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The Correlations between the Emission Probabilities of the More Intense Gamma Rays in ^{152}Gd and ^{152}Sm Following ^{152}Eu Decay

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The variance matrix between the emission probabilities of the strong gamma-rays following the β decay of ^{152}Eu were determined in a specially designed experiment. The emission probabilities are deduced from the decay scheme, whose branching-ratios and beta feeding fractions were fitted to the observed peak areas.

KEYWORDS: radioactivity, ^{152}Eu, efficiency calibration, emmission probabilities, covariances

I. Introduction

The full energy absorption efficiency for gamma-ray detectors can be conveniently calibrated with multigamma ray sources by a least-squares fit to the peak areas properly corrected for secondary detection effects like sum and pile-up. However, this procedure requires the covariance matrix of the gamma-ray emission probabilities of the calibration source, which cannot be easily deduced from the existing data if it can be determined at all. Therefore, we performed a specially designed experiment to determine this correlation matrix.

In order to determine the correlations between gamma-ray emission probabilities, we start calibrating both the total and the total energy absorption (peak) detector efficiencies. Then we proceed to determine the decay parameters: branching-ratios and beta feeding fractions, with their full variance matrix, by fitting the observed peak areas with appropriate formulas deduced from the decay scheme, taking into account the secondary detection effects, particularly summing. Finally, from these data and the decay scheme, we calculate the emission probabilities along with the variance matrix. Here, we present the results obtained by the application of this method to the more intense gamma rays of the β decay of ^{152}Eu.

II. Modeling the ^{152}Eu Decay

^{152}Eu decays by β^- to ^{152}Gd, and by electron capture and β^+ to ^{152}Sm. **Table 1** shows the number of transitions in each daughter nuclide, ranked by the emission probability value.

Table 1 Number of observed gamma transitions following the decay of ^{152}Eu, according the emission probability per decay.

Interval of emission probability	daughter ^{152}Sm	^{152}Gd	both
$P > 1\%$	9	5	14
$1\% > P > 0{,}1\%$	21	6	27
$0{,}1\% > P > 0{,}01\%$	25	18	43
$0{,}01\% > P$	19	14	33
all	74	43	117

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The appropriated gamma transitions for calibration purposes correspond to those with emission probabilities greater than 1%, which form a consistent set with energies varying from 122 to 1408 keV. However, the knowledge of the full decay scheme and many other gamma transitions is required to correct for the secondary detection effects and to determine the correlations between the emission probability values.

1. The Relation between Peak-areas and Decay Parameters

The number of counts in a full-energy peak observed with HPGe detectors depends on a number of quantities related to the decay scheme and also on many secondary detection effects. The formulas are well described in the literature.[1-5] We define the following symbols:

Ω = number of nuclei that decayed during the live counting time;

P_i = population of level i ;

f_i = fraction of decay to level i;

E_{ij}= energy of the transition from level i to level j;

$\alpha_{Eij} = \alpha_{ij}$ = internal conversion coefficient of transition E_{ij} between levels i,j;

$\varepsilon(E)$ = absolute full-energy-peak efficiency at energy E ;

$\varepsilon_t(E)$ = absolute efficiency for the absorption of any quantity of energy for gamma-rays at energy E ;

κ_{ij} = branching ratio of electromagnetic transition from level i to level j, relative to the decay of level i;

$\phi_{(ij)(kl)}$ = conditional probability of transition E_{kl} given that E_{ij} occurs, and

$W(E_{ij}, E_{kl})$ directional correlation of the transitions E_{ij} and E_{kl}, averaged over the detector's solid angle.

Note that the term *population* is being applied to the number of the daughter nuclei that were formed in level i during the observation, then $\sum P_i > \Omega$. However, $\sum f_i = 1$ and $\sum_{j=0}^{i-1} \kappa_{ij} = 1$. There is also a scheme dependent relation between $\phi_{(ij)(kl)}$ and the product of the branching-ratios of the involved transitions.

The zeroth-order term for the peak-area corresponding to the gamma-ray with energy E_{ij} decaying from level i is simply

$$N\left(E_{ij}\right) = P_i \kappa_{ij} \frac{\varepsilon\left(E_{ij}\right)}{1+\alpha_{ij}} \quad (1)$$

The first-order correction is due to sum between coincident gamma rays. It can give a positive contribution to the area of the peak located at the energy of the cross-over transition $E_{ik} = E_{ij} + E_{jk}$,

$$N\left(E_{ij}+E_{jk}\right) = P_i \kappa_{ij} \kappa_{jk} \frac{\varepsilon\left(E_{ij}\right)}{1+\alpha_{ij}} \frac{\varepsilon\left(E_{jk}\right)}{1+\alpha_{jk}} W\left(E_{ij}, E_{jk}\right) \quad (2)$$

The summing effect subtracts events in the peak-area corresponding to the gamma-ray with energy E_{ij} by an amount given by

$$\overline{N}\left(E_{ij} | E_{kl}\right) = \begin{cases} P_i \phi_{(ij)(kl)} \frac{\varepsilon(E_{ij})}{1+\alpha_{ij}} \frac{\varepsilon_t(E_{kl})}{1+\alpha_{kl}} W\left(E_{ij}, E_{kl}\right) & i > k \\ P_k \phi_{(kl)(ij)} \frac{\varepsilon_t(Eij)}{1+\alpha_{ij}} \frac{\varepsilon(Ekl)}{1+\alpha_{kl}} W\left(E_{kl}, E_{ij}\right) & i < k \end{cases} \quad (3)$$

for each gamma-ray E_{kl} in time coincidence with E_{ij}. The total loss due to summing effects is

$$\overline{N}(E_{ij}) = \sum_{kl} \overline{N}\left(E_{ij} | E_{kl}\right).$$

2. Application to ^{152}Eu

We took all the examples from the more intense gamma-ray transitions from the β^- decay of ^{152}Eu, which are presented in **Figure 1**. For the transitions with emission probabilities greater than 0.1%, the corresponding gamma peak areas were observed, and the decay parameters - branching-ratios and feeding fractions - fitted by using expressions derived from the equations 1-3 above. As example, the equation for the area of the peak corresponding to γ_{10-3} at 678 keV, correct to first-order in the detection effects, is:

$$A_{10-3} = \frac{\varepsilon\left(E_{10-3}\right)\Omega}{1+\alpha_{10-3}} \left(f_{10} + f'_{10}\right) \kappa_{10-3} \times \left(1 - \frac{\varepsilon_t\left(E_{3-1}\right)}{1+\alpha_{3-1}} - \frac{\varepsilon_t\left(E_{1-0}\right)}{1+\alpha_{1-0}}\right) \quad (4)$$

where W was taken equal to 1, and f'_i equals the sum of the absolute intensity of the transitions not observed, calculated from the decay scheme.

Besides the correction due to f'_i, the transitions that are not observed must be taken into account in the constraint on the branching-ratios sum, $\sum_{j=0}^{i-1} \kappa_{ij} = 1$, that is broken in the observed and unobserved parts,

$$\sum_{j,observed} \kappa_{ij} = 1 - \sum_{j,unobserved} \kappa_{ij}$$

Therefore, from all the ^{152}Gd levels observed, **Figure 1**, only four branching-ratios are free parameters to fit: κ_{13-1} ; κ_{10-1}; κ_{10-3} ; κ_{7-1} .

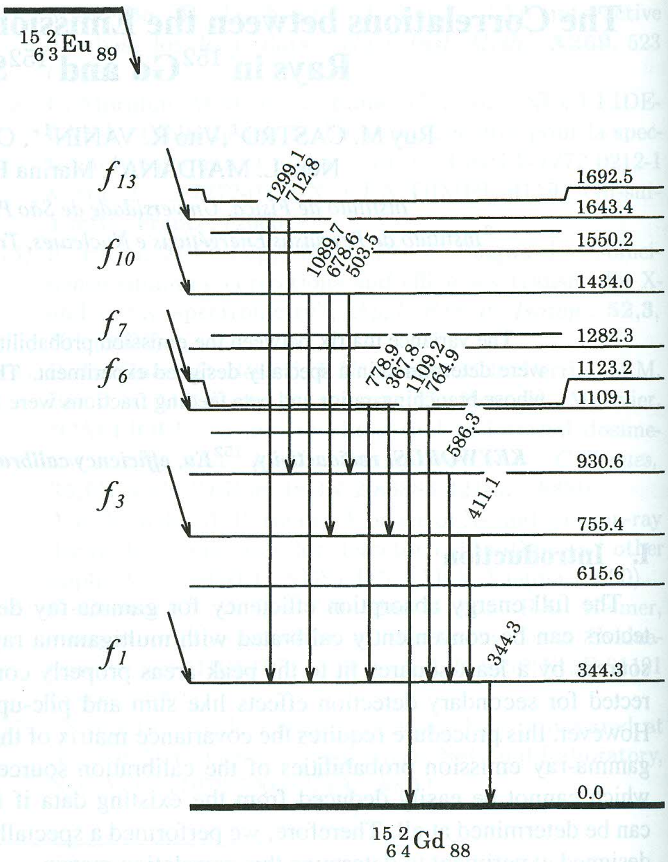

Fig. 1 Part of ^{152}Eu decay scheme, showing the gamma-ray transitions that furnished data for the fit and the beta feeding fractions considered as parameters.

III. The Detection System and the Efficiency Calibration

The system consists in a large HPGe detector (160 cm^3), with the source placed 20 cm away the detector capsule. Calibrated sources of ^{88}Y, ^{60}Co, ^{54}Mn with activities between 2 and 4 kBq calibrated in $4\pi - \beta\gamma$ detector were used, along with sources of ^{137}Cs,^{152}Eu and ^{133}Ba with ~ 30 kBq. The intention is to replace the low energy data of Eu and Ba with data coming from sources with simple decay schemes, like ^{57}Co and ^{51}Cr. We do understand that using strong sources, in particular of Eu and Ba, warrant bad results. However, this is the prototype of the experiment, which will provide a rough estimate of the correlations and not the emission probabilities.

The calibration function for the full energy peak was chosen as

$$\ln\left(\varepsilon\left(E\right)\right) = \sum_{k=0}^{4} c_k \ln\left(\frac{E}{E_b}\right)^k \quad (5)$$

where E_b = 1277.43 keV, chosen to reduce the correlation between the parameters c_k.

The total efficiency was determined from the total-to-peak ratio, measured with monochromatic sources, and the efficiency for the full energy peak.

IV. Determining the Quantities Related to the Decay Scheme

1. The Least-squares Procedure

All the gamma-ray transitions with emission probabilities greater than 0.1% were selected for observation except the 1109 keV (γ_{6-0}) that constitutes a doublet with the 1112 keV. Let us call N the number of observed peak areas and M the number of parameters to fit. We were left, therefore, with 11 equations for the peak areas of the transitions plotted as full lines in **Figure 1**, and 10 parameters: f_1 ; f_3 ; f_7 ; f_{10} ; f_{11} ; κ_{13-1} ; κ_{10-1}; κ_{10-3} ; κ_{7-1} ; and Ω, the number of disintegrations. The constraints: the total feeding by the β^- channel, and the sums of the branching-ratios for the levels 13, 10 and 7, were used to eliminate the parameters f_6 , and κ_{13-4} , κ_{10-4}, κ_{7-3}, respectively.

The model corresponding to the scheme in the figure and the assumed detection properties is expressed by formulas similar to (4), and can be fitted to the experimental data points by the design matrix

$$W_{\nu\mu} = \frac{\partial A(E_\nu)}{\partial q_\mu}\Big|_{\vec{q}} \ , \nu = 1,...\text{N and } \mu = 1,...\text{M}\,, \qquad (6)$$

where the parameter vector is $\vec{q}$, the first M-1 components being the decay scheme parameters f_i, κ_{ij}, and the last component $q_M = \Omega$. The matrix formulation for the least-squares method is explained in refs. [6] and [7].

The calculation of the variance matrix is a lengthy process. It has four components,

$$V_s = V_A + V_\varepsilon + V_t + V_\alpha, \qquad (7)$$

where V_A is the $M \times M$ matrix of the variances due to counting statistics, and V_ε, V_t and V_α are the variances due to the peak efficiency, the total efficiency and the conversion coefficients, respectively, propagated to the peak areas.

The fitting procedure is iterative because the formulas are not linear in the parameters, see formula (4). Once the iterative procedure converges, the variance matrix of the decay scheme data f_i and κ_{ij} is given by the upper left $(M-1) \times (M-1)$ sub-matrix of the variance matrix of the fitted parameters,

$$V_q = \left(W^t V_s^{-1} W\right)^{-1}.$$

2. Numerical Results

The disintegration data was obtained from a 20 kBq ^{152}Eu source, with the detector heavily shielded. The peaks were fitted with a gaussian plus exponential tails over a step and a second degree polynomial. The fit along the lines described above was performed, resulting in a chi-square value equal to 2.1 for the β^- part and 0.4 for the β^+ part, each one with one degree of freedom. In **Tables 2** and **3**, we list some of the decay parameters obtained with their standard deviations and the correlation matrix.

The overall precision is not good and also we do not have a reliable efficiency calibration. However, the results obtained are compatible with published data.

Table 2 Some of the ^{152}Eu β^- decay parameters obtained in the fit, with the correlation matrix. The first column presents the names of the parameters, the second the values with the standard deviations in parentheses. The rest of the table is the correlation matrix.

param.	value	correlation matrix				
		f_1	f_7	κ_{10-1}	κ_{10-3}	Ω
f_1	0.0731(17)	1.00	-0.98	-0.20	0.16	0.86
f_7	0.1436(12)	-0.98	1.00	0.09	-0.06	-0.81
κ_{10-1}	0.714(3)	-0.20	0.09	1.00	-0.94	-0.23
κ_{10-3}	0.1937(22)	0.16	-0.06	-0.94	1.00	0.18
Ω	123.2(11)E8	0.86	-0.81	-0.23	0.18	1.00

Table 3 Same of **Table 2** for ^{152}Eu β^+ decay.

param.	value	correlation matrix				
		f_{11}	f_{16}	κ_{9-1}	κ_{15-1}	κ_{15-9}
f_{11}	0.1222(12)	1.00	0.72	0.00	0.10	-0.09
f_{16}	0.02053(15)	0.72	1.00	-0.01	0.24	-0.24
κ_{9-1}	0.581(7)	0.00	-0.01	1.00	-0.04	0.04
κ_{15-1}	0.8473(11)	0.10	0.24	-0.04	1.00	-0.99
κ_{15-9}	0.1105(8)	-0.09	-0.24	0.04	-0.99	1.00

V. Determining the Emission Probabilities

With the decay scheme parameters of ^{152}Eu, all the gamma-ray transition probabilities can be determined by equations like the following, for the 411 keV gamma-ray:

$$I_{3-1} = \frac{\kappa_{3-1}}{1+\alpha_{3-1}}\Big\{ (f_3 + f_3') + (f_{10} + f_{10}')\,\kappa_{10-3} + \left(f_7 + f_7'\right)\Big(\sum_{j,observed} \kappa_{7j} - \kappa_{7-1}\Big)\Big\} \qquad (8)$$

The variance matrix can be deduced by using a propagation formula. The results are shown in **Tables 4** and **5**, for some of the gamma rays used for calibration purposes. Note that all the correlations are moderately important, which follows from properties of the decay scheme, reflected in the set of equations.

VI. Conclusion

We developed a method to determine the correlation matrix of the gamma-ray emission probabilities of the gamma rays following the decay of ^{152}Eu. In the experiment described here, we verified that the strong gamma rays form a very correlated data set. However, the emission probabilities do not constitute the simplest data set to be used when performing a precise efficiency calibration, when summing effects corrections are inevitable. Sum depends on the level feeding fractions from the parent nucleus and the branching ratios, which in turn form the easiest data set to determine when looking for

Table 4 Gamma-ray emission probabilities and correlation for some intense gamma-rays, calculated from the data on **Table 1**.

energy keV	intensity	correlation matrix 344	411	779	1089	1299
344	0.26712(18)	1.00	0.33	0.69	0.44	-0.29
411	0.0225(3)	0.33	1.00	0.46	0.73	-0.19
779	0.1354(12)	0.69	0.46	1.00	0.65	-0.37
1089	0.01817(9)	0.44	0.73	0.65	1.00	-0.27
1299	0.01685(9)	-0.29	-0.19	-0.37	-0.27	1.00

Table 5 Same of **Table 4**, for β^+.

energy keV	intensity	correlation matrix 121	964	1085	1212	1408
121	0.2862(4)	1.00	-0.23	-1.00	-0.64	-0.68
964	0.144(4)	-0.23	1.00	0.16	0.14	0.14
1085	0.1008(7)	-1.00	0.16	1.00	0.64	0.68
1212	0.01414(11)	-0.64	0.14	0.64	1.00	0.66
1408	0.2030(3)	-0.68	0.14	0.68	0.66	1.00

the gamma-ray emission probabilities. We conclude, therefore, that for efficiency calibration purposes it would be more convenient to determine and use the beta feeding fractions and the branching-ratios, with the respective variance matrix, being easy to transform the beta feeding fractions and the branching-ratios to gamma-ray emission probabilities.

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