Coincidence summing corrections in gamma-ray spectrometry

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1. Introduction

Gamma-ray spectrometry with high resolution detectors:

- peak energies => nuclide identification
- peak count rate => nuclide activity
  - Relative measurements

$$A = \frac{R(E)}{R_0(E)} \cdot A_0$$

R, R_0 =count rate for sample and standard
A, A_0 =nuclide activity in sample and standard

  - Measurements based on an efficiency calibration curve

$$A = \frac{R(E)}{\varepsilon(E) \cdot P_\gamma(E)} \cdot C$$

$P_\gamma(E)$ = gamma emission probability

$\varepsilon(E)$ = full energy peak efficiency (FEPE)

C = correction factors

  - FEPE calibration is based on the possibility to obtain $\varepsilon(E)$ for any energy from the measured values $\varepsilon(E_i)$ for several energies E_i ($\varepsilon(E)$ is a smooth function of energy E)
- relatively weak self-attenuation => no need for chemical preparation

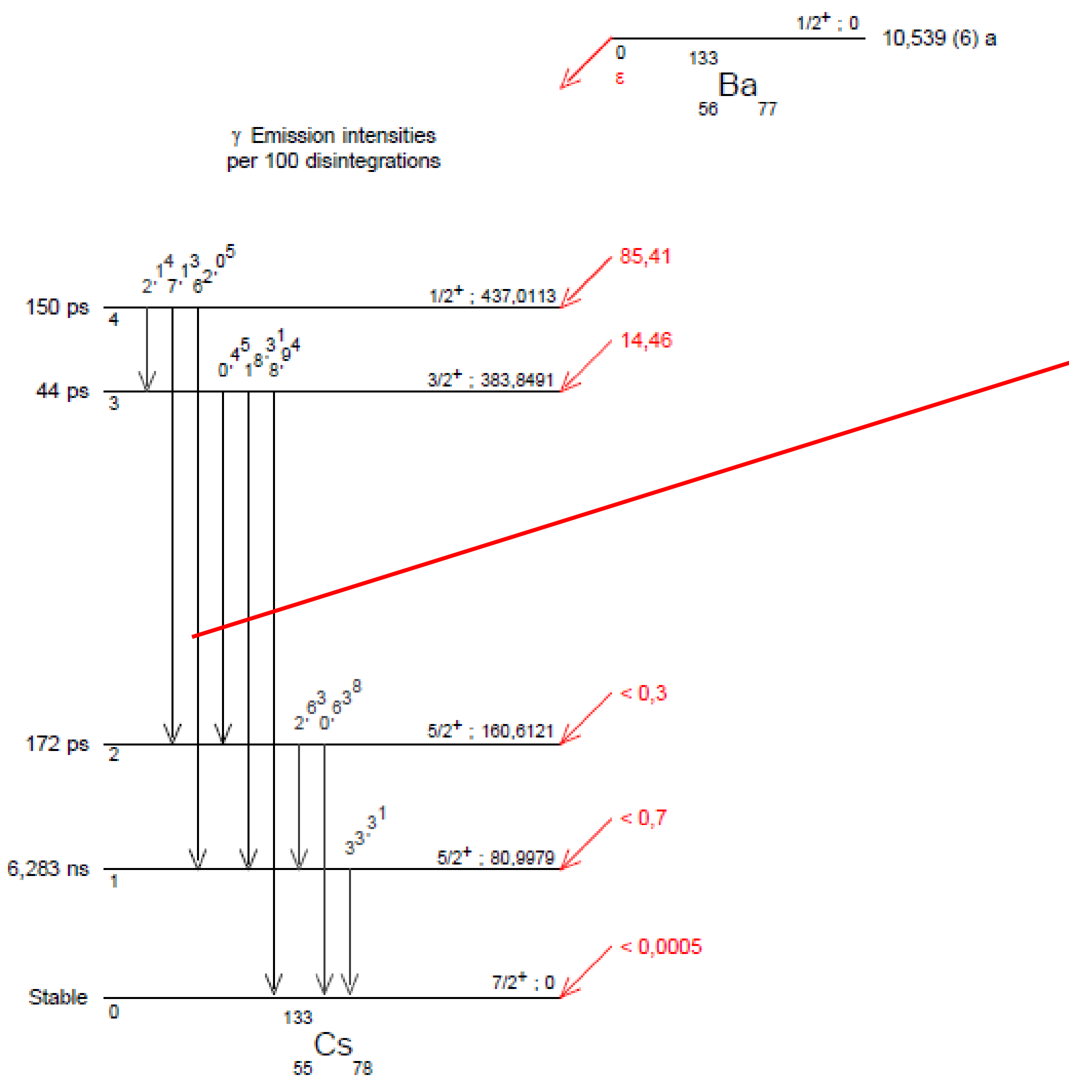
=> Gamma spectrometry: multielemental, nondestructive analysis method

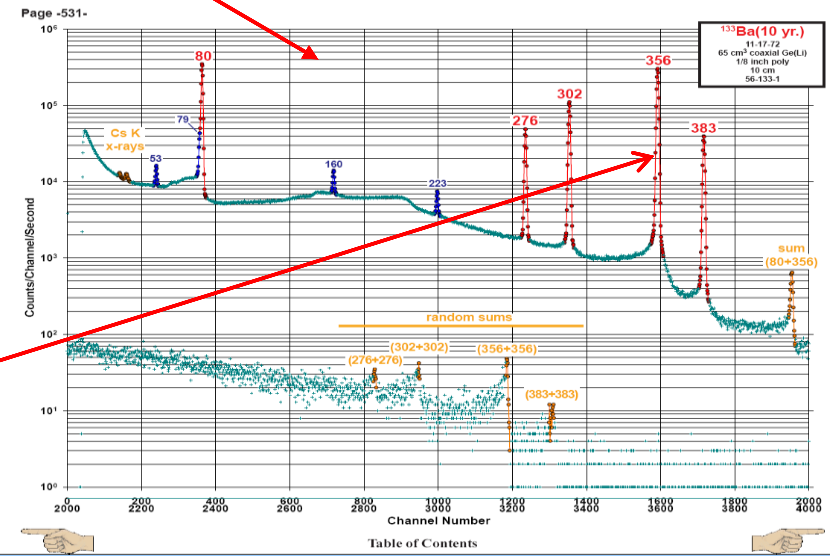
Low efficiency measurements

^{133}Ba

65 cm^3 Ge(Li)

Data source: Gamma-ray spectrum catalogue, INEEL





Source: Monographie BIPM-5 Table of Radionuclides Vol. 5 (2010)

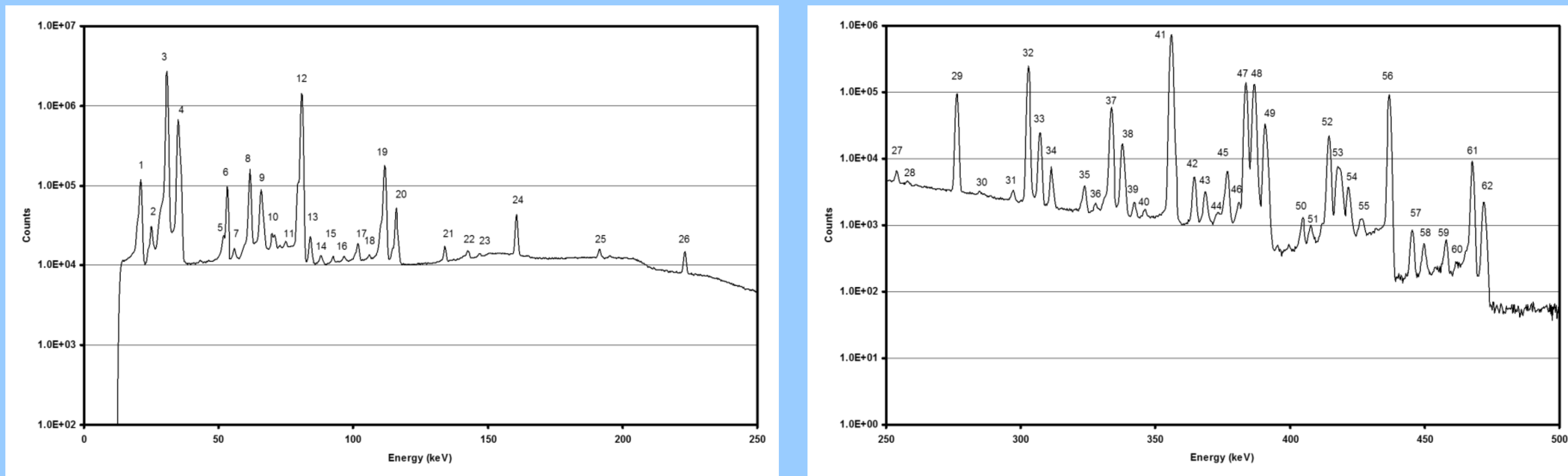
-One to one correspondence gamma emissions ⇔ peaks

-Peak count rate proportional to gamma intensity and to the efficiency $\varepsilon(E)$

- $\varepsilon(E)$ independent of nuclide, smooth function of energy E

High efficiency measurements

^{133}Ba – same nuclide as in previous figure – point source on endcap, n-type HPGe (spectrum split in two parts for better readability)



Spectrum of a ^{133}Ba point source measured in high efficiency conditions (Arnold and Sima, ARI 64 (2006) 1297)

- peaks not associated with gamma emission of ^{133}Ba

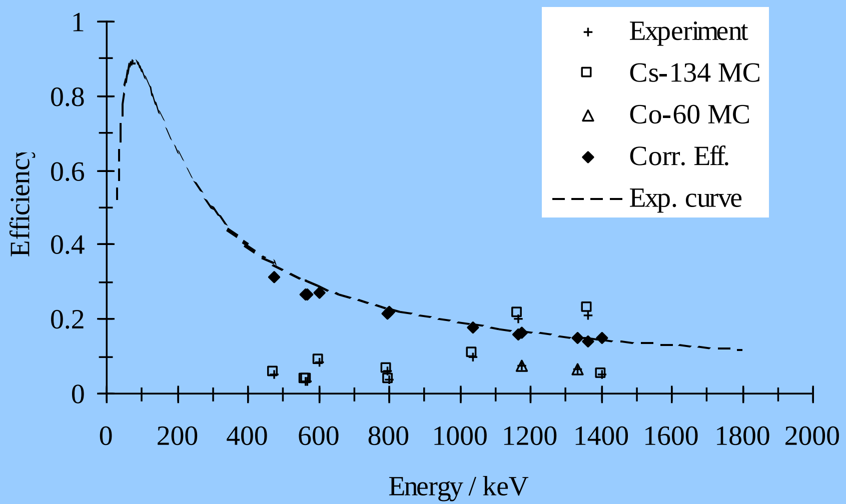
- distortion of the count rate in the peaks of the gamma photons of ^{133}Ba

=> the FEP efficiency from the efficiency calibration curve not appropriate

Coincidence summing effects responsible for the differences between the spectra measured in low and in high efficiency conditions

Coincidence summing effects extremely high for measurements with well-type detectors

- sum peaks very pronounced
- high distortion of the FEP efficiency due to coincidence summing



Well-type detector measurements
Apparent and corrected FEP efficiency

Source: Sima and Arnold, ARI 47 (1996) 889

Consequences:

- coincidence summing corrections required for the evaluation of the activity
- problems in nuclide identification (automatic analysis based on nuclide libraries)
  - a pure sum peak is erroneously attributed to a nuclide not present in the sample
  - a nuclide is not recognized due to an incorrect match of the count rate
  - incorrect activity evaluation due to unaccounted peak interferences

=> Need for coincidence summing corrections!

Why coincidence-summing effects are more important in present day measurements than years ago?

Features of present day applications of gamma-ray spectrometry:

- broad range of samples (volume, shape, matrix)
- demanding values of the detection limit, in short measurement time
- low and reliable uncertainties
- high throughput of the laboratory

Preferred measurement conditions:

⇒ Use of high efficiency detectors

⇒ Preference of high efficiency measurement conditions

⇒ When possible, measurement of volume samples

⇒ Automatic analysis of the spectra

Consequences:

- high efficiency detectors, close to detector measurement geometry: coincidence summing effects

=> nuclide dependent efficiency

- volume sources: self-attenuation effects

=> matrix dependent efficieny

=> intricate nuclide and matrix effects for volume sources

2. Physics of coincidence summing effects

Inability of the detector to record independently two photons very close in time
Coincidence resolving time of a HPGe spectrometer – microseconds

- charge collection, signal forming and analysis

Typical lifetime of excited nuclear states – nanoseconds or less

- photons emitted quickly one after the other in nuclear deexcitation cascades

Mean time between two successive decays of different nuclei for a source with *A* = 100 kBq – 10 microseconds; mean time between the registration of signals due to the radiations emitted by different nuclei – longer (not all decays recorded, due to efficiency)

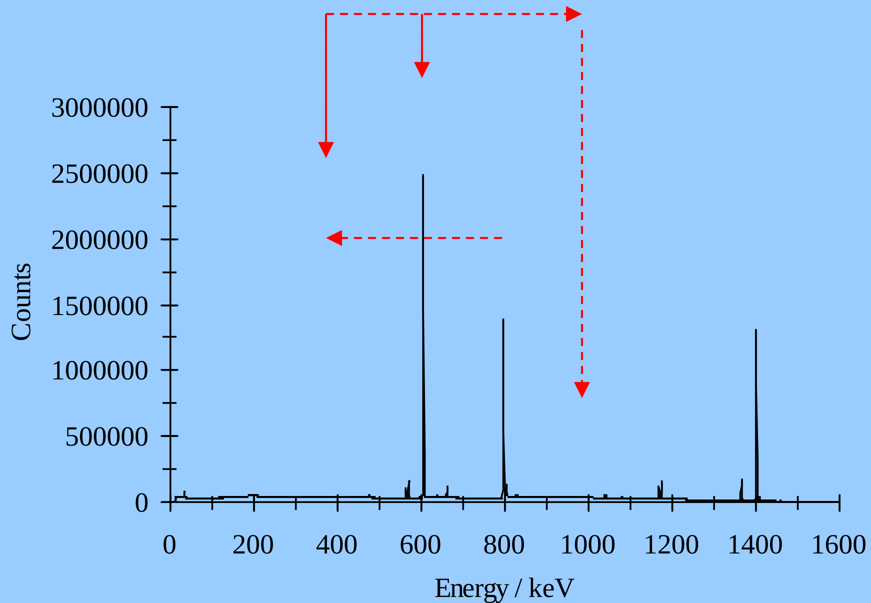
$\Rightarrow$ If n photons interact in the detector within the resolving time

$\Rightarrow$ a single signal delivered by the detector instead of n separate signals

$\Rightarrow$ the signal corresponds to a channel of energy

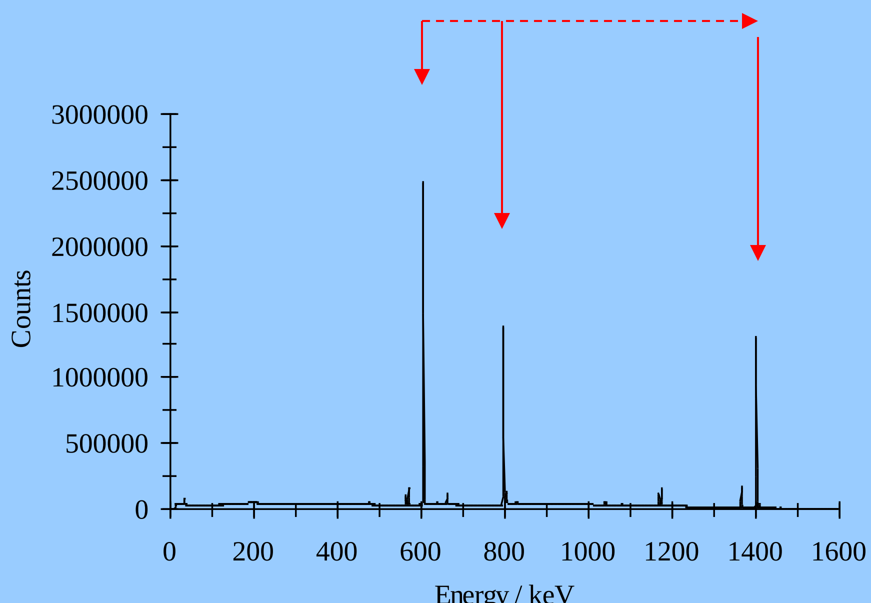
$$ED_{sum} = ED_1 + ED_2 + \ldots + ED_n$$

ED_k = energy deposited by the k-th photon (energy of the photon E_k) in the detector (in the peak, if $ED_k=E_k$ or in the total spectrum if $ED_k < E_k$)

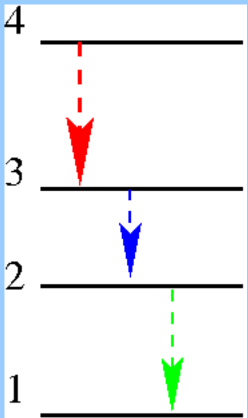


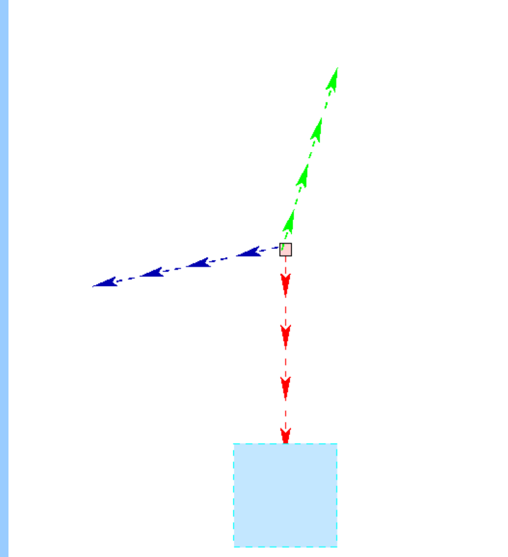
Coincidence losses from the 604 keV peak
The signal corresponding to complete absorption of 604 keV photon and incomplete absorption of the 796 keV photon added resulting in a single signal corresponding to an energy of 604+DE(796)

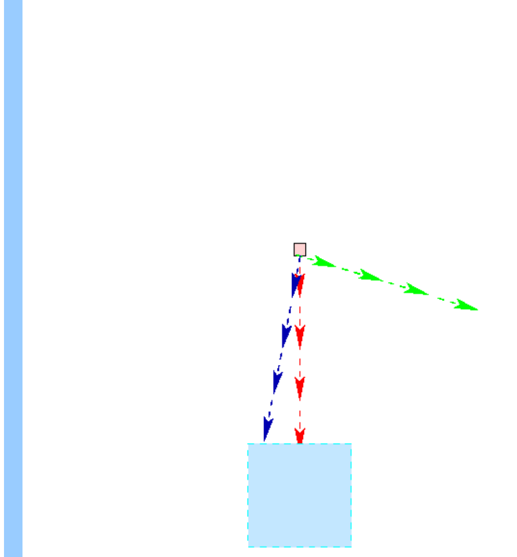
Spectrum of ^{134}Cs measured with a well-type detector

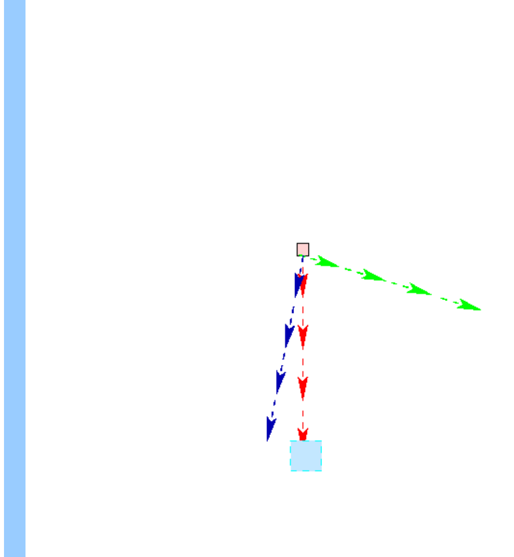


Sum peak at the energy 604+796=1400 keV
The signals corresponding to complete absorption of 604 and 796 keV photons are added resulting in a signal corresponding to an energy of 1400 keV
Probability of this event in comparison with the above:
$\varepsilon(796)$ in comparison with $\eta(796)$
ε = full energy peak efficiency
η = total efficiency



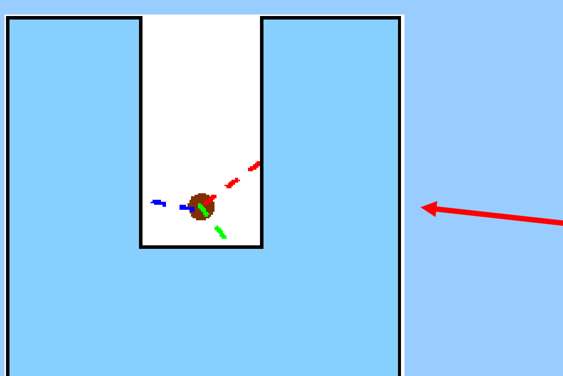


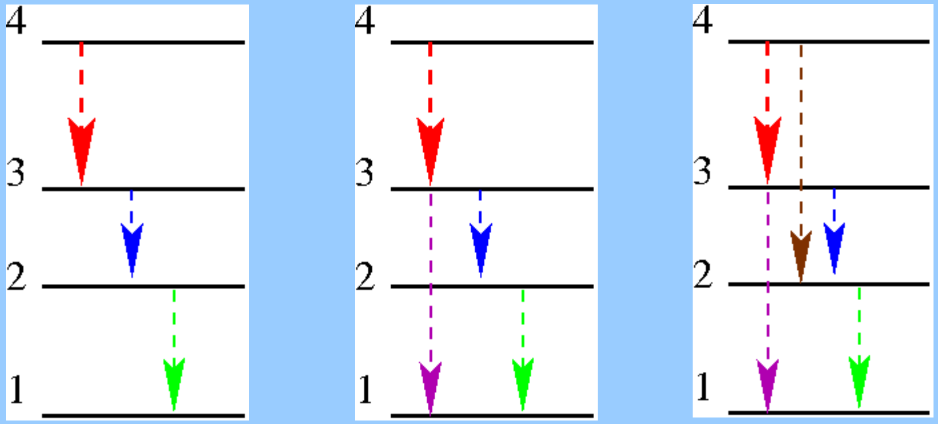




True Coincidences

- The three photons are emitted practically in the same time in various directions (with an angular correlation)
- Sometimes it happens that two photons interact with the detector (in closed end coaxial detectors the probability that three photons interact in the detector is much smaller than that for two photons)
- The probability of interaction depends on the detector dimensions and on the detector efficiency
- It is highest in the case of well-type detectors
- Coincidence summing effects – higher in high efficiency conditions (solid angle, intrinsic efficiency)



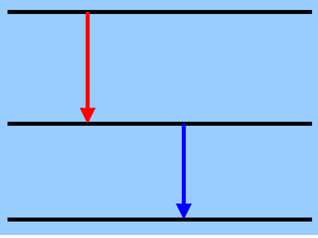


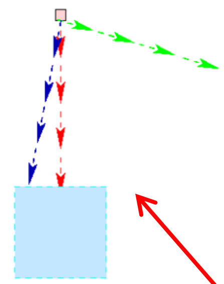
The groups of photons that are emitted together and their joint emission probabilities are different for the three decay schemes

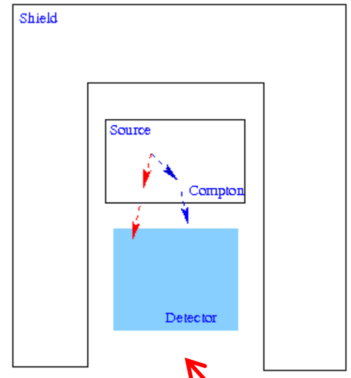
True Coincidences

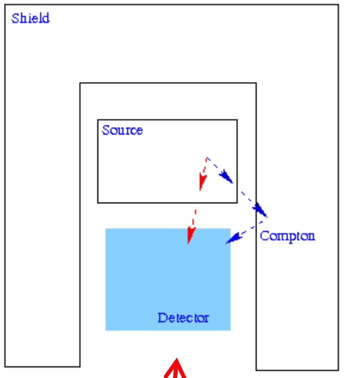
- Coincidence summing effects depend on the decay scheme and are specific to each transition

True Coincidences

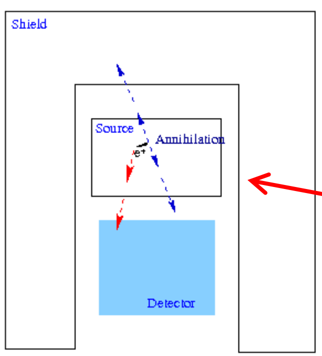


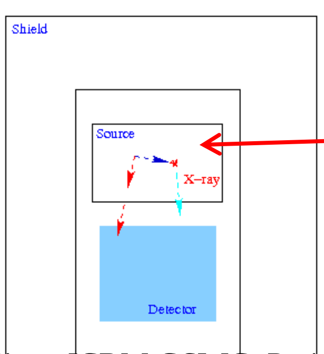




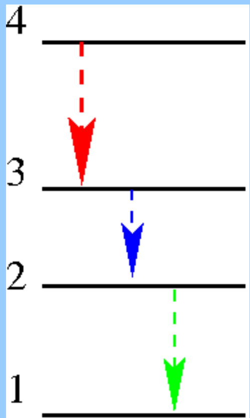


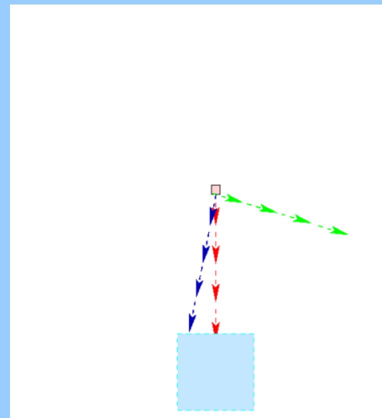
- Coincidence-summing affecting the "red" photon - Contributions from all the radiations following decay
  - gamma from transitions, X-rays (EC, conversion electrons)
  - radiation scattered in the source, shield
  - annihilation photons
  - X rays excited in the shield, in the matrix
  - beta particles, bremsstrahlung etc
- Relative contribution independent of A!

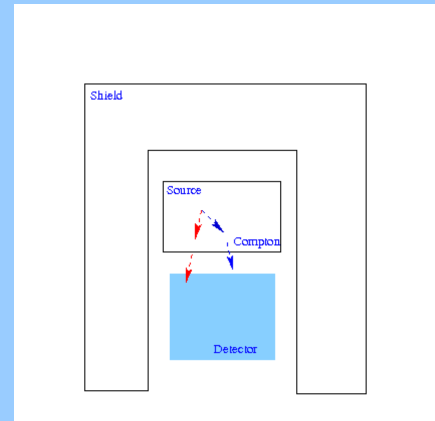


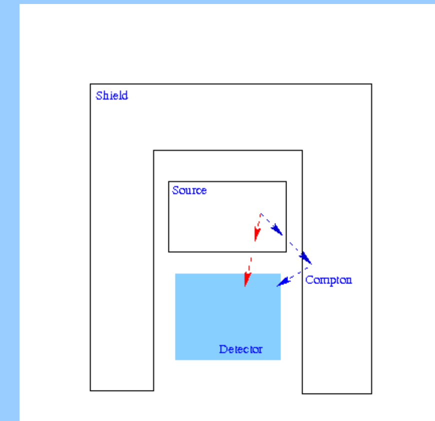


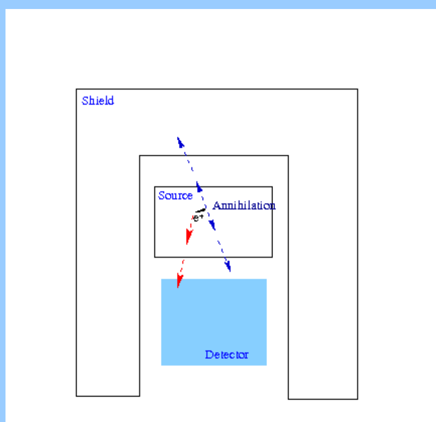
True coincidence losses from the peak depend on the total efficiency

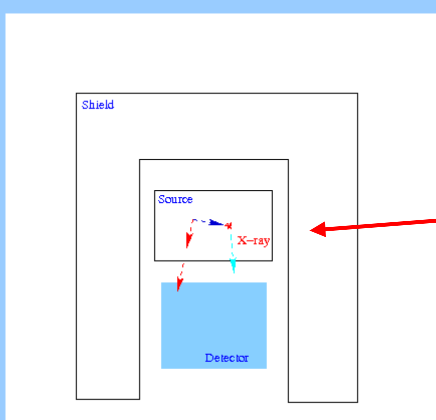












- Contributions to coincidence losses from the peak of the “red” photon due to the detection of the “blue” photon

  - direct gamma interaction with the detector

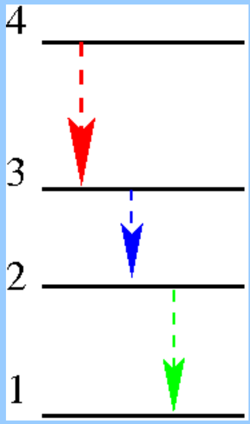
  - radiation scattered in the source, shield

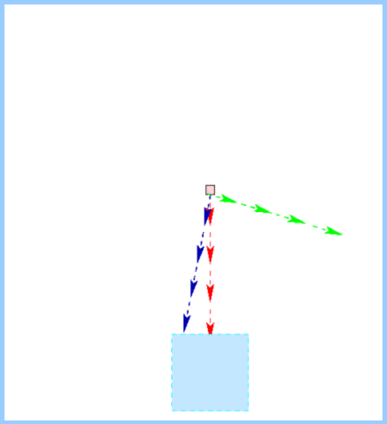
  - annihilation photons (in case of positron decay)

  - X rays excited in the shield, in the matrix

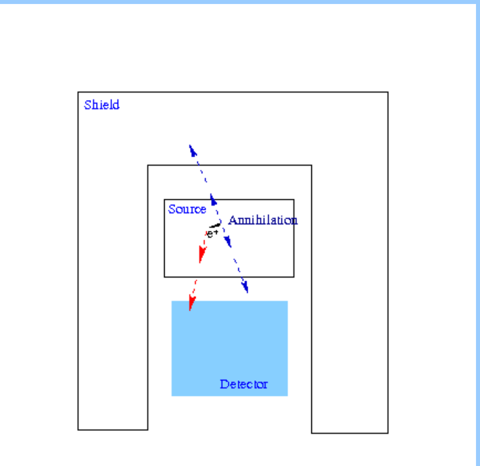
=> the same processes are responsible for the total efficiency of the “blue” photon in the absence of coincidences

Coincidence summing contributions to sum peaks depend on the peak efficiency

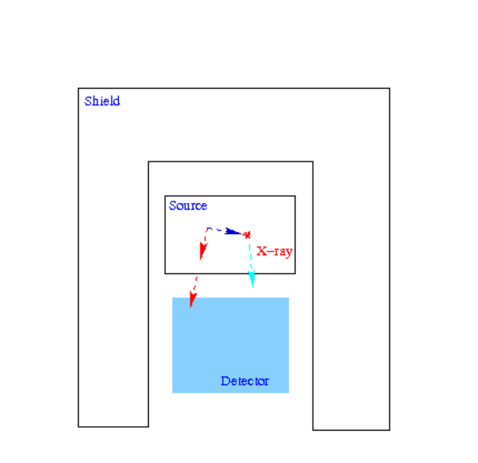




- Sum peak contribution of the "blue" and "red" photons: the same processes in which each photon would be registered in the peak in the absence of coincidence effects
- Sum peak of a gamma photon and an X-ray from EC decay of internal conversion: the same processes in which gamma and X would be registered in their peaks in the absence of coincidence effects



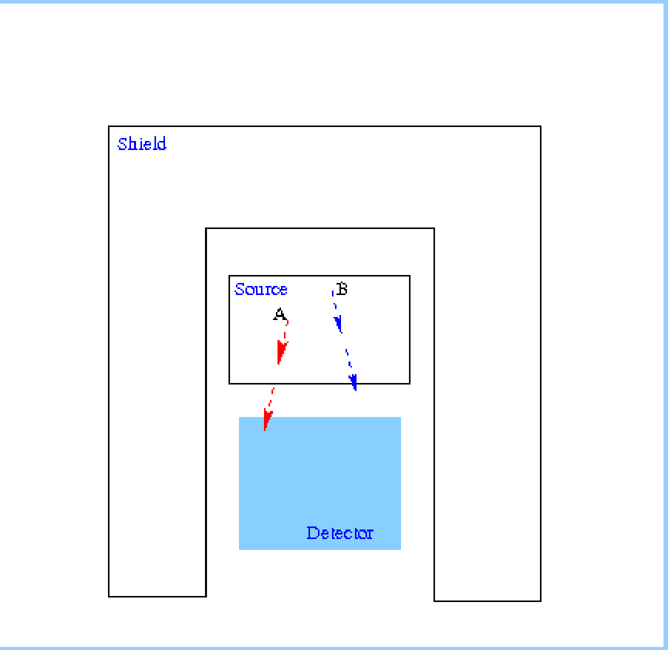
- Sum peak of a gamma photon and an annihilation photon: same processes in which each photon (gamma, annihilation) would be registered in the corresponding peak in the absence of coincidence effects



- Sum peak of a gamma photon and a matrix X-ray: same processes in which each photon (gamma, X) would be registered in the corresponding peak in the absence of coincidence effects

Random coincidences

Two different nuclei may emit radiations close in time one by the other by chance



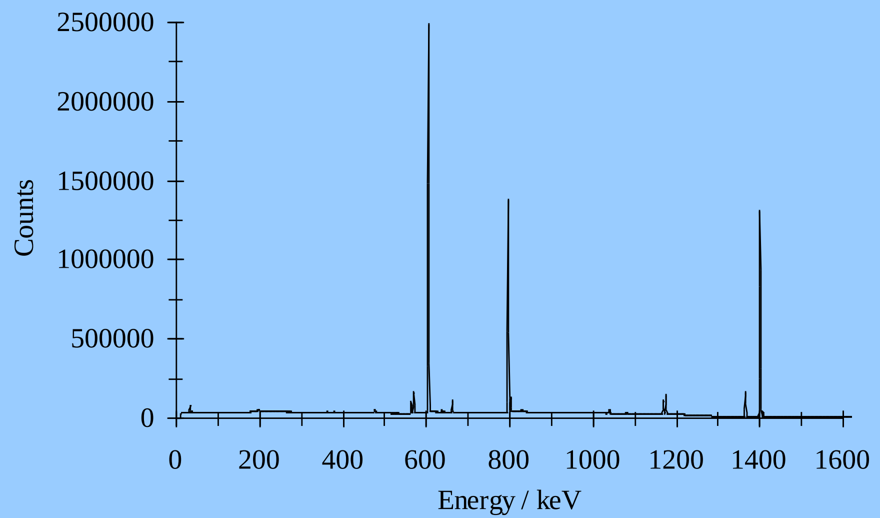
Important when the count rate is high
- The effect can be avoided by decreasing the count rate, e.g. measuring the source at big distances from the detector

- The displacement of the source far from the detector is not a good choice for low level samples; therefore for low level samples coincidence summing effects can not be avoided (true coincidence summing corrections are independent of the activity of the source)

Examples of coincidence-summing effects

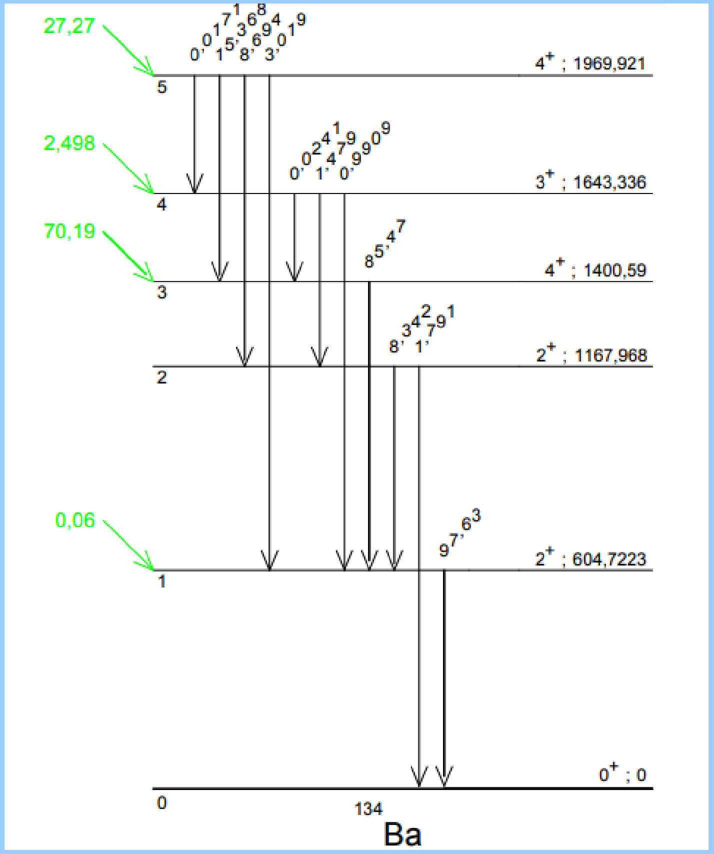
Coincidence with gamma photons

^{134}Cs - well-type detector (350 cm^3 crystal)



Source: Sima and Arnold, ARI 47 (1996) 889

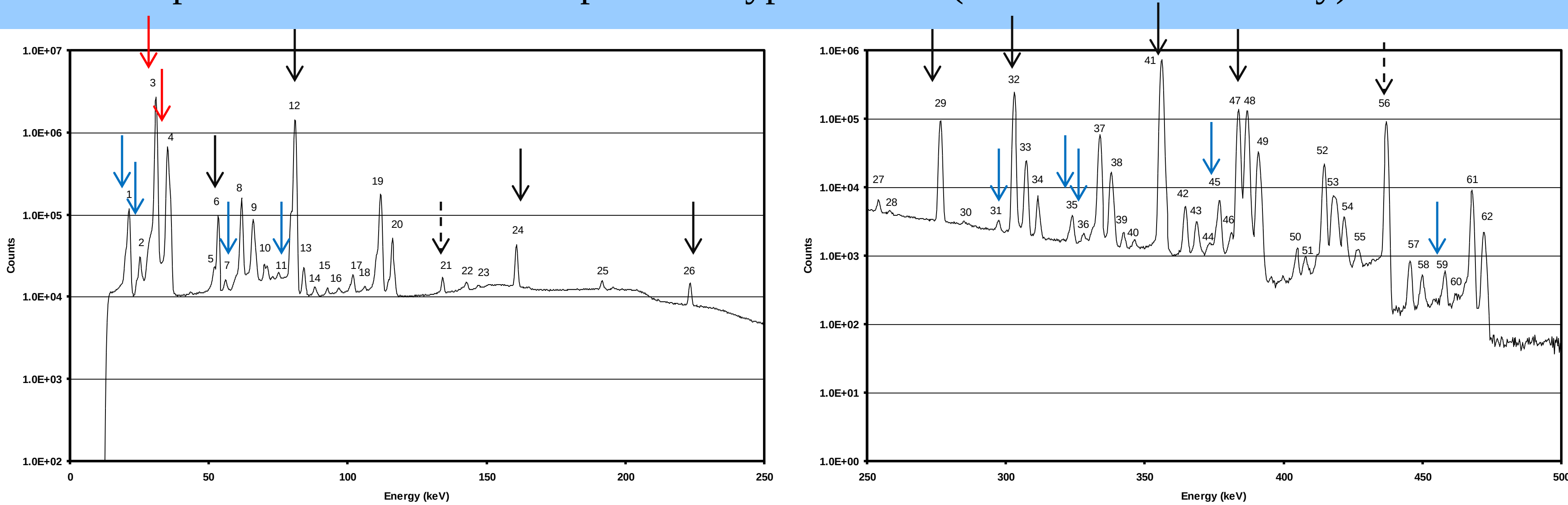
- 1400 keV peak completely due to coincidences (604+796 keV)
- The count rate in the 604 and 796 kev peaks is about 30% from the count rate in the absence of coincidence-summing ($F_C \cong 0.3$)
- $F_C \cong 0.15$ for 801 and 475 keV peaks, $F_C \cong 0.1$ for 563 and 569 keV peaks
- $F_C \cong 1.5$ for 1365 keV peak (summing-in dominates)



Source: Monographie BIPM-5 Table of Radionuclides Vol. 7 (2013)

Coincidence with X-rays

^{133}Ba - point source on the endcap of an n-type detector (25% relative efficiency)

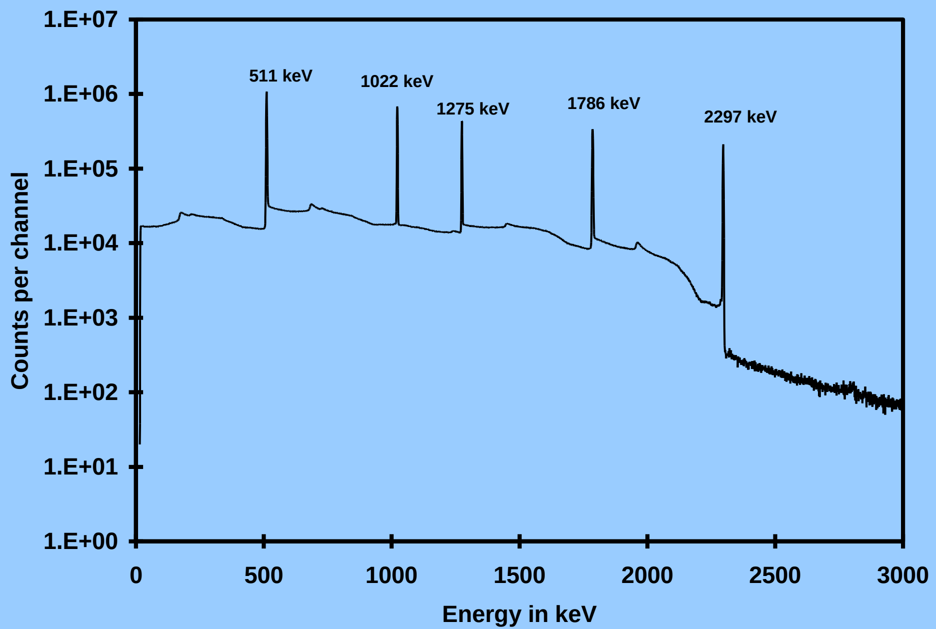


Source: Arnold and Sima ARI 64 (2006) 1297

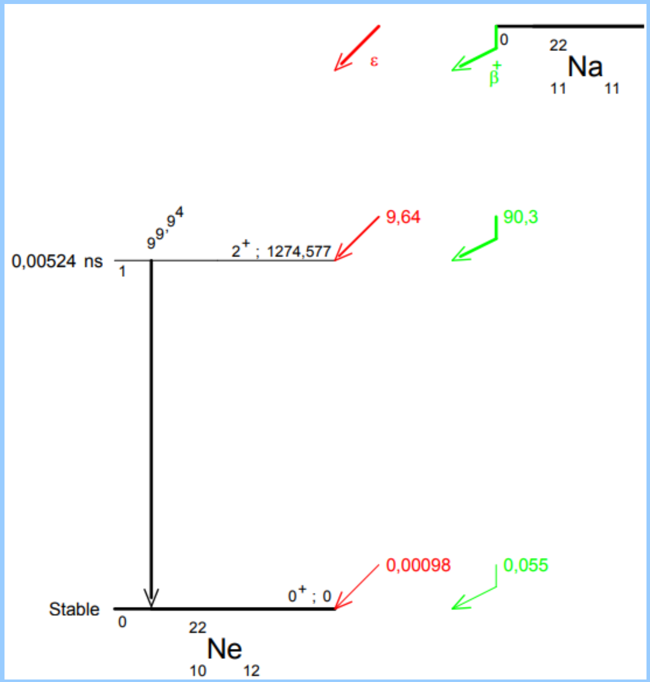
- Gamma peaks (black arrows): F_C between 0.5 and 1
- Sum peaks with gamma-rays (dashed black arrows), completely due to coincidences: most prominent 437 keV
- K_α and K_β X-ray peaks (red arrows): F_C about 0.7
- Escape peaks (blue arrows) – observed up to higher energies (e.g. 457 keV (peak 59), escape of Ge K_α X-ray in the event of simultaneous absorption of 356 and 81 keV gamma photons and 30.85 keV K_α X-ray)
- The other peaks – mostly due to summing events including at least one X-ray

Coincidence with annihilation photons

^{22}Na measured in well-type detector



Source: Sima and Arnold, ARI 53 (2000) 51

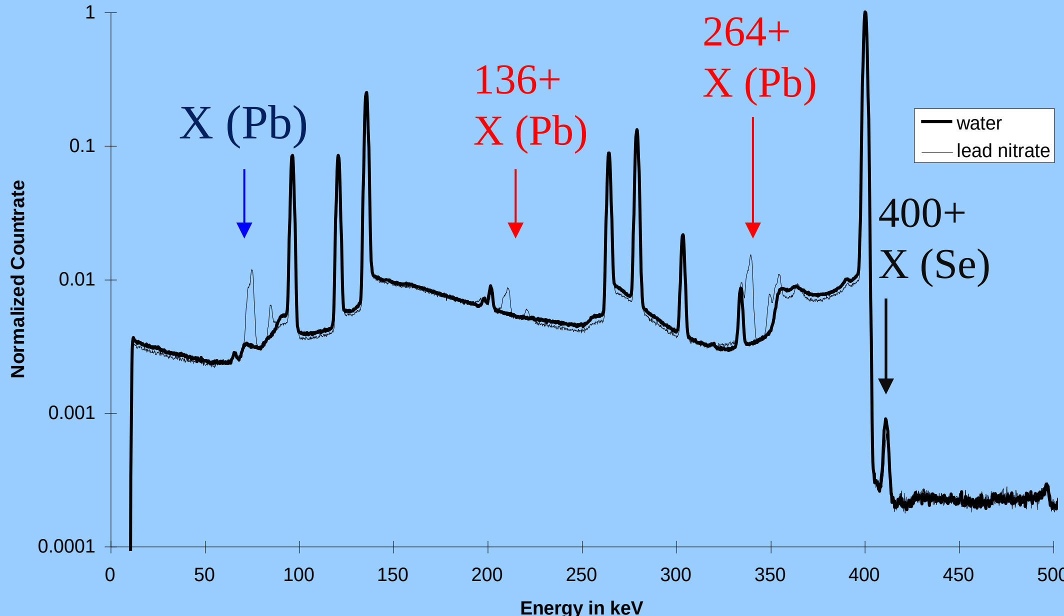


Source: Monographie BIPM-5 Table of Radionuclides Vol. 5 (2010)

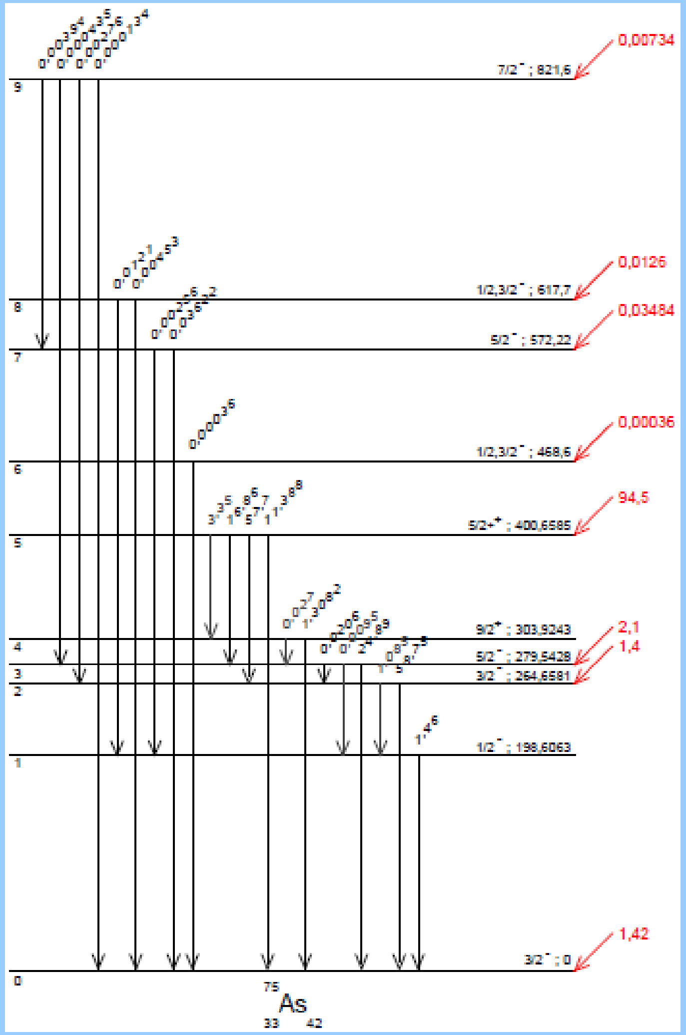
- F_C for main peaks: 0.17 for 1275 keV, 0.12 for 511 keV
- The other peaks due completely to coincidence-summing effects
- Triple coincidences very important:
  - the peak at 2297 keV results from summing of the 1275 keV photon and two annihilation photons (511 keV each)
  - Significant contribution to F_C for the other peaks

Coincidence with X-rays excited in the matrix of the source

^{75}Se measured in lead nitrate versus water in well-type detector



Source: Arnold and Sima, ARI 52 (2000) 725



Source: Monographie BIPM-5 Table of Radionuclides Vol. 5 (2010)

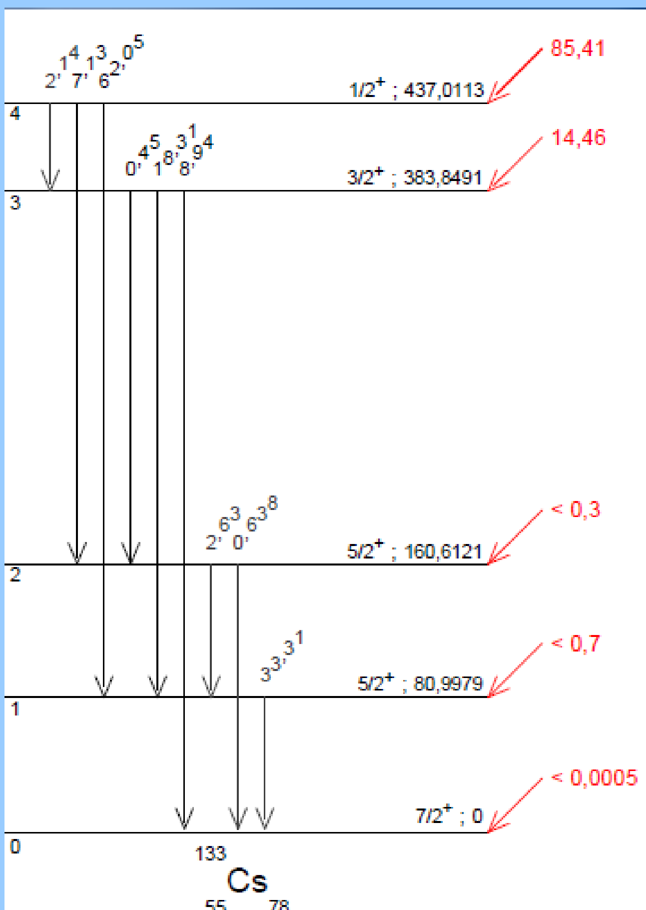
- In $Pb(NO_3)_2$ additional peaks with respect to the water sample
- Pb X-rays (blue arrow) excited by the γ rays of ^{75}Se
- Sum peaks of γ rays emitted by ^{75}Se and X-rays emitted by Pb (red arrows):
  - In the cascade 136 + 264 keV sum peaks at 339=264+K_α(Pb) (Pb excited by 136 keV) and smaller at 210=136+K_α(Pb) keV (Pb excited by 264)
  - Smaller effects in the 121+279 keV cascade

Intuitive calculation of coincidence-summing effects – point source

Example: the 302 keV photon emitted by ^{133}Ba (transition from level 3 to level 1)

In the absence of coincidence summing the peak count rate is:

$$R(302) = \varepsilon(302)\, P_{\gamma}(302)\, A$$



Source: Monographie BIPM-5 Table of Radionuclides Vol. 5 (2010)

But the 302 keV photon is always emitted together with other radiations, in several distinct decay cascades:

Cascades starting by decay on level 4:

1: K_{α}(EC4)+53(4=>3)+302(3=>1)+81(1=>0); probability p_1

2: K_{α}(EC4)+53(4=>3)+302(3=>1)+K_{α}(1=>0); p_2

3: K_{α}(EC4)+53(4=>3)+302(3=>1)+K_{β}(1=>0); p_3

4: K_{α}(EC4)+53(4=>3)+302(3=>1)+other(1=>0); p_4

5: K_{α}(EC4)+K_{α}(4=>3)+302(3=>1)+81(1=>0); p_5 …..

- Other cascades start on level 3, e.g.
  K_{α}(EC3) +302(3=>1)+81(1=>0)

$\Rightarrow$ in total 60 distinct cascades

Notations:

K_{α}(EC4)=K_{α} X-ray emitted after electron capture decay on level 4;

53(4=>3)=53 keV γ photon emitted in the transition 4=>3;

K_{α}(1=>0)= K_{α} X-ray emitted after conversion electron de-excitation in the transition 1=>0;

other(1=>0)=Non-detected radiations (L X-rays, Auger electrons) in the transition 1=>0

Direct contribution of the 302 keV photon to the peak count rate

From the point of view of the count rate in the 302 keV peak, two cases concerning the interaction in the detector of the radiations emitted in any of the 60 decay cascades:

- 302 keV photon is fully absorbed in the detector, and one or several radiations emitted in the cascade deposit whatever energy in the detector
  - ⇒ No contribution to the peak count rate (losses from the peak in comparison with the absence of coincidence-summing effects)
- 302 keV photon is fully absorbed in the detector, and no energy is deposited in the detector by any of the other radiations emitted in the cascade
  - => Contribution to the peak count rate
- The probability of contribution to the peak count rate is specific to each cascade
  - Probability of the cascade p_i
  - Probability of full energy absorption of 302 keV photon $\varepsilon(302)$
  - Probability of no energy deposition of other radiations $1-\eta(E)$

⇒ Contribution to the count rate in the 302 keV peak along the decay path 1

1: K_α(EC4) + 53(4=>3) + 302(3=>1) + 81(1=>0)

=> $R_1=p_1\cdot[1-\eta(K_\alpha)]\cdot[1-\eta(53)]\cdot\varepsilon(302)\cdot[1-\eta(81)]\cdot A$

$[1-\eta(K_\alpha)]$=probability that the K_α photon *does not deposit any energy* in the detector

- Similar contributions R_2, R_3, … R_{60} of all the other decay paths to the count rate in the 302 keV peak.

Sum peak effects

- Transition 3=>1 of 302 keV same energy as transitions 3=>2 (223 keV) and 2=>1 (79 keV)

$\Rightarrow$ Sum peak contribution of 223+79 keV to the 302 keV peak

$\Rightarrow$ Contributions along several distinct decay paths:

S1: K_α(EC4)+53(4=>3)+223(3=>2)+79(2=>1)+81(1=>0); probability p_{S1}

S2: K_α(EC4)+53(4=>3)+223(3=>2)+79(2=>1)+K_α(1=>0); p_{S2} ….

and other 58 decay paths (obtained from cascades 1, 2, … 60 by replacing 302(3=>1) with 223(3=>2)+79(2=>1))

- Rate of events corresponding to full energy absorption of 223 and 79 keV photons when other radiations *do not deposit any energy* in the detector on decay path S1:

$$R_{S1}=p_{S1}\cdot[1-\eta(K_\alpha)]\cdot[1-\eta(53)]\cdot\varepsilon(223)\cdot\varepsilon(79)\cdot[1-\eta(81)]\cdot A$$

$\Rightarrow$ These events add counts in the 302 keV peak

- Similar contributions on all the other paths

Final count rate in the 302 keV peak:

$$R(302)=R_1+R_2+\ldots+R_{60}+R_{S1}+R_{S2}+\ldots+R_{S60}$$

In the absence of coincidence summing:

$$R_0(302)= \varepsilon(302)\ P_\gamma(302)\ A$$

$\Rightarrow$ *Coincidence-summing correction factor:*

$$F_C(302)=R(302)/R_0(302)$$

Final expressions in the case of point sources:

$$F_C(E_i;X)=1-\sum_{j}\frac{p_{ij}}{p_i}\cdot\eta(E_j)+\sum_{j,k}\frac{p_{ijk}}{p_i}\cdot\eta(E_j)\cdot\eta(E_k)-\ldots$$

$$+\sum_{p,q}\frac{p_{pq}}{p_i}\cdot\frac{\varepsilon(E_p)\cdot\varepsilon(E_q)}{\varepsilon(E_i)}-\sum_{p,q,r}\frac{p_{pqr}}{p_i}\cdot\frac{\varepsilon(E_p)\cdot\varepsilon(E_q)\cdot\eta(E_r)}{\varepsilon(E_i)}+\ldots$$

p_i=probability of emission of radiation *i* per decay

p_{ij}=probability of emission of the pair of radiations *i* and *j* per decay

p_{ijk}=probability of emission of the triplet of radiations *i*, *j* and *k* per decay

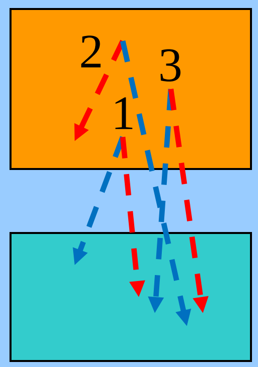
$\varepsilon(E)$=the full energy peak efficiency for energy E

$\eta(E)$=the total efficiency for energy E

- The correction terms in the first line (the sums) describe coincidence losses from the peak of energy E_i, due to simultaneous detection of radiation E_j (first sum) and E_j and E_k (second sum) and so on;
- The second line describes sum peak contributions to the peak of energy E_i, with $E_p+E_q=E_i$; the second term describes losses from the sum peak contribution due to the simultaneous detection of radiation E_r
- The correction factor depends on the nuclide X through p_i, p_{ij}, … and energies
- In the case of a point source ε and η are the efficiencies for the complete source, that can be directly measured

Extended sources

- Efficiencies depend on the emission point:



$\varepsilon^p(E,\vec{r})$=point source FEP efficiency, i.e. the probability that a photon emitted from the point $\vec{r}$ from the source produces a count in the peak of energy E;

$\eta^p(E,\vec{r})$=point source total efficiency, i.e. the probability that a photon emitted from a point $\vec{r}$ from the source produces a count in the spectrum

$$\varepsilon(E) = \frac{1}{V}\int_V \varepsilon^p(E,\vec{r})dV \qquad \eta(E) = \frac{1}{V}\int_V \eta^p(E,\vec{r})dV$$

$\varepsilon(E)$ and $\eta(E)$ represent the efficiencies for the complete source of volume V (averages of ε^p and η^p over the volume of the source)

Coincidence-summing effects:

- All efficiencies are affected simultaneously by the position of the emission point
- In the absence of coincidence effects, emission points 1 and 3 contribute to the peak of the “red” photon; emission point 2 does not contribute (photon absorption in the source)

⇒ Coincidence losses from the peak of the “red” photon due to the detection of the “blue” photon may occur in the case of emissions from the points 1 and 3, but not from point 2; however point 2 contributes to the total efficiency for the “blue” photon

⇒ Contrary to point sources, losses from the “red” peak **are not proportional** with the total efficiency η for the “blue” photon (η the total efficiency for the complete source)

Extended sources:

⇒ Products of efficiencies should be replaced by suitable integrals (Debertin and Schötzig, NIM 158 (1979) 471), for example:

$$\varepsilon(E_i)\cdot\varepsilon(E_j) \Rightarrow \frac{1}{V}\int_V \varepsilon^p(E_i,\vec{r})\cdot\varepsilon^p(E_j,\vec{r})dV$$

$$\varepsilon(E_i)\cdot\eta(E_j) \Rightarrow \frac{1}{V}\int_V \varepsilon^p(E_i,\vec{r})\cdot\eta^p(E_j,\vec{r})dV$$

$$\varepsilon(E_i)\cdot\eta(E_j)\cdot\eta(E_k) \Rightarrow \frac{1}{V}\int_V \varepsilon^p(E_i,\vec{r})\cdot\eta^p(E_j,\vec{r})\cdot\eta^p(E_k,\vec{r})dV$$

Coincidence-summing correction factors in the case of extended sources:

$$F_C(E_i;X) =$$

$$1-\sum_j \frac{p_{ij}}{p_i}\cdot\frac{\int_V \varepsilon^p(E_i,\vec{r})\eta^p(E_j,\vec{r})dV}{\int_V \varepsilon^p(E_i,\vec{r})dV}+\sum_{j,k}\frac{p_{ijk}}{p_i}\cdot\frac{\int_V \varepsilon^p(E_i,\vec{r})\eta^p(E_j,\vec{r})\eta^p(E_k,\vec{r})dV}{\int_V \varepsilon^p(E_i,\vec{r})dV}-..$$

$$+\sum_{p,q}\frac{p_{pq}}{p_i}\cdot\frac{\int_V \varepsilon^p(E_p,\vec{r})\varepsilon^p(E_q,\vec{r})dV}{\int_V \varepsilon^p(E_i,\vec{r})dV}-\sum_{p,q,r}\frac{p_{pqr}}{p_i}\cdot\frac{\int_V \varepsilon^p(E_p,\vec{r})\varepsilon^p(E_q,\vec{r})\eta^p(E_r,\vec{r})dV}{\int_V \varepsilon^p(E_i,\vec{r})dV}..$$

Complications with respect to the point source equation:

- Integrals of products of efficiencies do not have an experimental counterpart, whereas the efficiencies required in the case of point source calculation can be obtained experimentally
- The integrals are specific to each pair of photons (or higher multiplicity of photons): the coincidence losses due to the same photon of energy E_j from the peak of energy E_1 ($\sim \int_V \varepsilon(E_1,\vec{r})\eta(E_j,\vec{r})dV$) differ from the losses from the peak of energy E_2 ($\sim \int_V \varepsilon(E_2,\vec{r})\eta(E_j,\vec{r})dV$), contrary to the case of point sources (Sima and Arnold, ARI 53 (2000) 51)

Simplification: Quasi-point source approximation:

$\varepsilon^p(E,\vec{r}) \cong \varepsilon(E)$ for any $\vec{r}$ in the volume V of the source

$\eta^p(E,\vec{r}) \cong \eta(E)$ for any $\vec{r}$ in the volume V of the source

Validity:

- Small sources: the solid angle subtended by the detector from any point in the source approximately the same
- Low attenuation: the same attenuation (or negligible attenuation) in the sample for the photons emitted from any point within the source

Caution:

- Coincidence summing effects for a volume source **higher** than the effects evaluated on the basis of the quasi-point source approximation:

$$\frac{1}{V}\int_V \varepsilon(E_i,\vec{r})\varepsilon\left(E_j,\vec{r}\right)dV \geq \varepsilon(E_i)\varepsilon(E_j)$$

$$\frac{1}{V}\int_V \varepsilon(E_i,\vec{r})\eta\left(E_j,\vec{r}\right)dV \geq \varepsilon(E_i)\eta(E_j)$$

- For coincidence losses from the peak of energy E_i due to the detection of photon of energy E_j, an effective total efficiency $\eta^{eff}(E_j,E_i)$ is required instead of $\eta(E_j)$:

$$\eta^{eff}(E_j,E_i) = \frac{\int_V \varepsilon^p(E_i,\vec{r})\eta^p(E_j,\vec{r})dV}{\int_V \varepsilon^p(E_i,\vec{r})dV}$$

$$\eta(E_j) = \frac{1}{V}\int_V \eta^p(E_j,\vec{r})dV$$

- $\eta^{eff}>\eta$ because the weighting factor before η^p in the equation for η^{eff} increases the contributions of emission points close to the detector, where η^p is higher; both solid angle weighting and self-attenuation weighting contribute

⇒**Effective total efficiency always higher than common total efficiency**

⇒**The differences increase for lower E_i energies**

Example:

1000 cm^3 Marinelli beaker measured with a 50% relative efficiency HPGe

(Arnold and Sima, J. Radioanal. Nucl. Chem. 248 (2001) 365)

For E_i=50 keV the effective total efficiency for a water sample higher by 44% to 26% than the common total efficiency when E_j varies from 50 to 2000 keV

For E_i=1000 keV the same differences are by 25% to 16%

Solid angle weighting and self-attenuation weighting have roughly equal contributions

Higher differences are obtained for the term corresponding to sum peak contributions

Observation – Angular correlations

- In a cascade γ_i-γ_j, if γ_i is emitted in a given direction $\vec{n}_i$, the direction of emission $\vec{n}_j$ of the γ_j photon is generally not uniformly distributed
- The probability of the second photon to be emitted at an angle θ with respect to the direction of the first is given by the angular correlation function w(θ), depending on the spins and parities of the levels involved in transitions and on the multipole mixing ratios.
- Changes required in equations:
  - Point source:

$$\varepsilon(E_i)\varepsilon(E_j) \Rightarrow \frac{1}{(4\pi)^2}\int \varepsilon(E_i,\vec{n}_i)\varepsilon(E_j,\vec{n}_j)w(\theta(\vec{n}_i,\vec{n}_j))d\Omega_i d\,\Omega_j$$

θ = the angle between the directions $\vec{n}_i$ and $\vec{n}_j$, $d\Omega_i$ elementary solid angle around the $\vec{n}_i$ direction

$\varepsilon(E_i,\vec{n}_i)$= probability for a photon of energy E_i emitted in the direction $\vec{n}_i$ to be registered in the peak

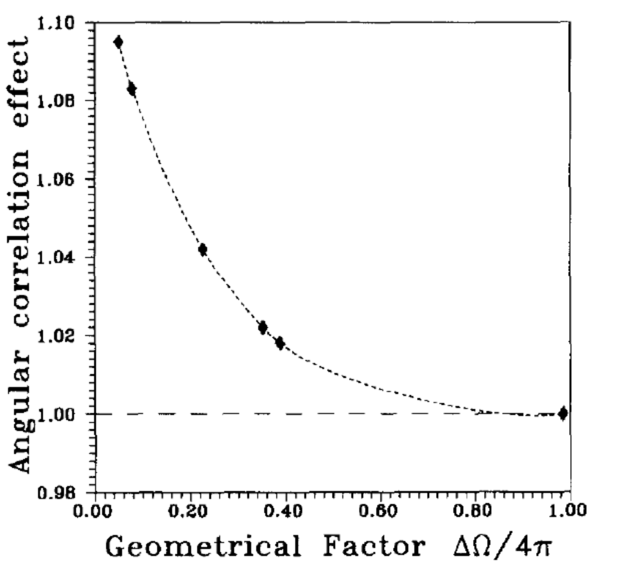
  - Volume source:
  - $\int \varepsilon^p(E_i,\vec{r})\,\varepsilon^p(E_j,\vec{r})dV \Rightarrow$

$$\frac{1}{(4\pi)^2}\int \varepsilon^p(E_i,\vec{r},\vec{n}_i)\varepsilon^p(E_j,\vec{r},\vec{n}_j)w(\theta(\vec{n}_i,\vec{n}_j))d\Omega_i d\,\Omega_j dV$$

  - $\varepsilon^p(E_i,\vec{r},\vec{n}_i)$ = probability for a photon of energy E_i emitted from the point $\vec{r}$ in the direction $\vec{n}_i$ to be registered in the peak

- Angular correlations are attenuated when the solid angle increases
  $\Rightarrow$ Small effects in the case of well-type detector measurements
- Angular correlation corrections are higher in the case of pure sum peaks

Sum peak of 2505 keV (1173+1332 keV) of ^{60}Co



Source: Sima, ARI 47 (1996) 919

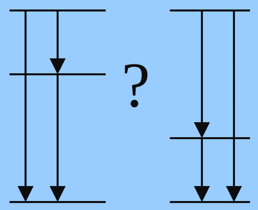
- Evaluation of angular correlation effect in the measurement of environmental samples: Roteta et al, NIMA 369 (1996) 665

3. Decay data

In the absence of coincidence summing the count rate in the peak of energy E depends on a single parameter of the decay scheme, $P_\gamma(E)$

- the uncertainty of the activity computing using $R(E)= \varepsilon(E)\, P_\gamma(E)\, A$ depends only on the uncertainty of this single parameter of the decay scheme
- $P_\gamma(E)$ can be measured relatively simply
- standardized values should be used for compatibility

In the presence of coincidence summing it is not sufficient to know $P_\gamma(E)$



- it is not sufficient to know all $P_\gamma(E_i)$ for all the emitted photons
- the complete decay scheme is required: p_i, p_{ij}, … angular correlation functions
- R(E) depends simultaneously on many parameters of the decay sheme => for the evaluation of the uncertainty of A the complete covariance matrix of the decay scheme parameters is required!

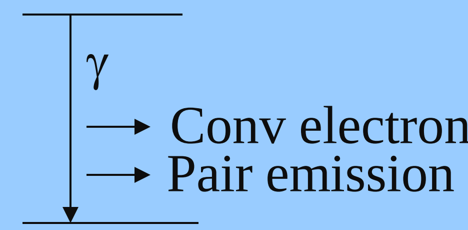
The preparation of the decay scheme of a nuclide => difficult

- only simple gamma spectra not sufficient
- combined measurements (X rays, conversion electrons, decay particles α, β, coincidence gating etc)

Definitions (Introduction –Table de radionucleides Note Technique LNHB 2011/53)

Gamma transition – total probability $P_g = P_\gamma + P_{ce} + P_{e+e-}$

Total probability=probability for γ emission + probability for conversion electron + probability of electron-positron pair emission



Conversion coefficient:

$$\alpha_t = \alpha_K + \alpha_L + \alpha_M + \ldots = P_{ce} / P_\gamma$$

(conversion on K, L, M, … atomic shells);

Internal pair conversion coefficient:

α_π relative emission probability of the pair (10^{-3} - 10^{-4})

Gamma emission probability in function of transition probability:

$$P_\gamma = P_g / (1+\alpha_t)$$

Conversion electron emission probability

$$P_{ce} = \alpha_t P_g / (1+\alpha_t)$$

Conversion electron emission from K atomic shell

$$P_{ceK} = \alpha_K P_g / (1+\alpha_t)$$

X-ray emission after the creation of a vacancy on the K shell:

- processes: emission of X-ray and emission of Auger electrons
    - Fluorescence yield ω_K=X-ray emission probability when the vacancy is filled
    - Auger emission probability $P_{Ak} = 1 - \omega_K$
- similar processes after creation of a vacancy on the L shell (or subshells)

Probability of a K X-ray emission in a de-excitation transition

$$P_{XK} = \omega_K \alpha_K P_g / (1+\alpha_t)$$

Probability of a K X-ray emission in a EC (electron capture) decay on the j-th level of the daughter:

$$P_{XK} = \omega_K P_{\varepsilon j} P_K$$

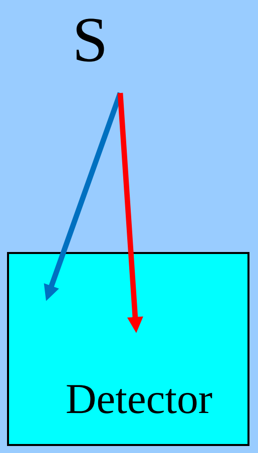
P_K – probability of electron capture from K shell if the electron capture transition was on the j-th level of the daughter nucleus (probability $P_{\varepsilon j}$)

International Committee on Radionuclide Metrology (ICRM):
Recommendations for the development of a consistent set of decay data

ICRM Recommendation for decay data:
- Use data from the Decay Data Evaluation Project (DDEP):
http://www.nucleide.org/DDEP_WG/DDEPdata.htm
- Careful and dedicated evaluation of data
- Periodic updates
- Monographie BIPM-5 Table of Radionuclides Vol. 1-8 (2004-2016)

For nuclides not included in the Monographie BIPM-5:
- BNL, ENSDF data libraries
- Nuclear Data Sheets

4. Efficiencies



Quasi-point source approximation:

Example – coincidence summing for photons E_i and E_j

- probability of completely absorbing both photons in the detector per one decay = probability of a count in the sum peak per decay:

$$p_{ij}\ \varepsilon(E_i)\ \varepsilon(E_j)$$

(angular correlation neglected)

- probability of completely absorbing E_i and incompletely absorbing E_j per decay = probability of losing a count from the peak of energy E_j per decay:

$$p_{ij}\ \varepsilon(E_i)\ \eta(E_j)$$

Peak and total efficiencies for the complete source are required for all the radiations emitted along all the decay paths of interest

Measurement of FEP efficiency ε – routinely done

Measurement of the total efficiency η – difficult

- sources emitting a single radiation required
- problems with background subtraction
- usually a low energy threshold, spectrum extrapolation to E=0 required
- preferably evaluation of the ratio P/T of the FEPE to the total efficiency

(Semkow et al., NIMA 290 (1990) 437; Korun and Martincic, NIMA 385 (1997) 511; Lépy, NIMA 579 (2007) 284)

- knowledge of the activity of the source not required
- weak dependence on the position of the emission point
- weaker dependence on energy than each of the efficiencies – better approximated

Computation of η or better P/T

- Monte Carlo
  - -problem: the effect of the dead layer – partially active:
    Arnold and Sima, ARI 60 (2004) 167
    Dryak et al., ARI 68 (2010) 1451
- Simplified procedures – De Felice et al., ARI 52 (2000) 745

Extended sources

- Integrals of products of efficiencies are required

Realistic computations – by Monte Carlo simulation (Décombaz et al., NIMA 312 (1992) 152; Sima and Arnold, ARI 53 (2000) 51; García-Talavera et al., ARI 54 (2001) 769; Berlizov, ACS Symp. Series 945 (2006) 183; Johnston et al., ARI 64 (2006) 1323)

- Correlated transport of the radiations emitted on a decay path
- Simultaneous evaluation of the ideal count rate in the peaks and of the real, coincidence-summing affected, count rate
- Variance reduction techniques can be implemented to improve the computation speed; e.g. for coincidence losses from the peak of a main photon (Sima et al., JRNC 248 (2001) 359):
  - Focused emission, attenuation approximation, forced first collision in the detector for the main photon;
  - Stop the simulation when the first interaction occurs in the detector for the accompanying photons

Calculations based on approximations:

a Decomposition of the volume in quasi-point source domains:

- Measurement: map of the point source efficiencies in the sample – tedious (Debertin and Schötzig, NIM 158 (1979) 471)
- Computations: De Corte et al., NIMA 353 (1994) 539; Kolotov et al., JRNC 210 (1996) 83; Dryak et al., ARI 70 (2012) 2130; Dias et al., 134 (2018) 205

b Numerical integration with efficiencies evaluated using approximations:

- Effective solid angle (Piton et al., ARI 52 (2000) 791; Lépy et al., 70 (2012) 2137)
- Virtual point detector (Rizzo and Tomarchio, ARI 68 (2010) 1448)
- other prescriptions (Korun and Martinčič, NIMA 355 (1995) 600; Kolotov and Koskelo, JRNC 233 (1998) 95; De Felice et al., ARI 52 (2000) 745)

c Third efficiency (LS) curve (Blaauw and Gelsema, NIMA 505 (2003) 311; Vidmar and Korun, NIMA 556 (2006) 543):

$$\int_V \varepsilon^p(E_i,\vec{r})\eta^p(E_j,\vec{r})dV \approx \sqrt{\int_V [\varepsilon^p(E_i,\vec{r})]^2 dV} \cdot \sqrt{\int_V [\eta^p(E_j,\vec{r})]^2 dV} = l(E_i)\,\varepsilon(E_i) l(E_j)\eta(E_j)$$

$$\int_V \varepsilon^p(E_p,\vec{r})\varepsilon^p(E_q,\vec{r})dV \approx \sqrt{\int_V [\varepsilon^p(E_p,\vec{r})]^2 dV} \cdot \sqrt{\int_V [\varepsilon^p(E_q,\vec{r})]^2 dV} = l(E_p)\varepsilon(E_p) l(E_q)\varepsilon(E_q)$$

Where $l^2(E) = \dfrac{\int_V [\varepsilon^p(E,\vec{r})]^2 dV}{\left[\int_V \varepsilon^p(E,\vec{r})dV\right]^2}$

$\Rightarrow$ Only 3 functions, detector and sample specific, are required for efficiency and coincidence-summing calculations: $\varepsilon(E)$, $\eta(E)$, $l(E)$

- $l(E)$ obtained from experimental spectrum in the presence of coincidence effects – Blaauw, or
- $l(E)$ computed by Monte Carlo using the effective solid angle method – Vidmar

Well-type detectors:

- small volume samples, big solid angle
- very high coincidence summing effects (Sima and Arnold, 47 (1996) 889; Blaauw, NIMA 419 (1998) 146; Wang et al., NIMA 425 (1999) 504; Laborie et al., ARI 53 (2000) 57; Jäderström et al., NIMA 784 (2015) 264; Britton and Davies, NIMA 786 (2015) 12)

=> Small volume: effective total efficiency close to the total efficiency

=> Useful analytical approximation for the total efficiency (Sima, NIMA 450 (2000) 98; refined by Pomme, NIMA 604 (2009) 584)

5. Methods for evaluation of coincidence summing corrections

- Coincidence summing correction factors: combination of
  - decay data parameters (p_i, p_{ij}, … or equivalent), and
  - efficiencies ε, η and products of efficiencies $\varepsilon_i\eta_j$, $\varepsilon_i\eta_j\eta_k$, … (point or quasi-point sources) or integrals of products of efficiencies $\int \varepsilon_i\eta_j dV$, $\int \varepsilon_i\eta_j\eta_k dV$, … (extended sources)
- Methods of evaluation:
  - How are the decay data prepared?
    - Deterministic methods, implicit or explicit evaluation of p_i, p_{ij}, …
    - Stochastic simulation of the decay processes
  - How are the efficiencies evaluated?
    - Experimental values (point and quasi-point sources)
    - Computed values (especially by Monte Carlo), including or not the correlations between factors (volume source integration of products of efficiencies, angular correlations)
  - How are the decay data and the efficiencies combined?
    - Complex coupling of decay data and of efficiencies
    - Evaluated independently, coupled in the final expressions

Deterministic description of the decay, with complex coupling of decay data and efficiencies

- First general methods proposed, compact formulation
- Probabilities of groups of photons p_i, p_{ij}, … not explicitly computed, intimately coupled with efficiencies
- Suitable for point sources and quasi-point source approximation
- Not appropriate for volume sources, products of efficiencies are included, not integrals of products of efficiencies
    - can be extended to volume sources by:
        - Implementation of the LS curve approximation
        - Decomposition of the volume in small domains and application of the quasi-point source approximation followed by suitable averaging

1. Recursive formulae (Andreev type; Andreev et al., Instr. Exp. Tech. 15 (1972) 1358)

Detector insensitive to X rays

$x_t(i,j)$ transition probability from level i to level j if level i is already populated

$$\sum_{j=1}^{i-1} x_t(i, j) = 1$$

$$a(i,j)=\frac{x_t(i,j)\cdot\varepsilon(i,j)}{1+\alpha_t(i,j)}$$ Probability of complete energy absorption in the transition if the initial level is already populated:

$$b(i,j)=x_t(i,j)\cdot\left[1-\frac{\eta(i,j)}{1+\alpha_t(i,j)}\right]$$ Probability of no signal in the detector following the transition if the initial level is already populated

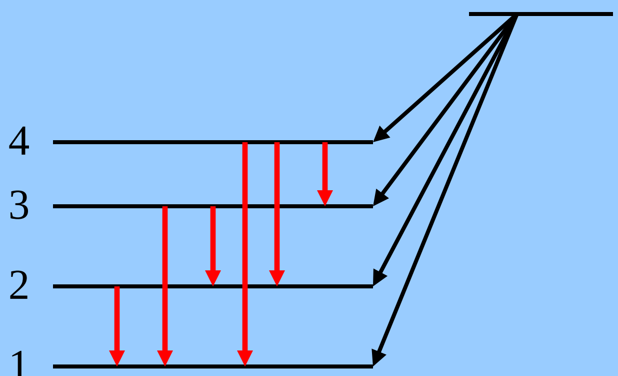
F(i) – probability of decay on level i

N(i) – probability of populating level i on any path without having any signal in the detector

M(k) – probability of any transitions from level k to the ground state without having any signal in the detector on these transitions (k populated)

A(i,k) – probability that the total energy of the transition from level i already populated to level k, in any possible sequences, is completely absorbed in the detector, i>k

$$N(i)=F(i)+\sum_{m=i+1}^{n}N(m)\cdot b(m,i),\quad N(n)=F(n)$$



$$M(k)=\sum_{l=1}^{k-1}b(k,l)\cdot M(l),\quad k=2,n;\quad M(1)=1$$

$$A(i,k)=a(i,k)+\sum_{j=k+1}^{i-1}a(i,j)\cdot A(j,k),\quad i=2,n;\ j=1,(i-1)$$

Probability of detecting the total energy in the transition from level i to level j per one decay is:

$$S(i,k) = N(i) \cdot A(i,k) \cdot M(k)$$

Extensions of the procedure and programs: Mc Callum and Coote, NIM 130 (1975) 189 – program; Debertin and Schötzig, NIM 158 (1979) 471 – inclusion of X rays, nuclide decay data (KORDATEN), program KORSUM; Morel et al., IJARI 34 (1983) 1115 – volume sources by transfer method, program CORCO; Jutier et al., NIMA 580 (2007) 1344 - inclusion of internal pair production

- Specific advantages:
  - Rigorous procedure for point sources, relatively simple programming
  - Fast computation if efficiencies are given in the input
- Specific disadvantages:
  - Complex coupling of the decay data with efficiencies – too long time if Monte Carlo simulation is applied for efficiency evaluation in the case of volume sources (evaluation of efficiencies and application of the recursive relations should be repeated for each emission point)
  - Does not account for sum peaks corresponding to non-connected transitions

2. Matrix formalism (Semkow et al., NIMA 290 (1990) 437)

- Idea: $a(i,j)$ considered the (i,j) element of a triangular matrix a; $b(i,j)$ similar
- Probability of transition from i to j in two successive transitions i to k and k to j with total energy absorption in the detector is $a(i,k)\,a(k,j)$. Probability of transition from i to j in two successive transitions for any k, i>k>j is:

$$\sum_{k=j+1}^{i-1} a(i,k)\cdot a(k,j) = (a\cdot a)(i,j) = (a^2)(i,j)$$

- Probability of transition between the same initial and final level by three successive transitions with total energy absorption in the detector is given by matrix a^3 and so on.
- Probability of the transition from any initial to any final level with all possible sequences of connected transitions and with the condition that the total energy is absorbed in the detector is given by a new matrix A:

$$A = a + a^2 + a^3 + \ldots + a^n$$

- Similarly the transitions from one level to another without any energy deposition is given by a matrix

$$B = 1 + b + b^2 + b^3 + \ldots + b^n$$

⇒ the matrix of the probability of detecting the complete energy in any possible transitions per one decay is given by:

$$S = N\,A\,M, \text{ where } N=\mathrm{diag}[(F\,B)_i] \text{ and } M=\mathrm{diag}(B_{i,1})$$

Extensions: Korun and Martinčič, NIMA 325 (1993) 478 – inclusion of X-rays
Vidmar and Korun, NIMA 556 (2006) 543 – inclusion of the LS curve, application to volume sources (EFFTRAN)

- Advantages in comparison with Andreev's procedure:
  - More convenient mathematical computation
  - Evaluation of the complete matrix of total energy deposition
- Disadvantages: similar with Andreev's procedure

3. Symbolic list manipulation – energy deposition (Novković et al., NIMA 578 (2007) 207)

- All decay paths are analyzed and energy deposition evaluated along each path by symbolic list manipulation
- Specific advantages:
  - Coincidence summing effects for all peaks (including all sum peaks, also with X-rays) can be evaluated
- Specific disadvantage:
  - In the case of complex decay schemes, many decay paths can contribute to the same energy – they should be grouped together by energy (attention to energy uncertainty in the decay data tables!)

Deterministic description of the decay, decoupling decay data and efficiencies

- Independent evaluation of the decay data and of efficiencies
  - Probabilities p_i, p_{ij}, … depend on decay scheme (on nuclide), but not on experimental setup – advantageous to evaluate them separately
  - Can be coupled with efficiencies obtained using various procedures:
    - Experimental values or values obtained using the transfer method (point and quasi-point sources, negligible angular correlations)
    - Monte Carlo or transfer method applied to each quasi-point source domain defined in the volume source (negligible angular correlations)
    - Full Monte Carlo for extended sources, including or not angular correlations
- Analytical evaluation of joint emission probabilities:
  - Faster than repeated Monte Carlo simulation of the decay scheme
  - Uncertainty of the results smaller in comparison with that obtained when the decay is simulated by Monte Carlo (statistical uncertainty due to random simulation of the decay avoided)
    - Especially important for low probability transitions
- Flexible and fast procedure

Deterministic calculation of joint emission probabilities

- First tables - Schima and Hopes, IJARI 34 (1983) 1109 – only pair coincidences
- General procedure - Sima and Arnold, ARI 66 (2008) 705

  => Efficient procedure of finding all the possible decay paths, based on graph theory methods

  - any decay scheme with less than 100 levels,
  - all coincidence orders (pair, triple, …) included
  - metastable states included
  - sum peaks with X-rays (two groups K_α and K_β) up to 10 X rays contributions included
  - contribution of positron annihilation included
  - identification of the transitions contributing to any peak based on transition levels, not grouping by energy
  - simple possibility to include angular correlation

  $\Rightarrow$ Implemented in GESPECOR

  - Full volume source simulation for obtaining the average values of the required products of efficiencies ($\int \varepsilon_i \eta_j dV$, $\int \varepsilon_i \eta_j \eta_k dV$, …)

Test of compatibility of the deterministic algorithms for the description of the decay – Kanisch et al., ARI 67 (2009) 1952

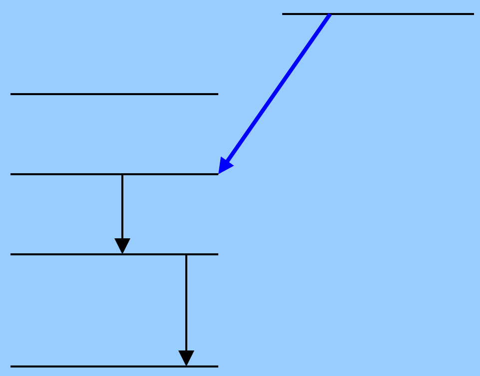
- Test based on the comparison of the coincidence summing correction factors for a point source, with given values of the efficiencies
- The algorithms of Andreev, Semkow, Sima and Arnold, Novković, and Vidmar and Kanisch were applied in the case of ideal decay schemes (perfectly balanced) and given values of efficiencies
- Results:
    - The algorithms of Sima and Arnold, Novković, and Vidmar and Kanisch are equivalent
    - The algorithms of Andreev and Semkow type are equivalent with the others except for the fact that they do not predict sum peaks for non-linked transitions and sum peaks with X-rays contributions

Monte Carlo simulation of the decay coupled with simulation of efficiencies

- Simulation of all processes from nuclide decay to energy deposition in the detector
- Natural coupling of decay and radiation transport
- All relevant radiations emitted can be included in simulation
    - Decay process:
        - Beta decay (β^-, $\beta+$): sample beta radiations (energy from spectrum, direction)
        - Electron capture decay: sample X-rays, Auger electrons (energy, direction)
    - De-excitation process:
        - Gamma photons: sample energy and direction
        - Conversion electrons: sample energy and direction, sample X-rays and Auger electrons (energy, direction)
        - Pair conversion: sample energy and directions of electron and positron (rarely needed)

Basics of simulation:

Simulation of the decay:



- Sample randomly the emission point (extended sources)
- Sample decay type
- Sample the level of the daughter nuclide using the branching ratios
- Sample the parameters of the radiations emitted (beta particles, or X ray emission and/or Auger electron emission in EC decays – atomic relaxation)
- Save the parameters of the relevant radiations

Simulation of de-excitation of the nucleus:

- Sample the final level of de-excitation transition using the transition probabilities
- Sample radiations emitted in the transition (γ, conversion electron and radiations emitted in atom relaxation, pair emission)
- Save the parameters of the emitted radiations
- Repeat sampling of the transitions until the final level is stable

Simulation of radiation transport for one decay

- Extract one radiation from particle bank
  - Follow the history of the particle and of all secondary radiations generated, until absorption or escape from the "world"
  - Evaluate the energy deposited in the detector
- Repeat until no particle remains in the bank: extract particle, transport it and evaluate the energy deposited by it and the secondary particles produced

Repeat N times (N = big number) the simulation of the decay, de-excitation and transport

Analyze and summarize the results

=> Normalized number of events in the peaks, in the presence of coincidence-summing effects

=> Coincidence-summing correction factors F_C: ratio of the normalized number of events in the peak to the normalized number of events in the same peak in the absence of coincidence summing (evaluated separately)

Procedures implemented in various general simulation codes:

- sch2for in GEANT3 - Décombaz and Laedermann, NIMA 369 (1996) 375
- Radioactive Decay Module in GEANT 4 - Agostinelli et al., NIMA 506 (2003) 250
- PENNUC in PENELOPE - García-Toraño et al., NIMB 396 (2017) 43

Applications – simulations of spectra, evaluation of efficiencies, computation of F_C, etc:

- Hurtado et al., IEEE TNS 56 (2009)1531 – includes study of sensitivity to Auger electrons
- Capogni et al., ARI 64 (2010) 1428 – comparison between sch2for and G4RadioactiveDecay
- Liu et al., ARI 137 (2018) 210 – GEANT 4 – includes emission times, random coincidences

- Angular correlations usually neglected

6. Uncertainties

- F_C depends on:
  - Efficiencies (ε, η, products of efficiencies or integrals of products of efficiencies)
  - Joint emission probabilities of groups of radiations (p_i, p_{ij}, …)
- Uncertainty components due to uncertainties of
  - Efficiencies
  - Parameters of the decay data

  Sima and Arnold, ARI 53 (2000) 51

- Uncertainties of the efficiencies:
  - Case of point and quasi-point sources and use of experimental efficiencies:
    - ⇒ Experimental uncertainties of ε, η; uncertainties required also for products of efficiencies
    - ⇒ Complication: correlations between efficiencies at different energies
  - All other cases – Monte Carlo simulation
    - ⇒ Evaluation of the sensitivity to detector model parameters
    - ⇒ Combine sensitivity values with reasonable uncertainty range of the values of the parameters

- Uncertainties of the joint emission probabilities p_i, p_{ij} … or of equivalent quantities
  - The values of p_i, p_{ij} … depend in a complex way on the parameters of the decay scheme
  - Simultaneous dependence on several parameters of the decay scheme

  => Complication: decay scheme parameters are correlated

  ⇒ Uncertainty evaluation should include the correlation between the decay parameters

  ⇒ Covariance matrix of the decay scheme parameters is required
    - Usually only variance of each parameter is available

- Analytical calculation of uncertainties generally too complex

- Best procedure: application of Monte Carlo simulation as recommended in the Supplement 1 to the Guide to the Expression of the Uncertainty in Measurement JCGM 101:2008

  Sima and Lépy, ARI 109 (2016) 493; Kastlander et al., 122 (2017) 174

Example: Effect of uncertainty of decay scheme parameters on F_C

- F_C for the 276 keV peak of a ^{133}Ba point source, measured with an n-type detector – Sima and Lépy, ARI 109 (2016) 493

⇒ Many parameters required simultaneously => covariances involved

⇒ Not feasible to evaluate analytically the distribution of F_C

⇒ Procedure:

  - Monte Carlo evaluation of ε(Ei), η(Ej), $\int \varepsilon^p(E_i, \vec{r}) \cdot \eta^p \left(E_j, \vec{r}\right) dV$, … for each group of radiations, with very good statistical uncertainty
  - Preparation of a big number of decay schemes of ^{133}Ba, randomly sampled on the basis of evaluated decay parameters and their uncertainties
  - Evaluation of F_C for each decay scheme
    - In order to find only the effect of the uncertainties of the decay scheme parameters, the same probabilities of detecting groups of radiations were applied for each decay scheme
  - Summarizing the distribution of F_C values

- ***Sampling of the decay scheme in the natural way (first decay transition, then sequential de-excitation transitions) not appropriate, would require describing strong correlations***

=> Special procedure of simulation of decay scheme parameters, based on evaluation procedure, for minimizing the effect of correlations

Procedure applied for sampling of the decay scheme – Sima and Lépy, ARI 109 (2016) 493 (thanks due to Marie-Martine Bé):

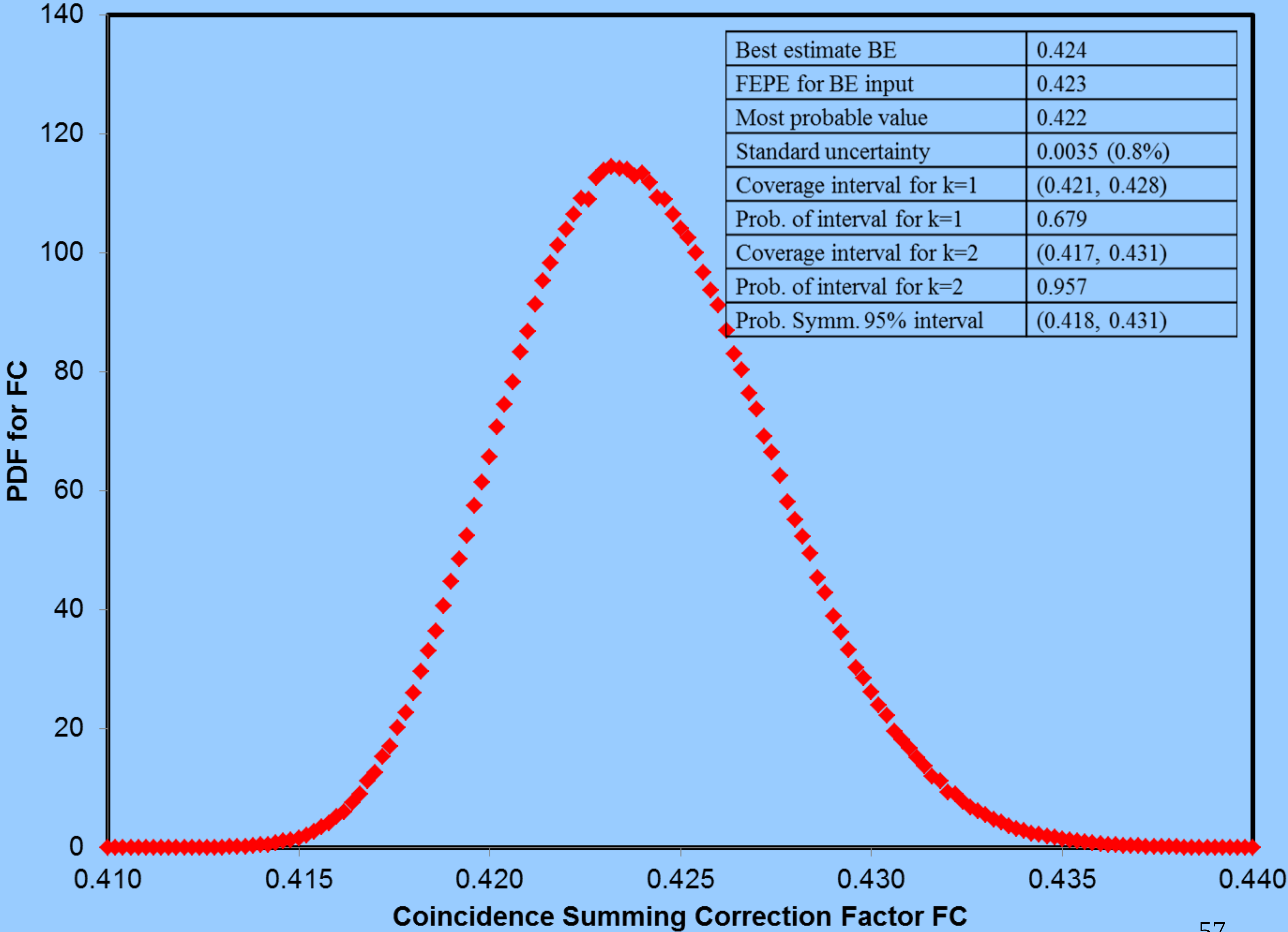
1. Gamma emission probabilities: sampled from normal distribution with parameters from DDEP
2. α_K and α_L sampled from normal distributions with parameters from DDEP (uncorrelated)
3. α_T sampled from normal distribution with parameters from DDEP; if in conflict with α_K and α_L a new value was sampled
4. Transition probabilities evaluated from gamma emission probability and α_T
5. Decay branching ratios evaluated from the balance of transition probabilities, starting from the highest level
6. P_K and ω_K sampled from normal distributions with parameters from DDEP

If unphysical values were obtained for a parameter, the value of that parameter was sampled again.

$\Rightarrow$ A sample decay scheme containing the data required for the evaluation of joint emission probabilities is obtained

$\Rightarrow$ Using the joint emission probabilities and the probability of interaction with the detector of the group of radiations a value of F_C is computed

$\Rightarrow$ The procedure was repeated 10^6 times to obtain the distribution of F_C values



Best estimate BE	0.424
FEPE for BE input	0.423
Most probable value	0.422
Standard uncertainty	0.0035 (0.8%)
Coverage interval for k=1	(0.421, 0.428)
Prob. of interval for k=1	0.679
Coverage interval for k=2	(0.417, 0.431)
Prob. of interval for k=2	0.957
Prob. Symm. 95% interval	(0.418, 0.431)

Source: Sima and Lépy, ARI 109 (2016) 493

6. Sum peak method

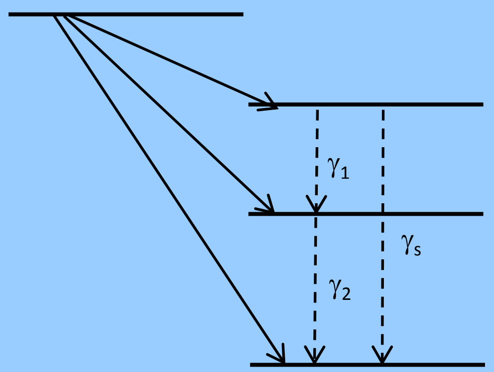
- For a simple decay scheme (e.g. ^{60}Co) the measurement of the peak count rates R_1 and R_2, of the sum peak count rate R_S and of the count rate in the total spectrum R_T allows the absolute determination of the activity and of the efficiencies - Brinkman et al., IJARI 14 (1963) 153; 433 – the sum peak method
  - 4 experimental data depending through 4 equations with particular expressions on 2 FEP efficiencies ε_1, ε_2 and 2 total efficiencies η_1, η_2
    => ε_1, ε_2, η_1, η_2 can be eliminated from equations => activity can be computed in function of count rates only
  - After obtaining the activity, the efficiencies ε_1, ε_2, η_1, η_2 can be calculated

$$R_1=p_1\varepsilon_1A-p_{12}\varepsilon_1\eta_2w_{\varepsilon,\eta}A=p_1F_{C1}\varepsilon_1A$$

$$R_2=p_2\varepsilon_2A-p_{12}\varepsilon_2\eta_1w_{\eta,\varepsilon}A=p_2F_{C2}\varepsilon_2A$$

$$R_S=p_S\varepsilon_SA+p_{12}\varepsilon_1\varepsilon_2w_{\varepsilon,\varepsilon}A=p_SF_{CS}\varepsilon_SA$$

$$R_T=(p_1\eta_1A-p_{12}\eta_1\eta_2w_{\eta,\eta}A)+(p_2\eta_2A-p_{12}\eta_1\eta_2w_{\eta,\eta}A)+p_{12}\eta_1\eta_2w_{\eta,\eta}A=p_1\eta_1A+p_2\eta_2A-p_{12}\eta_1\eta_2w_{\eta,\eta}A$$



p_i, p_{ij}=emission probabilities per decay

F_{Ci}=coincidence summing correction factor

$w_{\varepsilon,\eta}$ angular correlation factor: photon 1 in the peak, photon 2 in the full spectrum
$w_{\eta,\varepsilon}$: 1 in the spectrum, 2 in the peak; $w_{\varepsilon,\varepsilon}$ and $w_{\eta,\eta}$ both in the peak or in the spectrum

- First () in the formula for R_T: contribution to the spectrum of the cases when the first photon is absorbed, but the second is not; second () – the similar contribution of the second photon; last term the contribution to the total spectrum of the cases when both photons interact simultaneously with the detector
- Equations are valid for a point source

Case when $p_S=0$

- Equation for R_S becomes:

$$R_S=p_{12}\varepsilon_1\varepsilon_2 w_{\varepsilon 1,\varepsilon 2}A$$

- The other equations remain unchanged
- The system of equations can be solved with the result:

$$A=\left(\frac{R_1\cdot R_2}{R_S}+R_T\right)\cdot\frac{p_{12}\cdot w_{\varepsilon 1,\varepsilon 2}}{p_1\cdot p_2}\cdot\frac{1}{1+\delta}$$

δ = a small correction term (always neglected in the literature):

$$\delta=\frac{p_{12}}{p_1 p_2}\left[p_1\eta_1\left(w_{\varepsilon,\varepsilon}-w_{\eta,\varepsilon}\right)+p_2\eta_2\left(w_{\varepsilon,\varepsilon}-w_{\varepsilon,\eta}\right)-p_{12}\eta_1\eta_2\left(w_{\eta,\eta}w_{\varepsilon,\varepsilon}-w_{\varepsilon,\eta}w_{\eta,\varepsilon}\right)\right]$$

Standard formula for the sum peak method:

$$A = \left(\frac{R_1 R_2}{R_S} + R_T\right) \cdot \frac{p_{12} w_{\varepsilon,\varepsilon}}{p_1 p_2}$$

⇒ Absolute determination of activity (only $w_{\varepsilon,\varepsilon}$ depends on experimental conditions; can be computed rather accurately)

- Valid in the case of:
  - Point sources
  - Negligible dead time and pile-up effects
    - pile-up effects: Nemes et al., NIMA 898 (2018) 11

Uncertainties:

- Statistical uncertainties of count rates, especially of R_S
- Uncertainty of R_T due to:
  - Background subtraction
  - Spectrum extrapolation below the low energy cutoff
  - Problems if other nuclides are present in the spectrum (see below)
- Uncertainty of $w_{\varepsilon,\varepsilon}$
- Uncertainty of p_1, p_2, p_{12} – usually smaller than other contributions

Problems associated with background:

- Other nuclides present in the spectrum: ^{137}Cs and ^{134}Cs present in Fukushima samples
  - measurement of ^{134}Cs by the sum peak method (E_S=1400 = 604+796 keV) requires removal of ^{137}Cs contribution to the background
  - Ogata et al., ARI 134 (2018) 172 – contribution of ^{137} Cs to background estimated by using count rate in the 662 keV peak of ^{137}Cs and Total/Peak ratio for ^{137}Cs measured separately
- Elimination of the total count rate from the sum peak formula – extrapolation to large distances (equivalent to low values of R_1)
  - For R_T=>0, the standard sum peak formula becomes:

$$\left(\frac{R_1 R_2}{R_S} + R_T\right) \cdot \frac{p_{12} w_{\varepsilon,\varepsilon}}{p_1 p_2} \Rightarrow \left(\frac{R_1 R_2}{R_S}\right) \cdot \frac{p_{12} w_{\varepsilon,\varepsilon}}{p_1 p_2}$$

- Ogata et al., ARI 109 (2016) 354 – evaluation of activity by representing the modified expression

$$A' = \left(\frac{R_1 R_2}{R_S}\right) \cdot \frac{p_{12} w_{\varepsilon,\varepsilon}}{p_1 p_2}$$

as a function of R_1 and extrapolation towards R_1=>0 (then also R_T=>0)

- Another possibility:
  - Elimination of R_T from equations using computed values of F_C

$$A = \frac{R_1 R_2}{R_S} \cdot \frac{p_{12} w_{\varepsilon,\varepsilon}}{p_1 p_2} \cdot \frac{1}{F_{C1} \cdot F_{C2}}$$

- Not absolute determination of activity, because requires computed values of F_{C1} and F_{C2} (are sensitive to the uncertainties of the detector model and to the uncertainty of the source position – the latter uncertainty can be reduced using a certain correlation – Suvaila et al., ARI 81 (2013) 76)

Caution: not every pure sum peak can be used for the application of the sum peak method

⇒ The basic equations are not valid if other radiations are in coincidence with photons 1 and/or 2, e.g. in the equations for R_1, R_2, R_S losses due to coincidence effects with other photons should be included

⇒ The method is strictly valid only for nuclides with simple decay schemes

Note:

- For a nuclide with a complex decay scheme, emitting n photons, it is possible to have more than n sum peaks; then there are 2*n* unknowns (ε_i and η_i for each photon) and 2*n* or more experimental data (*n* normal peaks, *n* or more pure sum peaks) => absolute method of activity determination and of obtaining the efficiencies
  - Complex relations, solved numerically
  - If peak/total ratio can be accurately parameterized using a small number of unknown parameters, the required number of pure sum peaks may be lower than *n*
  - Semkow et al., NIMA 290 (1990) 437; Blaauw, NIMA 332 (1993) 493

- Equations valid for point sources

6. Summary

Coincidence summing effects are very important in present day gamma-spectrometric measurements:
- tendency to use high efficiency detectors
- tendency to choose close-to-detector counting geometries

The effects depend on the decay data of the nuclide, on the detector efficiency, on the sample

The effects are present both in the process of calibration and sample measurement

For activity assessment the coincidence summing correction factors F_C for principal peaks should be evaluated, several efficiency values and decay data parameters are required

- In the case of negligible coincidence-summing, the activity A can be obtained from the simple count rate (R) equation: $R(E) = \varepsilon(E)\, P_\gamma(E)\, A$
  - a single efficiency value $\varepsilon(E)$ (directly measured, interpolated from efficiency curve, or computed) and its uncertainty are required
  - a single parameter of the decay scheme, $P_\gamma(E)$, and its uncertainty are required

- In the presence of coincidence-summing effects, A can be obtained from the more complex count rate equation: $R(E)=F_C \cdot \varepsilon(E)\ P_\gamma(E)\ A$
    - Besides $\varepsilon(E)$ for the energy of the peak, also peak $\varepsilon(E_p)$ and total $\eta(E_j)$ efficiencies for all relevant groups of photons, their uncertainties and covariances, are required
        - Point sources: ε and η can be directly measured, interpolated from efficiency curves, or computed, η being more difficult to measure (better ε/η ratio)
        - Extended sources – efficiencies should be computed, do not have any experimental counterpart
    - Besides $P_\gamma(E)$ for the energy of the peak, all the joint emission probabilities for relevant radiations, their uncertainties and covariances are required

For spectrum analysis and nuclide identification (especially when using automatic procedures) all pure sum peaks should be properly assigned. Peak interferences should be removed

Presently there are tools that can be applied reliably for the evaluation of coincidence summing corrections