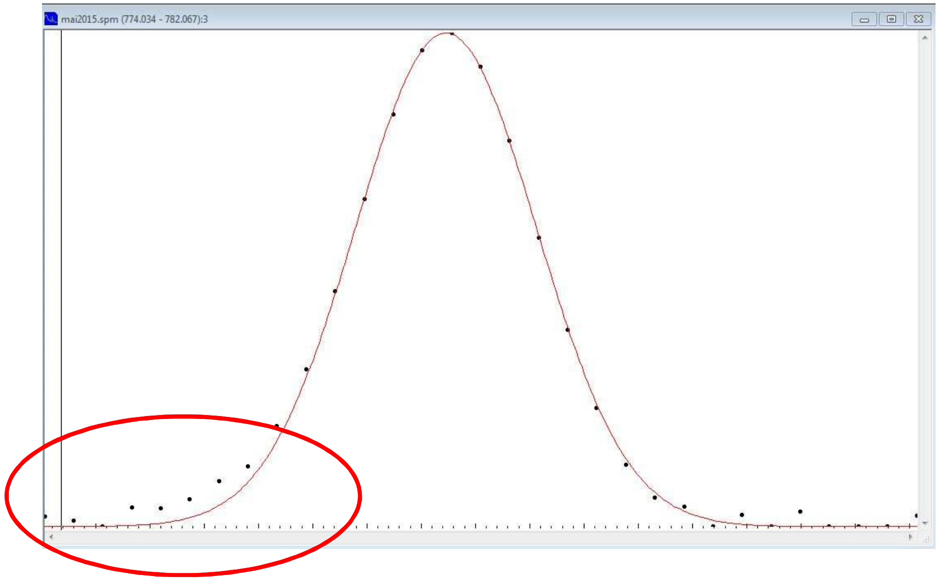
DIFFERENCE SUMMING/FITTING

MAESTRO: sum – background : 345 004 (805)

Fitting of Gaussian : 342 553 (819)

Fitting of Gaussian with left tail : 344 223 (821)



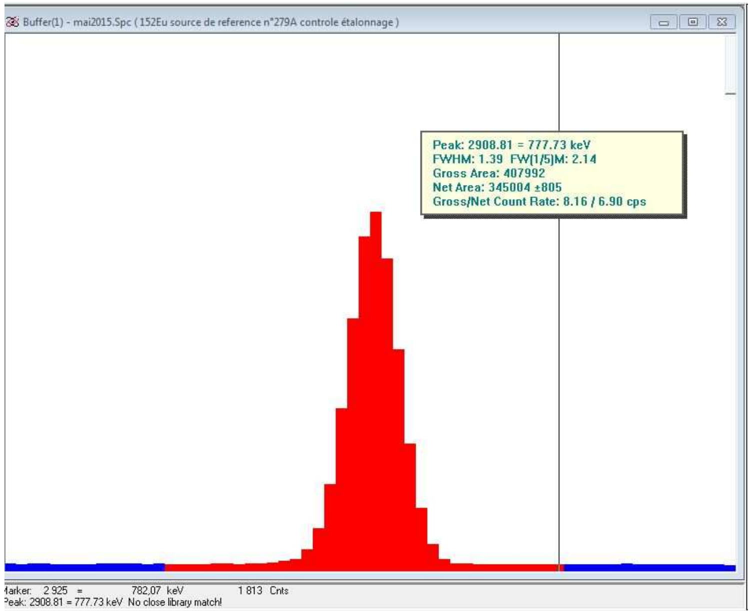


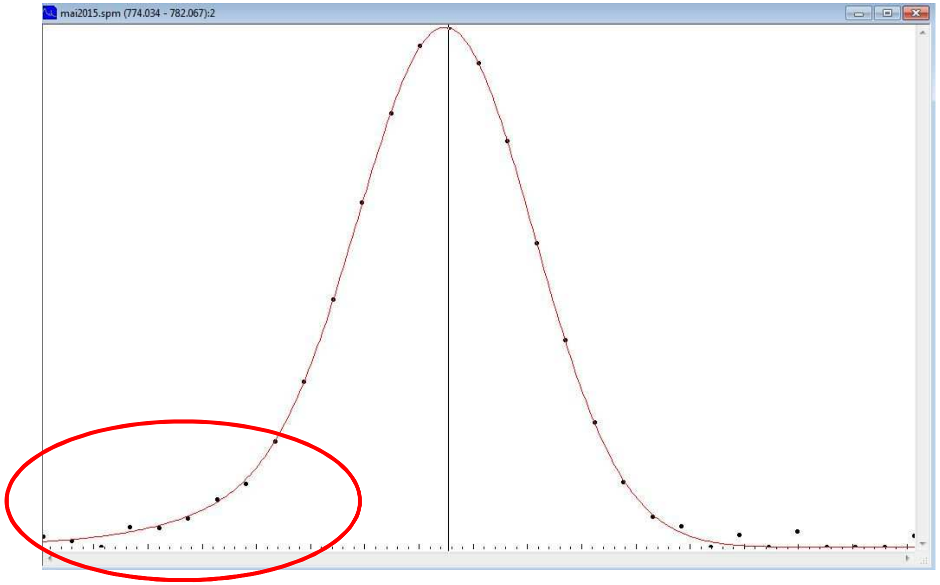
DIFFERENCE SUMMING/FITTING

MAESTRO: sum – background : 345 004 (805)

Fitting of Gaussian : 342 553 (819)

Fitting of Gaussian with left tail : 344 223 (821)



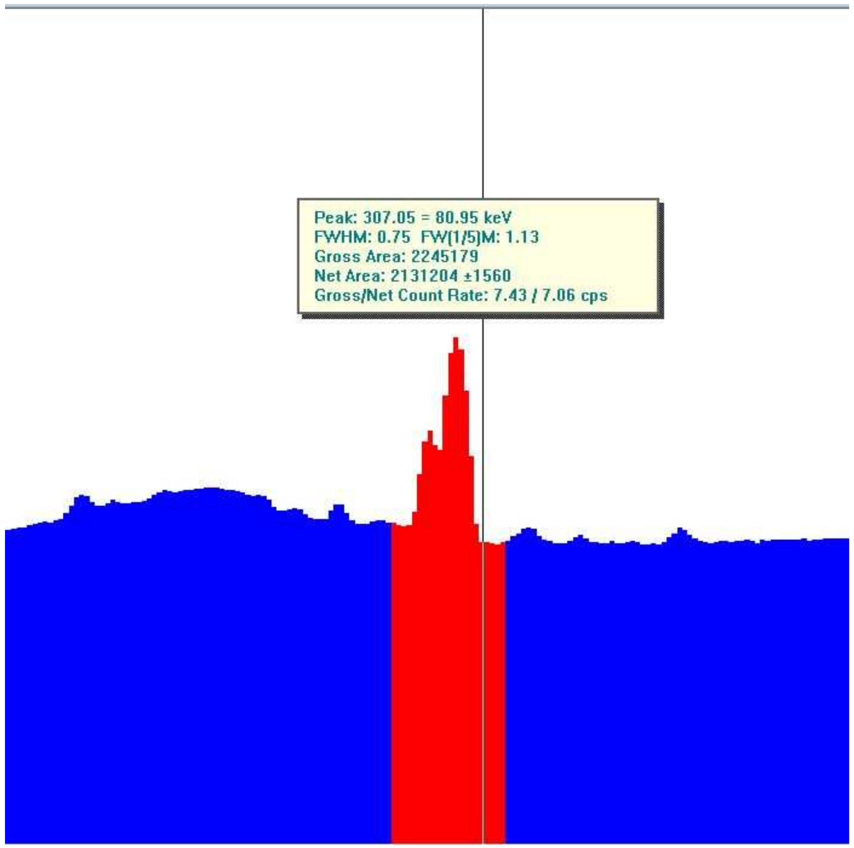


^{133}Ba : Doublet 79.61 -81.00 keV

MAESTRO: sum – background : 2 131 204(1 560)

Fitting of 2 Gaussian functions:

- 79.61 keV: 148 905
- 81.00 keV : 1 975 540
- Total : 2 124 445



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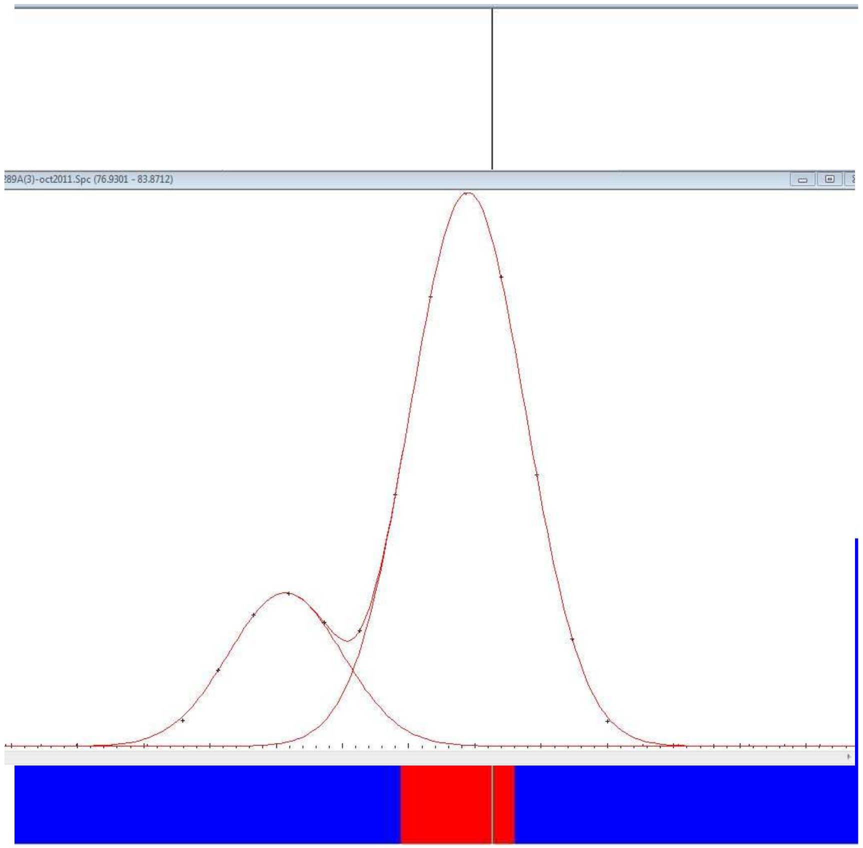


- Ratio of peak areas: 13.27
- Ratio of emission intensities: 12.67

79.61 keV : 2.63(19) -> 2.51 ?
81.00 keV : 33.31 (30)

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

Case of 511 keV region :

Example : ^{85}Sr

(Gamma at 514 keV)

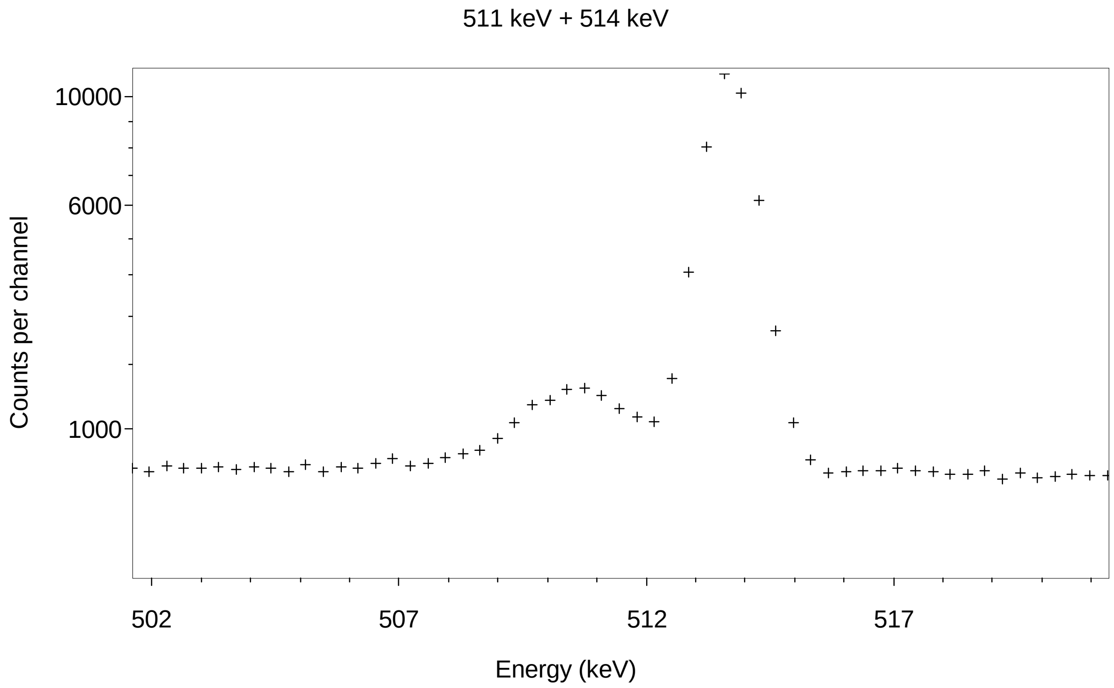
Net area 48500

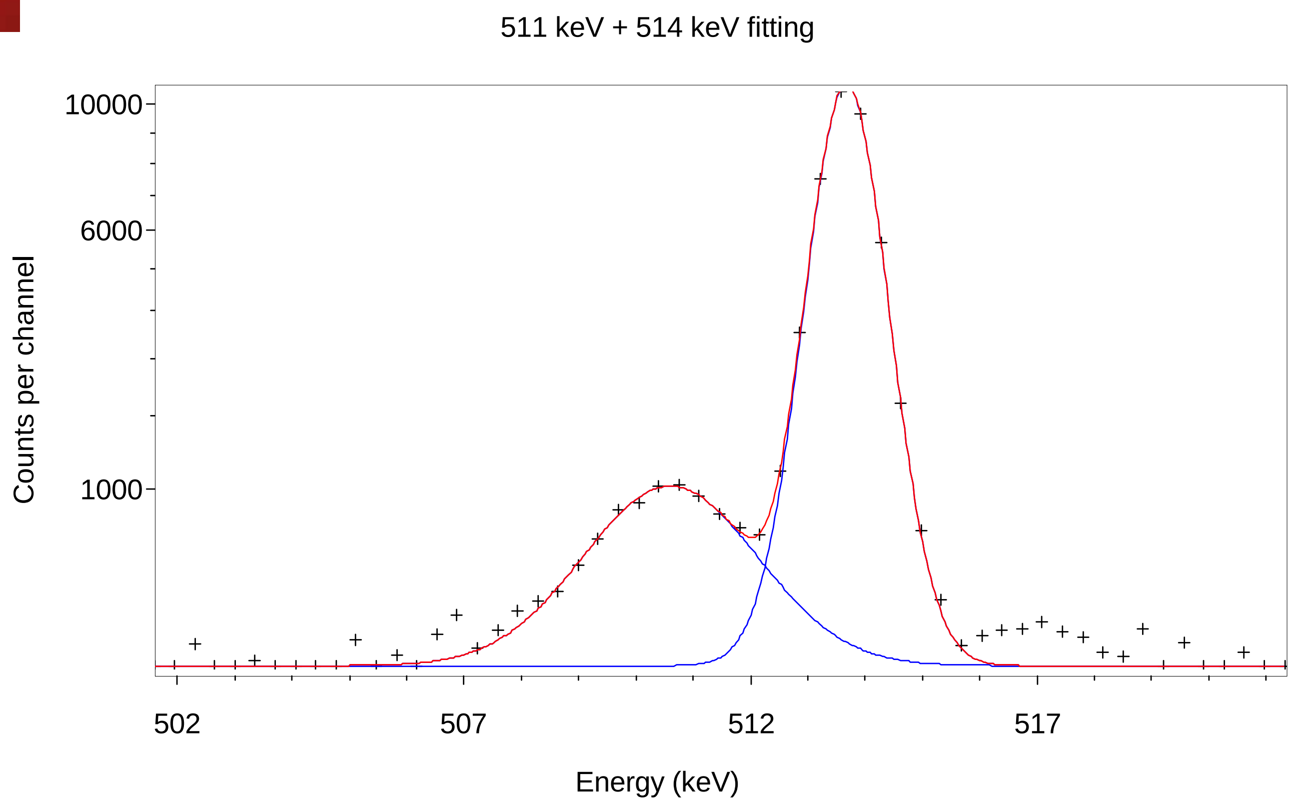
Amplitude 511 = 1000

Amplitude 514 = 10 000

Area of 511 = 4400

Area of 514 = 44 100



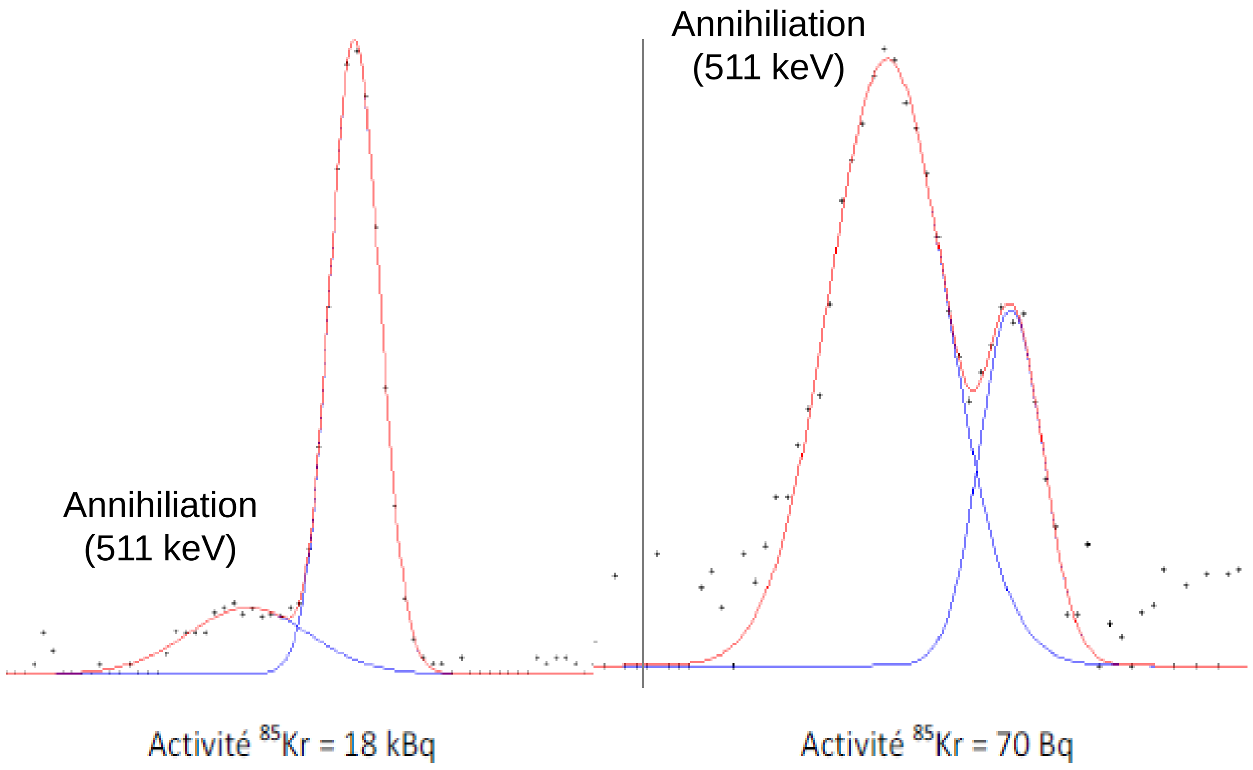


Fitting : 511 = 7730 514 = 40400

σ = 1.07 σ = 0.53

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

Exemple: ^{85}Kr 511-514 (or ^{85}Sr)



Low activity : requires to fix the Gaussian width (resolution calibration)

Photons = transitions between excited levels

Gamma = nuclear levels

X = atomic levels

For monoenergetic emission, there is a finite line shape

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

Transition width (Γ) = initial state energy width + final state energy width $\Gamma = \Delta E_i + \Delta E_f$

Energy levels = uncertainty (ΔE)

Heisenberg uncertainty principle : $\Delta E \, . \, \Delta t \geq h / 2\,\pi$

h (Planck constant) = 6.626 070 15 10^{-16} J.s (BIPM)

Δt uncertainty of the level half-life = $1/\lambda$ = 1.4427 $t_{1/2}$

($\lambda = \ln 2 / t_{1/2}$)

Examples : gamma lines

- $^{137}Cs \rightarrow {}^{137}Ba^{m}$: excited level = 661.7 keV
- level half-life = 2.552 min $\rightarrow$ $\Delta E \approx 3\ 10^{-18}$ eV

- ^{60}Co at 1 332,5 keV
- level half-life = 0.713 ps $\rightarrow$ $\Delta E \approx 6.5\ 10^{-4}$ eV

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

X-ray lines : some eV

Detector widening (Gaussian) : some hundreds of eV

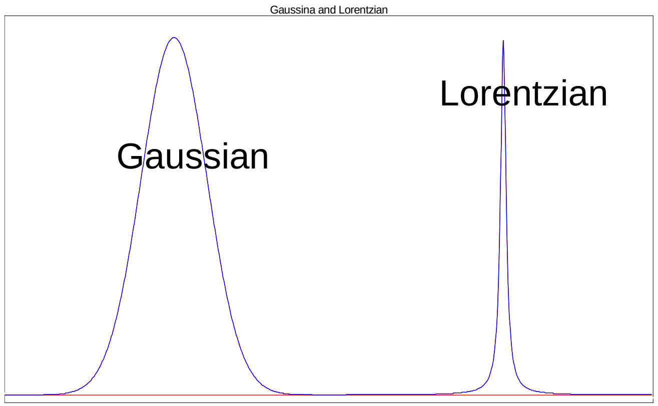
Element	Z	Energy (eV)	Level width (eV) K	L 3	$K\alpha 1$ linewidth Γ(eV)	σ (eV)	Γ / σ
Nickel	28	7,45	1,44	0,48	1,94	155	0,030
Cadmium	48	22,98	7,28	2,50	9,8	200	0,115
Lead	82	72,80	60,4	5,8	66,2	350	0,445
Uranium	92	94,65	96,1	7,4	103,5	415	0,587

Peak = *Lorentzian ⊗ Gaussian (Voigt profile)*

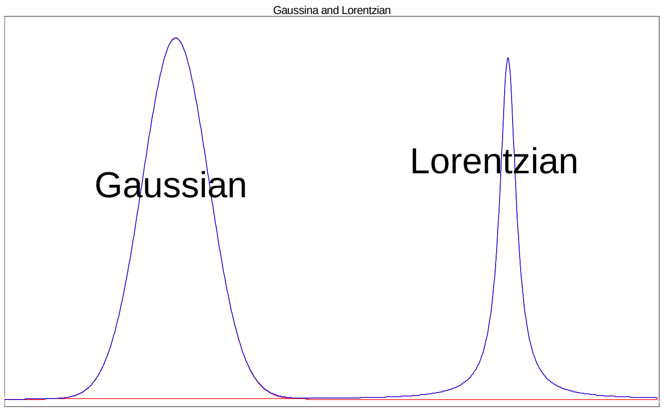
For high Z X-ray lines, the natural linewidth is not negligible versus the detector resolution

NATURAL LINEWIDTH

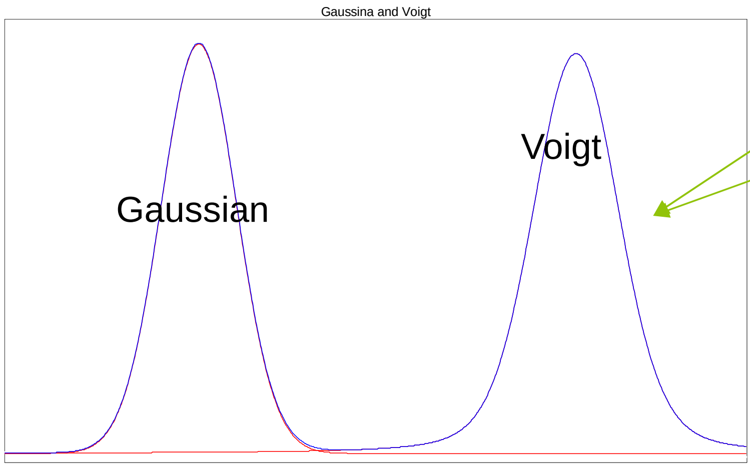
Peak processing



σ = 50 eV – Γ = 10 eV



σ = 50 eV – Γ = 30 eV



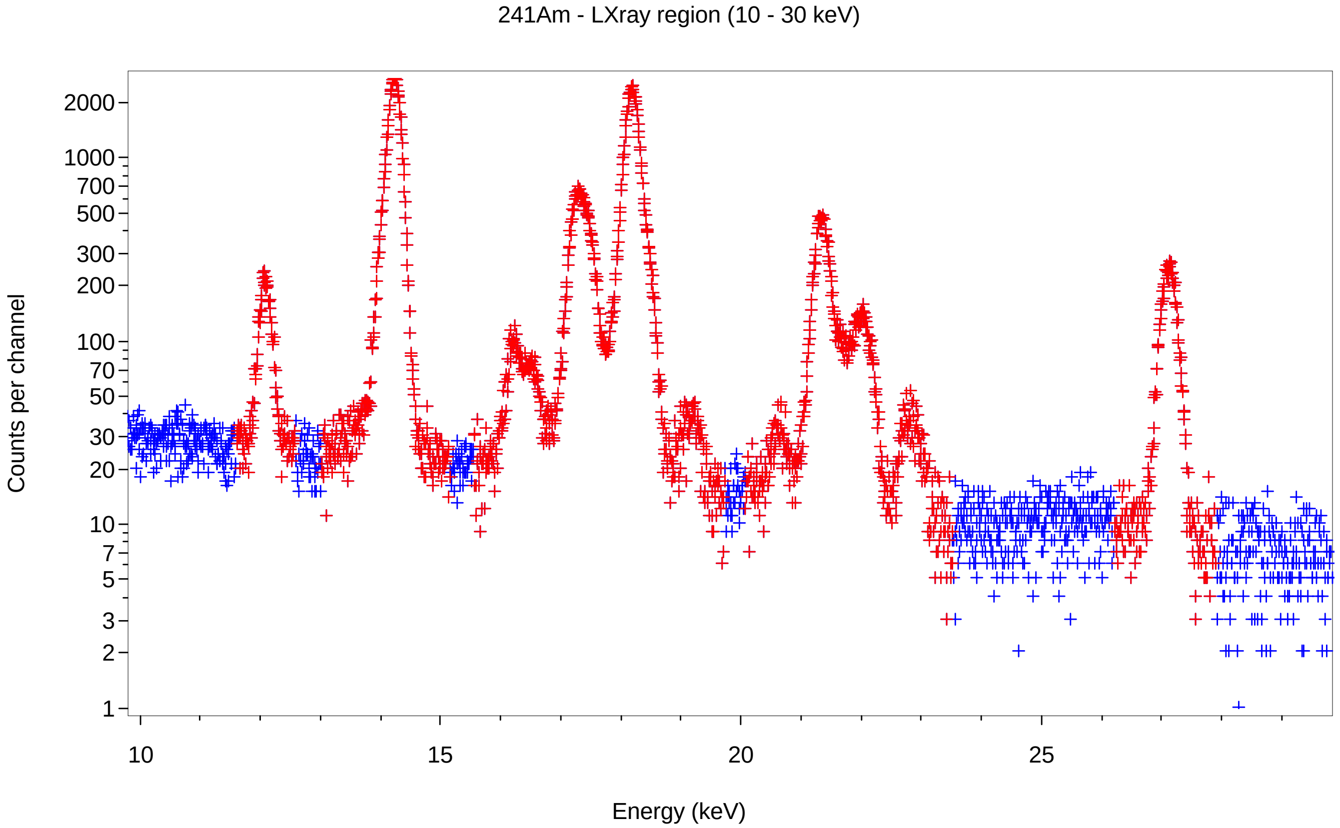
Peak = <u>*Lorentzian $\otimes$ Gaussian*</u>
<u>*(Voigt profile)*</u>

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

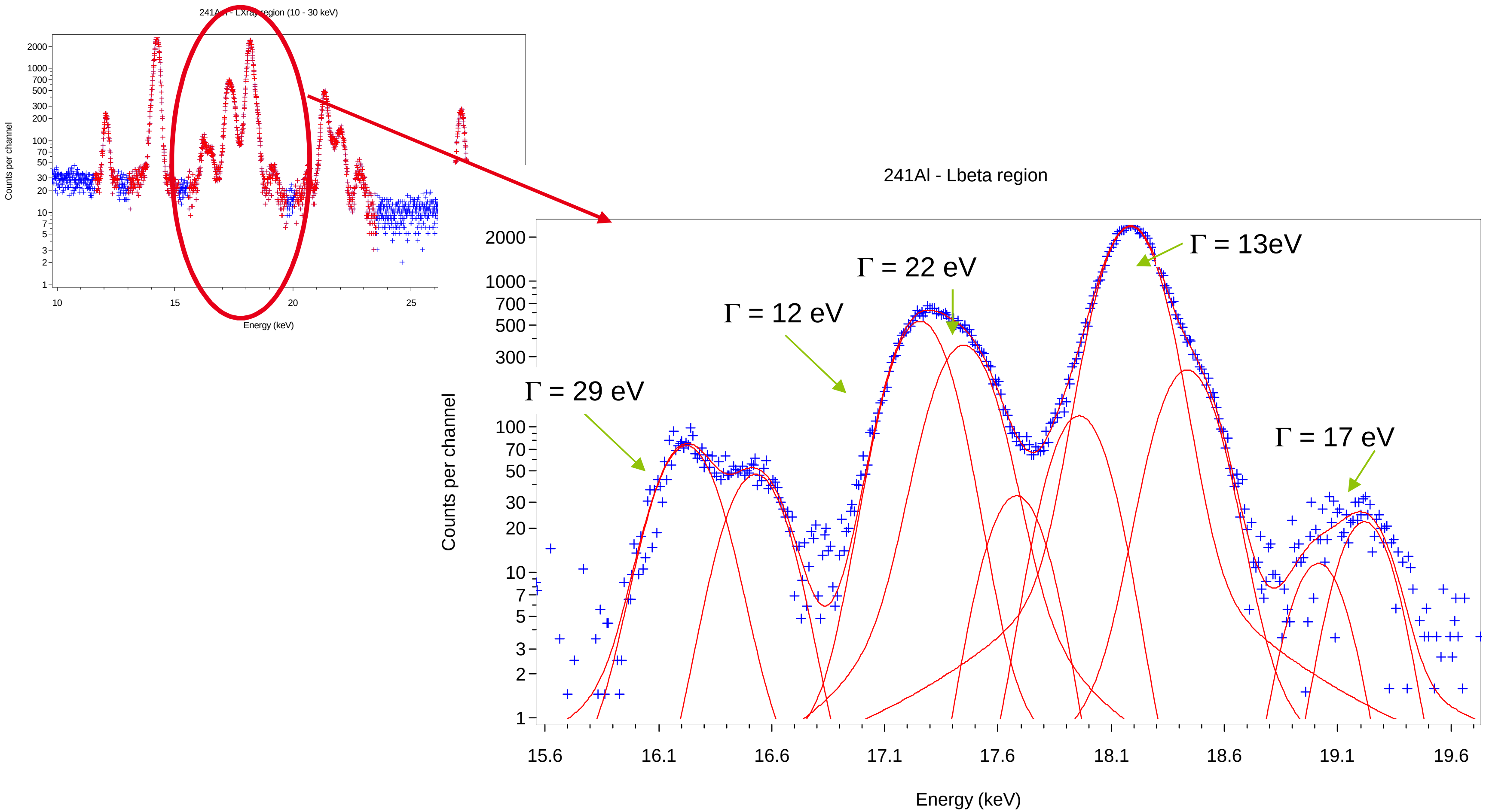
Peak fitting – X-rays

Example : ^{241}Am spectrum (10 – 30 keV region)

- Gamma at 59.54 keV (35.92%) and 26.34 (2.31%) keV highly converted
- Intense Np L X-rays in the 12-20 keV (37.66 %)



Processing of the Np L beta region



Peak fitting – Tailing

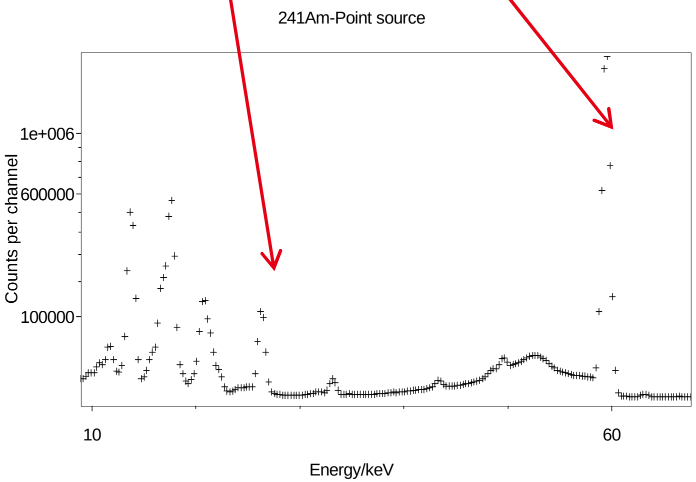
Detector + electronics effect (tailing can be included in the peak shape)

Scattering in the low-energy range : Is it part of the FEP ?

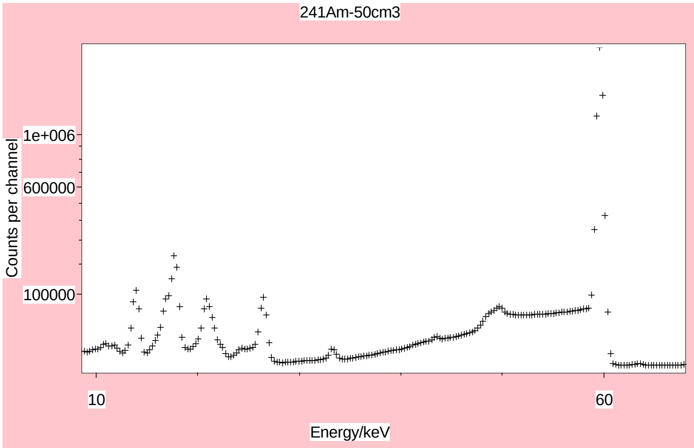
Peak fitting – Tailing

Experimental spectra obtained with calibration sources:

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



Point source at 10 cm



Volume source (50 cm^3) at 10 cm

Difference in scattering effect depending on the geometry

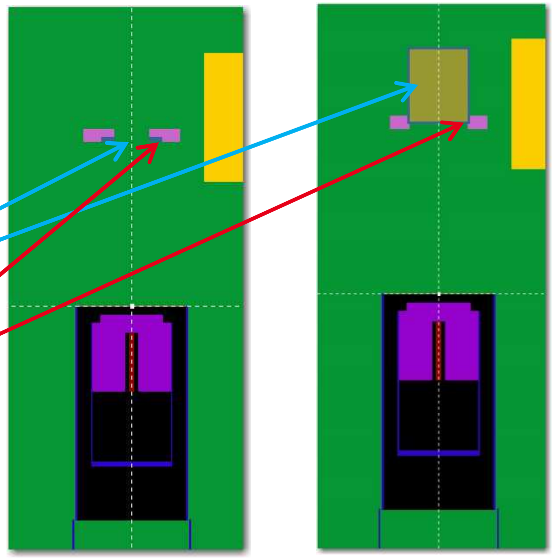
-> Monte Carlo simulation to identify main scattering sites

PENELOPE (Univ. Barcelona) : Monte Carlo simulation code
PENetration and Energy LOss of Positrons and Electrons

Result: Histogram of energy deposition in selected bodies (spectrum)

Modification of the code to follow each kind of interaction (ICOL)

If ICOL = 2 -> +1 in relevant body counter

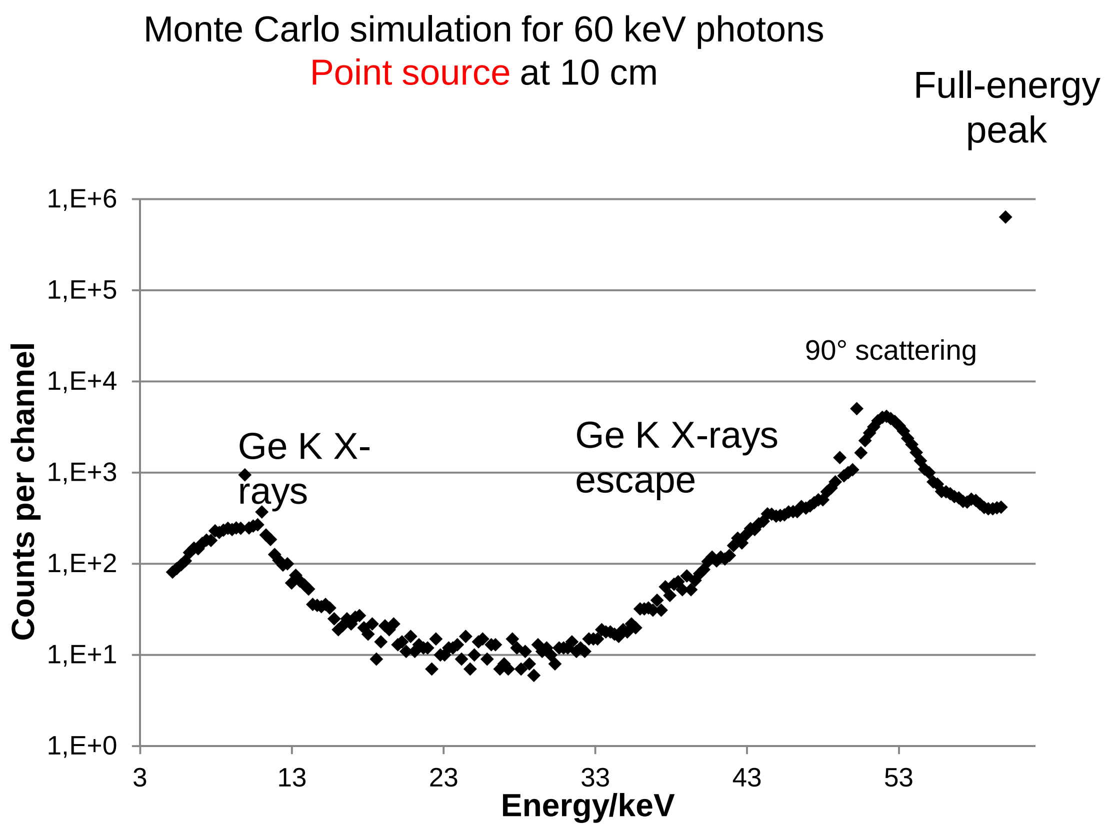


Body 18 : source material (or Mylar® film)

Body 19 : source container (or plastic ring)

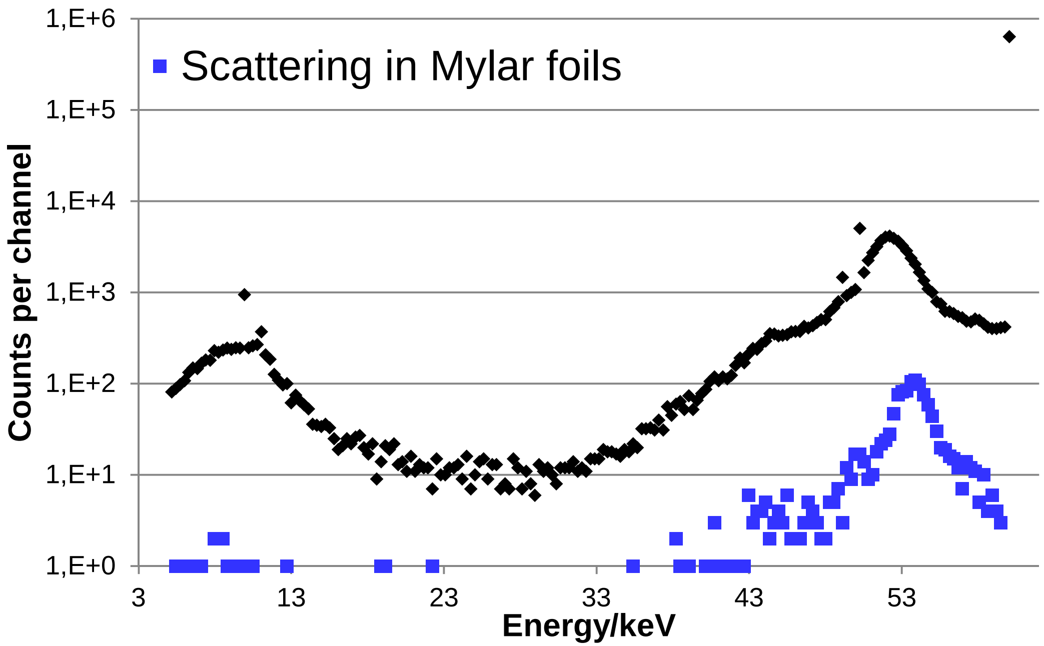
(Scattering angles around $\pi/2$)

Monte Carlo simulation



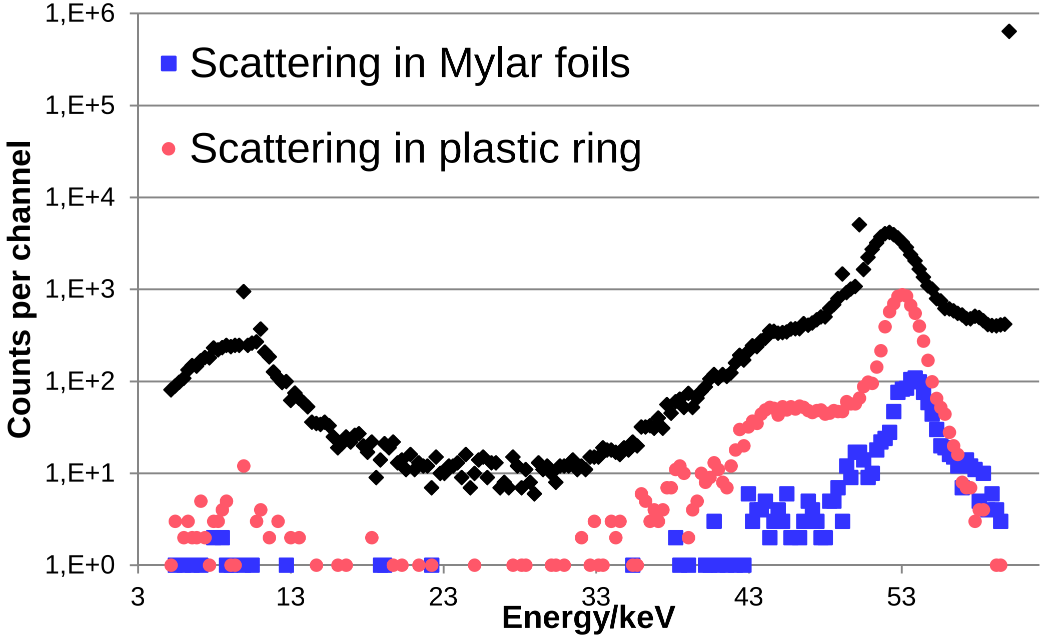
Monte Carlo simulation

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



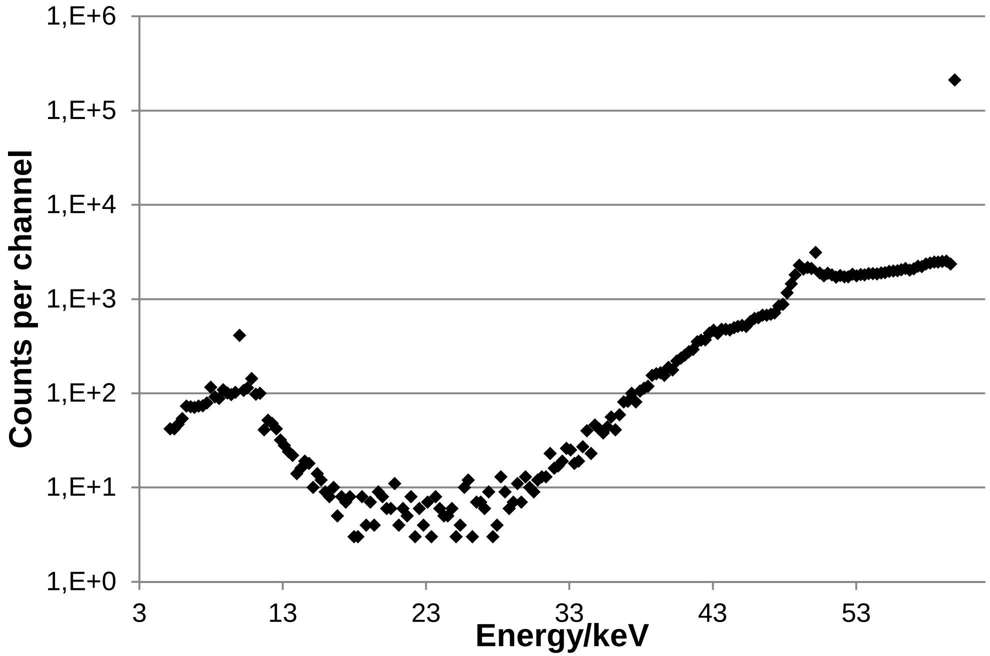
Monte Carlo simulation

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

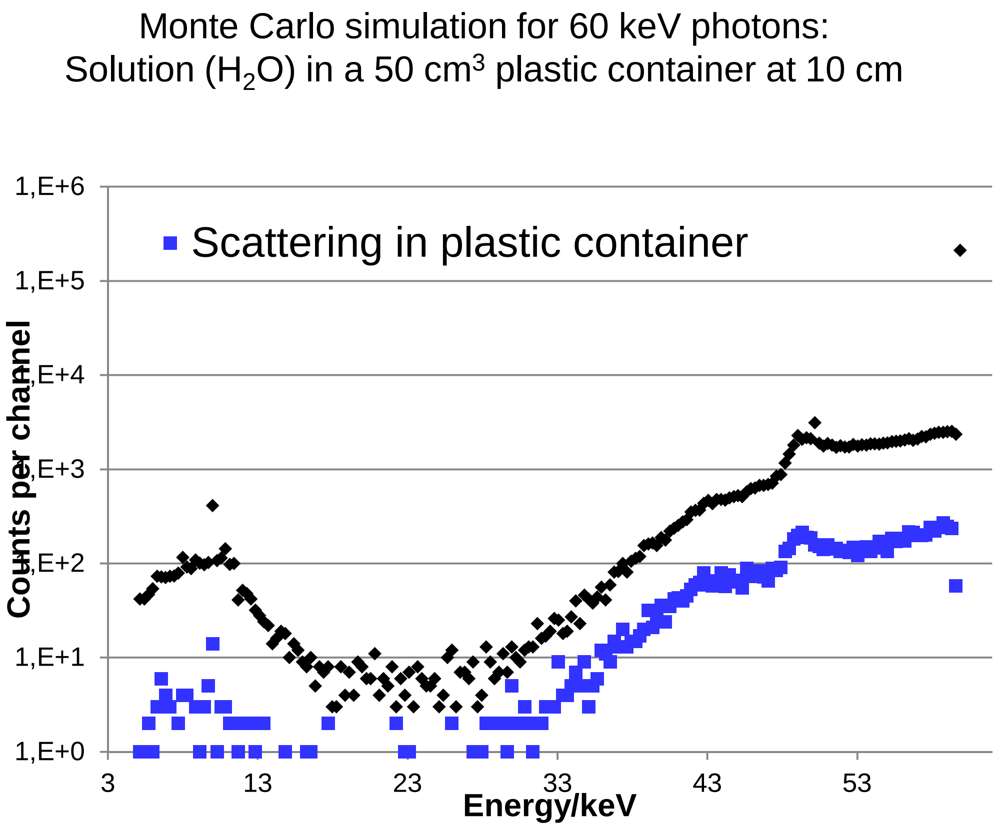


Volume effect

Monte Carlo simulation for 60 keV photons:
Solution (H_2O) in a 50 cm^3 plastic container at 10 cm



Volume effect



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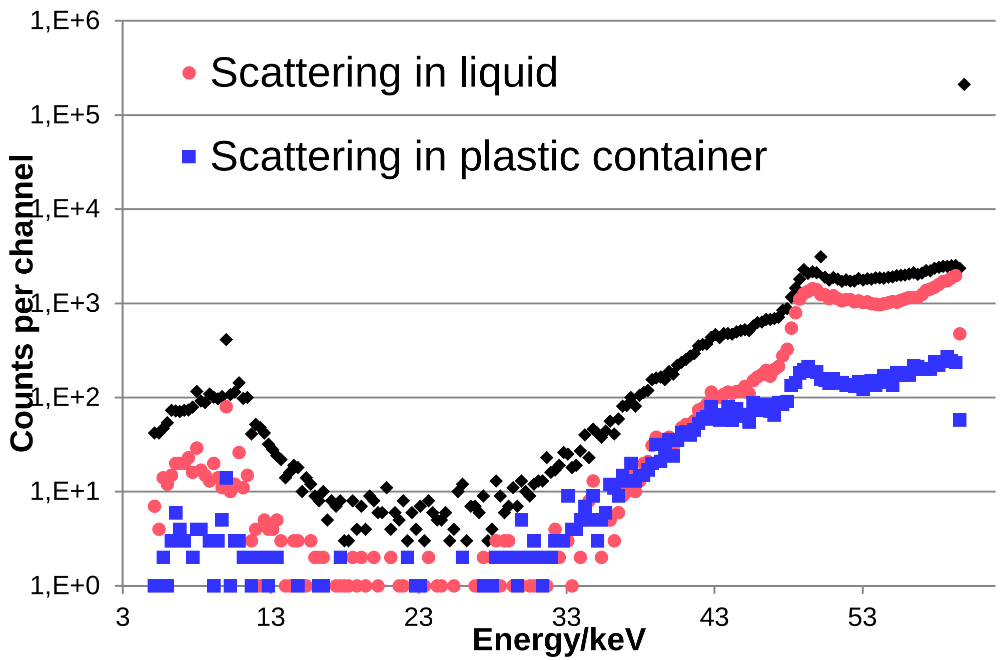
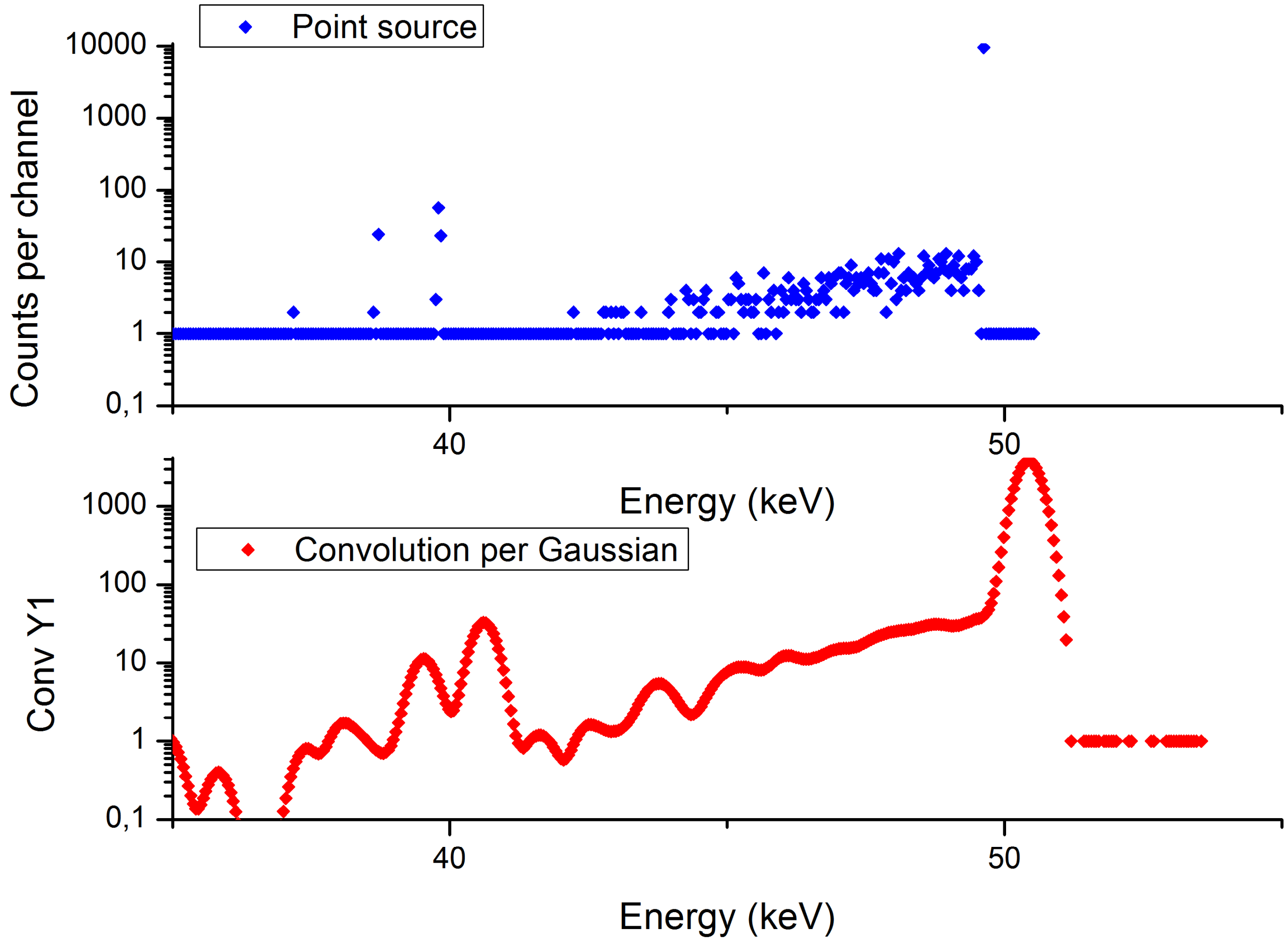
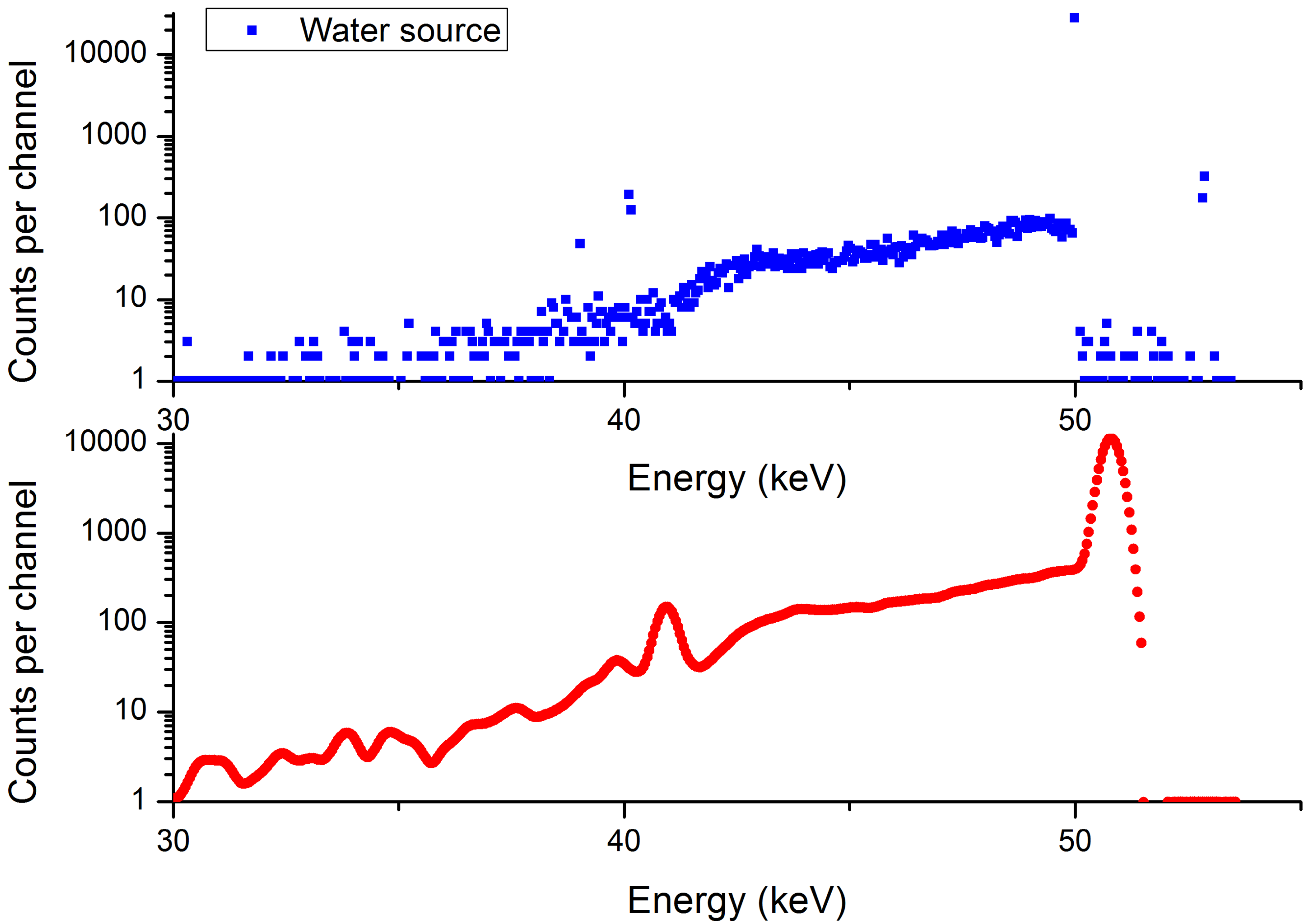


Peak fitting – Tailing

Detector response widening

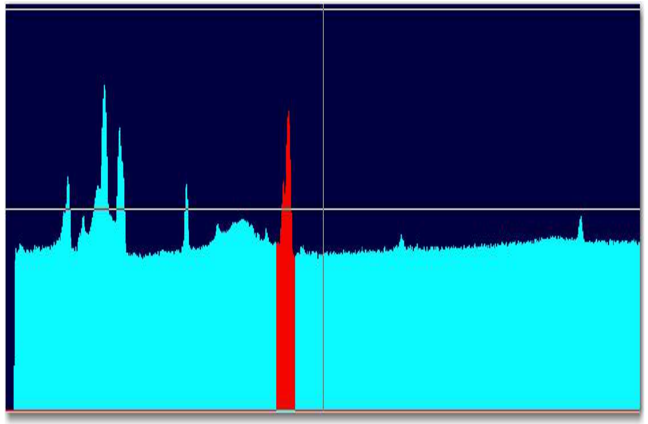


Detector response widening – Volume source

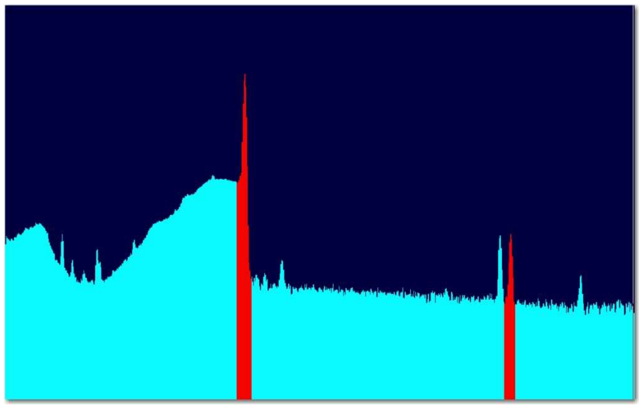


- Activity measurement of ^{133}Xe derived from:

Calibration with ^{133}Ba point source



- Efficiency transfer ?

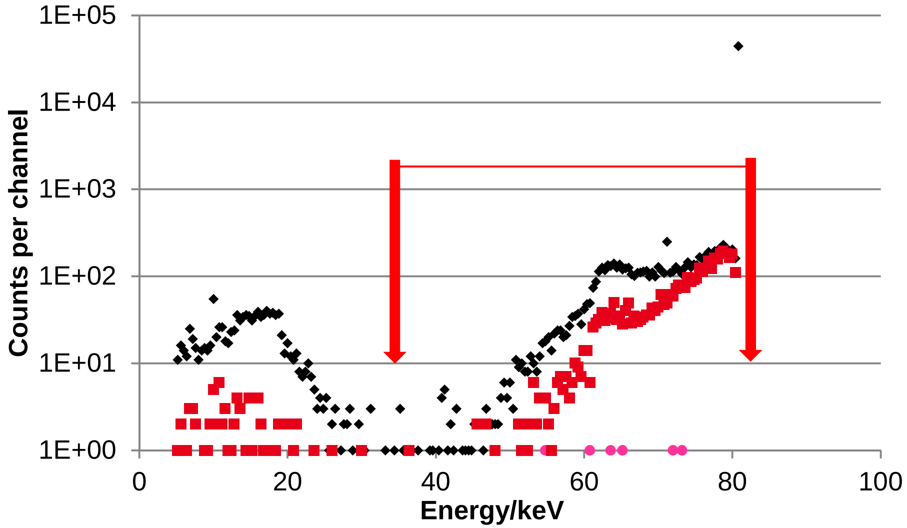


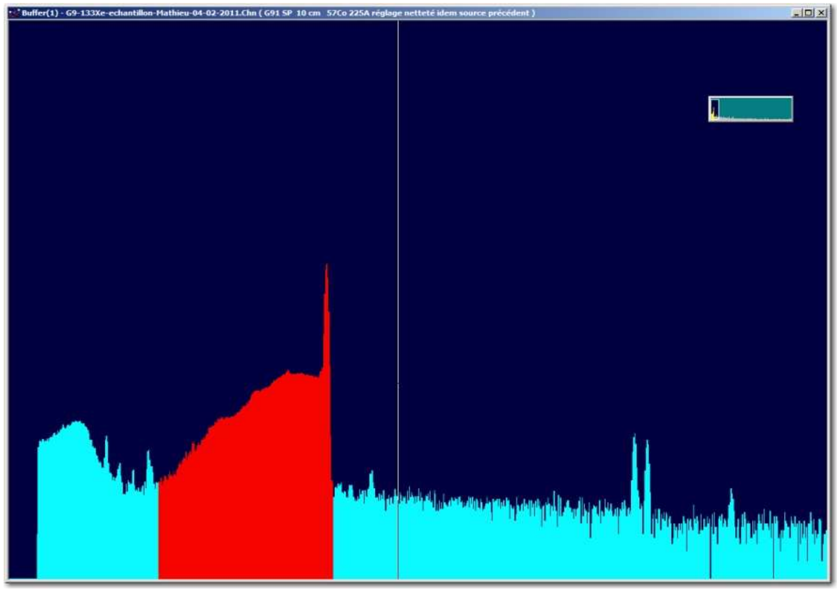
Peak fitting – Tailing

Suggestions on tailing due to scattering:

- Fit only the Gaussian part (more weight)

- For efficiency transfer : use MC results (FEP only or including scattering)





Your opinion ?

Thank you for your attention

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