Coincidence-summing based detector calibration

The magic of absolute efficiencies without certified sources

Nuclear data for Activation Analysis

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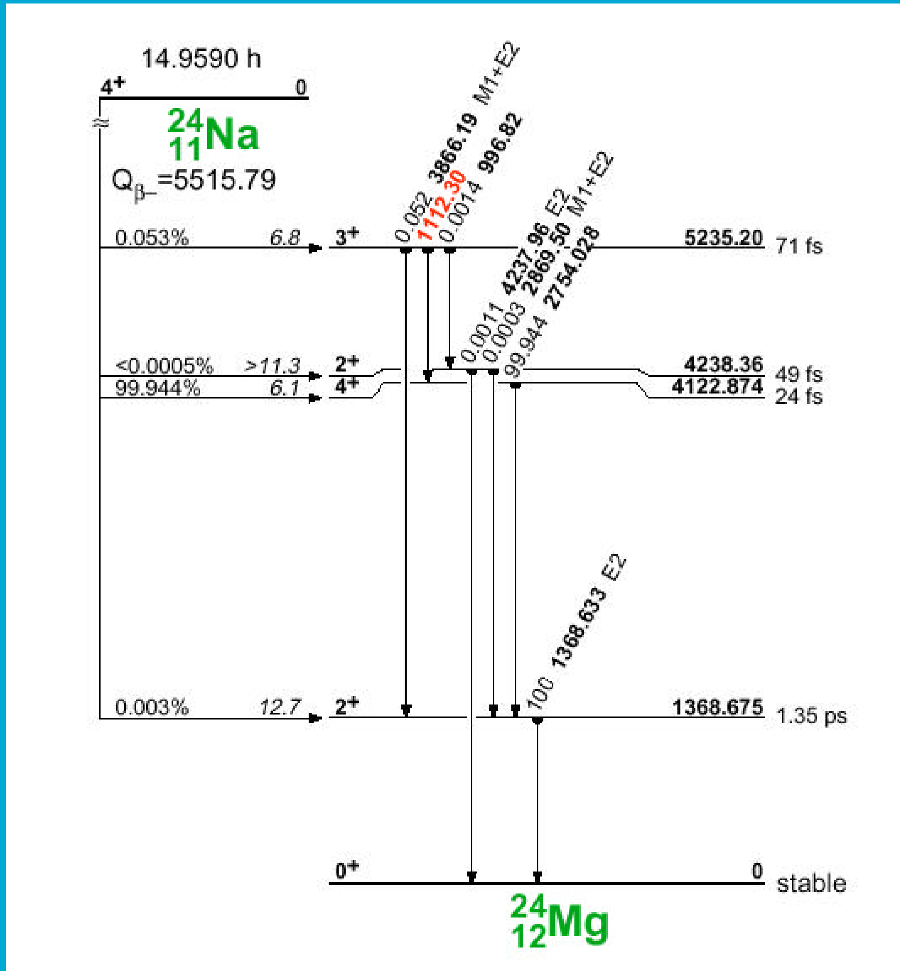
Overview

- Basics of coincidence summing
- How to determine efficiency curves in the presence of coincidence summing
- How to determine a source activity from scratch
- Applicability with Ge detectors
- Two advanced topics / applied examples
  - Do absolute peak areas exist?
  - The voluminous effect and the third curve

A simple decay scheme



Coincidence equations

$$A_{1368} = N\varepsilon_{1368}\left(1-\varepsilon_{t,2754}\right)$$

$$A_{2754} = N\varepsilon_{2754}\left(1-\varepsilon_{t,1368}\right)$$

$$A_{1368+2754} = N\varepsilon_{1368}\varepsilon_{2754}$$

the general equation

$$P_E = \sum_{itineraries\ where\ E=\sum_{i=1}^{M} E_i} \left\{\prod_{i=1}^{M}\varepsilon_i \prod_{j=1}^{N-M}(1-\varepsilon_{tj})\right\}$$

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How to calibrate in the presence of coincidence summing - method 1

- Measure the peak-to-total ratio curve separately.
- Count a calibrated, multi-gamma-ray source.
- Disregard the sum peaks.
- Each “normal” peak corresponds to a full-energy peak efficiency, so
- the number of questions is equal to the number of unknowns, so
- the full-energy peak efficiencies can be solved.

D.S. Andreev, K.I. Erokhina, V.S. Zvonov, I.Kh.Lemberg, Bull. Acad. Sci. USSR Phys.Ser. 37 (1973) 41-43
M. De Bruin, P.J.M. Korthoven, Radiochem.Radioanal. Letters 19 (1974) 153-156
T.M. Semkow, G. Mehmood, P.P. Parekh, M. Virgil, Nucl.Instr.Meth. in Phys.Res. A290 (1990) 437-444

How to calibrate in the presence of coincidence summing - method 2 - k_0-IAEA

- Measure the peak-to-total ratio curve separately using ^{137}Cs only[De Felice].
- Count a calibrated, multi-gamma-ray (> 4) source.
- Use a 6-th order polynomial with 4 degrees of freedom for the full-energy efficiency curve[Gunnink],
- the number of questions is equal or larger than the number of unknowns, so
- the full-energy peak efficiency curve can be fitted to the observed spectrum.

P. De Felice, P. Angelini, A. Fazio, R. Biagini, Appl. Radiat.Isot. 52 (2000) 745-752
Gunnink, R., Nucl. Instrum. Meth. Phys. Res. , Sect. A ,299, pp. 372-376, 1990.

How to determine a source activity from scratch

- We have *N* atoms decaying during our measurement, each emitting two photons simultaneously, with equal 100 % yields and energies E_1 and E_2
- We have a detector with energy resolution that detects these with efficiencies ε_1 and ε_2.
- We will then see three peaks in the spectrum, corresponding to E_1, E_2 and E_1+E_2, with net areas A_1, A_2, and A_{1+2}
- We can write and derive:

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How to determine a source activity from scratch

$$A_1 = N\varepsilon_1$$

$$A_2 = N\varepsilon_2$$

$$A_{1+2} = N\varepsilon_1\varepsilon_2$$

3 equations, 3 unknowns!

$$A_1 A_2 = N^2\varepsilon_1\varepsilon_2$$

$$\frac{A_1 A_2}{A_{1+2}} = \frac{N^2\varepsilon_1\varepsilon_2}{N\varepsilon_1\varepsilon_2} = N$$

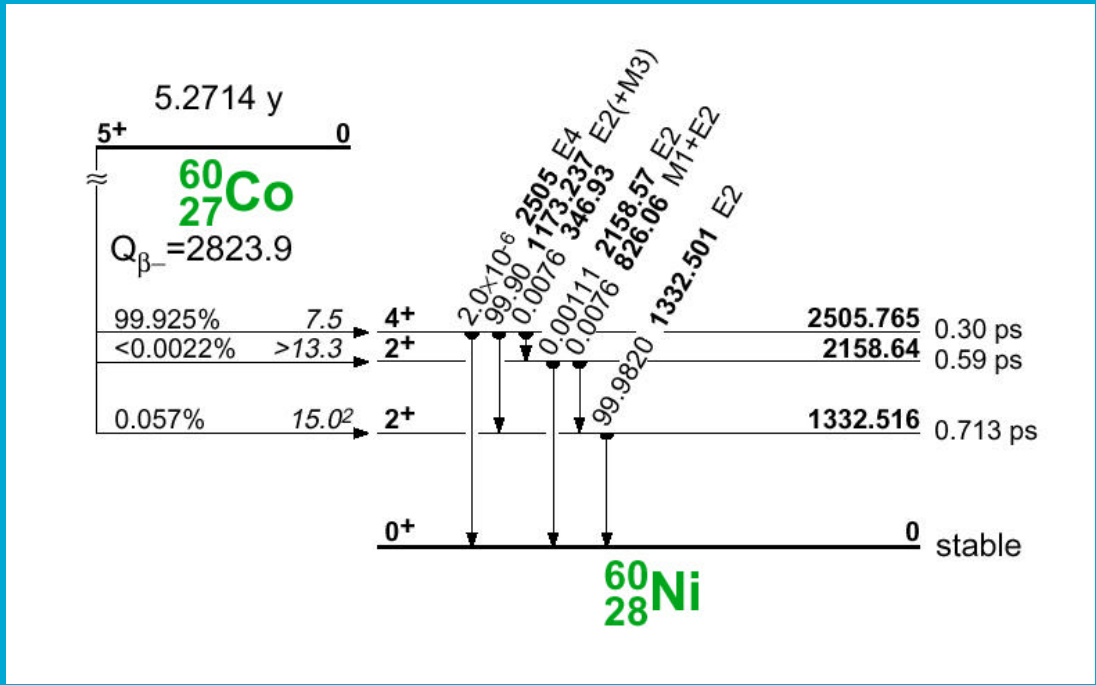
How to determine a source activity from scratch

$$N = \frac{A_1 A_2}{A_{1+2}} \text{ and } \varepsilon_2 = \frac{A_{1+2}}{A_1} \text{ and } \varepsilon_1 = \frac{A_{1+2}}{A_2}$$

So, knowing only the decay scheme of the radionuclide, we can get the source activity and the detection efficiencies from a single gamma-ray spectrum. A method traceable to physical principles!

G.A.Brinkman, A.H.W. Aten, J.Th.Veenboer, Appl.Radiat.Isot. 14 (1963) 153-157

Do such radionuclides exist?



Almost... but with cross-overs and angular correlations.

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Do such detectors exist for gamma-rays?

No, definitely not for higher energies!

Disappointment...

$$A_1 = N\varepsilon_1\left(1-\varepsilon_{t2}\right)$$
$$A_2 = N\varepsilon_2\left(1-\varepsilon_{t1}\right)$$
$$A_{1+2} = N\varepsilon_1\varepsilon_2$$

5 unknowns is too many

$$A_1 A_2 = N^2\varepsilon_1\varepsilon_2$$

$$\frac{A_1 A_2}{A_{1+2}} = \frac{N^2\varepsilon_1\varepsilon_2}{N\varepsilon_1\varepsilon_2} = N$$

How to determine source activity and both efficiency curves from scratch

- The number of equations is the number of single-photon peaks *T* plus the number of sum peaks, that is

$$\binom{T}{1}+\binom{T}{2}+\ldots\binom{T}{N}$$

- If all efficiencies and source activity are unknown, the number of unknowns is

$$1+2T$$

- So at T >= 3, the number of equations >= number of unknowns

More disappointment and solutions

- Triple sum peaks are very hard to detect indeed, not to mention higher-order ones...
- So we take a closer look at the unknowns:
  - We can parametrize the efficiency curve with Gunnink’s polynomial (4 unknowns)
  - We parametrize the p/t curve with a 2nd order polynomial on a log-log scale
  - With the source activity, that is 7 unknowns total
- At T=4, using only simple sum peaks, we’re there with 4+6 peaks
- So do we have candidate radionuclides?

Requirements for candidate nuclides

- Energies covering the whole energy-range of interest
- Many transitions without cross-overs
- Medium-complexity decay scheme (for computational speed)
- Medium half-life (not too much waste, not too much haste, reasonable count rates to avoid random summing)

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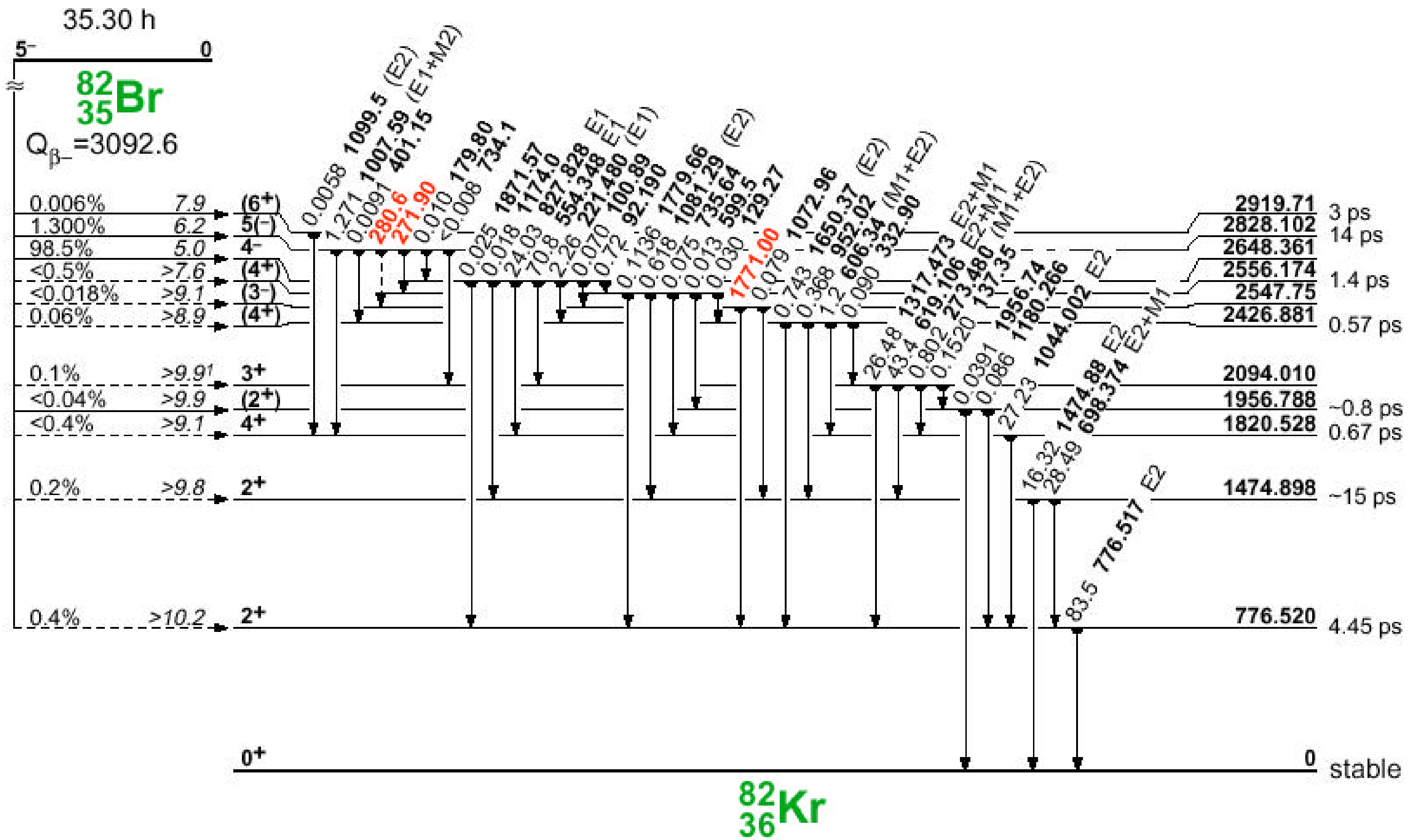
Suitable nuclide #1: ^{82}Br



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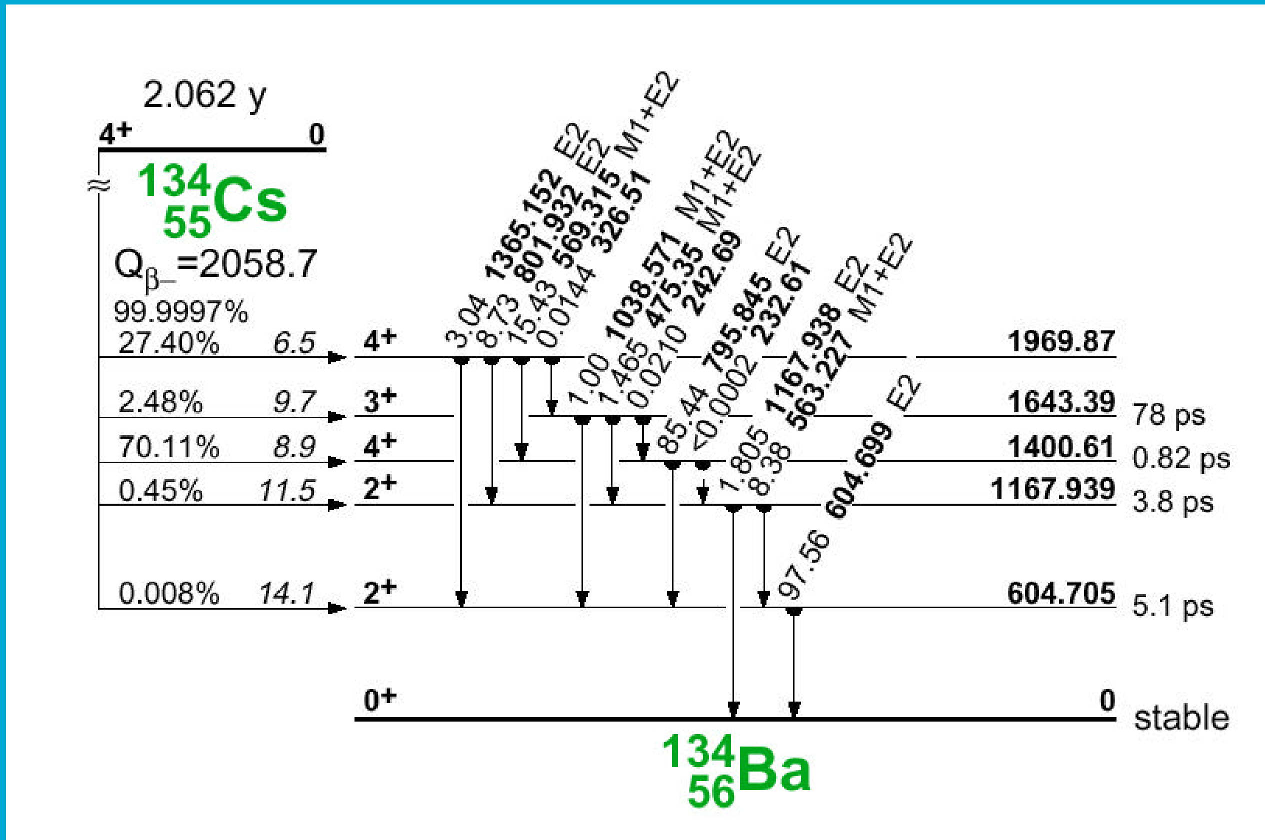
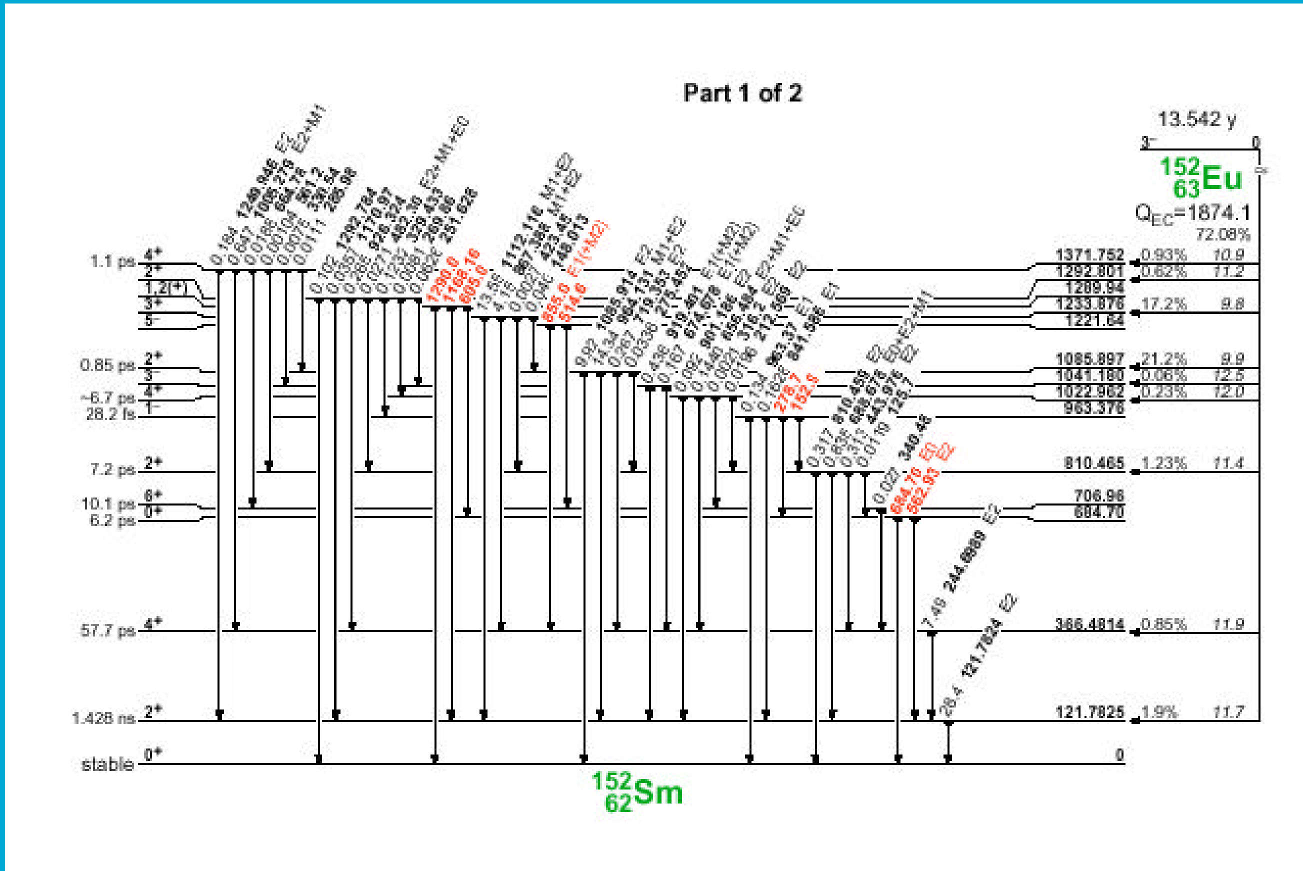
Suitable nuclide #2: ^{134}Cs



Only in conjunction with other nuclides to establish efficiencies at more energies

Suitable nuclide #3: ^{152}Eu



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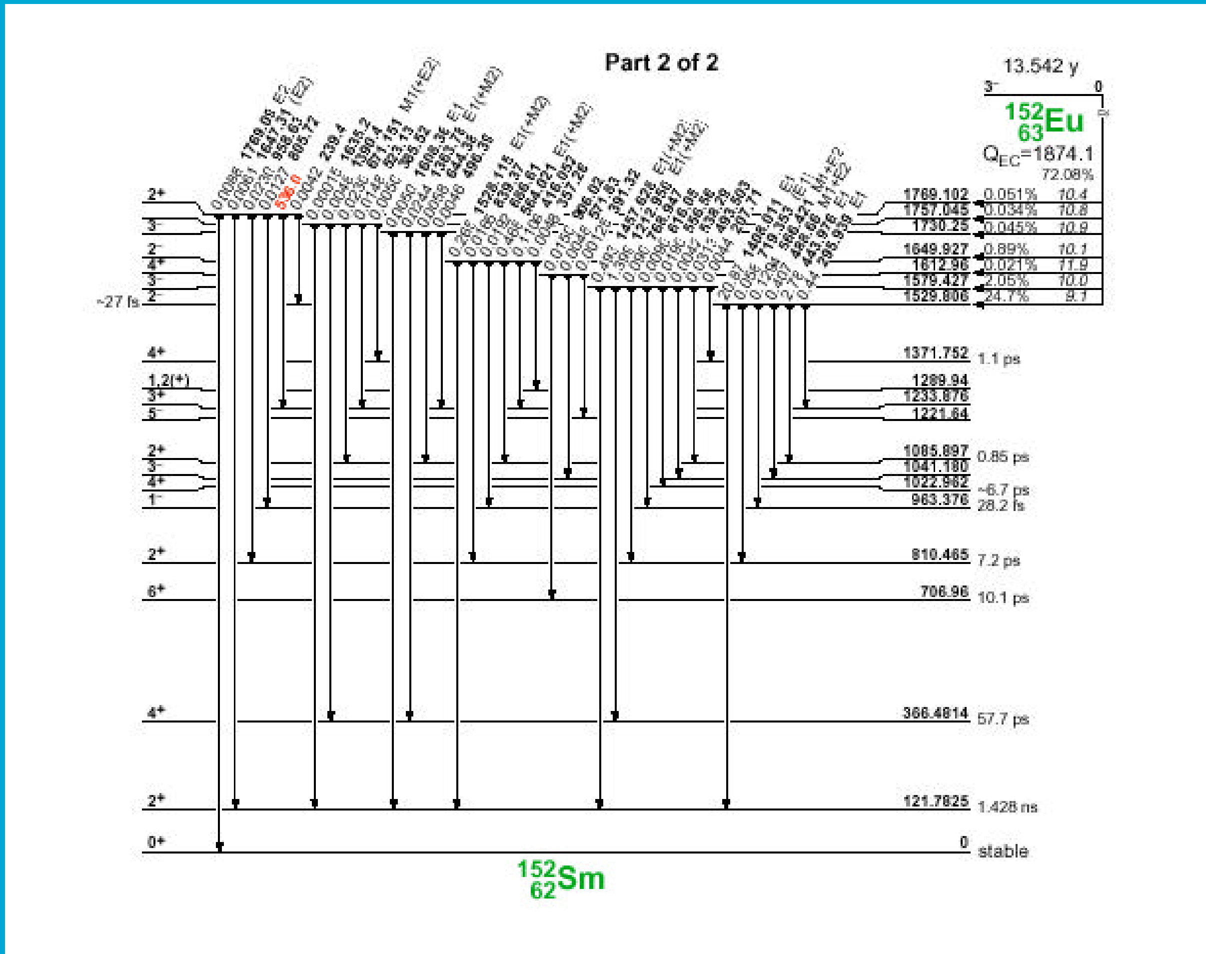
Suitable nuclide #3: ^{152}Eu



^{152}Eu pros and cons

- Hard to get in pure form, that is without ^{154}Eu.
- Complex decay scheme, but modern computers are fast
- Very nice coverage of low energies, from the X-rays around 40 keV up to 1408 keV. Higher energies missing.

The idea in a nutshell

- If we have a source containing suitable radionuclides, we can determine the source activity, full-energy peak efficiency curve and peak-to-total curve all at once.
- This was demonstrated for the first time in 1993

M.Blaauw, "The Use of Sources Emitting Coincident γ-Rays for Determination of Absolute Efficiency Curves of Highly Efficient Ge Detectors", Nucl.Instr.Meth. A332 (1993) 493-500

- The source activity is usually not determined very precisely due to strong covariances. With extreme counting statistics, better than 1 % precision in source activity is achievable.

A more stable method

- If we have a calibrated source containing suitable radionuclides, we can determine the full-energy peak efficiency curve and peak-to-total curve all at once.
- This was demonstrated for the first time in 1998

M. Blaauw, "Calibration of the Well-Type Germanium Gamma-Ray Detector Employing Two Gamma-ray Spectra", Nucl.Instr.Meth, A419 (1998) 146-153

- the strong covariances and instability disappeared,
- and since we have more equations than unknowns we can do even more...

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The Influence of Peak Area Determination Bias in Quantitative Gamma-ray Spectrometry in the Presence of True Coincidence Summing

or

There can be only one...

M. Blaauw, S.J. Gelsema

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Overview of the first advanced topic

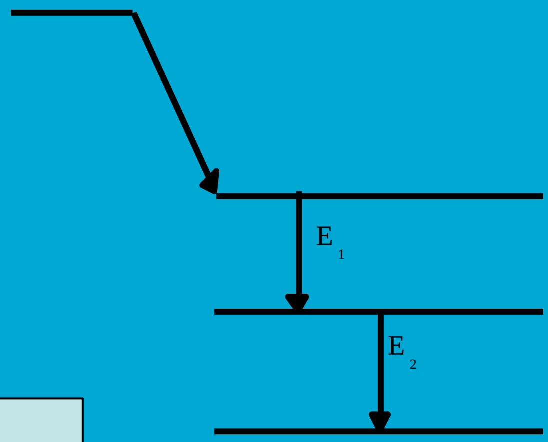
- Theory (no bias)
- Theory (bias)
- Experiment I: verification of 1 program
- Results I
- Experiment II: indirect verification of others
- Results II
- Conclusions / Implications

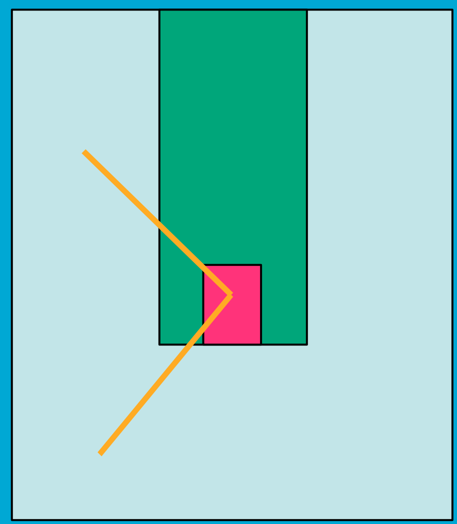
Theory (no bias)

$$A_{E_1} = N\varepsilon(E_1)(1-\varepsilon_t(E_2))$$

$$A_{E_2} = N\varepsilon(E_2)(1-\varepsilon_t(E_1))$$

$$A_{E_1+E_2} = N\varepsilon(E_1)\varepsilon(E_2)$$





Theory (biases)

- Given: Areas determined with program Y are a factor *f* higher than areas determined with program X.
- Results in: Peak efficiencies determined with program Y will be a factor *f* higher than those determined with X. Total efficiencies will be the same as determined with both.

Activity measurements:

program X

$$N_{X,E_1} = A_{X,E_1} / \varepsilon_X(E_1)(1 - \varepsilon_t(E_2))$$

$$N_{X,E_1+E_2} = A_{X,E_1+E_2} / \varepsilon_X(E_1)\varepsilon_X(E_2)$$

program Y

$$N_{Y,E_1} = A_{Y,E_1} / \varepsilon_Y(E_1)(1 - \varepsilon_t(E_2))$$

$$N_{Y,E_1+E_2} = A_{Y,E_1+E_2} / \varepsilon_Y(E_1)\varepsilon_Y(E_2)$$

For the ordinary peak, we get

$$N_{Y,E_1} = A_{Y,E_1} / \varepsilon_Y(E_1)(1-\varepsilon_t(E_2))$$
$$= f\,A_{X,E_1} / f\varepsilon_X(E_1)(1-\varepsilon_t(E_2))$$
$$= N_{X,E_1}$$

So far, so good: The results is identical for both programs. But for the sum peak we get

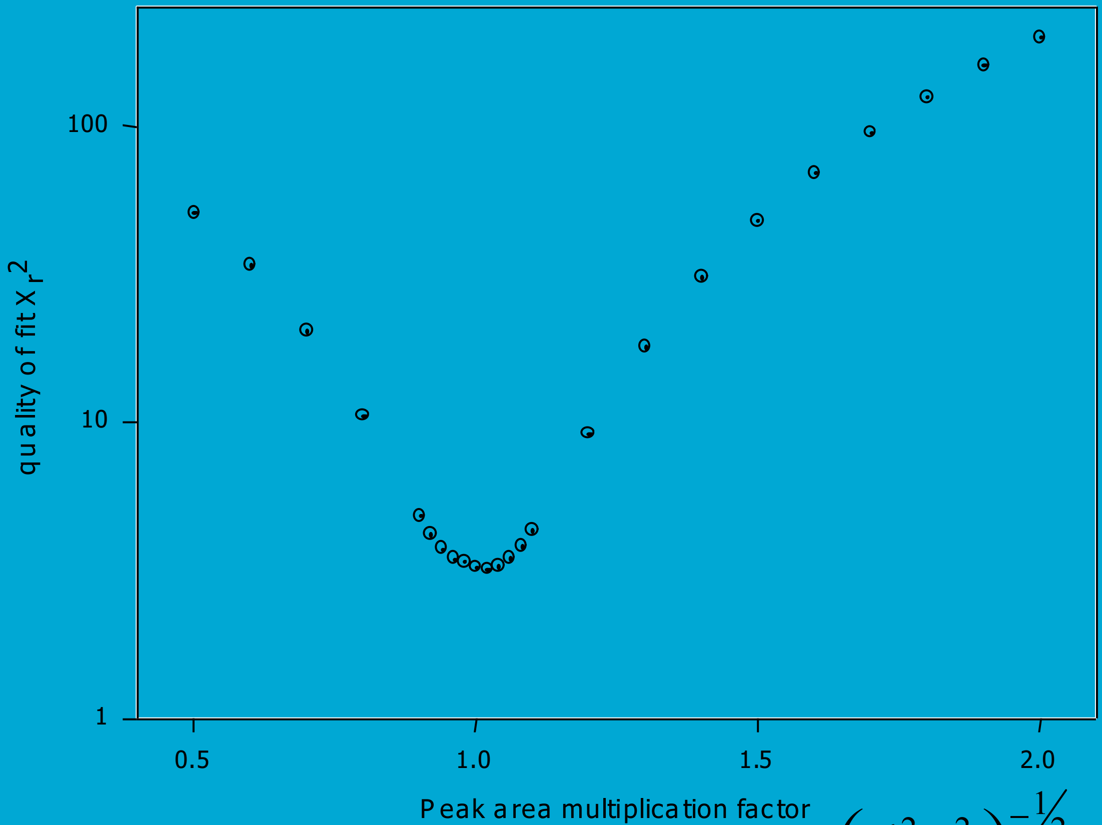
$$N_{Y,E_1+E_2} = A_{Y,E_1+E_2} / \varepsilon_Y(E_1)\varepsilon_Y(E_2)$$
$$= fA_{X,E_1+E_2} / f\varepsilon_X(E_1) f\varepsilon_X(E_2)$$
$$= N_{X,E_1+E_2} / f$$

Discrepancy! Can we tell which program is *right*? Yes, that must be the program for which $N_{E_1+E_2} = N_{E_1}$!

Experimental I

- ^{82}Br source with known activity (determined in calibrated, coincidence-free counting geometry) measured close to end cap of Ge(Li) coaxial detector.
- Peak areas determined with in-house software. This software was "calibrated" for peak areas using a clean ^{137}Cs peak without continuum and comparison of fit- and integration results.
- Peak areas multiplied with bias factors ranging from 0.5 to 2.0.
- Fit of peak efficiency and peak-to-total curve to measured peak areas attempted. Result: c_r^2 indicating internal consistency of peak areas and corresponding activities.

Results



With fit of parabola to minimum and $\sigma_f = \left(\frac{d^2 \chi_r^2}{df^2} \right)^{-1/2}$, we get the optimum factor f = 1.009 +/- 0.006.

Experimental II

- The “straight” 1995 IAEA test spectrum was analyzed with the in-house IRI software.
- The resulting areas were compared with the reference areas: The weighted average of the peak area ratios was computed.
- The resulting peak area ratio of 1.001 +/- 0.003 was compared to those obtained from the other programs tested in the 1995 IAEA intercomparison.

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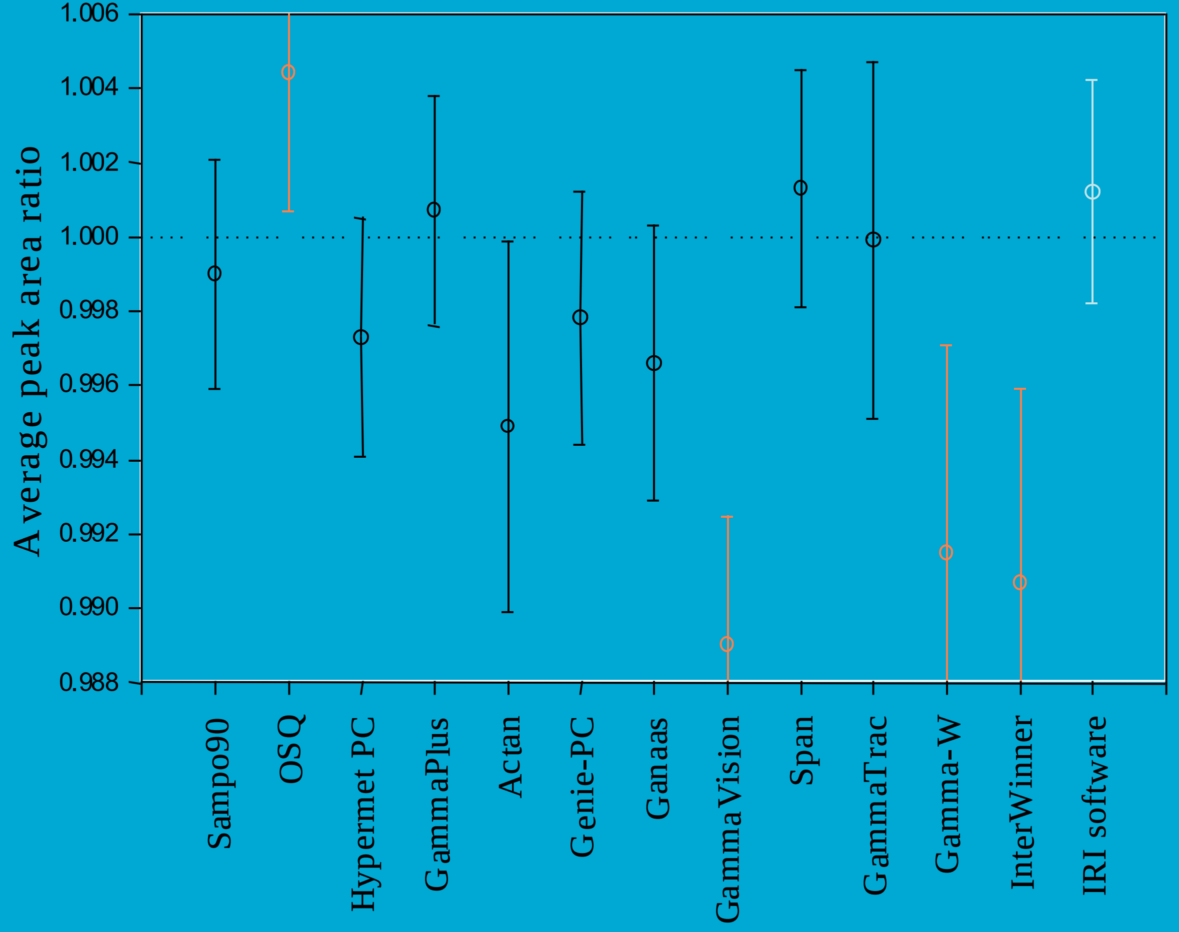
Results II


Average peak area ratio
1.006
1.004
1.002
1.000
0.998
0.996
0.994
0.992
0.990
0.988
Sampo90
OSQ
Hypermet PC
GammaPlus
Actan
Genie-PC
Ganaas
GammaVision
Span
GammaTrac
Gamma-W
InterWinner
IRI software

Conclusions / Implications

- ***There can be only one correct peak area***
- The tested program did not have a significant bias in this respect (and since this program was verified with the 1995 IAEA test spectra:)
- The following programs do not have a bias exceeding 1 % either: GammaTrac, Span, Ganaas, Genie-PC, Actan, GammaPlus, Hypermet PC, OSQ, and Sampo90.
- The following programs may have a bias of 1 % (areas are low): InterWinner, Gamma-W and GammaVision.
- Finally, *the ANSI standard for verification testing of programs for gamma-ray spectrum analysis should provide a procedure to test for the absence of peak-area-determination bias.*

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and more...

Cascade Summing in Gamma-Ray Spectrometry in Marinelli-Beaker Geometries: The Third Efficiency Curve

- Menno Blaauw, Sjoerd J. Gelsema

Overview of the second advanced topic

- The voluminous effect problem
- The solution
- The experiment
- Conclusions

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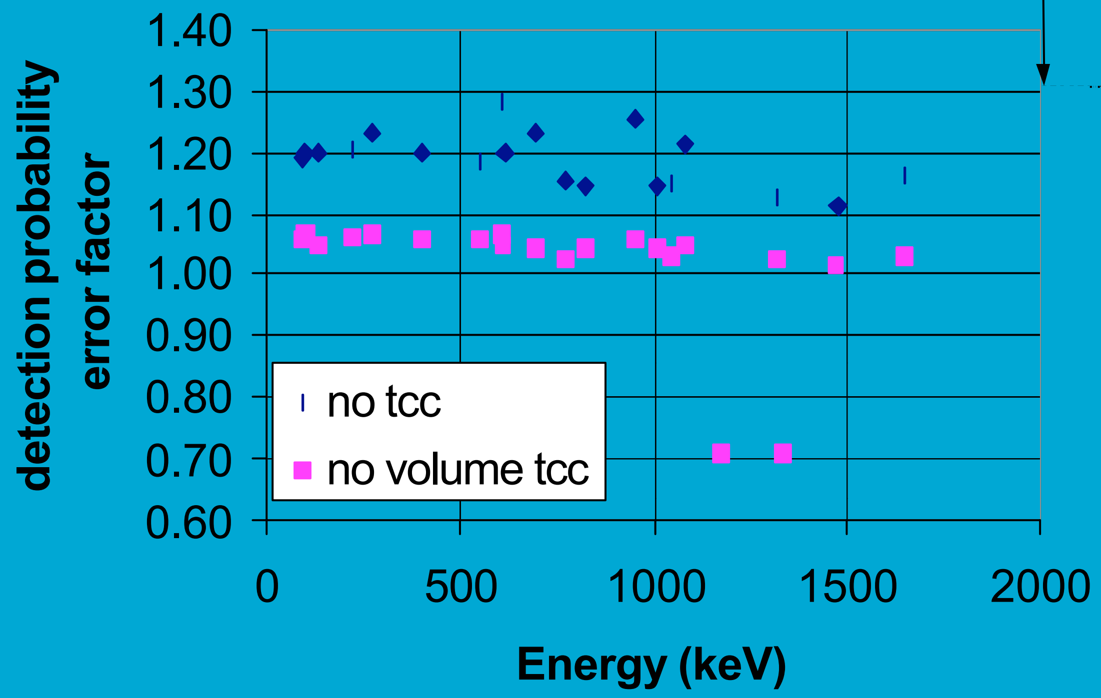
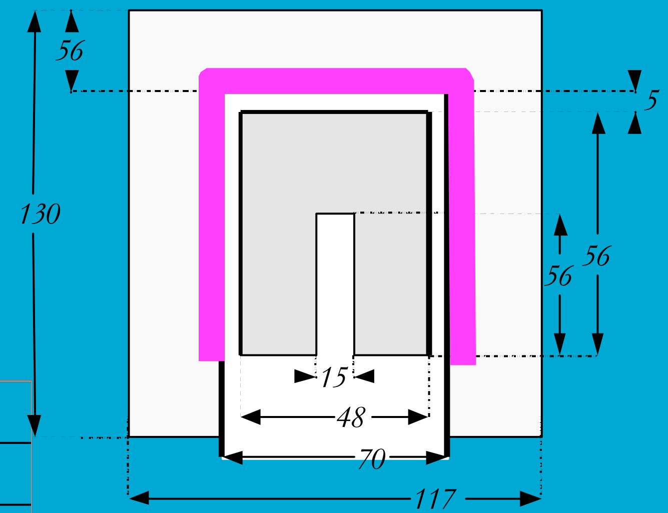
the problem





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the solution

$$P=\prod_{i=1}^{M}\varepsilon_i\prod_{j=1}^{N-M}(1-\varepsilon_{tj})$$ the general equation

$$\langle\varepsilon_1\varepsilon_2\rangle=\langle\varepsilon_1\rangle\langle\varepsilon_2\rangle+r\sigma_1\sigma_2$$ the voluminous correction to the product of the averages

$$l^2=\langle\varepsilon^2\rangle/\langle\varepsilon\rangle^2$$ definition of the sqr-to-lin ratio "*l*"

$$\langle\varepsilon_1\varepsilon_2\rangle=\langle\varepsilon_1\rangle\langle\varepsilon_2\rangle\left(1+r\sqrt{l_1^2-1}\sqrt{l_2^2-1}\right)$$ the voluminous correction again

$$\langle\varepsilon_1\varepsilon_2\rangle=l_1\langle\varepsilon_1\rangle l_2\langle\varepsilon_2\rangle$$ approx. the voluminous correction

$$\langle\varepsilon_1\varepsilon_2..\varepsilon_N\rangle=l_1\langle\varepsilon_1\rangle l_2\langle\varepsilon_2\rangle..l_N\langle\varepsilon_N\rangle$$ the approximation generalized

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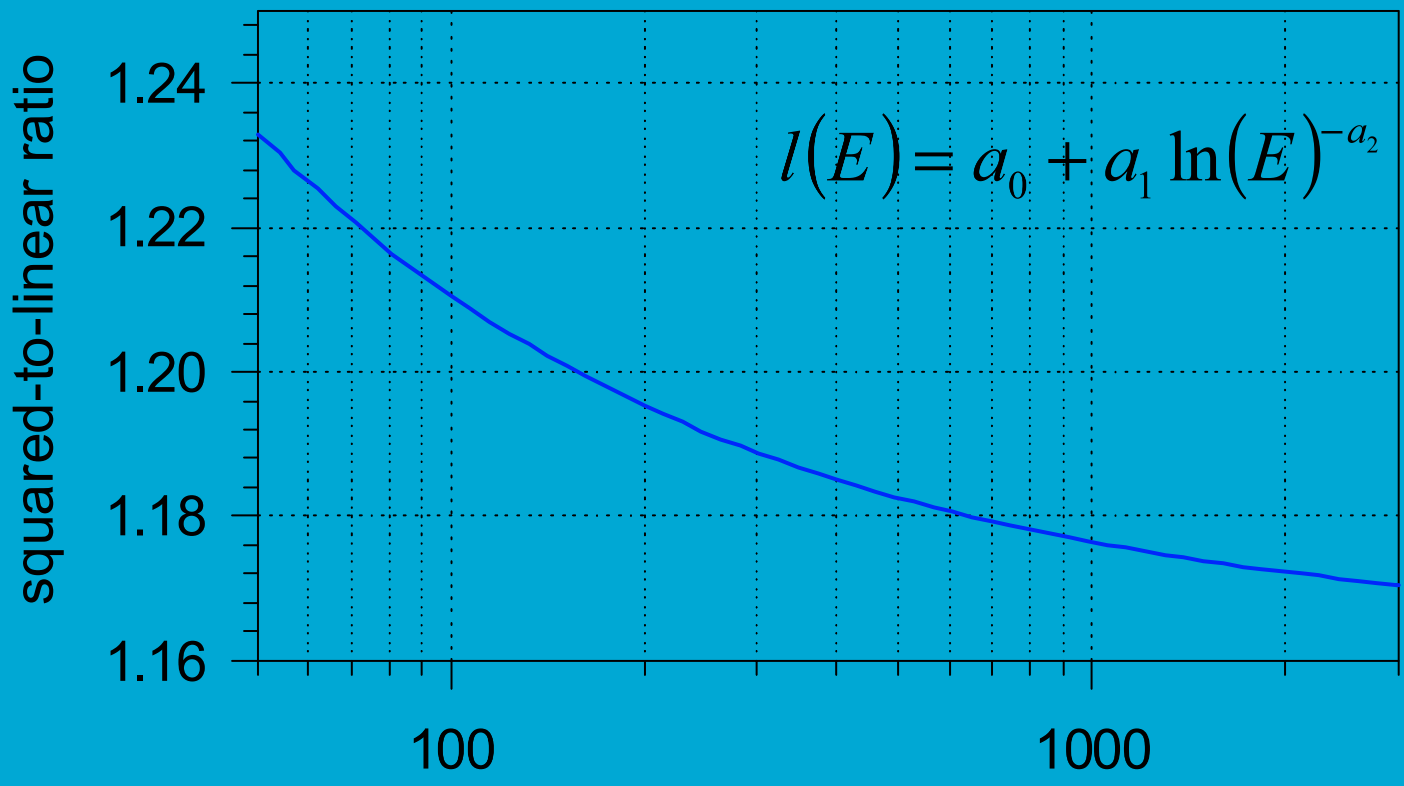
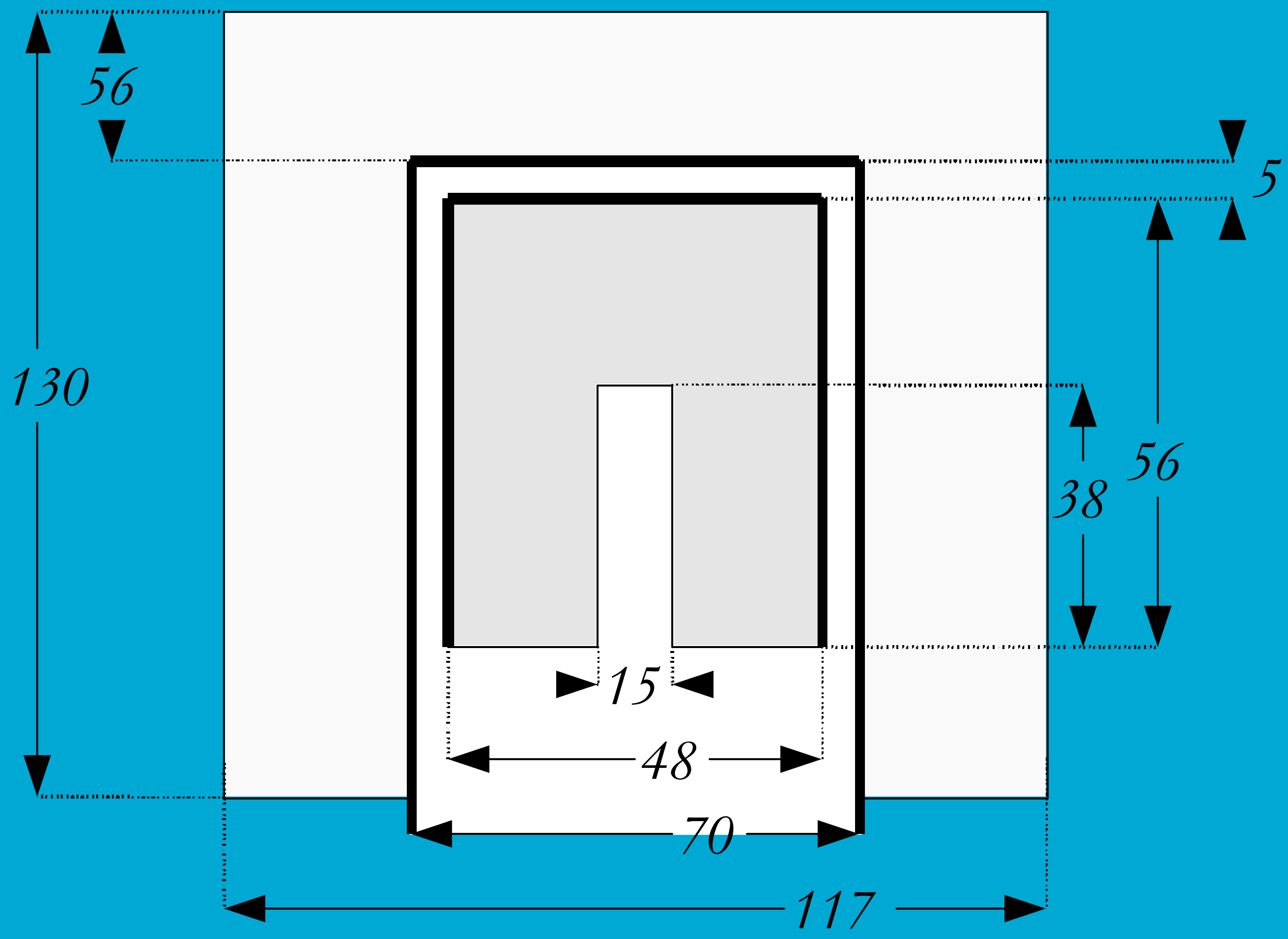
the shape of the solution: The third efficiency curve



The experiment



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The experiment

- Create source and measure calibration spectrum:
  - Amersham certified solution with ^{57}Co, ^{60}Co, ^{88}Y, ^{109}Cd, ^{113}Sn, ^{137}Cs and ^{139}Ce
  - activated NaBr solution (activity determined with point sources)
  - Measure the two, fit peaks and merge the lists of peaks
- Determine curves
  - Fit the three efficiency curves to the spectrum: Peak efficiency (4 pars), p/t ratio (2 pars) and stl ratio (3 pars).
- Test the curves
  - Get test source
  - Measure and interpret spectra
  - Compare activities found with known values.

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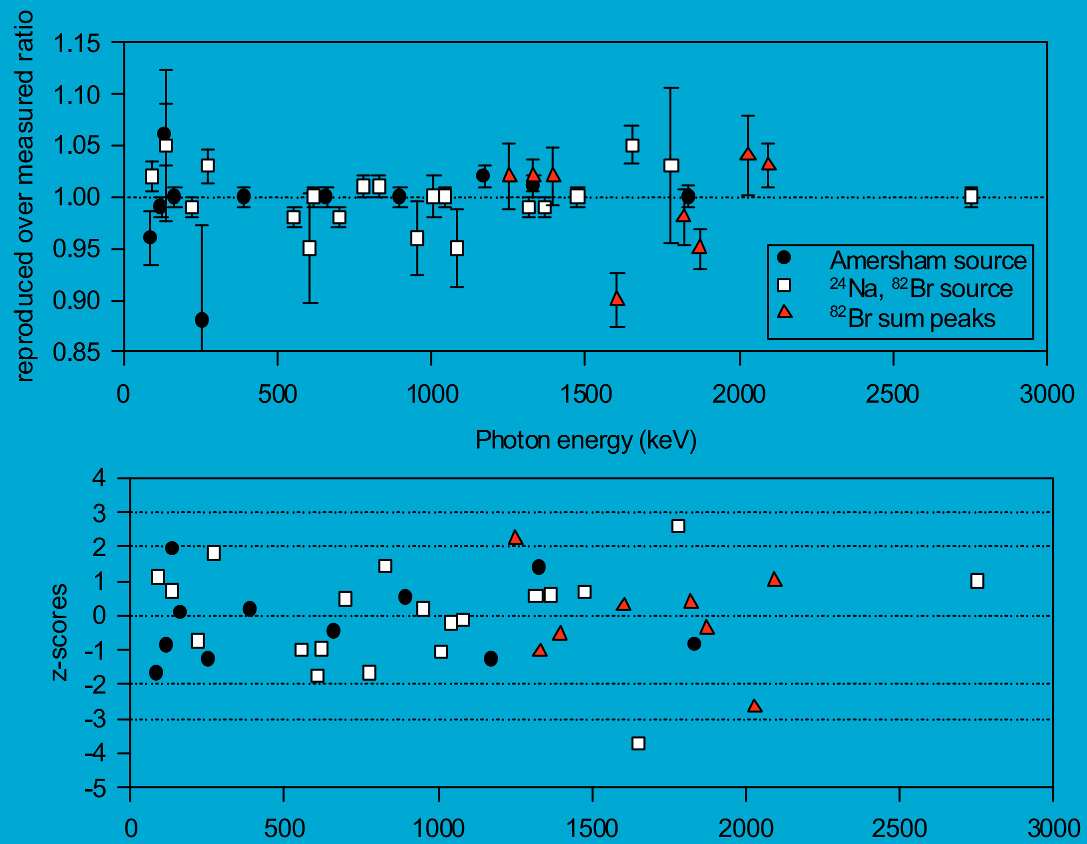
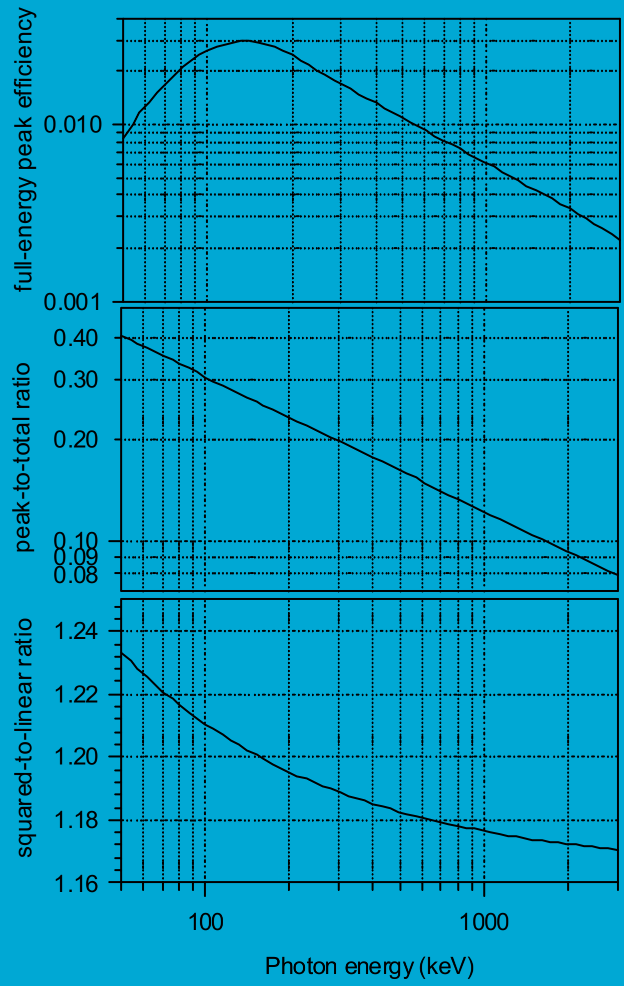
Fit results

$$X_r^2 = 2.2 \text{ at } 31 \text{ d.f.}$$





Test results

radionuclide	known activity	3-curve method	radionuclide-specific
^{40}K	186 ± 2	181 ± 4	
^{57}Co	137.3 ± 0.9	139 ± 0.7	136.1 ± 1.2
^{60}Co	116.7 ± 0.6	118.2 ± 0.5	116.7 ± 0.8
^{133}Ba	126.4 ± 0.9	129.6 ± 0.5	
^{134}Cs	129.8 ± 0.7	129.5 ± 0.5	
^{137}Cs	144.8 ± 0.9	144.0 ± 0.7	141.0 ± 1.2

(1 s.d. uncertainties)

Conclusions

- ☺ The third efficiency curve accounts for voluminous-source effects in cascade summing corrections
- ☺ The method has an accurucy of 1% or better, is much less laborious, and therefore can compete with radionuclide-specific calibration
- 😐 As yet, the third curve must be determined in the same geometry as the samples, but there is hope.

When is the voluminous effect relevant for INAA?

- The "voluminous effect" becomes significant in the case of two coincident gamma-rays when the detection efficiency for both energies involved cannot be considered constant over the sample volume
- since if one of the two is constant:

$$\left\langle \varepsilon_1 \varepsilon_2 \right\rangle = \left\langle \varepsilon_1 \right\rangle \left\langle \varepsilon_2 \right\rangle$$

- So in a well-type detector, things tend to be fine,
- But in a small capsule on top of the end cap, they may very well not be!

Conclusions

- It is possible to use point sources emitting coincident gamma-rays to determine efficiency curves, even close to the detector
- but
- It is wise to stay away from the end cap of the detector when measuring sizeable samples.
- and
- knowing the decay schemes, coincidence methods provide the tools to measure absolute activities and establish what “true” peak areas are
- and to verify decay schemes if the activity is known