International Atomic Energy Agency Technical Visit on

Coincidence summing and geometry correction in gamma spectrometry

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True Coincidence Summing Corrections - Theory

Octavian Sima

University of Bucharest, Romania

True Coincidence Summing Corrections - Theory

1. Introduction - high resolution gamma-ray spectrometry
   - low efficiency measurements
   - high efficiency measurements
   - need for coincidence summing corrections
2. Physics of coincidence summing effects
   - summing in
   - summing out
   - intuitive computation of coincidence summing
3. Decay data
4. Efficiencies
   - quasi-point source approximation
   - volume sources
5. Methods for evaluation of coincidence summing corrections
   - Recursive formulae (Andreev type)
   - Matrix formulation (Semkow type)
   - Deterministic approach
   - Random sampling of the decay path (=> separate lecture)
6. Application - GESPECOR
7. Summary

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 for 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)

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

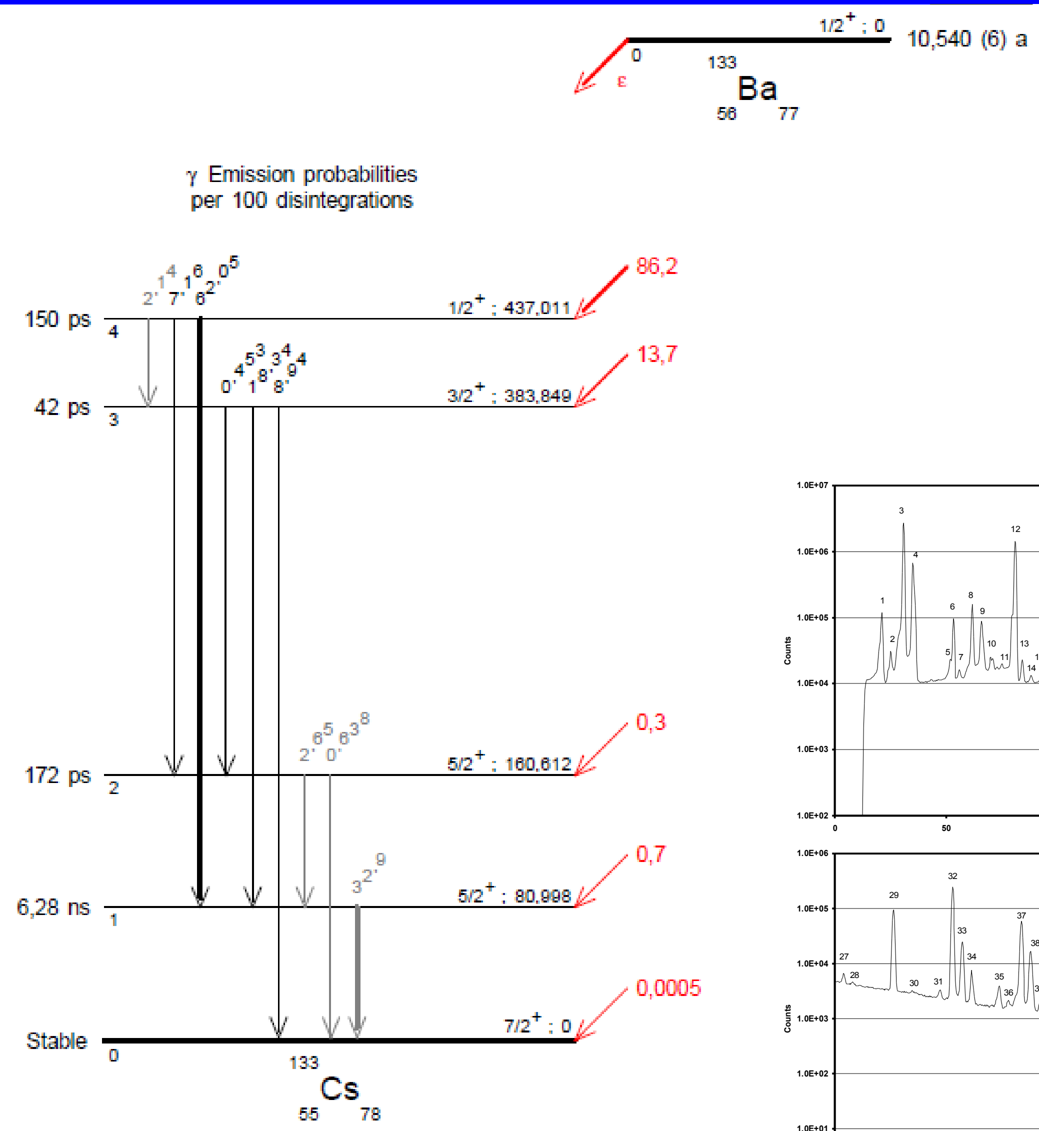
=> multielemental, nondestructive analysis

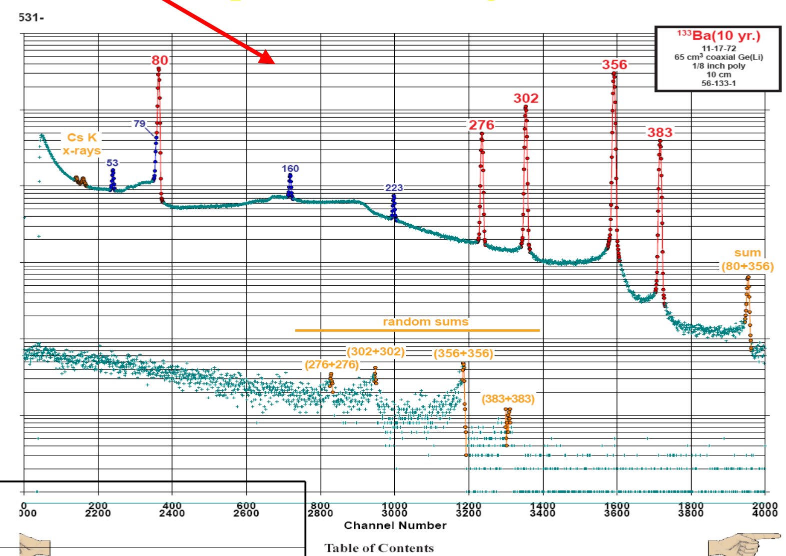
Low efficiency measurements

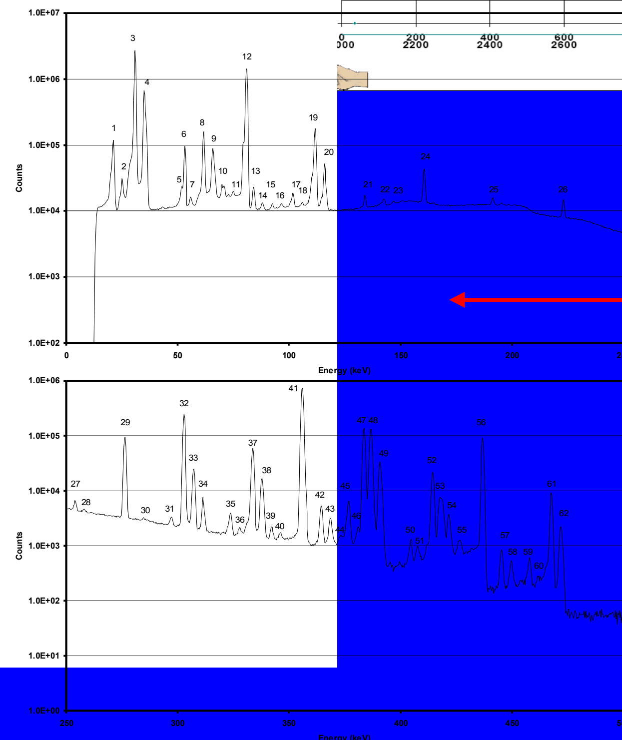
^{133}Ba

65 cm^3 Ge(Li)

Data source: Gamma-ray spectrum catalogue, INEEL







Close to detector measurement; 25% relative efficiency n-type HPGe

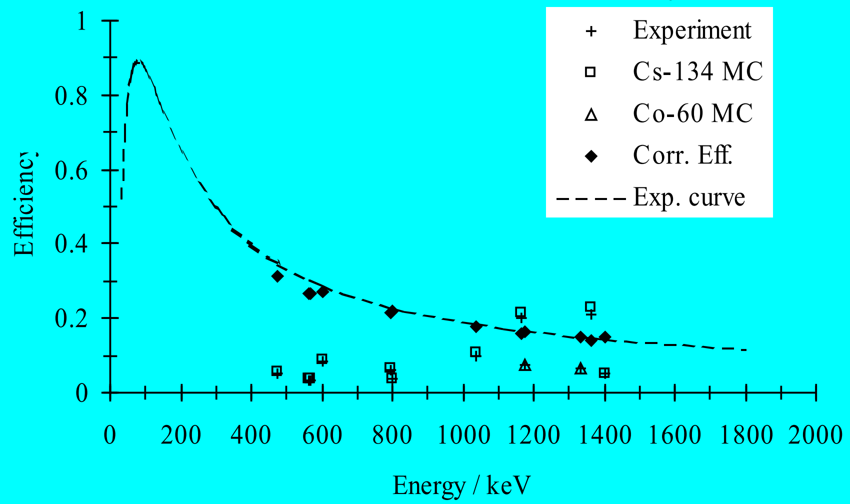
Source: Arnold and Sima, ARI 64 (2006) 1297

Data source: BIMP Monographie -5

Coincidence summing effects responsible for the difference between the spectra

- 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



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!

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 (efficiency)

⇒ n photons interact in the detector within the resolving time

⇒ a single signal of the detector instead of n separate signals

⇒ 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, $ED_k = E_k$ or in the total spectrum $ED_k < E_k$)

If $ED_k = E_k$ => in the absence of coincidence effects a count is recorded in the peak of energy E_k

=> due to the detection of the other photons, the count is no longer recorded in the peak

=> coincidence losses from the peak

=> the count rate in the peak of energy E_k is decreased due to coincidence losses

If each photon deposited the complete energy in the detector, $ED_1=E_1$, $ED_n=E_n$, then a count is recorded in the sum peak of energy $E_s=E_1+E_2+ \dots +E_n$

=> summing in contributions

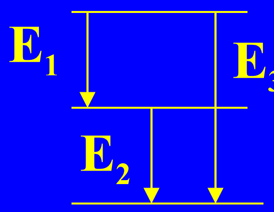
If the nuclide emits a photons with energy $E_s=E_1+E_2+ \dots +E_n$

=> additional signals in the peak

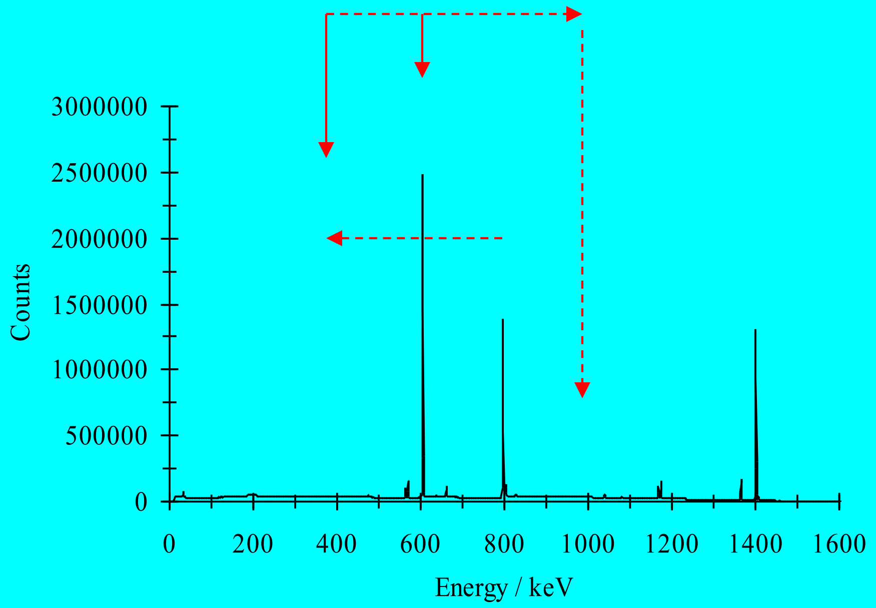
If the nuclide does not emit photons with energy $E_s=E_1+E_2+ \dots +E_n$

=> a pure sum peak appears

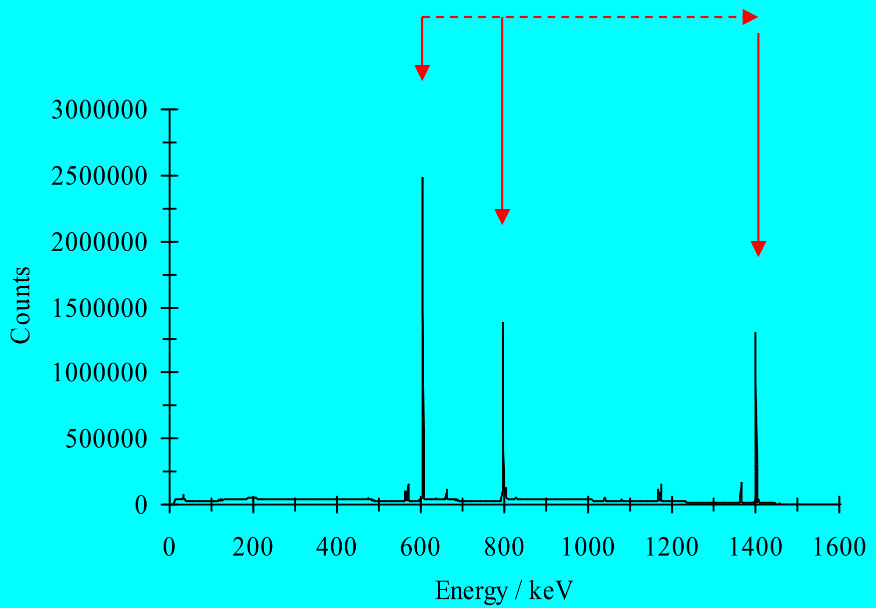
Ex: $\mathbf{E_1}$ and E_2 interact both with the detector:



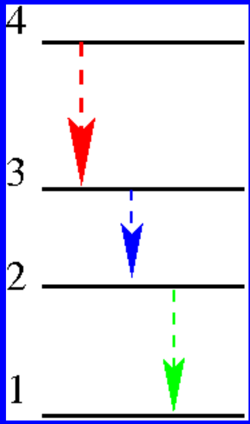
1. $ED_1=E_1$, $ED_2<E_2$ coincidence losses from E_1 peak
2. $ED_2=E_2$, $ED_1<E_1$ coincidence losses from E_2 peak
3. $ED_1=E_1$, $ED_2=E_2$ coincidence summing in the E_3 peak (pure sum peak with energy E_1+E_2 if no E_3 transition)
4. $ED_1<E_1$, $ED_2<E_2$ replacement of 2 counts with one in the Compton region of the spectrum

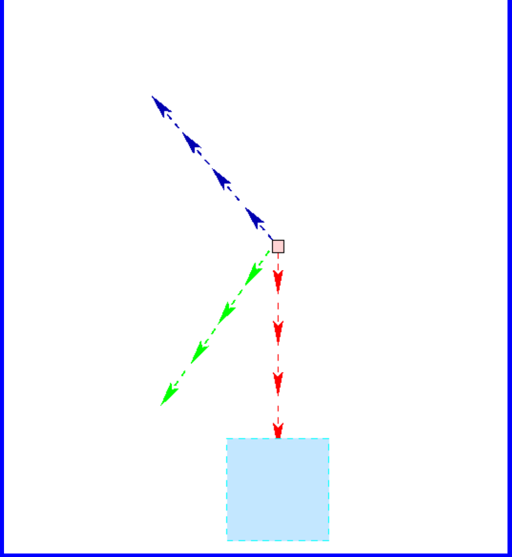


Coincidence losses from the 604 keV peak
The signals corresponding to complete absorption of 604 keV photon and incomplete absorption of the 796 keV photon added resulting in a single signal corresponding to 604+DE(796) energy



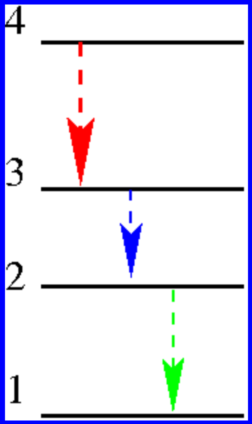
Sum peak at the energy 604+796=1400 keV
The signals corresponding to complete absorption of 604 and 796 keV photons are added resulting in a signal corresponding to an energy of 1400 keV
Probability of this event in comparison with the above:
ε(706) in comparison with η(796)

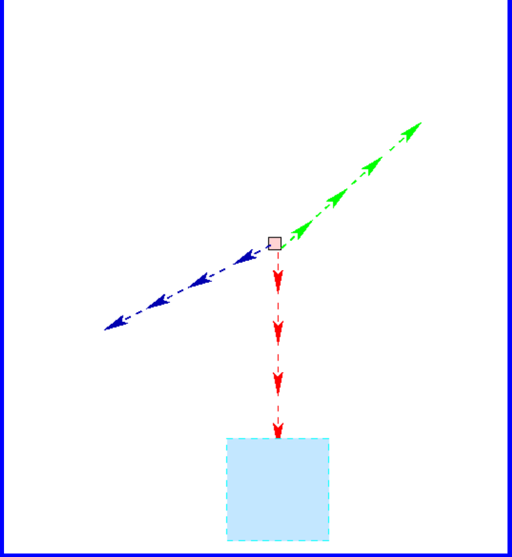




True Coincidences

- The three photons are emitted practically in the same time in various directions (with an angular correlation)





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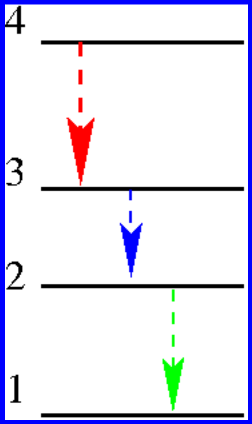

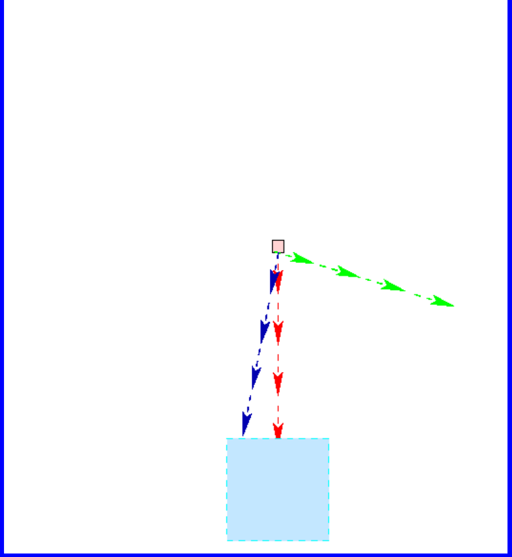

True Coincidences

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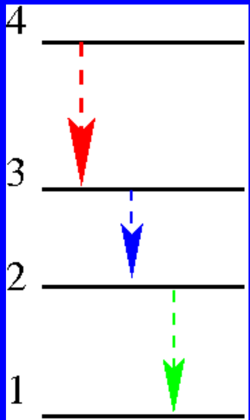
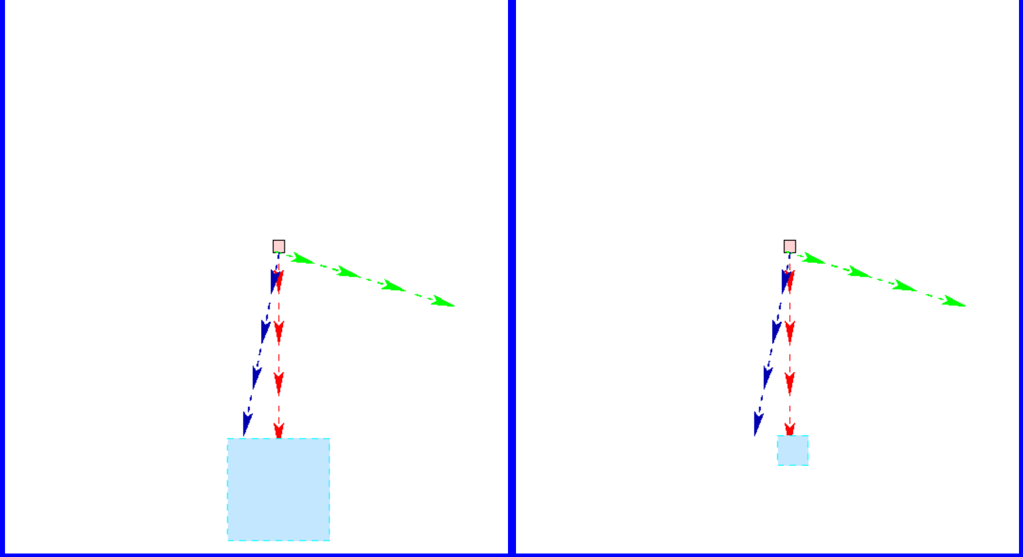
-Sometimes it happens that two photons interact with the detector (in closed end coaxial detectors the probability that three photons interact in the detector is much smaller than that for two photons)





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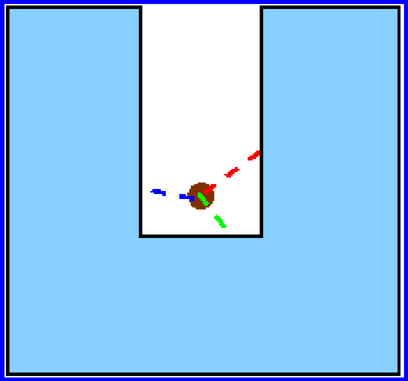
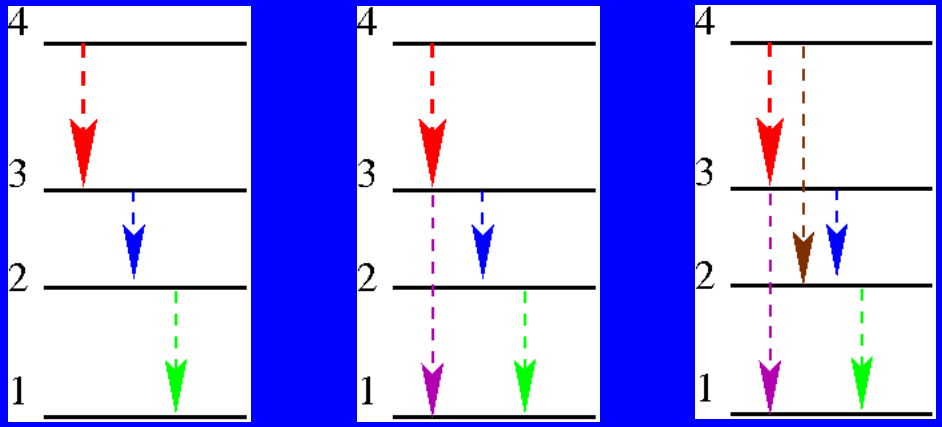


- The three photons are emitted practically in the same time in various directions (with an angular correlation)
-Sometimes it happens that two photons interact with the detector (in closed end coaxial detectors the probability that three photons interact in the detector is much smaller than that for 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)

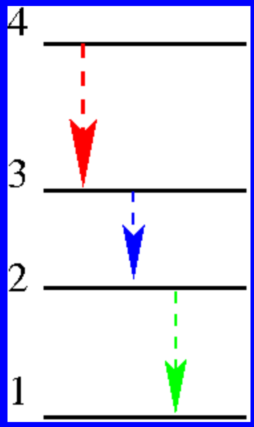


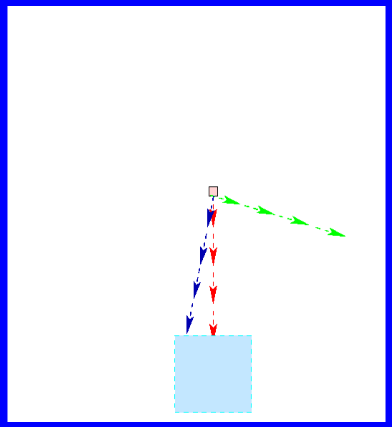
The groups of photons that are emitted together and their joint emission probabilities are different for the three decay schemes

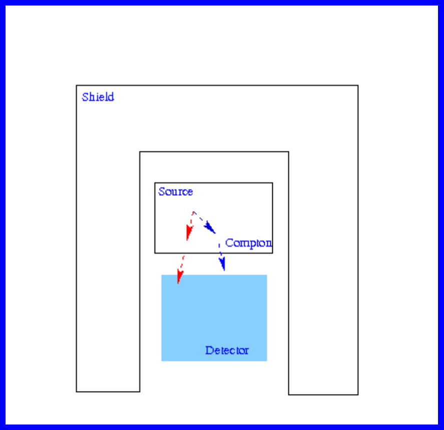
True Coincidences

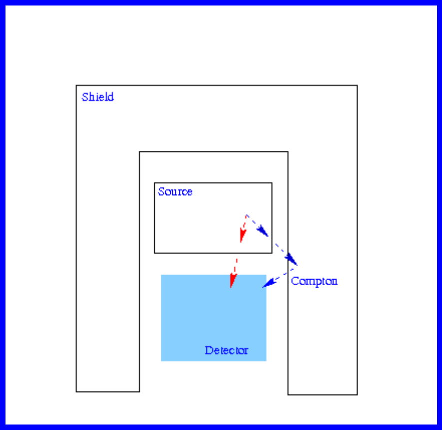
- Coincidence summing effects depend on the decay scheme and are specific to each transition

True Coincidences

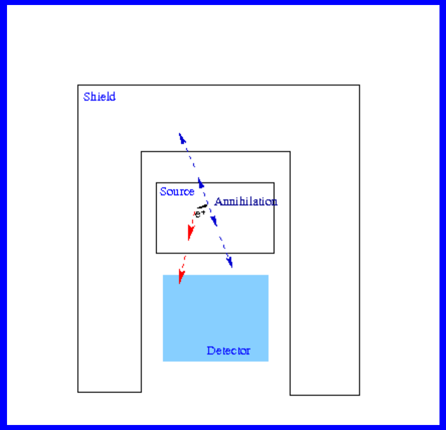








- Contributions to coincidence summing from all the radiations following decay

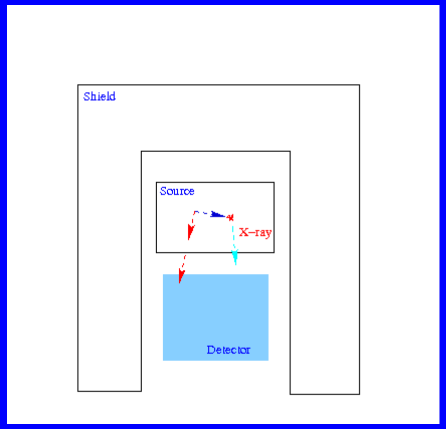


- gamma from transitions, X-rays (EC, conversion electrons)

- radiation scattered in the source, shield

- annihilation photons

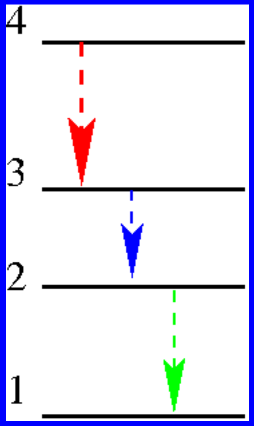
- X rays excited in the shield, in the matrix

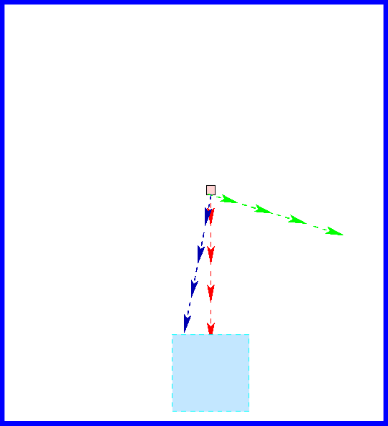


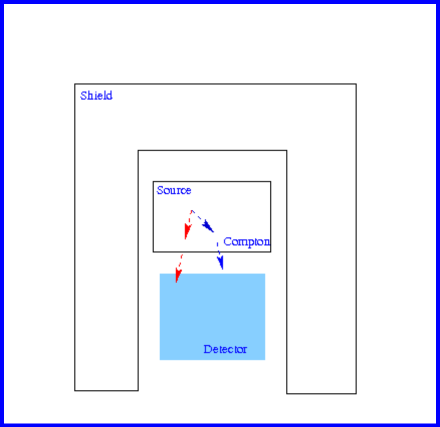
- beta particles, bremsstrahlung etc

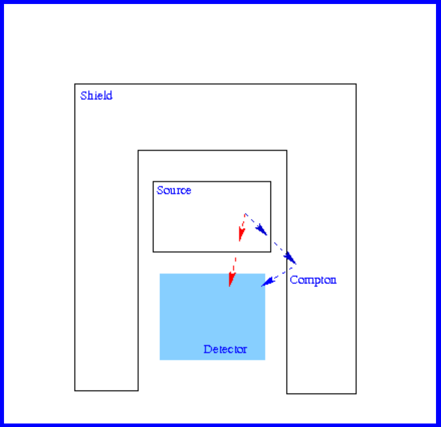
- Relative contribution independent of A!

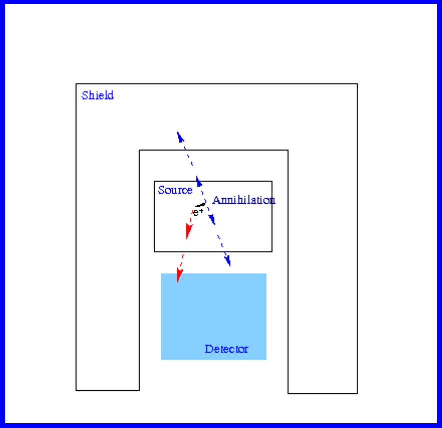
True coincidence losses from the peak depend on the total efficiency

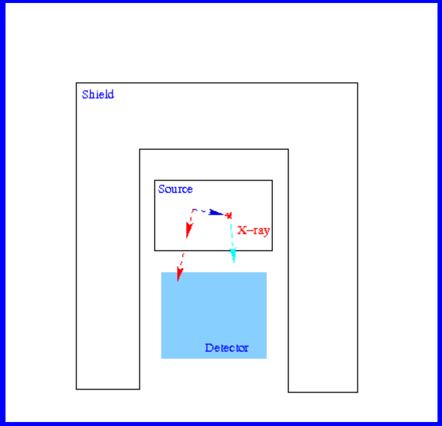












- Contributions to total efficiency of the “blue” photon in the absence of coincidences: the same processes as for coincidence summing

- direct gamma interaction with the detector

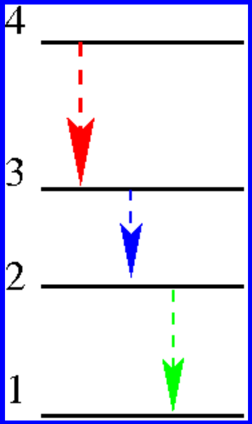
- radiation scattered in the source, shield

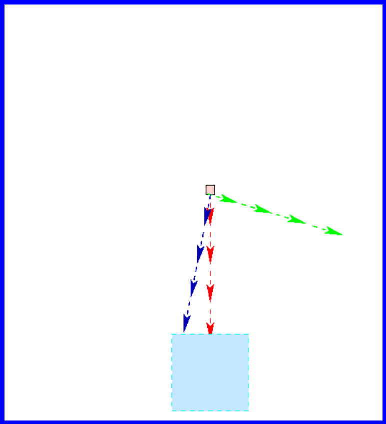
- annihilation photons (in case of positron decay)

- X rays excited in the shield, in the matrix

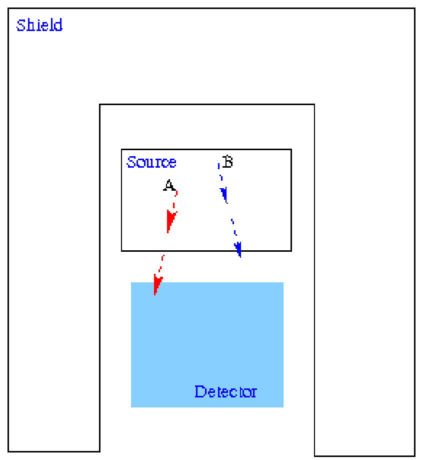
- beta particles, bremsstrahlung etc

Coincidence summing contributions to sum peaks depend on the peak efficiency





- Sum peak contribution of the “blue” and “red” photons is obtained in the same processes in which in the absence of coincidences each photon would be registered in the peak



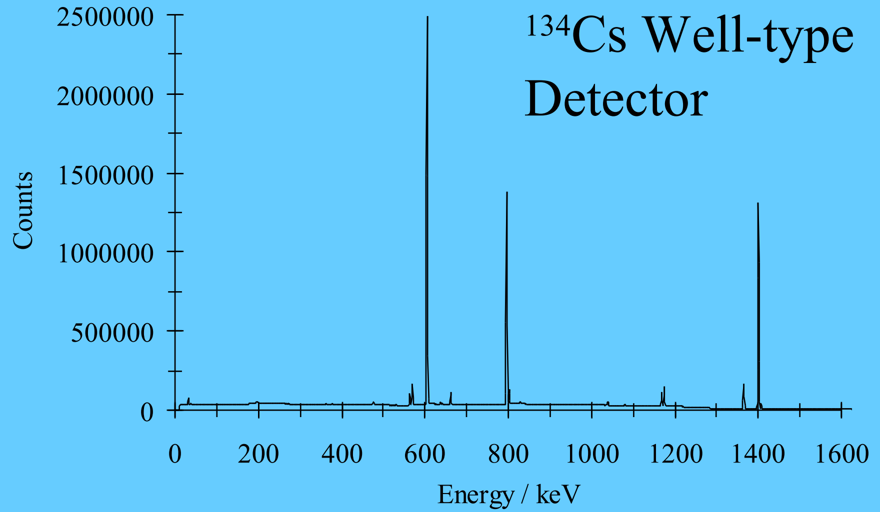
Random coincidences

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

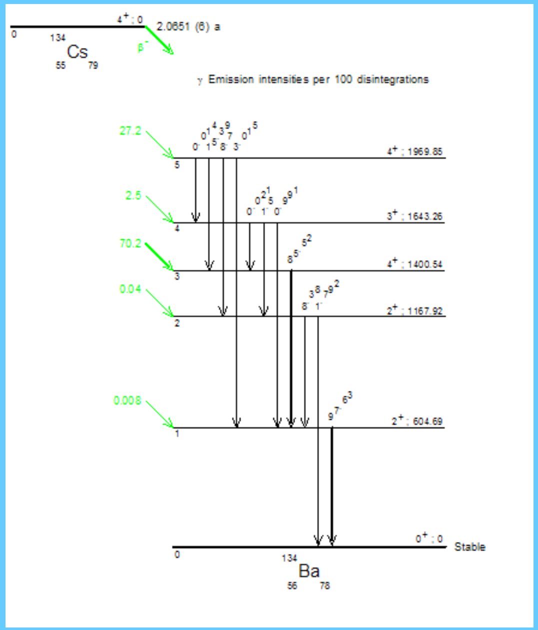
- Important when the count rate is high
- The effect can be avoided by decreasing the count rate, e.g. measuring the source at big distances from the detector

- 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)

Coincidence with gamma photons



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



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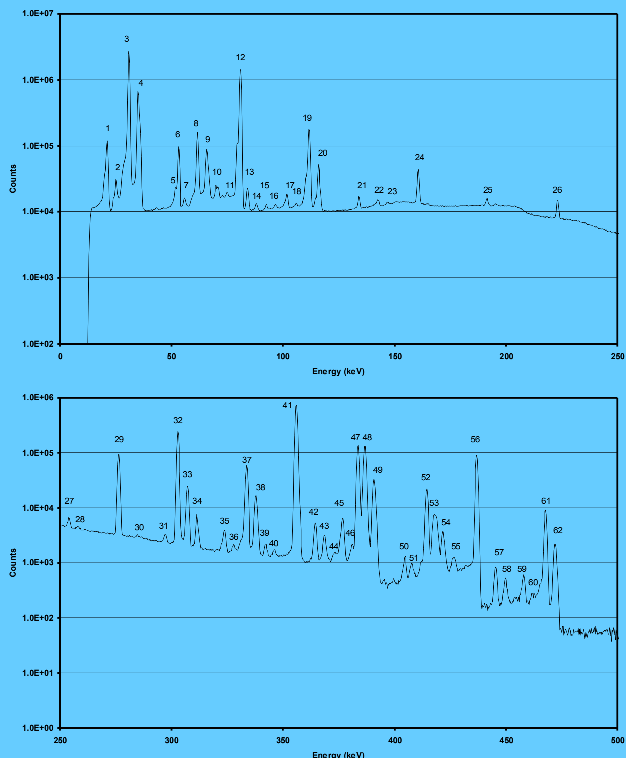
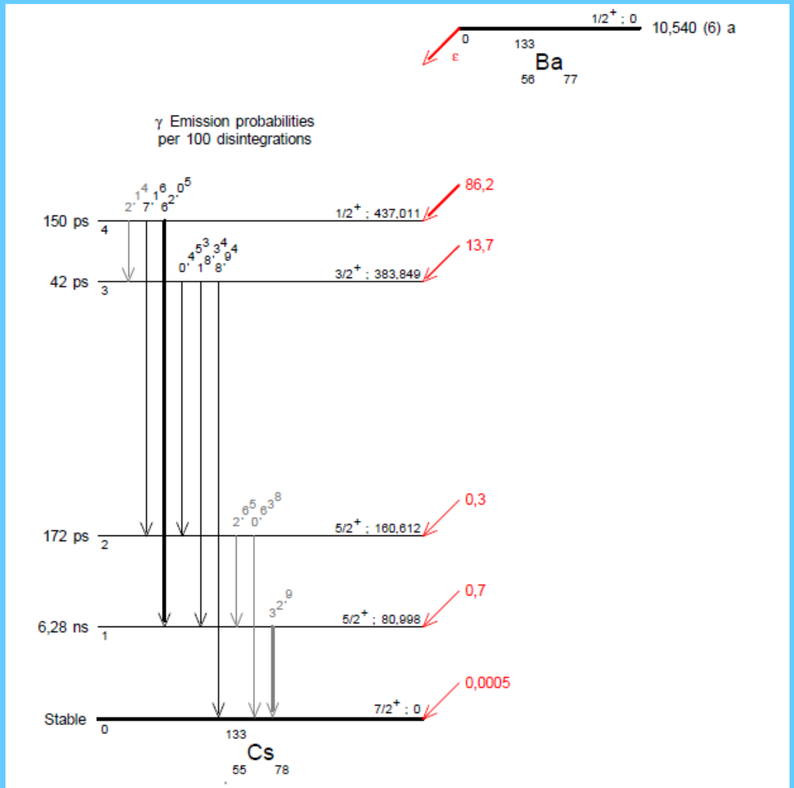
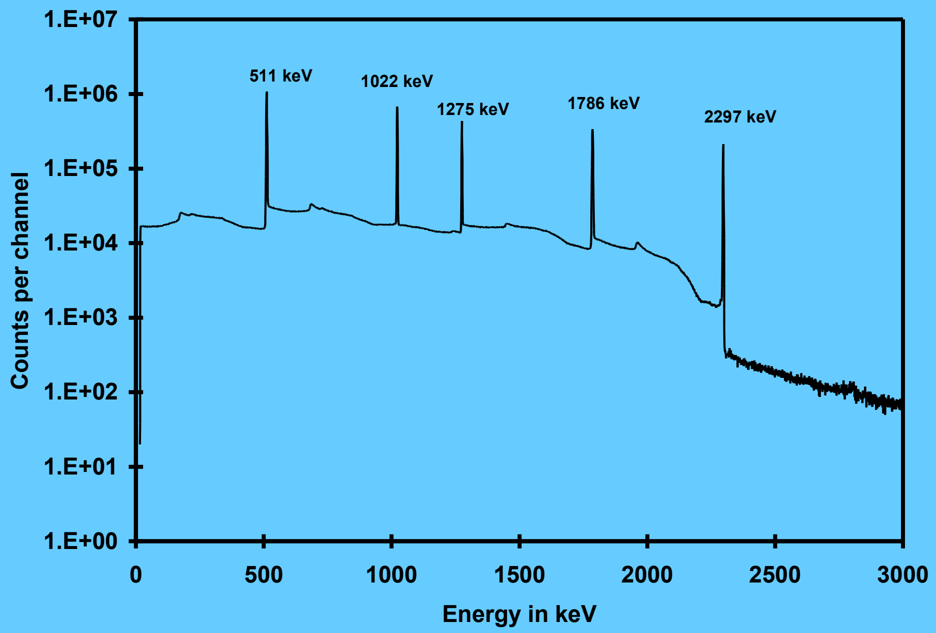


With X-rays

Source: Arnold and Sima ARI 64 (2006) 1297

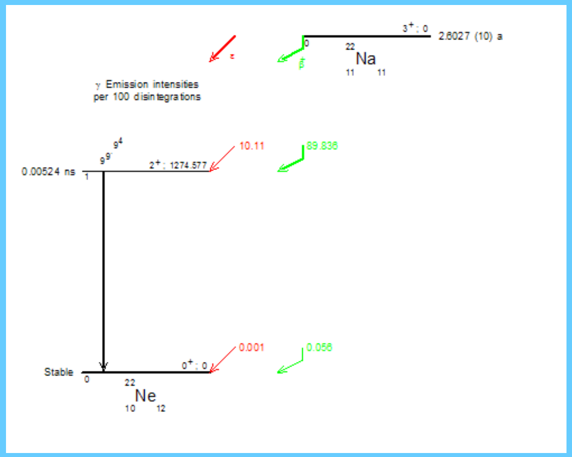




Source: Sima and Arnold, ARI 53 (2000) 51

Contributions of annihilation photons

^{22}Na measured in well-type detector



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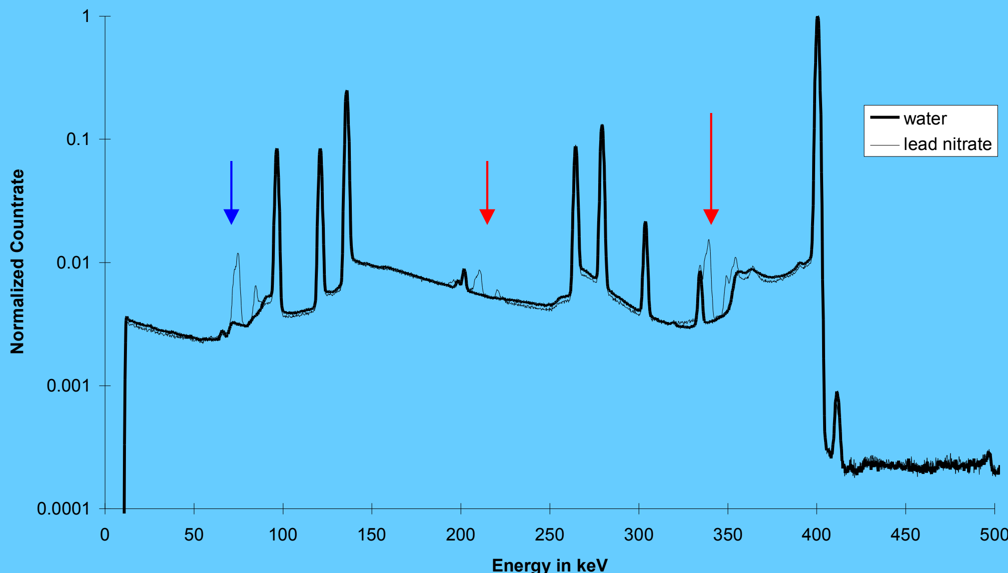
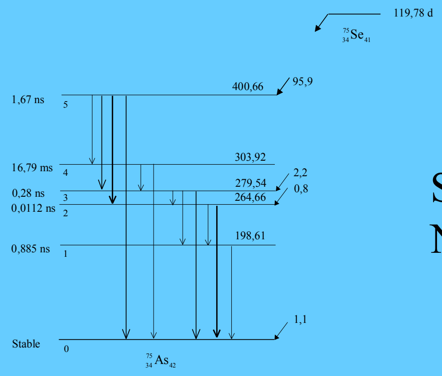


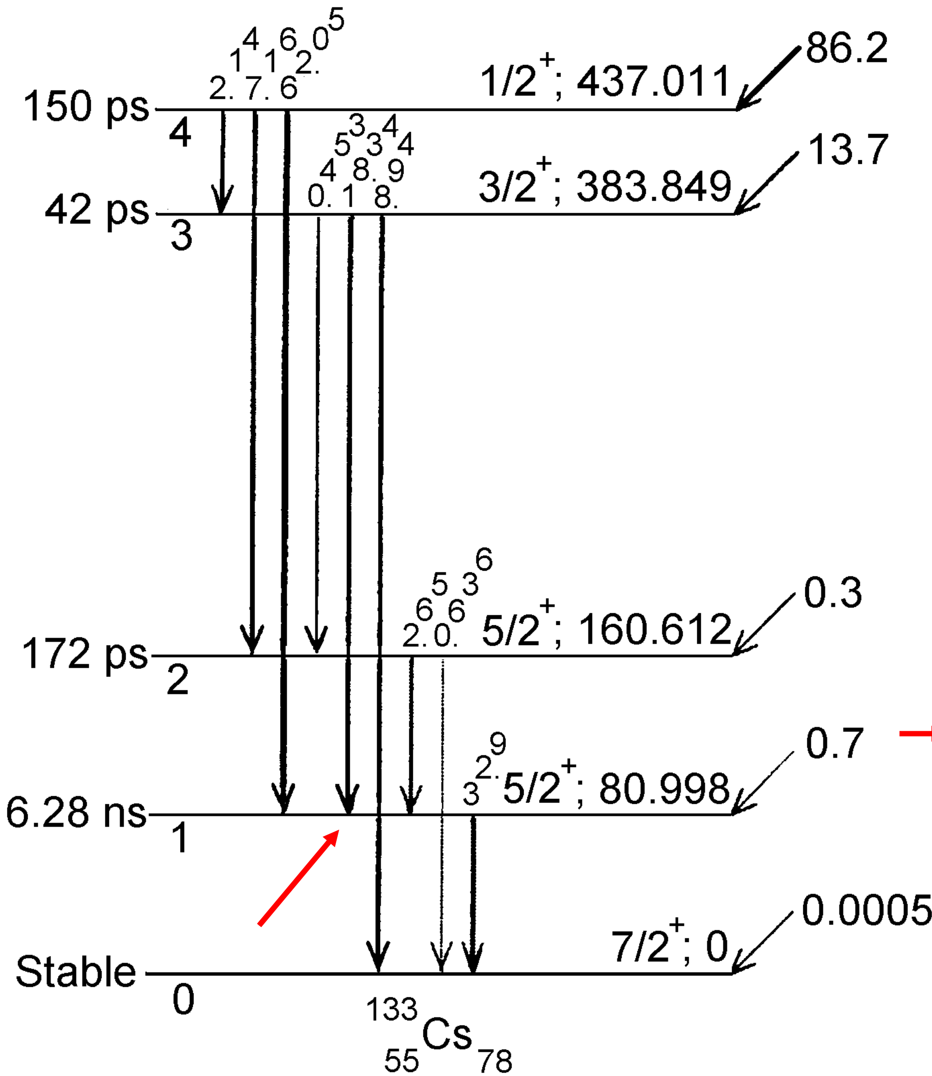
Contributions of matrix X-rays

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



Source: NUCLEIDE

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



Ba-133 EC decay

E (keV)	Iγ (per 100 Dis)
53.1622	2.14±0.03
79.6142	2.65 ±0.05
80.9979	32.9 ±0.3
160.6121	0.638 ±0.004
223.2368	0.453 ±0.003
276.3989	7.16 ±0.05
302.8508	18.34 ±0.13
356.0129	62.05 ±0.19
383.8485	8.94 ±0.06

Data source: Nucleide

302 keV; in the absence of coincidences $R(302) = \varepsilon(302)\, P_\gamma(302)\, A$

But: 302 keV photon is emitted together with other radiations!

1 K_α(EC4)-53-302-81

2 K_α(EC4)-53-302-K_α(81)

3 K_α(EC4)-53-302-K_β(81)

4 K_α(EC4)-53-302-other(81) (other => no signal in detector)

5 K_α(EC4)- K_α(53)-302-81

6 K_α(EC4)- K_α(53)-302- K_α(81)

And so on, ending with

48 other(EC4)-other(53)-302-other(81)

Other decay paths start by feeding the 383 keV level (EC3):

49 K_α(EC3)-302-81

50, 51, 52, 53, 54, 55, 56, 57, 58, 59

60 other(EC3)-302-other(81)

Each combination i has a specific **joint emission probability** p_i !

$$P_\gamma(302)=p_{Path\ 1}+p_{Path\ 2}+p_{Path\ 3}+\ldots+p_{Path\ 60}$$

The detector cannot resolve the signals produced by the photons emitted along a given decay path – a single signal, corresponding to the total energy delivered to the detector is produced

Each combination has a specific probability to contribute to the count-rate in the 302 keV peak, e.g. Path 1 (combination 1):

K_α(EC4)-53-302-81 =>

$$\varepsilon_{Path\ 1}=[1-\eta(K_\alpha)][1-\eta(53)]\,\varepsilon(302)\,[1-\eta(81)]$$

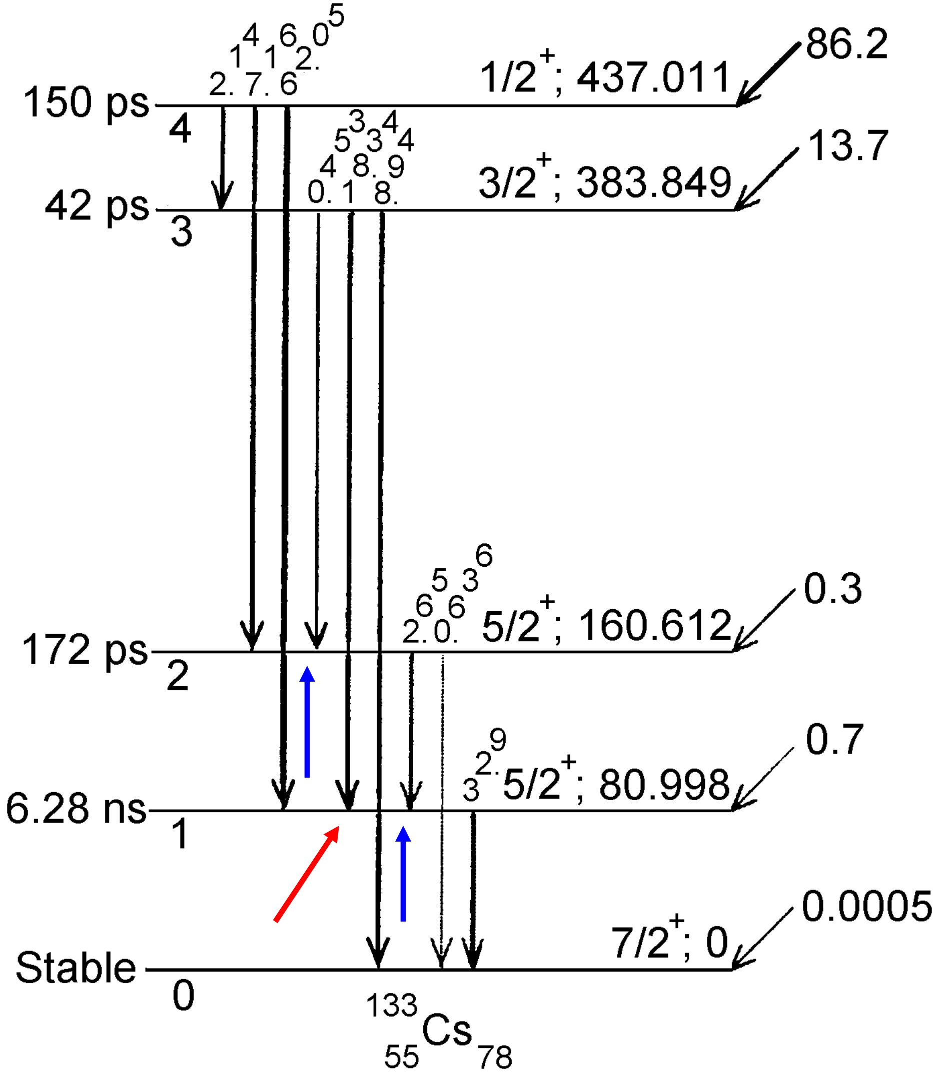
$\eta(E)$= total detection efficiency for photons of energy E

$\varepsilon_{Path\ i} < \varepsilon(302)$ => **coincidence losses from the 302 keV peak**

Volume sources: more complex – effective total efficiency is needed

Additional complication – angular correlation of photons

Ba-133 EC decay



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Data source: Nucleide

Sum peak contributions to the 302 keV peak:

Combinations like:

K_α(EC4)-53-223-79-81 contribute to the 302 keV peak

with a probability $[1- \eta(K_\alpha)][1-\eta(53)]\ \varepsilon(223)\ \varepsilon(79)\ [1-\eta(81)]$

Other 59 similar contributions

Generalization:

1. On each path including a direct transition with emission of the 302 keV photon:

$\varepsilon_{\text{Path } i} = \varepsilon(302)$ * Product of $[1- \eta(\text{radiation } k)]$

where radiation k corresponds to each of the radiations from the Path i that accompany the 302 keV photon

2. On each path with sum peak contribution (here only 223+79)

$\varepsilon_{\text{Path } j} = \varepsilon(s_1)^*..\ \varepsilon(s_p)$ * Product of $[1- \eta(\text{radiation } k)]$

where radiations $s_1 \ldots s_p$ corresponds to each of the radiations from the Path j contributing to the sum peak

3. The probability of a count in the 302 keV peak for one decay is the sum of the probability of each path with the probability of a count on that path:

$$\mathrm{Sum}\ (P_{\mathrm{Path\ n}} * \varepsilon_{\mathrm{Path\ n}})$$

n denotes any path 1 to 60 for direct gamma emission plus other 60 paths for sum peak combinations

4. Developing the products of efficiencies along each path and regrouping the terms

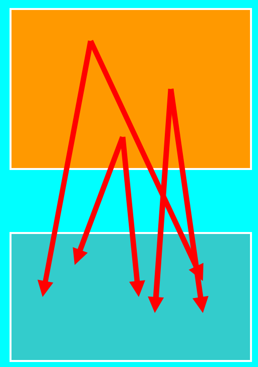
$\mathrm{Sum}\ (P_{\mathrm{Path\ n}} * \varepsilon_{\mathrm{Path\ n}}) = P_{\gamma}(302) * \varepsilon(302)$	direct contribution
$-\ \mathrm{Sum}\ [P_{\gamma(302),j} * \varepsilon(302) * \eta(j)]$	first order losses
$+\ \mathrm{Sum}\ [P_{\gamma(302),j,k} * \varepsilon(302) * \eta(j) * \eta(k)] - \ldots + \ldots$	higher order contrib.
$+\ P_{\gamma(223),\ \gamma(79)} * \varepsilon(223) * \varepsilon(79)$	first order summing in
$-\ \mathrm{Sum}\ [P_{\gamma(223),\ \gamma(79),j} * \varepsilon(223) * \varepsilon(79) * \eta(j)]$	1-st losses from S.P.
$+\ \mathrm{Sum}\ [P_{\gamma(223),\ \gamma(79),j,k} * \varepsilon(223) * \varepsilon(79) * \eta(j) * \eta(k)] - \ldots$	higher order losses from sum peak contribution

$P_{i,j}$, $P_{i,j,k}$ probability of emission of the group of photons (i,j), (i,j,k)

5. Volume sources

-Efficiencies depend on the emission point

-All efficiencies are affected simultaneously by the position of the emission point



$\Rightarrow$Products of efficiencies should be replaced by suitable integrals

Example:

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

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

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

In the presence of coincidence summing

$R(E) \neq \varepsilon(E)\ P_\gamma(E)\ A$, but

$R(E) = F_C\ \varepsilon(E)\ P_\gamma(E)\ A$

F_C = coincidence summing correction factor, depends on:

- **decay scheme parameters**
- **peak and total efficiency for the set of energies of all the photons**

The case of quasi-point sources:

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

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

In the correction formula the peak and total efficiencies for the complete source are required; they can be directly measured

The case of extended sources:

$$F_C(E_i;X) = 1 - \sum_j \frac{p_{ij}}{p_i} \cdot \frac{\int_V \varepsilon^P(E_i;\vec{r}) \cdot \eta^P(E_j;\vec{r})\, dV}{\int_V \varepsilon^P(E_i;\vec{r})\, dV} + \sum_{j,k} \frac{p_{ijk}}{p_i} \cdot \frac{\int_V \varepsilon^P(E_i;\vec{r}) \cdot \eta^P(E_j;\vec{r}) \cdot \eta^P(E_k;\vec{r})\, dv}{\int_V \varepsilon^P(E_i;\vec{r})\, dV} - \ldots$$

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

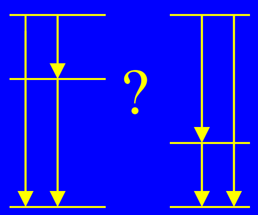
In this case integrals of efficiencies are required; they can not be expressed as directly measurable properties of the source and experimental configuration (Sima and Arnold, ARI 53 (2000) 51)

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 a 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
- 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)

International Committee on Radionuclide Metrology (ICRM)
President: Dr. Pierino De Felice
    http://physics.nist.gov/Divisions/Div846/ICRM/
Recommendations for the development of a consistent set of decay data

Decay Data Evaluation Project
    http://www.nucleide.org/DDEP.htm
Careful evaluation
Periodic updates

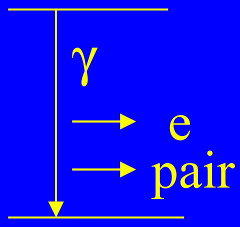
NUCLEIDE database and software (evolved from Tables des Radionucleides) – LNHB
    http://www.nucleide.org/

BIPM Monographie – 5
    http://www.nucleide.org/Publications/monographie_bipm-5.htm

BNL, ENSDF – larger number of nuclides, less dedicated evaluations
Nuclear Data Sheets
    http://www.nndc.bnl.gov/ensdf/

Definitions (Introduction –Table de radionucleides CEA–DETECS/LNHB)

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 deexcitation transition

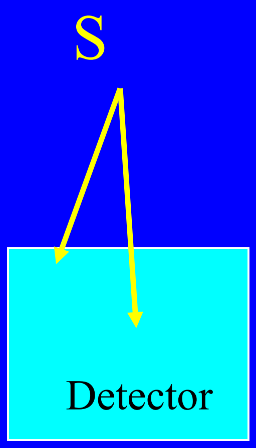
$$P_{XK} = \omega_K \, \alpha_K \, P_g \, / \, (1+\alpha_t)$$

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 $P_{\varepsilon j}$)

4. Efficiencies



Quasi-point source: E_i and E_j

- probability of completely absorbing both photons in the detector per one decay:

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

  (angular correlation neglected)

  = probability of a count in the sum peak per decay

- probability of completely absorbing E_i and incompletely absorbing E_j per decay:

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

  = probability of losing a count from the peak of energy E_j per decay

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 total efficiency η – difficult

- sources emitting a single radiation required
- preferably evaluation of the ratio P/T of the FEPE to total efficiency
    - 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

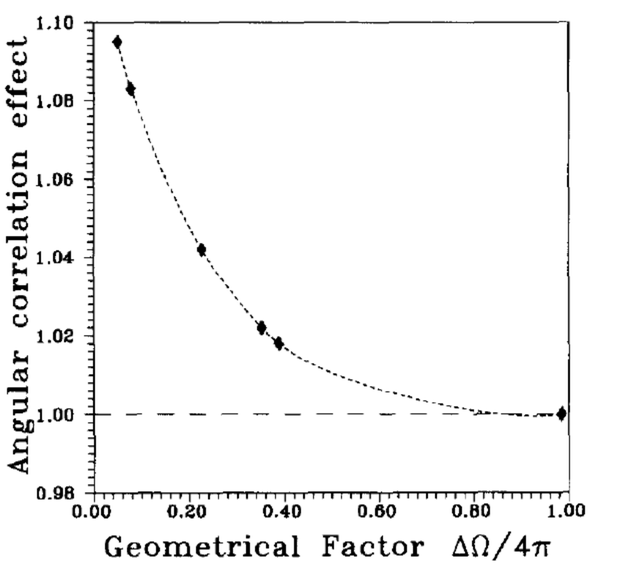
Computation of η or better P/T

- 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

Angular correlation effect

- origin
- magnitude higher for pure sum peaks
- environmental samples

(Roteta et al, NIMA 369 (1996) 665)



Sum peak 1173+1332 keV ^{60}Co

Source: Sima, ARI 47 (1996) 919

The required efficiencies are no longer directly accesible to measurement, e.g.

$$\varepsilon(E_i)\cdot\varepsilon(E_j) \Rightarrow \frac{1}{\Omega^2}\int_\Omega\int_\Omega \varepsilon^P(E_i;\vartheta_i,\varphi_i)\cdot\varepsilon^P(E_j;\vartheta_j,\varphi_j)\cdot w(\theta(\vartheta_i,\varphi_i,\vartheta_j,\varphi_j))d\Omega_i d\Omega_j$$

$\vartheta_i,\varphi_i =$ *polar angles of the direction of the i - th photon*

$\theta =$ *the angle between the directions of the two photons, function of* $(\vartheta_i,\varphi_i,\vartheta_j,\varphi_j)$

$\Omega =$ *the solid angle of the detector as seen from the source*

$w(\theta) =$ *the angular correlation function*

Extended sources

Integrals of products of efficiencies are required, e.g.

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

Effective total efficiency
Coincidence losses from E_i due to the detection of E_j

$$\frac{\int_V \varepsilon^P(E_p;\vec{r})\cdot\varepsilon^P(E_q;\vec{r})\,dV}{\int_V \varepsilon^P(E_i;\vec{r})\,dV}$$

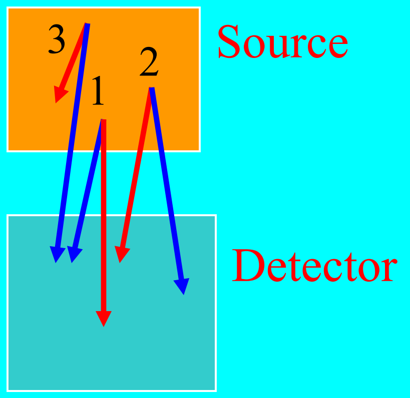
Contributions of E_p+E_q to the peak of energy E_i

$$\eta^{eff}(E_j;E_i)\quad ???\quad \eta(E_j)$$

Weighting factors different, strongly favoring close emission points for effective total efficiency, while for total efficiency a constant weighting factor:

$$\frac{\varepsilon^P(E_i;\vec{r})}{\int_V \varepsilon^P(E_i;\vec{r})\,dV}\quad \textit{instead of}\quad \frac{1}{V}$$

Weighting of the contribution of the “blue” photon due to the requirement of peak detection of the “red” photon:
-solid angle (the contribution of emission point 2 smaller than the contribution of point 1 to the effective total efficiency)
-self-attenuation (emission point 3 does not contribute to the effective total efficiency, but contributes to the usual total efficiency)



⇒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=50 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

Correct computations – by Monte Carlo simulation

Décombaz et al., NIMA 312 (1992) 152; Sima and Arnold, 53 (2000) 51; Laborie et al., ARI 53 (2000) 57; Sima, Arnold and Dovlete, J. Radioanal. Nucl. Chem., 244 (2001) 359; García-Talavera et al., ARI 54 (2001) 769; Berlizov, ACS Symp. Series 945 (2006) 183; Capogni et al., ARI 68 (2010) 1428.

Measurements – map of the point source efficiencies in the sample – tedious

Debertin and Schötzig, NIM 158 (1979) 471

Approximations:

$$\int_V \varepsilon^P(E_i;\vec{r})\cdot\eta^P(E_j;\vec{r})\,dV \approx \sqrt{\int_V \varepsilon^P(E_i;\vec{r})\cdot\varepsilon^P(E_i;\vec{r})\,dV}\cdot\sqrt{\int_V \eta^P(E_j;\vec{r})\cdot\eta^P(E_j;\vec{r})\,dV}$$

$$\int_V \varepsilon^P(E_p;\vec{r})\cdot\varepsilon^P(E_q;\vec{r})\,dV \approx \sqrt{\int_V \varepsilon^P(E_p;\vec{r})\cdot\varepsilon^P(E_p;\vec{r})\,dV}\cdot\sqrt{\int_V \varepsilon^P(E_q;\vec{r})\cdot\varepsilon^P(E_q;\vec{r})\,dV}$$

$$l^2(E)=\frac{\int_V \varepsilon^P(E;\vec{r})\cdot\varepsilon^P(E;\vec{r})\,dV}{\left[\int_V \varepsilon^P(E;\vec{r})\,dV\right]^2}$$

Blaauw and Gelsema, NIMA 505 (2003) 315

Third efficiency curve, or LS curve (ORTEC)

$$\int_V \varepsilon^P(E_i;\vec{r})\cdot\eta^P(E_j;\vec{r})\,dV \approx l(E_i)\cdot\varepsilon(E_i)\cdot l(E_j)\cdot\eta(E_j)$$

Blaauw and Gelsema evaluate the $l(E)$ curve on the basis of the analysis of an experimental spectrum with coincidence summing effects

Vidmar and Korun develop a Monte Carlo procedure for this evaluation; they apply EFFTRAN, based on the effective solid angle method (Vidmar and Korun, NIMA 556 (2006) 543)

Other methods:

-Use of a map of point source efficiencies and of a computed P/T ratio neglecting the changes due to the sample (Kolotov and Koskelo, JRNC 233 (1998) 95)

-Use of the transfer method for the evaluation of the efficiencies inside of the sample (Piton, Lepy et al., ARI 52 (2000) 791) – implemented in ETNA

-The application of the predictions of the virtual point detector model for the evaluation of the dependence of the efficiencies on the position (Rizzo and Tomarchio, ARI 68 (2010) 1448)

-Application of an exponential approximation for the dependence of the point source efficiencies on the emission point (Korun and Martinčič, NIMA 355 (1995) 600)

-Application of the quasi-point source formulae, with different prescriptions to the computation of the P/T, e.g. geometry independent, proportional with the ratio of attenuation coefficients (De Felice et al., ARI 52 (2000) 745)

Well-type detectors: very high coincidence summing effects
-small volume samples, big solid angle
=> 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

Decay data should be combined with proper efficiency values

Methods differ by the way in which decay data are evaluated and by the way in which they are coupled with efficiencies

Methods for the evaluation of decay data: deterministic and stochastic

Deterministic methods intimately coupling decay data and efficiencies

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:

$$b(i,j) = x_t(i,j) \cdot \left[1 - \frac{\eta(i,j)}{1 + \alpha_t(i,j)} \right]$$

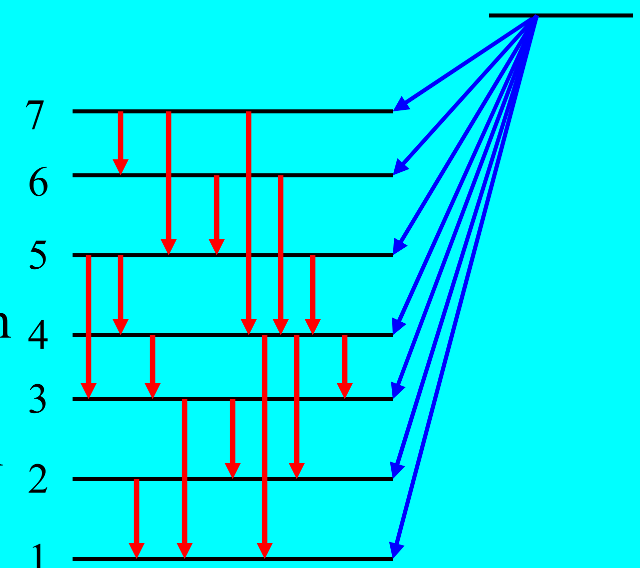
Probability of no signal in the detector following the transition if the initial level is already populated

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

Rigurous procedure, simple programming; disadvantage – complicate coupling of the decay data with efficiencies – not appropriate for volume sources and Monte Carlo simulation. Does not account for sum peaks corresponding to non-connected transitions

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

Ideea: *a*(*i,j*) 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

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

With $N=\mathrm{diag}[(F\ B)_i$ and $M=\mathrm{diag}(B_{i,1})$ the matrix of the probability of detecting the complete energy in any possible transitions per one decay is given by:

$$S= N\ A\ M$$

Extensions: Korun and Martinčič, NIMA 325 (1993) 478 – inclusion of X-rays

Advantage: mathematical computation; the complete matrix of total energy deposition; disadvantage: intrinsic coupling of decay data and efficiencies, does not account for sum peaks from non-connected transitions

Deterministic methods decoupling decay data and efficiencies

3. Deterministic calculation of joint emission probabilities
Schima and Hopes, IJARI 34 (1983) 1109 – for pair coincidences

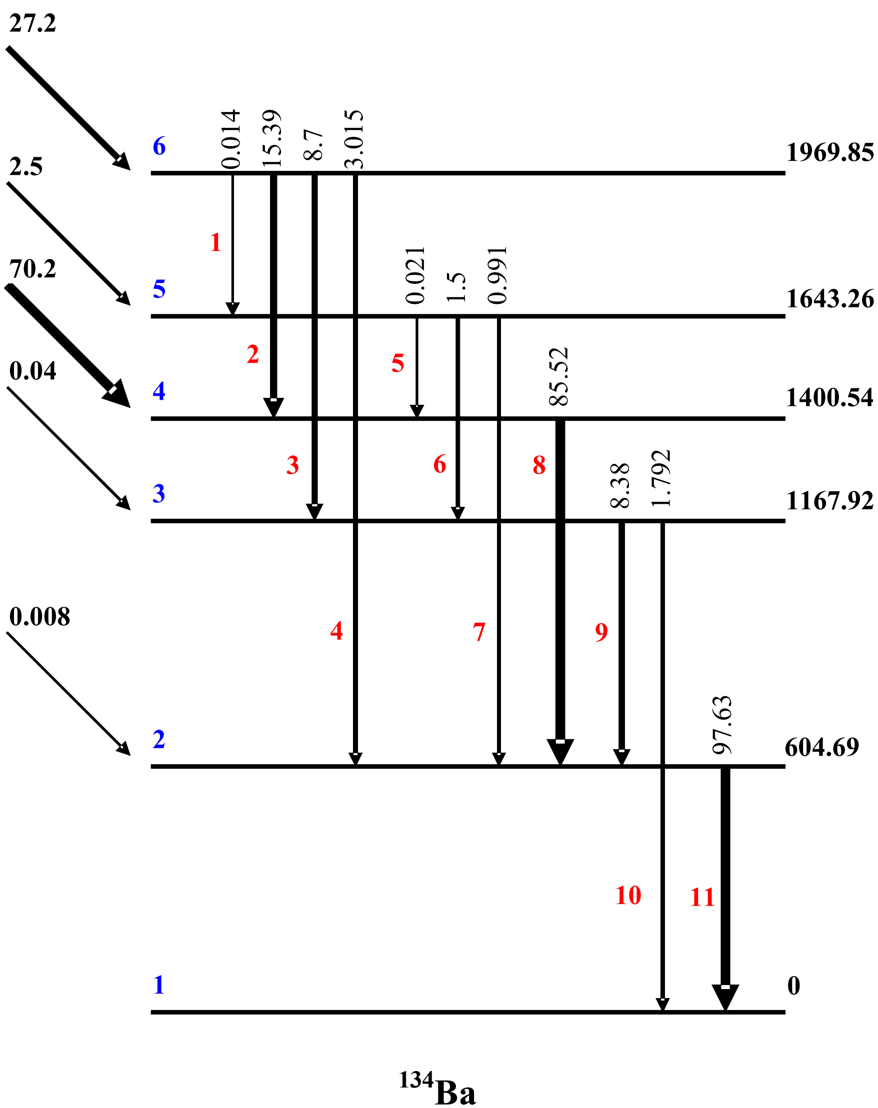
General procedure: Sima and Arnold, ARI 66 (2008) 705
- any decay scheme with less than 100 levels,
- all orders,
- grouping the contributions by transition levels, not by energy,
- inclusion of metastable states,
- inclusion of sum peaks with X-rays (two groups K_{α} and K_{β}) up to 10 X rays contributing,
- inclusion of the contribution of positron annihilation,
- simple possibility to include angular correlation

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

Advantage: decoupling of the decay scheme evaluation from the efficiencies => appropriate for Monte Carlo evaluation of the efficiencies, the decay data are analyzed and the emission probabilities are computed before Monte Carlo simulation

=> Implemented in GESPECOR

Example: ^{134}Cs



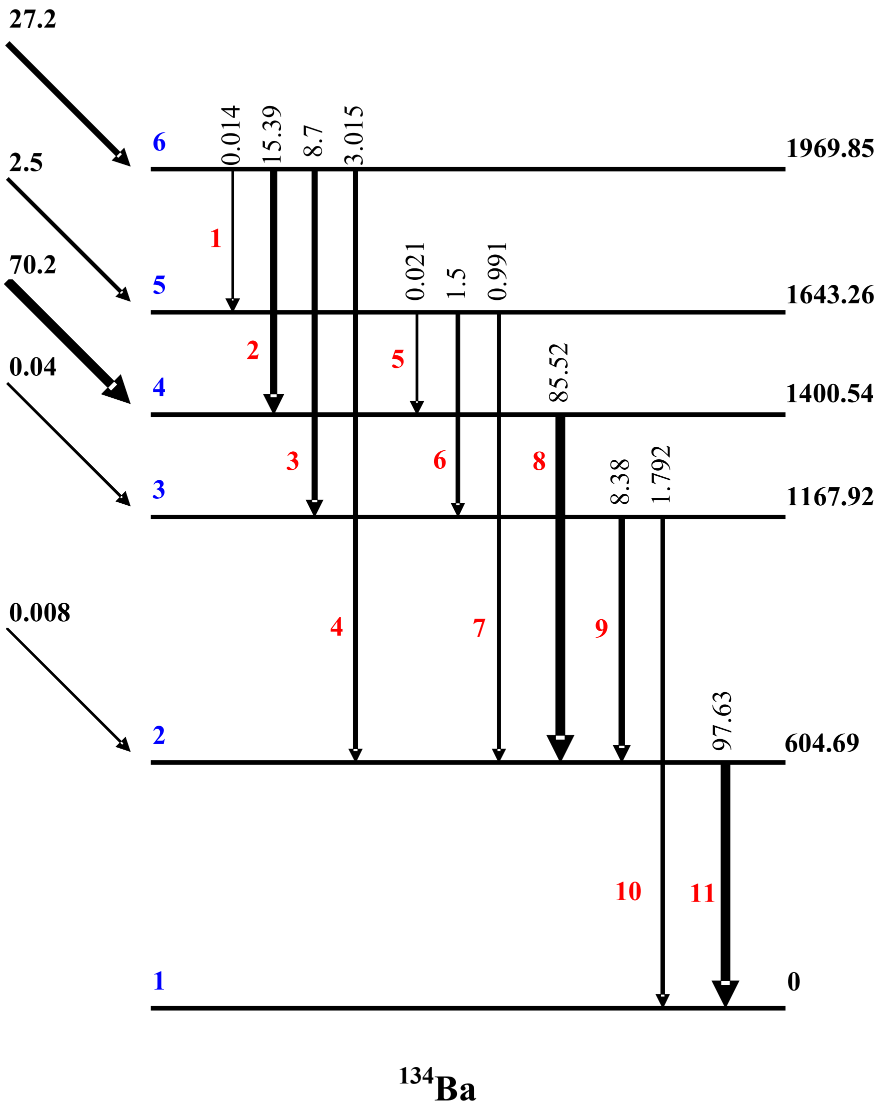
Problems:

1. Find all groups $M^{(n)}$ containing n γ photons that can be emitted together in the same decay act
2. Find all sets $S^{(n)}(k_1)$, $S^{(n)}(k_1,k_2)$, … of n photons that can be emitted together with the γ photon with index k_1 or with the pair (k_1,k_2) …
3. Find all sets $U^{(n)}(k_1)$, $U^{(n)}(k_1,k_2)$, … containing n photons in linked transitions between the same levels as the k_1 photon or the same levels as the k_1 and the k_2 photons …
4. Add K_α and K_β X-rays to the groups (when appropriate)

=> Efficient solution: search algorithm of the breadth-first type applied in solving graph theory problems.

=> Finding the sets defined above equivalent with finding the paths that satisfy specific conditions in an oriented graph.

Example: ^{134}Cs

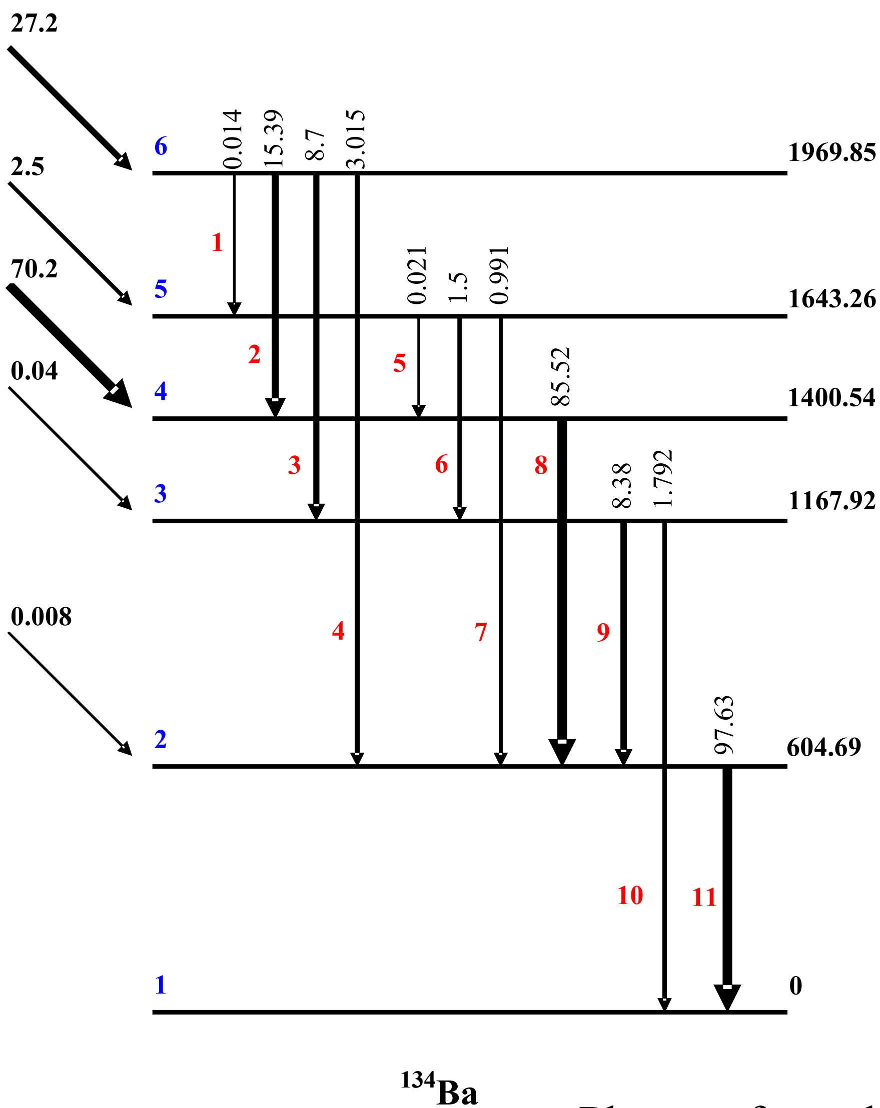


1. Find all groups $M^{(n)}$ containing n γ photons that can be emitted together in the same decay act

$M^{(1)}$ = {{1}, {2}, {3}, {4}, {5}, {6}, {7}, {8}, {9}, {10}, {11}}
$M^{(2)}$ = {{1,5}, {1,6}, {1,7}, {1,8}, {1,9}, {1,10}, {1,11}, {2,8}, {2,11}, {3,9}, {3,10}, {3,11}, {4,11}, {5,8}, {5,11}, {6,9}, {6,10}, {6,11}, {7,11}, {8,11}, {9,11}}
$M^{(3)}$ = {{1,5,8}, {1,5,11}, {1,6,9}, {1,6,10}, {1,6,11}, {1,7,11}, {1,8,11}, {1,9,11}, {2,8,11}, {3,9,11}, {5,8,11}, {6,9,11}}
$M^{(4)}$ = {{1,5,8,11}, {1,6,9,11}}

Efficient search algorithm

Example: ^{134}Cs



2. Find all sets $S^{(n)}(k_1)$, $S^{(n)}(k_1,k_2)$, … of n photons that can be emitted together with the γ photon with index k_1 or with the pair (k_1,k_2) …

If S belongs to the set $S^{(n)}(k_1)$, then $\{k_1,S\}$ belongs to $M^{(n+1)}$; if S belongs to the set $S^{(n)}(k_1,k_2)$, then $\{k_1,k_2,S\}$ belongs to $M^{(n+2)}$

Example: k=5

$M^{(2)}$ = {{1, 5}, {1,6}, {1,7}, {1,8}, {1,9}, {1,10}, {1,11}, {2,8}, {2,11}, {3,9}, {3,10}, {3,11}, {4,11}, {5,8}, {5,11}, {6,9}, {6,10}, {6,11}, {7,11}, {8,11}, {9,11}}

=> $S^{(1)}(5)$ ={{1}, {8}, {11}}

$M^{(3)}$ = {{1, 5,8}, {1, 5,11}, {1,6,9}, {1,6,10}, {1,6,11}, {1,7,11}, {1,8,11}, {1,9,11}, {2,8,11}, {3,9,11}, {5,8,11}, {6,9,11}}

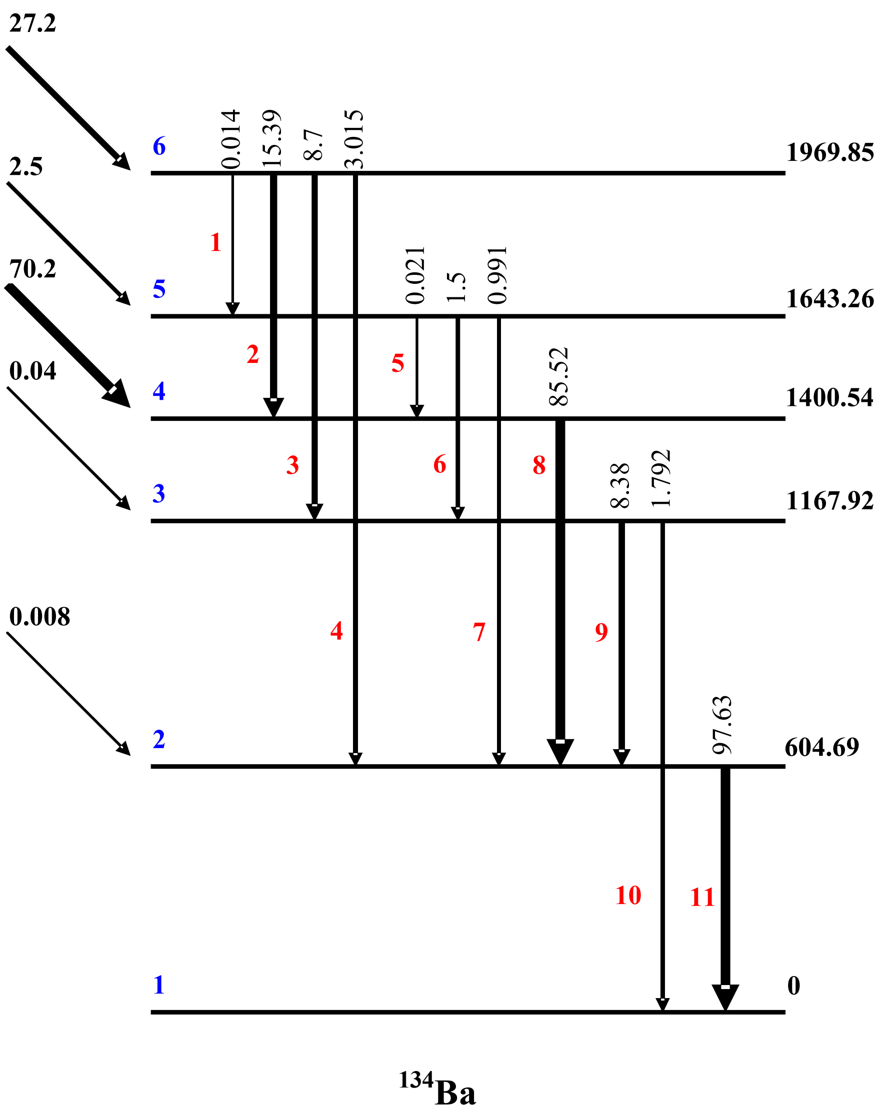
=> $S^{(2)}(5)$ ={{1,8}, {1,11}, {8,11}}

$M^{(4)}$ = {{1, 5,8,11}, {1,6,9,11}}

=> $S^{(3)}(5)$ ={{1,8,11}}

Photons from the set $S^{(n)}(k_1)$ give n-th order coincidence losses from the k_1 peak.

Example: ^{134}Cs



3. Find all sets $U^{(n)}(k_1)$, $U^{(n)}(k_1,k_2)$, … containing n photons in linked transitions between the same levels as the k_1 photon or the same levels as the k_1 and the k_2 photons …

Photon groups from $U^{(n)}(k_1)$ give sum peak contributions in the peak of k_1 photon, photon groups from $U^{(n)}(k_1,k_2)$ give sum peak contributions to the sum peak of the photons k_1 and k_2

Example: k=4

$U^{(2)}(4) = \{\{1,7\}, \{2,8\}, \{3,9\}\}$

$U^{(3)}(4) = \{\{1,5,8\}, \{1,6,9\}\}$

Losses from sum peaks:

If $\{l_1,l_2,..l_n\}$ belongs to the set $U^{(n)}(k_1)$, then $S^{(m)}(k_1)=S^{(m)}(l_1,l_2,..l_n)$; if $\{l_1,l_2,..l_n\}$ belongs to the set $U^{(n)}(k_1,k_2)$, then $S^{(m)}(k_1,k_2) = S^{(m)}(l_1,l_2,..l_n)$

The groups M, S, U are efficiently found even for complex decay schemes (100 levels) by an algorithm similar with algorithms developed for solving path problems in graph theory.

After finding the groups, the joint emission probabilities are easily computed.

The count rate in the peak of photon *i* in the case of a point source is:

$$R(E_i) = A \cdot p_i \cdot \varepsilon(i) - A \cdot \sum_{j \in S^{(1)}(i)} p_{i,j}\, \varepsilon(i)\eta(j) + A \cdot \sum_{j,k \in S^{(2)}(i)} p_{i,j,k}\, \varepsilon(i)\eta(j)\eta(k) - \ldots\ldots$$

$$+ A \cdot \sum_{q,r \in U^{(2)}(i)} \left[p_{q,r}\, \varepsilon(q)\varepsilon(r) - \sum_{j \in S^{(1)}(i)} p_{q,r,j}\, \varepsilon(q)\varepsilon(r)\eta(j) + \sum_{j,k \in S^{(2)}(i)} p_{q,r,j,k}\, \varepsilon(q)\varepsilon(r)\eta(j)\eta(k) - \ldots\ldots \right]$$

$$+ A \cdot \sum_{q,r,s \in U^{(3)}(i)} \left[p_{q,r,s}\, \varepsilon(q)\varepsilon(r)\varepsilon(s) - \sum_{j \in S^{(1)}(i)} p_{q,r,s,j}\, \varepsilon(q)\varepsilon(r)\varepsilon(s)\eta(j) + \sum_{j,k \in S^{(2)}(i)} p_{q,r,s,j,k}\, \varepsilon(q)\varepsilon(r)\varepsilon(s)\eta(j)\eta(k) - \ldots\ldots \right]$$

In the case of volume sources the products of efficiencies are replaced by integrals of the products of efficiencies

Other deterministic algorithms:

Novković et al., NIMA 578 (2007) 207, based on symbolic list manipulation

- in case of complex decay schemes, difficult to combine various decay paths that correspond in fact to transitions between the same levels

Test of several algorithms for compatibility:

Kanisch, Vidmar and Sima, ARI 67 (2009) 1952

- the algorithms of Sima and Arnold (GESPECOR), 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

Random simulation of the decay paths – see the separate lecture

6. Application - GESPECOR

Computation of coincidence summing corrections with GESPECOR:
Sima, Arnold and Dovlete, JRNC 248 (2001) 359; Sima and Arnold, ARI 53 (2001) 51

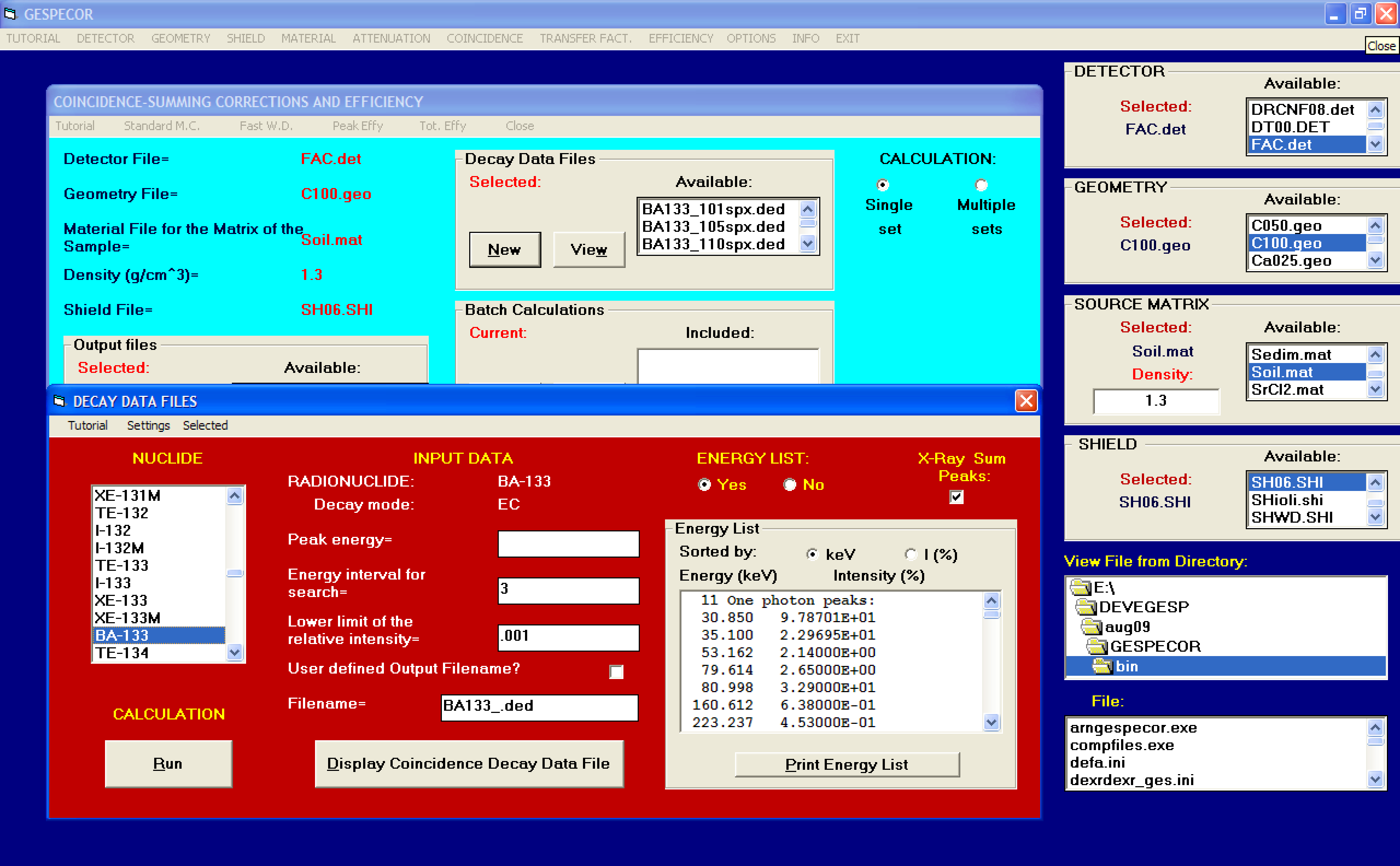
- automatic preparation of the required decay data for approx. 250 nuclides
    - deterministic method of computation (Sima and Arnold, ARI 66 (2008) 705)
    - includes all higher order terms
    - includes sum peaks with K X-ray contribution (Arnold and Sima, ARI 64 (2006) 1297)

- realistic description of the detector and sample, with arbitrary, but known, composition
    - closed end coaxial and well-type HPGE detectors
    - cylindrical and Marinelli beaker samples + many extensions

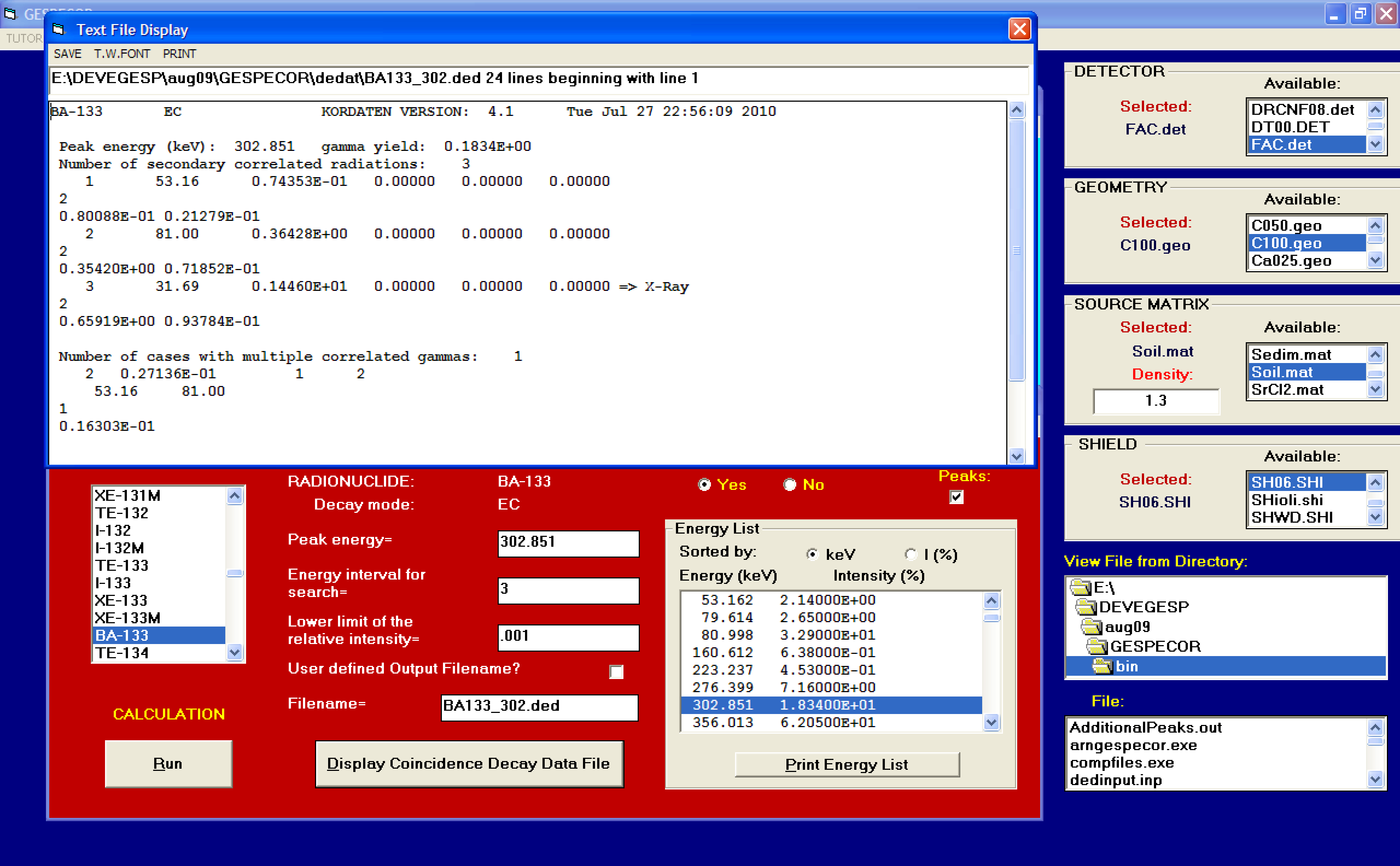
- peak oriented computation of the coincidence summing correction factors

- Computation of the efficiencies:
  - realistic Monte Carlo computation of the effective total efficiency and of the weighted peak efficiencies (Arnold and Sima, JRNC 248 (2001) 365)
    - powerful variance reduction techniques implemented
  - fast computation for quasi-point sources using for peak and total efficiencies input files instead of Monte Carlo computation
  - fast computation for well-type detectors based on an analytic formula for the total efficiency (Sima, NIMA 450 (2000) 98) and on measured peak efficiency
  - convenient possibility to implement angular correlation

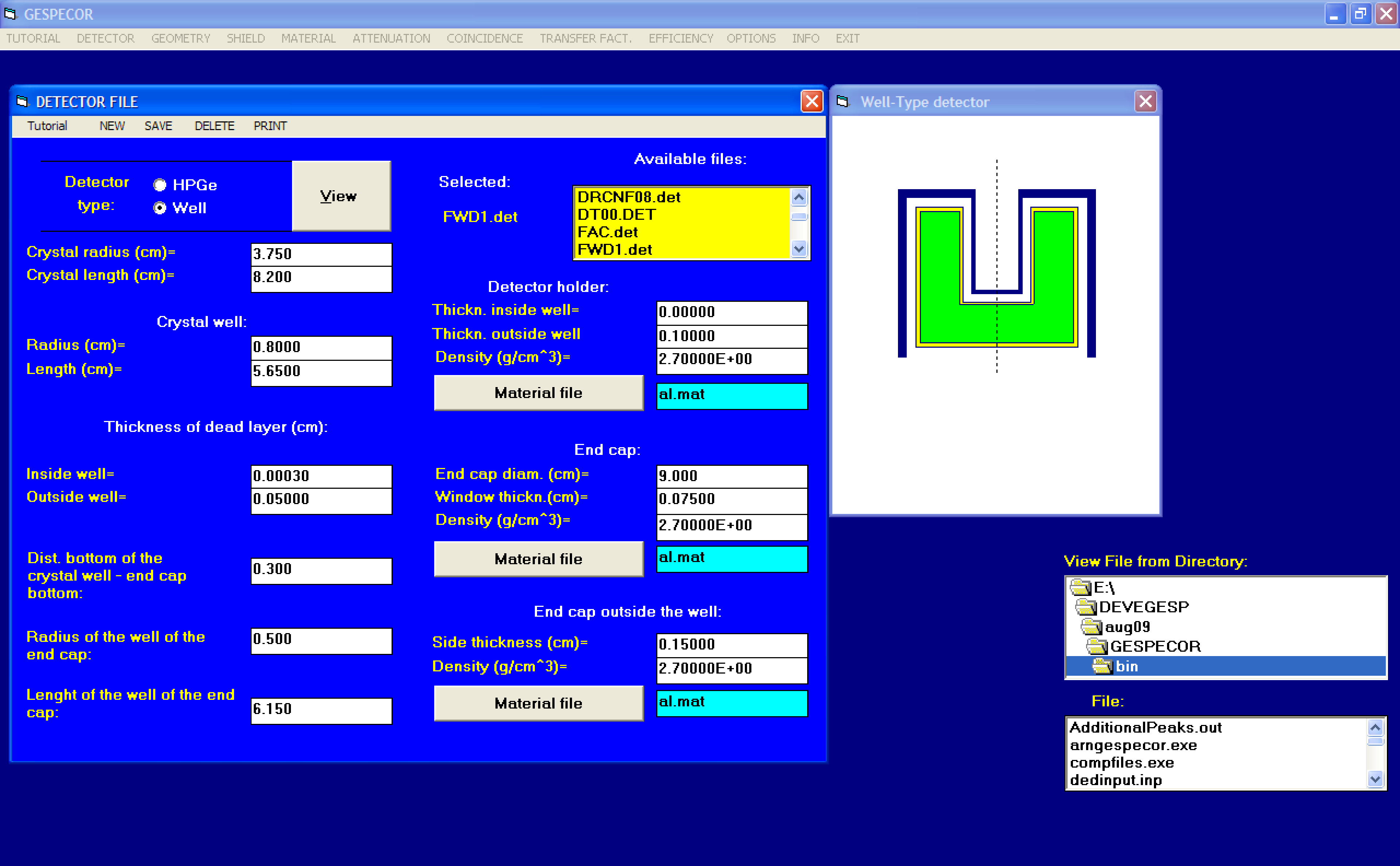
Validated both with respect to experimental data and with respect to other methods of computation (Kanisch et al., ARI 67 (2009) 1952)



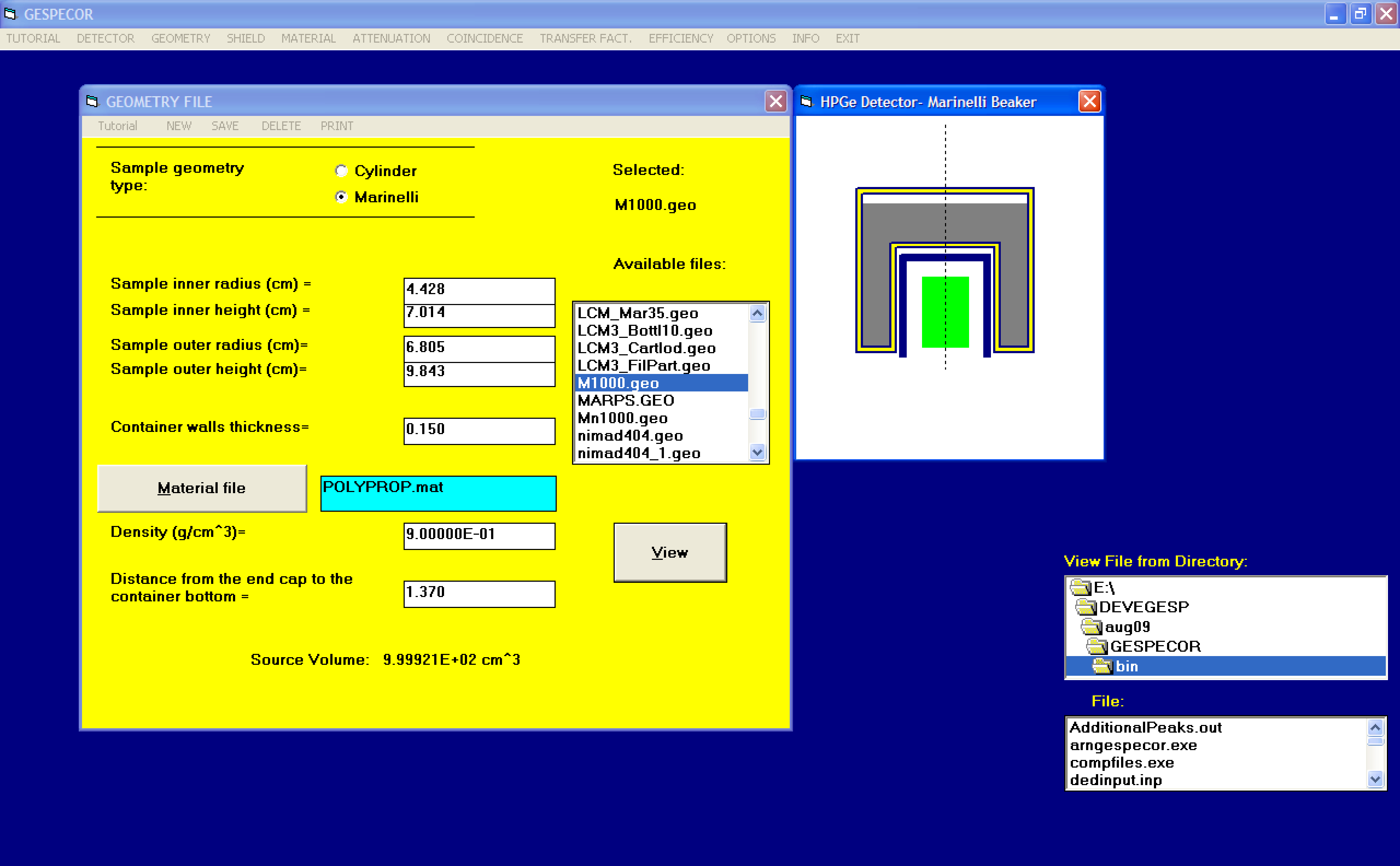
Selection of the nuclide for preparing the decay data file including joint emission probabilities for groups of photons



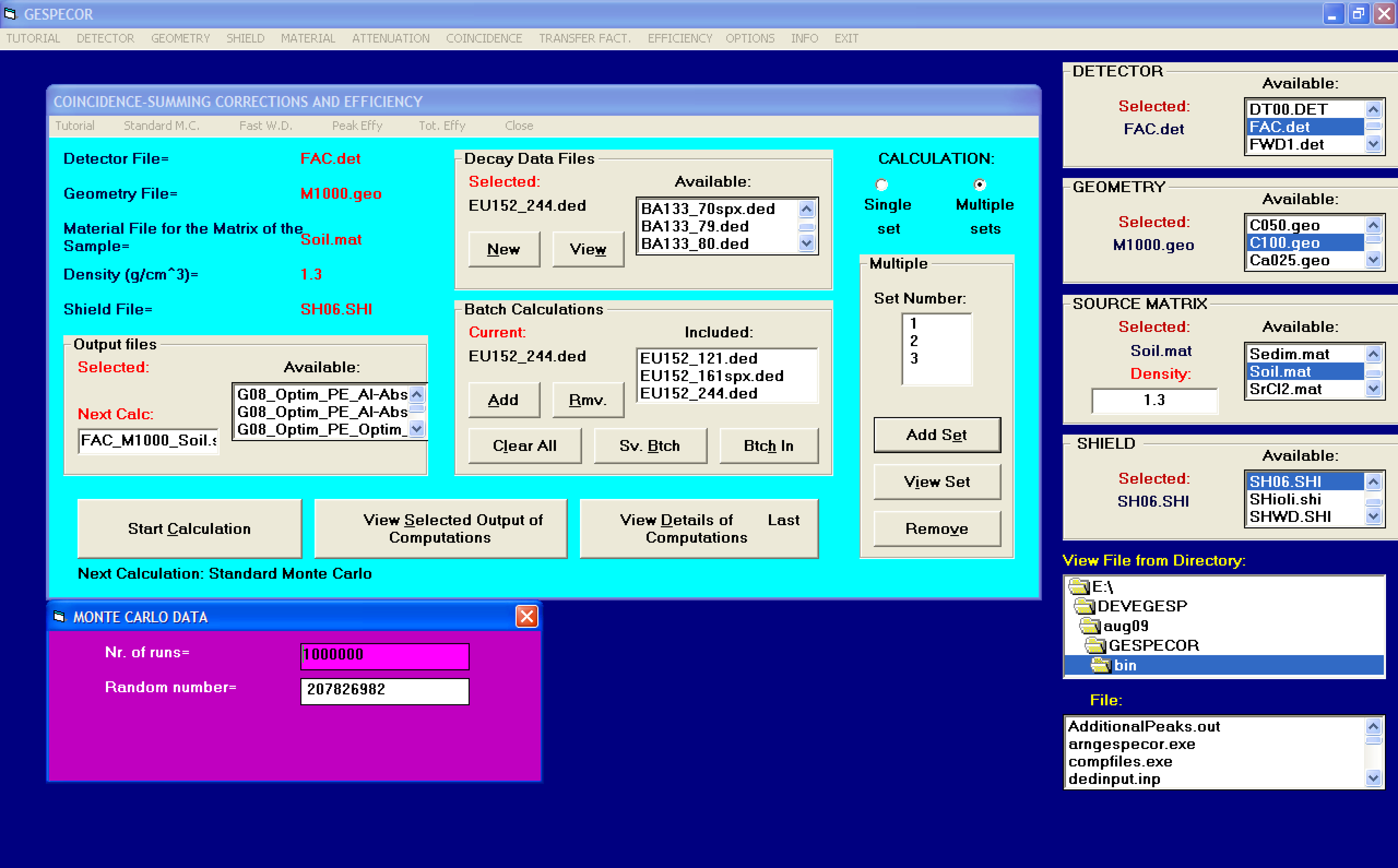
Part of the decay data file for the peak with energy 302 keV of ^{133}Ba



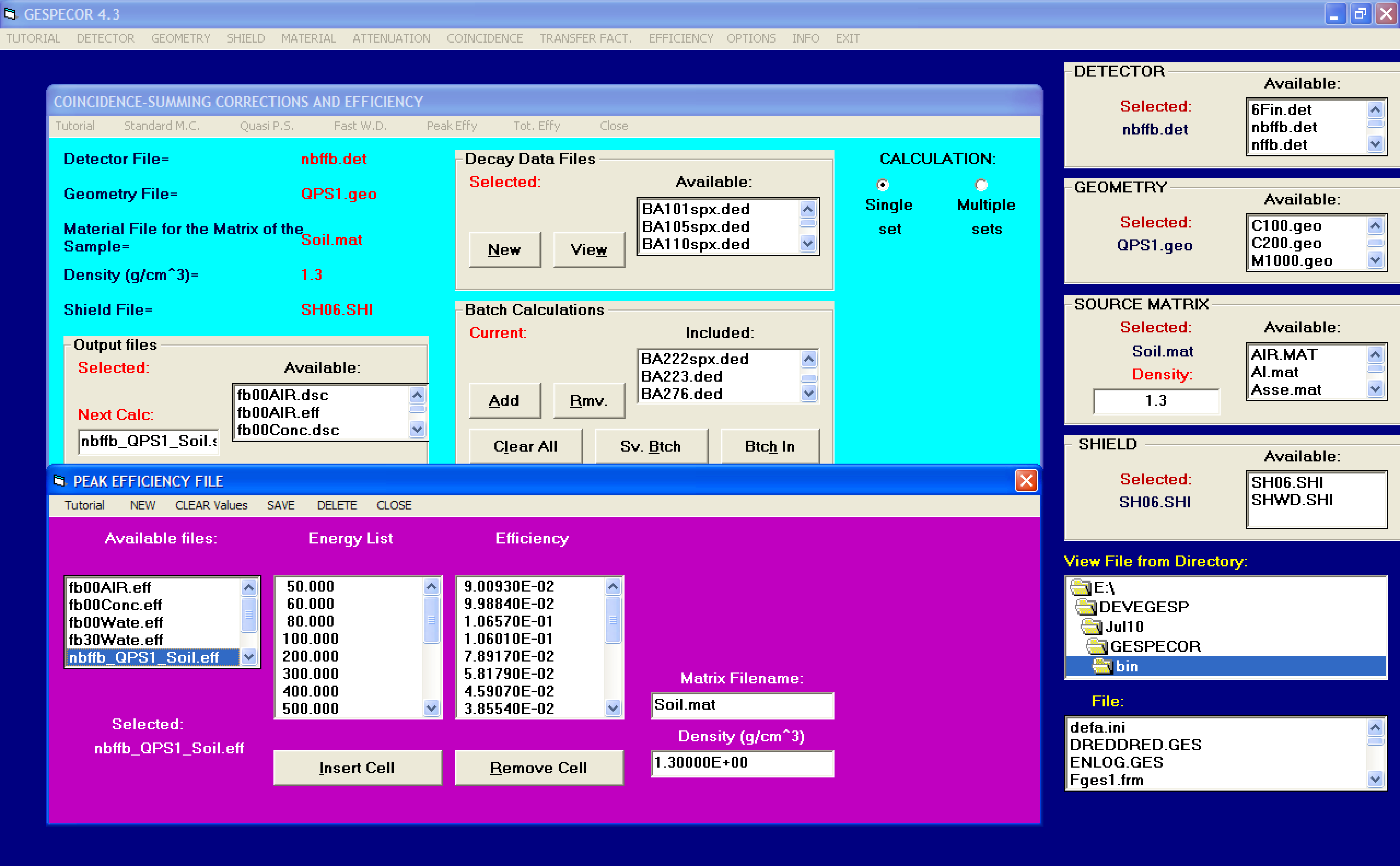
Detector data for a well-type detector



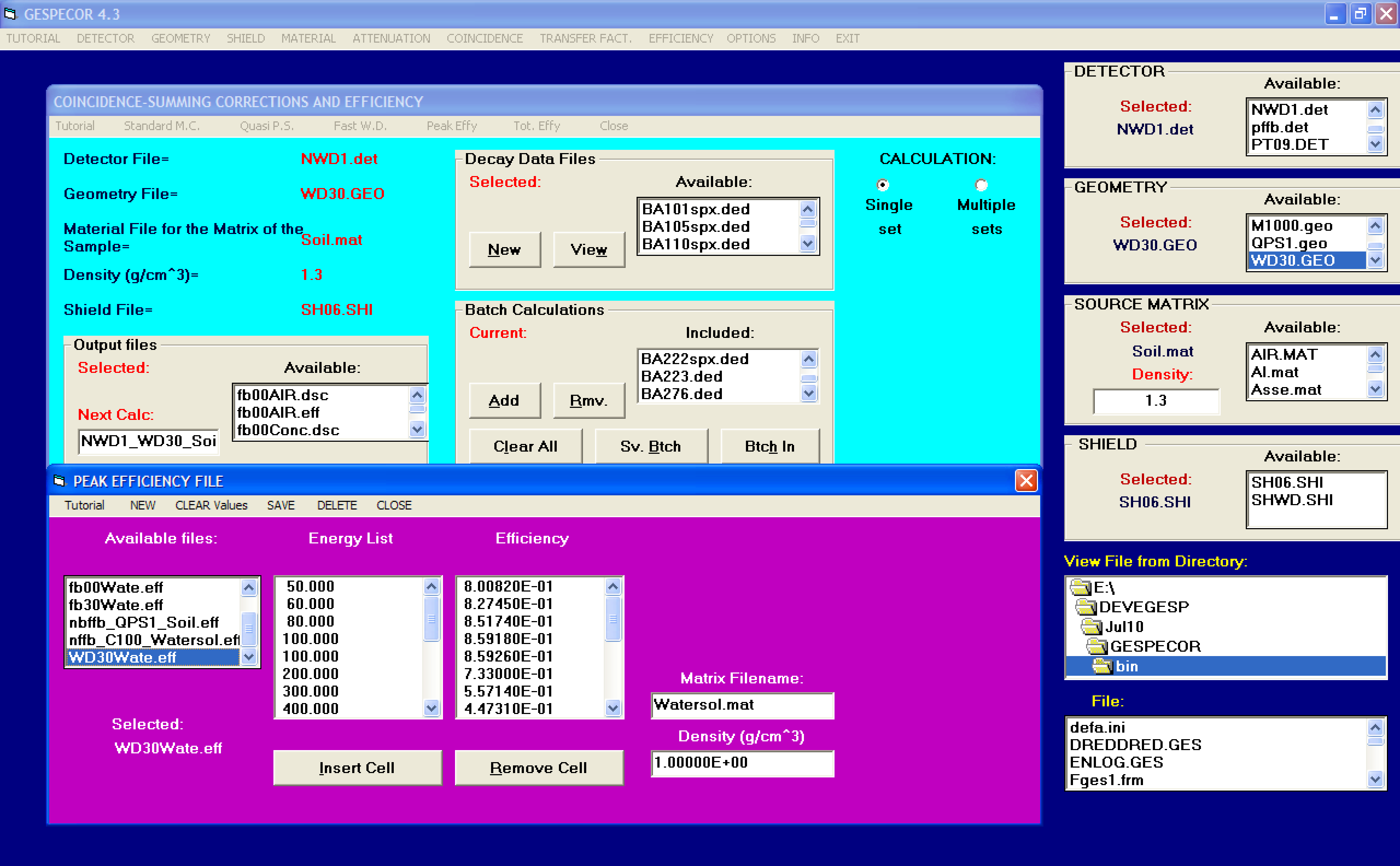
Geometry input file for a Marinelli beaker sample



Main interface of GESPECOR for standard computation of coincidence summing corrections



Selection of the input peak efficiency data file for the computation of the coincidence summing corrections in the case of quasi-point sources



Selection of the input peak efficiency data file for the computation of the coincidence summing corrections in the case of analytical computation of the total efficiency for well-type detectors

7. 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 for calibration and for measurement

For activity assessment the coincidence summing correction factors F_C for principal peaks should be evaluated

For spectrum analysis (especially 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